

SAFE HAVENS AND HOT SPOTS: IONIZING
RADIATION IN MARTIAN PERMAFROST AND ITS INFLUENCE
ON THE SURVIVAL OF ANCIENT LIFE

By

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July, 2022

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Major department: Physics

Permafrost on Mars is considered a primary target for the search for life. On a microscopic scale permafrost is highly non-uniform and the effective radiation dose absorbed by a microbe, and thus its survival probability, depends strongly on its microscopic location and its environment. The goal of this work was to determine how long microbes will survive in a dormant state under the influence of radiation from radionuclides in the Martian regolith, and how fast radiation will destroy the remnants of dead microbes. Our approach was to model the radiation environment that microbes are exposed to in the Martian subsurface using the Geant4 Monte Carlo programs *DosiSed* and *DosiVox*.

This dissertation describes the methodologies and results of this study. The reliability and precision of *DosiVox* and *DosiSed* simulations were rigorously tested on dose distributions measured with $\text{Al}_2\text{O}_3\text{:C}$ dosimeters buried in terrestrial samples and by reproducing published data. The optimum simulation parameters were determined for the infinite dimensional models for the Martian subsurface and dose rate distributions were determined.

Finally, the results and limitations of our calculated survival probabilities and probabilities of preservation of biomarkers in the Martian subsurface are presented and discussed.

Our result shows that microbes can survive for as long 50 billion years depending on their location and whether they are exposed to the full spectrum of the radiation or only gamma radiation. For microbes that are exposed to only gamma radiation, the most

radio-resistant of them can survive up to 350 billion years while those that are exposed to the full spectrum will not survive for more than 500 million years.

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A Dissertation

Presented to the Faculty of the Department of Physics
East Carolina University

In Partial Fulfillment of the Requirements for the Degree
Doctor of Philosophy in Biomedical Physics

by

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July, 2022

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ACKNOWLEDGEMENTS

I want to start by acknowledging my Lord and Saviour Jesus Christ for his overwhelming and relentless love, without which everything is nothing.

This journey began with me assuming I knew what I was getting into, but little did I know, literally and figuratively. I am writing this final note all thanks to my adviser Dr. Regina DeWitt. I cannot thank her enough or show gratitude enough for these past few years I have worked with her. Her patience, kindness, and warm personality make her an exemplary professor. Her guidance and commitment to excellence has made me a better student, researcher and teacher. She inspired me to think big, and never failed to demand from me what she saw I was capable of, even when I could not see it. Her dedication, effort and patience during the editing of this dissertation cannot be overemphasized.

My humble respect goes to my committee members, Dr. Michael Dingfelder, Dr. Martin Bier, Dr. Jae Won Jung, and Dr. David Mallinson. Many thanks for all the precious time taken out of your busy schedules to help my advancement and to provide guidance to improve this work.

A special thanks to Dr. Loïc Martin for giving us the non-public version of his MC code *DosiSed* and for patiently assisting as I broadened my understanding of Monte Carlo (MC) simulations with *DosiSed* and *DosiVox*.

My sincere thanks to all the great staff and faculties of the Physics department, as they have made my time as a Ph.D. student at ECU memorable. It is not always easy to be far away from home, but so many smiles paired with heart felt well-being check-ins from staff and faculty made it easier. I would also like to show my gratitude to Gene Oakley for his contribution to our beta dose rate attenuation paper.

Many thanks to my friends: Steven Yuvan, Nichole Libby, Joel Pogue, Tristan Gaddis, and Shiva Bhandari. These individuals were essential in making each day unique and positive. I have enjoyed the many fruitful collaborations and discussions about medical physics as well as life in general. They were the pillars that supported me mentally and emotionally during

the different stages of this Ph.D. candidacy and I believe God placed each one of them in my life for this reason. I'm so grateful for our friendship.

I want to also thank Dr. Nathan Carlson and Mark Strausbauch for assisting in the calibration of our beta source. A dear and special thank you to my church family, Greenville Christian Fellowship. Thank you to Pastor Blake Dameron, for the many prayers and loving friendship. I would also like to thank my men's group at Greenville Christian Fellowship. Tackling life's challenges together has made such a tremendous impact in my life and relationship with God.

I must especially appreciate and thank my mother, Mrs. Helen Osunkwor. She is my rock. Her support financially, spiritually and emotionally is the reason why I am here writing this today. My degree is dedicated to her. I cannot forget to also thank my immediate family, Louis Osunkwor, Adesuwa Osunkwor, and Lugard Igharo. Their contribution and encouragement are deeply appreciated.

Last but not the least, I would like to appreciate my gorgeous fiancée. She has made my world a little brighter and my life a little better. I am so glad and grateful to God for bringing such a person in my life. I'm excited about my future with her. I can't wait for June 12, 2022.

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Chapter 1

Introduction

1.1 Radiation and the possibility of life on Mars

The atmosphere of Mars is about 100 times thinner than Earth's. Due to the absence of a "thermal blanket", Mars cannot retain any heat energy. As a result, the average temperature on Mars, is about 210 K (- 62 °C). In the summer months, surface temperatures may reach a high of about 293 K (20 °C) at noon at the equator, and a low of about 120 K (- 153 °C) at the poles during the winter months [1]. This thin atmosphere which is mainly composed of 96% carbon dioxide and 2% nitrogen compared to Earth's 78% nitrogen and 21% oxygen has a surface pressure of around 6 mbar compared to Earth's 1,013.25 mbar. Consequently, water, a solvent that is critical for the origin and persistence of life, exists only as ice. Therefore, it can be argued that the Martian surface is a cold, dry, and barren desert and thus not favorable for most living things. However, in the past, Mars was warmer and wetter and thus more favorable to life [2–5]. The search for life on Mars is one of the most intriguing scientific endeavors of planetary exploration, and is indeed the main purpose of the Mars 2020 Perseverance Rover.

In addition to the cold and dry conditions on the surface, the thin atmosphere of Mars does not offer protection against solar ultraviolet (UV) rays. UV radiation that readily

inactivates cells, is also responsible for the highly oxidizing layer on the surface of Mars and is hypothesized to be an organic molecule destroyer [6]. On top of all these adverse conditions, ionizing radiation has been identified as the main limiting factor to the presence of life on Mars. Energetic charged particles, like solar energetic particles (SEP) and Galactic cosmic rays (GCR) reach and penetrate the Martian surface. Mars lacks an internal dynamo capable of generating a large global magnetic field like that of Earth to stop these charged particles. Generally, the radiation environment on Mars is dominated by GCR, SEP, and natural radioactivity. The destructive effects of GCR and SEP are confined to the upper 3 m of the Martian regolith due to the shielding properties of the regolith [7]. At depths below 3 m, the radiation environment is dominated by radioactivity.

While there have been numerous studies on how GCR and SEP affect the habitability of Mars in general [4, 7–11], only a few studies have been dedicated towards radioactivity as a habitability factor on Mars [12–14]. One reason for this is that the ultimate goal of exploring Mars is to send humans on Mars so that the real science can be done *in situ* [15, 16]. Therefore, a lot of resources have been put into studying the level of GCR and SEP at the surface and the properties of superficial Martian regolith as a potential source of shielding material when human exploration begins. Consequently, there is plenty of information on the chemical composition and radionuclide concentrations of the Martian surface but little or nothing on the deeper subsurface where the radiation environment is dominated by only radioactivity. However, Taylor et al. [17] argued that the surface measurements on Mars should reflect the bulk composition of the Martian crust based on similar observations on Earth.

Another reason is that dose rate from natural radioactivity is relatively low and thus considered to pose no immediate threat to survival of organisms. However, while this is true for active microorganism populations (organisms that can repair radiation damage) [18–27], dormant organisms cannot repair radiation damage. Over a long period of time, they can accumulate doses that are high enough to be lethal. In fact, there are some studies suggesting

that the dose rate due to radioactivity in Martian rocks and regolith is high enough to destroy dormant organisms after a few million years [7, 12, 28]. However, even if these dormant organisms accumulate a lethal dose, their relic DNA may still be preserved due to frozen conditions for several millions of years providing evidence that could reflect their existence.

1.2 Dissertation focus

The Mars 2020 Rover is seeking signs of ancient life. However, the question arises if it is theoretically possible to find living microorganisms or at least their biological traces. Goal of this dissertation is to contribute to answering this question. If there was an ancient biosphere on Mars, the continued presence of living organisms depends on the radiation environment and the time required to accumulate a lethal dose. In the same manner, the radiation environment also determines the timeframe for which detectable traces of DNA are preserved. This work addresses the question: how long will microorganisms survive in dormant state under the influence of radiation in the regolith of Mars, and how fast will radiation destroy traces of extinct life?

Based on the arguments above, surviving organisms or traces of their DNA are most likely preserved in the subsurface where the absorbed dose from natural radioactivity is the limiting factor. The approach used in this dissertation is to evaluate the habitability of Mars by modelling the radiation in the subsurface. The most suitable sites for modelling are the areas of Mars that have already been selected to have potential for life. Landing sites for rovers have been chosen due to the presence of minerals that suggest past wet condition on Mars. Therefore landing sites are good candidates for habitability models. Permafrost is a type of deposit that contains a mixture of ice and sediments and represents any ground that stayed frozen for more than 2 years. It is normally classified as part of the Martian polar environment. Its shielding property is considered similar to that of normal regolith and rocks. Regolith is any unconsolidated, loose, and heterogeneous deposit, covering solid rock.

It can be composed of dust, broken rocks, and other related materials but does not contain organic matter. Permafrost, however, contains lower concentrations of radionuclides [28] than conventional regolith and is thus also a good candidate for microbial survival.

The radiation environment in permafrost is highly non-uniform. Radioactive minerals are mixed with inert minerals. Ice content can also vary considerably. Some minerals are alpha emitters, others emit beta and gamma radiation only. Alpha radiation has a radiobiological effectiveness that is 20 times larger than that of beta or gamma radiation. It has also a very short range on the order of 10-20 μm . Beta radiation has a range on the order of few mm, gamma radiation on the order of tens of cm. As a result, the dose absorbed by microorganisms over time will depend on their locations, in pore spaces and on the outer surfaces of mineral grains, the ice content, mineral composition, and the concentration of radionuclides in the subsurface. There are radiation “safe havens” (low dose regions) and “hot spots” (high dose regions) in the radiation environment, which influence the survival of microbes and preservation of bio-markers. The microorganisms distributed in the subsurface experience a large variety of doses. In other words, they experience a dose distribution. Some microorganisms will survive considerably longer than others, biomarkers will not all be destroyed at the same rate. Information about the microscopic dose rate variation or distribution is currently lacking in the literature but is crucial in estimating the survival ability of microbes on Mars.

This dissertation provides a quantitative study of microscopic variations in the radiation environment through computer modeling of radiation transport at selected Martian locations. The approach chosen here is to focus on the microscopic dose rate distribution from alpha and beta radiation in the mineral matrix of the regolith. The contribution of gamma radiation to the total dose rate on a microscopic level is expected to be uniform due to its large range (~ 40 cm) compared to the dimension of interest (μm to mm scale).

Selected locations were modeled as mixtures of permafrost and microbes. The dose absorbed by the microbes across the entire matrix was calculated and the dose rate distributions

were recorded. The distributions were then used to calculate probabilities for survival and preservation of biological information. This dissertation therefore aims to answer the question: How do safe havens and hot spots in the radiation environment influence the survival of microbes and preservation of bio-markers?

1.3 Scope of the Work and Dissertation Outline

The work carried out in this dissertation can be divided into two main parts. The Monte Carlo Codes *DosiVox* and *DosiSed* were chosen to carry out the model calculations for the work presented here. The goal of the first part is to show that *DosiVox/DosiSed* are reliable tools for modelling complex sedimentary environments. $\text{Al}_2\text{O}_3\text{:C}$ dosimeters were buried in well-characterized sedimentary samples in the laboratory and luminescence dosimetry was used to experimentally obtain dose values and dose distributions for the dosimeters. The experimental setup was then modelled with *DosiVox* and *DosiSed*. In the second part, selected Martian environments were modelled. Dose rate distributions were obtained and survival probabilities for dormant micro-organisms on Mars were calculated.

The structure of this dissertation follows these two main parts. The initial chapters lay the theoretical framework and provide necessary background information. Chapter 2 discusses habitability of Mars. The radiation environment of Mars and its relevance to this dissertation is explored. The effect of ionizing radiation on organisms is examined. Some arguments for the possibility of life on Mars are presented, as well as previous results of survival of microorganisms and probability for the preservation of life on Mars based on the dose rate of ionizing radiation.

Chapter 3 introduces the general physics behind luminescence dosimetry and explains the optically and thermally stimulated luminescence (OSL/TL) process. Chapter 4 introduces the *DosiVox* and *DosiSed* codes that are based on Monte Carlo Geant4 simulations. The chapter explains the basics of Monte Carlo modelling in general, and modelling with Geant4

in particular. The creation of a sediment model in *DosiVox* and *DosiSed* is described and the necessary input parameters are discussed.

Chapters 5 and 6 present the materials and methodologies for this work and investigate the limitations of experimental work and model calculations. In Chapter 5, the dosimetric properties of the $\text{Al}_2\text{O}_3\text{:C}$ dosimeter are discussed, highlighting some advantages and challenges of this material and why it is the choice of dosimeter for this work. The measurement equipment is described. Experiments were performed to determine the best measurement protocols for the dosimeter material. In Chapter 6 the limitations of *DosiVox* and *DosiSed* are explored. Results reported in the literatures [29–32] are reproduced as a way to test the validity of the code. The influence of model parameters on the results is investigated to constrain input parameters.

A set of mineral mixtures with well-known properties such as grain sizes, mineral compositions, and radionuclide concentrations were prepared in the lab. These samples were studied by (1) measuring alpha and beta radiation separately for each sample using $\text{Al}_2\text{O}_3\text{:C}$ dosimeters and (2) running computer models for these samples and calculating the absorbed dose distributions of the dosimeters buried in the samples. Chapter 7 compares measured and modelled dose distributions to validate the model calculations.

Chapter 8 further investigates the validity of the model calculations. The previous simulations were carried out for containers of limited size, while the Martian subsurface is infinite in dimension. Different input parameters that are impacted by the simulation geometry are investigated. The best parameters for Martian subsurface simulations are selected.

Chapters 9 describe the simulations and results for the Martian subsurface. The deeper subsurface of Mars was investigated using published information about the various rover landing sites and Martian meteorites. The selected locations were modeled as matrix of spherical grains of various sizes, minerals, and radioactive contents. The microbial community in each location was modeled as randomly distributed 5-10 μm spherical grains

with appropriate chemical and physical properties. The effect of parameters such as ice content, chemical composition and radionuclide concentration was investigated and dose rate distributions were obtained. Survival probabilities of dormant microbes and the probability for the preservation of their biomarkers were calculated by applying theoretical models from the literature to the dose rate distribution results.

In conclusion, chapter [10](#) summarizes the relevant results, identifies the strengths and shortcoming of this work, and suggests improvements for future work.

Chapter 2

Habitability of Mars

The overall goal of this dissertation is to determine the survival probability for microorganisms in various Martian environments. However, the bigger question is, if life is possible on Mars in the first place. In this chapter, the possibility of ancient life on Mars is reviewed. The environmental conditions of Mars and their effect on its habitability are discussed. Due to the scope of this dissertation, the focus is on the subsurface. A study of Martian geochemistry and mineralogy is presented, starting with Martian meteorites that were discovered on earth, and followed by various models that have been proposed to characterize the Martian sedimentary environment. Furthermore, permafrost as a potential habitat for ancient microorganisms is discussed. Finally, this information is related to the radiation environment of Mars and how it motivates the objective of this dissertation.

2.1 The possibility of life on ancient Mars

All data that suggests the presence of life on Mars has been arguably inconclusive or at best probabilistic [6, 7, 33]. Under current conditions, the possibility of microbial life is known to be in particular limited by low temperatures, aridity, lack of biologically relevant organic molecules, and ionizing radiation [1, 6, 13, 34–36]. However, there is "evidence" that billions of years ago Mars was a warmer and wetter planet like Earth. These conditions would have

been suitable for microbial habitability. One evidence for a wetter and warmer Mars is Gale Crater. Gale Crater is the landing site of Curiosity, a rover sent to Mars in 2012. An extensive analysis of the rocks in the crater shows enrichment with mineral salts that suggest the existence of shallow briny ponds in the distant past. Also, the network of cracks in the slab of rock in the crater, generally referred to as the "Old Soaker" (see Fig. 2.1) may have formed from the drying of a mud layer 3.7 billion years ago [37]. Further evidence for the wet past of Mars is Jezero Crater, the landing site for the Mars 2020 rover, Perseverance. Data from the Mars Reconnaissance Orbiter suggest that the crater contains clay minerals (see Fig. 2.2). Water is essential for clay mineral formation.

Therefore, considering ancient Mars was warm and wet, the conditions were suitable for microbial life. If life formed, some ancient microorganisms might still exist, 3.7 billion years later, in the deeper subsurface of Mars. These microorganisms would have had to develop survival strategies through metabolic and physiological adaptations to the challenges presented by the extreme environmental conditions over geological time scale. Some microorganisms have been known to adapt by transforming into a dry state through a process called anhydrobiosis [38], or by spore formation, which is the dormant state of certain bacteria. In bacterial endospores on Earth, genes and pathways important to survive frozen conditions in ancient permafrost were detected. It was also observed that these genes increase in abundance with the age of the permafrost [39]. In *Bacillaceae* for example, the inner spore contains anhydrous cytoplasm and DNA, surrounded by a thick multilayered protective envelope. This type of adaptation allows dormant microbes to resist adverse conditions over extended periods of time [12]. Furthermore, most anhydrobiotic or spore forming microorganisms are also resistant to the effects of freezing to very low temperatures and the effects of ionizing radiation. Evidence of survival, possibly due to adaptation, has been found in studies of the survival response of radio-resistant bacterial strains to cosmic level ionizing radiation exposure under various frozen conditions [40–43]. One study reported an increase in radiation resistance at sub-zero temperature compared to higher temperature for the extremophile

Deinococcus radiodurans due to reduced intra-cellular diffusion of free radicals, which leads to a suppression of indirect biological radiation damage [42]. Another prime example of adaptation are the spores of *Bacillus Pumilus*, that are more resistant than vegetative cells to heat, desiccation, UV radiation, γ -radiation, H_2O_2 , and starvation. This bacterial strain has been found in extreme environments such as the interior of Sonoran desert basalt and the Mars Odyssey spacecraft after several pre-launch sterilization [44, 45]. This high resistance has been linked to their ability to produce small acid soluble spore proteins (SASP) that mitigate DNA damage [46].

Considering that the radiation environment in the deeper subsurface comprises only low level radiation from radionuclides and is completely shielded from the life-destroying effect of cosmic radiation, one can argue that these extreme environmentally-conditioned and radio-resistant microbes may survive over millions of years in the deeper subsurface. In the likely case of death, after accumulating low level radiation equivalent to a lethal dose. Their DNA, organic compounds, and mineral products may be preserved in the frozen conditions for even longer periods, which when discovered can be used as biomarkers indicative of their biological activity and bio-genesis [47]. These ancient dormant microorganisms and their survival probabilities are the focus of this work.

2.2 Habitable environments on Mars

The current surface of Mars is inhospitable due to cold temperatures and low atmospheric pressure. These two effects also combine to prevent the formation of water in its liquid state. Furthermore, the surface is continuously battered and bombarded by intense ionizing radiation that can deactivate any living organism or even complex organic molecules. Therefore, in order to search for life, one must look below the surface into the subsurface realm, where pockets of habitable conditions could potentially still harbor extant life and their biosignatures [33]. Studies on Earth have shown that fluids preserved within the Earth’s kilometer–deep

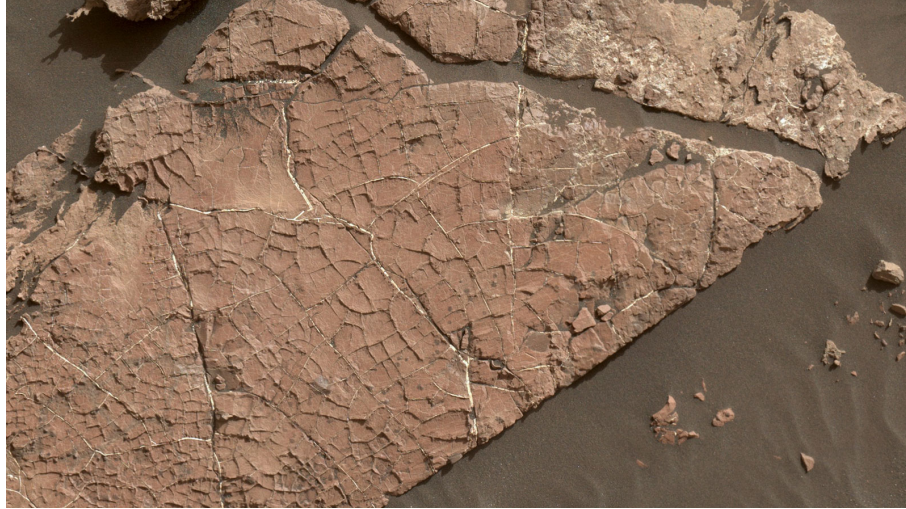


Figure 2.1: The "Old Soaker". A network of cracks is found in this Gale Crater rock. Image credit: NASA/JPL-Caltech/MSSS. [37]

subsurface contain significant quantities of biomass utilizing a wide range of redox reactions to drive their microbial metabolism [50]. Similar habitable subsurface environments could potentially have existed on ancient Mars [51] and may still exist on modern Mars [52]. In the deeper subsurface, planets like Mars with lower surface gravity than Earth are expected to have less compaction of habitable pore space and lower heat flow; hence a reduction in temperature constraints, and higher volume of fracture space that extends deeper into the crust compared to Earth [51, 53]. These conditions thereby increase the volume of supportable biomass and the odds for survival, as long as liquid water sufficient for redox energy production is also available [52]. Additionally, given the likelihood of dormancy for microorganisms in the deeper subsurface, the energy requirement is based on minimal metabolic activity, which can be derived from water-rock reactions including radiolysis and serpentinization [54]. Water molecules in pore spaces can be broken down by radiation from radioactive decay within host rocks, resulting in the production of dissolved reductants like H_2 and dissolved oxidants like H_2O_2 . The dissolved oxidants can further oxidize sulphide within the rock matrix to form dissolved sulphates [51, 54]. On Earth, sulphate-reducing microorganisms have been found in the deeper subsurface in ground water systems isolated for more than 10 million years [55]. Such Earth systems are direct proof that subsurface environments that contain radionuclides,

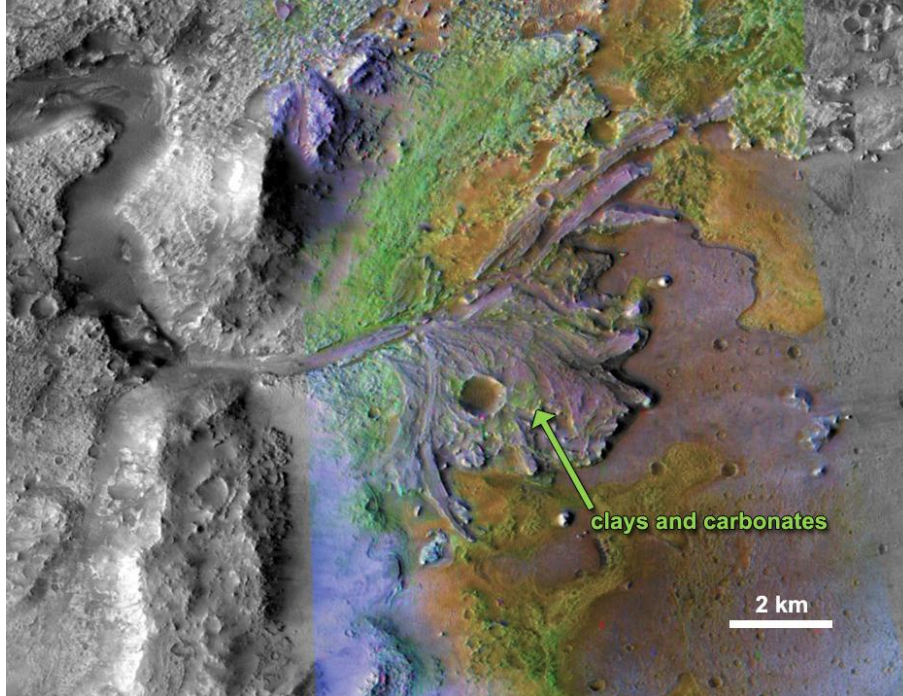


Figure 2.2: The Jezero Crater delta, sediments contain clay minerals and carbonates. This image was formed from the combination of information from the compact Reconnaissance Imaging Spectrometer for Mars and the Context Camera, both parts of NASA’s Mars Reconnaissance Orbiter. Image credit: NASA/JPL-Caltech/MSSS/JHU-APL. [48, 49]

liquid water, and sulfides can satisfy the minimal energy requirement for dormant biomes to exist.

On Mars, liquid water in the subsurface can occur (i) in the form of thin films of water in pore spaces and around the edges of grains [56], (ii) deeper subsurface groundwater layers where radiogenic heating and lithostatic pressures would allow liquid water to exist above freezing point [57], (iii) water from defrosted ice in the near subsurface due to warming during periods of higher obliquity [11], and (iv) mineralized brine in regions with high deposits of chloride salts like the Terra Cimmeria region [58] where dissolved salt molecules can depress the freezing point. However, most of the water on Mars is currently frozen or evaporated. Data collected by the Mars Odyssey spacecraft and the Thermal Emission Spectrometer (TES) confirmed the presence of perennial water ice in the top meter of the Martian polar regions [59]. In the south polar region, on top of a massive ice cap made up of bulk water ice, a large slab of dry ice (CO_2 ice) about 8 m thick was also detected from geomorphic and

spectral observation [60,61]. The north polar cap on the other hand is largely made up of water ice with a volume of about $1.5 \times 10^6 \text{ km}^3$ [62,63]. The combined polar ice volume is estimated to be about $3.1 \times 10^6 \text{ km}^3$ which is equivalent to the volume of the ice sheet in Greenland on Earth [63,64].

2.3 Martian Geochemistry and Mineralogy

2.3.1 Martian Meteorites

In the last 55 years, Mars exploration using orbiters, landers and rovers has provided a lot of information on the global mineralogy, composition and geomorphology of Mars. Most of this knowledge was obtained by studying Martian surface rocks in situ and remotely. However, these studies are limited in terms of the rigorousness of physical and geochemical sample parameters examined and the expertise, precision, and accuracy of analytical techniques available in Earth-based laboratories. Martian meteorites on the other hand are readily available on Earth and have thus gone through numerous and thorough investigation. They are therefore the only fully characterized Martian samples on Earth. The study of these meteorites has provided information on the timing and nature of emplacement and formation of magmatic rocks, the chemical and isotopic composition of the mantle, distribution and evolution of volatile compounds in and on Mars, environments, weathering and impact processes (see Fig. 2.3) [65].

Meteorites are generally formed by impact bombardment of planets by comets and other small bodies. When these impactors hit the ground, fragments of soil and rock are ejected at high velocities, some reaching escape velocity ($\sim 5 \text{ km/s}$; [66]), if the impactor is large and fast enough. These ejecta leave the planet and begin orbiting independently around the Sun, usually for timescales ranging from a thousand to several million years until they either impact another planetary body or are expelled out of our solar system forever [12]. Fragments of these ejecta form the Martian meteorites that have so far been recovered in

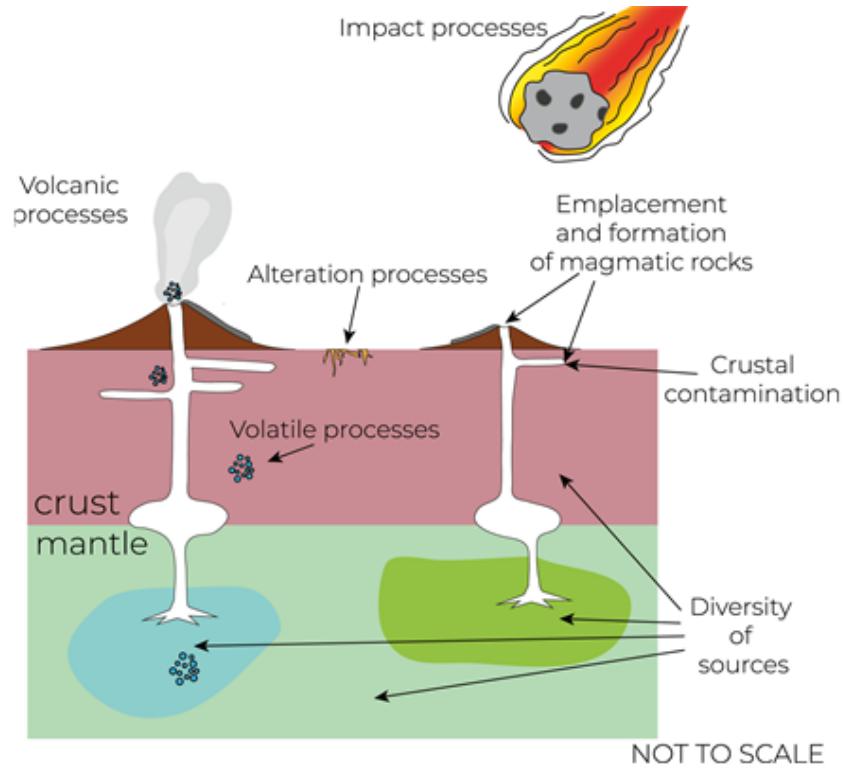


Figure 2.3: Conceptual diagram of the different kinds of information that Martian meteorites can provide about the Martian surface and interior. Blue bubbles represent highly volatile compounds, such as S, Cl, OH, H₂O, and CO₂. Reproduced with permission from AGU. [65, Fig. 1]

Antarctica, Brazil, Chile, France, India, Libya, Mali, Mauritania, Morocco, Nigeria, Oman, Tunisia, and USA [65, 67–69].

Martian meteorites can be broadly divided into two groups: SNCs (Shergottites, Nakhilites, and Chassignites) and non-SNCs. The SNCs are of the igneous family with mafic to ultramafic compositions ($\sim 4\text{--}30$ wt. % MgO). The non-SNCs include the orthopyroxenite Allan Hills (ALH) 84001 and the polymict breccia Northwest Africa (NWA) 7304 and its 16 paired meteorites; where the term "paired meteorites" is used to describe meteorites that originate from the same parent meteoroid but broke up into several pieces upon ejection from Mars or upon entry into the Earth's atmosphere. The oldest Martian meteorites are NWA 7034 which is as old as 4.5 Ga, followed by the ALH 84001 dated at 4.1 Ga [70]. The SNCs meteorites are considered relatively young, predominantly early to late Amazonian in age [65]. The first meteorites that were hypothesized to be from Mars were the Shergotty and Zagami [71]. In

fact, their chemistry, mineralogy, and ages suggest that they originate from a body large enough to still be volcanically active during the last half billion years. The first conclusive evidence for Martian origin is that gases encased in shergottites during shock impact match those measured for the Martian atmosphere [72]. Although, this has not been tested for all Martian meteorites, all known Martian meteorites have been confirmed to have similar bulk oxygen isotopic compositions, which is significantly different from earth (see Fig. 2.4). They are linked by a $\delta^{17}\text{O}$ mass-dependent fractionation line that is $\sim 3\%$ higher than terrestrial or lunar samples [73].

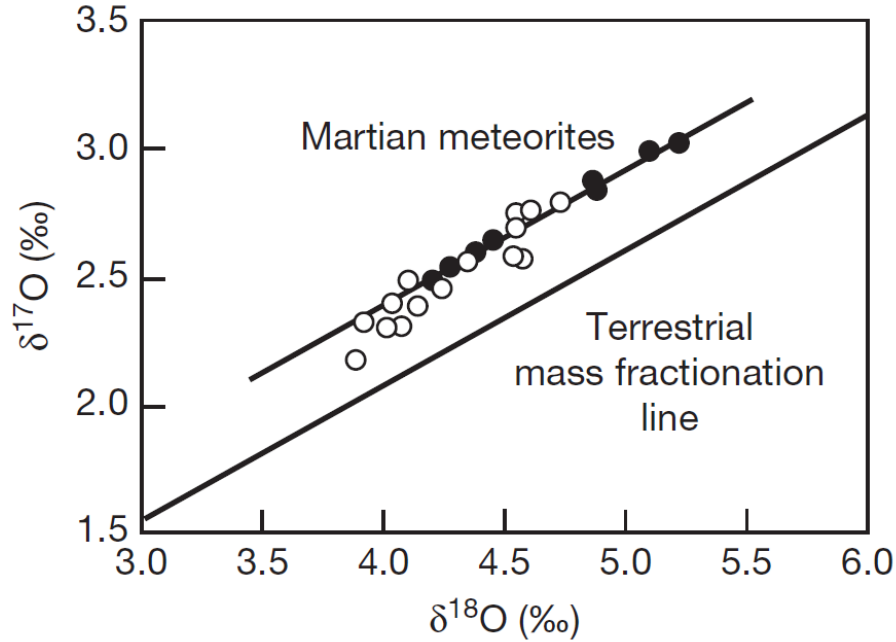


Figure 2.4: Distinct mass-fractionation line due to oxygen isotopic compositions of Martian meteorites compared to terrestrial samples. Open circles are data from [74]) and filled circles are from [75]. [72, Fig. 5]

Table 2.1: Compilation of physical and chemical properties of some Martian meteorites.

	Shergotty	Zagami	EETA 79001A	ALH 84001	Chassigny	Y 980459 ^d	QUE 94201	Nakhla
Location found ^a	Shergotty, India	Zagami, Nigeria	Elephant Moraine, Antarctica	Alan Hills, Antarctica	Chassigny, France	Yamato Mountains Antarctica	Queen Alexandra Range, Antarctica	Nakhla, Egypt
Year found ^a	1865	1905	1980	1984	1815	1998	1994	1905
Type ^b	Basaltic shergottite	Basaltic shergottite	Olivine-phyric shergottite	Orthopyroxenite	Chassignite	Olivine-phyric shergottite	Basaltic shergottite	Nakhlite
SiO ₂ ^c	51.3	50.5 ± 0.4	49.9 ± 0.9	52.8 ± 0.1	37.4 ± 1.1	49.9 ± 1.3	47.9	48.6 ± 1.7
Al ₂ O ₃ ^c	6.86 ± 0.51	6.05 ± 0.38	5.91 ± 0.60	1.29 ± 0.10	0.72 ± 0.30	5.17 ± 0.06	11.00 ± 1.10	1.68 ± 0.20
FeO ^c	19.4 ± 0.7	18.10 ± 0.5	18.4 ± 0.8	17.5 ± 0.5	27.3 ± 0.8	17.3 ± 0.2	18.5 ± 0.5	20.6 ± 1.5
MgO ^c	9.3 ± 0.2	11.3 ± 0.7	16.1 ± 0.8	25.0 ± 0.8	31.8 ± 0.8	18.70 ± 1.2	6.25 ± 0.07	12.1 ± 0.4
CaO ^c	9.60 ± 0.55	10.5 ± 0.6	7.26 ± 0.32	1.82 ± 0.38	0.66 ± 0.10	6.83 ± 0.61	11.4 ± 0.2	14.7 ± 0.7
Na ₂ O ^c	1.39 ± 0.19	1.23 ± 0.06	0.86 ± 0.05	0.14 ± 0.03	0.12 ± 0.02	0.65	1.58 ± 0.26	0.46 ± 0.10
K ₂ O ^c	0.17 ± 0.02	0.14 ± 0.01	0.04 ± 0.01	0.017 ± 0.006	0.04 ± 0.01	0.016 ± 0.004	0.045 ± 0.010	0.13
P ₂ O ₅ ^c	0.67 ± 0.14	0.5 ± 0.07	0.6 ± 0.07	0.014	0.071 ± 0.03			0.13
TiO ₂ ^c	0.82 ± 0.07	0.79 ± 0.04	0.70 ± 0.12	0.21 ± 0.01	0.08 ± 0.02	0.53 ± 0.05	1.84 ± 0.15	0.34
Cr ₂ O ₃ ^c	0.20 ± 0.02	0.33 ± 0.04	0.62 ± 0.04	1.14 ± 0.10	0.77 ± 0.15	0.70 ± 0.01	0.14 ± 0.01	0.26
MnO ^c	0.52 ± 0.02	0.5 ± 0.04	0.48 ± 0.02	0.46 ± 0.01	0.53 ± 0.11	0.48 ± 0.02	0.45 ± 0.02	0.49
S		0.19			0.026 ± 0.010			0.026
Total	100.25	100.13	100.87	100.39	99.52	100.27	99.11	99.49
Density (g/cm ³) ^e	3.30 ± 0.05	3.32 ± 0.04	3.39 ± 0.07	3.44 ± 0.02	3.61 ± 0.07	3.35 ± 0.05	3.27 ± 0.08	3.45 ± 0.03

^a Udry et al. (2020) [65]

^b McSween (2014) [72]

^c Lodders (1998) [68]

^d Shirai & Ebihara (2004) [76]

^e Baratoux et al.(2014) [77]

Some of the Meteorites were found in Antarctica. The Martian meteorites recovered from Antarctica are particularly important to this dissertation because Antarctica is a cold and extreme environment and is often used as a Mars analog. Thus, these meteorites are expected to be least affected by terrestrial weathering during their residence on Earth, compared to the ones found in hot deserts [76]. While this is a reasonable assumption, there has only been one sample to date, called Bounce Rock found in Meridiani Planum at the Martian surface, that matches the bulk composition of basaltic shergottite meteorites [78]. Generally, measurement of Martian surface rocks by rovers shows relatively higher SiO₂, alkalis, and lower MgO and CaO contents compared to that of Martian meteorites [65]. However, a new study based on terrestrial analog analyses reported that the APXS (Alpha Particle X-Ray Spectrometer) instrument on board *Spirit*, *Opportunity*, and *Curiosity* rovers overestimated elements like Al and S and underestimated elements like Mg due to matrix effects. So, the basaltic minerals at the Martian surface might be more similar to the meteorites than previously thought [79].

2.3.2 Subsurface bulk composition of Mars

To summarize the composition of an entire planet with all its variety in one set of numbers is challenging, to say the least. Several approaches have been used to estimate the bulk chemical composition of Mars. These approaches are based on cosmochemistry, the isotopic composition of Martian meteorites, or geophysical properties. The cosmochemical approach which involves defining the different components and determining their abundances on Mars [80–83]. An approach based on the isotopic compositions of Martian meteorites and chondrite groups that define the abundances of chondritic raw materials that accreted to Mars [67, 84, 85]. And finally, the geophysical approach that employs broad geophysical properties to establish mantle mineralogy [86–89]. The first approach will be discussed in further detail because it was used for one of the Martian simulations in this work.

Model based on cosmochemistry and nebular components

The Wänke & Dreibus model [80, 81, 90–92], is considered the most robust Martian bulk composition model. In this model, the bulk composition of Mars was estimated from elemental compositions of the Martian meteorites, with the assumption that Mn and all elements more refractory than Mn are present in chondritic proportions on Mars. This direct link of the Martian composition to samples from Mars is the reason why it is considered the standard model for bulk martian composition.

A reassessment of the composition of Mars from elemental data was carried out more recently by Taylor (2013) [82]. They used information from over 60 distinct Martian meteorites compared to only 10 used by Wänke & Dreibus in their work. This information includes chemical, petrologic, mineralogic, and isotopic data from the *Martian Meteorite Compendium*. The revised composition is presented in Table 2.2. The noteworthy feature of the revised composition is how similar it is to that of Wänke & Dreibus, despite all the additional improved geophysical, orbital and lander geochemical data considered. The model by Taylor

(2013) [82] was used for the simulations described in Chapter 9.

Table 2.2: Models for the subsurface bulk composition of Mars.

	WD(94) ^a	T(13) ^f
SiO₂	44.4	43.7
Al₂O₃	3.02	3.04
FeO	17.90	18.1
MgO	30.20	30.5
CaO	2.45	2.43
Na₂O	0.50	0.53
K₂O	0.04	0.04
P₂O₅	0.16	0.15
TiO₂	0.14	0.14
Cr₂O₃	0.76	0.73
MnO	0.46	0.44
Total	100.03	99.8
K (ppm)	305	309
Th (ppb)	56	58
U (ppb)	16	16

^a Wänke & Dreibus (1994) [81]

^f Taylor (2013) [82]

2.3.3 Permafrost on Mars

Generally speaking, permafrost is any ground that remains at or below 0°C for at least two years. Ice content of terrestrial permafrost can range from disseminated ice crystals in a soil matrix to several meters thick layer of nearly pure ice [13,93] (see Fig. 2.5). Permafrost is thus highly non-uniform on a macroscopic and microscopic scale. Its ground surface is typically composed of water, vegetation and ice. It is a general understanding that liquid water exists in permafrost in the form of very thin films on the surface of soil and ice particles [94]. The thickness of the water film is slightly affected by temperature variation and soil particle

size [22].

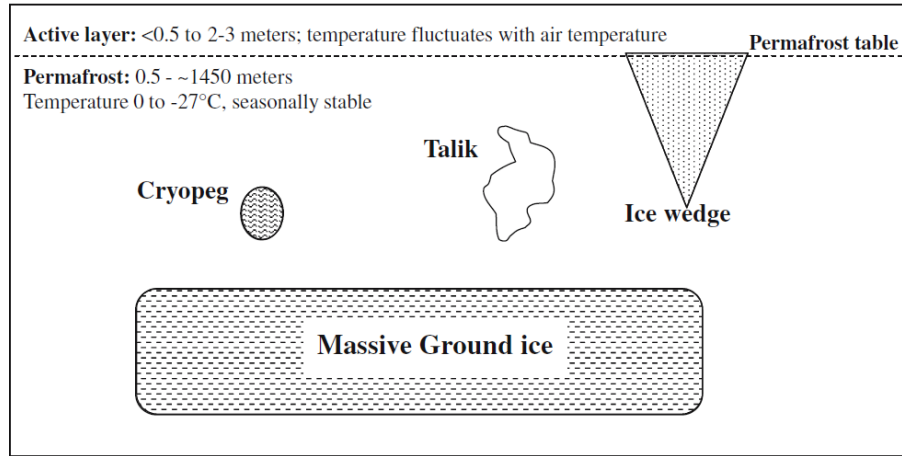


Figure 2.5: Schematic and conceptual diagram of the permafrost zone. Positions are generalized and do not reflect the actual depth or size. Reproduced with permission from Springer Nature. [13, Fig. 1]

Permafrost on Mars differs from that on Earth. Mellon and Jakosky (1993) [20] were the first to predict that interstitial ice which they called "pore ice" can occur in the Martian subsurface through a direct vapor deposition process. This mechanism which is uncommon on Earth was demonstrated in the laboratory by Hudson et al. (2009) [95]. Today we know that at least 30 percent of the Martian surface contains shallow ground ice [96, 97]. Ground ice on Mars has been a major target for research and exploration. Investigations of the Martian surface by orbiters, rovers and landers have identified multiple evidence for the presence of ground ice in mid- to high latitude near-surface Martian regolith [98]. Polygonally patterned landforms that occur over extensive portions on Mars have been attributed to the presence of ice-rich permafrost [99]. Massive ice deposits that far exceed the pore volume of the regolith have been detected using orbital remote sensing in the Martian subsurface [100]. These deposits have been hypothesized to come from atmospheric deposition such as frost or snow and subsequent burial by dust and debris [101]. Dundad et al. [102] also reported that erosion on Mars creates scarps that reveal ice sheets that extend to a depth of 100 m with distinct layers.

The form, structure and thickness of non-polar permafrost on Mars has been shown

to be latitude dependent [103] and similar to terrestrial glaciers [104]. Latitude-dependent permafrost comprises deposits ranging in thickness from tens to hundreds of meters in the high-latitude regions of Mars but becomes increasingly thinner and discontinuous in the mid latitudes, and absent in the equatorial regions [102, 105]. Furthermore, estimates based on subsurface radar data from the Mars Advanced Radar for Subsurface and Ionosphere Sounding (MARSIS) indicate that the ice/rock volume mixing ratio of the Southern and Northern hemisphere of Mars is about 50% and 100% respectively [106].

In the northern and southern high-latitude regions, cycles of orbital obliquity would have allowed this permafrost to melt and form liquid water for up to 100 days per year every ~ 50 kyr [4, 10], providing an excellent niche that could support life. Ice-cemented ground in the polar region of Mars may preserve organic matter and hold clues to the biological history of that planet [41, 107]. The polar regions and thus permafrost on Mars are therefore considered a primary target for the search for life [13, 108–110]. Terrestrial permafrost is an ideal test environment for permafrost on Mars. Permafrost as a microbial habitat and its microbial diversity have been studied for a wide variety of terrestrial permafrost. Detailed reviews have been published by Steven et al. (2006) [13] and Boetius et al. (2015) [111]. More recent work has been carried out on samples from study sites like the ‘Muot-da-Barba-Peider’ in Switzerland [112], Antarctica [110, 113] and Greenland [114, 115].

2.4 The radiation environment of Mars

The radiation environment of Mars consists of three populations of particles: Galactic cosmic rays (GCR), solar energetic particles (SEP) and emissions from natural radioactive nuclides in the regolith. Regolith is any unconsolidated, loose, and heterogeneous deposit, covering solid rock. It can be composed of dust, broken rocks, and other related materials but does not contain organic matter. Galactic cosmic rays (GCRs) are high energy particles with maximum energy around 500 MeV/nucleon. Most of the energies of this spectra are relatively

low except for the power law tail that reaches about 10^{20} eV at extremely low fluxes [1]. The GCR flux is modulated by the heliosphere and has a negative correlation with the solar cycle. The spectrum which depends on solar modulation is composed of 87% protons, 11% helium ions, 1% electrons, and 1% heavy ions. They can penetrate several meters into the Martian regolith before they are completely attenuated. Solar energetic particles (SEPs) are produced by solar flares and coronal mass ejections. They have fluxes that vary by several orders of magnitude and depend on the 11-year solar cycle. They are often composed of protons, but composition can vary significantly. They can be broadly divided into two classes depending on their energy per nucleon. The "soft spectrum" with energies below 150 MeV/nucleon that are not high enough to penetrate the Martian surface and the "hard spectrum" with energies much greater than 150 MeV/nucleon that can reach and penetrate the Martian surface [6, 7].

Cosmic radiation like the GCR and SEP, has been suggested to be the prime limiting factor for the survival of microbes on the surface of Mars [1, 116]. There are however some radio-resistant bacteria that possess a 100–1,000 times higher tolerance to ionizing radiation than other terrestrial microorganisms [117, 118] (Sterilization doses greater than 20 kGy have been found in laboratory experiments for *Deinococcus radiodurans* [28]). However, deeper in the subsurface, below 3 meters, the contribution of cosmic radiation decreases significantly. This has been reported by Dartnell et. al., (2007) [1]. The authors showed that more than 99% of the GCR and SEP flux are attenuated within the first 3 meters of the Martian regolith (see Fig. 2.6). In the deeper subsurface, the radiation environment is mostly dominated by the much lower and heterogeneous levels of natural radioactivity from radioactive isotopes (see Fig. 2.7) [7]. This work is focused on the deeper subsurface region at a depth below 3m, where the radiation field is dominated by natural radioactive nuclides. For the purpose of this work, this region will be referred to as the subsurface.

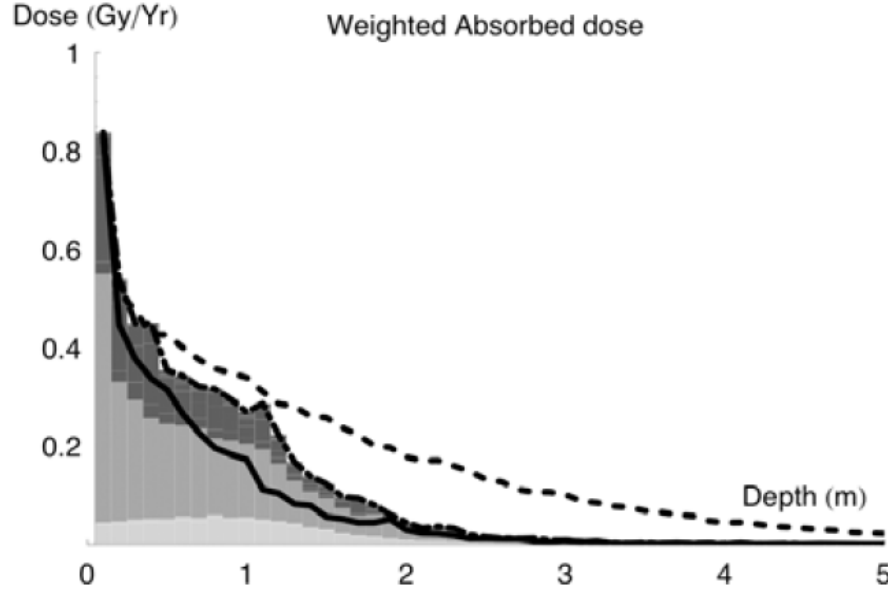


Figure 2.6: The decrease of biologically-weighted dose with depth. Three models of Martian "soil" were considered here. Dry homogeneous 2.8 g/cm^3 regolith (solid line), a 1 g/cm^3 block of pure ice (dashed line), and wet heterogeneous 1.1 g/cm^3 topsoil (2% wt) + 1.1 g/cm^3 topsoil (16% wt) + 3 g/cm^3 regolith (16% wt) (dot dashed line). The shades of gray represent the particles contribution; muon, e^+ , and e^- (light gray), proton (medium gray), and heavy ions (dark gray). Reprinted with permission from AGU. [1, Fig. 3 (bottom)]

Natural radioactive particles on Mars

Natural radioactivity is mainly due to ^{238}U , ^{235}U , ^{232}Th , and their daughter nuclides, as well as ^{40}K . The half-lives for ^{238}U , ^{235}U , ^{232}Th , and ^{40}K are 4.5×10^9 years, 0.7×10^9 years, 14×10^9 years, and 1.3×10^9 years, respectively. Although the abundances of potassium, thorium and uranium on Mars are not known with precision [12], They are expected to be about two orders of magnitude less than on Earth [119]. Based on the Shergotti Parent Body model for Mars, the abundances of potassium, thorium and uranium are 315 ppm, 5.6×10^{-2} ppm and 1.6×10^{-2} ppm, respectively [90]. The present isotopic abundances of the most common radionuclides are $\sim 100 \%$ for ^{232}Th , 99.2745 % for ^{238}U , 0.7200 % for ^{235}U , and 0.0117 % for ^{40}K [119].

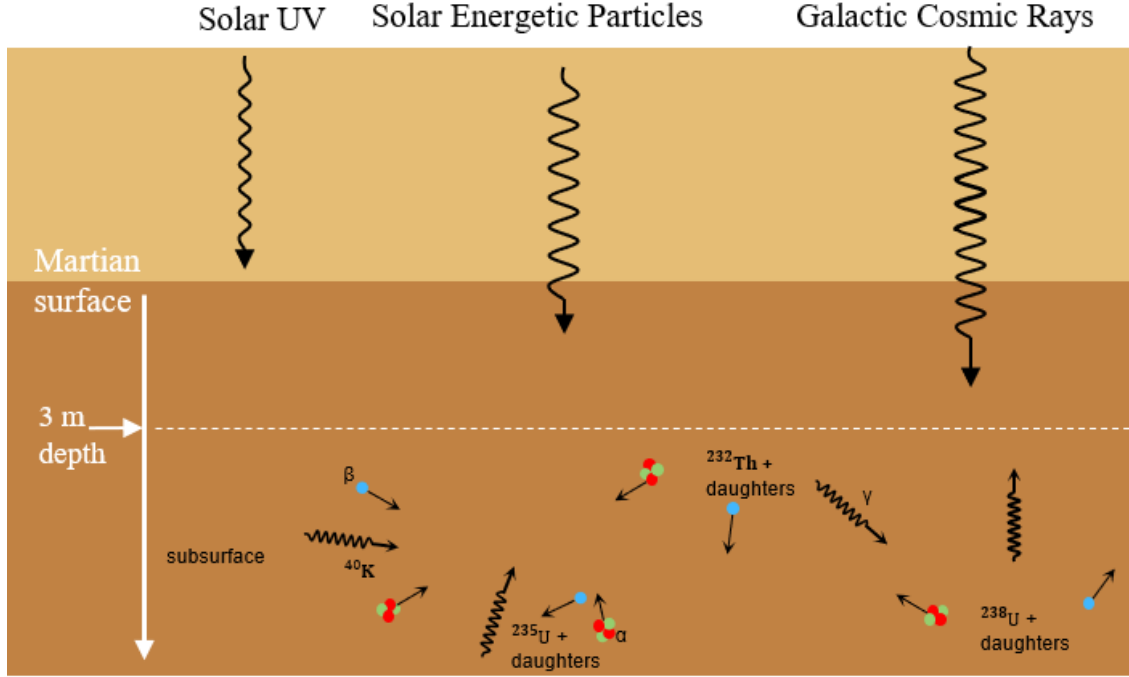


Figure 2.7: Schematic representation of the radiation environment of Mars.

The dose rate from radioactivity can be obtained from the formula:

$$\dot{D}(\text{Gy/year}) = \frac{[c \text{ (ppm)} \times E \text{ (MeV)/decay} \times \lambda \text{ (year}^{-1}\text{)} \times N_A]}{A \times 6.24 \times 10^{18}} \quad (2.1)$$

where $\dot{D}$ is the dose rate, c is the radionuclide concentration (where, the concentration in ppm must be multiplied by 10^{-6}), E is the energy per decay corrected for neutrino loss in β -decay and must be in unit MeV, λ is the decay constant, N_A is Avogadro's number ($6.022 \times 10^{23} \text{ mol}^{-1}$), A is the atomic mass of the nuclide in unit of g mol^{-1} and $6.24181 \times 10^{18} \frac{\text{eV}}{\text{J}}$ is the unit conversion factor to obtain the unit of dose rate in Gy/year. Thus, the dose rate is proportional to the concentration of the radionuclide and the energy release per decay.

As a baseline approximation, using average radionuclide concentrations ($^{40}\text{K} = 0.351 \%$, $^{232}\text{Th} = 0.670 \text{ ppm}$, and $^{238}\text{U} = 0.188 \text{ ppm}$) of the Martian rocks in three landing sites (Ares Vallis, Gusev Crater, and Gale Crater), the present total dose rate in the subsurface of Mars was calculated as $\sim 1.45 \text{ mGy/year}$ (where $\text{mGy} = 10^{-3}\text{Gy}$). When this dose rate is corrected for the biological effectiveness of alpha radiation using a quality factor of 20, the

present total dose rate becomes ~ 22.85 mGy/year. Based on this present value of dose rate, 4 billion years ago, the dose rate would have been ~ 37.35 mGy/year. One can thus assume that over the last 4 billion years, the average dose rate on the Martian subsurface is ~ 29.40 mGy/year. Based on this value, it will take about 1 million years to kill *B. Subtilis* bacterial spores.

Similar attempts have been made to estimate the dose rate in the Martian subsurface. For example, Kminek et al. [119] reported a dose rate calculated directly from the concentrations of each radioisotope and its decay parameters for the Martian subsurface. Their result ranges from 0.35 mGy/year 3.1 Ga ago to 0.13 mGy/year more recently due to radioactive decay over time. The concentration of radioisotopes on Mars that they used was based on composition of the Shergotty Martian meteorite. A Similar study by Mileikowsky et al. [12] reported an average dose rate of 0.4 mGy/year based on the average radioisotope concentrations of all the Martian meteorites available at the time. These seemingly low dose rates can nevertheless lead to high accumulated doses over geological time scales. For example, in 4 billion years, the accumulated dose can reach up to 1.6 MGy, a dose high enough to inactivate even the most radio-resistant microorganisms [42]. Freezing and extreme conditions on Mars imply that extant life will be left in the state of dormancy and thus cannot repair radiation damage like metabolically active cells can. In Martian permafrost with a mixture of minerals and ice, the key determinant for survival over a long period of time is thus accumulated dose and not the instantaneous dose rate. Therefore, based on their result, one might suspect that over geological time scales all of the microorganisms in dormant state will be inactivated and their bio-markers obliterated. [1].

2.5 Considerations relevant for the survival of microorganisms

The dose rates calculated by Mileikowsky et al. [12] and Kminek et al. [119] are based on average values. However, on the microscopic level, the Martian subsurface is basically a mixture of sedimentary grains of different sizes, shapes, and minerals. The average value can only paint a limited picture. This is because a sediment is made up of several grains, some radioactive and others non-radioactive. The radioactive grains will emit alpha, beta and gamma particles that will travel different distances in the sediment (10-20 μm for alpha, few mm for beta and tens of cm for gamma) [30]. If a microbe is close to an alpha emitter, it will receive a very high dose equivalent in one hit compared to one close to a beta or gamma emitter. This is because alpha radiation has a biological effectiveness that is 20 times more than beta and gamma radiation and thus causes more damage to cells. The relative biological effectiveness of radiation is defined as the dose of a standard radiation to produce a given biological effect divided by the dose of a test radiation to produce the same effect.

The dose rate received by microbes residing in the pore spaces or outer surfaces between grains depends on their location in the mineral matrix, i.e. the absorbed dose rate of each microbe depends on the microscopic dose rate inhomogeneity (or heterogeneity) existing in the sediment. The dose rate heterogeneity is a result of the distribution of radioemitter types and their range (alpha or beta particles), percent and composition of ice, grain size specific attenuation, and distribution of radionuclides. This dose rate inhomogeneity will result in a distribution of "hotspots" and "safe havens" in the microscopic realm of the sediment. Consequently, microbes close to a hotspot will receive a higher dose compared to those in a safe haven. And microorganisms in a safe haven will survive longer than those in a hotspot. Our approach is to determine this microscopic dose rate distribution in the cold Martian subsurface using the limited but better understanding of the Martian subsurface that has occurred in recent years. The goal is to use the microscopic dose rate distribution to give a

better prediction of how long it will take for the total inactivation of dormant cells and/or obliteration of their bio-markers.

Chapter 3

Luminescence Dosimetry

3.1 Introduction

The laboratory-based dose rate measurements in this work were carried out with $\text{Al}_2\text{O}_3\text{:C}$ luminescence dosimeters. Luminescence dosimetry is one of the most effective techniques for measuring dose rate in sedimentary environments [120, 121]. As mentioned in chapter 1, the objective of the laboratory-based measurements was to obtain experimental dose rates that were compared to computer simulated dose rates for well-defined sedimentary samples. This chapter discusses the general physics behind luminescence dosimetry and then relates it directly to optically and thermally stimulated luminescence. It provides a detailed review of the energy structure of crystalline insulators as the basis for luminescence and discusses the processes inside a crystal during luminescence. It also touches on the mathematical models and rate equations of the luminescence process, as well as the physics and the properties of $\text{Al}_2\text{O}_3\text{:C}$ as a luminescence dosimeter.

3.2 Physics of luminescence dosimetry

3.2.1 Energy levels in a perfect crystal

The first solids studied for crystal form and elastic behavior were ionic crystals. This study which was done using X-rays with the assumption that crystals are made up of positively and negatively charged ions reported the first values of crystal lattice energies and elastic properties [122]. A crystal is characterized by its regular, periodically repeated structure. The smallest unit of this structure is called a primitive cell. The periodicity of the crystal lattice is defined by the repeated pattern in the entire crystal due to the joining together of identical primitive cells. This periodicity makes the lattice invariant to translations over distances that are integer multiples of the lattice period [123].

The behaviour of a particle in a crystalline material was also studied and modeled by Schrödinger, using the well-known Schrödinger equation. Eq. (3.1) shows the time-independent Schrödinger equation that describes the movement of a single particle in an electric field.

$$\left[-\frac{\hbar^2}{2m} \nabla^2 + V(\mathbf{r}) \right] \Psi(\mathbf{r}) = E \Psi(\mathbf{r}) \quad (3.1)$$

where $\hbar$ is the reduced Planck constant ($\hbar = \frac{h}{2\pi}$), m is the particle's mass, ∇^2 is the Laplacian, V is the potential energy, Ψ is the quantum mechanical wave function, $\mathbf{r}$ is the position vector and E is a constant equal to the total energy of the system. Bloch [124] using Eq. (3.1), developed the theorem that the wave function solution from Eq. (3.1) in a periodic crystal lattice is given by:

$$\Psi_{\mathbf{k}}(\mathbf{r}) = u_{\mathbf{k}}(\mathbf{r}) \cdot \exp(i \mathbf{k} \cdot \mathbf{r}) \quad (3.2)$$

where $\mathbf{k}$ is the crystal wave vector, i is the imaginary unit, the factor $u_{\mathbf{k}}(\mathbf{r})$ carries the periodicity of the lattice, thus $\Psi_{\mathbf{k}}(\mathbf{r})$ is not just a plane wave but is instead a plane wave that is modulated by lattice or spatial periodicity and is called the Bloch function [123, 125, 126].

3.2.2 The Kronig-Penney model

The Kronig-Penney model is the simplest case of the Bloch function applied to a periodic lattice. It considers a one-dimensional periodic lattice with ion distance a , particle mass m , and length of the potential b resulting in the existence of band gaps [127]. The system in the Kronig-Penney model can be considered an infinite array of rectangular potential wells with height $(V_2 - V_1)$. See Figure 3.1.

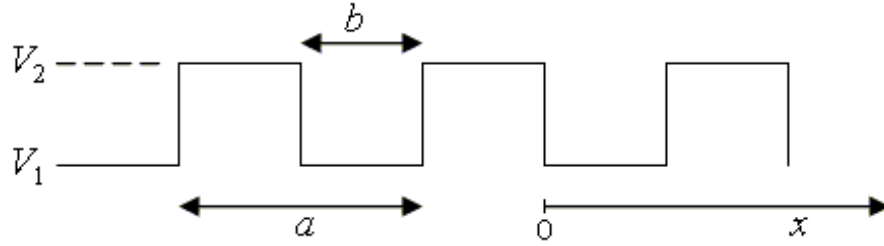


Figure 3.1: One-dimensional square wave potential with translational symmetry in its potential i.e. $V(x) = V(x + a)$. Adapted from Kollmitzer & Hadley [128] with permission. Copyright (2011), Elsevier.

substituting the Bloch function, Eq. (3.2) into Eq. (3.1) results in a non-trivial solution to this model. For the case where the potential V is zero except at atomic sites, where it is infinity (i.e. where the potential is modeled as a delta function), the non-trivial solution reduces to the expression:

$$\cos(ka) = \frac{P}{k_o a} \cdot \sin(k_o a) + \cos(k_o a) \quad (3.3)$$

where in the above equation, $k = \frac{2\pi}{\lambda}$, λ being the spatial period or simply the wavelength, $k_o^2 = \frac{2m|E|}{\hbar^2}$, and $P = \frac{mV_o a b}{\hbar^2}$. It is worth noting that not all possible values of $k_o a$ give a solution to Eq. (3.3) since $\cos(ka)$ lies between -1 and $+1$. Figure 3.2 depicts the right-hand side of Eq. (3.3) for a fixed value of $P = 3\pi/2$, as used in the original publication by Kronig & Penny [127]. The shaded regions represent the parts where a real solution exists for the right-hand side of Eq. (3.3). These regions are called the *allowed* regions. Considering the influence of P on the spectrum, if P vanishes, the forbidden zones disappear and we have a

continuous spectrum of energies from 0 to ∞ . This is the limiting case of free electrons (i.e. with zero potential energy). On the other hand, if P tends to infinity, the energy spectrum becomes discrete, corresponding to an electron confined between two impenetrable potential barriers and thus sentenced to eternal isolation.

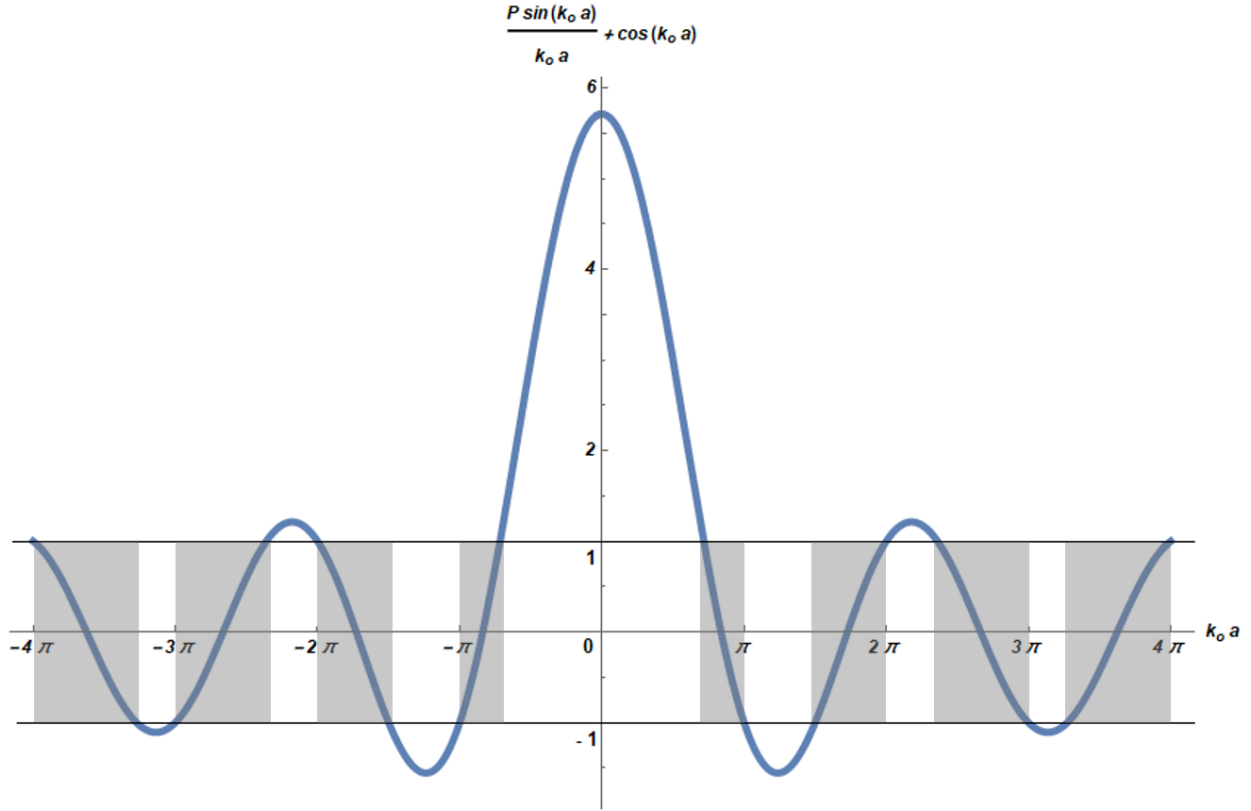


Figure 3.2: The right hand-side of Eq. (3.3) plotted against $k_o a$ for $P = 3\pi/2$. Note: in $k_o a$, only E varies, the other terms are constant. The grey-shaded regions represent portions with permissible values of $k_o a$ (*allowed* region). Thus dividing the plot into allowed regions and *forbidden* regions (with impermissible values of $k_o a$). Observe that the forbidden regions become narrower as the value $k_o a$ becomes larger. Adapted from Kittel [125] with permission. Copyright (2004), John Wiley and Sons.

One advantage of the Kronig-Penney model is that it is possible to analytically obtain expressions for the electron density of states. The expression is derived by considering that the Kronig-Penney potential exhibits translational symmetry (see Figure 3.1). The analytical expression for the density of states is presented in Eq. (3.4). For the detailed derivation, please refer to the paper by Kollmitzer & Hadley [128].

$$g(E) = \begin{cases} \frac{2}{\pi} \frac{dk}{dE} = \frac{2}{\pi} \frac{dk}{d\alpha} \frac{d\alpha}{dE} & |\alpha| < 2, \\ 0 & |\alpha| > 2, \end{cases} \quad (3.4)$$

Plotting the electron density of states function $g(E)$, (Eq. (3.4), with respect to energy E shows the existence of forbidden zones, that are directly associated with the potential V , where the density for electron states is zero. These zones are called band gaps. The zones between the band gaps are the bands (i.e. valence band and conduction band). The data used in Figure 3.3 was obtained from the TU Graz database.¹

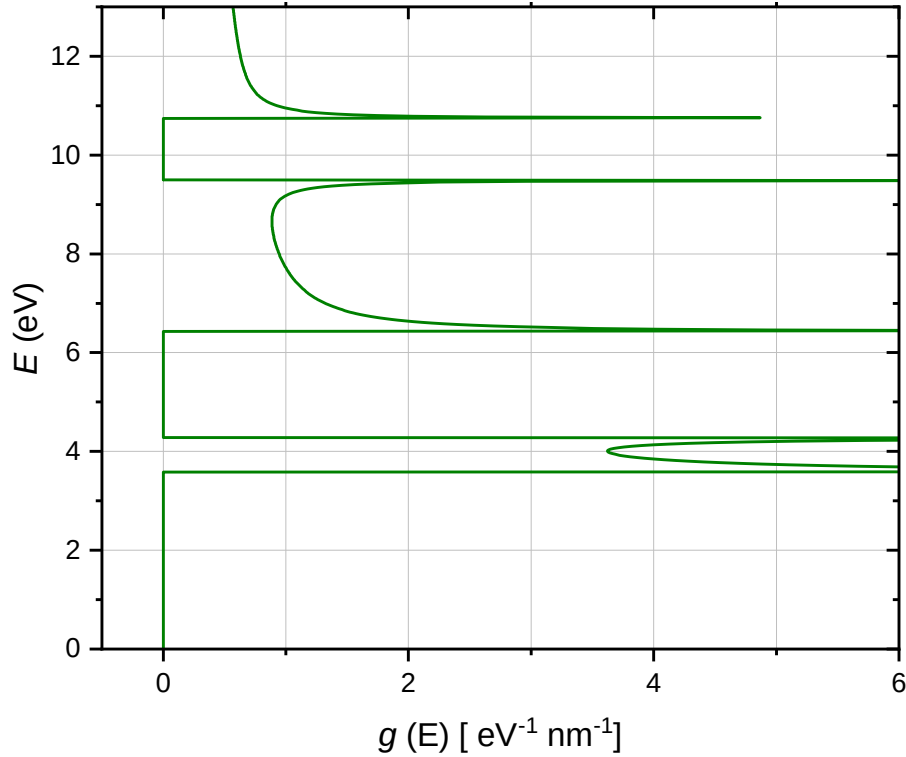


Figure 3.3: Density of states for the Kronig-Penney model. $g(E)$ has a value of 0 for some energies. The input parameters of the plot are: $V_1 = 0 \text{ eV}$, $V_2 = 5 \text{ eV}$, $a = 5 \times 10^{-10} \text{ m}$, $b = 1 \times 10^{-10} \text{ m}$, and $m = 9.1 \times 10^{-31} \text{ kg}$.

¹<http://lampx.tugraz.at/~hadley/ss1/KronigPenney/KronigPenney.php>, last accessed on May 4, 2022.

3.3 Thermally and Optically Stimulated Luminescence

The basic idea of luminescence dosimetry is centered on the two-band model, the valence band and the conduction band separated by the forbidden gap/zone [129]. The valence band is the highest filled energy band. Electrons "jump" out of this band and relocate to the conduction band when excited [125]. The conduction band on the other hand is the empty band above the valence band where electrons are considered to be free to migrate through the crystal lattice. The band gap is like the "payment" for this freedom, where the currency is energy. The required amount of energy is what distinguishes a metal from a semiconductor and an insulator. For example, for an insulator, this energy is greater than 2.5 eV (for reference, the thermal energy at room temperature is ~ 0.025 eV) [129].

Additionally, this gap as discussed in the previous section is sometimes called the "forbidden gap" because in a pure crystal no energy states exist in this gap. Charges can either be in the valence band or the conduction band at any time, and when they gain or lose energy equal to the band gap, they cross over this gap to the other band. However, in a crystal with lattice defects (imperfections and impurities), the defects act as localized energy levels within the forbidden gap. Irradiation of these crystals ionizes atoms creating electrons in the conduction band and holes in the valence band. The electrons from the conduction band are free to move and can diffuse into the vicinity of the defects in the forbidden gap, thereby becoming trapped in "electron traps" and the holes which are absent electrons can also diffuse into the forbidden gap and become trapped in "hole traps" (see Figure 3.4a).

The hole traps are more complex since they can also act as recombination centers for luminescence production [130]. The effectiveness of a given trap depends on its depth, E , below the conduction band. Traps with shallow depth can be emptied even by lattice vibrations at room temperature. The process of irradiation absorption for a crystal can be said to introduce some kind of perturbation causing the crystal to go from a state of thermodynamic equilibrium to a new state of meta-stability. And a return to equilibrium

happens when there is an absorption of external energy leading to stimulated relaxation. During stimulated relaxation, trapped electrons gain enough energy to be released from their traps and recombine with holes at the luminescence centers. This process results in the emission of light or luminescence, and is made possible through heating (thermoluminescence, TL) or by illumination (optically stimulated luminescence, OSL) [131]. Figure 3.4 outlines the various stages involved in the OSL/TL process.

For OSL, the stimulation energy source is light; infra-red, visible or UV. For TL, it is heat. The intensity of luminescence during stimulated relaxation depends on the rate at which the system achieves equilibrium, which, in turn, depends on the concentrations of trapped electrons. Some minerals have a large number of defects and the number of trapped charges is proportional to the amount of irradiation. A typical way to quantify the amount of radiation absorbed by the crystal is through recording the intensity of the luminescence as a function of time for OSL or as a function of temperature for TL, resulting in a characteristic luminescence-time curve (glow-curve for TL). For OSL, the luminescence intensity during stimulation depends on the stimulation time and wavelength of the stimulation. For TL, the luminescence intensity depends on the heating rate and temperature. The intensity also depends on the absorbed radiation dose. The area under the luminescence curve is considered to be related to the trapped charge, which in turn is proportional to the absorbed radiation dose. This is the basis for the use of OSL/TL in luminescence dosimetry. Examples of luminescence curves for OSL and TL are shown in Figure 3.5.

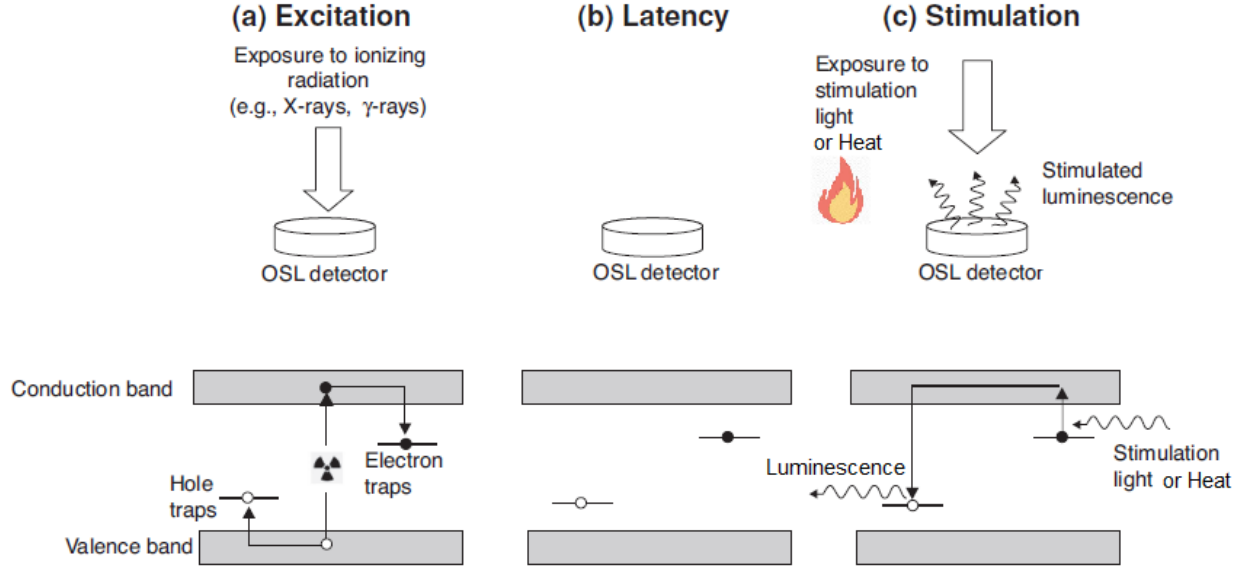


Figure 3.4: The various stages involved in the OSL and/or TL process: (a) excitation of the detector by irradiation; (b) latency period characterized by a metastable concentration of electrons and holes captured at the traps and (c) stimulation of the detector with light or heat, leading to recombination and emission of light. Adapted from [129, Fig. 2.1]

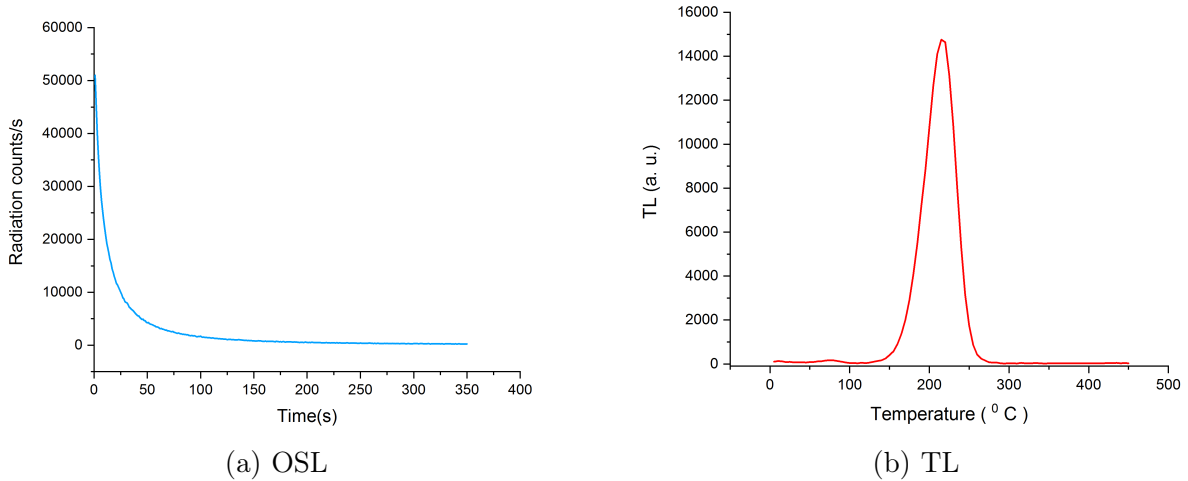


Figure 3.5: Typical experimental curves from stimulated luminescence of $\text{Al}_2\text{O}_3:\text{C}$ crystal.

3.3.1 Generalized mathematical description

Consider a crystal system that has been exposed to radiation and is thus in a meta-stable state. The total concentration of occupied meta-stable states in the system at a time t may be represented by [129]:

$$\mu(t) = \int_{\gamma_1} \int_{\gamma_2} \cdots \int_{\gamma_m} n(\gamma_1, \gamma_2, \dots, \gamma_m, t) d\gamma_1 d\gamma_2 \dots d\gamma_m, \quad (3.5)$$

where $n(\gamma_1, \gamma_2, \dots, \gamma_m, t)$ is the concentration of occupied states $1 \rightarrow m$, described by state parameters $\gamma_1, \gamma_2, \dots, \gamma_m$. In general $n(\gamma, t) = N(\gamma)f(\gamma, t)$, where $n(\gamma)$ and $N(\gamma)$ are the concentrations of occupied and available states respectively, $f(\gamma)$ is the occupancy of the state ($f = 1, \Rightarrow$ full state and $f = 0, \Rightarrow$ empty state). Also, the functions $n(\gamma)$ and $f(\gamma)$ are time-dependent.

The state parameters $\gamma_1, \gamma_2, \dots, \gamma_m$ determine the stability of the meta-stable state under the existing conditions of temperature and illumination intensity, i.e. they control the probability per unit time that the system will return to equilibrium. $n(\gamma_1, \gamma_2, \dots, \gamma_m, t)$ is the weighting function, or distribution that shows the concentration of occupied states possessing the parameters $\gamma_1, \gamma_2, \dots, \gamma_m$. Eq. (3.5) is a time-dependent and dose-dependent function since it increases during irradiation and decreases during stimulation.

In stimulated luminescence measurement, the emitted luminescence intensity during the returned of the system to equilibrium is recorded and observed. This luminescence intensity is proportional to the rate at which the meta-stable states decay, such that:

$$I(t) = \left| \frac{d\mu(t)}{dt} \right| = \mu^l(t)p(t), \quad (3.6)$$

where $p(t)$ is the probability per unit time of the decay of the meta-stable states $\mu^l(t)$. For first-order approximation ($l=1$), $\mu^l(t)$ can be approximated by $\mu(t) \approx n(t)$, where n is the total number of meta-stable states.

For optical stimulation, the probability p of optical ionization at a wavelength λ (with corresponding energy E) of the meta-stable state leading to a return of the system to equilibrium is given as:

$$p(E_o) = \Phi\sigma(E_o) \quad (3.7)$$

where Φ is the optical stimulation intensity and $\sigma(E_o)$ is the photo-ionization cross-section for the interaction of the meta-stable state with the incident stimulation photon. E_o is the threshold optical stimulation energy required to release charge and return the system to equilibrium. With reference to Eq. (3.5), we see that this corresponds to a case where $m = 1$ and $\gamma_1 = E_o$.

In the above representations, λ and Φ are all fixed values independent of time. For optical stimulation, the use of a fixed wavelength λ and a steady illumination intensity Φ to empty the traps results in a luminescence type known as *continuous wave OSL*, or CW-OSL.

3.3.2 The photo-ionization cross-section

The absorption of light of incident intensity $I_o(\lambda)$ at wavelength λ as a function of depth x in a solid is described by the Lambert-Beer Law, given as:

$$I(\lambda, x) = I_o(\lambda)\exp[-\alpha(\lambda)x], \quad (3.8)$$

where $\alpha(\lambda)$ is the absorption coefficient and $I(\lambda, x)$ is the intensity at depth x . The value of α depends on the optical absorption process occurring at the particular wavelength λ . A schematic representation of the various optical absorption transitions peculiar to wide-band-gap insulators like $\text{Al}_2\text{O}_3\text{:C}$ is presented in Figure 3.6. The type 1 transitions are called "defect" ionizations. An example is the ionization of the F-center in $\text{Al}_2\text{O}_3\text{:C}$ where absorption of a 6.05 eV photon will release an electron from the center to the conduction band (1a), and the electron subsequently recombines with a hole at a recombination center (1b) to produce TL and/or OSL [132]. Type 2 transitions are called "trap" ionizations. These are the transitions that make the $\text{Al}_2\text{O}_3\text{:C}$ crystal a very powerful luminescence dosimeter [132]. They result from the delocalization of electrons initially trapped during irradiation due to absorption of thermal or optical energy (2a) and the subsequent recombination of the electrons at radiative recombination centers (2b) resulting in light emission [129]. The

difference between transitions 1 and 2 is the magnitude of the transition energy required, as indicated by the size of the arrows. Lastly, the type 3 transitions are called inter-center transitions. They are due to movement of electron from radiation-induced defects to another intermediate energy level due to absorption of IR photon. This type of transition has been reported for minerals like feldspar [132].

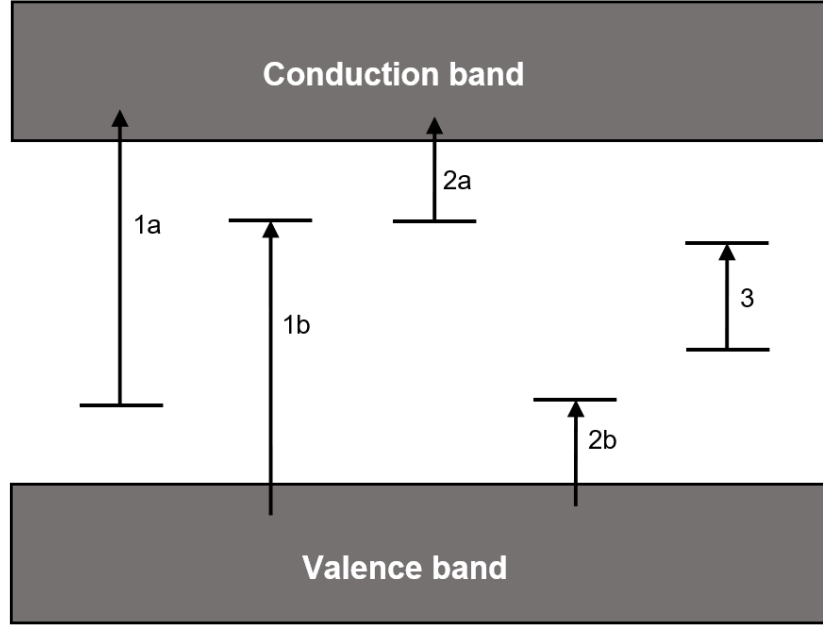


Figure 3.6: A schematic representation of the various optical absorption transitions peculiar to wide-band-gap insulators; (1a, 1b) defect ionization; (2a, 2b) trap ionization; (3) internal intra-center transition. Adapted from [132, Fig. 2.3] with permission. Copyright (2003), Elsevier.

The photo-ionization cross-section $\sigma(E_o)$ is arguably the most important parameter that dictates the type 2 transitions in Figure 3.6. The type 2 transitions are responsible for OSL/TL emission, photo-transfer effects, and optical bleaching of TL and/or OSL signals. They thus determine the stability of each trap during optical stimulation. The absorption coefficient for a defect-band optical transition at an optical stimulation energy $h\nu$ may be written as:

$$\alpha(h\nu) = n(E_o)\sigma(h\nu, E_o), \quad (3.9)$$

where $n(E_o)$ is the concentration of defects, each with an optical ionization threshold energy

E_o . The behaviour of the absorption coefficient $\alpha(h\nu)$ as a function of energy $h\nu$ should be edge-like in shape, since the absorption coefficient shape contains contributions from all the relevant continuum states in the band into which the electron is being excited [133]. Following the transition, the charge on the defect will change by one electronic charge and significant lattice relaxation may occur. This is also the case during the inverse process [134].

Some theoretical models for the energy dependence of $\sigma(h\nu, E_o)$ were presented by Bøtter-Jensen et al. [132] using a variety of assumptions that relate to potential energy in the vicinity of the defect. For a shallow electron trap where the electron can be considered quasi-free, the model is given as:

$$\sigma(h\nu, E_o) \propto \left[\frac{(h\nu - E_o)}{(h\nu)^5} \right]^{3/2} \quad (3.10)$$

For stable/deep traps like in wide-band-gap insulators, the model uses a delta-function potential for the defect. The energy dependence of the cross-section of the model is given as:

$$\sigma(h\nu, E_o) \propto \left[\frac{4(h\nu - E_o)E_o}{(h\nu)^2} \right]^{3/2}, \quad (3.11)$$

where σ reaches a maximum at $h\nu = 2E_o$, with σ increasing as $(h\nu - E_o)^{3/2}$ for $h\nu < 2E_o$, and as $(h\nu)^{-3/2}$ for $h\nu > 2E_o$.

In Eq. 3.11, the effective mass of the electron is assumed to be the same in the conduction band and in the trap. However, using the electron rest mass for the trap state and applying the parabolic band approximation leads to:

$$\sigma(h\nu, E_o) \propto \frac{(h\nu - E_o)^{3/2}}{h\nu [h\nu - E_o(1 - m_o/m_e)]^2} \quad (3.12)$$

The shapes of the three expressions for the photo-ionization cross-sections [132] presented above are shown in Figure 3.7 for a threshold energy $E_o = 3.0 \text{ eV}$, and $m_o/m_e = 2$. For each curve, $\sigma = 0$ for $h\nu \leq E_o$. The threshold optical ionization energy E_o is larger than the

thermal ionization energy (thermal trap depth, E_t) by an amount equal to the phonon energy, given as:

$$E_{ph} = S\hbar\omega_p, \quad (3.13)$$

where S is the Huang-Rhys factor and ω_p is the phonon vibration frequency [132, 135].

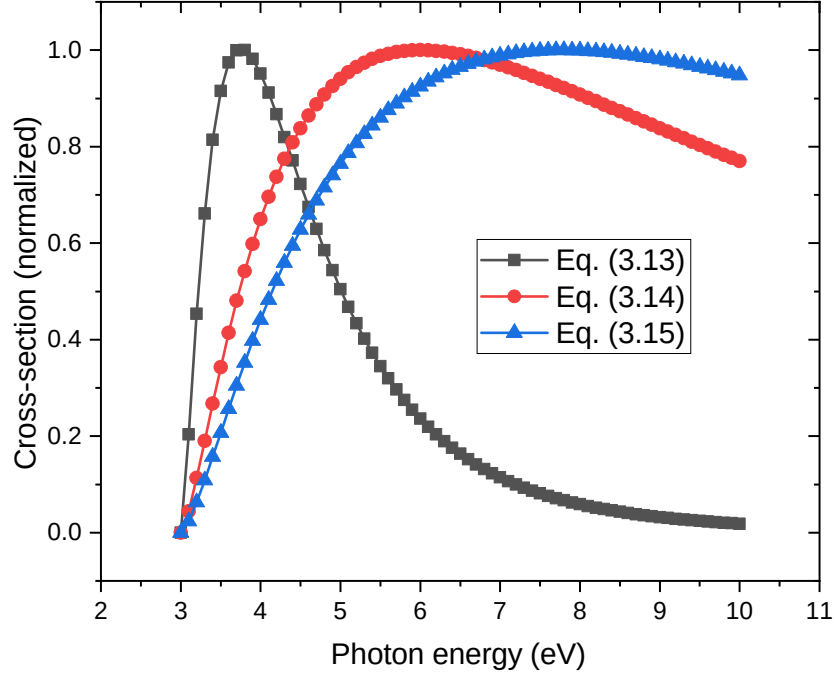


Figure 3.7: Comparison of the shapes of various expressions for the photo-ionization cross-section as a function of stimulation photon energy, obtained with a threshold energy $E_o = 3.0 \text{ eV}$, and $m_o/m_e = 2$. Each curve is normalized to a maximum of 1. Adapted from [132, Fig. 2.4] with permission. Copyright (2003), Elsevier.

For thermal stimulation, the stimulation is different from OSL because the temperature is increased at a constant linear rate. The probability p per unit time of thermal stimulation is thus given as:

$$p(t) = s \exp(-\Delta E/kT), \quad (3.14)$$

where ΔE is the energy difference between the trap and the conduction band, k is the Boltzmann's constant, T is the absolute temperature and s is the *attempt-to-escape* frequency factor. Therefore, Eq. 3.14 is basically saying the probability that an electron becomes detrapped is dependent on the energy difference ΔE , referred to as the energy depth of the

trap, the temperature T , and the the frequency factor s . The TL intensity can be obtained directly from this probability by using Eq. 3.14 on Eq. 3.6:

$$I(t) = pn(t) = n(t) s \exp(-\Delta E/kT) \quad (3.15)$$

Plotting this intensity as a function of the temperature give rises to the glow-curve mentioned earlier in Figure 3.5 b. As the temperature increases, the detrapping probability increases. The intensity initially increases, but reaches a local maximum as the rate of detrapping peaks, and then it drops to zero again. The fall of the intensity is due to depletion of trapped electrons. For a real material that contains multiple trap types located at various energy depths, a glow-curve may be made up of a number of glow-curve peaks, each corresponding to different trapping centers.

3.3.3 Models and rate equations

A series of non-linear, coupled rate equations can be used to model the transitions of charges between energy levels during irradiation and subsequent stimulation of a dosimeter. These equations are usually cumbersome and therefore several simplifying assumptions have to be introduced in order to arrive at analytical functions that describe the change of the luminescence intensity with time or temperature, and fundamentally, the dependence of the signal with absorbed dose. Such model must include the transport of electrons through the conduction band into the radiative recombination centers that trap holes. Some of these models are explored in the following sections based on the works and treatment of Kitis [130], Akselrod et al. [135], and Bøtter-Jensen et al. [132].

3.3.3.1 The OTOR model

This is the most common and the simplest energy band model [132,135]. The One-Trap/One-Recombination center (OTOR) model as the name suggests is a system that contains one type of electron

trap and one type of hole trap that acts as recombination center where recombination of electrons with the holes occurs, leading to luminescence emission (see Figure 3.8). This model comprises three stages: irradiation, relaxation and stimulation.

- **The irradiation stage:** during irradiation, charge carriers are produced. Electrons move to the conduction band and traps, while holes move to the valence band and recombination centers. So that the charge neutrality can be written as:

$$n_c + n = m_v + m, \quad (3.16)$$

where n and n_c (in cm^{-3}) are the concentrations of electrons in the traps and in the conduction band, respectively, and m and m_v (in cm^{-3}) are the concentrations of holes in the recombination centers and in the valence band, respectively. The differential equations describing the occupation of these energy states are:

$$\frac{dn}{dt} = n_c A_n (N - n), \quad (3.17)$$

$$\frac{dm}{dt} = m_v B (M - m) - n_c m A_m, \quad (3.18)$$

$$\frac{dn_c}{dt} = X - n_c A_n (N - n) - n_c m A_m, \quad (3.19)$$

$$\frac{dm_v}{dt} = \frac{dn_c}{dt} + \frac{dn}{dt} - \frac{dm}{dt}, \quad (3.20)$$

where N and M (in cm^{-3}) are the total concentrations of trapping states and recombination centers. A_n , A_m , and B (in $cm^{-3}s^{-1}$) are the trapping rate of electrons, recombination rate of holes and electrons at the recombination centers, and the trapping rate of holes, respectively. X (in $cm^{-3}s^{-1}$) is the dose-rate of excitation denoting the rate of production of electron-hole pairs due to exposure to radiation.

- **The relaxation stage:** this stage begins at the end of excitation and involves the settling of all charges in traps or recombination centers. So that n_c and m_v become

non-zero. Also, for this period, X will need to be set to zero in Eq. (3.18). If we consider thermal equilibrium at the end of this period, such that n_c and m_v are zero, then $n = m$ or put in another way, at the start of optical or thermal stimulation $n_0 = m_0$, where the subscripts "0" implies time $t = 0$ for the stimulation period.

- **The stimulation stage:** this stage involves the exposure to light or heat resulting in the release of trapped charges. During stimulation, it is assumed that trapped electrons transition to the conduction band but transitions of holes from and/or to the valence band do not occur so that $m_v \approx 0$ and Eq. (3.13) reduces to:

$$m = n_c + n \quad (3.21)$$

The other differential equations also change as follows:

$$\frac{dn}{dt} = n_c A_n (N - n) - np, \quad (3.22)$$

$$\frac{dm}{dt} = -n_c m A_m \text{ (since } m_v \approx 0), \quad (3.23)$$

$$\frac{dn_c}{dt} = np - n_c A_n (N - n) - n_c m A_m, \quad (3.24)$$

$$\frac{dn_c}{dt} = -\frac{dn}{dt} + \frac{dm}{dt}, \quad (3.25)$$

where p is the same time-dependent probability in Equations (3.7) and (3.14) that represents the stimulation mode.

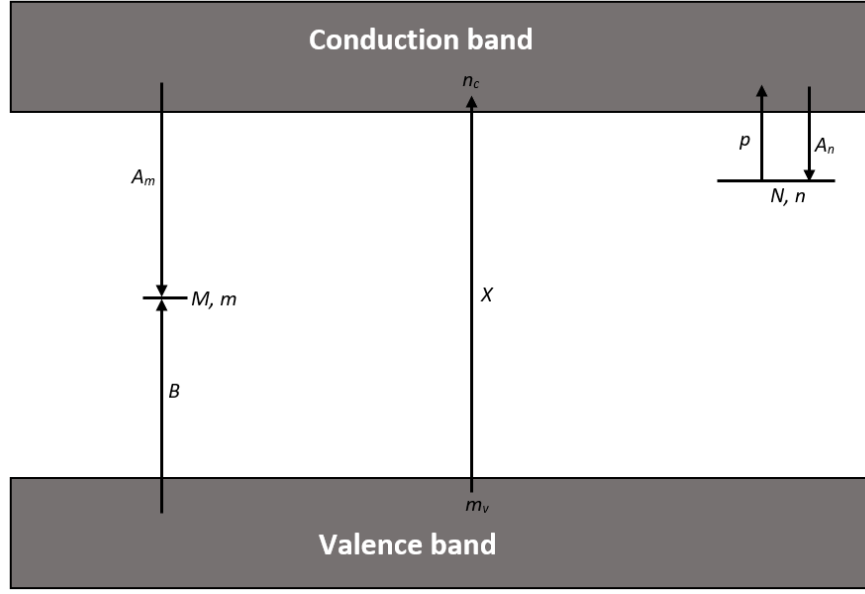


Figure 3.8: Schematic energy band representation for the one-trap-one-recombination center (OTOR) model.

In order to simplify the above differential equations, the quasi-equilibrium (QE) approximation is introduced. It is expressed as:

$$\left| \frac{dn_c}{dt} \right| \ll \left| \frac{dn}{dt} \right|, \left| \frac{dm}{dt} \right|. \quad (3.26)$$

An implication of this is n_c is always much less than n , so that $n \simeq m$ resulting in

$$\left| \frac{dn_c}{dt} \right| = 0, \quad (3.27)$$

and Equation 3.25 becomes

$$\frac{dn}{dt} = \frac{dm}{dt}, \quad (3.28)$$

which is related to the luminescence intensity $I(t)$ through the equation

$$I(t) = n_c m A_m = -\frac{dm}{dt} = -\frac{dn}{dt}, \quad (3.29)$$

Applying Eq. (3.27) to Eq. (3.24), will result to

$$n_c = \frac{np}{A_n(N - n) + mA_m}, \quad (3.30)$$

By replacing n_c in Eq. (3.29) by the right-hand side of Eq. (3.30), and since $n = m$, the following is obtained for the luminescence intensity:

$$I(t) = -\frac{dm}{dt} = -\frac{dn}{dt} = \frac{n^2 A_m p}{A_n(N - n) + nA_m}, \quad (3.31)$$

Eq.(3.31) is called the general one trap (GOT) equation. It was solved analytically by Kitis and Vlachos [136], 50 years after it was introduced [130].

The OTOR model can be expressed as first order kinetics for the case of weak or slow retrapping i.e. $A_n/A_m \ll 10^{-2}$ which implies that in the denominator of Eq. (3.31), $A_n(N - n) \ll nA_m$, leading to the first order differential equation stated in Eq. (3.6):

$$I(t) = -\frac{dm}{dt} = -\frac{dn}{dt} = np(t), \quad (3.32)$$

And for OSL, the solution of Eq. (3.32) is:

$$I_{OSL} = n_0 p \exp\{-tp\} = I_0 p \exp\{-t/\tau_d\}, \quad (3.33)$$

where n_0 and I_0 are initial concentration of electrons in the traps and the initial OSL intensity, respectively, and τ_d is the CW-OSL decay constant which is related to the photo-ionization cross section by $p = \Phi\sigma = 1/\tau_d$. This first order model leads to an exponentially decaying OSL intensity as the constant stimulation light intensity is applied to the sample. This leads to the eventual emptying of all traps and the OSL signal approaches zero (see example OSL curve in Figure 3.5a).

For TL, Eq. (3.32) can be further written as:

$$I(t) = -\frac{dn}{dt} = -\frac{dn}{dT} \frac{dT}{dt} = \frac{ns}{\beta} \exp\{-E/kT\}, \quad (3.34)$$

where a linear heating rate $\beta = dT/dt$ has been assumed, leading to the well known Randall and Wilkins [137] first order kinetic TL equation:

$$I_{TL} = \frac{n_0 s}{\beta} e^{-\{E/kT\}} \exp \left[-\frac{s}{\beta} \int_{T_0}^T e^{-\{E/kT'\}} dT' \right], \quad (3.35)$$

where n_0 is the initial concentration of electrons in the traps at time $t = 0$, T_0 is the initial temperature and T' is a dummy integration variable representing temperature.

In practice, OSL and TL curve shapes do not conform to this simple one trap, one recombination model. The simple reason is, in a real sample, there may exist multiple thermally and optically sensitive traps leading to two or more electron traps being emptied at the same time, each at its own rate with probably its own unique cross-section based on the stimulation conditions [130, 132, 135]. This case will be discussed briefly in the next section.

3.3.3.2 Multiple traps and recombination centers models

When there are for example two thermally or optically sensitive traps, with trapped charge concentrations n_1 and n_2 and stimulation rates p_1 and p_2 , the differential equations as given by [138] is:

$$\frac{dm}{dt} = -\frac{dn_1}{dt} - \frac{dn_2}{dt}, \quad (3.36)$$

and the solution is thus

$$I_t = n_{10} p_1 \exp\{-tp_1\} + n_{20} p_2 \exp\{-tp_2\}, \quad (3.37)$$

based on the superposition principle and assuming there are no interactions between the traps. This can obviously be extended to three or more sensitive traps, each depleting at their own unique rate during stimulation.

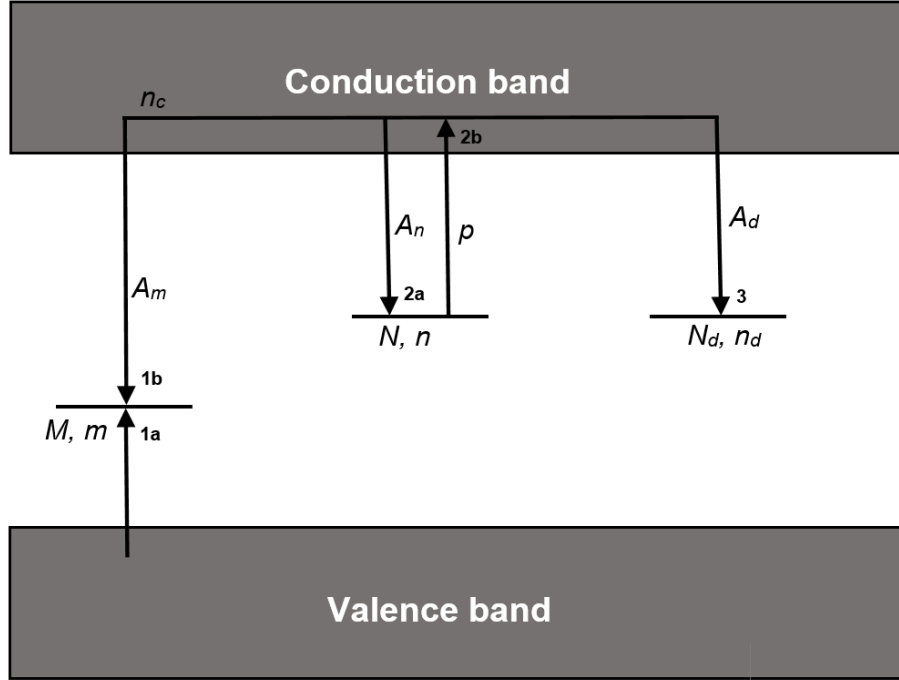


Figure 3.9: Schematic representation of the general transitions mechanism of multi-trap models. The transitions represented are 1a (hole trapping resulting in the creation of recombination center), 1b (recombination transitions), 2a (trapping of electrons in the dosimetry trap), 2b (optical stimulation transition), and 3 (capture of an electron by deep traps). For a real material, the three transitions can occur concurrently.

Whitney and McKeever [138] extended this to a distribution of traps. They used the one-dimensional Fredholm integral equation of the first kind to describe the OSL decay from a series of traps with distribution of threshold energies, E_o and stimulation photon energy $h\nu$. The equation is written as:

$$I_{OSL}(h\nu) = \int w(E_o) p(h\nu, E_o) dE_o = \Phi(h\nu) \int w(E_o) \sigma(h\nu, E_o) dE_o \quad (3.38)$$

where $w(E_o)$ is a weighting factor that defines the fraction of the OSL curve that is due to a particular trap.

Assuming the additional traps introduced into the energy band model are not optically sensitive, the OSL decay kinetics will still be affected because the optically insensitive traps can still trap charges released via optical stimulation. An example is shown in transition 3 of Figure 3.9, where a deep trap is able to capture free charges even though it is insensitive to

optical stimulation (too deep). In this model, the deep traps can be viewed as competitors to the recombination centers. The OSL intensity in this case is described by:

$$I_{OSL} = n_{10} p \exp\{-t/\tau\} - n_2 A_2 (N_2 - n_2) \quad (3.39)$$

where N_2 , n_2 and A_2 are the concentration of available traps, concentration of filled traps, and trapping rate or trapping coefficient, respectively for the deep trap.

This implies that the OSL intensity is reduced by an amount that depends on the relative rates of recombination and trapping into the deep, competing trap. The decay is no longer exponential since the second term in Eq. (3.39) is time dependent and approaches zero as $t \rightarrow \infty$ i.e. as the deep trap fills up. This description also applies to TL, with the only difference being the mode of stimulation. Other conditions or assumptions like the case where the competing centers are shallow and the case of multiple recombination centers can also be built into the multiple traps and recombination centers model. Detailed reviews of some of these cases can be found in the following literature [130, 132, 135, 138].

3.3.4 Luminescence model for $\text{Al}_2\text{O}_3\text{:C}$

The models described in the previous sections are for idealized materials. For a real material like $\text{Al}_2\text{O}_3\text{:C}$ (aluminum oxide doped with carbon), a more realistic model is required. This is because a real material contains multiple traps, some of which may be shallow, dosimetric or deep, and recombination centers that may be radiative (leading to luminescence production) or non-radiative. There may also be charge transfer processes involving the conduction band (current assumption) or not. In other words, real materials show a complexity of behavior which is too complicated and intricate to be captured by a single model. A relative realistic model for $\text{Al}_2\text{O}_3\text{:C}$ would be one that incorporates all the transition mechanisms in Figure 3.9. As a direct consequence of the trapping transition 2a, the concentration M , of the recombination centers increases with irradiation dose. However, this is not the case

for $\text{Al}_2\text{O}_3:\text{C}$ that has F^+ -centers recombination centers. The F^+ -center is a pre-existing oxygen vacancy occupied by one electron (see Section 5.2). During irradiation, transition 1b (recombination transition) in Figure 3.9 dominates due to the pre-existing F^+ -center concentration. Therefore, at high doses, the recombination center concentration decreases as dose increases [135, 139]. A detailed review of $\text{Al}_2\text{O}_3:\text{C}$ as a luminescence dosimeter will be presented in chapter 5.

3.4 Practical aspects of luminescence dosimetry

The goal of luminescence dosimetry is to obtain the dose in a material. The luminescence signal from luminescence dosimeters is assumed to be directly related to this dose. However, to retrieve accurate dose information, the response of the dosimeter must first be characterized by constructing a dose-response curve. The dose response curve is basically a graph of the measured signal versus the known dose or exposure (see Figure 3.10). For TL, the measured signal is obtained through heating the dosimeter via a metal plate by increasing the temperature of the plate linearly with time while measuring the luminescence signal via some combination of filters and a photo-multiplier tube (PMT). The actual signal is recorded as a function of the temperature of the plate and is commonly referred to as "glow curve". A portion of this glow curve, typically the area under the curve, is then related to the radiation exposure or dose. For OSL, the measured signal is obtained by shining light on the dosimeter. The source of the light can be a laser source or a light-emitting diode. The signal is detected as a function of the stimulation time. The stimulation in this case can be continuous, linear or pulsed with respect to time.

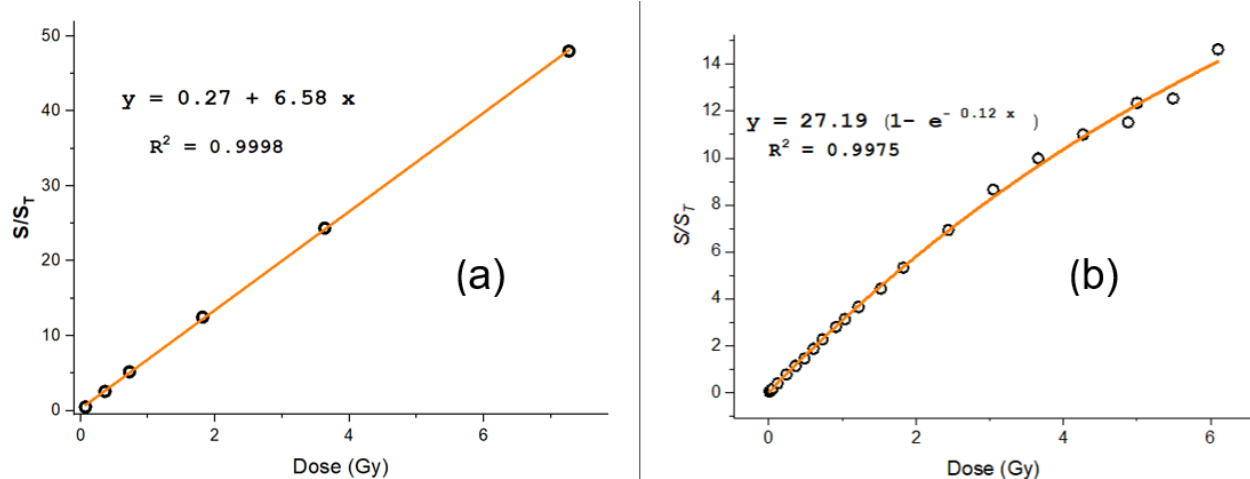


Figure 3.10: (a) OSL dose response curve for $\text{Al}_2\text{O}_3\text{:C}$ Luxel[®] dosimeters exposed to β radiation and (b) TL dose response curve for 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains exposed to β radiation. S/S_T represents the signal after normalization using a constant reference dose.

The dose response is typically created in the laboratory by exposing dosimeters to known doses. In a sedimentary environment, when a dosimeter is exposed to an unknown radiation dose from natural radioactivity, the absorbed dose is determined by comparing the "natural" luminescence signal to the laboratory-created dose response curve. The recovered dose is called the equivalent dose (D_e) and represents the laboratory dose that would produce a luminescence signal equal to the natural luminescence signal. An illustration of this is shown in Figure 3.11 for TL.

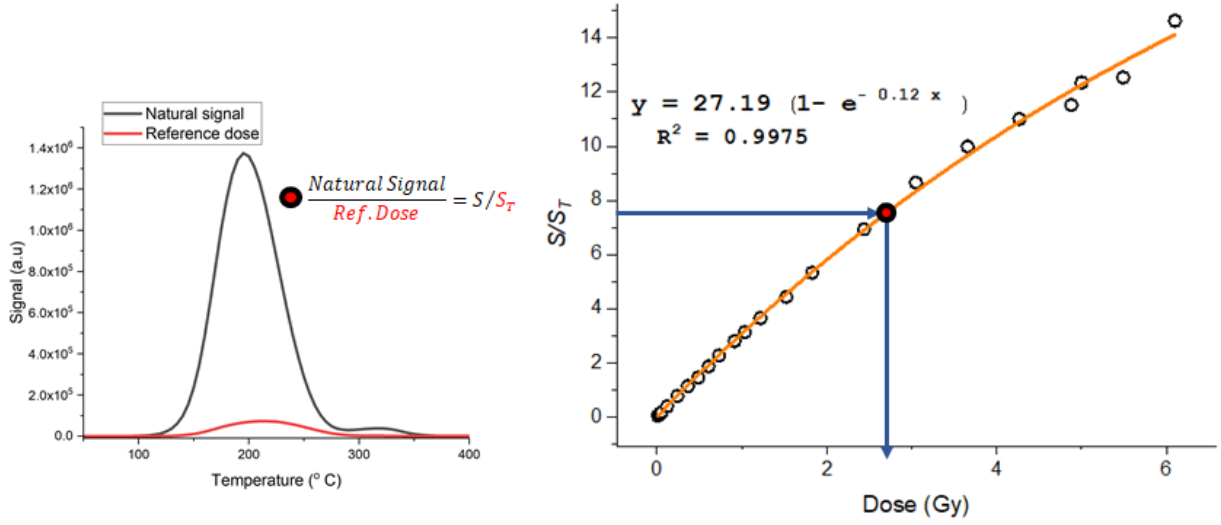


Figure 3.11: Illustration of determination of an unknown dose from the luminescence signal of a dosimeter irradiated from natural radioactivity using TL. This method of dose recovery can be used for any type of exposure.

The dose response of a dosimeter can either be linear or non-linear depending on the dose. A linear dose response describes a situation where the signal is proportional to dose. This is usually the case for low doses [139–141]. As dose increases, the response of the dosimeters tends towards non-linearity. This non-linear behavior has been associated with an increase in competition between traps (due to increased trap filling) and filling of recombination centers as the dose increases [140, 141]. This ultimately decreases the probability of dosimetric trapping and luminescence recombination for electrons, resulting in a gradual saturation of the dosimeter’s response [139]. Additionally, the dose response can also depend on the type and energy of the radiation, that is, the ionization density, as well as the dose rate [142].

This deviation from linearity can pose significant challenges in the use of these dosimeters. However, with proper characterization, these challenges can be managed to ensure high precision and accuracy. In this work, a rigorous characterization of the response of the luminescence dosimeters used was carried out. The result is presented and discussed in chapter 5.

Chapter 4

Modeling Radiation Transport in Sediments

4.1 Introduction

Radiation transport modelling in sediments is a key part of this dissertation. This chapter introduces the radiation transport codes used in this work: *DosiVox* and *DosiSed*. These codes are based on Monte Carlo (MC) Geant4 simulation toolkit. This chapter includes first a review of Monte Carlo radiation transport in general. Then Geant4, the most commonly used multipurpose radiation MC codes, is described in detail. Last but not the least, I review the architecture of *DosiVox* and *DosiSed*.

4.2 Monte Carlo simulations

The study of radiation transport problems was originally done using the Boltzmann transport equation that describes the change of macroscopic quantities in a thermodynamic system [30, 143]. The solution of the equation involves the determination of the probability density function of the macroscopic quantities in the entire six-dimensional phase space of the system using time derivatives of position and momentum [144–146]. The most common application

of the equation is a fluid where temperature gradients cause the transfer of heat from hotter regions to colder ones [144]. Although solving the Boltzmann equation is highly efficient for simple systems, the procedure becomes too challenging and complex when applied to limited geometries. In other words, numerical methods based on transport equations are only efficient when it comes to simple geometries, mainly for unlimited and semi-infinite media [146]. Monte Carlo (MC) simulation methods based on stochastic principles were developed as a powerful alternative to deal with transport problems. In MC methods, no transport equation is explicitly solved but rather, the average behaviour of a system is investigated through the Central Limit Theorem by simulating the movement of a large number of individual particles through a defined environment. With the right number of iterations, this effectively yields the same result as the solution to the Boltzmann transport equation [147], with the same interaction model, but is easier to implement for complex geometries. In fact, MC simulation in complex geometries is straightforward.

The main shortcoming of the MC method is its random nature: all the results are affected by statistical uncertainties, which can be reduced at the expense of increasing the sample population and, thus, the computation time. The first MC simulation of photon transport was done by Hayward and Hubbell (1954) [148]. They generated 67 photon histories using a desk calculator. Generally, for the interaction of ionizing radiation with matter, MC assumes that all probabilities (cross-sections) for interaction events in the life history of a particle are known. It involves following the primary particle and its secondaries throughout their life history. The interaction probabilities are used in each stage of the life history to determine the consecutive "state" of the particles until their energy is completely dissipated [149].

4.3 Geant4

Geant4 is arguably the most well-known multipurpose radiation MC toolkit mainly because of its flexibility when designing experiments and easy access to its physics libraries. It can

be defined as a toolkit for building simulations of particle-matter interactions using MC methods [150–152]. It started in 1993 at CERN, and is now being developed and managed through various international collaborations. The version used in this work is 10.1. As a toolkit, it comes with a full set of libraries written in C++ with functionalities for creating virtual geometry, and filling it with specified materials. The software system transports the particles emitted into the virtual geometry by simulating the particle interactions with matter based on the MC method. The data from the simulation interactions can be retrieved in various forms and interpreted.

The code libraries of Geant4 are accessible under a free license online (<http://geant4.web.cern.ch/geant4/>), and can be modified according to the user's need. It also allows users to add and integrate specific functions, other C++ codes and libraries in the construction of their simulation project, as long it is under the framework of Geant4. These are some of the advantages of Geant4 compared to MCNP that is more restrictive in use. MCNP is a general-purpose Monte Carlo N-Particle code that is also used for the transport of particles. It is also worth mentioning that Geant4 definitely benefits from an active development from its international collaboration and community. Geant4 is currently applied in a wide range of applications that include DNA damage simulations, cosmic radiation, archaeology, medical and space science.

There are three mandatory objects, called "user classes" that must be well defined for any Geant4 program.

1. The "DetectorConstruction" class, for constructing volumes for virtual geometry and defining the properties of the materials to fill the volume. The virtual geometry can be a detector or the simulation environment e.g. sediment.
2. The "PhysicsList" class, for specifying all necessary particles, all necessary interaction processes with the proper particles assigned, and the production (cut-off) ranges, as well as assigning them to their respective volume.

3. The "PrimaryGeneratorAction" class, for defining the details of the initial or primary particles (e.g. type, initial kinetic energy, position, direction, etc.). Detailed information on these and other classes can be found in the Geant4 toolkit manual [153,154].

In general, in Geant4 simulations, the class "DetectorConstruction" is used to create a virtual world, then primary particles are emitted inside the virtual world based on the parameters specified in the "PrimaryGeneratorAction" class. The simulation of each primary particle, from its emission to the complete absorption and the absorption of all its secondary particles due to interactions in the volume or due to escape from the volume is recorded as an "event". The "run" represents all the events in a simulation and is normally chosen at the start of the simulation.

4.4 *DosiVox* and *DosiSed*

DosiVox and *DosiSed* are two Geant4 codes for simulation of radiation transport in sedimentary environments (*DosiVox*: **D**osimetry in **V**oxelised volumes and *DosiSed*: **D**osimetry in **S**edimentary volumes). *DosiVox* is the version of the code that went public and has the advantage of being more user friendly, not requiring any major programming skill. *DosiSed*, on the other hand, was developed after *DosiVox* as a code that focuses mainly on random packing of grains. It has not gone public. However, it has the advantage of being more robust and flexible when it comes to modeling sediments as grains of different sizes and minerals. They both allow modelling of sedimentary samples by defining a "World" volume for simulation in a 3D grid (see Figure 4.1), where mineral compositions and associated radioactive contents (U, Th, K) can be defined in each voxel. The division of the volume into voxels results in a geometric grid produced by nested parametrization. Nested parametrization is a replication of the same geometry (grain packing and voxel) with the possibility of changing the materials inside each replicated unit. This ensures a considerable reduction in the time and memory required for geometry optimization and navigation during simulations [155]. Each element of

the grid; the voxel, is a homogeneous rectangular parallelepiped. It is also worth mentioning that since these codes were originally designed for archaeological sediments, the vertical Z-axis is oriented downwards, in order to directly represent the depth in a sediment.

In *DosiVox*, detectors are defined as "parallel worlds" and can be superimposed on the grid to record the energies of the primary emitted particles and energy deposited in each segment of the grid divided by the mass of the material present in the center of segment. The first of these detectors, the cylindrical probe (not available in *DosiSed*), is always present in every simulation but does not interact with the particles or the materials. In *DosiSed*, there is no such overlapping; all the grains are enclosed in their voxels using a Geant4 detector construction geometry called "regular world". Another detector present in both, called the solid detector is made up of random grain packing, and is used to model grains in a sedimentary environment (see Figure 4.2). This detector records all energy of interaction happening in the volumes it defines. It is also sensitive to all particles, each grain created acts as an independent detector recording the energy emitted and the dose it receives during the simulation. The user has the option of emitting particles from the grains or from the material surrounding the grains. More information on the use of detectors on DosiVox can be found in the cited literature [153, 155].

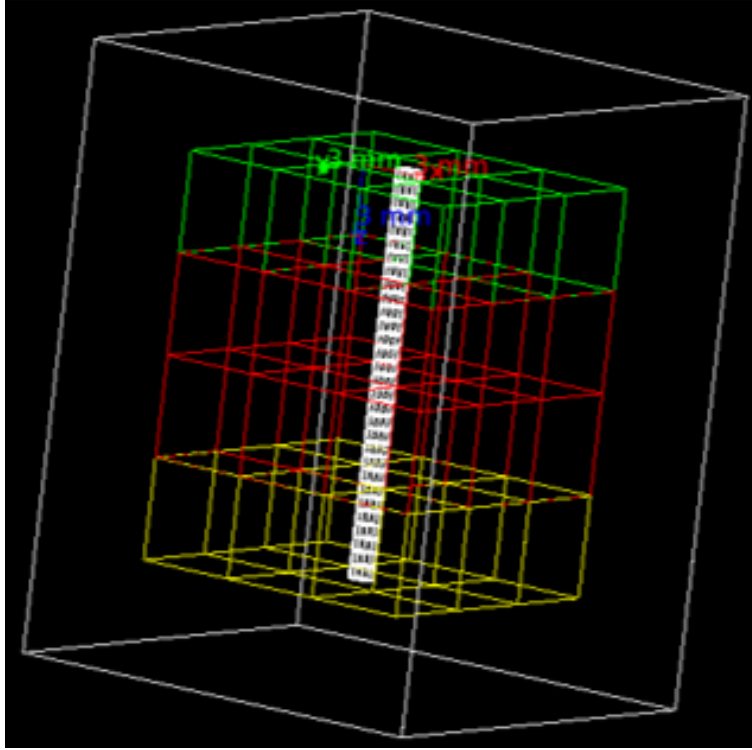


Figure 4.1: Example of a *DosiVox* voxel grid showing a $3 \times 3 \times 3$ voxels grid and the cylindrical probe at the center of the grid.

4.4.1 Modeling sedimentary grains by random grain fillings

The homogeneous spheres representation is one of the most common methods of modeling sedimentary grains [30]. Although, the congruity of this representation can be debated, mostly due to effects linked to the form [156] or internal heterogeneity [29, 157, 158], it remains however, one of the most effective and practical ways of studying dose-rate distributions in grains from a statistical point of view. This is the method for modeling grain distributions in *DosiVox* and *DosiSed*. In *DosiSed*, the detectors (grains) are created in the "regular world" equivalent in dimension to the defined voxel. In *DosiVox*, the grains are created in the parallel "DetectorWorld" world of a parallelepiped box that is superimposed on the "mass" world (where the materials are created) during simulations, thus leading to an overlapping of the volumes of the two worlds. The dimensions of the parallel world are defined by the user as a percentage ($\leq 100\%$) of the dimensions of a voxel.

The box is positioned at the location of a voxel chosen as detector, and it is possible to apply an offset to it to make sure it is within the voxel, when the size of the box is smaller than the size of the voxel. To ensure consistency in the choice of the point of emission of the primary particles, the box must not protrude from the voxel during the superposition of the parallel world with the mass world. In a situation where this occurs, the program will close with an error message announcing this.

Inside the box, spheres will be placed at random, in order to simulate a set of sedimentary grains (see Figure 4.2). The total number of spheres is determined by the size of the box, the grain volume fraction and the distribution of their sizes. The latter two being specified for each material by the user via a "pilot file" (see section 4.4.2). The component serving as grain material and its density are also specified for each material.

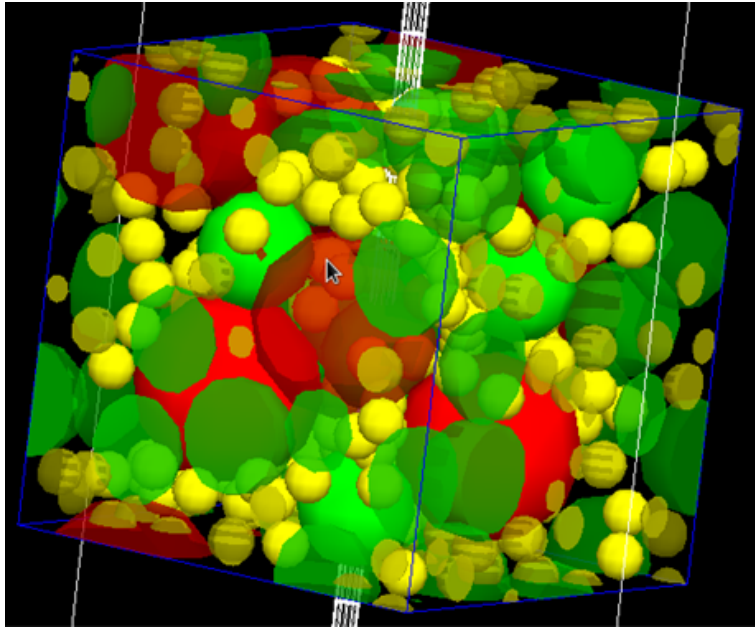


Figure 4.2: Example of *DosiVox* random packing of grains of different sizes in a defined voxel. The grains that appear transparent are those cut by an edge of the box or voxel that they fill (see Figure 4.3).

For *DosiVox*, it is important to mention that the concept of this detector is not to add grains on top of a material, but rather to create the grains out of the material with a level of modeling detail for which the grains, or at least part of them, are discernible from the

material matrix, and can thus explicitly interact with the simulated particles [153]. In the same vein, it was necessary to keep the same average absorption factor both inside the voxel containing the box of grains and the other voxels containing the same material. The mass of the material composing the grains is obtained from the constituents of the material defined in the detector voxel. The mass is calculated using the density and the volume fraction of the grains. A readjustment of the chemical composition and the density of the matrix is calculated according to Eqns. C.1 – C.4 listed in Appendix C, internally by the program. This implies that the bulk compositions of the materials in the model are some form of average compositions comprising the materials of the grains and the matrix specified by the user in the pilot file. These equations can also be used by the user as a second check for densities or compositions of the bulk materials in the simulation space. The equations are presented in Appendix C.

Random grain stacking is a common problem in mathematics [159], and the processing power of high-performance computing is normally required to solve it because of the number of calculation loops involved and the rigorous algorithm involved. Therefore, any calculation of random stacking of spheres like in the case of these codes, involves a number of mathematical loops increasing exponentially with the number of spheres and their compactness in the stacking volume, resulting in a proportionate increase in computing time. Considering current computer processors and the algorithm implemented in these codes, a computation time of several hours should be expected for a stack of the order of a thousand grains. This time can increase up to several days and even weeks when the volume fraction of grains requested exceeds 50%.

During random positioning of the grains, as long as the compactness is less than 35%, a non-overlap test to make sure the grains are not overlapping is sufficient after every random placement of a new grain. (see [153] for details of this and more information for the case where the compactness exceeds 35%). In the case where a placed grain protrudes from the box, the overlaps are tested as if the part coming out of the box entered it on the opposite face (see

Fig. 4.3). This concept, called "Periodic limits", makes it possible to obtain a homogeneous packing of grains in the whole volume, because all the grains are then statistically subjected to the same constraints of placement without edge effect (the loss of charge particle equilibrium at the edge of the model geometry), as long as the medium is virtually infinite.

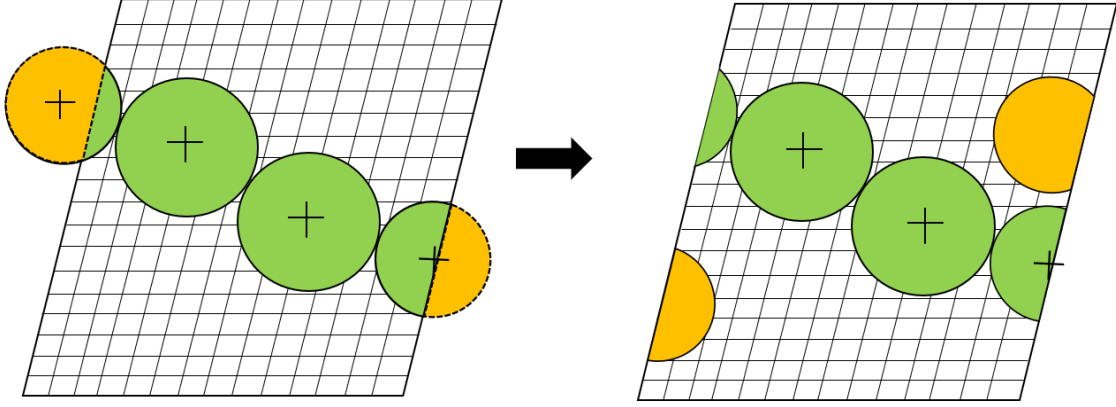


Figure 4.3: Schematic representation of the principle of periodic limits in a grain stack. The pieces of grain protruding from the box in which they are packed are transferred to the opposite sides, ensuring the statistical homogeneity of the packing.

The periodic limits in a parallelepiped, mathematically speaking, lead to a translation of the Cartesian coordinates of plus one, zero or minus one unit along each axis, the unit being the length of the side of the parallelepiped concerned. This effectively makes it possible to obtain a virtual replication of the parallelepiped according to a 3D mesh, where the overlap of each grain placed in the central volume with other grains in the central volume are tested as well as their overlap with grains in the replicated volume (see Fig. 4.4).

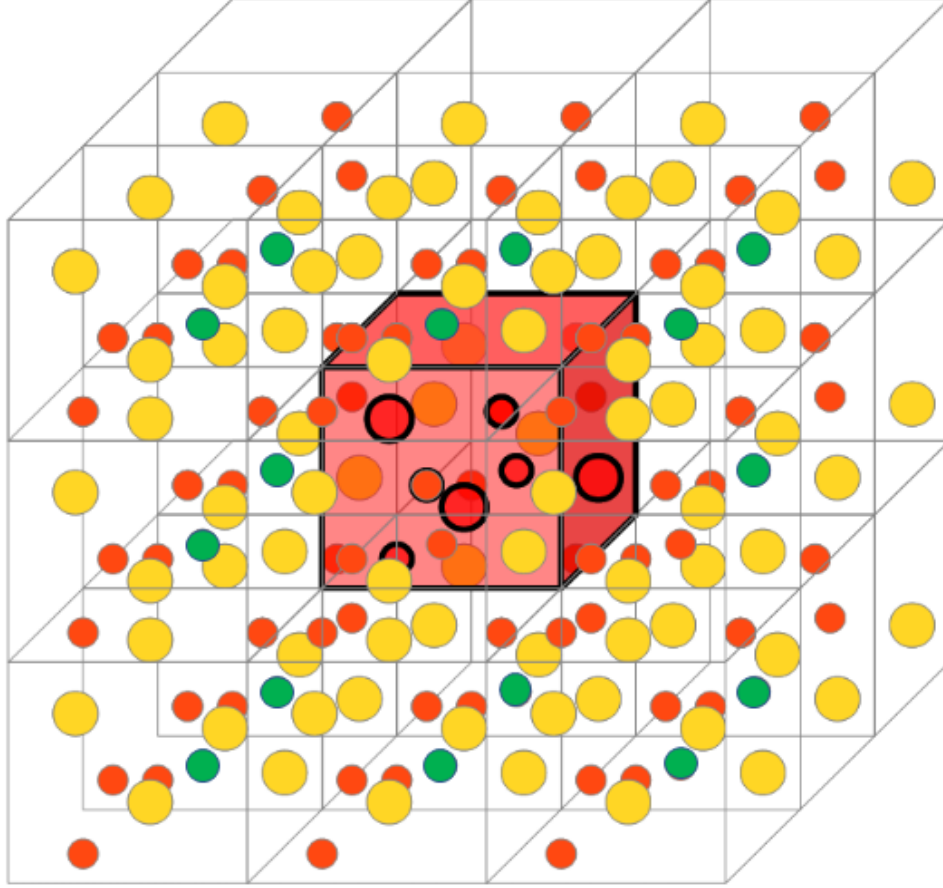


Figure 4.4: Schematic representation of the virtual geometry involved in the calculation of periodic limits. The periodic boundaries are created for stacking in the central volume, the other 26 adjacent volumes are mathematical replicas to perform the calculation. Adapted from [153, Fig. 7.8]

Another important aspect of these codes that needs to be discussed is the recording of dose deposited in each grain, since the grains can be used as detectors. In this regard, only whole grains, i.e. a grain not cut by the edges of the box can be used as a detector. Once grain positioning is completed, *DosiSed* (or *DosiVox*) counts the number of uncut grains (detector grains). If the number is zero, a new positioning algorithm is executed. However, this rarely happens, except where the requested grains are relatively large in size compared to the dimension of the box. This type of process is known to be effective and has been used for other programs like the PackLSD program [159]. The packLSD program is a robust molecular-dynamics code that was used for random grain packing calculations in Nathan's et.

al., (2003) [30] and Gu  rin et. al. (2012) [29].

4.4.2 The Pilot Text File

The pilot file or PTF in *DosiSed* (or *DosiVox*) is a file that contains a set of information organized according to certain rules. It is the most important part of the simulation for the user since it is what the user uses to create the model and to control the simulation of radiation. It comprises executable lines of codes with non-executable comments separated out on each line using the symbol `#`.

An example of the PTF of a model is presented in Table 4.1. The model is for a sample composed of a mixture of Thorite (ThSiO_4) grains, Aluminum oxide (Al_2O_3) grains, and Quartz (SiO_2) powder. A detailed description of all entry parameters can be found in Appendix C. Here it suffices to explain that the pilot text file is used to define (i) geometry, including voxel and grain size, (ii) the radioactive elements, concentrations and particles, and (iii) the chemical composition of the material.

Table 4.1: An example of Pilot Text File (PTF) for *DosiSed* program. The comments for each line of code are written after the # symbol in teal. The comments are not interpreted by the program. A detailed explanation of the different entries can be found in Appendix C

Line number	Executable code	Comment
1	Th_AlGr_Kb	# the name the result file is saved as
	# particle emission and properties #	
2	1000.	# ($\times 1000$) particles emitted
3	1	# 0 = alpha, 1 = beta, 2 = gamma
4	0	# radioactive elements, 0 = K, 1 = U-series, 2 = Th-series, 3 = User defined
5	3.0	# particles max range in mm
6	1.0	# cut-off value, in micrometer
	# voxelized geometry parameters #	
7	3.8	# cubic voxel size in mm
8	6 6 1	# number of voxels in the detector by X,Y and Z axis
9	0	# 0 = fixed medium, 1 = infinite medium
10	0	# 0 = random grain composition, 1 = recurrent grain composition
	# grains description #	
11	1.93	# total grain compacity in percent
12	2	# number of grain types
13	5	# number of grain sizes
14	# D	= spherical grain diameter in mm
15	# f	= volume fraction of this diameter
16	# Tn	= percent of grain type n in this diameter
17	D f T1 T2	
18	0.350 13.06 0 100	
19	0.300 13.05 0 100	
20	0.260 13.05 0 100	
21	0.090 30.42 100 0	
22	0.070 30.42 100 0	
23	Mtx T1 T2	
24	0 100 0	# relative percent of K in Mtx = matrix and Tn = Type n grain
25	0 100 0	# relative percent of U series in Mtx = matrix and Tn=Type n grain
26	0 100 0	# relative percent of Th series in Mtx = matrix and Tn = Type n grain
27	0 100 0	# relative percent of user defined radio-element in Mtx=matrix and Tn=Type n grain
	# material description #	
28	# new components	
29	1	# number of new component(s)
30	####	ID, name, density, number of atom type, list of atom symbol/number of atom of this symbol
31	18 thorite 5.35 3 Th 1 Si 1 O 4	
32	# material(s)	
33	0	# WF in the matrix, in percent
34	####	ID, name, density, number of atom type, list of component ID/mass fraction
35	0 def_matrix_mixture 1.129 1 1 100	# matrix
36	1 def_thorite 5.35 1 18 100	# type 1 grain material
37	2 def_alumoxide 3.97 1 2 100	# type 2 grain material

The structure of the PTF file for *DosiVox* is slightly different from that of *DosiSed* presented in Table 4.1. However, the functionality is the same. For another example of the PTF file and a detailed explanation of the line of codes see the *DosiVox* manual ². Also for free download of *DosiVox* see the link ¹.

¹<https://www.iram-at-crp2a.cnrs.fr/spip/spip.php?article144>, last accessed on May 6, 2022

Computer Requirement

Monte Carlo computer simulations require high computational power for smooth operation. For normal computers (core i5, 8GB RAM), statistically accurate simulations can be obtained at the expense of time. The speed of each simulation depends on the Random Access Memory (RAM) of the computer. It is also possible to run several parallel simulations in the limit of the computing cores available and to compile the results for each of them using multi-core computers. The simulations in this work were performed with a DELL Precision 3431 computer (Intel Core i9 with 64 GB RAM). This computer has the capacity to perform up to 16 parallel simulation at a time.

Chapter 5

Characterization of Dosimetric Materials and Development of Measurement Procedures

5.1 Introduction

One of the objectives of this dissertation is to measure the dose distribution of the natural background radiation in well-characterized sedimentary samples using highly sensitive $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains. These dose distributions are then compared to the microscopic dose distributions obtained from Monte Carlo computer modelling of the same samples. The goal of this comparison is to validate the computer simulations as a reliable tool for modelling microscopic sedimentary dose rates.

Natural background radiation is composed of alpha, beta, and gamma radiation emitted by radioactive nuclides in the mineral matrix of sediments. At the microscopic level, alpha radiation is the main component of local dose-rate variations in sedimentary environment due to its limited range (10–20 μm), followed by beta radiation. Gamma radiation on the other hand, can be considered uniform in the range that this study will cover ($\sim \mu\text{m}$ to

few cm). In trapped charge dating, one way to measure radiation in the laboratory is to prepare samples using natural sediments and to bury dosimeters in them for some duration to allow for radiation exposure of the dosimeters. Then, using luminescence dosimetry, the equivalent dose to the dosimeters can be estimated. However, when $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains are mixed with a sediment, the measured dose is the combined result of alpha, beta, and gamma radiation. Therefore it is necessary to develop methods to separate the components.

This chapter provides the practical and theoretical basis for the use of $\text{Al}_2\text{O}_3\text{:C}$ dosimeters for the dose measurement. It provides a detailed description of the material and the equipment used for OSL and TL measurements. It touches on the various forms of the dosimeters utilized to allow for separation of the alpha radiation dose from the beta radiation dose. The luminescence properties of the dosimeters are extensively characterized and discussed. Last but not the least, the chapter reports in detail, the development of robust and reliable measurement protocols that were used for all OSL/TL measurements in this dissertation.

5.2 $\text{Al}_2\text{O}_3\text{:C}$ as a dosimetric material

When $\text{Al}_2\text{O}_3\text{:C}$ was introduced as a TL dosimetric material [160], it enabled measurements that require a high level of sensitivity, particularly for environmental dosimetry using natural sediments. The dosimeter possesses a TL sensitivity that is 40–60 times greater than that of the commonly used LiF TLD–100 [120] making it ideal for low dose rate measurements. However, one of its draw-backs is thermal quenching, which causes the material’s TL sensitivity to vary strongly with higher heating rates. Another limitation of the material is its sensitivity to light which requires the material to be handled in light-tight conditions. However, it is this light limitation/optical nature of the material that makes it a powerful candidate for OSL dosimetry. Its use as an OSL dosimeter eliminates the need for heating and thus simplifies the measurement process.

$\text{Al}_2\text{O}_3\text{:C}$ crystals are grown for dosimetric purposes in a highly reducing environment at

low partial pressure of oxygen in the Stephanov Process [135]. This introduces stable oxygen vacancies. Occupancy of this vacancy by two electrons gives rise to a neutral F-center. When the vacancy is occupied by one electron, a positively charged F^+ -center with respect to the lattice is formed. These centers are associated with strong absorption and thus play a key role in the high luminescence output (TL and OSL) of $Al_2O_3:C$. Luminescence is produced by F-centers at 420 nm with a long lifetime (35 ms), on the other hand, emission from F^+ -centers is centered at 330 nm and has an extremely short lifetime (< 7 ns). OSL and TL sensitivity of $Al_2O_3:C$ can be increased by increasing the concentration of the F^+ -centers. The creation of F^+ -centers in $Al_2O_3:C$ during crystal growth in a highly reducing environment is a result of charge compensation. A divalent carbon ion substitutes a trivalent Al^{3+} -ion [160]. Figure 5.1 shows one possible configuration of this defect as presented by Akselrod et. al. [135].

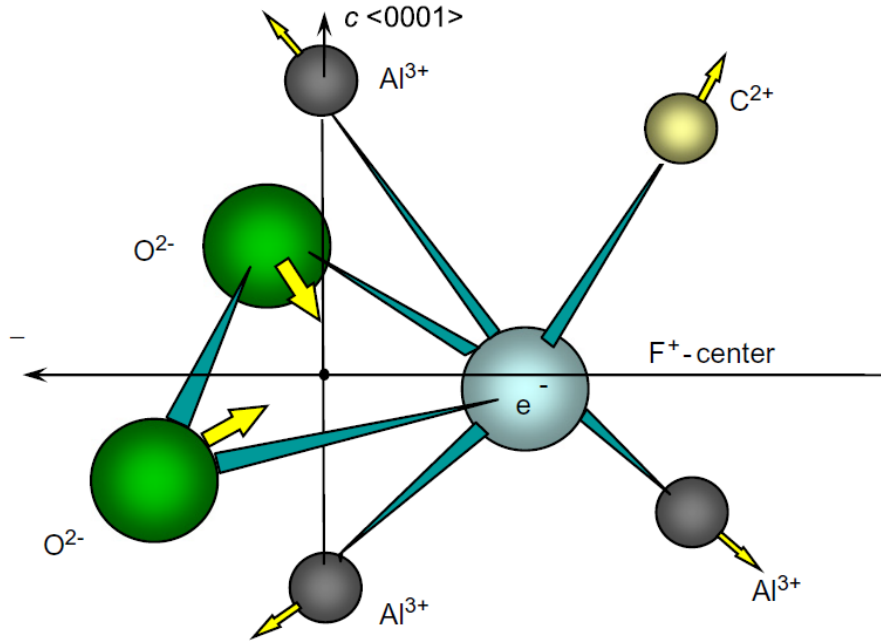


Figure 5.1: Suggested configuration of an F^+ -center defect created when trivalent Aluminum ions are substituted by divalent Carbon ions. Reprinted from [135, Fig. 12] with permission. Copyright (2007), Elsevier.

One fundamental advantage of $Al_2O_3:C$ as a dosimeter is that it has a wide energy band gap of 9.5 eV, allowing for the presence of deep and thermally stable traps and recombination

centers. Its dosimetric TL peak measured at a heating rate of 5 K/s is located at about 450K. Thermal activation energy obtained from TL peak analysis is about 1.5 eV. The electron trap associated with this TL peak is very stable at room temperature. The optical range of this trap is equal to 450 nm (2.75 eV). This suggests the range of wavelengths suitable for optical stimulation [135]. Blue light is considered the most efficient for OSL stimulation of $\text{Al}_2\text{O}_3\text{:C}$. However, because of the need to discriminate the stimulation light from the broad F- center emission centered at 420 nm (see Figure 5.2), green light with wavelengths longer than 500 nm is preferred.

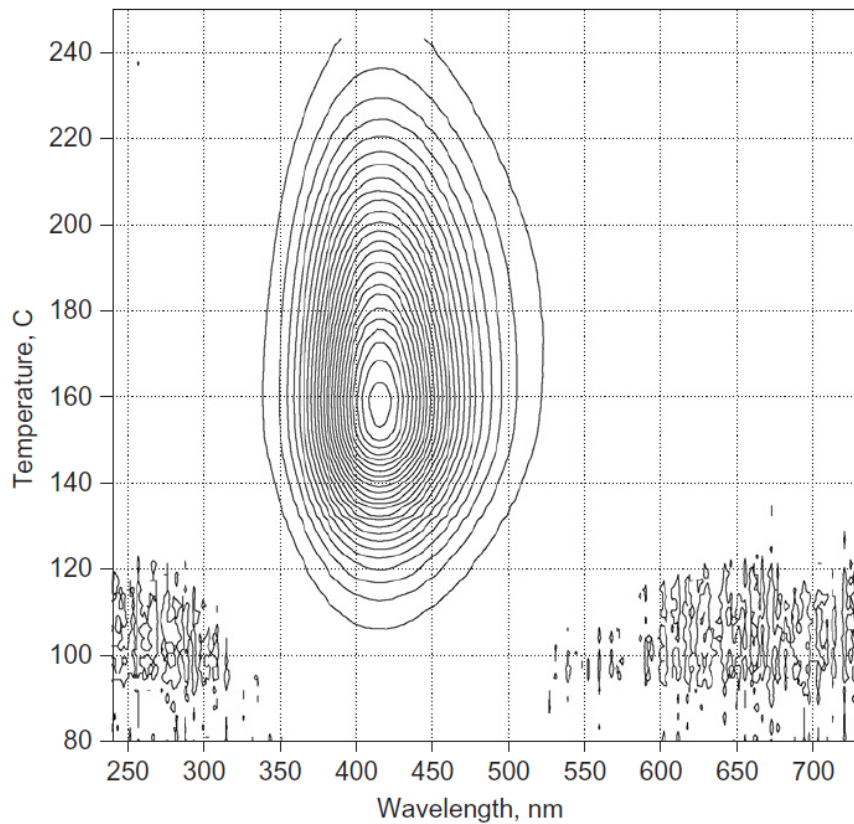


Figure 5.2: Contour plot of TL emission from an $\text{Al}_2\text{O}_3\text{:C}$ crystal at low 0.1 K/s heating rate. Reprinted from [135, Fig. 15] with permission. Copyright (2007), Elsevier.

Some of the features of $\text{Al}_2\text{O}_3\text{:C}$ that make it one of the most used dosimeters are:

- high sensitivity to radiation; approximately 60 times greater than LiF:Mg,Ti ;
- erasable and reusable via bleaching or annealing;

- low background noise and a $0.1 \mu\text{Gy}$ dose threshold;
- low fading during storage in the dark;
- wide dose range from 10^{-7} Gy to 10 kGy;
- relatively low effective atomic number ($Z_{eff} = 11.28$);
- linear dose response for doses less than 2 Gy.

Some drawbacks include; filling of deep traps during high dose irradiation which causes partial optical bleaching and regeneration of dose after optical bleaching. To avoid this, the use of high-temperature annealing to completely zero the dosimeter is recommended prior to use. The presence of both shallow and deep traps affects the performance of the material as an OSL dosimeter. Also, shallow traps introduce a temperature dependence to the OSL signal which can influence the shape of the decay curve. Finally, the competition between traps affects the sensitivity of the material which in turn leads to a non-linear dose response as dose increases [131, 135].

5.3 Materials and Methods

5.3.1 Dosimeters

One of the objectives of this dissertation is to measure α -, β -, and γ - dose distributions separately in well-defined sedimentary samples. This requires dosimeters in different forms because when a dosimeter is mixed with radioactive sediment, it absorbs the combined spectrum of α -, β -, and γ - radiation. Therefore, in this work, $\text{Al}_2\text{O}_3\text{:C}$ dosimeters were used in two forms:

Aluminum oxide ($\text{Al}_2\text{O}_3\text{:C}$) Luxel[®] tapes and 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains produced by Landauer, Inc., (Glenwood, IL., see Figure 5.3).

The Luxel[®] dosimeter tapes consist of crystalline powder grains (20 – 90 μm) sandwiched in a thin film between two polyester sheets for a total thickness of 0.3 mm [132, 161]. The tapes were cut into 6.3 mm diameter disks using a hole puncher for easy handling. The Luxel[®] tape was used to measure the combined spectrum of beta and gamma radiation since its outer polyester sheets are thick enough to absorb alpha radiation so that only beta and gamma radiation reaches the $\text{Al}_2\text{O}_3\text{:C}$ grains sandwiched between the sheets. The beta-only component of the dose was retrieved by taking advantage of the fact that, based on the range of gamma radiation, its radiation field can be considered uniform and thus can be measured via control samples as part of the uniform background radiation dose. To avoid melting of the polyester sheets during the heating required for TL, OSL was employed to measure the Luxel[®] cutouts.

The 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains on the other hand, measure the combined α -, β -, and γ - dose in sedimentary samples. A method was devised to separate the components. It is possible to desensitize the grains to alpha radiation, following the method reported by Akselrod et. al. (1993). The TL sensitivity of $\text{Al}_2\text{O}_3\text{:C}$ single crystals relies on the presence of oxygen vacancies (see section 5.2) and can be influenced by changing the amount of these vacancies in the outer layer [162]. To achieve this, Akselrod et. al. (1993) [162] annealed the crystals in air at 1200 °C for 30 min. Their result shows that this process led to the diffusion of oxygen and thus the destruction of oxygen vacancies up to a depth of 20 μm (range of alpha particles in the material).

In this work, a similar process was applied to the highly sensitive 250 – 355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains to remove alpha radiation sensitivity. While the "regular" $\text{Al}_2\text{O}_3\text{:C}$ grains were used to measure the combined spectrum, the alpha-desensitized $\text{Al}_2\text{O}_3\text{:C}$ grains were used to measure only the beta and gamma components of the radiation field as a way of studying the dose distribution without the effect of alpha radiation. The gamma component was separated out from these dose distributions following the same procedure used for the Luxel[®] dosimeters. The measurement was done via TL. The dosimeters were sieved from the sample, sorted out

with a needle and placed on sample cups using silicon oil.

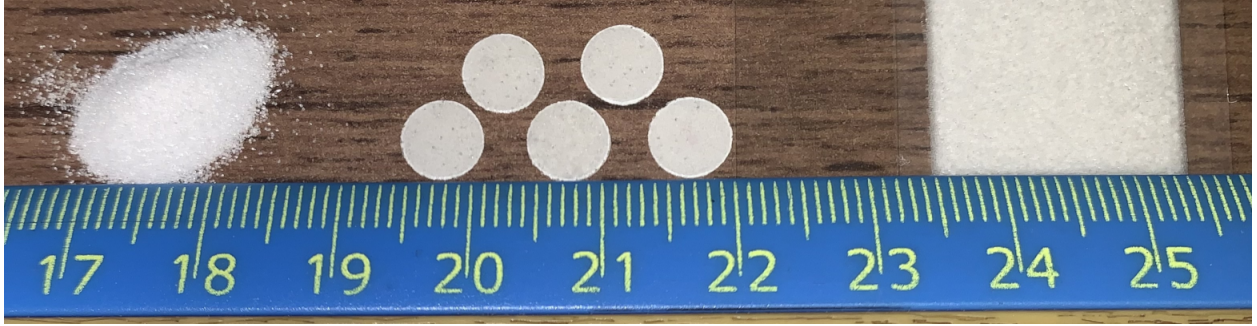


Figure 5.3: Dosimeters used in this work. From the left, 250–355 μm grains of $\text{Al}_2\text{O}_3\text{:C}$, 6.3 mm diameter round pieces of Luxel[®] tape, and a section of Luxel[®] tape.

5.3.2 Bleaching and annealing of dosimeters

The preparation of the dosimeters prior to measurement is required to remove any background signal accumulated during storage and transport to ensure that no previously trapped charges skew the results. The Luxel[®] dosimeters were thus prepared by bleaching for 8–12 hours. The bleaching unit consists of a halogen lamp and a filter. The light from the lamp is filtered using a Schott GG-495 (model FSQ-GG495) colored glass long-pass filter, 50.8 mm $\times$ 50.8 mm and thickness, (3.0 ± 0.1) mm [163]. It has a damage threshold of 30 W/cm². The 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains were prepared by heating in a muffled furnace to a temperature of 900 °C for 15 mins.

5.3.3 OSL/TL Reader

A reader is needed for OSL/TL signal measurements as well as irradiation of dosimeters in the laboratory. For this work, a Risø TL/OSL reader (model TL/OSL-DA-20) was used. This reader is essentially composed of three components: light detection system (photomultiplier tube, PMT), luminescence stimulation system (thermal and optical), and irradiation sources (alpha and beta). It allows measurement of both TL and OSL. A schematic drawing is shown in in Figure 5.4. One advantage of the Risø reader is that it is an automated reader

with built-in radiation sources, thus allowing readouts to be performed without removing the dosimeters from the reader for external annealing or irradiations, thereby eliminating uncertainties due to handling and the annealing processes.

Irradiation system

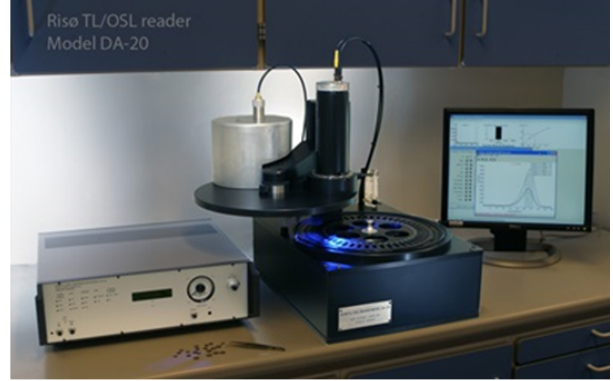
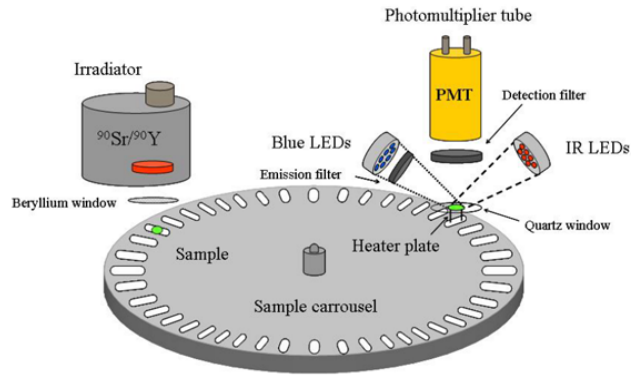
The irradiator system accommodates a ($^{90}\text{Sr}/^{90}\text{Y}$) beta source and a ^{241}Am alpha source. The beta source emits particles of maximum energy of about 2.27 MeV with a strength of about 1.48 GBq while the alpha source has a strength of about 10.7 MBq. The β -source of the Risø reader was calibrated against a 662 keV ^{137}Cs gamma source for the $\text{Al}_2\text{O}_3:\text{C}$ dosimeters used in this work (see detailed description of the experimental procedure used in Appendix A). The gamma equivalent dose rates at the measurement position in the reader are: (72.7 ± 1.5) mGy/s, (66.29 ± 1.85) mGy/s, and (68.76 ± 3.01) mGy/s for the Luxel[®] cutouts, 250–355 μm regular grains, and 250–355 μm alpha-desensitized grains respectively.

The beta source is mounted on a rotating, stainless steel wheel, that is actuated by air pressure and enclosed in a lead housing. It is rotated out of the housing to a position close to the sample during irradiation (open position) to and rotated back into the housing, when not irradiating (closed position) to significantly reduce leakage [164]. Time for irradiation is counted from the moment air pressure is applied, but it takes a small amount of time for the wheel to rotate fully out of the housing. This time called the off-set time or shutter time was calculated to be around 0.11s [164] and 0.24s [165] for their respective readers, suggesting that it might be different for every reader even when the source is mounted in the same way [165]. Notwithstanding, the off-set time is constant for all irradiations with the same reader and must be corrected for, especially for shorter irradiations. For longer irradiations, it may be considered negligible.

An unavoidable consequence of a reader system with a built-in source is that additional doses are unintentionally applied to dosimeters during readouts due to leakage radiation from the sources and cross-talk [165]. The leakage is defined as the radiation exposure to

dosimeters when the source is closed. A leakage dose rate of $0.08 \mu\text{Gy/s}$ (dosimeter material not mentioned) has been reported by Markey et al. [164] at the sample position directly under the source. A similar leakage dose rate of $0.06 \mu\text{Gy/s}$ was reported for $\text{Al}_2\text{O}_3\text{:C}$ chips by Kalchgruber et al. [165]. A relatively higher leakage dose rate of $0.69 \mu\text{Gy/s}$ at the sample position for $\text{Al}_2\text{O}_3\text{:C}$ chips was reported by Burbidge & Duller [166]. This higher dose rate is because they used the old Risø TL/OSL DA-10 reader for their measurement. Their result is however consistent with the leakage dose rate for all models prior to DA-15 [164, 165].

The irradiation cross-talk is defined as the radiation exposure to dosimeters in the vicinity but not directly under the irradiation source, when the source is open. This effect has been investigated by various authors for quartz and $\text{Al}_2\text{O}_3\text{:C}$ dosimeters and has been shown to be in the range of $0.006\text{--}0.25\%$ of the direct dose rate, depending on the specific reader and the irradiated sample [60, 165, 167–169]. As seen from Figure 5.4a, the reader comes with a carousel that can accommodate up to 48 samples. The distance between the centers of the sample positions is 1.7 cm, resulting in the cross-talk contributing dose to adjacent sample positions. To reduce this effect, dosimeters were placed on only every fourth position on the carousel, since the cross-talk has been shown to decrease with increasing distance from the source [60, 165]. The leakage dose rate is about 10^7 orders of magnitude less than the dose rate of direct irradiation in the reader, and was also shown to decrease with increasing distance from the source [165], so this effect was ignored.



(a)

(b)

Figure 5.4: (a) Schematic diagram and (b) picture of the Risø TL/OSL reader. Adapted from [170, Fig. 1.1]

Stimulation and Detection

The luminescence stimulation system consists of various light emitting diodes (LEDs): infrared (870 nm, 145 mW/cm²) [132], blue (470 nm, 80 mW/cm²) [132], and green (525 nm, 40 mW/cm²) [170]. These LEDs provide the stimulation light that enforces photoeviction of filled traps in the dosimeter. The sensitivity of the traps in our sample depends strongly on the wavelength of the stimulating light. Therefore, green stimulation was used for our OSL measurement to avoid the possibility of emptying the deep traps, which is possible with the energetic blue LEDs [171]. Additionally, for the OSL, the intensity of the emitted luminescence is many orders of magnitude smaller than the intensity of the stimulation light. In order to effectively discriminate between the two and also to prevent the stimulation light from reaching and destroying the PMT, appropriate filters were used.

The PMT (model EMI 9235QB; Electron Tubes Inc.) detects luminescence light and converts photons to electrons via its photo-cathode. The electrons are accelerated and amplified through a series of positively charged dynodes. The electrons arriving at the anode create current pulses. The number of pulses arriving during a pre-set time interval are added up and this number is then reported as "Signal" for the time-interval. The signal unit is "counts per channel" or, if the interval is equal to one second, "counts per second". To

separate stimulation light from luminescence, the UV transmitting and visible absorbing filter Hoya U-340, with bandwidth centered at 340 nm, was employed to filter the emission spectrum.

For TL measurement, there is no interference of stimulation light. The so-called blue filter pack consisting of a Schott BG-39 in combination with BG3 with a bandwidth centered around 390 nm [170] was used because it allows the passage of more signal compared to the U-340. All measurements were done with the insertion of either a 4 mm or 10 mm aperture, depending on the magnitude of signal expected. This was necessary to reduce the amount of light reaching the Photomultiplier tube (PMT), thus preventing signal saturation and potential damage of the PMT. Except, for low background radiation exposure where the signals are relatively weak and thus do not require the use of an aperture.

5.3.4 Attenuators

As mentioned in section 5.3.3, the dose for $\text{Al}_2\text{O}_3\text{:C}$ dosimeters is of the order of 100 mGy/s. However, environmental radiation is classified under low level background radiation and dose-rates are typically less than 100 $\mu\text{Gy/s}$ [166]. In routine environmental beta dose-rate assessment, dosimeters are buried for a period of few weeks to a year [172]. The cumulative absorbed dose measured even after a year has been reported to be comparable to the dose obtained after a 1s irradiation from the β -source of the reader [165]. Such short irradiation times are however not recommended since the rotating nature of the irradiation unit introduces a time off-set and thus adds uncertainty to the given dose [164].

Although it has been reported that $\text{Al}_2\text{O}_3\text{:C}$ shows a linear behavior from μGy to few Gy [140], it is not recommended to use larger irradiation times and to extrapolate the dose response to lower doses. Extrapolation is well known to lead to large errors in dose recovery, due to the relatively large impact of the uncertainty in the intercept fitting parameter. Therefore, a method of dose rate reduction for the built-in β source of the reader using attenuators was investigated. The result of this work has been published in the paper

Osunkwor & DeWitt [169]. A copy of this work is presented in Appendix B. The attenuators were used in this work, when smaller dose rate values were required for the measurements.

5.4 Developing an OSL measurement procedure for Al₂O₃:C Luxel[®] dosimeters

A comprehensive and systematic approach was taken to investigate some properties, as well as to quantify the response of our Luxel[®] dosimeter to ensure the accuracy of measured doses. The uniformity of the signals was quantified when different dosimeters are given the same dose. The effect of different bleaching modes on the response of the dosimeters and the overall precision of the dosimeter/reader combination was tested. The stability of the signal was measured, as the time between exposure and measurement increases, which is important because exposed dosimeters cannot be measured at the same time. Last but not the least, a dose response curve was obtained to allowing defining the relationship between exposure and measured signal.

5.4.1 Signal calculation

During measurement, the Luxel[®] dosimeters were stimulated for 500 s and the emitted signal was recorded using a UV filter. The stimulation time was chosen to ensure that the signals are reduced to background level at the end of the stimulation period as shown in Figure 5.5.

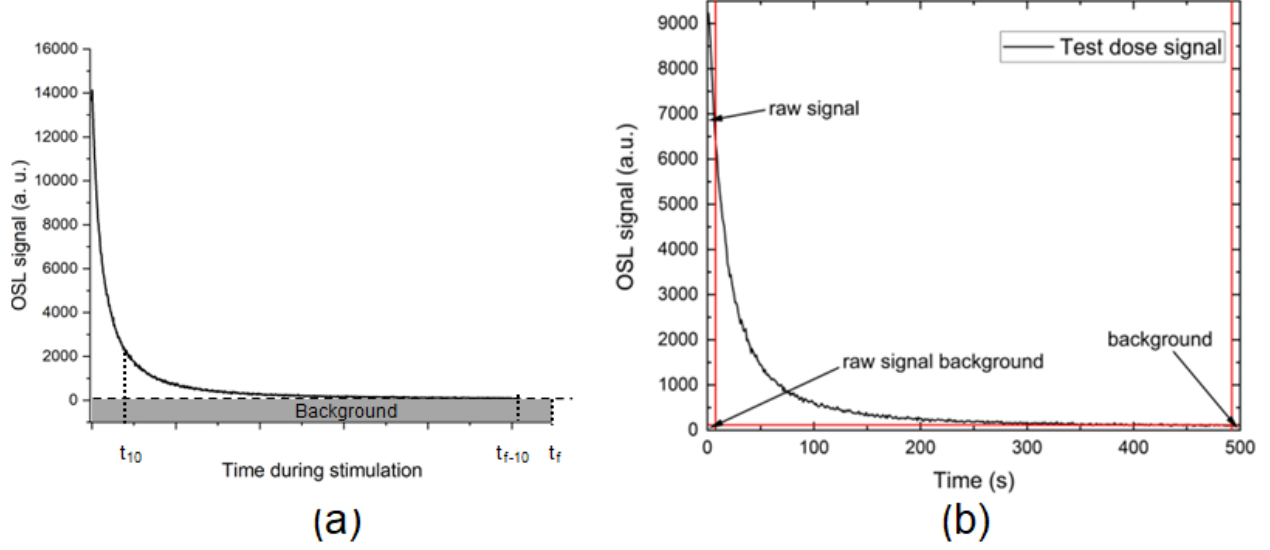


Figure 5.5: (a) OSL curve and details of signal calculation. The first dotted vertical line indicates the initial 10s during stimulation, the last two dotted lines on the right indicate the final 10s of the stimulation. The shaded area shows the level of background signal. (b) Sample OSL decay curve after irradiation with a test dose.

The signal for each time interval or "channel" C_i is recorded (compare section 5.3.3). After recording the OSL signal, the collected data was used to estimate the integrated OSL intensity for the first 10s of stimulation (I_{10} , Eq. 5.1), and the background of the signal was determined from the last 10s of stimulation (B , Eq. 5.2). The background was then subtracted from the OSL intensity for the first 10s (see Eq. 5.3). The background signal can be due to PMT noise, breakthrough LED light, and post-bleaching residual signals from dosimeter. It thus varies for each dosimeter, so that removing the background signal from the dosimeter ensures that results from different dosimeters are comparable.

The background-corrected signal S is calculated from the following equations:

$$I_{10} = \sum_{i=0}^{t_{10}} C_i \quad (5.1)$$

$$B = \sum_{i=t_f-10}^{t_f} C_i \quad (5.2)$$

$$S = I_{10} - B \quad (5.3)$$

The error in our signal can be calculated by using Poisson statistics so that the error in our signal is equal to the square root of the signal. The error in the background-corrected signal δS is thus expressed as:

$$\delta S = \sqrt{(\delta I_{10})^2 + (\delta B)^2} = \sqrt{I_{10} + B} \quad (5.4)$$

where $\delta I_{10} = \sqrt{I_{10}}$ and $\delta B = \sqrt{B}$.

5.4.2 Variation of OSL signal due to differences in dosimeter sensitivity

When different Luxel[®] cutouts are irradiated with the same dose, it does not necessarily result in the same signal. They have different “sensitivity”. Possible reasons for this are differences in dosimeter mass, differences in dosimeter sensitivity, and variations in reader’s sensitivity. When the dosimeters are buried and exposed to radiation, the resulting signal needs to be corrected for these differences in sensitivity by applying a fixed test dose. See Figure 5.6. The background-corrected signal obtained from "main" irradiation (in the reader or after burial in a sample) will be subsequently referred to as the OSL signal S , while the background-corrected signal from irradiation with a test dose (using the beta source of the Risø reader) is referred to as the test OSL signal S_T . For each exposed dosimeter, the ratio S/S_T is used to quantify the signal. The advantage of using the test dose is, it accounts for variations in parameters such as dosimeter sensitivity and dosimeter mass but also inter-measurement equipment sensitivity variation [141].

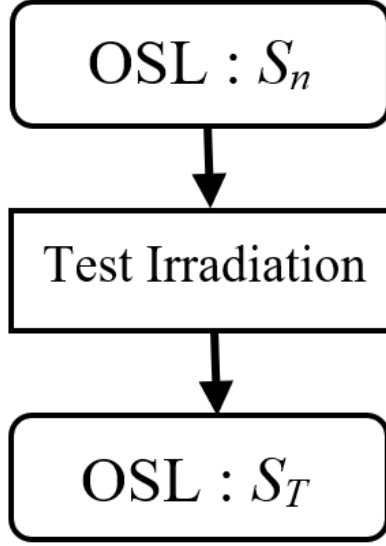


Figure 5.6: Summary of the general readout protocol after every irradiation of a dosimeter in this work. The subscript n represents the number of independent irradiations while subscript T represents the test dose.

The following procedure was used to quantify the variation in sensitivity of Al₂O₃:C Luxel[®] dosimeters:

- (i) bleach dosimeter using UV lamp (see section 5.3.2)
- (ii) β -irradiation of dosimeter for 8s
- (iii) OSL readout to obtain S (see section 5.4.1)
- (iv) β -irradiation of dosimeter for 4s (test-dose)
- (v) OSL readout to obtain S_T (see section 5.4.1)
- (vi) repeat steps (i)–(v) for 11 other dosimeters.

The OSL signal S and the test-dose normalized OSL signal S/S_T for the 12 dosimeters were normalized by their respective mean values for the twelve dosimeters. The results are presented in Figure 5.7. The result shows that the uncorrected signals are more susceptible to the dosimeter sensitivity variation compared to the test-dose normalized signal. For the

test-dose corrected signal, 100% of the points are better than 3% of the mean value while for the uncorrected signal, only 50% of the points are within 3% of the mean. Normalizing the OSL signal using a constant test-dose is one of the recommended ways to avoid the earlier mentioned variations and improve the precision of the measurement [173].

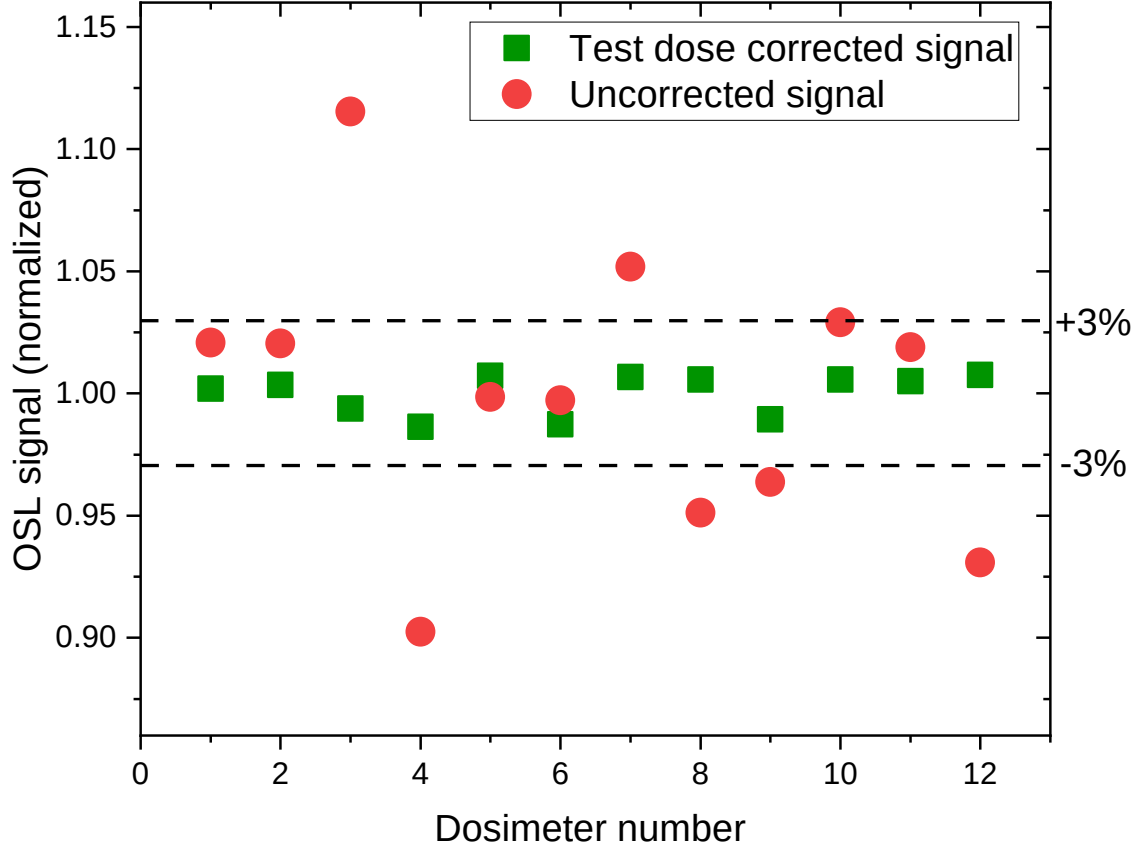


Figure 5.7: Variation of dosimeter sensitivity. Twelve $\text{Al}_2\text{O}_3\text{:C}$ Luxel[®] dosimeters were exposed to 8s β -irradiation. The red circles represent the uncorrected signal from the dosimeters while the green squares represent the 4s test-dose corrected signals of the dosimeters.

5.4.3 Effects of bleaching light on signal

Heating an OSL dosimeter is the most efficient method of zeroing the dosimeter after storage and/or transport and before use. However, due to the polyester covering of the $\text{Al}_2\text{O}_3\text{:C}$ Luxel[®] dosimeters, heating will result in the destruction of the dosimeter. Therefore, optical bleaching must be applied to zero each dosimeter before use. It has been suggested that the wavelength and power of the bleaching light used might affect the sensitivity of the dosimeter

by influencing the distribution of electron/holes in certain traps [174, 175]. For example, Umisedo et al. [171] suggested that depending on the energy of the bleaching light, there may be partial emptying of deep electron traps which can influence the competition dynamics between trapping centers during irradiation and stimulation, ultimately affecting sensitivity.

An experiment was devised to investigate the impact of the bleaching light source on dosimeter sensitivity. Impact was quantified via the signal variance. Two bleaching light options were tested for their effects on the OSL signal. The first is filtered light from the UV lamp, described in section 5.3.2 and the second are the green LEDs of the Risø reader. Dosimeters were bleached with the UV lamp for ~ 8 hours or with the green LEDs for ~ 8 min. Subsequently the dosimeters were irradiated with a constant dose and the resulting corrected signal was measured. The measurement procedure is summarized as follows:

- (i) bleach dosimeter with UV lamp for ~ 8 hrs or green LEDs for ~ 8 min
- (ii) β -irradiation of dosimeter for 8s (~ 0.58 Gy)
- (iii) OSL readout to obtain S
- (iv) irradiate dosimeter with a test-dose of 4s (~ 0.29 Gy)
- (v) OSL readout to obtain S_T
- (vi) obtain S/S_T
- (vii) repeat steps (i)–(vi) for 11 other dosimeters.

The signals were normalized to the mean value and the result is presented in Figure 5.8. There is no clear difference in the variance of post-UV and post-Green LED signals. The relative standard deviation for the post-UV signal is 0.83% compared to 0.79% for the post-Green LED signal. Therefore, we can conclude that both the UV lamp or the Green LEDs can be interchangeably used for bleaching our Luxel[®] dosimeters without introducing any additional uncertainty.

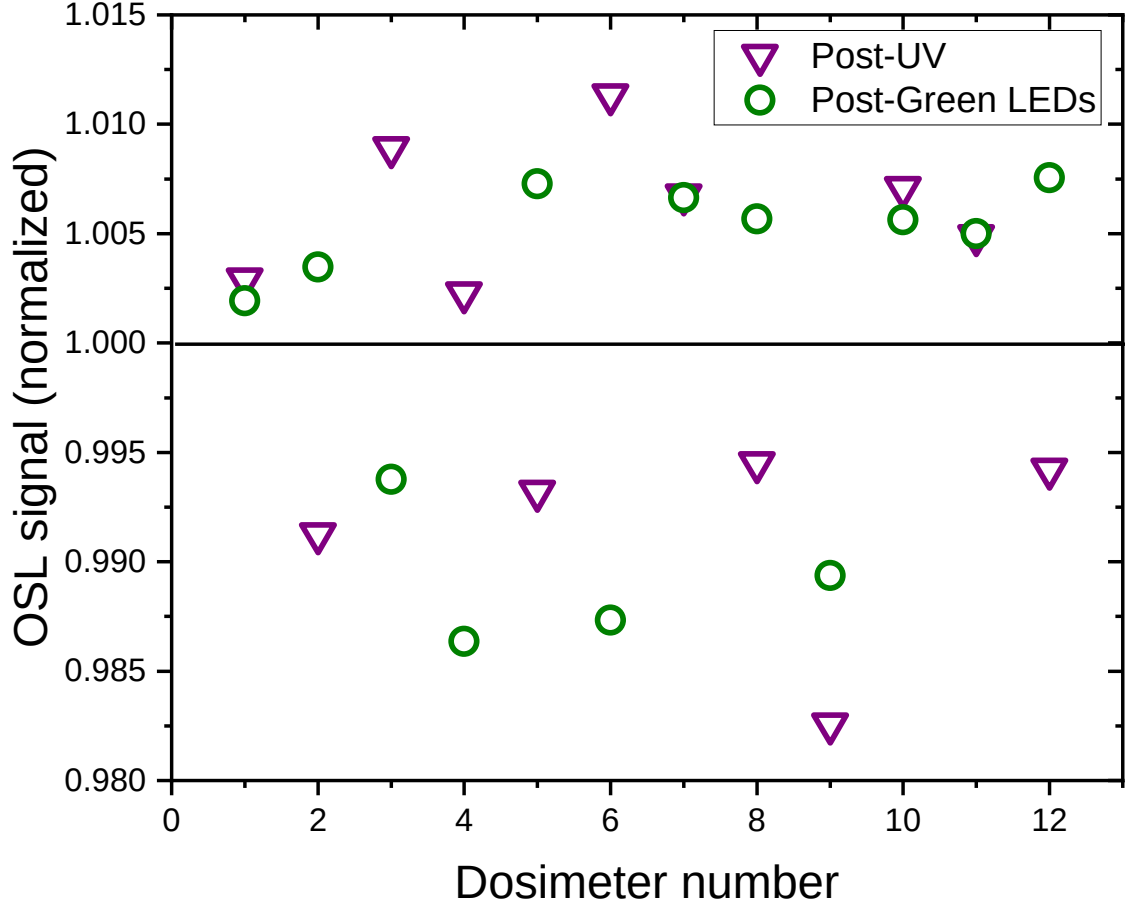


Figure 5.8: Effects of bleaching mode on the dosimeter signal

5.4.4 Effect of the dosimeter's position on the carousel

Measurements using the Risø reader involve placing each dosimeter on a cup that is in turn placed on a position in the carousel. The reader used in this work has 48 carousel positions. However, only few of these positions can be used at a time. Sufficient spacing between the dosimeters during test-dose irradiation is required in order to minimize radiation cross-talk [165] (see also section 5.3.3). The cross-talk in the reader has been reported to decrease symmetrically with increasing distance from an irradiated position [165]. When multiple dosimeters are placed on different positions on the carousel with sufficient spacing (see Figure 5.9a), a series of measurement/test-dose irradiation/measurement cycles will be required to complete the measurement of all the dosimeters. A test was designed to check

whether the cumulative cross-talk over all the measurement/test-dose irradiation/measurement cycles will bias some positions compared to others on the carousel. In other words, does the precise location of the dosimeter on the carousel affect its response ? The measurement procedure for this test is summarized as follows:

- (i) arrange 10 dosimeters on 10 different positions on the carousel (see Figure 5.9a)
- (ii) bleach dosimeter with Green LEDs for 500s
- (iii) β -irradiation of dosimeter for 15s (~ 1.1 Gy)
- (iv) OSL readout to obtain S
- (v) irradiate each dosimeter with a test-dose of 10s (~ 0.73 Gy)
- (vi) OSL readout to obtain S_T
- (vii) obtain S/S_T
- (viii) repeat steps (i)–(vii) for 2 other sets of 10 dosimeters.

The S/S_T values for the 10 dosimeters in each set were normalized by the S/S_T value for position 1 for the respective set. The result is presented in Figure 5.9b. The result does not show any correlation between the response of the dosimeters in the 3 sets and their respective positions on the carousel.

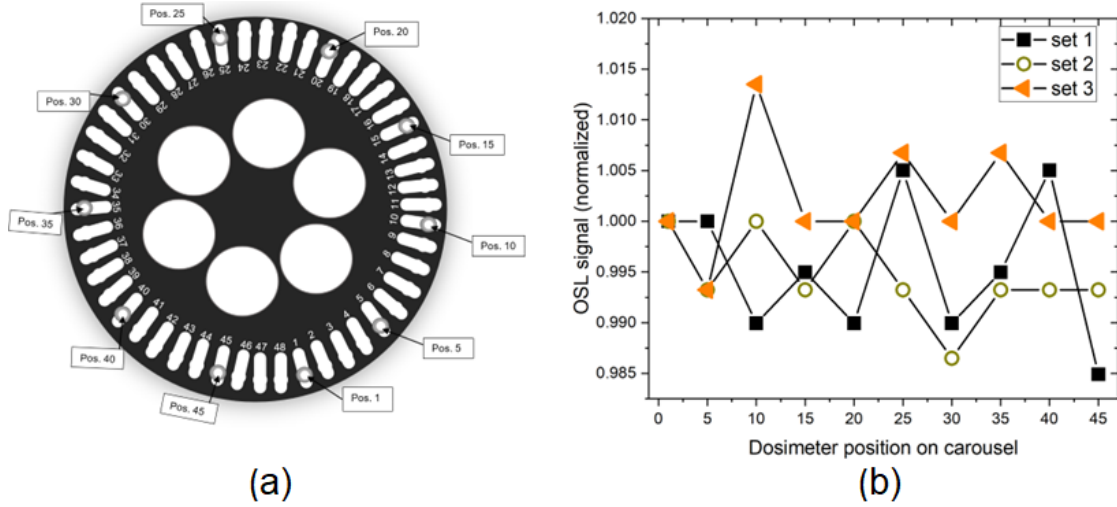


Figure 5.9: (a) Dosimeter measurement positions utilized and (b) dosimeter response vs. dosimeter position on the carousel.

5.4.5 OSL reproducibility

In all previous experiments, samples were irradiated and measured in the reader. However, in the dosimeter applications intended for this dissertation, exposure of dosimeters occurs externally via natural radiation from a mineral sample. The measurement is carried out subsequently with the reader by measuring the test-dose corrected signal from the dosimeters. It is common knowledge that the precision of an OSL dosimetry system can be affected by fluctuations in instrument performance, ambient conditions, and the stochastic nature of the radiation field. In order to evaluate the precision of the measurements made using the reader and the dosimeters, a reproducibility test for the readout of the OSL signals was performed and the associated experimental and relative standard deviations were estimated. A reproducibility test was also performed to evaluate the uniformity of the dosimeters since the equivalent dose to every dosimeter will be calculated from a single dose response curve.

This reproducibility investigation was particularly designed to be representative of experiments with external irradiation. Since no source for external radiation was available, all dosimeters were irradiated in the reader. However, to mimic external exposure, they were removed after irradiation, stored for some period, and then placed into the reader again for

measurement. Three measurement **scenarios** were considered to study these conditions:

1. Scenario 1 (Reference scenario): dosimeter was irradiated with a 15s β -dose in the reader and stays in the reader throughout the irradiation/measurement cycle, reader is not opened until measurement is completed. Measurement is done using a 10 mm aperture.
2. Scenario 2: dosimeter was irradiated with a 15s β -dose, removed from the reader following irradiation, stored for 5 min and re-placed in the reader for measurement. Measurement is done using a 4 mm aperture to simulate protection of the PMT from "high signal" expected from the "high dose". $\text{Al}_2\text{O}_3\text{:C}$ dosimeters are very bright; a signal equivalent to a test-dose of few seconds can easily saturate the PMT without an aperture [169].
3. Scenario 3: dosimeter was irradiated with a 15s β -dose but with an attenuator placed on top of the dosimeter to reduce the dose rate by a factor of 3.61 (see Osunkwor & DeWitt [169] for detailed description). The dosimeter is subsequently removed from the reader following irradiation, stored for 5 min and re-placed in the reader for measurement. Measurement of the main dose was done without an aperture because of its low signal intensity. A 10 mm aperture was later inserted before measuring the test-dose signal.

Each **scenario** was investigated by computing S/S_T values for 40 dosimeters. The results for the three scenarios are shown in Figure 5.10.

The relative standard deviations (ratio of standard deviation over the mean value) are 0.6%, 3.3%, and 3.4% for scenario 1–3 respectively, providing an indication of the uncertainty associated with the measurements. The relative standard deviation of 0.6% for the reference scenario 1 is in agreement with the 0.7% and 0.98% reported by Yukihiro et al. [141] and Vargas [176] respectively. The value is also in good agreement with the relative standard deviation observed for the bleaching experiments in section 5.4.3. The additional uncertainties

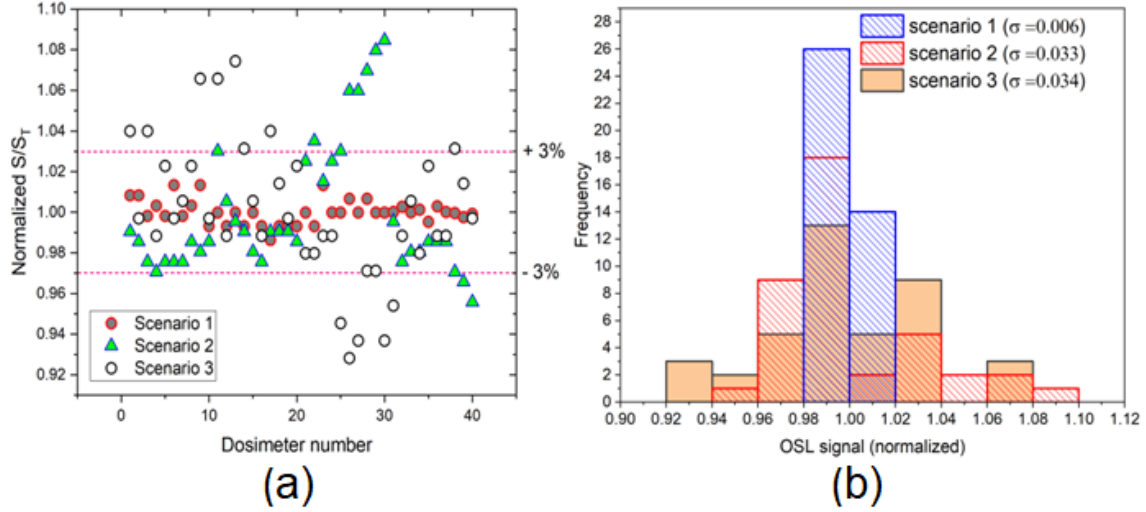


Figure 5.10: (a) Reproducibility of the test-dose corrected OSL signal for 40 dosimeters based on each scenario considered in section 5.4.5. The overall mean value and the $\pm 3\%$ lines are also depicted. (b) Histogram of the test-dose corrected OSL signal after 15s of exposure to β radiation for the three scenarios. Each value is normalized to the mean of its scenario distribution. The standard deviation σ for each scenario was obtained by the Gaussian fit of the distribution of each scenario.

seen in scenarios 2 and 3 are suspected to be partly due to handling between irradiation and measurement and partly due to the time delay between irradiation and measurement. A time-dependent increase in the UV luminescence of $\text{Al}_2\text{O}_3\text{:C}$ dosimeters has been observed by Yukihiro et al. [177]. The authors reported about 5% increase in the UV signal after 30 min storage between irradiation and measurement. However, from the publication, it is difficult to assess how much increase is expected for 5 min. Another possibility is signal fading, i.e., a loss of signal during storage. This will be investigated in the next section 5.4.6.

The results from the three scenarios indicate that removal of the dosimeter and/or opening the reader introduces some variability. A similar variation has been reported by Vargas [176]. She reported a 9% relative standard deviation for a reproducibility test on a single dosimeter that underwent twelve 1.26 Gy irradiation/measurement cycles when the reader was opened, and the dosimeter was exposed to room light from a weak red bulb between irradiation and measurement. Given that $\text{Al}_2\text{O}_3\text{:C}$ dosimeters also exhibit supralinearity with increase in dose history, it is quite possible that some of the variation in her result was due to a supralinear effect.

However, it is also important to note that the reported reproducibility in Figure 5.10 is the relative standard deviation and not the relative standard error. The relative standard errors, which are a more standardized way of reporting uncertainties, are 0.09%, 0.51%, and 0.54% for scenarios 1–3 respectively for our measurement. These numbers show that our signals are highly reproducible, even when the dosimeters are removed from the reader between irradiation and measurement.

5.4.6 OSL signal fading

In routine use of dosimeters in environmental dose monitoring, irradiation conditions, such as burial in the field, might make it impossible to measure the signal immediately after exposure to radiation. This implies that the duration between exposure and measurement might vary between measurements. The OSL signal of $\text{Al}_2\text{O}_3\text{:C}$ has been found to decrease with storage time for some samples. This phenomenon, called signal fading, has been reported in the literature. [140, 178, 179]. Therefore, it was imperative to characterize the fading properties of our dosimeters over different duration between exposure and measurement. The procedure for this investigation is summarized as follows:

- (i) bleach and irradiate 27 dosimeters with 15s β -irradiation (~ 1.1 Gy)
- (ii) readout 3/27 of the dosimeters immediately after exposure
- (iii) store the remaining dosimeters in a dark room
- (iv) perform readout of 3 dosimeters at a time for different post-exposure durations

Note: Every readout includes the 15s test dose β -irradiation (~ 1.1 Gy) to obtain the test-dose corrected signal.

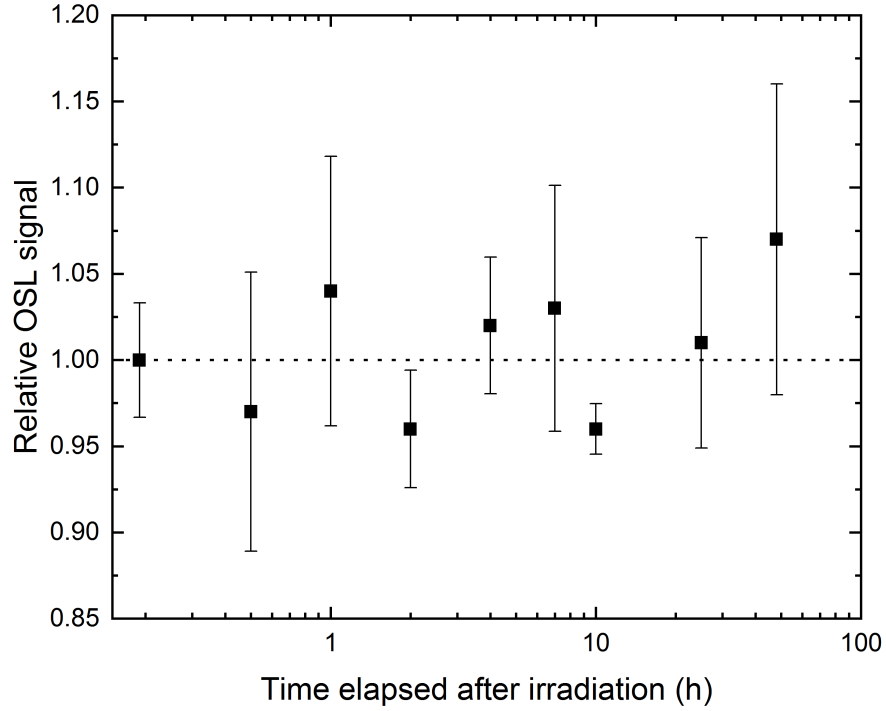


Figure 5.11: Relative OSL signal as a function of elapsed time since irradiation for the Luxel[®] dosimeter exposed to beta irradiation using the Risø reader. Each data point corresponds to the average of three dosimeters and the error bars are the relative standard deviations for the three dosimeters. The data was normalized to the first point measured directly after irradiation

The result for this test is reported in Figure 5.11. As shown in the plot, the signal does not seem to be increasing nor decreasing. The result seen here agrees more closely with the reports on fading of Luxel[®] dosimeters by Yukihiro et al. [173] and Vargas [176]. These authors mentioned that on average there was no significant fading for the dosimeter. Vargas [176] in addition, called into question the appropriateness of the term "fading", since the signal intensity is not decreasing with time but instead increasing at a rate that slows down with time. The increase in signal with time might be due to a time-dependent increase in the UV luminescence reported by Yukihiro et al. [177], which has been mentioned above. The high variance as seen from the error bars for the three dosimeters used for each dose point is suspected to be related to opening and closing the reader as part of the reading protocol (as observed from the reproducibility data in Figure 5.10). As far as measurement

in this work is concern, this result shows that once an externally irradiated dosimeter is retrieved, measurement can be performed within 24 to 48 hours without serious concern for signal fading.

5.4.7 Dose response and Dose Recovery

After every signal measurement, the signal has to be converted into an equivalent dose. For this purpose, the relationship between signal and dose must be established. In other words, the “dose response” of the dosimeter must be determined. In routine environmental dosimetry, the dosimeters are irradiated during burial and the test-dose corrected signal is then compared to an established dose response curve to determine the absorbed dose. The dose response and dose recovery of Luxel[®] dosimeters were investigated using the following protocol:

- (i) bleach dosimeter with UV lamp for ~ 8 hrs
- (ii) irradiate dosimeter with β dose #1
- (iii) remove from reader, store for 5 min
- (iv) OSL readout to obtain S
- (v) irradiate dosimeter with a test-dose of (~ 14.5 cGy)
- (vi) OSL readout to obtain S_T , then compute S/S_T
- (vii) repeat steps (i)–(vi) for β doses #2–6.
- (viii) repeat steps (i)–(vii) for 2 more dosimeters.

Step (iii), removing and storing the dosimeter from the reader after irradiation, was intended to make the dose response more consistent with the externally irradiated dosimeters for which it is intended. Dose values # 1-6 were 1s (0.07 Gy), 5s (0.36 Gy), 10s (0.72 Gy), 25s (1.81 Gy), 50s (3.63 Gy), 100s (7.26 Gy) respectively. Two dose response curves were

prepared with the given data. They are shown in Figure 5.12. The dose response was first fitted using a linear line with non-zero-intercept ($S = aD + b$; red line) with fit parameters $a = (6.58 \pm 0.04) \text{ Gy}^{-1}$ and $b = (0.31 \pm 0.12)$. A second linear fit with zero-intercept ($S = aD$; blue dotted line) with fit parameter $a = (6.640 \pm 0.001) \text{ Gy}^{-1}$ was also fitted to the dose response to remove the high variance associated with the intercept of the first fit, which may introduce additional uncertainties when low doses are recovered, i.e. points close to the intercept.

In addition, five dosimeters were irradiated with five additional β doses (1 dose per dosimeter) and the test-dose corrected signals were measured (steps (i)-(vi)). These dosimeters were used for the so-called dose recovery test. The signals from these additional dosimeters were compared to the dose response to obtain measured (aka. “recovered”) doses. These recovered dose values were compared to the known irradiation doses. The results are given in Table 5.1.

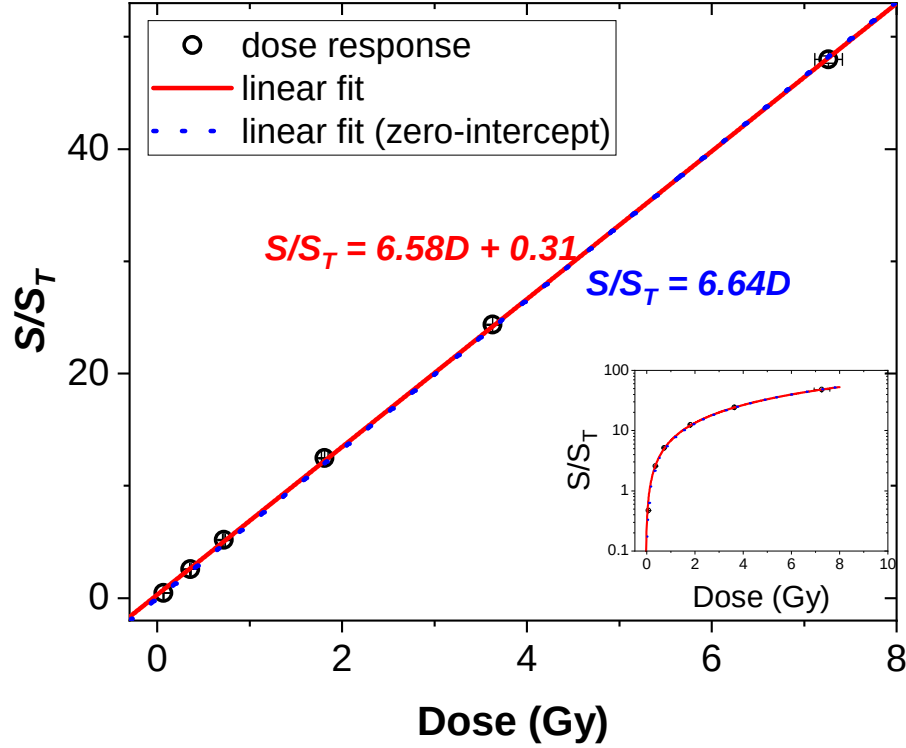


Figure 5.12: Dose response based on the test-dose corrected OSL signal S/S_T . The red line is a linear fit with the equation shown in the plot. The blue dotted line is a zero-intercept linear fit with equation also shown in the plot. The inset shows the same data and fits with the y-axis on a log-scale to emphasize the high degree of linearity even for low-dose points. Data points are mean values of three dosimeters. Error bars for S/S_T values indicate the combined uncertainty from counting statistics and uncertainty of the weighted mean, while the error bars for the dose indicate combined uncertainty from calibration of the source and the offset-time effect (0.08 ± 0.04)s.

Table 5.1: Dose recovery results for single dosimeters irradiated with known doses using the dose-response curves in Figure 5.12.

Known Dose (Gy)	S/S_T values	Linear fit ($a = 6.58, b = 0.3$)		Linear fit ($a = 6.64, b = 0$)	
		Recovered Dose (Gy)	Recovery Ratio	Recovered Dose (Gy)	Recovery Ratio
0.067 ± 0.002	0.477 ± 0.002	0.026 ± 0.001	0.388	0.072 ± 0.002	1.075
0.140 ± 0.003	0.996 ± 0.003	0.105 ± 0.005	0.750	0.150 ± 0.002	1.071
0.212 ± 0.005	1.545 ± 0.005	0.188 ± 0.010	0.887	0.233 ± 0.002	1.099
1.812 ± 0.037	12.460 ± 0.040	1.846 ± 0.081	1.019	1.876 ± 0.012	1.035
3.629 ± 0.075	24.290 ± 0.068	3.643 ± 0.068	1.004	3.658 ± 0.021	1.008

The dose response of Luxel[®] dosimeters based on the test-dose corrected signal (S/S_T)

is linear in the dose range tested in this work (see Figure 5.12). Akselrod et al. [161] and Yukihiro et al. [141], on the other hand, reported linearity in this dose range only for the uncorrected OSL signals, not for the test-dose corrected signals. It is quite interesting that they found linearity for the uncorrected OSL signals, given that the uncorrected OSL signal is expected to be more susceptible to sensitivity variation in the $\text{Al}_2\text{O}_3\text{:C}$ dosimeters as reported by Yukihiro et al [141] and Jursinic [140].

The recovered doses listed in Table 5.1 are compared to the known dose using the dose recovery ratio. The non-zero-intercept dose response curve seems to underestimate the low doses with deviations of up to 61% for the lowest dose recovered. However, as the dose increases, the recovery ratios become closer to 1, showing deviation of only 0.4% for the highest dose recovered. The zero-intercept dose response curve seems to perform better on average for all doses, although, it slightly overestimates the high doses recovered with a maximum deviation of 3.5%. In general, the result shows that on average, doses can be recovered with an uncertainty of about 2–10%. Based on these results, the recommendation is that the non-zero-intercept dose response is used for recovery of doses greater than or equal 0.3 Gy or for S/S_T values greater than or equal to 2. While the zero-intercept dose response is used for recovery of doses less than 0.3 Gy or for S/S_T values less than 2.

5.5 TL characteristics of $\text{Al}_2\text{O}_3\text{:C}$ grains

5.5.1 TL readout procedure

The $\text{Al}_2\text{O}_3\text{:C}$ grains were measured using TL by heating to 450 °C at a heating rate of 2 °C/s. The TL signal was collected using a blue filter combination (Schott BG-39 and BG3). The blue emission of $\text{Al}_2\text{O}_3\text{:C}$ is considerably brighter than the UV emission and with TL there is no stimulation light that needs to be suppressed. Similar to OSL, the number of PMT current pulses is recorded for a preset time interval, corresponding to a temperature interval, and displayed as signal or intensity. A constant test dose was also administered to

the grain(s) after measurement of the main signal to correct for possible changes in sensitivity, as discussed in section 5.4.2.

To retrieve information from the TL glow-curve, three methods were employed: (i) the use of the TL signal maximum intensity to represent the signal; (ii) the use of the integrated signal in the temperature range 100°C–300°C (i.e., TL area under the curve between 100 °C and 300°C); (iii) the use of the total integrated signal minus background (i.e., TL area minus background under the curve between 100 °C and 450°C). The signal integration was started at 100 °C to avoid the time-dependent, non-dosimetric low temperature (~ 54 °C) TL glow peak that has been reported for $\text{Al}_2\text{O}_3\text{:C}$ dosimeters [180]. The integration was extended to 450 °C for the third method to account for any dose-dependent changes in the shape and position of the TL dosimetric peak, as reported by McKeever et al. [181] and Agersnap et al. [182]. The three methods are illustrated in Figure 5.13.

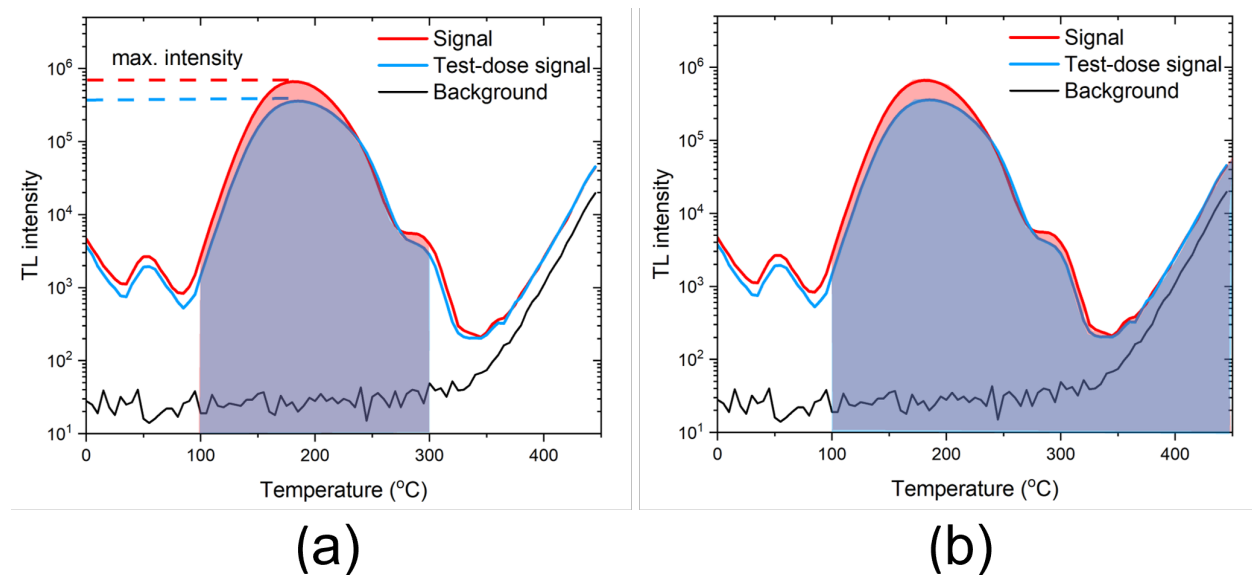


Figure 5.13: Schematic representation of the three methods used to evaluate the TL signal. (a) Method 1 based on maximum TL intensities and method 2 based on the 100°C–300°C TL area. (b) Method 3 based on the 100°C–450°C TL area. The signal and test-dose signal represent 0.66 Gy and 0.33 Gy β -doses. The dosimeter is an aggregate of 8–10 regular $\text{Al}_2\text{O}_3\text{:C}$ grains.

The signal from each method is divided by the corresponding test-dose signal to obtain a test dose-corrected signal. The signal can be from exposure in a natural sediment or exposure

from beta particles of the Risø reader. The test dose is given by exposing the grains to a constant dose using the β -source of the Risø reader.

5.5.2 Comparison of TL response of the regular and alpha-desensitized $\text{Al}_2\text{O}_3\text{:C}$ grains

The response of α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ grains to α - and β - radiation compared to the response of the regular (non-desensitized) $\text{Al}_2\text{O}_3\text{:C}$ grains was investigated using the following protocol:

- (i) divide the grains into two batches
- (ii) anneal the first batch at 1200 °C for 30 min in an external oven to α -desensitize them
- (iii) anneal the second batch at 900 °C for 30 min in an external oven to reset
- (iv) irradiate a set of aggregate grains (8-10 grains on a cup) with dose equivalent to 10s β radiation
- (v) 2 TL readouts to measure signal, then background
- (vi) irradiate the same set of aggregate grains with a dose equivalent to 5s β radiation
- (vii) 2 TL readouts to measure signal, then background
- (viii) irradiate the same set of aggregate grains with a dose equivalent to 1000s α radiation
- (ix) 2 TL readouts to measure signal, then background
- (x) irradiate the same set of aggregate grains with a dose equivalent to 500s α radiation
- (xi) 2 TL readouts to measure signal, then background
- (xii) repeat steps (iv)–(xi) for equal sets of aggregate grains from each batch

The 10s β and 1000s α radiation were used to study the response of the dosimeters to "main" exposure while the 5s β and 500s α radiation were used to study the dosimeter's response to test-doses or lower doses. The result of this investigation was prepared by using the protocol described in section 5.5.1. The ratio of α to β TL signal for the regular grains was compared to that of the α -desensitized grains. The result is presented in Figure 5.14.

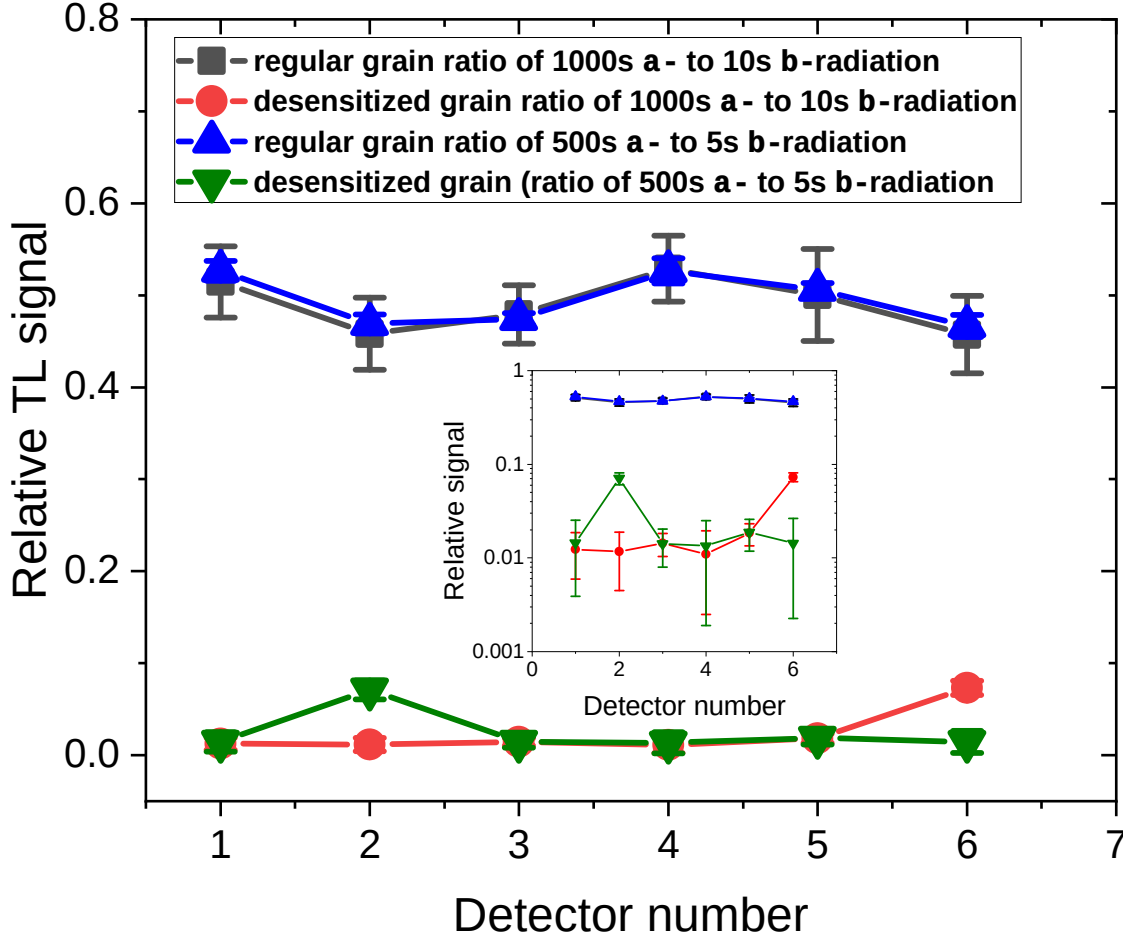


Figure 5.14: Comparing TL response of regular and α -desensitized 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains. The inset shows the same data set but with the y-axis on a log-scale to emphasize the error bars on the 500s α -to-5s β -signal ratios, which can not be seen from the main plot. Each data point is an average of the TL signal ratios of the three TL signal analysis methods discussed in 5.5.1. The Error bars represent the standard deviation.

The result in Figure 5.14 shows that the regular grains have a considerably higher ratio of α - to β - radiation induced TL signals than the desensitized grains, where the ratio is close to zero for the two dose regimes tested i.e., 1000s α /10s β and 500s α /5s β . This confirms

that the annealing of $\text{Al}_2\text{O}_3\text{:C}$ crystals at 1200 °C for 30 min is very effective at destroying oxygen vacancies in the superficial layer judging from insensitivity of the desensitized grains to α - radiation. It is quite interesting to note that the desensitized grains do not have zero sensitivity to α -radiation, but rather negligible sensitivity to α -radiation as can be seen from the inset. From the inset, we can also see that the level of desensitization is not uniform among all grains. This is probably due to the probabilistic nature of the heating process defined by the Boltzmann equation of heat transfer and the temperature gradient that might exist across the ceramic container used to pack the grains. A temperature gradient can occur since multiple grains were packed into a ceramic container during heating. It is also possible that grains further to the bottom of the container were not exposed to enough oxygen to deplete all oxygen vacancies. This might not be the case if a single grain is heated at a time, but of course that will be extremely time consuming.

5.5.3 TL reproducibility of single grains

In this work, the dose distributions in well-defined samples were determined by burying $\text{Al}_2\text{O}_3\text{:C}$ grains in the samples. The dosimeters were left in the samples for a duration that allows for sufficient exposure of the grains to the natural radioactivity from the samples. Each grain was then retrieved and measured using TL. The equivalent doses for all the grains were calculated from their respective TL signals using the same dose response curve. A reproducibility test was thus important to evaluate the uniformity of the single dosimeter grains. The TL signal reproducibility of the regular and α -desensitized 250–355 μm single $\text{Al}_2\text{O}_3\text{:C}$ grains was investigated using the following procedure:

- (i) anneal the grain at 900 °C for 15 min in an external oven to reset
- (ii) irradiate grain with a dose equivalent to 10s β radiation
- (iii) 2 TL readouts to measure signal, then background
- (iv) irradiate grain with a test-dose equivalent to 5s β radiation

(v) 2 TL readouts to measure signal, then background

(vi) repeat steps (i)–(v) for 11 more regular and 12 α -desensitized grains

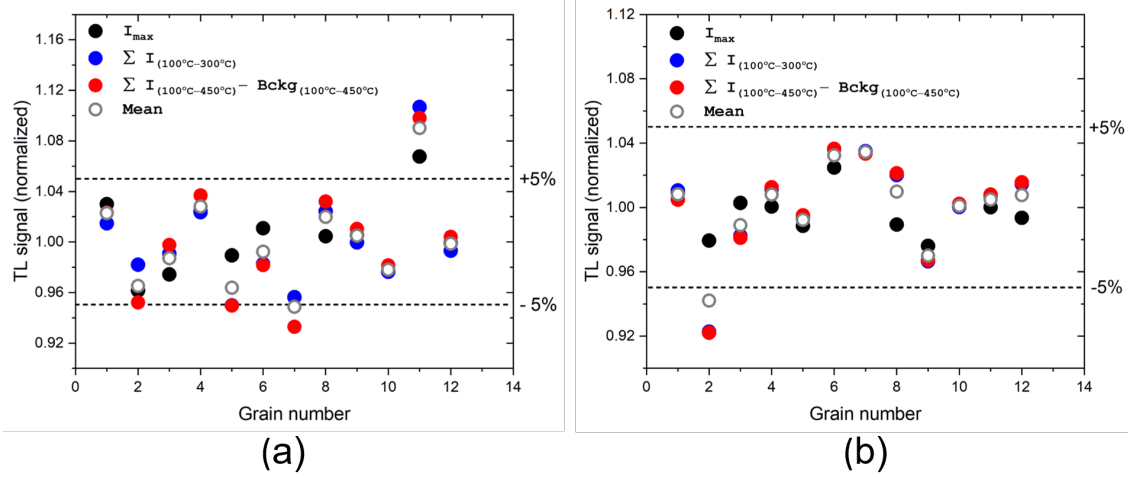


Figure 5.15: Test-dose corrected TL signal reproducibility of $\text{Al}_2\text{O}_3\text{:C}$ grains. Each grain was exposed to 10s β radiation followed by measurement. The reproducibility is presented based on the three TL data analysis methods and their overall mean. (a) Reproducibility of α -desensitized grains. (b) Reproducibility of regular grains. The data points in each method were normalized to the respective mean of each method. The dashed lines represent $\pm 5\%$ variation from the respective mean.

Table 5.2: TL signal reproducibility result for $\text{Al}_2\text{O}_3\text{:C}$ grains. All TL signals were test-dose corrected. Signals were calculated using the three different methods mentioned above. The reported values are the mean and standard deviations for the 12 grains. Listed are also the mean and standard deviation for all the signal calculation methods.

Protocol	α -desensitized grain	regular grain
Peak	1.79 ± 0.05	1.81 ± 0.03
TL area, $100^\circ\text{C}-300^\circ\text{C}$	1.65 ± 0.06	1.72 ± 0.05
TL area, $100^\circ\text{C}-450^\circ\text{C} - \text{Bckg}$	1.63 ± 0.07	1.72 ± 0.05
Mean value of the 3 protocols	1.69 ± 0.06	1.75 ± 0.04

The result of the reproducibility test is presented in Figure 5.15 and Table 5.2. The TL signals reported are the test-dose corrected signals i.e., they were corrected using the signal

from the 5s β test-dose administered to each dosimeter. The signals were computed using the three methods of signal analysis described in section 5.5.1. Figure 5.15 shows the TL signals normalized by the mean TL signal for each method. While Table 5.2 shows the absolute values of the TL signals for each method.

A higher variance was seen for the α -desensitized grains where irrespective of the TL signal analysis method, at least 1 dosimeter showed more than 5% deviation from the mean value (see Figure 5.15a). For regular grains, almost all the data are well within 5% of the mean. On average, the regular grains also show a standard deviation of 2.3% compared to 3.6% for desensitized grains (see Table 5.2). In all, the grains show good TL signal reproducibility with a maximum standard deviation of 7%. This result is comparable to the 3% reported by Kalchgruber et al. [183] for 200–300 μm $\text{Al}_2\text{O}_3\text{:C}$ grains and better than the 8.3% reported by Pogue (2021) [184] for $\text{Al}_2\text{O}_3\text{:C}$ chips. The result also shows that none of the methods performed significantly better than the others. So, for the purpose of this dissertation, the mean of the three methods will be utilized for TL signal calculations.

5.5.4 Dose Response

Two standard dose responses were built using single α -desensitized grains and single regular grains. The dose response was used to determine the equivalent dose of each grain exposed to natural radiation in our sample. The dose response of $\text{Al}_2\text{O}_3\text{:C}$ grains was tested using the following procedure:

- (i) anneal the grain at 900 °C for 15 min in an external furnace to reset
- (ii) irradiate grain with β dose #1.
- (iii) 2 TL readouts to measure signal, then background
- (iv) irradiate grains with a test-dose equivalent to 5s β radiation
- (v) 2 TL readouts to measure signal, then background

(vi) repeat steps (i)–(v) for 2 more regular grains and 3 α -desensitized grains

(vii) repeat steps (i)–(vi) for β doses #2–25

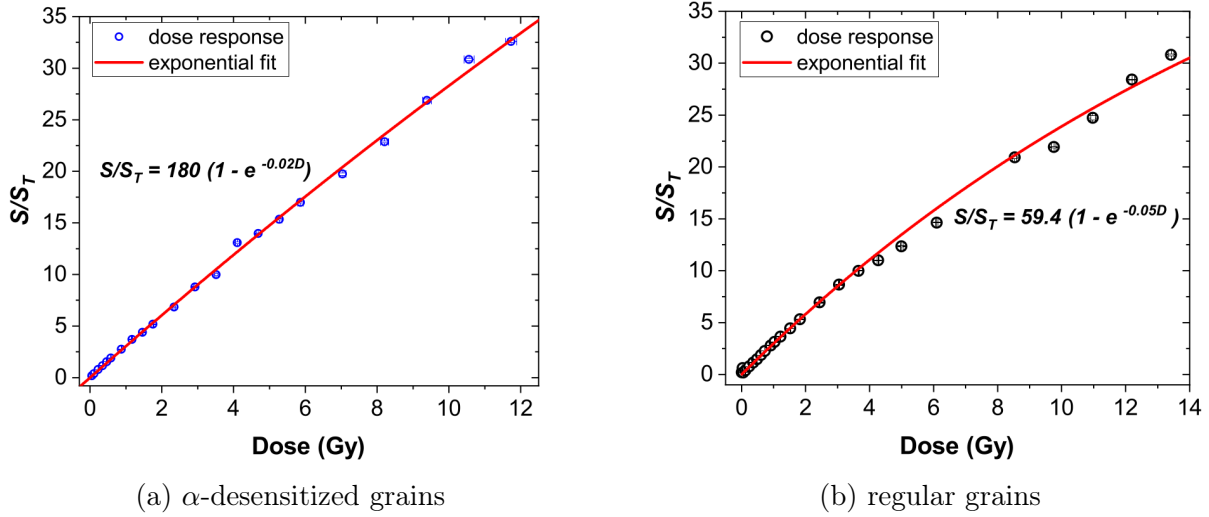


Figure 5.16: TL dose response (growth curve) for (a) α -desensitized grains and (b) regular grains. The growth curves were fitted with saturating exponentials. The fit parameter D_o , which characterizes the shape of the curves is 50 Gy and 20 Gy for α -desensitized and regular grains respectively. Data points are mean values of three dosimeters each. Error bars for S/S_T values indicate the uncertainty of the weighted mean, while the error bars for the dose indicate the combined uncertainty from calibration of the source and the offset-time effect of (0.08 ± 0.04) s.

The dose responses are presented in Figure 5.16. The growth curves are linear at lower doses but signal saturation sets in for higher doses. The growth curves were thus fitted by a saturating exponential of the form:

$$S/S_T = S/S_{T\infty} (1 - e^{-D/D_o}), \quad (5.5)$$

where $S/S_{T\infty}$ is the maximal attainable value for the test-dose corrected signal and D_o is a parameter which is related to the shape of the curve. It defines the dose that will result in 64% of the maximum test-dose corrected signal. The D_o values of the growth curves in Figure 5.16 imply that the growth curves are still growing albeit at a decreasing rate. However, we are not interested in higher doses since the dose range of interest in this work is from 0 to

10 Gy. It must be stressed that these curves are only presented to show the general dose response behavior of our dosimeters and were not used as is, for the purpose of dose recovery. In this work, dose recovery for dosimeters irradiated externally or buried in sediments was done by linear or exponential interpolation between test-dose corrected signals bracketing the test-dose corrected signal of the dose that needs to be recovered. In other words, the dose response curves in Figure 5.16 were broken into multiple smaller linear or exponential dose response curves with shorter dose ranges. Examples of these dose response curves are presented in Figures 5.17 and 5.18.

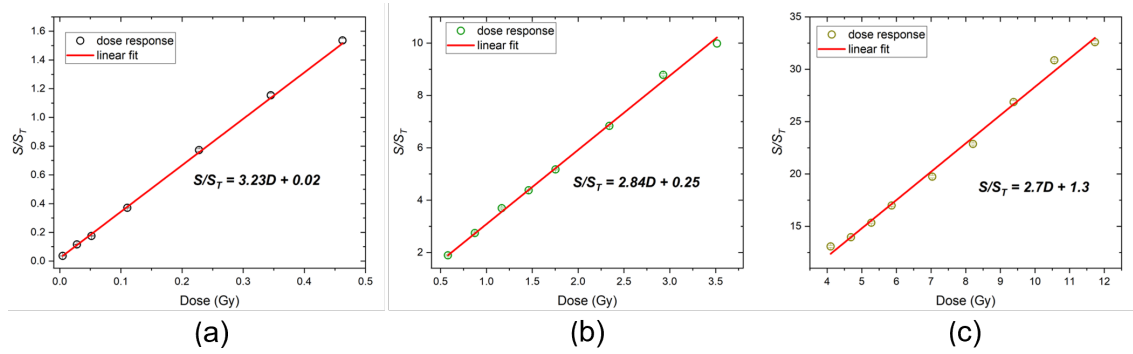


Figure 5.17: TL dose responses for α -desensitized grains, determined by breaking the dose response in Figure 5.16a into 3 smaller fractions. (a) Dose response from 0–0.5 Gy. (b) Dose response from >0.5–3.5 Gy. (c) Dose response from >3.5–12 Gy. The fit for each dose response is represented by a red line. The equations for the fits are also shown.

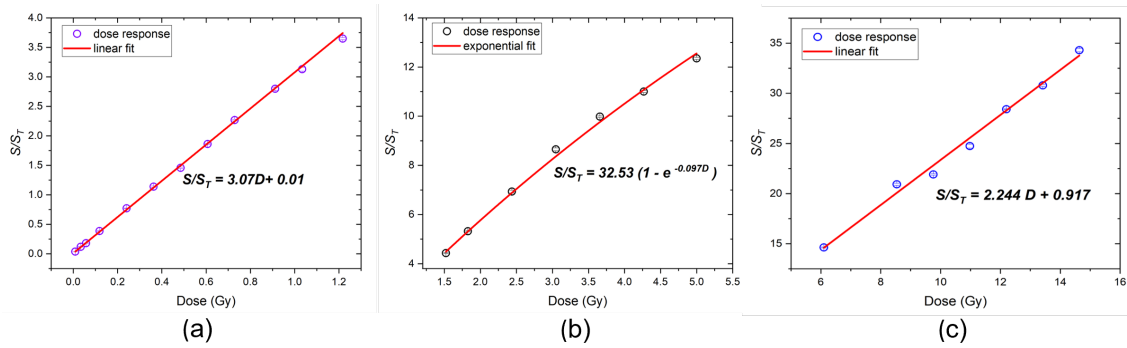


Figure 5.18: TL dose responses for regular grains, determined by breaking the dose response in Figure 5.16b into 3 smaller fractions. (a) Dose response from 0–1.2 Gy. (b) Dose response from >1.2–5.0 Gy. (c) Dose response from >5–15 Gy. The fit for each dose response is represented by a red line. The equations for the fits are also shown.

The shorter dose-range dose responses presented in Figures 5.17 and 5.18 are all linear

except for Figure 5.18b where a saturating exponential fit was used. In deciding which dose response to use for dose recovery, the test-dose corrected signal S/S_T calculated for a dosimeter with an unknown dose is simply compared against the y-axis of the plots to decide which dose response will be appropriate for dose recovery. These dose response curves in Figures 5.17 and 5.18 were thus used to calculate equivalent doses for all dosimeter grains exposed to radiation in this work.

5.5.5 Dose Recovery for single grains exposed to known beta doses

As a method of testing the accuracy of the growth curves presented in Figures 5.17 and 5.18, some single grains were exposed to known doses of β radiation and an attempt was made to recover the known doses from their test-dose corrected signals. The results are presented in Table 5.3.

Table 5.3: Dose recovery results for seven single grains irradiated with known doses of β radiation. Error bars represents 2σ uncertainty.

Known Dose (Gy)	regular grains		α -desensitized grains	
	Recovered Dose (Gy)	Recovery Ratio	Recovered Dose (Gy)	Recovery Ratio
0.26 ± 0.01	0.27 ± 0.02	1.04	0.28 ± 0.03	1.08
0.53 ± 0.03	0.52 ± 0.04	0.98	0.54 ± 0.05	1.02
0.59 ± 0.03	0.57 ± 0.04	0.97	0.60 ± 0.06	1.02
1.06 ± 0.06	1.06 ± 0.08	1.00	1.08 ± 0.10	1.02
1.12 ± 0.06	1.04 ± 0.11	0.93	1.13 ± 0.11	1.01
2.12 ± 0.12	2.09 ± 0.17	0.99	2.18 ± 0.21	1.03
3.31 ± 0.18	3.26 ± 0.25	0.98	3.35 ± 0.31	1.01

Error bars represent 2 sigma uncertainties. The uncertainties on the known doses are the uncertainty in source calibration, while the uncertainties on the recovered doses are the measurement uncertainties. The result shows that the recovered doses agree with the known doses within their error limits. On average, the dose recovery ratio for known doses

recovered using the dose response curves are (0.98 ± 0.03) and (1.03 ± 0.02) for the regular and α -desensitized grains respectively. The maximum deviation for dose recovery is 7% and 8% for the regular and α -desensitized grains respectively. In general, the result shows that doses can be recovered with an uncertainty of about 0–8%.

5.5.5.1 Measured dose distributions for single grains exposed to beta radiation only

In section 5.5.3, reproducibility tests were presented for the test-dose corrected signals of 12 regular and 12 α -desensitized 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains. The variance in signals was calculated. In this section the test-dose corrected signals were compared to the dose response and the equivalent doses for those grains were determined. The resulting dose distributions are presented in Figure 5.19a for regular and desensitized grains. Figure 5.19b shows for direct comparison the same distributions, normalized to their respective mean values.

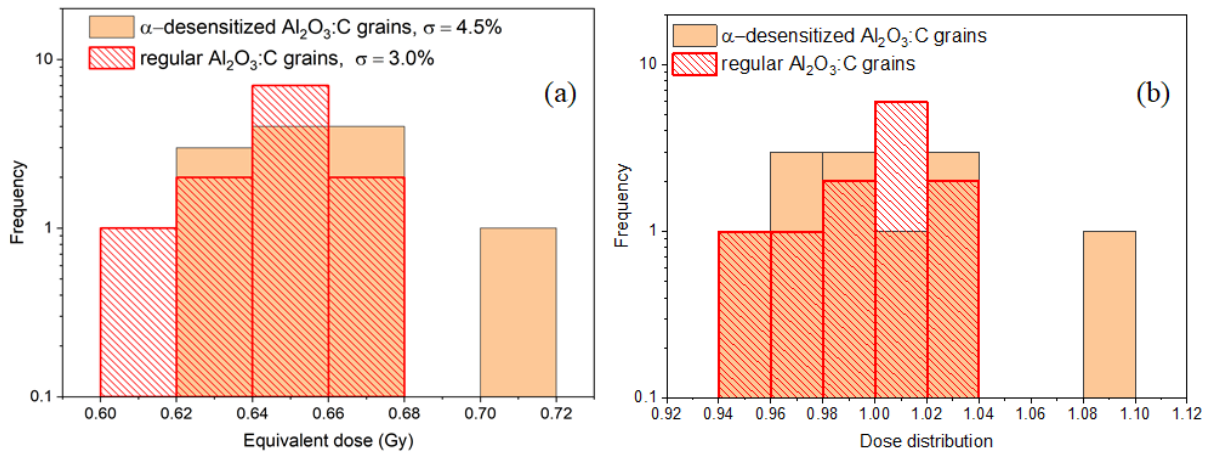


Figure 5.19: Comparison of (a) absolute dose distribution and (b) relative dose distribution for 12 regular and 12 α -desensitized 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains exposed to a 10s β -dose. The estimated statistical uncertainty of individual measurements was 3–5%.

Using their respective dose response curves, the mean dose recovered from the regular grains was (0.65 ± 0.02) Gy compared to (0.66 ± 0.03) Gy for the α -desensitized grains. The grains were irradiated with 10s β irradiation so that the actual doses given were (0.66 ± 0.04) Gy and (0.69 ± 0.06) Gy respectively, showing that the mean recovered doses are not only

the same but also agree with the given doses within their error limits.

While the mean recovered doses are the same, as can be seen from Figure 5.19b, the distributions themselves are not the same. The spread represented as relative standard deviation for the α -desensitized grains is 4.5% compared to 3% for the regular grains. In the dose distribution of the α -desensitized grains, one of the dose points was confirmed to be an outlier at the 0.1 level using the Grubbs's test (see Figure 5.20). This explains the higher standard deviation of the distribution. When this dose point was removed, the spread was 3% like that of the regular grains. Nevertheless, the overall trend is that dose recovery was accurate and the dosimeters showed good uniformity since their standard deviations are within the measurement uncertainties.

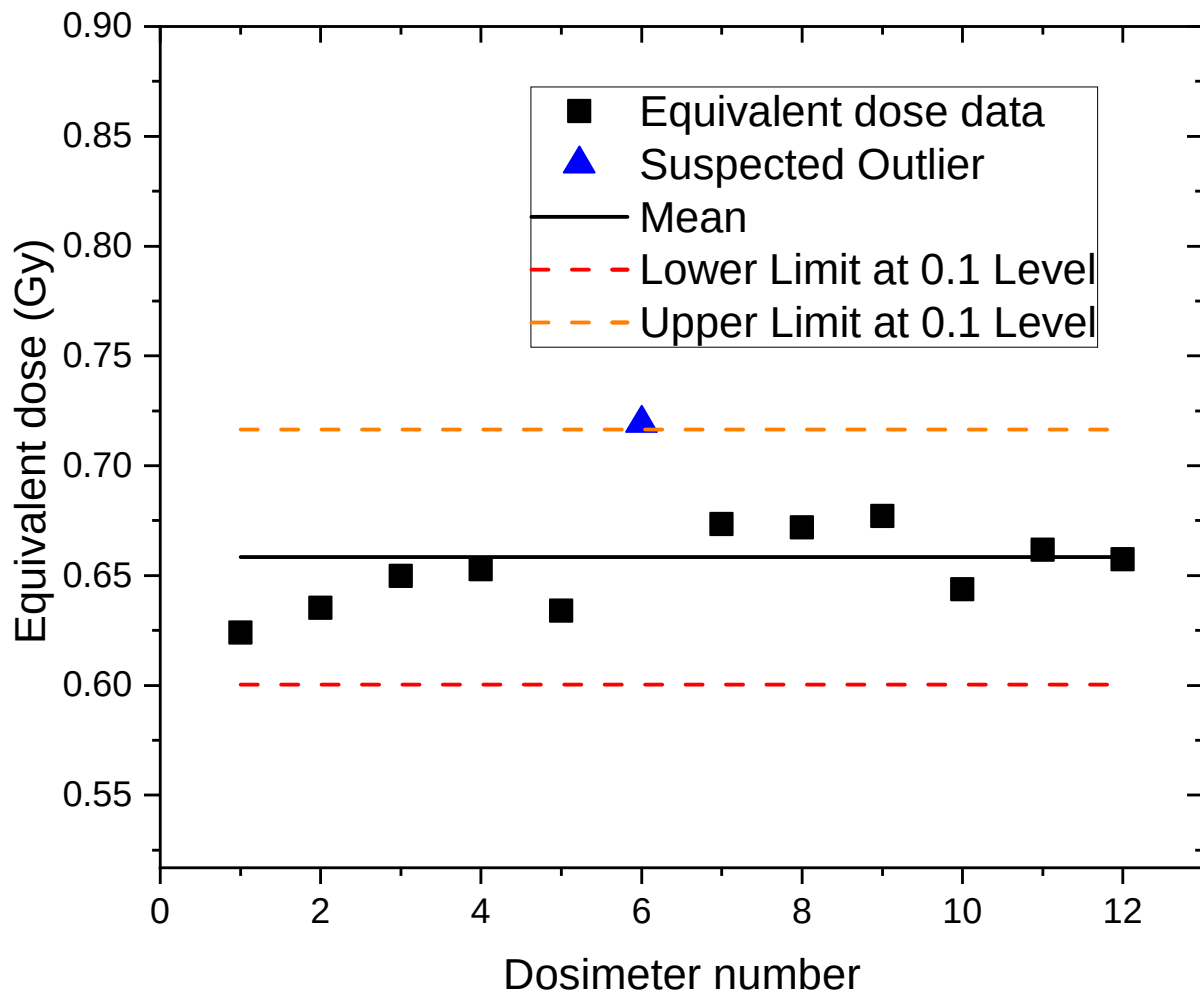


Figure 5.20: Grubbs's Test showing that 0.719 Gy data point in the equivalent doses of the α -desensitized grains is an the outlier at the 0.1 level

5.5.5.2 Measured dose distributions for single grains exposed to mixed-field alpha and beta radiation

Grains buried in sediments will be exposed to both, beta and alpha radiation. Thus, another test was conducted to investigate the effect of mixed radiation, i.e., the effect of α -radiation in addition to β -irradiation, on the dose distribution of the dosimeter grains. The investigation was carried out using the following procedure:

- (i) anneal the grain at 900 °C for 15 min in an external furnace to bleach
- (ii) irradiate grain with dose equivalent to 10s β radiation
- (iii) irradiate grain with dose equivalent to 500s α radiation
- (iv) 2 TL readouts to measure signal, then background
- (v) irradiate grains with a test-dose equivalent to 5s β radiation
- (vi) 2 TL readouts to measure signal, then background
- (vii) repeat steps (i)–(vi) for 11 more regular grains.
- (viii) repeat steps (i)–(vi) for 12 α -desensitized grains
- (ix) recover the dose for each dosimeter using their test-dose corrected signal and the dose response.

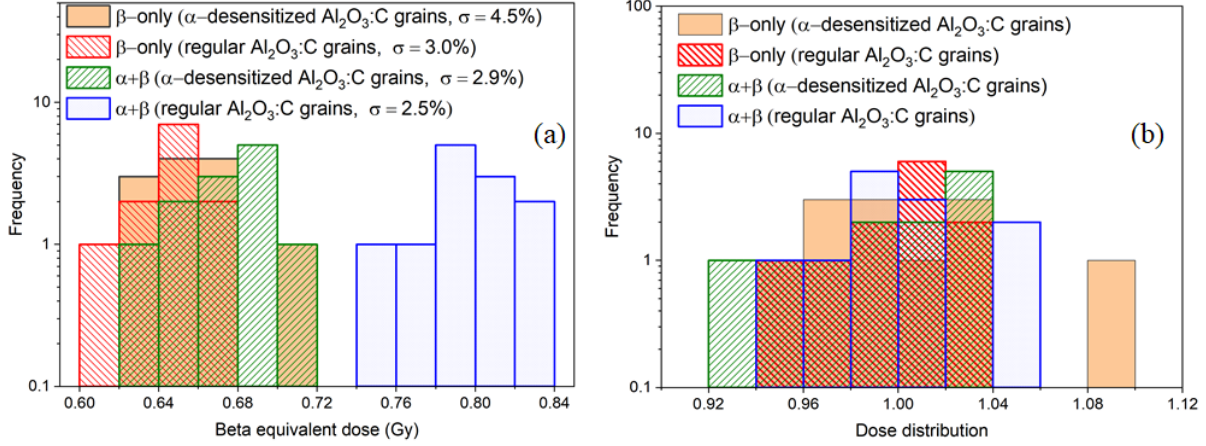


Figure 5.21: Comparison of dose distribution for 12 regular grains exposed to 10s β irradiation (red), 500s α irradiation in addition to 10s β irradiation (blue), and 12 α -desensitized grains exposed to 10s β irradiation (brown), and 500s α irradiation in addition to 10s β irradiation (green). The estimated statistical uncertainty of individual measurements was 3-5%. (a) Measured doses. (b) Same distributions normalized by their respective mean values.

Figure 5.21a compares the dose distributions for beta-only and mixed radiation fields for both, regular and desensitized grains. For easier comparison the distributions were normalized by their respective mean values (Figure 5.21b). When exposed to 500s α radiation in addition to 10s β radiation, the mean dose to the regular grains was (0.79 ± 0.02) Gy compared to (0.68 ± 0.02) Gy for the α -desensitized grains. The additional α -particle irradiation causes a 22% increase in measured dose for the regular grains, but only a 3% increase for the α -desensitized grains. The latter is statistically not significant as it agrees within error limits with the beta-only value, confirming their negligible sensitivity to alpha radiation. The standard deviations for the new dose distributions were also within measurement uncertainties.

Additionally, the decrease in standard deviations for the α -desensitized grains may be used to justify our initial argument that the suspected data point with almost a +12% deviation is indeed an outlier, since it was not seen in the new distribution. Possible explanations for the outlier and the 3–5% measurement uncertainty are counting statistics, reader stability and uneven heating of grains during readout. The temperature is monitored for the heating plate, not for the grain itself. Thermal lag between cup and heating plate as well as grain and cup can introduce signal variation.

5.6 Summary

In this chapter the important characteristics of our dosimetric materials which include $\text{Al}_2\text{O}_3\text{:C}$ 250–355 μm dosimeter grains and Luxel[®] dosimeters, and the Risø OSL/TL reader have been described. The precision with which we can measure dose from a single dosimeter has been determined. Dose responses for all the dosimeter types were built and tested. The uniformity of dose distribution when several dosimeters were irradiated with the same dose was also calculated. Last but not the least, reliable measurement procedures were developed and tested for all the dosimeters.

For Luxel[®] dosimeters, an OSL measurement protocol was designed, that accounts for variation between dosimeters and corrects the OSL signal using test-dose. This is discussed in section 5.4. The measurement reproducibility was approximately 3.5%. The dose recovery accuracy, for dosimeters irradiated with known doses, using a standard dose response curve was about 2–10%. Similarly, a TL measurement protocol based on test-dose corrected signals was also devised for single $\text{Al}_2\text{O}_3\text{:C}$ grains. This is discussed in section 5.5.1. The measurement reproducibility was better than 4%. The dose recovery accuracy, for dosimeters irradiated with known doses, using a standard dose response curve was about 0–8%.

In conclusion, we have demonstrated that our dosimetric system is reliable, and capable of measuring dose distribution precisely and accurately under the studied irradiation conditions.

Chapter 6

Characterization of the Monte Carlo models

Sediment is mineral or organic material transported and deposited under the influence of wind, water or glaciers. Thus, sediments are highly non-uniform and are composed of a variety of minerals, of different grain sizes and shapes. Modelling these complex environments is only possible in a simplified version and through approximations. The Geant4 Monte Carlo (MC) programs *DosiSed* and *DosiVox* were used in this work to simulate radiation transport in sediments. The theoretical framework of the software packages has already been discussed in Chapter 4. The goal of this chapter is to validate the numerical simulation approximations employed and to quantify the uncertainty associated with the results.

First, the method of creating sediment and dosimeter models in *DosiSed* and *DosiVox* as well as the approach taken to analyze data from the models are discussed. Some preliminary models are presented to characterize the reliability and precision of results. Finally, some simple well-known examples of dose attenuation in layers and grains are simulated and results are compared to tabulated empirical data.

6.1 Basic Considerations for Simulating Sediments and Dosimeters with DosiSed and DosiVox

DosiSed and *DosiVox* are C++ programs based on the Geant4 toolkit that can be used to simulate particle-matter interaction using Monte Carlo techniques (see Chapter 4 for more details). They allow modelling sediments by constructing a voxelised 3D space, i.e., the volume of interest is divided into smaller fragments called "voxels". The mineral compositions, associated radioactivity as well as grain size distribution can be defined in each voxel [155]. The emission of either alpha-, beta-, or gamma- particles from the radioactivity in the sample is then simulated and the corresponding dose deposited as well as the energy emitted is recorded by different detector types. Some advantages of these programs are that they have a user-friendly interface and do not require elaborate programming knowledge. However, *DosiVox* is still relatively new and has seen only a few usages for advanced modelling [155, 185–187]. For *DosiSed*, on the other hand, no published results were found, yet it is arguably more robust for grain modelling.

The differences between *DosiVox* and *DosiSed* are in the output result and the extent to which voxel properties can be manipulated. For *DosiVox*, the mineral properties of each individual voxel can be manipulated. Thus the total volume can be filled with voxels of different properties. However, it is not possible to obtain dose values for the entire 3D-space. Instead, two options exist to "detect" the absorbed dose values in a limited volume: (i) a cylindrical probe that is divided into segments is inserted into the 3D-space; or (ii) one single voxel can be divided into multiple smaller voxels. In each case, the dose absorbed in each of the segments or subvoxels is recorded.

DosiSed, on the other hand, does not allow manipulation of mineral properties per individual voxel. A single voxel is randomly filled with grains of defined size and composition; identical voxels are then stacked in 3D-space to fill the entire space with mineral grains (see section 4 for details). In the output dose values are reported for each single grain of a defined

material and grain size in the entire simulation space. The $\text{Al}_2\text{O}_3\text{:C}$ Luxel[®] dosimeters have the shape of disks. Their respective samples were therefore modeled using *DosiVox* and the cylindrical probe. Samples with the 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains were modeled with *DosiSed*.

Sediment samples were approximated with the following simplified composition. A limited number of mineral types was selected. Each mineral was modelled to consist of spherical grains of selected appropriate sizes. The grains were randomly packed and distributed in each voxel that makes up the model (3D volume of the sample). The total number of grains per type of mineral depends on the volume fraction of the mineral in the sample.

6.2 Model Creation and Data Analysis

For each simulation in *DosiSed/DosiVox*, the user begins with a pilot file where all the model parameters must be specified. This pilot file (section 4.4.2) can be created using a graphical tool provided alongside the software. It can also be constructed manually by editing one of the template pilot files provided. This was the approach taken in this work. In the pilot file, the geometry, the various materials and their mineral information are specified along with their radio-isotope concentrations. In our case, the radioactive materials were given a radio-isotope concentration of 1 ppm for U and Th (1% for K) instead of their actual concentrations, for easy manipulation of pilot files from one model to another. In a single simulation run, only one type of radiation (alpha, beta or gamma) and one radioisotope (U, Th or K) can be simulated. Thus, multiple simulations are required to obtain the result of alpha, beta and gamma radiations for all the radioisotopes in a sample. The result is then scaled later with the actual radioisotope concentration. Doing this also allowed us to save the additional time required in specifying manually the radioisotope concentration for each voxel that makes up the model. One reason why we could do this is because we had one radio-emitter per sample in our complex models. If there were multiple radio-emitters per sample, then we would have to specify per voxel. The energy spectra used for β emissions

of K, U-, and Th-series were taken from Guérin [188], while for α emission, the spectra were adopted from ENSDF (Evaluated Nuclear Structure Data Files, Brookhaven National Laboratory).

At the end of the simulation two results are reported: the dose (D) absorbed in each grain or voxel as well as the energy (E) emitted per unit mass of the radioactive grain or voxel in the model. This energy emitted per unit mass represents the infinite matrix dose, which is the dose that every grain in the model would absorb if the model constituted a homogenous material and if it had infinite size relative to the range of all particles emitted in the model. Let us consider a single simulation run, e.g., alpha emission of the Th-series. The dose rate can be calculated from the absorbed dose fraction of the infinite matrix dose (D/E) as follows:

$$\dot{D}_\alpha = c_{Th} \frac{D_{\alpha,Th}}{E_{\alpha,Th}} F_{\alpha,Th}, \quad (6.1)$$

where c is the average radioisotope concentration in ppm or percent for the radioactive mineral in the sample. F is the energy conversion factor in $\text{Gyka}^{-1}\text{ppm}^{-1}$. The F values allow direct conversion of radioactive element concentrations (in ppm or %) into dose rate. They have been tabulated for the three radiation types and radioactive isotopes considered in this work. For Th- and U-series, these factors were taken from [29] while for ^{40}K , the updated factors were taken from [189].

The total dose rate for the Th-series can be calculated after 3 simulation runs:

$$\dot{D}_{Th} = \sum_{i=1}^3 c_{Th} \frac{D_{i,Th}}{E_{i,Th}} F_{i,Th}, \quad (6.2)$$

where i represents the radiation type (alpha, beta, and or gamma).

Because of the Monte-Carlo technique used, our raw data from *DosiSed/DosiVox* does not account for time. Eq. 6.1 & Eq. 6.2 are thus used to obtain the dose rate $\dot{D}$ (in Gy/a) per voxel/subvoxel/grain.

The total dose rate for Th, U, and K, on the other hand can be calculated after 8

simulations using:

$$\dot{D} = \sum_{i=1}^3 c_{Th} \frac{D_{i,Th}}{E_{i,Th}} F_{i,Th} + \sum_{i=1}^3 c_U \frac{D_{i,U}}{E_{i,U}} F_{i,U} + \sum_{i=1}^2 c_K \frac{D_{i,K}}{E_{i,K}} F_{i,K}, \quad (6.3)$$

^{40}K emits beta and gamma particles only, so that there are only two components in the sum.

To calculate the total D_E , the respective values for all radiation types and radioisotopes have to be added and then multiplied by t . It is given as:

$$D_E = \dot{D} t, \quad (6.4)$$

where t is the length of time (in years) for which the dosimeter was exposed to radiation.

The uncertainty follows from the individual uncertainties by applying the standard laws of error propagation as follows:

$$\delta D_E = D_E \sqrt{\left(\frac{\delta c_{i,j}}{c_{i,j}}\right)^2 + \left(\frac{\delta D_{i,j}}{D_{i,j}}\right)^2 + \left(\frac{\delta E_{i,j}}{E_{i,j}}\right)^2 + \left(\frac{\delta t}{t}\right)^2} \quad (6.5)$$

Where applicable, the error in the radioisotope concentrations δc resulted from gamma spectrometry. The exposure time for buried dosimeters was measured in days. A conservative estimate of the error was performed to obtain an error of 1.1% (i.e., 4 days/365 days). For a voxel or subvoxel, the errors for D and E are computed by the program as the standard deviations of the dose deposited in, and energy emitted from, the voxel or subvoxels respectively. For a single grain, the errors are obtained from the width of the dose distribution in the case where multiple grains of that same mineral are given uniform dose.

6.3 Testing the Reliability and Precision of the Monte Carlo simulations

6.3.1 Complementarity of external and internal β dose rates: proof of radiation equilibrium

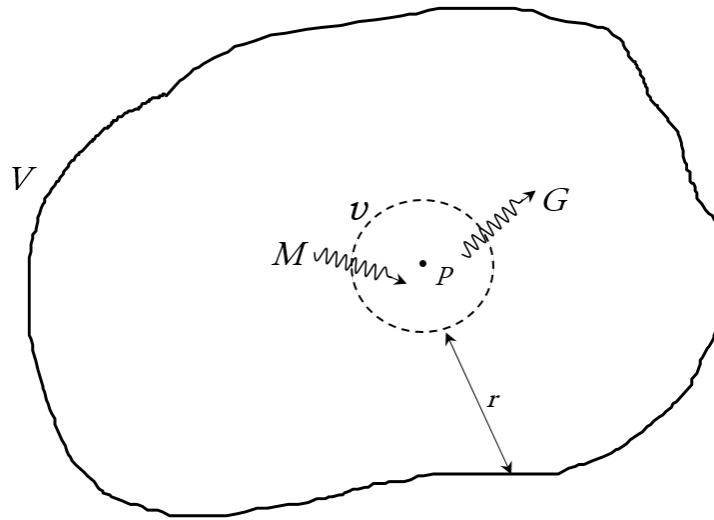


Figure 6.1: Radiation equilibrium existing at a point P within an infinite-matrix volume V . The radiation energy G carried out of the volume v by a charged particle is balanced by the radiation energy M carried into the volume v as long as the distance r is greater than the range of energy deposition of the particles. Adapted from [30, Fig. 1.2]

A common assumption that is made in dose rate calculation for trapped charged dating is the existence of an infinite-matrix (IM) in the sediment. The presence of an IM means that a state of radiation equilibrium exists in the sediment so that dose rate is uniform throughout the sediment and the *absorbed dose* (dose = energy per unit mass) in a volume is equal to the energy per unit mass *emitted* from the volume [30]. Figure 6.1 depicts radiation equilibrium where the energy of radiation G carried out of the volume v is balanced by the energy of radiation M carried into the volume v for both directly and indirectly ionizing radiation. For the IM assumption to be valid, the following conditions must be satisfied:

1. The atomic composition and density of the sediment is homogeneous
2. The radioactivity is homogeneous in the sediment
3. The dimension of the sediment is greater than the effective range of the most penetrating radiation, including any generated bremsstrahlung x-rays in the case of β -radiation.

A direct implication of this is, in the case where radioactivity is uniform in the matrix and in the grain, the grain receives the infinite matrix dose rate. However, assuming only the grain is radioactive and the matrix is not radioactive, the dose rate received by the grain is a fraction f of the IM dose rate. This fraction f is called "self-dose" or "self-dose value". If we consider the reverse case, where the grain is not radioactive, but only the matrix is, the grain receives a fraction $(100 - f(\%))$ of the IM dose rate. This fraction $(1-f)$ is called the "attenuation fraction" or "attenuation factor". If the two models are run independently, one expects therefore that these two fractions should add up to 100%. A single grain embedded in a sedimentary environment, as opposed to a uniform matrix, experiences additional dosimetric effects induced by neighboring grains. However, the complementarity effect should still hold in sedimentary environments as well. The complementarity effect is therefore a simple test that can be performed to investigate the reliability of a Monte Carlo Code. This effect was thus tested to investigate the reliability of *DosiVox* and *DosiSed* using a model previously presented by Martin et. al. [190].

The test involves creating a representative sediment using random packing of spherical grains. The model is made up of $3 \times 3 \times 3$ cubic voxels, each 5 mm in size for a total grid size of 15 mm $\times$ 15 mm $\times$ 15 mm. Each cubic voxel was packed with 400 μ m diameter silica (quartz) grains. The space between the grains, the so-called matrix, was filled with silica (SiO_2 , density: 2.65 g/cm³). The grains represented 30% of the voxel's volume (see Figure 6.2). The total number of grains was approximately 1000. The entire voxel is homogeneous in terms of material. Two types of simulations were done separately:

- type 1 – emission of β -particles from the grains, matrix is inert

- type 2 – emission of β -particles from the surrounding voxel (matrix), grains are inert.

Only the β -spectrum of ^{40}K was simulated. The dimensions of the simulation space ($15\text{ mm} \times 15\text{ mm} \times 15\text{ mm}$) compared to the range of β particles ($\sim 2\text{ mm}$) ensure that the IM condition is satisfied. For both simulation type, only grains in the central voxel were considered to avoid edge-effects. The doses deposited in the grains as a percent of their respective infinite matrix dose rates were recorded and compared. In the case where the grains were radioactive (type 1), the average deposited dose in the grains was $(36.7 \pm 1.4)\%$ of the infinite matrix dose rate. The average deposited dose in the grains due to emission from the surrounding matrix (type 2) was $(62.8 \pm 1.8)\%$ of the infinite matrix dose rate. The sum of the two dose rates is 100% within error limits, and thus satisfies the superposition principle. This result also agrees with the result reported by Martin et. al. [190].

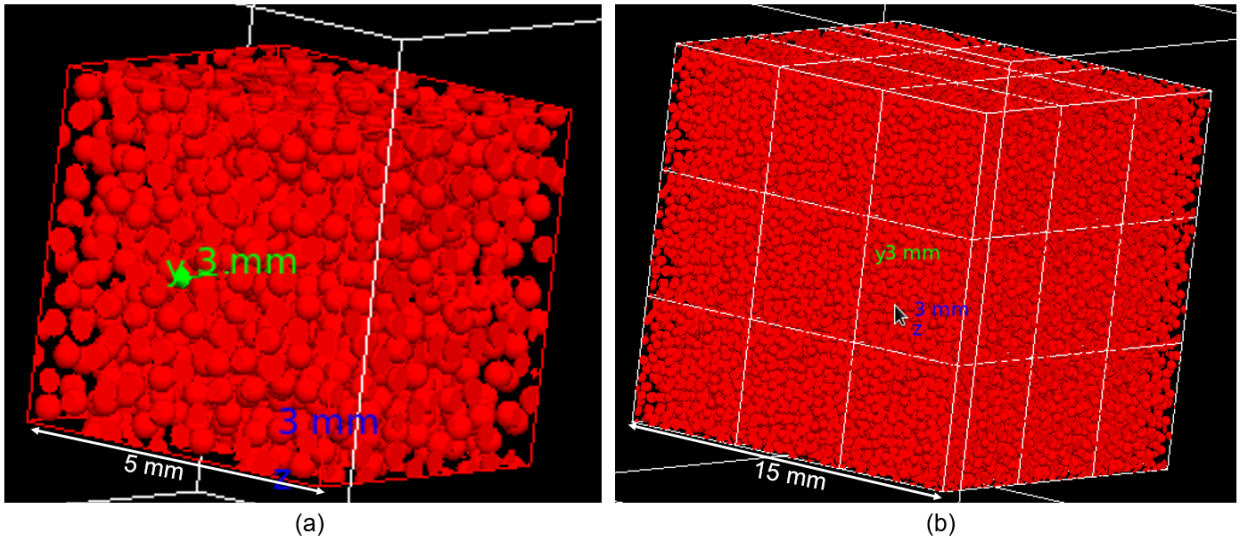


Figure 6.2: (a) The central voxel filled with $400\text{ }\mu\text{m}$ diameter quartz grains that were also specified as detectors. (b) The entire $3 \times 3 \times 3$ cubic voxels grid. The extra voxels in the grid are replicas of the central voxel but do not act as detectors. They were packed around the central voxel to satisfy the IM condition. The number of particles simulated is 10^7 .

6.3.2 Effect of the number of simulated particles on the result

Considering that Monte Carlo simulation is based on the probabilistic principle, the result of each simulation depends on whether the number of particles simulated is sufficient to

represent the behavior of the system realistically. Unfortunately, simulation time increases with the number of particles simulated. Therefore, one has to find a balance between minimal statistical uncertainty and reasonable computational time. In this section, a dimensionally similar model like the one described in section 6.3.1 was used to study the effect the number of simulated particles has on the statistical uncertainties of the result. However, unlike the model in section 6.3.1, the β -spectrum of the Th-series was simulated from the matrix while the grains are kept inert. Only grains in the central voxel were considered to avoid edge-effects. The average deposited dose fraction as a percent of the infinite matrix dose and the corresponding **standard deviations** were calculated for different numbers of simulated particles ranging from 10^5 to 10^{10} . The deposited dose fraction of the infinite matrix dose rate in this case is simply the *attenuation factor* of the grain since the radiation is emitted externally from the grain. The statistical uncertainties for each number of simulated particles are represented by the standard deviations of the corresponding attenuation factors. The result is shown in Figure 6.3.

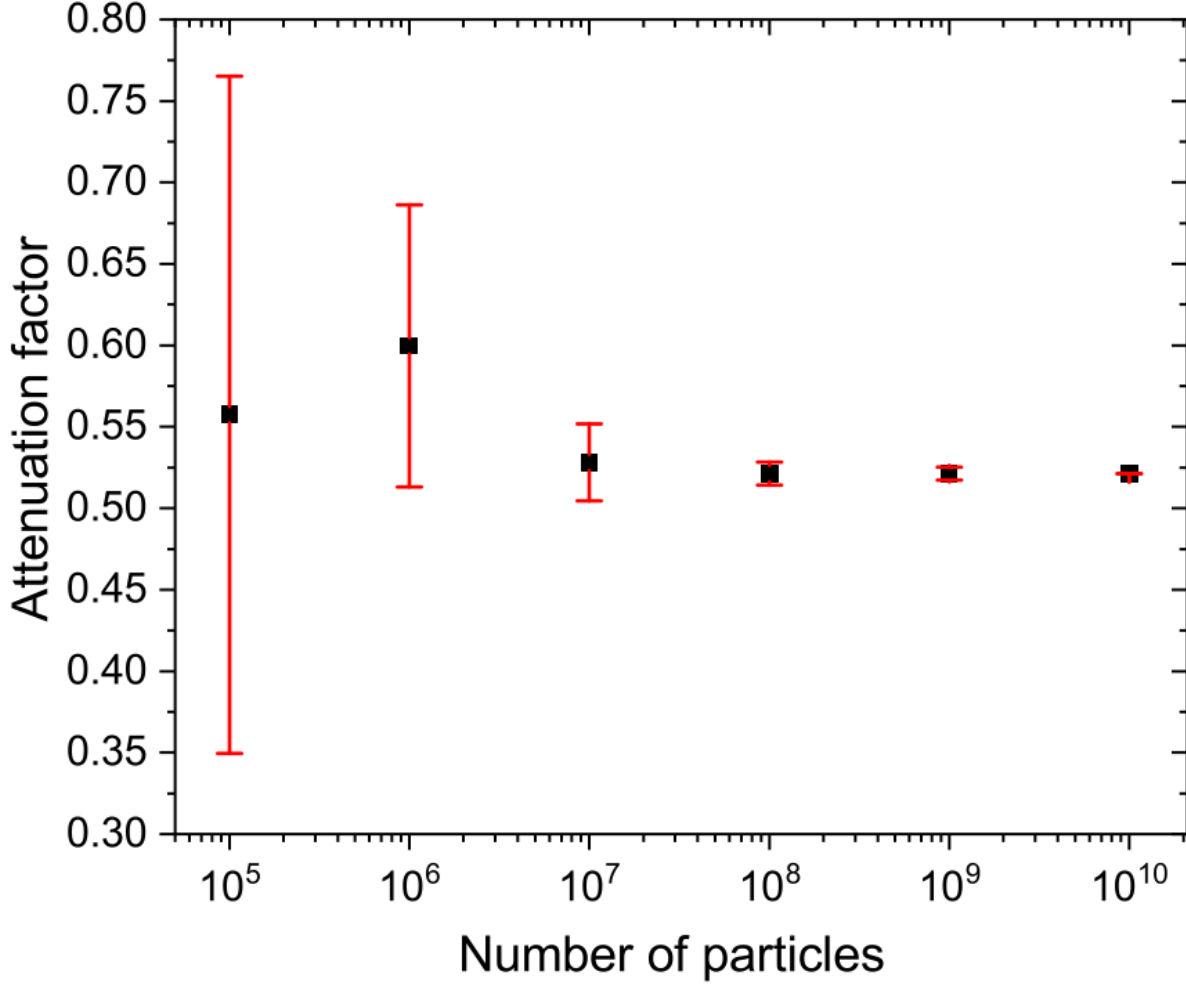


Figure 6.3: Effect of number of simulated particles on the statistical uncertainties of the result. This result represents the attenuation factors calculated for the case of emission of β -particles (Th-series) from the matrix.

The result shows that the standard deviations for 10^5 , 10^6 , 10^7 , 10^8 , 10^9 , and 10^{10} simulated particles, were 37%, 14.5%, 4.5%, 1.4%, 0.8% and 0.02% respectively. A simulation of 10^7 particles and above is enough to reduce the statistical uncertainty to less than 5%. The simulation time for 10^7 particles is about 15–30 min for the 64 GB RAM computer used. The uncertainty associated with 10^{10} particles is 0.02%. However, it takes about a month to run this simple model here. For a complex model, it can take about 2–3 months to run, and is therefore not feasible.

6.3.3 Beta self-dose values

6.3.3.1 Quartz and feldspar

In a natural sedimentary environment, a homogeneous infinite-matrix (IM) sediment like the one discussed in section 6.3.1 does not exist. This is because mineral grains attenuate radiation in a manner that depends on their chemical compositions, sizes and shapes. This introduces heterogeneities into the uniform infinite matrix. To quantify this effect, Mejdahl [191], using the principle of energy conservation and the superposition theorem (i.e., attenuation factor + self dose value = 1, under IM condition), followed by Fain et al. [156], and Brennan [157] calculated the self-dose factors for quartz grains, as a function of their size and shape. The approach consists of considering a radioactive quartz grain in an infinite homogeneous inert matrix. In this section, β self-dose factors for spherical quartz and feldspar grains of different sizes were calculated. The results were compared to similar calculations performed by Guérin et al. [29] using Geant4, Nathan [30], using MCNP and the dose point kernels method [29, 30], and Mejdahl [191]. Dose point kernels (DPK) are radial distributions of dose from a point source within a uniform material. DPKs are some of the most common numerical methods for calculating integrated dose through an infinite homogeneous body with an extended source [30, 192].

An example of the model used for these calculations is shown in Figures 6.4 and 6.5. For quartz, the matrix material chosen was silica, i.e, SiO_2 with a density of 2.65 g/cm^3 . The grain material is also quartz (SiO_2) with a density of 2.65 g/cm^3 . The quartz grain constitutes a radioactive sphere within the non-radioactive silica matrix. For feldspar, the matrix material chosen was also silica, i.e, SiO_2 with a density of 2.65 g/cm^3 . The grain material is potassium feldspar (KAlSi_3O_8) with a density of 2.60 g/cm^3 . The potassium feldspar grain constitutes a radioactive sphere within the non-radioactive silica matrix. The diameter of grains considered here ranges from 20 to $6000 \mu\text{m}$.

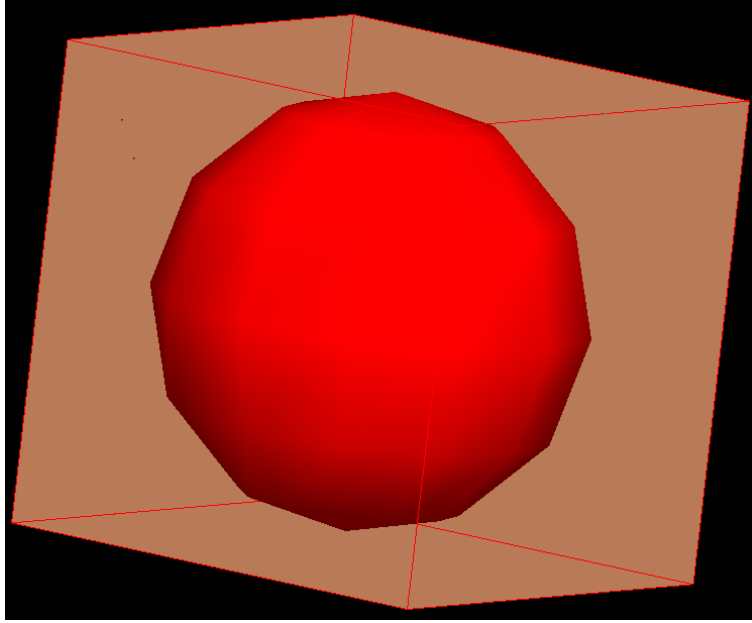


Figure 6.4: The model for the self-dose factor calculation. The grain (red) and the matrix (brown) are composed of the same material for quartz and different materials for feldspar.

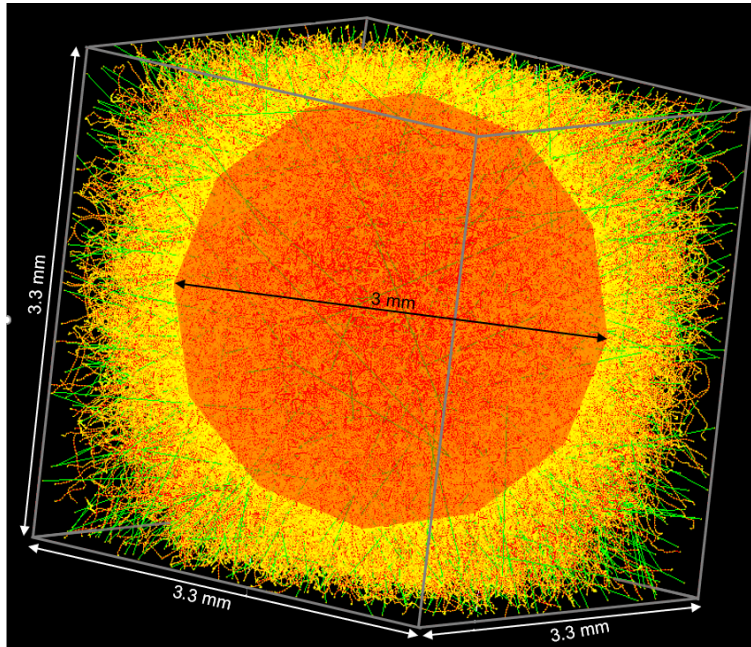


Figure 6.5: Beta spectrum emission (from ^{40}K) model of a 3 mm diameter grain enclosed in a 3.3 mm cubic voxel. This is an example of the model used to calculate the self-dose factors in Table 6.1. The self-dose represents the amount of radiation absorbed by the grain that is also emitting it. For the 3 mm quartz grain, the self-dose for ^{40}K is approximately 73% of the IM dose.

Table 6.1: Beta self-dose values (f) calculated with *DosiVox* and *DosiSed* for spherical quartz and potassium feldspar grains of different sizes. The internal radionuclide concentrations were 1% for K, 1 ppm for Th and 1 ppm for U. The results are compared with values reported in the literature.

Grain size (mm)	<i>Result</i>	Quartz ($d = 2.65 \text{ g cm}^{-3}$)				Feldspar ($d = 2.60 \text{ g cm}^{-3}$)		
		Nathan ^a	DPK	Mejdahl	Guérin ^b	<i>Result</i>	DPK	Guérin ^b
<i>Potassium</i>								
0.02	0.007 ± 0.001	NA	0.008	0.007	0.007	0.007 ± 0.000	0.008	0.007
0.06	0.022 ± 0.001	NA	0.022	0.021	0.022	0.021 ± 0.000	0.021	0.021
0.10	0.037 ± 0.001	0.037	0.036	0.035	0.037	0.036 ± 0.000	0.035	0.035
0.20	0.075 ± 0.001	0.076	0.074	0.070	0.075	0.073 ± 0.000	0.071	0.072
0.30	0.115 ± 0.001	0.116	0.112	0.105	0.114	0.115 ± 0.001	0.108	0.110
0.50	0.193 ± 0.000	0.196	0.189	0.175	NA	0.190 ± 0.000	0.183	NA
1.00	0.375 ± 0.001	0.378	0.368	0.337	0.372	0.371 ± 0.000	0.358	0.360
2.00	0.614 ± 0.001	0.615	0.606	0.571	NA	0.610 ± 0.001	0.596	NA
6.00	0.861 ± 0.001	0.864	0.859	0.837	NA	0.859 ± 0.001	0.855	NA
<i>Thorium</i>								
0.02	0.049 ± 0.000	NA	0.055	0.058	0.048	0.048 ± 0.000	0.054	0.047
0.06	0.100 ± 0.000	NA	0.104	0.109	0.098	0.098 ± 0.000	0.103	0.097
0.10	0.135 ± 0.000	0.133	0.139	0.143	0.134	0.133 ± 0.000	0.137	0.131
0.20	0.201 ± 0.000	0.198	0.203	0.206	0.200	0.199 ± 0.000	0.200	0.196
0.30	0.249 ± 0.000	0.245	0.250	0.251	0.247	0.246 ± 0.000	0.247	0.243
0.50	0.321 ± 0.000	0.317	0.321	0.320	NA	0.319 ± 0.001	0.317	NA
1.00	0.452 ± 0.001	0.444	0.449	0.443	0.449	0.448 ± 0.001	0.443	0.441
2.00	0.620 ± 0.001	0.611	0.616	0.604	NA	0.617 ± 0.001	0.609	NA
6.00	0.850 ± 0.001	0.837	0.849	0.825	NA	0.848 ± 0.000	0.844	NA
<i>Uranium</i>								
0.02	0.039 ± 0.000	NA	0.040	0.037	0.038	0.038 ± 0.000	0.040	0.037
0.06	0.072 ± 0.000	NA	0.073	0.071	0.072	0.071 ± 0.000	0.072	0.056
0.10	0.096 ± 0.000	0.094	0.096	0.096	0.095	0.094 ± 0.000	0.094	0.093
0.20	0.144 ± 0.000	0.142	0.143	0.146	0.143	0.142 ± 0.000	0.140	0.140
0.30	0.187 ± 0.000	0.184	0.184	0.189	0.185	0.184 ± 0.000	0.181	0.181
0.50	0.259 ± 0.000	0.257	0.255	0.263	NA	0.256 ± 0.000	0.251	NA
1.00	0.398 ± 0.001	0.393	0.392	0.400	0.395	0.394 ± 0.001	0.385	0.387
2.00	0.575 ± 0.001	0.569	0.568	0.568	NA	0.571 ± 0.000	0.560	NA
6.00	0.823 ± 0.003	0.817	0.821	0.803	NA	0.822 ± 0.001	0.816	NA

^a calculated using MCNP code for unetched spherical grains [30]

^b calculated using a different Geant4 code [29]

^{NA} data not available

For each grain size chosen, a voxel size is used that is just big enough to allow only a single grain. This ensures that the position of the grain in repeated simulations for a specific grain size is the same. The number of simulated particles is 10^8 . 10 simulations were performed for each grain size. Five with *DosiVox* and five with *DosiSed*. The self-dose values are average values from the 10 data points for each grain size. The errors represent the standard deviation of the 10 simulation results. They are reported in Table 6.1 in the column *Result*. There was no significant difference between the *DosiVox* and *DosiSed* results since the spectra data are the same. This is evident from the very low standard deviations. The self-dose values increase with grain size. This is expected because as the grain size increases, the total distance travelled by the particle inside the grain also increases. Dose deposition occurs along the travel path of charged particles, resulting in the well-known exponential dose fall-off of charge particles.

The results show good agreement with the results in the literature. The maximum deviation (11%) was seen between our results and the results by Mejdahl [191] for the ^{40}K self-dose value of the 1 mm quartz grain. Mejdahl [191] used data from Berger [193, 194] that was scaled from water to quartz by a process introduced by Cross [195]. This data has been since updated by Brennan [196] using DPKs generated directly in quartz. The updated data was used for the DPK-method calculated self-dose values by Nathan [30]. Our results, when compared to the updated DPK, MNCP, and Geant4 results have a maximum deviation of about 6%, 2%, and 4% respectively. All the maximum deviations are seen for potassium. A possible explanation for this and the deviations in the updated DPK-method result is the use of a different spectrum for potassium in our dose calculations and the introduction of a systematic error during smoothing and interpolation of the monoenergetic DPK to form composite DPK [30].

6.3.3.2 Beta self-dose values for $\text{Al}_2\text{O}_3\text{:C}$

Given that our dosimeter of choice for this work is $\text{Al}_2\text{O}_3\text{:C}$, the β self-dose factors for spherical Al_2O_3 grains of different sizes in a homogeneous matrix of Al_2O_3 were calculated as well. The results were compared to the tabulated values obtained by Nathan [30], using the dose point kernel method. The comparison is presented in Table 6.2. The uncertainties represent the standard deviation of self-dose values from 5 runs, each run being a simulation of 10 million particles.

Table 6.2: Beta self-dose values (f) calculated for spherical Al_2O_3 grains of different sizes. The internal radionuclide concentrations were 1% for K, 1 ppm for Th and 1 ppm for U. The results are compared with values reported in the literature

Grain size (μm)	Uranium series		Thorium series		Potassium series	
	<i>Result</i>	Nathan ^a	<i>Result</i>	Nathan ^a	<i>Result</i>	Nathan ^a
5	0.019 ± 0.001	NA	0.023 ± 0.001	NA	0.003 ± 0.000	NA
10	0.031 ± 0.001	0.028	0.039 ± 0.004	0.037	0.005 ± 0.000	0.005
20	0.049 ± 0.004	0.043	0.064 ± 0.003	0.058	0.011 ± 0.001	0.009
60	0.089 ± 0.004	0.077	0.125 ± 0.006	0.110	0.032 ± 0.001	0.024
100	0.119 ± 0.005	0.101	0.169 ± 0.008	0.147	0.054 ± 0.004	0.040
200	0.184 ± 0.008	0.152	0.246 ± 0.011	0.214	0.112 ± 0.005	0.081
260	0.217 ± 0.010	NA	0.280 ± 0.013	NA	0.146 ± 0.007	NA
300	0.238 ± 0.011	0.196	0.301 ± 0.013	0.262	0.169 ± 0.008	0.123
350	0.262 ± 0.012	NA	0.325 ± 0.015	NA	0.198 ± 0.009	NA
500	0.330 ± 0.015	0.271	0.388 ± 0.017	0.336	0.283 ± 0.013	0.208
1000	0.493 ± 0.022	0.413	0.541 ± 0.024	0.470	0.507 ± 0.023	0.397

^a estimated using dose point kernel by [30]

^{NA} data not available

The results show reasonable agreement for grains of smaller sizes but as the grain size increases, the deviation increases in a random manner. A maximum deviation of 28% was seen for the ^{40}K self-dose value of the 0.2 mm grain. The results by Nathan [30] were based on the updated DPK method. It is not quite clear why our results do not agree with these results within error limits. However, just like the case for quartz, Nathan [30] mentioned in his work that the smoothing and interpolation of the monoenergetic DPK to form composite DPK

introduces systematic errors to their result. See Nathan [30] for more details. Additionally, it was not clear from the work by Nathan [30] how the dose point kernels of Al_2O_3 were determined. If it was by scaling the DPKs of quartz, then one can argue that this might have introduced additional systematic errors to their result.

6.3.4 Alpha self-dose values

6.3.4.1 Calculation for quartz

Alpha radiation has a much shorter range than beta radiation. An example model for α -self dose calculation reported in Martin et al. [31] is reproduced with some modifications. The model geometry is presented in Figure 6.6. The reproduction of this model is only for comparison, and for testing the reliability of alpha dose rate modelling for grains using *DosiVox* and *DosiSed*. The model in Martin et al. [31] is composed of a single grain surrounded by a uniform clay matrix with dimensions greater than the maximum range of alpha particles, to ensure that infinite matrix conditions are satisfied. The clay is composed of 55% in mass of SiO_2 , 35% of Al_2O_3 , and 10% of Fe_2O_3 , with a density of 1.8 g/cm^3 [197]. The grain material is quartz with a density of 2.65 g/cm^3 . The matrix is represented as a radioactive spherical shell whose internal radius is equal to the grain radius. The external radius is equal to the sum of the grain radius and 1.2 times the maximum range of alpha particles. The grain itself is non-radioactive (see Figure 6.6a). The alpha particles were simulated from the U- and Th-series in secular equilibrium, in other words, the radionuclides are decaying at the same rate at which they are being produced. The "alpha attenuation factors" were calculated as the dose deposited in the grain divided by the infinite matrix dose for $5 \text{ }\mu\text{m}$ to $400 \text{ }\mu\text{m}$ quartz grains.

In this work, the grain was modelled as a radioactive sphere within a non-radioactive cubic clay matrix (see Figures 6.6b and 6.7. for the model geometry). Radiation was emitted from the grain to obtain the self-dose of the grain. The self-dose values were calculated by

dividing the absorbed dose by the infinite matrix dose from the grain. This was repeated for 5 runs, each run being a simulation of between 1 million to 1 billion alpha particles, and the average was calculated. Attenuation factors were calculated from the self-dose values using the superposition theorem (i.e., $1-f$; see section 6.3.1). The diameters of the grains considered range from 1 to 1000 μm . The obtained α -dose attenuation factors were then plotted in Figure 6.8 for Th and Figure 6.9 for U in comparison to the α -dose attenuation factors reported by Martin et al. [31] and Bell [32].

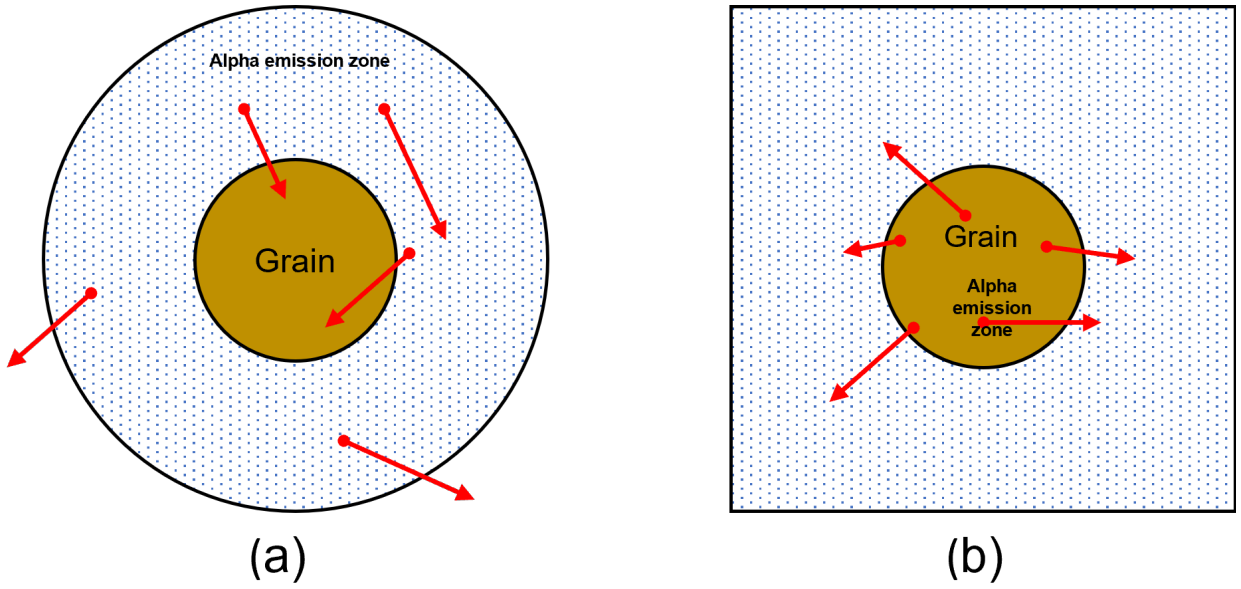


Figure 6.6: Simulation geometry of the pottery sherd model used to simulate alpha (a) attenuation factors [31] and (b) self-dose values (this work).

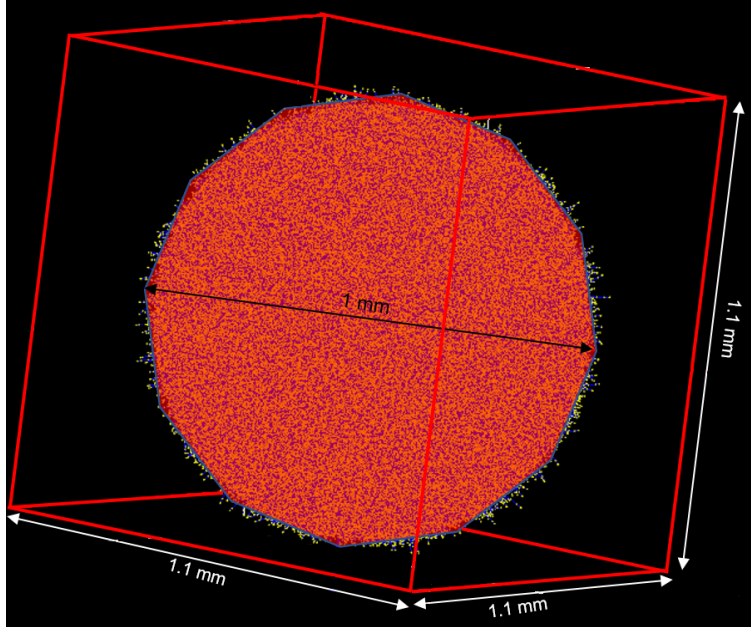


Figure 6.7: Example model for calculating α self-dose values (U-series). A 1 mm diameter grain is enclosed in a 1.1 mm cubic voxel.

The values calculated with our model were very reproducible irrespective of the grain size as can be seen from the associated error which was estimated as twice the standard deviation of the 5 runs because it was too small and because Martin et al. [31] also reported his error as twice the standard deviation. In our simulation, there was no need to scale the size of the cubic matrix to account for the range of alpha particles, in order to ensure IM conditions. This is because the grain is both the emitter and the dosimeter. This reduced the simulation time significantly without compromising the accuracy of the result. It must be stated that this approach can only be applied when simulating self-dose. On the other hand, if it were the attenuation factor that was simulated as was done in Martin et al. [31], then the size of the voxel would have to be adjusted accordingly based on the maximum range of alpha particles in order to satisfy IM conditions.

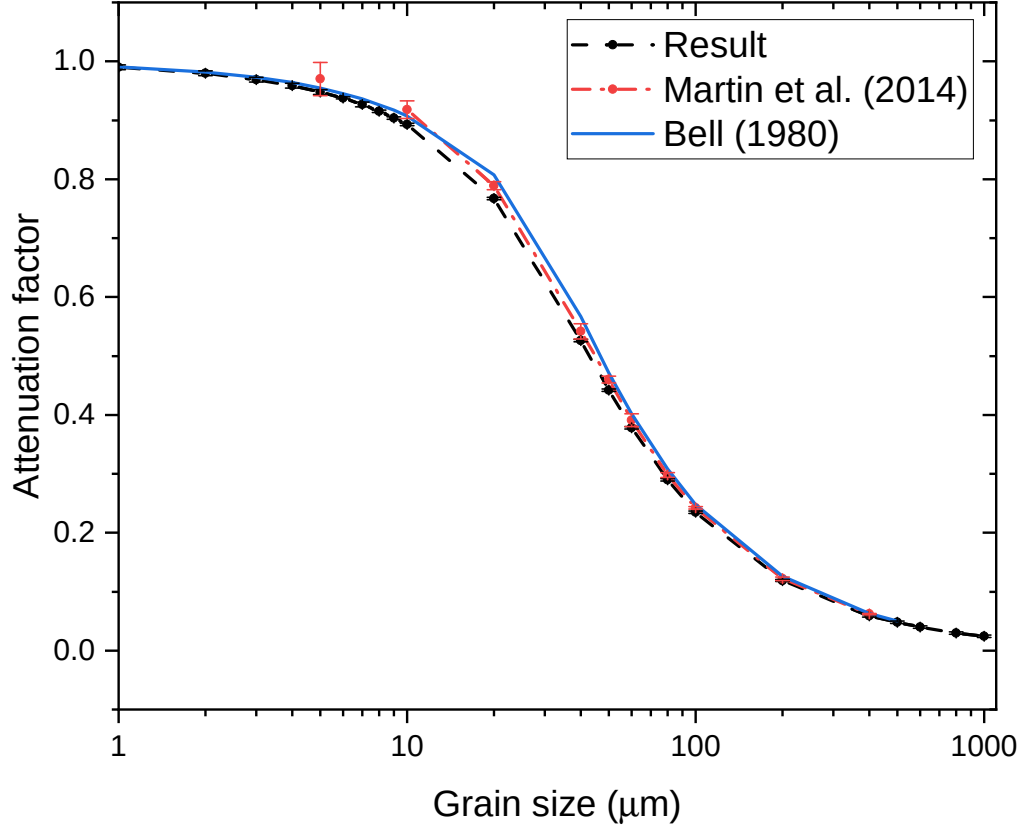


Figure 6.8: Alpha attenuation factors for a spherical quartz grain in an infinite clay matrix from Th-series in secular equilibrium.

These attenuation factors calculated by Martin et al. [31] and Bell [32] were also converted to self-dose factors and compared to the results obtained in this work (see Table 6.3). The self-dose values increase with grain sizes, approaching unity as the grain size becomes 1 mm. The range of alpha particles in quartz is $\sim 20 \mu\text{m}$, so that for $40 \mu\text{m}$ grains, the self-dose factor should be around 50%. This is in agreement with the self-dose factors for $40 \mu\text{m}$. The values cannot actually reach 100% because alpha particles that are emitted near the surface of the grain will still deposit some of their dose outside the grain.

For both the U-series and Th-series, agreement is better for larger grains than for smaller grains. This might be due to the difference in the models used. In Martin et al. [31], the α -radiation was emitted from a spherical shell bounding the spherical grain. The accuracy and precision of the simulation depends strongly on the probability of an alpha particle striking the grain. This probability is higher for larger grains compared to smaller grains.

Consequently, for their model, the result for the larger grains are more precise and accurate. For this work, α -radiation was emitted randomly from any position within the grain, including positions at the center of the grain up to positions near the surface of the grain.

These two different approaches might be the cause of the slight differences in self-dose factor. The simulation results also agree with Bell (1980) especially for the smallest and largest grains. However, there seem to be significant differences for grains with intermediate diameters. This might be due to an update of the physics models used for describing the interaction of α -particles with matter. Furthermore, there might be a difference in the stopping power used by Bell in his calculation compared to that used by Geant4 [31]. These trends are also presented visually in Figure 6.8 and Figure 6.9. Note that the figures show attenuation factors (not self-dose values) as a function of grain diameters.

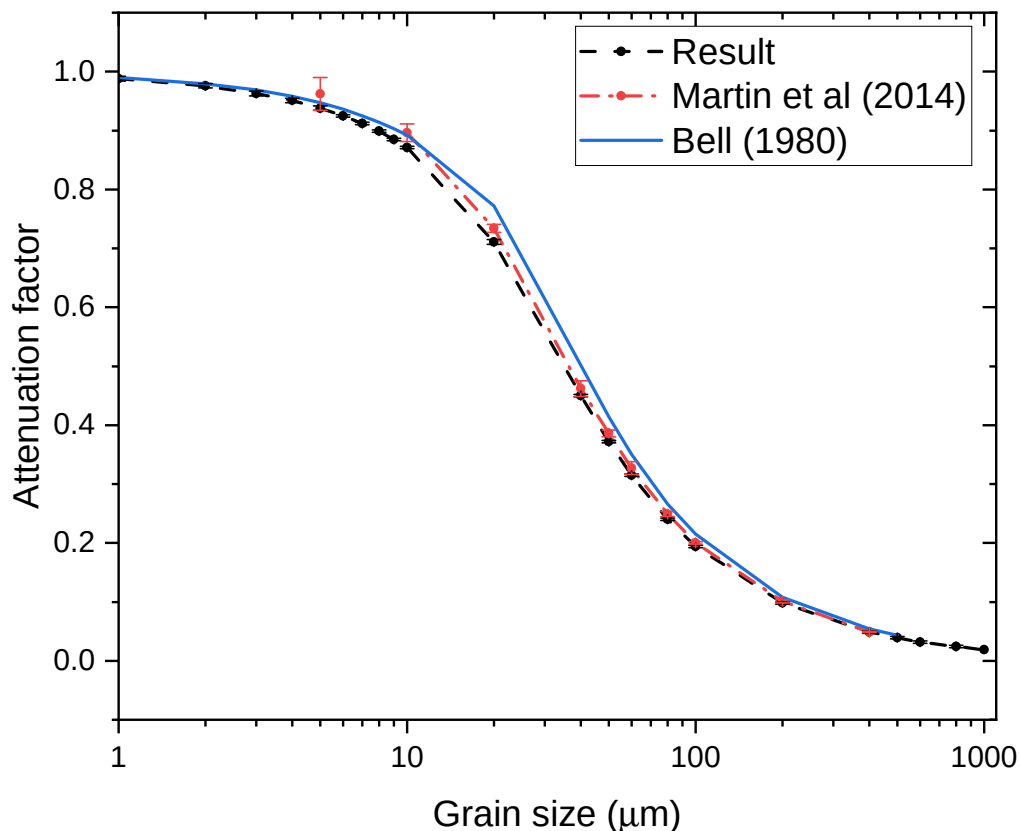


Figure 6.9: Alpha attenuation factors for spherical quartz grain in an infinite clay matrix from U-series in secular equilibrium.

Table 6.3: Alpha self-dose values (f) calculated for spherical quartz grains of different sizes. The internal radionuclide concentrations were 1% for K, 1 ppm for Th and 1 ppm for U. The results are compared with values reported in the literature.

Grain size (μm)	<i>Result</i>	Uranium series		<i>Result</i>	Thorium series	
		Martin et. al.(2014) ^a	Bell (1980)		Martin et. al.(2014) ^a	Bell (1980)
1	0.012 ± 0.004	NA	0.010	0.010 ± 0.004	NA	0.009
2	0.024 ± 0.004	NA	0.021	0.020 ± 0.004	NA	0.018
3	0.037 ± 0.004	NA	0.031	0.031 ± 0.004	NA	0.027
4	0.049 ± 0.004	NA	0.042	0.041 ± 0.004	NA	0.036
5	0.062 ± 0.004	0.038 ± 0.028	0.053	0.052 ± 0.004	0.030 ± 0.028	0.045
6	0.075 ± 0.002	NA	0.064	0.062 ± 0.002	NA	0.055
7	0.088 ± 0.002	NA	0.075	0.073 ± 0.004	NA	0.064
8	0.101 ± 0.002	NA	0.086	0.085 ± 0.002	NA	0.074
9	0.115 ± 0.002	NA	0.097	0.096 ± 0.002	NA	0.083
10	0.129 ± 0.002	0.104 ± 0.015	0.108	0.107 ± 0.002	0.082 ± 0.015	0.093
20	0.289 ± 0.004	0.266 ± 0.007	0.228	0.233 ± 0.002	0.211 ± 0.007	0.193
40	0.550 ± 0.002	0.538 ± 0.013	0.498	0.474 ± 0.002	0.458 ± 0.013	0.433
50	0.628 ± 0.002	0.614 ± 0.006	0.586	0.558 ± 0.002	0.540 ± 0.006	0.529
60	0.685 ± 0.002	0.673 ± 0.011	0.650	0.622 ± 0.002	0.609 ± 0.011	0.598
80	0.760 ± 0.002	0.751 ± 0.004	0.734	0.710 ± 0.002	0.702 ± 0.004	0.692
100	0.806 ± 0.002	0.800 ± 0.002	0.785	0.765 ± 0.002	0.758 ± 0.002	0.752
200	0.902 ± 0.002	0.899 ± 0.003	0.892	0.881 ± 0.002	0.878 ± 0.003	0.874
400	0.951 ± 0.002	0.951 ± 0.001	0.946	0.941 ± 0.002	0.938 ± 0.001	0.937
500	0.961 ± 0.002	NA	0.957	0.952 ± 0.002	NA	0.950
600	0.968 ± 0.002	NA	NA	0.960 ± 0.002	NA	NA
800	0.976 ± 0.002	NA	NA	0.970 ± 0.002	NA	NA
1000	0.981 ± 0.000	NA	0.978	0.976 ± 0.002	NA	0.975

^a calculated from attenuation factors obtained using a different Geant4 code

^b calculated from attenuation factors obtained theoretically

^{NA} data not available

6.3.4.2 alpha self-dose value calculation for $\text{Al}_2\text{O}_3\text{:C}$

A Similar calculation for α self-dose factors was performed for spherical Al_2O_3 grains of different sizes in a homogeneous matrix of Al_2O_3 . This result is presented in Table 6.4. The uncertainty listed is the standard deviation of self-dose values from 5 runs, each run being a simulation of 10 million particles.

Table 6.4: Alpha self-dose values (f) calculated for spherical Al_2O_3 grains of different sizes. The internal radionuclide concentrations were 1 ppm for Th and 1 ppm for U.

Grain size (μm)	Uranium series	Thorium series
5	0.093 ± 0.004	0.077 ± 0.004
10	0.199 ± 0.004	0.164 ± 0.004
20	0.432 ± 0.002	0.355 ± 0.004
60	0.781 ± 0.002	0.734 ± 0.002
100	0.867 ± 0.002	0.838 ± 0.002
200	0.934 ± 0.002	0.919 ± 0.002
250	0.947 ± 0.002	0.936 ± 0.002
260	0.949 ± 0.002	0.938 ± 0.002
300	0.956 ± 0.002	0.946 ± 0.002
350	0.962 ± 0.002	0.954 ± 0.002
500	0.974 ± 0.002	0.968 ± 0.002
1000	0.987 ± 0.002	0.984 ± 0.002

In Table 6.4, the self-dose value increases with grain sizes as expected, approaching unity as the grain size becomes 1 mm. The range of alpha particles in Al_2O_3 should be less than quartz, i.e., less than 20 μm . This implies that the denser Al_2O_3 will have a higher self-dose for the same grain size. This can be seen clearly when Table 6.4 is compared to Table 6.3.

6.4 Summary

In this chapter, we have been able to show that *DosiVox* and *DosiSed* are reliable Monte Carlo simulation tools for modelling radiation distribution in non-homogeneous sedimentary environments. In order to produce results with minimal uncertainty, 10^7 or more particles must be simulated. The uncertainty decreases with increasing number of simulated particles.

However, the number of particles that can be simulated is limited by time. All results compared to tabulated data shows good agreement. In this chapter, new results like the α - and β -dose rate attenuation factors for Al_2O_3 are considered a contribution to knowledge.

The characterization of reliability and precision of the Monte Carlo simulations with *DosiVox* and *DosiSed* presented in this chapter is only a preliminary step. A second and arguably more important step will be comparing the results of *DosiVox* and *DosiSed* obtained in complex sedimentary cases with experimental measurements. This will require the use of detectors for experimental dose measurement. Therefore any such comparison will require careful uncertainty analysis for both the simulation and experimental dose determination. This arduous but interesting task will be tackled in the next chapter.

Chapter 7

Validating the Monte Carlo models: Comparing Measured and Modelled Dose Distributions

7.1 Introduction

One of the major objectives of this dissertation is to calculate the microscopic dose rate distribution in the Martian subsurface. Presently, computer modelling represents the best possible method to estimate dose rates on the Martian subsurface given that direct measurement is not possible. However, a drawback to this solution for dose rate determination on a planet like Mars is the question of how to validate the result, since direct samples from Mars are not yet available on Earth. In this work, we seek to validate our Martian result by first performing what we want to do for Mars on terrestrial samples. In other words, if we can show that experimental and model result agrees for terrestrial samples, we can make a case for the validity of our Martian simulation result.

The reliability and precision of our MC codes were already tested with some basic tests, by comparing our results to results from other work (see Chapter [6](#)). The conclusion was,

our MC codes are reliable and produce precise results. These were however very simple models. Therefore, further verification of the reliability and precision of our codes in complex terrestrial sedimentary environments, i.e., non-uniform sediments is still required to justify its use to simulate the environment on Mars.

To do this, dose distributions were measured directly in well-defined samples and compared to modelled or simulated dose distributions of the same samples. We mixed samples using carefully selected minerals of known grain sizes, chemical compositions, and radionuclide concentrations. We measured the dose distributions in the samples via dosimeters buried in them and then compared the experimental dose distribution to a simulated dose distribution obtained from modelling the samples using our MC codes. This chapter presents the detailed description of the materials and methods, as well as the results and discussions of this crucial validation part of the work.

7.2 Description of Samples

To compare measured results with simulations, suitable samples had to be devised. The following considerations were taken into account:

1. Sample must be non-uniform to obtain a distribution. This is achieved by using different minerals and grain sizes.
2. Sample must be simple enough for easy modelling. The mineral diversity of each sample was limited to a maximum of 3 minerals. The grain size distribution was kept fairly large ($\geq 45 \mu\text{m}$). Modelling grains $\leq 45 \mu\text{m}$ can cause a significant increase in simulation time.
3. Each sample must contain only one radioactive mineral for easy representation of radionuclide concentrations in the model

Based on these considerations, samples that comprise an inert matrix and radioactive grains

were created.

The samples are mixtures of Thorite, fluorapatite, or Thorianite grains and fine quartz powder with weight fractions ranging from 6–26% and ~ 40 –80% respectively (see Table 7.1 & 7.2). The Thorium, Uranium and for fluorapatite also, Potassium bearing minerals provide radiation hotspots for β - and α -radiation while the quartz is inert and serves as matrix. The quartz consisted of 99.9% pure SiO_2 powder ($< 2 \mu\text{m}$) purchased from Alfa Aesar. The concentrations of radioelements in the minerals were obtained via gamma spectrometry and are presented in Table 7.3. The type and weight (in%) of radioactive grains in each sample were carefully selected to study the effect of "hot" and "cold" samples. Samples with Thorite and Thorianite are considered "hot" compared to samples with fluorapatite. Additionally, the mass of radioemitters in each sample was varied to account for low and high dose rate. About 1.8–30 cm^3 of the different mixtures were packed into separate cylindrical containers in the laboratory.

$\text{Al}_2\text{O}_3\text{:C}$ Luxel[®] were either placed on top of the mixture in the container or buried at some depth in the mixture to ensure electron equilibrium for the dosimeters. Luxel[®] was used to measure β - radiation as a way of testing for dose uniformity since Luxel[®] is made up of aggregated grains. Other containers included the regular 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains to measure alpha and beta radiation or the α -desensitized 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains to measure beta radiation only. The gamma radiation from the small sample volumes was assumed to be uniform and thus obtained together with the background radiation in the room via control samples consisting of pure quartz.

The naming convention utilized in Table 7.1 is as follows: Samples with names that have "GS" are samples with $\text{Al}_2\text{O}_3\text{:C}$ grains as dosimeters. Samples with names that have "LS" are samples with Luxel[®] as dosimeters. Samples with names that have "IM" are infinite matrix samples and were packed in the cylindrical containers shown in Figure 7.1, with internal diameter (32.2 ± 0.2) mm and height (37.5 ± 0.2) mm. The dosimeters were buried far enough from the surfaces to ensure charged particle equilibrium for the dosimeters in every

direction. The other samples qualify as "non-infinite matrix samples". In the "non-infinite matrix" samples, the Luxel[®] were placed directly on top of the mixture in the container. Al₂O₃:C grains were initially mixed with samples in the smaller cylindrical containers shown in Figure 7.2, with internal diameter (26.4 ± 0.1) mm and height (12.0 ± 0.1) mm. Samples were filled in the container to a height of 3–9 mm. These small heights are expected to result in the loss of charged particle equilibrium in the z-direction, so that these samples are also termed non-infinite matrix samples.

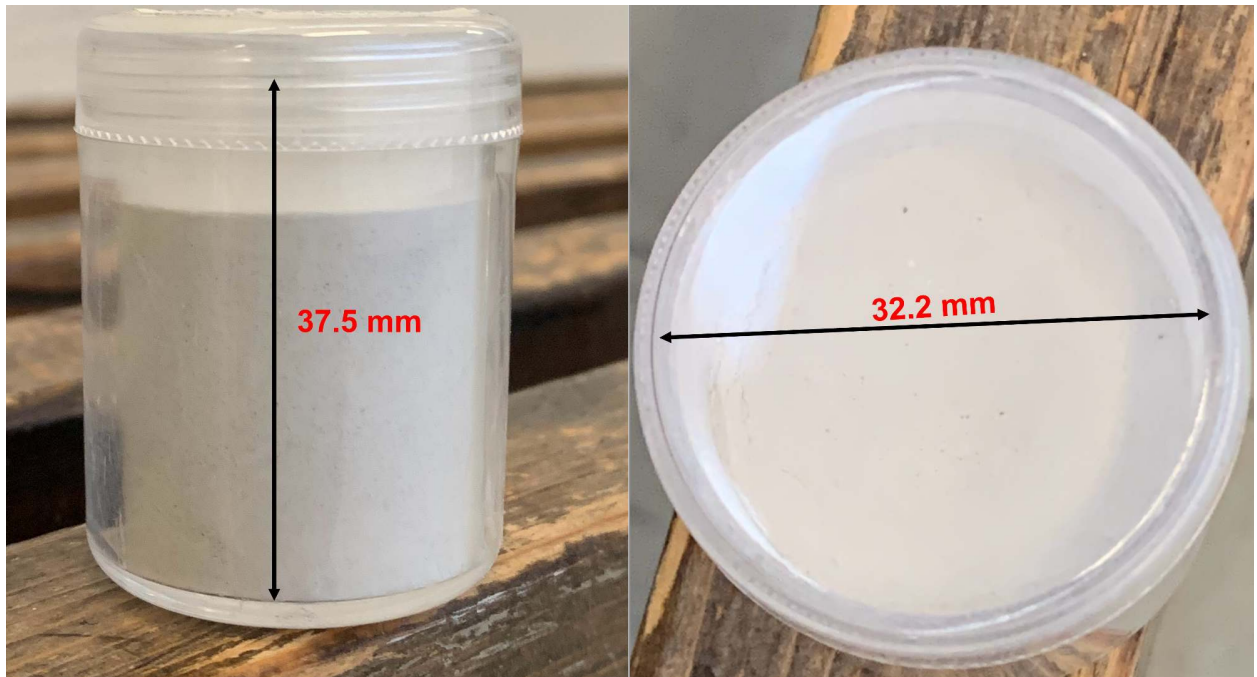


Figure 7.1: The infinite matrix sample container with internal diameter of about 32.2 mm and height of about 37.5 mm without the cap.



Figure 7.2: The non-infinite matrix sample container with internal diameter of about 26 mm and height of about 12 mm without the cap. Most of the samples in the container had heights of ~ 3 –9 mm.

Table 7.1: Proportion (in wt.%) of each material used to produce samples

	Thorite	Thorianite	Fluorapatite	Luxel [®]	Al ₂ O ₃ :C	Quartz
IMGS010	6.9				1.2	91.9
IMGS011	7.0				1.2 ^a	91.8
IMLS020		18.8		0.3		80.9
GS100	5.7				10.9	83.4
GS101	8.5				10.8 ^a	80.7
GS120	20.8				9.9	69.3
GS121	21.2				11.8 ^a	67.0
GS200		10.6			11.7	77.7
GS201		11.5			6.7 ^a	81.9
GS220		30.7			6.7	62.6
GS221		37.0			6.1 ^a	56.9
GS300			10.3		11.9	77.8
GS301			11.0		13.0 ^a	76.0
GS320			28.1		8.5	63.4
GS321			26.0		10.7 ^a	63.3
LS610	6.5			3.4		90.1

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Table 7.1: – continued from previous page

	Thorite	Thorianite	Fluorapatite	Luxel [®]	Al ₂ O ₃ :C	Quartz
LS612	28.6			2.5		68.9
LS620		18.6		0.5		80.9
LS622		41.2		2.0		56.8
LS630			15.8	2.9		81.3
LS632			41.0	2.0		57.0

^a α -desensitized Al₂O₃:C grains

Table 7.2: Physical Properties of simulants

	Volume (cm ³)	Bulk density (g cm ⁻³)	Particle density (g cm ⁻³)	Porosity (%)
IMGS010	30.54	1.18	2.76	57.3
IMGS011	30.54	1.20	3.01	56.3
IMLS020	25.42	1.55	3.07	49.3
GS100	1.99	1.20	2.83	57.8
GS101	1.65	1.37	2.88	52.5
GS120	1.85	1.48	3.07	52.0
GS121	1.82	1.51	3.10	51.3
GS200	1.71	1.28	3.00	57.2
GS201	1.70	1.31	2.96	55.7
GS220	1.65	1.74	3.51	50.4
GS221	1.63	1.72	3.73	53.9
GS300	1.59	1.31	2.81	53.3
GS301	1.89	1.29	2.82	54.3
GS320	2.35	1.33	2.86	53.6
GS321	2.09	1.46	2.87	49.2
LS610	1.71	1.23	2.74	55.3
LS612	1.79	1.93	3.82	48.2
LS620	1.89	1.55	3.07	49.3
LS622	1.79	1.93	3.82	49.0
LS630	1.72	1.40	2.70	48.6
LS632	2.04	1.70	2.84	39.9

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Table 7.2: – continued from previous page

Volume (cm ³)	Bulk density (g cm ⁻³)	Particle density (g cm ⁻³)	Porosity (%)
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In Table 7.2, the bulk density (in g cm⁻³), which is sometimes defined as the degree of compaction in a sample was calculated as the total mass of the dry minerals in the sample divided by the volume occupied by the sample (this volume includes air-gaps). The mass of each mineral in the sample was measured in *grams* using a digital precision balance and summed to get the total mass. The volume occupied by the sample was calculated geometrically using the formula for the volume of a cylinder ($V = \pi r^2 h$), where h is the height of the sample in the container and r is the internal radius of the container, calculated as half the internal diameter of the container. The particle density was calculated as the total mass of the dry minerals divided by the total volume of the dry minerals in the sample. The volume of each dry mineral in a sample was individually calculated by dividing the mass of the mineral in the sample by the density of the mineral (i.e., $V = m/\rho$). The total volume of the dry minerals in a sample is thus the sum of the volume of all the dry minerals (i.e., $V = \sum_{i=1}^n V_i$). The porosity (in %) is the fraction of the total volume that represents the pore space. For a dry sample, the pore space represents the volume occupied by air, i.e., the air-gaps. It was calculated as 1 minus the ratio of the bulk density and particle density; porosity = 1-(Bulk density/Particle density).

7.3 Methods

7.3.1 Experimental dose measurement

Prior to "burial", Luxel[®] and dosimeter grains were reset by bleaching or heating as described in Chapter 5. The samples with the dosimeters were stored for a period of 5–8 months.

Table 7.3: Radioisotope concentrations in the various radioactive minerals that were used for the samples

Material	Uranium (ppm)	Thorium (ppm)	Potassium (%)
Thorite	$(486.0 \pm 3.5)\text{E3}$	$(23.1 \pm 3.8)\text{E3}$	
Thorianite	$(36.59 \pm 0.28)\text{E3}$	$(323.2 \pm 4.2)\text{E3}$	
Apatite	$(1.65 \pm 0.02)\text{E2}$	$(33.57 \pm 0.71)\text{E2}$	3.57 ± 0.31

During this time, the buried dosimeters were exposed to radiation from the radionuclides in the samples. After the storage period, the Luxel[®] dosimeters were retrieved from the container, cleaned with distilled water and placed on the sample cups for measurement. The grains were recovered by sieving, since the dosimeter grains had grain sizes distinctly different from the matrix and the radioactive minerals. The individual grains were picked with a needle and placed on the sample cups using silicon oil. The signals were measured using the procedures in Table 7.4 and following the methodologies discussed in Chapter 5. The S/S_T values were then compared to the respective dose responses (Chapter 5, sections 5.4.7 and 5.5.4) to obtain dose values.

Table 7.4: Complete measurement protocols for dosimeters buried in samples

General measurement protocol for Luxel®
Retrieve dosimeter from sample after storage, wash with distilled water and/or clean and dry
OSL readout to obtain S
irradiate dosimeter with 2s β -irradiation (test-dose)
OSL readout to obtain S_T
calculate S/S_T
General measurement protocol for grains
Retrieve grains from sample after storage, wash with distilled water and/or clean and dry
2 TL readouts to measure signal, then background
irradiate grains with a dose equivalent to 5s β -radiation (test-dose)
2 TL readouts to measure signal, then background
calculate test-dose corrected signal

7.3.2 Modelling sample and dosimeter geometry

Even though the samples represent relatively simple mineral mixtures, there are various approaches for modelling the dose rate distribution using *DosiVox* and/or *DosiSed*. In *DosiVox/DosiSed*, a sample is divided into a voxelized grid with dimensions the size of the sample in the container. In our case, the sample container is cylindrical. However, in *DosiVox* or *DosiSed*, the voxels are rectangular or more specifically parallelepiped in shape. Therefore, the samples were approximated with models of different levels of complexities to characterize the dependence of the result on the size and geometry of the sample in the container and size and shape of the dosimeter. The various approaches considered and the assumptions made in the construction of these models are listed in detail in Appendix D.

7.3.3 Grain modeling of minerals in sediment

For both *DosiVox* and *DosiSed*, one can either represent the sediment as uniform materials of different compositions [155, 186, 187] or as grains of different minerals [31, 155, 185, 186]. The grain model is preferred for heterogeneous samples. The dose absorbed by a grain depends on the energy emitted in its environment. The radiation often has to pass through other grains before reaching a grain of interest. The attenuation coefficients vary between minerals, so that the energy arriving at the grain position depends on its neighbors. In uniform materials these effects disappear. For grain modeling, the software uses the homogeneous sphere representation, which is considered one of the most effective ways of studying dose rate distributions in sedimentary grains from a statistical standpoint [30]. In *DosiVox*, spherical grains of specific minerals are randomly packed into voxels for which the minerals were defined. For *DosiSed*, spherical grains are randomly packed into a sedimentary volume based on the composition of minerals present in the volume. Even though the grains for our samples were sieved, they still contain a whole distribution of grain sizes. For example, the dosimeters were sieved with a 250 μm and a 355 μm sieve, and any grain size in between these values can likely be found in the sample. It is impossible to model every grain size in the sample due to time constraints and computational limitation. Therefore, some grain sizes in the sample were selected to be representative of the grain size distribution in samples. See Table 7.5 for the real grain size distribution and the simulated grain sizes for each sample. For each sample, the simulated grain sizes were equally distributed. Additionally, *DosiSed* was used for modelling the samples with $\text{Al}_2\text{O}_3\text{:C}$ grains as dosimeters. Since the grain packing model involves creation of grains only, the cylindrical sample geometry was approximated with the equivalent perimeter model (EP) or the Equivalent Surface Area-to-Volume Ratio (ESAVR) model. Details of the models are described in Appendix D. Figure 7.3 shows an example of the grain packing model used in *DosiSed*.

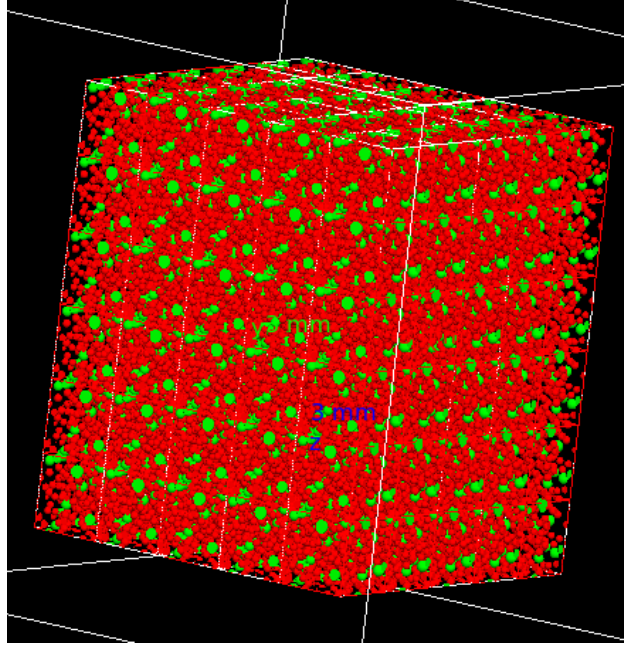


Figure 7.3: Example of Equivalent perimeter (EP) model used in *DosiSed* for sedimentary grain modeling.

Table 7.5: Grain size distribution approximation for Models

Sample (s)	Real grain size (μm)	Modelled grain size (μm)	Mineral grains
IMGS010, IMGS011	45 – 250	45, 63, 90, 125, 150, 180, 212, 250	Thorite
IMLS020	45 – 250	45, 63, 90, 125, 150, 180, 212, 250	Thorianite
GS100, GS101	> 125 – 150	130, 150	Thorite
GS120, GS121	> 63 – 90	70, 90	Thorite
GS200, GS201	> 125 – 150	130, 150	Thorianite
GS220, GS221	> 90 – 125	100, 125	Thorianite
GS300, GS301	> 150 – 180	160, 180	Apatite
GS320, GS321	> 180 – 212	190, 212	Apatite
LS610, LS612	> 150 – 180	160, 180	Thorite
LS620	45 – 250	45, 125, 250	Thorianite
LS622	> 250 – 355	260	Thorianite
LS630, LS632	> 250 – 355	260	Apatite
Grain dosimeters	> 250 – 355	260, 300, 350	$\text{Al}_2\text{O}_3\text{:C}$

7.3.4 Simulation of radiation in models

Each radiation-type (α , β , and γ) emitted from the radioactive grains or uniform material was simulated separately. This is repeated for each radio-element (Th, U, and ^{40}K) in the minerals of the sample (See Table 7.3). The final dose rate distribution of the detectors in the sample is the sum of all the dose rate contributions (Eq. 6.1 & Eq. 6.2) per radiation-type per radio-element. In order to be able to accurately sum the dose rates/particle/radio-element for each detector, all simulations were done with a fixed geometry per model so that the detector position remains fixed over simulations of different particles and multiple runs. As a result of this, only the emission of particles was left random.

7.3.5 Correction for alpha efficiency

The regular $\text{Al}_2\text{O}_3\text{:C}$ grains absorb alpha, beta and gamma radiation. In luminescence dosimetry, the measured signal (or luminescence) from the dosimeter is considered proportional to the absorbed radiation (see Chapter 3). So that the dose measured is called the equivalent dose and not the absorbed dose. It is the dose that is equivalent to the signal, not necessarily to the absorbed dose. Alpha radiation, unlike beta and gamma has high ionization density, which can lead to saturation of the trapping centers in the dosimeter, consequently resulting in luminescence signals that are not proportional to the energy deposited in the material [198]. In other words, if the same dose of alpha, beta and gamma radiation is administered to this dosimeter, alpha will result in much less signal compared to beta and gamma. This phenomenon, expressed as alpha efficiency or a -value has been thoroughly investigated for $\text{Al}_2\text{O}_3\text{:C}$ in the following references [199–202].

For a mixed radiation field with each radiation-type delivering the same dose, the measured luminescence signal will be due to a combination of alpha, beta, and gamma doses. The signal contribution to the total signal, from each radiation-type will depend on the a -value.

For example, consider a mixed radiation field of 10 Gy beta and 10 Gy alpha radiation. If the 10 Gy beta results in a signal of 1000 counts, then the 10 Gy alpha will result in $(1000 \times a\text{-value})$ counts. For an a -value of 0.1, the 10 Gy alpha signal is only 100 counts. The total signal measured for the total dose of 20 Gy will be 1100 counts instead of 2000 counts, the contribution from each radiation-type is unknown. For an unknown dose from a mixed field with a signal equal to 1100 counts, the measured signal is compared to a dose response. This will result in an equivalent dose of 11 Gy, not 20 Gy.

The simulation on the other hand, uses a dose scoring system, such that the doses deposited in a detector volume (dosimeter) are tallied within the volume, until the simulations of all charged particles in the system are completed. This method of dose computation does not account for saturation effects in the scoring volume. In this work, the dose from alpha and beta radiation are simulated separately and then added up later. The absorbed dose due to beta only from the simulation should agree with the equivalent dose from measurement. The combined alpha and beta absorbed dose from the simulation, on the other hand, will not agree with the combined equivalent dose measured. Since nothing can be done to remove this effect from the measurement, we are left with the option of correcting the simulated distribution. This is performed by first multiplying the alpha dose distribution from the simulation with the a -value, before adding it to the beta dose distribution to obtain the combined dose distribution that will agree with the measured equivalent dose distribution.

7.4 Experimental and simulated gamma uniformity for the dosimeters

The simplest method for comparing measured and modelled dose distributions uses a uniform radiation field. Thus, an investigation was designed to measure the uniformity of Al_2O_3 dosimeters when they are uniformly irradiated with gamma radiation from a 662 keV ^{137}Cs gamma source. A cylindrical ceramic (Al_2O_3) sample container was filled with 250–355 μm

$\text{Al}_2\text{O}_3\text{:C}$ grains and positioned 35 cm from the source. The air-gaps in the container were filled with Al_2O_3 powder. The Al_2O_3 container serves as build-up for the radiation beam and the Al_2O_3 powder ensures charged particle equilibrium in the container so that all the $\text{Al}_2\text{O}_3\text{:C}$ grains will receive a uniform dose. See also Appendix A for more details. After irradiation with a 60.64 Gy dose, the grains were measured using TL to obtain the measured dose distribution. A model of the investigation was also created and the simulated dose distributions of the grains with respect to different numbers of simulated particles were also obtained. The experimentally measured and simulated dose distributions are compared in Figure 7.4.

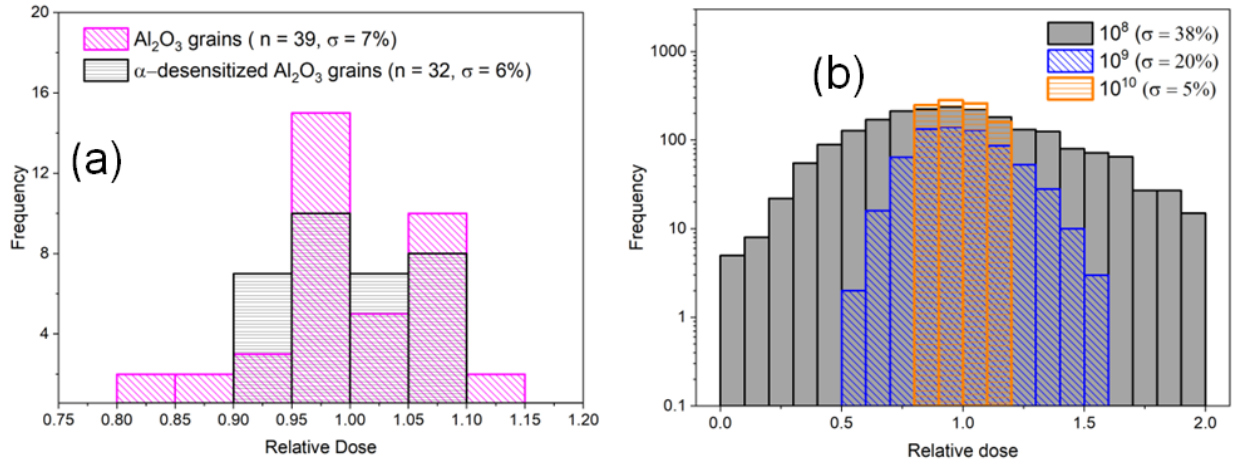


Figure 7.4: (a) Dose distribution of 39 $\text{Al}_2\text{O}_3\text{:C}$ and 32 α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ grains measured after gamma irradiation and normalized to the mean value. The estimated statistical uncertainty of individual measurements was 3-4% (b) Effect of the number of simulated gamma particles on the dose distribution of 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains irradiated using gamma radiation from a 662 keV ^{137}Cs gamma source.

Standard deviations of 7% and 6% of the mean doses were observed for the regular $\text{Al}_2\text{O}_3\text{:C}$ and α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ grains respectively. This result is only approximately 2 times higher than the expected distribution based on 3–4% TL measurement uncertainty. Unlike α - and β -radiation, the measured dose for gamma radiation should be independent of grain size and thus present a uniform distribution dominated only by measurement uncertainties. The additional uncertainties observed might be due to perturbation of the secondary electron equilibrium due to any remaining air gaps in the container. It could also be due to the small

number of grains studied. The result reported here is better than similar results of gamma irradiation of Al_2O_3 grains by Kalchgruber et al. [183], where a standard deviation of 20% of the mean dose was observed. Their broad distribution is probably due to the use of radiation cross-talk as test dose during readout, which is highly dependent on the exact positioning of the grain on the measurement cup. A deviation of 30–80% from expected values has been reported for recovered doses when cross-talk is used for test dose during readout [169].

To define a uniformity baseline for our dosimeters, the contribution of the measurement uncertainties (3–4%) was removed from the experimental standard deviation of the gamma irradiated dosimeters (6–7%) using the error propagation formula in Eq. 7.1, resulting in a standard deviation of about 5.5%. This 5.5% will be used as our uniformity baseline for the dosimeters. Comparing this to our simulated dose distribution, we see that it takes a simulation of 10^{10} particles to achieve the experimental uniformity baseline. A possible explanation for this is that the container is tiny compared to the entire solid angle around the source. In the model, the container is ~ 1.0 cm in diameter and the source is 35 cm from the container, same as the experimental setup. Only a fraction of the photons hits the container. The fraction of photons hitting the container can be estimated by calculating the geometric efficiency of a detector of radius ~ 0.5 cm positioned 35 cm from a source. The geometric efficiency is ~ 0.005 %. So that even with 10^{10} particles, only about 5×10^5 particles hit the container.

$$\delta Uniformity = \sqrt{(\delta Experimental \ SD)^2 - (\delta Measurement \ SD)^2} \quad (7.1)$$

where SD means standard deviation or uncertainty.

7.5 Comparison of measured and simulated doses for the Luxel dosimeters

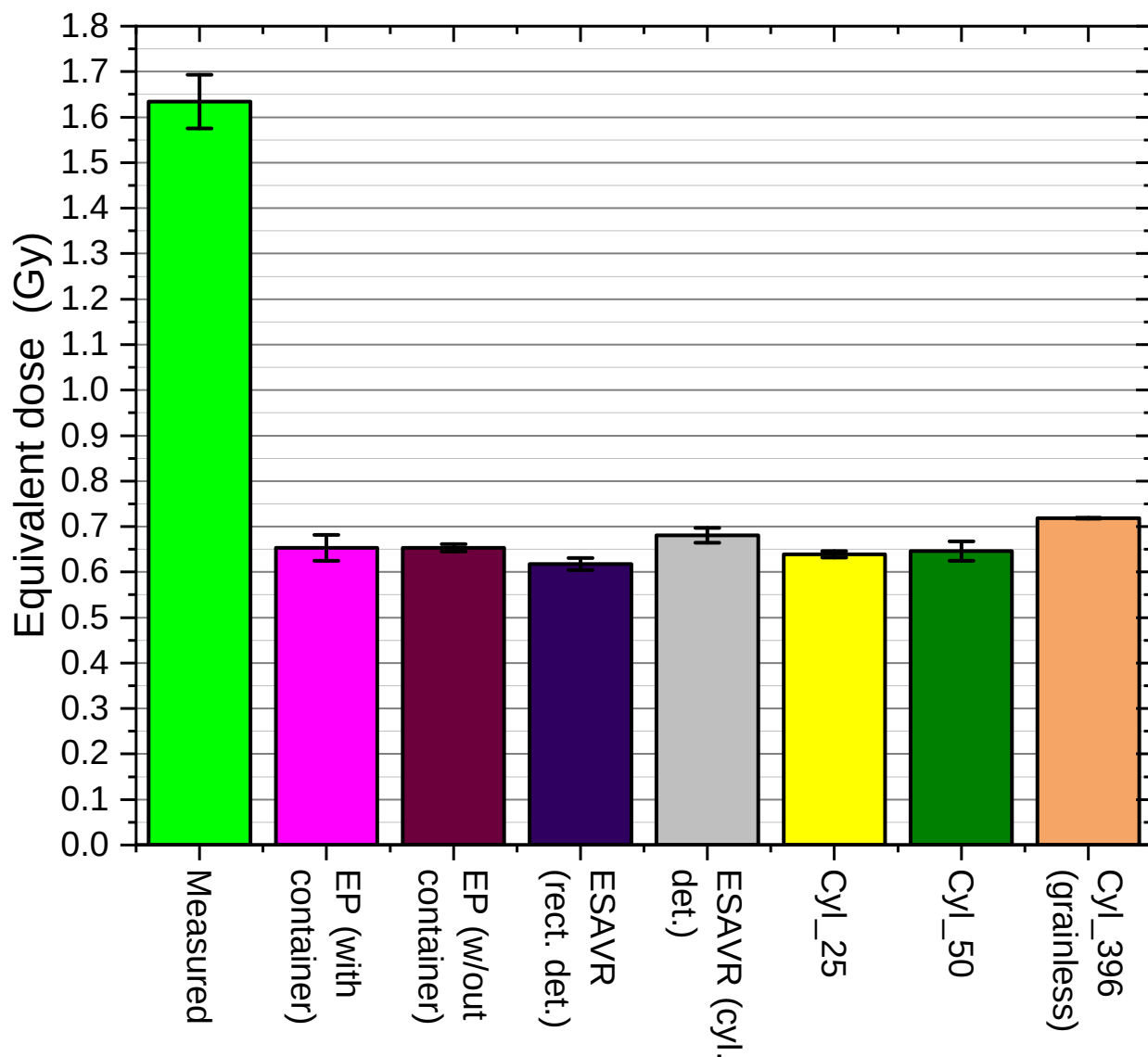


Figure 7.5: Beta equivalent dose of the infinite matrix sample IMLS020 measured with Luxel[®] dosimeters. For the experimental values the error weighted mean of 4 dosimeters and the associated uncertainty was calculated. The uncertainties are 1σ values and include measurement errors. The uncertainties of the equivalent doses from the models also represent the error of the weighted mean of 4 dosimeters (1σ) and include the uncertainty due to the numbers of simulated particles.

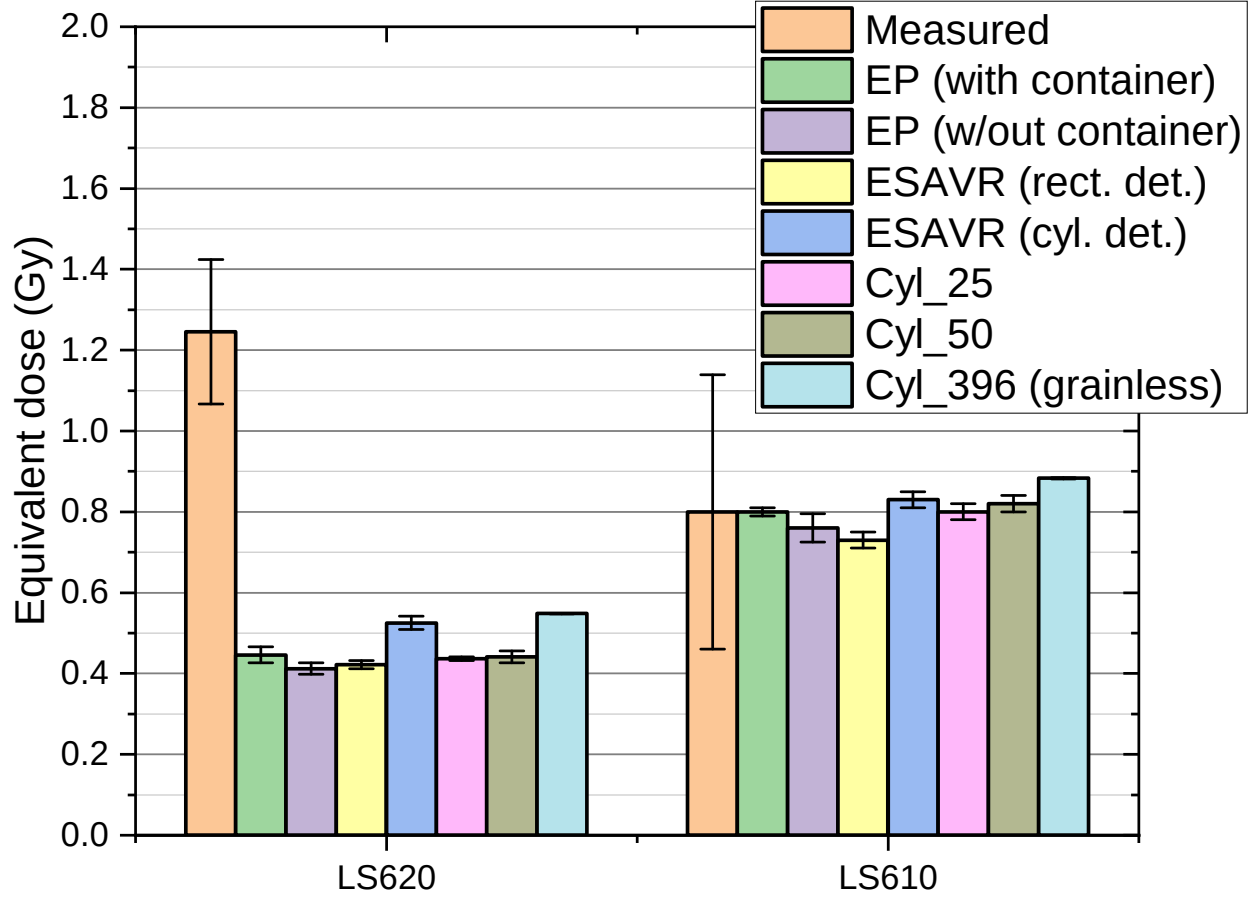


Figure 7.6: Beta equivalent doses for non-infinite matrix samples measured with Luxel[®] dosimeters. The samples were considered low dose rate (low proportions of radioemitter). For the experimental values the error weighted mean of 5 dosimeters and the associated uncertainty was calculated. The uncertainties are 1σ values and include measurement errors. The uncertainties of the equivalent doses from the models also represent the error of the weighted mean of 5 dosimeters (1σ) and include the uncertainty due to the numbers of simulated particles. For a description of the various simulation models see Appendix D.

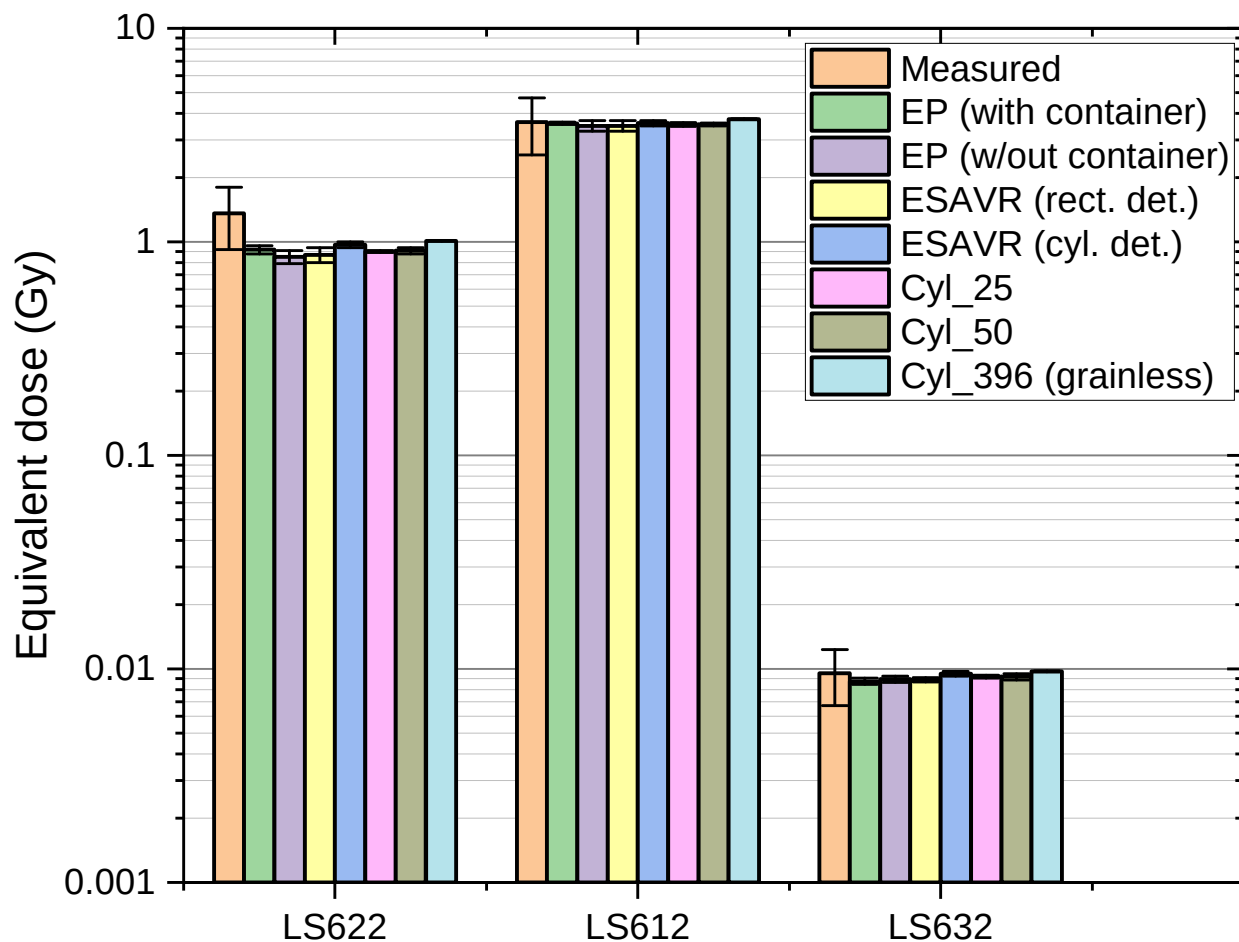


Figure 7.7: Beta equivalent doses for non-infinite matrix samples measured with Luxel[®] dosimeters. The samples were considered high dose rate (high proportions of radioemitter). For the experimental values the error weighted mean of 5 dosimeters and the associated uncertainty was calculated. The uncertainties are 1σ values and include measurement errors. The uncertainties of the equivalent doses from the models also represent the error of the weighted mean of 5 dosimeters (1σ) and include the uncertainty due to the numbers of simulated particles.

The Luxel[®] dosimeters are large and contain hundreds of individual dosimeter grains. During measurement the signal from the entire dosimeter is recorded, which represents an average of the doses absorbed by all grains in the dosimeter. Thus, little variation is expected between the dosimeters. For each sample 4 or 5 dosimeters were measured and their error weighted mean and the associated uncertainty were calculated. The contribution from cosmic and γ were measured via separate dosimeters in control samples and subtracted from the measured equivalent dose. For the simulated containers, 4 or 5 dosimeters were simulated and their

error weighted mean and the associated uncertainty were also calculated.

Figures 7.5, 7.6, and 7.7 show the comparison between experimentally measured doses from the Luxel[®] dosimeters and the dose values calculated from simulations. To enable this comparison, the equivalent doses of the dosimeters (after background subtraction) were assumed to be exclusively due to β -radiation from the radioemitters in the sample. The simulation models are described in Appendix D.

The laboratory measured equivalent doses agree with the simulated doses within the 1σ uncertainty margin for all samples except IMLS020, LS620, and LS622 where the measured dose is about 1.5–3 times more than the simulated dose. These three samples all contain the same radioemitter, thorianite, which leads us to suspect that the differences in the measured and simulated equivalent doses might be due to a systematic error. The thorianite minerals used in the samples were assumed to be pure and no analysis was done to verify this. The grains were obtained from a small hand-specimen that was crushed and sieved and which showed different coloration. It is not clear that the entire specimen consisted of thorianite. Any impurities in the thorianite that might have changed its density or elemental composition will result in a systematic error in the simulated result since the density and elemental composition were specified based on theoretical values in the model. The measured results would indicate that the grains had a higher amount of radionuclides than obtained with gamma spectrometry from a subsample. The various simulation models all show good agreement and are consistent.

The large error bars on the measured equivalent doses for the non-infinite matrix samples can be attributed to loss of electron equilibrium in those samples. Although Monte Carlo simulation can model dose in build-up areas, the dose is strongly location-dependent and thus any slight difference in the location of the dosimeters might result in a widely different dose. It must be stated that it was impossible to know the exact location of the dosimeters on top the mixture or how much shift might have occurred during handling and/or storage prior to measurement. These effects are not reflected in the simulation, where the dosimeters

remained in a fixed position.

Lastly, using the dose rate conversion factors in Guérin et al. [29] and calculating the dose rate in the samples from the radionuclide concentrations in Table 7.3, the infinite matrix (IM) dose rate in each sample was found. On multiplying the dose rate by the exposure duration (burial time) for each dosimeter, the IM dose absorbed by each dosimeter was determined. These infinite matrix doses represent the theoretical maximum dose the dosimeters can absorb in the sample, i.e., assuming no loss of electron equilibrium, and no build-up, nor build-down of dose due to the quartz matrix. These infinite matrix doses are presented in Table 7.6, along with the simulated and measured doses. From the table, the measured and simulated doses were either less or close to the IM dose, except for the measured doses of the samples with systematic errors. In general, the equivalent doses of the non-infinite matrix samples were less than the IM dose, while the equivalent dose of the infinite matrix sample was roughly equal to the IM dose.

Table 7.6: Comparison of experimental and simulated beta equivalent doses for all samples with Luxel[®] dosimeters. For the experimental values the error weighted mean of 4 or 5 dosimeters and the associated uncertainty was calculated. The uncertainties are 1σ values and include measurement errors. The uncertainties of the equivalent doses from the models also represent the error of the weighted mean of 4 or 5 dosimeters (1σ) and include the uncertainty due to the numbers of simulated particles. All dose values are in the unit of Gy. IM indicates the theoretical value calculated from the nuclide concentrations.

Sample	Measured	EPWC	EPWOC	ESAVRRD	ESAVRCD	Cyl_25	Cyl_50	Cyl_396	IM dose
IMLS020	1.63 ± 0.06	0.65 ± 0.03	0.65 ± 0.01	0.62 ± 0.01	0.68 ± 0.02	0.64 ± 0.01	0.65 ± 0.02	0.718 ± 0.001	0.79 ± 0.13
LS622	1.36 ± 0.44	0.92 ± 0.04	0.85 ± 0.06	0.87 ± 0.07	0.97 ± 0.03	0.90 ± 0.01	0.91 ± 0.03	1.012 ± 0.001	1.40 ± 0.23
LS620	1.25 ± 0.18	0.45 ± 0.02	0.41 ± 0.01	0.42 ± 0.01	0.53 ± 0.02	0.436 ± 0.005	0.44 ± 0.01	0.548 ± 0.001	0.79 ± 0.13
LS612	3.63 ± 1.09	3.59 ± 0.05	3.50 ± 0.16	3.53 ± 0.17	3.60 ± 0.10	3.55 ± 0.08	3.55 ± 0.06	3.760 ± 0.02	7.95 ± 1.31
LS610	0.80 ± 0.34	0.80 ± 0.01	0.76 ± 0.04	0.73 ± 0.02	0.83 ± 0.02	0.80 ± 0.02	0.82 ± 0.02	0.883 ± 0.002	1.81 ± 0.30
LS632	0.010 ± 0.003	0.009 ± 0.000	0.009 ± 0.000	0.009 ± 0.000	0.010 ± 0.000	0.009 ± 0.000	0.009 ± 0.000	0.010 ± 0.000	0.017 ± 0.003

EPWC EP w/container , *EPWOC* EP w/out container, *ESAVRRD* ESAVR w/rectangular detector, & *ESAVRRD* ESAVR w/cylindrical detector

7.6 Comparison of experimental and simulated dose distributions for dosimeter grains

The goal of this section was to test the accuracy of microscopic dose rate modelling with *DosiSed*. Equivalent doses measured using dosimeter grains were compared to equivalent doses obtained from simulations for various samples. The results are presented here. Additionally, the use of the single grain TL measurement protocol presents an opportunity for the direct investigation of microscopic dose rate inhomogeneity in natural sediments. Spatial dose-rate variations in natural sediment can occur due to the different ranges of alpha and beta particles, grains size specific attenuation, as well as non-uniform distribution of radioactive nuclides in the sedimentary minerals. All this and other relevant implications will also be discussed.

All grain-detector simulations with *DosiSed* were performed using a rectangular container, which has the same perimeter as the measured cylindrical container (EP model, see Appendix D). Detailed sample descriptions can be found in Table 7.1, 7.2, 7.3, and 7.5.

7.6.1 Dosimeters buried in samples under infinite matrix conditions

Samples IMGS010 and IMGS011 are mixtures of thorite (ThSiO_4 , 6.9–7 wt.%; density = 5.35 g/cm³) and quartz (SiO_2 , density = 2.65 g/cm³). Two of the larger, infinite-matrix containers (Figure 7.1) were filled each with about 30.5 cm³ of the mixture. About 0.4 g of 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ and α – desensitized $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains were carefully mixed with about 36 g of sample material to obtain IMGS010 and IMGS011, respectively. The proportion of dosimeter grains compared to the sample without the dosimeter was to ensure that the grains do not influence the dose rate i.e., to create an infinite matrix condition for the dosimeters. The dosimeters were buried in the sample for a period of 5 months. Regular grains absorb both alpha and beta radiation. As described in section 7.3.5, the modelled dose distributions for the regular grains must therefore be corrected for the alpha efficiency.

Alpha-desensitized grains record beta radiation only and no such correction is necessary. For the modelled dose distribution, the number of simulated particles was 10^8 . The resulting measured and modelled dose distributions are presented in Figure 7.8 and 7.9.

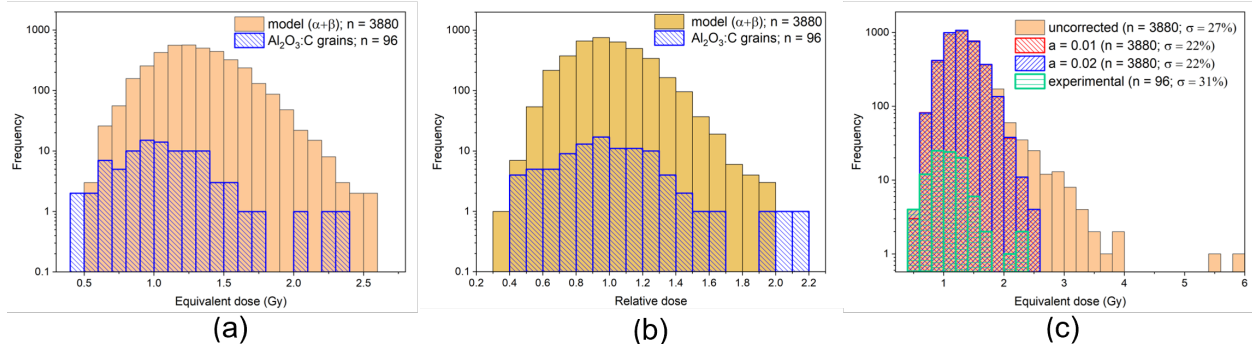


Figure 7.8: Comparison of experimental to simulated dose distributions for regular $\text{Al}_2\text{O}_3:\text{C}$ dosimeter grains (alpha + beta) buried in sample IMGS010 with 6.9% thorite by proportion. (a) Absolute equivalent dose distribution with the simulated distribution corrected for alpha efficiency using an a -value of 0.01 (b) Same as (a), with each distribution normalized to its respective mean. (c) Effect of correction for alpha efficiency. For the orange distribution, the alpha and beta doses of each grain were added and no further correction was applied. The alpha components of the red and blue distributions were corrected for alpha efficiency with two different a -values. The estimated uncertainty of individual measurements was 3–7 %. The uncertainty of the simulated equivalent dose per dosimeter is about 10%. Therefore, a bin size of 0.1 was selected for easy comparison of the distributions.

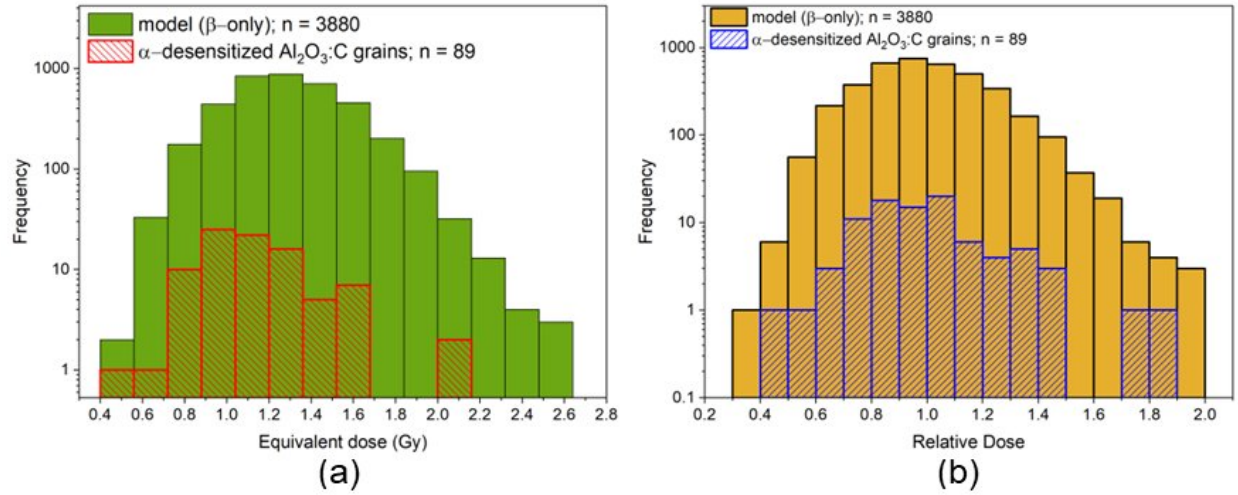


Figure 7.9: Comparison of measured to simulated dose distributions for α -desensitized $\text{Al}_2\text{O}_3:\text{C}$ dosimeter grains buried in sample IMGS011 with 7.0% thorite by proportion. (a) Absolute equivalent dose distribution (b) Same as (a), with each distribution normalized to its respective mean. The simulated distribution represents β -particles only. The estimated uncertainty of individual measurements was 5–12 %. The uncertainty of the simulated equivalent dose per dosimeter is about 10%. Therefore, a bin size of 0.16 was selected for easy comparison of the distributions.

At first glance, it seems like the experimental equivalent dose distribution of the regular $\text{Al}_2\text{O}_3:\text{C}$ grains (Figure 7.8a) is slightly shifted to the left (towards lower dose) compared to the simulated dose distribution. However, after normalizing the dose distributions to its respective mean values (mean is represented as 1) for better visualization of the spread (Figure 7.8b), we see that the two distributions have similar shape. On the other hand, the equivalent dose distribution of the α -desensitized $\text{Al}_2\text{O}_3:\text{C}$ grains (Figure 7.9a) shows very good agreement with the simulated dose distribution. For the $\text{Al}_2\text{O}_3:\text{C}$ dosimeter grains, the mean dose for the 96 measured grains is (1.09 ± 0.34) Gy, and (1.30 ± 0.28) Gy for the 3880 simulated grains. For the α -desensitized $\text{Al}_2\text{O}_3:\text{C}$ dosimeter grains, the mean dose for the 89 measured grains is (1.13 ± 0.27) Gy, and (1.30 ± 0.28) Gy for the 3880 simulated grains. The overlap in margin of error also confirms that the distributions are the same. The measured and simulated doses also agree with the expected infinite matrix doses: $\sim (1.22 \pm 0.20)$ Gy and $\sim (1.19 \pm 0.20)$ Gy for the regular and desensitized grains respectively. These IM doses were calculated directly from dose rate conversion factors and the radionuclide

concentrations. They represent theoretical maximum doses in the samples.

Furthermore, the measured and simulated relative distributions were compared using the Mann-Whitney non-parametric test. The result shows that the two distributions are NOT significantly different at the 0.05 level, with a p-value of 0.588 and 0.647 for Figures 7.8b and 7.9b respectively. The standard deviations of the experimental and simulated dose distributions of the α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ grains are also very close at 24% and 22% respectively. The only small difference in the dose distributions can be seen for the regular grains when we compare the standard deviations of the experimental dose distribution (31%) to that of the simulated dose distribution (22%). Interestingly, the standard deviation of the uncorrected simulated dose distribution for the regular grains (7.8c) is closer to that of the experimental dose distribution at 27%. Which suggests that the highlighted discrepancy might be due to over-correction of the alpha part of the simulated dose distribution because of the really small a-values used. An a-value of 0.01 [183] or 0.02 [202] completely removes the contribution of the alpha radiation. This is reflected in the observation that there is no difference between the mean doses and standard deviations of the simulated a-value corrected $\alpha+\beta$ -dose distribution and the β -only dose distribution. Finally, the slight increase in the standard deviation of the experimental dose distribution (24%) compared to the simulated beta-only distribution (22%) for the alpha-desensitized grains can be explained by the effect observed in section 5.5.2, that the alpha-desensitized grains are NOT 100% insensitive to alpha radiation, i.e., they have a very minimal sensitivity to alpha radiation that varies from one grain to another. A summary of the result is presented in Table 7.7.

Table 7.7: Comparison of dose distributions for samples IMGS010 and IMGS011

	Experimental	Simulation	
		<i>a-value</i> = 0.01 ^b , 0.02 ^b	uncorrected
Sample IMGS010; Dosimeter: regular Al₂O₃:C; Radiation: $\alpha+\beta$			
Mean (Gy)	1.09 ± 0.34	1.30 ± 0.28	1.35 ± 0.37
Standard deviation (%)	31	22	27
Dose Rate Inhomogeneity (%)	30.2	21.6	26.6
Sample IMGS011; Dosimeter: α-desensitized Al₂O₃:C; Radiation: β only			
Mean (Gy)	1.13 ± 0.27		1.30 ± 0.28
Standard deviation (%)	24		22
Dose Rate Inhomogeneity (%)	23.0		21.6

^a Kalchgruber et al. 2003 [183]^b Osvey & Ranogajec-Komor, 1999 [202]

Additionally, microscopic dose rate variation or dose rate inhomogeneity in a sedimentary environment can be due to the inhomogeneous distribution of radionuclides in the sediment, different ranges of alpha and beta particles, attenuation of beta particles and grain size variation. Therefore an attempt was made to estimate the portion of the observed distributions that is due to dose-rate inhomogeneity in the samples measured. To do this, one must first realize that our experimental dose distribution is a combination of dose rate inhomogeneity and measurement uncertainties, while our simulated dose distribution is a combination of dose rate inhomogeneity and uncertainties due to the numbers of simulated particles. If we remove the contributions by measurement uncertainties for the experimental distributions and the contributions due to the numbers of particles for the simulation, we will be left with only the spread due to dose rate inhomogeneities. For example, the 31% and 24% standard deviations measured for Al₂O₃:C and α -desensitized Al₂O₃:C in the samples are due to measurement uncertainties and dose-rate in-homogeneity. If we remove the standard deviation of 6.5% measured for dosimeters in a uniform gamma radiation field, that also includes measurement uncertainties, the remaining approximately 30% and 23% must be due

to dose-rate in-homogeneity in the samples.

The 22% standard deviation simulated for both regular $\text{Al}_2\text{O}_3\text{:C}$ and α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ grains in the samples represents uncertainties due to the numbers of simulated particles and dose-rate in-homogeneity. The uncertainty associated with the 10^8 simulated particles is 4.5%, thus leaving a standard deviation of about 21.6% associated with simulated dose-rate in-homogeneity. Comparing, the experimental and simulated dose rate inhomogeneities (see Table for 7.7), we see very good agreement for α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ grains. For the regular grains, the simulated dose rate inhomogeneity is about 9% less than the experimental dose rate inhomogeneity when the distribution is corrected for alpha efficiency and about 4% less when the distribution is not corrected for alpha efficiency. Another possible reason for this discrepancy apart from the one mentioned earlier is that for the simulation, spherical grain geometry was assumed. Real Al_2O_3 grains have been reported to exhibit non-spherical shapes like oval, rectangular, and some irregular forms [203, 204]. This will ultimately affect the effective size of each grain and thus the experimental dose rate variation especially for alpha particles. There is also the statistical error contribution due to the difference in the number of simulated grains compared to the number measured. In general, the dose rate inhomogeneity which represents the microscopic variation of dose rate in the samples are between 20–30% of the mean value for 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains.

Finally, separate plots of the simulated beta-only and alpha-only (uncorrected) distributions for Al_2O_3 dosimeter grains are presented in Figure 7.10 to show how the spread of the simulated alpha-only distribution compared to the simulated beta-only distribution.

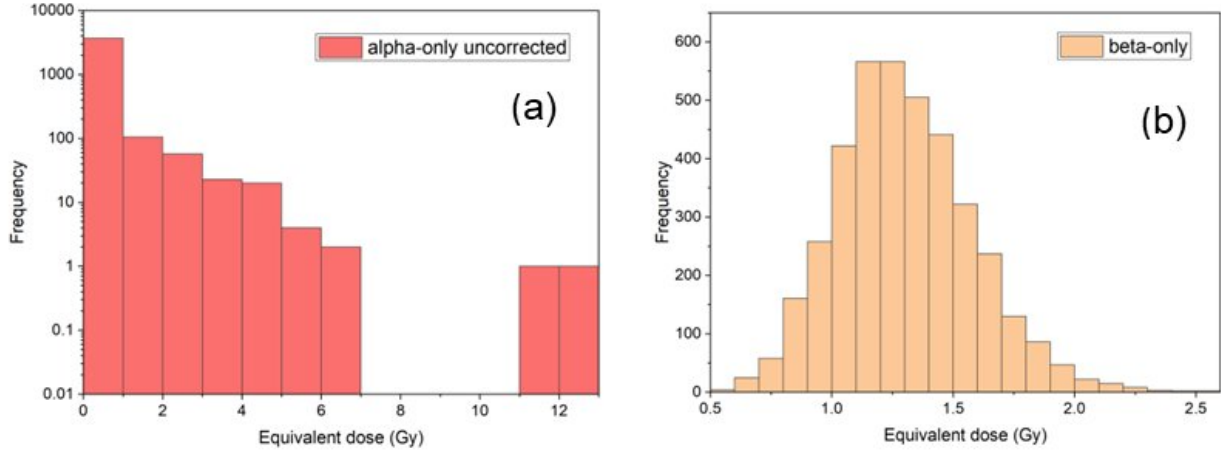


Figure 7.10: Beta-only vs. uncorrected alpha-only dose distribution for regular $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains buried in sample IMGS010, showing the spread of the two distributions.

7.6.2 Samples with small amounts of radioemitters

The measured and simulated dose distributions for the low dose rate samples i.e., samples with small amount of radioemitters will be presented here. These samples were packed into the smaller cylindrical container and the actual height of the samples in the container ranges from 0.3–0.9 cm. Thus the infinite-matrix condition is not fulfilled. The dose distributions of the individual samples are discussed here. A discussion comparing distributions from all samples can be found at the end of this chapter.

Samples GS100 and GS101

Samples GS100 and GS101 are mixtures of thorite (ThSiO_4 , density = 5.35 g/cm^3) and quartz. The specific information about samples GS100 and GS101 are presented in Table 7.8. The result of dose distribution for these samples are presented in Figure 7.11.

Table 7.8: Specific sample information for samples with small amounts of radioemitters.

Sample	Thorite wt.%	Volume (cm^3)	Fill height (cm)	Grain-type	$\text{Al}_2\text{O}_3\text{:C}$ mass (g)
GS100	5.7	1.99	0.37	regular	0.26
GS101	8.5	1.65	0.35	α - desensitized	0.27

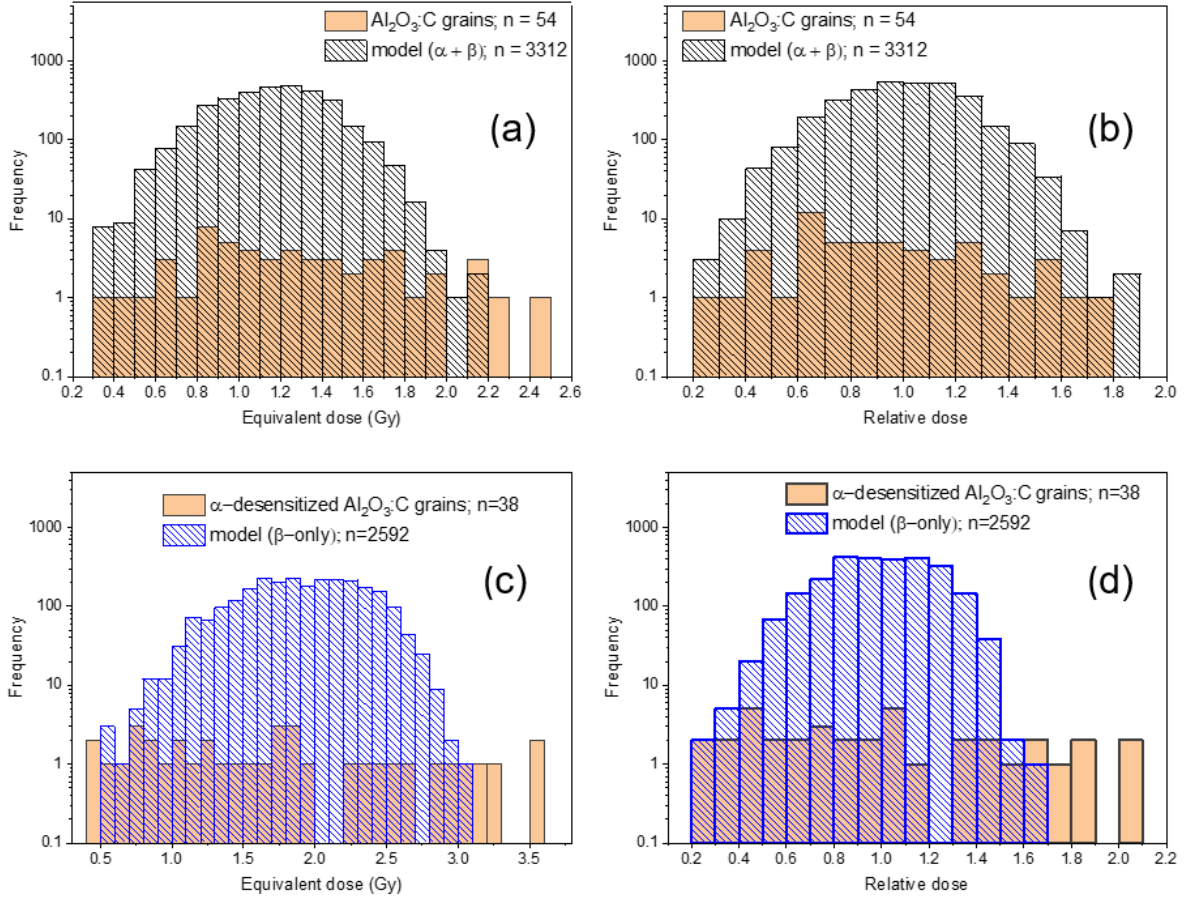


Figure 7.11: Comparison of experimental to simulated dose distribution for regular $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains in sample GS100 (a, b) and GS101 (c,d) with 5.7% and 8.5 % thorite by proportion respectively. (a) and (c) Absolute equivalent dose distributions with the simulated distributions corrected for alpha efficiency using an a -value of 0.01. (b) and (d) Same as (a) and (c) respectively, but with each distribution normalized to its respective mean. The simulated distribution represents the combined dose from $\alpha + \beta$ -particle simulations. The estimated uncertainty of individual measurements was 3–6 % for GS100 and 4–11 % for GS101. The uncertainty of the equivalent dose per dosimeter from the simulation is about 10% including the uncertainty associated with the 10^8 particles simulated. Therefore, a bin size of 0.1 was selected for easy comparison of the width of the distributions.

In Figures 7.11, the mean dose measured by the 54 regular $\text{Al}_2\text{O}_3\text{:C}$ grains is (1.27 ± 0.50) Gy, while the mean dose calculated from the 3312 grains simulation is (1.16 ± 0.27) Gy. For the α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains, the mean dose for the 38 measured grains is (1.75 ± 0.92) Gy, and (1.91 ± 0.41) Gy for the 2592 simulated grains. The overlap in margin of error confirms that the mean values of the distributions are the same. Furthermore,

the measured and simulated relative distributions were compared using the Mann-Whitney non-parametric test. The result shows that for sample GS101 with α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ grains as dosimeters, the two distributions are NOT significantly different at the 0.05 level, with a p-value of 0.344. For GS100 on the other hand, with regular $\text{Al}_2\text{O}_3\text{:C}$ grains as dosimeters, the parametric test says the two distributions are significantly different with a p-value of 0.013. The measured and simulated doses also agree with the expected infinite matrix doses: $\sim (1.67 \pm 0.28)$ Gy and $\sim (2.42 \pm 0.40)$ Gy for the regular and desensitized grains respectively. These IM doses represent theoretical maximum doses in the samples.

However, the relative standard deviation of the experimental dose distribution for sample GS101 is 53%, which is 2 times that of the simulated dose distribution at 21%. For sample GS100, the relative standard deviation of the experimental dose distribution is 39% compared to 23% for the simulated dose distribution. One possible explanation for some of the discrepancies in the standard deviations is the really small number of measured grains compared to the number of simulated grains. Statistically, the precision of a measurement increases with the sample size. Unfortunately, it was impossible to measure more grains since each grain is measured by picking it with a needle and placing in the measurement cup. This time consuming endeavor greatly limited the number of grains that can be feasibly measured. The use of a single grain measuring device would have made this less time consuming but this will require the purchase of the device, which requires money.

Samples GS200 and GS201

Samples GS200 and GS201 are mixtures of thorianite (ThO_2 , density = 9.7 g/cm^3) and quartz. The specific information about samples GS200 and GS201 are presented in Table 7.9. The result of dose distribution for these samples are presented in Figure 7.12.

Table 7.9: Specific sample information for samples with small amounts of radioemitters.

Sample	Thorianite wt. %	Volume (cm ³)	Fill height (cm)	Grain-type	Al ₂ O ₃ :C mass (g)
GS200	10.6	1.77	0.31	regular	0.26
GS201	11.5	1.70	0.31	α -desensitized	0.15

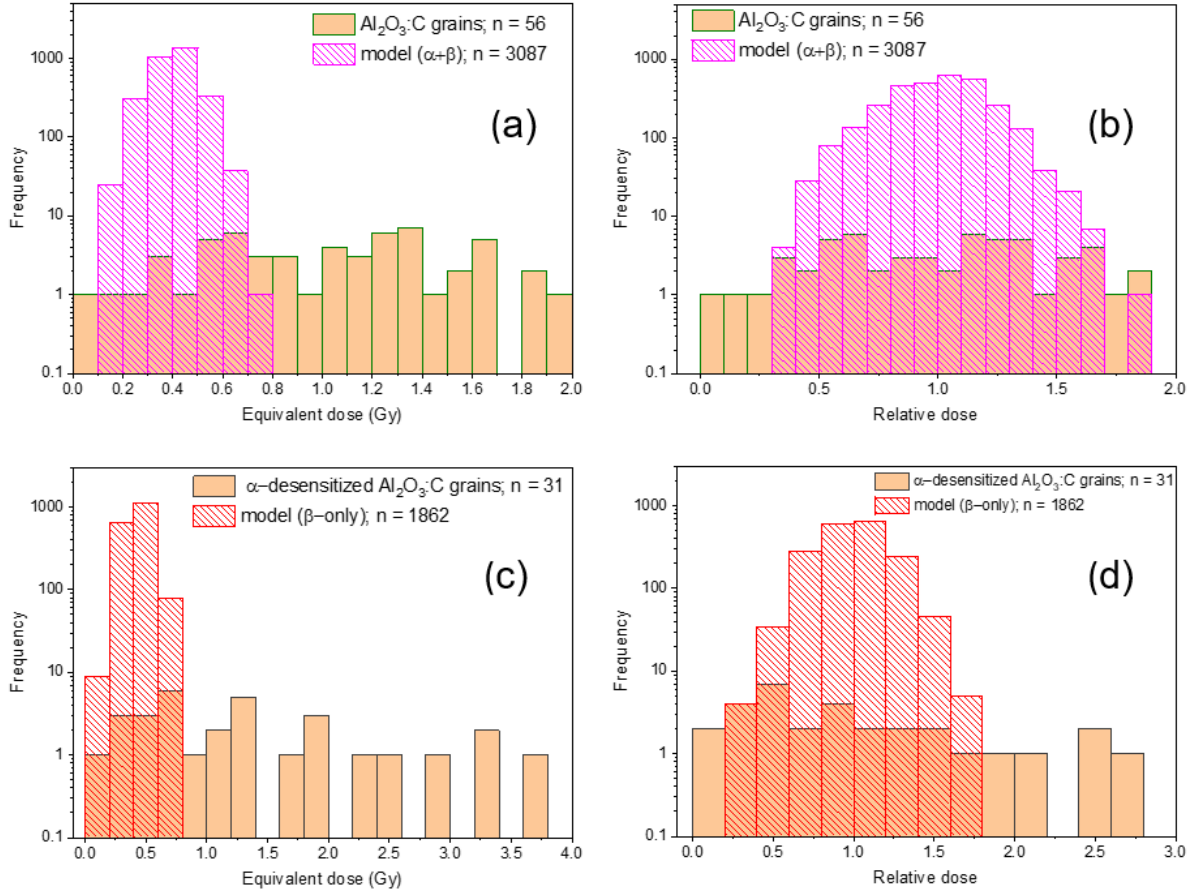


Figure 7.12: Comparison of experimental to simulated dose distribution for regular Al₂O₃:C dosimeter grains in sample GS200 (a, b) and GS201 (c,d) with 10.6% and 11.5 % thorianite by proportion respectively. (a) and (c) Absolute equivalent dose distributions with the simulated distributions corrected for alpha efficiency using an a -value of 0.01. (b) and (d) Same as (a) and (c) respectively, but with each distribution normalized to its respective mean. The simulated distribution represents the combined dose from $\alpha+\beta$ -particle simulations. The estimated uncertainty of individual measurements was 3–4 % for GS200 and 5–11 % for GS201. The uncertainty of the equivalent dose per dosimeter from the simulation is about 10% including the uncertainty associated with the 10^8 particles simulated. Therefore, a bin size of 0.1 and 0.2 was selected in (a,b) and (c,d) respectively, for easy comparison of the width of the distributions.

In Figures 7.12, the mean dose measured by the 56 regular $\text{Al}_2\text{O}_3\text{:C}$ grains is (1.03 ± 0.48) Gy, while the mean dose calculated from the 3087 grains simulation is (0.41 ± 0.08) Gy. For the α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains, the mean dose for the 31 measured grains is (1.34 ± 0.96) Gy, and (0.44 ± 0.09) Gy for the 1862 simulated grains. There was no overlap in margin of error for the regular $\text{Al}_2\text{O}_3\text{:C}$ grains. For the α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ grains, there was an overlap but the experimental mean dose is 3 times more than the mean dose from the simulation. Interestingly, the Mann-Whitney non-parametric test shows that the two distributions are NOT significantly different at the 0.05 level, with a very high p-value of 0.748 and a very low p-value of 0.075 for Figure 7.12b and 7.12d respectively.

This result at first glance seems absurd but the explanation for this is that there was a systematic error in the definition of the density and mineral compositions of the thorianite minerals used. This systematic error was already reported in section 7.5 with the Luxel[®] dosimeters where we saw also found that the measured dose were approximately 1.5–3 times the simulated dose. The relative standard deviation of the experimental dose distribution for sample GS200 is 47%, which is ~ 2 times that of the simulated dose distributions at 20%. For sample GS201, the relative standard deviation of the experimental dose distribution is 72% compared to 20% for the simulated dose distribution. The measured and simulated doses were also comparable to the expected infinite matrix doses: $\sim (0.45 \pm 0.07)$ Gy and $\sim (0.49 \pm 0.08)$ Gy for the regular and desensitized grains respectively.

Sample GS300 and GS301

Samples GS300 and GS301 are mixtures of fluorapatite ($\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$, density = 3.15 g/cm^3) and quartz. The specific information about samples GS300 and GS301 are presented in Table 7.10. The result of dose distribution for these samples are presented in Figure 7.13.

Table 7.10: Specific sample information for samples with small amounts of radioemitters.

Sample	Thorianite wt. %	Volume (cm ³)	Fill height (cm)	Grain-type	Al ₂ O ₃ :C mass (g)
GS300	10.3	1.59	0.29	regular	0.25
GS301	11.0	1.89	0.35	α -desensitized	0.32

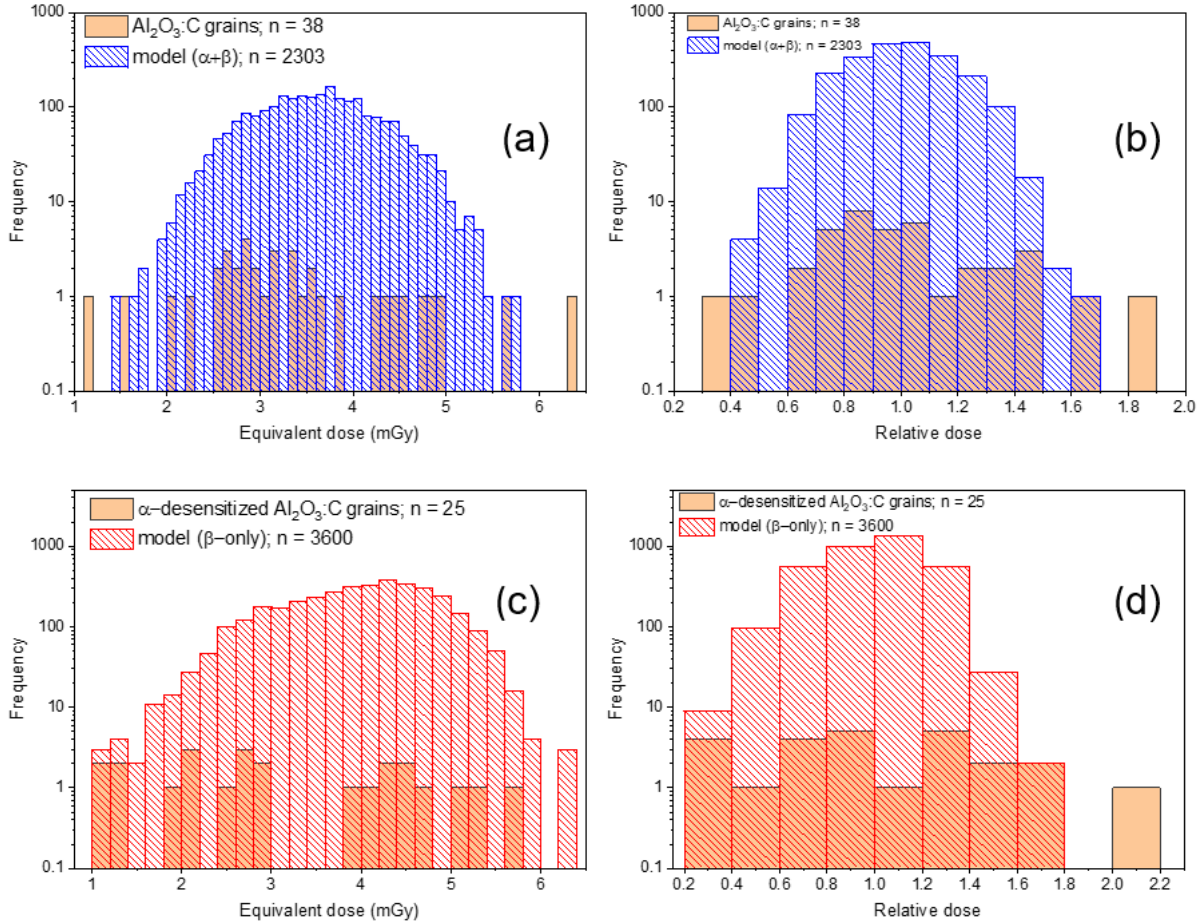


Figure 7.13: Comparison of experimental to simulated dose distribution for regular Al₂O₃:C dosimeter grains in sample GS300 (a, b) and GS301 (c,d) with 10.3% and 11.0 % fluorapatite by proportion respectively. (a) and (c) Absolute equivalent dose distributions with the simulated distributions corrected for alpha efficiency using an a -value of 0.01. (b) and (d) Same as (a) and (c) respectively, but with each distribution normalized to its respective mean. The simulated distribution represents the combined dose from $\alpha+\beta$ -particle simulations. The estimated uncertainty of individual measurements was 6–12 % for GS300 and 6–15 % for GS301. The uncertainty of the equivalent dose per dosimeter from the simulation is about 10% including the uncertainty associated with the 10^8 particles simulated. Therefore, a bin size of 0.1 and 0.2 was selected in (a,b) and (c,d) respectively, for easy comparison of the width of the distributions.

In Figures 7.13, the mean dose measured by the 38 regular $\text{Al}_2\text{O}_3\text{:C}$ grains is (3.36 ± 1.06) mGy, while the mean dose calculated from the 2303 grains simulation is (3.60 ± 0.65) mGy. For the α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains, the mean dose for the 25 measured grains is (3.32 ± 1.60) mGy, and (3.97 ± 0.81) mGy for the 3600 simulated grains. The overlap in margin of error confirms that the distributions are the same. The Mann-Whitney non-parametric test shows that the two distributions are NOT significantly different at the 0.05 level, with a p-value of 0.363 and 0.736 for Figure 7.13b and 7.13d respectively. The relative standard deviation of the experimental dose distribution for sample GS300 is 32%, which is ~ 2 times that of the simulated dose distributions at 18%. For sample GS301, the relative standard deviation of the experimental dose distribution is 48% compared to 20% for the simulated dose distribution. The measured and simulated doses are also comparable to the expected infinite matrix doses: $\sim (4.69 \pm 0.77)$ mGy and $\sim (4.93 \pm 0.81)$ mGy for the regular and desensitized grains respectively. Note that this infinite matrix doses are the theoretical maximum doses attainable by the dosimeters in these samples.

7.6.3 Samples with large amounts of radioemitters

The measured and simulated dose distributions for the high dose rate samples i.e., samples with large amounts of radioemitters will be presented here. Similar to the previous section, these samples were filled into the smaller cylindrical container and the infinite-matrix condition is not fulfilled. The dose distributions of the individual samples are discussed here.

Sample GS120 and GS121

Samples GS120 and GS121 are mixtures of thorite and quartz. The specific information about the samples are presented in Table 7.9. The result of dose distribution for these samples are presented in Figure 7.14.

Table 7.11: Specific sample information for samples with small amounts of radioemitters.

Sample	Thorianite wt. %	Volume (cm ³)	Fill height (cm)	Grain-type	Al ₂ O ₃ :C mass (g)
GS120	20.8	1.85	0.34	regular	0.27
GS121	21.2	1.82	0.33	α -desensitized	0.32

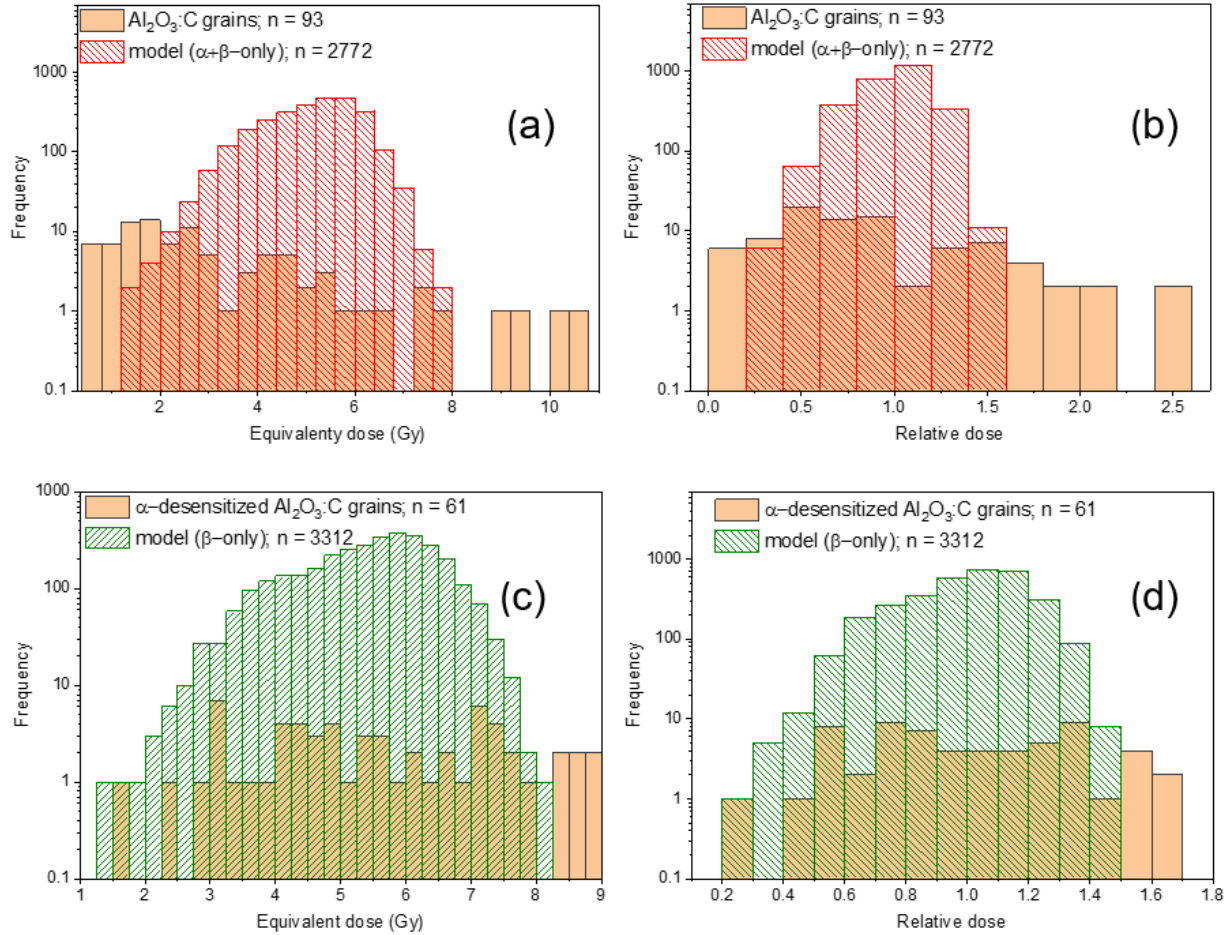


Figure 7.14: Comparison of experimental to simulated dose distribution for regular Al₂O₃:C dosimeter grains in sample GS120 (a, b) and GS121 (c, d) with 20.8% and 21.2 % thorite by proportion respectively. (a) and (c) Absolute equivalent dose distributions with the simulated distributions corrected for alpha efficiency using an α -value of 0.01. (b) and (d) Same as (a) and (c) respectively, but with each distribution normalized to its respective mean. The simulated distribution represents the combined dose from $\alpha+\beta$ -particle simulations. The estimated uncertainty of individual measurements was 3–7 % for GS120 and 4–8 % for GS121. The uncertainty of the equivalent dose per dosimeter from the simulation is about 10% including the uncertainty associated with the 10^8 particles simulated. Therefore, a bin size of 0.4 and 0.25 was selected in (a, b) and (c, d) respectively, for easy comparison of the width of the distributions.

In Figures 7.14, the mean dose measured by the 93 regular $\text{Al}_2\text{O}_3\text{:C}$ grains is (2.98 ± 2.23) Gy, while the mean dose calculated from the 2772 grains simulation is (5.08 ± 0.96) Gy. For the α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains, the mean dose for the 61 measured grains is (5.52 ± 1.87) Gy, and (5.43 ± 1.01) Gy for the 3312 simulated grains. For the α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains, the overlap in margin of error confirms that the distributions are the same. For the regular $\text{Al}_2\text{O}_3\text{:C}$ grains, there was an overlap but the error in the mean dose of the experimental dose distribution is 75%. This is very high compared to 19% for the simulated dose distribution.

Additionally, the Mann-Whitney non-parametric test shows that the experimental and simulated dose distributions for the α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains are NOT significantly different at the 0.05 level, with a very high p-value of 0.860 (see Figure 7.14d). For Figure 7.14b, the test shows that the two distributions are significantly different. The relative standard deviation of the experimental dose distribution for sample GS121 is 34%, which is ~ 2 times that of the simulated dose distributions at 19%. The measured and simulated doses also comparable to the expected infinite matrix doses: $\sim (5.70 \pm 0.94)$ Gy and $\sim (5.67 \pm 0.93)$ Gy for the regular and desensitized grains respectively. They represent theoretical maximum doses in the samples.

Sample GS220 and GS221

Samples GS220 and GS221 are mixtures of thorianite (ThO_2 , density = 9.7 g/cm^3) and quartz. The specific information about the samples are presented in Table 7.12. The result of dose distribution for these samples are presented in Figure 7.15.

Table 7.12: Specific sample information for samples with small amounts of radioemitters.

Sample	Thorianite wt. %	Volume (cm^3)	Fill height (cm)	Grain-type	$\text{Al}_2\text{O}_3\text{:C}$ mass (g)
GS220	30.7	1.65	0.30	regular	0.19
GS221	37.0	1.63	0.30	α - desensitized	0.17

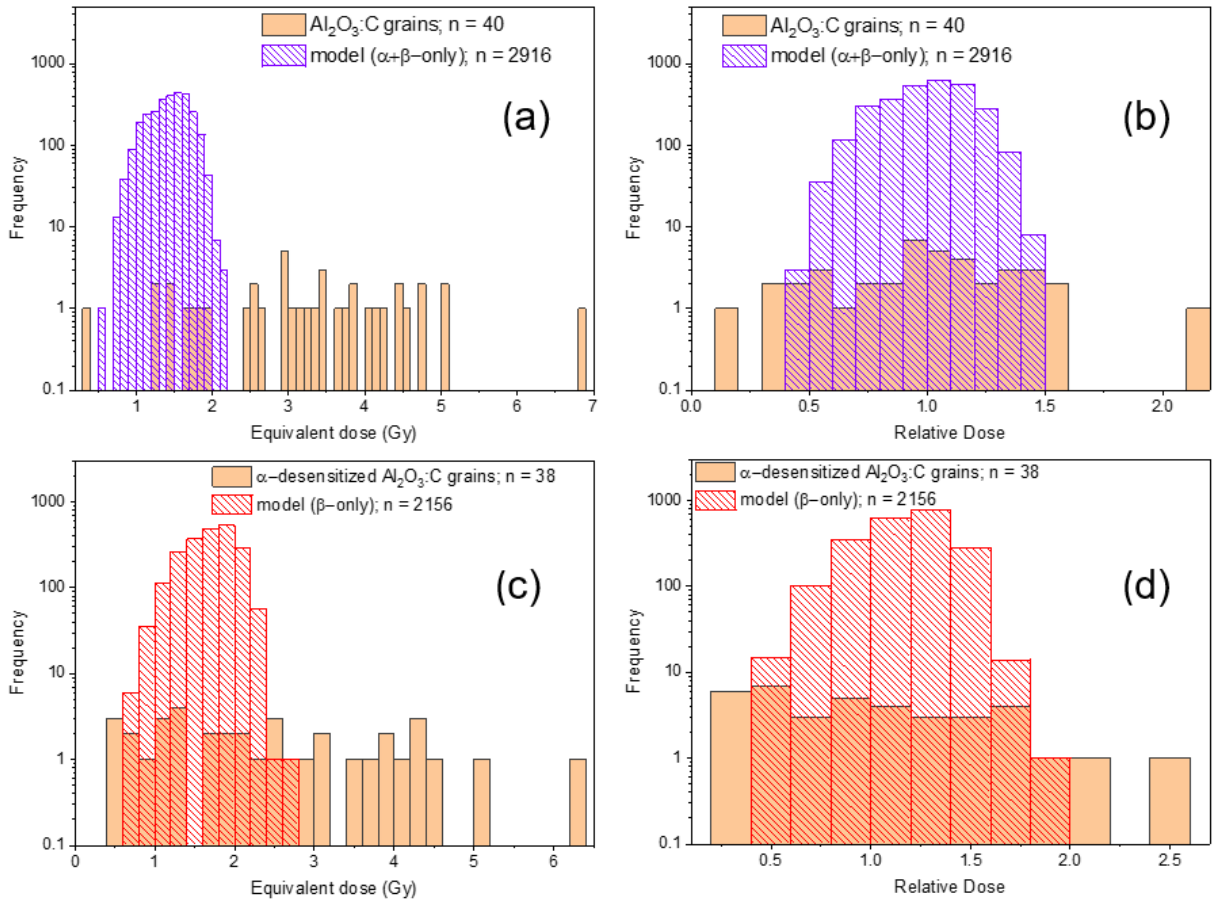


Figure 7.15: Comparison of experimental to simulated dose distribution for regular $\text{Al}_2\text{O}_3:\text{C}$ dosimeter grains in sample GS220 (a, b) and GS221 (c,d) with 30.7% and 37.0 % thoriantite by proportion respectively. (a) and (c) Absolute equivalent dose distributions with the simulated distributions corrected for alpha efficiency using an a -value of 0.01. (b) and (d) Same as (a) and (c) respectively, but with each distribution normalized to its respective mean. The simulated distribution represents the combined dose from $\alpha+\beta$ -particle simulations. The estimated uncertainty of individual measurements was 3–6 % for GS220 and 4–6 % for GS221. The uncertainty of the equivalent dose per dosimeter from the simulation is about 10% including the uncertainty associated with the 10^8 particles simulated. Therefore, a bin size of 0.1 and 0.2 was selected in (a,b) and (c,d) respectively, for easy comparison of the width of the distributions.

In Figures 7.15, the mean dose measured by the 40 regular $\text{Al}_2\text{O}_3:\text{C}$ grains is (3.21 ± 1.29) Gy, while the mean dose calculated from the 2916 grains simulation is (1.44 ± 0.26) Gy. For the α -desensitized $\text{Al}_2\text{O}_3:\text{C}$ dosimeter grains, the mean dose for the 38 measured grains is (2.45 ± 1.45) Gy, and (1.69 ± 0.31) Gy for the 2156 simulated grains. Like in the case of the other thoriantite samples, we also see here the experimental distribution shifted

to a higher dose compared to the simulated due to possibly a systematic error in mineral and density description of thorianite. For the regular $\text{Al}_2\text{O}_3\text{:C}$ grains, the relative standard deviation of the mean dose of the experimental dose distribution is 40%, about $2 \times$ more than the 18% for the simulated dose distribution. For the α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains, the relative standard deviation of the experimental dose distribution is 59%, which is ~ 3 times that of the simulated dose distributions at 18%.

Additionally, the Mann-Whitney non-parametric test shows that the experimental and simulated dose distributions for the regular $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains are NOT significantly different at the 0.05 level, with a very high p-value of 0.974 (see Figure 7.15b). For Figure 7.15d, the test shows that the two distributions are significantly different. The measured and simulated doses are also comparable to the expected infinite matrix doses: $\sim (1.39 \pm 0.23)$ Gy and $\sim (1.67 \pm 0.28)$ Gy for the regular and desensitized grains respectively.

Sample GS320 and GS321

Samples GS320 and GS321 are mixtures of fluorapatite ($\text{Ca}_{10}(\text{PO}_4)_6\text{F}_2$, density = 3.15 g/cm^3) and quartz. The specific information about the samples are presented in Table 7.13. The result of dose distribution for these samples are presented in Figure 7.16.

Table 7.13: Specific sample information for samples with small amounts of radioemitters.

Sample	Thorianite wt.%	Volume (cm^3)	Fill height (cm)	Grain-type	$\text{Al}_2\text{O}_3\text{:C}$ mass (g)
GS220	28.1	2.35	0.43	regular	0.26
GS221	26.0	2.09	0.38	α - desensitized	0.33

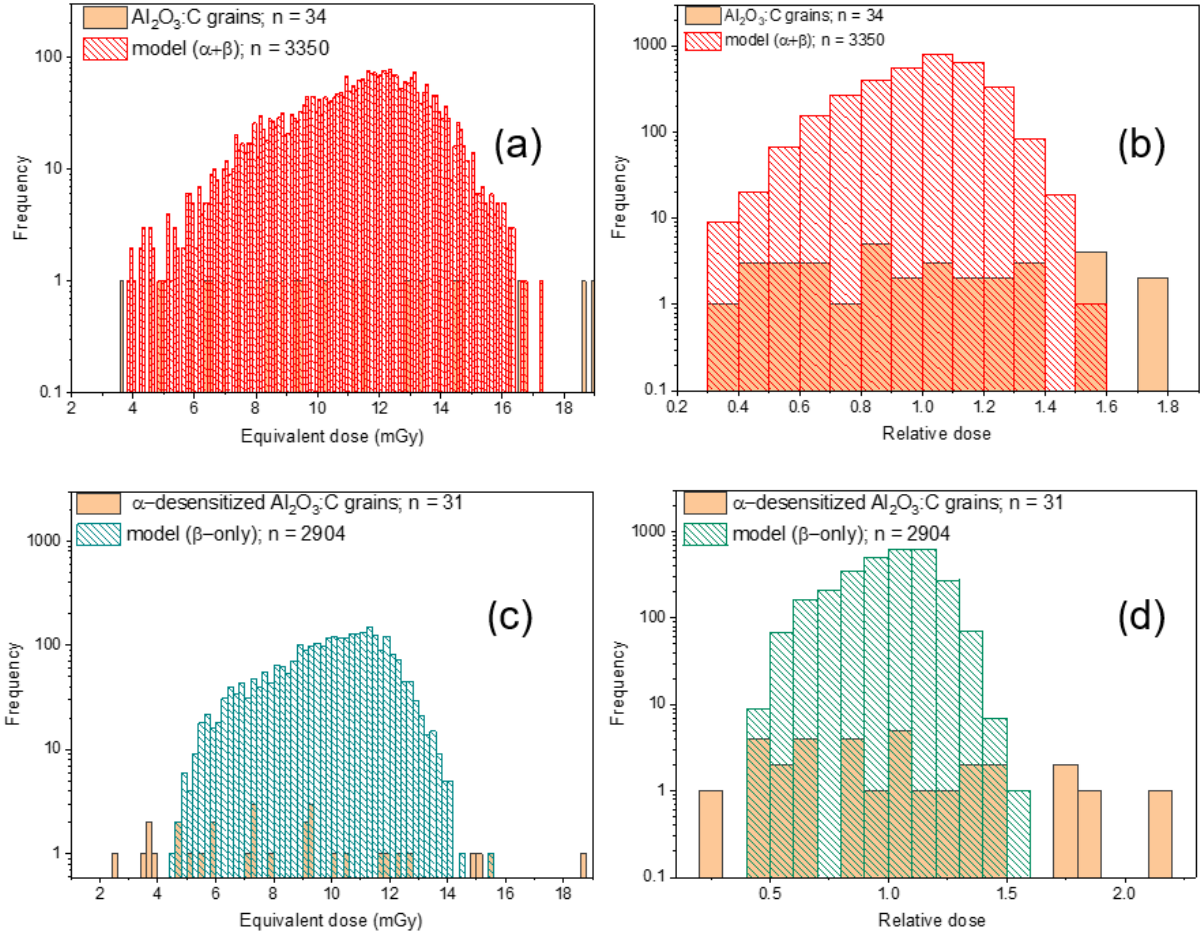


Figure 7.16: Comparison of experimental to simulated dose distribution for regular $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains in sample GS320 (a, b) and GS321 (c, d) with 28.1% and 26.0 % fluorapatite by proportion respectively. (a) and (c) Absolute equivalent dose distributions with the simulated distributions corrected for alpha efficiency using an α -value of 0.01. (b) and (d) Same as (a) and (c) respectively, but with each distribution normalized to its respective mean. The simulated distribution represents the combined dose from $\alpha+\beta$ -particle simulations. The estimated uncertainty of individual measurements was 5–15 % for GS320 and 5–20 % for GS321. The uncertainty of the equivalent dose per dosimeter from the simulation is about 10% including the uncertainty associated with the 10^8 particles simulated. Therefore, a bin size 0.1 was selected for easy comparison of the width of the distributions.

In Figures 7.16, the mean dose measured by the 34 regular $\text{Al}_2\text{O}_3\text{:C}$ grains is (10.80 ± 4.35) mGy, while the mean dose calculated from the 3350 grains simulation is (11.34 ± 2.14) mGy. For the α -desensitized $\text{Al}_2\text{O}_3\text{:C}$ dosimeter grains, the mean dose for the 31 measured grains is (8.55 ± 4.07) mGy, and (9.97 ± 1.84) mGy for the 2904 simulated grains. The overlap in margin of error confirms that the distributions are the same. Furthermore, the

Mann-Whitney non-parametric test shows that the two distributions are NOT significantly different at the 0.05 level, with a p-value of 0.682 and 0.410 for Figure 7.16b & d. The relative standard deviation of the experimental dose distribution for sample GS320 is 40%, which is ~ 2 times that of the simulated dose distributions at 19%. For sample GS321, the relative standard deviation of the experimental dose distribution is 48% compared to 18% for the simulated dose distribution. The measured and simulated doses also agree fairly with the expected infinite matrix doses: $\sim (12.79 \pm 2.11)$ mGy and $\sim (11.65 \pm 1.92)$ mGy for the regular and desensitized grains respectively. They represent theoretical maximum doses in the samples.

7.7 Summary of dose distribution comparison

Table 7.14: Average doses and standard deviations for measured and modelled dose distributions for dosimeter grains. The expected infinite matrix doses (IMDs) calculated directly from dose rate conversion factors are also presented for comparison.

Sample	Experimental (Gy)	Simulation (Gy)	Expected IMD (Gy)
IMGS010	1.09 ± 0.34	1.30 ± 0.28	1.22 ± 0.20
IMGS011	1.13 ± 0.27	1.30 ± 0.28	1.19 ± 0.20
GS100	1.27 ± 0.50	1.16 ± 0.27	1.67 ± 0.28
GS101	1.75 ± 0.92	1.91 ± 0.41	2.42 ± 0.40
GS120	2.98 ± 2.23	5.08 ± 0.96	5.70 ± 0.94
GS121	5.52 ± 1.87	5.43 ± 1.01	5.67 ± 0.93
GS200	1.03 ± 0.48	0.41 ± 0.08	0.45 ± 0.07
GS201	1.34 ± 0.96	0.44 ± 0.09	0.49 ± 0.08
GS220	3.21 ± 1.29	1.44 ± 0.26	1.39 ± 0.23
GS221	2.45 ± 1.45	1.69 ± 0.31	1.67 ± 0.28
GS300	$(3.36 \pm 1.06) \times 10^{-3}$	$(3.60 \pm 0.65) \times 10^{-3}$	$(4.69 \pm 0.77) \times 10^{-3}$
GS301	$(3.32 \pm 1.60) \times 10^{-3}$	$(3.97 \pm 0.81) \times 10^{-3}$	$(4.93 \pm 0.81) \times 10^{-3}$
GS320	$(10.80 \pm 4.35) \times 10^{-3}$	$(11.34 \pm 2.14) \times 10^{-3}$	$(12.79 \pm 2.11) \times 10^{-3}$
GS321	$(8.55 \pm 4.07) \times 10^{-3}$	$(9.97 \pm 1.84) \times 10^{-3}$	$(11.65 \pm 1.92) \times 10^{-3}$

A summary of the experimental and simulated dose distributions is presented in Table 7.14. Most of the experimental and simulated dose distributions show good agreement. Exceptions are samples GS200, GS201, GS220, and GS221 where the experimentally measured dose

is about 2–3 times the simulated dose. This difference is most likely due to a systematic error in the density and mineral description of the radioemitter thorianite used in all these simulations.

The limited number of measured grains compared to simulated grains might have affected the accuracy of our mean doses, especially for samples where the number of measured grains were less than 50. However, based on the Central Limit Theorem, 30 observations are the minimum requirement for good statistics [205, 206]. For our samples, the minimum number of measured grains was 31. Now, while the number of data points considered can affect the accuracy (mean) of a distribution, it has little effect on the spread of the distribution [207]. In this work, this spread was reported directly as standard deviation. The use of standard deviation instead of standard error to report the spread ensures that the number of measured grains has minimal effect on the spread of our distributions.

Additionally, the result shows that there is no noticeable difference between the dose values of dosimeters buried in samples with high and low proportions of the radioemitters. Samples were initially divided into high and low amount of radioemitters in order to study the effect of dose rate. The result shows that the response of the dosimeters is either independent of dose rates or the statistical uncertainties of our result are bigger than the dose rate effect on the dosimeters if any. Similarly, some samples were devised with "hot" radioemitters like thorite and thorianite while others were prepared with "cold" radioemitters like apatite. This was done to check whether our measurement/simulation protocols can be able to distinguish between "hotspots" and "coldspots". The result confirms that our measurement/simulation protocols can clearly differentiate between high dose and low dose regions as can be seen from Tables 7.6 and 7.14, where both the Gy and mGy doses were reported, compared and also show good agreement.

Furthermore, the maximum absorb-able doses in our samples were calculated as the infinite matrix dose for each sample. These infinite matrix doses are also presented in Tables 7.6 and 7.14 for $\text{Al}_2\text{O}_3\text{:C}$ Luxel[®] and grains respectively. The measured and simulated doses

were either less or close to the IM dose as expected, with the exception of the measured doses that suffer from systematic errors.

In general, the experimental and simulated dose distribution show a better agreement for samples under infinite matrix conditions i.e., samples IMGS010 and IMGS011 compared to the samples where the infinite matrix condition is violated in the z-axis (vertical direction of the sample). This is evident from the extent of overlap and agreement in standard deviations and dose rate inhomogeneity for samples IMGS010 and IMGS011 compared to the other samples. The large differences in standard deviations and consequently dose rate inhomogeneity for the non-infinite matrix condition samples can be attributed to the loss of electron equilibrium in the z-axis of the container. The height of the samples in the container were approximately 3.5 mm. For perspective, the range of a 2 MeV beta particle in Al_2O_3 , ThSiO_4 , and SiO_2 are ~ 2.5 mm, 1.9 mm, and 3.8 mm respectively. Therefore, the farther a dosimeter is from the center of the sample, the more its dose is affected by the loss of electron equilibrium.

To conclude, we have shown that the dose distributions and microscopic dose rate variations of a sediment can be calculated accurately from Monte Carlo simulations. We have also confirmed that single grain luminescence dosimetry is a reliable technique for obtaining spatial dose rate variation in sedimentary environments. Some limitations of this result are the low sensitivity of OSL for measurement of very low doses and the inability to localize the dosimeters in the sample. Another limitation is the total number of particles that can be simulated for each model due to the linear relationship between simulation time and numbers of simulated particles

Chapter 8

Simulations Under Infinite-Matrix Conditions

8.1 Introduction

In the previous chapters the model calculations were validated by comparing experimental dose distributions with results from simulations. In all cases the sample dimensions were limited to a container of known dimensions. The overall goal of this dissertation is to simulate the radiation environment in the Martian subsurface. There the "sample" dimension is infinite and the simulated dose rate distribution (the dose rate is simply dose normalized by time) is supposed to be representative of the specified location on Mars. Therefore, the number of simulated particles will not be the only factor that might affect the uncertainties in our result.

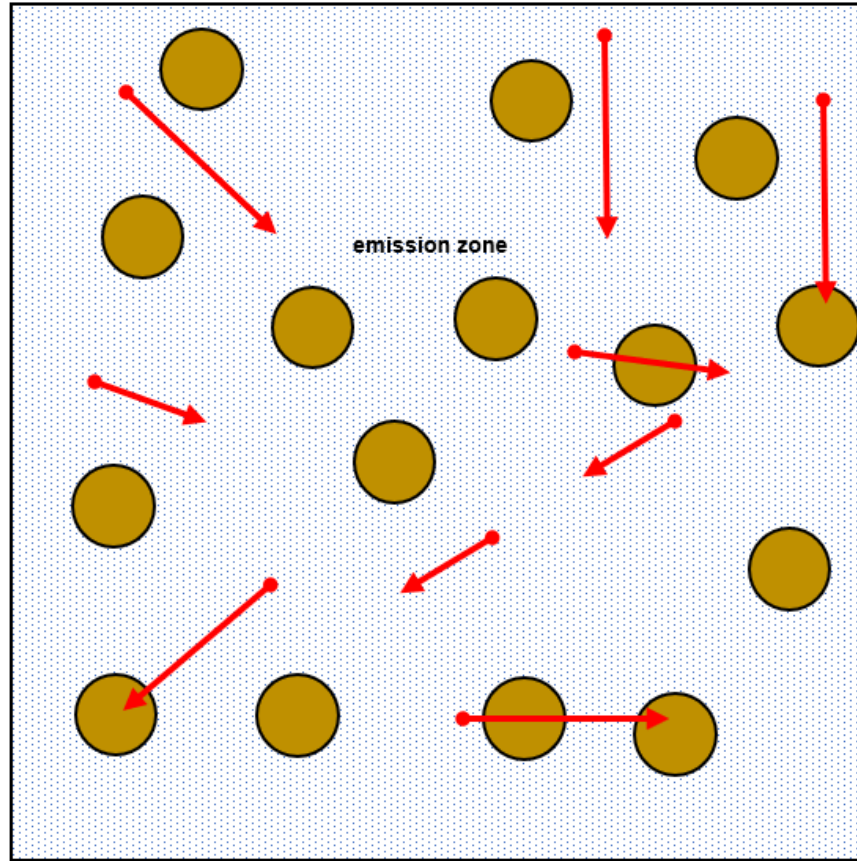


Figure 8.1: Schematic representation of the infinite matrix model with a uniform solid matrix surrounding $260\ \mu\text{m}$ $\text{Al}_2\text{O}_3\text{:C}$ grains (brown spheres). The red arrows represent charged particles.

Infinite matrix (IM) samples were modelled with *DosiSed* since the dosimeters will be modelled as grains. The model has the shape of a cube. The cube is composed of multiple spherical grains surrounded by a radioactive, uniform matrix of a specific mineral (see Figure 8.1.). The dimensions of the matrix are calculated from the sum of the "dosimeter voxels" and 1.2 times the maximum range of the simulated particles. The dosimeter voxels are the voxels whose grains are used as dosimeters. They can also be defined as the volume of interest, i.e., the volume where the dose information is needed. When an infinite matrix condition is defined, doses are only calculated in the detector voxels and not in all the voxels that make up the model (i.e., not in the voxels used to enforce the infinite matrix condition). The external "non-dosimeter voxels" are exactly the same as the dosimeter voxels in mineral

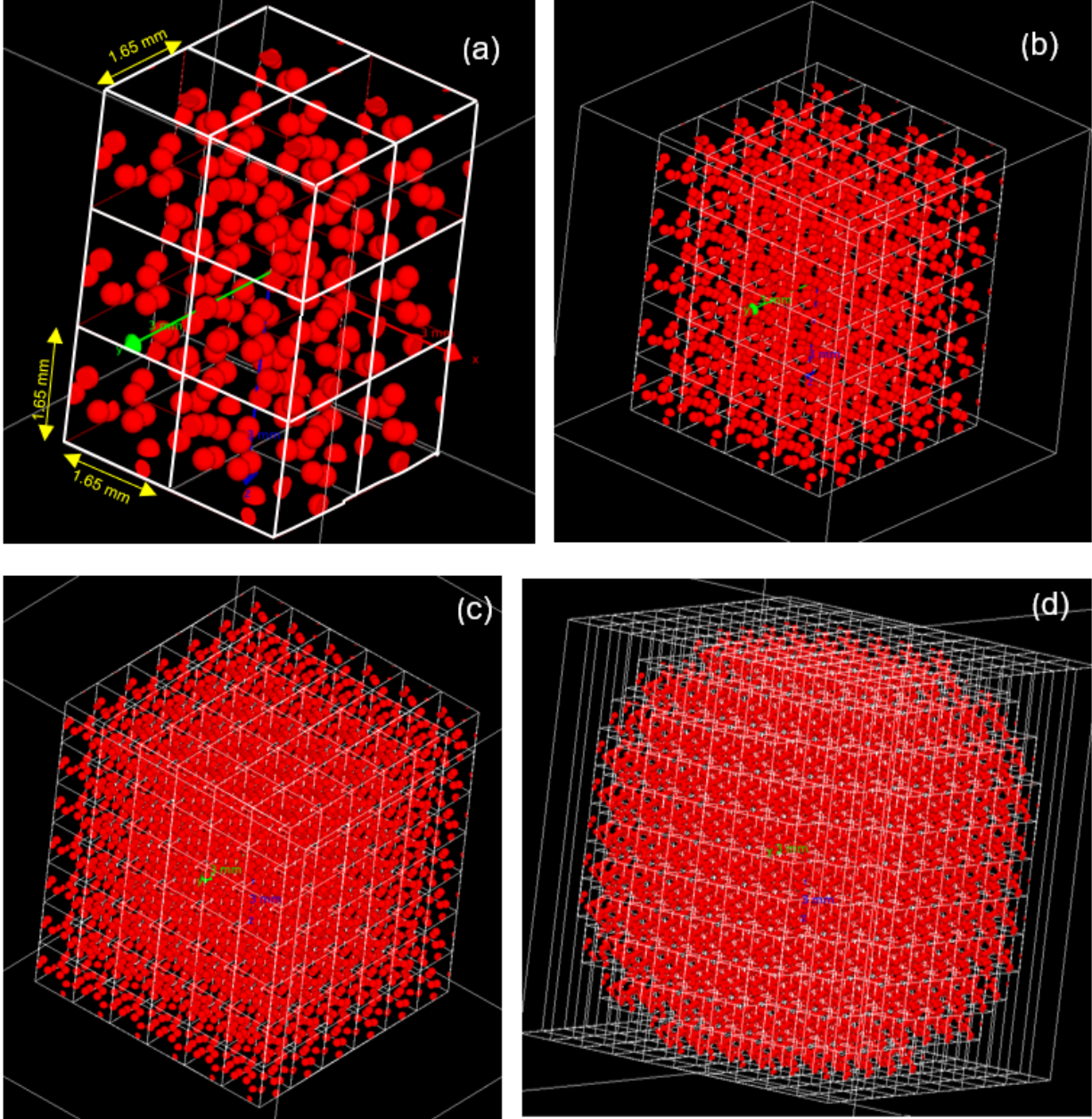


Figure 8.2: Infinite matrix condition implementation in *DosiSed* by specifying the max. range, (r) of the simulated particles. The side length, l of the cubic voxel is 1.65 mm. (a) dosimeter voxels ($2 \times 2 \times 3$); IM condition not specified. (b) for $r = 1.5$, the dosimeter voxels are padded with $([1.2 \times 1.5]/1.65 = 1.09 \sim 1)$ one extra voxel in every direction, making it $4 \times 4 \times 5$. (c) for $r = 3.0$, the dosimeter voxels are padded with $([1.2 \times 3.0]/1.65 = 2.18 \sim 2)$ 2 extra voxels in every direction, making it $6 \times 6 \times 7$. (d) for $r = 5$, the dosimeter voxels will be padded with $([1.2 \times 5]/1.65 = 3.63 \sim 4)$ with 4 extra voxels in every direction, making it $10 \times 10 \times 11$. Notice that as the number of non-dosimeter voxels increases, the model becomes more of a sphere.

compositions except they are only used to enforce the infinite matrix conditions. To achieve this in *DosiSed*, the user must specify the maximum range of the simulated particle via the pilot file (see Section 4.4.2). This number in mm is then multiplied by 1.2 and then divided by the cubic voxel size and rounded up to a whole number to determine how many voxels are used to pad the "dosimeter voxels" in every direction to ensure that charged particle equilibrium is maintained even at the edges of the dosimeter voxels. A demonstration of this is shown in Figure 8.2.

In the model, radiation is emitted from the matrix, while the inert $260\text{ }\mu\text{m Al}_2\text{O}_3$ grains serve as the dosimeters. Given that the matrix is uniform, the dose rate distribution of the dosimeters should be equal to or better than the experimental definition of uniformity as measured via gamma irradiation of the dosimeters in Section 7.4. The uniformity of the dose rate distribution will be tested by varying one of the simulation parameters at a time and then quantifying the effect on the dose rate distribution. The effect in each case will be calculated as the relative standard deviation or the coefficient of variation of the distribution. This coefficient of variation in unit of percent is the standard deviation divided by the mean of the distribution. The reference for the coefficient of variation is the experimentally established uniformity baseline of 5.5%.

The simulation parameters tested are listed in Table 8.1. The voxel size is used to describe the side length of the each cubic voxel. The volume fraction is defined as the volume of the dosimeters (grains) in each voxel divided by the total volume of the voxel. In other words, the volume fraction is defined per voxel, and is replicated in every voxel including the dosimeter and non-dosimeter voxels. The total volume is sum of the volumes of all dosimeter voxels. This sum does not include the non-dosimeter voxels. It represents the dose volume. It can be calculated by multiplying the total number of voxels in the dosimeter voxels by the volume of each voxel.

Table 8.1: Simulation parameter space tested using infinite matrix condition

Parameter name	Values tested
No of simulated particles	1E7, 5E7, 1E8, 5E8, and 1E9
Volume fraction (%)	1.63, 5, 10, 20, 30 and 40
Voxel size: side length of cubic voxel (mm)	1, 1.5, 3, 5, 7.5, 15
Total volume (mm ³) based on n ³	where n = 1, 8, 12, 20, and 30
Grain size (μm)	5 – 350

It is important to mention that in testing the simulation parameters, the maximum ranges of the simulated particles were fixed. They are; 3 mm for beta particles, 30 μm for alpha particles and 3 mm for the Compton electrons of gamma particles. This ensures that irrespective of the parameter tested, the infinite matrix condition is always satisfied for the charged particles.

8.2 Quantifying the coefficient of variation for an infinite simulation space

8.2.1 Effect of the number of simulated particles on the uniformity of beta-dose rate distributions

In Section 6.3.2 it was shown that the number of simulated particles has an impact on the result of Monte Carlo simulation and thus introduces an uncertainty to the results. Thus, in a first step, this effect was quantified for an infinite-matrix condition. This was done by varying the simulated number of particles using the infinite matrix model described in the introduction. In this investigation in particular, other fixed simulation parameters are: voxel size (1.5 mm), volume fraction (1.63% first, and then 16.5% later), total volume (7.5 mm $\times$

7.5 mm $\times$ 7.5 mm), and grain size (260 μ m). The model is presented in Figure 8.3. Three different spectra were considered, Th-series, U-series and ^{40}K . The Th- and U-series beta emission was from a uniform thorite matrix (ThSiO_4 , density = 5.34 g/cm 3). The ^{40}K beta emission was from a uniform potassium chloride matrix (KCl , density = 1.98 g/cm 3). Given that the grain size of the detector is small compared to the range of beta particles in $\text{Al}_2\text{O}_3\text{:C}$ (~ 2 mm), a narrow dose rate distribution is expected.

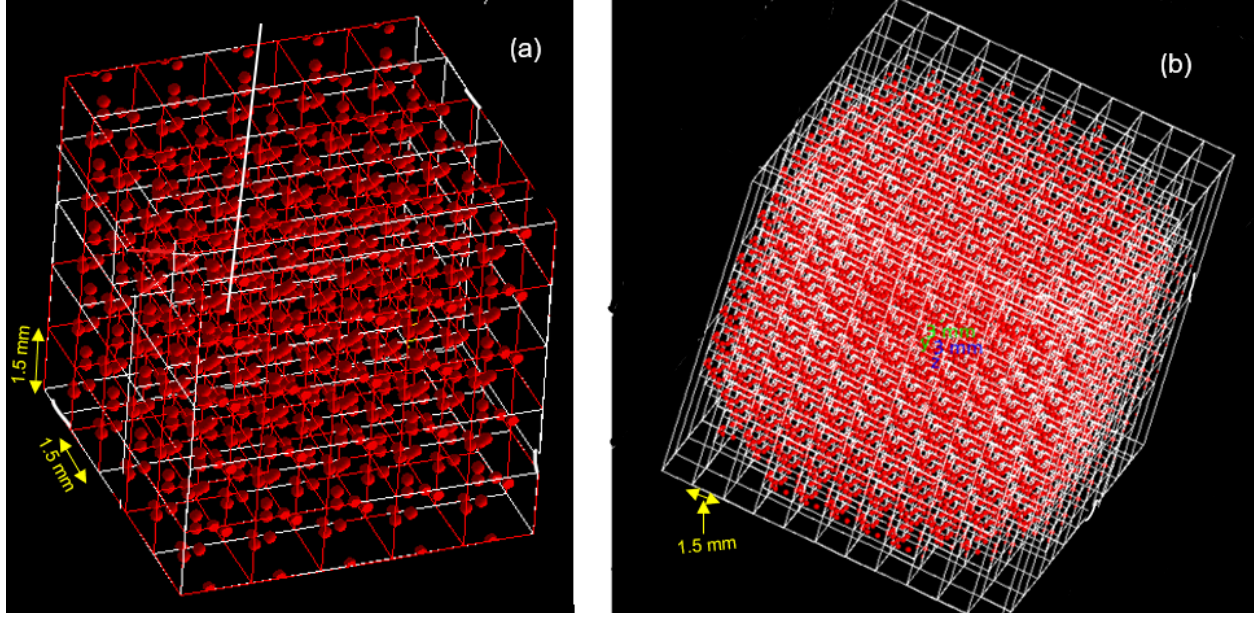


Figure 8.3: IM model for testing the effect of the number of simulated particles. (a) the dosimeter voxels and (b) IM condition implementation. In this model, voxel size = 1.5 mm, volume fraction = 1.63%, total volume (7.5 mm) 3 and grain size = 260 μ m.

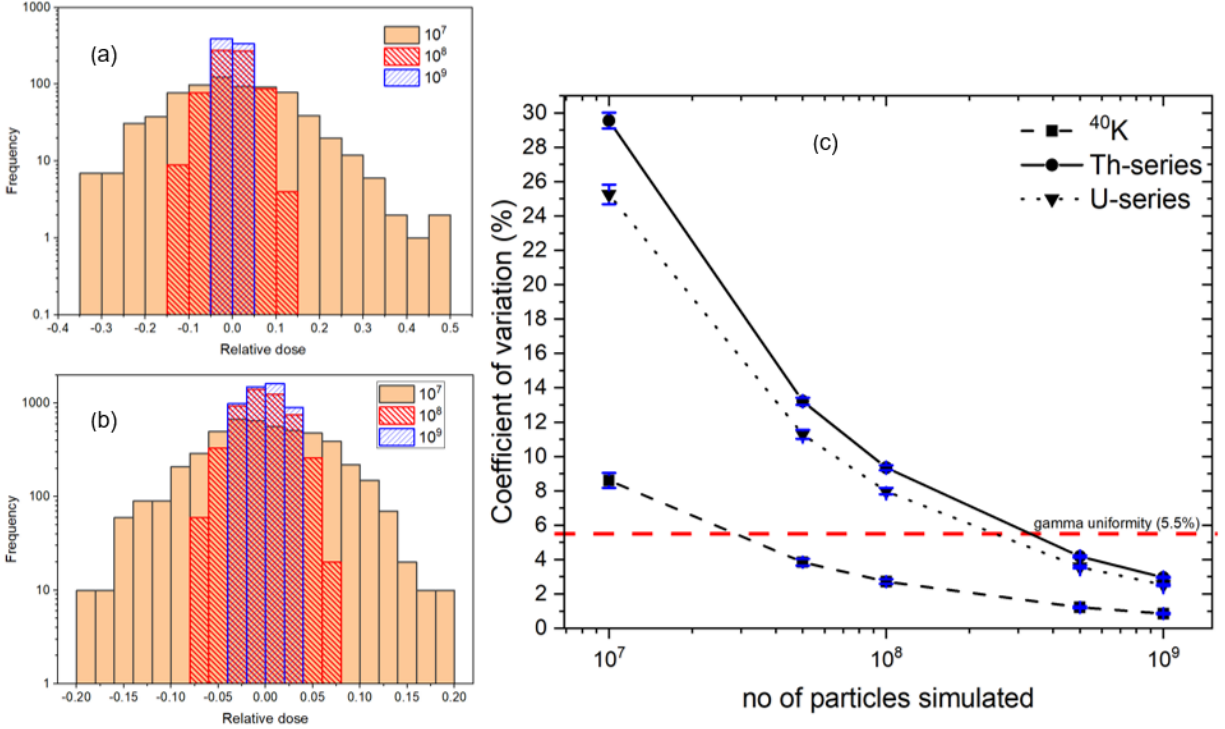


Figure 8.4: Effect of number of beta simulated particles on the dose rate distribution of 260 μm $\text{Al}_2\text{O}_3\text{:C}$ grains surrounded by a uniform matrix of ThSiO_4 or KCl . (a) shows an example of the ^{40}K dose rate distribution when the dosimeter grains represent 1.63% of the total volume. (b) shows an example of the ^{40}K dose rate distribution when the dosimeter grains represent 16.5% of the total volume. (c) The coefficient of variation versus the number of simulated particles. The red dash line represents the gamma uniformity baseline (5.5%). The error bars represent the standard deviation of the coefficient of variations of the two volume fractions of dosimeter considered.

From the distribution in Figure 8.4 the number of particles required to achieve or better the uniformity baseline of 5.5% for the ^{40}K , Th- and U-series is 5×10^8 . As the number of particles increases, the coefficient of variations (percent standard deviations of the mean) also decrease. The overall trend is that of $1/\sqrt{N}$ as expected for Poisson statistics. The overall coefficient of variations for ^{40}K are less than those for the Th- and U-series. It took only 5×10^7 particles to meet the uniformity baseline for ^{40}K . This might be due to the density difference between the dosimeter and the uniform matrix. For example, for thorite matrix, with density 5.34 g/cm³, which is greater than the density of the dosimeter (3.97 g/cm³), there will be build-down (dose fall-off due to change in attenuation and scatter when you go

from a region of higher density to one of lower density) of dose whenever radiation goes from thorite to Al_2O_3 dosimeters. For KCl, on the other hand, there will be a build-up since KCl (1.98 g/cm^3) has a lower density compared to the dosimeter. Additionally, the back-scatter contributions at the matrix/dosimeter interface are also different due to density differences. In fact, back-scatter at the dosimeter/thorite interface is higher than that of the dosimeter/KCl interface. Also, the Th- and U-series have a higher portion of their spectra in the low energy range and back-scattering at an interface increases as energy decreases. All these likely contributed to more broadening of the Th- and U-series dose rate distribution. Additionally, Figures 8.4a and b show the beta dose rate distributions for uniformly distributed ^{40}K with different volume fractions of dosimeter grains compared to the total volume considered (i.e., 1.63% and 16.5%). The coefficient of variations for the distributions are slightly different and were used to calculate the error bars in Figure 8.4c. The influence of the volume fractions of the dosimeter was further investigated in 8.2.3

8.2.2 Characterizing the coefficient of variations for alpha particle dose rate distributions

The same simulations as described in 8.2.1 were also carried out for α -particles. Given the size of the dosimeter grains compared to the range of alpha particles ($20 \mu\text{m}$), it was interesting to see how the coefficient of variations compare to the gamma uniformity. Apart from the fact that the alpha radiation only affects the outer shell of the grains, the alpha field is expected to be uniform for this case as well. The result is presented in Figure 8.5.

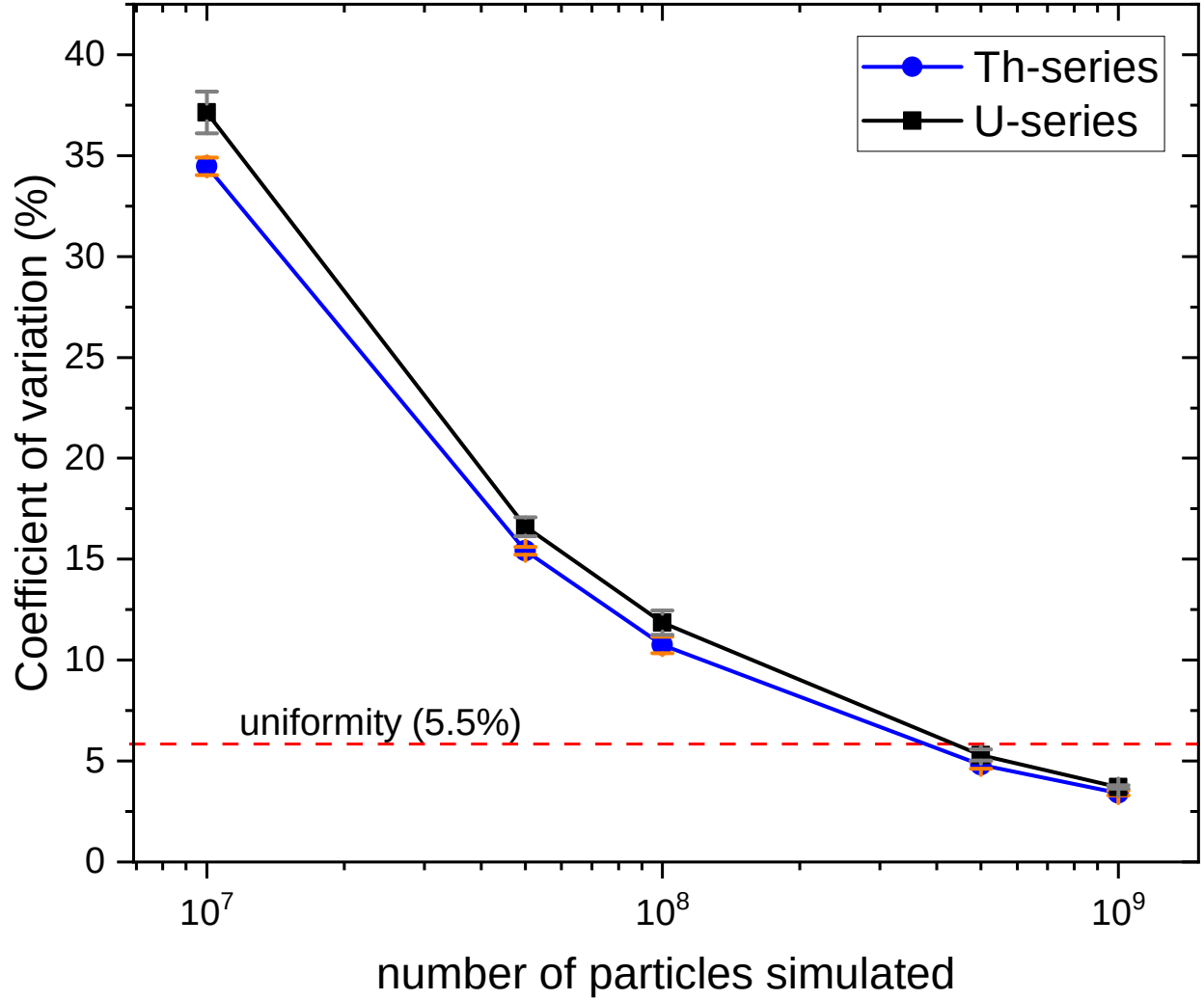


Figure 8.5: Effect of number of simulated alpha particles on the alpha dose rate distribution of $260 \mu\text{m}$ $\text{Al}_2\text{O}_3\text{:C}$ grains surrounded by a radioactive matrix under infinite matrix conditions. The red broken line is the gamma uniformity baseline (5.5%). The error bars represent the standard deviation of the coefficient of variations for 3 runs of the same simulation.

For the simulation of 10^8 particles, the beta uniformity for the Th- and U- series was $\sim 9\%$ & 8% , while the alpha uniformity is $\sim 12\%$ & 11% respectively. The alpha radiation simulation also achieves gamma uniformity when 5×10^8 particles are simulated, which is surprising when we consider the range of alpha particles compared to the size of the dosimeter grains, one would expect a broader distribution compared to beta. However, I suspect the approximation of the dosimeter grains as spheres contributes to narrowing the distribution. For a real sediment, with irregularly-shaped grains, the effect of grain size and shape will

likely result in a broader distribution for the alpha radiation. The alpha dose rate distribution will be strongly dependent on the surface of the grains and the entry angle.

The large number of particles required to achieve alpha radiation uniformity for all three spectra implies very long computational times. For example, the computational time for 10^8 particles ranges from 12–24 hours depending on the level of complexity. For 10^9 , it will take 10 times the time and for 10^{10} , the simulation time will increase by 100 (~ 2 –4 months) and so was not tested.

8.2.3 Effect of the volume fractions of dosimeter on the uniformity of beta-dose rate distribution

Figure 8.4 illustrated that the relative volume occupied by the inert dosimeter grains can influence the dose rate distribution. For this investigation, the same simulations as described in 8.2.1 with beta particles were carried out by varying the volume fraction of dosimeters (grains). The number of dosimeter grains can influence the dose rate distribution in two contrasting ways. A smaller volume fraction of dosimeters will preserve uniformity, and thus the infinite matrix condition, so that one would expect a lower coefficient of variation for fewer dosimeters. On the other hand, a larger number of dosimeters is expected to result in more dosimeter-particle interactions and thus less uncertainty. This effect was tested for dosimeter volume fractions from 1-40% with 5×10^8 simulated particles in all cases. Selected dose rate distributions are shown in Figure 8.6a. The relative standard deviations are shown in Figure 8.6b.

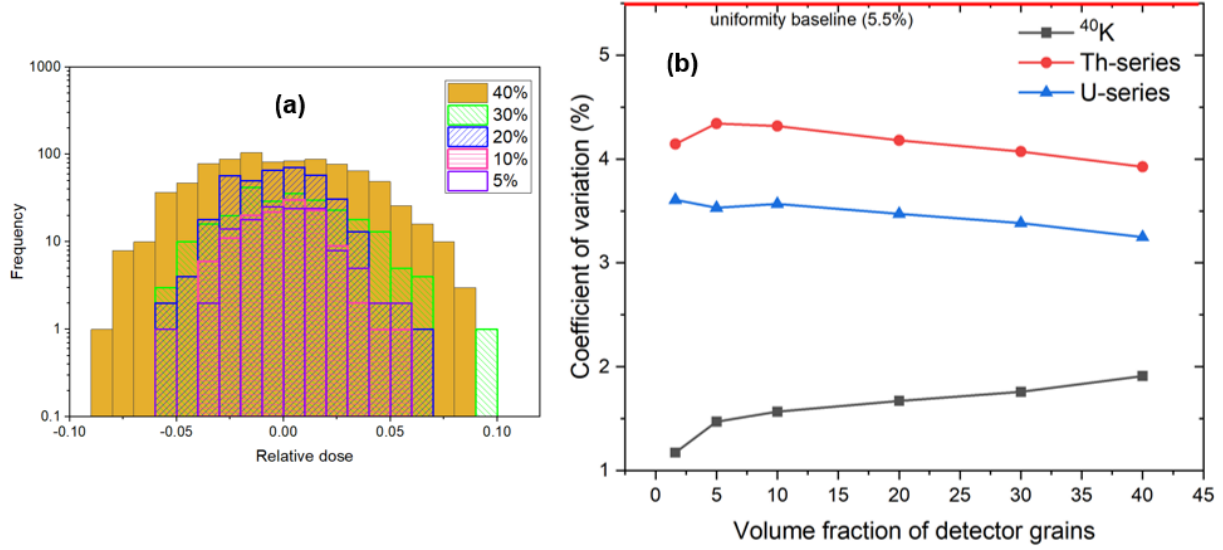


Figure 8.6: Effect of volume fractions of detector grains on the dose rate distribution obtained from $260\ \mu\text{m}$ $\text{Al}_2\text{O}_3:\text{C}$ dosimeter grains in a uniformly radioactive matrix. The number of simulated particles was 5×10^8 . (a) Examples of selected relative dose rate distributions for ^{40}K with each case normalized to their respective mean. (b) The coefficient of variations of the distributions versus volume fractions. The red line is the uniformity baseline (5.5%).

We see a slow decrease in the coefficient of variations for the Th- and U-series and a slow increase in the coefficient of variations for ^{40}K as the volume fraction increases. For the Th-series, we also see a maximum which is not visible for the U-series. The slow increase for ^{40}K could be as a result of disequilibrium as the number of dosimeter increases. KCl has a lower density compared to the dosimeters so beta particles travel further in the matrix so that equilibrium is established for smaller number of dosimeters. But as the number of dosimeters increases, this equilibrium is slowly disturbed because of increase in the matrix/dosimeter interfaces (regions of increase attenuation). For U and Th, the opposite effect might be happening. However, for 5×10^8 simulated particles, all the coefficient of variations were below the gamma uniformity baseline. Also, the differences between the results from the various volume fractions are small compared to the coefficient of variations.

To conclude, considering the results of Figures 8.4 and 8.6, simulations of 10^7 particles can result in coefficient of variation of up to 9% for ^{40}K , 25% for U-series and 30% for Th-series, indicating that 10^7 particles should not be used for dose rate distribution comparison. In

order to keep coefficient of variations below the uniformity baseline for all spectra, 5×10^8 or more particles must be simulated.

8.2.4 Effect of voxel size and total volume on the beta-dose rate distributions

As mentioned in section 4.4.1, sedimentary grain modelling using MC simulation involves random grain stacking or packing of spherical mineral grains into a box that represents the simulation space (dosimeter voxels). Packing the grains into a single voxel (using 1 large voxel as the dosimeter voxel) or using a combination or stacking of multiple smaller voxels to represent the dosimeter voxels (see illustration in Figure 8.7) can affect the simulation time and potentially affect the coefficient of variations. This effect was investigated via beta-dose rate distributions, to determine the optimum voxel size. To achieve this, the total volume is kept constant ($15 \text{ mm} \times 15 \text{ mm} \times 15 \text{ mm}$; 3375 mm^3) while the voxel sizes are varied. The volume is composed of a uniform ThSiO_4 or KCl matrix surrounding Al_2O_3 grains. Other fixed parameters are volume fraction (1.63%) and grain size ($260 \mu\text{m}$). Some of the voxel sizes simulated are shown in Figure 8.7. It must be stressed that only the dosimeter voxels are presented in Figure 8.7, the actual simulation was performed using the infinite matrix condition. The result is shown in Figure 8.8. Note: For easy representation, the voxel size is defined as the side length of the cubic voxel in mm.

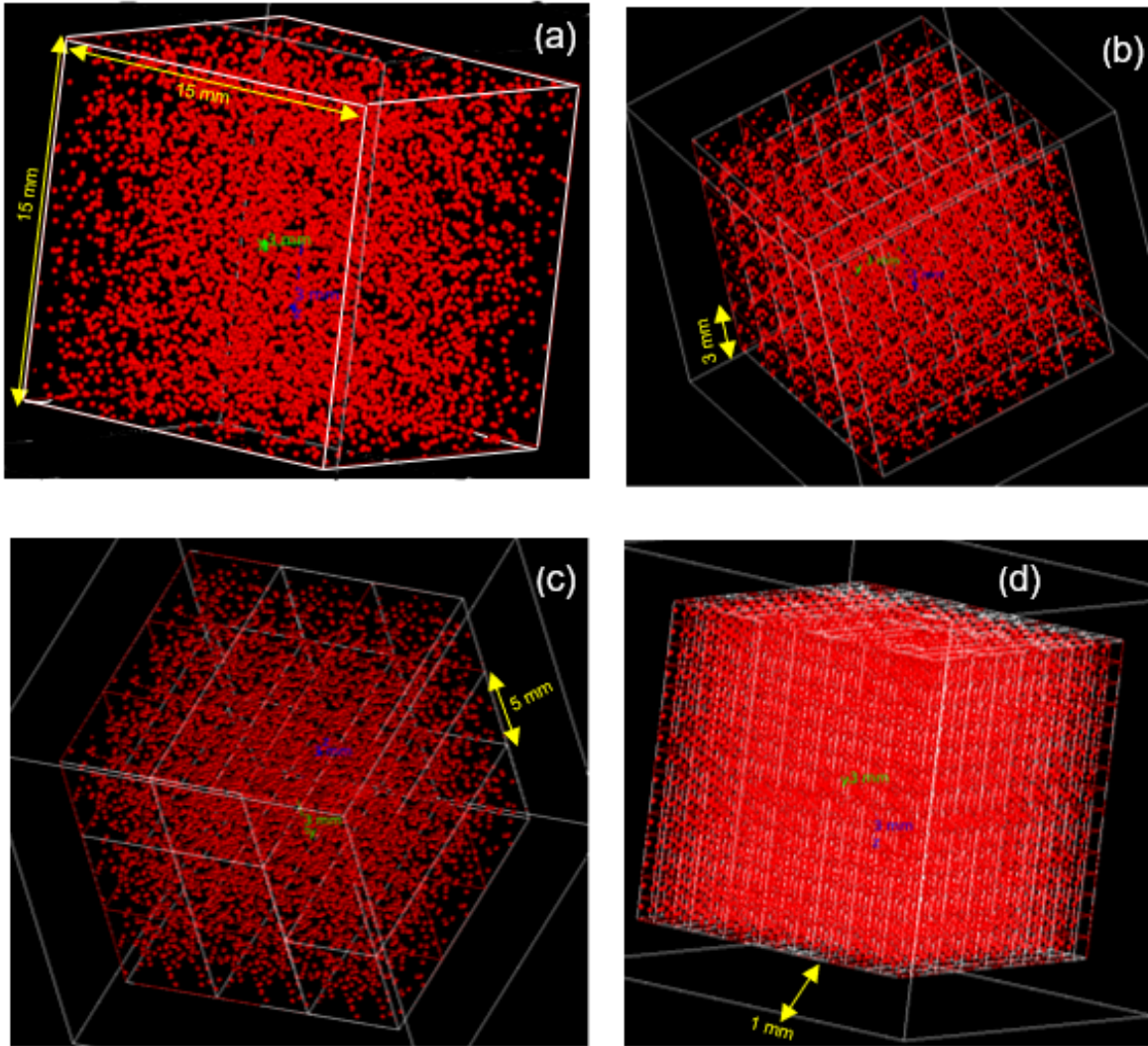


Figure 8.7: Models for investigating the effect of voxel sizes. These models are the "dosimeter voxels" alone before the implementation of the IM condition. The IM condition is implemented in the actual simulations. (a) Single $(15 \text{ mm})^3$ voxel for a total volume of 3375 mm^3 . (b) $5 \times 5 \times 5$ voxels with dimensions $(3 \text{ mm})^3$ for a total volume of 3375 mm^3 . (c) $3 \times 3 \times 3$ voxels with dimensions $(5 \text{ mm})^3$ for a total volume of 3375 mm^3 . (d) $15 \times 15 \times 15$ voxels with dimensions $(1 \text{ mm})^3$ for a total volume of 3375 mm^3 .

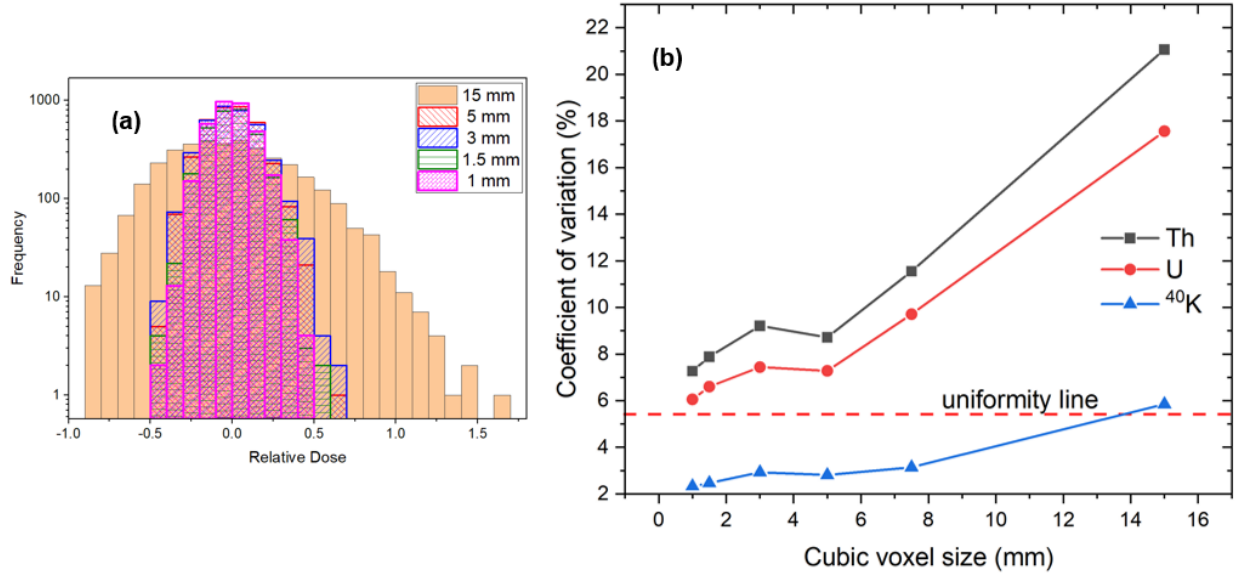


Figure 8.8: Effect of voxel sizes on the dose rate distribution of $260\ \mu\text{m}\ \text{Al}_2\text{O}_3\text{:C}$ grains mixed with a radioactive matrix under infinite matrix conditions. The number of simulated particles was 5×10^8 . The total volume of the dosimeter voxel is $15\ \text{mm} \times 15\ \text{mm} \times 15\ \text{mm}$. (a) Examples of selected dose rate distributions for ^{40}K with different voxel sizes (b) The coefficient of variations versus voxel sizes. The red broken line represents the gamma uniformity baseline (5.5%).

The result shows that as the voxel size increases the coefficient of variations also increase. This result is difficult to explain because one would expect the same total number of grains irrespective of the voxel size. A possible explanation for this is radiation is simulated in each voxel, the emission when the voxels are smaller are more targeted (smaller space to emit radiation randomly) compared to the bigger random space available for a bigger voxel. This will thus increase the chance of hitting a grain in a smaller voxel compared to a bigger voxel. Another possibility is that for the same number of particles, the beta particles are losing all their energies before traversing the voxel (2 mm particle in a 15 mm voxel). This will increase the discrepancies in the dose absorbed by the grains. For a smaller voxel comparable to the range of the particles, the grains receive a uniform dose. If this is true then simulating a longer ranged particle like gamma will prove it. This was not investigated further.

The result also shows that for the volume considered ($15\ \text{mm} \times 15\ \text{mm} \times 15\ \text{mm}$), the coefficient of variations for ^{40}K are within the baseline except for a voxel size of 15 mm. For

Th & U, the coefficient of variations are greater than the uniformity even for the smallest voxel size considered. Figure 8.6 on the other hand, which was calculated for a $7.5 \text{ mm} \times 7.5 \text{ mm} \times 7.5 \text{ mm}$ volume, showed coefficient of variations within the uniformity baseline for even Th & U. Thus it can be concluded that the coefficient of variation also depend on the total volume of the simulation space. Therefore, the effect of the total volume on the coefficient of variation was investigated. The same simulation as above was carried out using a fixed voxel size of 2 mm. The total volume was changed by increasing the number of voxels in each direction. For example, an $8 \text{ mm} \times 8 \text{ mm} \times 8 \text{ mm}$ volume is created by arranging four 2mm voxels in each direction ($4 \times 4 \times 4 = 64$ voxels in total). The result is presented in Figure 8.9.

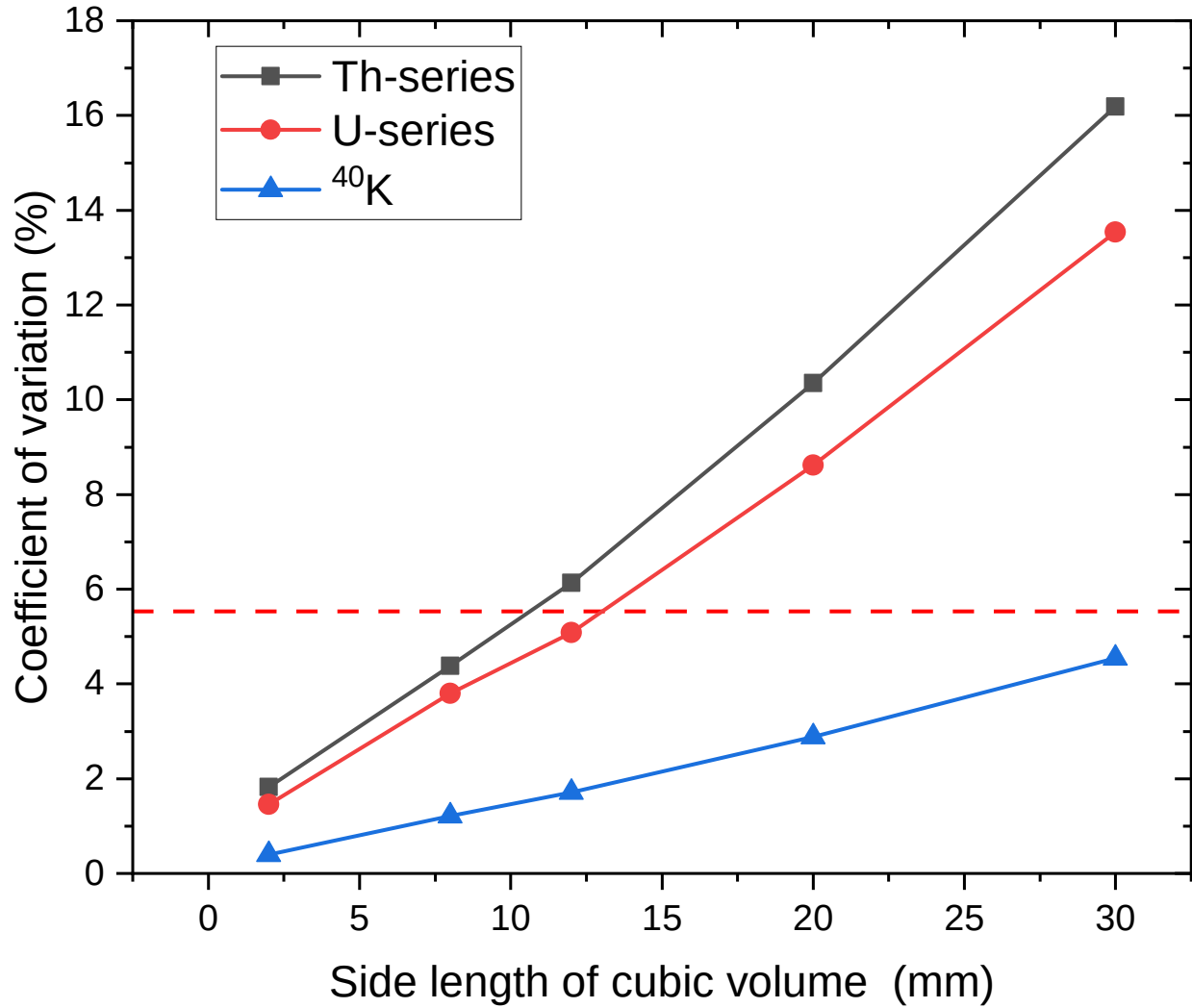


Figure 8.9: Effect of the total volume on the beta dose rate distribution of $260 \mu\text{m Al}_2\text{O}_3\text{:C}$ grains mixed with a radioactive matrix under infinite matrix conditions. The number of simulated particles was 5×10^8 . The voxel size is 2mm. The coefficient of variations are plotted on the y-axis. The volume is represented by the side length of the cube that makes up the volume. For example, a cubic volume of $8 \text{ mm} \times 8 \text{ mm} \times 8 \text{ mm}$ is represented with a side length of 8 mm. The red broken line represents the gamma uniformity baseline (5.5%).

As suspected, the statistical uncertainty increases with increase in total volume. This might be due to the decrease in the number of simulated particles to volume ratio as the volume increases while the number of simulated particles stays the same. The implication of this is each dosimeter will be hit by fewer particles. Thus the statistics gets worse.

8.2.5 Effect of the dosimeter grain size on the beta radiation dose rate distribution

When dosimeters of different grain sizes are exposed to a uniform beta radiation field, each dosimeter is expected to absorb a dose that is equivalent to its attenuation of the uniform dose, where the attenuation is a function of its grain size. Every dosimeter of equal grain size would exhibit a uniform dose rate distribution. This effect was investigated for dosimeters of various grain sizes mixed with the same radioactive matrix considered in the other tests (with a volume of $10\text{ mm} \times 10\text{ mm} \times 10\text{ mm}$, voxel size of 2 mm , and a dosimeter volume fraction of 1.63%). The grains are exposed to beta radiation from the uniform matrix. The coefficient of variations for the various grain sizes are shown in Figure [8.10](#). A total of 5×10^8 particles were simulated in each case.

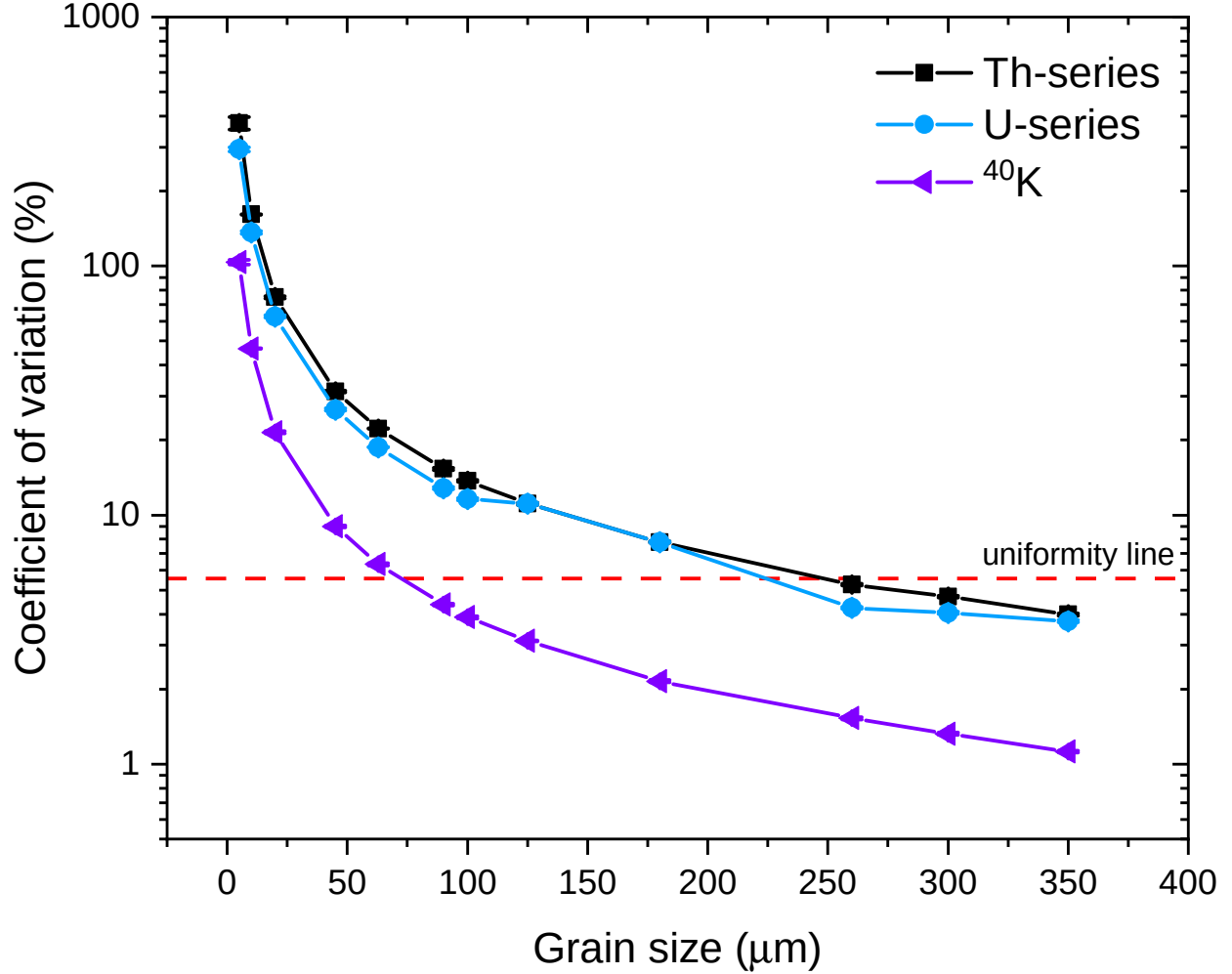


Figure 8.10: Effect of dosimeter grain size on the broadening of the dose rate distribution. The number of simulated particles was 5×10^8 . The volume of the dosimeter voxels is 10 mm $\times$ 10 mm $\times$ 10 mm, voxel size is 2 mm, and the volume fraction is 1.63%. The red dashed line is the gamma uniformity baseline (5.5%). The error bars represent the standard deviations of 3 runs of the same simulation.

The result in Figure 8.10 shows that the coefficient of variation increases with decrease in grain size. For a 5 μ m grain, the coefficient of variation for Th, U, and, ⁴⁰K are about 390%, 300% and 100% of the mean dose rates compared to $\sim$ 5%, 4% and 2% of the mean dose rates for 260 μ m grains. This large increase in coefficient of variation can be attributed to the decrease in the probability of a grain getting hit by a particle during the simulation as the grain size decreases and therefore more particles will need to be simulated to compensate for this decrease in probability. However, the result presented here is for 5×10^8 particles, any

further increase in the number of particles will result in a longer simulation time ranging from 2 months to 2 years. Another reason for this high uncertainty is because of the limitations of the MC condensed history used. For a small sized grain like $5\text{ }\mu\text{m}$, a very low cut-off value $\sim 0.1\text{ }\mu\text{m}$ is required to improve statistics. In this investigation, the cut-off value was $2\text{ }\mu\text{m}$ because of time constraints.

This result is very important because for the Martian subsurface simulations, the dosimeters will be modelled as microbes or biomarkers (DNA and amino acid) with effective sizes of $1\text{--}10\text{ }\mu\text{m}$. The large coefficient of variation seen from this result means that the dose rate distribution results that will be obtained for the microbes will be dominated by high levels of uncertainties and will thus not be representative of the real microscopic dose rate variation. Given that the simulation of more particles is not an option because of the time limitation, an alternate option will be to simulate the microbes as larger grains and then use grain size attenuation factors, calculated from the Al_2O_3 self-dose factors in Section 6.3, to correct the dose rate distribution to the required grain size, i.e., $5\text{ }\mu\text{m}$. This approach will be tested in the next section.

8.3 Converting dose rate distributions inter–grain size using attenuation factors

In this section we investigate the accuracy of inter–grain size dose rate distribution conversion. If we can show that one can convert a dose rate distribution obtained for a particular grain size to a dose rate distribution for another grain size while preserving the microscopic dose rate variation, then we can apply this to simulate the dose rate distribution of the tiny microbes ($\sim 5\text{ }\mu\text{m}$) on Mars. In other words, the simulated dose rate distributions of smaller grains are dominated by high levels of uncertainty from the simulation itself, which compromise the microscopic dose rate variations inherent to the sediment. Our solution is to simulate the microbes as larger grains ($260\text{ }\mu\text{m}$), and then use attenuation factors to convert the dose rate

distribution to that of 5 μm (grains representing microbes). Figure 8.11 shows the result of one of this test for 20 μm and 260 μm grains. Figure 8.12 shows the result for 20 μm and 100 μm grains. The conversion process, using the 20 μm and 260 μm grains for the U-series is demonstrated as follows:

First, the dose rate to each 20 μm grain is calculated from the fraction of the infinite matrix dose deposited in the grain (D/E), the energy conversion factors (F) of Guérin et al. [29] for (β , U), and the radionuclide concentration (c) of the radioactive matrix thorite.

$$\dot{D}_{20} = c \frac{D}{E} F = 485957 \frac{(2.75E - 4)}{(1.3E - 4)} 0.1457 \text{ mGy/a} = 149777.55 \text{ mGy/a} = 150 \text{ Gy/a} \quad (8.1)$$

The above dose rate is for one of the 20 μm grains. Next, the same process is applied to calculate the dose rate to one of the 260 μm grain

$$\dot{D}_{260} = c \frac{D}{E} F = 485957 \frac{(7.60E - 5)}{(1.3E - 4)} 0.1457 \text{ mGy/a} = 41393.07 \text{ mGy/a} = 41 \text{ Gy/a} \quad (8.2)$$

The Al_2O_3 self-dose factors presented in Section 6.3 are used to calculate attenuation factors for each grain size based on the superposition principle. For example, the beta self-dose factor for 20 μm and 260 μm are 0.049 and 0.217 respectively. Their attenuation factors are simply $(1 - 0.049) = 0.951$ and $(1 - 0.217) = 0.783$ respectively. To convert the $\dot{D}_{260}$ to $\dot{D}_{20}$,

$$\dot{D}_{20_{corrected}} = \frac{(0.951)}{(0.783)} \dot{D}_{260} = \frac{(0.951)}{(0.783)} 41 \text{ Gy/a} = 49.8 \text{ Gy/a} \sim 50 \text{ Gy/a} \quad (8.3)$$

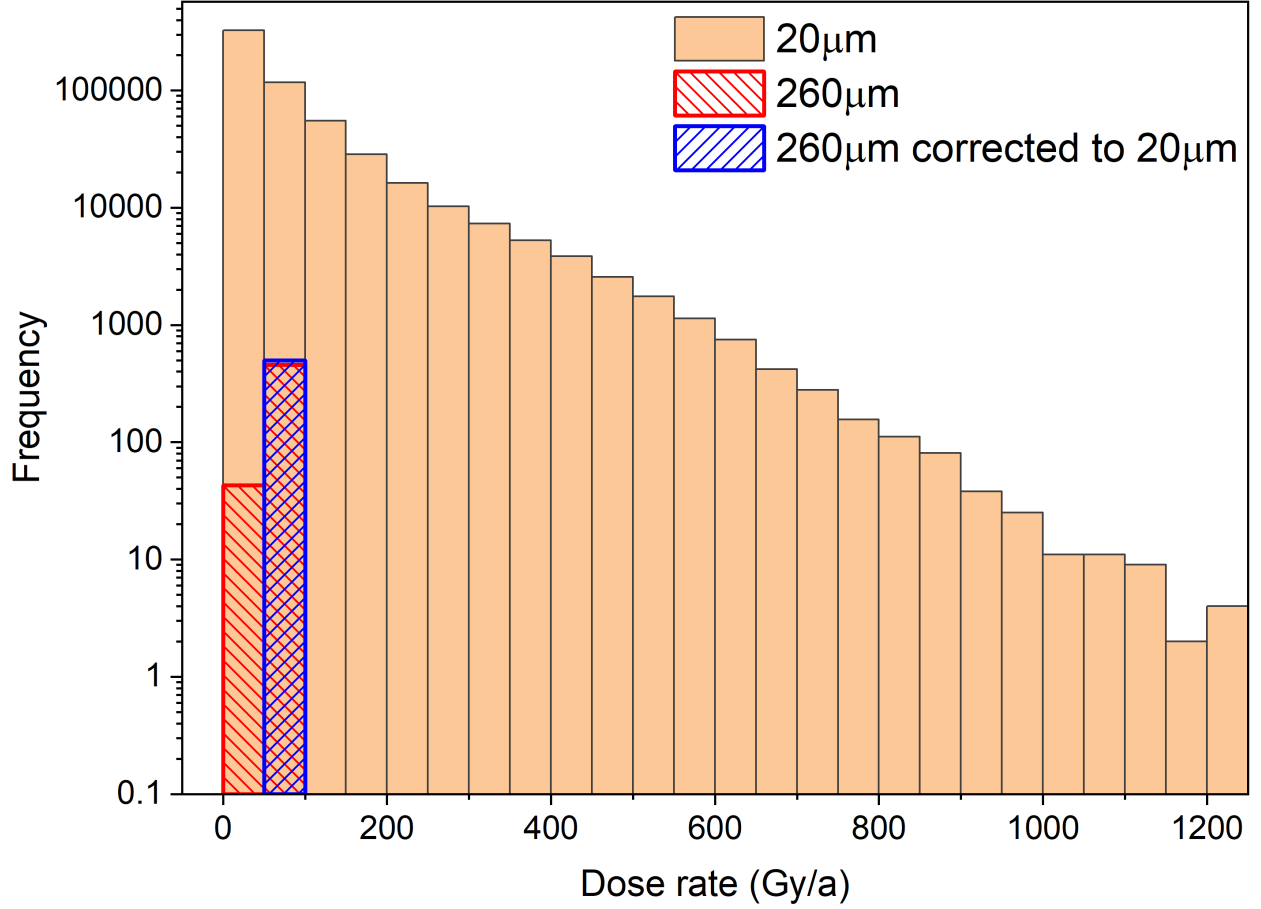


Figure 8.11: Comparison of a simulated 20 μm grain dose rate distribution to a simulated 260 μm grains dose rate distribution, which was then converted to a 20 μm grain dose rate distribution using beta attenuation factors. The simulated spectrum is the U-series. The dosimeters are Al_2O_3 grains. The number of simulated particles was 10^8 .

In Figure 8.11, the mean dose rate for the simulated 20 μm dose rate distribution is (70.9 ± 99.6) Gy, i.e. a standard deviation of about 140%. The mean dose rate for the 260 μm grains dose rate distribution converted to 20 μm grains dose rate distribution is (69.7 ± 6.7) Gy, i.e., a standard deviation of 9.61%. The mean dose rate for the original 260 μm dose rate distribution is (57.3 ± 5.5) Gy.

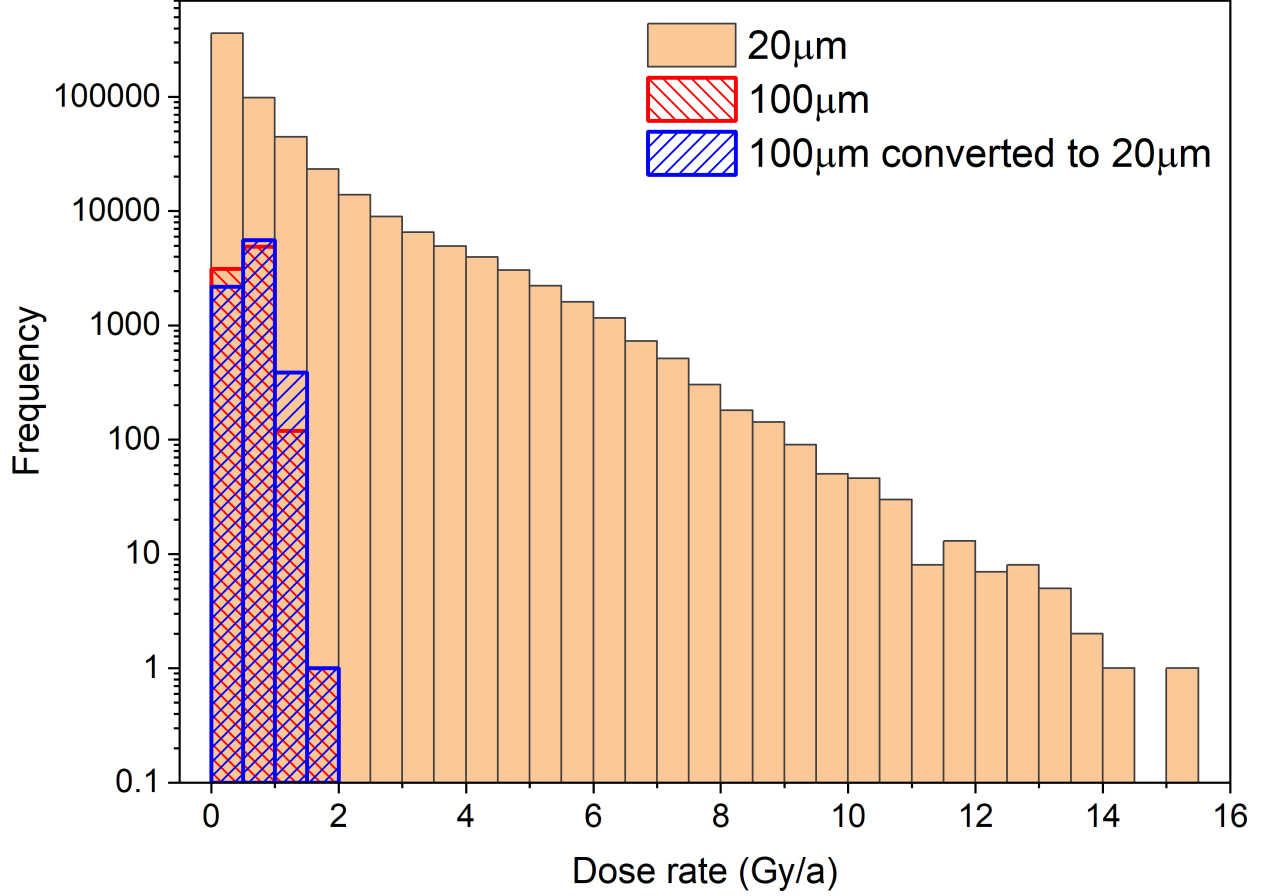


Figure 8.12: Comparison of a simulated 20 μm grains dose rate distribution to a simulated 100 μm grains dose rate distribution, which was then converted to a 20 μm grains dose rate distribution using beta attenuation factors. The simulated spectrum is the Th-series. The dosimeters are Al_2O_3 grains. The number of simulated particles was 10^8 .

In Figure 8.12, the mean dose rate for the simulated 20 μm dose rate distribution is (0.64 ± 1.08) Gy, i.e. a standard deviation of about 169%. The mean dose rate for the 100 μm grains dose rate distribution converted to 20 μm grains dose rate distribution is (0.63 ± 0.20) Gy, i.e., a standard deviation of 31%. The mean dose rate for the original 100 μm dose rate distribution is (0.56 ± 0.18) Gy. The agreement between the mean dose rates of the simulated 20 μm dose rate distribution and the 20 μm grains dose rate distribution obtained from conversion using grain-size attenuation factors shows that the inter-grain dose rate distribution conversion method is accurate for the mean dose rate. However, it is the width of the dose rate distribution that is of interest in this work and not just the mean. But the width of the simulated distribution (169%), dominated by simulation uncertainties cannot

be compared to the width of the calculated distribution. Therefore, a comparison needs to be made between grain sizes with distributions that are not dominated by the simulation uncertainties. For example, 300 μm or 350 μm and 260 μm . This result and the result of the comparison with other grain sizes are presented in Tables 8.2 and 8.3.

In the tables, the simulated fraction of the infinite matrix dose deposited i.e. (D/E) for a specific grain size is listed under "simulated". The (D/E) distribution of that specific grain size calculated from 260 μm grains using attenuation factors are listed under "calculated". The mean of the distributions, as well as the standard deviations are listed. To obtain the dose rate, you simply multiply (D/E) by the respective energy conversion factor and radionuclide concentration (see Eq. 8.1)

Table 8.2: Comparison of beta dose simulated to beta dose calculated from 260 μm grains simulated beta dose using attenuation factor correction for Al_2O_3 dosimeters of different grain sizes. The dose is represented as dose fractions.

Grain size (μm)	Th		U		K	
	Simulated	Calculated	Simulated	Calculated	Simulated	Calculated
350	0.66 ± 0.07	0.68 ± 0.09	0.73 ± 0.05	0.73 ± 0.08	0.67 ± 0.03	0.67 ± 0.06
300	0.70 ± 0.07	0.70 ± 0.09	0.76 ± 0.07	0.76 ± 0.09	0.69 ± 0.03	0.70 ± 0.06
260	0.72 ± 0.08	0.72 ± 0.10	0.78 ± 0.07	0.78 ± 0.09	0.72 ± 0.04	0.72 ± 0.06
100	0.90 ± 0.23	0.88 ± 0.10	0.85 ± 0.27	0.83 ± 0.11	0.82 ± 0.12	0.79 ± 0.07
90	0.86 ± 0.30	0.84 ± 0.11	0.91 ± 0.26	0.88 ± 0.10	0.83 ± 0.14	0.80 ± 0.07
45	0.87 ± 0.31	0.82 ± 0.07	0.94 ± 0.56	0.92 ± 0.11	0.91 ± 0.64	0.90 ± 0.13
20	0.96 ± 1.62	0.94 ± 0.13	0.96 ± 1.35	0.95 ± 0.11	0.91 ± 0.77	0.83 ± 0.07

Table 8.3: Comparison of alpha dose simulated to alpha dose calculated from 260 μm grains simulated alpha dose using attenuation factor correction for H_2O dosimeters of different grain sizes

Grain size (μm)	Th		U	
	Simulated	Calculated	Simulated	Calculated
260	0.18 ± 0.01	0.18 ± 0.01	0.14 ± 0.01	0.14 ± 0.01
200	0.23 ± 0.01	0.23 ± 0.02	0.19 ± 0.02	0.19 ± 0.02
100	0.42 ± 0.07	0.39 ± 0.03	0.36 ± 0.06	0.34 ± 0.03
20	0.83 ± 0.63	0.87 ± 0.07	0.83 ± 0.67	0.85 ± 0.07

In Tables 8.2 and 8.3, the standard deviations of the calculated dose fraction distribution (i.e calculated from grain size attenuation factors) for grain sizes like 350, 300, 260 or 200 μm that are least affected by the simulation uncertainties show reasonable agreement with the simulated values for both beta and alpha radiation. For the smaller grains, it is impossible to tell if the width of the calculated distributions represents the real width of the simulated distribution without the influence of the simulation uncertainty. However, based on the reasonable agreement of the calculated standard deviations with the simulated standard deviations for grains larger than or equal to 200 μm , we can conclude that the use of attenuation factors is a good approximation for obtaining the simulation-uncertainty-free dose rate distribution for smaller grains like 5 μm , without having to simulate 10^{10} or more particles which can take anywhere from 6–12 months to be completed with our current computer.

8.4 Conclusions relevant for Martian simulations

For the simulation of the microscopic dose rate distribution in the subsurface of Mars, our simulation space must fulfill infinite-matrix conditions. In this chapter the effect of different geometry-related simulation parameters on the results, specifically on the standard deviations (coefficient of variations) of the dose rate distributions was evaluated.

The results presented in this chapter have shown that the precision of the Monte Carlo (MC) simulated microscopic dose rate variation using *DosiSed* depends on the following parameters:

1. The number of simulated particles
2. The volume fraction of the dosimeter grains compared to the total volume of the defined simulation space or voxel(s)
3. The voxel size
4. Total volume of the dosimeter voxel
5. Dosimeter grain size

The impact of voxel size and volume was unexpected as the method of defining infinite matrix conditions is independent of the voxel size and volume but depends on the range of charged particles. It must be stressed that while the effect of the simulation geometry is very strong on the spread or standard deviation of the dose rate distribution, it is almost negligible when it comes to the mean dose rate of the distribution. This trend can be seen from the attenuation factor conversion results presented.

A set of simulation geometry parameters was established to ensure that the microscopic dose rate variation is not or is as little as possible compromised by geometry-related parameters. Parameters were selected for which the result for infinite matrix condition is close to or within the 5.5% level of uniformity established from experimental gamma irradiation of $\text{Al}_2\text{O}_3\text{:C}$

dosimeters. The set of optimum simulation geometry parameters are summarized in Table 8.4. The established optimum simulation geometry parameters were used for modelling the subsurface of Mars in Chapter 9.

Table 8.4: Simulation Geometry Parameters used to simulate the radiation distribution in the Martian subsurface

Parameters	Value
Volume fraction (%)	1–5
Voxel size (mm ³)	$1 \times 1 \times 1 - 5 \times 5 \times 5$
Total volume (mm ³)	$< 20 \times 20 \times 20$
No of simulated particles	5×10^8

For the effect of grain sizes on the simulation, a drastic increase in statistical uncertainties was observed as the grain size decreases. Given the small size of the microbes ($\lesssim 5\mu\text{m}$), this poses a big problem for the Martian subsurface. Because of time constraints, it was impossible to run realistic simulations for grains $\lesssim 5\mu\text{m}$ which require simulating at least 10^{10} or more particles. To tackle this problem, a solution was devised. The solution involves simulating the dose rate distribution at a larger grain size and using grain size attenuation factors to convert the dose rate distribution of the larger grain with minimum statistical uncertainty to a dose rate distribution of the required smaller grain size.

The result shows that the expected mean of a $5\mu\text{m}$ dose rate distribution can be correctly calculated from corrected $260\mu\text{m}$ dose rate distribution. The main goal of our simulation, however, is to obtain dose rate distributions and not mean dose rates. The result also shows that the expected width of the simulated dose rate distribution for grain sizes $\geq 200\mu\text{m}$ can be correctly calculated from attenuation-factor-corrected $260\mu\text{m}$ dose rate distribution. While this result was encouraging, the difference in grain sizes between $\geq 200\mu\text{m}$ and $\leq 5\mu\text{m}$ is not trivial. Alpha particles for example, come to a full stop in $260\mu\text{m}$ grains but pass

through $5\mu\text{m}$ grains. This effect might become exacerbated in real sediment where grains come in irregular shapes and sizes so that the incident angle becomes also a factor.

Nevertheless, based on the precision of the dose rate distribution conversion for larger grains, our conclusion is that the microscopic dose rate variation of the microbes ($\sim 5\mu\text{m}$ in size) residing in the pore spaces in the Martian subsurface can be approximated by first simulating the microbes as $260\mu\text{m}$ grains and then using grain size attenuation factor to convert the microscopic dose rate variation to that of $5\mu\text{m}$. It must be stressed that the use of the correction factors for the microbes on Mars is based on the results obtained at grain sizes more than 50 times larger than the size of microbes and thus must be interpreted as such.

Chapter 9

Choosing Model Parameters for the Martian Subsurface and Calculating Survival Probabilities for Microbes

9.1 Introduction

In chapter 8, optimum parameters for enforcing infinite matrix conditions for the Monte Carlo simulations were determined. It was found that the small grain size of microbes is the biggest challenge, as the small microbe “grains” have a small probability of being hit by simulated particles, resulting in bad statistical precision of the dose distributions. One way to tackle this problem would be to simulate more particles, i.e. in the order of billions and trillions. However, due to time constraints, this was not an option in the framework of this dissertation. To circumvent the grain size problem, an approach was suggested.

It involves simulating the dose rate distribution at a larger grain size and using grain size attenuation factors to convert the dose rate distribution of the larger grain with some statistical uncertainty to a dose rate distribution of the required smaller grain size. It was found that average dose values agree with expected values, however, the approach is limited

by the inability to verify the precision of the dose rate distributions. In this chapter final simulation results for the Martian subsurface are presented, but due to the use of conversion factors, the dose rate distributions must be treated as an approximation.

Before performing the final simulations for Mars, it was necessary to select the parameters that describe the chemical composition and radioactivity of the Martian subsurface. The parameters are described in the first part of this Chapter. The final goal of this work is to obtain survival probabilities for microbes and probabilities for the preservation of bio-markers. The second part of this chapter describes the framework for converting simulated dose rate distributions into probability distributions.

9.2 Simulation Parameters for the Martian subsurface

So far, no samples from the surface of Mars have ever been returned to Earth. The available information, obtained from meteorites and Mars missions, has been discussed in detail in Chapter 2. This section describes the selected input parameters for chemical compositions and radioactivity.

9.2.1 Parameters for chemical composition

Table 9.1 summarizes the chemical compositions and physical properties that were selected as input parameters for the simulations. The following materials and their chemical compositions were chosen:

1. The meteorites Shergotty, which represents the most common class of meteorites, and ALH 84001, which is one of the oldest meteorites.
2. Five scenarios that are based on the mineral compositions of distinct rock types analyzed at three different rover landing sites. They represent (i) basaltic andesite (a mixture of basalt and andesite) from Ares Vallis (Pathfinder); (ii and iii) olivine-rich basaltic regolith from Gale Crater (Curiosity), the sulphur-rich "Paso Robles" and one silicate

rich; (iv) samples from Gusev Crater (Spirit) which show enrichment with mineral salts that suggest the past existence of shallow salty ponds; (v) dust from Gale Crater (similar in composition to Gale Crater)

3. A scenario to capture the "bulk composition" of Mars. This is based on the model for the bulk silicates on Mars proposed by Taylor [82].

Mitranov et al. (2004) [208] estimated the water-ice contents for numerous locations on Mars based on neutron measurements using the Mars Odyssey spacecraft. They showed that the water-ice content in the subsurface can reach up to 53% by mass. Based on these ice content estimations, and in order to represent northern and southern polar regions on Mars, the simulated scenarios are a mixture of 50% ice and 50% of the sedimentary material by weight. The only exception was the scenario for sediments dominated by dust, which consist of 5% Martian dust (composition analyzed by the Curiosity rover) and 95% ice by weight. Dust (particle size $< 63 \mu\text{m}$) is pervasive on Mars as suspended particles in the atmosphere, and as air fall deposits trapped in ice due to melting and freezing of ice coated by the air fall deposits [209]. This scenario was used to represent the north polar regions of Mars.

Table 9.1: Bulk chemical compositions and physical properties of the Martian meteorites and landing-site regoliths/rocks simulated. For the “North Pole” simulations 95% ice content and 5% sediment were assumed. For all other simulations the ratio was 50%-50%. Percentage is (% wt).

Mineral Oxides	Meteorites		Landing sites					Bulk
	Shergotty ^a	ALH 84001 ^a	Pathfinder Balsatic Andesite ^b	Gusev Crater Paso Robles ^c	Silica-rich Gusev Crater ^c	Rocknest Gale Crater ^c	Martian dust North pole ^d	Silicate Mars ^e
SiO ₂	51.17	52.60	57.05	21.82	46.37	42.94	39.83	43.79
Al ₂ O ₃	6.86	1.28	11.00	4.13	9.97	9.45	9.03	3.05
FeO	19.35	17.43	15.70	21.02	15.76	19.22	22.28	18.14
MgO	9.28	24.90	1.51	5.54	9.07	8.70	8.42	30.56
CaO	9.58	1.81	8.09	6.85	6.01	7.30	7.31	2.43
Na ₂ O	1.39	0.14	2.46	1.64	3.19	2.72	2.79	0.53
K ₂ O	0.17	0.02	1.36	0.19	0.45	0.49	0.48	0.04
P ₂ O ₅	0.67	0.01	0.95	5.62	0.93	0.94		0.15
TiO ₂	0.82	0.21	0.69	0.62	0.8	1.19	1.07	0.14
Cr ₂ O ₃	0.20	1.14		0.04	0.29	0.49		0.73
MnO	0.52	0.46	0.55	0.25	0.31	0.41	0.43	0.44
Cl			0.32	0.55	0.51	0.69	1.09	
SO ₃			0.32	31.73	6.34	5.46	8.45	
Density (g/cm ³)	3.30	3.44	2.80	3.25	3.25	2.81	2.81	2.90

^a Lodders (1998) [68]

^b Wänke et al. (2001) [92]

^c Ramkissoon et al.(2019) [210]

^d Berger et al. (2016) [209]

^e Taylor (2013) [82]

9.2.2 Radionuclide concentrations

The Mars Odyssey gamma ray spectrometer (GRS) was the first to do direct measurements of elemental concentrations of the entire Martian crust [211]. The principle radionuclides on Mars are the ²³⁸U, ²³⁵U, and ²³²Th decay series and ⁴⁰K. The U and Th-series emit alpha, beta and gamma radiation. ⁴⁰K emits only beta and gamma radiation. In this work, ⁴⁰K and ²³²Th concentrations were obtained from the GRS Mars Odyssey data (see Figure 9.1) [17, 211, 212]. These data comprise the abundances and distribution of K and Th measured from the detection of characteristic gamma ray photons emitted from the upper few tens of centimeters of the Martian surface.

Taylor et al. [17] argued that this result can be generalized to the deeper subsurface.

The U concentrations have not been obtained directly from GRS data due to its weak

signal-to-noise ratio [212, 213]. In this study, U concentrations were calculated from the GRS Th concentrations based on a U/Th ratio of 0.28; a value characteristic of most SNC meteorites [214, 215]. This method of obtaining U concentration has been previously applied by Dzaugis et al. (2018) [213] to study radioisotope induced H₂ production in the subsurface of Mars through water radiolysis.

For the meteorites considered, the Th, U, and K concentrations were taken from Lodders (1998) [68, 72]. For the bulk composition of mars (BSM), the concentrations were calculated as an average of concentrations from Wänke & Dreibus [81], Taylor [82] and Yoshizaki & McDonough [83]. See Table 9.2 for a summary of the radioisotope concentrations for each meteorite and location.

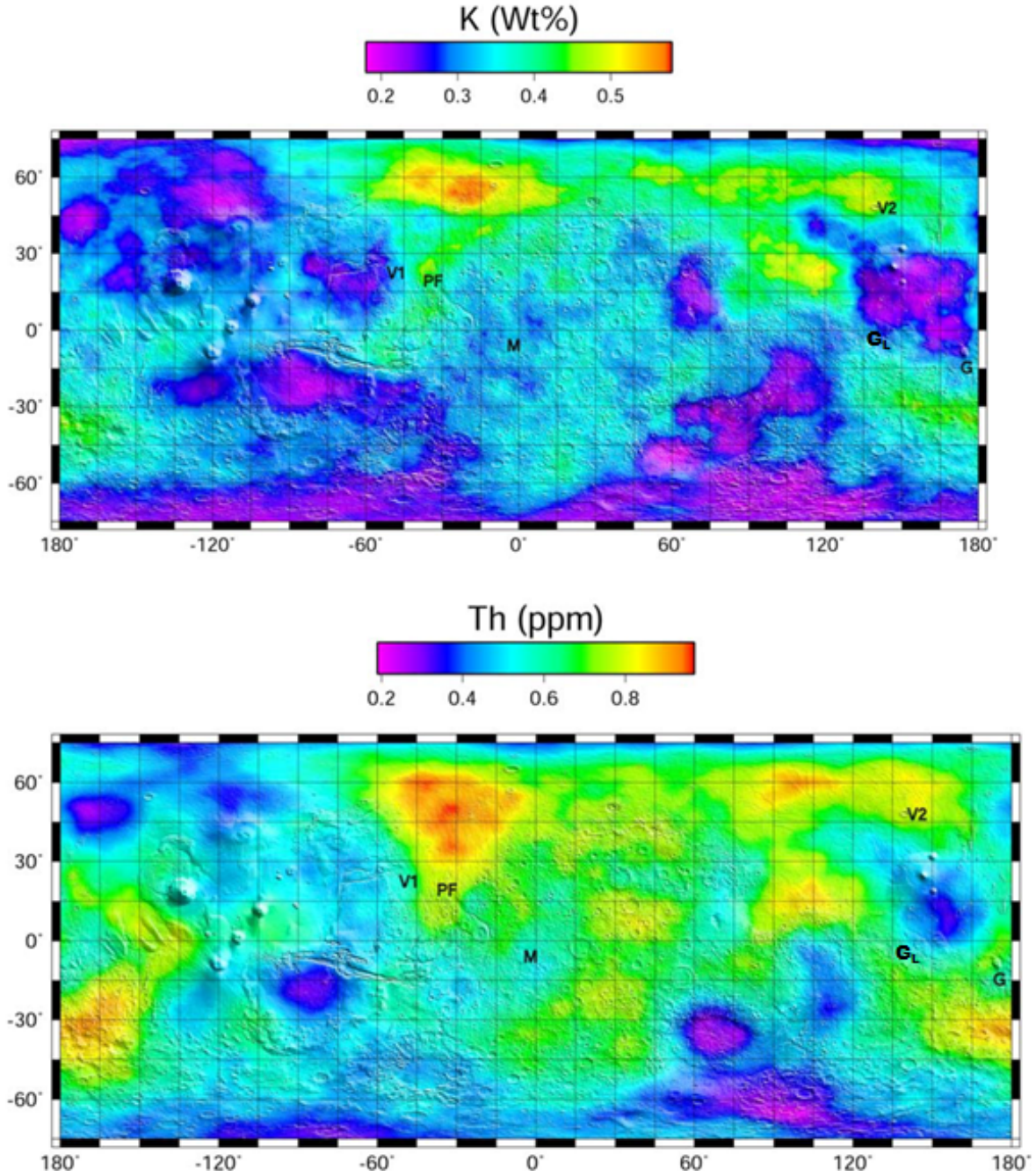


Figure 9.1: Mars radionuclide concentration map for Th and K. The concentrations were measured by the Mars Odyssey Gamma Ray Spectrometer [212]. The landing sites for some missions are also indicated: V1, Viking 1 in Chryse Planitia; V2, Viking 2 in Utopia Planitia; PF, Pathfinder in Ares Vallis; M, Opportunity in Meridiani Planum; G, Spirit in Gusev Crater, and G_L , Curiosity in Gale Crater (Fig. 1. from [17] reproduced with permission from AGU Journal).

Table 9.2: Radioisotope concentrations of the Martian meteorites and some landing sites on Mars used for the model calculations. The Th and K concentrations for the landing sites were measured by Mars Odyssey Gamma Ray Spectrometer [212] while their corresponding U concentrations were calculated using a constant U/Th ratio of 0.28.

Regolith	Uranium (ppm)	Thorium (ppm)	Potassium (%)
Shergotty ^a	0.11 ± 0.02	0.38 ± 0.08	0.144 ± 0.011
ALH 84001 ^a	0.011 ± 0.002	0.035	0.014 ± 0.005
Pathfinder Basaltic Andesite	0.219 ± 0.020	0.783 ± 0.072	0.436 ± 0.030
Gusev Crater Paso Robles	0.176 ± 0.017	0.629 ± 0.060	0.330 ± 0.020
Silica-rich Gusev Crater	0.176 ± 0.017	0.629 ± 0.060	0.330 ± 0.020
Rocknest Gale Crater	0.167 ± 0.017	0.598 ± 0.060	0.286 ± 0.020
Martian dust, Northern pole	0.048	0.17 ^b	0.077 ^b
Bulk Silicate Mars ^c	0.017 ± 0.001	0.061 ± 0.005	0.032 ± 0.003

^a Lodders (1998) [68]

^b Dzaugis et al. (2018) [213]

^c Mean of Wänke & Dreibus (1994) [81], Taylor (2013) [82], and Yoshizaki & McDonough (2020) [83]

9.2.3 Summary

A comparison of elemental compositions shows that Shergotty has more Al and Ca, but less Mg than ALH 84001, otherwise they are very similar in chemical composition. Shergotty has about 10 times more K which means it has also 10 times as much ^{40}K . The landing sites on the other hand, are all very similar in composition. There are minor differences in Al, Fe, Mg and Ca. Basaltic andesite has generally a higher nuclide concentration, Gale Crater has the lowest. In general, the nuclide concentrations of the landing sites are much higher than for the meteorites. Martian dust, as expected has a very low nuclide concentration which is diluted by the high ice content. The scenario based on bulk composition of Mars (BSM) has the lowest concentration of radionuclides.

It is not directly obvious from Table 9.2 that the radioactivity is dominated by K abundances given that the K/Th ratio on Mars is 5300 compared to 360 and 2900 for the Moon and Earth respectively. This implies that the microscopic beta dose rate in the Martian subsurface is predominantly due to beta particles from ^{40}K . The total microscopic dose rate

is a combination of all charged particles and gamma photons, but it is dominated by the alpha particles from Th-, and U-series and the beta particles from ^{40}K . The contribution from gamma radiation is expected to be very small at the microscopic level.

9.3 Methodologies for obtaining Survival probabilities/ DNA preservation probabilities

9.3.1 Survival probability

Survival probability is the probability that a microorganism survives over a given time frame under given circumstances. Survival probability can be considered for two groups; active organisms that are able to repair radiation damage, and dormant organisms that accumulate radiation damage without the mechanism for repair. The survival of the first group is limited by the intensity of the dose, i.e., the dose at an instant, and the second group by its accumulated dose over a long period of time. Furthermore, it has been suggested that active cells that are constantly regenerating are more sensitive to radiation compared to dormant cells that are not active [216]. This means that dormant cells might actually be more resistant to radiation. In a systematic study using *Bacillus anthracis*, the dose required for complete inactivation of dormant cells (spores) was reported to be about 15 times more than that of active cells [217]. Unfortunately, many studies on microbial community survival do not distinguish between active, dead, and dormant cells. However, this is of particular importance especially in ancient permafrost, where subzero temperatures preserve DNA from dead organisms and dormancy is the number one survival strategy [216].

In environments like Mars, survival probability can be modelled using some of the most radio-resistant microorganisms that are dormant. Very good candidates are the *Bacillus pumilus* SAFR-032 bacteria strain that are notoriously resistant to unfavorable conditions such as low or no nutrient availability, extreme desiccation, H_2O_2 , UV, gamma-radiation, or

chemical disintegration [218]. In fact, they have been found in extreme environments such as Sonoran desert basalt and the Mars Odyssey spacecraft after intensive sterilization in preparation for launch [45]. Another well-known radioresistant microbe is the *Deinococcus radiodurans*. This microbe has been reported to survive gamma radiation doses as high as 90kGy when irradiated under martian simulated conditions [42, 219].

Kminek et al, (2003) [119] in their study, calculated the survival probability of two bacterial spore species (dry *Bacillus thuringiensis* and *Salibacillus marismortui*) irradiated with γ rays from a ^{137}Cs source under martian conditions at a dose rate of 13.3 Gy/min.

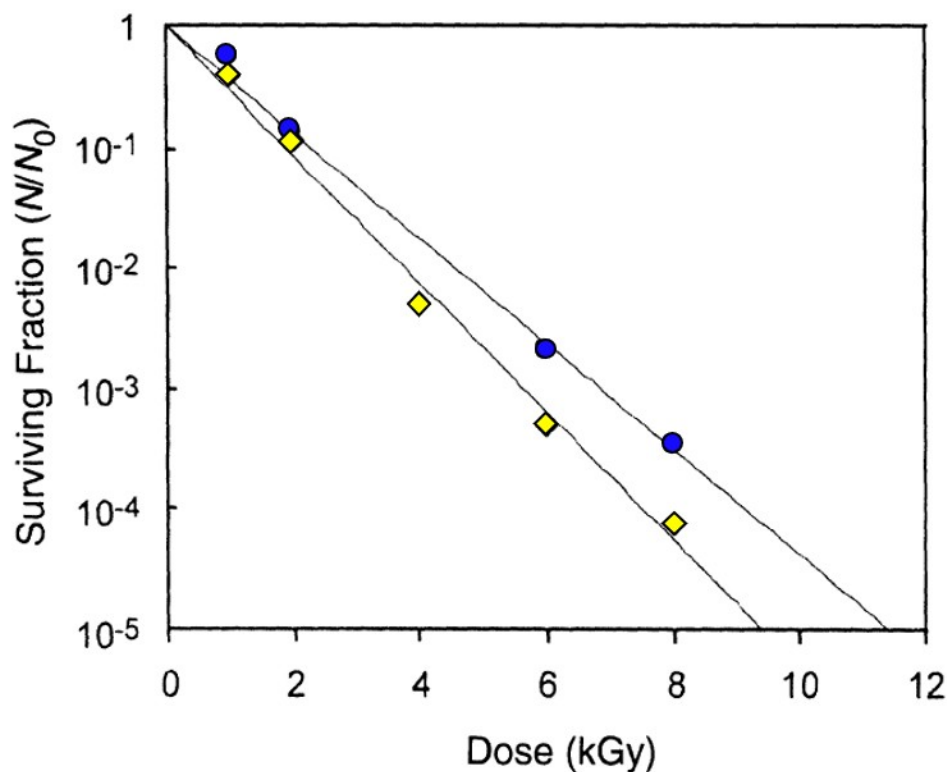


Figure 9.2: Representative dose-survival curve for dry *B. thuringiensis* spores (●) and *S. marismortui* spores (◆) in a pH 4 solution. Adapted from [119, Fig. 1]

The dose-survival curve is presented in Fig. 9.2. The relationship between the dose and the surviving fraction was expressed using a regression curve (best-fit lines on Fig. 9.2) based on a first-order exponential equation that has been used previously by Webb et al. [220]:

$$\ln (N/N_0) = -kD + n, \quad (9.1)$$

where N is the number of colonies that survived after irradiation, N_0 is the number of colonies prior to irradiation, k is the so-called inactivation constant (in units kGy^{-1}), D is the radiation dose (kGy), and n is the extrapolation number.

The inactivation constant k is used to represent the sensitivity of the cell type. It is equal to the slope of the regression line in Figure 9.2. The inverse of k is defined as D_{37} or D_0 and corresponds to the radiation dose required to reduce the dormant cell or colony population to 37% of the initial population prior to irradiation. At this dose, each cell has, on average, received one lethal hit, and those that survive must have been balanced by other cells taking two or more lethal hits [42].

Table 9.3 lists the temperature-corrected radiation inactivation constant k , and the resulting lethal dose values for the microbial strains considered in this dissertation. Some of the values were directly taken from the literature, others required analysis of published data. Details can be found in Appendix E.

The survival time of a microbe was calculated as:

$$\text{Survival time (yr)} = \text{Lethal Dose (Gy)} / \text{Dose Rate (Gy/yr)} \quad (9.2)$$

In other words if we know the dose rate a microbe is exposed to, then one can calculate how long it will take for the microbe to accumulate a dose commensurate to its lethal doses, assuming dormancy.

Table 9.3: Radiation Inactivation Constant k , for some dry microbial and some low temperature frozen microbial strains, and their resulting lethal dose values.

Microbes	Temp. corr. k (kGy ⁻¹)	Lethal dose (kGy)
<i>B. subtilis</i>	0.938 ± 0.028	24.97 ± 0.74
<i>B. thuringiensis</i>	0.926 ± 0.009	25.29 ± 0.25
<i>B. pumilus</i> SAFR-032	0.113 ± 0.082	207.26 ± 150.74
<i>Brevundimonas</i> sp. MV.7	1.902	12.31
<i>Rhodococcus</i> sp. MV.10	4.318	5.42
<i>Pseudomonas</i> sp. MV.27	10.271	2.28
<i>D. radiodurans</i>	0.242 ± 0.164	96.78 ± 65.61

9.3.2 Preservation of biological information

Complex organic molecules or biological signatures that might allow tracing relict Martian life can be progressively destroyed by accumulated dose [32]. The effects of ionizing radiation on biological molecules are typically studied under two conditions. (1) In dilute aqueous solution, where the effect is entirely due to production of highly reactive diffusable radicals from the action of ionizing radiation on water. And (2) in the dry state, where the effect is highly dependent on the number and localized distribution of the energy loss events, with the main process being the formation of free radicals by the loss of hydrogen atoms as observed from electron spin resonance experiments. In general, irradiation of biological macro-molecules such as proteins, nucleic acids, lipids, and polysaccharides results in both non-random and random fragmentation and aggregation which ultimately leads to the inactivation of such molecules in a cell [221]. At low temperatures in frozen conditions, fragmentation in the form of breakage of the polypeptide backbone is the major damage seen [222, 223]. Filali-Mouhim

et al. (1997) [222] demonstrated that an accumulated dose of 70 kGy (1 Gy/s dose rate) was enough to break lysozyme to at least 11 different fragments. It has also been reported that radiation reduces the activity of an enzyme exponentially and that there is a correlation between the molecular mass of enzymes and their sensitivity to radiation [221, 224, 225]. The percentage of molecular bonds P broken by radiation was modelled by Mohler, 1958 [226] as a function of radiation dose.

$$P = 10^{-4} GMD \quad (9.3)$$

where D is dose in Gy, M is the molecular mass in atomic units and G is a number of molecules inactivated per 100 eV of absorbed energy. The value for G ranges from 0.03 to 3.0 and is dependent on dose rate [28, 221, 227]. Pavlov et al. (2002) using Eq. 9.3 concluded that total destruction of macro-molecules in the surface layers of Mars requires about 1 billion years. However, there is evidence that fragmentation induced by radiolysis in protein is dependent not on molecular mass but on the surface area of the protein exposed to the solvent. This is based on observation that, given the same dose, denatured proteins with larger surface area exposed to solvent compared to native ones experience more fragmentation [228].

Furthermore, Kminek and Bada (2006) modelled the radiolytic decomposition of dry amino acids after exposure to gamma rays by the same exponential function as in Eq. 9.1. In this case, N is the amino acid abundance after irradiation, N_0 is the amino acid abundance before irradiation, k_R is the radiolysis constant in MGy^{-1} ($10^6 \text{ Gy} = 1 \text{ MGy}$), and D is the radiation dose in MGy. The result also showed that the calculated radiolysis constants are linearly proportional to the molecular weight of the studied amino acids. Pavlov et al. [229], used the linear dependence of the radiolysis constant and molecular weights, and extended the range from 100–200 g/mol [230], to 50–500 via linear extrapolation of the original data.

In this work, two categories of organic compounds will be considered: "light" organic compounds with mass equal to or less than 200 g/mol (i.e., $k_R = 0.12$) and "heavy" organic compounds with mass greater than 200 g/mol (i.e., $k_R = 0.68$). Examples of "light" organic

compounds are amino acids such as Alanine, Aspartic acid, Glycine, Glutamic acid, Histidine etc. Organic compounds such as RNAs, DNAs and proteins are "heavy" organic compounds.

To determine how long it will take for radiation to destroy evidence of these organic materials in the subsurface of Mars, Eq. 9.2 and Eq. 9.1 were combined and rearranged to obtain:

$$\text{Time (yr)} = \frac{-\ln(N/N_0)}{k_R \dot{D}} \quad (9.4)$$

Where $\dot{D}$ is the dose rate and $N/N_0 = 10^{-3}$. The latter is based on the assumption that organic materials become undetectable, when their initial abundance has been reduced by a factor of 1000 [229].

9.3.3 An Example: Calculation of survival probabilities for microbes and detection probabilities for biomarkers on Mars

To illustrate an example, let's take a hypothetical dose rate distribution for 100 microbes (Figure 9.3).

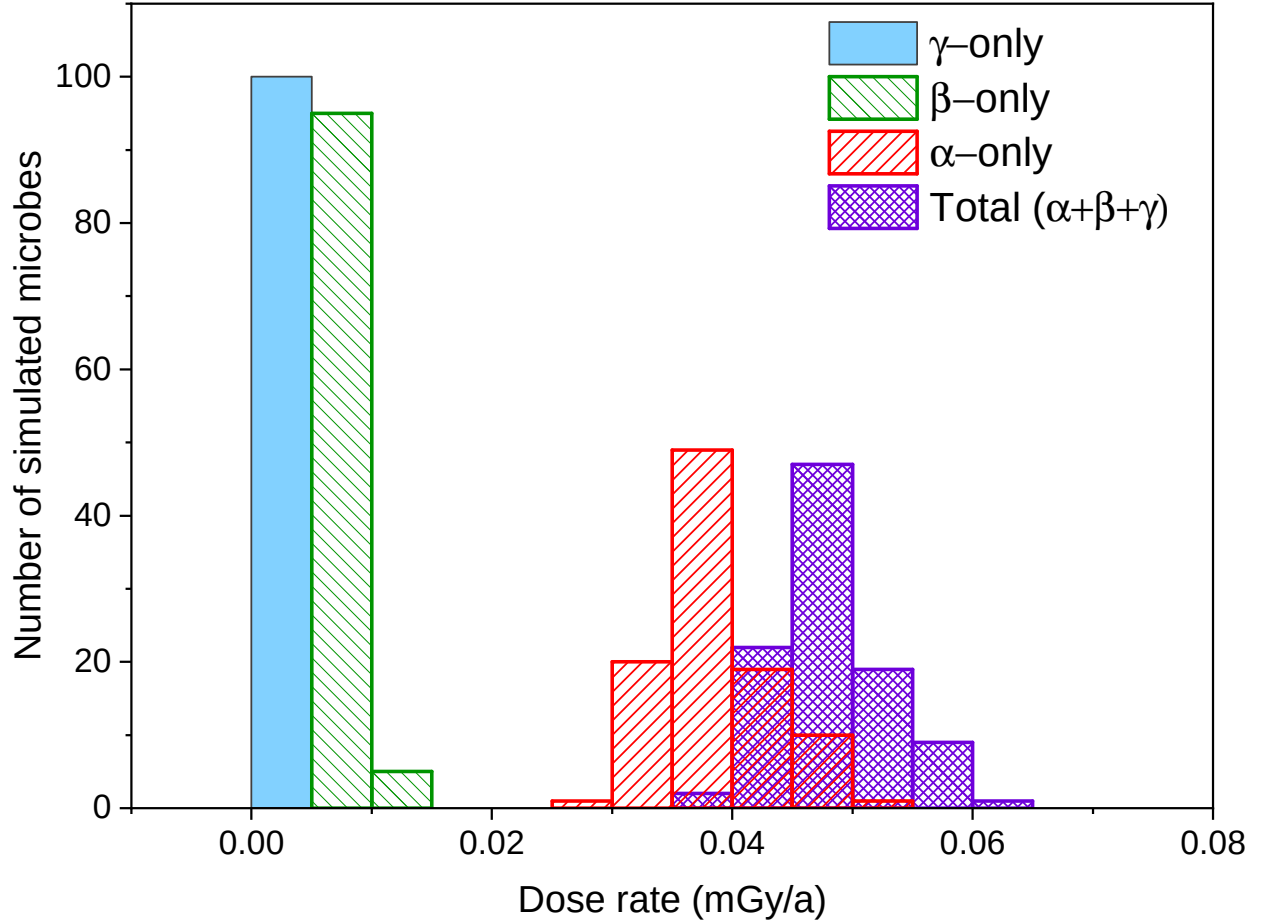


Figure 9.3: Hypothetical dose rate distributions used in the example.

The dose rate distributions were obtained by performing each particle (α , β , and γ) simulation with the same sediment setup, so that each sedimentary grain and microbe was always in the exact same spot over multiple runs of the simulation. The alpha, beta or gamma dose rate values for each microbe were therefore obtained separately. The dose rate values for the three particles were then added for each microbe individually and the resulting distribution of the total dose rate was obtained. Two total dose rate distributions were calculated. One where the dose rate values for alpha, beta and gamma are added directly. And another one that accounts for the quality factor of alpha radiation. This was done by weighting the alpha portion of the dose distribution with a quality factor of 20 before adding it to beta and gamma. This correction accounts for the higher biological effectiveness of alpha radiation in causing damage to cells compared to beta and gamma radiations.

Each of the dose distributions in Figure 9.3 was divided into smaller bins using frequency counting. For example, the gamma dose rate distribution for the 100 microbes can be binned as shown in Table 9.4.

Table 9.4: Demonstration of how the survival probabilities are calculated from dose rate distributions

Bin center	Frequency Count	Cumulative Count	Cumulative Frequency	Time (billion years)	Survival probabilities
6.75E-5	1	1	0.01	369.93	0.00
7.25E-5	1	2	0.02	344.41	0.01
7.75E-5	11	13	0.13	322.19	0.02
8.25E-5	14	27	0.27	302.67	0.13
8.75E-5	14	41	0.41	285.37	0.27
9.25E-5	17	58	0.58	269.95	0.41
9.75E-5	17	75	0.75	256.10	0.58
1.025E-4	9	84	0.84	243.61	0.75
1.075E-4	7	91	0.91	232.28	0.84
1.125E-4	5	96	0.96	221.96	0.91
1.175E-4	3	99	0.99	212.51	0.96
1.225E-4	1	100	1	203.84	0.99
Initially, there were 100 microbes which is equivalent to a survival prob. of 1					1

In Table 9.4, column 1 shows the bin centers. In column 2 and 3, the frequency counts for each bin and the cumulative counts are calculated. The cumulative frequency is calculated by dividing each cumulative count by 100, i.e. the total number of microbes simulated. This cumulative frequency is also used as survival probability (See column 6). The survival time in billion years is calculated by dividing the lethal dose for a specific bacterial strain (Table 9.3) by the bin center of the dose rate.

For example, in the beginning, there were 100 microbes, which is equivalent to a survival probability of 1. Out of the 100 microbes, one was exposed to the highest dose rate of 1.225E-4 mGy/a. At this dose rate, it will accumulate its lethal dose of 24.97 kGy (*B. subtilis*) relatively faster than the other microbes. It will reach its lethal dose in 203.84 billion years. Once this time is reached, the survival probability becomes 0.99. Similarly, three microbes were exposed to a dose rate of 1.175E-4 mGy/a. At this dose rate, they will accumulate the lethal dose of 24.97 kGy (*B. subtilis*) in 212.51 billion years (~ 9 years later after the first microbe "died"). Once this time is reached, the three microbes become "dead" also, and only 96 microbes will be remaining. The survival probability becomes 0.96. This will continue until all the microbes are dead.

The same approach is used for probability of preservation of the bio-marker. The respective radiolysis constants k_R are used on Eq. 9.4 to obtain the time in years.

9.4 Dose distributions for Martian subsurface scenarios

9.4.1 Model Parameters

In this work, the microbes were modelled as water (H_2O , density = 1 g/cm³) since most cells are made up of at least 70% of water [231]. Biomarkers like DNA and amino acids were also modelled as water but with a density of 1.7 g/cm³ and 1.22 g/cm³ respectively [231–233]. The model used is similar to the one presented for infinite matrix simulations in Section 8.1. The microbes are modelled as 260 μm spherical grains with a density of 5.2×10^2 to 6×10^4 cells/g based on the abundance of culturable bacteria in Antarctic Dry Valley permafrost reported by Gilichinsky [234]. The "microbial grains" are surrounded by matrix and sedimentary grains from the selected compositions discussed in Section 9.2. A break-down of how the 50% ice and 50% sediment composition by weight were distributed between matrix and sediment in the model is shown schematically in Figure 9.4. The grain size distribution was taken from the report by De Vera et al. [235], where they presented the grain size distribution of the Martian regolith in terms of early acidic (P-MRS) and late basic (S-MRS) compositions. Based on the breakdown in Figure 9.4, 70% of the radiation was simulated from the coarse grains while the remaining 30% was simulated from the matrix due to the 30% of the sediment included in the matrix. The IM conditions discussed in Section 8.1 were enforced during the simulation.

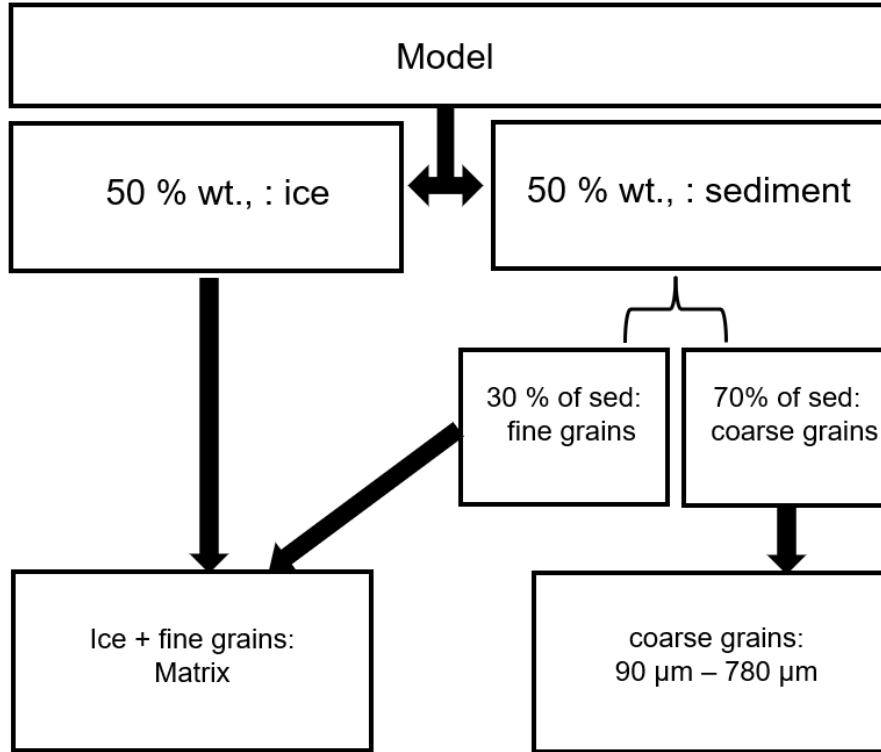


Figure 9.4: Breakdown of the model.

See Appendix C for sample bulk model parameter calculations using the densities and chemical compositions of the microbes, sediment and ice. The goal of this section is to present and discuss the result of the simulated equivalent dose rate distribution of microbes and biomarkers in the different Martian subsurface scenarios discussed in Section 9.2. After modelling the microbes and biomarkers as $260\ \mu\text{m}$ grains, their dose rate distributions were converted to those of $5\ \mu\text{m}$ microbes and biomarkers respectively using the attenuation factors in Table 9.5. The Table also contains attenuation factors for other grain sizes in case one needs to do conversion between grain sizes other than $260\ \mu\text{m}$ and $5\ \mu\text{m}$. The procedure is described in Section 8.3.

Table 9.5: Alpha and beta attenuation factors for H_2O , used for conversion of $260 \mu\text{m}$ distributions into distributions for $5 \mu\text{m}$ grains.

Grain size (μm)	Beta			Alpha	
	U	Th	K	U	Th
5	0.992 ± 0.045	0.990 ± 0.045	0.999 ± 0.045	0.970 ± 0.044	0.975 ± 0.044
100	0.940 ± 0.043	0.919 ± 0.041	0.984 ± 0.044	0.350 ± 0.016	0.397 ± 0.018
200	0.912 ± 0.041	0.878 ± 0.039	0.968 ± 0.044	0.191 ± 0.009	0.233 ± 0.011
260	0.898 ± 0.040	0.857 ± 0.039	0.958 ± 0.043	0.148 ± 0.007	0.181 ± 0.008
350	0.879 ± 0.040	0.831 ± 0.037	0.944 ± 0.042	0.110 ± 0.005	0.135 ± 0.006

9.4.2 Microscopic dose rate distributions for the Martian subsurface

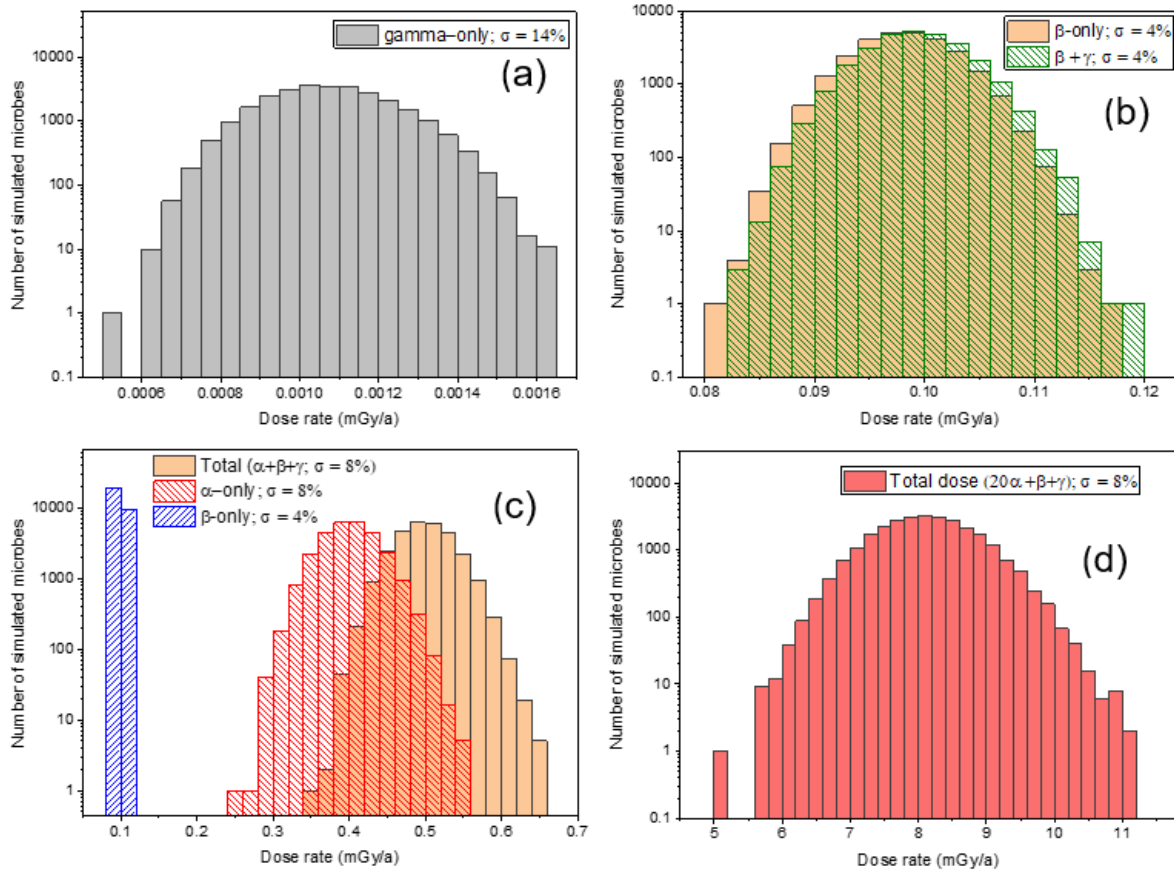


Figure 9.5: Microscopic dose rate distribution of microbes in the shergotty-type Martian subsurface permafrost. (a) Gamma dose rate distribution only. (b) Beta only and Beta + Gamma dose rate distributions. (c) Alpha only, beta only and total sum of alpha + beta + gamma dose rate distributions. Here the dose rate values were added directly. The total dose rate distribution in (d) was obtained by multiplying the alpha radiation portion of the dose distribution by 20 to account for the alpha quality factor.

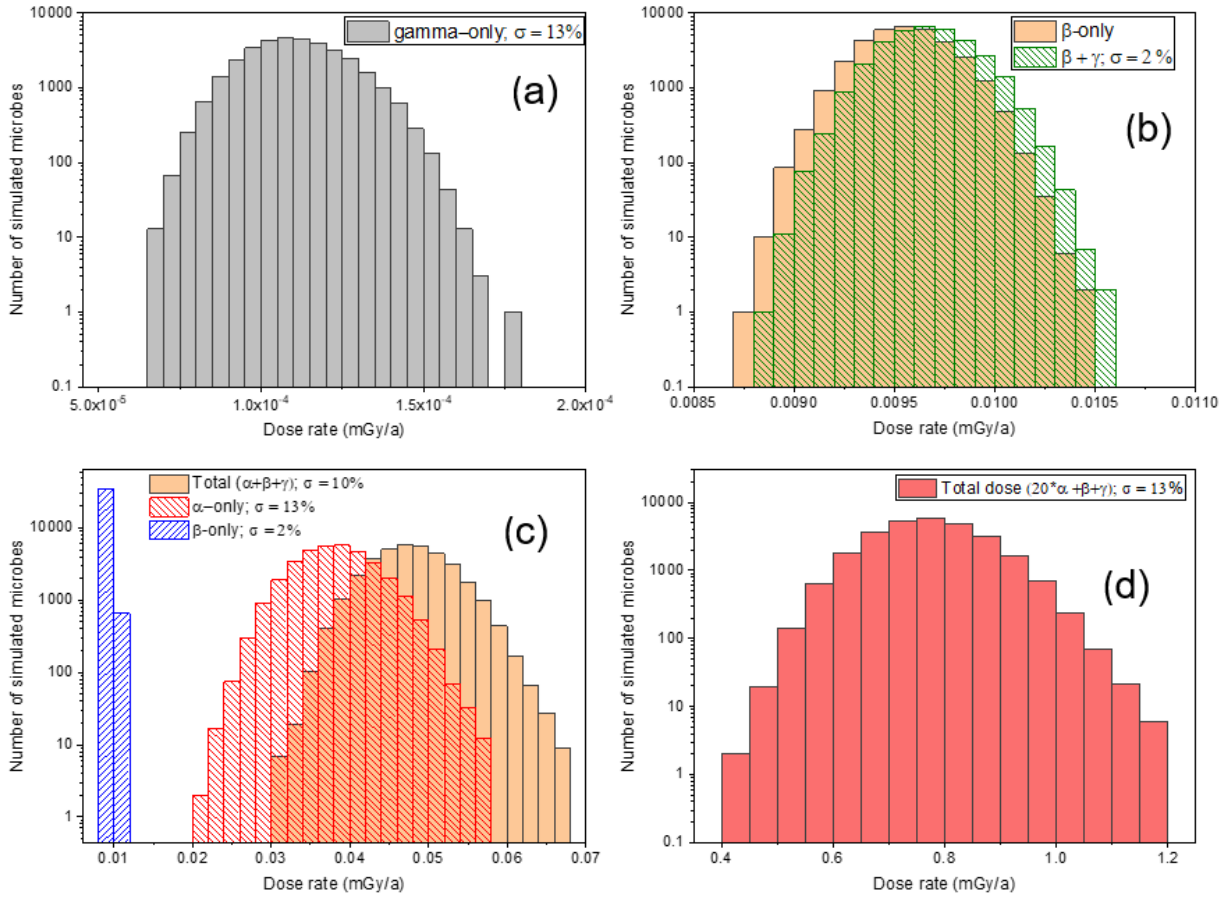


Figure 9.6: Microscopic dose rate distribution of microbes in the ALH 84001-type Martian subsurface permafrost. (a) Gamma dose rate distribution only. (b) Beta only and Beta + Gamma dose rate distributions. (c) Alpha only, beta only and total sum of alpha + beta + gamma dose rate distributions. Here the dose rate values were added directly. The total dose rate distribution in (d) was obtained by multiplying the alpha radiation portion of the dose distribution by 20 to account for the alpha quality factor.

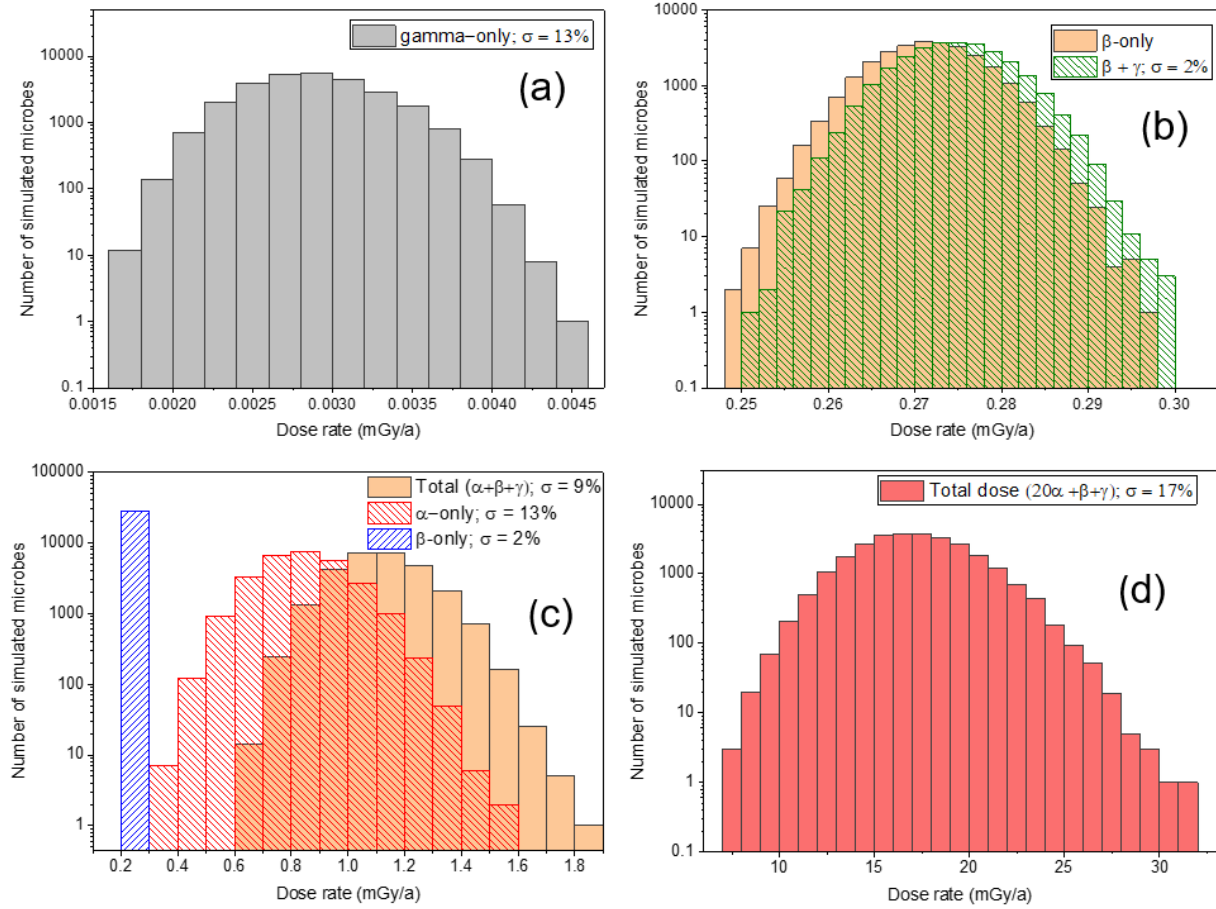


Figure 9.7: Microscopic dose rate distribution of microbes in the basaltic andesite-type Martian subsurface permafrost. (a) Gamma dose rate distribution only. (b) Beta only and Beta + Gamma dose rate distributions. (c) Alpha only, beta only and total sum of alpha + beta + gamma dose rate distributions. Here the dose rate values were added directly. The total dose rate distribution in (d) was obtained by multiplying the alpha radiation portion of the dose distribution by 20 to account for the alpha quality factor.

The dose rate distributions of the Martian scenarios considered are presented in Figures 9.5 to 9.7 here, and Figures F.1 to F.5 in Appendix F. The highest dose rate was seen for the basaltic andesite composition, the lowest was for the scenario based on bulk composition on Mars. As expected, the dose rate depends strongly on the radionuclide concentrations. The elemental compositions of the different scenarios seem to have a minimal effect of the distributions. A comparison between the distributions from the meteorite compositions shows that the Shergotty-type (Figure 9.5) is 10 times higher than the ALH 84001-type. This difference is also a reflection of their radionuclide concentrations. The negligible effect of the elemental

compositions can be seen clearly for the two different compositions from Gusev Crater; sulphur-rich (Figure F.1 and silica-rich Figure F.2) with the same radionuclide concentrations. The graphs show a similar distribution. The distribution for the Gale Crater-type scenario is also comparable to the Gusev Crater type. This is expected given that these two locations are relatively close together on Mars and only have slightly different radionuclide concentrations. The simulated dose rate in the north polar region of Mars due to dust trapped in ice is about two times more than that due to the bulk composition of Mars.

The dose rate due to alpha particles is higher than beta and gamma. The lowest dose rate distribution contribution is due to gamma. This is expected at the microscopic level because the recoil electrons released by the photon may deposit their energy outside the volume. The dose rate with the highest spread is the gamma dose rate. This is surprising considering that the gamma dose is expected to be uniform. An explanation for this spread is the simulation uncertainty due to the number of particles simulated. The total dose rate when alpha particles are corrected for their dense ionization track was 20x higher compared to the total uncorrected dose rate.

The survival curves produced from these distributions for some selected microbes are presented in the next section.

9.4.3 Survival probabilities

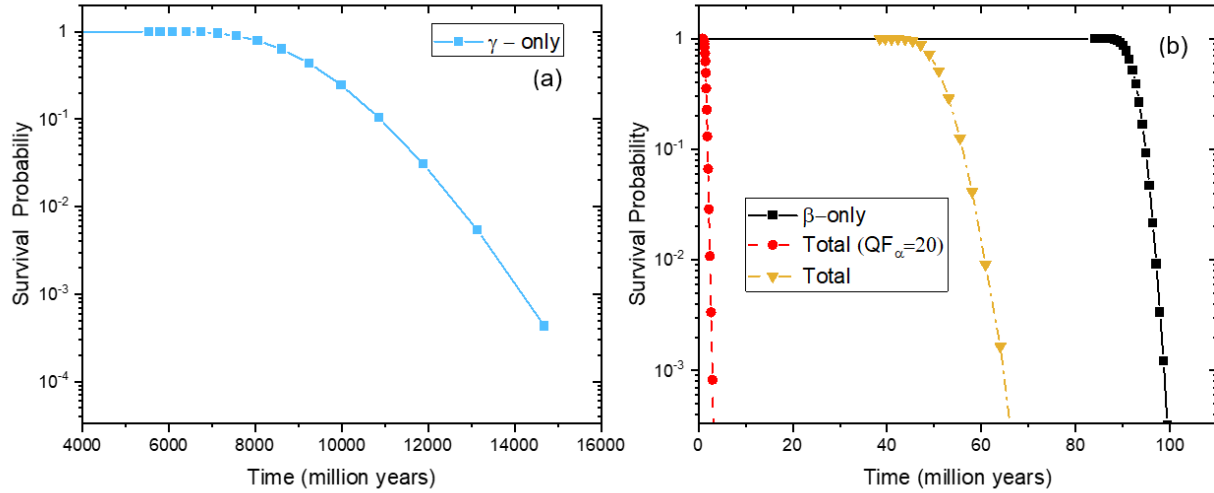


Figure 9.8: Calculated surviving probabilities of dry *B. subtilis* spores in the shergotty-type subsurface permafrost regions of Mars. The data points were calculated from the histogram and the lines are plotted to guide the eye.

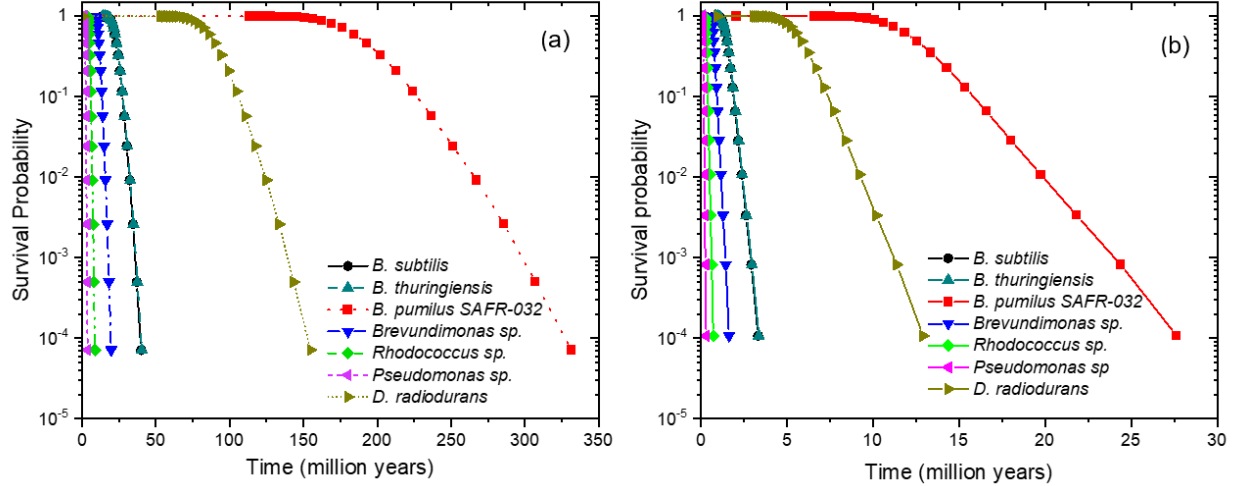


Figure 9.9: Calculated Survival probability of different microbes in the subsurface permafrost regions of Mars with shergotty-type composition as a function of time in billion years. The survival probability curves are based on (a) The total dose rate uncorrected for alpha quality factor and (b) the total dose rate corrected for alpha quality factor by multiplying the alpha part of the distribution by 20. The data points were calculated from the histogram and the lines are plotted to guide the eye.

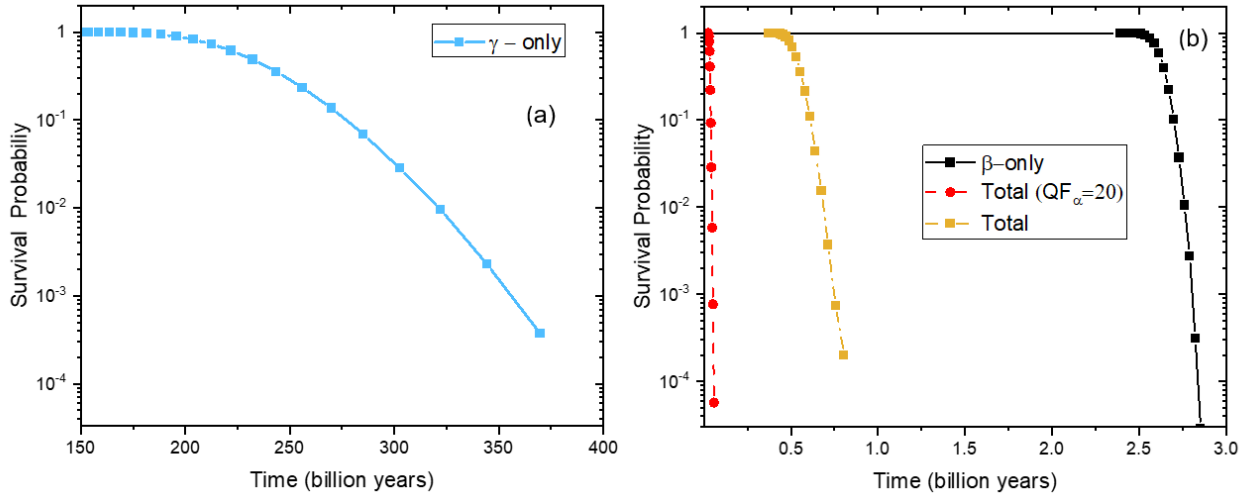


Figure 9.10: Calculated surviving probabilities of dry *B. subtilis* spores in the ALH 84001-type subsurface permafrost regions of Mars. The data points were calculated from the histogram and the lines are plotted to guide the eye.

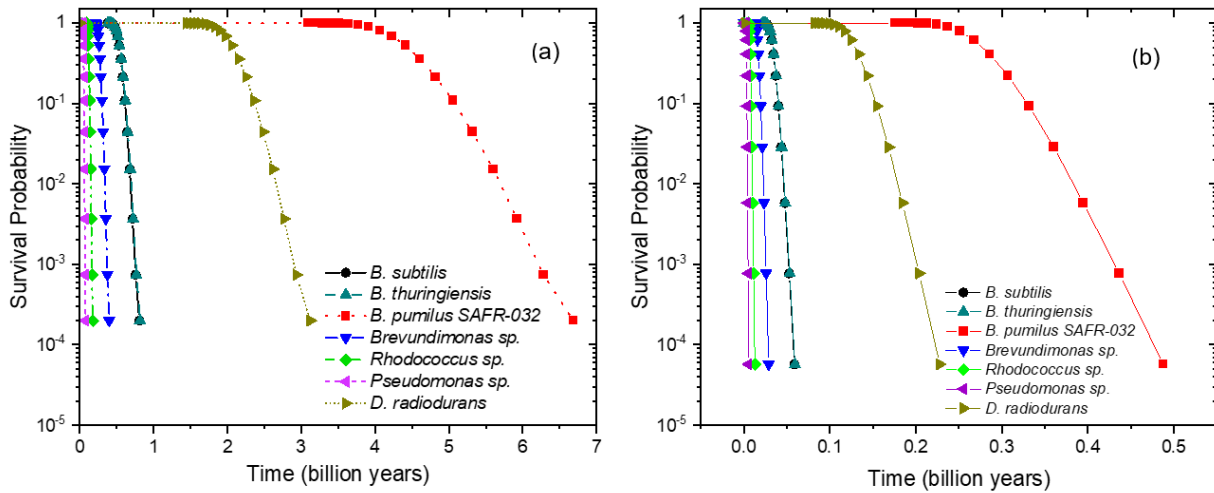


Figure 9.11: Calculated Survival probability of different microbes in the subsurface permafrost region of Mars with ALH 84001 rock/regolith composition as a function of time in billion years. The survival probability curves are based on (a) The total dose rate uncorrected for alpha quality factor and (b) the total dose rate corrected for alpha quality factor by multiplying the alpha part of the distribution by 20. The data points were calculated from the histogram and the lines are plotted to guide the eye.

The survival probabilities calculated from the dose rate histograms are shown in Figures 9.8 to 9.11 here and Figures F.6 to F.15 in Appendix F. These survival probabilities are for dormant microbes that cannot repair radiation damage, allowing us to calculate their survival

times using their simulated dose rate distributions as their dosage accumulation rates. The survival probabilities are presented as the time required to produce an approximately 10^4 reduction in the total number of microbes simulated.

Based on the alpha-corrected total dose rate distributions in this work, the most "radio-resistant" microbe on Mars will only survive a maximum period of 500 million years (in ALH 84001 composition-type regions). For the other regolith types apart from BSM, the survival time is much less, ranging from just 25 million years in Gusev-type regolith to 85 million years in the north polar region. The survival probability on the other hand without the alpha quality factor correction, increases by approximately a factor of 20.

Some limiting cases were also considered, using *B. subtilis* as a reference. On the microscopic scale, not all microbes might receive the full spectrum of radiation. For example, some microbes might only be exposed to gamma radiation. The results show that such microbes (*B. subtilis*), can survive for about 15 to 350 billion years. The latter number seems extreme. It was obtained for the survival of microbes in the ALH 84001 meteorite-type region of Mars. If this composition is currently present on Mars and has similar radionuclide concentrations, the result suggests that dormant microbes can still be existing there on Mars today. However, no meteorite has been matched to its actual location on Mars to date, except for the shergottite-like Bounce Rock sample that was found in Meridiani Planum on the Martian surface [78]. For the limiting case of microbes that were exposed to only beta radiation, or beta plus gamma radiation (dose rate distributions not different from beta only), the results suggest that such microbes can survive for about 100 million to 2.8 billion years.

Furthermore, out of the microbes whose survival probabilities were calculated, the *B. pumilus* SAFRR-032 showed a survival that is twice that of the well-known radioresistant *D. radiodurans* and more than six times that of *B. subtilis*. This is in agreement with experimental observations reported by Belbe et al. [236] and Gioia et al. [46].

9.4.4 Probabilities for Biomarkers and DNA preservation

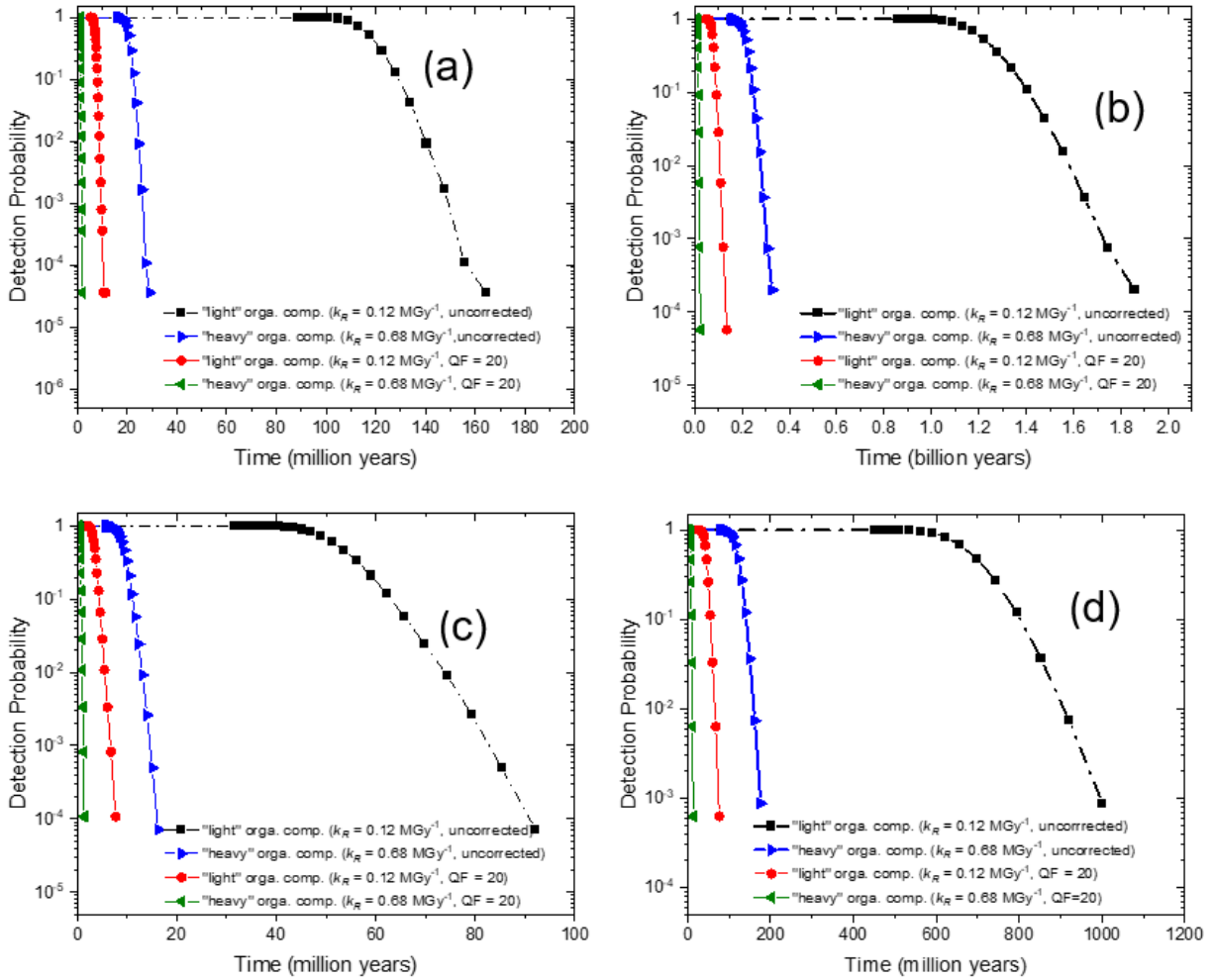


Figure 9.12: Detection probability of light and heavy organic molecules in Martian subsurface permafrost based on alpha quality-uncorrected and corrected dose rate distributions. The data points were calculated from the dose rate histogram and the lines are plotted to guide the eye. The result is presented for different regions of Mars with (a) Shergotty-type regolith/rock compositions. (b) ALH 84001-type regolith/rock compositions. (c) Basaltic Andesite-type regolith/rock compositions. (d) Martian bulk silicate (BSM) compositions.

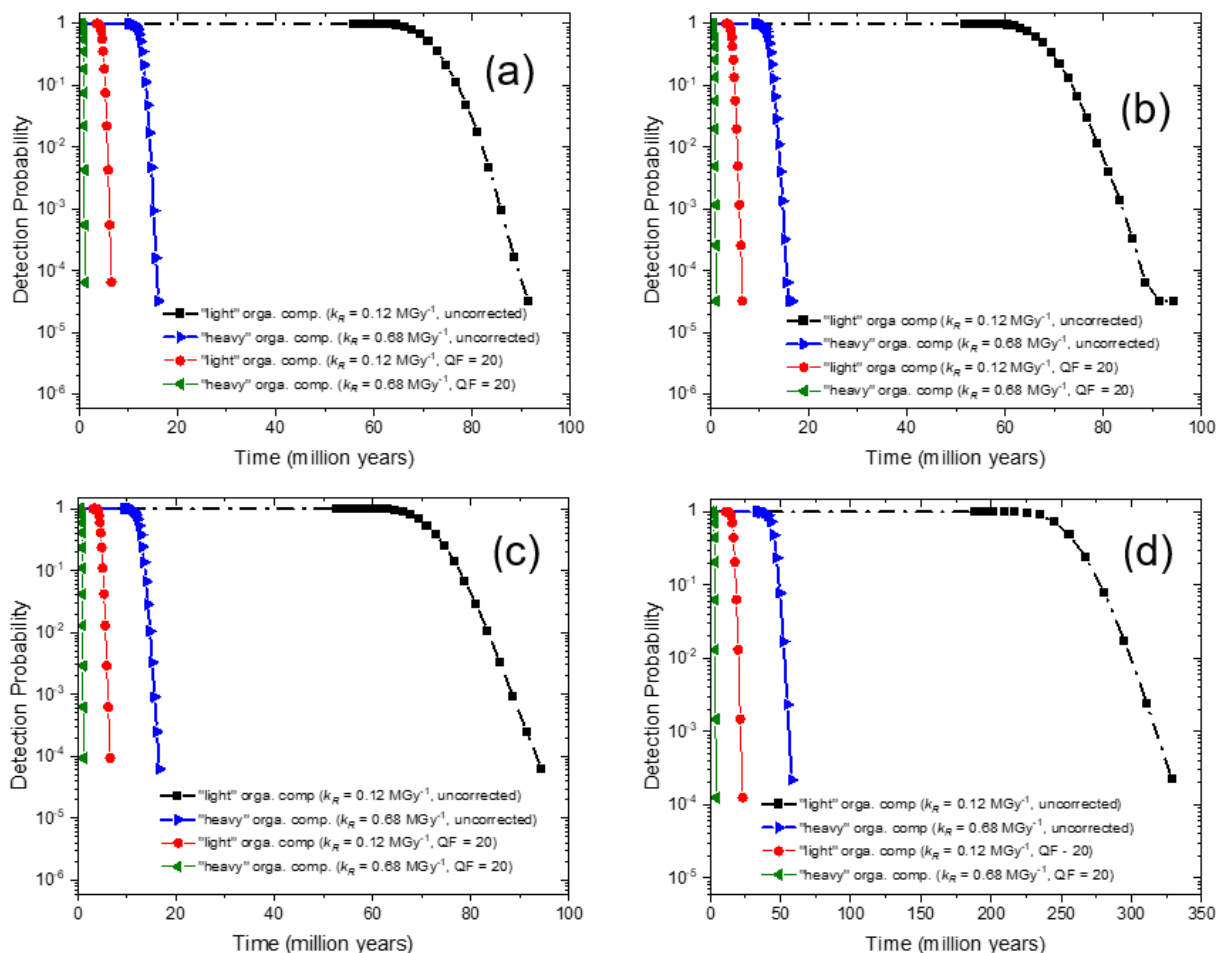


Figure 9.13: Detection probability of light and heavy organic molecules in Martian subsurface permafrost based on alpha quality-uncorrected and corrected dose rate distributions. The data points were calculated from the dose rate histogram and the lines are plotted to guide the eye. The result is presented for different regions of Mars with (a) sulphur-rich Gusev Crater-type regolith/rock compositions. (b) silica-rich Gusev Crater-type regolith/rock compositions. (c) Rocknest Gale Crater-type regolith/rock compositions. (d) compositions of ice and dust similar to the north pole.

The probabilities for DNA and biomarker preservation were plotted as detection probabilities calculated from the dose rate histograms and are shown in Figures 9.12 and 9.13 for all the subsurface models.

The Figures show the time of exposure needed to produce a 1000-fold reduction in the amount of organic molecules that remains after radiation reaches lethal level and kills the microbes. Results from our calculations suggest that it will take 10–100 million years depending on the location on Mars, to obliterate (or at least render undetectable) organic

compounds with masses less than 200g/mol. This is for the case when the radiation is corrected for alpha quality factor. For organic compounds with masses larger than 200g/mol, this number is even less. This is in agreement with the result of Pavlov et al. [229]. They reported that it will take less than 50 million years to destroy organic molecules in the top cm of Martian surface rocks.

This calculation using 1000-fold reduction was based on the Mars Science Laboratory (MSL) Curiosity rover organic molecules detection sensitivity. The sensitivity of future devices is most likely going to be better. Additionally, the detection probability plots presented were based on the assumption that each microbe absorbed the complete radiation spectrum (α , β , and γ). Biological markers that are exposed to only part of the spectrum are expected to be preserved considerably longer. For example, organic compounds exposed to only gamma radiation could be preserved for up to 900 billion years.

9.5 Summary

In this chapter the dose rate distributions of microbes and biomarkers in eight Martian subsurface permafrost scenarios were simulated. The microbes and biomarkers were modelled as 260 μm H_2O grains surrounded by a uniform matrix and grains, both of which were radioactive.

The models represented different chemical compositions, grain size distributions, radionuclide concentrations, and ice contents. The simulated dose rate distributions from the models were converted to 5 μm "microbial grains" by using attenuation factor corrections. The results of dose rate distributions obtained via these conversion factors were considered an approximation. The average dose rate values are likely close to what is obtainable with direct simulations of 5 μm grains but the actual distributions are likely different. The distributions for the eight scenarios are very similar, with dose rate values that depend on the radionuclide concentrations. This is in agreement with the observation by Taylor et al. [17] that K/Th does

not vary much on the surface of Mars in spite of evidence for rock-water interactions affecting K and Th concentrations. They argued that the small variation might be an indication that the products of weathering were highly acidic, which makes both K and Th mobile [17].

In conclusion, the results show that ancient dormant microbes that are surviving the harsh environmental conditions on Mars by residing in the Martian subsurface permafrost can survive for a maximum period of ~ 500 million years. The maximum time for the detection of their remnants after death is ~ 100 million years.

Chapter 10

Conclusions and Future Directions

10.1 Conclusions

The goal of this work was to determine how long microbes will survive a dormant state under the influence of radiation in the Martian regolith, and how fast radiation will destroy their remains after death. Our approach was to model the radiation environment that microbes are exposed to in the Martian subsurface using the Geant4 Monte Carlo (MC) programs *DosiVox* and *DosiSed*. The reliability and precision of the simulation results were investigated by comparing to published data. The models were also tested on dose distributions measured with $\text{Al}_2\text{O}_3\text{:C}$ dosimeters buried in terrestrial samples.

The optimum simulation parameters were determined for the infinite dimensional models for the Martian subsurface and dose rate distributions were presented. The dose rates calculated were further analyzed and survival probabilities for some selected microbes and probabilities for preservation of biomarkers for some organic compounds were obtained.

In conclusion, if microbes once existed on Mars, and are currently residing in dormant state in the subsurface permafrost regions of Mars, their survival is limited by the amount and type of radiation they are exposed to from the natural radioactivity of the Martian regolith. Based on the results presented in this dissertation, the microbes can survive for as

long 50 billion years depending on their location and whether they are exposed to the full spectrum of the radiation or only gamma radiation. For microbes that are exposed to only gamma radiation, the most radio-resistant of them can survive up to 350 billion years while those that are exposed to the full spectrum will not survive for more than 500 million years.

10.2 Limitations of this work

In our Martian subsurface simulation models, a number of assumptions and simplification have been made. Regardless of the logic behind the assumptions and simplifications, the results presented here for the Martian subsurface permafrost must be taken as what they are: approximations. Their significance is to demonstrate the orders of magnitude involved, as well as the relative differences due to changes in parameters. Some assumptions are discussed here:

- ***Conditions of early Mars:*** The whole premise of this dissertation is based on the evidence that 3.8 billion years ago liquid water existed on Mars. This would allow for ancient microbes to have existed. Mileikowsky et al. [12] argued that Mars' distance from the Sun, as well as the nature of its greenhouse gases would have made it difficult for the planet to produce and sustain a surface temperature of 0°C or more that is required for liquid water to exist on its surface. However, there are several studies that support the idea of a "wet" Mars [3, 237–239].
- ***Subsurface Martian models:*** Subsurface Martian models: Our model assumes that the density and porosity of the sediments in every location considered is constant radially and vertically and can be represented by an average value. It also assumes that the density does not change over geological time scales. However, the effect of density is minor when the results from different locations are compared. In fact, our dose rate models did not account for differences in elemental compositions due to global processes in different Martian geological periods (Amazonian, Hesperian, and Noachian) [65].

- **Radionuclide concentrations:** The GRS data used in this work is the most exhaustive record of radionuclide concentrations on Mars. However, it is geographically low resolution. The data according to the author Taylor [82] was obtained by splitting the Martian surface into bin sizes with each bin equivalent to the size of North Carolina (personal communication with Taylor). Therefore, even though we present results for microscopic dose rate variations, our calculations do not take into account micro-scale variability in radionuclide concentrations at distances smaller than the GRS footprints.
- **Inactivation constants and survival probabilities:** There was no result for the survival of *B. pumilus* SAFR-032 under exposure to gamma radiation. Thus the inactivation constant used was calculated based on their response to UV relative to the response of *B. subtilis*. This calculation which was done using the slopes of their survival curve resulted in an error that is about 70% of the mean. The radiolysis constants were either calculated from experimental survival curves or taken from tabulated results. There is a big variation in the inactivation constants reported in the literature for, for example, *D. radiodurans* [28, 40, 42]. The inactivation constants used for this work did not account for damage to micro-organisms or cells in general due to secondary oxidation processes like the production of reactive oxygen species. For example, regardless of the exact composition of the Martian subsurface, energetic charged particles can produce O_2^- and OH^- from the interaction with SiO_2 or FeO minerals and ice. These free radicals can cause indirect damage to organic molecules. This also applies to the radiolysis constants used.
- **Radiolysis constants and DNA preservation probabilities:** The extrapolated result of the linear dependence of radiolysis constant vs. molecular mass to account for light and heavy organic molecules taken from Pavlov et al. [229] was based on the original experimental data by Kminek & Bada [230], which covers only 50–200 g/mol. The linear dependence was extrapolated to 500 g/mol [229] without additional experiments [229].

A justification for the linearity is that the cross section of the molecular breakup by the energetic particle is proportional to the geometric size of the target molecule and the geometric size is proportional to the molecular mass (at least for organic molecules that contains mostly C and H) [229].

10.3 Suggestions for future work

There are several improvements in our models that could improve the accuracy of our result and make our models closer to reality.

In our Martian models, the sedimentary grains were uniformly radioactive. A model of a mix of inert and radioactive grains would be more representative of what is found in nature. In such model, there should be grains that do indeed only receive beta and/or gamma dose. This will require researching the actual Th, U, and ^{40}K bearing minerals on Mars and using the compositions of Martian regolith that are reflective of this.

All our Martian scenarios were homogeneous mixtures. Inhomogeneous scenarios such as ice wedges or inert layers (thin layers of ice) in between or underlying dry rocks can be considered more realistic. This will require some modifications in the Monte Carlo codes. Currently this can be done with *DosiVox* but the challenge is the addition of the α , β , and γ dose rates for each grain because in *DosiVox*, the position of each grain is not specified due to the "parallel world" geometry applied to superimposed dosimeter grains with the "mass" world, where materials are created (see Section 4.4 for more details).

Another idea will be the incorporation of the Geant4-DNA extension of the Geant4 Monte Carlo simulation toolkit into the *DosiVox* and *DosiSed* codes if possible. This will allow for simulating the effect of radiation damage on DNA and bio-markers on the microscopic level directly.

Finally, the simulations of "microbial grains" with dimensions the size of bacteria with large number of simulated particles to obtain the actual dose distributions. This will require

running several parallel simulations using a computer with at least 250GB RAM with multi-core capabilities. A cheaper option is time. The frame-work and characterization of these codes for infinite matrix simulations have been performed in this work. Using those results, one can at least perform simulations for 1–5 μm grains within a year, as long as Microsoft Windows does not force-update the computer during the period.

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Appendix A: Calibration of the Risø β -source against a ^{137}Cs gamma source

The irradiations performed in this work for dose response studies and calibration purposes were carried out with the built-in $^{90}\text{Sr}/^{90}\text{Y}$ β -source of the Risø reader. The dose rate delivered at the dosimeter position in the reader was obtained through calibration of the built-in source against a 662 keV ^{137}Cs gamma source. To carry this out, an Al_2O_3 ceramics container, was filled with 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains and Luxel[®] dosimeters and was positioned 35 cm from the source (see Figure [A.1](#)). The air-gap in the container was filled with Al_2O_3 powder. The Al_2O_3 container serves as build-up material for the radiation beam and the Al_2O_3 powder ensures that charged particle equilibrium is maintained while the beam travels through the container, so that all the $\text{Al}_2\text{O}_3\text{:C}$ grains will receive a uniform dose. Because the dosimeters are light-sensitive, the Al_2O_3 container was wrapped with black tape. The gamma dose rate in dry air at the container position (35 cm from the gamma source) was obtained by using two-large volume ionization chamber survey meters that have been previously calibrated in a standard lab. The goal of this section is to describe the methods used to convert the dose in dry air to dose to the $\text{Al}_2\text{O}_3\text{:C}$ dosimeters inside the container.



Figure A.1: Gamma Irradiation setup for the calibration of beta source

The method employed is cavity theory, specifically the Burlin cavity theory which is a formulation of cavity theory for cavities with sizes that are intermediate between small and big cavities [240]. In such cavities, the total dose contribution is due to interactions of both electrons and photons inside the cavity [241]. Our Al_2O_3 container including all the dosimeters inside it can be approximated as a cavity placed in a medium (dry air). Thus cavity theory can be used to determine the dose to the cavity theoretically from the dose to air at the cavity position assuming the cavity was absent. This involved accounting for the photon beam interactions as well as secondary electron production in both media. Cavity theory therefore also depends on the geometry of the cavity, particularly the mean path length through the cavity [242].

Spherically shaped dosimeters are the ideal geometry for cavity theory. The dosimeter container however is cylindrical and not spherical so two-dimensional approximations were applied to account for the dependence of cavity theory on the size and geometry of the dosimeter. The first is an approximation of the dosimeter container as a sphere with surface

area equal to that of the of the cylindrical container, and a sphere with volume equal to that of the of the cylindrical container. Using these approximations, we start by first defining the cavity theory relationship for the cavity and the medium :

$$\frac{D_{cav}}{D_{med}} = d \frac{(\bar{S}/\rho)_{cav}}{(\bar{S}/\rho)_{med}} + (1 - d) \frac{(\mu_{en}/\rho)_{cav}}{(\mu_{en}/\rho)_{med}}, \quad (\text{A.1})$$

where D_{med} is the dose to the medium (air), D_{cav} is the dose to the cavity ($\text{Al}_2\text{O}_3\text{:C}$ dosimeters container). $(\bar{S}/\rho)$ is the average mass stopping power of the Compton electron spectrum produced from the photon. This was calculated by determining the mean electron energy of the 662 keV photon using the Klein-Nishina cross section and energy-transfer cross section obtained from the appendix in Attix [240]. The average mass stopping power was then obtained from the mean energy by using the NIST ESTAR stopping-power tables for electron data ¹. (μ_{en}/ρ) is the mass energy absorption coefficient, calculated from NIST Standard Reference Database 126 ². The parameter d accounts for the relationship between the mean electron fluence arising in the cavity wall $\bar{\Phi}_w$ compared to the initial electron fluence $\bar{\Phi}_w^e$ and can be calculated from the expression [240]:

$$d = \frac{\bar{\Phi}_w}{\bar{\Phi}_w^e} = \frac{1 - e^{-\beta L}}{\beta L} \quad (\text{A.2})$$

where L represents the mean chord length, calculated as four times the cavity volume divided by its surface area. The parameter β describes the build-up and build-down of electron fluence as a function of distance into the cavity [242]. It can be obtained from the relation:

$$\beta = -\frac{\ln(0.04)}{t_{max}} \quad (\text{A.3})$$

where t_{max} is the maximum depth of penetration for the electron and the value of 0.04 was based on the result of Jannssens et al. (1974) [243]. It is important to note that for the derivation of cavity theory, the cavity is an ionization chamber filled with air and the medium is typically a material other than air, for example water. Thus dose to air is converted to

dose to medium. However, in our case we are doing the reverse, our cavity is $\text{Al}_2\text{O}_3\text{:C}$ and our medium is air. We know the dose to air D_{air} via measurement using a survey meter, and we need to obtain the dose to $\text{Al}_2\text{O}_3\text{:C}$. Therefore the dose to $\text{Al}_2\text{O}_3\text{:C}$, $D_{\text{Al}_2\text{O}_3\text{:C}}$ can be simply derived from Eq. A.1 as:

$$D_{\text{Al}_2\text{O}_3\text{:C}} = D_{air} \left[d \frac{(\bar{S}/\rho)_{\text{Al}_2\text{O}_3\text{:C}}}{(\bar{S}/\rho)_{air}} + (1 - d) \frac{(\mu_{en}/\rho)_{\text{Al}_2\text{O}_3\text{:C}}}{(\mu_{en}/\rho)_{air}} \right], \quad (\text{A.4})$$

where d was ~ 0.02 for both the volume and surface area approximation. The other parameters used in the above equation to calculate $D_{\text{Al}_2\text{O}_3\text{:C}}$ are presented in Table A.1. There was negligible difference between the result of volume and surface area approximation. The calculated gamma dose to the $\text{Al}_2\text{O}_3\text{:C}$ dosimeters in the Al_2O_3 cylindrical container is (60.64 ± 1.37) mGy.

Table A.1: Parameters used to calculate $D_{\text{Al}_2\text{O}_3\text{:C}}$

Parameters	Dry Air	$\text{Al}_2\text{O}_3\text{:C}$
$(\bar{S}/\rho)$ (MeV cm ² /g)	2.233 ± 2.051	1.16 ± 0.27
(μ_{en}/ρ) (cm ² /g)	2.931×10^{-2}	2.892×10^{-2}
t_{max} (mm)		0.515
Density (g/cm ³)	0.0013	3.97

Finally to obtain the dose rate of the built-in $^{90}\text{Sr}/^{90}\text{Y}$ β -source of the Risø reader, the signals of the gamma irradiated dosimeters were measured and further converted to their $^{90}\text{Sr}/^{90}\text{Y}$ β -source exposure-duration-equivalence. i.e., the amount of exposure (in unit of seconds) from the β -source that produces a signal equal to the signal measured from the gamma irradiated dosimeters. The known gamma dose (60.64 ± 1.37) mGy, was then divided by this β -source exposure-time-equivalence to obtain the gamma equivalent dose rate of the built-in $^{90}\text{Sr}/^{90}\text{Y}$ β -source in (in mGy/s).

Appendix B: Beta dose rate reduction for
the built-in $^{90}\text{Sr}/^{90}\text{Y}$ sources of Risø
TL/OSL automated readers

Beta dose rate reduction for the built-in $^{90}\text{Sr}/^{90}\text{Y}$ sources of Risø TL/OSL automated readers

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Received: October 14, 2021; in final form: November 30, 2021

Abstract

Environmental dosimetry requires measuring doses, which are often below the range accessible with the commonly used built-in β -source of Risø TL/OSL readers. $\text{Al}_2\text{O}_3:\text{C}$ Luxel[®] dosimeters were used to investigate irradiation cross-talk and attenuators as means to reduce the dose rate of the source. Irradiation cross-talk was found to be highly effective in reducing the dose rate but was not reproducible and resulted in dose recovery deviations of up to 80%. Four attenuator materials (PTFE, microscope coverslip glass, wine glass, and ABS plastic) were investigated with regard to their dose reduction capabilities. All materials resulted in good dose recovery results and allow building a standard reference dose response curve. Microscope cover glass reduces the dose rate by a factor of 3 for each 1 mm thickness. The material is transparent and does not have to be removed during measurement and is therefore recommended as attenuator of choice for dose rate reduction.

Keywords: Dose rate reduction, Risø source, $\text{Al}_2\text{O}_3:\text{C}$, OSL, Attenuators

1. Introduction

$\text{Al}_2\text{O}_3:\text{C}$ dosimeters have become one of the most widely used luminescence dosimeters for environmental dose-rate assessment. The appeal of this dosimeter stems from its high sensitivity to radiation and linear behavior over a wide dose range from μGy to several Gy (Akselrod et al., 1990; Yuki-hara & McKeever, 2008). $\text{Al}_2\text{O}_3:\text{C}$ has been used in various

studies for retrospective dosimetry (McKeever et al., 1995; Bøtter-Jensen et al., 2000; Discher et al., 2021) and measurement of β dose rates to natural sediments (Göксу et al., 1999; Burbidge & Duller, 2003; Kalchgruber et al., 2002; Kalchgruber & Wagner, 2006; Durcan et al., 2015; Smedley et al., 2020). To determine the environmental dose absorbed by the dosimeters during burial, the signal is first measured in the laboratory, followed by irradiation of dosimeters for calibration (Kalchgruber & Wagner, 2006). A common instrument for this measurement and subsequent irradiation is the Risø TL/OSL-DA reader. The reader is often equipped with a 1.48 GBq $^{90}\text{Sr}/^{90}\text{Y}$ β -source, that delivers a dose rate of the order of 100 mGy/s to a sample when the source is open. Considering that signals from $\text{Al}_2\text{O}_3:\text{C}$ dosimeters are very bright a dose rate of this magnitude can easily saturate the photomultiplier tube (PMT) of the reader after a few seconds of irradiation. The light reaching the PMT can be reduced unless one uses by using an aperture to reduce the amount of light reaching the PMT. Alternatively, one can correct for the dead time loss that causes saturation, in which case, there is a risk, however, of damaging the PMT. Furthermore, environmental radiation is classified under low level background radiation and dose-rates are typically less than 100 $\mu\text{Gy/s}$ (Burbidge & Duller, 2003). In routine environmental beta dose-rate assessment, dosimeters are buried for a period of few weeks to a year (Kalchgruber & Wagner, 2006). The cumulative absorbed dose measured even after a year has been reported to be comparable to the dose obtained after a 1s irradiation from the β -source of the reader (Kalchgruber et al. (2002)). Such short irradiation times are however not recommended since the rotating nature of the irradiation unit introduces a time offset and thus adds uncertainty to the given dose (Markey et al., 1997). Although $\text{Al}_2\text{O}_3:\text{C}$ shows a linear behavior from μGy to few Gy it is not recommended to use larger irradiation times and to extrapolate the dose response to lower doses. Extrapolation is well known to lead to large

errors in dose recovery, due to the relatively large impact of the uncertainty in the intercept fitting parameter.

To reduce the dose rate of the beta source, [Burbidge & Duller \(2003\)](#) used bremsstrahlung radiation that is produced by beta particles from the source in closed position. This option greatly reduced the dose rate to about $(0.687 \pm 0.003) \mu\text{Gy/s}$ at the dosimeter position and is therefore an effective method for assessing very low doses in the range of 10 μGy to about 1 mGy. However, when the goal is to assess medium doses $\sim 1 - 100$ mGy, bremsstrahlung requires long exposure times under the closed source; for example, it will take 4 hours to obtain a dose of 10 mGy. Such long exposure times are often not feasible when instrument time is limited. Additionally, this leakage radiation was reported to be maximum at a position different from the position directly under the source (see Fig.1) and asymmetric due to the asymmetric scattering of radiation ([Kalchgruber et al., 2002](#)). The radiation affects more than a quarter of all positions on the turntable and additional positions are impacted by leakage radiation from the alpha source for readers which have an alpha source installed as well. Consequently, it is only practical to irradiate and analyze one or two dosimeters at a time.

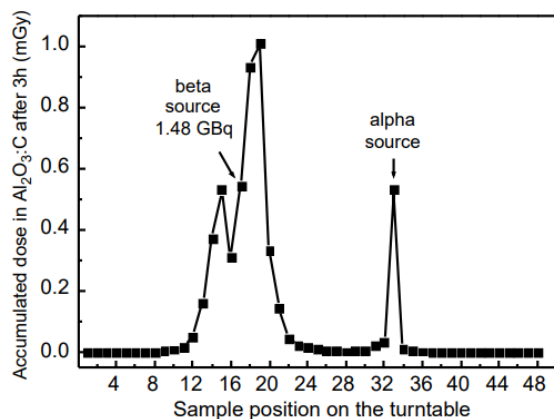


Figure 1. Accumulated dose due to leakage radiation in 3 h at various carousel positions in a Risø TL/OSL-DA-15 reader measured with $\text{Al}_2\text{O}_3\text{:C}$ dosimeters, with both beta and alpha sources installed. The position underneath the beta source indicated on the plot is not the position of maximum dose. The position under the alpha source is also a hotspot as indicated on the plot (Fig. 4. from [Kalchgruber et al. \(2002\)](#))

Other options offered by the manufacturer include purchasing a second, weaker source or the reduction of dose rate by increasing source-sample distance using a separately sold dose rate kit. The kit consist of an aluminum filter, a brass ring and an aluminum spacer that can reduce the dose rate by a factor of ~ 10 . However, this dose rate kit requires removing the built-in source from the irradiator for installation. The radiation safety regulations in many institutions require that a radiation safety officer perform this kind of procedure and thus might not be feasible if inserting or removing the kit is required several times a week.

A third option is the use of irradiation cross-talk. Irradia-

tion cross-talk is defined as radiation exposure to dosimeters in the vicinity but not directly under the irradiation source, when the source is open. This effect has been investigated by various authors for quartz and $\text{Al}_2\text{O}_3\text{:C}$ dosimeters and has been shown to be in the range of 0.006–0.25 % of the direct dose rate, depending on the specific reader and the irradiated sample ([Bøtter-Jensen et al., 2000](#); [Kalchgruber et al., 2002](#); [Bray et al., 2002](#); [Thomsen et al., 2006](#)). Yet another option for dose rate reduction, which to our knowledge has not been investigated in detail, involves placing an attenuator directly on top of the dosimeter.

This paper investigates the latter two options of reducing the dose rate of the built-in β -source of the Risø reader: i) irradiation crosstalk and ii) attenuators. The major limitation with the absorber is the small distance between the bottom of the cup and the lid. This distance which can range from $\sim 1.8 - 2.5$ mm is reader specific. To this end, different materials of different thicknesses were investigated to arrive at the best option for the attenuator. The overall goal is to build a reference dose response curve that can be used for dose assessment of dosimeters buried in sediment for several weeks to a year.

2. Materials and methods

2.1. Dosimeters

The dosimeters used in this work are the $\text{Al}_2\text{O}_3\text{:C}$ Luxel[®] dosimetry tapes cut into 6.0 mm diameter round pieces using a hole puncher (Fig.2). The Luxel[®] tapes are produced by Landauer Inc. and consist of 20 - 90 μm $\text{Al}_2\text{O}_3\text{:C}$ grains sandwiched between two polyester sheets, resulting in a thickness of about 0.3 mm ([Akselrod et al., 2000](#); [Bøtter-Jensen et al., 2003](#)). Prior to use, the dosimeters were bleached for 8-12 hours with light from a halogen lamp filtered with a Schott FSQ-GG-495 colored glass long-pass filter (thickness 3.0 mm) as recommended by [Sawakuchi et al. \(2008\)](#) to remove any background signal accumulated during storage and transport, if applicable. Between irradiation and measurement, the dosimeters were handled in a dark room with subdued red light for visibility and to prevent any bright light from resetting the dosimeters and placed in stainless steel cups.

2.2. Attenuators

Attenuators of different materials were investigated in this work as a way of reducing the dose rate of the β -source of the Risø reader. Four different attenuator materials were tested for their effect on the dose rate:

- a 10 mm diameter disk of clear glass, type “Libbey midtown white wine glass” produced by LIBBEY GLASS. The thickness of the disk is 1.4 mm and its density is 2.27 g/cm^3 .
- two sets of round coverslips (microscope cover glass, density 2.42 g/cm^3), 12 mm diameter and 8 mm diameter respectively. Six of the 12 mm coverslips were

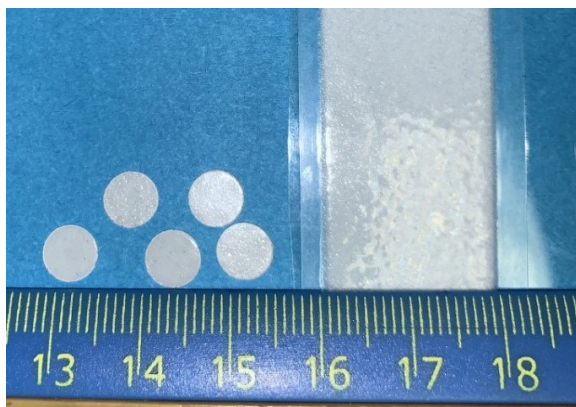


Figure 2. OSL Luxel® round punch-out and tape. The round punch-out was obtained by using a hole punch on the tape. The thickness of the dosimeter is 0.3 mm and diameter is 6.0 mm.

stacked to get a thickness of 1.4 mm (including the transparent tape used to hold the stack together). The actual thickness of the stack without the tape is 1.2 mm. Similarly, ten 8 mm coverslips were stacked to get a thickness of 1.9 mm and held together by glass glue. The use of a tape was not feasible in this case because of the smaller diameter. The coverslips are produced by BIPEE, China.

- Acrylonitrile-Butadiene-Styrene (ABS) (density: 1.17 g/cm³) disks, 8.2 mm diameter with a recess of diameter 6.3 mm and depth 0.3 mm to hold the Luxel dosimeter (Fig. 5c). The thickness of the disk at the rim is 1.8 mm and 1.5 mm above the dosimeter in the recess. The material is produced and sold in the form of rods by US Plastic Corp®, Lima, OH.
- Teflon® (natural virgin PTFE, density 2.20 g/cm³) disks with 8.2 mm diameter and thicknesses of 1.4 mm and 1.9 mm respectively. Teflon is produced in the form of rods by ePlastic®, San Diego, CA.

Photos of all the attenuators are shown in Fig. 4. The wine glass disk and the 12 mm diameter microscope coverslip glass stack were placed on top of the sample cup as shown in Fig. 3 (left) and 5a, leaving a small air gap of ~0.4 mm between attenuator and dosimeter. The maximum total height of the combination is about 2.3 mm, which is the allowable distance between the bottom of the cup and the lid for this specific model of Risø reader. The 8-mm cover glass, and the ABS and Teflon disks were in direct contact with the dosimeters (Figs. 3b and 5b, c).

2.3. Irradiation and readout equipment

All measurements were performed with a Risø TL/OSL-DA-20 automated reader produced by DTU Physics, Denmark. The reader is equipped with a built-in nominal 1.48 GBq ⁹⁰Sr/⁹⁰Y source with a beryllium window located between the irradiator and the measurement chamber, which acts as a vacuum interface for the measurement chamber



Figure 3. Absorber and sample cup combinations: (a) 12 mm diameter microscope coverslip glass stack on top of the sample cup with a dosimeter and (b) 1.9 mm PTFE disk placed inside the sample cup directly on top of the dosimeter.

(Markey et al., 1997). The dose rate to Al₂O₃:C Luxel® in the position directly under the source is 72.7 mGy/s (±3%). Green LEDs (at 525 nm, 68 mW/cm²) were chosen for optical stimulation of the dosimeters. Due to the high sensitivity of Al₂O₃:C, a 10 mm aperture was used in combination with 7.5 mm thick U-340 filters (transmission 340 ± 40 nm) in front of the PMT to reduce the possibility of over-saturation. The dosimeters were placed on stainless steel cups mounted on a turntable with 48 sample positions. Only every 5th position was used to prevent radiation crosstalk from affecting the dosimeters.

2.4. General read-out procedure

The measurement procedure is as follows. Irradiation steps varied with experiment and are described in detail below.

1. Irradiation with beta source:

- (a) indirect irradiation using cross talk (see section 3.1)
- (b) direct irradiation, with or without attenuator (see section 3.2)

2. Remove attenuator, where applicable

3. OSL with green diodes for 500 s at 30°C, record one data point every 1s; signal S

4. Test dose irradiation

- (a) indirect irradiation using cross talk (see section 3.1)
- (b) direct irradiation, without attenuator (see section 3.2)

5. OSL with green diodes for 500 s at 30°C, record one data point every 1s; signal S_T

The test dose accounts for variation in parameters such as dosimeter sensitivity, dosimeter mass, and equipment sensitivity (Murray & Wintle, 2000; Yuki-hara et al., 2005). OSL intensity was obtained from the integrated signal of the first 10 s of stimulation, and the background from the last 10 s of stimulation. The test-dose corrected signal S/S_T and its uncertainty is calculated for each dosimeter. Each dosimeter is only used once.

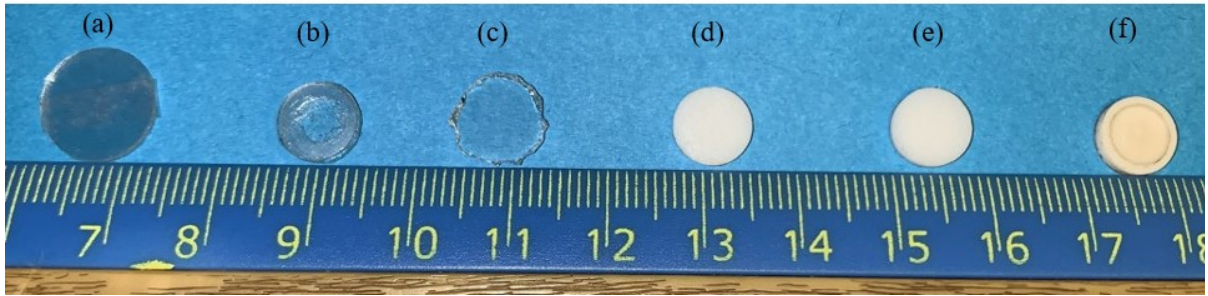


Figure 4. Absorber materials used in this study. From the left, (a) microscope coverslip glass stack (diameter = 12 mm, thickness = 1.4 mm), (b) microscope coverslip glass stack (diameter = 8 mm, thickness = 1.9 mm), (c) wine glass disk (diameter = 10 mm, thickness = 1.4 mm), (d) PTFE disk (diameter = 8.2 mm, thickness = 1.4 mm), (e) PTFE disk (diameter = 8.2 mm, thickness = 1.9 mm), and (f) ABS disk (diameter = 8.2 mm, thickness = 1.8 mm, also showing the 0.3 mm recess for the dosimeter).

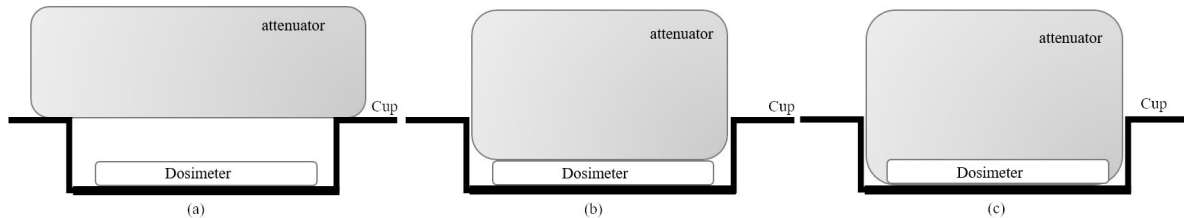


Figure 5. Three Configurations of sample cup-dosimeter-attenuator combination. Configuration (a) was used for the 12mm diameter coverslip glass stacks and the 10 mm diameter wine glass, since their diameters exceeded the 8.3 mm diameter of the recess in the sample cup. Configuration (b) was used for the PTFE disks and the 8 mm diameter coverslip stack, and (c) was used for the ABS disk with recess.

3. Results

3.1. Indirect irradiation with radiation cross-talk

3.1.1 Estimation of cross-talk dose-rate

In our first set of experiments, we investigated the use of next-position irradiation ("cross-talk") as described by Kalchgruber et al. (2002), as a method of extending the range of the dose-response curve to low doses in the μGy region. No attenuators were used for this set of experiments. The position adjacent to each dosimeter was irradiated with specific doses ranging from 200–3000 s (3 dosimeters per dose point) and subsequently implementing the readout procedure described in section 2.4 using a test dose of 250 s, also via cross-talk. A linear fit was used to determine the resulting dose response. In a second step, 3 separate dosimeters were irradiated directly for 1 s. Their 250 s indirect test-dose corrected signal was compared to the dose response. The cross-talk irradiation time equivalent to 1 s direct irradiation was determined to be 2078 s ($\pm 8\%$) (see Fig. 6). In other words, the cross-talk dose rate of the Risø reader is about 0.05% that of direct irradiation.

3.1.2 Effect of dosimeter positioning on reproducibility

The purpose of dose-response curves is generally for dose recovery. For environmental dosimetry the dosimeters are irradiated during burial and the test-dose corrected signal is then compared to an established dose response curve to determine the absorbed dose. Radiation cross-talk changes considerably between adjacent positions (Fig. 1). Dosimeters buried

in sediments and then placed in the reader for measurement might not all be in exact same position on the cup. There is also the possibility of random shift in the position of the dosimeter in the cup during measurement procedure. To test the impact of positioning on our results, 3 dosimeters were irradiated with cross-talk doses of 250 s, 450 s and 800 s

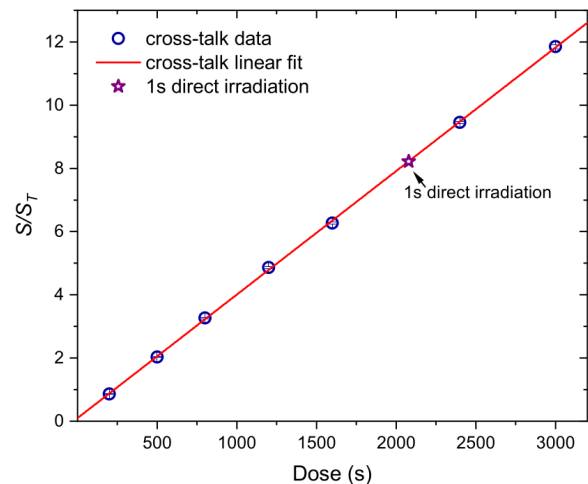


Figure 6. Reference dose-response curve of $\text{Al}_2\text{O}_3:\text{C}$ dosimeter built up with the β -source of the Risø reader (Risø TL/OSL-DA-20) using cross-talk for indirect irradiation. The red line indicates the linear fit of the irradiation cross-talk S/S_T vs. equivalent dose data while the purple star symbol represents the 1s direct irradiation data.

Protocol	Known Dose	Recovered Dose	Recovery Ratio
	(s)	(s)	
Dosimeter “stayed in reader” between irradiation and measurement	250	254 ($\pm 4\%$)	1.016
	450	455 ($\pm 2\%$)	1.011
	800	802 ($\pm 2\%$)	1.003
Dosimeter removed, stored for 4h between irradiation and measurement	250	107 ($\pm 9\%$)	0.428
	450	585 ($\pm 2\%$)	1.300
	800	1483 ($\pm 1\%$)	1.854

Table 1. Accuracy of dose recovery when dosimeters remain in the reader after irradiation, versus brief removal and re-insertion of dosimeters.

respectively. The three dosimeters were read immediately without opening the reader using the readout procedure. Another set of 3 dosimeters, used to mimic external irradiation, were removed from the reader subsequent to irradiation and stored for 4 hours in a dark room to simulate transport and storage, before they were read using the readout procedure. The doses recovered from these dosimeters using the dose response curve were compared to the known doses and results are given in Table 1

Table 1 shows that the measurement procedure is suitable for recovering doses with a precision of better than 2%, and that random shifts during measurement have little impact on the results. For the dosimeters that were removed from the reader and stored for 4 hours, measured doses show large deviations up to 85%. Yuki-hara & McKeever (2006), reported that UV emission from $\text{Al}_2\text{O}_3\text{:C}$ increases with the time elapsed between irradiation and measurement. In order to remove the effect of wait time, a similar test, but with an equal wait time of 5 min for all dosimeters was conducted. In this test, 10 dosimeters were irradiated with radiation cross-talk for 500 s and the signal was measured using the readout procedure, including test-dose application via cross-talk. For 5 out of the 10 dosimeters, the reader was not opened, but there was a 5 min delay between irradiation and measurement. The other set of 5 dosimeters was irradiated in the reader, removed and stored for 5 minutes, before signal measurement and test-dose correction. For each of the two sets, consisting of 5 dosimeters each, the average recovered dose and its standard error was calculated. The relative standard error for dosimeters removed from the reader is about 19% compared to 0.6% when there was no removal of the dosimeter, again showing that removal of dosimeters from the reader has a significant impact on the reproducibility and accuracy of dose recovery.

3.1.3 Summary of cross-talk results

Table 1 shows that our recovered doses differ from the known dose by about 30–85% when the dosimeter is removed from the reader and stored for 4 hours before measurement, compared to 2–4% for dosimeters that remained

in reader throughout irradiation and measurement with no wait time in between. For the latter, it is impossible to isolate what part of this deviation is due to removal of the dosimeters and what part is due to increase in UV signal with time after irradiation. According to Yuki-hara & McKeever (2006), the UV signal increase after 4 hours is about 33%. While this can explain some of the deviation seen, it is important to mention that for the 250 s irradiation, the recovered dose was about 57% less than the known dose. This cannot be explained by the UV effect. Additionally, the test by Yuki-hara & McKeever (2006) that showed the increase of UV signal with measurement time after irradiation was performed with 20 Gy. We do not know if this effect also occurs for very low doses like those measured here and to what extent. Furthermore, our second test eliminated effects of wait-time, but it also showed poor reproducibility when the dosimeters were removed, further indicating that removal of the dosimeter introduces some variability to the result

3.2. Dose rate reduction using attenuators

In a second set of experiments the dose-rate of the source was reduced by placing the absorber materials described in section 2.2 between source and dosimeter. For this set of experiments direct irradiation was used as opposed to the cross-talk used above. Unless otherwise noted, the attenuators used for this investigation are the 1.9 mm PTFE disk, 1.2 mm microscope coverslip glass stack, 1.4 mm wine glass disk and the 1.8 mm ABS plastic disk.

3.2.1 Dose rate reduction factor and reproducibility

To measure the amount by which the attenuator reduces the dose rate, i.e. the “dose rate reduction factor,” a dosimeter was exposed to a fixed dose of 10 s without the attenuator and then the readout procedure as described in section 2.4 was performed with a test-dose of 4 s (given without the attenuator) to obtain (S_1/S_T) . In a second step, the same dosimeter was irradiated with the attenuator inserted for (S_2/S_T) . The ratio $k = (S_1/S_T)/(S_2/S_T)$ corresponds to the *dose rate reduction factor* of a specific attenuator used, where S_1 represents the signal from exposure without the attenuator ma-

Attenuator description	k	$k_t(mm^{-1})$
10 mm diameter, 1.4 mm thick Wine Glass disk	4.11 ± 0.04	2.94 ± 0.04
12 mm diameter, 1.2 mm thick Microscope Coverslip stack	3.61 ± 0.03	3.02 ± 0.04
8.2 mm diameter, 1.4 mm thick PTFE disk	3.34 ± 0.04	2.39 ± 0.03
8.2 mm diameter, 1.5 mm thick ABS plastic disk	2.07 ± 0.04	1.38 ± 0.03
8.2 mm diameter, 1.9 mm thick PTFE disk	5.33 ± 0.07	2.81 ± 0.04
8 mm diameter, 1.9 mm thick Microscope Coverslip stack	5.97 ± 0.10	3.14 ± 0.06

Table 2. Dose rate reduction factors k for the absorber materials investigated. The mean was calculated from 6 dosimeters per attenuator material, the uncertainties represent the standard error. k_t is the dose reduction factor per thickness

terial, where S_1 represents the signal from exposure without the attenuator and S_2 represents the signal from exposure with the attenuator. The results for the various attenuators is presented in Table 2. For easier comparison the reduction factor per 1 mm thickness, k_t is listed as well

To test reproducibility in absorber placement, for each absorber material 10 dosimeters were irradiated for 10 s. The same absorber stack was used in each case to eliminate possible variation between stacks of the same material. The test dose corrected signals, normalized to the average value for each material are presented in Fig 7. Table 3 lists relative standard deviations for all materials, including experiments without absorber and results for an experiment with 10 different coverslip stacks.

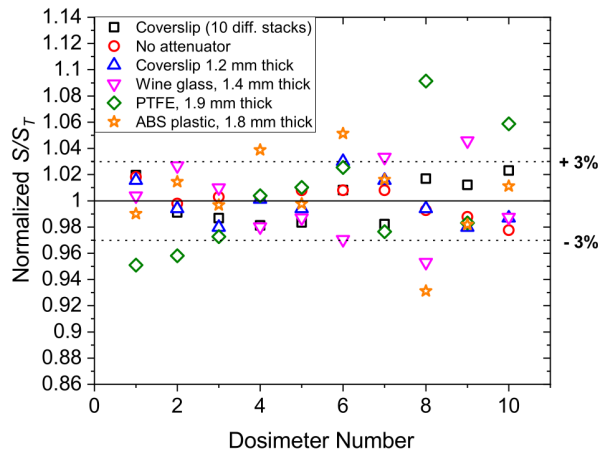


Figure 7. Absorber reproducibility. The same absorber stack was used for 10 dosimeters per material. Test dose corrected signals were normalized to the average value for each material. For comparison, data obtained without absorber and data obtained with 10 different coverslip stacks are shown as well. The normalization line (bold line) and the $\pm 3\%$ spread from the mean lines (dotted lines) are also indicated

Attenuator description	$\frac{\sigma}{\mu}(\%)$
PTFE, 1.9 mm	1.4
Coverslip 1.2mm thick	0.6
Wine glass, 1.4 mm	0.9
ABS plastic, 1.8 mm	1.0
No attenuator	0.4
Coverslip (10 different stacks)	0.5

Table 3. Absorber reproducibility. Relative standard deviations for 10 dosimeters per material. Data obtained without absorber and data obtained with 10 different coverslip stacks are shown as well.

3.2.2 Dose response/Calibration curve and dose recovery

The overall goal of this study is to use the built-in source of the Risø reader to build a dose response that will be used as reference curve for externally irradiated dosimeters. Dose response curves for the different attenuators were built, by exposing three dosimeters per given dose to 10 different doses in the range 1–100 s (with attenuators). The actual dose given to each dosimeter will depend on the attenuator used. Attenuators were removed after irradiation and before measurement, since some of the attenuators are opaque. Dose responses for uncorrected and test-dose corrected signals are shown in Fig. 8. Yukihiro et al. (2005) and Akselrod et al. (2000), reported linearity only for the uncorrected dose response in this dose range. In our case, both responses are linear in this dose range. Although, the corrected dose response shows a better linearity compared to the uncorrected dose response.

The accuracy in dose determination using the dose response curves in Fig. 8 was assessed by giving known doses to 4 dosimeters each (with the attenuator in place), then measuring the dose as if the dosimeters had absorbed an unknown burial dose. Doses were recovered based on S/S_T values and also the uncorrected S values. The recovered dose is then compared to the known dose using dose recovery ratio. The dose recovery results are presented in Table 4.

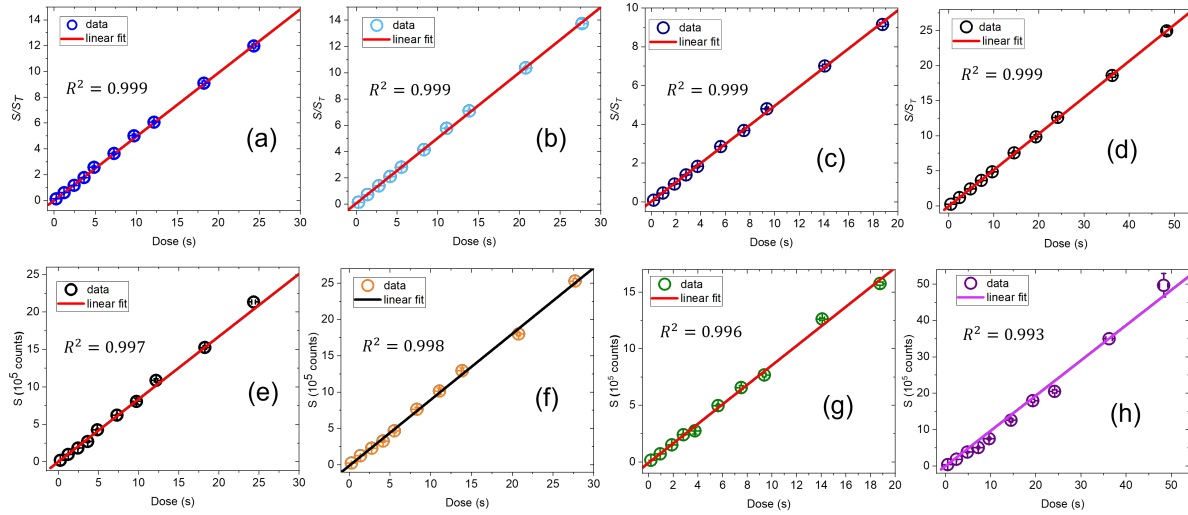


Figure 8. Test dose corrected (upper row) and uncorrected (lower row) dose responses of $\text{Al}_2\text{O}_3\text{:C}$ irradiated with wine glass attenuator (a and e), microscope coverslip glass (b and f) PTFE (c and g) and ABS (d and h). Each data point represents the average and standard error for 3 dosimeters. The adjusted R^2 is also represented to show the goodness of the fit

Attenuator	Known	test-dose corrected		uncorrected	
	Dose (s)	Recovered dose (s)	Recovery Ratio	Recovered dose (s)	Recovery Ratio
PTFE	8	8.30 ± 0.06	1.04	7.83 ± 0.10	0.98
	17	17.97 ± 0.12	1.06	17.57 ± 0.23	1.03
	43	44.84 ± 0.28	1.04	40.62 ± 0.53	0.95
	77	81.59 ± 0.51	1.06	78.07 ± 1.01	1.01
Coverslip glass	8	8.18 ± 0.05	1.02	7.50 ± 0.08	0.94
	17	17.22 ± 0.10	1.01	15.62 ± 0.16	0.92
	43	44.36 ± 0.25	1.03	44.16 ± 0.45	1.03
	77	73.81 ± 0.42	0.96	68.87 ± 0.70	0.89
ABS plastic	8	7.88 ± 0.03	0.99	7.14 ± 0.18	0.89
	17	16.90 ± 0.07	0.99	15.20 ± 0.38	0.89
	43	42.02 ± 0.16	0.98	38.88 ± 0.96	0.90
	77	76.35 ± 0.29	0.99	78.64 ± 1.95	1.02
Wine glass	8	7.65 ± 0.05	0.96	6.50 ± 0.078	0.81
	17	16.63 ± 0.10	0.98	15.64 ± 0.16	0.92
	43	42.91 ± 0.24	1.00	44.52 ± 0.46	1.04
	77	74.95 ± 0.43	0.97	67.87 ± 0.69	0.88

Table 4. Summary of dose recovery for irradiation with attenuator. Average recovery ratios for the test-dose corrected data are (1.05 ± 0.01) for PTFE, (1.01 ± 0.02) for coverslip glass, (1.01 ± 0.02) for ABS, and (0.98 ± 0.01) for wine glass.

The goal is to compare externally irradiated dosimeters with the calibration curves. It was therefore imperative to test if calibration curves built with attenuators can be used to recover doses administered without an attenuator. 12 dosimeters were irradiated without attenuator to assess if the attenuator itself influences the dose recovery. It is important to note that the test dose in the readout procedure must be the same

as the test dose in the readout procedure used to build the dose response curve. Using the known dose reduction factors (see Table 2) and the test-dose corrected dose response curves (Fig. 8), measured and given doses were compared (Table 5).

Known Dose (s)	PTFE		coverslip glass		wine glass		ABS plastic	
	Recovered dose (s)	Recovery ratio	Recovered dose (s)	Recovery ratio	Recovered dose (s)	Recovery ratio	Recovered dose (s)	Recovery ratio
1	1.00 ± 0.02	1.00	0.95 ± 0.02	0.95	0.97 ± 0.02	0.97	0.97 ± 0.06	0.97
2	2.08 ± 0.04	1.04	1.98 ± 0.04	0.99	2.03 ± 0.03	1.02	2.01 ± 0.13	1.01
3	3.20 ± 0.06	1.07	3.05 ± 0.06	1.02	3.12 ± 0.05	1.04	3.1 ± 0.2	1.03
4	4.22 ± 0.07	1.06	4.02 ± 0.08	1.01	4.12 ± 0.07	1.03	4.1 ± 0.3	1.02
5	5.33 ± 0.09	1.07	5.07 ± 0.10	1.01	5.19 ± 0.08	1.04	5.15 ± 0.33	1.03
6	6.26 ± 0.11	1.04	5.96 ± 0.12	0.99	6.10 ± 0.10	1.02	6.06 ± 0.38	1.01
7	7.41 ± 0.13	1.06	7.04 ± 0.14	1.01	7.22 ± 0.12	1.03	7.17 ± 0.45	1.02
8	8.45 ± 0.15	1.06	8.03 ± 0.16	1.00	8.23 ± 0.13	1.03	8.17 ± 0.52	1.02
9	9.76 ± 0.17	1.08	9.28 ± 0.18	1.03	9.51 ± 0.15	1.06	9.44 ± 0.60	1.05
10	10.63 ± 0.19	1.06	10.11 ± 0.20	1.01	10.36 ± 0.17	1.04	10.29 ± 0.65	1.03
11	11.74 ± 0.21	1.07	11.16 ± 0.22	1.01	11.44 ± 0.18	1.04	11.35 ± 0.72	1.03
12	12.67 ± 0.22	1.06	12.05 ± 0.24	1.00	12.35 ± 0.20	1.03	12.26 ± 0.78	1.02

Table 5. Dose recovery results for dosimeters irradiated without attenuator using the dose-response curves in Fig 8, produced from the S/ST data for irradiation with attenuators. The average recovery ratios are (1.05 ± 0.01) for PTFE, (1.00 ± 0.01) for coverslip glass, (1.03 ± 0.02) for wine glass, and (1.02 ± 0.01) for ABS plastic.

4. Discussion

4.1. Dose rate reduction using irradiation cross-talk

The irradiation cross-talk on the position adjacent to the irradiated position as measured here is about 0.05% of the direct dose. This is a little higher than the 0.04% reported by Kalchgruber et al. (2002) for Al₂O₃:C dosimeters and 8 times higher than the 0.006% reported by Bray et al. (2002) for quartz discs. But it is about 3 times lower than the 0.17% reported by Bøtter-Jensen et al. (2000) for quartz coarse grain. This result confirms previous observation by Kalchgruber et al. (2002) that values of cross-talk depend on the individual reader, the sample holder and the dosimeter material used.

Dose recovery tests resulted in excellent agreement between given and measured dose, when the reader remained closed between irradiation and measurement (Table 1). However, Table 1 also illustrates that a reference dose response built with irradiation cross-talk is not suitable for externally irradiated dosimeters. When dosimeters are removed from the reader between irradiation and measurement, recovered doses deviate 50-80% from the expected values. Recovered doses were well below and also well above the expected values.

This high variability in measurement response is suspected to be partly due to the increase in UV luminescence signal with time of measurement after irradiation that was reported by Yukihiro & McKeever (2006), due to the 4-hour wait period before the dosimeter was read, which was done to simulate external exposure of the dosimeters. However, even this effect doesn't fully explain the huge deviation observed as can be seen from the 19% compared to 0.6% reproducibility observed for the case where the dosimeters

were taken out of the reader before measurement compared to when they were not even when the wait time before measurement were the same. Similar variation has been reported by Vargas (2011). She reported a 9% relative standard deviation for a reproducibility test on a single dosimeter that underwent twelve 1.26 Gy irradiation/measurement cycles when the reader is opened, and the dosimeter is exposed to room light from a weak red bulb between irradiation and measurement. Our result for a similar test using 10 dosimeters yielded a reproducibility of 1%. Vargas also reported high variability for a fading test where the dosimeter was taken out of the reader and kept in a dark case for a period before measurement. She concluded that this effect is due to change in the position of the dosimeter on the cup from one irradiation-measurement cycle to another (Vargas, 2011).

It can be argued that in the case of cross-talk, where scattered radiation is used, the dose has a significant dependence on the exact position of the dosimeter in the sample holder. We therefore recommend that irradiation cross-talk, when considered as an option for dose rate reduction, should be tested for reproducibility and dose recoverability first.

4.2. Dose rate reduction using attenuators

Dose rate reduction with attenuators depends as expected on the density and thickness of the material (Table 2). The microscope coverslip glass stack provides the highest dose rate reduction per thickness at a factor of 3 per mm, followed by the wine glass, then the PTFE. The lowest dose rate reduction per thickness is that of the ABS rod which also has the lowest density among the materials investigated. The microscope coverslips and wine glass have the additional advantage of being transparent compared to the PTFE and ABS disks that are opaque. A transparent attenuator will

allow OSL measurements to be conducted without opening the reader to remove the attenuator before measurement. i.e., measurement will be automatic, requiring no user intervention until completion. Additionally, the microscope coverslip glass is the cheapest material investigated (US\$10 for a pack of 100).

Reproducibility test for all attenuators presented in Fig.7. shows that 83% of the S/S_T values are within $\pm 3\%$ of the mean. The overall relative standard error of the distribution is 0.5%. Yukihiro et al. (2005) reported 0.7% for Luxel[®] dosimeters, and Burbidge & Duller (2003) reported 2.1% for $Al_2O_3:C$ chips. This proves that the use of the attenuator doesn't introduce any substantial uncertainty to the measurement process. The relative standard error when a single coverslip stack was used for 10 dosimeters and when 10 different coverslip stacks were used are 0.6% and 0.5% respectively. The average thickness of the 10 coverslip stacks was calculated to be (1.40 ± 0.02) mm. This indicates that the coverslip stacks exhibit uniform response, even though there might be a slight nonuniformity in the glue used to bind the slips into a stack.

Attenuators allowed building a dose response for lower doses than usually accessible with the most commonly used built-in source. Dose response of $Al_2O_3:C$ dosimeters is more linear and accurate for all attenuator materials when test-dose corrected signals were used compared to when the uncorrected signal was used, as evident from the dose recovery result in Tables 4 and 5. The results in Table 4 indicate that on average, the dose response curve based on the test-dose normalized signal S/S_T yields better dose recovery results than that based on the uncorrected signal S . On average, the dose recovery ratios of doses recovered using S/S_T data is 1.00 ± 0.01 with a maximum of 1.05 ± 0.01 (for PTFE) compared to 0.94 ± 0.02 with a maximum of 0.91 ± 0.05 (for wine glass) for the uncorrected signals. Dose responses from S/S_T data have a slightly higher adjusted R^2 value (0.999) than those of the S data (0.993–0.998). Additionally, the use of a test-dose for $Al_2O_3:C$ Luxel[®] corrects for sensitivity variations between dosimeters, when they are punched out from the same Luxel[®] strip. Dose recovery from direct irradiation, yielded promising results as well. A higher deviation was generally seen for the PTFE attenuator. On average, the dose recovery ratio for the direct irradiation are 1.05 ± 0.01 for PTFE, 1.00 ± 0.01 for microscope coverslip, 1.03 ± 0.01 for wine glass, and 1.02 ± 0.01 for ABS plastic.

5. Conclusion

Irradiation crosstalk and attenuators were investigated as methods for reducing the dose rate of the built-in β -source of the Risø TL/OSL reader. For the Risø TL/OSL system used in this study, the irradiation crosstalk dose rate is about 0.05% of the value for direct irradiation. While, it was possible to extend the dose range to lower doses, dose recovery tests for dosimeters removed and re-inserted in the reader resulted in deviations of 50–80%. One possible explanation was attributed to an increase in UV luminescence with mea-

surement time after irradiation. Another possible reason for the large deviations is a change in the position of the dosimeter on the cup during handling of the carousel which led to the conclusion that response to cross-talk irradiation might be sensitive to dosimeter position. However, more tests need to be performed to quantify the positional dependence of cross-talk irradiation.

The use of low-cost attenuator materials, though limited in thickness, provides a cheap and reliable alternative for dose rate reduction for the Risø TL/OSL reader. Four attenuator materials were investigated: PTFE, microscope coverslip glass, wine glass and ABS plastic. All materials resulted in good dose recovery results and allowed building a standard reference dose response. Microscope cover glass reduces the dose rate by a factor of 3 for each 1 mm thickness. The material is transparent and does not have to be removed during measurement and is therefore recommended as attenuator of choice for dose rate reduction.

Acknowledgments

The authors would like to thank Dr. Kristina Thomsen for her detailed review and for helpful suggestions that improved the clarity of the manuscript.

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Appendix C: Additional Information on *DosiVox* and *DosiSed*

C.1 Monte Carlo radiation transport

The Monte Carlo (MC) simulations involve the use of a sequential collection of random walks to generate a life story of a particle and all its progeny [143]. A random walk is a Markov chain of stochastic events describing the passage of a particle through a material. Each step in the random walk can be related to behavior of the particle and, importantly, there is no dependence on any previous steps. The calculations that determine each step is based on the database of differential cross sections that are used to generate probability distribution functions that determines the mean free path, type of interaction, the energy loss and angular deflection in a step or event as well as possible secondary particle production [143].

A particle life story is followed until it either fall below a defined energy or exit the geometry. During the particle’s “existence”, results are tallied across specified regions. The result may include energy loss over certain distance and/or energy flux [30]. There are various Monte Carlo simulation codes for beta and gamma radiation transport, MCNP, EGS, ITS, PENELOPE, and GEANT. They all share similar characteristics of the ability to model complex physical systems, the treatment of particle interactions, the scoring of specified particle properties like dose deposition or energy loss, and the setting of physical constraints. Although, each code has specific advantages, or might be better suited for specific

applications. For example, the Geant4 libraries allow for the robust simulations of low energy electromagnetic physics and hadronic physics processes. It is also reliable for dosimetry in the case of archaeological and geological studies as reported by Gu  rin et al. [29].

It is beyond the scope of this dissertation to discuss in detail the physics involved in electron/photon transport modelling – that would only be appropriate in a more theoretical study. However, some concepts that are important in the approaches employed in Geant4 to solving the physical system will be discussed briefly. In Geant4, the physics models used to perform simulations are grouped into the Geant4 low energy electromagnetic physics constructor class [244]. This low energy physics class is derived from the PENELOPE (PENetration and Energy Loss of Positrons and Electrons) code from Baro et al. [245], since it is more accurate for low energy electrons than other physics class [246]. This PENELOPE code can accurately calculate the energy of secondary electrons produced by primary particles (including alpha particles) along their path of interaction. The accuracy of the Geant4 physics for alpha particles has been investigated in detail by [247].

In coupled electron and gamma models, electron transport computation is significantly more costly than gamma radiation transport due to the comparatively large number of interactions involved. The very high number of interactions is due to the high collisional cross-section that is associated with the long-range nature of Coulomb interactions. For larger initial energies, and thick geometries, the average number of collisions experienced by an electron until it is effectively stopped becomes very large, and detailed simulation is very inefficient [143]. To circumvent this, most of the MC codes use multiple-scattering theories, which allow the simulation of the global effect by combining many collisions in a track segment of defined electron steps. This method of simulation was referred to as “condensed histories” by Berger [147]. The multiple-scattering theories implemented in condensed simulation algorithms are only approximate and may lead to systematic errors, which can be made evident by the dependence of the simulation results on the adopted step length [248].

The PENELOPE code contains arguably the most accurate condensed history electron

transport algorithms of the major codes [249] and maintains physical accuracy of electron transport down to a few hundred eV according to its authors. It uses a mixed condensed history algorithm that partitions the treatment of “hard” and “soft” interactions. Hard interactions are infrequent catastrophic events leading to large energy loss and/or track perturbation and they are individually treated in detail, while soft interactions are collectively simulated using condensed history algorithms. A user defined energy loss or deflection cut-off parameter is used to separate hard and soft interactions. This parameter is kept relatively small to reduce the contribution of the soft component on the transport but high enough to maintain an acceptable level of computational efficiency. It is also important to mention that due to the nature of multiple-scattering theories and/or their implementations in the simulation, using a very short cut-off parameter may introduce spurious effects in the simulation results [143].

The accuracy of the MC simulation results thus depends on the quality of the implementation of the physics and the cut-off value (assuming all model input parameters are exact). The cut-off value is generally defined to be much smaller than the size of the detector and the range of the simulated particles. In a practical sense, the simulation uses this value to decide whether to simulate the interactions of a secondary particle. If the range of the secondary particle calculated is less than or equal to this value, the simulation of the particle is replaced by a local dose deposition. However, if the range of the calculated secondary particle is more than the cut-off value, the interaction of the secondary particle is simulated. Therefore, it is preferable to have a small cut-off value in order to represent more accurately the dosimetric system, but that requires a significant increase in simulation time. A compromise between accuracy and simulation speed must thus be investigated.

C.2 General architecture of Geant4 used in *DosiVox* and *DosiSed*

This section will give some basics of the functioning of Geant4 to aid understanding of its principles as it was applied in the programs *DosiVox* and *DosiSed*.

As mentioned in chapter 4, Geant4 is a set of class libraries written in C++. Each class defines the creation of objects that perform specific functions, such as creation of a virtual volume or calculating the energy deposited by an interacting particle. Data inside a class are called functions, while the possible operations are methods. This architecture, called "object oriented programming" allows Geant4 to create custom-made simulation programs using functions and objects predefined in classes from the libraries. A class can get its characteristics from another class i.e. a class can be called inside other classes. It is also possible to create new classes to handle specific tasks. *DosiVox* and *DosiSed* discussed in section 4.4, was built by the creation of new classes within the Geant4 class framework [153].

For successful running of simulations, the user can follow the creation rules of the Geant4 class structure, as well as specific construction instructions in Geant4, established by the physics of the particle-matter interaction in the libraries, in addition to the interdependence that exists between the different classes. Geant4 basically consists of 17 class categories and the interdependence of these classes is shown in Figure C.1

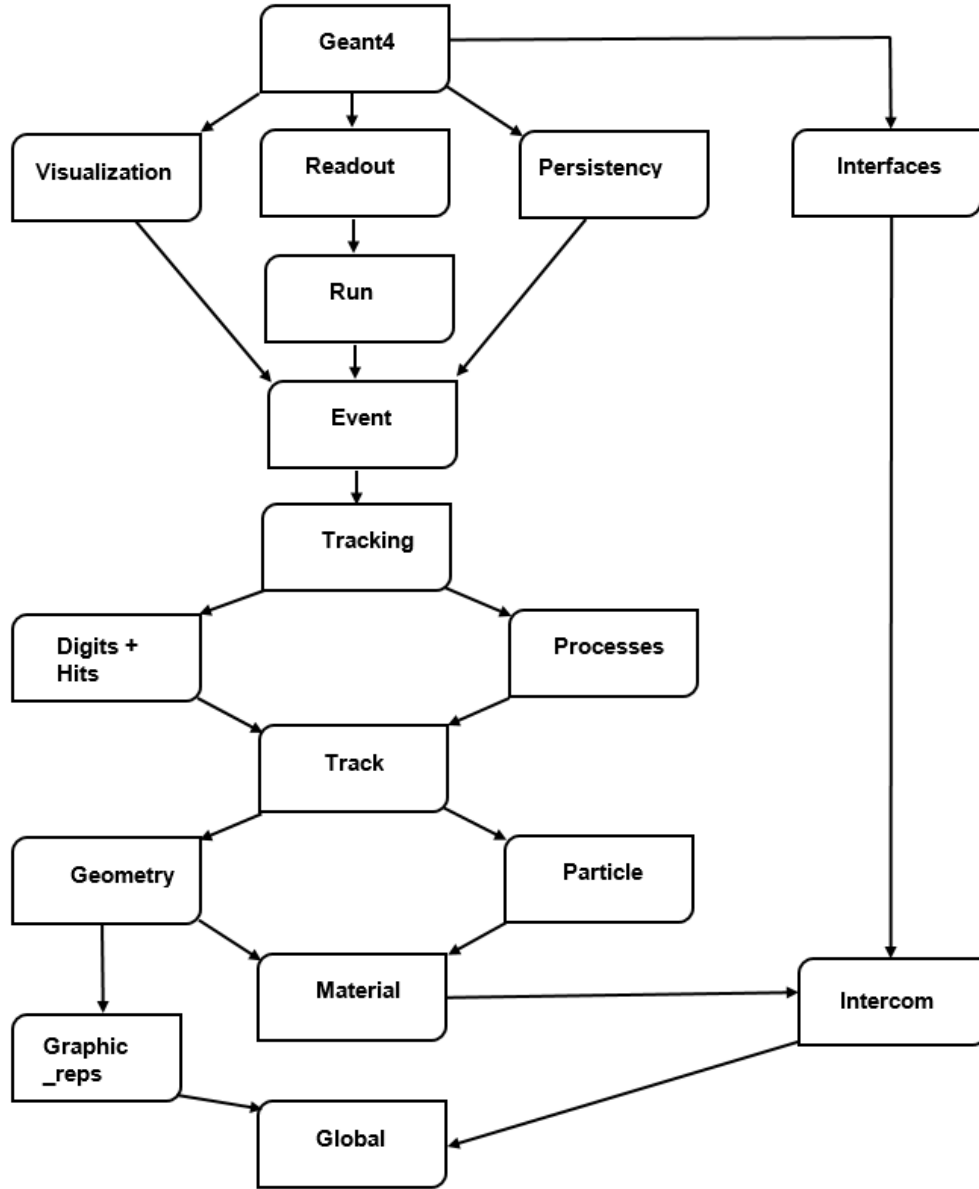


Figure C.1: The interdependence of the Geant4 class categories. Each category uses the category its arrow points to.

C.3 Equations used by Dosivox to adjust chemical composition and matrix density

$$\rho_{\text{wm}} = (\rho_{\text{w}} - f_{\text{g}} \times \rho_{\text{g}}) / (1 - f_{\text{g}}) \quad (\text{C.1})$$

$$f'_{\text{wm}} = WF \times \rho_{\text{d}} / (1 - f_{\text{g}} \times \rho_{\text{wm}}) \quad (\text{C.2})$$

$$f'_{\text{xm}} = f'_{\text{x}\bar{x}} / (1 - \rho_{\text{g}} \times f_{\text{g}} / \rho_{\text{d}}) \quad (\text{C.3})$$

$$f'_{\text{gm}} = (f'_{\text{x}\bar{x}} - \rho_{\text{g}} \times f_{\text{g}} / \rho_{\text{d}}) / (1 - \rho_{\text{g}} \times f_{\text{g}} / \rho_{\text{d}}) \quad (\text{C.4})$$

where, ρ_{w} is the average density of the wet material (matrix, grains, and water);

ρ_{wm} is the density of wet matrix (matrix and water only, without the grains);

f_{g} and ρ_{g} are the volume fraction of grains and density of grains respectively;

ρ_{d} is the average density of the dry material (matrix and grains);

WF is the water content of the material, expressed as a percentage of the mass of dry material;

f'_{wm} is the mass fraction of water in the matrix;

$f'_{\text{x}\bar{x}}$ is the mass fraction of any component x considered in the average material;

f'_{xm} is the mass fraction of any component x , considered in the matrix;

f'_{gm} is the mass fraction of the remaining component in the matrix from which the grains are created from;

C.4 *DosiSed* Pilot Text File

The following rules must be adhered to when making the pilot file:

- (i) metric unit of mm, g/cm³ and % must be used for depth or length, density, and proportion specification respectively.
- (ii) different data on a line must be separated by at least one space.
- (iii) scientific notations like 10E5, 10⁵, or 10^5 cannot be interpreted.
- (iv) use a single line break to separate sections. Multiple line breaks between sections will terminate the PTF.

Table C.1: An example of Pilot Text File (PTF) for *DosiSed* program. The comments for each line of code are written after the # symbol in teal. The comments are not interpreted by the program.

Line number	Executable code	Comment
1	Th_AlGr_Kb	# the name the result file is saved as
	# particle emission and properties #	
2	1000.	# ($\times 1000$) particles emitted
3	1	# 0 = alpha, 1 = beta, 2 = gamma
4	0	# radioactive elements, 0 = K, 1 = U-series, 2 = Th-series, 3 = User defined
5	3.0	# particles max range in mm
6	1.0	# cut-off value, in micrometer
	# voxelized geometry parameters #	
7	3.8	# cubic voxel size in mm
8	6 6 1	# number of voxels in the detector by X,Y and Z axis
9	0	# 0 = fixed medium, 1 = infinite medium
10	0	# 0 = random grain composition, 1 = recurrent grain composition
	# grains description #	
11	1.93	# total grain compacity in percent
12	2	# number of grain types
13	5	# number of grain sizes
14	# D	= spherical grain diameter in mm
15	# f	= volume fraction of this diameter
16	# Tn	= percent of grain type n in this diameter
17	D f T1 T2	
18	0.350 13.06 0 100	
19	0.300 13.05 0 100	
20	0.260 13.05 0 100	
21	0.090 30.42 100 0	
22	0.070 30.42 100 0	
23	Mtx T1 T2	
24	0 100 0	# relative percent of K in Mtx = matrix and Tn = Type n grain
25	0 100 0	# relative percent of U series in Mtx = matrix and Tn=Type n grain
26	0 100 0	# relative percent of Th series in Mtx = matrix and Tn = Type n grain
27	0 100 0	# relative percent of user defined radio-element in Mtx=matrix and Tn=Type n grain
	# material description #	
28	# new components	
29	1	# number of new component(s)
30	###	ID, name, density, number of atom type, list of atom symbol/number of atom of this symbol
31	18 thorite 5.35 3 Th 1 Si 1 O 4	
32	# material(s)	
33	0	# WF in the matrix, in percent
34	###	ID, name, density, number of atom type, list of component ID/mass fraction
35	0 def_matrix_mixture 1.129 1 1 100	# matrix
36	1 def_thorite 5.35 1 18 100	# type 1 grain material
37	2 def_alumoxide 3.97 1 2 100	# type 2 grain material

There are three mandatory objects, called "user classes" that must be well defined for any Geant4 program. The "DetectorConstruction", "PhysicsList", and the "PrimaryGeneratorAction" class. In general, in Geant4 simulations, the class DetectorConstruction is used in creating a virtual world, then primary particles are emitted inside the virtual world based on the parameters specified in the PrimaryGeneratorAction class. The simulation of each primary particle, from its emission to the complete absorption and the absorption of all its secondary

particles due to interactions in the volume or due to escape from the volume is recorded as an "event". The "run" represents all the events in a simulation and is normally chosen at the start of the simulation. During the simulation, the energy of each interaction and the volume where it happened is obtained at the "steppingAction" class after each particle step. This information is send to the "RunAction" class for result gathering. The RunAction class is used to define certain actions to be performed before, during or after a run. In *DosiVox* and *DosiSed* codes, this class and its functions were used to accumulate and order the doses during simulation, and then write the results in output files. See Figure C.1 to see the interdependence of some of the classes mentioned above. For detailed information on the architecture of Geant4 consult the following cited literatures [153, 154].

PTF explanation

In Table 4.1, the header: Line number, Executable code and Comment are not part of the PTF. It was only included to allow for easy reference of the different sections of the PTF for the explanation that follows:

- Line 1: This is the name with which simulation result will be saved as. The simulation result can be found in a **result** folder inside the *DosiSed* folder. For the simulation in this work, the suffix **Kb** in the result name was used to identify the radionuclide and particle simulated; K for ^{40}K and b for β particle.
- Line 2: This number times 1000 represents the number of particles emitted from the radioactive materials in the simulated. For example when the number in line 2 is 100, the total number of particles emitted during the simulated will be 10^5 particles.
- Line 3: This is where you specify the type of primary particle to emit during simulation (Note: the secondary particles can be any sort of particles resulting from the interaction and defined in the PhysicsList). Only one particle can be simulated or emitted at a

time. For example, this PTF file was used to simulate β particles only. To simulate α particles, this line will be changed to 0.

- Line 4: This is where you specify the radionuclide for the emission of radiation. In this case ^{40}K was selected. You have the option of simulating the β and γ spectra of U-series (^{238}U and ^{235}U) and Th-series under secular equilibrium and ^{40}K , as well as the α spectra of U-series and Th-series via their emission spectra files that are stored in the *DosiSed* folder `data/spectra`. The **User defined** option 3 can be used to specify any other energy spectrum. See the *DosiVox* manual ¹ for more details on this. Also note that line 3 and 4 are used to fully specify the particle and radionuclide types. This is important because only one particle/radionuclide combination can be simulated at a time.
- Line 5: This is where you specify the maximum range of the primary particle simulated in mm. This is important for infinite matrix simulation. It is used to tell the code how much extra voxels to use to pad the dosimeter voxels to make sure electron equilibrium is not lost at the edges of the dosimeter voxels. If the value is shorter than the actual particle range, then the volume around the grid will not be large enough to be considered as infinite for the particle and the dose rate in the detector may be underestimated. The number of additional voxels used is calculated by multiplying the maximum range specified in line 5 by 1.2 and dividing by the voxel size.
- Line 6: This is where you specify the cut-off value for secondary particles simulation in μm . The simulation uses this value to decide whether or not to simulate the interactions of a secondary particle. Based on this value, if the range of the secondary particle calculated is less than or equal to this value, the simulation of the particle is replaced by a local dose deposition. However, if the range of the calculated secondary particle is more than the cut-off value, the interaction of the secondary particle is simulated. it is

¹https://www.iram-at-crp2a.cnrs.fr/spip/IMG/zip/dosivox_manuals-3.zip, last accessed on May 6, 2022

preferable to have a small cut value in order to represent more accurately the dosimetric system, but that require increased simulation time as more secondary particles will be generated. A compromise between accuracy and simulation speed must be investigated.

- Line 7: This is where you specify the dimension of the voxel that will be used to define the volume of the sample or simulation space. Each voxel must be a cube and the value here represents the side length of the cubic voxel. For example, a cubic voxel size of 3.37 mm means a cubic voxel volume of $3.37 \text{ mm} \times 3.37 \text{ mm} \times 3.37 \text{ mm}$.
- Line 8: This is where you specify how many of the voxels specified in Line 7 will be used to create the volume of the sample or simulation space. This is done by specifying the number of voxels in x-, y-, and z-axis. In *DosiSed*, all the voxels specified here will be used as detectors also. In other words, the dose deposition in these voxels and the grains inside these voxels will be compiled into a result file.
- Line 9 and 10: Line 9 is used to tell the program to add extra voxels to the detector voxels specified in Line 8 in order to ensure that the dose deposition calculations for the detector voxels are based on the infinite matrix conditions i.e., do not suffer from loss of electronic equilibrium at the edges of the detector voxel. To do this use 1 for infinite medium. If you're interested in simulating only the detector voxels then use 0 for fixed medium. Line 10 is used to specify whether to randomized the grain compositions when there are more than one grain types of the same size. Randomized grain composition provides more diversity in the model (following the defined size and composition of grain). The periodic grain composition ensure that grains overlapping the voxel edge conserve the same material from a voxel to another, which is preferable if there are grains not significantly smaller than the voxel size.
- Line 11: This is used to specify the compacity of the total grain in the volume in %. It can be calculated as the total volume of grains divided by the total volume of the sample. For example, in the case above, the total volume of the sample is due to volume

of ThSiO_4 grains, Al_2O_3 grains, SiO_4 powder, and air gaps. This volume was calculated using the dimension of the container and the height of the sample in the container. Subsequently, the compacity was calculated as the volume of ThSiO_4 plus Al_2O_3 grains divided by the total volume. The volumes of ThSiO_4 and Al_2O_3 were calculated by dividing their mass by density. This compacity is replicated for each detector voxel.

- Line 12 : This is used to specify the number of grain types in the sample. For example, in this case the only ThSiO_4 and Al_2O_3 minerals are grains. So the the number is 2.
- Line 13 : This is used to specify the total number of grain sizes in the sample. For example, in this case the grain sizes of the ThSiO_4 mineral were 70 and 90 μm . The grain sizes of the Al_2O_3 mineral were 260, 300 and 350 μm . So the number is 5.
- Line 14–22 : This is used to specify the volume fractions of all the grain types and sizes that make up the total grain volume. If all the grains are of the same material and same size then this number should be 100. However, for grains of different materials and sizes, its the total volume fraction that should equal to 100. In the case of the pilot file in Table 4.1, the grains are of two different materials and 5 different sizes. The volume fraction of the ThSiO_4 grains (T1) was calculated by dividing the volume of the ThSiO_4 in the sample by the total volume. This was about 60.84%, and since two grain sizes were simulated for ThSiO_4 ; 90 μm and 70 μm . Dividing 60.84% by 2 gives 30.42% as the volume fraction of each grain size. The volume fraction of each Al_2O_3 grains size was also calculated in the same manner.
- Line 23–27 : These are used to specify the mineral(s) or portion(s) of the sample to emit radiation from and the type of radionuclide (U, Th or K or user defined) in %. For example in this case, the sample can be subdivided into T1 (ThSiO_4), T2 (Al_2O_3), and matrix (SiO_4 + airgaps). The radioemitter is ThSiO_4 while the other parts of the sample is inert. Therefore the relative percent of radioemitter in the sample is 0, 100, 0 for Matrix, T1, and T2 respectively. In the case where more than one portions or

minerals of the sample is radioactive, one can give equal weight to the portions but the total weight must be 100%.

- Line 28–31 : This is used to specify how many new components will be defined and also to defined the new component(s). Components are defined by a density and a chemical composition, and can be mixed in different mass proportion to create the materials used in the simulation. *DosiSed* comes with a predefined list of components. The list comprises 18 basic components referenced by their index, from 0 to 17, see the *DosiVox* manual ² for this list. Therefore, a new component can be created starting with index 18. There is no limitation to the number of new components that can be added. To define a new component follow the layout in line 30 and 31 where ThSiO_4 was defined.
- Line 32–37 : These are used to define the materials or mineral compositions of the sample. Line 33 gives water content of dry mass of the sample in %. Line 35 is used to define the material that makes up the matrix, in this case it is SiO_4 + airgaps. The density of the matrix (1.129 g/cm^3) in line 35 was calculated as $(m_{\text{Total}} - m_{\text{ThSiO}_4} - m_{\text{Al}_2\text{O}_3})/V_{\text{Total}}$, where m is mass and V is volume. Finally, in line 36 and 37, ThSiO_4 and Al_2O_3 were defined.

C.5 *DosiSed* Result file

An example of the result from *DosiSed* is presented below.

²https://www.iram-at-crp2a.cnrs.fr/spip/IMG/zip/dosivox_manuals-3.zip, last accessed on May 6, 2022

1	infinite matrix dose			0.000000017359 gray					
2	summary for all detectors								
3	average dose in the matrix (gray):						1.85E-09		
4	diam/mm	Gr_type	Gr_number	Av_Dose	STDEV				
5	0.35	1	72	1.14E-08	3.84E-08				
6	0.3	1	144	1.33E-08	7.49E-08				
7	0.26	1	180	4.05E-09	1.36E-08				
8	0.09	1	13392	5.49E-09	1.74E-07				
9	0.07	1	29268	4.48E-09	2.04E-07				
10									
11	summary by voxel detector								
12	voxel_No	diam/mm	Gr_type	Gr_number	Av_Dose	STDEV			
13	0	0	matrix	1.97E-09					
14	0	0.35	1	2	0	0			
15	0	0.3	1	4	2.03E-07	3.52E-07			
16	0	0.26	1	5	0	0			
17	0	0.09	1	372	1.23E-08	1.91E-07			
18	0	0.07	1	813	2.67E-09	7.60E-08			
19	1	0	matrix	2.47E-09					
20	1	0.35	1	2	1.59E-08	1.59E-08			
21	1	0.3	1	4	0	0			
22	1	0.26	1	5	0	0			
23	1	0.09	1	372	0	0			
24	1	0.07	1	813	4.29E-09	8.63E-08			
.									
.									
.									
.									
25	results by grain detector								
27	voxel_No	grain_No	Det_No	diam/mm	X/mm	Y/mm	Z/mm	Gr_type	Dose/gray
29	0	2	0	0.35	-0.83834	-0.35466	0.288597	1	0
30	0	3	1	0.35	-0.11367	0.898445	0.597607	1	0
31	0	4	2	0.3	-0.6963	-0.82204	0.362374	1	8.13E-07
32	0	9	3	0.3	1.03055	-0.36428	0.679597	1	0
33	0	10	4	0.3	-1.11917	0.837803	0.778023	1	0

The following describes the content of the result file from a *DosiSed* simulation.

- Line 1: This is sum of the energies carried by the emitted particles by unit of mass in a volume. It represents the infinite matrix dose in the case of an infinite and homogeneous medium. For a medium where the particles are emitted from only some part of the medium, this value must be appropriately corrected. The easiest way to do this is to a difference of mass correction. For example if the mass of the radioactive portion is 2 g and the total mass is 5 g, then the infinite matrix dose can be corrected using $(IMD \times 5g/2g)$. It is expressed in the unit of Gy.
- Line 3 Reports the mean dose in the matrix.
- Lines 4–9: output a summary of the results for all grains in the simulation. The first column "diam/mm" outputs the diameter of the grains in mm. The column "Gr_type" reports the mineral of the grain based on their assigned number in the pilot file. The column "GR_number" reports the total number of the grains simulated for each size and mineral defined in the pilot file. The columns "Av_Dose" and "STDEV" as the names indicate, report the mean and standard deviations of each size and mineral defined in the pilot file. The unit is in Gy.
- Lines 11–24: Report everything that Lines 4–9 reported but for each voxel in the simulation.
- Lines 25–33 Report everything that Lines 4–9 reported, but for each grain in the simulation. It does not report standard deviation or mean since each line of result is for a single grain. It also reports the position of the grain in the simulation in x-, y-, and z- coordinate system.

C.6 Simulation of the mixing of inert and radioactive grains

These results represent some simulation tests that were done as part of chapter 6 but could not be reported there because of page limitation.

C.6.1 Variation of dose rate from a radioactive grain to an inert one in a two-grain system

In sedimentary deposits, radioactive and inert grains occur next to each other. The grains are exposed to different dose rates. The differences in dose rate when two grains with different radioactivity are placed next to each other are investigated. No comparable results have been published in the literature. However, the result from such a simulation is expected to not violate the grain self-dose threshold. The model used here contains two quartz grains arranged side by side in the z-direction, separated by a negligible gap. The upper grain is radioactive while the lower one is inert. The diameter of both grains was 0.3 mm. The grains and their surrounding environment inside their voxels were made up of siliceous material with density 2.65 g/cm³. The beta dose rate variation for the ⁴⁰K, U- and Th-series spectra are investigated for the two-grain system by computing the fractional dose deposited in each grain (see Figure C.2). A second model was also set-up to investigate the α dose rate variation. The model is similar to the first except, except that it consists of 0.02 mm diameter grains and only the U- and Th-series spectra were simulated since ⁴⁰K does not produce α radiation. Figure C.3 shows an example of the two-grain system model with α radiation.

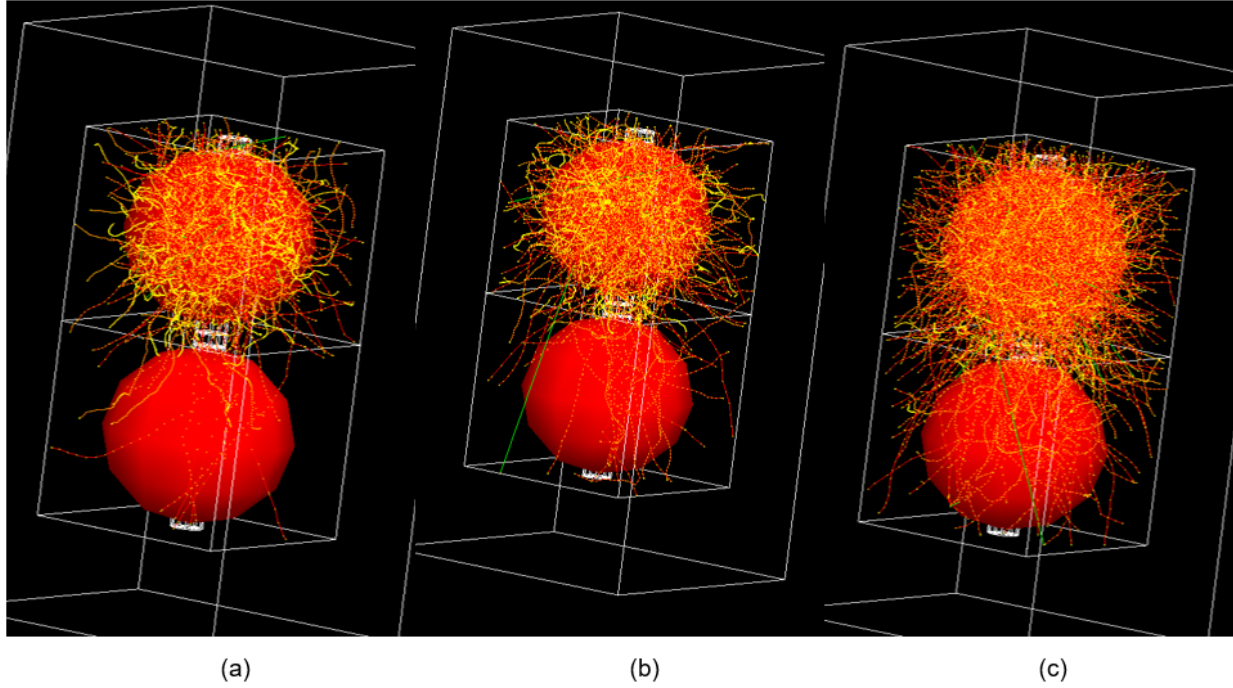


Figure C.2: Model of beta spectrum emission from (a) Th-series, (b) U-series, and (c) ^{40}K . Grain size is 0.3 mm. The top grain is radioactive while the bottom grain is inert.

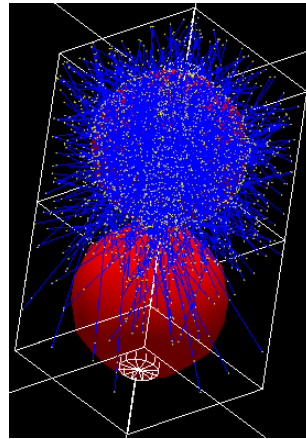


Figure C.3: Model of alpha spectrum emission from U-series. Grain size is 0.02 mm. The top grain is radioactive while the bottom grain is inert.

The results of these models are presented and discussed below:

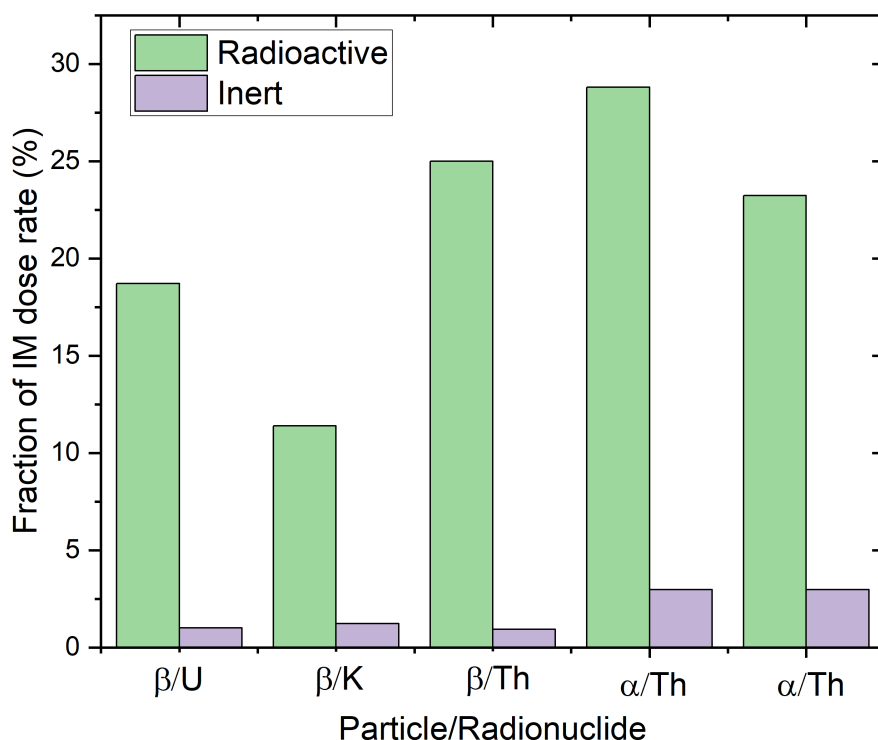


Figure C.4: Absorbed dose fractions in two grains placed next to one another. One of the grains is radioactive and the other is non-radioactive. The beta dose fractions were calculated for $300\mu\text{m}$ grains while the alpha dose fractions were calculated for $20\mu\text{m}$ grains.

The simulated beta and alpha dose fractions for the two grains are shown in Figure C.4. It can be seen that the amount of beta radiation attenuated by the radioactive grain is equivalent to the self-dose factors reported in Table 6.1, i.e., approximately 18.7%, 11.4%, and 25% for the U-series, ^{40}K , and Th-series respectively. The values for α radiation are in good agreement with the 29% and 23% for U-series and Th-series respectively reported in Table 6.3. The inert grains, on the other hand, received a considerably smaller dose rate compared to the radioactive grains, suggesting a sharp dose fall-off outside the radioactive grains and because most of the radiation is emitted in directions other than that of the inert grain. The fractional absorbed dose rates to the 0.3 mm inert grains, due to beta radiation are also considerably less than the fractional absorbed dose rates to the 0.02 mm inert grains due to alpha radiation. This is possibly due to the fact that the alpha particle range in SiO_2 is comparable to the sizes of the inert grain considered for alpha radiation. The alpha particle

will therefore end its path in the inert grain, thus depositing a high dose via the so-called "Bragg peak". The Bragg peak occurs immediately before an ionizing particle come to rest.

C.6.2 Dose profile inside an inert grain positioned next to a radioactive grain

Aitken [197] investigated the γ -dose rate profile for two adjacent soil layers, where one is inert and the other radioactive. He determined dose rate versus depth in each of the layers. His work has been reproduced by various authors [155, 157, 250]. The investigation here was designed to mimic this previous study, albeit for a single inert grain next to a radioactive grain and for alpha and beta radiation instead of gamma radiation. A vertical depth profile that cuts through the inert grain was used to study the dose profile in the grain. In this model, the diameters of the radioactive grain and the inert grain are kept the same due to the voxel size limitation of voxelized modelling. The dose rate profiles were computed using the cylindrical probe detector of *DosiVox*. The results are presented in Figures C.5, C.6, and C.7.

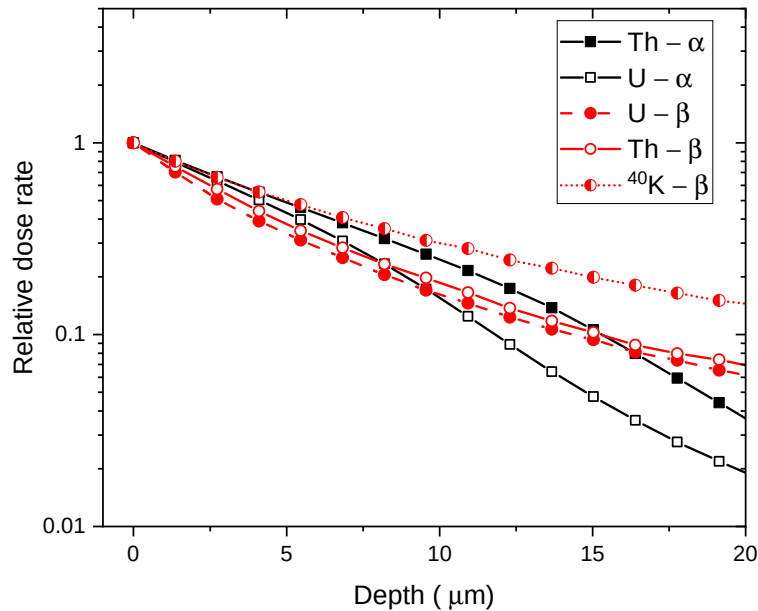


Figure C.5: Dose rate depth-profile in a 20 μm inert grain next to a 20 μm radioactive grain. Values were normalized to the surface dose rate of the inert grain.

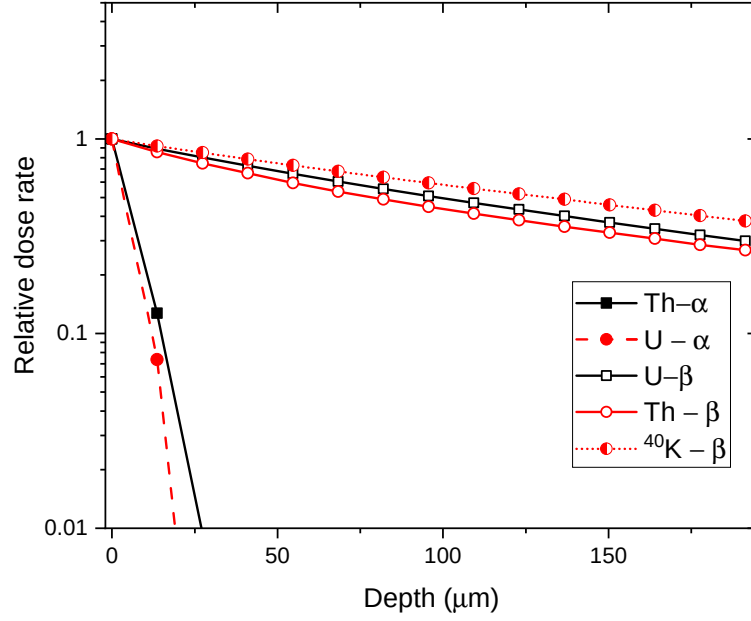


Figure C.6: Dose rate depth-profile in a 200 μm inert grain next to a 200 μm radioactive grain. Values were normalized to the surface dose rate of the inert grain

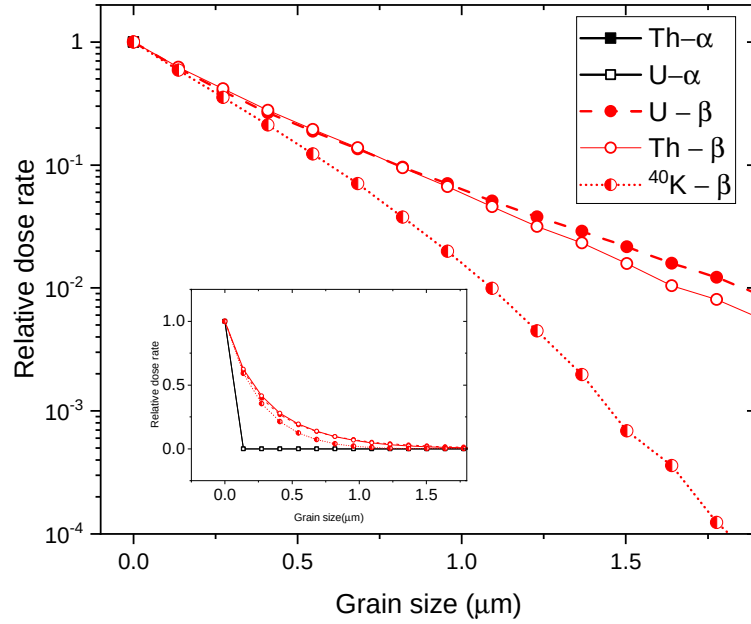


Figure C.7: Dose rate depth-profile in a 2 mm inert grain next to a 20 mm radioactive grain. Values were normalized to the surface dose rate of the inert grain. The inset allows the visualization of the dose rate profile due to alpha particles, which cannot be seen on log-linear plot.

In Figure C.5 to Figure C.7, The general trend observed is a faster fall-off of alpha dose compared to beta dose. This is due to the heaviness, i.e. the higher mass of alpha particles

compared to beta particles. A heavy particle has a denser ionization track and loses its energy faster than a light particle like beta. Alpha particles thus have a lower range when compared to beta in any material. It is quite interesting to see from the 20 μm grain in Figure C.5, that the dose fall-off in the first 10 μm layer of the grain is very similar for alpha and beta particles. One would expect that the beta dose rate in the top μm , and even the entire 20 μm grain layer would be fairly constant. This result proves the contrary. A possible explanation for this is the relatively high stopping power of low energy electrons in quartz. This can be visualized in Figure C.8. The plot and data were retrieved from the NIST Stopping Power & Range Tables for Electrons, Protons, and Helium Ions ³. The stopping powers of electrons and helium are shown for quartz. Notice that at low energies, the stopping powers are very similar. The rate of energy loss per unit mass, at every point along the track of a particle is proportional to the total stopping power. As the beta particles, that comprise a spectrum of electrons of different energies, enter the grain, the dose deposition at the outer layer will be mainly due the low energy part of the spectrum.

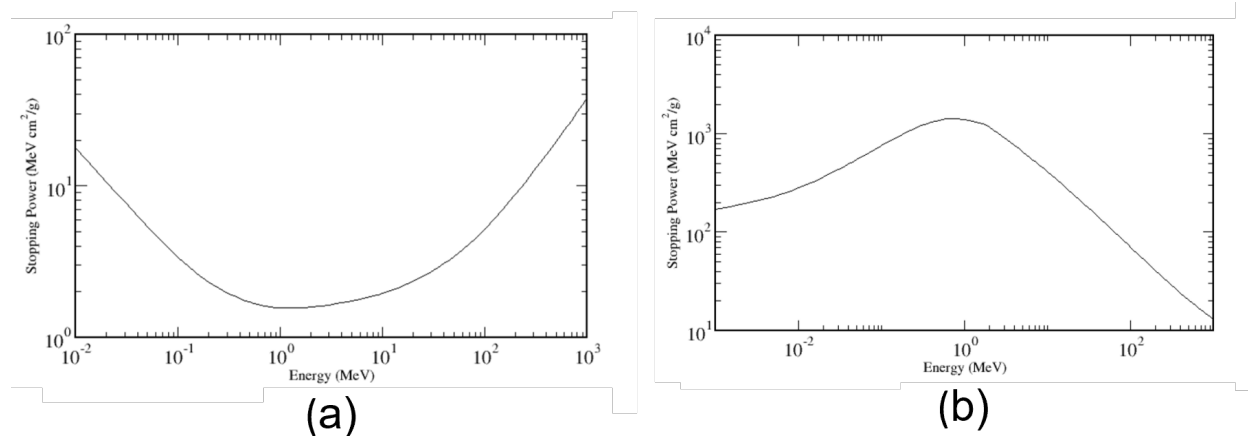


Figure C.8: Stopping power of (a) electron and (b) helium, in a quartz (silicon dioxide) medium.

As the grain size increases, representing deeper regions in the grain, we start seeing a clear distinction between the alpha and the beta dose rate profiles. In fact for the 2 mm grain, the dose profile for alpha was a single data point. In general, the dose falls off with

³<https://www.nist.gov/pml/stopping-power-range-tables-electrons-protons-and-helium-ions>, last accessed on May 14, 2022

depth in the inert grain. The rate of fall-off depends on the effective energy of the spectrum, the particle type and the size of the grain. This result is comparable to the result of β - and α -dose rate attenuation in inert silica layers near a radioactive layer [190, 251, 252]. All authors reported a depth dependent dose fall off for both particles.

Appendix D: Approaches and Assumption Applied to Model Sample and Dosimeter Geometry in Chapter 7

D.1 Model 1: Cylindrical model of container/sample using *DosiVox*

This approach seeks to construct the container and the sample as realistic as possible. It involves constructing the 3D image of the cylindrical container as well as the sediment inside it using ImageJ. The image was then converted to a sequence of text images representing slices of the sample and its container. Each slide is subdivided into voxels representing the mineral mixture in that slice of the sample. This model was investigated for the samples with Luxel[®] as dosimeters. Three different cylindrical models were created. In the first, the $x - y$ dimension of the sample and its container was divided into a grid of 25×25 voxels, the second was divided into a grid of 50×50 voxels for an improved resolution and finally, for the third, it was divided into a grid of 396×396 voxels using subvoxelization for fine resolution. As a result of the very small voxel size of the third model (~ 0.07 by 0.07 mm), the grain model was omitted and instead the sample was modelled as a uniform mix of quartz and radioactive minerals. The z -direction is used to represent the height of the model. The number of voxels in this direction thus depends on the height of the sample in the container.

The size of each voxel in this direction is chosen to be 0.3 mm to represent the thickness of Luxel[®] dosimeters. This is to ensure that a cluster of voxels in the $x - y$ direction can be used to represent a single Luxel[®]. See Figure D.1.

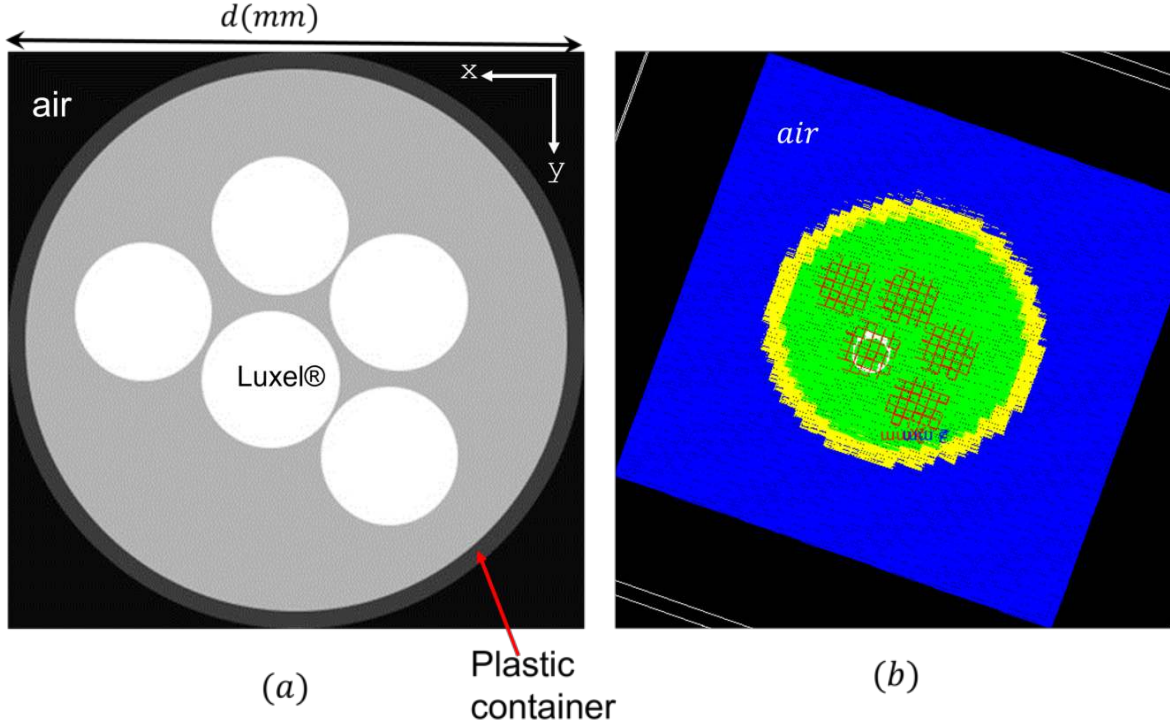


Figure D.1: 2D slice of one of the cylindrical models showing a slice of the sample where the Luxel[®] dosimeters were carefully arranged. (a) is the drawing of the sample slice using ImageJ. (b) is the 50×50 voxel grid representation of the sample slice in *DosiVox*

D.1.1 Model 2: Equivalent Perimeter (EP) model

Given that modelling in *DosiVox* is constrained to grids with cubic voxels, a model with rectangular or cubic geometry is much easier and straightforward to construct than a model with cylindrical or spherical geometry. However, our container and therefore sample including the Luxel[®] dosimeter are all cylindrical. Additionally, the β -dose deposited in our sample and/or detector will be dependent on the diameter and height of our cylindrical container due to the range of beta particles. Thus, one approach investigated is to approximate the cylindrical containers with a rectangular container of the same height, without compromising dose deposition based on the range of beta particles in the sample. To achieve this, the

cylindrical container was modelled as a rectangular container, where the rectangle is equivalent in perimeter to the perimeter of the circular cross section of the cylinder. This model is defined as the Equivalent Perimeter (EP) model (See Figure D.2). One implication of this approximation is that the total volume and mass of the sample might be less. However, this should not affect the result of the dose distribution since dose is energy normalized by mass. This model was also applied to the Luxel[®] dosimeter, i.e., the Luxel[®] dosimeter was represented as a rectangular disk instead of a circular disk. The height of the rectangular model will be the same as the height of the sample in the cylindrical container. A simplified form of this model involves just the sample in the container without including the container walls. Including the container walls is expected to account for any potential backscatter radiation from the internal surface of the container for dosimeters close to the surface. However, this is expected to be negligible since the containers are made of plastic. This simplified model was also used in *DosiSed* to model the geometry of samples with Al₂O₃:C grains as dosimeters. In the models where the container walls were included, the walls were modelled as High Density Poly Ethylene (HDPE) mineral with the chemical formula C₂H₄, and density 0.94 g cm⁻³.

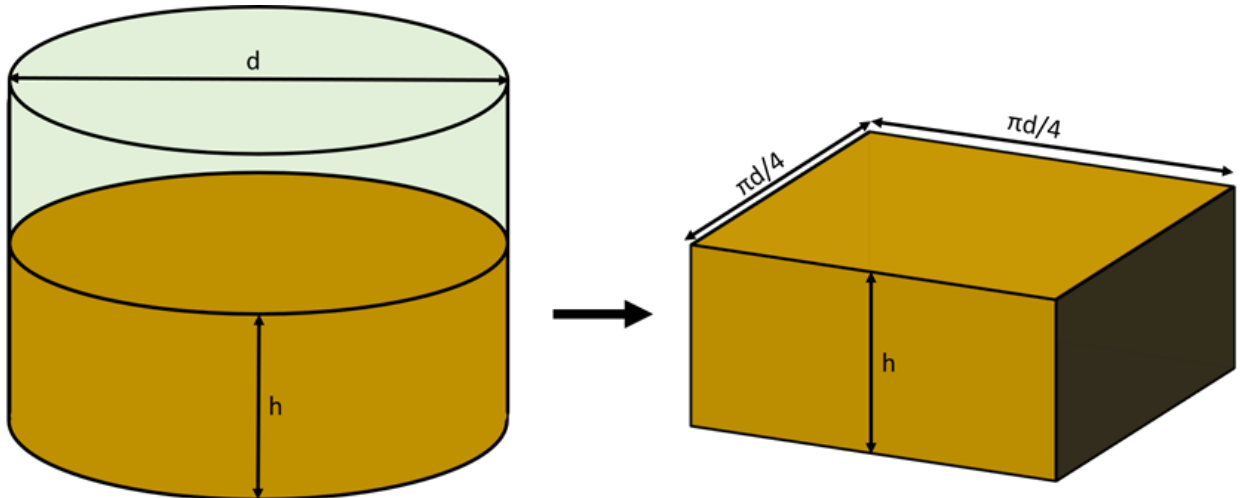


Figure D.2: 3D schematic representation of the Equivalent Perimeter (EP) model

D.2 Model 3: Equivalent Surface Area- to-Volume Ratio (ESAVR) model

In this case, the cylindrical container was approximated with a rectangular container that has the same surface area to volume ratio as the cylindrical container. The ESAVR model is expected to account for edge effects that the EP model is susceptible to, and that influence dose deposition. In this model, the cylindrical Luxel[®] dosimeter was modeled in two ways: (1) as a rectangular disk of side length equal to the diameter of the Luxel[®] and thickness equal to the thickness of Luxel[®]. (2) As a cylindrical disk that matches the exact dimensions of Luxel[®] using the subvoxelized option of *DosiVox* (see Figure D.3).

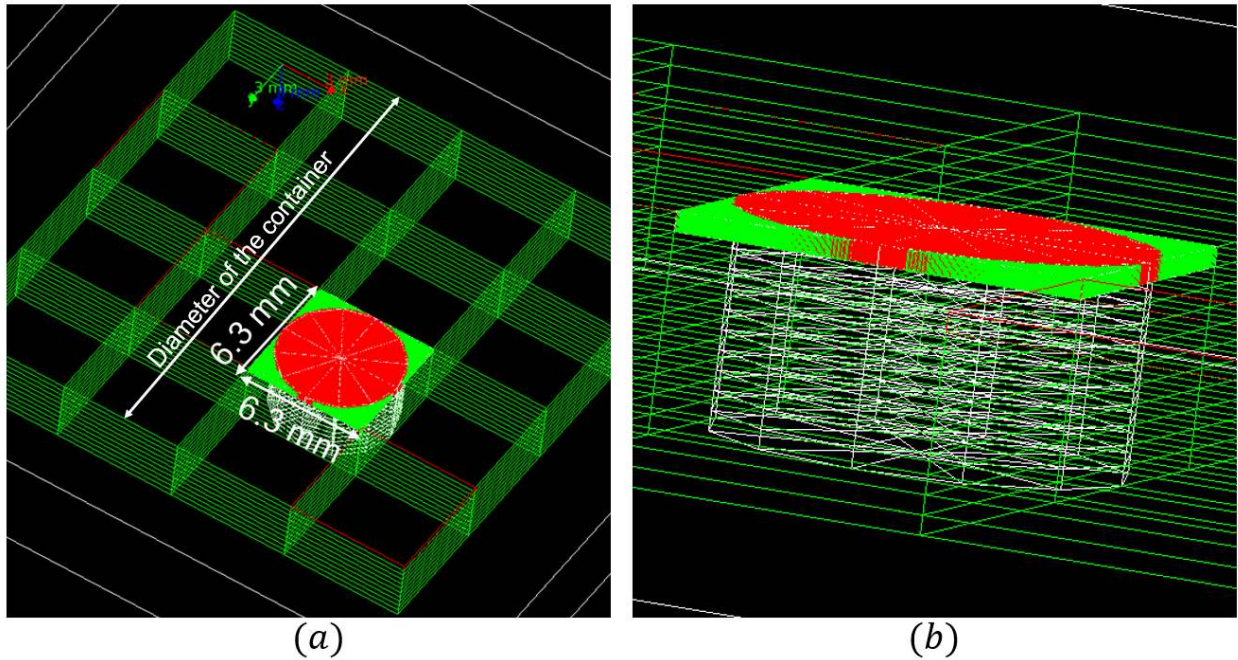


Figure D.3: (a) 3D view of a simplified ESAVR model. (b) Close-up view of the Luxel[®] dosimeter modeled to exact dimension using subvoxelization

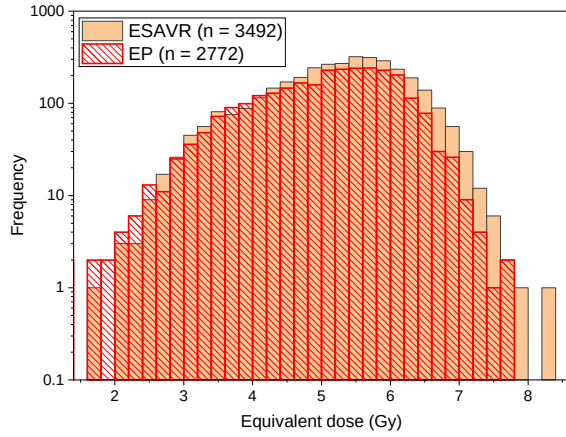
The following abbreviations are used to describe the result of these models in chapter 7 and in the following:

- EP (with or without container); the equivalent perimeter model where the sample container is included in the model or not included

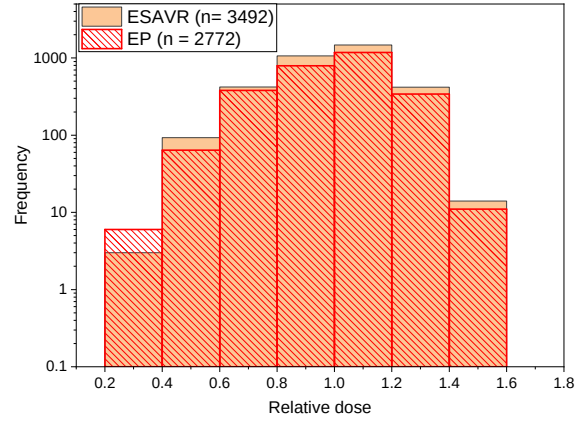
- Cyl_25, _50 and _396; cylindrical models where the sample is divided into a grid of 25×25 , 50×50 , or 396×396 voxels in the x-y direction respectively
- ESAVR (rect. or cyl. det.); the equivalent surface area-volume ratio model where the detector is modelled as a rectangle (EP model for detector) or as cylindrical (real shape).

D.3 Comparing Models: EP vs. ESAVR

Various approaches were used to simulate granular sediments and dosimeters with *DosiSed* (see section 7.3.3). In a first step results from the EP and ESAVR models were compared. In both models the cylindrical container is approximated as a rectangular container, but dimensions of the containers are different for the two models. Sample GS120 (large mass fraction of Thorite in a quartz matrix mixed with regular 250–355 μm $\text{Al}_2\text{O}_3\text{:C}$ grains) was modeled using the EP and ESAVR model. The dose distributions are shown in Figure D.4. The results show that there is good agreement between the two models except for slight differences due to edge effects, i.e., loss of electron equilibrium for grains closer to the surface. These grains experiences lower doses compared to grains farther from the surface. As can be seen from the distributions, ESAVR is slightly shifted towards higher doses compared to EP. This is because for ESAVR the surface-volume ratio is matched to the real sample and thus represents a bigger volume compared to the EP model where only the perimeter was matched. This can also be seen from the number of detector grains in the ESAVR model 3492 compared to 2772 for the EP model. However, due to the larger volume and thus increased number of grains, the ESAVR model requires considerably longer simulation time. The mean dose for the ESAVR simulated grains is (5.23 ± 0.96) Gy, and in agreement with the (5.08 ± 0.96) Gy for the EP simulated grains. Due to time constraints, all further grain-detector simulations with *DosiSed* were performed using the EP model.



(a) equivalent dose



(b) relative dose distribution

Figure D.4: EP vs. ESAVR model for sample GS120. (a) Equivalent dose distribution (b) Same distributions normalized by their respective mean values.

Appendix E: Calculation of the inactivation constant and lethal dose

Table E.1: Radiation Inactivation Constant k , for some dry microbial and some low temperature frozen microbial strains

Microbes	Irradiation Temp. (K)	k (kGy ⁻¹)	References
<i>B. subtilis</i>	310	1.01 ± 0.03^d	[119]
<i>B. thuringiensis</i>	310	1.00 ± 0.01^d	[119]
<i>B. pumilus</i> SAFR-032	193	0.11 ± 0.08	[46]
<i>Brevundimonas</i> sp. MV.7 ^A	193	1.85	[42]
<i>Rhodococcus</i> sp. MV.10 ^A	193	4.20	[42]
<i>Pseudomonas</i> sp. MV.27 ^A	193	9.99	[42]
<i>D. radiodurans</i>	193	0.24 ± 0.16^c	[42]

^d dry samples

^A Antarctic bacterial strains

^c calculation from the plot in the reference paper

The inactivation constant of *B. pumilus* SAFR-032 reported in the Table E.1 was calculated from that of *B. subtilis* using their relative response to UV based on data reported by Gioia et al [46]. The inactivation constant for *radiodurans* was calculated directly from the slope of its gamma dose survival curve reported by Dartnell et al [42]. For *radiodurans* and *B.*

pumilus SAFR-032, the inactivation constants were calculated by breaking the slope of their survival curves into smaller portions and then taking the average of the slope. The slopes that were averaged included the shoulder (where slope is $\sim$ zero). The data used for these calculations are presented in Figure E.2. Some tabulated radiation inactivation constants for other microorganisms studied under simulated Martian conditions are also presented in Table E.1.

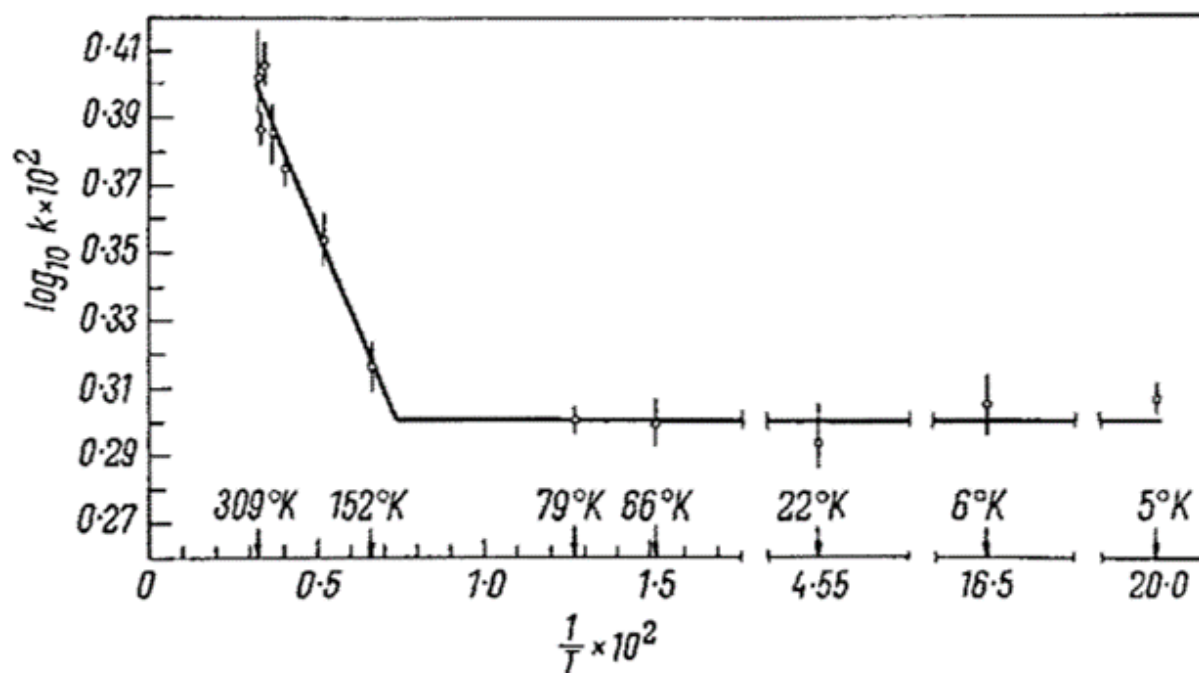
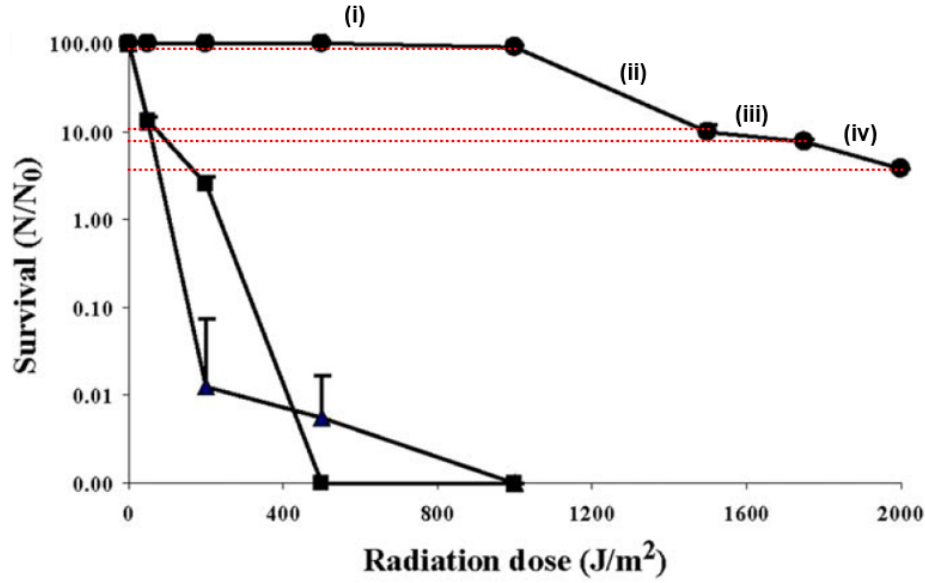
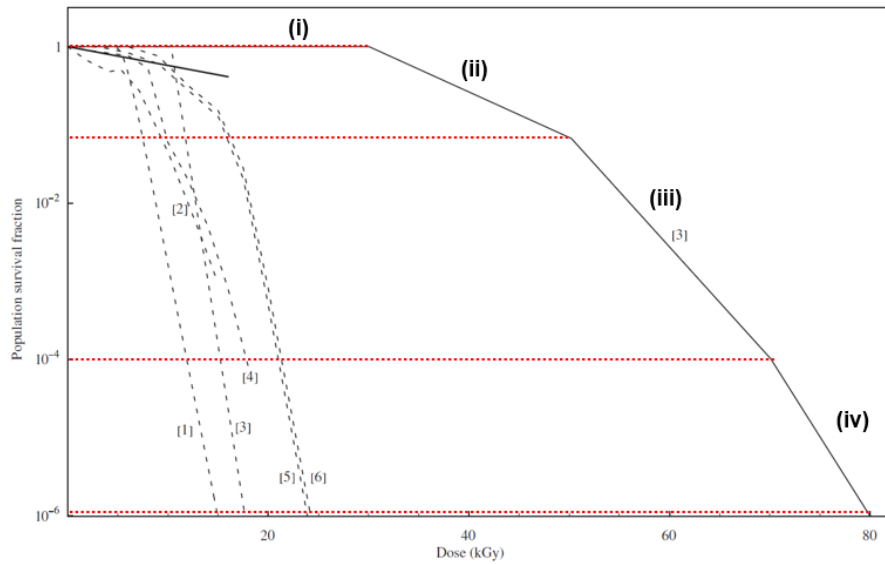


Figure E.1: Relationship between inactivation constant and the temperature of irradiation for dry microbial spores. Adapted from [220, Fig. 2]



(a)



(b)

Figure E.2: (a) Resistance of *B. pumilus* SAFR-032 and *B. subtilis* spores to UV radiation. This data was first used to calculate the relative survival of *B. pumilus* SAFR-032 and *B. subtilis* to UV radiation. The relative survival was then used to calculate the inactivation constant of *B. pumilus* SAFR-032 from the tabulated *B. subtilis* inactivation constant. (b) Solid line on the plot shows the survival of *radiodurans* as a function of gamma-dose in kGy. The inactivation constants for *radiodurans* and the relative survival for *B. pumilus* SAFR-032 were calculated by taking the average of the slopes indicated as (i), (ii), (iii), and (iv) on the plots. Figure adapted from [6, 46].

These inactivation constants were all determined from exposure to γ rays. Knowing the

inactivation constant of a microorganism and the dose rate of the radiation it is exposed to, one can in principle calculate the surviving fraction of that microorganism as a function of time in million years. However, the radiation inactivation constants of dry *B. subtilis* reported in the literature are in the range of $0.71\text{--}1.01\text{ kGy}^{-1}$ [119,253]. This difference is attributed to the difference in temperature during irradiation. Dry microbial spores show a temperature dependence in their radiation sensitivity (see Fig. E.1). The dry *B. subtilis* sample from Table E.1 was irradiated at 310 K, the temperature-corrected inactivation constant for a Martian subsurface temperature of 220 K is $\sim 0.90\text{ kGy}^{-1}$ based on Figure E.1. The temperature-corrected inactivation constants for the other spores listed in Table E.1 are presented in Table 9.3. Detailed reports of the dependence of bacteria irradiation survival on temperature conditions can be found in the following publications [42,219,254].

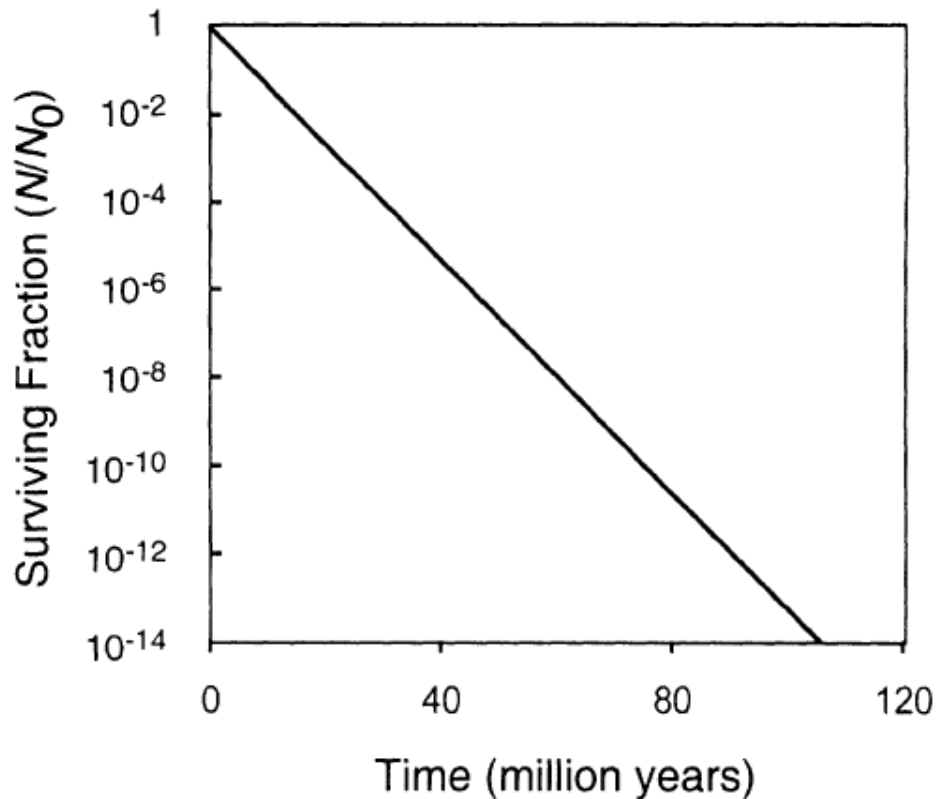


Figure E.3: Calculated surviving fraction of dry bacterial spores in the Martian subsurface as a function of time in million years using a dose rate of $0.13\text{--}0.35\text{ mGy/a}$. The starting point is 3.1 billion years ago. Variation in the radiation inactivation constant have no strong impact on the survival time [119, Fig. 3].

The lethal dose of some microbes have been reported in the literature [28, 40, 42, 219, 236, 254–258]. Most of these results were determined experimentally. The result by Pavlov et al. [28], however was based on the calculation of sterilization dose from radiation inactivation constants using Eq. 9.1 (in chapter 9). They determined a sterilization dose of about 20 kGy for *D. radiodurans* (corresponding to $N/N_0 = 10^{-8}$) using a radiation inactivation constant of $k=1 \text{ kGy}^{-1}$ and $n=5$. Based on this method, the lethal dose of various microbes can also be calculated directly from their temperature-corrected inactivation constants. The lethal doses of some microbes considered in this work, calculated using Eq. 9.1 and the temperature corrected inactivation constants in Table E.1 are presented in Table E.2.

Table E.2: Radiation Inactivation Constant k , for some dry microbial and some low temperature frozen microbial strains, and their resulting lethal dose values.

Microbes	Temp. corr. k (kGy^{-1})	Lethal dose (kGy)
<i>B. subtilis</i>	0.938 ± 0.028	24.97 ± 0.74
<i>B. thuringiensis</i>	0.926 ± 0.009	25.29 ± 0.25
<i>B. pumilus</i> SAFR-032	0.113 ± 0.082	207.26 ± 150.74
<i>Brevundimonas</i> sp. MV.7	1.902	12.31
<i>Rhodococcus</i> sp. MV.10	4.318	5.42
<i>Pseudomonas</i> sp. MV.27	10.271	2.28
<i>D. radiodurans</i>	0.242 ± 0.164	96.78 ± 65.61

Appendix F: Results of Martian subsurface simulations

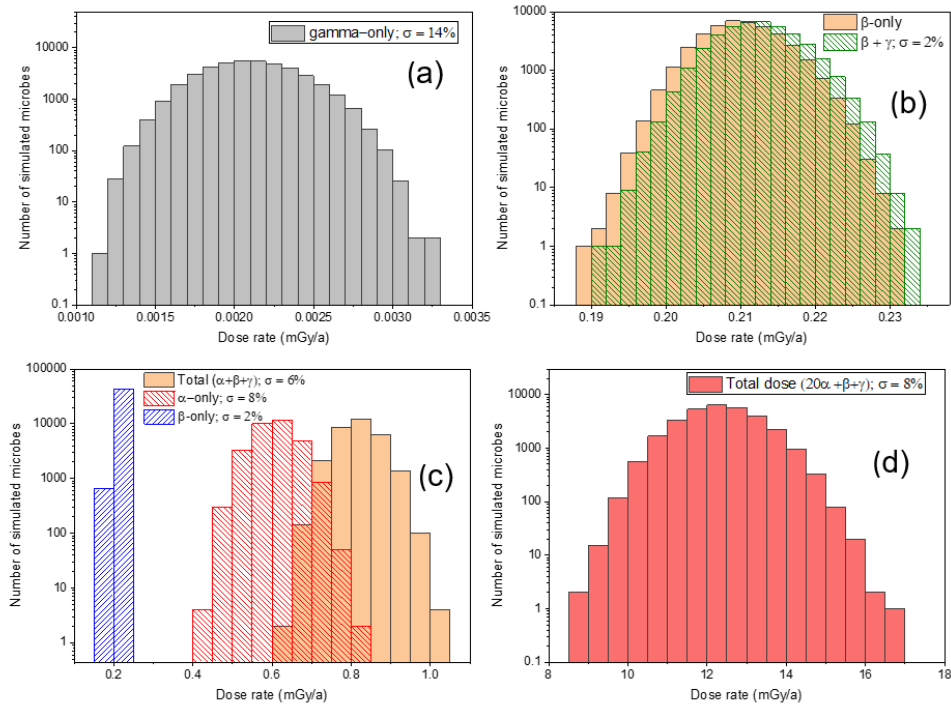


Figure F.1: Microscopic dose rate distribution of microbes in the sulphur-rich Gusev crater-type Martian subsurface permafrost. (a) Gamma dose rate distribution only. (b) Beta only and Beta + Gamma dose rate distributions. (c) Alpha only, beta only and total sum of alpha + beta + gamma dose rate distributions. Here the dose rate values were added directly. The total dose rate distribution in (d) was obtained by multiplying the alpha radiation portion of the dose distribution by 20 to account for the alpha quality factor.

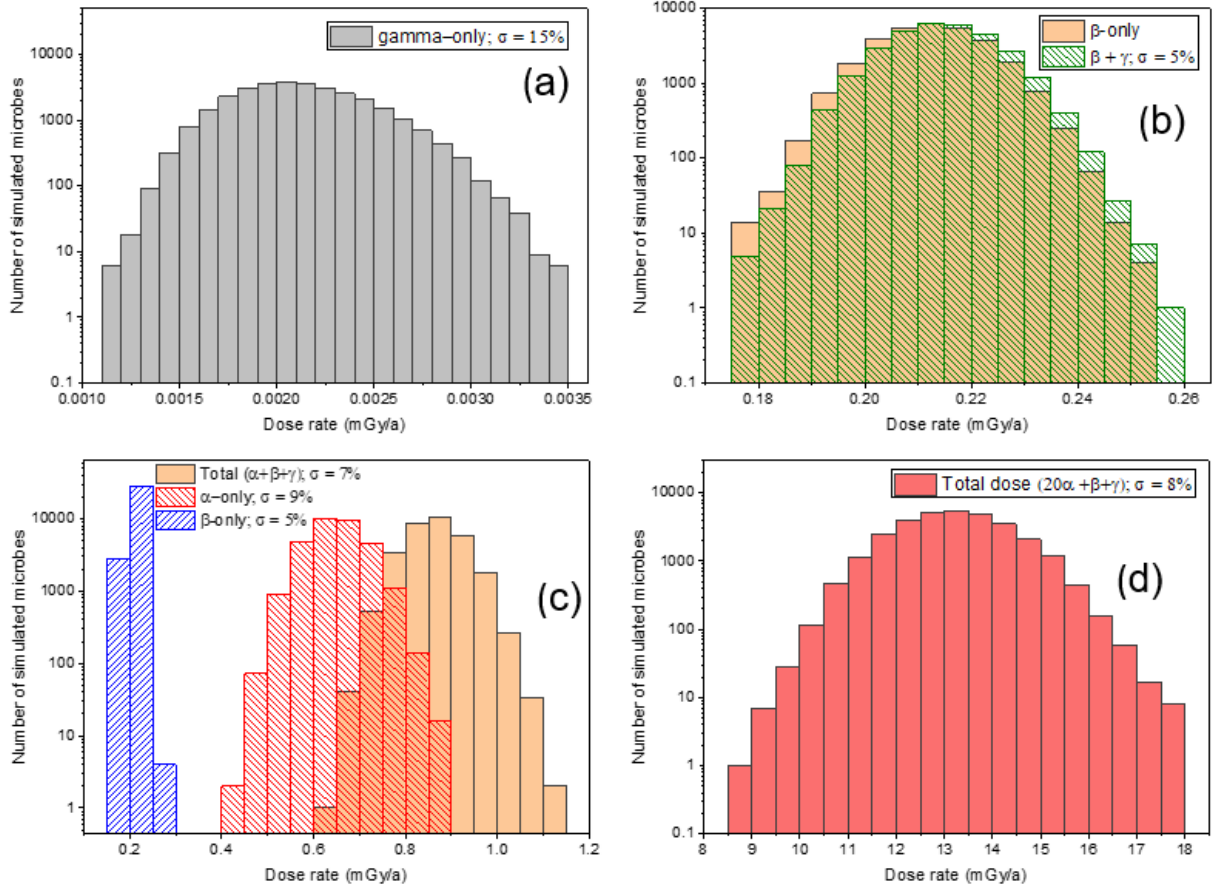


Figure F.2: Microscopic dose rate distribution of microbes in the silica-rich Gusev crater-type Martian subsurface permafrost. (a) Gamma dose rate distribution only. (b) Beta only and Beta + Gamma dose rate distributions. (c) Alpha only, beta only and total sum of alpha + beta + gamma dose rate distributions. Here the dose rate values were added directly. The total dose rate distribution in (d) was obtained by multiplying the alpha radiation portion of the dose distribution by 20 to account for the alpha quality factor.

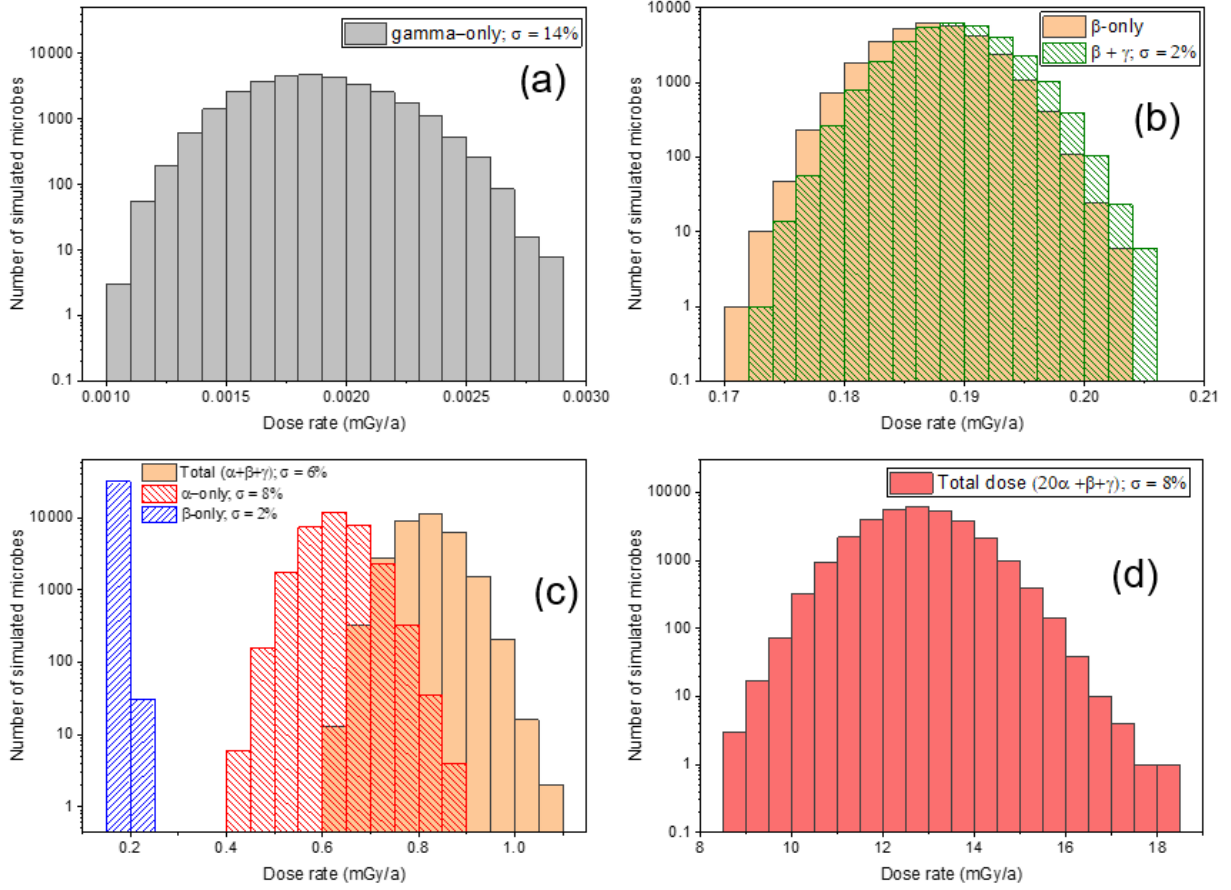


Figure F.3: Microscopic dose rate distribution of microbes in the Rocknest Gale crater-type Martian subsurface permafrost. (a) Gamma dose rate distribution only. (b) Beta only and Beta + Gamma dose rate distributions. (c) Alpha only, beta only and total sum of alpha + beta + gamma dose rate distributions. Here the dose rate values were added directly. The total dose rate distribution in (d) was obtained by multiplying the alpha radiation portion of the dose distribution by 20 to account for the alpha quality factor.

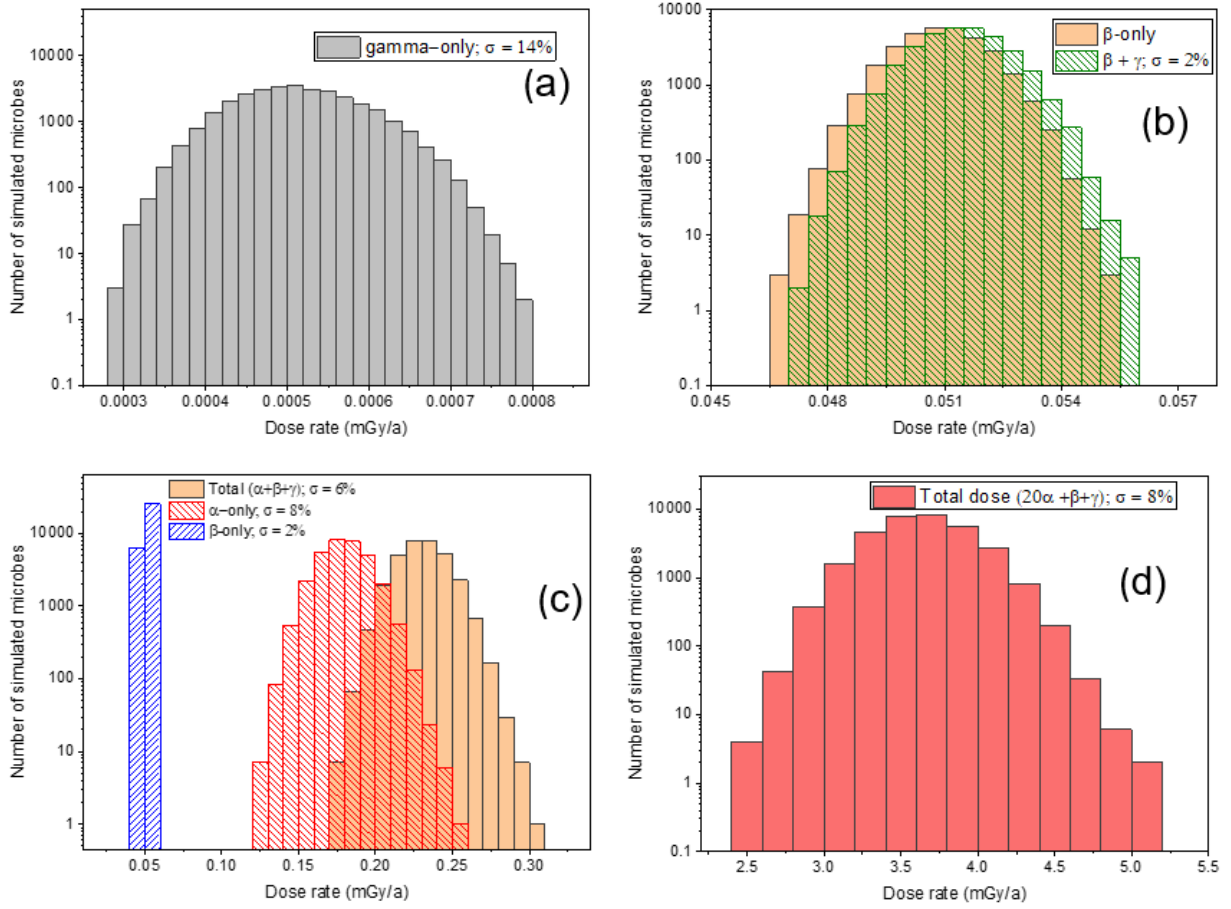


Figure F.4: Microscopic dose rate distribution of microbes in the North polar region on Mars due to Martian dust. (a) Gamma dose rate distribution only. (b) Beta only and Beta + Gamma dose rate distributions. (c) Alpha only, beta only and total sum of alpha + beta + gamma dose rate distributions. Here the dose rate values were added directly. The total dose rate distribution in (d) was obtained by multiplying the alpha radiation portion of the dose distribution by 20 to account for the alpha quality factor.

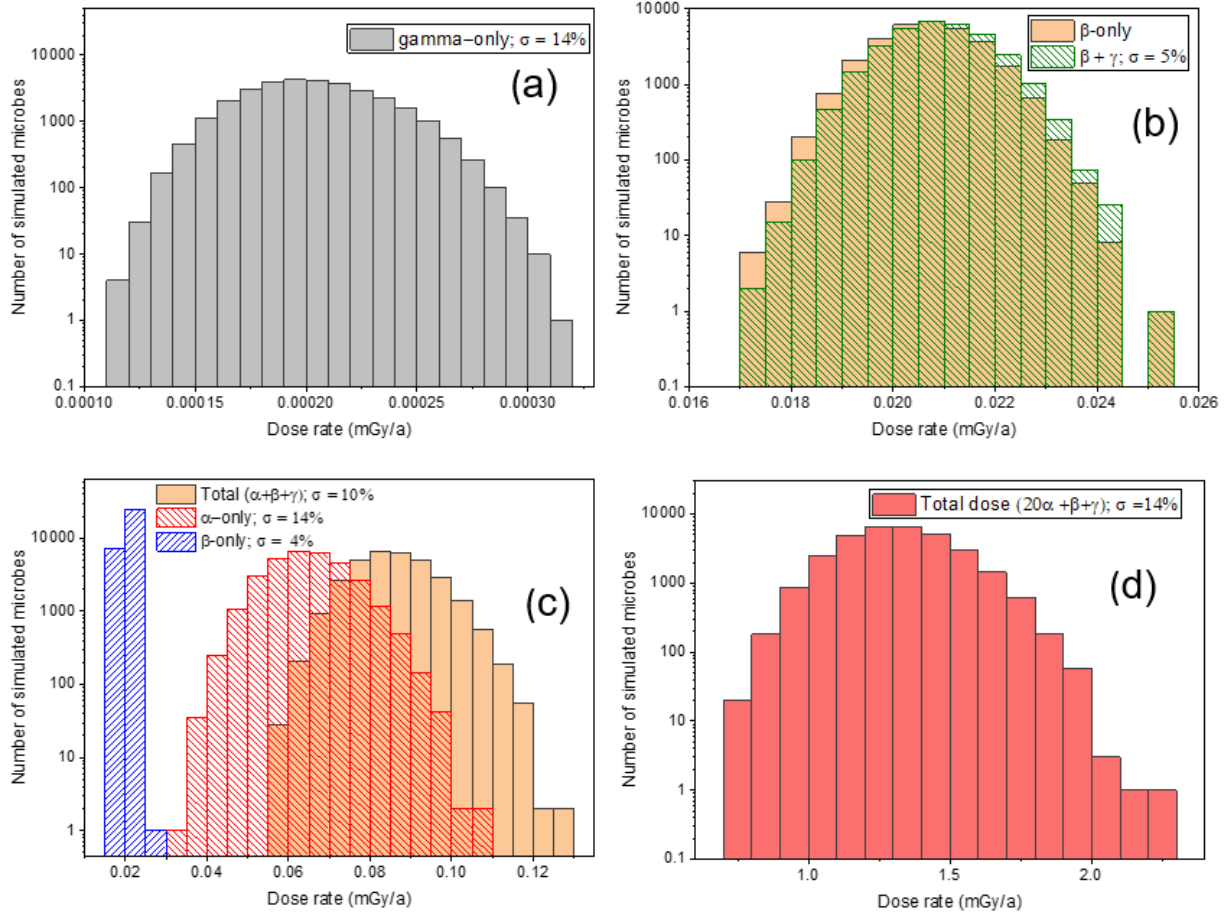


Figure F.5: Microscopic dose rate distribution of microbes calculated using the bulk composition of Mars. (a) Gamma dose rate distribution only. (b) Beta only and Beta + Gamma dose rate distributions. (c) Alpha only, beta only and total sum of alpha + beta + gamma dose rate distributions. Here the dose rate values were added directly. The total dose rate distribution in (d) was obtained by multiplying the alpha radiation portion of the dose distribution by 20 to account for the alpha quality factor.

Remaining survival probabilities results

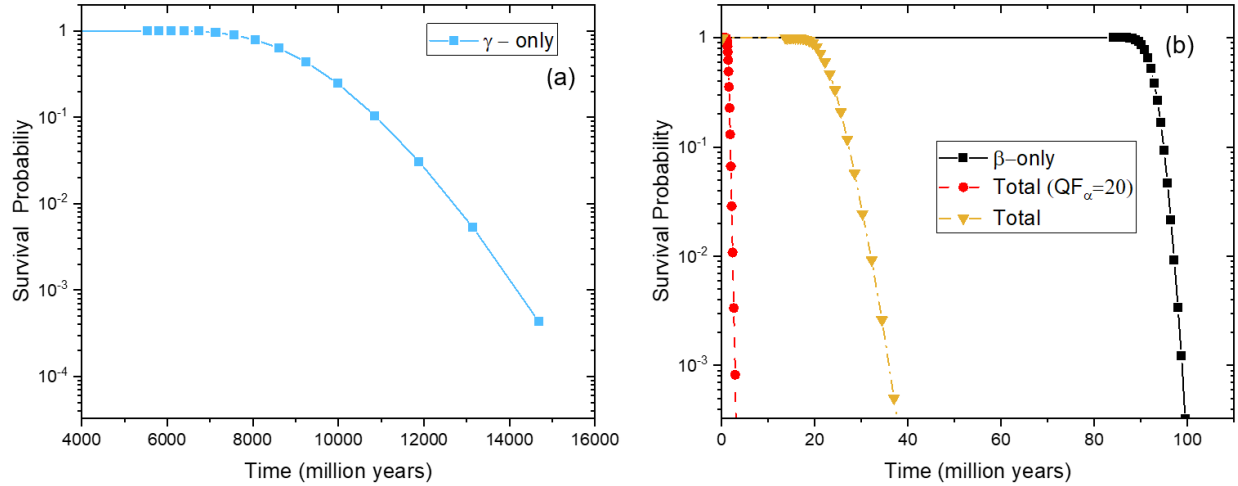


Figure F.6: Calculated Survival probability of dry *B. subtilis* spores in the subsurface permafrost regions of Mars with balsatic andesite rock/regolith composition as a function of time in years. The data points were calculated from the histogram and the lines are plotted to guide the eye.

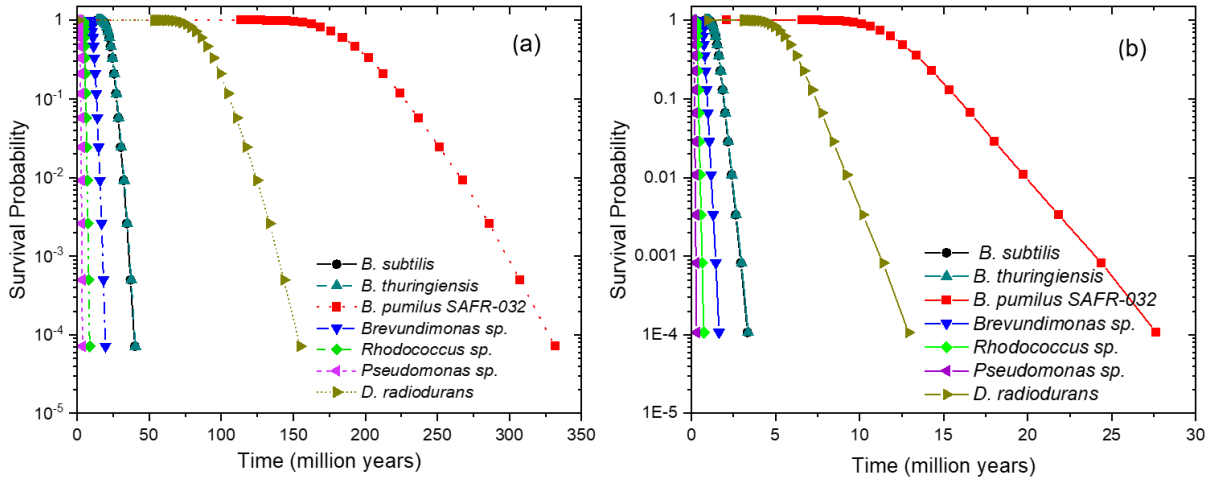


Figure F.7: Calculated Survival probability of different microbes in the subsurface permafrost region of Mars with balsatic andesite rock/regolith composition as a function of time in years. The survival probability curves are based on (a) The total dose rate uncorrected for alpha quality factor and (b) the total dose rate corrected for alpha quality factor by multiplying the alpha part of the distribution by 20. The data points were calculated from the histogram and the lines are plotted to guide the eye.

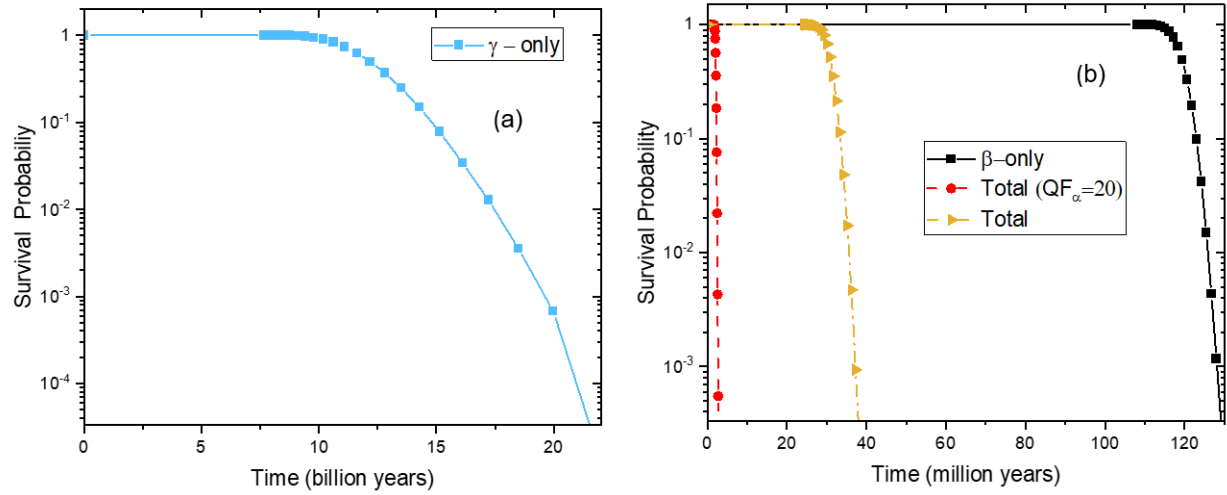


Figure F.8: Calculated Survival probability of dry *B. subtilis* spores in the subsurface permafrost regions of Mars with sulphur-rich Gusev crater rock/regolith composition as a function of time in years. The data points were calculated from the histogram and the lines are plotted to guide the eye.

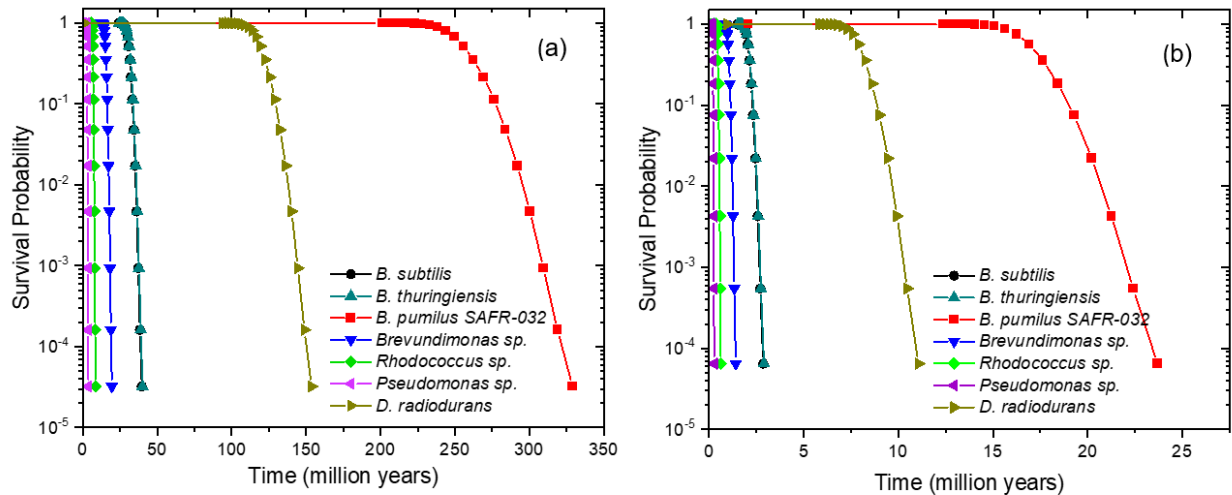


Figure F.9: Calculated Survival probability of different microbes in the subsurface permafrost region of Mars with the sulphur-rich Gusev crater rock/regolith composition as a function of time in years. The survival probability curves are based on (a) The total dose rate uncorrected for alpha quality factor and (b) the total dose rate corrected for alpha quality factor by multiplying the alpha part of the distribution by 20. The data points were calculated from the histogram and the lines are plotted to guide the eye.

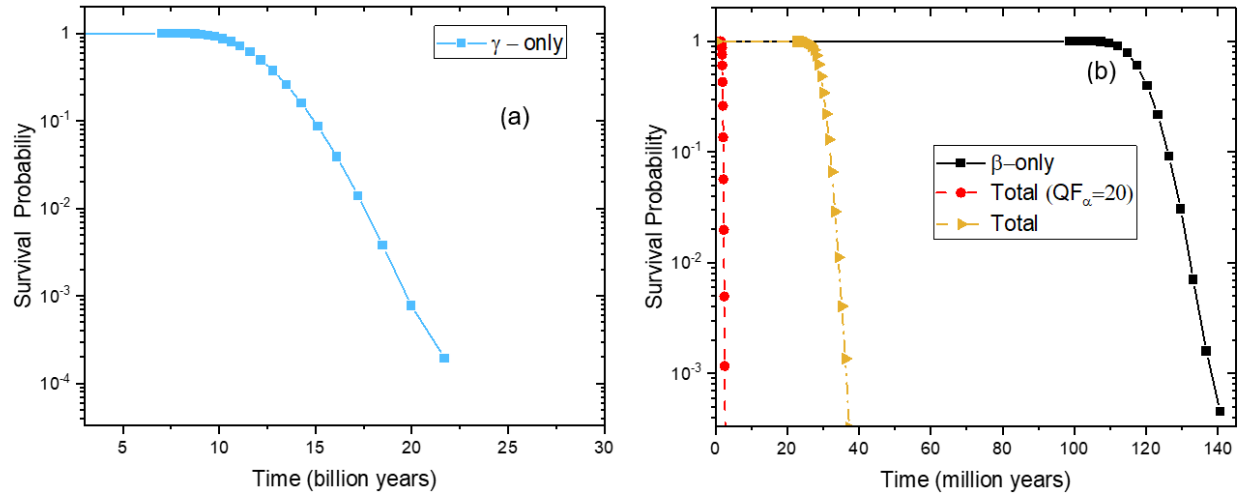


Figure F.10: Calculated Survival probability of dry *B. subtilis* spores in the subsurface permafrost regions of Mars with silica-rich Gusev crater rock/regolith composition as a function of time in years. The data points were calculated from the histogram and the lines are plotted to guide the eye.

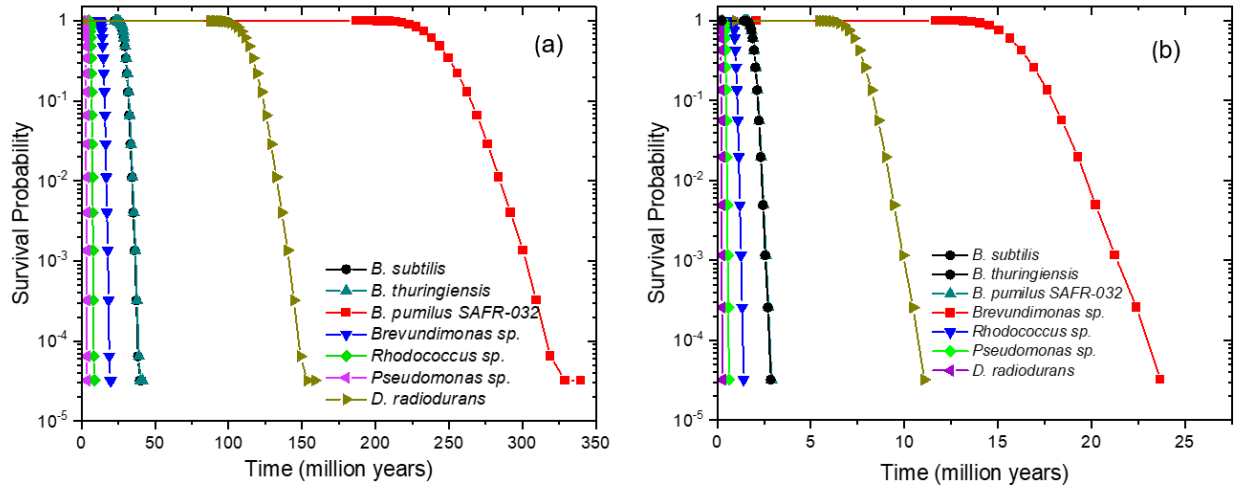


Figure F.11: Calculated Survival probability of different microbes in the subsurface permafrost region of Mars with the silica-rich Gusev crater rock/regolith composition as a function of time in years. The survival probability curves are based on (a) The total dose rate uncorrected for alpha quality factor and (b) the total dose rate corrected for alpha quality factor by multiplying the alpha part of the distribution by 20. The data points were calculated from the histogram and the lines are plotted to guide the eye.

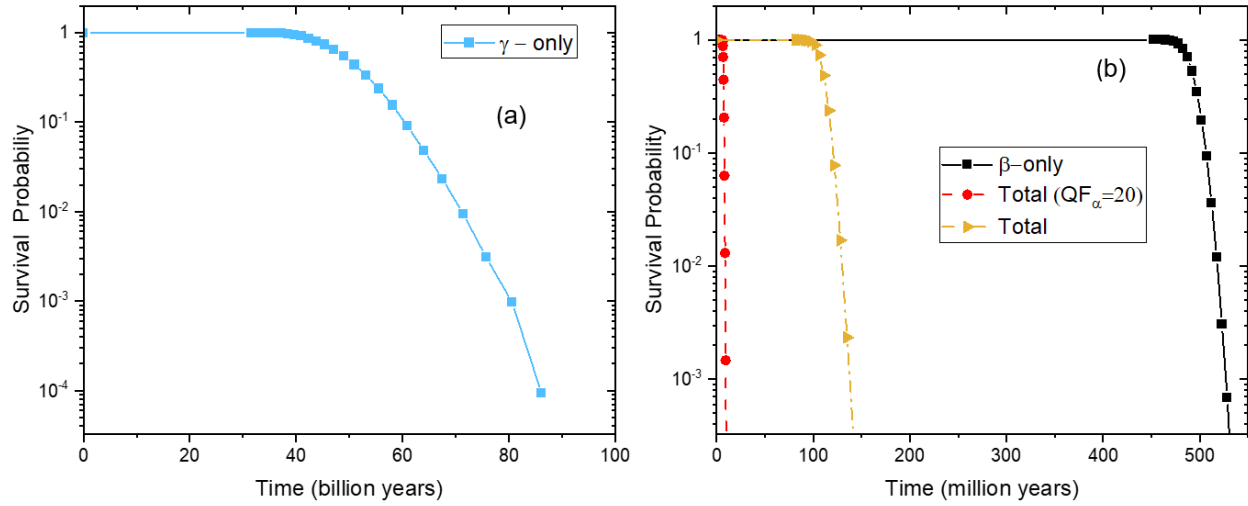


Figure F.12: Calculated Survival probability of dry *B. subtilis* spores in the North polar region of Mars due to Martian dust with Gale crater regolith composition as a function of time in years. The data points were calculated from the histogram and the lines are plotted to guide the eye.

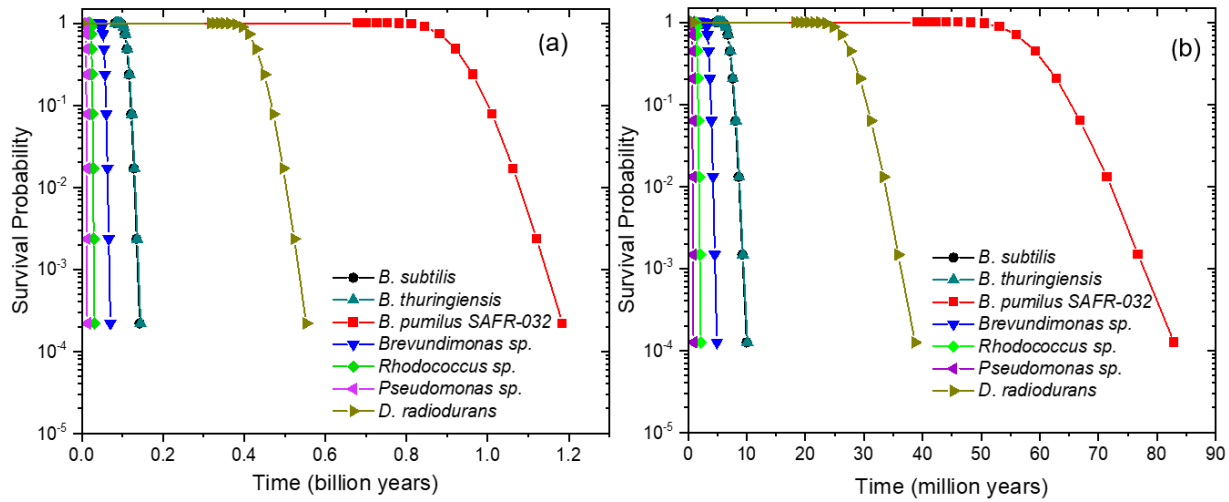


Figure F.13: Calculated Survival probability of different microbes in the North polar region of Mars due to Martian dust with Gale crater regolith composition as a function of time in years. The survival probability curves are based on (a) The total dose rate uncorrected for alpha quality factor and (b) the total dose rate corrected for alpha quality factor by multiplying the alpha part of the distribution by 20. The data points were calculated from the histogram and the lines are plotted to guide the eye.

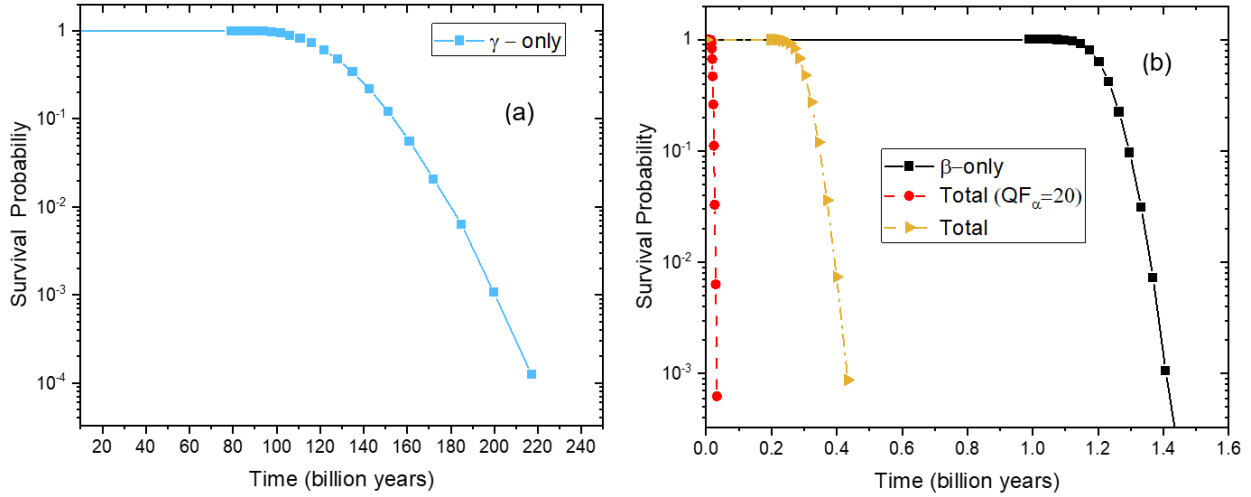


Figure F.14: Calculated Survival probability of dry *B. subtilis* spores based on the bulk composition of Mars as a function of time in years. The data points were calculated from the histogram and the lines are plotted to guide the eye.

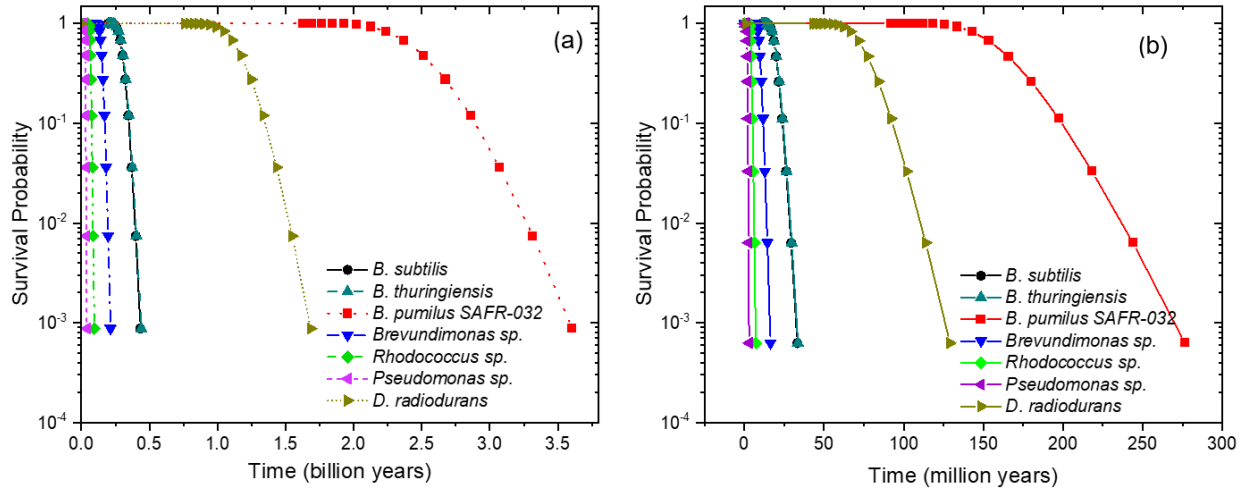


Figure F.15: Calculated Survival probability of different microbes based on the bulk composition of Mars as a function of time in years. The survival probability curves are based on (a) The total dose rate uncorrected for alpha quality factor and (b) the total dose rate corrected for alpha quality factor by multiplying the alpha part of the distribution by 20. The data points were calculated from the histogram and the lines are plotted to guide the eye.

