DETERMINATION OF BREATHING RESISTANCE ACROSS ACTIVATED CARBON FIBER RESPIRATOR CARTRIDGES USING SINUSOIDAL AIRFLOW

by

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Activated carbon fibers are considered good alternative adsorbents in respirators because of their high adsorption capacities, light weight, and fabric form. In order to further assess such application, the pressure drop across Activated carbon fibers in respirators must be adequately low to allow comfortable breathing of the wearer. This study investigated the pressure drop, PD across Activated carbon fibers in respirator cartridges using realistic breathing patterns. Two forms of Activated carbon fibers, cloth, ACC and felt, ACN were tested at three different surface areas: 1000, 1500 and 2000 g/m^2. Each Activated carbon fiber was placed in respirator cartridge at three different layers and subjected to two patterns of breathing: 20 litres per minute by 20 breaths per minute, and 68 litres per minute by 20 breaths per minute from a dynamic breathing machine supplied with purified air at 23°C and 50% relative humidity. PD determination was carried out in a customized Teflon testing chamber. Woven ACC was composed of closely knitted fibers while the unwoven ACN was composed of gradually distributed fibers. The individual effect of Activated carbon fibers forms, layers, breathing patterns, and surface areas was significant on inhalation and exhalation PD. Inhalation and exhalation PD were not significantly different, *P*=0.446 and *P*=0.736, respectively between the two forms. Increasing airflow in a single respirator cartridge to 68 LPM significantly increased inhalation PD, *P*<0.001

and exhalation PD, $P<0.001$ compared to PDs measurements obtained from 20 x 20 breathing pattern. Differences in inhalation and exhalation PD of the surface areas were not significant, $P=0.647$ and $P=0.665$. Increasing the Activated carbon fibers layers significantly increased both inhalation PD, $P=0.016$ and exhalation PD, $P<0.001$. This observation suggest an optimum layer for Activated carbon fibers’ use in respiratory protection also, respirator certification testing using 68 litres per minute sinusoidal airflow may simulate strenous activities in work place better. It is concluded that Activated carbon fibers’ forms, number of layers, breathing pattern and surface area are all important individual factors in designing a breathable respirator cartridge.

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A Thesis

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DEDICATION

I dedicate this thesis to Al-Rahman for his countless blessings and bountiful mercy on my life and that of my family. To my parents for their strong support also, to Ayuub and Azeemah for the hectic trips, lonely days and nights.

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LIST OF SYMBOL/ABBREVIATIONS

ACF: Activated Carbon Fiber

ACN: Activated Carbon Fiber Felt

ACC: Activated Carbon Fiber Cloth

ACF Forms: The two forms of ACF, ACN and ACC.

ACF Types: ACF forms designated by their surface area.

GAC: Granular Activated Carbon

PD: Pressure drop in millimeter H_2O across ACF cartridges

LPM: Air flow in liters per minute.

BPM: Breathing frequency in breaths per minute.

Surface Area: Specific total surface area of ACF in square meters per gram; related to the porosity of the ACF material, which increases as the degree of ACF activation increases

SEM Images: Scanning Electron Microscope Images of ACFs

I. CHAPTER 1: INTRODUCTION

All over the world, respirators are used to protect workers from hazardous air pollutants that they are exposed to in their workplaces. In the United States (U.S.), the National Institute for Occupational Safety and Health (NIOSH) is the federal agency responsible for certifying respirators (Oestenstad, Elliott, & Beasley, 2007), while the Occupational Safety and Health Administration (OSHA) sets the Permissible Exposure Limits (PELs) for certain hazardous airborne pollutants in the workplace.

OSHA also regulates the respiratory protection program through 29 CFR 1910.134 that lists the elements of a workplace respiratory protection program: adequate respiratory protection, worker training, fit testing, medical evaluation, respirator maintenance, proper respirator use and respiratory protection program assessment (Jannsen, 2001). In spite of regulations, a significant number of workers do not comply with the respirator use requirements. This has been attributed to a number of factors, such as bulkiness of respirators, and discomfort and difficulty in breathing during respirator use (Fukakusa et al., 2011; Salazar et al., 2001). These setbacks have necessitated the search for more convenient respirator designs that fulfill regulatory expectations.

Granular activated carbon (GAC) is currently the standard adsorbent for vapors and gases in respirators. However, because of its containment need and heaviness, alternative adsorbents like activated carbon fibers (ACFs) are being sought (Balanay et al., 2011). Activated carbon fibers have not been used in commercially available respirators for adsorbing gaseous pollutants at significant concentrations, but they have been used extensively in the adsorption of pollutants in various applications like carbon dioxide (CO_2) capture, environmental cleaning, and waste water treatment, and their high adsorption capacity has been reported (Balanay et al., 2011; Tsai et al., 2008).

The goal of this study is to aid in the design of efficient, lighter, and more comfortable respirator cartridges composed of activated carbon fibers that will improve the ability of workers to breathe during strenuous work tasks under a variety of environmental conditions and offer adequate protection against volatile organic compounds, such as toluene and benzene. Knowledge from previous studies on the adsorption capacity and other important advantages of activated carbon fibers suggests that designing respirator cartridges and filters made of different forms of ACF adsorbents (i.e. cloth and felt) will enable us to achieve this goal (Balanay et al., 2011).

II. LITERATURE REVIEW

Respiratory Protection against Airborne Pollutants

Respiratory protective devices (RPDs) are used to protect workers and other members of the public from hazardous substances including toxic gases, vapors and particulates. In the workplace, RPDs are the alternative protection provided for workers when engineering control strategies are unavailable or inadequate to protect against respiratory hazards (Fukakusa et al., 2011). Unprotected exposure to respiratory hazards in the work place have been attributed to respiratory diseases including asthma, bronchitis, lung cancer, and chronic obstructive pulmonary disease (Beckett, 2000; Fukakusa et al., 2011).

Besides worker protection, RPDs are also used to maintain worker productivity (Harber et al., 2011). According to Crump (2007), the protective cability of a respirator depends on the type of respirator, workplace environment, and the characteristics of the pollutant of interest.

Along with OSHA's respiratory protection program requirements, NIOSH recommends respiratory protection devices, although many workplaces do not comply with these recommendations (Bureau of Labor Statistics, 2001). The recommendations by NIOSH include having a written program for respirator use, cartridge change out schedule, conducting air sampling to determine the type of respirators appropriate in the workplace, conducting respirator use training, and having a trained respirator program administrator (NIOSH, 2005b). In a study by Greskevitch et al. (2007), a survey of respirator use in crop production facilities showed that 73% of facilities did not have a written respiratory protection program, 21% were not aware of sampling for air pollutants, and 29.5% did not conduct respirator training for their workers.

Factors Affecting Worker Compliance on Respirator Use

For respirators to provide adequate protection of workers against airborne pollutants, they must be used consistently, usually for long hours, and in compliance with respirator use guidelines. Unfortunately, this is not the case in many workplace settings and this is related to non-compliance among workers. A study by Fukakusa et al. (2011) showed that compliance with RPD use at work among workers suffering from respiratory illness was positively associated with convenient location, safety training, fit testing, and age of the patient, while RPD use was negatively associated with shortness of breath and nasal stuffiness. The same study indicated that respirator use was also associated with worker awareness of hazards. Another study (Doney et al. 2007) indicated that metal workers working with paint vapors, solvents, welding fumes, and silica dust were more likely to comply with respirator use because of the well-known hazards associated with their workplaces. Worker concern about work exposure to hazards and adequate fit testing and training also have a positive influence on respirator use; conversely, discomfort, visual obstruction, fatigue, poor communication and faulty design of work environment were negatively associated with respirator use (Salazar et al., 2001). In order to improve respirator use, it is important to address the factors responsible for impeding respirator use compliance, including discomfort and lack of awareness about respiratory hazard exposure (Jahangiri, Motovagheh & Khavvaj, 2009).

The type of respirator has been shown to influence worker compliance with its use. Two types of respirators used in the workplace are the elastomeric half-face mask (HFM) dual-cartridge respirator and the N95 filtering respirator. The HFM is a tight-fitting air-purifying type of respirator for particulates, gaseous pollutants, or both (Chandler, 1998). On the other hand, the N95 respirator is a non-oil resistant filtering face piece respirator that is capable of filtering

95% particles that have diameter greater than or equal to 0.3 microns (NIOSH, 2005b). According to Harber et al. (2011), there was no significant difference between HFM and N95 respirator use on the impact on work task performance among tested workers. However, the use of HFM increases anxiety among workers, while the use of lighter N95 filtering face piece had no observed effect (Wu et al., 2011). This implies that lighter weight respirators protecting against gases and vapors, similar to N95, may increase RPD use compliance and consequently improve worker protection against airborne hazards.

Volatile Organic Compound Adsorption in Respirators

Volatile organic compounds (VOCs) are important group of pollutants frequently encountered in the workplace. VOCs are important to human health because of their potential toxicity, including mutagenicity and carcinogenicity (USEPA, 1990; Kostianen, 1995). Commonly encountered VOCs include toluene, benzene, and tetrachloroethylene (Geeta & Rao, 2009). A common method of removing VOCs in respirators is by adsorption unto granular activated carbon (GAC). Although GAC is currently used as the standard respirator adsorbent for VOCs, its granules require containment, along with other disadvantages, necessitating research into more efficient alternatives like ACFs (Balanay et al., 2011). As mentioned previously, activated carbon fibers (ACFs) are more effective in adsorbing hazardous gases and vapors (including VOCs) but their use in air purifying respirators is understudied.

ACFs are expected to offer an affordable and versatile option for the capture of VOCs in respirators, as demonstrated by successful use in the adsorption of VOCs in other applications such as wastewater treatment (Wang, Feng, & Yu, 2007) and air pollution control (Das et al., 2004; Huang et al., 2003; Lorimier et al., 2005). “For any given VOC, ACFs’ adsorptive capacity is represented by an adsorption isotherm, of the amount of adsorbed VOC to the equilibrum pressure at constant temperature" (Geeta & Rao, 2009).

Differences between Granular Activated Carbon and Activated Carbon Fiber as Adsorbents

Activated carbon fibers (ACFs) are manufactured from polymeric fibers, with diameters between 10 and 20 µm, that are prepared from novoloid, polyacrylonitrile (PAN), rayon precursors, and pitch, which are carbonized and activated (Lo, 2002). The uniform activation of these small-diameter polymeric fibers results in ACFs of narrow pore size distribution (Feng et al., 2005). ACFs are newer forms of porous carbon materials that have more merits than the conventional granular carbon (Nabais et al., 2006). Unlike granular activated carbon (GAC), ACF forms (i.e. woven cloth and unwoven felt) are easier to use and handle. Likewise, they are characterized by higher adsorption capacity, larger surface area, and higher adsorption rate from gas or liquid phase compared to GAC (Nabais et al., 2006).

From assessments of the differences in critical bed depths and adsorption capacity for toluene, GAC was demonstrated to have a lower adsorption capacity compared to ACF cloth (ACFC) and ACF felt (ACFF) with similar surface area (Balanay et al., 2011). Although the same study showed that GAC had a much higher (275%) critical bed depth than average ACFC it has a lower critical bed depth compared to ACFF. Additionally, ACFC had the higher surface area and the highest adsorption capacity while also having the lowest critical bed depth (Balanay et al., 2011). Moreover, ACF had higher adsorption rate than GAC when challenged with chloroform, acetone, and acetonitrile (Tsai et al., 2008).

Adsorption Capacity of Activated Carbon Fibers

Studies have demonstrated the adsorption capacity of ACFs, and their uses as an alternative adsorbent in respirators have been suggested. ACFC was found to be a good adsorbent for metals in the gaseous state (Liu, 2007). Similarly, ACFs are good adsorbents for toluene, with ACFC of 2000 m^2/g surface area having the lowest critical bed depth and highest adsorption capacity for toluene (Balanay et al., 2011).

A study by Tsai et al. (2008) compared the adsorption capacity of ACF, commercial activated carbon and sludge-derived adsorbent for VOCs (e.g. chloroform, acetone, and acetonitrile) and showed that higher surface area and smaller fiber diameter in ACFs resulted in higher adsorption capacity compared to the other adsorbents tested. The small pore size in ACFs also gave it a smaller diffusion coefficient for volatile organic pollutants in the range 10^{-8} to 10^{-7} cm^2 s^{-1} compared to that on commercial activated carbon and sludge-derived adsorbents (Tsai et al., 2008). Moreover, Figueiredo et al. (2011) showed that adsorption capacity of ACFs increased with increasing ACFs micropore volumes, and the ACFs tested were found to have high adsorption capacity for phenol, particularly CO_2 activated carbon fibers.

Pressure Drop across Respirators

In air filtration, resistance refers to the static pressure drop across the filter at a given face velocity (reference). When applied to respiratory protection, pressure drop is also referred to as breathing resistance and is the difference in static pressure between two pressure points located before and after the air cleaning media of the respirator (i.e. filter or adsorbent materials)(reference). The pressure drop test is an important part of certification tests required in the US for respirators, as it is translated as the breathability of a respirator. According to NIOSH respirator certification requirements, the maximum initial inhalation resistance across chemical-cartridge respirators for gases, vapors, or gases and vapors is 40 millimeter of water (mm H_2O), while the maximum exhalation resistance is 20 mm H_2O. Certification testing is done on complete respirators assembled onto a headform or test fixture, using a constant airflow of 85 liters per minute (LPM) (NIOSH, 2005a). Inhalation and exhalation pressure drop tests can be carried out under fixed continuous airflow or sinusoidal airflow simulating normal breathing pattern in humans (Kaufman & Hastings, 2005). Adsorption of toxic vapors and gases on ACF are primarily studied using fixed airflow rates (Nir, Suzin, & Kaplan, 2002).

Despite meeting certification requirements, previous studies have shown that many respirators make it difficult for workers to breathe normally during strenuous working conditions (Torbjorn, 2002). Respirator pressure drops can be attributed to airborne contaminant leakage, exposure, sorbent loading, and workers performance (Janssen & Weber, 2005 & 2006; Cho & Yoon, 2012; Caretti et al., 2006). Inhalation and exhalation respirator resistance have been shown to affect workers' performance, and inhalation resistance provides a good estimate of airflow in respirators during hard work (Caretti et al., 2006). Studies have shown that increasing pressure drop does not increase faceseal leakage or contaminant exposure in respirators, provided that there is an adequate respiratory protection program, including fit testing, in the workplace (Janssen & Weber, 2005 & 2006). A recent study of the acceptable breathing resistance in air purifying respirators found that during long term work lasting up to 1 hour, work of breathing per tidal volume (breathing resistance) of below 0.9 Kpa was within tolerance level, while 80% of the tested population tolerated breathing resistance of 1.3 Kpa between 10 to 15 minutes of exercise at respiratory minute ventilation of 110 liters per minute (Shykoff & Warkander, 2011). Another recent study on the impact of N95 respirators on breathing resistance showed an average increase of 126% inspiratory and 122% expiratory flow resistances in human subjects during N95 respirator use (Lee & Wang, 2011).

In contrast, increasing welding fume loads on respirator filters in laboratory and the workplace resulted in increased pressure drop across the respirator filters of particulate respirators (Cho & Yoon, 2012). Visual study and chemical analysis of welding fumes on the respirator filters also showed that fumes were trapped mainly in the first and second layers of the filter, and no fume was present in the fourth layer of the respirator filter (Cho & Yoon, 2012).

Despite the importance of pressure drop in ensuring breathability of respirator during its usage, few studies have focused on this aspect of respirator design. Research on the variation of pressure drop in air purifying respirators is limited, while research on pressure drop in respirators using activated carbon as adsorbent is not available at present. Although ACFs have shown potential as excellent adsorbents in respirators based on the critical bed depth and adsorption capacity, pressure drop across these ACFs in respirators may be a concern. The ACFC types in particular are much denser than the ACFF types because of their tightly woven fibers that may restrict airflow (Balanay, 2014).

Breathing Patterns among Workers

It is widely believed that the main reason behind workers' non-compliance with respirator use is due to difficulty in breathing while using respirators during work tasks (Morgan, 1983). According to Qiu & Wang, (2012), an individual at rest has airflow of 6 to 7 liters per minute (LPM) which is increased to 70 to 120 LPM during exercise. During maximum exercise, minute ventilation is increased to 150 LPM and breathing frequency is increased to 40 to 50 breaths per minute (Levitzky, 2003). Although expired minute ventilation of 40 LPM is the

NIOSH recommended breathing pattern for sinusoidal airflow, using a minute ventilation of 135 LPM was recommended in testing respirators in order to achieve a close representation of human respiratory pattern during high-intensity physical exercise (Coyne et al., 2006).

A study of the effect of different respirator types on the breathing pattern of healthy male workers showed that firefighters using self-contained breathing apparatus (SCBA) pressure-demand industrial respirators have a significantly lower breathing rate and longer expiratory time than construction workers that used an airline apparatuses to protect them from air pollutants (Louhevaara et al., 1986). However, the observed difference in breathing patterns was not related to the respiratory protection used, but to the workload (Louhevaara et al., 1986). The aforementioned study suggests that workload may affect breathing ability during respirator use. However, firefighter training includes instructions for slow and deep breathing; hence this may have contributed to differences in their observed breathing rate compared to construction workers.

Kaufman & Hastings (2005) showed that U.S. Marine Corps from the Chemical Biological Incident Response Force (CBIRF) may encounter high respiratory flow rate during their chemical protection activities, causing increased contaminant load and recovery resistance in respirators. It was discovered that asymmetric double sigmoidal airflow showed a closer representation to respiratory measurements than sinusoidal airflow models (Kaufman & Hastings, 2005). According to Harber & SooHoo (1984), breathing resistance is a constraint to the use of air-purifying respirators, resulting in modification of breathing pattern among users. It is unusual for voluntary breathing modification to occur, a process that is expected to reduce breathing frequency when work rate is increased (Yasukouchi & Serita, 1990).

Comparing two types of SCBA respirators with varying breathing resistance showed that, for the respirator with higher breathing resistance, worker tolerance and response time were significantly shorter than for workers wearing a respirator with lower breathing resistance (Qiu & Wang, 2012). Tolerance time is duration of work when work rate is increased; it measures tolerance capacity (Qiu & Wang, 2012). This shows that increasing breathing resistance in respirators will reduce the length of time workers are able to work and therefore affects their productivity. On the other hand, minute respiration and breathing frequency increased with increasing respirator breathing resistance (Qiu & Wang, 2012). This further emphasizes the need to design respirators with reduced breathing resistance that will increase worker comfort and productivity.

Employing constant airflow rates in studies of the adsorption capacity of an activated carbon respiratory protective device is technically simpler but often associated with an exaggerated respirator service life (Tanaka et al., 1996a; Tanaka et al., 1996b; Suzin, Nir & Kaplan, 2000). However, using sinusoidal airflow simulates human breathing and gives a more realistic measurement of the service life of respirators. A comparison of breakthrough time of activated carbon canisters when challenged with dimethyl-methyl phosphate (DMMP) during constant airflow and sinusoidal flow showed that the use of sinusoidal breathing simulation air flow resulted in 4% and 6% shorter breakthrough time compared to canisters supplied with steady air flow at breathing frequencies of 30 and 40 LPM, respectively (Suzin, Nir & Kaplan, 2000; Nir, Suzin, & Kaplan, 2002). This indicates that sinusoidal airflow gives a more accurate estimation of respirators' service life when compared with fixed continuous airflow.

III. HYPOTHESIS AND STUDY OBJECTIVES

The main hypothesis of this study is that the pressure drop across the ACF cartridge is dependent on the physical form (cloth vs felt), specific surface area (1,000 vs 1,500 vs 2,000 m^2/g), the layer thickness of the ACF materials, and the breathing pattern across the ACF materials. The following are the specific measurable hypothesis:

Hypothesis 1: The pressure drop across ACF cartridge is significantly different between the ACF cloth and ACF felt.

Hypothesis 2: The pressure drop across ACF cartridge is significantly different between the 20 x 20 and 68 x 20 breathing patterns.

Hypothesis 3: The pressure drop across ACF cartridge is significantly different among specific surface areas of 1000, 1500 and 2000 m^2/g.

Hypothesis 4: The pressure drop across ACF cartridge is significantly different among low, medium and high layer thickness.

The purpose of this research study was to determine the breathing resistance across activated carbon fibers in respirator cartridges using sinusoidal airflow. The specific aims are to:

1) Characterize commercially available ACFs by forms, types and fiber organization
2) Determine the breathing resistance across ACF in respirator cartridges by ACF type, surface area and number of layers using sinusoidal air flows

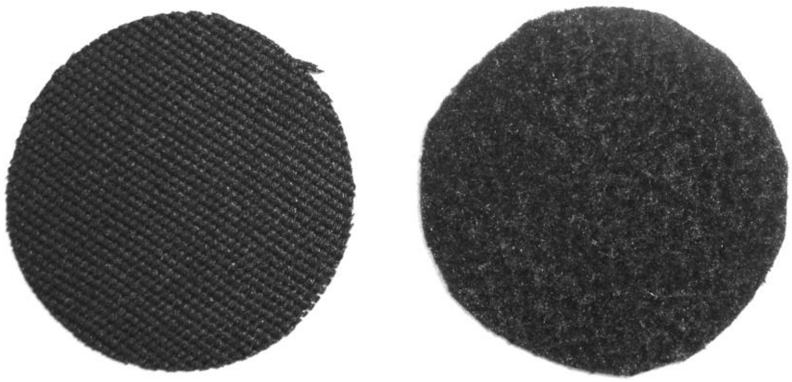
IV. SIGNIFICANCE OF THE STUDY

ACFs have prospective uses as adsorbents in respirator cartridges due to their light weight, adequate containment, and high adsorption capacity. Therefore, they are a suitable adsorbent that can be used to design comfortable and efficient respirators. The pressure resistance of ACFs must be low enough to allow breathing to progress. Therefore, it is important to determine the pressure drop across ACF respirator cartridges to assess suitability in respiratory protection applications. We have conducted preliminary pressure drop tests using either constant or sinusoidal airflows. In sinusoidal models, lung expansion during the first few inhalations gradually increases, giving a better representation of human breathing than in continuous airflow models. The data obtained from this study may be used to optimize respirator cartridge design for workers' protection and comfort in future research studies.

V. METHODOLOGY

Materials

Two forms of ACF were tested as adsorbent materials: unwoven ACF felt (ACN) and woven ACF cloth (ACC) (Figure 1). For each ACF form, three manufacturer-specified surface areas (1000 m^2/g, 1500 m^2/g and 2000 m^2/g) were tested, with a total of six types of ACFs analyzed. ACF types were designated as shown in Table 1 based on their form and surface area. The ACFs were obtained from American Technical Trading, Inc. (Pleasantville, NY) and were manufactured from novoloid, a phenol aldehyde-based fiber. The average thickness of the ACN layers was 0.22 cm, while that for ACC layers was 0.11 cm.



ACF Cloth (ACC) ACF Felt (ACN)

Figure 1. Forms of Activated Carbon Fiber (ACF)

Table 1. Denotations of ACF Types Based on ACF Characteristics			
Form	**Nominal* Surface Area, m^2/g**		
	1000	**1500**	**2000**
Cloth	ACC10	ACC15	ACC20
Felt	ACN10	ACN15	ACN20

*Manufacturer-specified

The ACFs were cut into three inch discs, with a diameter similar to that of a cartridge container, using a customized stainless steel cutter. These ACF discs were treated overnight in a Precision Compact Model 665 oven (Thermo Scientific, Marietta, OH) at 200°C to remove excess moisture and volatile impurities on the adsorbent in order to prevent interference. After oven treatment, the ACF discs were placed in a desiccator for about 10 minutes and then weighed (in grams) with the Voyager Pro analytical balance (Ohaus Corp., Parsippany, NJ), and the thickness (in cm) was measured using a Vernier caliper. The ACF discs were then placed in a cartridge for pressure drop testing.

Pressure Drop Testing

ACF adsorbents of different forms, surface areas and number of layers were placed in respirator cartridges and tested for pressure drop. Table 2 shows the designation of each cartridge type based on ACF forms, types (based on surface area), and number of layers. These cartridges were placed in a customized cylindrical Teflon test chamber and challenged with sinusoidal airflow at a constant temperature of 23^0 C and relative humidity of 50%. Two breathing patterns were used for each cartridge type, with flow rates and breathing frequencies as shown in Table 3. The breathing patterns were be produced using a dynamic breathing machine (Warwick Technology Ltd, U.K.), which used a simple sine wave output to simulate the required breathing patterns.

Table 2. ACF Types and Number of Layers	
ACF Type	**Number of Layers**
ACN10	5
ACN15	5
ACN20	5
ACN10	7
ACN15	7
ACN20	7
ACN10	9
ACN15	9
ACN20	9
ACC10	3
ACC15	3
ACC20	3
ACC10	5
ACC15	5
ACC20	5
ACC10	7
ACC15	7
ACC20	7

Table 3. Breathing Patterns According to Flow Rate and Breathing Frequency		
Breathing Pattern	**Flow Rate (liters per minute, LPM)**	**Breathing Frequency (breath per minute, bpm)**
1	20	20
2	68	20

The flow rate values were based on the NIOSH recommended minute ventilation of 40 LPM for sinusoidal air flow and the minute ventilation of 135 LPM used in the Coyne et al. (2006) study. Cartridges utilized in this study are usually used in pairs in dual cartridge respirators, with each cartridge receiving half of the total airflow. Thus, the flow rate values used in this study were half of the above-mentioned NIOSH-recommended minute ventilation rates because a single respirator cartridge was tested at a time. An air compressor equipped with air filtering units (Parker Hannifin Corp., Haverhill, MA) was used to supply clean, dry air, which was preconditioned at a constant temperature of 23°C and relative humidity of 50% using a Miller-Nelson Model HCS-501-100 instrument (Assay Technology, Livermore, CA). A reagent grade water purification system (Aqua Solutions, Inc., Jasper, GA) was used to supply purified water to the Miller-Nelson unit for relative humidity control. The preconditioned air was passed through an air tank where it was stored briefly. The ACF cartridges were tested for pressure drop in the Teflon test chamber, wherein the temperature and relative humidity were monitored using a HOBO Model U14-002 temperature and relative humidity datalogger (Onset Computer Corp., Pocasset, MA). Pressure drop was measured using a DP-Calc Model 5825 micromanometer (TSI Inc., Shoreview, MN). Pressure drop measurements, in millimeters of water (mmH_2O), across the ACF cartridges were obtained every second for 20 minutes, resulting to a total of 1,200 pressure drop data points per test. Each test was conducted in duplicate.

After collecting all data, the peak pressure drop measurements for both inhalation and exhalation per test were obtained from the pressure drop curves by determining the minimum and maximum pressure drops, respectively. Considering the set-up for the determination of differential pressure, the exhalation pressure drop showed positive values but the inhalation pressure drop showed negative values. Thus, the absolute values of the minimum pressure drop measurements were obtained for the inhalation pressure drop. Peak pressure drop measurements were averaged for duplicate tests. The pressure drop measurements were compared among the different ACF forms, breathing patterns, surface areas and number of layers to determine the factor that will give the lowest breathing resistance. Figure 2 shows the schematic diagram of the experimental set-up for pressure drop testing using sinusoidal air flow. Considering the number of ACF types, layers and breathing patterns tested, a total of 72 pressure drop tests (6 ACF types x 3 layers x 2 breathing patterns x 2 duplicates) were conducted.

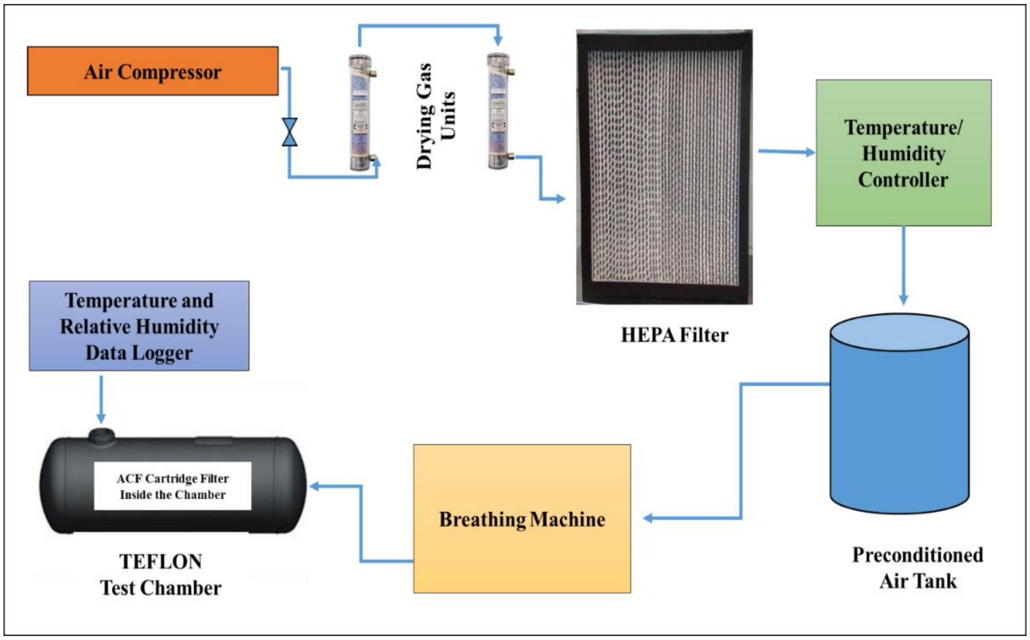


Figure 2. Experimental Setup for Pressure Drop Testing Using Sinusoidal Air Flow

Characterization of Fiber Organization

ACF types were analyzed using a Model FEI Quanta 200 Mark 1 scanning electron microscope (SEM) at the Imaging Core Facility at the Department of Biology, East Carolina University. Images of the ACFs were obtained at three magnifications (50x, 200x and 800x) to illustrate fiber arrangement in ACF samples (Figures 3-5).

Data Analysis

The Statistical Package for the Social Sciences (SPSS version 20, SPSS Institute, Chicago IL) was used to analyze the data. The inhalation and exhalation pressure drop measurements across ACF respirator cartridges by ACF forms, different layers, surface areas and breathing patterns were compared using boxplots as visualizing tool. One-way Analysis of variance (ANOVA) was used to determine significant differences in average peak inhalation and exhalation pressure drop by ACF forms, breathing pattern, surface area and number of layers as predictor variables. The means of inhalation and exhalation PDs obtained using the two breathing patterns, three surface areas and three layer arrangements were compared between the two ACF types using one-way ANOVA. In addition, the primary effects and interaction effects of ACF types, breathing patttern, surface areas and layers on inhalation and exhalation PD were determined using two-way ANOVA. From these, the effects of the study variables on inhalation and exhalation PD may be employed in designing effective and breathable respirator cartridges. Significance was accepted at $P \leq 0.05$ for all analyses.

VI. RESULTS

Characterization of Fiber Organization

The ACF images obtained from the SEM analysis illustrate the fiber arrangement in ACF samples at three magnifications: 50x, 200x and 800x (Figures 3, 4 and 5, respectively). At 50x magnification, the fiber organizations of the ACFs by form were dissimilar: ACC was made of closely woven strands of fibers, while ACN contained non-woven, casually distributed fibers. At 200x magnification, the random distribution of ACN fibers was profound, and a closer image of ACC's definite fiber weaving was obtained. Images at 800x magnification displayed individual fiber strands, with ACCs showing fibers that are closely bunched together and ACNs showing more spaces around each fiber. At all magnifications, ACC samples showed a much denser form compared to the ACN.

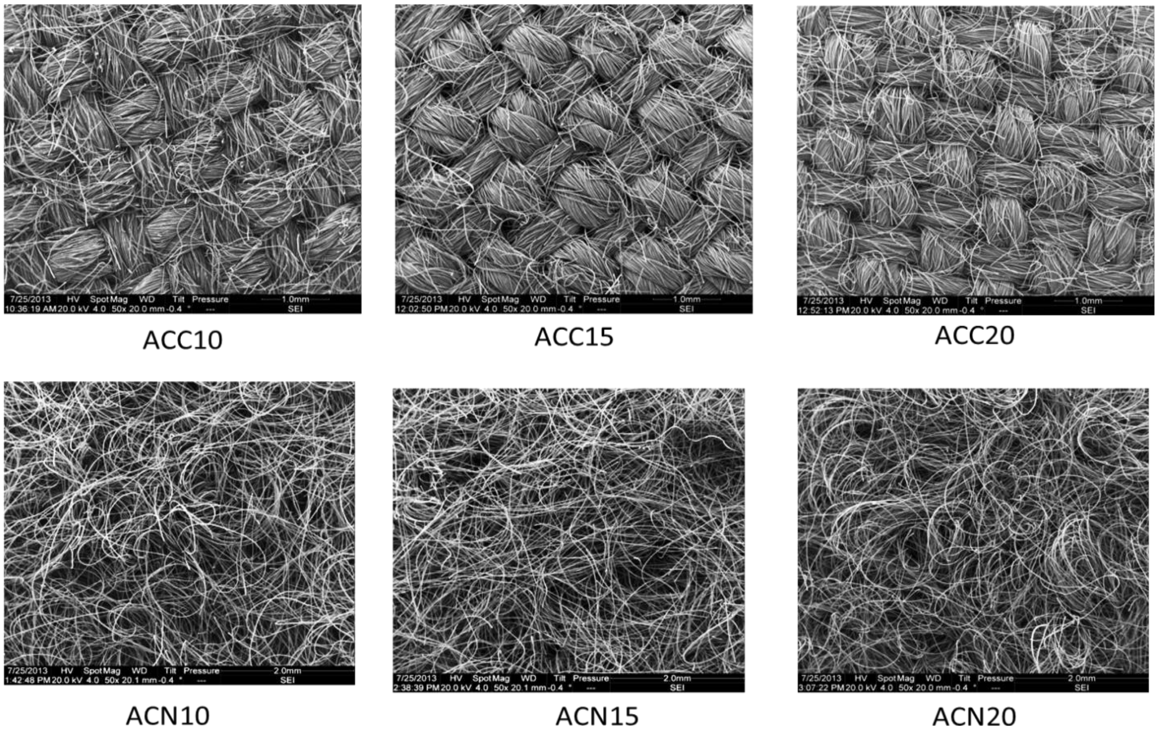


Figure 3. SEM Images of Activated Carbon Fiber Types at 50x Magnification

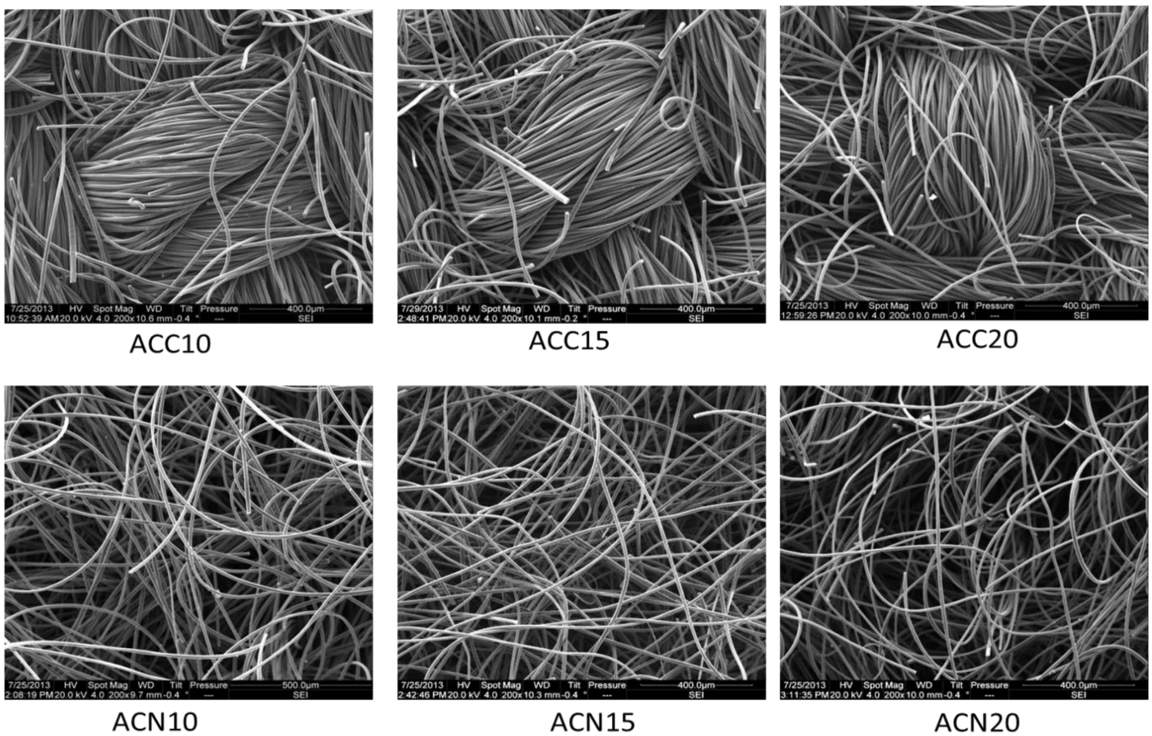


Figure 4. SEM Images of Activated Carbon Fiber Types at 200x Magnification

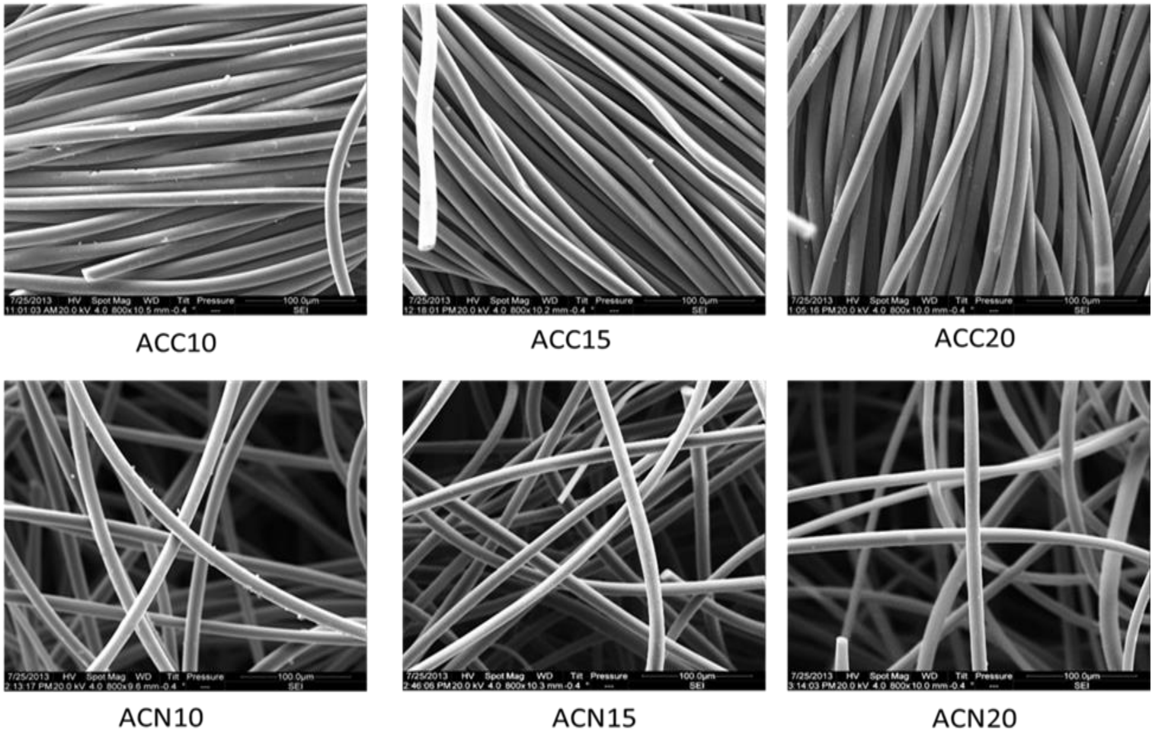


Figure 5. SEM Images of Activated Carbon Fiber Types at 800x Magnification

Pressure Drop (PD) Measurements

Pressure drop (PD) measurements (mmH_2O) across the ACF cartridges were obtained every second for 20 minutes, resulting in a total of 1,200 PD data points per test as shown in pressure drop curves. Figure 6 shows a sample of a pressure drop curve for ACN210 at five layers using 20 x 20 breathing pattern. The peak inhalation and exhalation PD data were derived from these pressure drop curves for all ACF types by ACF form, surface area, and number of layers using the two breathing patterns as shown in Appendix A.

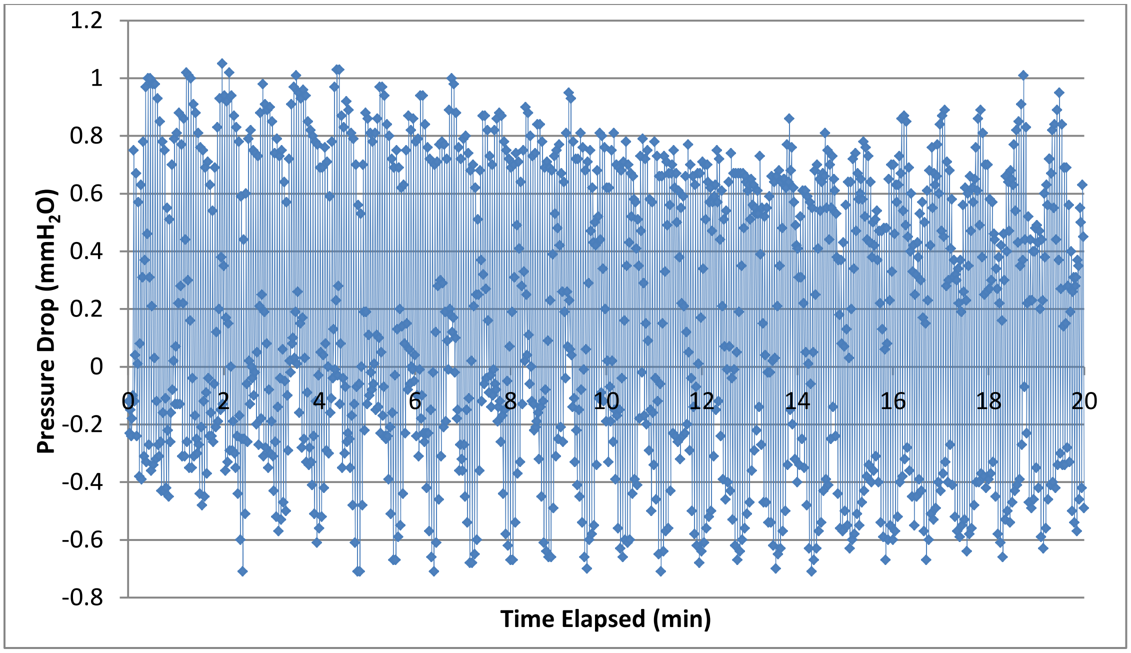


Figure 6. Sample Pressure Drop Curve for ACN10 at five Layers Using 20 x 20 breathing Pattern

Table 4 shows the mean inhalation and exhalation pressure drop values by the different factors investigated in this study, which are the ACF form, specific surface area, layer thickness and breathing pattern. Significant differences (p-values) within the study factors were also shown.

Table 4. Mean inhalation and exhalation pressure drop values and significance by study factors

Factors		Inhalation PD $\bar{x} \pm SD$	*P*	Exhalation PD $\bar{x} \pm SD$	*P*
ACF FORM	ACN	2.315 ± 1.435	0.446	3.262 ± 2.058	0.736
	ACC	2.055 ± 1.439		3.446 ± 2.535	
Breathing	20 x 20	1.016 ± 0.490	< 0.001	1.296 ± 4.505	< 0.001
	68 x 20	3.354 ± 1.054		5.413 ± 1.327	
Surface area	1000	2.267 ± 1.449	0.647	3.434 ± 2.395	0.665
	1500	1.962 ± 1.365		3.020 ± 2.092	
	2000	2.326 ± 1.514		3.609 ± 2.439	
Layers	Low	1.404 ± .9507	0.016	2.447 ± 1.659	< 0.001
	Medium	2.183 ± 1.252		3.294 ± 2.092	
	High	2.968 ± 1.608		4.322 ± 2.699	

Effect of ACF Forms on Inhalation and Exhalation Pressure Drop

Inhalation PD data per ACF form were pooled together, regardless of other variables, to determine the mean PD (N = 36). The mean inhalation PD of ACN was 2.315 ± 1.435 mmH_2O and that of the ACC was 2.055 ± 1.439 mmH_2O (Figure 7), which was not significantly different (P = 0.446, Appendix B, Table B1). Similarly, the mean exhalation PD of ACN (3.262 ± 2.058 mmH_2O) was not significantly different (P = 0.736) from that of the ACC (3.446 ± 2.535 mmH_2O) (Figure 8). The primary effects of ACF forms on both inhalation and exhalation PD were statistically significant (P < 0.001 in both cases) (see Appendix D1 and D2 respectively).

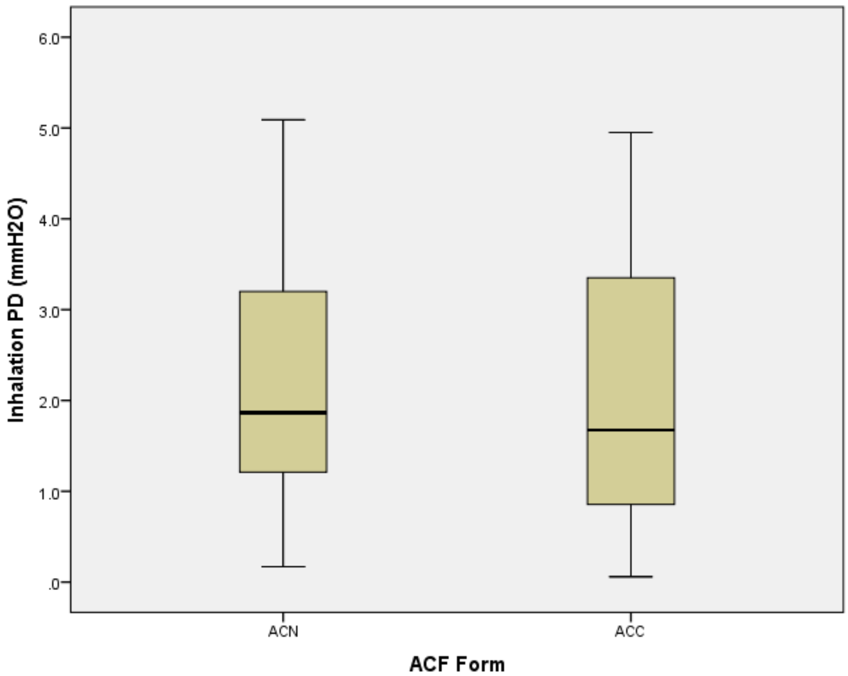


Figure 7. Inhalation Pressure Drop by ACF Form

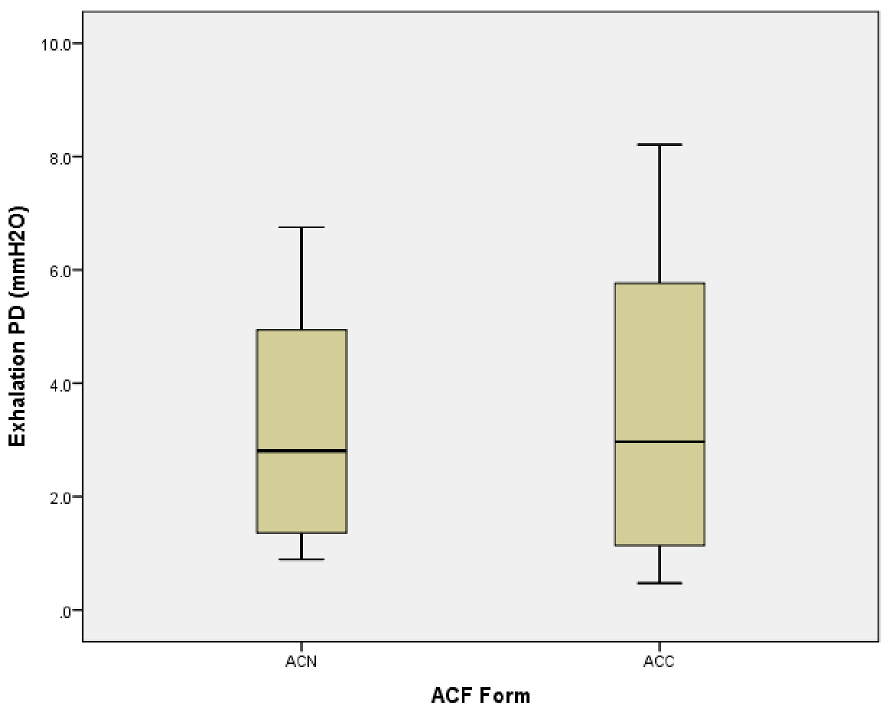


Figure 8. Exhalation Pressure Drop by ACF Form

Effect of Breathing Pattern on Inhalation and Exhalation Pressure Drop

Inhalation PD data per breathing pattern (N = 36) and exhalation PD data per breathing pattern (N = 36) were pooled together, regardless of other variables, to determine the mean PD. Increasing air flow in the 68 x 20 breathing pattern caused an increase in inhalation and exhalation PD compared to inhalation and exhalation PD obtained from the 20 x 20 breathing pattern. The mean inhalation PD for the 20 x 20 pattern was 1.016 ± .490 mmH_2O, and that of the 68 x 20 pattern (3.354 ± 1.054 mmH_2O) was significantly higher ($P < 0.001$) (Figure 9). Similarly, the mean exhalation PD for the 20 x 20 pattern was 1.296 ± 4.505 mmH_2O, and that of the 68 x 20 pattern (5.413 ± 1.327 mmH_2O) was significantly higher ($P < 0.001$) (Figure 10). For both inhalation and exhalation PD, the 68 x 20 breathing pattern resulted in higher PD values than the 20 x 20 pattern. The primary effect of breathing patterns on both inhalation and exhalation PD were statistically significant ($P < 0.001$ in both cases, Appendix Tables D1 and D2 respectively). Appendix B (Tables B2) shows the mean and significance of breathing pattern on inhalation and exhalation PD.

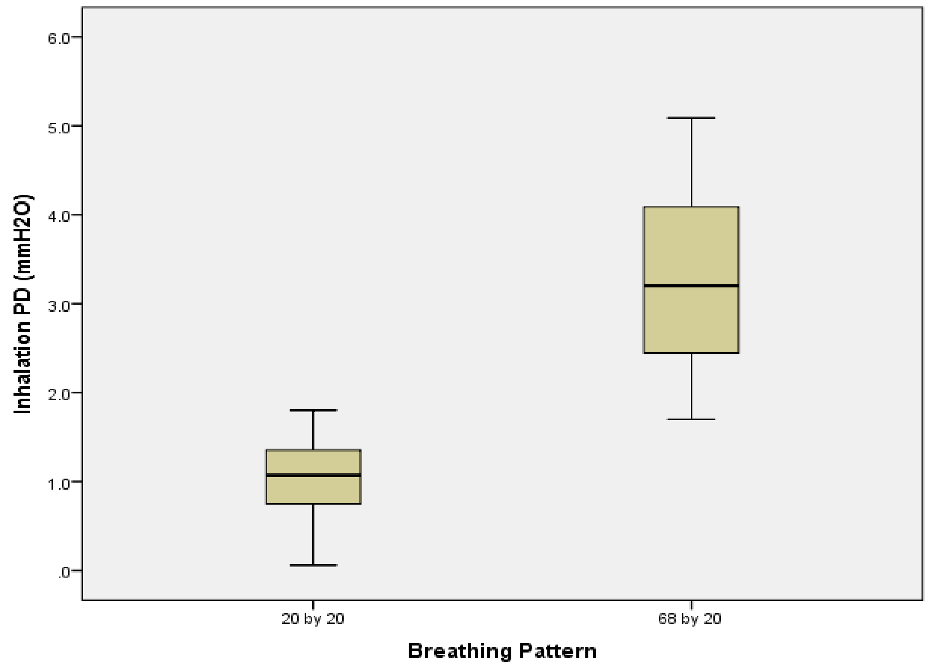


Figure 9. Inhalation Pressure Drop by Breathing Pattern

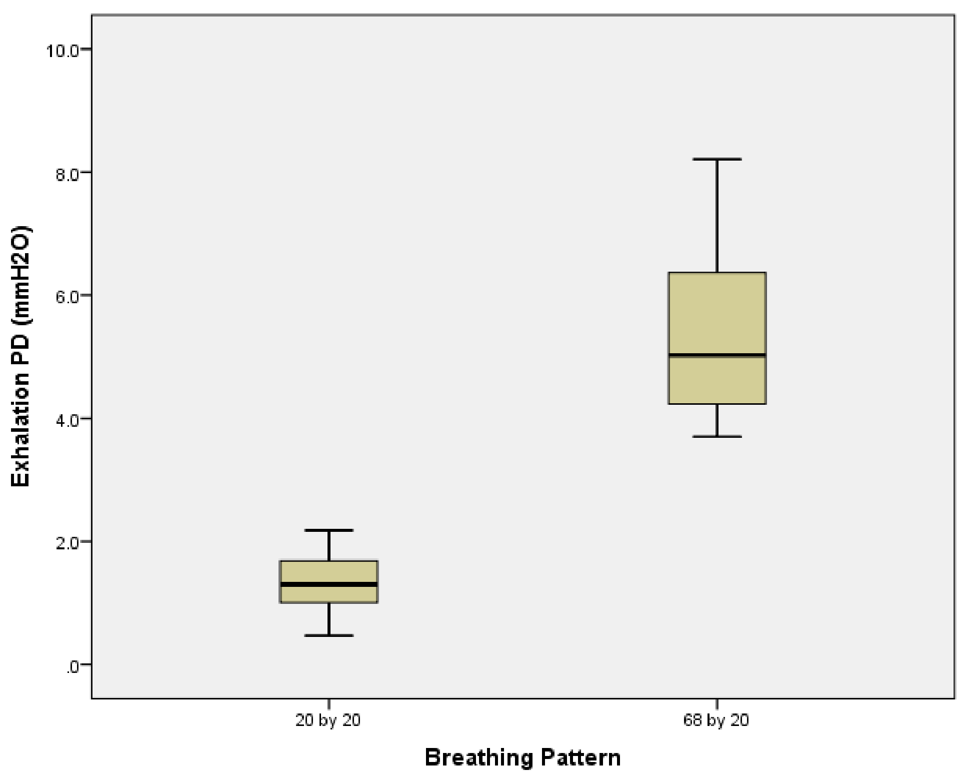


Figure 10. Exhalation Pressure Drop by Breathing Pattern

Inhalation Pressure Drop by ACF Forms and Breathing Pattern

Inhalation PD data per ACF form and breathing pattern were pooled together, regardless of other variables, to determine the mean PD (N = 18). Figure 11 shows the mean inhalation PD by ACF form and breathing pattern. Using the 20 x 20 breathing pattern, the mean inhalation PD for ACC (0.852 ± 0.489 mmH_2O, ranging from 0.180 to 1.650 mmH_2O) was significantly lower ($P = 0.043$) than that for ACN (1.179 ± 0.444 mmH_2O, ranging from 0.170 to 1.800 to mmH_2O). However, using the 68 x 20 breathing pattern, the mean inhalation PD values between ACC (3.258 ± 0.978 mmH_2O, ranging from 1.700 to 4.950 to mmH_2O) and ACN (3.450 ± 1.145 mmH_2O, ranging from 1.930-5.090 mmH_2O) were not significantly different ($P = 0.593$). There was an increase in the spread of recorded PD values compared to using the 20 x 20 pattern. For both ACF forms, the 68 x 20 breathing pattern had a significantly higher ($P < 0.001$) inhalation PD compared to the 20 x 20 pattern.

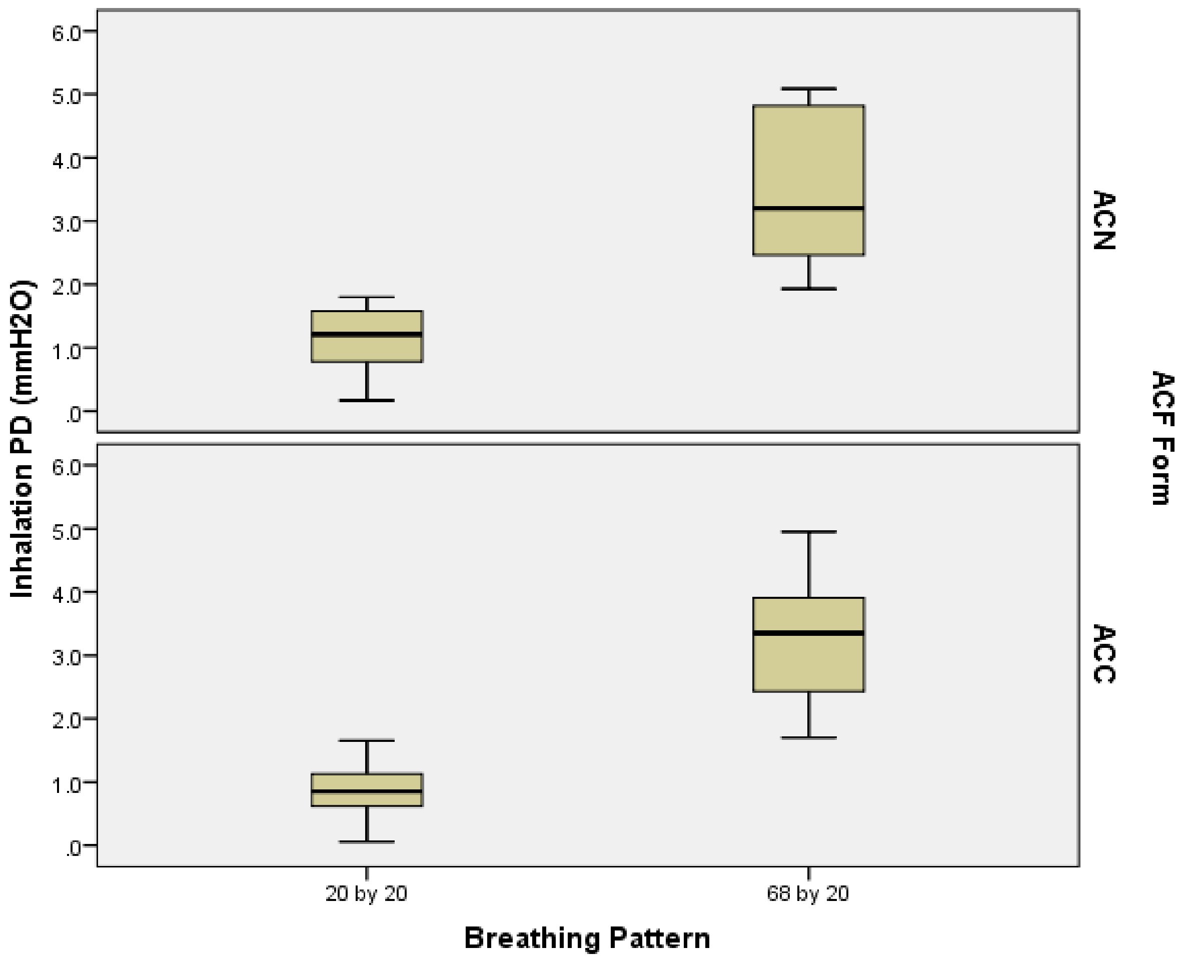


Figure 11. Inhalation Pressure Drop by ACF Form and Breathing Pattern

Exhalation PD and Breathing Pattern by ACF Forms

Exhalation PD data per ACF form and breathing pattern were pooled together, regardless of other variables, to determine the mean PD (N = 18). Figure 12 shows the mean exhalation PD by ACF form and breathing pattern. With the breathing pattern of 20 x 20, the mean exhalation PD in ACN was 1.397 ± 0.343 mmH_2O (ranging from 0.890 to 1.920 mmH_2O) while that for ACC was 1.194 ± 0.528 mmH_2O (ranging from 0.470 to 2.180 mmH_2O), which are not significantly different (P = 0.179). Likewise, when the 68 x 20 breathing pattern was used, the mean exhalation PD of ACN (5.127 ± 1.113 mmH_2O, ranging from 3.700 to 6.750) was not significantly different (P = 0.200) from that of ACC (5.699 ± 1.487, ranging from 3.760 and 8.210). Appendix C (Tables C1 and C2) presents the mean and significance of inhalation and exhalation PD by ACF forms and breathing pattern.

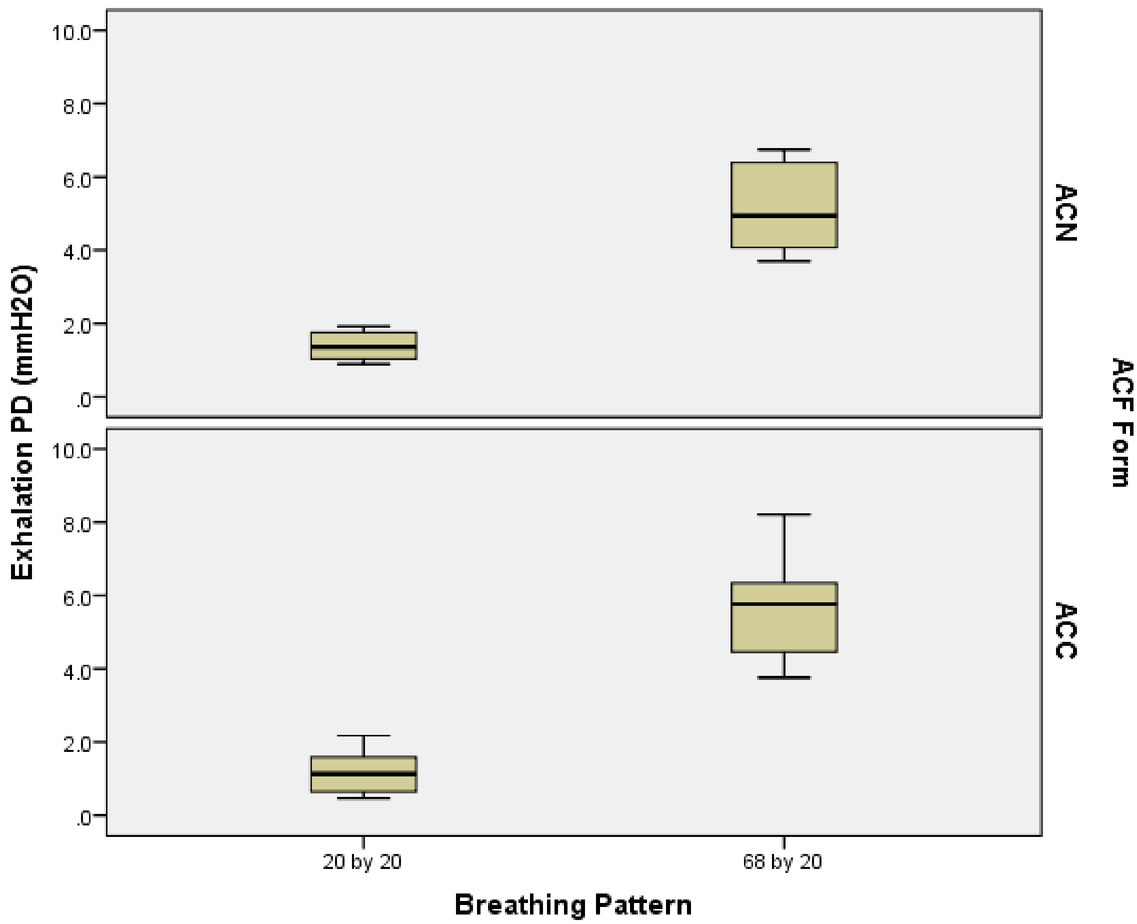


Figure 12. Exhalation Pressure Drop by ACF Form and Breathing Pattern

Effect of Surface Area on Inhalation and Exhalation Pressure Drop

Inhalation PD data per surface area (N = 24) and exhalation PD per surface area (N = 24) were pooled together irrespective of other variables to determine the mean PD. The structure and fiber organization of ACFs at the three surface areas under investigation influenced inhalation and exhalation PD. The mean inhalation PD at 1000 m^2/g (2.267 ± 1.449 mmH_2O), 1500 m^2/g (1.962 ± 1.365 mmH_2O) and 2000 m^2/g (2.326 ± 1.514 mmH_2O) were not significantly different (P = 0.647). Likewise for exhalation PD, the mean PD at 1000 m^2/g (3.434 ± 2.395 mmH_2O), 1500 m^2/g (3.02 ± 2.092 mmH_2O) and 2000 m^2/g (3.609 ± 2.439 mmH_2O) were not significantly different (P = 0.665). Despite this, surface area of 1500 m^2/g reported the least inhalation and exhalation PD in both cases. The primary effect of surface area on inhalation PD and exhalation PD were both significant (P < 0.001 in both cases) (Appendix D, Tables D1 and D2, respectively). Appendix B (Tables B3) shows the means and significance of surface areas on inhalation and exhalation PD.

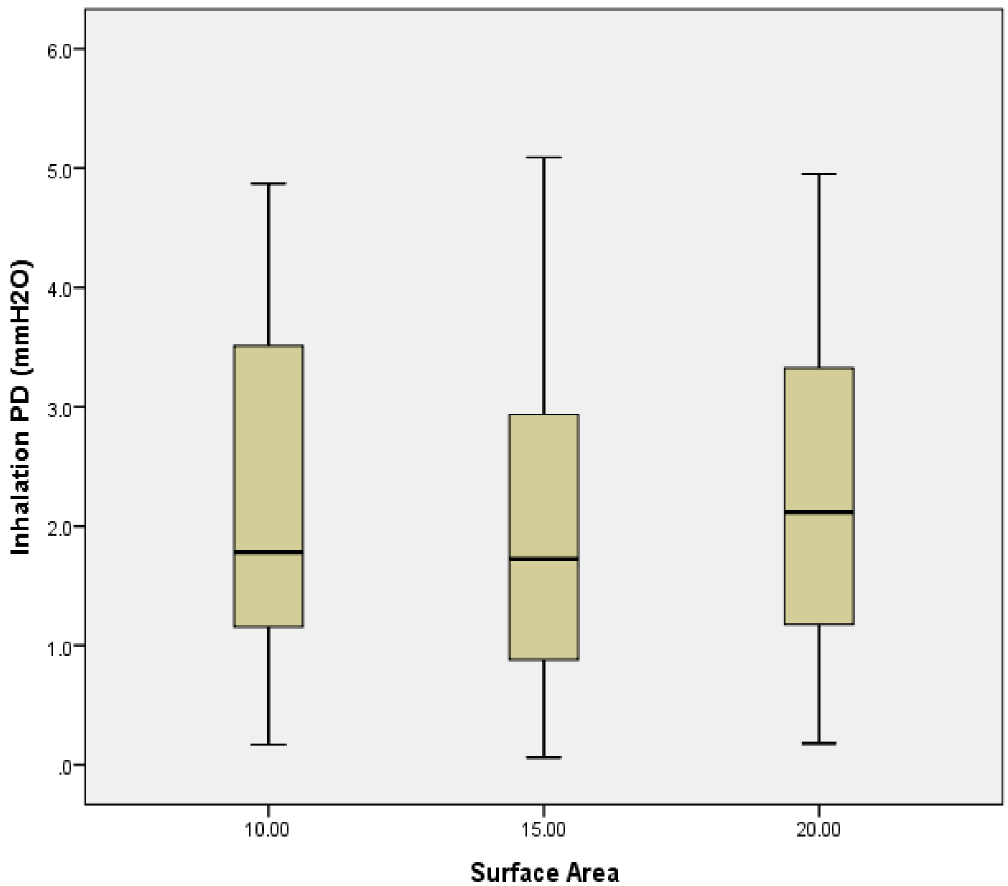


Figure 13. Inhalation Pressure Drop by Surface area

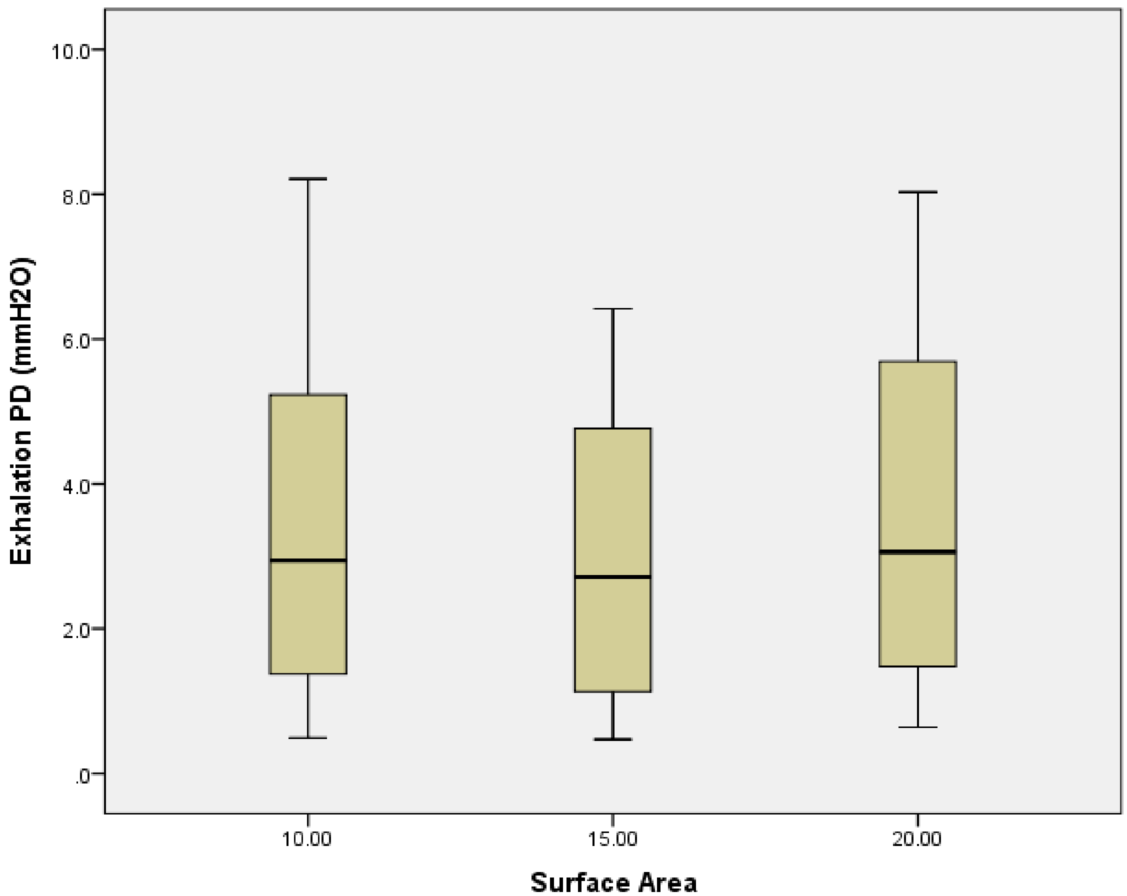


Figure 14. Exhalation Pressure Drop by Surface Area.

Inhalation PD and Surface Area by ACF Forms

Each ACF form was tested at three surface areas: 1000, 1500 and 2000 g/m^2. Inhalation PD data per ACF form and surface area were pooled together, regardless of other variables, to determine the mean PD (N = 12). Figure 9 shows the mean inhalation PD by ACF form and surface area. At surface area of 1000 g/m^2, the average inhalation PD for ACN (2.229 ± 1.487 mmH$_2$O, ranging from 0.170-4.860 mmH$_2$O) was not statistically significant (P = 0.901) from that for ACC (2.305 ± 1.475, ranging from 0.690 to 4.870 mmH$_2$O). Using a surface area of 1500 and 2000 g/m^2 resulted in lower inhalation PDs in ACC than ACN. At a surface area of 1500 g/m^2, the average PD for ACN was 2.263 ±1.480 mmH$_2$O (ranging from 0.730 to 5.090 to mmH$_2$O) while for ACC was 1.662 ± 1.229 mmH$_2$O (ranging from 0.060 to 3.670 mmH$_2$O) but this observed difference was not significant (P = 0.291). At a surface area of 2000 g/m^2, the mean PD values for ACN and ACC were 2.453 ± 1.455 mmH$_2$O (ranging from 0.680 to 4.890 to mmH$_2$O) and 2.199 ± 1.625 mmH$_2$O (ranging from 0.180 to 4.950 mmH$_2$O), respectively. Similarly, the observed difference was not statistically significant (P = 0.691) among all the ACF types; ACC15 had the lowest PD (1.662 mmH$_2$O) while ACN20 had the highest PD (2.453 mmH$_2$O). As the surface area increased for both ACF types, no apparent trend on the inhalation PD was observed.

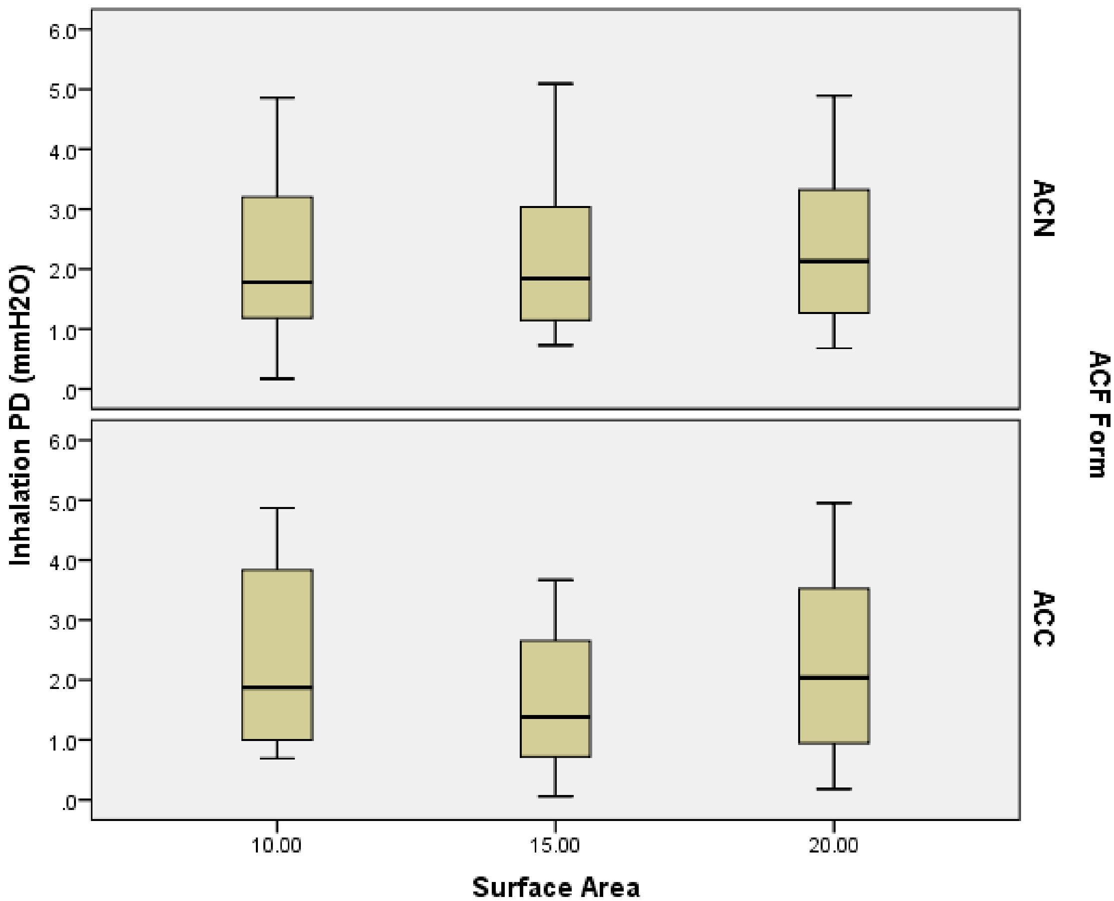


Figure 15. Inhalation Pressure Drop by ACF Form and Surface Area

Exhalation PD and Surface Area by ACF Forms

Exhalation PD data per ACF form and surface area were pooled together, regardless of other variables, to determine the mean PD (N = 12). Figure 10 shows the mean exhalation PD by ACF form and surface area. Using a surface area of 1000 g/m^2 , the mean ACN exhalation PD was 3.223 ± 2.076 mmH_2O (ranging from 1.040 to 6.530 mmH_2O), and it was not significantly different (P = 0.676) from the mean ACC exhalation PD (3.645 ± 2.755 mmH_2O, ranging from 0.490 to 8.210 mmH_2O). Also, at surface area of 1500 g/m^2, mean exhalation PD for ACN (3.152 ± 2.088 mmH_2O, ranging from 0.890 to 6.420) and for ACC (2.888 ± 2.181 mmH_2O, ranging from 0.470 to 6.420 mmH_2O) was not statistically different (P = 0.765). PD results from using an area of 2000 g/m^2 followed the same trend with results obtained from using surface area of 1000 g/m^2. For ACN20, the mean exhalation PD was 3.412 ± 2.185 mmH_2O (ranging from 1.000 to 6.750 mmH_2O), and was not significantly different (P = 0.701) from that for ACC20 (3.806 ± 2.754, ranging from 0.640 to 8.030 mmH_2O). Both ACF forms recorded lowest average exhalation PD at surface area of 1500 g/m^2 (i.e. ACN15 and ACC15), with ACC15 recording the least value. Similar to the inhalation PD, no apparent trend on the exhalation PD was observed as the surface area increased for both ACF types. Appendix tables C3 and C4 show the mean and significance of inhalation and exhalation PD values by ACF form and surface area.

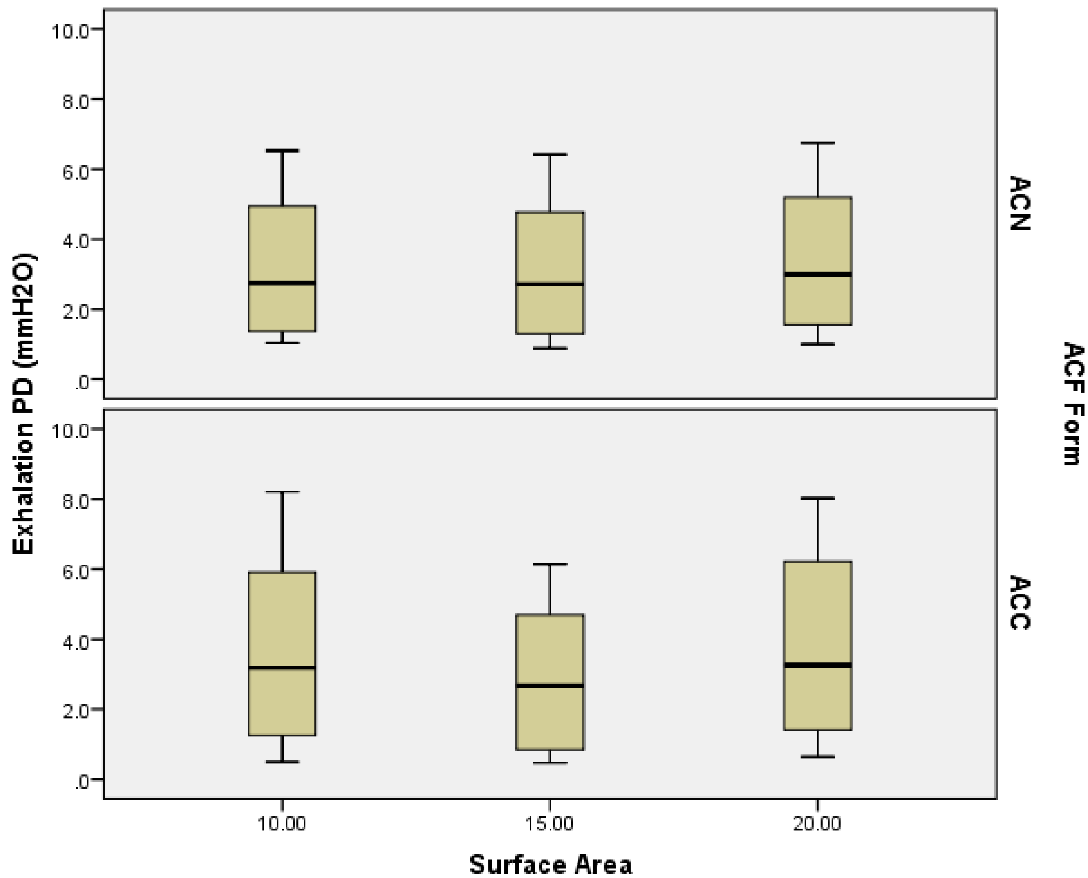


Figure 16. Exhalation Pressure Drop by ACF Form and Surface Area

Effect of ACF Layer Thickness on Inhalation and Exhalation PD

For this analysis, ACF layer arrangement was categorized by thickness into low, medium, and high based on the number of ACF layers. This corresponds to three, five and seven layers for ACC and five, seven, and nine layers for ACN. Inhalation PD per layer thickness (N = 24) and exhalation PD data per layer thickness (N = 24) were pooled together regardless of other variables to calculate the mean PD. The mean inhalation PD at low ACF layers was 1.404 ± .9507 mmH_2O, at medium ACF layers was 2.183 ± 1.252 mmH_2O, and at high ACF layers was 2.968 ± 1.608 mmH_2O (Figure 17). The observed difference in inhalation PD per layer thickness was statistically significant ($P = 0.016$). For exhalation PD, the mean PD at low ACF layers was 2.447 ± 1.659 mmH_2O, at medium ACF layers was 3.294 ± 2.092 mmH_2O, and at high ACF layers was 4.322 ± 2.699 mmH_2O (Figure 18). The difference in exhalation PD per layer was also statistically significant ($P < 0.001$). Inhalation and exhalation PD increased with increasing layers. The primary effect of ACF layers on inhalation and exhalation PD was significant ($P < 0.001$, Appendix Tables D1 and D2) in both cases. Appendix Table B4 shows the mean and significance of ACF layers on inhalation and exhalation PD.

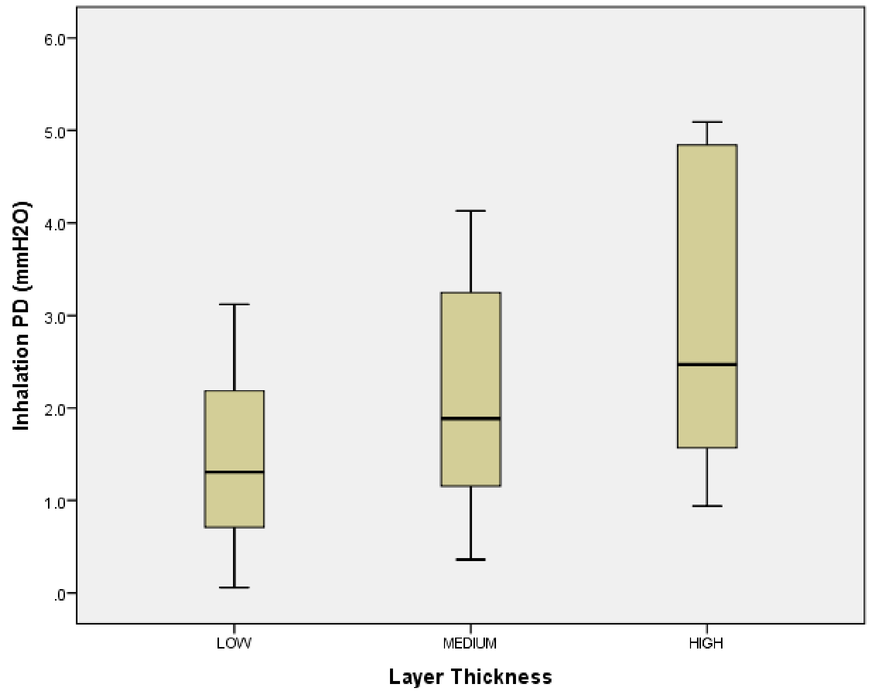


Figure 17. Inhalation Pressure Drop by Layer Thickness

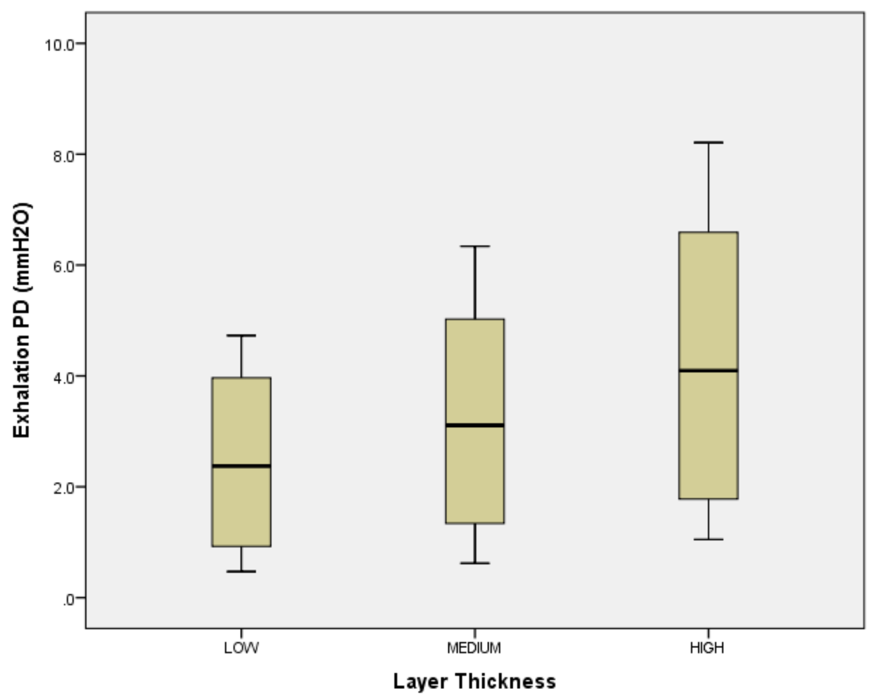


Figure 18. Exhalation Pressure Drop by Layer Thickness

Inhalation PD and Layer Thickness by ACF Forms

For the analysis, the number of layers per ACF form was categorized by thickness into low, medium and high: 3, 5 and 7 layers for ACC and 5, 7 and 9 layers for ACN, respectively. Inhalation PD data per ACF form and layer thickness were pooled together, regardless of other variables, to determine the mean PD (N = 12). Figure 19 shows the mean inhalation PD by ACF form and layer thickness.

Among the layer thickness, ACN of low layers showed a mean inhalation PD of 1.474 ± 0.908 mmH_2O (ranging from 0.170 to 3.120 mmH_2O) while ACC of low layers showed a mean inhalation PD of 1.334 ± 1.027 mmH_2O (ranging from 0.180 to 2.860 mmH_2O), but this difference was not significant statistically (P = 0.727). With medium ACF layers, the average inhalation PD for ACC (2.171 ± 1.481, ranging from 0.360 to 4.130 mmH_2O) was only slightly different from that of ACN (2.194 ± 1.042 mmH_2O, ranging from 1.070 to 3.370 mmH_2O) and therefore, was not statistically significant (P = 0.965). With high ACF layers, the average inhalation PD for ACN 3.276 ± 1.687, (ranging from 1.560 to 5.090 mmH_2O) was not statistically significant (P = 0.360) than ACC average inhalation PD 2.661 ± 1.535 (ranging from 0.940 to 4.950 mmH_2O). Among the thickness layers by ACF form, the mean inhalation PD was lowest for ACC with low layers (1.334 mmH_2O). Increasing ACF layers increased inhalation PD in both ACC and ACN types.

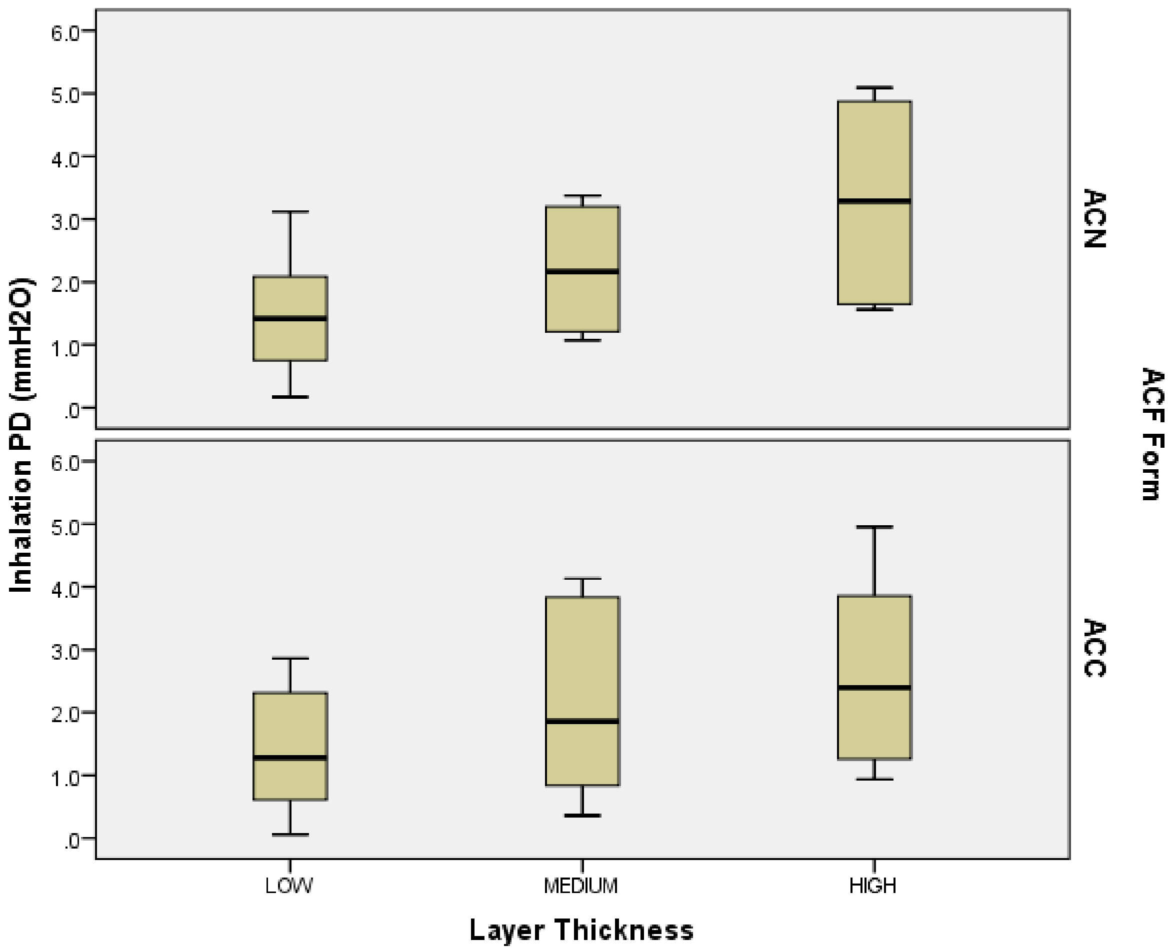


Figure 19. Inhalation Pressure Drop by ACF Form and Layer Thickness

Exhalation PD and Layer Thickness

Exhalation PD data per ACF form and layer thickness were pooled together, regardless of other variables, to determine the mean PD (N = 12). Figure 20 shows the mean exhalation PD by ACF form and layer thickness. At low layers, the mean exhalation PD for ACN at 2.457 ± 1.523 mmH_2O (ranging from 0.890 to 4.240 mmH_2O) was not significantly higher (P = 0.977) than ACC mean exhalation PD of 2.437 ± 1.854 mmH_2O (ranging from 0.470 and 4.730 mmH_2O). At both medium and high layers, the mean ACC exhalation PD was more than that of the ACN. At medium layers, ACN showed a mean exhalation PD of 3.188 ± 1.866 mmH_2O (ranging from 1.270 to 5.280 mmH_2O) while ACC showed a mean PD of 3.400 ± 2.376 mmH_2O (ranging from 0.620 to 6.340 mmH_2O), but this observed difference in PD between ACFs at this layer was however not statistically significant (P = 0.811). Likewise, for high layers of ACN, the mean exhalation PD was 4.142 ± 2.474 mmH_2O (ranging from 1.680 to 6.750 mmH_2O) is not significantly different (P = 0.751) from the mean ACC exhalation PD of 4.503 ± 3.007 mmH_2O (ranging from 1.050 to 8.210 mmH_2O). Both ACF forms recorded the lowest mean exhalation PD at low layers, with ACC only slightly lower than ACN. Similarly with inhalation PD, the exhalation PD in both ACC and ACN types increased as the ACF layers increased. Appendix tables C5 and C6 tabulates the mean and significance of inhalation and exhalation PD values by ACF form and layer thickness.

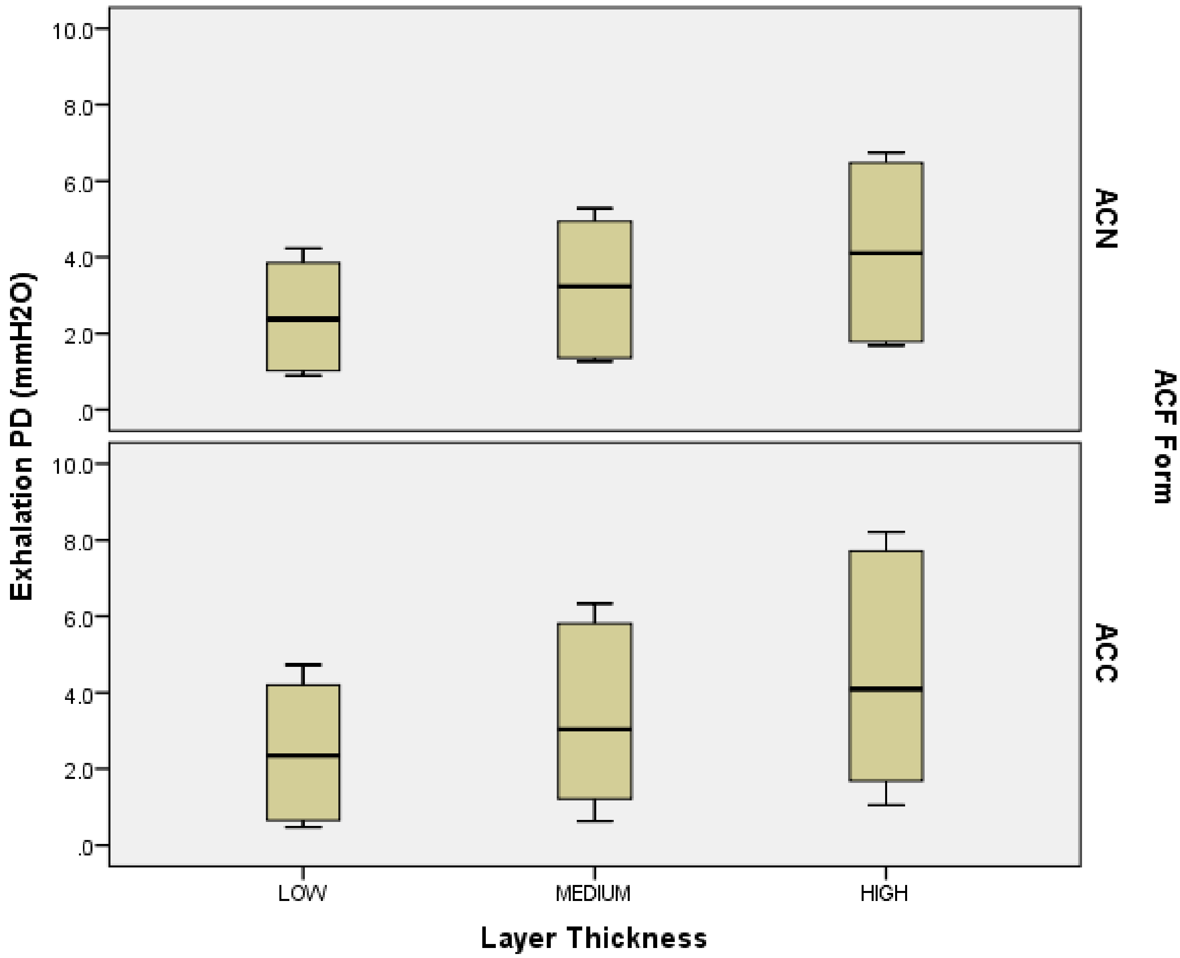


Figure 20. Exhalation Pressure Drop by ACF Form and Layer Thickness

Interaction of Study Variables

Interaction of ACF forms, layers, surface areas and breathing pattern were observed to show the effect of each individual factor, as well as the effect of the combined factors on both inhalation and exhalation PDs. These interactions demonstrate the importance of the individual and combined effects of the study variables in designing effective and efficient respirators.

For inhalation PD, the primary effect of ACF forms, layers, breathing patterns and specific surface areas were all significant. The following two-way interactions were significant: forms × area ($P = 0.005$), layers × breathing pattern ($P < 0.001$), and forms × layers ($P = 0.010$). Likewise, the two-way interaction between breathing pattern and area was almost significant ($P = 0.051$). In contrast, the interaction of forms × breathing pattern, and layers × area were not significant ($P = 0.396$ and $P = 0.670$). The effect of breathing pattern and layers were significant ($P < 0.01$) for each ACF form, while the effect of area and layers, and area and breathing pattern were not significant for each ACF form ($P = 0.873$ and $P =0.462$, respectively). Also, the effect of area and breathing pattern were the same for each layer ($P = 0.216$), but the effect of area, breathing pattern and layers were not significantly different for each form of ACF ($P = 0.721$).

For exhalation PD, the primay effects of ACF form, layers, breathing pattern, specific surface area were all significant. The following two-way interactions were significant: form × breathing pattern ($P < 0.001$), form × area ($P < 0.001$), layers × breathing pattern ($P < 0.001$), and breathing pattern × area ($P < 0.001$). However, the following interactions were not significant: form and layers ($P = 0.016$), and layers and area ($P = 0.040$). The effects of breathing pattern and layers on the two forms of ACF were not significant ($P = 0.505$). Also, the effects of area and layers on the forms of ACF were not significant ($P = 0.078$), as well as the effects of breathing pattern and area on layers ($P = 0.554$). In contrast, the effect of area and breathing pattern on forms of ACF was significant ($P < 0.001$). The interaction between ACF forms, layers, breathing pattern and area are not significant (*P* = 0.510). Appendix D shows the SPSS Statistical Output for the determination of interaction among study variables for both inhalation and exhalation PDs.

VII. DISCUSSION

Factors Influencing Pressure Drop across Respirator Cartridges

The primary outcome of each of the four factors on pressure drop across ACF respirator cartridges was investigated: ACF form, ACF surface area, ACF layer thickness and breathing pattern. The individual effects of each of these factors were found to be significant on inhalation and exhalation PD across the tested respirator cartridges this validates the main hypothesis.

Despite the remarkable difference in the fiber organization of the ACF forms has shown in the SEM images (figures 3, 4, and 5), based on the ACF arrangement in the current experiment, the mean inhalation PD of the gradually distributed ACN was not significantly different from the mean inhalation PD of the closely knitted ACC. Similarly, the mean exhalation PD of the ACN was not significantly different from ACC. This result nullifies the (first) hypothesis that pressure drop across ACF cartridge is significantly different between the ACF cloth and ACF felt. This implies that either of the ACF forms may be breathable adsorbent in respirator cartridge. Therefore, a desirable choice can be made based on other important adsorbent characteristics, such as the absorption capacity and breakthrough time.

For both inhalation and exhalation settings, PD values were significantly lower when the 20 x 20 breathing pattern was used compared to the 68 x 20 breathing pattern. This observation validates the (secound) hypothesis that the pressure drop across ACF cartridge is significantly different between the 20 x 20 and 68 x 20 breathing patterns. In both breathing patterns, the breathing frequencies (20 breaths per minute) were the same but the flow rate was higher in the second breathing pattern (i.e., 20 vs. 68 LPM). Thus, the 68 by 20 breathing pattern represents a worker performing a more strenuous activity, resulting to deeper breathing. This observation supports the recommended airflow of 135 LPM using sinusoidal breathing pattern for respirator testing and certification of dual cartridge respirators by Coyne et al. (2006). Both studies suggest that varying airflow impacts PD differently across ACF respirators and that the worker's physical activity and other factors that may influence breathing pattern may affect the acceptability of pressure drop across ACF respirators. Further investigation of this impact will be necessary to determine the breathing pattern that can be employed in designing effective respirators in varying physical activities.

Considering the three specific surface areas investigated 1000 m^2/g, 1500 m^2/g and 2000 m^2/g, inhalation PD was least at 1000 m^2/g for ACN and 1500 m^2/g for ACC, with specific surface area of 1500 m^2/g having lesser PD values among tested surface areas. Nevertheless, there was no statistically significant difference in inhalation PD per specific surface area. This indicated that the difference in inhalation PD was not important among the three surface areas investigated. Similarly, the low mean exhalation PD of ACF at surface area of 1500 m^2/g was not important because the exhalation PD of ACFs at three surface areas were not significantly different. The results obtained nullifies the (third) hypothesis that the pressure drop across ACF cartridge is significantly different among specific surface areas of 1000, 1500 and 2000 m2/g. Supposedly, the three surface areas that were examined will not affect the breathing resistance of the two ACF forms. ACF types with high surface areas were previously shown to have the highest adsorption capacity compared to those with lower surface areas (Balanay et al., 2014). Therefore, the use of high surface area ACFs may be desirable in designing a breathable respirator that will be both convenient and efficient in airborne toluene protection and will have longer service lives. Inhalation and exhalation PD determination of ACF at higher layers may be required to validate this finding.

Increasing the number of ACF layers increased both the mean exhalation and inhalation PD significantly in ACF cartridges. Results validate the (fourth) hypothesis that the pressure drop across ACF cartridge is significantly different among low, medium and high layer thickness. This is important to consider in designing the thickness of ACF respirators because increased layer thickness may result in unbreathable respirators which may negatively impact workers' respirator use compliance. Since higher adsorbent thickness corresponds to higher adsorption capacity, the optimum layer balance for high adsorption capacity and breathable pressure drop must be determined.

It is crucial to determine the factors affecting the pressure drop across ACF materials because understanding such factors will aid in the design of ACF respirators and will keep both inhalation and exhalation PDs as low as possible without compromising its ability to protect workers from airborne pollutants. A study by Caretti et al. (2006) showed that increasing inhalation PD result in significant linear decreases in work performance regardless of exhalation PD likewise, increasing exhalation PD was found to reduce work performance though insignificantly. In addition, previous study showed that increased inhalation resistance reduced physical performance and comfort of subjects (Lerman et al, 1983). Caretti et al. (2001) reported significant reduction in work performance of subjects as exhalation PD increased. Consequently, ACF form, breathing pattern, surface area and layer arrangement that give the lowest inhalation and exhalation PDs should be used in respirator certification testings and designs.

Interaction Effect of Breathing Pattern on Pressure Drop across ACF respirator cartridges

For inhalation PD, both ACF forms recorded their highest average PD when the 68 x 20 breathing pattern was used with ACC respirator cartridges, recording significantly lower (P =

0.043, Appendix Table C1) inhalation PD per breathing pattern per ACF form than ACN respirator cartridges. In spite of this, the interaction between ACF forms and breathing pattern was not statistically significant for inhalation PD. On the other hand for exhalation PD, the effect of breathing pattern on ACF forms was significant, and the 68 x 20 breathing pattern recorded higher exhalation PD values than the 20 x 20 pattern. Exhalation PD was least for ACC cartridges when the 20 x 20 pattern was used while ACN recorded a lower PD than ACC when the 68 x 20 breathing pattern was used. Previous studies (Coyne et al., 2006) reported that dual cartridge respirators tested using sinusoidal airflow of 135 LPM simulate real life situations, since single cartridge was tested in this study, high average inhalation and exhalation PD reported for airflow of 68 LPM suggest that an ACF respirator with low mean inhalation and exhalation PD at this airflow will be breathable during real life hard work.

Interaction Effect of Surface Area on Pressure Drop across ACF Respirator Cartridges

The interaction of ACF types and surface area was significant for both inhalation and exhalation PD. At a surface area of 1000 g/m^2, ACC had higher average inhalation PD than ACN but ACC recorded lower mean inhalation PD than ACN at areas of 1500 g/m^2 and 2000 g/m^2, surface area of 1500 g/m^2 recorded the least average inhalation PD values. For exhalation PD, ACN had a lower PD than ACC at area of 1000 g/m^2 and 2000 g/m^2 while ACC recorded lower PD than ACN at 1500 g/m^2. The mid-grade surface area of 1500 g/m^2 showed the lowest mean exhalation PD values.

Interaction Effect of Layers on Pressure Drop across ACF Respirator Cartridges

The interaction of ACF types and number of layers was significant for inhalation and exhalation PD. Using low and high layers, ACC had a lower inhalation PD than ACN. ACC's

PD was only slightly lower than ACN when medium number of layers was used. For exhalation PD, ACN reported lower PD with medium and high numbers of layers while ACC had a lower PD with low layers. In general, inhalation and exhalation PD increased with increasing ACF layers for both ACN and ACC respirator cartridges and low layers of ACC resulting in the lowest inhalation and exhalation PD values. In this study, however, ACN low layers and ACC low layers does not correspond to the same number of layers. For example, low layers mean three layers for ACC and five layers for ACN. Moreover, the two ACF forms differ in thickness and weaving density (as demonstrated by the SEM images), which are the two factors that affect PD across any material. Future investigations may involve testing different ACF forms at the same bed depth or same mass of material.

NIOSH Requirements on Respirator Pressure Drop Testing

In the US, NIOSH is the federal agency responsible for carrying out respirator certification. The maximum inhalation resistance across dual carriage chemical cartridge respirators is 40 mmH_2O and that of exhalation resistance is 20 mmH_2O using a constant air flow of 85 LPM. Although the mean inhalation and exhalation PD per ACF form was much lower than these recommended values, it is not appropiate to use the continous air flow standard for PD measurement derived from sinusoidal air flow. Studies have showed that continuous air flow does not reflect workers everyday experience and more studies have emphasized the need for certification tests that simulate real life experience (Coyne et al., 2006). Consequently, research on inhalation and exhalation PD using sinusoidal airflow to test breathing resistance in respirators is important. For this to be effective, NIOSH need to set standards for inhalation and exhalation pressure drop using sinusoidal air flow.

VIII. CONCLUSIONS

ACF forms, breathing patterns, surface area and layer have significant effects on inhalation and exhalation PD. Therefore, ACF forms, breathing patterns, surface area and layer are important factors in designing a breathable respirator cartridge. Although inhalation and exhalation PD values obtained from the 20 x 20 breathing pattern were significantly lower compared to the 68 x 20 pattern, the latter simulate real life situations better and it is therefore more realistic to test respirators using this pattern. A surface area of 1500 g/m^2 resulted in least mean inhalation and exhalation PD values compared to 1000 and 2000 g/m^2 but this was not significantly different from those of the other surface areas tested. This implies that ACF with larger surface area may be employed in designing respirators with high adsorption capacity. Lower numbers of ACF layers resulted in significantly lower PD across ACN and ACC respirator cartridges. Increasing inhalation and exhalation PD reported across ACF cartridges with increasing ACF layers suggest an optimum layer for ACF use in designing an efficient and comfortable respiratory protection device. The ability of ACC in recording insignificantly lower breathing resistance than ACN was also observed. Based on this study and the observed results, ACC of specific surface area 1500 g/m^2 may be recommended for designing respirator cartridges with ACF adsorbent.

Studies have emphasized the need to change airflow pattern in respirator certification testing to sinusoidal airflow in order to adequately simulate work place conditions better. Therefore, it is important for NIOSH to develop maximum inhalation and exhalation pressure drop standards for sinusoidal flow against which respirator inhalation and exhalation pressure drop can be tested for acceptability. Moreover, further investigation of inhalation and exhalation

PD at higher layers of ACC and ACN similar to what is obtained in commercial respirators should be carried out using a realistic breathing pattern.

Lastly, ACF form, breathing pattern, surface area and layer arrangement that give the lowest inhalation and exhalation PDs may be used in respirator certification testings and designs.

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X. APPENDICES

Appendix A. Mean Inhalation and Exhalation Pressure Drop (PD) Values by Study Variable and ACF Forms

Appendix B. SPSS Statistical Output for Inhalation and Exhalation Pressure Drop (PD) Tests of Individual Study Variables

Appendix C. SPSS Statistical Output for Test of Study Variables on Inhalation and Exhalation Pressure Drop (PD) by ACF Forms

Appendix D. SPSS Statistical Output for Tests of Between-Subjects Effects

APPENDIX A: Mean Inhalation and Exhalation Pressure Drop (PD) Values by Study Variables

Table A1. Inhalation Pressure Drop (mmH_2O) Breathing Pattern and ACF Form

	20 by 20			68 by 20		
	Both Forms (N=36)	ACN (N=18)	ACC (N=18)	Both Forms (N=36)	ACN (N=18)	ACC (N=18)
Mean	1.016	1.179	0.852	3.354	3.450	3.258
Median		1.210	0.855		3.200	3.350
Std. Deviation	0.490	0.444	0.489	1.054	1.145	0.978
Minimum		0.170	0.180		1.930	1.700
Maximum		1.800	1.650		5.090	4.950

Table A2. Exhalation Pressure Drop (mmH_2O) by Breathing Pattern and ACF Form

	20 by 20			68 by 20		
	Both Forms (N=36)	ACN (N=18)	ACC (N=18)	Both Forms (N=36)	ACN (N=18)	ACC (N=18)
Mean	1.296	1.397	1.194	5.413	5.127	5.699
Median		1.360	1.135		4.940	5.765
Std. Deviation	0.451	0.343	0.528	4.964	1.113	1.487
Minimum		0.890	0.470		3.700	3.760
Maximum		1.920	2.180		6.750	8.210

Table A3. Inhalation Pressure Drop (mmH_2O) by Surface Area (m^2/g) and ACF Form

	1000			1500			2000		
	Both Forms (N=24)	ACN (N=12)	ACC (N=12)	Both Forms (N=24)	ACN (N=12)	ACC (N=12)	Both Forms (N=24)	ACN (N=12)	ACC (N=12)
Mean	2. 267	2.229	2.305	1.962	2.263	1.662	2.326	2.453	2.199
Median		1.780	1.875		1.840	1.385		2.130	2.040
Std. Deviation	1.449	1.487	1.475	1.365	1.480	1.229	1.514	1.455	1.625
Minimum		4.860	4.870		5.090	3.670		4.890	4.950
Maximum		0.170	0.690		0.730	0.060		0.680	0.180

Table A4. Exhalation Pressure Drop (mmH_2O) by Surface Area (m^2/g) and ACF Form

	1000			1500			2000		
	Both Forms (N=24)	ACN (N=12)	ACC (N=12)	Both Forms (N=24)	ACN (N=12)	ACC (N=12)	Both Forms (N=24)	ACN (N=12)	ACC (N=12)
Mean	3.434	3.223	3.645	3.02	3.152	2.888	3.609	3.412	3.806
Median		2.750	3.175		2.715	2.675		2.995	3.260
Std. Deviation	2.395	2.076	2.755	2.092	2.088	2.181	2.439	2.185	2.754
Minimum		1.040	0.490		0.890	0.470		1.000	0.640
Maximum		6.530	8.210		6.420	6.140		6.750	8.030

Table A5. Inhalation Pressure Drop (mmH_2O) by Layer Thickness and ACF Form

	Low			Medium			High		
	Both Forms (N=24)	ACN (N=12)	ACC (N=12)	Both Forms (N=24)	ACN (N=12)	ACC (N=12)	Both Forms (N=24)	ACN (N=12)	ACC (N=12)
Mean	1.404	1.475	1.334	2.183	2.194	2.171	2.968	3.276	-2.661
Median		1.420	1.280		2.165	1.860		3.285	-2.395
Std. Deviation	0.951	0.908	1.027	1.252	1.042	1.481	1.608	1.687	1.535
Minimum		3.120	2.860		3.370	4.130		5.090	-4.950
Maximum		0.170	0.180		1.070	0.360		1.560	-0.940

Table A6. Exhalation Pressure Drop (mmH_2O) by Layer Thickness and ACF Form									
	Low			Medium			High		
	Both Forms (N=24)	ACN (N=12)	ACC (N=12)	Both Forms (N=24)	ACN (N=12)	ACC (N=12)	Both Forms (N=24)	ACN (N=12)	ACC (N=12)
Mean	2.447	2.457	2.437	3.294	3.188	3.400	4.322	4.142	4.503
Median		2.375	2.350		3.230	3.035		4.110	4.095
Std. Deviation	1.659	1.523	1.854	2.092	1.866	2.376	2.699	2.474	3.006
Minimum		0.890	0.470		1.270	0.620		1.680	1.050
Maximum		4.240	4.730		5.280	6.340		6.750	8.210

Appendix B. SPSS Statistical Output for Inhalation and Exhalation Pressure Drop (PD) Tests of Individual Study Variables

B1. INHALATION AND EXHALATION PD BY ACF FORMS

Descriptives

		N	Mean	Std. Deviation	Std. Error	95% Confidence Interval for Mean		Minimum	Maximum
						Lower Bound	Upper Bound		
Inhalation PD	ACN	36	-2.314722	1.4348290	.2391382	-2.800199	-1.829246	-5.0900	-.1700
	ACC	36	-2.055278	1.4387167	.2397861	-2.542069	-1.568486	-4.9500	.1800
	Total	72	-2.185000	1.4325885	.1688322	-2.521642	-1.848358	-5.0900	.1800
Exhalation PD	ACN	36	3.262222	2.0582227	.3430371	2.565820	3.958625	.8900	6.7500
	ACC	36	3.446389	2.5353841	.4225640	2.588538	4.304239	.4700	8.2100
	Total	72	3.354306	2.2947164	.2704349	2.815074	3.893537	.4700	8.2100

ANOVA

		Sum of Squares	df	Mean Square	F	Sig.
InhalationPD	Between Groups	1.212	1	1.212	.587	.446
	Within Groups	144.502	70	2.064		
	Total	145.714	71			
Exhalation PD	Between Groups	.611	1	.611	.114	.736
	Within Groups	373.256	70	5.332		
	Total	373.866	71			

B2. INHALATION AND EXHALATION PD BY BREATHING PATTERN

Descriptives

		N	Mean	Std. Deviation	Std. Error	95% Confidence Interval for Mean		Minimum	Maximum
						Lower Bound	Upper Bound		
Inhalation PD	20 by 20	36	-1.015833	.4895968	.0815995	-1.181489	-.850178	-1.8000	.1800
	68 by 20	36	-3.354167	1.0542956	.1757159	-3.710889	-2.997444	-5.0900	-1.7000
	Total	72	-2.185000	1.4325885	.1688322	-2.521642	-1.848358	-5.0900	.1800
Exhalation PD	20 by 20	36	1.295556	.4505327	.0750888	1.143117	1.447994	.4700	2.1800
	68 by 20	36	5.413056	1.3265806	.2210968	4.964205	5.861906	3.7000	8.2100
	Total	72	3.354306	2.2947164	.2704349	2.815074	3.893537	.4700	8.2100

ANOVA

		Sum of Squares	df	Mean Square	F	Sig.
Inhalation PD	Between Groups	98.420	1	98.420	145.674	.000
	Within Groups	47.294	70	.676		
	Total	145.714	71			
Exhalation PD	Between Groups	305.169	1	305.169	310.953	.000
	Within Groups	68.698	70	.981		
	Total	373.866	71			

B3. INHALATION AND EXHALATION PD BY SURFACE AREA

Descriptives									
		N	Mean	Std. Deviation	Std. Error	95% Confidence Interval for Mean		Minimum	Maximum
						Lower Bound	Upper Bound		
Inhalation PD	10.00	24	-2.26708	1.449271	.295831	-2.87905	-1.65511	-4.8700	-.1700
	15.00	24	-1.96208	1.365172	.278664	-2.53854	-1.38562	-5.0900	.0600
	20.00	24	-2.32583	1.513846	.309012	-2.96507	-1.68659	-4.9500	.1800
	Total	72	-2.18500	1.432588	.168832	-2.52164	-1.84835	-5.0900	.1800
Exhalation PD	10.00	24	3.43416	2.395139	.488909	2.42278	4.44554	.4900	8.2100
	15.00	24	3.02000	2.092311	.427091	2.13649	3.90350	.4700	6.4200
	20.00	24	3.60875	2.439214	.497902	2.57876	4.63874	.6400	8.0300
	Total	72	3.35430	2.294164	.270434	2.81507	3.89353	.4700	8.2100

ANOVA						
		Sum of Squares	df	Mean Square	F	Sig.
Inhalation PD	Between Groups	1.830	2	.915	.439	.647
	Within Groups	143.884	69	2.085		
	Total	145.714	71			
Exhalation PD	Between Groups	4.389	2	2.195	.410	.665
	Within Groups	369.477	69	5.355		
	Total	373.866	71			

B4. INHALATION AND EXHALATION PD BY LAYERS

Descriptives									
		N	Mean	Std. Deviation	Std. Error	95% Confidence Interval for Mean		Minimum	Maximum
						Lower Bound	Upper Bound		
Inhalation PD	LOW	24	-1.40416	.9507293	.194066	-1.805624	-1.002709	-3.1200	.1800
	MEDIUM	24	-2.18250	1.252268	.255618	-2.711287	-1.653713	-4.1300	-.3600
	HIGH	24	-2.96833	1.608360	.328305	-3.647484	-2.289182	-5.0900	-.9400
	Total	72	-2.18500	1.432588	.168832	-2.521642	-1.848358	-5.0900	.1800
Exhalation PD	LOW	24	2.44666	1.659394	.338722	1.745966	3.147367	.4700	4.7300
	MEDIUM	24	3.29416	2.092279	.427084	2.410674	4.177659	.6200	6.3400
	HIGH	24	4.32208	2.698725	.550875	3.182511	5.461655	1.0500	8.2100
	Total	72	3.3543	2.29471	.27043	2.815074	3.893537	.4700	8.2100

ANOVA						
		Sum of Squares	df	Mean Square	F	Sig.
Inhalation PD	Between Groups	29.360	2	14.680	8.705	.000
	Within Groups	116.354	69	1.686		
	Total	145.714	71			
Exhalation PD	Between Groups	42.336	2	21.168	4.406	.016
	Within Groups	331.530	69	4.805		
	Total	373.866	71			

Appendix C. SPSS Statistical Output for Test of Study Variables on Inhalation and Exhalation Pressure Drop (PD) by ACF Forms

TABLE C1: INHALATION PD BY ACF FORM AND BREATHING PATTERN

Breathing Pattern		Sum of Squares	Df	Mean Square	F	Sig.
20 BY 20	Between Groups	.964	1	.964	4.412	.043
	Within Groups	7.426	34	.218		
	Total	8.390	35			
68 BY 20	Between Groups	.331	1	.331	.291	.593
	Within Groups	38.573	34	1.135		
	Total	38.904	35			

TABLE C2: EXHALATION PD BY ACF FORM AND BREATHING PATTERN

Breathing Pattern		Sum of Squares	df	Mean Square	F	Sig.
20 BY 20	Between Groups	.372	1	.372	1.879	.179
	Within Groups	6.732	34	.198		
	Total	7.104	35			
68 BY 20	Between Groups	2.941	1	2.941	1.705	.200
	Within Groups	58.652	34	1.725		
	Total	61.594	35			

TABLE C3: INHALATION PD BY ACF FORM AND SURFACE AREA

Surface Area		Sum of Squares	df	Mean Square	F	Sig.
10.0	Between Groups	.035	1	.035	.016	.901
	Within Groups	48.274	22	2.194		
	Total	48.309	23			
15.0	Between Groups	2.166	1	2.166	1.171	.291
	Within Groups	40.699	22	1.850		
	Total	42.865	23			
20.0	Between Groups	.385	1	.385	.162	.691
	Within Groups	52.325	22	2.378		
	Total	52.710	23			

TABLE C4: EXHALATION PD BY ACF FORM AND SURFACE AREA

Surface Area		Sum of Squares	df	Mean Square	F	Sig.
10.0	Between Groups	1.067	1	1.067	.179	.676
	Within Groups	130.877	22	5.949		
	Total	131.944	23			
15.0	Between Groups	.416	1	.416	.091	.765
	Within Groups	100.273	22	4.558		
	Total	100.689	23			
20.0	Between Groups	.932	1	.932	.151	.701
	Within Groups	135.912	22	6.178		
	Total	136.845	23			

TABLE C5: INHALATION PD BY ACF FORM AND LAYER

LAYERS	Sum of Squares	df	Mean Square	F	Sig.

Low	Between Groups	.118	1	.118	.125	.727
	Within Groups	20.672	22	.940		
	Total	20.789	23			
Medium	Between Groups	.003	1	.003	.002	.965
	Within Groups	36.065	22	1.639		
	Total	36.068	23			
High	Between Groups	2.269	1	2.269	.872	.360
	Within Groups	57.228	22	2.601		
	Total	59.497	23			

Table Caption

TABLE C6: EXHALATION PD BY ACF FORM AND LAYER

LAYERS		Sum of Squares	df	Mean Square	F	Sig.
Low	Between Groups	.002	1	.002	.001	.977
	Within Groups	63.330	22	2.879		
	Total	63.333	23			
Medium	Between Groups	.269	1	.269	.059	.811
	Within Groups	100.417	22	4.564		
	Total	100.686	23			
High	Between Groups	.781	1	.781	.103	.751
	Within Groups	166.731	22	7.579		
	Total	167.512	23			

APPENDIX D: SPSS Statistical Output for Tests of Between-Subjects Effects

TABLE D1: INHALATION PRESSURE DROP STATISTICAL OUTPUT FOR TEST OF EFFECTS

Tests of Between-Subjects Effects					
Dependent Variable: INHALATION PD					
Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	141.688[a]	35	4.048	36.201	.000
Intercept	343.744	1	343.744	3073.871	.000
TYPES2	1.212	1	1.212	10.835	.002
LAYERS	29.360	2	14.680	131.272	.000
Breathing Pattern	98.420	1	98.420	880.107	.000
AREA	1.830	2	.915	8.184	.001
TYPES2 * LAYERS	1.179	2	.589	5.270	.010
TYPES2 * Breathing Pattern	.083	1	.083	.739	.396
TYPES2 * AREA	1.374	2	.687	6.143	.005
LAYERS * Breathing Pattern	5.020	2	2.510	22.444	.000
LAYERS * AREA	.265	4	.066	.593	.670
Breathing Pattern * AREA	.726	2	.363	3.244	.051
TYPES2 * LAYERS * Breathing Pattern	.994	2	.497	4.443	.019
TYPES2 * LAYERS * AREA	.137	4	.034	.305	.873
TYPES2 * Breathing Pattern * AREA	.176	2	.088	.789	.462
LAYERS * Breathing Pattern * AREA	.681	4	.170	1.523	.216
TYPES2 * LAYERS * Breathing Pattern * AREA	.233	4	.058	.520	.721
Error	4.026	36	.112		
Total	489.458	72			
Corrected Total	145.714	71			
a. R Squared = .972 (Adjusted R Squared = .946)					

Between-Subjects Factors			
		Value Label	N
TYPES2	1.00	ACN	36
	2.00	ACC	36
LAYERS	1.00	Low	24
	2.00	Medium	24
	3.00	High	24
Breathing Pattern	20 BY 20		36
	68 BY 20		36
AREA	10.00		24
	15.00		24
	20.00		24

TYPES2- Two ACF forms

TABLE D2: EXHALATION PRESSURE DROP STATISTICAL OUTPUT FOR TEST OF EFFECTS

Tests of Between-Subjects Effects					
Dependent Variable: EXHALATION PD					
Source	Type III Sum of Squares	df	Mean Square	F	Sig.
Corrected Model	372.150[a]	35	10.633	223.008	.000
Intercept	810.098	1	810.098	16990.614	.000
TYPES2	.611	1	.611	12.805	.001
LAYERS	42.336	2	21.168	443.972	.000
Breathing Pattern	305.169	1	305.169	6400.458	.000
AREA	4.389	2	2.195	46.028	.000
TYPES2 * LAYERS	.442	2	.221	4.634	.016
TYPES2 * Breathing Pattern	2.703	1	2.703	56.687	.000
TYPES2 * AREA	1.805	2	.902	18.924	.000
LAYERS * Breathing Pattern	11.653	2	5.827	122.207	.000
LAYERS * AREA	.534	4	.134	2.802	.040
Breathing Pattern * AREA	1.054	2	.527	11.049	.000
TYPES2 * LAYERS * Breathing Pattern	.066	2	.033	.696	.505
TYPES2 * LAYERS * AREA	.438	4	.109	2.296	.078
TYPES2 * Breathing Pattern * AREA	.644	2	.322	6.75 6	.003
LAYERS * Breathing Pattern * AREA	.146	4	.037	.767	.554
TYPES2 * LAYERS * Breathing Pattern * AREA	.160	4	.040	.837	.510
Error	1.716	36	.048		
Total	1183.965	72			
Corrected Total	373.866	71			
a. R Squared = .995 (Adjusted R Squared = .991)					

Between-Subjects Factors			
		Value Label	N
TYPES2	1.00	ACN	36
	2.00	ACC	36
LAYERS	1.00	LOW	24
	2.00	MEDIUM	24
	3.00	HIGH	24
Breathing Pattern	20 BY 20		36
	68 BY 20		36
AREA	10.00		24
	15.00		24
	20.00		24

TYPES2- Two ACF forms