

## **ABSTRACT**

JAFARI, MEHRAN. Structure and Filtration Properties of PLA Meltblown Air Filters. (Under the direction of Dr. Eunkyong Shim and Dr. Behnam Pourdeyhimi).

Among nonwoven technologies meltblowing is capable of producing self-bonded fibrous media with fiber diameter ranging between 2 and 5  $\mu\text{m}$  and unique characteristic of meltblown webs such as high coverage and low pore size makes it very well suited for filtration applications. Polypropylene is the most commonly used polymer in this technology as it combine great rheological properties and adaptability to the process with excellent physical properties. Rising concerns over environmental issues and sustainability of petroleum based has encouraged and facilitated application of biodegradable polymer in last decades. This study aims to address processability and structure of Poly (lactic) acid (PLA) meltblown media and investigating mechanical and electrostatic filtration performance to establish material-process-structure-property relationship of PLA meltblown filter media. Without a doubt fiber formation and web structure in meltblown process highly dependent polymer properties and processing conditions. PLA grades with different properties and different meltblowing systems were employed to establish relationship between structure and property relationship. Morphology of PLA fibers modified by incorporating nucleating agent to the polymer melt in the meltblown process. Mechanical and electrostatic filtration properties of PLA filter media and relationship between web structure and fiber properties on filtration performance analyzed. The last part of this research is dedicated to comparative study of filtration performance of PP and PLA by designing similar media structure and analyzing mechanical and electrostatic filtration and charge decay performances.

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Structure and Filtration Properties of PLA Meltblown Air Filters

by  
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## **DEDICATION**

This dissertation is dedicated to my parents Ghanbar Jafari and Marzieh Taheri, my lovely sisters Behnaz and Fatemeh and my brother Mohsen and my niece and nephew Elena and MohammadReza for their endless love, support and encouragement.

## **BIOGRAPHY**

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# **CHAPTER 1**

## **INTRODUCTION**

Nonwovens are highly engineered fabrics that can be defined as sheets or webs from directionally or randomly oriented fibers or filaments entangled together mechanically, thermally or chemically. Nonwoven technologies allows to produce fiber and web at much higher speed and lower cost than traditional textile processes. Structure and properties of nonwoven fabrics can be engineered to meet specific functions in automotive industry, medical apparel, and filter media (Russell, 2006).

Meltblowing is considered as one of the most significant developments in nonwoven production technology (Yu et al., 2015). In this process molten polymer is injected through micro-scale size orifices of a die. Molten polymer after emerging from the die are exposed to high-velocity hot air jets. Molten streams of polymer will be attenuated by the drag force imposed by air jets and create fibers with much lower diameter than die orifices diameter. Meltblown process is a one step process to produce fabric having fiber diameter ranging from 1-5 $\mu\text{m}$  (Albrecht, Fuchs, & Kittelmann, 2006; Dutton, 2008). Meltblown fabrics exhibit special properties such as high surface area per unit weight because of fineness and enormous amount fiber, excellent barrier properties due to low porosity and pore size distribution (Y. Lee & Wadsworth, 1990). These characteristics of meltblown fabrics make them ideal candidate for high quality air filtration media. Nowadays meltblown process is widely used to produce different types of filter media such as cartridge filters, cleanrooms filter, and face mask. Filtration is considered as the largest segment application of meltblown fabrics followed by disposable medical products, wipes, diaper, and feminine hygiene products.

There is no doubt that it is urgent to remove various suspended particles and contaminants from the air stream. Particulate matters such as microorganisms, fine dusts, and pollen are issue



of concern as they cause or contribute to serious illnesses (Kampa & Castanas, 2008). Providing clean air with minimum amount of contaminants has been always challenging from both academic and industrial point of view. According to filtration theories, improving filtration efficiency of filter media can be done by use of fine fibers in filters structure and applying electrostatic charge on filter media (electret filters). Finer fibers enhance filtration efficiency by improving mechanical filtration mechanism such as diffusion and interception but at same time finer fibers lead to higher pressure drop across the filter media. Imparting electrostatic charge into filter media increases filtration efficiency with no change in the structure (Barrett & Rousseau, 1998). Electret filters benefit from electrostatic capture mechanism by creating electrostatic field at vicinity of fibers acting on both neutral and charged particles an attracting them toward fibers (Van Turnhout, Adamse, & Hoeneveld, 1980). The stronger the electric filed the higher the capturing efficiency. Reliability and performance of electret filters significantly depends on chargeability and charge stability of fibers in filter media over lifetime of the filter. Therefore in addition to structure of filter media, electret properties of polymer should be considered. Many thermoplastic polymers are used in meltblown process to produce air filter media but among them, polypropylene (PP) stands as the most commonly used polymer in this process. Indeed, of PP properties such as low melt viscosity or fluidity and great physical and electret properties combined with low cost and versatility makes it perfectly adapted to meltblowing technology and air filtration application (Rungiah et al., 2017a). However the major drawback of PP and other conventional plastic materials is that they are petroleum based and not from renewable resources. Petroleum based polymer and their products are non-degradable and last for many years in the environment (Yu

et al., 2015). The non-degradability, non-renewability, emissions of greenhouse gases and the diminishment of petroleum resources in production process have limited the versatility and applications of these polymers. Concerns over environmental burden and sustainability associated with petroleum based materials and strong demand to improve relationship between environment and industry in recent decades has motivated many engineering and scientific efforts to develop and modify biodegradable and eco-friendly polymers. Polylactic Acid (PLA) is linear thermoplastic aliphatic polyester derived from 100% renewable resources such as starch and sugar which are biodegradable, recyclable, and compostable (Avérous, 2008; Drumright, Gruber, & Henton, 2000). PLA is considered as potential alternative to substitute petroleum based polymers. In addition to advantages regarding biodegradability of PLA, it exhibits excellent mechanical properties comparable to those of petroleum based materials. In the past because of high manufacturing cost application of PLA was restricted to medical applications such as tissue implants and surgical suture. By advance in technology and engineering of PLA and decrease in price combined with adaptability of PLA with current technologies and growing demand for green products has expanded application of PLA in different fields such as biodegradable textiles, technical textiles, agriculture and filtration (Vink et al., 2004). Lactide has two active isomers with different optical configuration, L-lactide and D-lactide, Figure 1. Stereochemical composition of PLA significantly affects its crystallization behavior and mechanical properties. Stereochemical composition can be controlled during polymerization to produce poly(L-lactide acid) (PLLA), poly(D-lactide acid) (PDLA), and poly(D, L lactide acid) (PDLLA). Ratio of D and L isomer affects crystalline behavior of PLA. PLA with more than 90% L-isomer tend to crystallize (Avérous, 2008).

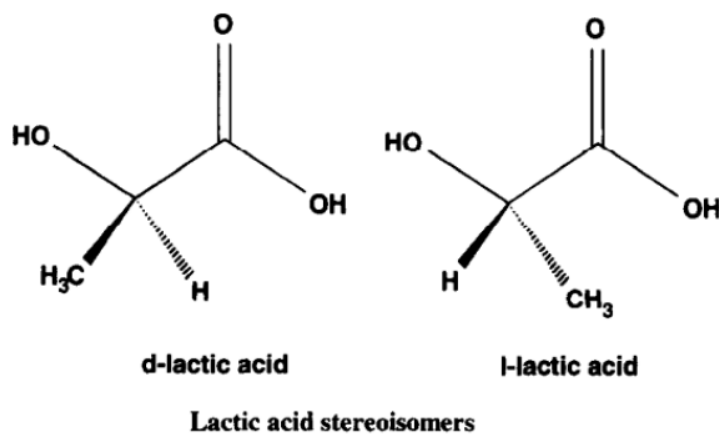


Figure 1-1 Configurations of two lactic acid isomers

PLA as a thermoplastic polymer can be used in meltblown process to replace petroleum based polymers such as PP. However, substituting PLA with conventional plastic polymers such as PP as a meltblown fabric faces some difficulties. Beside differences in physical properties, rheological properties of PLA is not fully adapted for meltblown process as and possess larger fiber size distribution and therefore lower solidity and larger pore size distribution (Rungiah et al., 2017a).

There is no extensive study on processability of PLA in meltblown technology and effect of material and processing parameters on fiber and web structure and filtration performance of PLA meltblown filter media. This study aims to address material-process-property relationship of PLA meltblown media and its air filtration properties. Without a doubt PLA properties such as chemical composition and melt viscosity would affect fiber and web properties and thus filtration performance. Fiber and web properties will be analyzed and the relationship between structure of fiber and web with mechanical and electrostatic filtration properties will be

compared. Effect of fiber morphology on electrostatic capture mechanism in terms of chargeability and charge stability of PLA filter by incorporation nucleating agent. Filtration performance PLA and PP media will be compared and analyzed to view possibility of replacing PLA with PP in air filtration application.

In the proceeding chapters, Chapter 2 contains literature review on filtration theories including mechanical and electrostatic filtration mechanism, different charging methods as well as PLA properties. Chapter 3 explains effect of melt viscosity and PLA chemical composition in terms of L/D ratio on fiber and web formation in Biax meltblown system and filtration performance of PLA meltblown webs. In chapter 4, effects of incorporating nucleating agent into fibers on fiber morphology and web structure and electrostatic filtration properties of PLA were investigated. Chapter 5 includes structure property relationship of low melt viscosity PLA in meltblowing process on conventional meltblown system and traditional Exxon die system. Chapter 6 compares structure and property of PLA and PP meltblown media along with their filtration performance. Chapter 7 recaps this research with a conclusion and recommendations for possible future works.

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## **CHAPTER 2**

### Literature Review

## 2.1 Electrets

In order to understand electrets first we should understand concept of dielectric materials. “A dielectric material (dielectric for short) is an electrical insulator that can be polarized by an applied electric field. When a dielectric is placed in an electric field, electric charges do not flow through the material as they do in a conductor, but only slightly shift from their average equilibrium positions causing dielectric polarization (“Dielectric,” 2016).” The polarization will orient positive and negative charge opposite toward together. An electret is a dielectric material possessing quasi-permanent electrical charge which means the time constants characteristic for the decay of the charge are much longer than the time periods over which studies are performed with the electret(Jasper, Mohan, Hinestroza, & Barker, 2007; Sessler, 1987). Based on their application dielectric materials can be classified as passive or active dielectrics. Application of passive dielectric materials is in insulation which are able to store charge for long time and the active dielectric materials applications includes control of electric charges by storing the charge and releasing them with the appropriate excitation, such as light or electric field.

According to the literature Gray (1732) was the first one mentioning the electret properties different dielectric materials like waxes and resins. More than a century later, Faraday (1839) studied and theorized electret properties as “dielectric which retains an electric moment after the externally applied field has been reduced to zero”(Kuzmany, Mehring, & Roth, 2012). In 1892, Heaviside came up with the word “Electret” to name dielectric materials after assuming permanent polarization charge on such materials and ever since then the term Electrets used universally. In 1919, systematic research conducted by Japanese physicist Eguchi. He



produced formed electrets by his own technique: the thermal method, and find out that charge on both side of electrets changed signs from opposite polarity “*hetero-charge*” to the same polarity “*homo charge*” to that of the electrode. In 1950’s, focus of research turned from thick plate of wax or similar substances to thin film polymers like PTFE and PVDF. In 1962, introducing the first polymer film electret microphones was a turning point in application of electret polymers and since then polymer electrets were used in sensors, transducers, and air filtration applications (Sessler, 1987).

Electret filters are fibrous filters consisting of dielectric materials with a quasi-permanent electrical charge(Hwang, Park, Bae, & Jung, 2014). Nowadays electret filters have wide range of applications including surgical face mask, air-purifying respirator, heat, ventilation, and air-conditioning filters, high efficiency particulate air filters, to name a few (Thakur, Das, & Das, 2014). The oldest type of electret filters is resin wool filter or Hanssen filter. It was invented by Hanssen in 1930s and it was the first military respirators filter. He found out that efficiency of respirator filters enhanced by mixing resin powder and wool fibers (J. C. Kim, Otani, Noto, Namiki, & Kimura, 2005). Years later Turnhout suggested use of electret fibers in air filtration and the product called as Filtrete™ which was charged differently and to a higher density. Bipolar and very stable charge of Filterete filters enabled them to have higher filtration properties (Van Turnhout et al., 1980). Polypropylene (PP), polycarbonate, polyurethane, and polyethylene are fibrous polymers with extremely low electrical conductivity and are commonly employed as air filter materials (Antoniou, Tabti, Plopeanu, & Dascalescu, 2010).

## 2.2 Electret Filters

Demand for air filters with high filtration efficiency and low pressure drop is growing as the air we breathe today is extremely polluted due to suspended particles and contaminants like beach sand, carbon black, pollen, coal dust, atmospheric dust, bacteria, radioactive fall-out, tobacco smoke, virus, diesel soot, pesticide, to name a few. Without a doubt some of these particles should be removed from the air we breathe since particles less than 1  $\mu\text{m}$  can go through inside human body and cause serious health problems. Most of air filter are composed of fibrous materials which are essentially made of either synthetic (polypropylene, polyethylene, polyester, polystyrene) or natural (cotton, wool) fibers. Fibrous filters capture particles through several mechanisms which are well-known and active in every fibrous filter.

Mechanisms are as follows:

- Interception
- Impaction
- Diffusion

Among all mentioned mechanism of filtration, Interception is the only one that can be named real collection mechanism. In this mechanism, a particle following the streamline gets captured by a filter fiber when it comes within one particle radius of the filter fiber, as shown in figure 2-1. Interception is one of the main mechanisms of filtration for particles between 0.1 and 1  $\mu\text{m}$ .



Figure 2-1 Interception in air filter media (NIOSH, 2003)

In inertial impaction mechanism, particles will no longer follow the streamline through fibrous media due to the High inertia, figure 2-2. This mechanism is main governing mechanism in capturing particles greater than 1  $\mu\text{m}$  (Naomi H. Harley & Lev S. Ruze, 2012).



Figure 2-2 Inertial impaction in air filter media(NIOSH, 2003)

The diffusion mechanism is based on the Brownian motion of the particles. Brownian motion is the random movement of particles in gas or liquid. Brownian motion governs the particle trajectory when the particle is extremely small compared to the fiber size. During diffusion, as shown in figure 2-3, particles do not follow their streamline, and have a random path. This random path is longer than the streamline path; so the probability of these small particles coming in contact with fiber surface is very high (Hinds, 1982; Naomi H. Harley & Lev S. Ruze, 2012; NIOSH, 2003). This diffusion mechanism is considered to be the main governing mechanism for particle less than 0.1  $\mu\text{m}$  in filtration process. Particles greater than 0.3  $\mu\text{m}$  do not have significant Brownian motion in air; therefore, their removal is not related to the diffusion mechanism(Naomi H. Harley & Lev S. Ruze, 2012).



Figure 2-3 Brownian diffusion in air filter media(NIOSH, 2003)

These mentioned mechanisms are referred as mechanical filtration mechanisms. Their overall performance in fractional efficiency could be seen in figure 2-4. Since effectiveness collection efficiency of mechanical filtration mechanism depends on particle size there is a minimum efficiency which is referred as most penetration size or MPPS. At this point which generally range from 0.1 to 0.3  $\mu\text{m}$  particles are too large to be captured by diffusion and too small for interception(Naomi H. Harley & Lev S. Ruze, 2012) .

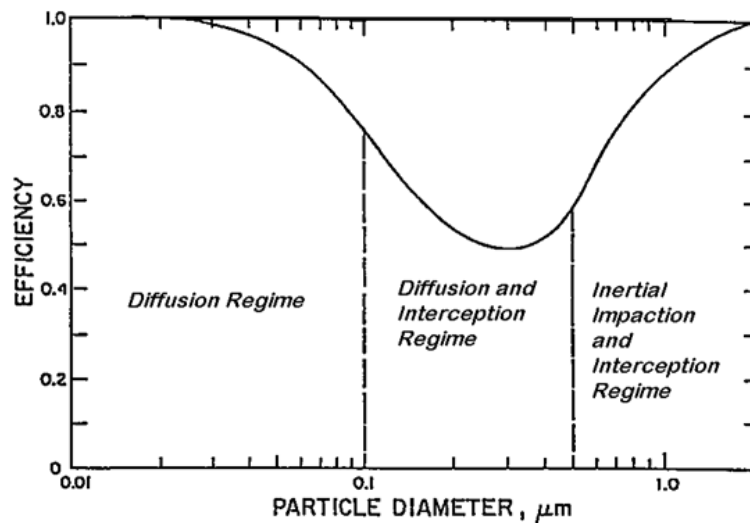


Figure 2-4 Fractional collection efficiency versus particle diameter for a mechanical filter (NIOSH, 2003)

Electret filter materials in addition to mechanical filtration mechanism benefit electrostatic filtration mechanism that attracts particles toward fibers. Electrically charged fibers in fibrous

filters enhance filtration efficiency by acting in two different ways on particles. It can attract particles by either Coulomb or polarization force acting on charged and neutral particles respectively. Charged particles will be attracted toward the fibers due to columbic force. If particles are not charged electric field generated by electrically charged fibers in their vicinity induces dipole in particles, Because of the nonuniform electric field across the particle, the particle becomes polarized, the magnitude would depend on particle volume, and polarized particles will be attracted toward filter media (Oh, Jeon, Jung, & Jung, 2002).

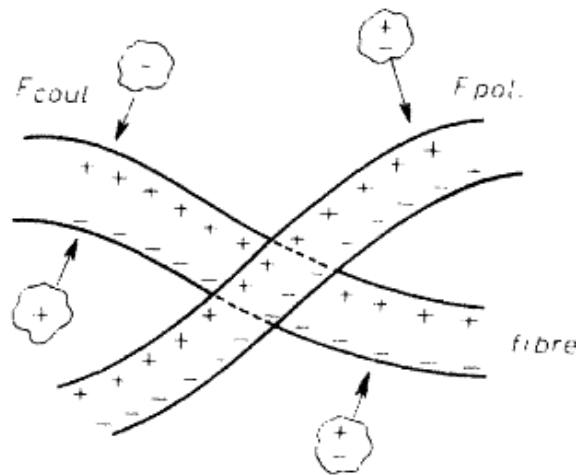


Figure 2-5 Two charged particles on left are attracted by Coulomb force, whereas two uncharged particles on right are converted into dipoles and attracted by polarization force (Van Turnhout et al., 1980).

Attraction of charged particles due to columbic force depends on the electric field on the surface of fiber which is directly related to the level and configuration of electret filters. The greater the charge, the greater will be the electric field produced and the particle collection efficiency by electric forces (Oh et al., 2002).

The advantage of electrostatic filtration mechanism over mechanical ones in capturing particle is that they can act on particle at quite some distance from fibers and so they can have much

opener structure in comparison non electret filter with same efficiency which mean less air resistance or lower pressure drop that is very important economically.

### 2.3 Performance of electret air filters

Filter performance is usually described and evaluated by their filtration efficiency which is the most important factor in filters. Based on application of filter, for achieving desired efficiency, type of filter media used in filter will be determined. In practice, filtration efficiency can be defined as the ratio of number of particles captured by filter to total number of particles (Leung, Hung, & Yuen, 2010)

$$E = \frac{N_{in} - N_{out}}{N_{in}} \quad (2-1)$$

Obviously higher filtration efficiency is more desirable. Beside filtration efficiency, pressure drop across the filter is another parameter used to characterize performance of filters. Pressure drop of filters directly affects energy consumption during filtration process because we always need to push air through the filter media. The pressure drop is defined by the difference of pressure in the upstream side of the filter and the pressure in the downstream side of the filter. Evidently, the lower pressure drop the better performance of filter. To consider effect of both filtration efficiency and pressure drop other indicator call Quality Factor, QF, is used. The quality factor of filter is defined as follows:

$$QF = \frac{-\ln[(1 - E)]}{\Delta p} \quad (2-2)$$

The  $QF$  is also known as the figure of merit and used in previous studies (Kalayci *et al.*, 2006; Wang *et al.*, 2008a, b).

In general, filtration efficiency of fibrous filters is determined by single fiber efficiency, geometry of fiber in filter, and internal structure of filter. Many scientists have proposed different theoretical and numerical models in order to predict filtration efficiency of filters. The theoretical aerosol penetration of particles through the filters is normally expressed in terms of single fiber efficiency (Hinds, 1982):

$$P = \exp \left[ \frac{-4\alpha\chi E_{\Sigma}}{\pi d_f (1 - \alpha)} \right] \quad (2-3)$$

Where  $\alpha$  is packing density,  $\chi$  is filter thickness,  $E_{\Sigma}$  single fiber efficiency,  $d_f$  fiber diameter. As discussed before the electret filter has advantage of electrostatic filtration and combines both mechanical and electrostatic filtration. As total filtration efficiency of electret filter can be expressed as a summation of mechanical filtration efficiency and electrostatic filtration efficiency:

$$E_{Total} = E_{Mechanical} + E_{Electrostatic} \quad (2-4)$$

Filtration performance due to electrostatic capture mechanism can be expressed as:

$$E_{Electrostatic} = E_s + E_o \quad (2-5)$$

Where  $E_c$  and  $E_o$  are single fiber efficiency for coulombic and dielectrophoretic capture mechanisms respectively. Therefore, for electret filter overall filtration efficiency can be defined as:

$$E_{Total} = E_{Mechanical} + E_s + E_o$$

By testing electret filters with neutral particles polarization responsible for capturing particles and single fiber efficiency would be combination of mechanical and dielectrophoretic efficiency. Therefore, the single fiber efficiency due to dielectrophoretic force can be obtained as (Romay, Liu, & Chae, 1998):

$$E_o \cong E_{Total} - E_{Mechanical} \quad (2-6)$$

Similarly, Coulombic force and mechanical mechanisms are effective in testing same filter with singly charged particles. Filtration efficiency will be combination of columbic force and mechanical filtration efficiency. Therefore, single fiber efficiency due to coulombic capture can be calculated as (Romay et al., 1998):

$$E_p \cong E_{Total} - E_{Mechanical} \quad (2-7)$$

Filtration efficiency of electret filters in addition to physical and structural properties of filter depends on charge characteristics of fibers. Charged media produced by triboelectrification has a higher charge density and filtration efficiency than corona charged media (Tsai, Schreuder-Gibson, & Gibson, 2002). Meltblown media has higher efficiency than spunbonded media mainly due to fiber size since the Columbic and dielectrophoretic effects are also known to be higher in case of smaller fibers as they have larger surface area and can carry more charges, providing stronger electrostatic attraction (Huang et al., 2013; Walsh & Stenhouse, 1996). As mentioned previously, filtration efficiency is also affected by structures of filter. Higher packing density of electret also indicates more filtering efficiency as mechanical filter but effect of packing density is more notable since higher packing density provides more amount of filtering materials and larger surface area which means higher charge carriers in the filter (Huang et al., 2013). Face velocity is another parameter affecting filtration performance



of electret filters. Generally face velocity affects aerosol particles  $< 1 \mu\text{m}$  since increase in face velocity results in shorter the retention time for particles which means lower chance of collision of particles with fibers due dominant filtration mechanisms in this size range, diffusion and electrostatic attraction (Huang et al., 2013; Wei, Chun-Shun, Cheong-Ki, & Chao, 2006). Also electret filters with higher thickness are known to have better efficiency. MPPS of electret filters tend to vary with increase in thickness since particles with high electrical mobility are more likely to be collected by the front layer of an electret filter. The particles penetrating the first layer would be those with lower electrical mobility and would more likely penetrate the following layer, thus providing the shift of the penetration curve and the MPPS change. Lowkis et al. reported that higher thickness and lower diameter leads to better filtration performance of polypropylene filters (Lowkis & Motyl, 2001).

As mentioned charge on fibers affects particle capture efficiency of filters. Huang et al. reported that with increase in fiber charge density aerosol penetration decreases remarkably (Huang et al., 2013). Wei et al. reported that smaller particles are affected by coulombic force and larger ones by polarization force and when fiber charge density is weaker penetration of smaller particles are lower than larger ones since coulombic force is dominant and when the fiber density is stronger penetration of smaller particles is higher than larger ones as polarization force and the inertial impaction and interception effects begin to dominate (Wei et al., 2006).

However, MPPS tends to decrease with increasing fiber charge (from  $0.3 \mu\text{m}$  to  $0.04 \mu\text{m}$  ( $\delta = 1.2 \times 10^{-4} \text{ C/m}^2$ )) and seems that excess charge of fibers is effective on particles large than  $0.04 \mu\text{m}$ , figure 2-6.

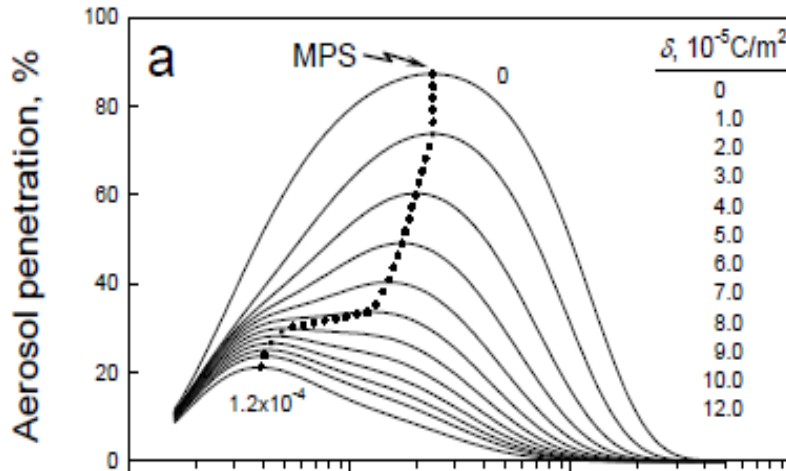


Figure 2-6 Effect of fiber charge density on aerosol penetration (Huang et al., 2013)

Filtration performance of electret filters is also highly affected by the physical properties of the particles. Particles penetration behavior could be affected by their charge density. Increase in charge density of particles results in lower penetration through the filter (Fjeld & Owens, 1988; Kanaoka, Emi, Otani, & Iiyama, 1987). And the effect of charge density is higher for smaller particles (Fjeld & Owens, 1988). Also, penetration through an electret filter declines as the dielectric constant of the aerosol increases since aerosol with a higher dielectric constant can be more easily polarized by the charged fiber of the filter (S. Yang & Lee, 2005).

Morphology of particle deposition on electret and non-electret filters studied by oh et al. comparing particles deposition on neural fibers and fibers with different charge density and also charged and uncharged particles with different sizes. They find out that for neutral fiber, small particles deposited on entire perimeter of fiber and for lager particles mostly dendrite deposition takes place. However, in case of electret fibers particles tend to deposit on entire perimeter of fiber and to be more uniform with the increase in fiber charge density, figure 2-7.

Walsh et al reported that lower face velocity would result in more uniform deposition of particles on electret filters(Walsh & Stenhouse, 1997).

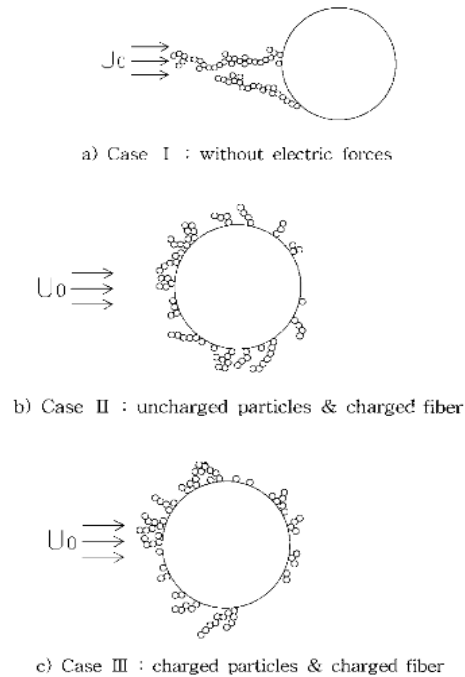


Figure 2-7 particle deposition morphology depending

There are two main common classifications for electret filters, one based on method which materials become electrically charged and the other is based on the nature of the charge (microscopic level) they are carrying. Electret filters based on charging process could be categorized as corona charged electret, triboelectrically charged electret, and induction charged electret. According to nature of carried charge by electret, they can be classified as real-charge electrets or dipolar-charge electrets, which charge in former one is embed or injected within electret for example exposing the material to a charged plasma or corona and in later one

permanent dipoles are formed by orientation of the polar groups at molecular level by heating material to higher temperature and cooling them in presence of external electric field.

## **2.4 Charging mechanisms**

### **2.4.1 Corona Charging**

Corona discharge is the most frequently used method for the electric charging of the filtration media. In general, corona charging techniques are considered as best and easiest ways of electret filter production (Fedel, 2012; Yahiaoui et al., 2013). The corona discharge is self-sustained, i.e., no external source of ionization is needed to maintain the current. Corona discharge method is consist of asymmetric electrodes such as a needle or a wire and a blunt electrode (called collecting electrode or counter electrode ) that high potential difference between the two electrodes would create Corona discharge phenomena. Electric field around stressed electrode would strongly reinforce and free electrons generated by natural background radiation will be driven toward anode. As a result, the air which is normally neutral becomes ionized and resulting ions are accelerated toward lower electric field. If an insulating polymeric fibrous material is placed on the receiver electrode then an electrostatic charge is developed on the polymeric fibrous material (Thakur, Das, & Das, 2013). Availability of free electrons plays an important role in ionizing the gas in vicinity of stressed electrodes since they can transfer all the energy achieved from electric field to the gas molecules. Corona discharge is one of the last steps in the air filter manufacturing process (Antoniou et al., 2010).

Corona process can be positive, negative, and bipolar based on polarity of electrodes. Different ions can be produced in positive and negative coronas which is highly dependent to the type of gas. In air, positive corona would generate hydrated ions with general formula of  $(H_2O)_nH^+$ ; n will increase by relative humidity. At lower rates of humidity other types of ions like  $(H_2O)_nNO^+$  and  $(H_2O)_n(N_2O)^+$  become dominant. In case of negative coronas based on relative humidity at 50% and 10% humidity,  $CO_3^-$  and  $(H_2O)_nCO_3^-$  would be dominant species respectively.

Figure 2-8 illustrates the positive corona process in a point-to-plane geometry. The ionization region is constrained to a small volume close to the discharge electrode. Remaining space between electrodes is called Drift Region which is distinguishing feature of corona discharge in comparison with other discharge forms. In this region, the ions and electrons drift in electric field without additional ionization as they have too low energy and density to ionize or react with other ionized particles (Goldman, Goldman, & Sigmond, 1985).

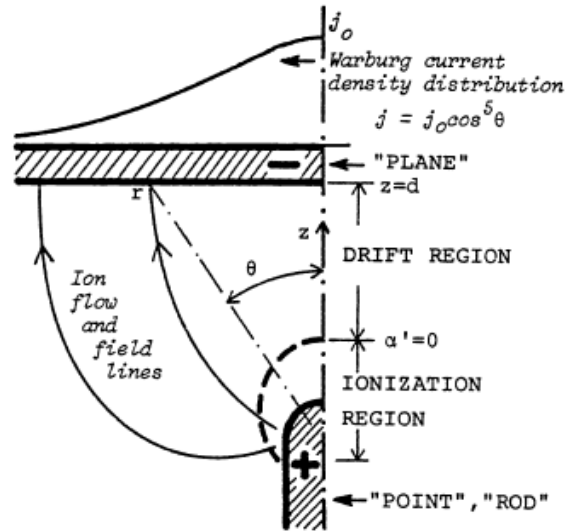


Figure 2-8 typical point-to-plane corona geometry (Goldman et al., 1985)

In the corona discharge method, the key process variables are known to be the amount and duration of the voltage applied to the corona electrode. Tabti et al. reported that with the increase in applied voltage the surface potential of the electret fibrous filters was initially remained unchanged, but once applied voltage surpasses a certain threshold, sudden increase in surface potential occurs, figure 2-9. The threshold voltage was reported to vary based on the charging polarity however for the same material and under identical environmental (Tabti, Mekideche, Plopeanu, Dumitran, Herous, et al., 2010).

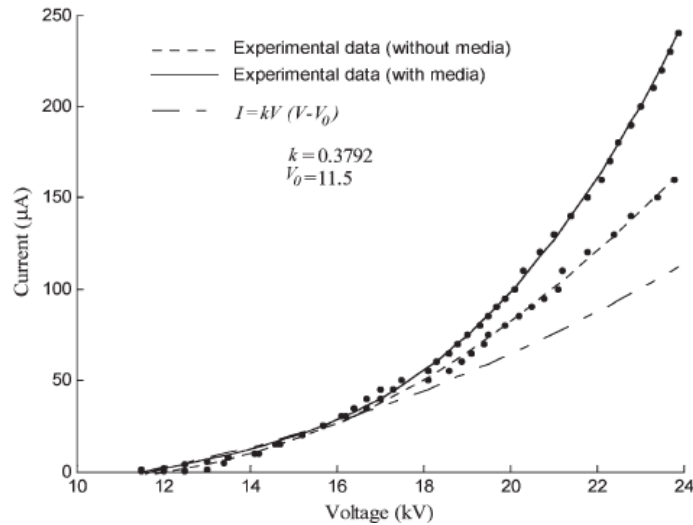


Figure 2-9 current- voltage characteristic of nonwoven media (Tabti, Mekideche, Plopeanu, Dumitran, Herous, et al., 2010) .

Das et al. experimentally investigated effect of applied voltage and charging time on surface potential of polypropylene meltblown nonwoven and found out that at higher applied voltage the rate of charge accumulation on the fabric was much higher (Das, Thakur, & Pradhan, 2012). Same result observed by Dascalescu et al for higher applied voltages causing higher initial surface potential. However applying higher voltages does not necessarily result in higher surface potentials charging quality (Dascalescu et al., 2010). Tabi et al. observed higher applied voltage leads to higher surface charge decay. Kacprzyk et al. reported that there is an optimum value for applied voltage which with increases in applied voltage the surface potential first increases to a maximum value and then starts to decrease (Kacprzyk & Mista, 2006). To reach to the maximum surface potential of electret filters determination of optimum voltage seems necessary. Higher charging time was found to be more favorable in order to get higher surface potential, Figure 2-10 (Das et al., 2012). But, Nifuku et al. observed that the initial surface

potential reached to saturation within a few seconds of charging time (Nifuku, Zhou, Kisiel, Kobayashi, & Katoh, 2001).

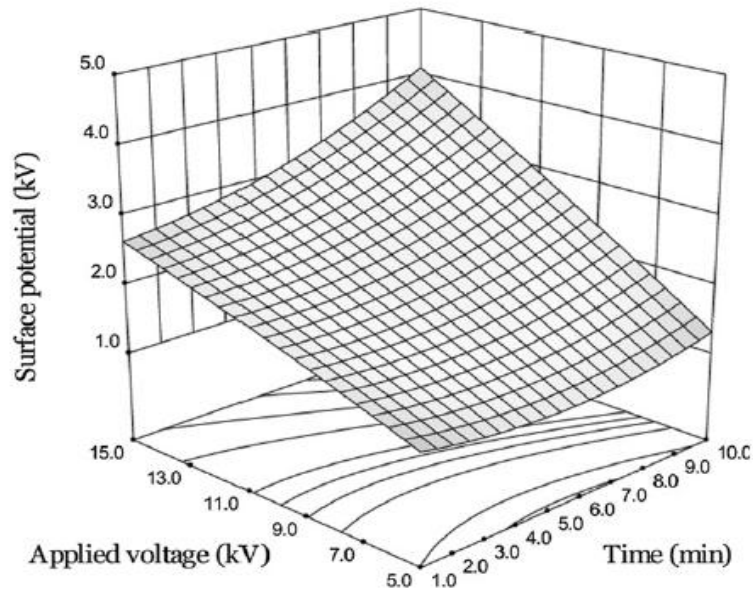


Figure 2-10 effect of applied voltage and charging time on surface potential (Das et al., 2012)

Temperature and heating condition of corona charging process affects initial surface potential and charge stability of nonwoven materials, figures 2-11, 2-12. These effects can be explained by two competing physical mechanism. At higher temperatures, the injected charge at surface of nonwoven media may travel to deeper traps. Charge mobility would increase by temperature which could increase chance of charge going deep into the media. In addition, reduction in moisture content of slows down the charge decay, as water increase the conductivity and favorites the neutralization of charge (Dascalescu, Plopeanu, Antoniu, & Mekideche, 2009; Tabti, Mekideche, Plopeanu, Dumitran, Antoniu, et al., 2010; Zhang et al., 2011).



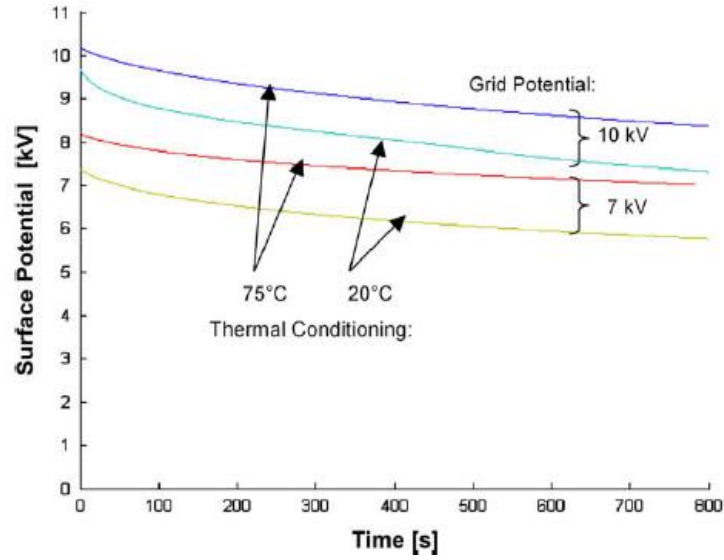


Figure 2-11 Effect of thermal conditioning on surface potential decay (Dascalescu et al., 2009)

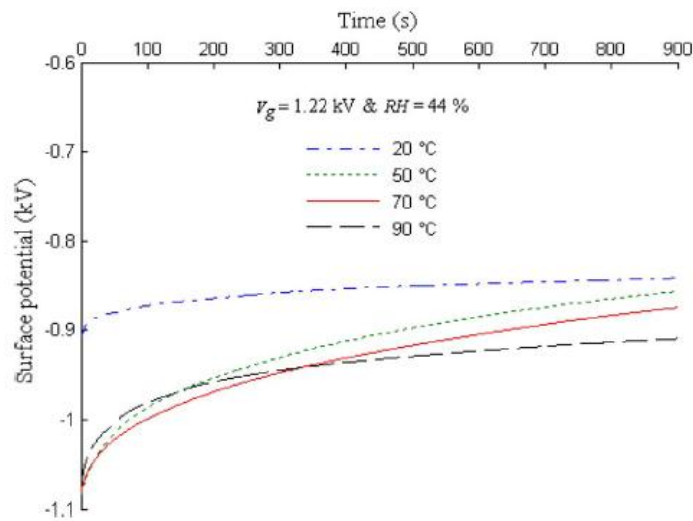


Figure 2-12 Surface potential curve of corona charged nonwoven fabric at different temperatures (Dascalescu et al., 2009)

Charging distance (Distance between emitting electrodes and filter media) also affects surface potential of filters. Das et al. observed that the higher distances between emitting electrodes

was, the lower was surface potential due to higher chance of charge loss in longer distances. Thakur et al. considered effect of electrodes distance in combination with time on initial surface potential and observed slight decrease in initial surface potential with increase electrode distance (Thakur et al., 2014). Further, they find out change in charging time cause slightly higher change in the initial surface potential. They conclude that generally higher applied voltage in combination with higher charging time and lower electrode distance is more favorable for higher initial surface potential for electret filters (Thakur et al., 2014). Short distances also can cause problems like spark generation and nonuniform charging.

#### **2.4.2 Triboelectrification**

Triboelectric charging involves the rubbing or intimate contact of two dissimilar dielectric materials. When two materials make contact with each other, some electrical charge transfers from one material to the other. One material gains an excess of electrons and becomes negatively charged and the other material loses an equal amount of electrons, and becomes positively charged (P. A. Smith, East, Brown, & Wake, 1988; Tsai et al., 2002). Tribocharged filter media are produced by processing a blend of fibers that have a large triboelectric potential difference. The fibers are brushed against each other, resulting in charged fiber surfaces (P. A. Smith et al., 1988).

When surfaces of different materials are triboelectrically charged, it is possible to determine which one of the pair becomes positively and which becomes negatively charged from triboelectric series (Park, 2000). The triboelectric series is arranged according to their tendency of electron accepting or donating characteristic. Materials with tendency to donate electron are at the top and it goes down progressively to the materials that accept electrons.

Table 2-1 Triboelectric series

a) +	Glass
more positive	Mica
	Polyamide (Nylon 6,6)
	Rock salt (NaCl)
	Wool
	Fur
	Silica
	Silk
	Aluminum
	Poly(vinyl alcohol) (PVA)
	Poly(vinyl acetate) (PVAc)
	Paper
	Cotton
	Steel
	Wood
	Amber
	Poly(methyl methacrylate) (PMMA)
	Copper
	Silver
	Gold
	Poly(ethylene terephthalate) (Mylar)
	Epoxy resin
	Natural rubber
	Polyacrylonitrile (PAN)
	Poly(bisphenol A carbonate) (Lexan,
	Poly(vinylidene chloride) (Saran)
	Polystyrene (PS)
	Polyethylene (PE)
more negative	Polypropylene (PP)
	Poly(vinyl chloride) (PVC)
- ↓	Polytetrafluoroethylene (Teflon, PTFE)

This charging is known to be composed of kinetic and equilibrium components. Energy dissipation that occurs when two dissimilar materials are rubbed together results in arise of the kinetic component. It basically leads to frictional heating at the area of contact and this leads to transfer of charges between the materials (Park, 2000).

Concept of electron exchange between polymer surfaces has been challenging topic among different researchers. Whitesides and McCarty refuted electron transfer in tribo charging since removal of an electron from non-ionic polymers such as polypropylene, polyester and polyethylene would require large amount of energy which rubbing action cannot provide that.

They proposed a model in which charging of fibrous structure would occur due to transfer of ions and not due to transfer of electrons. According to them, existence of interface of water in the form of moisture between polymers eases transfer of ions as electret fibrous filter charged by triboelectrification was made up of polymers which were non-ionic. In other research, Pence et al. considered effect of relative humidity on electrification. They observed increase in contact electrification till 40% relative humidity and after that there was no electrification. The decrease in contact electrification at high humidity was conducted to increased surface conductivity. In addition, there was no contact electrification at relative humidity of 0% which confirms role of water in triboelectrification of non-ionic polymers (McCarty & Whitesides, 2008).

### **2.4.3 Liquid Contact**

Charge generation through liquid contact is another method to produce electret filters. Liquid contact charging consists of treating fibrous webs with aqueous or non-aqueous polar liquid. In most of methods relating to liquid charging fibrous web is saturated with aqueous polar or dielectric liquid and then dried. In some methods there is pre wetting step which is believed to enhance the charge density of fibrous web and resulting in better filtration performance (Thakur et al., 2013). Figure 2-13 illustrates method introduced by Rousseau and Eitzman for fibrous web by aqueous polar liquid. As shown, the fibrous web is subjected to pre-wetting process by passing through series of rollers in a section containing wetting liquid. After that web is directed to next vessel containing an aqueous polar liquid and once the web is saturated with the liquid it could be send to drying system (Part 31 on figure 2-13). An important factor affecting quality of charging is surface tension of the polar liquid. In order to charge the web,

surface tension of aqueous web should be higher than surface energy of fibrous web. 1 to 10 seconds 1 second to 5 minutes are preferable times to contact with wetting liquid and aqueous polar liquid (Eitzman, Rousseau, Jones, & Angadjivand, 2002, WO 26778 A1, 2001).

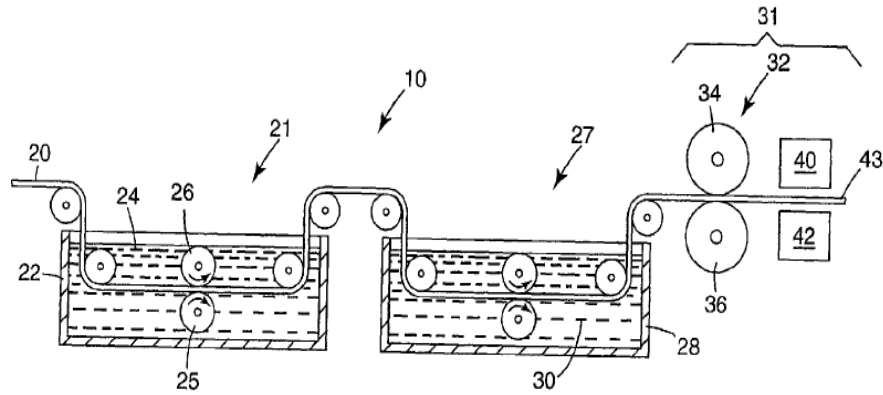


Figure 2-13 schematic of liquid charging process (WO 26778 A1, 2001)

#### 2.4.4 Hydrocharging

Hydro charging method which brought up by Coufal and Grygier in 1992 is a method of charging the polymeric dielectric media by striking a jet of fluid with at least velocity of 100 m/sec (Coufal & Grygier, 1994). Based on their patent interaction of high speed jet of fluid and the surface of dielectric material can create high electric potential. This method exploits tribocharging principles and tribocharging occurs when high velocity uncharged fluid impinging surface of dielectrics and net negative or positive charged is left on the surface. The maximum obtainable surface potential depends on many parameters like the dielectric surface conductivity and thickness as well as the fluids' velocity, dielectric constant, vapor pressure and electrical conductivity. Fluid could either be gas or liquid. Liquids are known to be more efficient due to their higher dielectric constant which results in more tribocharging. One year

later Angadjivand et al. disclosed a method of hydrocharging Polypropylene melt blown electret filters by using water high speed jets in hydroentangling process (Angadjivand, Jones, & Meyer, 1996). As it's shown in figure 2-14 hydroentangling water jets(part 12 on figure 2-14) provides water high pressure water jet impinging the fabric (part 10 in figure 2-14). They claimed that hydroentangling at pressure greater than 170kPa increased electret filter characteristics of meltblown polypropylene web. They also conducted a comparative study on effect of hydrocharging of corona treated webs and find out after corona treatment increases the Quality Factor when compared to webs treated only with corona charging (Angadjivand et al., 1996).

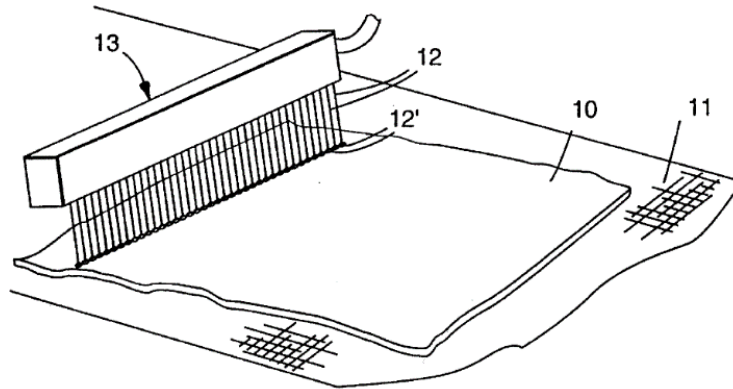


Figure 2-14 Hydro charging schematic (Goel, 2003) .

## 2.5 Evaluation of charge characteristics of electret filter media

Charge characteristics of electret filter are evaluated by several methods that are categorized in two groups: direct measurement and indirect measurement methods. Directs methods

measure surface potential, charge density, and electric field characteristics of fibers in filter media. Main direct methods used for evaluation of electret filter materials are thermally stimulated discharge current, electrostatic force microscopy, electrostatic voltmeters for surface potential measurements. Indirect methods monitor and evaluate the effect of presence of electrostatic charge on performance of materials such as filtration efficiency in filter media.

### **2.5.1 Thermally stimulated discharge current (TSDC)**

The technique of thermally stimulated discharge current (TSDC) has proved to be one of the most useful techniques in studying the charge storage and charge decay processes (Gun'ko et al., 2007; Mudarra & Belana, 1997)(Kumar, Pal, Kumar, & Ahirwar, 2014). Charge decay in this technique is studied by heating the electret at a constant rate instead of time since the charge decay measurement is time consuming at room temperature. Charges and dipoles are immobile at low temperatures in electrets and when they are heated they become mobile quickly and this way measurement time shortens considerably. In this method first electret is heated to a higher glass transition temperature,  $T_p$  (polarization temperature). An electric field is applied at this temperature which generates alignment of dipoles. Next step is rapid cooling of sample to the room temperature or below in the field which makes all the charges and dipoles frozen and immobile. The process continues by a short circuit for a few minutes and then reheated at a linear rate (1 to 3°C per min) and the generated discharge current is measured by an electrometer as a function of temperature, figure 2-15. TSDC plotted spectrum usually shows three or four peaks which are attributed to dipoles and space charges (Gun'ko et al., 2007).

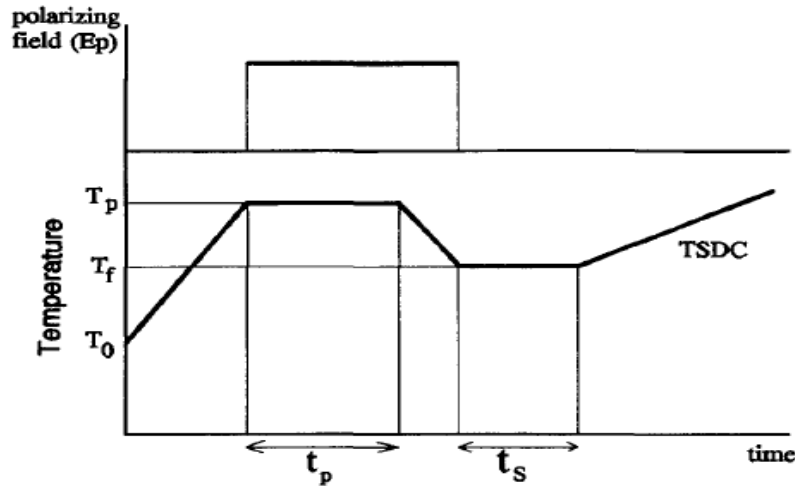


Figure 2-15 Sketch of TSDC measurements (Gun'ko et al., 2007)

### 2.5.2 Electrostatic Force Microscopy

Electrostatic Force Microscopy (EFM) is analytical technique of evaluation of electrostatic charge created by contact process and discharge in electret materials by providing local information of the electrical properties materials at the nanoscale such as surface potential and charge distribution (Kikunaga, Toosaka, Kamohara, Sakai, & Nonaka, 2011). In this technique, interaction of a conductive tip of instrument and sample surface maps the attractive and repulsive forces between them as electrostatic forces (coulombic, polarization, etc) affects the resonance frequency of the vibrating tip. In the EFM measurement process two scans are conducted to minimize the topographic effects on measurements. During first scan, by intermittent contact of tip with surface of sample topographical profile of specimen is obtained. Then for second scan the tip is set in certain distance from the sample surface and at this constant distance second pass of scan collects electrical information from the sample. During



second scan the tip is biased and parallel to the topography, figure 2-16, and electrostatic gradient images are generated by processing oscillation phase of vibrating cantilever(J. Kim, Jasper, & Hinestroza, 2006).

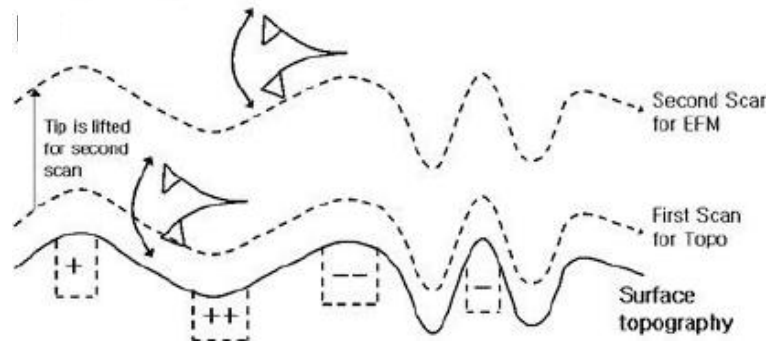


Figure 2-16 Scanning process in EFM technique.

Electric properties, tip geometry, and tip-surface separation distance influence tip-surface interaction during the measurement. The planar surface geometries are most ideal samples to be tested by EFM technique. In addition, in case of dielectric materials measurement is even more complicated since total force is contributed not only by the tip-induced polarization charges but also the surface/volume trapped charges (J. Kim et al., 2006).

### 2.5.3 Surface potential decay measurement

Experimental and theoretical studies have shown possible deterioration of polymeric materials when electric charges are accumulated on them. Lack of charge stability and charge decay over a period of time highly affects performance of electret fibrous filter media for long term applications. Charge decay can be monitored by measuring surface potential of electret filter media in certain intervals. Surface potential decay (SPD) measurement techniques now are widely used by researchers and industry to characterize electric charges on dielectric material

surface due to their reliability and low cost. Electrostatic voltmeter is mostly used to measure SPD in which the probe is adjusted in certain distance from fabric (usually several millimeters) The measurement of surface potential is common way to evaluate and monitor charge of electret filters. It is mostly observed with respect to time and actually the charge decay rate is the filter is being evaluated which is a key parameter in understanding the charge storage and transport in electrets. Several methods has been adapted in order to measure surface potential of electret filters (Goel, 2003).

## **2.6 Poly(lactic) acid**

Nowadays with huge advances in polymer technology, wide variety of synthetic polymers are available and used to produce different product ranging from water bottles to aerospace industry. Most of these polymers which are considered as raw materials for many applications are petroleum based in which human lives are dependent to polymers in many aspects. Conventional polymeric plastic material can stay in environment for very long time and do not degrade naturally and cause pollution. Also production of conventional plastic materials consumes fossil fuels which also adds CO<sub>2</sub> and other toxic gasses to the air we breathe.

In recent decades many research has been done in order to replace petroleum based polymers with eco-friendly and sustainable and degradable materials. These materials in addition to have the ability to degrade and compost in natural environment should meet requirements regarding the specific applications.

Poly (lactic acid) is an aliphatic polyester derived form 100 percent renewable resources such as corn and rice (Blackburn, Zhao, Farrington, & Johnson, 2006; E. Kim et al., 2009). WallaceH. Carother documented the first attempts of polymerization of lactides in 1932. Latter

on many research and studies was conducted in order to produce high molecular weight PLA to achieve adequate properties such as mechanical and thermal properties to be able to compete with conventional petroleum based polymers. In recent decades with rising environmental concerns need for bio friendly and bio degradable polymers in order to replace petroleum based polymers has led many research and studies to improve properties, polymerization and mass production of PLA.

In 1970's, application of PLA product in terms of direct contact by biological fluid got approved by Us food and drug administration (FDA).

PLA has many advantageous over conventional plastic material and also other bio polymers available in the market. Advantages of PLA are its renewability and availability from sustainable resources, biocompatibility and compostability, processability and energy saving as well as good mechanical and thermal properties (Garlotta, 2001; Tábi, Sajó, Szabó, Luyt, & Kovács, 2010; Xiao, Wang, Yang, & Gauthier, 2012).

PLA degradation happens by hydrolysis of ester bond with no need of enzymes and degradation rate varies based on environmental condition, PLA properties and size and shape of PLA product(Garlotta, 2001). PLA degradation products are H<sub>2</sub>O and CO<sub>2</sub> that are non-toxic and also do not cause any harm to human which makes PLA suitable for biomedical applications.

In terms of energy consumption, production of PLA consumes less fossil fuel than conventional plastic materials(Vink, Rabago, Glassner, & Gruber, 2003). Based on Association of Plastic Manufactures in Europe report on fossil fuel consumption of petroleum-based polymers, production of PLA requires 25-55% less fossil fuel energy indicating a high

potential in benefiting environment comparing to conventional plastics (Blackburn et al., 2006).

### 2.6.1 Chemistry of Poly(Lactic Acid)

PLA chemistry requires processing and polymerization of lactic acid monomer which is a simple chiral,  $\text{HOCH}_2\text{CHCOOH}$ , having two enantiomers, L- and D-lactic acid, figure 2-17. Proportion of these two enantiomers in the structure determines the properties of PLA and different ratio of LA and D isomers can result in different properties (Lopes, Jardini, & Maciel Filho, 2012). Therefore, wide range of PLA polymer can be produced to meet the requirements of the end use application (Gupta, Revagade, & Hilborn, 2007; Lim, Auras, & Rubino, 2008; Lunt, 1998).

L and D isomer difference can be detected by their effect on polarized light. In case of L isomer, the plane of polarized light is rotated in clockwise direction and D isomer is rotated in counterclockwise direction.

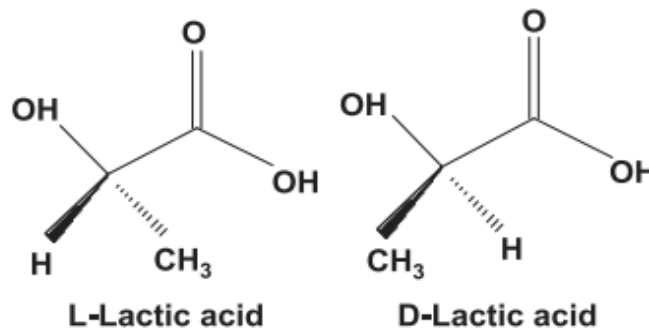


Figure 2-17 Optical isomers of lactic acid (Gupta et al., 2007)

## **2.6.2 Synthesis of PLA**

There are two major methods to achieve high molecular weight PLA in which lactic acid is used as a main substance, direct polycondensation and ring opening polymerization.

### **2.6.3.1 Condensation polymerization**

This method comparing to ring opening polymerization is cheaper and simpler in term of processing steps (Lopes et al., 2012). Polycondensation polymerization can be solution or melt polycondensation based on the solvent used in polymerization process. Solution polycondensation an organic solvent which is able to dissolve PLA without any other interaction. In this method water should be removed from process in order to reach high molecular weight. Molecular weight up to 300000 g/mol can be achieved by proper catalyst and condition (Xiao, Wang, Yang, & Gauthier, 2012) .

In melt polycondensation , the reaction can take place without organic solvent however it essential to keep the reaction temperature above melting point and under optimal condition molecular weight of 500000 g/mol can be reached (Moon, Taniguchi, Miyamoto, Kimura, & Lee, 2001).

### **2.6.3.2 Ring Opening Polymerization**

Ring opening is considered as the most common and effective route to achieve high molecular weight of poly lactic acid. In 1932 Carothers used this method to polymerize lactic, however the high molecular weights were not produced until DuPont in 1957 developed a technique to increase purity of lactide(Carothers, Dorough, & Natta, 1932; Garlotta, 2001).

Basically in this method a cyclic monomer will be propagated usually by a initiator such as ions or catalysts (Hu, Daoud, Cheuk, & Lin, 2016). In case of PLA cyclic dimer of lactide acid is the reactive center of propagation. By controlling the residence time and the temperatures

and proper catalyst type and concentration, desirable ratio and sequence of D and L lactic acid in the structure of PLA could be achieved and molecular weight could be controlled (Kricheldorf, Kreiser-Saunders, Jürgens, & Wolter, 1996; Lunt, 1998). Based upon the type of catalyst the polymerization process can be ionic, anionic, and coordination insertion and the polymerization can be processed in melt, bulk, or solution (Gupta et al., 2007; Lopes et al., 2012; Penczek, Duda, Szymanski, & Biela, 2000).

### **2.6.5 Crystallization behavior**

Crystallization in any polymeric system is very important because it affects structural formation of polymer chains and therefore properties such as mechanical and thermal (Zhai, Ko, Zhu, Wong, & Park, 2009). Therefore, it is necessary to understand crystallization behavior of PLA not only for crystalline structure but also for final mechanical and physical properties of PLA products.

As mentioned PLA has a chiral structure that allows to different composition and enantiomers namely D and L (Kulinski & Piorkowska, 2005). They both have the ability to crystallize but dimers of different chirality in the polymer structure limits the crystallization. PLA is known to have a slow crystallization rate comparing to other thermoplastics and PLA products show low crystallinity or amorphous structure and which limits its application in extrusion and injection molding that need to reach high level of crystallinity in short range of time (Shi, Zhang, Phuong, & Lazzeri, 2015). Based on literature, for pure sample of PLA crystallization half time takes around 17-45 minutes and it varies depending on temperature and molecular weight (Ren, 2011). In addition, degree of crystallinity of PLA highly depends on the ratio of

L and D enantiomers in the structure and can determine the polymer be either amorphous or semi crystalline (Lim et al., 2008). PLA with more than 90% L isomer in the structure tend to crystallize and less than that amount polymer will be amorphous (Shi et al., 2015). Even though presence of D isomer may affect degree of crystallinity, stereocomplex PLA, Having both L and D enantiomers, in comparison with pure PLLA or PDLA shows better thermal resistance and mechanical properties as well as higher melting temperature. The improvement is attributed to the formation of crystals in which PLLA and PDLA chains packed side by side within  $3_1$  helix.

Crystalline behavior of polymers as mentioned can play an important role in reaching certain properties. In addition to mechanical and thermal properties, crystallinity highly affects dimensional stability of PLA which is important for certain applications such as fibers and nonwoven materials. In case of fibers, shrinkage is related to crystalline and amorphous phase in the structure and the higher the amorphous phase is the higher shrinkage is going to be as in that region with stress and proper temperature polymer chains will retract and deform. (P. B. Smith, Leugers, Kang, Hsu, & Yang, 2001). It's been suggested that a stable crystalline units in the structure is needed to decrease or avoid shrinkage since it limits chains motion and their ability to shrink (Aou, Kang, & Hsu, 2005). Smith et al. also suggested that lower D isomer content in PLA structure would result in less shrinkage. Aou et al. proposed that in there is critical degree of crystallinity for PLA fibers, around 40%, which is needed in the structure in order to control and suppress shrinkage. Therefore, it is essential to understand and control crystallization of PLA fiber for end use applications (Aou et al., 2005).

In addition, crystalline structure is known to affect chargeability and charge stability of polymers which is crucial for electret applications such as air filter media. Charge trap and charge storage life time is influenced by morphology and structure of polymer. Crystalline and amorphous interface and boundaries is considered as deep traps that play important role in keeping charge in the polymer (A. A. Guzhova, Galikhanov, Kuznetsova, Petrov, & Khairullin, 2016). Charges also are trapped at defect in crystallites and impurities in the polymer structure. Increase in crystallinity and crystal size enhances chargeability and charge stability (Nath & Perlman, 1989).

Crystallinity of semicrystalline polymers such as PLA can be improved by addition of nucleating agent into structure since it enhances nuclei generation during crystallization process of polymers (Kawamoto, Sakai, Horikoshi, Urushihara, & Tobita, 2007). Regarding slow crystallization behavior of PLA it is shown that addition of nucleating agent can reduce the crystallization half time by more than one order of magnitude (Ke & Sun, 2003). Battezzato et al. studied incorporation of talc in PLA and concluded that addition of talc not only increased the crystallization rate of PLA but also improved the thermal and mechanical properties of PLA. Also the crystallization rate increased slightly with increase of talc content in the blends (Battezzato, Bocchini, & Frache, 2011). Ublekov et al investigated effect of effect of montmorillonite on crystallinity of PLA/montmorillonite nanocomposites and indicated that crystallinity increased at montmorillonite concentration higher than 5 wt% (Ublekov, Baldrian, Kratochvil, Steinhart, & Nedkov, 2012) . Concentration higher than 5 wt% . it is reported that application of other nucleating agents such as  $CaCO_3$ ,  $TiO_2$ , and also



improved crystallization rate of PLA (Buzarovska & Grozdanov, 2012; Liao, Yang, Yu, & Zhou, 2007).

### 2.6.6 Electrical properties of Poly Lactic (acid)

Polylactic acid is a linear aliphatic polyester and some of its physical properties such as electrical properties are similar to conventional polyesters (Urbaniak-Domagala, 2013). Electrical properties of poly lactic acid can be characterized by high resistivity, electric polarization and static charge.

In 2003, Shinyama and Fujita studied electrical properties of PLA and compared with general synthetic plastics such as PE, polyester and PP. Measurement was done at room temperature and based on their results PLA has higher volume resistivity ( $5 \times 10^{18}$ ) comparing to other synthetic polymers (Shinyama & Fujita, 2003) (table 2-2). They also investigated and compared the relative permittivity ( $\epsilon_r$ ), and dielectric loss tangent ( $\tan \delta$ ) of polymers. Results indicated that PLA has relative permittivity values between PE, PP, and polyester. Regarding  $\tan \delta$ , in comparison to PP and PE, PLA has higher values but smaller than polyester. According to the results they concluded that electrical properties of PLA is similar to those of other synthetic polymeric materials and have potential to be used as insulating materials (Shinyama & Fujita, 2003).

Table 2-2 Comparison of electrical properties of PLA, PE, polyester and PP (Shinyama & Fujita, 2003).

Items	PLA	PE	Polyester	PP
$\rho$ ( $\Omega \cdot \text{cm}$ )	$>5 \times 10^{18}$	$>10^{16}$	$>10^{16}$	$>10^{16}$
$\epsilon_r$	1 (kHz)	2.46~2.59	2.3	3.2
	100 (kHz)	2.43~2.65	—	—
$\tan \delta$	1 (kHz)	$2.6 \sim 3.0 \times 10^{-3}$	$2 \times 10^{-3}$	$5.0 \times 10^{-3}$
	100 (kHz)	$4.9 \sim 9.0 \times 10^{-3}$	—	—

In recent years, several researches have been conducted in order to investigate in order to examine dielectric and electrical properties of PLA for different application and in some cases modification has been done to improve electret behavior of Poly lactic acid.

Wieslawa investigated electrical properties of a low crystalline PLA film under DC filed with different range of temperature and tested their surface charge density for different intervals (up until 4 month after charging) concluded that PLA film polarized at electric filed with intensity larger than 16.7 MV/m showed acceptable electret properties, table 2-3, (Urbaniak-Domagala, 2013).

Table 2-3 surface charge densities of PLA

Side of sample	Time after terminating charging	Surface charge densities, $q_s$ , $\mu\text{C}/\text{m}^2$				
		$E_p = 8.3 \text{ MV/m}$	$E_p = 16.7 \text{ MV/m}$	$E_p = 25.0 \text{ MV/m}$	$E_p = 33.3 \text{ MV/m}$	$E_p = 41.7 \text{ MV/m}$
s1	$t = 1 \text{ h}$	+1.7	+2.6	+9.9	+20.6	+30.0
	$t = 1 \text{ day}$	-0.9	-1.7	+6.0	+15.6	+20.6
	$t = 4 \text{ months}$	-1.3	-4.3	+3.4	+4.3	+13.0
s2	$t = 1 \text{ h}$	-4.3	-4.3	-10.3	-20.6	-26.0
	$t = 1 \text{ day}$	-1.7	-4.3	-8.6	-13.0	-20.6
	$t = 4 \text{ months}$	-3.0	-6.6	-6.6	-6.0	-11.0

Guzhova et al. studied electret properties of PLA film treated with corona with positive and negative charging at two different temperature. They find out that the charge decay nature for both negative and positive corona charging follows almost same trend because of similar charge carrier traps. In addition, increase in temperature (above  $T_g$ ) enables PLA dipole groups to be able to move and ordered dipole break down and escape, figure 2-18. They also found out that PLA film treated with negative corona charging at 90 °C showed most stable surface potential values (A. Guzhova, Yovcheva, & Viraneva, 2015).

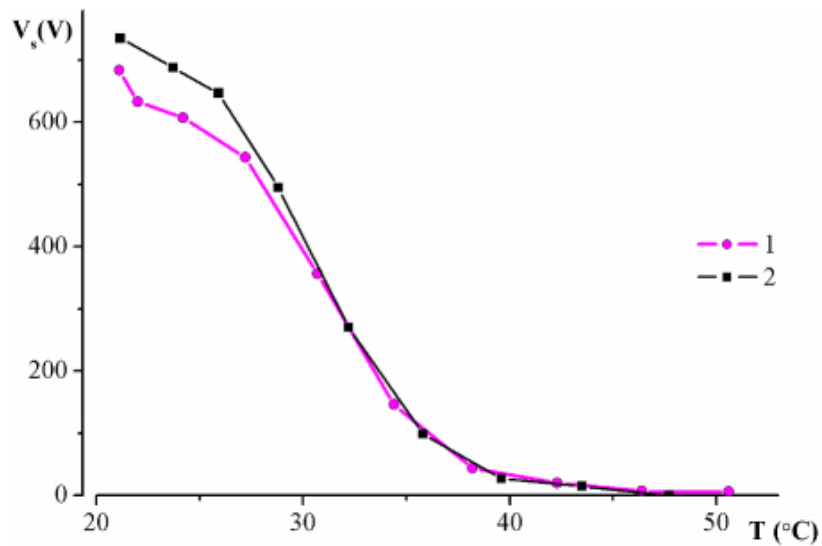


Figure 2-18 Surface potential vs. Temperature of thermally stimulated surface potential relaxation for PLA electrets films negatively (1) and positively (2) (A. Guzhova et al., 2015).

In another study, Guzhova et al investigated charge depth on corona charged poly(lactic) acid and used fine filler particles to improve electret properties of PLA. They explained charge depth behavior of PLA using of homo and hetero-charge distribution model in polymer corona electrets in which bulk polymer is divided to three levels (figure 2-19). According to this model, polarization of polar group in PLA molecule is dominant at first level because of surface oxidation during corona charging however at second level injected charges are dominant. At third level which is more depth in bulk polar groups become polarized by corona charging system but formation of polar group is dependent to mobility polymer macromolecules at the certain condition. They also used  $\text{BaTiO}_3$  in order to improve electret properties of polylactic acid. They concluded that addition of 2%  $\text{BaTiO}_3$  enhanced charged stability and shifted sharp

decay region since it can create new structural elements that can act as charge carrier traps (A. Guzhova & Galikhanov, 2015).

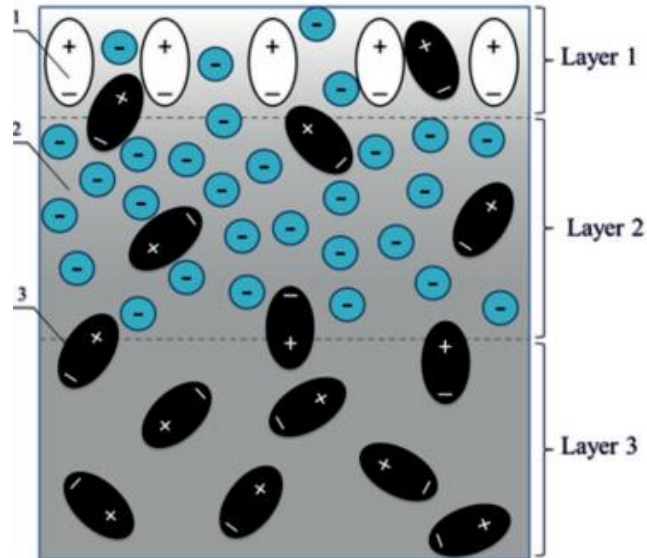


Figure 2-19 model of homo- and hetero-charge distribution in PLA (A. Guzhova & Galikhanov, 2015)

Several researchers such as Nakagawa et al. investigated electrical properties of polylactic acid in order to examine insulation characteristic of this polymer to be used as insulation materials. Nakagawa et al. compared electrical properties of PLA with XLPE and result showed that the volume resistivity and dielectric loss tangent of PLA were found to be almost the same as those of XLPE. Measured dielectric constant of PLA (3.0) was larger than those of XLPE (2.4) which could be attribute to polar carbonyl groups in PLA structure. Meanwhile this value is smaller than those values of PVC (3.5) which is currently being used for insulation electric wires. Impulse break down of PLA was 1.3 times higher (figure 2-20) than XLPE and space charge accumulation was one half (Nakagawa, Nakiri, Hosoya, & Tajitsu, 2004). However this

excellent insulation properties of PLA is highly temperature. PLA shows excellent electrical insulation characteristics up to 70 °C but after that temperature because of poor thermal resistance its insulation properties starts to deteriorate (Oi, Shinyama, & Fujita, 2012).

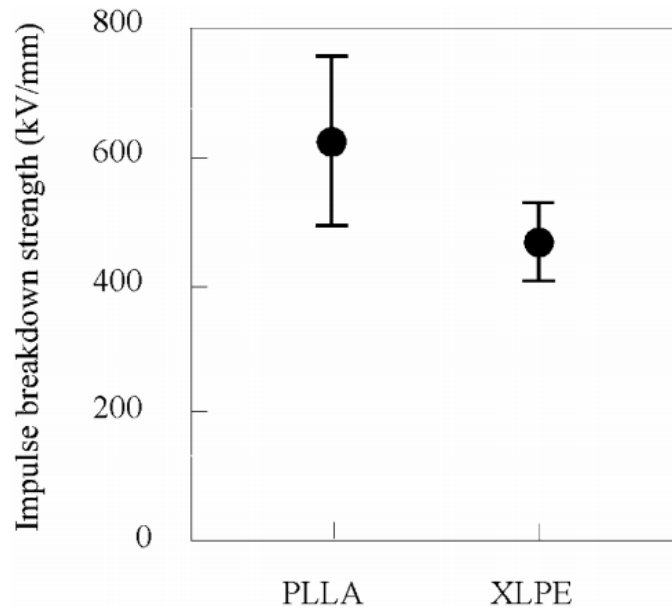


Figure 2-20 Impulse breakdown strength of PLA and XLPE (Nakagawa et al., 2004).

Effect of crystallinity on dielectric properties of PLA investigated by Hikosaka and his colleagues. PLA films with different crystallinity were achieved by heat treating them for different time intervals and conduction current was measured in each sample by applying DC electric field at 20, 50, 80°C. At lower temperatures (below  $T_g$ ) the conduction current did not affected by change in crystallinity but at 80°C in which polymer is in rubbery state (above  $T_g$ ) the conduction current become larger because of more free volume helping transportation of charge carriers, figure 2-23. Amorphous samples showed higher conductivity as in amorphous

regions molecular motion is higher and micro-Brownian motion assist transport of charge carriers (Hikosaka, Ishikawa, & Ohki, 2011)

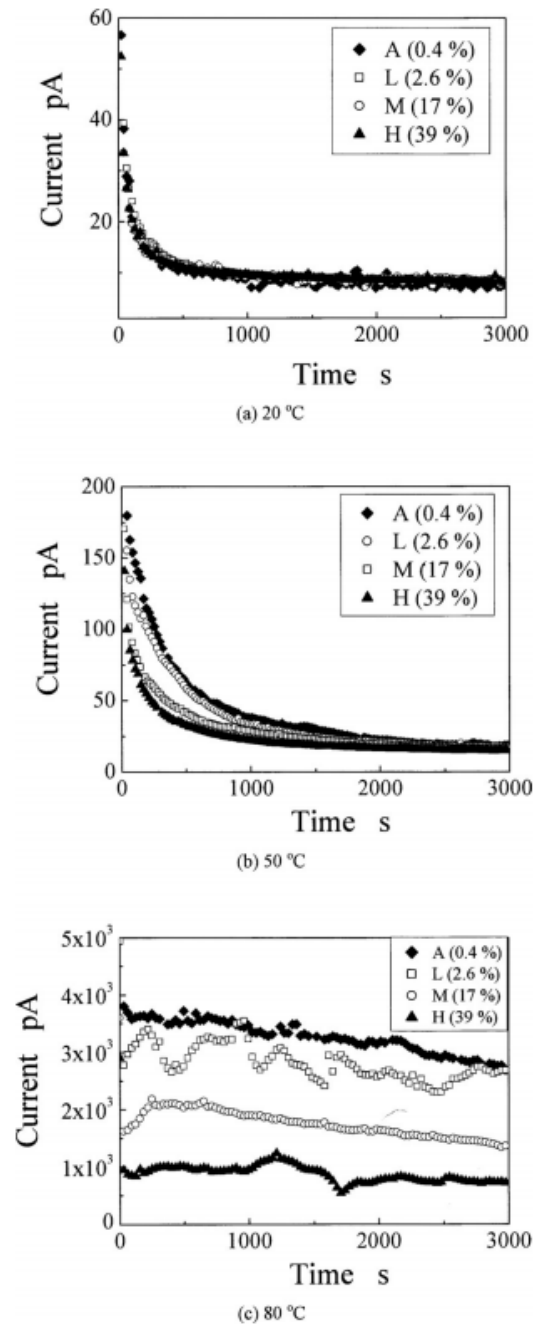


Figure 2-21 Effect of crystallinity on conduction currents at 20, 50, 80 °C (Hikosaka et al., 2011).

### **2.6.7 PLA meltblown nonwovens**

Nowadays PLA with different properties and molecular structure is available in the market. Natureworks LLC is considered as a leading company in the developing and innovating PLA polymer for wide range applications including medical packaging, textile and nonwovens. Regarding nonwovens, several grades of PLA with different properties such as melt flow index, L/D ratio, and molecular weight are developed and can be used in different nonwoven technologies. Meltblowing is considered as one the most advance nonwoven technologies in which polymer resins will be transformed to nonwoven fabrics in a one step process. In this process thermoplastic polymer resins will be melted and extruded through a die with micrometer size orifices. High velocity hot air will attenuate the extruded molten polymers right after coming out of die and form fine fibers ranging 1 to 5 $\mu$ m. These fibers will be collected on a collector where there is no need for bonding of fibers to make the nonwoven fabric since in the gap between tip of die and collector fibers get entangled because of the air turbulent caused by hot air stream (Bresee, Ko, & others, 2003; Dutton, 2008; Wehmann & McCulloch, 1999). Meltblown fabric are used in Filtration, medical, hygiene and wipes.

Few research has been done so far on PLA meltblown fiber and web. In 2001, Muller and Krobjilowski investigated processability of meltblown fabrics made of biodegradable polymers such as PLA (Muller & Krobjilowski, 2001). For a polymer to be process in meltblown system need to have low melt viscosity as molten polymer has to be extruded through micro-scale size orifices and suitable processing temperature. Unlike most of the petroleum based thermoplastics that can be processed in meltblown system application of biodegradable

polymers faces difficulties because of high melt viscosity (Muller & Krobjilowski, 2001). They found PLA was processable over wide range of temperature except over 230 °C because of thermal overload.

Liu et al. studied effect of meltblowing process parameters and web formation of PLA meltblown web (Liu, Cheng, & Cheng, 2010). By changing hot air temperature from 250°C to 290 °C they were able to change fiber diameter and create PLA meltblown webs with different structure in terms of porosity and thickness and tested their filtration efficiency. They found out that with increase in hot air temperature filtration efficiency decreased as it led to larger fiber diameter.

Yu et al. looked into property of PLA/Tourmaline blends and meltblown webs. Addition of tourmaline slightly increased crystallinity of PLA meltblown fibers. Results indicated that PLA meltblown samples with tourmaline had larger pore size and distribution. They also tested filtration efficiency of PLA meltblown webs and concluded that addition of tourmaline increased surface charge density efficiency of electret PLA filters (Yu et al., 2015).

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## **CHAPTER 3**

### **Effect of PLA Properties on Structure and Filtration Performance of PLA Filter Media**

### **3.1 Introduction**

PLA is the most common thermoplastic polymer used to produce biodegradable materials, derived from 100% renewable and sustainable sources such as corn, sugar, and vegetables. PLA degradation products are non-toxic with no harm to human (Avérous, 2008; Nampoothiri, Nair, & John, 2010). Raising concerns over environmental issues caused by conventional petroleum-based polymers have been driving force to develop and manufacture commodities from biopolymers. Among commercially available biodegradable polymers PLA is considered as the most promising alternative to conventional synthetic polymers as it exhibits good mechanical properties comparable to synthetic polymers produced from petroleum sources and facilitate application of PLA in current processing technologies.

Among nonwoven technologies, meltblowing is of most promising and cost efficient nonwoven process to produce fabrics with high barrier properties for filtration and separation with micro or nano sized fibers (Dutton, 2008). In this process, solid polymer is converted to fibers and fabric in one step process in which molten polymer will be extruded through micrometer size holes of a die and pushed toward the collecting drum or belt. Regarding raw material, PP is the most common polymer used in meltblown technology because of its compatible rheological properties with requirements of meltblowing system and great physical properties (Rungiah et al., 2017a; Yu et al., 2014, 2015). However the drawback for PP and other synthetic petroleum-based polymer is that they are not eco-friendly and come from non-renewable resources.

PLA is linear aliphatic polyesters and functional group of its monomers do not have internal plane symmetry. Moreover, PLA characteristics highly depends on its chemical composition (Chariyachotilert, 2011). PLA has two optically active isomers: L-Lactide and D-lactide. Depending on stereoregularity, three different types of PLA can be produced. These are L and D lactic acid and mixture of D-L lactic acid (Kolstad, 1996). It is established that PLA properties can be manipulated to a large extent by controlling the ratio and distribution of these two isomers in the structure. In general, commercially available PLA grades in the market are copolymers of poly(L-lactic acid) and poly(D-lactic acid). It is known that mixture of both L- and D-lactide acid provides better mechanical properties and thermal resistance than pure form each isomers. However, combination of both isomers in the structure leads to decrease in crystallinity (Auras, Harte, & Selke, 2004). Higher ratio of D-lactide in structure results in lower crystallinity as it interrupts regularity of crystal structure of poly(L-Lactide). To achieve semi crystalline structure more than 90% L-lactic acid in the mix is required (Auras, Harte, & Selke, 2004).

In the past application of PLA was limited to medical because of high manufacturing cost. However, by advance in technology and reduction in production cost PLA has attracted a lot of interest in manufacturing biodegradable textiles. Thermoplastic nature of PLA plus acceptable mechanical properties as well as biodegradability makes this polymer very interesting for fibers and nonwoven products such as disposable medical, hygiene, and filtration products.

Crystallization behavior of polymer play an important role on physical and mechanical properties as well as electret properties of a fiber. Therefore, the study of crystallization properties of PLA is important because crystallization affects not only physical properties but also the final characteristic of fibers for end use application such as air filtration. In this chapter to investigate effect of PLA properties on fiber morphology and fiber formation in meltblowing process, we looked into effect of chemical composition of PLA in terms of ratio of D and L isomer in the structure of PLA on crystallinity of PLA meltblown fibers on the lab scale Biax design meltblowing unit. Three grades of PLA with different D-isomer content in the structure and MFR were chosen to be processed in meltblown system and material-structure –property relationship studied.



## 3.2 Experiments

### 3.2.1 Material

PLA 6100 and 6202 with different melt flow index and D-isomer content kindly provided by Natureworks LLC (Minnetonka, MN) were chosen to investigate effect of PLA properties on processing and fiber production. Based on the technical information provided by the Natureworks LLC, PLA 6100 has the lowest D-isomer content in its structure. Table 3-1 summarizes properties of each PLA.

Table 3-1 Properties of different grades of PLA

Polymer	Melt index (g/10 min at 210 C)	Melt density (g/cm <sup>3</sup> at 230 C)	Specific Gravity (g/cm <sup>3</sup> )	Relative Viscosity	D-isomer content (%)
PLA 6100	24	1.08	1.24	3.1	0.5-0.7
PLA 6202	15	1.08	1.24	3.1	2.0-2.2

### 3.2.2 Melt blowing Process

Lab scale meltblown system designed by Biax FiberFilm Corporation at the nonwoven institute research laboratory of NC State University utilized to produce PLA meltblown webs. Biax meltblown die system was chosen because it is able to process high melt viscosity polymers due to special die design comparing to traditional dies. Figures 3-1 and 3-2 illustrate schematics of traditional dies and Biax design die. Unlike traditional dies (with V shape design), Biax die has multiples rows of small capillaries. Each capillary is surrounded by air holes attenuating the molten polymers to fine fibers. Extruding of higher melt viscosity polymers through micro-scale size holes will need higher feeding pressure to push the molten polymer to flow and in V shape die pressure cannot exceed the certain pressure as die may take apart. However, in Biax design die because of its flat design and multiple rows capillary processing and handling higher pressure comparing to traditional dies is possible.

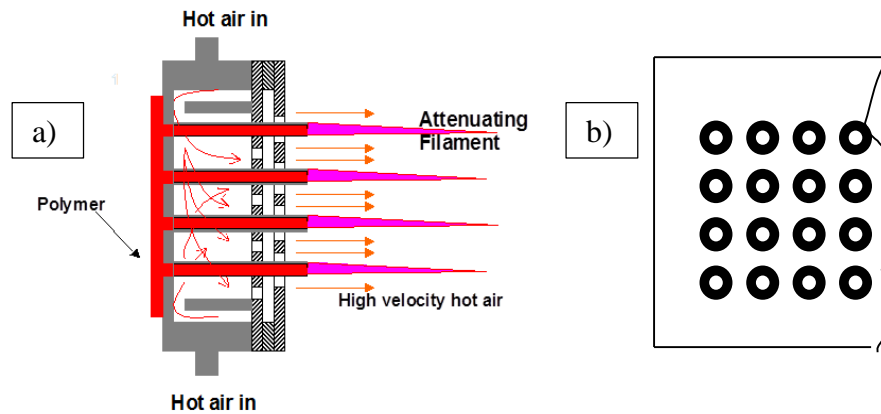


Figure 3-1 Schematics of Biax design die (Courtesy Biax Fiberfilm Corp). a) Cross section view and air distribution design b) Front view of Die.

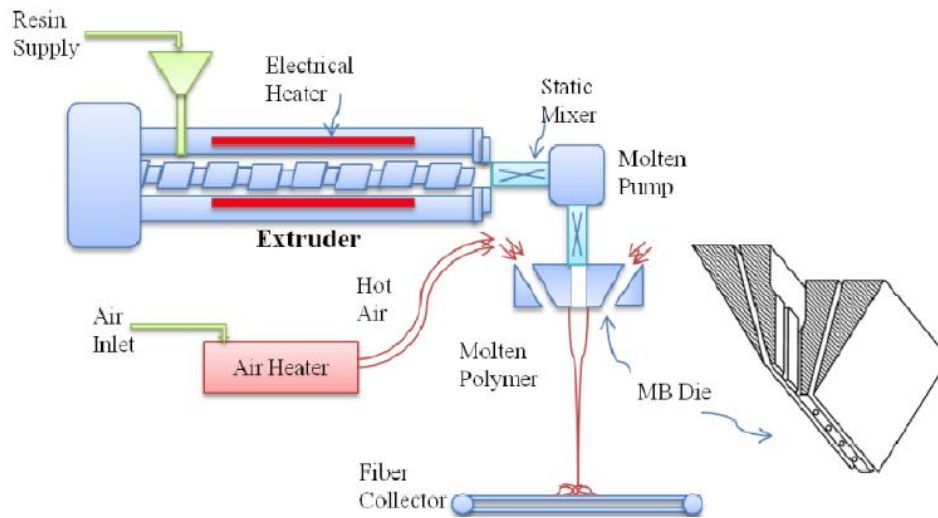


Figure 3-2 Schematic drawing of traditional meltblown system and die (not to scale)

At first step, PLA chips were dried for 8 hrs at 80 °C in a vacuum dryer in order to eliminate the moisture to minimize hydrolysis during the meltblowing. The meltblowing system has 15

inch die and 368 holes in capillaries. After drying, PLA chips were fed to the extruder which heated up to 200-270 °C in order to create webs and fibers with different properties. Processing conditions such as polymer throughput, air pressure and die to collector distance could be adjusted. Three levels of air pressure and 2 levels of extruder rpm were chosen to produce fiber with web with different structure. Produced webs collected on collecting drum at the die to collector distance of 25 cm and basis weight was kept constant 20 g/m<sup>2</sup>. Table 3-2 summarizes processing conditions in meltblowing system.

Table 3-2 Processing condition of PLA meltblown filter media

SAMPLE ID	POLYMER	PUMP RATE (RPM)	THROUGHPUT (GHM)	AIR PRESSURE (PSI)
F1	6100	50	0.2	8
F2	6100	80	0.3	8
F3	6100	50	0.2	11
F4	6100	80	0.3	11
F5	6100	50	0.2	14
F6	6100	80	0.3	14
P1	6202	50	0.3	8
P2	6202	50	0.3	11
P3	6202	50	0.3	14

### 3.2.3 Fiber diameter

Morphology and web structure meltblown fabrics were investigated by Scanning Electron Microscopy. Fiber size measurement carried out by Image J software and for each sample 200

random measurements were conducted in order to get average fiber size and fiber size distribution.

### 3.2.4 Differential scanning calorimetry

Crystalline structure of PLA polymers and fibers were investigated using DSC in temperature ranging from 10 to 190 °C. Crystallization behavior of all the samples carried out at 3 steps, heating, cooling, and heating. First in order to eliminate thermal history samples were heated to 190 °C at the rate of 10 °C/min then quenched to ambient temperature. At the third step samples heated again to 190 °C at the rate of 10 °C/min to investigate melting behavior. Thermal properties such as glass transition temperature ( $T_g$ ), melting peak ( $T_m$ ), cold crystallization temperature ( $T_{cc}$ ), and enthalpy of crystallization while heating ( $\Delta H_{cc}$ ) and melting enthalpy ( $\Delta H_m$ ) were determined. Degree of crystallinity is calculated by:

$$\chi_c(\%) = \frac{\Delta H_m - \Delta H_{cc}}{H_{100\%}} \times 100$$

Where  $H_{100\%}$  for PLA is 93 J/g (Ke & Sun, 2000).

### 3.2.5 Filtration properties analysis

PLA meltblown filters were exposed to 0.3  $\mu\text{m}$  monodispersed DOP (dioctyl phthalate) particles at face velocity of 5.3 cm/s. Two particle counters before and after filter count the number particles of and calculate filtration efficiency. Filtration efficiency is defined as:

$$E = 1 - \frac{C_{down}}{C_{up}} \quad (1-1)$$

Where  $C_{down}$  and  $C_{up}$  are the number of particles at filter downstream and upstream, respectively. Automated filter tester machine, TSI 3160, utilized to measure filtration performance in which certain particle size is generated by a collision type atomizer and a

classifier, classifies the particles based on their electrical mobility and after neutralization particles are fed into the filter holder.

### **3.2.6 Isopropyl alcohol treatment (Discharging)**

Isopropyl alcohol solution used to remove the electrostatic force from electret filters. Isopropyl alcohol does not change physical properties of the web and only removes the charge in media which can be confirmed by testing pressure drop before and after IPA treatment. All the samples were immersed in IPA solution for 2hrs and dried for 24 hrs. After discharging, mechanical filtration mechanisms will be only capturing mechanisms (Seeberger, 2011).

### **3.2.7 Corona charging**

Corona charging of PLA meltblown filters carried out at different charging voltage, charging time, and charging distance in order to determine the optimal charging condition. PLA meltblown samples were charged at three different level of 10 kV, 15 kV, and 22 kV for 5 sec, 60 sec, and 300 sec from an electrode distance of 5cm and 10 cm.

## **3.3 Result and discussion**

### **3.3.1 Thermal properties**

Heating and cooling curve of both PLA 6100 and 6202 are shown in figure 3-3 and 3-4. Thermal properties of heating process are summarized in table 3-3. PLA physical, thermal, and barrier properties are influenced by its solid state morphology and crystallinity. The glass transition temperature  $T_g$  of two polymers was around 59 °C. Figure 3-3 displays thermograms for the first cooling scan of the polymers. Exothermic peak only observed for PLA 6100. PLA 6202 higher D-isomer content exhibited no crystallization peak upon cooling because of very slow crystallization rate.

Table 3-3 Crystallinity and melting peak of different PLA grades

Polymer	$T_m$ (°C)	$T_g$ (°C)	$T_{cc}$ (°C)	$\Delta H_m$ (J/g)	$\Delta H_c$ (J/g)	Crystallinity (%)
PLA 6100	181	59.8	104	54.8	29.7	26
PLA 6202	172	59.4	116	32	27.3	6

Regarding first heating scan shown in figure 3-4, both polymers displayed cold crystallization exotherm with different cold crystallization temperature  $T_{cc}$ . Presence of the exothermic peak reflecting cold crystallization in both PLA meltblown fibers can be explained by nature of meltblowing process and PLA crystallization behavior. PLA is known to have a slow crystallization half time and for pure PLA it may take up to 17- 45 min to crystallize depending temperature, stereochemical structure and molecular weight (Harris & Lee, 2008). Meltblowing process is a fast process that in few seconds polymer melt is blown and laid down on a collector. As result due to Low crystallization rate and high cooling rate in meltblowing process polymer chains do not have enough time move and orient which is necessary for crystal formation. Hence as PLA meltblown fibers are heated above glass transition temperature unlocked and non-oriented polymer chains will be able to move and form crystals. Lack of orientation has negative effect on structural properties and dimensional stability. Comparing cold crystallization exotherms, PLA 6100 showed lower  $T_{cc}$  narrower peak wide indication better crystallization of PLA 6100. Degree of crystallinity can be determined by sum of enthalpies of cold crystallization and melting. Figure 3-4 shows that with decrease in L isomer (decrease in optical purity) melting peak becomes smaller. Endothermic peak associated with melting occurred at 167 and 176 for PLA 6202 and 6100, respectively.

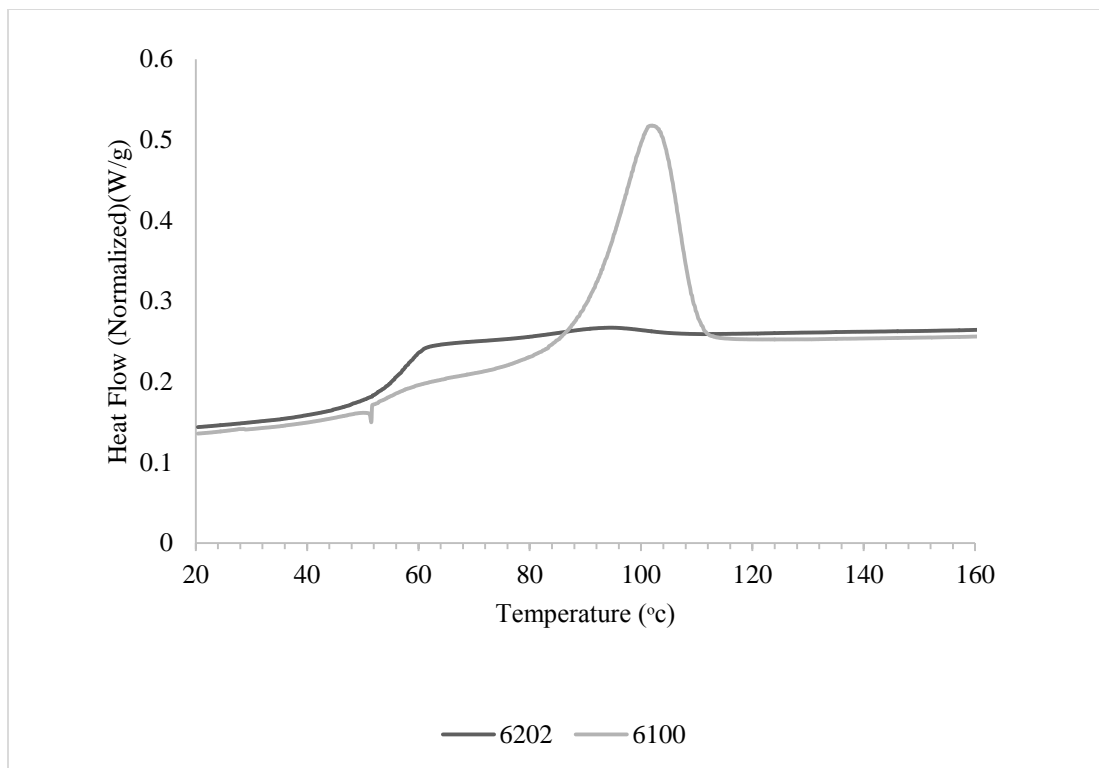


Figure 3-3 DSC Cooling thermogram of three different grades of PLA polymers

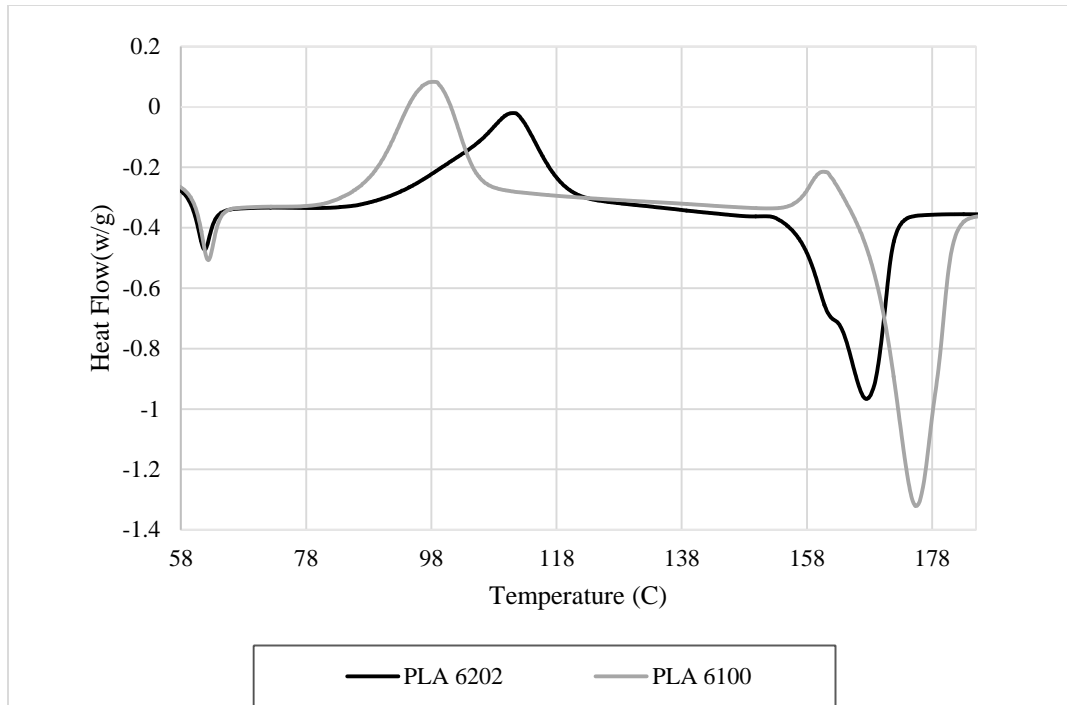


Figure 3-4 DSC heating thermogram of three different grades of PLA meltblown samples (50 rpm, 8psi, 25 cm DCD).

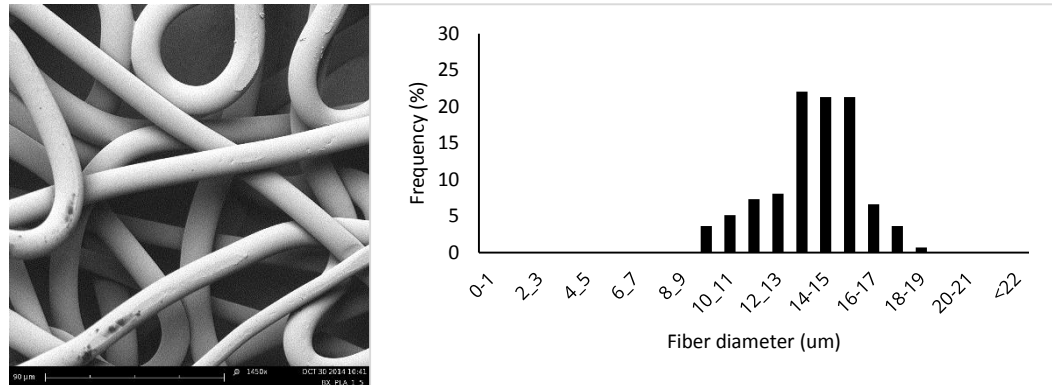
Regarding the rapid cooling rate and quick lay down process, polymer with higher crystallization and faster initiation rate will result in fibers with higher crystallinity.

### 3.3.2 Fiber size distribution

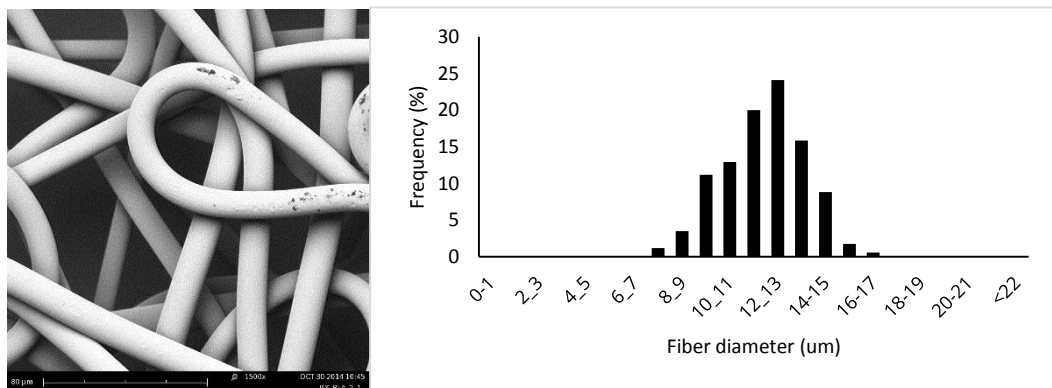
Fiber size can be controlled by meltblowing processing parameters. In order to see effect of processing condition on fiber size in meltblowing process we changed air pressure and extruder rpm rate. Three levels of air pressure psi and two level of extruder rpm, 50 and 80 rpm were used in the manufacturing process but 80 rpm was only achievable for PLA 6100 which had the higher MFR.



Figure 3-5 shows SEM images and fiber size distribution of PLA meltblown fibers with same processing condition. Fiber sizes for both polymers are relatively large comparing to typical meltblown fiber (1-5  $\mu\text{m}$ ) because of melt viscosity.



(a)



(b)

Figure 3-5 SEM images of all PLA meltblown fibers with fiber size distribution, a) PLA 6202 b) 6100 Processing condition 50 rpm, 11 psi.

Figure 3-6 shows effect of air pressure on average fiber size. As mentioned three levels air pressure utilized to see how fiber diameter could change with varying air pressure. For both

polymers, average fiber diameter decreased with increase in air pressure. Air pressure determines the amount of attenuation force of fiber since it affects the air velocity applied to the fibers. The higher air velocity the higher the air drag force on fibers and thus fiber diameter will decrease.

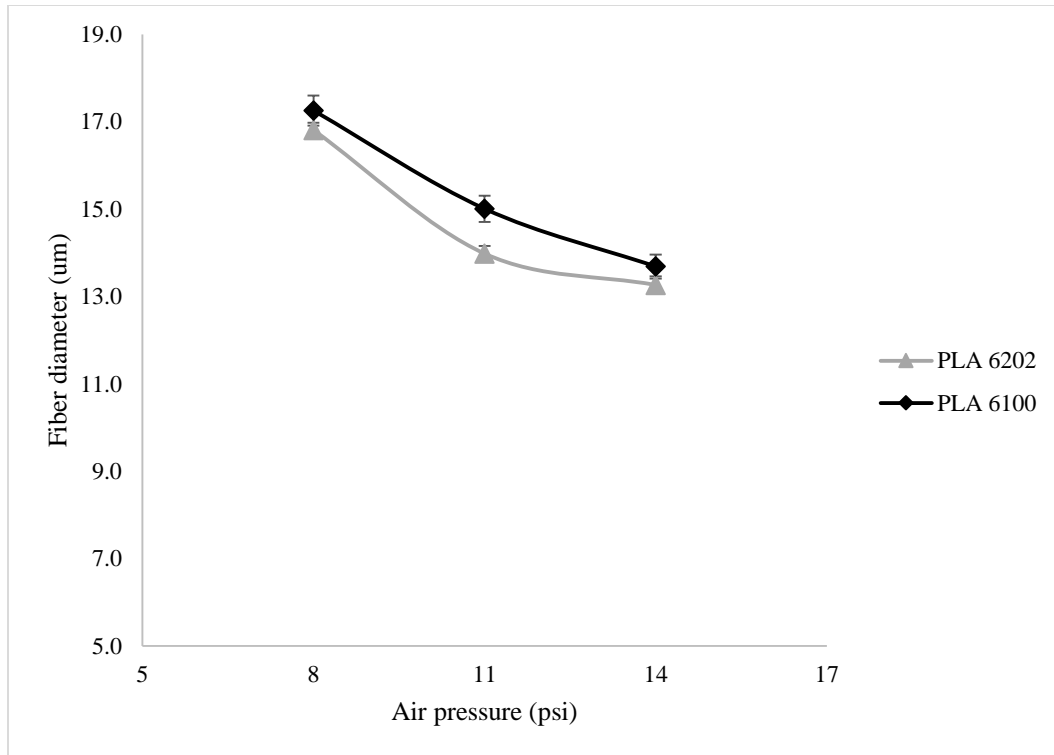


Figure 3-6 Effect of air pressure on fiber diameter (throughput: 6100: 0.3 ghm, 6202: 0.3 ghm,).

Figure 3-7 shows effect of extruder rpm on average fiber diameter. Increase in extruder rpm mean increase in amount of molten polymer fed into the system (higher throughput). As mentioned because of rheological and process restriction changing extruder rpm (throughput) was only possible for PLA 6100. As expected with increase in throughput average fiber

diameter increased by approximately 30%. With lower throughput attenuation on the fibers increases as same air drag force will be applied to lower amount of molten polymer and fiber diameter decreases.

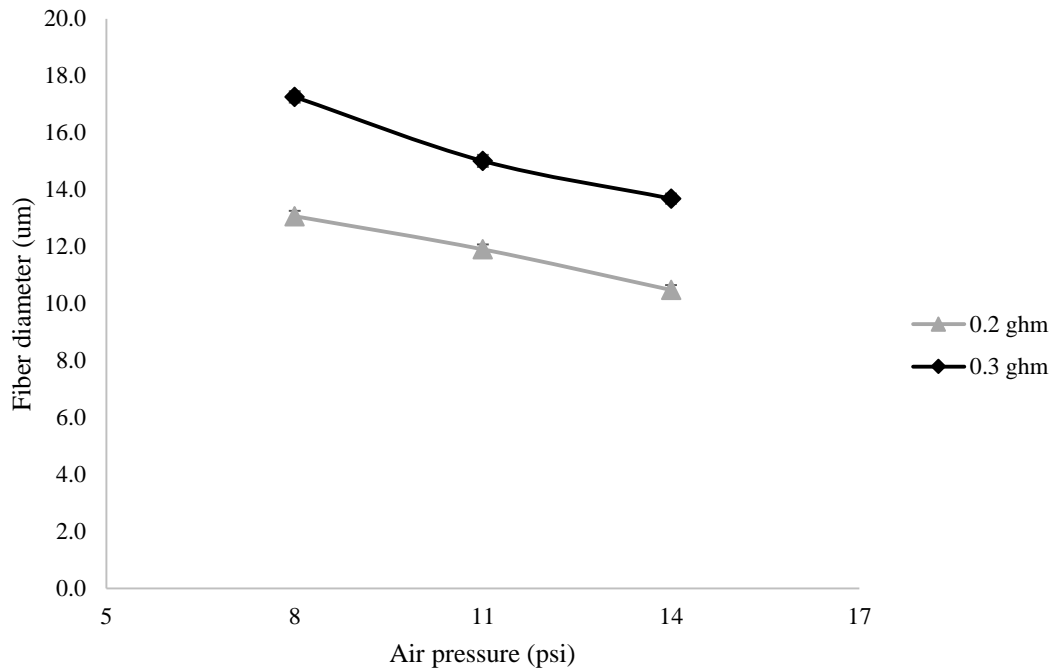


Figure 3-7 Effect extruder rpm on fiber diameter of PLA6100 meltblown fiber.

### 3.3.3 Filtration property of PLA meltblown webs

Figure 3-8 displays filtration performance for as received PLA 6202 meltblown filters with different fiber diameter and thickness. As indicated previously, with increase in air pressure in meltblowing process fiber diameter decreases. Smaller fibers will enhance mechanical filtration mechanisms with increase in fiber surface area as well as compactness filter media which results in higher chance of collision between fibers and particle. Major drawback of

finer fibers is increase in pressure drop. In addition, with increase in filter thickness there will be more fibers in the structure and therefore more sites for particles to be deposited and as a result filtration efficiency increases. Figure 3-8 shows that for all the samples with different fiber diameter, filtration efficiency result are very close and mainly differ in pressure drop. Filters with smaller fiber size in the structure are supposed to have higher filtration efficiency as well as higher pressure drop. Results are indicating use of lower fiber diameter in the filter media did improve filtration efficient and imposed higher-pressure drop instead.

Table 3-4 physical properties of PLA 6202 filter media

Sample	Polymer	Fiber diameter ( $\mu\text{m}$ )	Solidity
P1	6202	17	0.04
P2	6202	14	0.042
P3	6202	13	0.054

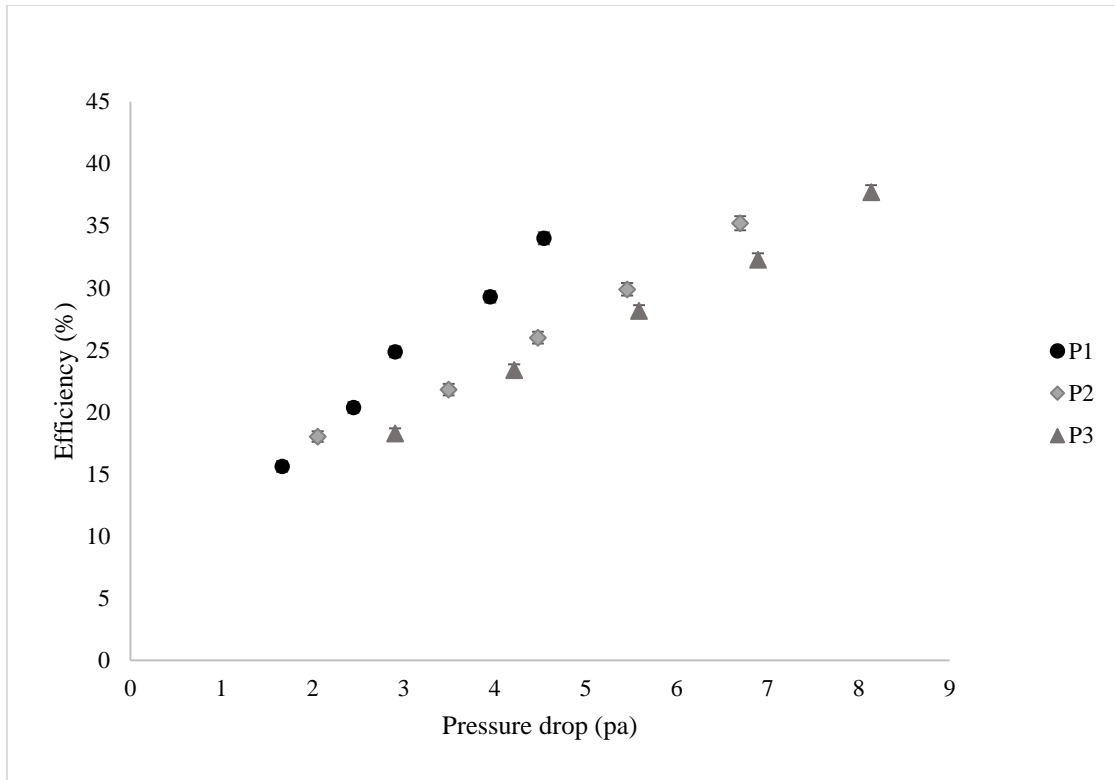


Figure 3-8 Filtration efficiency vs Pressure drop of filters with different thickness and fiber size.

Table 3-5 summarizes properties of different PLA 6100 filter media. As in production different extruder rpm (50 and 80 rpm) was possible for PLA 6100 polymer, meltblown webs with different fiber diameter at constant air pressure in processing condition obtained. Filtration performance of PLA 6100 filter media is show in figure 3-9.

Table 3-5 physical properties of PLA 6202 filter media

Sample	Polymer	Fiber diameter ( $\mu\text{m}$ )	Solidity
F1	6100	13.1	0.040
F2	6100	17.3	0.049
F3	6100	11.9	0.069
F4	6100	15.0	0.048
F5	6100	10.5	0.081
F6	6100	13.7	0.049

F2, F4, and F6 samples with throughput have larger average fiber sizes in their structure. Comparing the filtration performance of Sample F2 with highest average fiber diameter and F5 with lowest fiber diameter it can be seen that filtration efficiency result are almost similar to each other but differ in pressure drop dramatically. For PLA 6100 filters as well as PLA 6202 filters with larger fibers size have almost same filtration efficiency as filters with smaller fiber size but they have lower pressure drop.

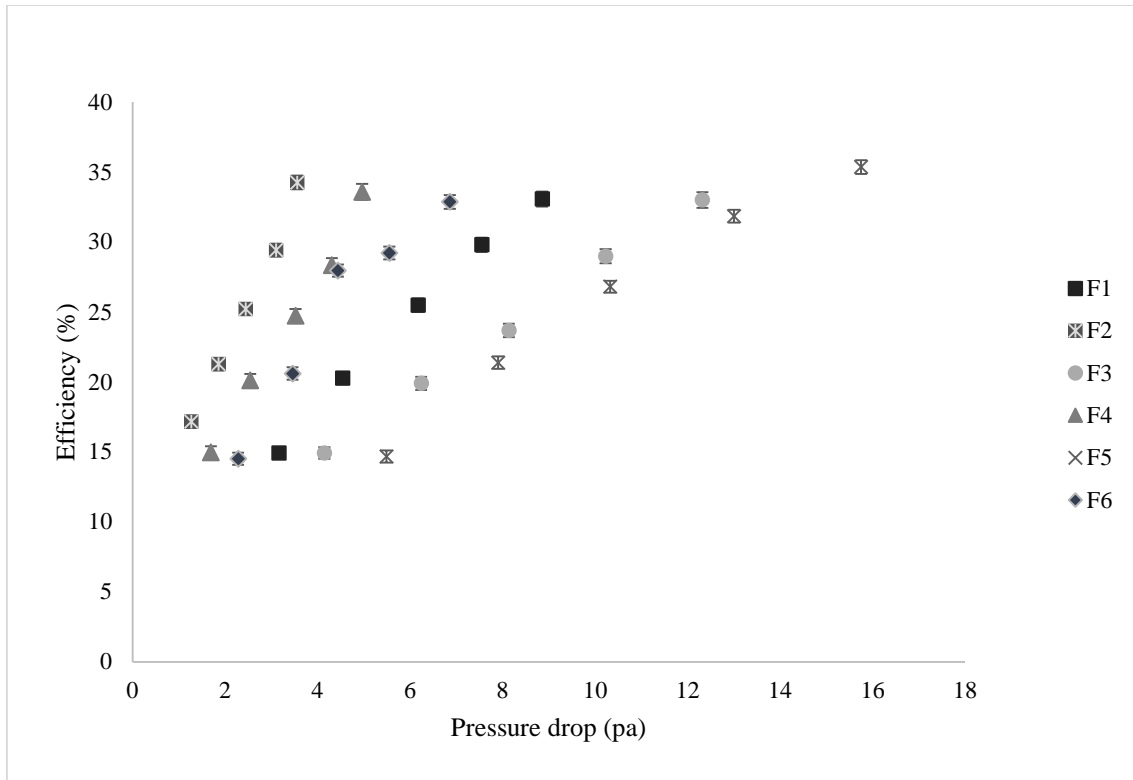


Figure 3-9 Filtration efficiency vs Pressure drop of filters with different thickness and fiber size (PLA 6100)

The difference is more clear when quality factor of filters are compared. Figure 3-10 compares filter quality factor of as received PLA 6100 filter media with different through put. As filters have almost same filtration efficiency according to equation (2-2) filter with lower pressure drop will have better quality as a result filters with larger fiber have much better quality factor. Since results do not comply mechanical filtration principles (described in section 2-2), in order to see the high efficiency for filters with larger size is purely because of mechanical filtration efficiency, filter media were discharged to eliminate possible electrostatic charged in fibers. Presence of electrostatic in fiber increases filtration capture efficiency by acting on both charged and neutral particles.

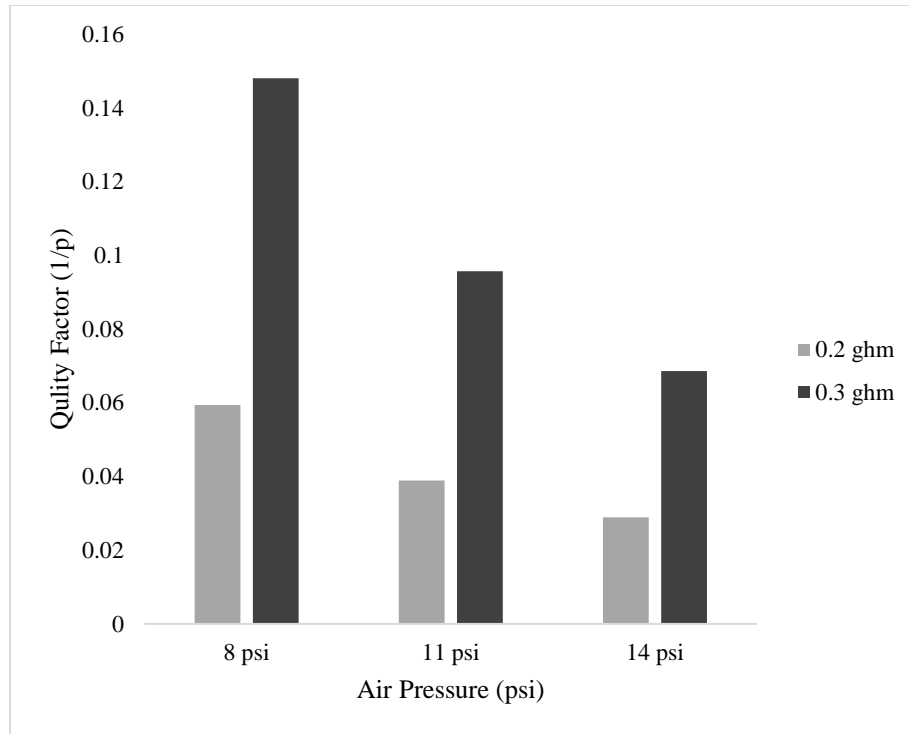


Figure 3-10 Filter quality factor of PLA 6100 meltblown media.

Filtration performance of discharged samples are shown in figure 3-11. Filtration efficiency drop after discharging shows that in addition to mechanical capturing mechanism, electrostatic capture mechanism affected filtration performance of as-received PLA meltblown media. Since there has not been in any external electrostatic field to apply charge on filters, PLA fibers are getting charged in the middle of meltblowing process possibly because of triboelectrification .



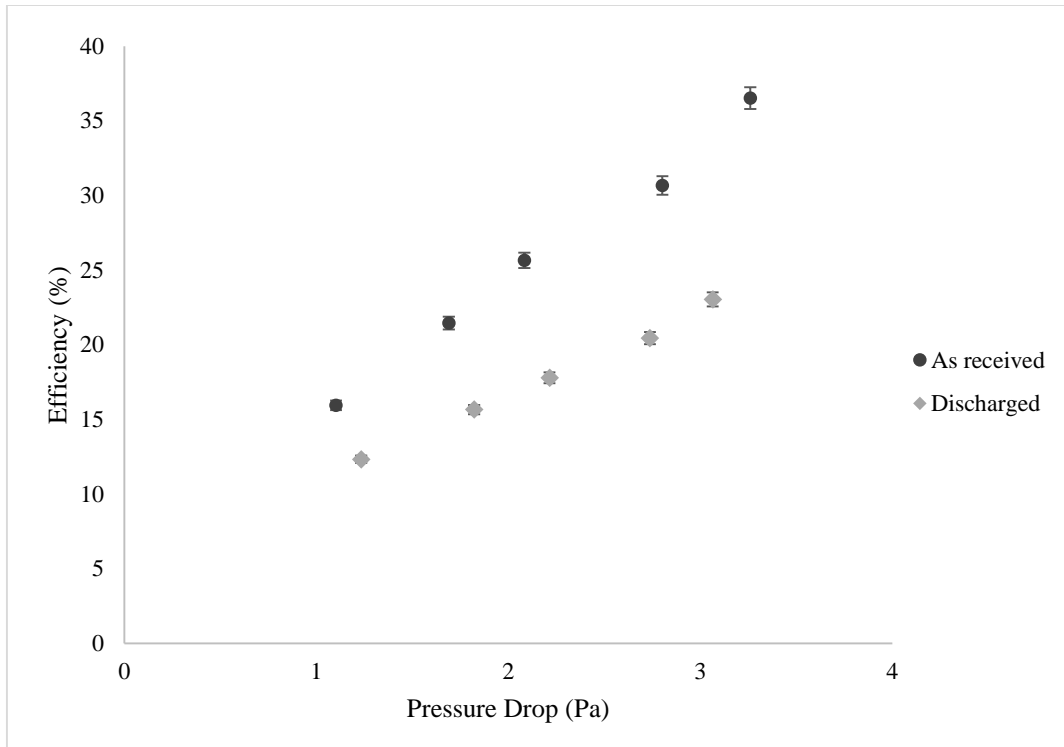


Figure 3-11 Filtration performance of as received and discharged (F2 sample).

### 3.3.4 Corona charging of PLA meltblown filter

It was shown that PLA meltblown are getting charge in manufacturing process enhancing filtration efficiency. In order to examine chargeability of PLA fibers finding optimum charging condition PLA filters treated with corona charging technique. Different charging parameters in corona charging namely applied voltage, charging time, and charging distance were examined to determine the optimum condition for charging and chargeability of fibers assessed based on their filtration efficiency of samples.

Figure 3-12 shows interactive effect charging time and charging voltage. It can be seen that with increase in applied voltage filtration efficiency of samples increases because of higher charge density and electrostatic force in filter media. At higher voltages higher amount of

ionized air will be driven toward the sample and therefore more charges will be deposited on fibers. With increase in charging filtration efficiency increases due to dripping of higher amount of charge on meltblown fibers. However, at higher applied voltage there is no change in filtration efficiency with increase in charging time.

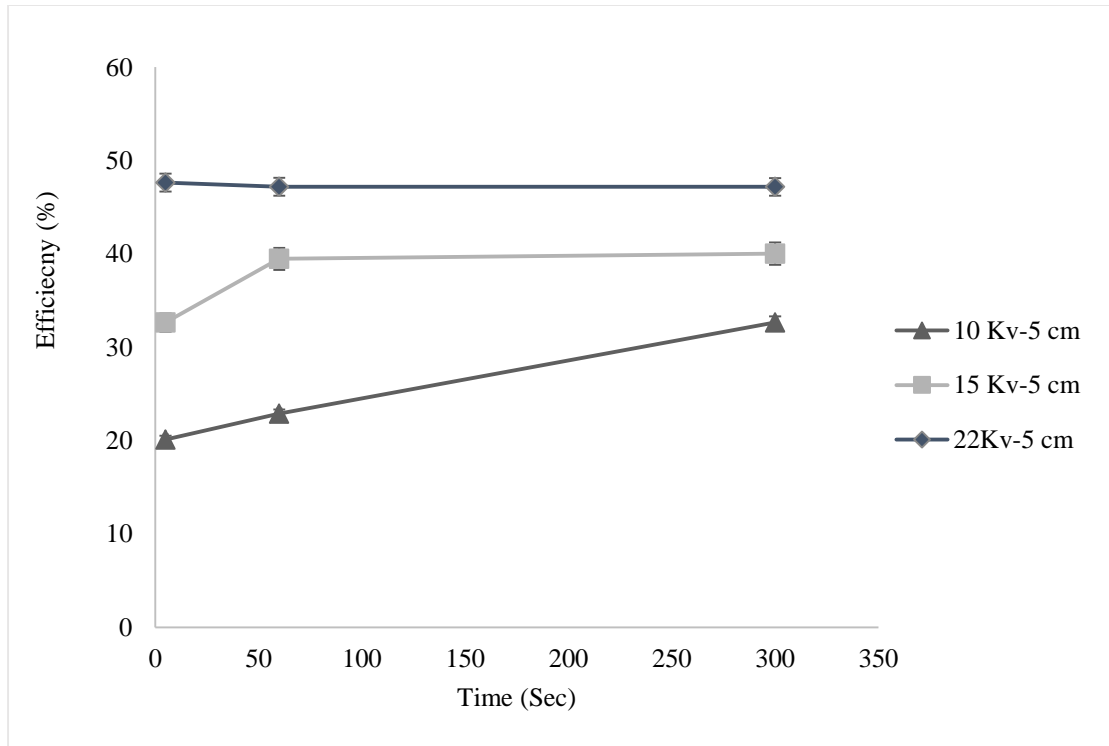


Figure 3-12 Effect of applied voltage and charging time on filtration efficiency (PLA 6100, 40 GSM)

Combined effect of charging distance and charging voltage displayed in figure 3-13. With increase in applied voltage at any distance filtration efficiency increases. However with increase in charging distance efficiency decreases as there will be more charge loss in the gap between two electrodes.

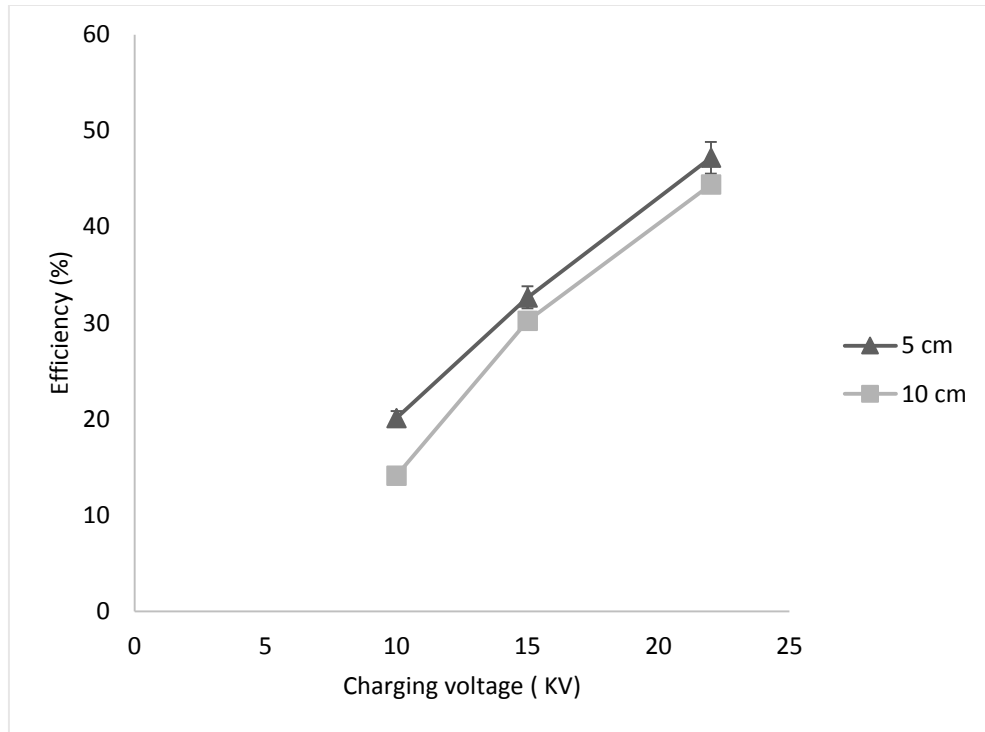


Figure 3-13 Effect of applied voltage and charging Distance on filtration efficiency (PLA 6100, 40 GSM)

Effect of charging time and charging distance for low and high-applied voltage displayed in figure 3-14. It can be seen that charging time and charging distance do not affect filtration efficiency at high voltages and PLA meltblown fibers charge density reaches to maximum at very beginning of charging process.

Results indicates that the applied voltage is the most influential parameters in corona charging of PLA meltblown fibers. At high-applied voltage after initial 5 seconds time has no influence in charging or filtration efficiency of PLA fibers and lower charging distance is more favorable. Regarding the results, corona charging of PLA fibers at 22 kV applied voltage at the distance of 5 cm for 5 sec can be considered as optimum charging condition.

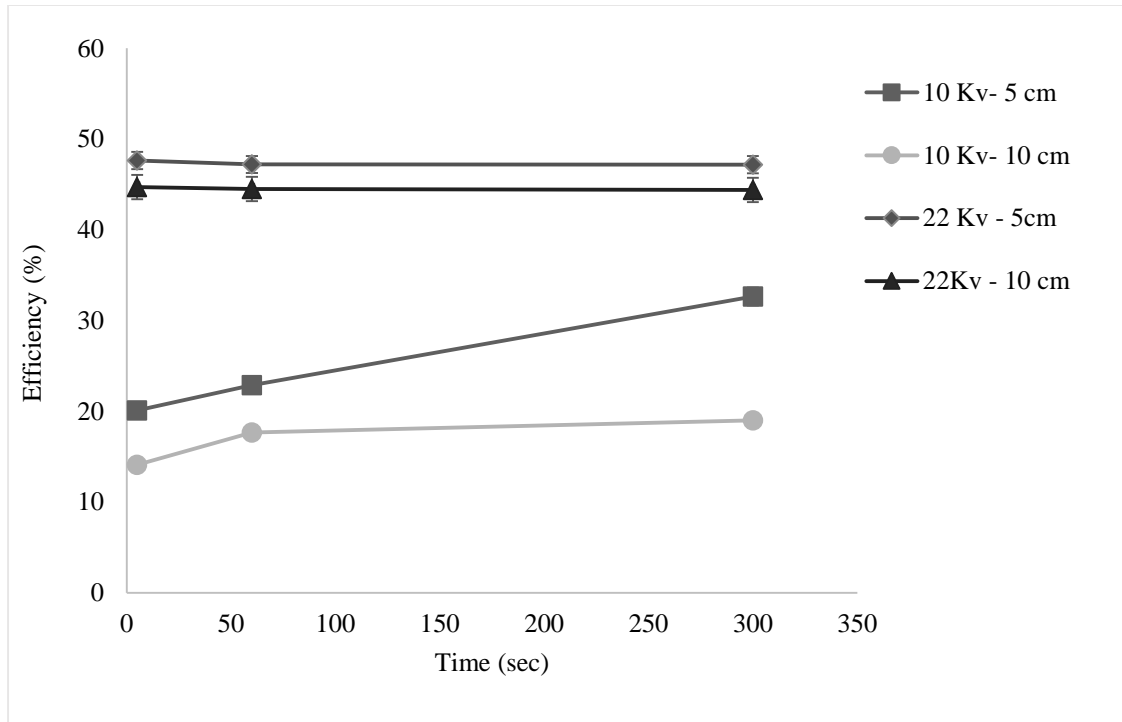


Figure 3-14 Effect of Charging distance and charging time on filtration efficiency (PLA 6100, 40 GSM)

### 3.4 Conclusion

Processability and effect of PLA polymer properties and fiber properties and fiber formation were investigated. PLA meltblown fibers made of PLA 6100 with lower D-isomer content showed acceptable range of crystallinity. Regarding processability in Biax meltblown system PLA 6100 with lower melt viscosity (higher melt flow index) performed better. Filtration efficiency results indicated that as-received PLA meltblown webs without presence of electrostatic field are getting charged in the production process for both polymer grades and resulting in higher filtration efficiency. PLA meltblown fibers reach to saturation level in few

when they are exposed to high voltage corona charging and additional charging time does not improve filtration efficiency of PLA meltblown webs.

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## **Chapter 4**

### **Effect of Nucleating Agent on Filtration Properties of PLA Meltblown Electret Filters**

## 4.1 Introduction

Electret material and their ability to retention of charge for long time makes them suitable for different applications such as air filters. Electret filters received significant of attention during last decades as high capturing efficiency of these filters proposed an effective and energy efficient, and economic way to air filtration (Barrett & Rousseau, 1998; Choi, Park, Kim, Kim, & Lee, 2015). Uncharged filter media capture particles through mechanical filtration mechanisms such as interception, impaction, and diffusion. To increase and enhance mechanical capture mechanisms fiber size should be decreased(K. W. Lee & Liu, 1980). Finer fibers in filter structure results in higher pressure drop across filter which is not favorable for filtration. Electret filters take advantage of electrostatic capture mechanism an addition to mechanical mechanisms and increase the efficiency without imposing extra pressure drop. Electrostatic mechanism allows reach high filtration efficiency without significant change in filter structure such as fiber diameter, thickness, or solidity and causing higher-pressure drop (Kanaoka et al., 1987). For many applications pressure drop cannot exceed from certain threshold. Regardless of physical and operational factor, charge density that and electret fiber can carry plays an important role in capturing efficiency. The higher the charge density on fiber the higher the electrostatic field acting on particles and thus higher the capturing efficiency. Moreover, another key element of electret filters is their ability to keep the charge in the fibers during lifetime of the filter as lack of charge stability in fibers after initial filtration efficiency will result in a mechanical filters with higher penetration since the structure is more open. Different studies has been conducted to improve chargeability and charge stability of electret filters (Cartwright, Davies, Swingler, & Vaughan, 1996; Tanaka et al., 2003) .



Modifying the fiber morphology using different types of additive is one of the most common approaches (A. A. Guzhova, Galikhanov, Gorokhovatsky, et al., 2016).

Additives such as nucleating agents has been wildly used to improve electret properties of polymeric materials. Nucleating agent (NA) are used to modify and improve formation of crystals in polymeric materials such as fibers. Slow crystallization rate of PLA results in amorphous and semi-crystalize structure and addition of NA not only can improve physical properties of PLA fibers but also electret properties as NA can affect size and quantity of crystals. Smaller crystal in the structure will provide more boundaries and interfaces between crystals and amorphous regions which are considered as charge trap sites in the structure and also creating deeper rap in the crystals which helps in retention of charge for longer time (Mohmeyer et al., 2007).

In this regard in order to improve electret filtration performance of PLA meltblown webs in terms on electrostatic capture efficiency and charge stability over lifetime we incorporated NA into polymer melt in meltblowing process. In chapter 3, it was shown that PLA 6100 with lower D-isomer content and melt viscosity showed better crystallization and processability on mini scale BIAX meltblown system. Therefore, we chose PLA 6100 and produced meltblown fiber and web but on pilot scale Reicofl meltblown system with BIAX deign die. Different NA concentration up to 1% were added and morphology of fibers were studied. Effect of NA on electrostatic filtration was studied using corona charging instrument and filtration efficiencies compared. Effect of fiber morphology on charge stability of electret filters investigated at high temperature and relative humidity up to 24hrs filtration test repeated at different intervals and results compared to analyze decay rate.

## **4.2 Experiments**

### **4.2.1 Material**

In pervious chapter, performance three different grades of PLA in meltblowing system examined and among them only PLA 6100 showed acceptable range of crystallinity and better processability due to lower melt viscosity. Therefore, in order to investigate effect of NA on morphology of PLA meltblown fiber, PLA 6100 with MFR value of 24 (10 min at 210 °C) provided by Natureworks LLC used to produce meltblown filter media.

CTL01 from Polyvol used as a NA to modify the fiber structure. Nucleating agent initially prepared in form of masterbatch at 20% (w/w) to provide better uniformity. The prepared masterbatch to extrude PLA fiber with NAs with concentration of 0.1%, 0.2%, 0.5%, and 1%.

### **4.2.2 Meltblown web production**

Meltblown webs were produced with pilot scale Reicofil meltblown in the Nonwovens Institute. In order to be able to process PLA 6100 we installed BIAX design die system on meltblowing unit because of processability issues such as high pressure n system. BIAX design die is able to handle high pressures and process high melt viscosity polymers.

### **4.2.3 Fiber Diameter Measurements**

In order to analyze the fiber morphology and measure the fiber size distribution, piece of each sample were sputter coated with a thin layer of gold and magnified images of fibers were taken at 1500X by a scanning electron microscope (Phenom FEI). More than 150 fiber diameter measurements were taken using Image J software from 15 pictures for each meltblown samples and average fiber size and fiber size distribution determined.

#### 4.2.4 Differential scanning calorimetry

Crystalline structure of PLA polymers and fibers were investigated using DSC in temperature ranging from 10 to 190 °C. Crystallization behavior of all the samples carried out at 3 steps, heating, cooling, and heating. First in order to eliminate thermal history samples were heated to 190 °C at the rate of 10 °C/min then quenched to ambient temperature. At the third step samples heated again to 190 °C at the rate of 10 °C/min to investigate melting behavior. Thermal properties such as glass transition temperature ( $T_g$ ), melting peak ( $T_m$ ), cold crystallization temperature ( $T_{cc}$ ), and enthalpy of crystallization while heating ( $\Delta H_{cc}$ ) and melting enthalpy ( $\Delta H_m$ ) were determined. Degree of crystallinity is calculated by:

$$\chi_c(\%) = \frac{\Delta H_m - \Delta H_{cc}}{H_{100\%}} \times 100 \quad (4-1)$$

Where  $H_{100\%}$  for PLA is 93 J/g.

#### 4.2.5 Air Permeability Measurement

Air permeability of meltblown samples were measured using Textest air permeability tester according to ASTM D 737, in analytical lab of the Nonwovens Institute in North Carolina State University. 10 replications of each sample were tested and average reported.

#### 4.2.6 Discharging

In order to study mechanical filtration and electrostatic capture efficacy separately, any charge and electrostatic effect should be eliminated. Therefore, discharging as received filter media after basic filtration performance measurements were carried out by immersion in isopropyl alcohol (IPA) bath for two hours and then dried under hood in room temperature for 24 hours.

#### **4.2.7 Corona Charging**

Corona charging technique utilized to charge meltblown samples. Sample placed between two electrodes at the optimum condition that described in chapter 3. Samples charged for 5 seconds at the voltage of +22 kV with charging distance of 3 cm.

#### **4.2.8 Filtration Performance Measurement**

Filtration performance of meltblown webs such as capture efficiency and pressure drop studied using 0.3  $\mu\text{m}$  monodisperse DOP (dioctyl phthalate) particles at the face-velocity of 5.3cm/s. Automated filter tester machine, TSI 3160, utilized to measure filtration performance in which certain particle size is generated by a collision type atomizer and a classifier, classifies the particles based on their electrical mobility and after neutralization particles are fed into the filter holder.

In order to evaluate, filter is installed on the filter holder and two condensation particle counters at upstream and downstream count the number of the particles and aerosol penetration or capture efficiency is calculated. Pressure drop is also calculated by mass flow meter. For each meltblown media six samples were cut and filtration performance were measured and the average of results were reported.

### **4.3 Result and Discussion**

#### **4.3.1 Web properties**

Figure 4-1 shows SEM images of selected samples containing NA and control sample. SEM images show that Nucleating agent is uniformly distributed within PLA fibers with no agglomeration detected on fibers.

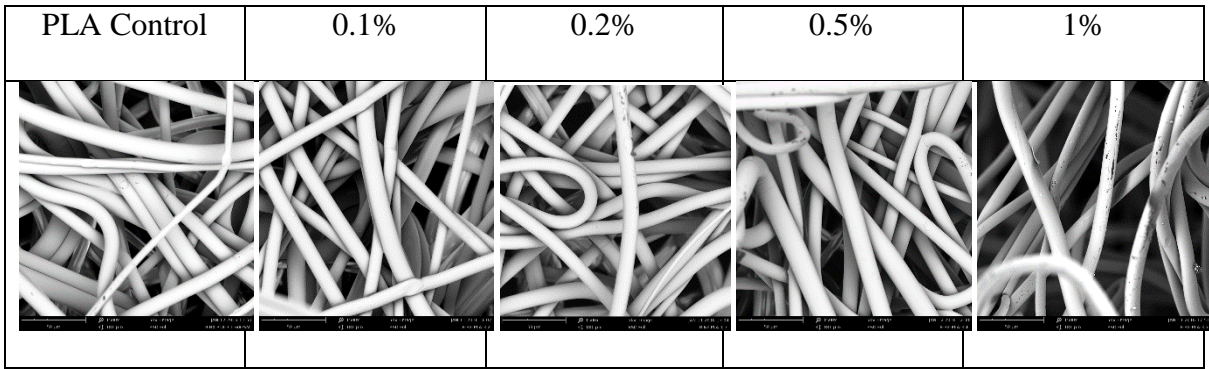


Figure 4-1 SEM images of PLA fiber containing nucleating agent

Measuring average fiber diameter and its distribution for each sample we realized that addition of NA in the meltblowing process did not significantly affect fiber size. Figure 4-2 compare average fiber size for control PLA and fibers containing NA and figure 4-3 displays fiber size distribution for control and 1% nucleating agent. In general having similar range average diameter and distribution (basic structure) is necessary to compare filtration performance of meltblown media. Fiber size plays an important role in mechanical capture efficiency and with having similar average fiber diameter and distribution would be helpful in studying effect of additive in electrostatic capture efficiency.

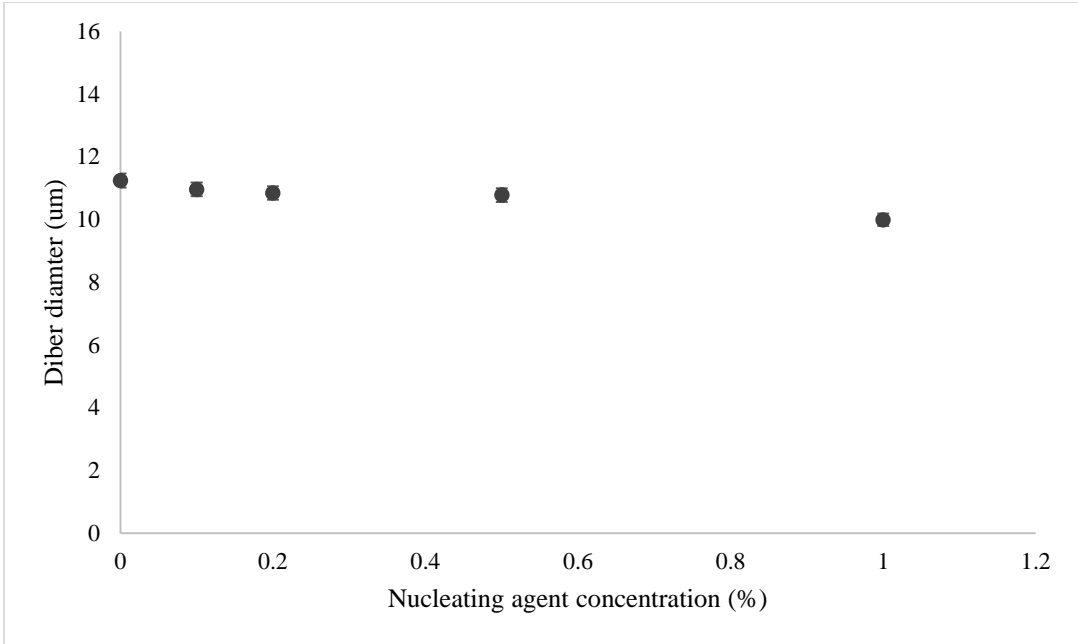


Figure 4-2 average fiber diameter of PLA meltblown fibers containing nucleating agent

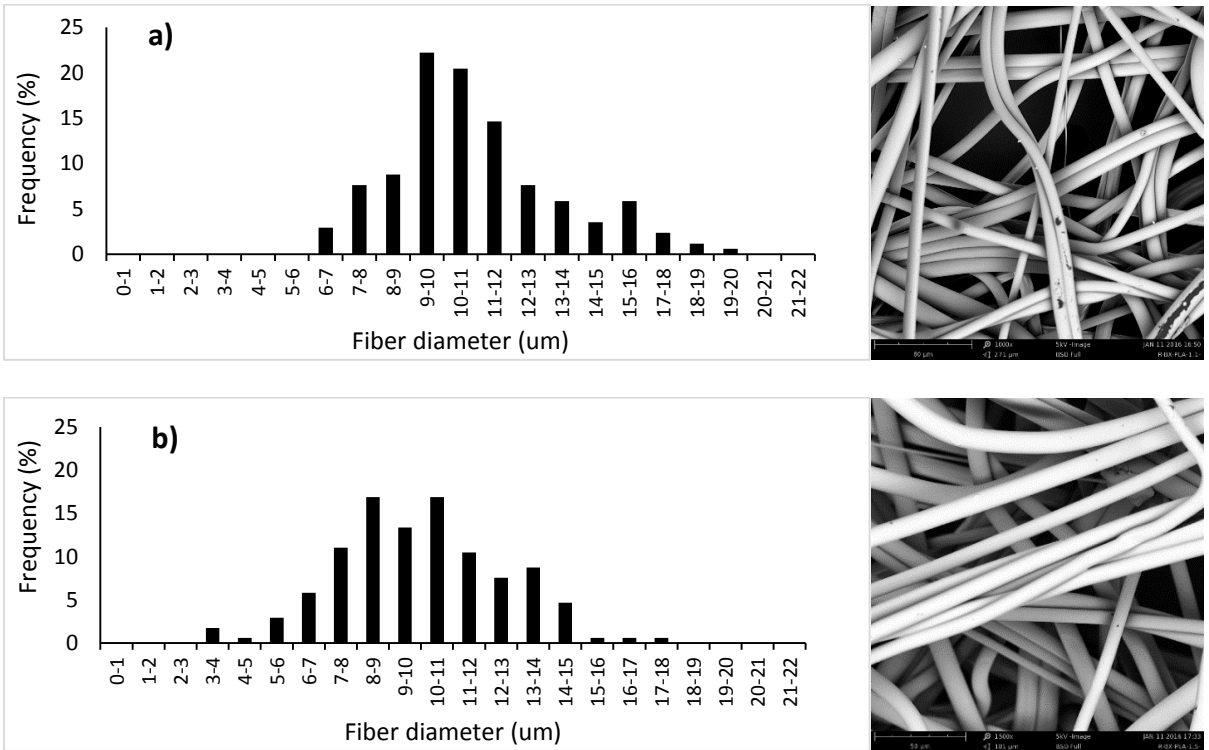


Figure 4-3 fiber size distribution of PLA containing nucleating agent: a) 0% b) 1%.

### 4.3.2 Thermal properties

Formation of crystals in the structure initiates with nucleation that can be described as aggregation of ordered polymer chains in molten state performing as crystal growth centers. Nucleating agents in the polymer melt act like aggregation center and increase the rate of crystallization. PLA does have a slow crystallization rate and meltblowing is a very fast process in which polymers are melted, extruded, and cooled down before collection (Li & Huneault, 2007). Consequently, polymer chains do not have enough time to move and orient and form crystals before cooling down below  $T_g$  where polymer chains are lock and immobilized. In order increase rate of crystallization, nucleating agent utilized.

Thermal properties of meltblown webs consisting pure PLA and PLA/NA are listed in table 4-1. All thermal properties of control and PLA/NA fibers obtained from first heating and cooling curves.

Table 4-1 Thermal characteristics of PLA containig nucleating agent

Sample	$\Delta H_m$ (J/g)	$\Delta H_c$ (j/g)	Crystallinity (%)	$T_m$ (°c)	$T_c$ (°c)	$T_g$ (°c)
PLA control	53.304	28.44	26	179	109	56
0.1% NA	53.90	25.99	30	173	110	58
0.2 % NA	55.2	27.45	29.8	172	110	55
0.5% NA	50.26	21.38	31.2	172	109	55
1% NA	55.97	21.7	36.9	172	107	53.3

Figure 4-4 and figure4-5 show DSC melting and cooling thermogram for all samples. It can be seen that addition of NA did not significantly affect glass transition temperature and  $T_g$  values are about the same except for PLA with 1% NA that slightly decreased, table 4-1. Reduction in  $T_g$  shows that nucleating agent was able to facilitate segmental mobility of PLA chains (Fehri, Cinelli, Coltelli, Anguillesi, & Lazzeri, 2016).

Addition of NA affected crystallinity of PLA meltblown fibers. By crystallinity rate achieved by adding 1% NA around 37%. Increasing NA content can lead to more nuclei density nad so higher crytalizablity of polymer. In addition empathy of cold crystallization decreased by incorporation of NA.

Samples with 0.5% and 1% NA showed double melting peaks. Based on literature appearance of double melting peaks which is common in semi-crystalline polymers such PLA can be attributed to formation of different crystal types or different morphology regarding size and thickness of crystals (Quero, Müller, Signori, Coltelli, & Bronco, 2012). As mentioned before NA could change quantity and size of crystals.

By looking at DSC cooling thermogram, we found out NA was not efficient enough to affect onset crystallization temperature showing for almost all the fibers crystallization time was similar, figure 4-5. Higher crystallinity of PLA fibers with 1% NA indicates higher rate of crystallization in the cooling process before collection.



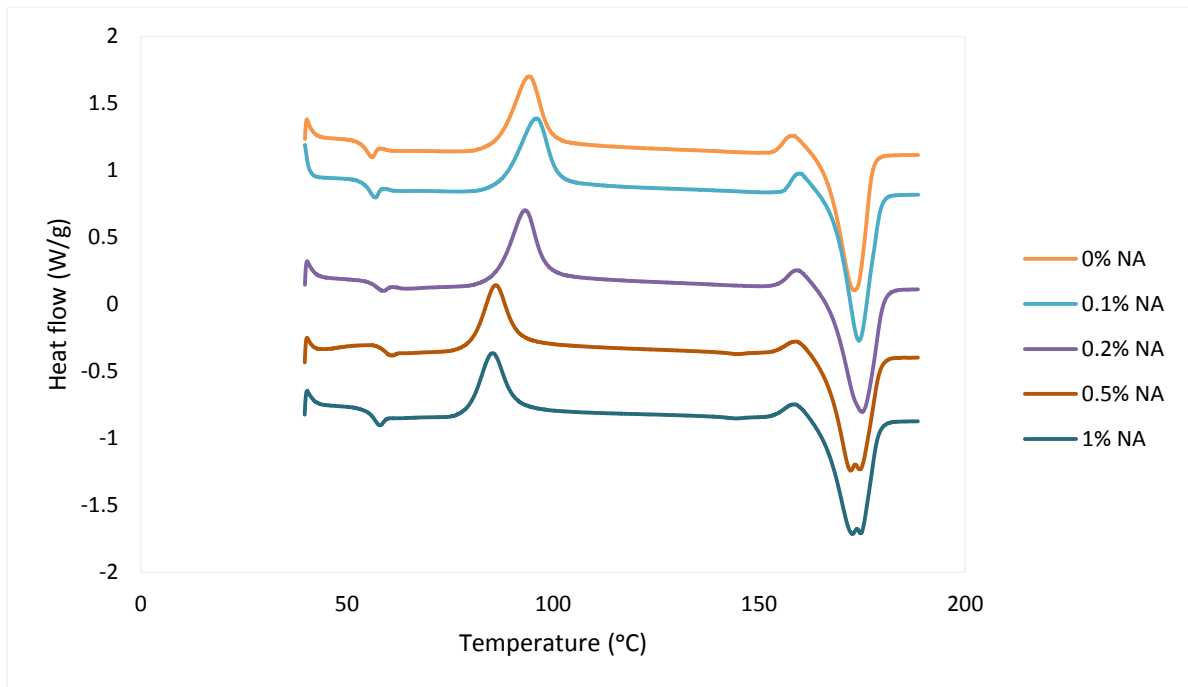


Figure 4-4 DSC heating thermogram of PLA fibers with different percentage of nucleating agent in the structure

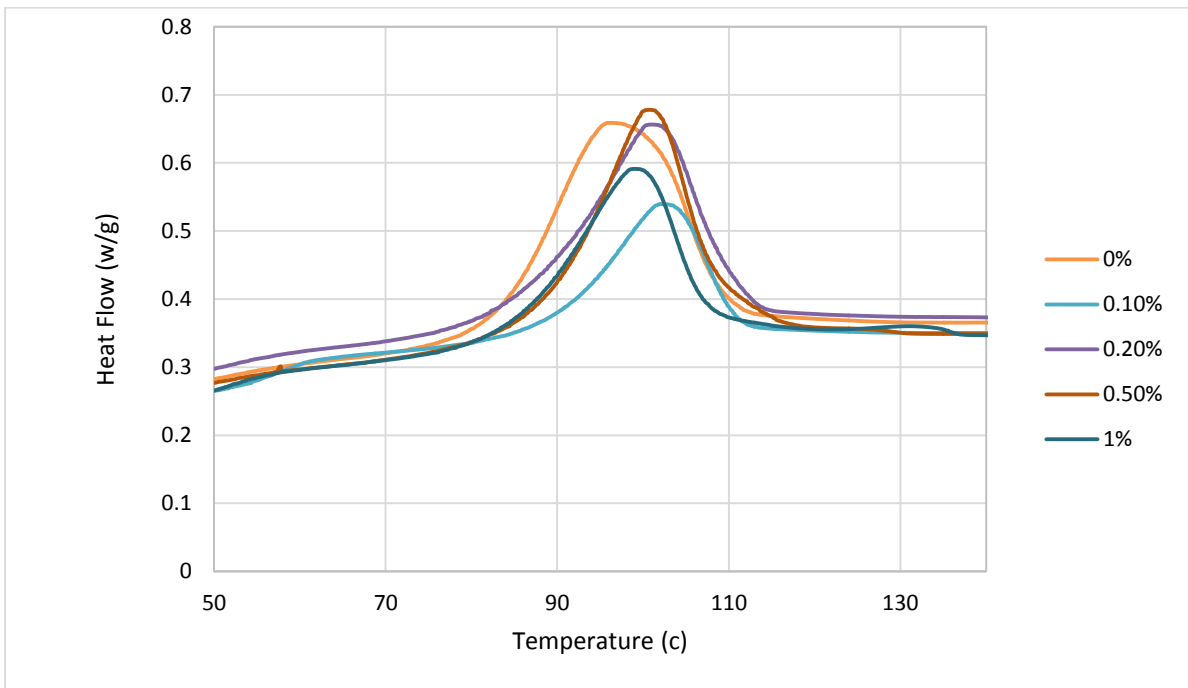


Figure 4-5 DSC cooling thermogram of PLA fibers contain nucleating agent

### 4.3.3 Air permeability

Addition of NA also influenced on the structure of the meltblown webs. Increasing in concentration of NA resulted in higher air permeability of meltblown webs. Previously mentioned that NA did not significantly affect fiber diameter and their distribution. However, incorporation of NA into fiber structure leads to higher density of fibers which influences on solid volume fraction (solidity) of resulted web. In meltblowing process producing samples is performed as weight feed base, meaning different melt density results in different volume of feed in production process. If the polymer matrix in the molten state possess higher density, in order to reaching same basis weight in the process lower volume of feed was processed per unit of time. Therefore, lower amount of fibers will be deposited on collector and so webs will have rather open structure (higher fiber-to-fiber distance). **Figure** 4-6 shows air permeability of meltblown samples. With increase in concentration of NA, meltblown samples show higher air permeability. Higher air permeability is desired for air filtration to have lower pressure drop.

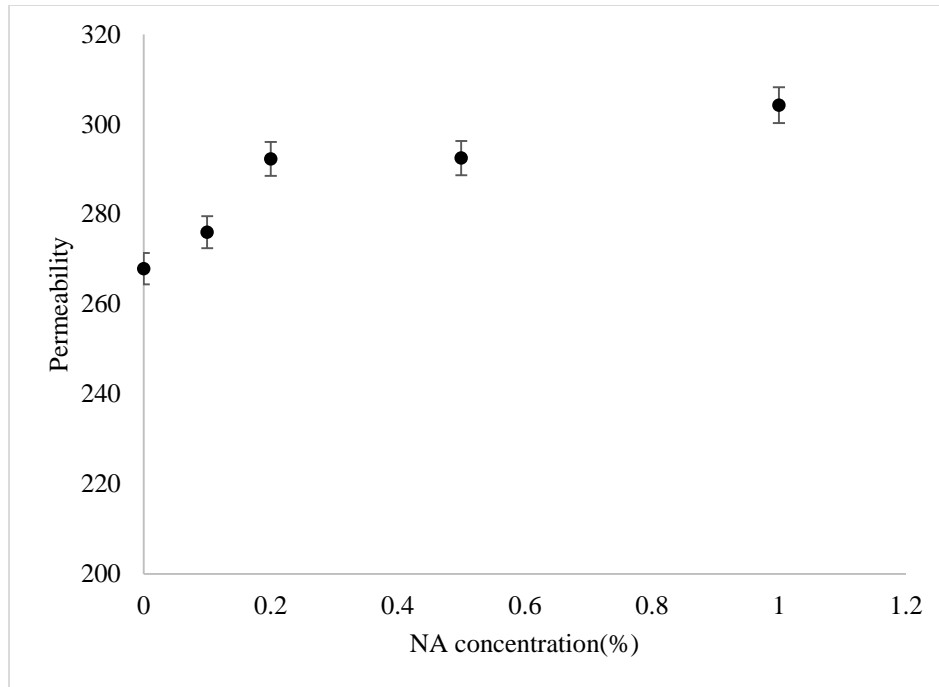


Figure 4-6 Air permeability of PLA meltblown web containing nucleating agent.

#### 4.3.4 Filtration performance

As discussed NA in the fibers not only affects fiber properties but also web properties. Fiber and web properties both affect the mechanical and electret filtration efficiencies. Filtration results of as received PLA control and PLA/NA meltblown web are shown in figure 4-7. Filtration properties of all the samples are similar to each other except for sample containing 1% NA. As mentioned addition of NA and increasing fiber density resulted in web with more open structure and higher air permeability. Despite open structure still showing even higher filtration efficiency comparing to other samples showing better quality of filter.

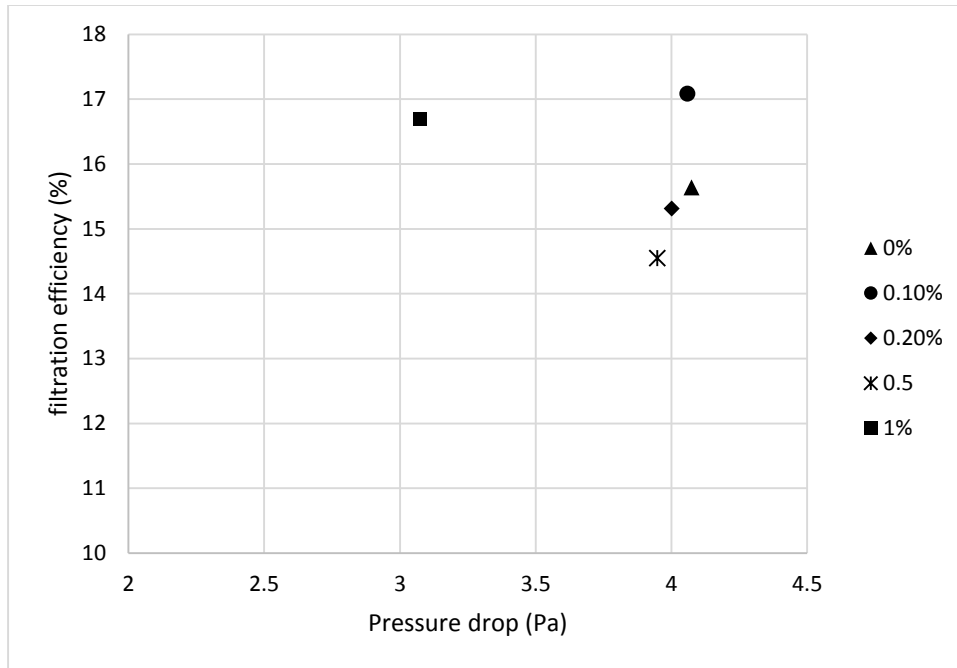


Figure 4-7 as received filtration performance of PLA web containing nucleating agent.

Filtration efficiency of discharged PLA control and PLA/Na samples are shown in figure 4-8. Efficiency of all sample with and without NA decreased. As no external electrostatic field was involved before testing filtration efficiency of meltblown samples, presumably during manufacturing process electrostatic charge induced to the fibers through triboelectrification.

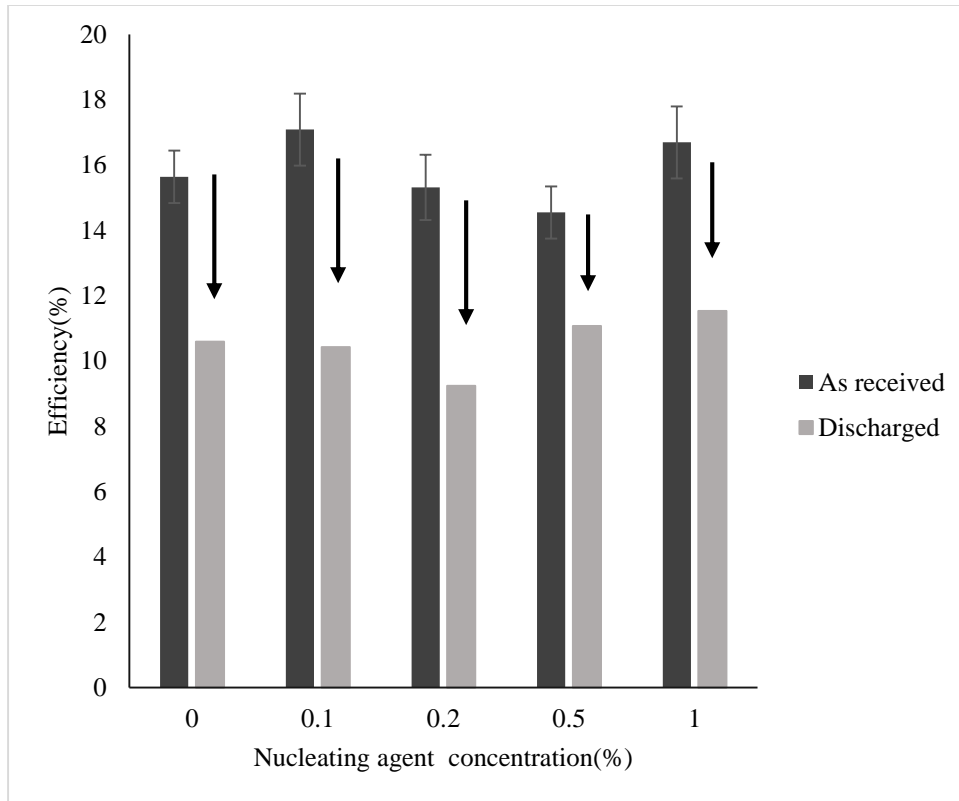


Figure 4-8 filtration efficiency of discharged and as received PLA web with nucleating agent.

As mentioned before addition of NA did not significantly affect average fiber diameter and their distribution, which make the comparison of change in electrostatic filtration more accurate since mechanical filtration would be similar. In figure 4-9 filtration results for charged samples are shown. Applying electrostatic charge enhanced filtration efficiency of samples regardless of NA concentration. PLA filter media with 1% NA in the structure is posed better filtration performance having higher filtration efficiency and lower pressure drop. Since mechanical filtration efficiencies were almost similar, we can conclude that addition of one percent NA into the fibers and increasing rate of crystallinity improved electrostatic capture mechanism of PLA meltblown fiber.

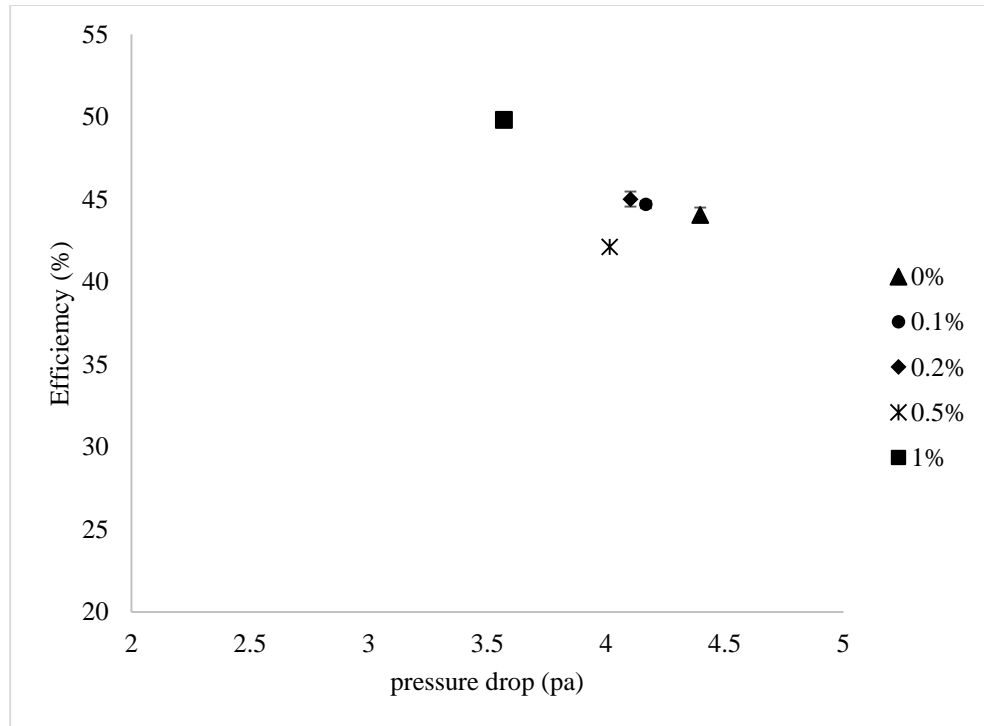


Figure 4-9 Efficiency vs pressure drop of corona charged PLA meltblown samples containing nucleating agent

Figure 4-10 illustrates the quality factor values after and before charging. Increasing nucleating concentration less than 1% did not significantly affect quality of filters before and after charging. Tangible difference occurred at 1% as the samples with 1% NA showed higher filtration efficiency and lower pressure drop which. Increase in electrostatic filtration efficiency can be attributed to higher charge density of fibers. NA increases rate of crystallinity and reducing size of crystals therefore new deep traps will be produced at the amorphous-crystalline interface resulting in stronger electrostatic field at the vicinity of fibers.

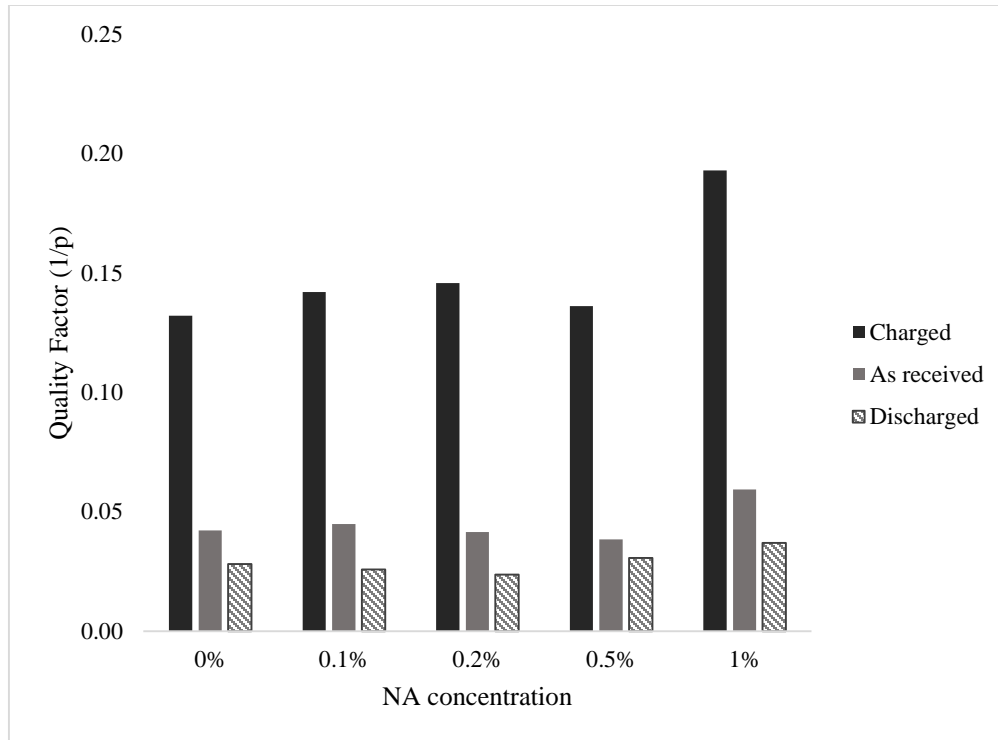


Figure 4-10 Quality factor of as received, charged, and discharged PLA meltblown media with different NA concentration.

One of the most characteristic of webs and fibers as electret filters is their ability to keep the charge in structure during lifetime of filter as electric field generated by charged fibers acts on particles. Charge stability in fibers will indicate the durability of filter performance. In order to analyze charge stability of fiber, accelerated decay method at high temperature used to characterize charge decay performance of the fibers since at lower temperatures polymer chains and molecules are immobile decay rate would be too low and thus time consuming. Electric charges are known to be have temperature dependent mobility within the bulk and over the surface and higher temperature will lead to ease of decay. In addition, water molecules or charged particles in the air or DOP particles in the filtration test can neutralize charges. It

should be noticed that charge decay caused by charged particle and testing materials are relatively small as the exposure time is just few minutes. Hence, filter samples were put in the environmental chamber under certain temperature and humidity and at different intervals up to 24 hrs filtration efficiency as a measure of surface potential evaluated. Loss in efficiency equals to charge loss and less electrostatic capture mechanism. Figure 4-11 shows charge decay performance PLA filters in terms of filtration efficiency. Filtration efficiency of PLA sample with 1% NA comparing to control sample after initial drop follows a steadier trend which means charges are more stable.

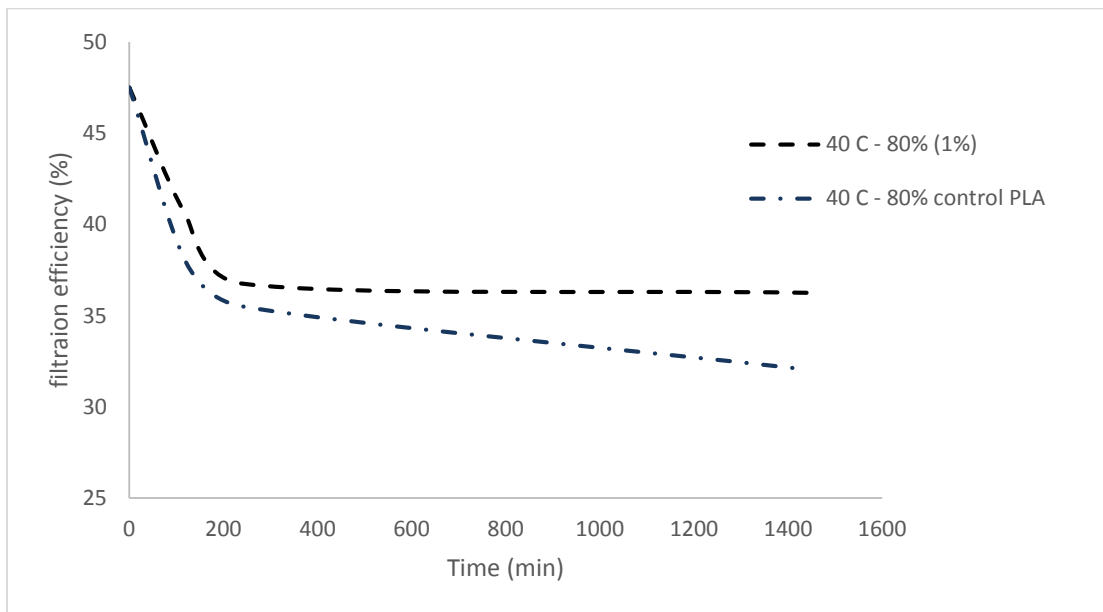


Figure 4-11 charge decay performance of control sample and sample containing 1% nucleating agent



#### 4.4 Conclusion

Influence of nucleating agent addition on PLA on fiber thermal and physical properties as well as electrostatic filtration performance was investigated. Addition of 1% nucleating agent improved rate of crystallization and crystallinity of fibers with 1% NA agent increased up to 37% comparing to control PLA (26%). Incorporation of 1% NA into fiber enhanced their electret behavior. Chargeability of PLA meltblown fibers improved by addition one 1% NA and filters showed higher electrostatic capture efficiency comparing to control samples. Accelerated decay test was performed to analyze charge stability of PLA electret filters. Charge stability of PLA/Na fibers improved and decay rate after 24hrs was lower than those of control samples.

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## **CHAPTER 5**

Process-Structure-Filtration Property relationship of low viscosity PLA via  
traditional Exxon die system

## 5.1 Introduction

Structure and properties of final nonwoven fabric is affected by material and processing condition. Many studies investigated effect of structural properties and nonwoven characteristics for different applications such as air filtration (Albrecht et al., 2006; Huang et al., 2013). Parameters such as basis weight, web thickness, fiber diameter, and solidity affect performance of filter media. Among these parameters, fiber diameter plays a significant role influencing on web structure and filtration capturing mechanisms (mechanical and electrostatic) . Lower fiber diameters results in larger surface area and smaller pore size that are critical in filtration application (Gądor & Jankowska, 1999).

Meltblowing as one of the most significant developments of nonwoven technology is a unique one-step process in which polymer is transformed into fiber within the range of 1-5  $\mu\text{m}$  with high coverage and small pore size that makes it suitable for air filtration applications (Yu et al., 2015). In this process, molten polymer is extruded through micro-scale holes of die and jets of hot air attenuates the streams of molten polymers. Many thermoplastic polymers are used in meltblown system but among them polypropylene is the most common polymer due to relatively lower price and availability in different composition for producing different products. In addition, PP possess low melt viscosity which is an essential requirement for polymer in meltblowing process as it should be able to flow through small holes (Dutton, 2008).

Increasing environmental concerns regarding the harms of plastic materials to the environment in recent years, biodegradable materials has received more attention in academic and industrial point of view. Among biodegradable material PLA is considered as promising polymeric material for replacement of petroleum-based synthetic plastic because of its better mechanical and thermal properties (Gao, Wang, Wang, & Zhang, 2016).

Processing of biodegradable polymers in meltblowing system such as PLA is in developing stage as the rheological properties imposes strong restrictions (Muller & Krobjilowski, 2001). Meltblowing process requires low molecular weight or low melt viscosity polymer to produce uniform and fine fibers. The melt viscosity is inversely proportional to the melt flow rate (MFR).

In this chapter, we aim to investigate effect of melt viscosity or melt flow rate (MFR) and processing parameters on fiber and web formation in meltblown system. In addition, mechanical and electret filtration performance of PLA meltblown web investigated.

## **5.2 Experiments**

### **5.2.1 Material**

PLA 6260 with MFR value of 65 (10 min at 210 °C) kindly provide by Natureworks LLC. According to information provided by the company, PLA 6260 comparing to other PLA grades used in this research had lower molecular weight and similar range of D- isomer content to PLA 6100.

### **5.2.2 Meltblown production**

The Pilot scale Reicofil-Exxon die 35 (Hole/Inch) meltblowing line of The Nonwoven Institute at North Carolina State University utilized to produce PP and PLA meltblown media.

PLA chips were dried for 8 hrs at 80 °C in a vacuum dryer in order to eliminate the moisture to minimize hydrolysis during the melt blowing.

In meltblowing process, in order to achieve different fiber diameter distribution and solidity we changed polymer throughput, airflow, and die to collector distance and kept basis weight constant (40 gm<sup>-2</sup>). For both PLA and PP, polymer throughput were kept at 0.3 and 0.6 g/hole/min. Airflow of 800, 1200, 1500, and 1900 (m<sup>3</sup>/hr) used to impact on fiber size distribution, Table 5-1.

Table 5-1 Processing condition of PLA 6260

Sample	DCD (mm)	Throughput (ghm)	Air (m <sup>3</sup> /hr)	Basis weight (gsm)
P1	25	0.6	800	40
P2	25	0.6	1200	40
P3	25	0.6	1500	40
P4	25	0.6	1900	40
P5	25	0.3	800	40
P6	25	0.3	1200	40
P7	25	0.3	1500	40
P8	25	0.3	1900	40
P9	15	0.6	800	40
P10	15	0.6	1200	40
P11	15	0.6	1500	40
P12	15	0.6	1900	40
P13	15	0.3	800	40
P14	15	0.3	1200	40
P15	15	0.3	1500	40
P16	15	0.3	1900	40

### 5.2.3 Fiber diameter measurements

In order to analyze the fiber morphology and measure the fiber size distribution, piece of each sample were sputter coated with a thin layer of gold and magnified images of fibers were taken at 1500X by a scanning electron microscope (Phenom FEI). More than 150 fiber diameter measurements were conducted using Image J software from 15 pictures for each meltblown samples.

### 5.2.4 Fiber microstructure

Crystalline structure of PLA polymers and fibers were investigated using DSC in temperature ranging from 10 to 190 °C. Crystallization behavior of all the samples carried out at 3 steps, heating, cooling, and heating. First in order to eliminate thermal history samples were heated to 190 °C at the rate of 10 °C/min then quenched to ambient temperature. At the third step samples heated again to 190 °C at the rate of 10 °C/min to investigate melting behavior. Thermal properties such as glass transition temperature ( $T_g$ ), melting peak ( $T_m$ ), cold crystallization temperature ( $T_{cc}$ ), and enthalpy of crystallization while heating ( $\Delta H_{cc}$ ) and melting enthalpy ( $\Delta H_m$ ) were determined. Degree of crystallinity is calculated by:

$$\chi_c(\%) = \frac{\Delta H_m - \Delta H_{cc}}{H_{100\%}} \times 100 \quad (5-1)$$

### 5.2.5 Air Permeability measurement

Air permeability of meltblown samples were measured using Textest air permeability tester according to ASTM D 737, in analytical lab of the Nonwovens Institute in North Carolina State University. 10 replications of each sample were tested and average reported.



### **5.2.6 Discharging**

In order to study mechanical filtration and electrostatic capture efficacy separately, any charge and electrostatic effect should be eliminated. Therefore, discharging as received filter media after basic filtration performance measurements were carried out by immersion in isopropyl alcohol (IPA) bath for two hours and then dried under hood in room temperature for 24 hours.

### **5.2.7 Corona charging**

A lab scale negative corona discharger (Mystic Marvels, Model NIP-7E) was used to charge samples. Sample were put between two electrodes at the optimum condition that described in chapter 3. Samples were charged for 5 seconds at the voltage of +22 kV with charging distance of 3 cm.

### **5.2.8 Filtration performance measurement**

Filtration performance of meltblown webs such as capture efficiency and pressure drop studied using  $0.3 \mu\text{m}$  monodisperse DOP (dioctyl phthalate) particles at the face-velocity of 5.3cm/s. Automated filter tester machine, TSI 3160, utilized to measure filtration performance in which certain particle size is generated by a collision type atomizer and a classifier, classifies the particles based on their electrical mobility and after neutralization particles are fed into the filter holder.

In order to evaluate, filter is installed on the filter holder and two condensation particle counters at upstream and downstream count the number of the particles and aerosol penetration or capture efficiency is calculated. Pressure drop is also calculated by mass flow meter. For each meltblown media six samples were cut and filtration performance were measured and the average of results were reported

## 5.3 Result and Discussion

### 5.3.1 Fiber Diameter

In many applications including filtration, fiber size is a key parameter and it is important to be controlled to reach desired properties. Fiber size in meltblown process is affected by polymer properties and processing parameters. Processing condition such as throughput, airflow and DCD affect fiber diameter. Regarding polymer properties, molecular weight and melt viscosity affect fiber diameter. In chapter 3, we indicated that PLA meltblown fibers due to high melt viscosity of selected PLA grades showed higher average fiber diameter (around  $10\mu\text{m}$ ) which was higher than typical melt blown and for filtration application in comparison to convectional polymer used in meltblown process.

Figure 5-1 shows effect of processing parameters on average fiber size distribution of PLA 6260 meltblown fibers. Increasing in airflow resulted in decreasing average fiber diameter. The reduction in fiber diameter is mainly because of higher drag forces on molten jet of polymer and higher attenuation before lay down. Also with increase in throughput, average fiber diameter increases. With keeping other processing parameters constant except throughput, decreasing throughput or polymer mass flow results in lower average fiber size since the same amount of drag force from the air jets is functioning on smaller polymer mass.

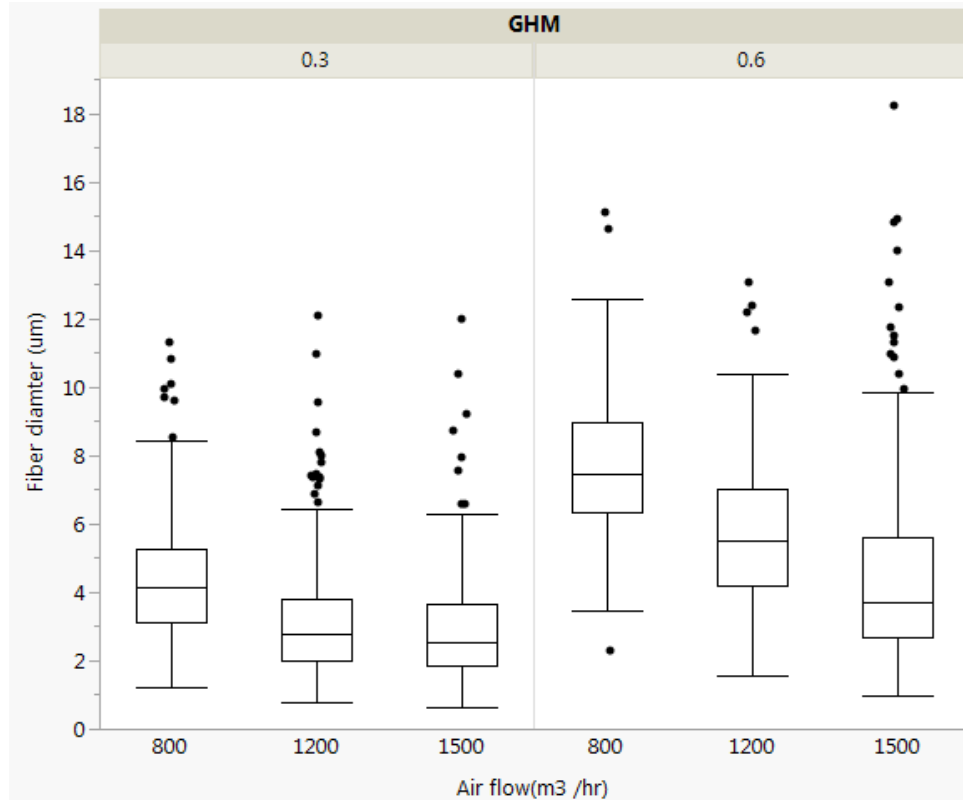


Figure 5-1 effect of throughput and air flow on average fiber diameter of PLA 6260 meltblown media ( DCD=15 cm, 40 gsm)

Figure 5-2 represents effect of DCD on fiber average fiber size of PLA meltblown fibers with different processing condition. As it is shown dependency of average fiber size on DCD in comparison to throughput and airflow is relatively low. Research indicates that majority of stressed acting of molten polymer occurs in few centimeters of after coming out of die therefore increasing DCD cannot highly affect fiber diameter (Dutton, 2008). However, as fibers produced in meltblowing often do not solidify before lay down on the collector fiber diameter continuous to change and fibers attenuate due to air drag force.

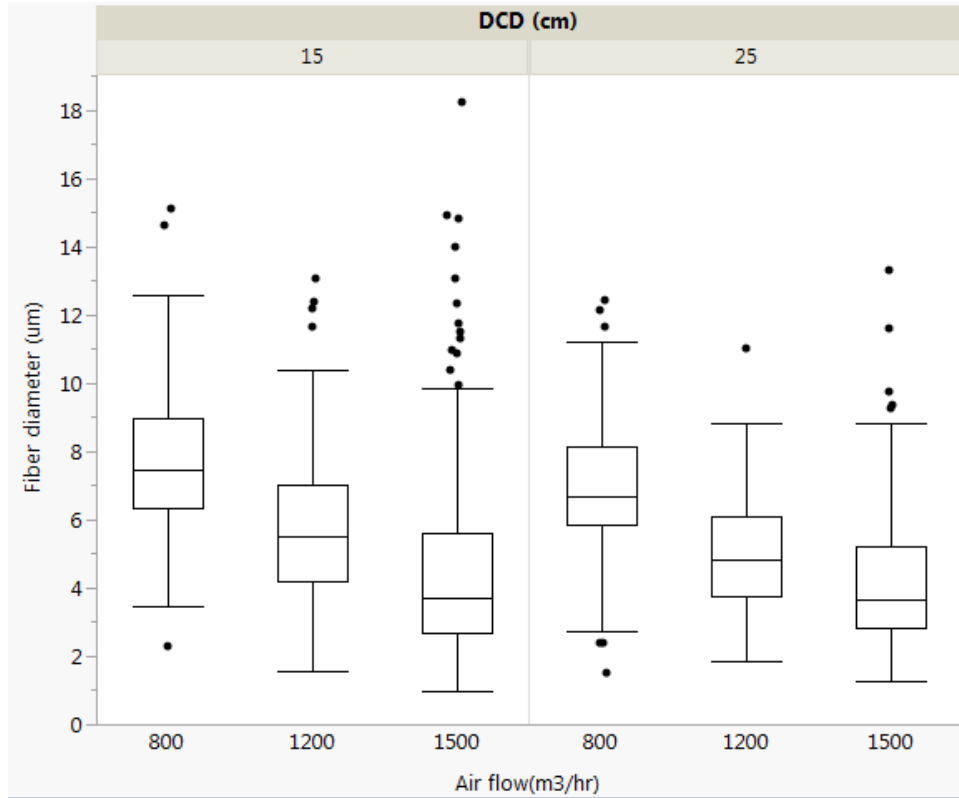


Figure 5-2 Effect of DCD on average fiber diameter of PLA 6260 meltblown media (0.6 ghm, 40 gsm)

### 5.3.2 Effect of processing condition on air permeability on solidity of PLA meltblown media

Air permeability of nonwoven media is strongly affected by structure and composition of the nonwoven web. In filtration application higher air permeability is required in order to have lower pressure drop. Air permeability of nonwoven media is inversely proportional to the thickness of the media in which with increase in thickness air permeability will decrease (Ozturk, Nergis, & Candan, n.d.). In addition, air permeability of nonwoven fabric is affected by pore size in the structure, which is function of fiber diameter. Figure 5-3 shows effect of processing condition on air permeability of PLA meltblown media. As it can be noticed with

higher airflow and lower throughput (polymer mass flow rate) air permeability decreases. Higher airflow and lower throughput results in lower fiber diameter. Smaller fiber size results in smaller pores in the structure hence at the constant basis weight air permeability decreases.

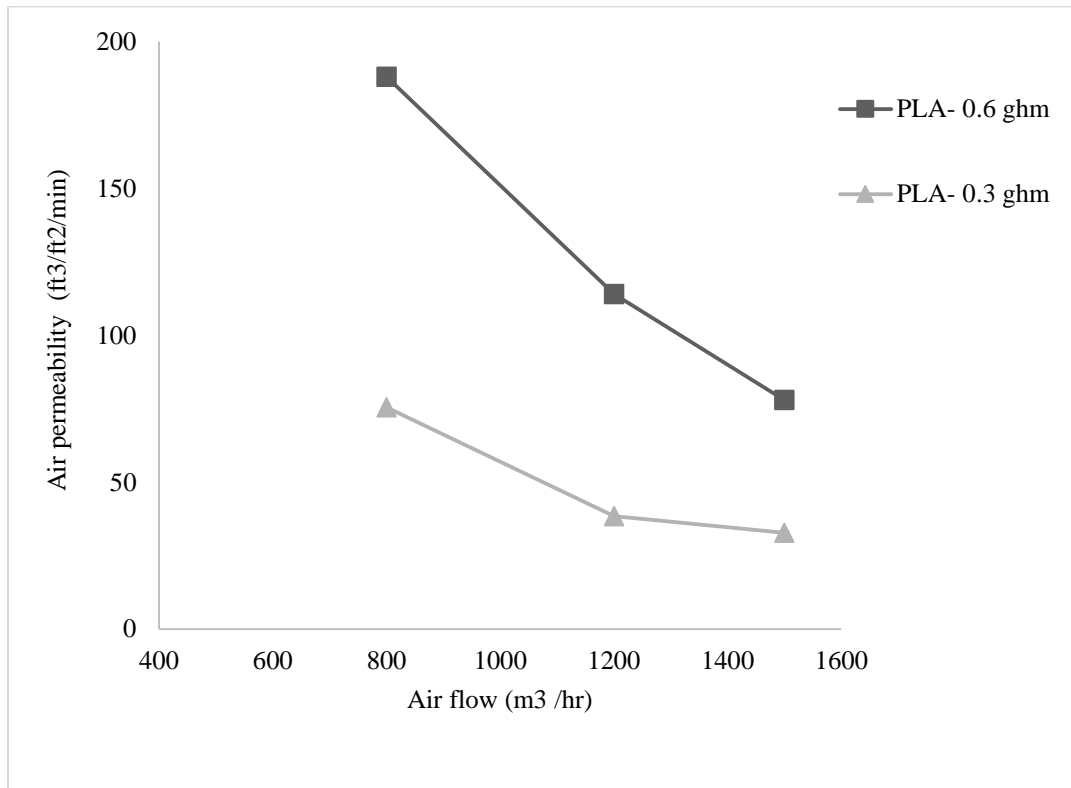


Figure 5-3 Effect of processing condition on air permeability of PLA 6260 meltblown media ( DCD=15 cm, 40 gsm)

Figure 5-4 shows effect of processing condition or fiber diameter on solidity or solid volume fraction. As fiber size decreases, solidity of nonwoven media increases as more fibers can be placed next to each other at same volume and overlap of fiber on top of each other will result in lower thickness at the constant basis weight.

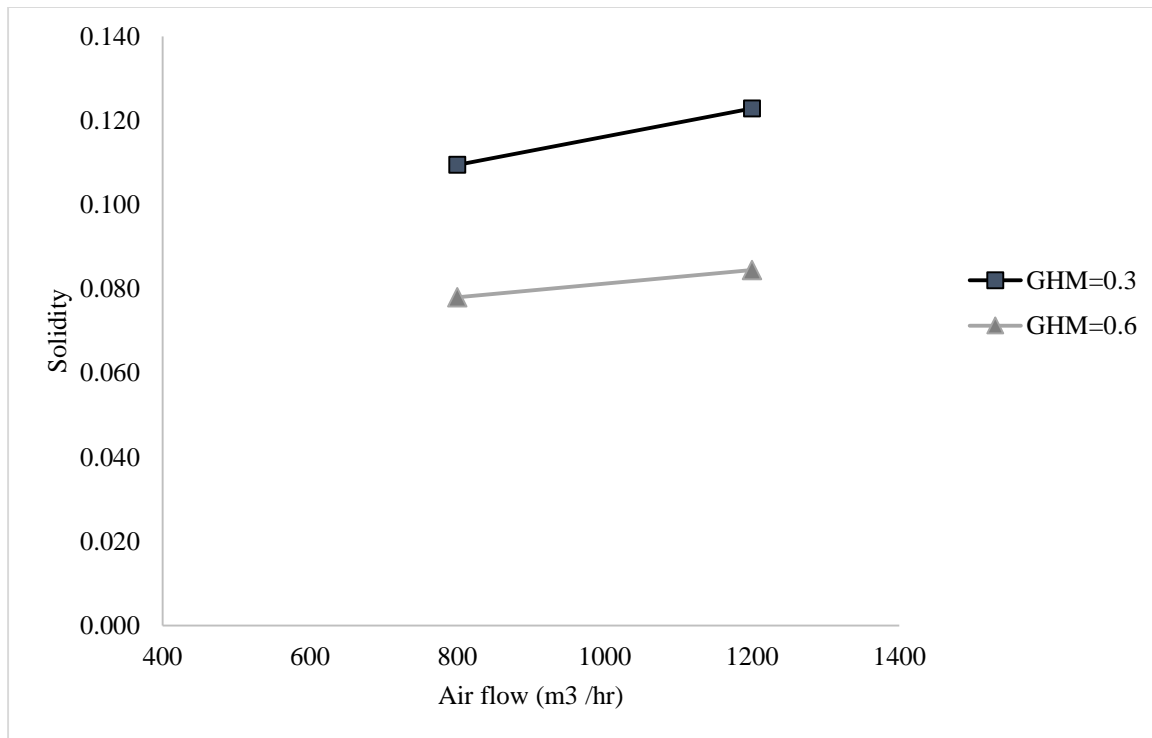


Figure 5-4 Effect of processing condition on solidity of PLA 6260 meltblown media

### 5.3.3 Filtration performance

In order to investigate filtration performance of PLA meltblown webs at first we focused on filtration efficiency of as-received filter media to determine if mechanical capture efficiency or combination of mechanical and electrostatic efficiency are functioning in filtration process as triboelectrification may occur and induce charge to samples. To make sure the filtration efficiency of as-received samples is only mechanical efficiency discharging of samples was conducted and filtration efficiency measured.

Figure 5-5 illustrates filtration efficiency of as-received and discharged samples. Regarding the filtration efficiency of discharged samples, we observed that efficiency loss after discharging is large enough to consider electrostatic charge interfering and affecting filtration

efficiency of as received PLA meltblown media. Change in filtration efficiency is more tangible for filters with larger average fiber size in the structure. As mentioned with larger fiber, solidity decreases and air permeability increases. Lower solidity of nonwoven media equals to lower amount fiber and according to mechanical filtration capture mechanisms leads to lower chance of collision between of particles and fibers. Therefore, when filters with larger fibers are equipped to electrostatic capture mechanism filtration efficiency improves drastically as particles would be pulled toward fibers and helps mechanical capture mechanisms which are not effective enough comparing to media with smaller fiber size. Therefore as it can be seen from figure 5-5, the efficiency drop for filter media with larger average fiber size is much higher than filters with smaller fiber size as they have stronger mechanical capture mechanism.

Effect of induced charge on efficiency during process as well as structure of filter media on pressure drop of PLA meltblown media can be seen more clearly in their filtration quality factor. The filtration quality of samples shown in figure 5-6 has been calculated and depicted based on the following equation (Wang, Kim, & Pui, 2008):

$$Q_F = \frac{-\ln(P)}{\Delta P} \quad (5-3)$$

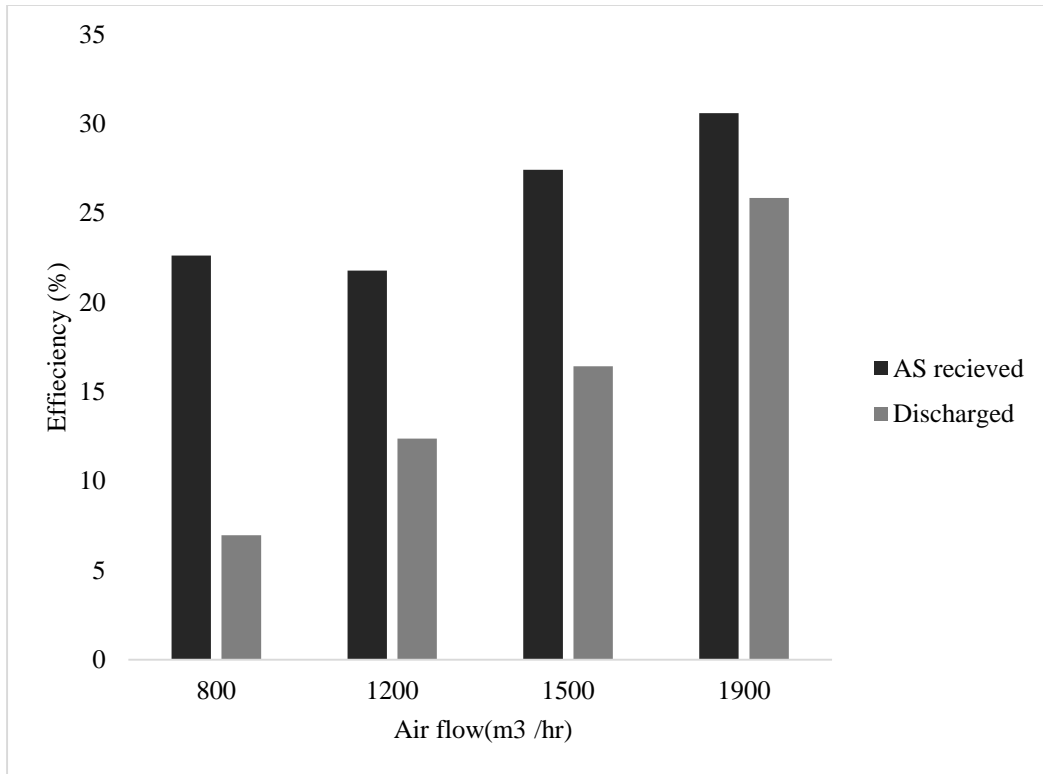


Figure 5-5 Filtration efficiency of PLA meltblown media as received and discharged (40 gsm, DCD= 25 cm, 0.6 ghm)

As received Filtration efficiency result of PLA meltblown samples are within range of 22-31% and relatively similar. However, because of different average fiber size and structural composition, pressure drop is expected to be different. Therefore, according to equation 5-3 samples with lower pressure drop will result in better filter quality factor, figure 5-6.



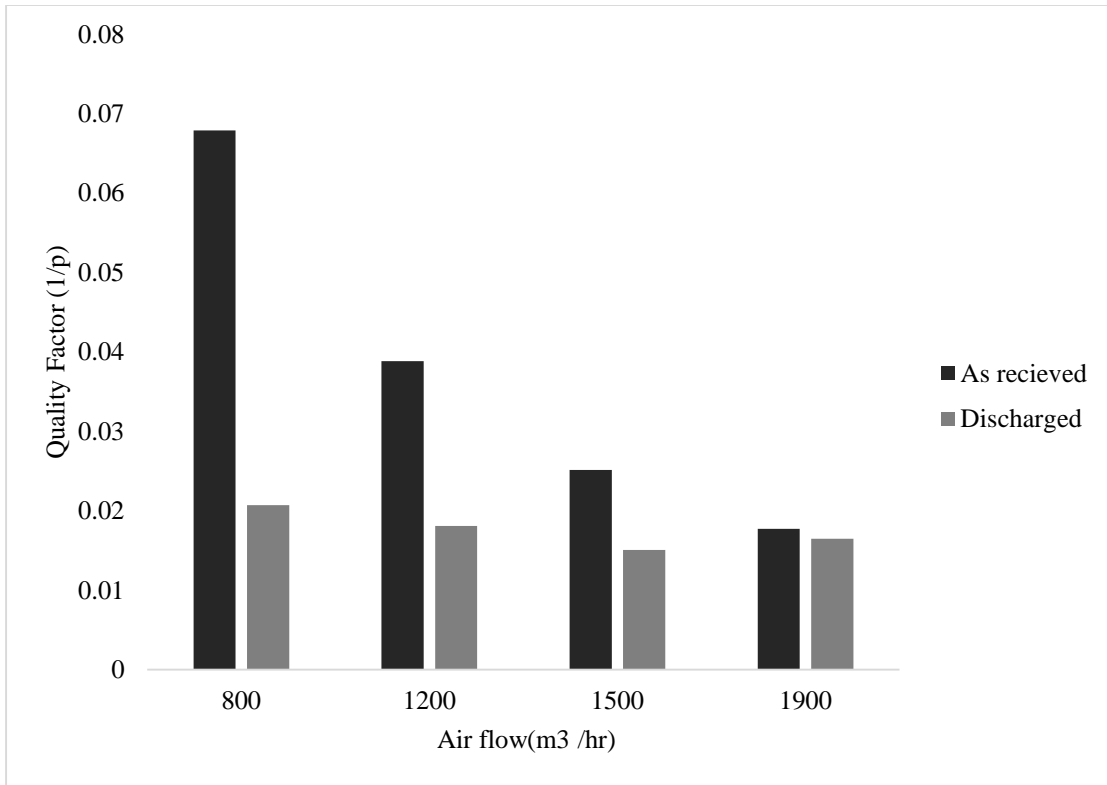


Figure 5-6 Quality factor of PLA meltblown media as received and discharged (40 gms, DCD= 25 cm, 0.6 ghm)

### 5.3.4 Electret filtration performance

As previously mentioned, finer fibers in filter media enhance mechanical filtration efficiency by increasing solidity and providing larger surface area. Larger surface area also benefits electrostatic capture mechanism as fibers with larger surface area could carry higher amount of charges resulting in stronger electrostatic field and thus higher attraction of particles. Figure 5-7 shows filtration efficiency of PLA filter media after and before applying electrostatic charge. Filtration efficiency of PLA filter media increases with decrease in fiber diameter.

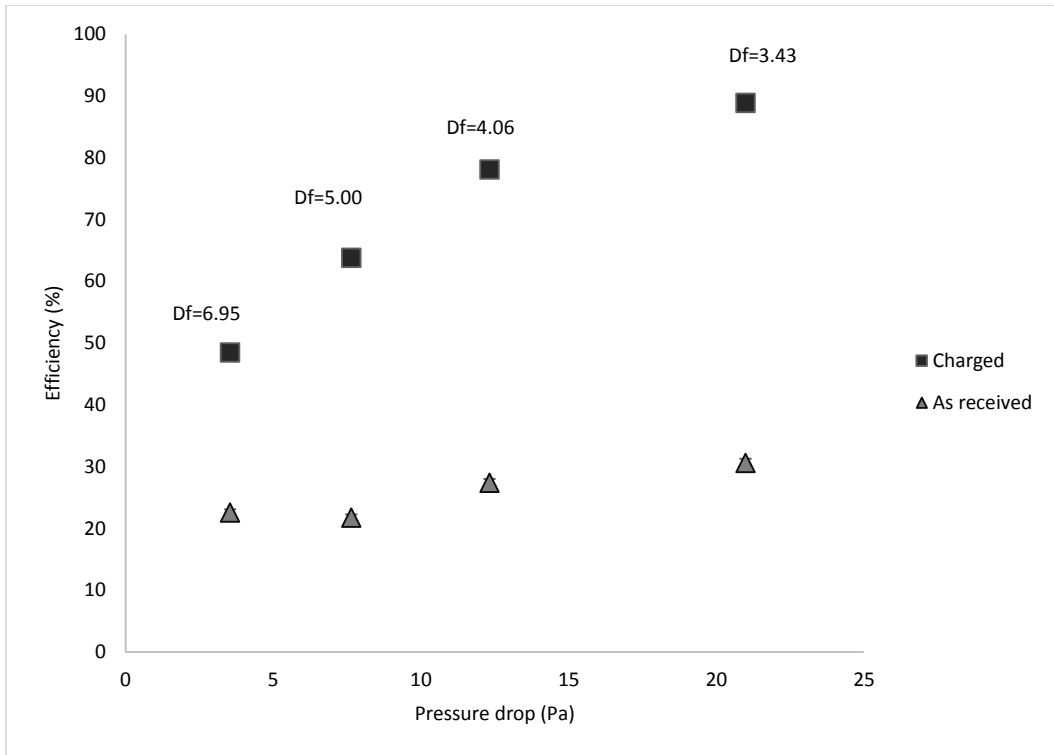


Figure 5-7 Effect of charging and fiber diameter on filtration efficiency of PLA 6260 meltblown media (0.6 ghm, DCD=25, 40 gsm)

On the other hand, decrease in fiber diameter and increasing surface area results in higher pressure drop across the filter. Figure 5-8 shows quality factor for filter with different average fiber diameter before and after charging. Figure 5-8 indicates that for 0.3 $\mu$ m, quality factor increases with increase in fiber diameter meaning that increase in filtration efficiency due to higher surface area provided by smaller fibers is not enough to overcome and offset the extra pressure drop caused by the same smaller fiber size even with assistance of electrostatic attraction.

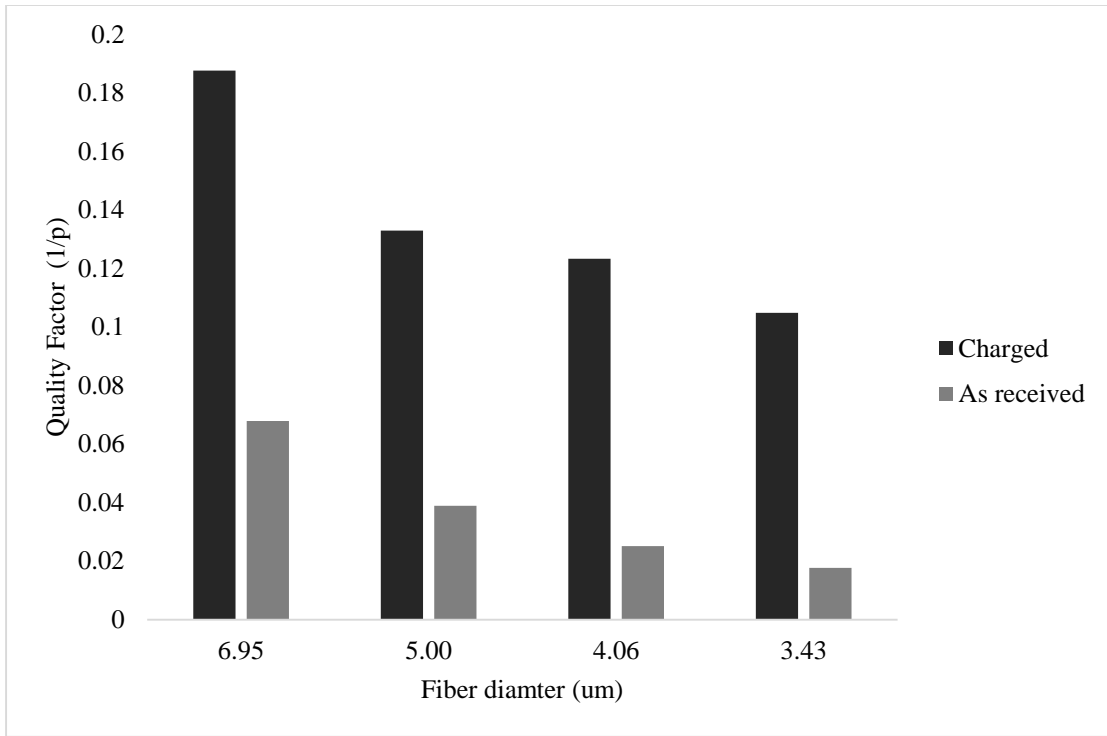


Figure 5-8 Quality factor of as received and charged PLA 6260 meltblown media with different fiber diameter.

## Conclusion

Effect of melt viscosity on morphology, fiber diameter and web properties and filtration performance were analyzed. It is found that lower melt viscosity PLA resulted in lower average fiber diameter within the range of typical meltblown fibers. Both PLA with similar range of D-isomer in the structure resulted in similar crystallization behavior and crystallinity. Fiber diameter affects solidity and air permeability of resulted web and thus filtration performance. Smaller fiber size results in higher surface area and lower fiber to fiber distance in the structure which increase the mechanical and electret filtration efficiency. However, the higher filtration efficiency comes at a cost of higher pressure drop reducing quality factor of resulted filter media.

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## **CHAPTER 6**

### **COMPARATIVE STUDY OF FILTRATION PERFORMANCE OF POLYLACTIC ACID AND POLYPROPYLENE MELTBLOWN AIR FILTER MEDIA**

## 6.1 Introduction

Over the past 20 years demand in filtration and separation industry has been growing and expanding steadily beyond economic situations (E Gregor, 2015). As previously mentioned meltblown technology is able to manufacture nonwoven fabric with microfibers in the structure with low cost and diversity of final products. . Most of polymeric material used in meltblown technology are petroleum based and more than 90% percent of meltblown products are made of polypropylene (Yu et al., 2015). Application of polypropylene for filtration application has been widely investigated (Song, Zhou, Wang, Kang, & Cheng, 2012; Z. Z. Yang, Lin, Tsai, & Kuo, 2002). Having low melt viscosity, low dielectric constant, and low cost compared to other synthetic materials make polypropylene suitable for meltblowing process and filtration application (Dutton, 2008). On the other hand, environmental concerns is driving the industry towards application of use of sustainable materials such as PLA.

In previous chapter, processability of PLA polymer in meltblown system and effect of polymer properties such as chemical structure, nucleating agent, and melt viscosity as well as meltblowing processing parameters on fiber and web formation investigated. In addition, filtration performance of PLA meltblown media regarding effect of all above-mentioned parameters analyzed.

In this chapter, we aim to compare structure and filtration performance PLA and PP meltblown air filter media in terms of mechanical and electrostatic efficiency and charge decay behavior over the time. In order to compare filtration properties of filter media made of different polymers it is necessary to have similar structure such as fiber diameter and solidity. PLA inherently possess high melt viscosity which results in higher range of fiber diameter. In order

to produce PP fiber similar to PLA fibers, we chose PP meltblowing grade with relatively low MFR (higher melt viscosity) to produce fiber and media similar to PLA media.

## **6.2 Experiments**

### **6.2.1 Material**

In order to compare filtration performance of PP and PLA filter media, both media must have relatively similar structure such as fiber diameter which a very important parameter in filtration according to capturing mechanisms. Low molecular weight PLA associate with lower melt viscosity and MFR value of 65 ( 10 min at 210 °C) provided by Natureworks LLC and PP 650W with MFR values of 500 supplied by Exxon chemical used to produce meltblown filter media.

### **6.2.2 Meltblown filter media production**

The Pilot scale Reicofil-Exxon die 35 (Hole/Inch) meltblowing line of The Nonwoven Institute at North Carolina State University utilized to produce PP and PLA meltblown media. PLA chips were dried for 8 hrs at 80 °C in a vacuum dryer in order to eliminate the moisture to minimize hydrolysis during the melt blowing.

In meltblowing process, in order to achieve different fiber diameter distribution and solidity we changed polymer throughput, airflow, and die to collector distance and kept basis weight constant (40 gm<sup>-2</sup>). For both PLA and PP, polymer throughput were kept at 0.3 and 0.6 g/hole/min. Airflow of 800, 1200, 1500, and 1900 (m<sup>3</sup>/hr) used to impact on fiber size distribution, table 6-1 and table 6-2.



Table 6-1 PLA meltblown filters processing condition.

Sample	DCD (mm)	Throughput (ghm)	Air (m <sup>3</sup> /hr)	Basis weight (gsm)
P1	25	0.6	800	40
P2	25	0.6	1200	40
P3	25	0.6	1500	40
P4	25	0.6	1900	40
P5	25	0.3	800	40
P6	25	0.3	1200	40
P7	25	0.3	1500	40
P8	25	0.3	1900	40
P9	15	0.6	800	40
P10	15	0.6	1200	40
P11	15	0.6	1500	40
P12	15	0.6	1900	40
P13	15	0.3	800	40
P14	15	0.3	1200	40
P15	15	0.3	1500	40
P16	15	0.3	1900	40

Table 6-2 PP meltblown filters processing condition

Sample	DCD (mm)	Throughput (ghm)	Airflow (m <sup>3</sup> /hr)	Basis weight (gsm)
N1	25	0.3	800	40
N2	25	0.3	1200	40
N3	25	0.3	1500	40
N4	25	0.3	1900	40
N5	25	0.6	800	40
N6	25	0.6	1200	40
N7	25	0.6	1500	40
N8	25	0.6	1900	40
N9	15	0.6	800	40
N10	15	0.6	1200	40
N11	15	0.6	1500	40
N12	15	0.6	1900	40
N13	15	0.3	800	40
N14	15	0.3	1200	40
N15	15	0.3	1500	40
N16	15	0.3	1900	40

### 6.2.3 Fiber Diameter Measurements

In order to analyze the fiber morphology and measure the fiber size distribution, piece of each sample were sputter coated with a thin layer of gold and magnified images of fibers were taken at 1500X by a scanning electron microscope (Phenom FEI). More than 150 fiber

diameter measurements were taken using Image J software from 15 pictures for each meltblown samples and average fiber size and fiber size distribution determined.

#### **6.2.4 Air Permeability Measurement**

Air permeability of meltblown samples were measured using Textest air permeability tester according to ASTM D 737, in analytical lab of the Nonwovens Institute in North Carolina State University. 10 replications of each sample were tested and average reported.

#### **6.2.5 Discharging**

In order to study mechanical filtration and electrostatic capture efficacy separately, any charge and electrostatic effects should be eliminated. Therefore, discharging as received filter media after basic filtration performance measurements were carried out by immersion in isopropyl alcohol (IPA) bath for two hours and then dried under hood in room temperature for 24 hours.

#### **6.2.6 Corona Charging**

Corona charging technique was used to charge meltblown samples. Sample were put between two electrodes at the optimum condition that described in chapter 3. Samples were charged for 5 seconds at the voltage of +22 kV with charging distance of 3 cm.

#### **6.2.7 Filtration Performance Measurement**

Filtration performance of meltblown webs such as capture efficiency and pressure drop studied using 0.3  $\mu\text{m}$  monodisperse DOP (dioctyl phthalate) particles at face-velocity of 5.3cm/s. Automated filter tester machine, TSI 3160, utilized to measure filtration performance in which certain particle size is generated by a collision type atomizer and a classifier, classifies the

particles based on their electrical mobility and after neutralization particles are fed into the filter holder.

In order to evaluate, filter is installed on the filter holder and two condensation particle counters at upstream and downstream count the number of the particles and aerosol penetration or capture efficiency is calculated. Pressure drop is also calculated by mass flow meter. For each meltblown media six samples were cut and filtration performance were measured and the average of results were reported.

## **6.3 Result and Discussion**

### **6.3.1 Solidity and air permeability**

Nonwoven media is able to create a stable structure with low solid volume fraction which translates to high air permeability. Possessing high air permeability is a key element for air filtration application to have low resistance against airflow.

It is crucial to mention that each polymeric raw material has its own properties and behavior and resulted meltblown webs would have different characteristics. Melt viscosity or onset of crystallization can affect properties of final web in terms of fiber diameter or solidity. In addition to intrinsic properties of each polymer, processing parameters of each polymer in terms of machine setting is different. For example die temperature and hot air temperature varies based on the polymer melting point. Unlike melt spinning that a physical drawing attenuates the fibers, in meltblowing air drag force is responsible for attenuating fibers after coming out of spinneret and thus to some extent meltblowing is not a controllable process and change in the processing condition such as die/air temperature, polymer mass flow (throughput), airflow, and DCD results in web with different properties.

Table 6-3 PP and PLA web properties

Sample	Fiber diameter ( $\mu\text{m}$ )	Thickness ( $\mu\text{m}$ )	Solidity
P10	5.8	324.4	0.093
P11	4.7	293.4	0.102
P12	3.7	293.7	0.105
N10	5.1	531.7	0.087
N11	3.9	373	0.117
N12	3.4	340.4	0.133

Table 6-3 summarizes characteristic of chosen PP and PLA air filter media. Comparing PP and PLA samples with similar basis weight (40 gsm). It can be seen that despite similar average fiber diameter and processing condition all PP samples have higher thickness and solidity than PLA samples. Higher thickness of PP media can be attributed to density difference and mechanical properties of PP and PLA polymers. In Meltblown process, adjustment of basis weight is based on polymer weight feed into extruder at a given time. Regarding density of PLA ( $1.26 \text{ g/cm}^3$ ) and PP ( $0.9 \text{ g/cm}^3$ ) in order to reach same basis weight, higher volume of feed was sent per unit of time for PP as it has lower density comparing to PLA. Therefore, PP web would have higher amount of fibers in the structure at the same volume of fabric comparing to PLA to reach the targeted basis weight resulting in higher web thickness. Considering similar average fiber size of the samples and higher amount PP fibers meltblown web will have lower fiber-to-fiber distance in the structure leading to higher solid volume fraction of resulted web. Figure 6-1 shows solidity difference of both polymers at different

processing condition. It can be observed PP media exhibits higher solid volume fraction comparing to PLA media.

Mechanical properties of each polymer also affects deformation of fibers in the structure web during lay down on the collector (Tausif, O'Haire, Pliakas, Goswami, & Russell, 2016). In meltblown process molten polymers between die tip and the collector are hit by high-velocity hot air jets to attenuate and form microfibers. In addition to attenuating fiber high velocity air will compress fiber of on the collector resulting in more entanglement of fiber (Rungiah et al., 2017b). Depending on the mechanical properties and deformation of polymer the entanglement rate would be different. Comparing mechanical properties of PLA and PP, PLA possess higher flexural rigidity and is stiffer than PP (Pickering, 2008 ). Higher stiffness result in higher resistance to bending of fibers whilst they are laying down on the collector under influence of high-velocity air. Subsequently entanglement of fiber would be lower for PLA resulting in larger pore size distribution. Larger pore size and lower solidity results in higher air permeability of filter media. In addition, PLA and PP differ in their glass transition temperature which results in different solidification of fibers from rubbery state to solid state. PLA and PP have glass temperature around 58 °C and below 0 °C respectively. Therefore, molten PLA fiber will solidly faster than PP fibers and result in less entangled and diffused fibers in the web .

Figure 6-2 displays air permeability of PP and PLA meltblown media with similar fiber diameter and processing condition.

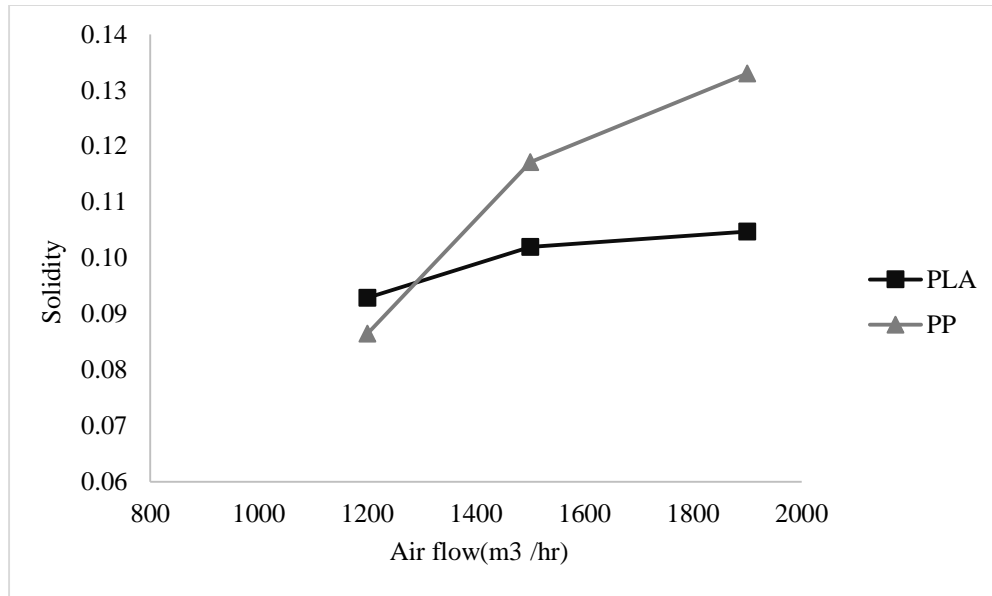


Figure 6-1 Solidity of PLA and PP meltblown webs (DCD=15, 0.6 ghm, 40 gsm)

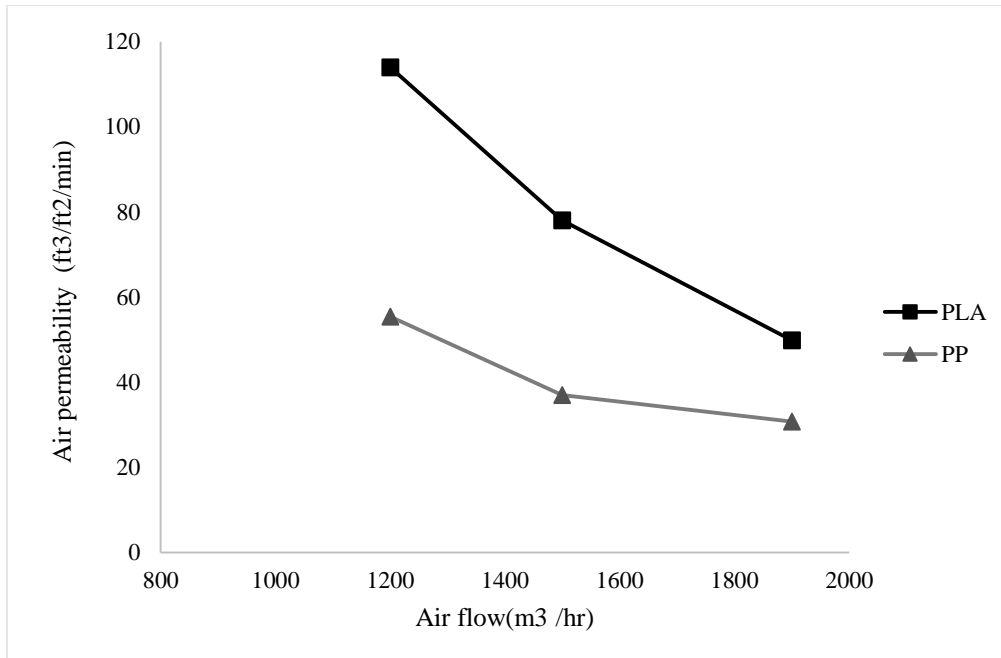


Figure 6-2 Air permeability of PLA and PP meltblown webs (DCD=15, 0.6 ghm, 40 gsm)

### 6.3.2 Filtration performance

It is crucial to mention that each polymeric raw material has its own properties and behavior and resulted meltblown webs would have different characteristics. As previously discussed, PP and PLA have different intrinsic properties and reaching to similar meltblown web structure would be difficult and comparison of functionality of resulted webs would not be accurate. In particular in filtration application, structure of web significantly affects the performance of the media as fiber diameter, pore size, and solidity play important role in capturing particles as well as influencing on pressure drop across the filter media.

In order to be able to compare filtration performance of PP and PLA meltblown air filter media, regarding the difficulties and almost impossibility of creating identical structure and properties,



similarity of fiber diameter and processing parameter were chosen as a measure of similarity. As mentioned previously were able reach finer fiber for PLA 6260 in the range of typical meltblown and close to those of PP 650W although webs structure such as air permeability and solidity were different.

Figure 6-3 shows as received filtration performance of PP and PLA meltblown media with almost similar fiber diameter and processing condition at different thicknesses. Different layers of meltblown media stacked on top of each other to create filters with different thickness without affecting packing density of webs. All the meltblown filter media exposed to  $0.3 \mu\text{m}$  DOP particle at a face velocity of 5.3 cm/s. Table 6-4 summarizes the web and fiber properties of chosen PP and PLA media. It was shown that PLA meltblown media had higher air permeability in comparison PP media. This difference is more obvious and tangible by looking at pressure drop at different thicknesses. Comparing PP and PLA media with similar fiber diameter at any thickness it was observed that PLA media have lower pressure drop which can be attributed to lower solidity or solid volume fraction of filter media and thus lower air resistant through filter. Although PLA media showed lower pressure drop we observed PLA media is having much higher filtration efficiency. Based on mechanical filtration mechanisms, the lower the fiber diameter the higher the filtration efficiency since the chance of collision between fiber and particles increases. However, PLA meltblown media with similar range of fiber diameter and lower solidity have higher capture efficiency. This gap in efficiency is more obvious for PLA and PP media with larger fibers. Comparing filtration performance of P10 and N10 samples we can see that P10 at any thickness is having filtration efficiency of twice of those of N10 and almost half amount of pressure drop indicating better performance of PLA

media. Considering lower solidity and higher permeability of PLA media is filtration efficiency results do not comply with filtration theories and mechanism.

Table 6-4 Characteristic of PLA and PP meltblown media

Sample	Fiber diameter (um)	Thickness (um)	Solidity	Air permeability (m3 /hr)
P10	5.8	324.4	0.093	114
P12	3.7	293.7	0.105	50
N10	5.1	531.7	0.087	55.4
N12	3.4	340.4	0.133	30.7

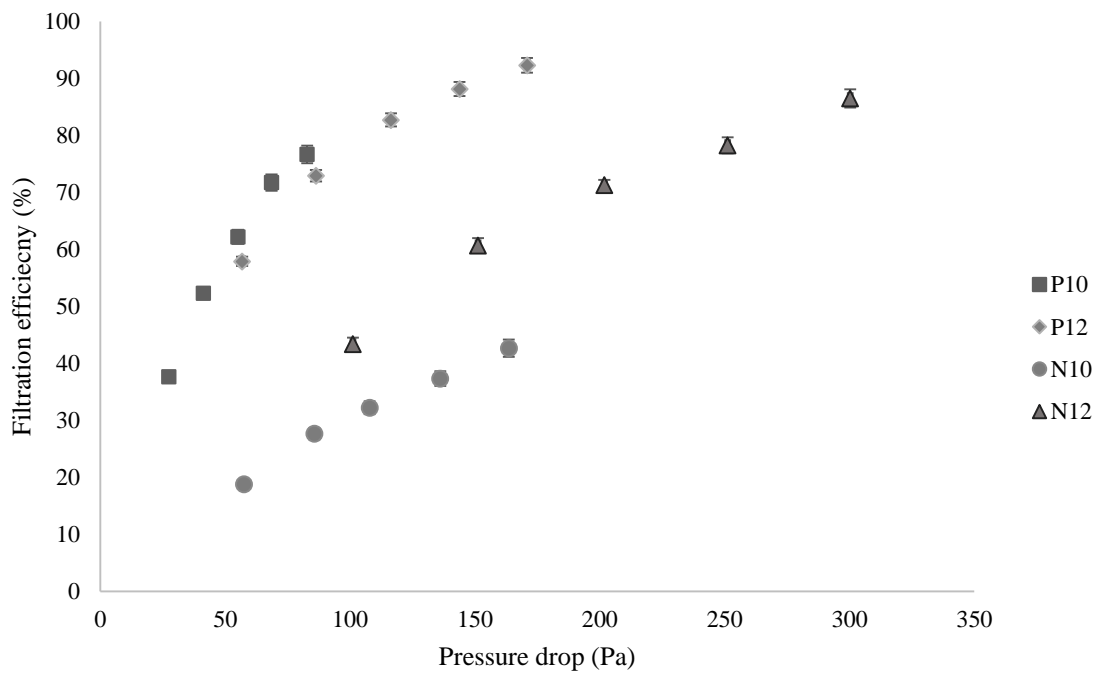


Figure 6-3 Filtration efficiency vs Pressure drop of PLA 6260 and PP 650W meltblown filters.

The quality factor of PP and PLA filter media highlights the difference between performances of these filters. Quality factor is important regarding energy usage and cost benefits. In

addition, in some applications such as respirators high air resistance may cause physiological problem for the users. Quality factor of meltblown filters of both polymers is shown in figure 6-4. As received PLA media showing better quality than PP filter media due to higher filtration efficiency and lower pressure drop.

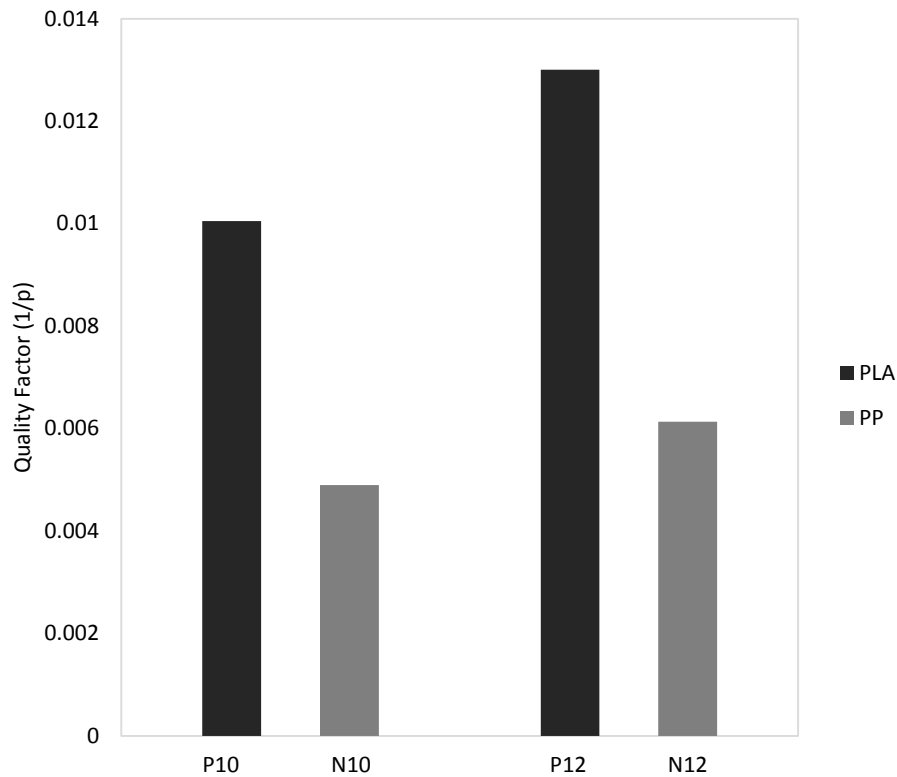


Figure 6-4 Quality factor of PLA 6260 and PP 650W meltblown media.

In chapter 3 it was shown that electrostatic charge during the meltblowing process enhanced filtration efficiency of PLA filter media. Presence of electrostatic charge improves filtration efficiency with no change in pressure drop which allows designing filters with more open structure and still high filtration efficiency and lower pressure by acting on not only charge particles but also neutral ones. Electret filtration performance of fibers depends on the injected

charge and polarization. PP and PLA are both non-polar polymers and therefore electrostatic properties are dependent on injected charge and their ability to trap the charge in their structure. Electret filtration performance PLA and PP meltblown media investigated by inducing charge to fibers using corona charging technique. As mentioned comparing filtration performance of meltblown media is difficult particularly when different polymers are involved. To be able to compare electret filtration performance of PLA and PP media we chose samples with same processing condition and similar fiber diameter as fiber size play an important role in air filtration process. Table 6-5 summarizes the web properties of chosen samples.

Table 6-5 characteristics of PLA and PP meltblown media

Sample	Fiber diameter (um)	Thickness (um)	Solidity
P13	4.4	299.7	0.105
P14	3.1	269.6	0.115
P15	3.0	278.9	0.113
N13	3.6	389.4	0.113
N14	3.2	311.4	0.140
N15	2.8	316.5	0.144

Figure 6-5 shows as received and electret filtration performance of PLA and PP meltblown air filters. As summarized in table 6-5, PP media has higher solid volume fraction and thickness resulting in higher air resistance and pressure drop. As figure 6-5 displays charging enhances filtration efficiency of samples dramatically regardless of polymer type or fiber structure.

As received filtration, efficiency of both samples were quite similar even though PLA samples had more open structure. Considering electrostatic filtration efficiency, it can be seen that PLA filter media is relatively performing better. Specially for samples with larger fiber size in which electrostatic filtration mechanism is more dominant comparing to samples with smaller fiber size as lower fiber diameter improves mechanical filtration efficiency. Comparing as received and electret filtration efficiencies of P13 and N13, after applying electrostatic charge P13 showing higher filtration efficiency and lower pressure drop indicating it is performing better than N13 samples when challenged by 0.3 micron particles. The same results extend to P14 and P15. Regarding structural difference of PLA and PP media used in the study it appears that PLA media is able to create stronger electric field and therefore stronger electrostatic filtration mechanism and higher filtration efficiency. According to this result we can say PLA fibers are showing better or at least same chargeability as PP fibers.

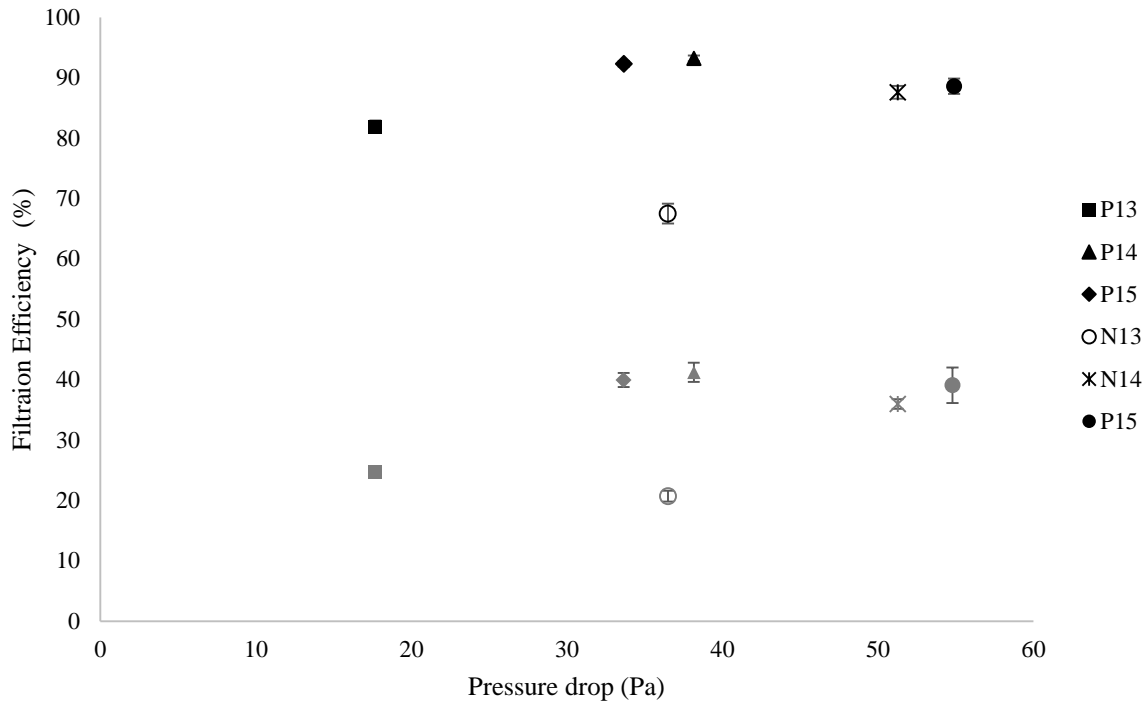


Figure 6-5 filtration efficiency vs Pressure drop of charged PLA 6260 and PP 650W meltblown.   
 Corona Charged       As received

A desired filter is the one that is able to keep the charge and so electrostatic mechanism for a long period and hence a high filtration efficiency. However, the strong electric field at the vicinity of fibers tends to weaken over the time and operation condition. Different studies have investigated effect of different parameter on charge decay and lifetime of electret filters. Filtration efficiency of electret filter decreases over the time because of decrease in electrical components and neutralization of charged fibers by particles and environmental condition (Ieda, Sawa, & Shinohara, 1967; Plopeanu et al., 2011). Humidity can reduce filtration efficiency of filters by weakening the electric field (Herous, Nemamcha, Remadnia, & Dascalescu, 2009). There is not an established method to assess and evaluate charge decay of

electret filters. Study of charge decay at ambient condition is time consuming since at lower temperature polymers chains and molecules are immobile and no tangible change can occur in that condition (Kilic & others, 2012) . Therefore, in order to study charge decay behavior of fibers and accelerate rate of charge loss we tried to accelerate the process high temperature. At elevated temperature charges regain their freedom of motion and their mobility will lead to ease of decay. To compare PP ad PLA filter media we chose filter with similar fiber diameter and electret filtration efficiency and put them in the environmental chamber at the temperature of 40 °C and 80% relative humidity. Table 6-6 summarizes PP and PLA filter media characteristics.

Table 6-6 PP and PLA filter media characteristics

Polymer	Thickness (um)	Solidity	Df (μm)	Efficiency (%)	$\Delta P$ (pa)
P15	278.9	0.115	3	92	39.5
N15	316.5	0.144	2.8	91	58.7

Figure 6-6 shows initial electrostatic filtration performance and charge stability over the time. Here filtration efficiency is used as a measure of charge stability evaluation in which change in filtration efficiency is considered as change in the strength of the electric filed generated by electret fibers. For both filter media at the beginning of exposure to higher temperature and humidity initial charge loss or drop in filtration efficiency is attributed to neutralization of surface charges and shallow trapped charges which can be conducted away by water

molecules. For both, filtration efficiency decrease with an initial sharp slope within first 6 hrs of exposure to high temperature and relative humidity. Efficiency decreased by 10% of initial value but after the rapid decay at the beginning both samples maintained their efficiency and charge in the structure. Both polymers are hydrophobic and they have similar crystalline structure (helix) so the deep charging sites in the structure could be similar. Comparing trend in both filter media we can see that in term of charge decay or losing filtration efficiency over the time, these two media are following similar trend and behave similar to each other. Again it should be noted that structure of selected PLA media is more open due to lower solidity meaning lower amount of charged fibers.

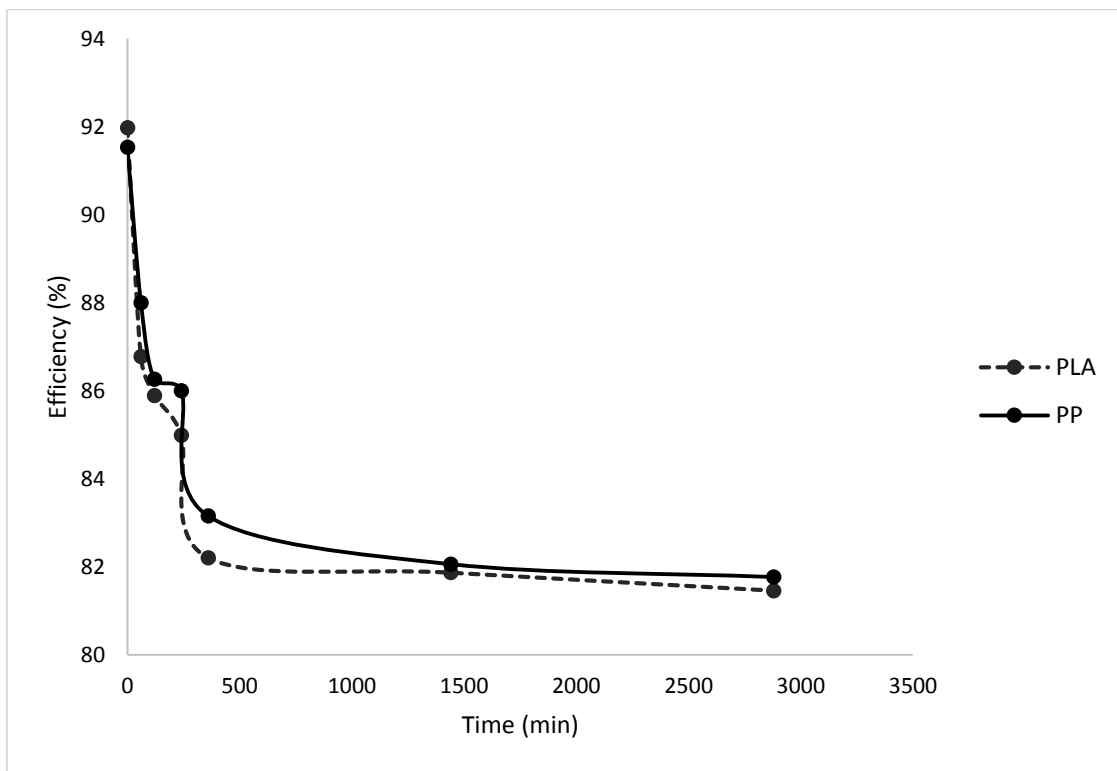


Figure 6-6 charge decay behavior PLA and PP meltblown filters over time.



## **Conclusion**

By choice of rather high melt viscosity PP meltblown grade and low melt viscosity PLA we produced PP and PLA meltblown web with similar range of fiber diameter. Although we were able to reach similar range of fiber diameter, structure of web varies as polymers have different thermal behavior. PLA meltblown webs tend to have higher air permeability and lower solidity comparing to PP webs which is favorable for filtration application. Higher air permeability of webs results in filters with lower pressure drop. Regarding capturing efficiency, since as-received PP meltblown media is getting charged in production process is showing better filtration efficiency. Lower pressure drop and higher capturing efficiency leads to better filter quality of PLA meltblown webs. Regarding electret filtration efficiency result it can be concluded that PLA fiber are able to create stronger electrostatic field which leads to stronger electrostatic capture mechanism and higher chargeability. In addition, both polymers are showing similar behavior in charge stability and decay rate over time in elevated temperature and relative humidity.

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## **Chapter 7**

### **OVERALL CONCLUSION AND RECOMMENDATION FOR FUTURE WORK**

## 7.1 Overall conclusion

Processability of different grades of PLA in meltblown system and effect of PLA properties as well as meltblowing processing parameters on fiber and web formation investigated. Melt viscosity and chemical composition of PLA plays a key role and fiber diameter and morphology of PLA fibers, respectively. Processability and versatility of resulted web is highly affected by melt viscosity and fiber size distribution PLA filter media. PLA with lower D-isomer content resulted in better crystallinity of fibers. Filtration efficiency results indicated that as-received PLA meltblown webs without presence of electrostatic field are getting charged probably due to triboelectrification.

Addition of nucleating agent into fibers improved crystallinity of PLA fibers. Moreover, it was shown that incorporation of 1% NA into fibers resulted in higher air permeability of media (more open structure). Despite higher air permeability and loose structure, filter media with 1% NA showed better electrostatic filtration efficiency and quality factor indicating NA was effective in improving chargeability of PLA meltblown fibers. Charge stability of PLA fibers improved by incorporation of NA into structure of fiber.

Low melt viscosity PLA showed better processability on meltblown system with traditional Exxon die system. PLA filter media with wide variety of fiber size distribution within the typical meltblown range and structure achieved.

PLA meltblown media compared to PP media with similar processing condition and average fiber size showed similar Mechanical and electrostatic filtration performance but lower pressure drop because of different physical and mechanical properties of PLA and resulted in better quality factor.

## **7.2 Recommendation for future work**

- Further study on crystallization properties by incorporation of polymer inclusions such as Cyclodextrins into fibers and investigating electret properties of PLA fibers.
- Novel bimodal structure of PLA meltblown media for super-efficient filters with low-pressure drop.
- Characterizing process-structure property relationship and filtration performance of PLA/PP bicomponent meltblown filter media.