eXPRESS Polymer Letters Vol.2, No.1 (2008) 49–56
Available online at www.expresspolymlett.com
DOI: 10.3144/expresspolymlett.2008.7

Development of structure and properties during thermal calendering of polylactic acid (PLA) fiber webs

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Received 4 August 2007; accepted in revised form 8 November 2007

Abstract. Due to its thermoplastic and biodegradable nature, poly(lactic acid) (PLA) holds good promise in its increasing use in the form of fibers for medical, agricultural, apparel, upholstery, hygiene, and other applications. Most of the research being done on PLA fibers is to understand their production by melt spinning, solution spinning, and the structure-property relationships during fiber formation. Nonwovens are one of the important forms of the materials into which PLA polymer can be converted to create many useful products. Thermal bonding is the most widely used bonding technique employed to impart strength, and other useful characteristics to the nonwovens. However, there is limited research done to study the behavior of PLA fibers during thermal bonding of nonwovens. Hence the research was carried out to investigate the thermal bonding of nonwovens made from PLA staple fibers. The PLA fibers were carded and then calendered at different temperatures. The webs were characterized for their structure and properties. The observed results are discussed with respect to the investigated processing conditions.

Keywords: *thermal properties, mechanical properties, PLA fiber, thermal calendering*

1. Introduction

With ever increasing use of synthetic polymeric fibers, and their adverse impact on the environment, degradation behavior of the polymer at the end of the life cycle is gaining more importance. This is where the conventional polymers such as polyethylene terephthalate (PET), nylon, and polypropylene used for fiber manufacture have negative impact on the environment since these polymers are not biodegradable. The availability of landfills is becoming scarce due to the increasingly large amounts of waste that is being dumped. The incinerator emissions are also of growing concern in the era of global warming. One of the ways to reduce the negative impact on the environment and keeping the earth greener for longer time is by recycling these products that would otherwise go into landfills. Recycling is easier said than done due to the cost and quality implications involved with many nonwoven products. Under these circumstances, biodegradable polymers hold good promise in various applications so that at the end of the products' life cycle, they will degrade on their own with no harm to the environment. The biodegradable polymers break down in physiological environments by macromolecular chain scission into smaller fragments, and finally into simple stable end-products [1].

Today, a variety of biodegradable polymers are available in the market, both natural and synthetic, such as polysaccharides, proteins, polycaprolactone (PCL), polyhydroxybutyrate-valerate (PHBV) and polyesters such as PLA. PLA is one of the most promising biodegradable polymers due to its good mechanical properties that stem from L-lactic acid, thermoplastic processability, biocompatibility and

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biodegradability. In most applications, it exhibits very good durability as compared to some of the other biodegradable polymers [2]. Compared to PET, it crystallizes faster and to a greater extent. By controlling the isomer content i. e. L-form, D-form and Meso form in the polymer, relatively wider range of properties can be obtained in the fibers. Extensive review of PLA fibers was carried out by Gupta *et al*. [3] covering fiber manufacturing methods, properties development, and various applications. The review elucidates the point that much of the research is confined to fiber manufacture and its properties development. The limited research published on nonwovens processing of PLA fiber includes melt blown nonwovens made out of PLA polymer by Khan *et al*. [4], manufacture of PLA nonwoven fabrics using CO_2 laser thinning method by Akaoka and Suzuki [5], and preparation of cellulosic nonwoven based composites made from PLA staple fibers by Kamath *et al*. [6].

A review published by Dharmadhikary *et al*. [7] discusses thermal bonding of nonwoven fabrics from several fibers. However, there is no report on the behavior of PLA fibers into nonwoven processes, especially during thermal bonding. Bhat *et al*. [8] have shown the importance of thermal bonding process variables in the development of structure-properties in the nonwovens made from polypropylene fibers. Since thermal bonding process is very important in the production of nonwovens, this study was conducted to understand the behavior of PLA fibers during thermal bonding. The main objective of this research was to investigate the effect of bonding temperature during thermal calendering on the structure and properties of PLA nonwoven webs.

2. Experimental details

2.1. Materials and processing

PLA staple fibers were provided by Fiber Innovations Technology (FIT), Johnson City, TN. The fibers were converted to nonwoven webs using the laboratory model SDS Atlas carding machine. The schematic of the carding machine is shown in Figure 1. Weighed amount of PLA staple fibers was fed to the feed section of the carding machine to produce the webs with basis weight of approximately 35 grams per square meter (gsm). Sequences

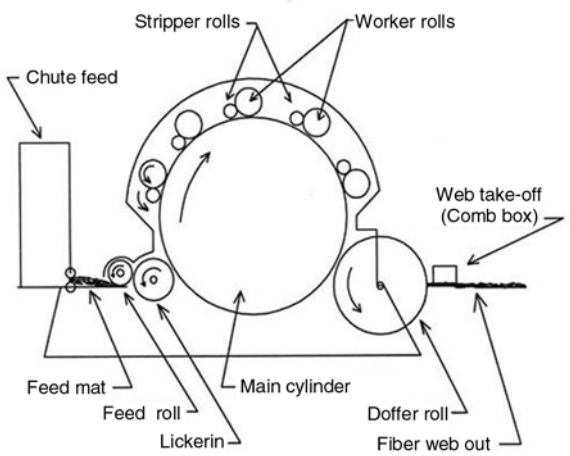

Figure 1. Schematic of the carding machine [9]

of rollers located in the machine individualize the fibers and lay them parallel in the machine direction. The continuous web of fibers coming out of the doffer was made to roll on the rotating drum. When all the material fed was deposited onto the drum completely, the card was stopped and the web was cut. Several webs of 120 cm × 30 cm dimension were thus produced, each with a basis weight of 35 gsm and used for thermal calendering. The carded webs were thermally bonded at series of temperatures above the glass transition temperature and below the melting point of the fibers. The calender rolls speed and pressure were kept constant throughout the experiment. The schematic of the thermal bonding process consisting of a pair of rollers, which are heated to required temperature, through which nonwoven web is passed under desired pressure is shown in Figure 2. The bottom roll is smooth and the top roll is embossed with diamond patterns. Besides the bonding temperature and pressure, the raised patterns on the top rolls

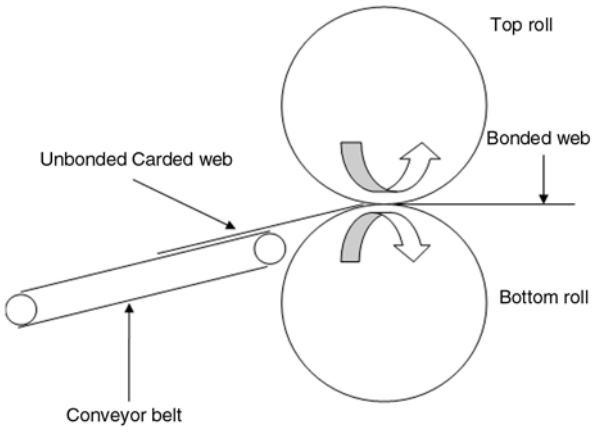

Figure 2. Schematic of the thermal calendering

determine the degree of bonding. Five different calender roll temperatures 130, 137, 140, 145 and 150°C were used, and the calendar speed and pressure were maintained at the same value for all the samples.

2.2. Characterization

Fibers were characterized for tensile and thermal properties. Tensile properties were determined using the Thwing Albert tensile tester with a load cell of capacity 2 kg. Five samples, each as a bundle of 10 fibers, were tested and average of the five readings is reported. Young's modulus was calculated at 5% extension. The gauge length was 25 mm and the test speed was 300 mm/min.

Melting temperature and percent crystallinity of the original fibers and webs were determined using the Mettler DSC at a heating rate of 10°C/min. Heat of fusion for 100% crystalline PLA polymer was taken as 93.7 J/gm [10] for estimating the crystallinity of the samples.

The calendered webs were characterized for various properties such as basis weight, thickness using the ASTM standard D5729-97 [11], thermal analysis by DSC, tensile strength and elongation using the ASTM standard D5035-95 [12], air permeability using the ASTM standard D737-96 [13], tear strength using the ASTM standard D5734-95 [14], optical micrographs and photographs by scanning electron microscopy (SEM).

3. Results and discussion

3.1. Fiber properties

The fiber properties are listed in Table 1. The melting temperature of PLA fiber was determined to be around 164°C (Figure 3). Hence the calendering temperatures were chosen to be below 164°C. The original fiber crystallinity as determined by DSC was around 53.3%.

Table 1. PLA fiber properties

Fiber property	value
Denier [den]	3
Staple length [mm]	76
Tensile strength [gm/den]	2.8
Peak elongation [%]	64.35
Young's modulus [gm/den] (at 5% extension)	31.07
Melting temperature [°C]	164

3.2. Web properties

The basis weights of the webs were found to be around 40 gsm. The change in web thickness against calendering temperature is as shown in Figure 4. As the calendering temperature was increased, the thickness of the webs showed a decreasing trend. At constant calendering pressure and constant calender speed, higher roll temperature lead to better compaction of the web and hence decrease in nonwoven web thickness. This is evident from the optical micrographs shown in Figure 5 which were taken at the same magnification for all the webs. At lower calendering temperatures, the bond point appeared to be hazy due to the inability to focus on the entire thicker bond point simultaneously. With increase in calendering temperature, the picture of bond point became more clear over the entire bond point.

3.3. Thermal properties

DSC study was done to analyse the effect of bonding temperature on the degree of crystallinity

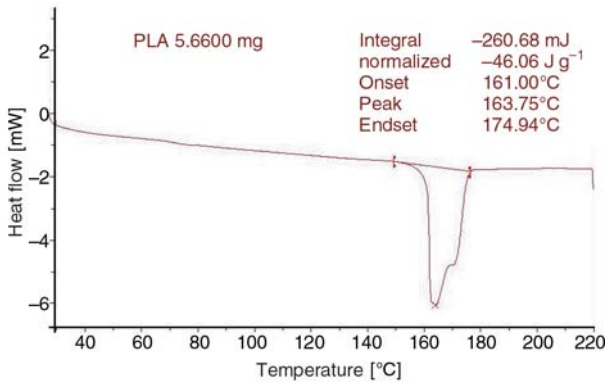

Figure 3. DSC thermogram of original PLA fiber

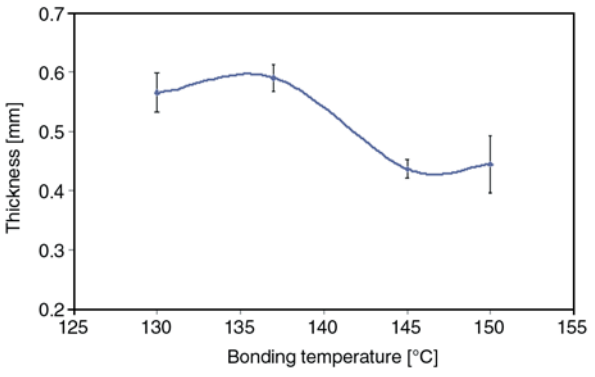

Figure 4. Thickness of the calendered PLA webs

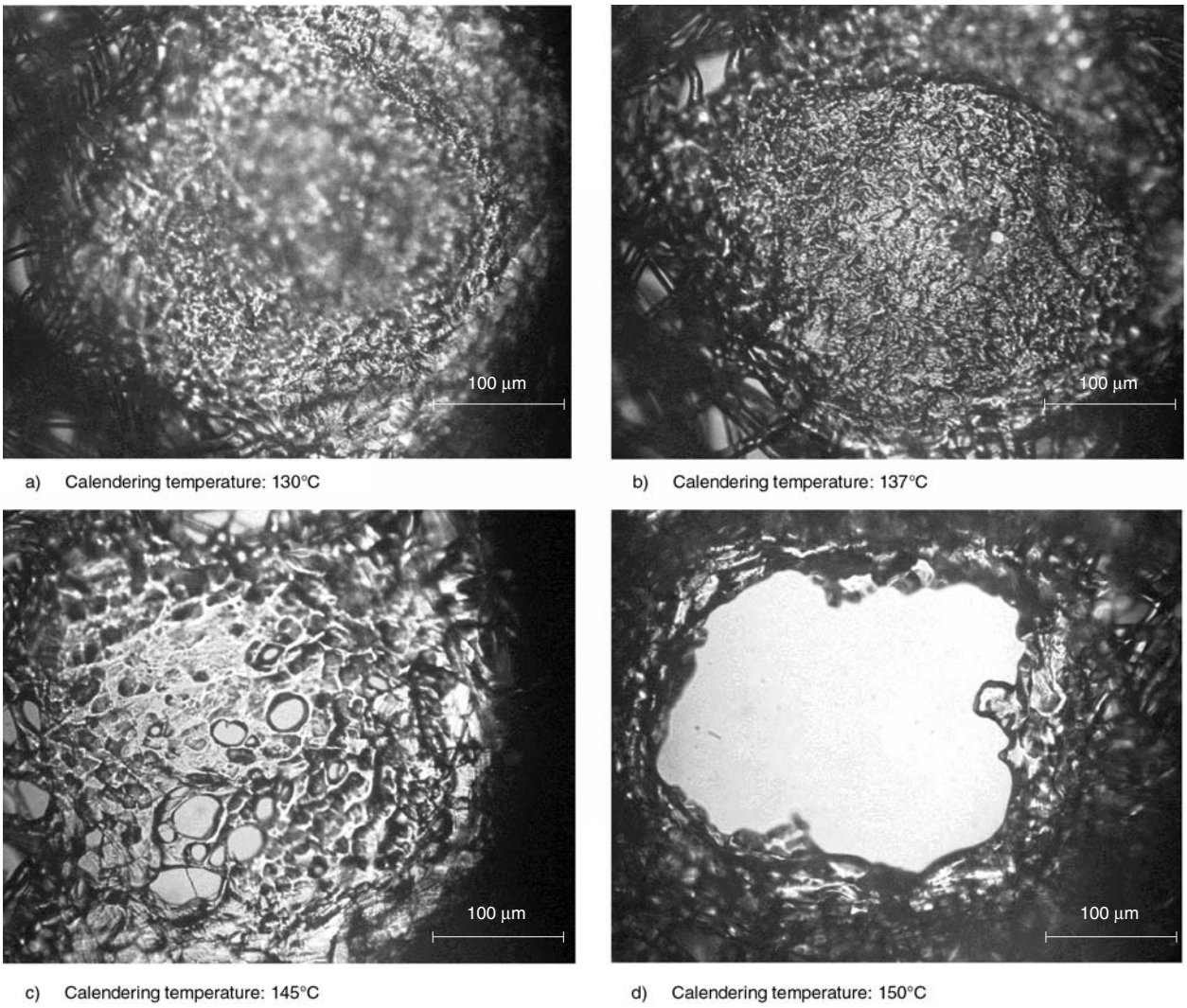

Figure 5. Optical Micrographs of the calendered PLA webs at the bond points

developed in the fibers after thermal calendering and is depicted in Figure 6. The crystallinity of the fibers before thermal calendering was 53.3%. Upon thermal calendering, crystallinity was lower than that in the fiber before calendering for all the bonding temperatures. Similar observations have been reported by Chand *et al*. [15] on thermal bonding of polypropylene webs. They had observed that the crystallinity of the polypropylene webs was increasing if the initial fiber crystallinity was lower and decreasing if the starting fiber crystallinity was very high. Obviously PLA fibers have high crystallinity before calendering and hence during calendering there is reduction in crystallinity due to partial melting and recrystallization in the unoriented conditions. Further there appears to be a decrease in crystallinity with increase in calendering temperature. Mezghani *et al*. [16] reported that

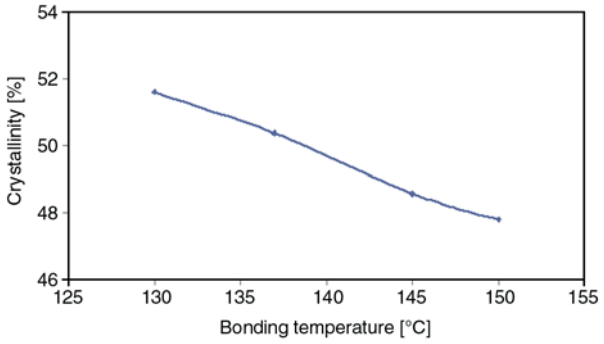

Figure 6. DSC crystallinity vs. calendering temperature of the calendered PLA webs

in the absence of molecular orientation in the melt, the crystallization kinetics of PLA become so slow that the polymer develops amorphous structure. In the present study, at various bonding temperatures, the molecular chains might have tried to relax upon partial melting. As is evident from the optical

micrographs in Figure 5, at higher calendering temperatures holes are apparent in the bond point region, most probably due to relaxation within the fibers, and further shrinkage of the webs. Such stress relaxation might have led to the loss of some of the orientation present in the original fibers and thereby slowering the crystallization kinetics leading to decrease in crystallinity with calendering temperature.

3.4. Tensile properties

The tensile strength of bonded webs was found to increase with increase in calendering temperature with the maximum at 145°C calendering temperature. The tensile strength increased sharply between 137 and 145°C (Figure 7) and above 145°C, the strength started dropping. This behavior shows that 145°C is the optimum calendering temperature for PLA under the given processing conditions. The tensile strength was higher in the machine direction (MD) than in the cross direction (CD) due to better fiber orientation in the web along the MD. The thickness data and the optical micrographs suggest that at lower calendering temperatures the web compaction at the bond point is not good, and exhibits poor bond integrity and tensile strength. With increase in calendering temperature, the bond point compactness was found to be improving, with better bond integrity, leading to increase in tensile strength. Further, optical micrographs clearly indicate that at a bonding temperature of 150°C, the bond quality is very poor. At this temperature, most of the fibers in the bond region were seen to be melted, and due to the stress relaxation by these fibers, the webs shrunk towards the edge of the bond point, creating hole in the bond region. This observation implied that beyond 145°C, over-bonding occurred and led to drop in tensile strength. Of course, the optimum bonding temperature will be shifted slightly, when calendar speed is altered due to change in residence time. Such optimum calendering temperature was also reported for thermally bonded polypropylene nonwovens produced through spun bonding process by Bhat *et al.* [17]. It has been shown that the calendaring pressure has a minimum effect above the minimal value [18].

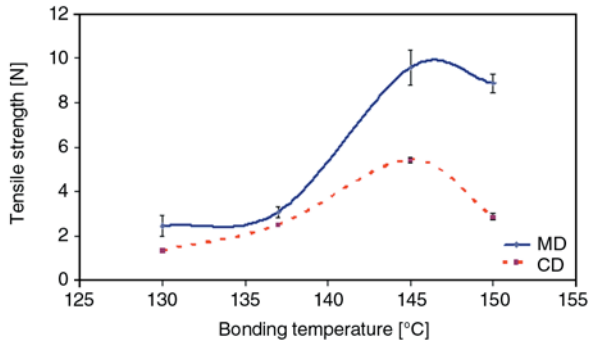

Figure 7. Tensile strength vs. calendering temperature of the calendered PLA webs

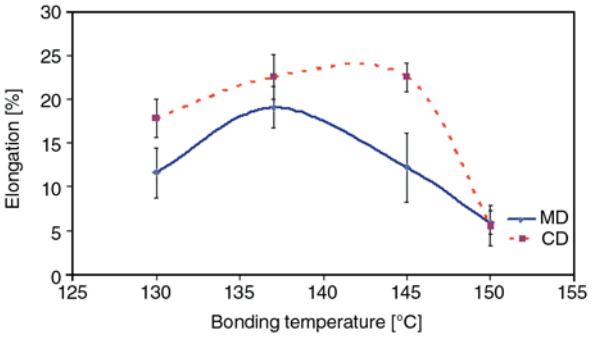

Figure 8. Elongation vs. calendering temperature of the calendered PLA webs

Referring to Figure 8, peak elongation showed a slight increase, and then a decreasing trend with increasing bonding temperature. This is because with increase in bonding temperature, the bond points became stiffer reducing the web elongation. Elongation was found to be higher in the CD than in the MD for the same reasons discussed above for tensile strength.

3.5. Tear strength

The tear strength of the bonded webs in the MD (Figure 9), improved with increasing calendering temperatures reaching its maximum at 145°C. At 150°C, the tear strength values were much lower indicating 145°C as the optimum calendering tem-

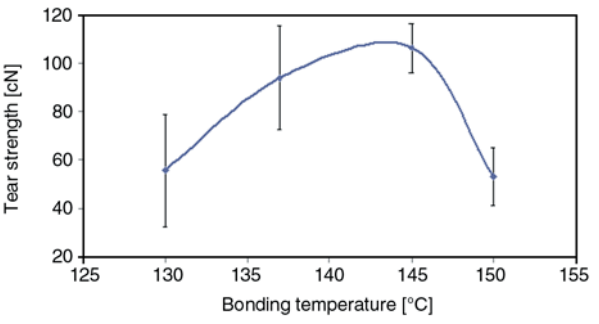

Figure 9. Tear strength vs. calendering temperature of the calendered PLA webs

perature. Comparing Figure 9 with Figure 7, it is seen that tear strength results correlate with the tensile strength results. Such a correlation between tear strength and tensile strength results has been reported by Bhat *et al*. [8] in thermal bonding of polypropylene spunbonded webs. Improved bond integrity up to 145°C and over bonding at 150°C as discussed in above section, might be associated with the observed behavior of web tear strength with respect to bonding temperature. Primentas *et al*. [19] discussed that the mechanism by which tear resistance of the fabric can be improved is by the movement of individual fibers or yarns such that they form bundle at the tip of the tear, share the load and resist the tear propagation further. It is reported that in the tear test, yarns which were tested for tear resistance actually failed in tension [20]. Improving bond integrity increases load sharing between the fibers and hence increase in tear strength is exhibited. However, upon over bonding, the webs became stiffer and hamper the fiber movements to adjust into a bundle, thus reducing the load sharing ability of the fibers in the bonded structure leading to drop in tear strength.

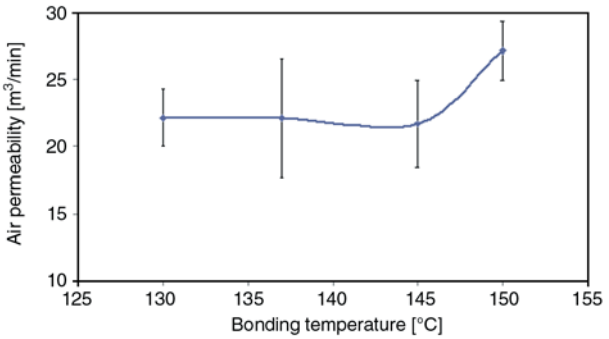

Figure 10. Air permeability vs. calendering temperature of the calendered PLA webs

3.6. Air permeability

Thermally bonded carded webs showed almost the same air permeability with increasing calendering temperatures up to 145°C as shown in Figure 10. However for webs calendered at 150°C, there was a significant increase in air permeability. Since the

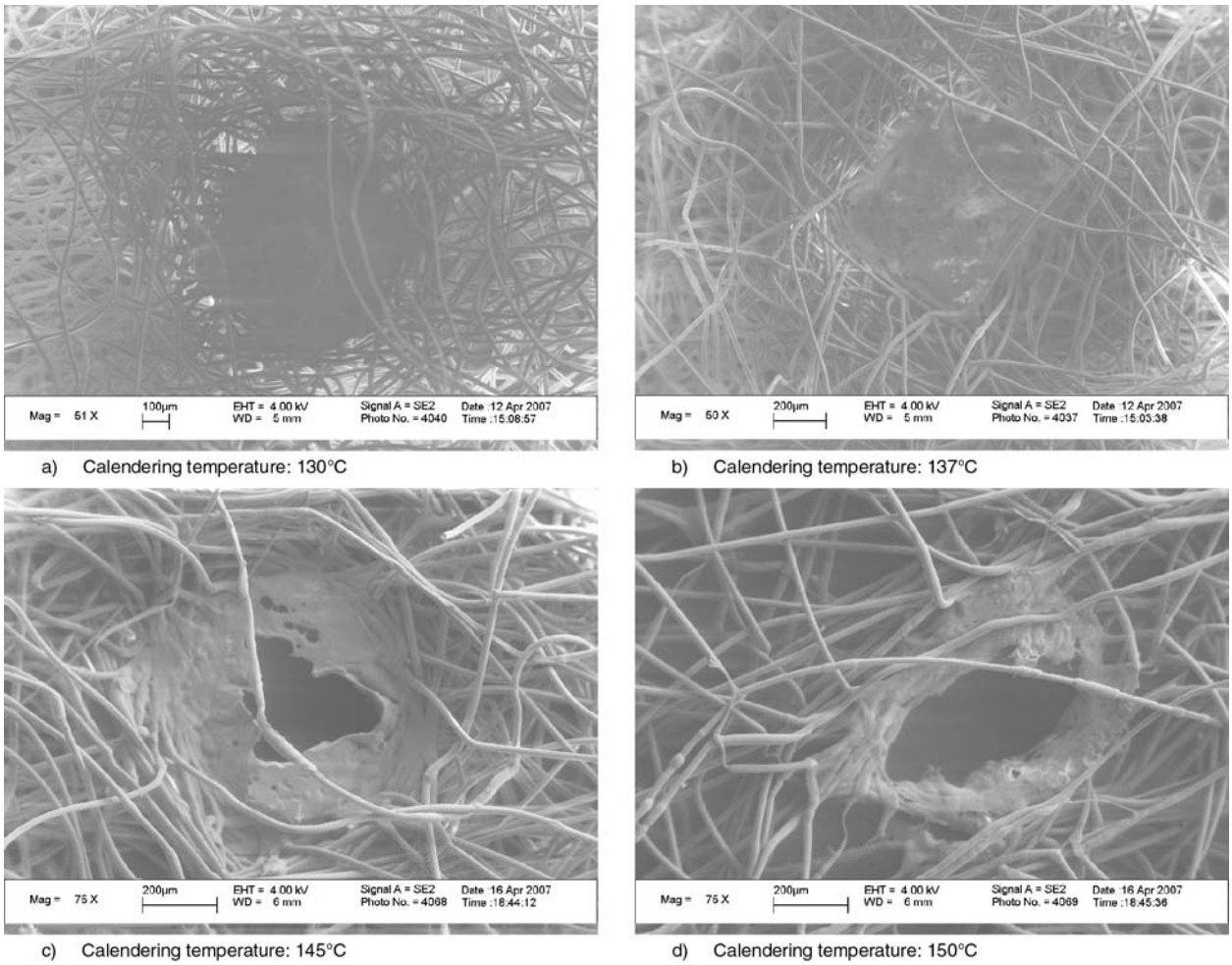

Figure 11. SEM Micrographs of the calendered PLA webs

basis weight of the webs is the same, it is expected that the air permeability remain more or less at the same level. The significant increase in air permeability at 150°C could be explained with the help of the structure at the bond points of the webs as seen in the optical micrographs in Figure 5 and in SEM photographs in Figure 11. Webs produced at 150°C showed big holes at the center of the bond points which could have formed due to the fact that as fibers approach their melting temperature of 164°C, they tend to relax more and more from the built in stresses in the fiber. Such stress relaxation caused the softened material to shrink and move towards the boundary of the bond points, thus leaving a hole in the center. Shrinkage of the softened polymer, and holes formation appeared to be taking place above 140°C, and was seen to be growing in size from 145 to 150°C. Such openness in the bond point area can explain the significant increase in air permeability at 150°C. In a study on the effect of bonding temperature on specialty elastomeric polyolefin spunbonded webs, Dharmarajan *et al*. [21] had observed the hole formation in the case of the webs produced from 100% elastomeric polyolefins. However, they did not discuss the reasons for formation of holes.

4. Conclusions

PLA staple fibers can be converted into nonwoven webs using the carding process and a subsequent thermal calendering. Selection of proper calendering conditions, especially the temperature, is important to produce stronger nonwovens. The optimum calendering temperature was found to be 145°C for the PLA carded webs produced under these conditions. The phenomenon of relaxation, as evidenced from hole formation was observed at higher calendering temperatures. This may be the combined effect of loss of molecular orientation, slower crystallization kinetics, and hence a decrease in crystallinity with increase in calendering temperature. The hole formation at bond point affected the characteristics of nonwoven webs such as air permeability and strength. Overall, strong webs can be produced by carding and thermal calendering, as long as optimum calendering conditions are selected.

Acknowledgements

Authors sincerely acknowledge the support from Fiber Innovations Technology, Johnson City, TN, for supplying PLA fibers used in this study.

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