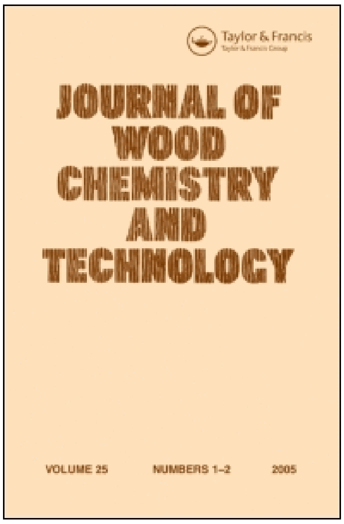

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Electrospinning of Technical Lignins for the Production of Fibrous Networks

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Abstract: Electrospinning is an effective strategy to produce micron and sub-micron diameter fibrous networks from a variety of polymeric systems. Using seven different technical lignins the effect of lignin structure on fiber formation by electrospinning was studied. Surprisingly, none of the technical lignins could be electrospun into continuous fibers, although beaded fiber formation was observed for the softwood Kraft lignin system at high concentration ($>$50 wt%). However, the addition of poly(ethylene oxide) dramatically affected the electrospinning behavior and fiber formation. For all of the technical lignins a clear transition from electrospray or beaded fibers to uniform fibers was observed upon addition of poly(ethylene oxide); the lignin concentration dependent on poly(ethylene oxide) content. In all of the systems a linear increase in fiber diameter with increasing lignin concentration was observed. At the same concentration, the various lignin solutions had varying viscosities and different electrospinning behavior, that is, fiber diameter and ability to form uniform fibers, suggesting lignin specific structures and intermolecular interactions are influencing solution properties and electrospinning behavior. In fact, specific viscosity versus concentration plots reveal scaling exponents', $\eta \sim c^{7.4\text{–}7.8}$ consistent with a branched polymer participating in intermolecular interactions such as hydrogen bonding or association complexes.

Keywords: Lignin, PEO, electrospinning, fibers, viscosity, spinnability

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INTRODUCTION

Lignin-based carbon fibers are an attractive potential application for technical lignins, which are produced in large quantities by the pulp and paper industry.[1–7] Although widely studied, technical lignin-based carbon fiber prepared by conventional melt or solution spinning processes yield relatively weak carbon fibers compared to those derived from petroleum-based pitch and poly(acrylonitrile) (PAN) precursors.[8] One strategy to improve lignin-based carbon fiber properties is to reduce the fiber diameter, which reduces defects and improves molecular orientation along the fiber axis.[7,9–11]

Electrospinning is a well-known process to produce nanometer to micron diameter fibers from polymer solutions using an electric field.[12–14] Linear, high molecular weight polymers in good and theta solvents can readily form networks of entanglements that can deform yet remain intact during electrospinning. Entanglement networks promote fiber formation.[15–17] In certain cases, such as inelastic[18] or unentangled polymer solutions, there is a tendency to form fibers with beads-on-string morphology or break into droplets instead of forming uniform fibers.

In studies on electrospinning of Alcell lignin, it has been reported that a coaxial spinneret system with a sheath layer of solvent (ethanol) was required to stabilize the fluid droplet at the tip of the spinneret during electrospinning.[19–20] Electrospray was observed below a critical sheath fluid flow rate. The tendency to electrospray is consistent with the fact that technical lignins have heterogeneous, branched structures and are degraded to low molecular weight during biomass delignification. However, it is clear from published results that technical lignins have the ability to form fibers by electrospinning when instabilities are dampened.

The addition of poly(ethylene oxide) (PEO) has been shown to facilitate fiber formation of other biopolymers from solution by electrospinning. These include cellulose acetate,[21] keratin,[22,23] alginate,[24] and Chitosan.[25] Furthermore, it has been demonstrated that PEO forms a miscible blend with technical lignins including kraft and Alcell lignins,[26–29] and that PEO improves the ease of fiber formation by fusion spinning.[7] Improved processibility was presumably related to a change in the hydrogen bonding or other systems of interactions resulting from the incorporation of PEO.[29,30] Therefore, it was expected that incorporation of PEO would facilitate electrospinning of technical lignin solutions. Herein we report the effect of lignin concentration, PEO addition and processing parameters on electrospinning of seven different technical lignins.

EXPERIMENTAL

Polymers

Softwood kraft lignin (Indulin-AT, SKL), hardwood kraft lignin (HKL), and sulfonated kraft lignin (Kraftsperse 25M, SL) were obtained from MeadWestvaco

(Glen Allen, VA, USA). Kraft lignins were repeatedly washed with aqueous HCl (pH 2) to exchange sodium counterions, and freeze-dried before electrospinning. Hardwood organosolv lignin, Alcell (HOL) was obtained from Aldrich (Oakville, ON) and used as received. Softwood organosolv lignin (SOL) was prepared by a previously reported protocol.[31] Pyrolytic lignin (PL) was precipitated from bio-oil (Dynamotive Energy Systems, Vancouver, BC) as previously described[32] and subjected to an initial heat treatment at 160°C for 30 minutes under reduced pressure to remove volatile contaminants.[7] Lignosulfonate (Starflo dye dispersant, LS) was obtained from Georgia Pacific (Bellingham, WA). Poly(ethylene oxide) (PEO) with an average molecular weight of 6×10^5 g/mol was obtained from Union Carbide (Houston, TX) and N,N'-Dimethylformamide (DMF) was obtained from Fisher Scientific (Ottawa, ON) and used as received. Weight average molecular weight (M_w) of SKL, HKL, SOL, HOL, and PL were determined using gel permeation chromatography (GPC) and multi-angle laser light scattering (MALLS) (Dawn Helios II, Wyatt Technologies, Inc., Santa Barbara, CA) of acetylated samples dissolved in THF (flow rate = 0.5 mL/min at 35°C).

Solutions

Electrospinning solutions were prepared as follows: The appropriate amounts of lignin and PEO were weighed and the dry powders were mixed using a spatula. The appropriate volume of solvent (DMF for SKL, HKL, HOL, SOL, PL, and water for SL, LS) was then added to reach the desired total polymer concentration, which ranged from 10–50 wt%. Vials containing solutions were then sealed tightly, vortexed for 1 minute, and heated in an oil bath at 80°C. The solutions were again vortexed for 2 minutes after 30 and 60 minutes of heating, and again for 3–4 minutes after 2 hours of heating and allowed cool at room temperature before electrospinning. Because the solutions were optically opaque, the mixing was evaluated by qualitative observation of the flow behavior inside the vial containing the solution to ensure that there were no undissolved solids. Electrospinning was then carried out as described below.

Electrospinning

Electrospinning was carried out in a vertical orientation using a 1 mL syringe fitted with a flat-tip 18 G needle as a spinneret connected to the positive terminal of a high voltage power supply (Glassman High Voltage, Inc., High Bridge, NJ). The operating voltage was varied from 9–14 kV. An aluminum foil collector was placed 14–20 cm below the spinneret and was connected to ground. A syringe pump (New Era Pump Systems, Inc. Wantagh, NY) operating at a flow rate of 0.03 mL/min supplied the polymer solution to the spinneret.

Viscosity Measurements

The viscosities of the lignin solutions with and without added PEO were measured as a function of shear rate over a range of 1–2000 s^{-1} using an AR2000 shear rheometer (TA Instruments, Grimsby, ON) with a cone and plate configuration (60 mm diameter geometry with a 2^{o} cone angle) Specific viscosity was calculated using the mean viscosity over the shear rate range of 10–100 s^{-1}, denoted η, and the relationship $\eta_{sp} = (\eta_{sp} - \eta_{s})/\eta_{s}$, where η_{s} is the solvent viscosity determined by capillary viscometer at 25°C ($\eta_{s} = 8 \times 10^{-4}$ Pa·s for DMF).

Imaging

Fiber morphology was characterized initially by optical microscopy using an Olympus BX41 microscope and/or by SEM analysis (Hitachi S3000N) using gold coated samples, magnification of 500–5000×, working distance of 10–15 cm, and an accelerating voltage of 5 kV. Fiber and particle diameter distributions were generated from SEM images by measuring 15–30 diameters per image using the ImageJ software package (U.S. National Institutes of Health). Diameters are reported as the mean ± standard deviation based on measurements of 100–200 fibers.

RESULTS AND DISCUSSION

Solutions of each technical lignin in DMF (SKL, HKL, HOL, SOL, PL) or water (LS and SL) were prepared over a range of concentrations from 10–50 wt%. Unfortunately, attempts to electrospin the various lignin solutions failed; none were capable of uniform fiber formation, only producing electrospray. However, the SKL/DMF system did display visible evidence of beaded fiber formation at high SKL concentration. Figure 1 shows SEM images of material produced from 40 wt% (Figure 1a) and 50 wt% (Figure 1b) SKL/DMF solutions. Increasing the concentration of SKL from 40–50 wt% clearly shows the transition from electrospray (Figure 1a) to electrospinning with the formation of beaded fibers (Figure 1b). Unfortunately, further increasing the lignin concentration above 50 wt% resulted in highly viscous solutions which produced uneven jetting and caused large droplets to be emitted onto the collector. Nevertheless, it is apparent that the transition to fiber formation is favored by increasing the polymer concentration and solution viscosity that is known to promote fiber uniformity.[15–17]

It is clear that technical lignin structure and properties affect the ability to form fibers, since one of the lignins was able to form beaded fibers and the others electrosprayed. Interestingly, the SKL had the highest viscosity

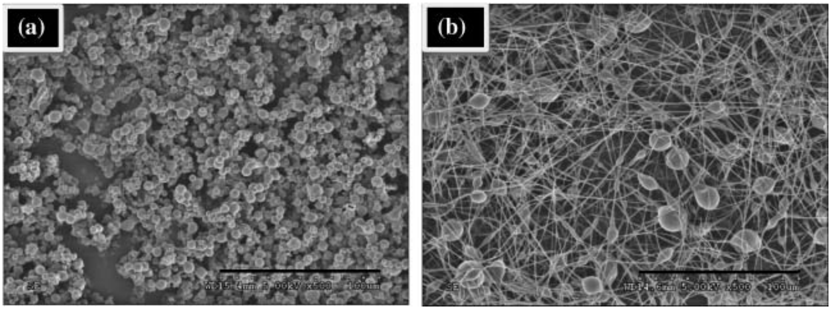

Figure 1. SEM images of SKL solutions electrospun at 40% (a), and 50% (b). Scale bar = 100 μm (500× magnification).

compared to the other lignins at the same concentration dissolved in DMF. Table 1 shows the viscosity for the various lignin/DMF solutions at 30, 40, and 50 wt% concentrations. There is a clear concentration dependence on viscosity, wherein all of the lignin solutions increase in viscosity by an order of magnitude over the range of 30 wt% (~10 mPa s) to 50 wt% (~100 mPa s) with the exception of SKL which increased by over 2 orders of magnitude (~20 to ~2000 mPa s). Since the lignins are of similar molecular weights (~2200–3500 M_w), the large difference in the SKL system may be a result of the differences in the molecular structure and intermolecular interactions as compared to the other lignins.[29, 30] We are currently investigating this further.

It is evident from the literature that poly(ethylene oxide) (PEO) can be used to facilitate biopolymer electrospinning.[21–25] However, in the development of lignin-based carbon fibers it was observed that PEO levels greater than 5 wt% led to fibers fusing together during thermal processing.[7] As a result, the effect of PEO addition on lignin electrospinning was evaluated at lignin:PEO mass ratios of 99:1 and 95:5 from solutions with total concentrations of 20–50 wt%, where concentration in wt% is expressed as% mass of (lignin + PEO) with respect to total solution mass. Figure 2 shows SEM images of electrospun

Table 1. Molecular weights and viscosities of lignin/DMF solutions at 30, 40, and 50 wt%

Lignin	M_w (g/mol)	Dispersity (M_w/M_n)	$\eta_{30\%,\,\mathrm{DMF}}$ ($mPa^*s \pm s.d.$)	$\eta_{40\%,\,\mathrm{DMF}}$ (mPa*s ± s.d.)	$\eta_{50\%,\,\mathrm{DMF}}$ ($mPa^*s \pm s.d.$)
SKL	3700	8.4	24 ± 1.0	158 ± 8	2341 ± 58
HKL	2500	7.6	12 ± 0.4	53 ± 3	427 ± 29
SOL	2200	6.6	7 ± 0.2	22 ± 1	94 ± 1
HOL	2300	7.9	8 ± 0.3	28 ± 0.4	178 ± 4
PL	2600	7.5	6 ± 0.1	21 ± 0.4	127 ± 2

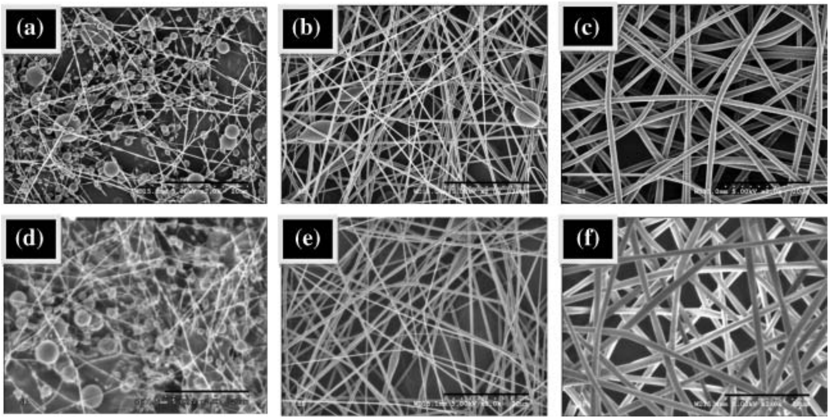

Figure 2. SEM images of 95:5 and 99:1 SKL:PEO fibers electrospun from solutions at different concentrations. (a): 95:5, 20 wt%, (b): 95:5, 25 wt%, (c): 95:5, 30 wt%, (d): 99:1, 30 wt%, (e): 99:1, 35 wt%, (f): 99:1, 40 wt%. Scale bar = 20 μm (2000× magnification).

SKL:PEO solutions. At a lignin:PEO ratio of 95:5, the 20 wt% solution produced beaded fibers (Figure 2a). Increasing the solution concentration to 25 wt% resulted in nearly uniform fibers with a few beads (Figure 2b), while larger diameter uniform fibers were obtained by increasing the concentration to 30 wt% (Figure 2c). Decreasing the SKL:PEO ratio to 99:1, resulted in higher concentrations being required to obtain uniform fibers, but the concentrations were still lower than the 50 wt% required to form beaded fibers for SKL without PEO. At the 1% PEO content, the 30 wt% solutions formed mostly beaded fibers (Figure 2d), the 35 wt% solutions formed fibers with a few beads (Figure 2e), and the 40 wt% solutions formed uniform fibers (Figure 2f). Further increasing the total concentration resulted in increasing fiber diameters, and ultimately the fibers appeared fused at their points of contact, suggesting that solvent evaporation was incomplete.

It is clear that the addition of PEO enables the continuous electrospinning of lignin fibers; in the absence of PEO the 40 wt% solutions of SKL (Figure 1a) or any other lignin in solution only electrosprayed. Moreover, increasing the PEO content reduced the total polymer concentration required for fiber formation. Since lower polymer concentration is typically correlated with reduced fiber diameter,[15–17] we initially expected that lower concentration might produce smaller fibers. However, 95:5 SKL:PEO fibers produced from 30 wt% solutions had similar diameters (1363 ± 234 nm) to those produced from 40 wt% solutions of the 99:1 SKL:PEO mixture (1318 ± 251 nm). It seems that in this case increasing the relative amount of PEO may counterbalance the tendency to reduce the fiber diameter arising from lower total polymer concentration. A more complete characterization of the effect of PEO content on fluid properties and resulting fiber diameter is warranted, and is the subject of current and future work.

Since a SKL:PEO mass ratio of 99:1 was sufficient for fiber formation, the effect of PEO addition on the electrospinning of the other six technical lignins was carried out using the 1% PEO content. For all technical lignins a clear transition from electrospray or beaded fibers to uniform fibers was observed with increasing total concentration. Figure 3 presents the SEM images of all of the lignin fibers electrospun from the 99:1 lignin:PEO solutions. All of the lignin:PEO solutions were electrospinnable at the same total polymer concentrations where the lignin solutions without PEO only electrosprayed. Interestingly, the concentration of PEO in the 99:1 lignin:PEO electrospinning solutions was $\leq$0.5 wt%, substantially lower than the minimum PEO concentration (5 wt%) required to electrospin PEO from water or DMF. These observations suggest that interactions between PEO and lignin influence the ability to form fibers by electrospinning.

Table 2 lists the fiber diameters and standard deviations of the electrospun fibers produced from the different 99:1 lignin:PEO solutions. For almost all of the lignin:PEO solutions, a concentration of 40 wt% was sufficient to form uniform fibers, the exception was the SOL:PEO solution, which required a slightly higher concentration. The water-based systems, LS and SL, formed uniform fibers at the lowest total concentration of all the lignins. At 30 wt% the LS produced uniform electrospun fibers, albeit quite large in diameter. We

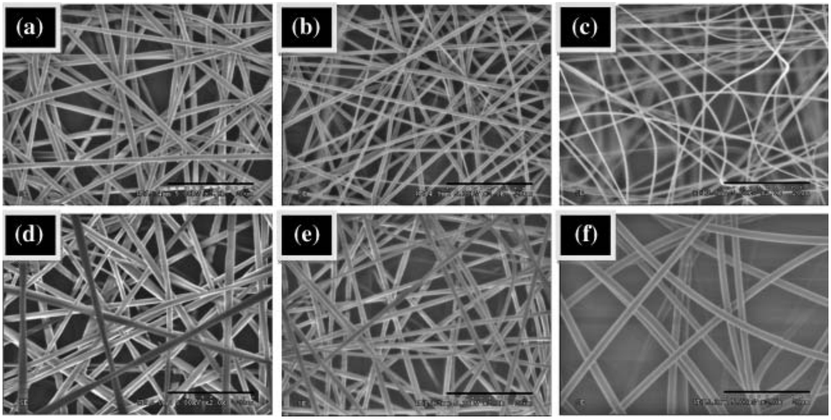

Figure 3. SEM images of lignin fibers electrospun from 99:1, lignin:PEO solutions using different technical lignins. (a): HKL 40 wt%; (b): PL 40 wt%; (c): SL 30 wt%; (d): SOL 50 wt%; (e): HOL 40%; (f): LS 30 wt%. All scale bars = 20 μm (2000× magnification).

Table 2. Diameters ± standard deviation of 99:1 lignin:PEO fibers produced by electrospinning (N = 200)

Lignin:PEO system	Concentration (%)	Diameter (nm)
SKL	40	1318 ± 251
HKL	40	1085 ± 188
SOL	50	1517 ± 415
HOL	40	1135 ± 171
PL	40	912 ± 176
LS	30	1645 ± 371
SL	30	702 ± 186

speculate that this is due to the higher molecular weight of lignosulfonates as compared to other technical lignins.[33,34] As expected the lower molecular weight SL formed smaller fibers in the same solvent although with a few beads (Figure 3c). From our experiments it was evident that while all systems formed fibers, the fiber morphology depended on the technical lignin, as well as the operating parameters. In all of the systems, the higher concentration solutions (DMF and H_2O) required a larger collection distance in order to allow fiber solidification. In some cases, average fiber diameters differed slightly when different voltage, flow rate, and collector distance were used. The values reported in Table 2, were obtained using the same operating parameters in DMF (10 kV, 0.03 mL/min, 14 cm) and water (14 kV, 0.03 mL/min, 20 cm), respectively.

It has been reported that fiber diameter scales with concentration.[15–17] Therefore, the relationship of fiber diameter with concentration was investigated for the various lignin:PEO systems. Figure 4 shows the fiber diameter versus concentration for the 99:1 SKL:PEO system; an essentially linear increase in fiber diameter with concentration was observed. Fiber diameters for all the different systems were in the range of roughly 200 nm to over 5 microns, with the largest fibers being produced from aqueous solutions at higher concentration. Fibers with diameters around 1 micron and above were usually observed to be relatively free of bead defects, but below 1 micron bead frequency increased with decreasing diameter. Although quite large relative to other electrospun nanofibers, which can be smaller than 100 nm in diameter,[12–14] the diameters of the lignin fibers produced here by electrospinning are roughly 25–50 times smaller than those obtained by fusion spinning.[7] Moreover, they were produced from seven different technical lignins, including one (SKL) that was shown to have poor thermal processibility in previous work.[7]

It is well known that the morphology of electrospun fibers depends on the fluid properties such as viscosity, relaxation time, surface tension, and conductivity.[35,36] The solution properties are functions of the polymer-solvent system, chain entanglements, and/or specific intermolecular interactions such

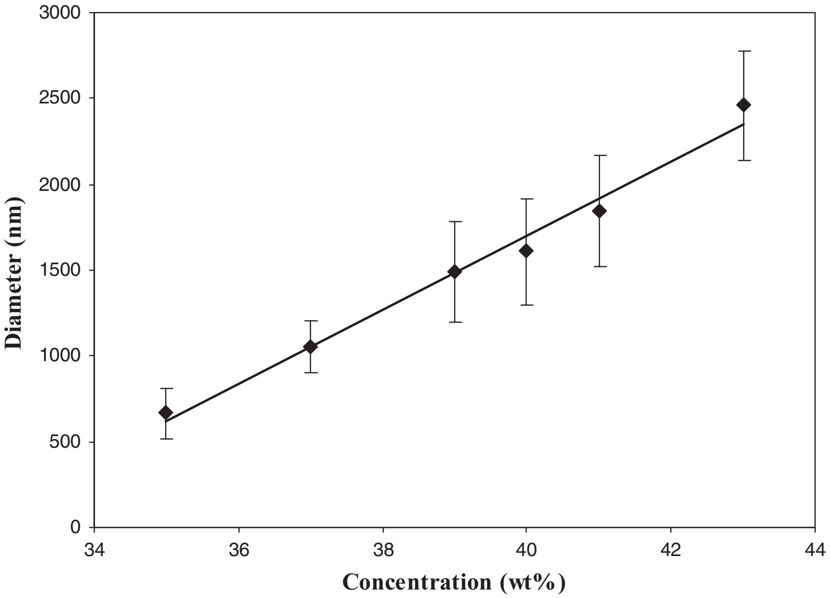

Figure 4. Plot of fiber diameter versus concentration for the 99:1 SKL:PEO system. Diameters are reported as mean ± standard deviation, N = 100 at each concentration.

as hydrogen bonding[37] or associative interactions.[38] Previous investigators have used viscosity data to characterize the relationship between polymer concentration and entanglements, and reported correlations of viscosity with fiber diameter and electrospinning behavior.[15–18,23,25,35,36]

To investigate the scaling relationship of viscosity with concentration, a logarithmic plot of specific viscosity versus concentration was generated for SKL and 99:1 SKL:PEO blends (Figure 5). There is a clear increase in slope at approximately 25–30 wt% for both the SKL and SKL:PEO systems. Based on the literature, this may represent a threshold between regimes above which interactions or entanglements between chains become more important.[15–17] The intersection of two extrapolated linear regressions ($R^2 = 0.97$–0.98) over the ranges 10–25 and 30–45% for solutions with and without PEO were calculated to compare the relative threshold of the upturn in viscosity. The intersection for SKL alone was 28 wt%, while the intersection for SKL:PEO was 27 wt%. The observed difference between SKL and SKL:PEO is small or negligible, but would be consistent with a slightly lowered entanglement threshold in the presence of PEO. Clearly the concentration must be above this threshold to form beaded fibers for SKL with or without PEO, although a higher concentration is required for SKL alone.

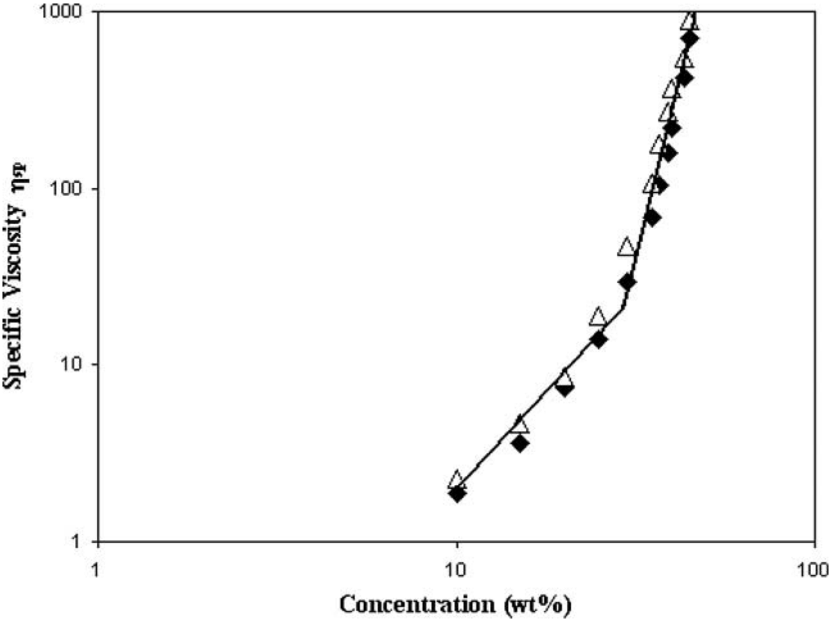

Figure 5. Plot of specific viscosity versus concentration for SKL and SKL:PEO systems. Δ = SKL:PEO, ♦ = SKL.

The shape and slope of the plots were very similar for SKL and SKL:PEO, although the SKL:PEO had slightly higher values of specific viscosity. The calculated fits produced dependencies of $\eta \sim c^{2.2}$ and $c^{2.3}$ from 10–25 wt% and $\eta \sim c^{7.8}$ and $c^{7.4}$ from 30–45 wt% for SKL and SKL:PEO, respectively. The calculated values are higher than values expected based on theory for linear polymers in good or theta solvents in the semi-dilute range (1.3 in the unentangled and 3.7–4.7 in the entangled regime),[39] although higher values than predicted have been reported.[15,25,37,38] Notably, such values have been observed in systems involving hydrogen bonding[37] and association complexes.[25,38] Similarly, higher values can also be expected as solvent quality decreases[17,39] and also, depending on the branch chain length and concentration, in the case of branched polymers.[40] The higher observed values for the SKL and SKL:PEO solutions are therefore consistent with the fact that technical lignins are branched macromolecules which assume compact conformation in solution,[41,42] and participate in hydrogen bonding and associative interactions.[43] Furthermore, based on the Mark-Houwink-Sakurada parameter α, which has been measured to be around 0.1 in DMF at 318 or 350 K,[41] DMF is not a good or theta solvent for Kraft lignins. The addition of PEO increases the viscosity by an average of 36% but does not dramatically change the scaling behavior of viscosity with concentration. These observations confirm that the specific viscosity is primarily due to the lignin component, with PEO adding a relatively minor contribution. This observation makes sense since lignin accounts for 99% of the solid content in solution. Importantly for the present work, the plots for SKL with or without PEO are similar while their electrospinning behaviors are different. This observation suggests shear viscosity does not completely describe the differences with or without PEO in regards to the ability to form fibers. For example, the difference in viscosity between the SKL solution at 50 wt% (which formed beaded fibers) and the 99:1 SKL:PEO solution at 40% (which formed uniform fibers) was roughly ~2 versus 0.4 Pa·s. All other lignin:PEO systems displayed similar trends, in that lower viscosity lignin:PEO solutions were capable of forming fibers while higher viscosity lignin solutions electrosprayed.

To further clarify the role of viscosity, electrospinning was also carried out from solutions of the same viscosity and using the same operating parameters with different technical lignins and PEO (mass ratio 99:1) to investigate the effect of different lignins on fiber diameter. At a viscosity of 8.6×10^{-2} Pa*s, PL at 43 wt% and HOL at 42 wt% with PEO had similar diameters (1850 nm and 1700 nm, respectively). On the other hand, there were clear differences when comparing other lignins at the same viscosity. For example, SKL solutions at 35 wt% with PEO at the same viscosity formed fibers with average diameters of 750 nm, while the average diameters for the HKL solutions at 39 wt% was 1250 nm. Another difference was that the SKL fibers had a few beads while the HKL fibers were more uniform. In addition, LS and SL solutions with PEO at the same viscosity (30 and 40 wt%, respectively, $\eta = 9.0 \times 10^{-2}$ Pa*s) formed fibers with very different diameters. The LS fibers were roughly 1600 nm, while the SL fibers were much larger in diameter, and over a wide range of 5–10 microns. As mentioned previously, LS is expected to have higher molecular weight compared to SL, and this is confirmed by the higher concentration of the SL:PEO solution (40 wt%) required to match the viscosity of LS:PEO at 30 wt%. These observations indicate that the fiber diameter depends on the fluid properties at a particular concentration, which depend on polymer structure and solvent. Different technical lignins are therefore expected to have a slightly different relationship between their fluid properties with concentration, corresponding to slightly different diameters at a given concentration depending on the structure of the technical lignin and solvent.

These observations are consistent with the work of Yu and coworkers, who demonstrated no correlation of the dimensionless viscosity, or Ohnesborge number, with the ability to form fibers from elastic PEG–PEO aqueous solutions.[18] The same authors showed that elasticity as characterized by fluid relaxation time and steady elongational viscosity was critical in the prevention of jet breakup during electrospinning of their system. Our lignin:PEO solutions bear a resemblance to the aqueous Boger fluids prepared by Yu and coworkers in that both contain a relatively small concentration of higher molecular weight PEO with another oligomeric species (0.1–0.2 vs. 0.5 wt% PEO or less in our study). The oligomeric species in the work of Yu et al. was PEG, and in the present work was replaced by technical lignins, which are generally complex, polydisperse mixtures of branched polyaromatic oligomeric and polymeric species. It is logical based on the report of Yu et al to hypothesize that the addition of relatively high molecular weight PEO to lignin in solution increases the fluid elasticity, since it was demonstrated that small amounts of high molecular weight PEO can dramatically influence fluid elasticity and electrospinnability without significantly altering the viscosity.[18] Dilute solutions of high molecular weight polymers in viscous solvents or in the presence of oligomeric species are often highly elastic in elongational flow.[44–46] Furthermore, it is known that real material responses to shear can be different than the responses to extension.[40] Since electrospinning is a case of strong elongational flow with high strain rate,[47] nonlinear rheological behavior (extensional thickening) would be expected to occur with elastic fluids during electrospinning.[48] If the addition of PEO to lignin solutions significantly increased the fluid elasticity while only slightly affecting the viscosity, shear viscosity measurements might not completely describe the electrospinning behavior of lignin:PEO solutions, since these measurements provide no information on extensional strain hardening. In future work we hope to characterize the extensional properties of lignin solutions to allow a more complete analysis of the relationships between lignin structure, fluid properties, and electrospinning behavior.

CONCLUSION

Seven different technical lignins were readily electrospun into fibers through the addition of PEO (1–5 wt%); in the absence of PEO, none of the lignins could be processed into uniform fibers, although beaded fiber formation was observed for the SKL system at high concentration ($>$50 wt%). As with other polymer systems, a linear increase in fiber diameter with increasing lignin concentration was observed. However, at the same concentration, the various lignin solutions had varying viscosities and different electrospinning behavior, that is, fiber diameter and ability to form uniform fibers. Similar results were found using lignin:PEO solutions with the same viscosity. Together these results suggest lignin specific structures and intermolecular interactions are influencing solution properties and electrospinning behavior. Further support of this was observed from the scaling exponents calculated based on specific viscosity versus concentration plots, which at beyond 27–28 wt% concentration $\eta \sim c^{7.4\text{–}7.8}$, consistent with a branched polymer participating in intermolecular interactions such as hydrogen bonding or association complexes. On the other hand, lignin:PEO solutions produced uniform fibers at viscosities much lower than those of lignin alone that only electropsprayed, suggesting viscosity plays one, but not the only key role in determining electrospinnability.

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