Fabrication and Properties of Alkaline Lignin / Poly (Vinyl Alcohol) Blend Membranes

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Membranes were prepared from alkaline lignin and poly(vinyl alcohol) (PVA) by a film casting method, and their properties were evaluated. These blend membranes can aptly be termed as green by nature as they are totally non-toxic and eco-friendly. The optimal mass ratio was determined based on the mechanical properties of membranes. The microstructure, mechanical properties, oxygen and carbon dioxide transmission, light transmittance, and thermal stability of the membranes were investigated. The blend membrane exhibited better thermal stability and barrier performance to oxygen and carbon dioxide than a PVA membrane due to the incorporation of alkaline lignin. Both tensile strength and elongation at break were increased with increasing mass fraction of alkaline lignin up to 15%, at which point the maximum of tensile strength and elongation at break were 43.65 MPa and 211.6%, respectively. The blockage of visible light at 600 nm was 62.36%, which was an improvement of 314.90% compared with the PVA membrane (13.36%). Based on this result, we suggest that the mechanical properties of blend membrane containing 15% alkaline lignin are excellent and better than PVA membrane (40.26 MPa, 179.37%). Thus, an alkaline lignin/PVA blend membrane was judged to be potentially suitable as an eco-friendly packing material.

Keywords: Alkaline lignin; PVA; Blend membrane; Structural characterization

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INTRODUCTION

The development of biodegradable composite materials for a wide range of applications has played an important role in recent years. Environmental pollution, especially by packaging materials, which are used in large quantities, has stimulated research for biodegradable composite materials (Fomin and Guzeev 2001). To this end, materials such as agricultural residues and bioprocessing byproducts are obvious sources of low cost biomacromolecule materials.

Lignin, a natural polymer found in wood, is an amorphous and polyaromatic polyol present in large amounts in plants and wood tissues. Because of its biodegradability, renewability, and low cost, the development and application of lignin have been of interest for many researchers (Funaoka 1999; Ibrahim *et al.* 2011; Yuan and Huang 2011; Wang *et al.* 2010). Commercially, lignin is obtained as a by-product of 'wood free' papermaking (Guigo *et al.* 2010; Suhas *et al.* 2007). Lignin is utilized as a stabilizer for plastics and rubber. It is also used in the formulation of dispersants, surfactants, and adhesives or modifiers to improve the performance of the composite by blending it with other polymers (Pouteau *et al.* 2004; Simionescu *et al.* 1993; Stewart 2008; Stoica-Guzun *et al.* 2011).

Poly(vinyl alcohol) (PVA) is a hydrophilic polymer that is nontoxic, water-soluble, and biodegradable and has excellent mechanical strength (Ozaki *et al.* 2005; Park *et al.* 2001; Pereira *et al.* 2000). Therefore PVA is usually used for special applications in packaging, such as water-soluble films in agriculture. It is also used as an emulsifier and a stabilizer for colloid suspensions and as a sizing agent and coating in the textile and paper industries (Chiellini *et al.* 2003; Yamaoka *et al.* 1995). Also PVA has film-forming capacity and could be used in blends and composites with other polymers or inorganic materials, most of which have wide applications (Bai *et al.* 2007; Karthikeyan 2005; Lebrun *et al.* 2004; Tang *et al.* 2008).

The miscibility between lignin and polar polymer materials such as PVA and PVC is favorable to the formation of hydrogen bonding at the interface. Thus the blends exhibit suitable interfacial binding force (Yue and Chen 2010). Kubo and Kadla (2003) have indicated that PVA is a polymer miscible with lignin, and a high degree of inter-molecular interaction takes place between lignin and the PVA chains. Although both miscibility and morphology of lignin/PVA blends have been studied, the effect of alkaline lignin blending with PVA on the properties of membrane has not been reported.

In this study, alkaline lignin was prepared from wheat straw alkaline lignin pretreated by sodium hydroxide solution. Then the alkaline lignin and PVA were used to prepare the blend membranes by the film casting method. The aim of the work was to find method to modify lignin and make the modified alkaline lignin disperse in PVA domain more easily. A further goal was to demonstrate the influences of addition of alkaline lignin on the membranes properties.

EXPERIMENTAL

Chemicals and Reagents

Wheat straw alkaline lignin (Table 1) was isolated from black liquor obtained from the kraft pulping process (Shandong, China) and dried in a vacuum oven prior to use. Poly (vinyl alcohol) 1788 (PVA1788), M_w 84,000 to 89,000 g/mol, 86 to 89% hydrolyzed, were purchased from Shandong Chemicals Company (Shandong, China). Sodium hydroxide (analytical reagent grade) was purchased from Tianjin Yongda Chemical Company (Tianjin, China). The C_9 empirical formula and the C_9 displayed formulas were as follows:

Empirical formula: $C_9H_{3.75}O_{3.82}(OCH_3)_{0.75}$

Displayed formula: $C_9H_{2.88}O_{2.84}(OCH_3)_{0.75}(OH_{phenol})_{0.36}(OH_{alcohol})_{0.45}(CO)_{0.052}(COOH)_{0.064}$

Table 1. Analysis of Functional Groups of Wheat Straw Alkaline Lignin

Functional group	Total hydroxyl group	Total acidic group	Carbonyl group	Carboxyl group	Phenol hydroxyl group	Alcohol hydroxyl group
Content (mmol/g)	4.08	4.10	2.41	0.86	1.80	2.28

Preparation of Alkaline Lignin

The wheat straw alkaline lignin was dissolved in sodium hydroxide aqueous solution (0.1 mol/L), resulting in a solution of 5 wt % wheat straw alkaline lignin. The solution was filtered and then dried with an experimental spray drier to obtain refined alkaline lignin.

Fabrication of the Membranes

Alkaline lignin, PVA, and distilled water were put into a 250 mL 3-mouth flask, homogenized by controlling the stirring time for 3 h in the 90 ^{o}C constant water bath, followed by treatment in an ultrasonic bath for a further 20 min. The mixture was defoamed in a vacuum drying oven (0.06 MPa vacuum degree), placed on a clean plate, and finally the blended solution was dried at room temperature for 24 h. For all membranes, the total weight of solute was kept constant, and the concentration of solution was 4% (w/w); the alkaline lignin/PVA (0/100, 10/90, 15/85, 20/80, 25/75, 30/70) membranes were successively denoted by L-0, L-1, L-2, L-3, L-4, L-5. When visually dry, the membranes were peeled from the plastic substrate and had a thickness of 30 to 60 μm.

Mechanical Properties

According to Chinese standard GB/T13022-91, the membrane samples were cut into rectangular strips (150 mm × 15 mm). All samples were conditioned at 50% RH and 23 ^{o}C for 90 h before the test. The thickness of membranes was measured using a digital micrometer (±0.001mm), averaging 10 different positions. Tensile strength and elongation at break of membranes were measured with an intelligent electronic universal testing machine (LDX-200, Beijing Landmark Packing Material Co., Ltd. China) at the speed of 33 mm/min. Five different rectangular strip samples were measured in each group to calculate their average.

Scanning Electron Microscopy (SEM)

After surface cleaning and liquid nitrogen fracture, imaging of the membranes surface and fractured cross section was performed with environment scanning electron microscopy (Quanta200, FEI Co., Ltd. USA).

Thermogravimetric Analysis (TGA)

The thermal stability of the membranes was evaluated using thermogravimetric analyzer (TG209F3, Netzsch Scientific Instruments Co., Ltd., Germany). The weight changes of membrane samples were recorded under flowing nitrogen (20 mL/min) atmosphere at a heating rate of 10 ^{o}C/min, ranging from 30 ^{o}C to 800 ^{o}C.

Differential Scanning Calorimetry (DSC)

Membrane samples were analysed by differential scanning calorimetry (DSC204, Netzsch Scientific Instruments Co., Ltd., Germany) under flowing nitrogen (20 mL/min) atmosphere at 10 ^{o}C/min ranging from 30 ^{o}C to 300 ^{o}C.

Light Transmittance

Rectangular strip samples (30 mm × 12 mm) were prepared, and then regular light transmittances were measured at wavelengths from 200 to 800 nm by double-beam UVspectrophotometer (PERSEE TU-1901, Beijing Purkinje General Instrument Co., Ltd. China), obtaining the transmittance data from the spectrum.

Oxygen and Carbon Dioxide Transmission

Based on Chinese standard GB/T 2918-1998, three circle samples were prepared in each group and conditioned at 50% RH and 23 °C for 48 h before the testing. Then the oxygen and carbon dioxide transmission of samples was measured using a VAC-VBS pressure difference gas permeameter (Labthink, Jinan, China).

RESULTS AND DISCUSSION

Physical and Mechanical Properties

The effects of different quantities of alkaline lignin on the tensile strength and elongation at break of membranes are shown in Fig. 1. The resulting blend membranes were brown, translucent, smooth, and a little crisp. According to the Chinese standard GB/T13022-91 that the samples were conditioned at 50RH and 23 °C for 90 h, the samples were more flexible.

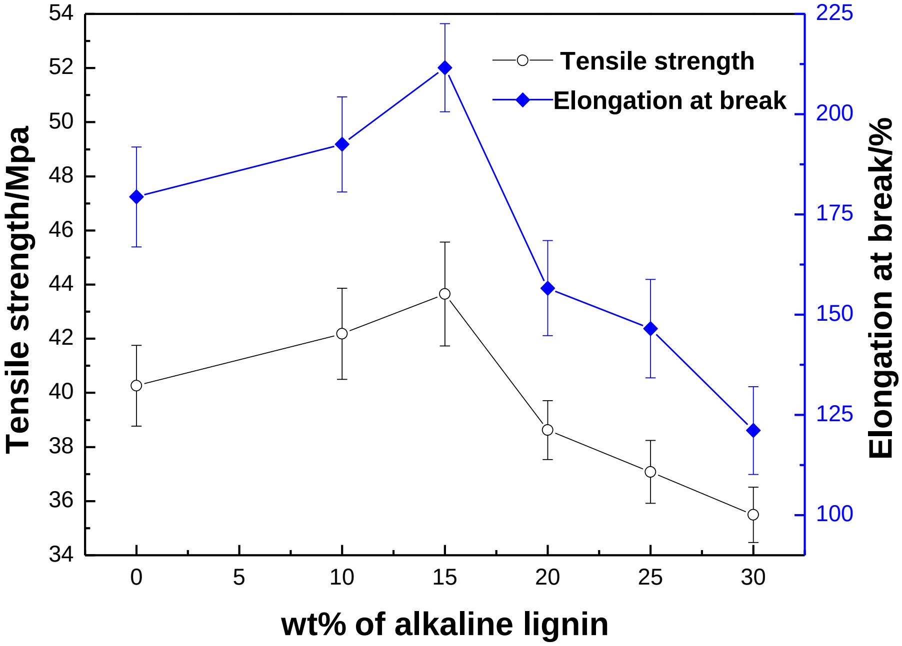

Fig. 1. Effect of quantity of alkaline lignin on tensile strength

As shown in Fig. 1, with the quantity of lignin increasing, tensile strength increased before the mass fraction of alkaline lignin reached 15%, and then it decreased. The maximum tensile strength was 43.65 MPa when the mass fraction of alkaline lignin was 15%. Compared with PVA, the tensile strength was increased by 8.42%. The improvement of tensile strength may be caused by several factors. For instance, the interaction between alkaline lignin (with its network structure) and long PVA chains results in great resistance to the movement of PVA chains, forming an entangled structure. Besides, the formation of strong intermolecular hydrogen bonds between the hydroxyl groups of PVA and lignin also enhances the tensile strength of the membrane (Kubo and Kadla 2003). With the mass of alkaline lignin increasing, a part of alkaline lignin and PVA chains can bind tightly, but another part of alkaline lignin cannot be coupled with PVA tightly, and only act as infillings in the network structure of alkaline lignin. This may lead to the reduction of mechanical property of blend membranes.

Meanwhile, the maximum elongation at break was 211.6%, corresponding to an improvement of 17.97% compared to the PVA membrane. Alkaline lignin molecules with network structure combine with PVA chains compactly, when the blend membranes are drawn, the alkaline lignin molecules stretch gradually, which may lead to the raising of elongation at break. The membranes were more pliable after the conditioning according to the Chinese standard GB/T13022-91, and this may have contributed to the improvement of ductility of the blend membrane. Also, it can be concluded that the reason for the reduction of elongation at break of the blend membranes at higher lignin contents may be due to the low ductility of lignin.

The optimal tensile strength and elongation at break of blend membrane were 43.65 MPa and 211.6%, respectively. This indicates that the blend membrane containing 15% alkaline lignin possessed excellent mechanical properties.

Morphology by SEM

Figure 2 presents micrographs of the membranes.

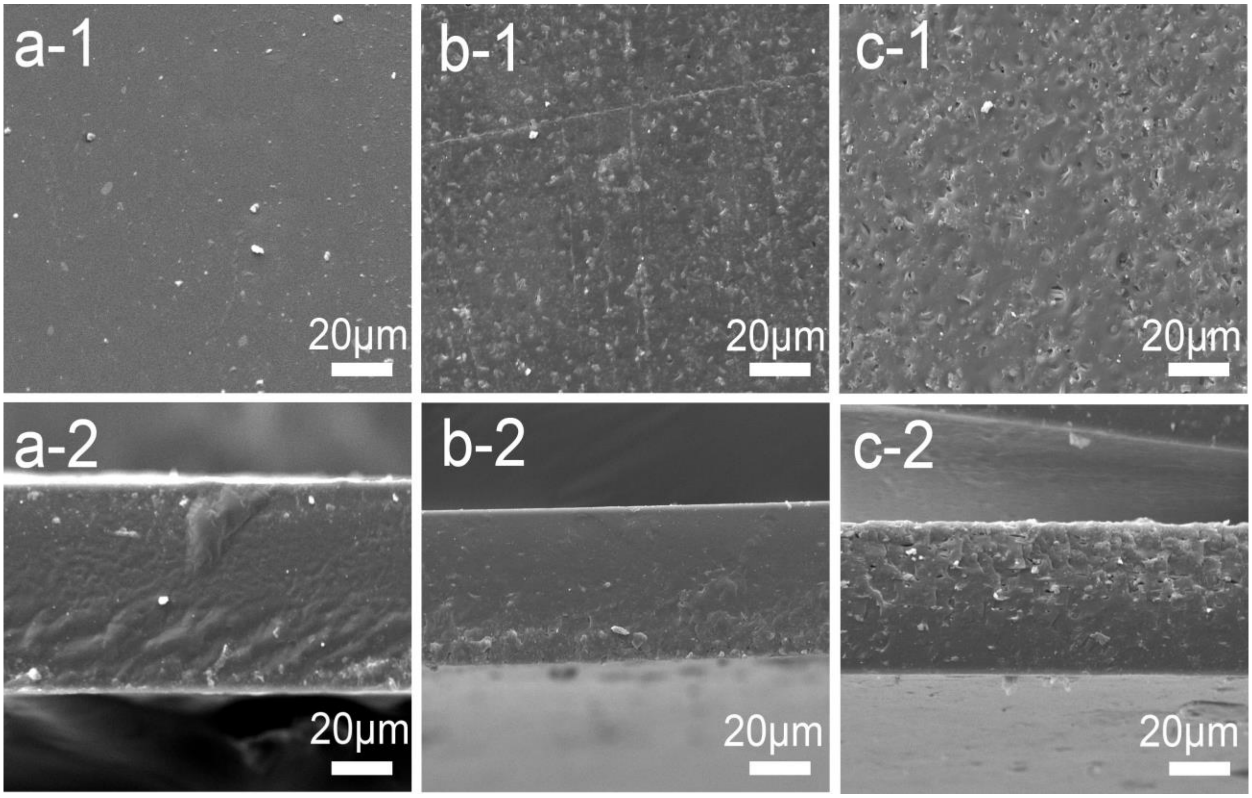

Fig. 2. SEM micrographs of the membranes: membrane L-0 (×1000; a-1 surface, a-2 cross section); blend membrane L-2 (×1000; b-1 surface, b-2 cross section); blend membrane L-5 (×1000; c-1 surface, c-2 cross section)

From parts b-1 and b-2 of the figure, corresponding to a 15 wt % of lignin in the blend membrane, it can be seen that alkaline lignin particles were well dispersed in PVA domain and there was little conglomeration in PVA continuous phase. The miscibility between lignin and polar PVA was expected to be favorable, and the surface was smooth, though there was a little sodium salt in the membrane. When the mass ratio of alkaline lignin reached 30%, some irregular grooves formed on the surface of the membrane, as is evident in micrographs c-1 and c-2. The fractured cross-section of this membrane showed poor dispersion of alkaline lignin in PVA. This may explain the mechanical properties of the blend membranes discussed earlier.

Thermal Stability Analysis (TGA)

Figures 3 (a) and (b) show TGA results for the membranes. There were four main mass loss areas in the membranes. The temperature range 30 ^{o}C to 178 ^{o}C corresponded to the loss of water, with *ca*. 4% of physically absorbed water molecules desorbed from the pure PVA membrane. The second weight loss of about 75% occurred between 270 and 350 ^{o}C; this was attributed to the decomposition of PVA. The mass loss rate of pure PVA membrane reached a maximum 285 ^{o}C. The mass loss continued to increase between 350 ^{o}C and 470 ^{o}C, a range in which PVA is known to form polyene (Morancho *et al*. 2011); the corresponding weight loss was *ca*. 94%. Carbon loss occurred above 470 ^{o}C, and there was no significant quality change in this temperature range. The residuals after the heating cycle were about 2.5%. The blend membrane L-2 containing 15% alkaline lignin exhibited a similar trend line compared to the pure PVA membrane, but there were two distinct differences: The weight loss rate of blend membrane L-2 in the second mass loss area (245 ^{o}C to 326 ^{o}C) was lower than that of the pure PVA membrane. Meanwhile the maximum temperature of mass loss rate of blend membrane L-2 was 305 ^{o}C, which was an improvement compared with pure PVA membrane 285 ^{o}C. Besides, the decomposition product of pure PVA membrane was volatile and the amount of residue was only 2%. But the decomposition product of blend membrane L-2 was more stable, thus leaving more than 10% as residue. These results indicate that the thermal stability of membrane L-2 was enhanced with the incorporation of alkaline lignin.

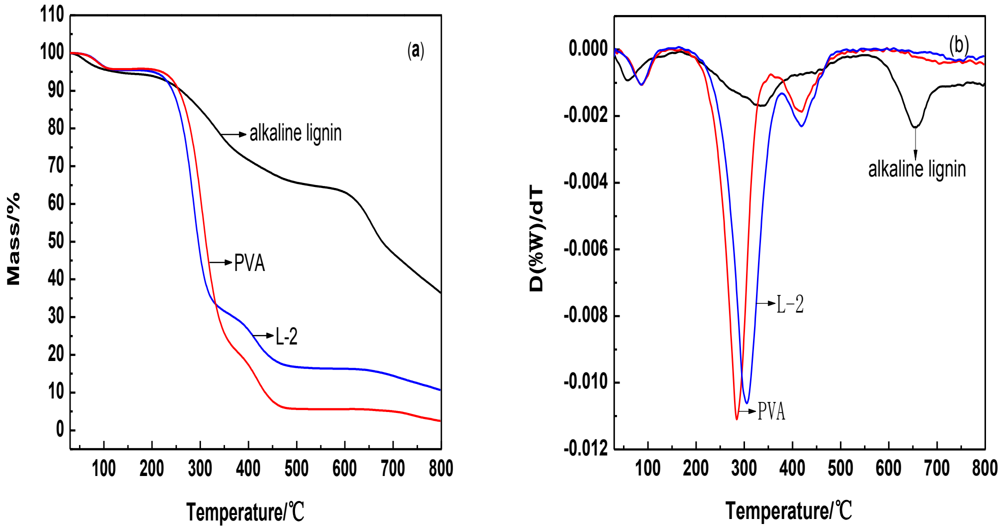

Fig. 3. TG and DTG diagram of alkaline lignin, PVA and blend membrane L-2: (a) TG; (b) DTG

Differential scanning calorimetric measurements were carried out for pure PVA and blend membrane L-2, and the resulting thermograms are presented in Fig. 4. The DSC thermogram of pure PVA membrane and blend membrane L-2 showed a similar T_g at 80 ^oC. The melting process took place between 205 and 270 ^oC, and the corresponding thermal degradation started at 270 ^oC. When alkaline lignin was incorporated into the PVA film, it can be seen that the thermal degradation onset was shifted to a higher temperature while the melting peak was shifted to a lower temperature. These behaviours are better visualized in the peak temperature data shown in Table 2. The melting point depression is indicative of favoured interactions between the polymer chains, when the components of the blend are amorphous or semi-crystalline polymers, as in this case (Castro *et al.* 2003). On the other hand, the PVA thermal degradation peak was broadened by the presence of alkaline lignin, indicating that the mechanisms of the thermal degradation are different from that of pure PVA membrane.

TG and DTG, together with DSC analysis showed that because of the incorporation of the alkaline lignin, the membrane L-2 exhibited better thermal stability than the pure PVA membrane. Similar behavior has been reported by Bittencourt *et al.* 2005 in the case of lignin/PVA blends.

Table 2. Temperature at the Minimum of the Melting and the Thermal Degradation Peaks for PVA Membrane and Blend Membrane L-2

Blend membrane alkaline lignin/PVA	$T_{melting}/^oC$	$T_{decomposition}/^oC$
0/100	240	270
15/85	234	275

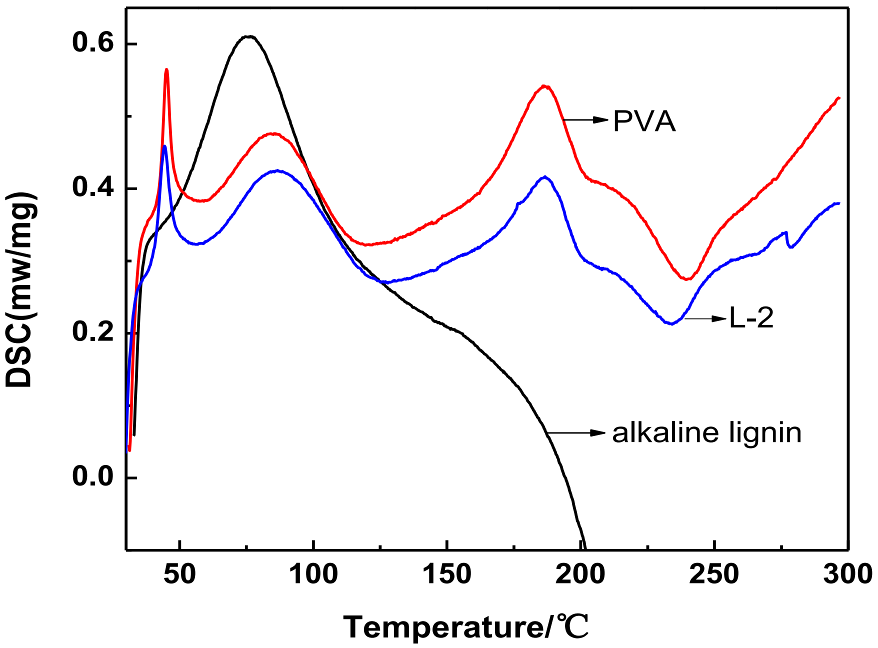

Fig. 4. DSC thermograms of alkaline lignin, PVA, and blend membrane L-2

Light Transmittance Analysis

Figure 5 presents the UV-visible spectra of PVA and blend membranes. It can be observed that the transmittance of the blend membranes decreased significantly with the increasing of the lignin content in the membranes. Also, the barrier performance of the blended membrane containing alkaline lignin 15% to visible light at 600 nm was 62.36%, representing an improvement of 314.90% compared with PVA membrane (13.36%). The average absorption of ultraviolet light of the blend membrane containing alkaline lignin 15% near the ultraviolet region (200 to 400nm) was above 97%, furthermore reaching 100% for L-4 and L-5, which was much higher than the PVA (1.22% to 80.27%). These results can be attributed to the fact that the membrane is brown and translucent because of the addition of alkaline lignin. Moreover, alkaline lignin can absorb ultraviolet radiation and play a role as a barrier to light (Fernandes *et al.* 2006). Thus, the blend membrane containing alkaline lignin 15% has been shown to exhibit excellent light insulation compared with a pure PVA membrane.

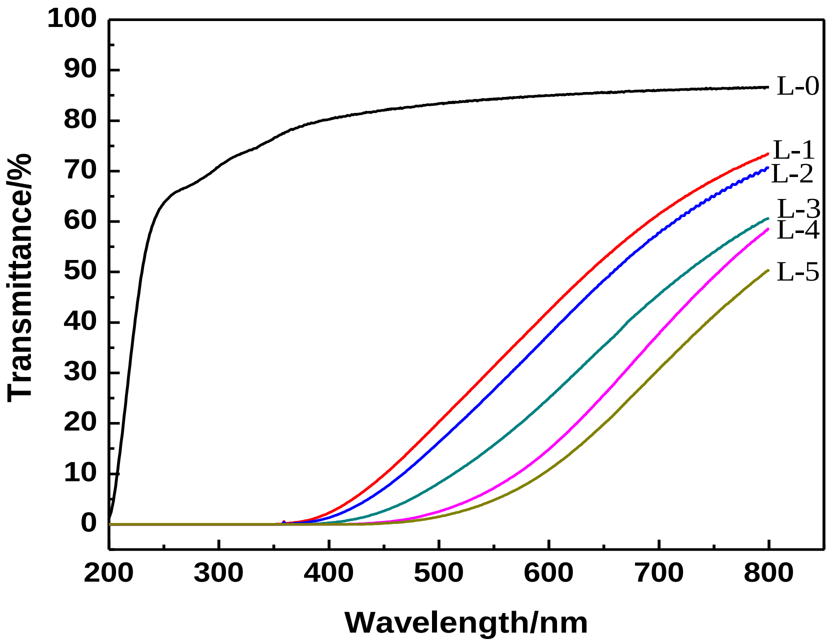

Fig. 5. UV-visible spectra of the membranes

Oxygen and Carbon Dioxide Transmission

- The oxygen and carbon dioxide transmission of blend membranes showed an irregular tendency in Table 3, but all blend membranes presented better oxygen and carbon dioxide-barrier properties compared with PVA. Also, the insulation of oxygen and carbon dioxide of blend membrane L-2 improved 111.58% and 170.26% compared to pure PVA, respectively. This demonstrates that the addition of alkaline lignin decreases the oxygen and carbon dioxide transmission of blend membranes. PVA membranes already possess good gas barrier performance (Kim *et al.* 2011). Alkaline lignin with its amorphous structure in the membranes may be expected to provide more tortuous and lengthy paths for diffusion of oxygen and carbon dioxide, thus further enhancing the gas barrier properties of the blend membrane.
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Table 3. Oxygen and Carbon Dioxide Transmission of the Membranes

Membrane notation	L-0	L-1	L-2	L-3	L-4	L-5
Oxygen transmission Carbon dioxide transmission ($10^{-5}\cdot cm^3/m^2\cdot d\cdot Pa$)	6.58 5.27	6.41 4.28	3.11 1.95	1.63 1.91	3.72 2.16	2.15 1.63

CONCLUSIONS

Blend membranes of alkaline lignin and PVA were prepared, and their properties were compared to those of neat PVA membrane. It was concluded that:

1. The mechanical properties of blend membranes can be improved compared to neat PVA membrane because of the addition of alkaline lignin. The maximum of tensile strength and elongation at break were 43.65 MPa and 211.6%, respectively when the mass fraction of alkaline lignin was 15%. This demonstrates that the optimum mass ratio of alkaline lignin for preparing the blend membrane was 15%, at which the blend membrane exhibited the best mechanical properties.
2. SEM analysis revealed that the blend membrane containing 15% alkaline lignin presented a favorable miscibility. Alkaline lignin higher than 30% gave a poor membrane formation.
3. TG and DSC analyses of blend membrane containing alkaline lignin 15% suggested the intermolecular interactions between alkaline lignin and PVA chains were favored and suggested that the blend membrane L-2 had good thermal stability.
4. The transmittance analysis indicated that the blend membranes achieved excellent light insulation. And the oxygen transmission $3.11\times10^{-5}\cdot cm^3/m^2\cdot d\cdot Pa$ and carbon dioxide transmission $1.95\times10^{-5}\cdot cm^3/m^2\cdot d\cdot Pa$ showed the blend membrane L-2 had good oxygen and carbon dioxide-barrier performance.
5. Meanwhile the addition of alkaline lignin can reduce the cost of the fabrication of the blend membranes significantly. All these results indicate that the blend membrane may be of interest as eco-friendly packing material.

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