

## PREPARATION AND PROPERTIES OF LIGNIN-EPOXY RESIN COMPOSITE

Quanfu Yin, Weijun Yang, Chengjun Sun, and Mingwei Di\*

A cross-linked biomass-polymer composite with a lignin content of up to 60% was prepared by blending lignin with an epoxy resin and polyamine using a hot press molding process. The characteristics of the curing reaction of lignin with epoxy resin were studied using DSC and FTIR analysis. The effect of molding temperature and molding pressure on the mechanical properties and microstructure of the lignin/epoxy resin composite was also studied by SEM, DMA, and TG analyses. The results showed that the epoxy resin can be cured by lignin, and the curing temperature for the blends can be reduced by the introduction of a polyamine cure agent. The properties of the composite, such as bending strength, impact strength, glass-transition temperature, and thermal stability, were evidently influenced by the molding process. A good interfacial combination was formed between lignin and epoxy resin. Increasing the molding temperature and pressure proved beneficial to achieve a better interfacial combination for the composite, and the degree of ductile fracture was increased in the fracture surface of the composite.

*Keywords:* Lignin; Epoxy resin; Composites; Mechanical properties; Microstructure; Molding process

*Contact information:* Key Laboratory of Bio-Based Material Science & Technology (Ministry of Education), Northeast Forestry University, Harbin 150040 China;

\* Corresponding author, [dimingwei@126.com](mailto:dimingwei@126.com)

### INTRODUCTION

Lignin is considered to be one of the most promising future organic resources because it is renewable and second only to cellulose in abundance among naturally occurring polymers. It is generally accepted that the most important chemical functional groups in the lignin molecule structure include the aromatic, hydroxyl, methoxyl, carbonyl, and carboxyl groups (Wang *et al.* 2010; Yuan and Huang 2011). Thus, lignin can be used as an organic, raw, bio-based material for the synthesis and modification of polymer composites. Lignin has several advantages, such as sufficient sources, low cost, biodegradability, and renewability. Therefore, its development and application have been of interest for many researchers (Bhat *et al.* 2009; Feldman *et al.* 1991; Funaoka 1999; González *et al.* 2009; Ibrahim *et al.* 2011; Nonaka *et al.* 1997; Stewart 2008). In the past, however, most research has been carried out with liquefied lignin used for partial substitution of the phenols involved in the synthesis of polymer materials (Mansouri *et al.* 2011; Park *et al.* 2008). In addition, the high reactivity of polyphenolic structures is included in the molecular composition of lignin, and this is beneficial to the use of lignin as an additive to reduce the cost of production of composite materials or as modifiers to improve the performance of the composite by blending with other polymers (Pouteau *et al.* 2004; Simionescu *et al.* 1993). Until now, there has been a lack of effective and sufficient utilization of lignin.

Enzymatic hydrolysis lignin is a novel form of lignin that is isolated from the enzymatic hydrolysis residues of the biomass. As a by-product of biomass conversion industry, enzymatic hydrolysis lignin possesses greater chemical activity and applicability than lignosulfonate or kraft lignin (Jin *et al.* 2011; Xie *et al.* 2011; Cheng and Liu 2006). This is because the enzymatic hydrolysis process of biomass is carried out under relatively mild conditions; thus, many functional groups such as phenolic hydroxyl and alcoholic hydroxyl were well preserved. Therefore, enzymatic hydrolysis lignin could be used to modify polymer materials by directly reacting with formaldehyde, epichlorohydrin, isocyanate, and so on.

The preparation process of blending two or more polymers has been considered to be one of the most promising methods to achieve new materials with good performance, as compared with their chemical modification. Combining the synthetic polymers with natural materials provides ways to reduce costs and offers benefits from the combined properties. The association by blending is an interesting way to overcome some of the most important weaknesses of polymer, such as poor mechanical properties and low degradability. Blending artificial polymers with natural polymers can obtain new polymer materials with superior performance, while reducing the amount of chemical synthetic polymers in use. Compared with the synthesis or modification of new polymer materials, these methods can reduce costs and save time (Doherty *et al.* 2011). In this study, a cross-linked biomass-polymer composite based on enzymatic hydrolysis corn straw lignin with a lignin content of up to 60% was prepared by blending lignin with thermosetting epoxy resin. The effect of enzymatic hydrolysis lignin on the curing behavior of epoxy resin was studied by DSC and FTIR analysis. The effect of molding temperature and molding pressure on the mechanical properties and microstructure of lignin/epoxy resin composite was also studied by SEM, DMA, and TG analyses.

## EXPERIMENTAL

### Materials

Enzymatic hydrolysis corn straw lignin with a bulk density of 0.6 g/cm<sup>3</sup> was provided by Songyuan Laihe Chemicals Co., Ltd. of China. It was dried at 50 °C for 24 hours in a vacuum oven before blending. Bisphenol, an epoxy resin with an epoxy value of 0.51, was provided by Nantong Xingchen Synthetic Material Co., Ltd. of China. The curing agent polyamine trademarked TY-200 was provided by Tianjin YanHai Chemical Co., Ltd. of China.

### Preparation of the Composite

According to the mass ratios shown in Table 1, the lignin, epoxy resin, and polyamine were blended with a two-roll mill at the speed of 20 rpm and temperature of 20 to 30 °C for 10 to 30 min. The mixed materials were removed and put into a mold for hot pressing at a certain temperature and pressure. The hot pressing time was chosen according to the thickness of the composite materials and maintained at 2 to 5 min/mm. Cold pressing was carried out at a certain temperature after hot pressing. Finally, the composites were demolded and removed for further testing.

**Table 1.** Blending Mass Ratios of Lignin, Epoxy Resin, and Polyamine

Tests	Lignin	Epoxy Resin	Polyamine
DSC and FTIR Tests	1	1	-
	1	1	0.5
SEM, DMA, and TG Tests	6	2	2

### Tests and Analyses

The bending strength of the composite was tested according to the Chinese standard GB/T 9341-2000 with a CMT-6104 Electronic Universal Testing Machine provided by Shenzhen Ruigeir Instrument Co., Ltd. of China. The sample bars measured 80 mm × 12.7 mm × 4 mm (length × width × thickness) and were prepared for testing. The test was performed at a speed of 2 mm min<sup>-1</sup> with a span of 64 mm. The Notched Izod impact properties of the composite were tested according to the Chinese Standard GB/T 1043-93 in a XJJ-50 Charpy impact testing machine provided by Chengde Experiment Machine Co., Ltd. of China. The dimensions of the samples were 80 mm × 10 mm × 4 mm (length × width × thickness), and the test was performed at a speed of 3.5 m s<sup>-1</sup> on the samples with a span of 60 mm. The impact energy was 1 J. All measurements were performed under ambient conditions with a relative humidity of approximately 50%. At least five samples were tested for each composite formulation.

A NETZSCH 204 model differential scanning calorimetry (DSC) system provided by NETZSCH Scientific Instruments Co., Ltd. of Germany was used to analyze the curing reaction of lignin with epoxy resin at heating rates of 5 °C/min. The FTIR spectrum analysis was obtained by a Nicolet 560 Fourier transform infrared spectrograph provided by Nicolet Co., Ltd. Each spectrum was recorded in a frequency range of 400 to 4000 cm<sup>-1</sup> using a potassium bromide (KBr) disc. The KBr was previously oven dried to reduce the interference of water. The microscopic structure of lignin/epoxy resin composite was analyzed and characterized by a DMA 242C dynamic mechanical analysis (DMA) instrument and a TG 209-F3 thermogravimetric analysis (TG) instrument provided by NETZSCH Scientific Instruments Co., Ltd. of Germany, and a QUANTA 200 scanning electron microscope (SEM) with a working distance of about 10 mm at 12.5 kV provided by FEI Co., Ltd. of the USA. The DMA measurements were carried out in three-point bending mode, using rectangular specimens with dimensions of 50 mm × 10 mm × 2 mm over a temperature range of 25 °C to 180 °C with a heating rate of 5 °C/min under nitrogen flow. The samples were scanned at a fixed frequency of 2.5 Hz with a static force of 0.2 N and a dynamic force of 0.8 N. The composite specimen (with dimensions of 50 mm × 10 mm × 2 mm) for SEM examination was immersed in liquid nitrogen for 5 minutes for quick freezing and was then immediately broken by an impact force. A fractured surface was selected for SEM examination. All SEM samples were coated with approximately 10 to 20 nm of gold before examination.

## RESULTS AND DISCUSSION

### Curing Reaction of Epoxy Resin with Lignin

The epoxy resin curing reaction is exothermic. Thus, DSC analysis can determine whether the epoxy resin was cured, according to the heat variation in the epoxy resin

blends. The heat changes for the lignin-epoxy resin blends (1:1) and the lignin-epoxy resin-polyamine blends (1:1:0.5) with increasing temperature are shown in Fig. 1.

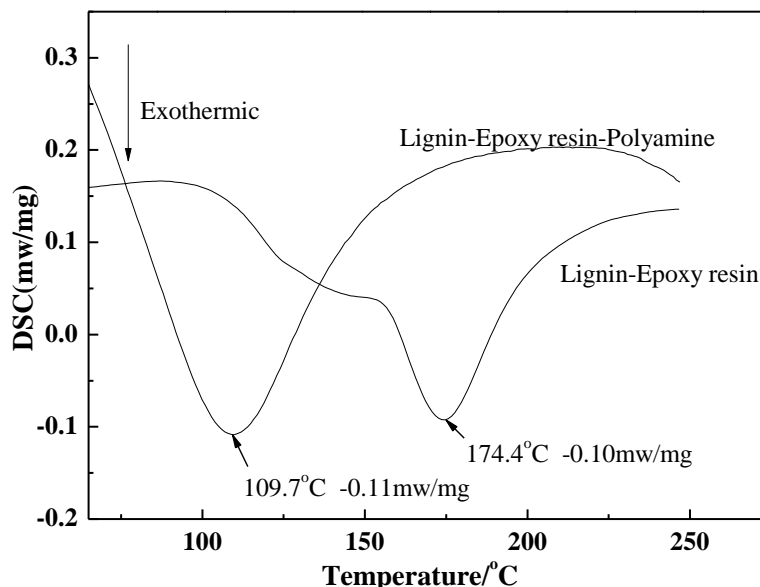
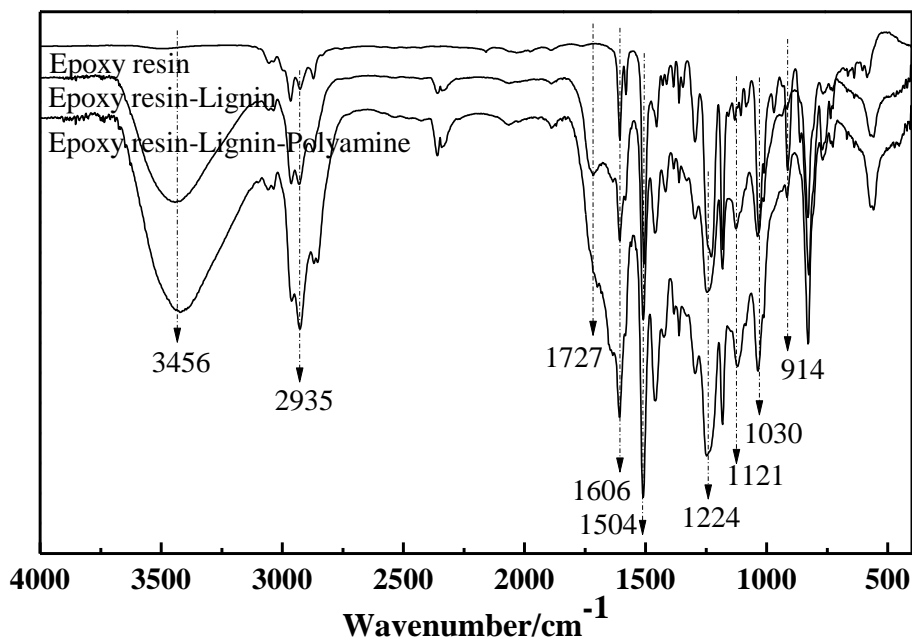


Fig. 1. DSC curves for two blends at a heating rate of 5 °C/min

Figure 1 shows that only one exothermic peak was observed in the DSC curve for the lignin-epoxy resin blends in the temperature range selected for the experiment, and that the peak temperature was 174.4 °C. It shows that the epoxy resin can be cured by enzymatic hydrolysis corn straw lignin. One reason for this is that the curing reaction was catalyzed by the polyphenol structure of lignin, and the other reason is that the carboxyl or hydroxyl groups contained within the enzymatic hydrolysis lignin react with the epoxy resin to make the epoxy resin cure. The lignin-epoxy resin blends had only one complete exothermic peak after the addition of the curing agent polyamine, and the peak temperature was 109.7 °C. The phenomenon was ascribed to the fact that the polyamine did not disturb the curing of the epoxy resin; rather, it reduced the curing temperature. Thus, the lignin-epoxy resin blends could be thermoformed at lower temperatures through the addition of the polyamine curing agent. In addition, as a flexible curing agent with a long molecule chain, polyamine can compensate for the inherent brittleness that is characteristic of cured blends of lignin and epoxy resin, and the performance of the blends was improved consequently.

The FTIR spectra for epoxy resin, the lignin-epoxy resin blends (1:1), and the lignin-epoxy resin-polyamine blends (1:1:0.5) are shown in Fig. 2. FTIR spectrum analysis was employed to characterize the raw materials and analyze the changes in the epoxy resin structure during its curing reaction, mainly to observe the introduction of new functional groups or disappearances of existing ones. From the FTIR spectrum of uncured epoxy resin presented in Fig. 2, it can be seen that the stretching vibration absorption for epoxy group is 914  $\text{cm}^{-1}$ . The FTIR spectrum of lignin was characterized by an O-H band at 3456  $\text{cm}^{-1}$  and an intense C-H band at 2935  $\text{cm}^{-1}$ , which are typical vibrations of methoxyl groups. The absorption of carbonyl groups manifested as a band at 1727  $\text{cm}^{-1}$ . The absorption of the C-H vibration of the aromatic ring was at 1606  $\text{cm}^{-1}$  and 1504  $\text{cm}^{-1}$ . The peaks at 1224  $\text{cm}^{-1}$  and 1121  $\text{cm}^{-1}$  correspond to C-O with an aromatic ring and the ether bond (-O-) at 1030  $\text{cm}^{-1}$ .



**Fig. 2.** FTIR spectra for epoxy resin, epoxy resin-lignin blends, and epoxy resin-lignin-polyamine blends

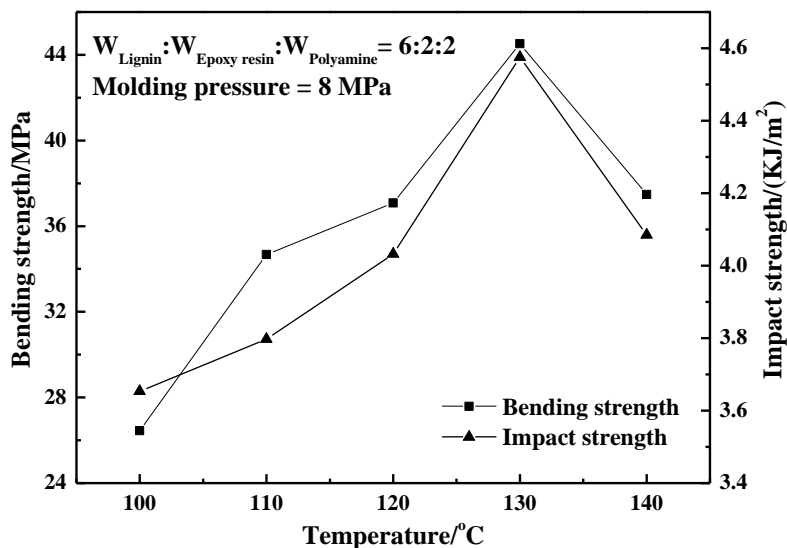
After blending epoxy resin with lignin, the introduction of groups in the epoxy resin structure was indicated by the presence of bands at  $3456\text{ cm}^{-1}$  and  $1727\text{ cm}^{-1}$ . The peaks at  $1121\text{ cm}^{-1}$  were also significantly enhanced. In addition, there was no stretching vibration absorption for the epoxy group at  $914\text{ cm}^{-1}$ . After the addition of the polyamine curing agent, the FTIR spectrum of the main peak for lignin-epoxy resin-polyamine blends was not changed, and the absorption peak for the epoxy group at  $914\text{ cm}^{-1}$  disappeared similarly.

The results for DSC and FTIR analysis showed that the epoxy resin can be cured by enzymatic hydrolysis corn straw lignin, and there may be a chemical bond between active groups within the lignin and epoxy or hydroxyl groups during the curing process. This provided the possibility to form a good interfacial combination between lignin and epoxy resin during the preparation of blends, which is beneficial to the properties of the lignin-epoxy resin composite.

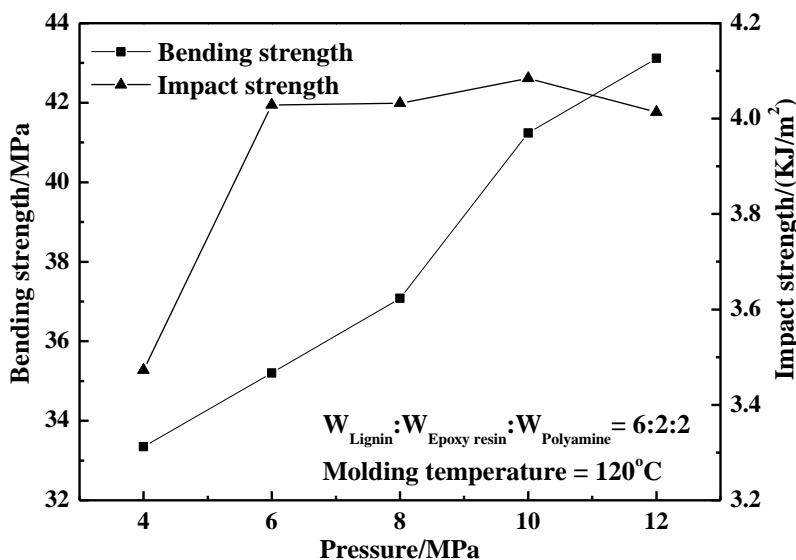
### Mechanical Properties

The effects of molding temperature and molding pressure on the bending strength and impact strength of lignin-epoxy resin-polyamine blends are shown in Fig. 3 and 4, respectively.

As shown in Fig. 3, with an increase of molding temperature, the bending strength and impact strength for lignin/epoxy resin composite initially increased, then declined. The values of bending strength and impact strength reached a maximum of  $44.52\text{ MPa}$  and  $4.58\text{ KJ/m}^2$  at the molding temperature of  $130\text{ }^{\circ}\text{C}$ . The reason for this is that a higher molding temperature contributed to improved fluidity of the epoxy resin, which made the epoxy resin fill the interspace between the lignin flour more uniformly, thereby improving the contiguity between lignin and epoxy resin.



**Fig. 3.** Effects of molding temperature on bending strength and impact strength of lignin epoxy resin-polyamine blends



**Fig. 4.** Effects of molding pressure on bending strength and impact strength of lignin-epoxy resin-polyamine blends

Meanwhile, with the increase in molding temperature, the curing degree for epoxy resin was increased and the activity of lignin was further enhanced, which benefitted the chemical bonding between lignin and epoxy resin, and consequently the cross-linking density of the composite increased. However, an excessively high molding temperature caused the composite to degrade and become brittle, especially the lignin within the composite, which negatively influenced its mechanical properties.

It can be seen from Fig. 4 that the bending strength for the composite increased with an increase in molding pressure, so that the bending strength reached 43.12 MPa when the molding pressure was set at 12 MPa. The impact strength of the composites increased initially and then remained at a stable value. This may be because the low molding pressure decreased the externally acting force, which disturbed the impregnation of epoxy resin into lignin and eliminated residual gas within the composite. With the

increase of molding pressure, the impregnation penetrability of epoxy resin increased, and the molecular interaction force among lignin, epoxy resin, and polyamine was also enhanced. Simultaneously, the residual gas within the composite was more easily eliminated from the blends. When the molding pressure increased further, the compaction rate of lignin-epoxy resin-polyamine blends became saturated, which helped to achieve a suitable balance of the mechanical properties of the composite.

### DMA Analysis

Figure 5 shows the effects of molding temperature on the loss tangent ( $\tan\delta$ ) for lignin/epoxy resin composite. The weight ratio for the constituents of the composite  $W_{\text{Lignin}}:W_{\text{Epoxy resin}}:W_{\text{Polyamine}}$  was 6:2:2, and the molding pressure was set at 8 MPa.

DMA analysis is a useful means of revealing the microscopic relaxation movement of the polymer molecules. It is well known that the  $\alpha$  peaks in the loss tangent ( $\tan\delta$ ) versus temperature curves are closely related to the glass transition. In general, an increase in glass transition temperature ( $T_g$ ) and a decrease in the  $\alpha$ -transition peak height can be related to the increase of cross-linking density in amorphous polymers. It can be seen from Fig. 5 that under the molding temperature of 130 °C, the  $T_g$  temperature of the blends increases and the peak  $\tan\delta$  value decreases with the increase in molding temperature, indicating that the cross-linking density of the blends increases. On the contrary, with a further increase in the molding temperature, the  $T_g$  descends and the peak  $\tan\delta$  increases, revealing that the cross-linking density decreases.

It can be concluded that reasonably increasing the molding temperature would be conducive to enhancing the cross-linking density, and when the molding temperature exceeds 140 °C, the cross-linking density for the composite would be reduced due to the degradation of lignin within the composite. This result is consistent with what was obtained by the above mechanical properties analysis.

Figure 6 shows the effect of molding pressure on the loss tangent ( $\tan\delta$ ) for the lignin/epoxy resin composite. The weight ratio for the constituents of the composite  $W_{\text{Lignin}}:W_{\text{Epoxy resin}}:W_{\text{Polyamine}}$  was 6:2:2, and the molding temperature was set at 120 °C.

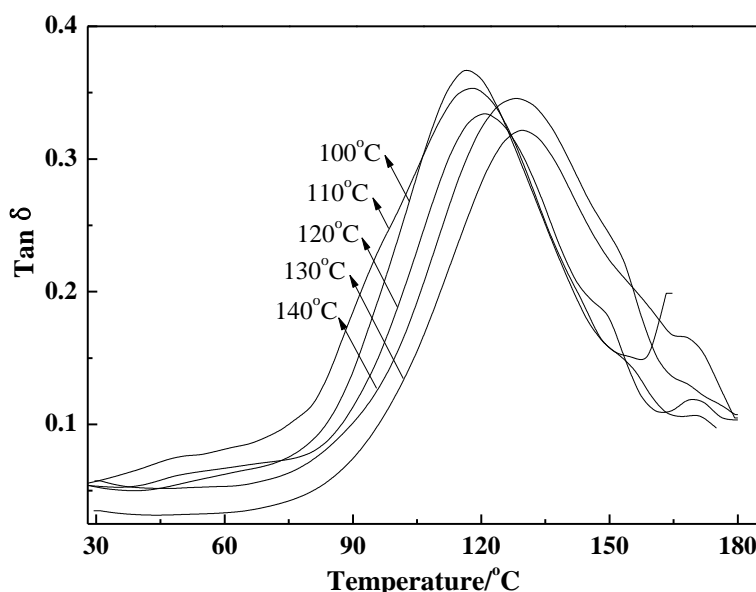
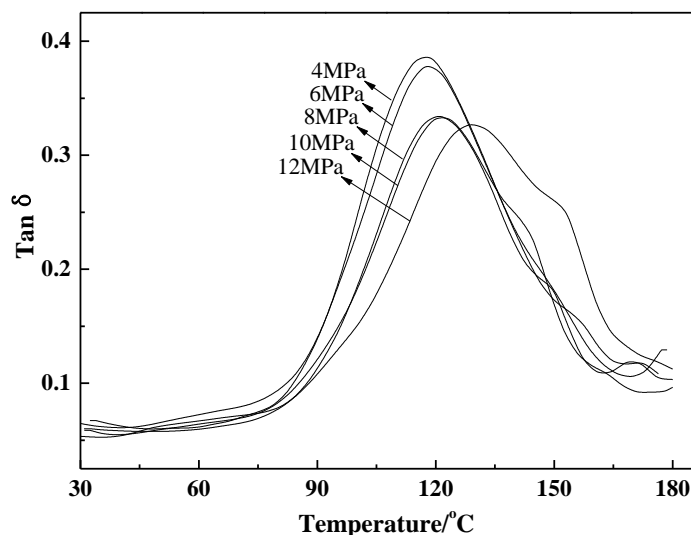


Fig. 5. Effect of molding temperature on  $\tan\delta$  of the lignin-epoxy resin composite

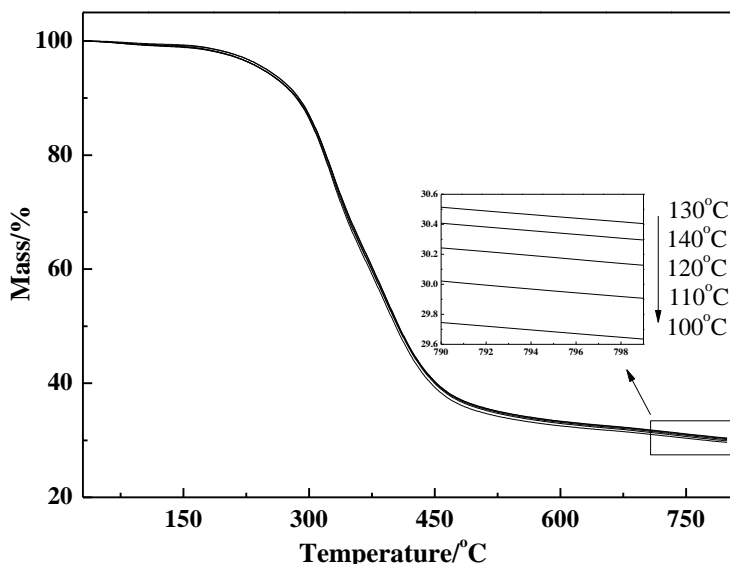


**Fig. 6.** Effect of molding pressure on  $\tan\delta$  of the lignin-epoxy resin composite

Figure 6 illustrates that the  $T_g$  temperature of the blends increases and the peak  $\tan\delta$  value decreases with increasing molding pressure, which indicates that the cross-linking density of the blends increases with the increase of molding pressure. The reason for this is that the higher molding pressure is beneficial to the molecular interaction forces among lignin, epoxy resin, and polyamine. As a result, the interfacial combination between lignin, epoxy resin, and polyamine also improved. This result is also consistent with the results obtained by the above mechanical properties analysis.

### TG Analysis

Figures 7 and 8 exhibit the influence of molding temperature and pressure on the thermal stability of lignin-epoxy resin composite, respectively. The data from TG analysis of lignin-epoxy resin composite under various molding temperatures and molding pressures are also shown in Tables 2 and 3, respectively.

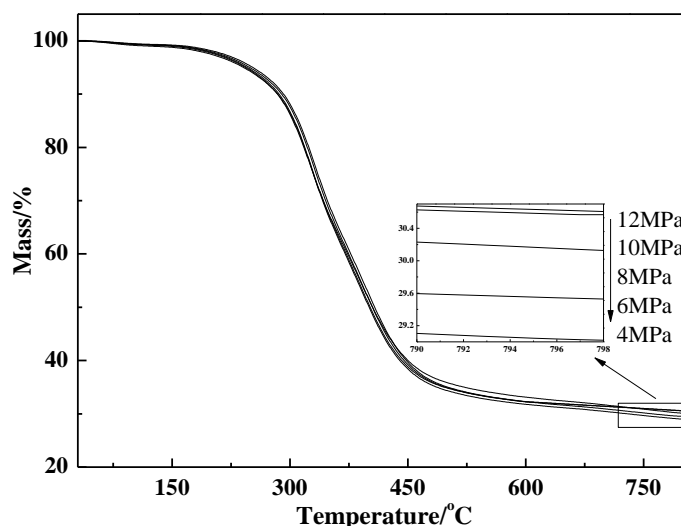


**Fig. 7.** Effect of molding temperature on the thermal stability of lignin-epoxy resin composite



**Table 2.** Data from TG Analysis of Lignin-Epoxy Resin Composite under Various Molding Temperatures

Molding temperature/°C	Residual quantity of 50%/°C	Residual quantity/%
100	403.3	29.64
110	404.8	29.90
120	405.1	30.12
130	406.0	30.40
140	406.2	30.29

**Fig. 8.** Effect of molding pressure on the thermal stability of lignin-epoxy resin composite**Table 3.** Data from TG Analysis of Lignin-Epoxy Resin Composite under Varying Molding Pressure

Molding pressure/MPa	Residual quantity of 50%/°C	Residual quantity/%
4	400.7	29.01
6	402.0	29.53
8	405.1	30.12
10	406.9	30.57
12	407.8	30.61

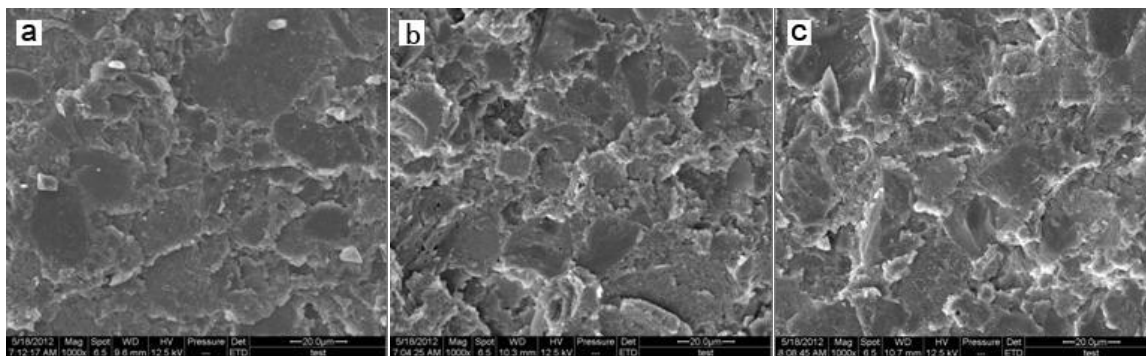
It can be seen from Fig. 7 and Table 2 that, for the lignin-epoxy resin composite, the weight loss temperatures at the weight loss rate of 50% increased with an increase of molding temperature, and the residual weight of the composite showed a tendency to increase initially and then decrease. For the thermal stability of the lignin-epoxy resin composite under varying molding pressure shown in Fig. 8 and Table 3, it can be seen that both the weight loss temperatures at the weight loss rate of 50% and the residual weight for the lignin-epoxy resin composite increased with an increase in molding pressure.

The results of TG analysis indicated that proper molding temperature and molding pressure would help the curing and cross-linking reactions of epoxy resin. The cross-linking density of the composite also consequently increased, which strengthened the

thermal stability of the lignin-epoxy resin composite. These results further proved the data obtained by the above mechanical properties analysis and DMA analysis.

### SEM Observation

Figure 9 shows the fracture morphology for the lignin-epoxy resin composite made under the molding conditions 4 MPa/110 °C, 8 MPa/110 °C, and 8 MPa/130 °C. It illustrates that the lignin can be distributed in the epoxy resin matrix uniformly. There was also no obvious existing interphase, which explained the good interfacial combination between lignin and epoxy resin. It is also a guarantee that the composite has better mechanical properties.



**Fig. 9.** The fracture morphology for lignin-epoxy resin composite a) 4 MPa/110 °C; b) 8 MPa/110 °C; c) 8 MPa/130 °C

However, the degree of interfacial combination between lignin and epoxy resin could be influenced distinctly by different molding conditions. Under the conditions of lower molding pressure and molding temperature, as shown in Fig. 9a, the fracture morphology for the lignin-epoxy resin composite was relatively smooth and characterized by the fracture surface with a certain degree of brittle fracture. The fracture morphology for the composite hot pressed under the higher molding pressure and molding temperature became obviously rough, as shown in Fig. 9b and 9c. Especially for the composite hot pressed under the molding temperature of 130 °C and molding pressure of 8 MPa, the fracture surface shown in Fig. 9c had many traces of tearing that exhibited obvious ductile fracture.

The viscosity of epoxy resin was sensitive to temperature, and the viscosity of blends of lignin and epoxy resin was reduced as the molding temperature increased. Thus, the fluidity of blends and the wettability of epoxy resin to lignin were improved, which improved the dispersibility of lignin into the epoxy resin matrix. Accordingly, the interfacial combination for the lignin-epoxy resin composite was strengthened. Meanwhile, the higher molding pressure was conducive to improving the fluidity of epoxy resin. The lignin was then pressed into the epoxy resin matrix under mechanical force, and as a result, the mechanical entanglement was enhanced and the interfacial combination for lignin-epoxy resin composite was also strengthened. The analysis results were effectively proved by SEM observation.

## CONCLUSIONS

1. Epoxy resin was able to be cured together with enzyme hydrolysis lignin under high temperature, but the curing temperature for the lignin-epoxy resin blends could be reduced by the introduction of a polyamine cure agent. The lignin-epoxy resin composite with a lignin content of up to 60% was prepared by blending lignin with epoxy resin and polyamine using a hot press molding process.
2. The bending strength, impact strength, glass-transition temperature, and thermal stability for the lignin-epoxy resin composite showed a trend of initially increasing and then decreasing with an increase in molding temperature. Optimum performance by composite materials was achieved at the molding temperature of 130 °C.
3. The bending strength, glass-transition temperature, and thermal stability for the lignin-epoxy resin composite improved gradually with an increase in the molding pressure, while the impact strength increased initially and then decreased.
4. A good interfacial combination was formed between lignin and epoxy resin, and the degree of interfacial combination was directly affected by the molding process. Increasing the molding temperature and pressure proved helpful to achieve a better interfacial combination for the composite, and the degree of ductile fracture increased in the fracture surface of composite.

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