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Curing behavior and adhesion properties of epoxy resin blended with phenol-liquefied *Cryptomeria japonica*

Wen-Jau Lee¹ · Chen-Ling Kang¹ · Yi-Chun Chen¹ · Zheng-Ying Wu¹

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Abstract

Epoxy resin is one of the most important thermosetting polymers. Most of the commercial epoxy resins are prepared by reacting bisphenol A with epichlorohydrin. However, epoxy resins have the drawback of being expensive. How to reduce manufacturing cost is an important issue. Solvent liquefaction is an effective method to convert biomass from solid to liquid. Phenol-liquefied wood contains a large amount of phenol structure which has the potential to react with the epoxide group of epoxy resin. In this study, wood of *Cryptomeria japonica* was liquefied in phenol with HCl and H₂SO₄ as a catalyst and named as LW-C and LW-S, respectively. These phenol-liquefied woods were mixed with epoxy resin to prepare blended resins. The curing behavior, thermal properties and wood bonding performance of blended epoxy resins with different weight mixing ratios were investigated. The results show that the gel time of blended resins can be shortened when compared with the neat epoxy resin, especially for those mixed with LW-S. DSC analysis shows blended epoxy resin prepared by mixing with LW-C has a heat flow variation similar to that of neat epoxy resin but a lower peak temperature with less heat released. Blended epoxy resins mixed with LW-C can be used as a wood adhesive to achieve a dry bonding strength similar to that of neat epoxy resin. These results indicated liquefied wood has the potential to prepare epoxy resin. However, the wet bonding strength is still necessary to further improve.

1 Introduction

Petroleum is the most important resource for mankind, being extensively employed as the raw material for energy and various petrochemical products. However, it is a non-renewable resource. In addition, the utilization of petroleum will increase the concentration of carbon dioxide in the air, inducing serious global warming. Therefore, finding alternative resources is of great urgency. Plants are the most plentiful natural resource on earth and are also renewable. However, their solid nature restricts their application. Solvent liquefaction is an effective method to convert them from solid into liquid. The liquefied lignocellulose can be used as raw materials for preparing various synthetic polymers depending on the kind of solvent employed (Pan 2011).

Phenol and polyethylene glycol are two common agents used in liquefying lignocellulose. During the process of solvent liquefaction under acidic condition, wood components

would undergo hydrolysis, degradation and decomposition to become small fragments or monomolecular compounds, followed by reaction with phenol or polyethylene to form a derivative and dissolve in the free solvent. The liquefied products are complex mixtures of free solvent, biomass fragments and derivatives, and other decomposition compounds (Kurimoto et al. 1999; Yamada et al. 2007; Zou et al. 2009). Most of the phenol-liquefied lignocellulose is used for preparing the phenol–formaldehyde resins (PF) which include novolak-type PF resins (Lee and Chen 2008; Mustata 2011) and resol-type PF resins (Alma and Basturk 2006; Lee et al. 2012a, b). On the other hand, polyethylene glycol-liquefied lignocellulose is used as the raw material in various polyurethane resin (PU) products, such as foams (Chen and Lu 2009; Wang et al. 2008; Yao et al. 1996; Yu and Lee 2014), films (Kurimoto et al. 2000, 2001; Lee et al. 2015) and adhesives (Lee and Lin 2008; Ugovšek and Sernek 2013a, b; Ugovšek et al. 2013; Tohmura et al. 2005).

Epoxy resin is one of the most important thermosetting polymers. It has the characteristic of low shrinkage during hardening. Once fully hardened, the cured epoxy resins have good water resistance, heat resistance, chemical resistance, strong mechanical and adhesion properties,

✉ Yi-Chun Chen
chenyc@nchu.edu.tw

¹ Department of Forestry, National Chung-Hsing University, Taichung 402, Taiwan

and better insulation and electrical properties. Therefore, they are widely used in adhesives, coatings, encapsulants and matrices in reinforced composites. At present, most of the commercial epoxy resins are based on the reaction between bisphenol A and epichlorohydrin. However, both are petrochemical products. In addition, bisphenol-A is often undesirable because it can mimic the body's own hormones and may lead to negative health effects (Okada et al. 2008). Therefore, using liquefied lignocelluloses to replace part of the bisphenol A in the preparation of epoxy resins has been tried by some researchers.

Kishi et al. (2006, 2011) synthesized wood-based epoxy resins by reacting resorcinol liquefied wood and polyethylene glycol-liquefied wood with epichlorohydrin. They pointed out the epoxy functionality of the resins would be a dominant factor for crosslink density and the properties of the cured resins. Wu and Lee (2010) liquefied *Dendrocalamus latiflorus* in phenol and polyhydric alcohol. The liquefied bamboos were reacted with bisphenol A and epichlorohydrin to prepare copolymer epoxy resins. The results showed epoxy resins prepared with phenol-liquefied bamboo had higher reactivity and more heat released than those with polyhydric alcohol-liquefied bamboo during curing and had curing behavior similar to neat epoxy resin, even when 50% of bisphenol-A was replaced with liquefied bamboo.

In addition, blending epoxy resin with polyhydric alcohol liquefied wood was also conducted. Kobayashi et al. (2000, 2001) developed a new epoxy resin system prepared by blending epoxy resin with polyethylene glycol-liquefied wood. The results showed liquefied wood was incorporated into epoxy resin to form a homogeneous polymer network. Xie and Chen (2005) prepared an epoxy resin by mixing the ethylene carbonate liquefied bagasse with diglycidyl ether of bisphenol A. The triethylenetetramine was added as a curing agent and the resin was completely cured at 100 °C. They indicated this resin displayed higher adhesive shear strength and better thermal stability than a commercial epoxy resin. Asano et al. (2007) used polyethylene glycol as a solvent to liquefy ozone-treated wood and blended the liquefied products with oily epoxy resin (diglycidyl ether of bisphenol A) and water-soluble epoxy resin (polyethylene glycol diglycidyl ether). They found the resin mixed with oily epoxy resin and cured by citric acid had a tensile strength similar to commercial plastics. Wu and Lee (2011) investigated the curing behavior and adhesion properties of epoxy resin blended with polyhydric alcohol-liquefied wood. The results showed blended epoxy resins could cure at room temperature with an exothermic reaction and had good dry bonding strength when triethylenetetramine was added as a curing agent. However, the wet bonding strength could be improved if the glued strips were set under higher temperature (60 and 90 °C). However, the properties of epoxy resin blended with phenol-liquefied wood were still not found.

Cryptomeria japonica is one of the most important coniferous artificial plantation species in Taiwan. However, due to the income from the harvest being less than their cost, many *Cryptomeria japonica* trees have exceeded the appropriate cutting age, resulting in tree growth pausing and large areas of aging forest. Developing high value-added *Cryptomeria japonica* products has become an important issue. Therefore, in this study, *Cryptomeria japonica* wood was chosen as the raw material and the phenol-liquefied wood was blended with epoxy resin to prepare a novel adhesive system. The curing behavior and adhesion properties of different blended epoxy resins were investigated.

2 Experimental

2.1 Materials

Wood powders of *Cryptomeria japonica* (Japanese cedar) with a dimension passing 20-mesh were used as the raw material for liquefaction. Wood strips of *Alnus formosana* (Formosan alder) with the dimensions of 260 × 25 × 10 mm³ and equilibrium moisture content of 12% were used as an adherent for the bonding strength test. Chemicals such as phenol, sulfuric acid (H₂SO₄) and hydrochloric acid (HCl) were used for wood liquefaction. Bisphenol A (Hayashi Chemical Ind.), epichlorohydrin (Tedia Chemical Co.) and sodium hydroxide (NaOH) were used for synthesizing the epoxy resin. Triethylenetetramine (TETA, Hayashi Chemical Co.) that has the active hydrogen equivalent weight (AHE) of 24.4 g eq⁻¹ was used as a crosslinking hardener. All these chemicals were reagent grade and used without further purification.

2.2 Wood liquefaction

The wood powders with a dimension passing 20-mesh were oven-dried at 105 °C for 12 h prior to liquefaction. The weight ratio of phenol to wood was 3/1, and the dosage of HCl and H₂SO₄ (with the concentration of 36 and 95%, respectively) added were 10 and 5% (based on the weight of phenol), respectively. The liquefaction was conducted in a 1000 mL four-neck round-bottom glass reactor equipped with stirrer, thermometer, and reflux condenser under constant stirring for 60 min at 110 and 130 °C for those using HCl and H₂SO₄ as the catalyst, respectively. The liquefied products that used HCl and H₂SO₄ as the catalyst were named LW-C and LW-S, respectively. The residue content, viscosity, pH and nonvolatile content of liquefied products were measured as previously reported (Lee et al. 2012a).

2.3 Synthesis of epoxy resin

The mixture of epichlorohydrin and bisphenol A with a molar ratio of 5/1 was charged into a four-necked glass reaction flask, and the synthesis was kept at 110 °C for 4 h under continuous stirring and gradually dripping of 1 mol NaOH (40%_(aq)) followed by another 1 h of heating. The byproduct of NaCl was removed by filtration, while the water and unreacted epichlorohydrin were removed by reduced pressure distillation. The viscosity and epoxy equivalent weight (EEW) of the epoxy resin were 1096 cps and 269.3 g eq⁻¹, respectively.

2.4 Curing test of blended epoxy resins

The blended epoxy resins were prepared by mixing epoxy resin with liquefied wood with weight ratios of 100/30, 100/50, and 100/70. TETA was used as a crosslinking hardener. For the curing test, an equivalent epoxy resin and TETA that was calculated based on the EEW and AHE, i.e. 10 part of TETA per hundred parts of epoxy resin (10 phr), were mixed and stirred continuously until cured. The mixture was continued to be stirred until the resin cured which was defined as the gel time. The maximum exothermic temperature during curing reaction was monitored with an electronic thermometer. After being fully cured, the resin was ground into powders and immersed in acetone for 24 h, followed by filtration and drying. The percentage of weight retention after acetone immersion was used to indicate the gel degree of cured resins.

2.5 DSC thermo-setting analysis of blended epoxy resins

The thermo-setting behavior of epoxy resin and blended epoxy resins mixed with TETA was performed with a differential scanning calorimeter (DSC; Perkin-Elmer DSC-7). The sample was heated from 30 to 250 °C at a heating rate of 10 °C min⁻¹ under an atmosphere of N₂.

2.6 FTIR analysis of cured resins

Spectrum analysis was carried out by mixing cured epoxy resin powders with KBr (1/100; wt/wt) and detected with Fourier transform infrared spectroscope (FTIR; Mattson Genesis II) using a diffuse reflectance mode and a deuterated triglycine sulfate detector with a wavenumber between 650 and 4000 cm⁻¹ at a resolution of 8 cm⁻¹ and scanned 16 times.

2.7 TGA thermo-degradation of cured resins

For TGA analysis (Perkin-Elmer Pyris 1), about 5 mg of the sample was put into a platinum sample pan and heated from 50 to 700 °C at a heating rate of 10 °C min⁻¹ under an atmosphere of N₂ with a flow rate of 20 mL min⁻¹. Curves of weight loss (TG) and derivative TG (DTG) were plotted. The onset temperatures and weight loss percentage at each thermal degradation stage were calculated.

2.8 Bonding strength of blended epoxy resins

Epoxy resin and blended epoxy resins mixed with 10 phr of TETA were applied to strips of *A. formosana* at a rate of 200 g m⁻². The glued strips were cured at room temperature under a pressure of 1.5 MPa for 24 h. The compression shear bonding strength was done under dry and wet conditions, according to the CNS 5809 standard by means of a universal strength testing machine (Shimadzu UEH-10) at a loading speed of 1 mm min⁻¹. For the wet bonding strength, specimens were immersed in boiling water for 4 h, followed by drying in an oven at 60 °C for 20 h, and finally immersed in boiling water again for 4 h. The wood failure of the testing samples was estimated by the naked eye to the nearest 5% of shear area. Eight specimens were tested for each test set.

3 Results and discussion

3.1 Properties of liquefied wood

During the liquefaction period, wood components will undergo hydrolysis, degradation and decomposition to become small fragments or monomolecular compounds followed by reaction with phenol, and the formed derivatives are then dissolved in the free phenol (Lin et al. 2001a, b, 2004).

Table 1 shows the properties of the phenol-liquefied woods. Among which, LW-C and LW-S were liquefied with HCl and H₂SO₄ as the catalyst, respectively; and both of them resulted in acidic products. The residue and non-volatile contents can be used as indicators for the efficiency of liquefaction. The residue content for LW-C and LW-S are 3.9 and 2.2%, respectively. The weight percent of wood in

Table 1 Properties of phenol-liquefied woods

Liquefied wood	Catalyst	Residue content (%)	Viscosity (cps)	pH	Nonvolatile content (%)
LW-C	HCl	3.9	4510	0.46	43.6
LW-S	H ₂ SO ₄	2.2	7650	0.51	56.9

the initial stage of liquefaction for LW-C and LW-S were 23.3 and 24.1%, respectively. However, the nonvolatile content for LW-C and LW-S are higher than those, indicating some of the phenol combined with wood components to form derivatives. Comparing LW-C and LW-S, the latter has more nonvolatile content than the former. As mentioned above, wood liquefied with H₂SO₄ as a catalyst has a better efficiency in the conversion than that with HCl, i.e. it more effectively promotes the decomposition of wood components and their derivatization with phenol. On the other hand, LW-S has a viscosity higher than that of LW-C, 7650 cps versus 4510 cps. It may be due to the LW-S containing more derivatives than LW-C, which has a larger molecular weight and leads the liquefied wood with higher viscosity.

3.2 Curing characteristics of blended epoxy resins

The curing reaction of epoxy resins can take place at room temperature when TETA is added as a crosslinking hardener, which will display an exothermic phenomenon during the curing period. Table 2 shows the viscosity and curing characteristics of epoxy resin blended with liquefied woods. The viscosity of neat epoxy resin is 1096 cps but it increases with the amount of liquefied wood added. Comparing LW-C and LW-S, blended resins mixed with LW-S have the viscosity higher than that of LW-C.

The gel time for the neat epoxy resin is 21.0 min after 10 phr of TETA was added and the maximum temperature reaches 43.4 °C during the curing reaction. While being mixed with liquefied woods, the blended resins show a shorter gel time with a higher maximum temperature as compared with the neat epoxy resin, indicating they have higher reactivity. In the case of that mixed with 30 parts of LW-S, the mixture gel at 3.5 min reaches a temperature of 127.8 °C. This result indicates the phenol-liquefied wood promotes the curing reaction of epoxy resins, which may be due to the aromatics existing in the phenol-liquefied wood providing a synergistic effect with amine for the curing reaction (Gao and Yu 2003). In addition, the curing reaction

may also be influenced by the acidity of blended resins. In a previous study, it was found that the higher the pH value (from 0.5 to 9.0) of phenol-liquefied wood, the longer the gelation time of the blended epoxy would be (Kang and Lee 2017). However, the curing time prolonged when the addition amount of the liquefied wood is further increased, such as from 30 parts to 50 parts and 70 parts. This means the curing reaction is relaxed by the exceeded liquefied wood that did not participate in the curing reaction.

When comparing LW-C and LW-S added epoxy resins, the latter has shorter gel time with higher maximum temperature than that of the former. These results are similar to a previous study that used LW-C and LW-S as raw materials for preparing resol-type PF resins (Lee et al. 2012a). It may be due to the LW-S having more derivatives with larger molecular weight and higher viscosity than LW-C.

The weight retention of cured resins after acetone immersion can be used as an index of gel degree. As can be seen in Table 2, the neat epoxy resin has weight retention of 85%. However, it is lower for blended epoxy resins and continuously decreases as the amount of liquefied wood increases. Nevertheless, the values are larger than the weight percentage of epoxy resin in blended epoxy resins, indicating some of the liquefied woods participated in the polymerization of the curing reaction.

Based on the above description, the gel time for epoxy resin blended with LW-S is too short to be applied to gluing operation. Therefore, only 50 part of LW-S-added epoxy resin (100/50; wt/wt) was selected for subsequent testing and the results are used for comparison with LW-C-added epoxy resin.

3.3 DSC thermo-setting of blended epoxy resins

DSC analysis has been used as an effective tool to analyze the curing process of thermosetting polymers. Figure 1 shows the heat flow variation of epoxy resin and blended epoxy resins prepared by mixing epoxy resin with liquefied wood (100/50; wt/wt) during DSC heat scanning.

Table 2 Curing characteristics of epoxy resin blended with liquefied woods

Epoxy (part)	Liquefied wood		Viscosity (cps)	Gel time (min)	Maximum temp. (°C)	Weight retention ^a (%)
	Kind	Amount (part)				
100	–	–	1096	21.0	43.4	85.0
100	LW-C	30	1994	8.0	51.4	80.2
100	LW-C	50	2997	10.5	52.0	79.2
100	LW-C	70	3870	12.0	47.5	72.6
100	LW-S	30	3906	3.5	127.8	79.3
100	LW-S	50	5040	5.0	86.8	74.6
100	LW-S	70	7540	5.5	100.8	72.3

^aAfter acetone immersion

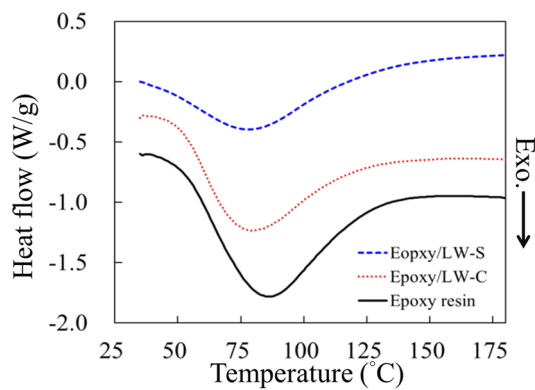


Fig. 1 DSC curves of epoxy resin and blended epoxy resins; the weight ratio of epoxy resin to liquefied wood is 100/50

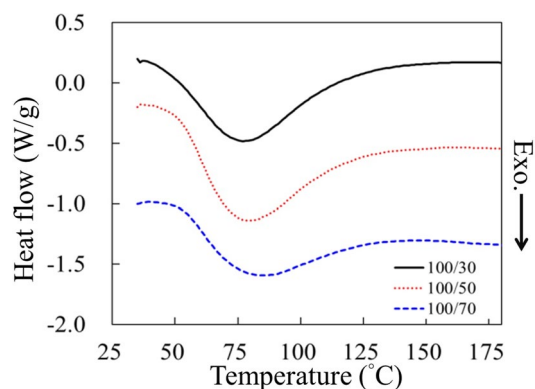


Fig. 2 DSC curves of blended epoxy resins with different weight ratios of epoxy resin to liquefied wood (LW-C)

All display a broad exothermal peak caused by the heat released due to the crosslinking reaction. Among which, blended epoxy resin prepared with LW-C has a heat flow curve similar to neat epoxy resin. However, it differs from that prepared with LW-S, which shows less heat released and lower exothermic peak height. As mentioned in previous discussions, epoxy resin mixed with LW-S has a faster curing reaction than that with LW-C. This may result in faster molecular growth and succeed by restricting molecular motion, which leads to a lower degree of polymerization (Kobayashi et al. 2000; Pérez et al. 2011).

Figure 2 shows the DSC curves of blended epoxy resins prepared with different amounts of LW-C. The exothermic phenomenon for the resin with a weight ratio of 100/70 is more moderate than others.

Table 3 shows the thermo-setting parameters calculated according to the exothermic peak. The neat epoxy resin has a peak temperature of 84 °C with a released heat of 285 J g⁻¹. Blended epoxy resins have a lower peak temperature than neat epoxy resin, indicating the fastest curing reaction for blended resins occurred at temperatures lower than that of the neat epoxy resin. However, they have less heat released than neat epoxy resin, indicating they underwent less crosslinking reaction during the curing reaction. Comparing the blended epoxy resins mixed with LW-C and LW-S, using LW-S as a raw material leads to significantly less heat release than that with LW-C when the weight ratio of epoxy resin to liquefied wood is 100/50. On the other hand, the released heat obviously decreased for LW-C at a weight ratio of 100/70.

3.4 FTIR analysis of cured resins

Figure 3 shows the FTIR spectra of liquefied *C. japonica* wood, cured epoxy resin and blended epoxy resins. The cured Epoxy/LW-C has a spectrum similar to that of cured pure epoxy resin, indicating that the main structure of the cured epoxy resin was maintained even with the addition of LW-C. However, the intensity of the peaks appears at 2800–3000 cm⁻¹ (asymmetric and symmetrical stretching vibration of CH₂), 1106 cm⁻¹ (stretching vibration of C–H of the substituted benzene), 1037 cm⁻¹ (symmetrical stretching vibration of C–O–C of benzyl-alkyl ether) and 827 cm⁻¹ (out of plane vibration of C–H of 1,4-substituted benzene) for Epoxy/LW-C decreases (Cherdoud-Chihani et al. 2003). On the other hand, the peak of 754 and 692 cm⁻¹ that was attributed to out-of-plane deformation vibration of ortho-substituted aromatic ring and the mono-substituted aromatic ring (i.e. phenol) of LW (Pan et al. 2009), respectively, becomes more obvious. This result illustrates LW is introduced into the molecular structure of cured epoxy resins.

Table 3 DSC thermo-setting parameters of epoxy resin blended with liquefied woods

Epoxy (part)	Liquefied wood		Onset (°C)	Peak (°C)	End (°C)	Delta H (J/g)
	Kind	Amount (part)				
100	–	–	51	84	126	285
100	LW-C	30	46	77	119	178
100	LW-C	50	51	76	120	209
100	LW-C	70	52	79	123	116
100	LW-S	50	41	80	124	132

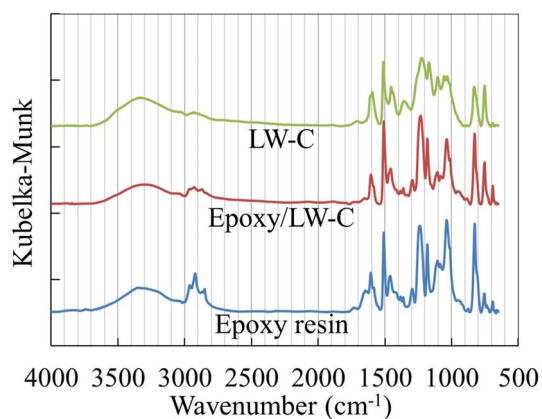


Fig. 3 FTIR spectra of liquefied *C. japonica*, epoxy resin and blended epoxy resins; the weight ratio of epoxy resin to liquefied wood is 100/50

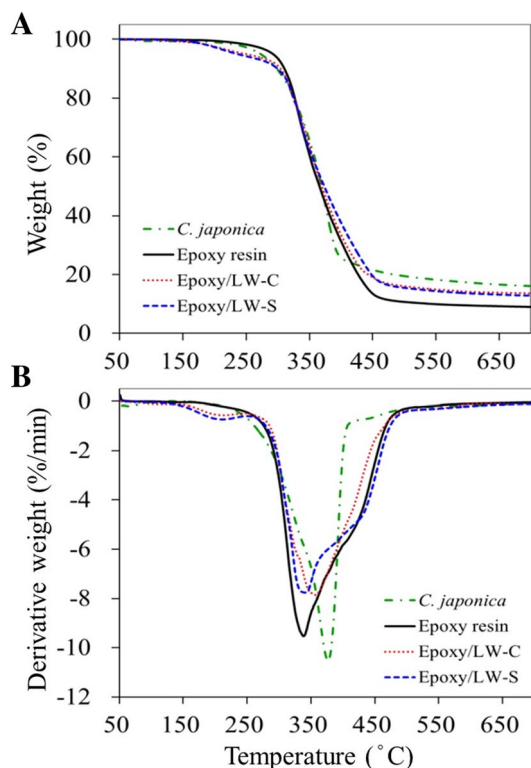


Fig. 4 TG (a) and DTG (b) curves of *C. japonica*, epoxy resin and blended epoxy resins; the weight ratio of epoxy resin to liquefied wood is 100/50

3.5 TGA thermo-degradation of cured resins

Figure 4 shows the TG and DTG curves of *C. japonica* wood, cured epoxy resin and blended epoxy resins. Their respective thermal degradations can be divided into three stages. Table 4 shows the initial temperature (onset) and

weight loss for each stage. For *C. japonica* wood, it has an onset temperature of 216, 298 and 416 °C for each thermal degradation stage with weight losses of 10.1, 62.8 and 10.7%, respectively. The weight loss in the first stage is due to the thermal degradation of hemicellulose. The rapid weight loss at temperatures between 298 and 416 °C is caused by thermal degradation of cellulose and lignin. Then, the moderate weight loss contributes to the rearrangement of the aromatic structure and the dehydrogenation of char materials (Byrne and Nagle 1997; Sjöström 1993). The obvious weight loss for the neat epoxy resin appears at temperatures between 300 and 440 °C, which is similar to the results of Laza et al. (2008). When mixing with liquefied wood, the weight loss begins at a lower temperature. This may be due to liquefied woods containing some components that cannot participate in the crosslinking reaction, which has lower heat resistance and will be degraded at a lower temperature (Lin et al. 2001b, 2004). However, the TG curves shift to a higher temperature at temperatures over 330 °C, indicating blended epoxy resins have better heat resistance than neat epoxy resin. This is because LW-added epoxy resin contains the structure of lignin that has better heat stability and can provide the blended resins with higher heat resistance. In addition, the lower peak height as shown in DTG curves also proves that LW-added epoxy resins have better heat resistance than that of neat epoxy resin. When comparing Epoxy/LW-C and Epoxy/LW-S, Epoxy/LW-C has the peak temperature shifts to higher value, which means Epoxy/LW-C has better heat resistance than Epoxy/LW-S. This is because Epoxy/LW-C underwent a more complete crosslinking reaction than Epoxy/LW-S as mentioned in discussion of DSC thermo-setting analysis.

3.6 Bonding strength of blended epoxy resins

Table 5 shows the bonding strength of epoxy resin and blended epoxy resins. The dry bonding strength and percent of wood failure of epoxy resin are 11.2 MPa and 100%, respectively. Although LW-C and LW-S had similar pH value (0.46 vs. 0.51), blended epoxy resins mixed with LW-C have a dry bonding strength similar to the neat epoxy resin, even when the added amount reaches 70 parts but the blended epoxy resin mixed with LW-S has worse dry bonding strength than neat epoxy resin. According to statistical analysis, there was no significant difference between epoxy resin and blended epoxy resins mixed with LW-C, while mixed with LW-S was significantly different. As mentioned above, epoxy resin mixed with LW-S provides a short gel time which may cause worse wettability and penetration. In addition, DSC analysis showed it has a lower degree of polymerization. Both of these results may be the reason why the blended epoxy

Table 4 TG parameters of *C. Japonica*, epoxy resin and blended epoxy resins

Sample	1st stage		2st stage		3st stage		Char yield (700 °C) (%)
	Onset temp. (°C)	Weight loss (%)	Onset temp. (°C)	Weight loss (%)	Onset temp. (°C)	Weight loss (%)	
<i>C. Japonica</i>	216	10.1	298	62.8	416	10.7	16.0
Epoxy resin	227	6.8	301	77.2	440	7.0	9.0
Epoxy/LW-C ^a	167	8.3	296	71.9	445	6.2	13.6
Epoxy/LW-S ^a	167	9.6	297	73.1	463	4.4	12.8

^aThe weight ratio of epoxy resin to liquefied wood is 100/50

Table 5 Bonding strength of epoxy resin blended with liquefied woods

Epoxy (part)	Liquefied wood		Bonding strength (MPa) ^a	
	Kind	Amount (part)	Dry	Wet ^b
100	–	–	11.2 ± 1.7 ^c (100)	5.5 ± 0.9 ^c (60)
100	LW-C	30	12.7 ± 1.0 ^c (90)	2.7 ± 2.6 ^d (10)
100	LW-C	50	10.7 ± 3.1 ^{cd} (45)	3.3 ± 1.4 ^d (5)
100	LW-C	70	12.4 ± 1.9 ^c (85)	2.6 ± 2.1 ^d (5)
100	LW-S	50	7.9 ± 1.8 ^d (10)	0.3 ± 0.2 ^e (0)

^aGroups with same letters in column indicate that there is no statistical difference ($p < 0.05$) between the samples according to Duncan's multiple range test. The values in parentheses are percent of wood failure. Eight specimens were tested for each test set

^bRepetitive boiling water soaking

resin mixed with LW-S had lower bonding strength than that mixed with LW-C, rather than the effect of pH value.

Meanwhile, the wet bonding strength of blended epoxy resins is less than that of the neat epoxy resin, regardless of whether it is mixed with LW-C or LW-S. This result is similar to Kobayashi et al. (2001) who prepared the epoxy resins by blending them with polyethylene glycol-liquefied wood. This may be due to liquefied woods containing some components that cannot participate in the crosslinking reaction (Lin et al. 2001b, 2004), which reduced the cross-link density of the cured resin and resulted in the glue layer being more easily hydrolyzed and destroyed during the process of repeated boiling water immersion. In addition, the worse wettability and penetration due to high-viscosity and rapid gelation of blended epoxy resin may be another factor leading to their wet bonding strength being lower than that of the epoxy resin.

4 Conclusion

Wood powders of *C. japonica* were liquefied in phenol with HCl and H₂SO₄ as a catalyst. The blended epoxy resins were prepared by mixing epoxy resin with liquefied woods at different weight ratios. The results show wood liquefied with H₂SO₄ as a catalyst has better efficiency than that with HCl. Epoxy resins can cure at room temperature when TETA is added as a crosslinking hardener. While mixing with liquefied woods, the gel time can be shortened, especially for those mixed with LW-S. However, the weight retention of cured resins decreases as the amount of liquefied wood increases. DSC analysis shows blended epoxy resin prepared with LW-C has a curing behavior similar to neat epoxy resin but the peak temperature shifts have better heat resistance than neat epoxy resin at a temperature above 330 °C. Blended epoxy resins mixed with LW-C can be used as a wood adhesive which has the dry bonding strength similar to neat epoxy resin but less wood failure.

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