# Polydimethylsiloxane Containing Isocyanate Group-Modified Epoxy Resin: Curing, Characterization, and Properties

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**ABSTRACT:** A synthesized polydimethylsiloxane containing an isocyanate group was used to improve the flexibility and to reduce the internal stress of epoxy resin cured with MDA (4,4'-methylene dianiline). The effect of polysiloxane content on the curing kinetics of a novolac-type epoxy modified with an isocyanate group was investigated. It was found that the modified epoxy resin showed significant improvement in impact strength. The polysiloxane containing isocyanate groups effectively depressed the internal stress of cured epoxy resins by reducing the flexural modulus and the coefficient of thermal expansion, while the glass transition temperature was increased. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 73: 2739–2747, 1999

Key words: polydimethylsiloxane; isocyanate group; epoxy resin

# INTRODUCTION

Epoxy resins are widely used in various industrial applications, such as high-performance protective coatings, structural adhesives, encapsulation of semiconductor devices, etc. However, they are relatively brittle and have poor impact resistance. Toughening of epoxy resins with reactive diluents and/or oligomers has been an active field of research.<sup>1-4</sup> Extensive investigation on the improvement in flexibility of epoxy resin has resulted in better understanding of structure–properties relationships. Many investigations have concentrated on flexibility and lowering the internal stress of epoxy resins using elastomers. It is known that the

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factors influencing the flexibility of modified epoxy resin are the particle size, distribution, and the content of elastomer in the epoxy resin matrix.<sup>5–8</sup> In this study, a polydimethylsiloxane containing an isocyanate group was prepared to modify epoxy resins that were then cured by MDA (4,4'-methylene dianiline). The effect of polysiloxane content on the curing kinetics of novolac-type epoxy modified with polysiloxane containing isocyanate end groups was investigated. Thermal and mechanical properties of polydimethylsiloxane containing isocyanate groupmodified epoxy networks were also studied.

#### **EXPERIMENTAL**

#### Materials

The *o*-cresol-formaldehyde novolac epoxy resin (ESCN-195XL, epoxy equivalent weight EEW = 200 g/Eq) used in this study was supplied by the Sumitomo Co., Japan. The 4,4'-methylene di-

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aniline (MDA) used as a curing agent, with an amine equivalent weight of 50, was supplied by the Tokyo Chemical Industry Co., Japan. The  $\alpha,\omega$ -diol polydimethylsiloxane (Tegomer 2111), with a hydroxyl content of 5.6 wt %, was purchased from the Goldschmidt AG Co., Germany. Phenyl isocyanate and *m*-xylylene diisocyanate were supplied by the Tokyo Chemical Industry Co., Japan. Stannous octoate (T-12) was used as a catalyst in the synthesis of isocyanate-terminated polydimethylsiloxane.

# Preparation of Polydimethylsiloxane Containing an Isocyanate Group

 $\alpha,\omega$ -Diol polydimethylsiloxane (0.5 mol) with 0.2 wt % stannous octoate was heated under nitrogen atmosphere at 80°C in a 500-mL four-necked round-bottomed flask equipped with a capped  $CaCl_2$  drying tube and a thermometer. For the reaction, phenyl isocyanate (0.5 mol) was added dropwise to the mixture under a nitrogen atmosphere. The reaction completion was confirmed by the disappearance of the characteristic NCO peak (at 2270 cm<sup>-1</sup>) in the IR spectrum. Then *m*-xylylene diisocyanate (0.5 mol) was added dropwise to the mixture. The reactants were stirred and maintained at 80°C until the absorption peak of the OH group in the IR spectrum disappeared. The synthesis of PDMS-based urethane prepolymer is shown in Scheme 1.



## Preparation of Polydimethylsiloxane Containing an Isocyanate Group-Modified Novolac-Type Epoxy

The novolac-type epoxy resins, modified with 0, 5, 10, 15, 20, and 25 phr polydimethylsiloxane con-

taining an isocyanate group, were prepared by a hot-melt method. The novolac-type epoxy resin was heated to 120°C, and then vigorously stirred for 6 h. The polydimethylsiloxane containing an isocyanate group was added at this stage while maintaining the reaction temperature at 120°C for 2 h, until the mixture was completely miscible and homogeneous. Epoxy resin was placed in a vacuum oven at 100°C for 30 min to remove residual moisture. MDA was then placed in an oven at 100°C for 30 min to heat until the MDA was melted. Following this process, MDA and a polysiloxane-modified novolac epoxy resin were mixed carefully, until homogeneous and clear, without forming any bubbles. The mixture was degassed again under vacuum for 2 min, poured into hot molds with proper sample shapes, and cured in an oven at 160°C for 180 min. After curing, samples were cooled down to room temperature and kept in a desiccator for further characterization and properties testing.

### **Study on Curing Kinetics**

Samples for measuring the thermal data were prepared by mixing epoxy resin with a 24.75 phr curing agent (MDA), followed by milling to obtain fine powder. Small samples (5–10 mg) of powder were then placed in a DSC cell, and the normal scanning procedures were carried out with a Du-Pont 2100 DSC Thermal Analyzer, coupled with a TA 2000 data analysis system. Dynamic DSC scanning was carried out up to 300°C at a heating rate of 10°C/min, under nitrogen atmosphere.

DSC has been used extensively for the characterization of curing kinetics for thermosetting resins,<sup>9–12</sup> such as epoxy and unsaturated polyesters. The data from the dynamic DSC measurements are analyzed by following the kinetic equation :

$$d\alpha/dt = k\alpha^m (1-\alpha)^n \tag{1}$$

where  $\alpha$  is the fractional conversion, k(T) is rate constant, and m and n are constants describing the order of the reaction.

The fractional conversion at various temperatures was calculated by  $\alpha = H_T / H_{\text{total}}$ , where  $H_{\text{total}}$  is the total amount of exothermic heat evolved, and  $H_T$  is the heat evolved at temperature T. Temperature dependence of the constant k(T) may be described by the Arrhenius expression, as shown in eq. (2)

$$k(T) = A \exp(-Ea/RT)$$
(2)



**Figure 1** IR spectra of  $\alpha, \omega$ -diol polydimethylsiloxane and polydimethylsiloxane with an isocyanate group.

where A is a pre-xponential factor,  $E_a$  is activation energy (J/mol), R is the gas constant, and T is temperature (K).

#### **Characterization and Properties of Cured Resin**

The IR spectra were recorded with a Perkin-Elmer IR spectrophotometer operated with a resolution of 2.4 cm<sup>-1</sup> in the transmission mode. <sup>1</sup>H-NMR spectra were recorded on a Bruker AM-300 MHZ-NMR with deuterated CDCl<sub>3</sub> solvent, and tetramethylsilane was used as an internal standard. The Izod impact strengths of the unmodified and modified epoxy resins were measured by means of the Izod Impact Test following the specifications of ASTM-D256. The dimensions of the specimens were  $63 \times 10 \times 3.2$  mm. The flexural properties of cured resins were tested using a Shimadzu AGS-500 Universal Testing Machine. Flexural strength and modulus were obtained at a crosshead speed of 1.3 mm/min according to ASTM-D790. A three-point bending system was chosen, and rectangular bar specimens,  $60 \times 25$  $\times$  3.2 mm, were molded directly by a transfer molding process. The coefficient of thermal expansion (CTE) was measured with a DuPont 943 thermal mechanical analyzer in accordance with ASTM E831-86. A specimen of 5 mm in length was used at a heating rate of 5°C/min. Normally, the thermal expansion increases with temperature, and the CTE values were calculated from the slope. An abrupt change in slope of the expansion curve indicates a transition of the material from one state to another. The moisture pickup was determined by placing preweighed  $25 \times 25 \times 3.2$ -mm cured specimens in boiling water for 72 h. The specimens were removed and were cooled to ambient temperature and then wiped dry and weighed to determine the weight gain.



**Figure 2** <sup>1</sup>H-NMR spectra of  $\alpha, \omega$ -diol polydimethylsiloxane and polydimethylsiloxane with an isocyanate group.

# **RESULTS AND DISCUSSION**

# Preparation of Polydimethylsiloxane Containing an Isocyanate Group

The synthesized polydimethylsiloxane containing an isocyanate group was identified by IR spectral measurements. Figure 1 shows the IR spectra of  $\alpha, \omega$ -diol polydimethylsiloxane and polydimethylsiloxane containing an isocyanate group. The completion of an isocyanate-terminated prepolymer formation was confirmed by the disappearance of the hydroxyl group absorption peak at 3500 cm<sup>-1</sup> and the appearance of the absorption peak of urethane at 3316 cm<sup>-1</sup> (—NH) and 1736 cm<sup>-1</sup> (C=O).

Figure 2 shows the <sup>1</sup>H-NMR spectra of  $\alpha, \omega$ -diol polydimethylsiloxane and polydimethylsiloxane containing an isocyanate group. The <sup>1</sup>H-NMR spectra of  $\alpha, \omega$ -diol polydimethylsiloxane showed absorption peaks at 0.04 ppm (CH<sub>3</sub> in PDMS); 0.5, 1.31, 1.51, and 3.6 ppm (CH<sub>2</sub> in PDMS); 1.69 ppm (OH in PDMS). Polydimethylsiloxane containing an isocyanate group reacts rapidly with deuterated CDCl<sub>3</sub> solvent and produces an inter-

ference in the absorption of NMR. Two drops of methanol, which will react with the isocyanate group, were added to this solution and also con-



Figure 3 Dynamic runs of DSC in the temperature range between  $30-300^{\circ}$ C at a heating rate of  $10^{\circ}$ C/min. ESCN-195XL/MDA/polysiloxane (a) 100/24.75/0, (b) 100/24.75/5, (c) 100/24.75/10, (d) 100/24.75/15, (e) 100/24.75/20,(f) 100/24.75/25. (phr = part per hundred parts of resin.)

		Polysiloxane (phr)						
Epoxy Resins		0	5	10	15	20	25	
ESCN-195XL	$\begin{array}{c} T_p \; (^{\circ}\mathrm{C}) \\ \Delta H \; (\mathrm{J/g}) \end{array}$	$193.40\ 356.4$	$190.96 \\ 343.9$	$193.51 \\ 330.1$	$192.59 \\ 321.0$	$194.90 \\ 302.3$	$193.65 \\ 288.4$	

Table ITypical Curing Characteristics of Polysiloxane-Modified Epoxy Resins from DSCMeasurements at a Heating Rate of 10°C/min

 $T_p$  is peak temperature,  $\Delta H$  is heat of cure reaction.

phr = part per hundred parts of resin.

tribute to the carbamate peak. The <sup>1</sup>H-NMR spectra of polydimethylsiloxane containing an isocyanate group showed absorption peaks at 5.03 and 6.62 ppm (H in carbamate); 7.0–7.3 ppm (aromatic proton in phenyl isocyanate); 7.16 ppm (aromatic proton in *m*-xylylene diisocyanate), and 3.69 ppm (CH<sub>3</sub> in the urethane group).

# Curing Kinetics of Polydimethylsiloxane Containing an Isocyanate Group-Modified Novolac-Type Epoxy Resin System

The curing behavior of epoxy resin depends upon the curing conditions. The curing conditions can greatly affect not only the curing kinetics but also the physical properties of the resulting material. In this study, various contents of polydimethylsiloxane containing an isocyanate group-modified epoxy resin with a constant amount of the curing agent were used to simplify the system for the kinetic study.

Typical DSC analyses for the novolac type epoxy resin modified with 0, 5, 10, 15, 20, and 25 phr polydimethylsiloxane containing an isocyanate group, and cured by MDA with a dynamic mode of the heating rate of 10°C/min, are shown in Figure 3. A single exothermic peak was observed for the self-polymerization process of the epoxy resin. The area under the DSC curve represents the magnitude of exothermic heat. The curing characteristics of the novolac-type epoxy resin modified with 0, 5, 10, 15, 20, and 25 phr polydimethvlsiloxane containing an isocyanate group and cured by MDA are summarized in Table I. A higher peak of the curing temperature and a lower curing enthalpy of modified epoxy than those of the unmodified epoxy were observed. This may be attributed to the necessity of a higher temperature being required for the larger molecules with attached siloxane segments to align the oxirane groups for self-polymerization and to lower the probability of etherification.<sup>11,12</sup> The curing reaction rate of epoxy with the curing agent is directly related to the probability of mutual diffusion of the reaction molecules. This probability generally decreases as the viscosity of the reaction mixture and the epoxy equivalent weight value of the epoxy increase. In other words, the probability would decrease when a foreign, relatively inactive, substance is present in the reacting mixture.

The data of kinetic parameters by DSC scans of isothermal curing reactions for the novolac-type epoxy resin modified with 0, 5, 10, 15, 20, and 25 phr polydimethylsiloxane containing an isocyanate group and cured by MDA are calculated from

 Table II
 Kinetic Parameters of the Novolac-Type Epoxy Resin/MDA/Polysiloxane Blends Obtained

 by Using the Ozawa and Kissinger's Equations

Novolac-Type Epoxy Resin/ MDA/Polysiloxane	m	n	Log A (1/min)	$E_a$ (KJ/mol)
$\begin{array}{c} 100/24.75/00\\ 100/24.75/05\\ 100/24.75/10\\ 100/24.75/15\\ 100/24.75/20\\ 100/24.75/25\\ \end{array}$	$egin{array}{c} 0.252 \pm 0.032 \ 0.292 \pm 0.036 \ 0.278 \pm 0.028 \ 0.273 \pm 0.027 \ 0.269 \pm 0.038 \ 0.285 \pm 0.029 \end{array}$	$\begin{array}{c} 1.48 \pm 0.069 \\ 1.68 \pm 0.084 \\ 1.55 \pm 0.062 \\ 1.57 \pm 0.062 \\ 1.45 \pm 0.078 \\ 1.59 \pm 0.064 \end{array}$	$5.43 \pm 0.071 \ 5.08 \pm 0.075 \ 5.35 \pm 0.062 \ 5.52 \pm 0.063 \ 5.28 \pm 0.082 \ 5.34 \pm 0.064$	$\begin{array}{c} 47.8 \pm 0.50 \\ 44.5 \pm 0.53 \\ 47.0 \pm 0.44 \\ 48.4 \pm 0.45 \\ 46.5 \pm 0.59 \\ 46.9 \pm 0.46 \end{array}$



**Figure 4** The preexponential factor (*A*) of polysiloxane-modified epoxy resins obtained by using the Ozawa and Kissinger's equations.

the Ozawa and Kissinger equations,<sup>10</sup> as shown in Table II. From Table II, the order of reaction mis lower than 1 and n is higher than 1 for modified or unmodified epoxy resin; hence, the curing reaction of epoxy may be a self-polymerization process. Furthermore, the magnitudes of  $E_a$  and logA exhibit the same tendency at various polysiloxane contents.

Plots of preexponential factor (logA) vs. polysiloxane contents for the novolac-type epoxy resins modified with 0, 5, 10, 15, 20, and 25 phr polydimethylsiloxane containing an isocyanate group and cured by MDA are shown in Figure 4. The data of  $E_a$  and logA exhibit a lower value for epoxy resin modified with polysiloxane than those of unmodified epoxy resin. The epoxy resin modified with 5 phr polysiloxane shows the lowest values ( $E_a = 44.5 \pm 0.53$  KJ/mol, logA = 5.08  $\pm$  0.075 L/min), while epoxy resin modified with 15 phr polysiloxane shows the highest values ( $E_a$ = 48.4  $\pm$  0.45 KJ/mol, logA = 5.52  $\pm$  0.063 L/min). The crosslinked epoxy modified with polysiloxane is plasticized by the polysiloxane oligomeric molecules. Once the cure reaction reaches the gel point, the reacting species in the neat epoxy would have less molecular mobility than those modified by polysiloxane, as in the epoxy modified with the polysiloxane system. For this reason, the retardation effect on the curing of epoxy modified with polysiloxane was relatively small. It is known that the polysiloxane particle in epoxy resin tends to phase separation, which will retard the reaction of epoxy molecules with curing agent molecules and increase the probability of mutual diffusion of the reaction molecules.

Consequently, when epoxy resin was modified with 15 phr polysiloxane, it showed the highest values of  $E_a$  and logA, which may be caused by polysiloxane molecules starting to form more phase-separated particulates.

# Thermal Properties of Polydimethylsiloxane Containing an Isocyanate Group-Modified Epoxy

The  $T_{g}$ s of the novolac-type epoxy resin modified with 0, 5, 10, 15, 20, and 25 phr polydimethylsiloxane containing an isocyanate group and cured by MDA, are shown in Table III. One can see that the  $T_{g}$ s of the cured modified epoxy resin are higher than those of the cured unmodified epoxy resin from Table III, except for that of the epoxy resin modified with 5 phr polysiloxane. It is proposed that the polydimethylsiloxane containing an isocyanate group and the epoxy/MDA curing system reacted and resulted in the reduction of  $T_{a}$ of the epoxy-rich phase. This result reveals that most of the polydimethylsiloxane containing an isocyanate group may be forming the oxazolidone structure via epoxy ring opening with the isocyanate groups of urethane at a higher isocvanate group content in polydimethylsiloxane. In addition, raising the polydimethylsiloxane containing isocyanate group content encourages immiscibility between the modifier and matrix. As a result, the flexural modulus does decrease with increasing the modifier, and one may observe the nature of the phase separation in a siloxane-modified epoxy system.<sup>13</sup>

TGA is usually utilized to study the thermal stability of polymeric materials. The values of



**Figure 5** TGA curves of ESCN-195XL/MDA/polysiloxane (a) 100/24.75/0, (b) 100/24.75/5, (c) 100/24.75/10, (d) 100/24.75/15, (e) 100/24.75/20, (f) 100/24.75/25 (phr) blend at a heating rate of 20°C/min in air.

	Polysiloxane (phr)					
	0	5	10	15	20	25
$T_g$ (°C) Flexural strength (Mpa)	$215.60 \\ 52.23$	$\begin{array}{c} 214.34\\ 49.71 \end{array}$	$216.69 \\ 45.29$	$\begin{array}{c} 218.24\\ 48.36 \end{array}$	$217.34 \\ 32.23$	$\begin{array}{c} 217.12\\ 26.52 \end{array}$
Flexural modulus (Mpa) Impact strength, Izod (ft-lb/in) Misture absorption (%) <sup>a</sup>	$2459.6 \\ 0.5257 \\ 1.56$	$2331.8 \\ 0.5631 \\ 1.37$	$2240.0 \\ 0.6075 \\ 1.58$	$2043.5 \\ 0.5849 \\ 1.45$	$1551.8 \\ 0.5386 \\ 1.72$	$1402.3 \\ 0.5378 \\ 1.73$

Table III Characterization and Properties of Polysiloxane-Modified Epoxy Resins Cured with MDA

<sup>a</sup> Moisture absorption (%) =  $[(W - W_0) * 100\%]/W_0$ .

W: weight of sample after standing at 100°C water for 72 h.

 $W_0$ : weight of sample after dried at 100°C for 3 h.

weight retention of the epoxy-rich phase of the novolac-type epoxy resin modified with 0, 5, 10, 15, 20, and 25 phr polydimethylsiloxane containing an isocyanate group and cured by MDA, are shown in Figure 5 and Table IV. From Figure 5, the weight retention of polydimethylsiloxane containing AN isocyanate group-modified epoxy resin was found to be similar to that of unmodified epoxy resin. For epoxy resins in air at 800°C, the small effect of polydimethylsiloxane containing an isocyanate group on the weight retention may be due to the increase in the amount of carbonized residue of epoxy resins modified with polydimethylsiloxane containing isocyanate groups.

Table V shows the internal stress of the polysiloxane-modified epoxy resin cured with MDA from the flexural modulus by the thermal expansion coefficient. The thermal expansion coefficient in the glassy state below the  $T_g$  was taken from 75 to 125 °C, and that above the  $T_g$  was taken from 225 to 275 °C. Increasing the isocyanate group content in the polydimethylsiloxane may cause an increase of the thermal expansion coefficient. The maximum thermal expansion coefficient occurs for the novolac-type epoxy resin modified with 5 phr polydimethylsiloxane containing an isocyanate group and cured by MDA.

# Physical Properties of Polydimethylsiloxane Containing an Isocyanate Group-Modified Epoxy

Figure 6 shows the flexural properties of the polysiloxane-modified epoxy resin cured with MDA. The flexural strength and modulus of the cured epoxy resin was considerably reduced for the epoxy resin modified with polydimethylsiloxane containing an isocyanate group. It is suspected that increasing the polysiloxane content results in incomplete mixing caused by gross incompatibility between the polydimethylsiloxane with the isocyanate group and the novolac-type epoxy resin prior to cure.

The internal stress of the cured epoxy resin was calculated from the flexural modulus by the thermal expansion coefficient. Table V shows that the internal stresses of the cured modified epoxy resins are higher than that of the cured unmodified epoxy resin, except for the epoxy resin modified with 5 phr polysiloxane. For polydimethylsiloxane containing an isocyanate group content above 5 phr, the inter-

Table IV Weight Retention of Polysiloxane-Modified Epoxy Resins Cured with MDA from TGA Measurements at a Heating Rate of 20°C/min in Air

Weight Retention (%)	Polysiloxane (phr)						
	0	5	10	15	20	25	
$387^{\circ}C^{a}$	90.78	89.97	88.80	90.73	89.25	89.14	
569°С <sup>ь</sup> 800°С	$33.76 \\ 1.553$	$28.48 \\ 3.148$	$33.10 \\ 3.167$	$\begin{array}{c} 32.55\\ 3.449 \end{array}$	$\begin{array}{c} 32.94\\ 3.802 \end{array}$	$23.44 \\ 3.806$	

<sup>a</sup> The primary weight retention rate from TGA calculated on weight retain differential with temperature.

<sup>b</sup> The secondary weight retention rate from TGA calculated on weight retain differential with temperature.

		Polysiloxane (phr)						
	0	5	10	15	20	25		
$\overline{T < T_g}$								
Modulus								
(MPa)	2459.6	2331.8	2240.0	2043.5	1551.8	1402.3		
CTE $(\mu m/m^{\circ}C)$	217.1	316.3	212.9	228.9	257.2	265.5		
Stress (Pa/°C)	533.98	737.55	476.90	467.76	399.12	372.31		
$T > T_{a}$								
Modulus								
(MPa)	2459.6	2331.8	2240.0	2043.5	1551.8	1402.3		
$CTE (\mu m/m^{\circ}C)$	302.40	402.30	365.90	320.90	342.10	411.70		
Stress (Pa/°C)	743.78	938.08	819.62	655.76	530.87	577.33		

Table VInternal Stress of Polysiloxane-Modified Epoxy Resins Cured with MDA from the Multiplyof Flexural Modulus by the Thermal Expansion Coefficient

nal stress of the cured epoxy resin tends to decrease with increasing the isocyanate group content in the polydimethylsiloxane.

Figure 7 shows that the Izod impact strength of the cured modified epoxy resin is higher than that of the cured unmodified epoxy resin. The maximum Izod impact strength was the epoxy resin modified with 10 phr polysiloxane. The Izod impact strength of the epoxy resin modified with 10 phr polysiloxane was increased 15.6% compared to that of the unmodified epoxy resin.

Moisture absorbed can exert a serious debilitating effect in that it not only plasticizes the epoxy resin, causing a lowering of the  $T_g$  and affecting mechanical properties, but also leads to a deterioration in a high-temperature load-bearing capacity. The moisture absorption data are summarized in Figure 8. For the epoxy resin modified with 20 phr polysiloxane or less, its moisture absorption was lower than for unmodified epoxy resin.

# CONCLUSION

In this study, polydimethylsiloxane containing an isocyanate group from  $\alpha, \omega$ -diol polydimethylsiloxane, phenyl isocyanate, and *m*-xylylene diisocyanate were synthesized. Novolac-type epoxy resin modified with 15 phr polysiloxane showed the highest value of  $E_a$  and logA, which may be caused by polysiloxane molecules starting to form more phase-separated particulates. A process was developed to incorporate stable dispersed polydimethylsiloxane containing isocyanate group particles in an epoxy resin matrix that not only greatly reduces the stress of the cured epoxy resin



**Figure 6** Flexural properties of polysiloxane-modified epoxy resin cured with MDA.



**Figure 7** Impact strength of polysiloxane-modified epoxy resin cured with MDA.



**Figure 8** Moisture absorption of polysiloxane-modified epoxy resin cured with MDA.

but also increases the  $T_g$  of the cured epoxy resin. The most suitable content of polysiloxane-modified epoxy resin is 15 phr. Epoxy resin modified with 15 phr polysiloxane exhibits good thermal and mechanical properties and lower moisture absorption than the others.

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