

# Epoxidized Natural Rubber-Bonded Para Rubber Wood Particleboard

B. Thongnuanchan, K. Nokkaew, A. Kaesaman, C. Nakason

Department of Rubber Technology and Polymer Science, Faculty of Science and Technology, Prince of Songkla University, Pattani Campus, Thailand

Para rubber wood particleboard (PB) was prepared using NR based adhesives and hot pressing processes. Two types of NR based adhesives were used; epoxidized natural rubber (ENR) and unmodified NR. The ENR latex was prepared using *in situ* performic epoxidation. Molecular weight of the ENR molecules was then reduced by incorporating of a reducing agent; sodium nitrite solution. A chain scission reaction and epoxidation simultaneous occurred via a chain-scission parallel epoxidation mechanism. Sulphur and multifunctional amine (i.e., hexamethoxymethylmelamine) curing systems were used to cure the adhesives. It was found that the hexamethoxymethylmelamine gave the PB with higher tensile strength than that of the sulphur vulcanization system. Furthermore, the tensile strength increased with increasing concentration of citric acid in the compounding formulation. Adhesion of the ENR adhesive with Parawood sawdust was observed to improve by reducing molecular weight of the ENR molecules. That is, the highest tensile strength of the PB was observed for the ENR adhesive with the lowest  $M_n$  (i.e.,  $1.10 \times 10^5$ ). This may be attributed to the adhesive with lower molecular weight exhibited greater ability to wet or cover the wood particle surfaces. As a consequence, greater chemical interaction between the adhesive and the wood particles was observed. POLYM. ENG. SCI., 47:421–428, 2007. © 2007 Society of Plastics Engineers

## INTRODUCTION

With an increasing environmental concern and forest protection, a search is on for alternative sources of wood materials. Particleboard (PB) and medium-density fiberboard are practically inexpensive alternatives to solid wood. They have been currently become one of the main leading building materials. By definition a PB is a structural material made of wood fragments, such as chips or shavings that are mechanically pressed into sheet form and bonded together with resin

adhesives. The formaldehyde based resins have been generally used for manufacturing of the PB. These types of resins are considered as a probable human carcinogen because they emit formaldehyde gas during manufacturing process and service life. Inhalation of even small amounts of the gas causes increasing risks of contracting lung and nasal cancer [1]. Formaldehyde emissions in the environment and worker's exposure at a manufacturing site, are also major health and environmental safety concern.

Natural rubber have been widely used as adhesives for particularly non-polar adherends or those where a strong mechanical strength involved in the adhesion mechanism [2]. However, wood material contains highly polar ingredients of various chemical compositions, such as cellulose, hemicellulose, and lignin. Therefore, polar based adhesives are more suitable for wood or PB manufacturing. To improve adhesion of natural rubber adhesive on polar wood surfaces, epoxidized natural rubber (ENR) is possible to be used instead of un-modified natural rubber adhesives. Preparation of wood adhesive from ENR is one of the promising way to obtain high performance polymeric materials from a renewable resource. This type of adhesive is possible to be used instead of the formaldehyde based resins. The properties of ENR depend on degree of epoxidation or level of epoxide groups in the ENR molecules [3]. For instance, the glass transition temperature ( $T_g$ ) increased with an increasing level of epoxide groups. Also, the rubber becomes increasingly oil resistance and melt viscosity as increasing the epoxide content of the ENR [4–6]. Adhesion properties of ENR based adhesive have been improved by reducing its molecular weight [7]. ENR can be cured or crosslinked using a multifunctional amine compound, such as *p*-phenylenediamine by utilizing the epoxide groups as crosslinking sites [8]. The crosslinking reaction of ENR can also be performed via a sulphur curing system [3]. However, with the multifunctional amine system, good chemical bonds between the crosslinking agents and hydroxyl groups in both wood and ENR molecules are possible. Therefore, physical properties of the final product may be further improved.

In this work, rubber wood based PB was prepared using Para wood sawdust and ENR latex adhesive via a hot pressing process. A novel multifunctional amine curing system

Correspondence to: C. Nakason; e-mail: ncharoen@bunga.pn.psu.ac.th  
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(i.e., hexamethoxymethylmelamine) was used to cure the ENR adhesive. The sulphur curing system was also exploited to compare the results. Furthermore, influence of molecular weight of ENR molecules and level of epoxide groups in the molecules on the properties of the rubber wood based PB was investigated.

## EXPERIMENTAL

### *Materials*

High ammonia (HA) concentrated natural rubber latex used for a preparation of ENR, manufactured by Yala Latex Co. (Yala, Thailand). The non-ionic surfactant used to stabilize the latex during epoxidation was Teric N30 (alkylphenol ethoxylate) which was manufactured by Huntsman Corp. Australia Pty. (Ascot Vale Vic, Australia). The formic acid used as a reactant for the preparation of ENRs was manufactured by Fluka Chemie (Buchs, Switzerland). The hydrogen peroxide used as a coreactant for the preparation of the ENRs was manufactured by Riedel De Haën (Seelze, Germany). The oxidizing agents used to reduce the molecular weight of ENR molecules via a redox reaction were hydrogen peroxide and sodium nitrile, which were manufactured by Riedel De Haën (Seelze, Germany) and Merck (Darmstadt, Germany). Hexamethoxymethylmelamine used as a crosslinking, manufactured by Qindao Hualu Melamine Co. (Hong Kong, China). Citric acid used as a catalyst for multifunctional amine curing system, manufactured by Riedel De Haën (Seelze, Germany). The sulphur with a purity of 99.5% used as a vulcanizing agent was manufactured by Ajax Chemical Co. (Samutprakarn, Thailand). The zinc oxide with a purity of 99.7% used as an activator was manufactured by Global Chemicals Co. (Bangkok, Thailand). Zinc diethyldithiocarbamate (ZDEC) used as accelerator for sulphur vulcanization system was manufactured by Shanxian Chemical Co. (Shandong Province, China). The polyphenolic additive, Wingstay<sup>®</sup>L, used as an antioxidant was manufactured by Eliokem (Ohio, USA).

### *Preparation of ENR*

The ENR latices were prepared using high-ammonia concentrated latex with a dried rubber content (DRC) of ~60%. The latex was first diluted to have a DRC of ~40% and then stabilized against coagulation by incorporating of 10% solution of a non-ionic surfactant (i.e., Teric N30). Predetermined quantities of hydrogen peroxide and formic acid were then added to form in situ performic epoxidation. Details for the preparation process of the ENRs are described elsewhere [3]. The reaction temperature was kept at 50°C for 2 h. The latex was then cooled down to a room temperature. Various concentrations (i.e., 0.25, 0.50, 0.75, 1.0, and 3.0 phr) of reducing agent (NaNO<sub>2</sub>) were each introduced into the latex to reduce the molecular weight of ENR molecules. The solution was added slowly to avoid

an excessive development of heat and nitrogen oxide by-product. The reaction was then allowed to proceed at this temperature for a pre-determined time. Samples were drawn out at various time intervals to analysis of epoxide content using FT-IR technique [3]. A number-average molecular weight (i.e.,  $\bar{M}_n$ ) of the ENR molecules was also determined using Gel Permeation Chromatograph (GPC), model Waters 1525 (Waters Corporation, USA).

### *Preparation of Sawdust*

Para wood sawdust was obtained from local Para wood manufactures in Pattani Province, Thailand. Sawdust was first sieved through a mechanical screen device with a capacity of 20-mesh. Therefore, Para wood sawdust particles with the sizes of 20-mesh and smaller were used to prepare the PB. It was later dried in a hot-air oven at 135°C to reach 6% equilibrium moisture content and used without further treatment.

### *Preparation of Latex Adhesives*

ENR and NR latices based adhesives were prepared using compounding formulations, as shown in Table 1. Two types of vulcanization systems were exploited to prepare the adhesives. These include a multifunctional amine curing system using hexamethoxymethylmelamine and sulphur curing system. Influence of sulphur-to-accelerator (ZDEC) ratios on properties of PB was studied using various ratios at 0.3, 1.0, 3.3, 7.5, and 12.0, respectively. Furthermore, various ratios of hexamethoxymethylmelamine to citric acid were also each studied at 0.5, 1.0, 2.0, 5.0, and 10.0, respectively. Wood rosin emulsion was first prepared by dissolving in toluene. Subsequently, ammonium oleated solution was incorporated and thoroughly stirred for 30 min. The wood rosin emulsion was later incorporated into the latices. The curing agents and other chemical ingredients were added according to the details in Table 1. The mixture was stirred for 30 min and kept a room temperature for about 24 h.

### *Preparation of Particleboard*

Dried Para wood sawdust was mixed with the latex adhesive at a weight ratio of sawdust to adhesive = 60:40. The mixtures were then dried in a hot air oven for 48 h at 50°C and later cut into small pieces using plastic grinder machine. The PB was later prepared by compression molded of the dried mixture of sawdust and adhesive at 150°C and a maximum pressure of 11 MPa for 10 min. It was then further vulcanized in a hot air oven at 170°C until reaching a maximum tensile strength. Mechanical properties of the PB were later determined according to the ASTM D 1037. These include the determination of the tensile strength parallel to surface and modulus of rupture. The tensile strength specimens were cut into a dog bone shape of 51 × 254 mm<sup>2</sup> with a gauge length of 38 mm wide and 51 mm long. Dimension of the modulus of rupture specimens

TABLE 1. Formulation of NR and ENR latex adhesives.

Ingredients	Formulation (phr)					
	A	B	C	D	E	F
NR	100	—	—	100	—	—
ENR-20	—	100	—	—	100	—
ENR-30	—	—	100	—	—	100
50% Sulphur	Variable <sup>a</sup>	Variable <sup>a</sup>	Variable <sup>a</sup>	—	—	—
50% ZDEC	Variable <sup>a</sup>	Variable <sup>a</sup>	Variable <sup>a</sup>	—	—	—
50% Hexamethoxymethylmelamine	—	—	—	5	5	5
Citric acid	—	—	—	Variable <sup>b</sup>	Variable <sup>b</sup>	Variable <sup>b</sup>
50% Wood rosin	15	15	15	15	15	15
50% ZnO	5	5	5	—	—	—
50% Wing stay L	1	1	1	1	1	1

<sup>a</sup> Sulphur-to-ZDEC ratios at 0.75/2.5, 1.5/1.5, 2.5/0.75, 15/2.0, and 30/2.5, i.e., at 0.3, 1.0, 3.3, 7.5, and 12.0, respectively.

<sup>b</sup> Hexamethoxymethylmelamine-to-citric acid at 5/10, 5/5, 5/2.5, 5/1, and 5/0.5, i.e., at 0.5, 1.0, 2.0, 5.0 and 10.0, respectively.

were  $75 \times 204 \text{ mm}^2$ . The mechanical test was performed at  $(27 \pm 2)^\circ\text{C}$  at a crosshead of 1 mm/min using the Hounsfield Tensometer model H 10 KS (Hounsfield Test Equipment Co. UK). Moisture resistance was also investigated by measuring water absorption and thickness swelling according to ISO 2380 using a PB specimen of  $300 \times 300 \text{ mm}^2$ . The tests specimens were conditioned at  $(27 \pm 2)^\circ\text{C}$  and  $\sim 65\%$  relative humidity for 48 h. The specimens were later submerged horizontally in distilled water at a temperature of  $(27 \pm 2)^\circ\text{C}$ . The water absorption capacity and thickness swelling were then determined. For both moisture resistance and mechanical tests, five specimens of PB were exploited.

## RESULTS AND DISCUSSION

### Characterization of ENR

Figure 1 shows infrared spectra of ENRs prepared using a typical epoxidation and chain-scission parallel epoxidation reactions. It can be seen that a FTIR spectrum of a typical ENR product based on a normal epoxidation at  $50^\circ\text{C}$  differed from the one obtained from a chain-scission parallel

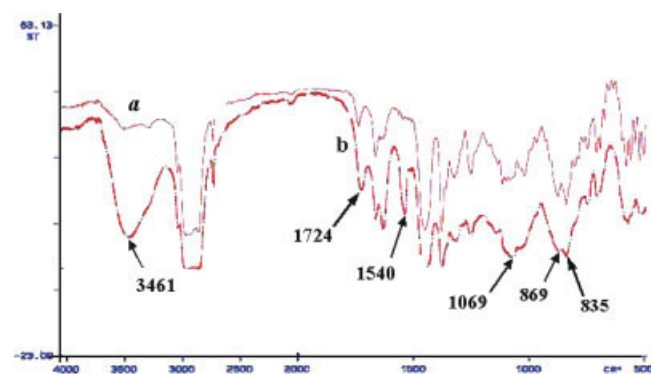


FIG. 1. Infrared spectra of ENRs compared between a typical epoxidation and chain-scission parallel epoxidation; (a) ENR-30 prepared by in situ epoxidation, (b) ENR-30 prepared by chain-scission parallel epoxidation. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com)]

mechanism at a room temperature (i.e., Fig. 1b). That is, the presence of new characteristic weak to medium bands for unsaturated aliphatic nitro group at  $1540 \text{ cm}^{-1}$  region [4]. Beside the main characteristic bands of epoxide ring at  $870$  and  $1250 \text{ cm}^{-1}$ , the other distinct bands of ring-opened structures were also observed in the ENR prepared by a chain-scission parallel mechanism. The majority of ring-opening products were cyclic ether, carbonyl groups, and molecules containing hydroxyl groups. The most outstanding among them was the OH stretching vibration at the wave numbers of  $3200\text{--}3500 \text{ cm}^{-1}$  region. This can be ascribed as the occurrence of ring-opened secondary products because of high acidic conditions [9, 10]. In addition, the chain-scission together with simultaneous epoxidation led to decrease of the molecular weight with higher OH content product. Similar results were also observed elsewhere [4].

### Effect of a Reducing Agent

Figure 2 shows effect of concentration of a reducing agent (i.e.,  $\text{NaNO}_2$ ), on a level of epoxide groups in the ENR molecules. In the typical in situ epoxidation, the reaction proceeds

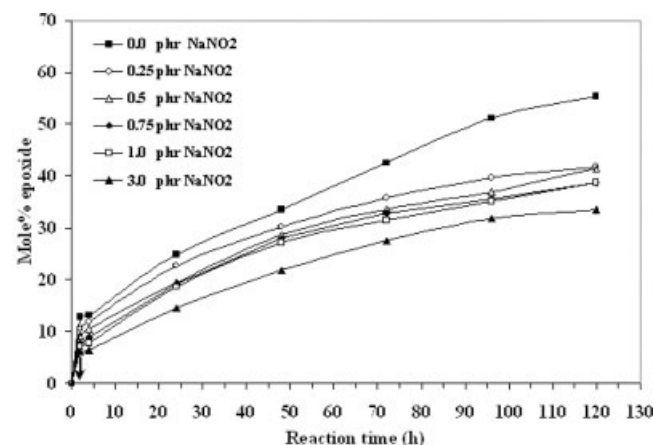


FIG. 2. Effect of concentration of a reducing agent ( $\text{NaNO}_2$ ) on a level of epoxide groups in ENR molecules.

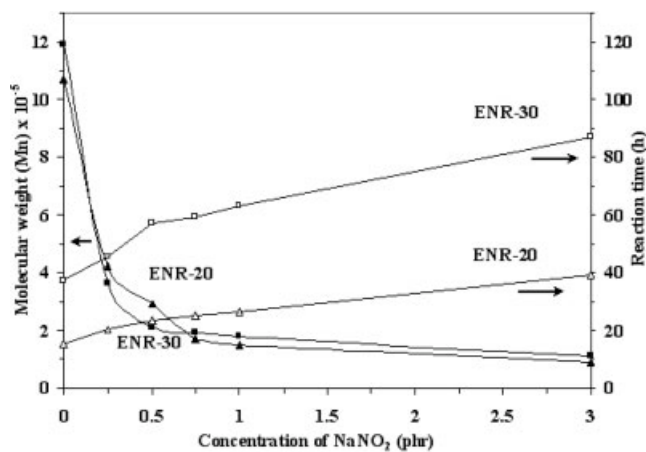


FIG. 3. Influence of concentration of a reducing agent ( $\text{NaNO}_2$ ) on a number-average molecular weight ( $\bar{M}_n$ ) and reaction time to reach levels of epoxide groups to 20 and 30% mole epoxide (i.e., ENR-20 and ENR-30, respectively).

rapidly in the first 2 h at 50°C. After the reducing agent was introduced, the latex was cooled down to a room temperature. The moment of the introduction of the reducing agent was indicated by a small arrow in Fig. 2. After an introducing of the  $\text{NaNO}_2$ , the epoxidation parallel with ENR chains scission took place at a room temperature. This caused an increasing trend of epoxide content and a decreasing trend of average molecular weight of the ENR molecules. In Fig. 2, it is also seen that the rate and extent of epoxidation were dependent upon the concentration of  $\text{NaNO}_2$  incorporated. The lower concentration of  $\text{NaNO}_2$  caused the greater reaction rate and the final epoxide content in the ENR molecules.

For further study, the ENR with 20 and 30 mol% epoxide (i.e., ENR-20 and ENR-30, respectively) were selected to prepare. Figure 3 shows the influence of concentrations of  $\text{NaNO}_2$  on the number average molecular weight ( $\bar{M}_n$ ) and reaction time used to prepare the ENR to reach levels of epoxide groups at 20 and 30 mole% epoxide. It is seen that an increasing in concentration of  $\text{NaNO}_2$  caused abruptly decreasing trend of number average molecular

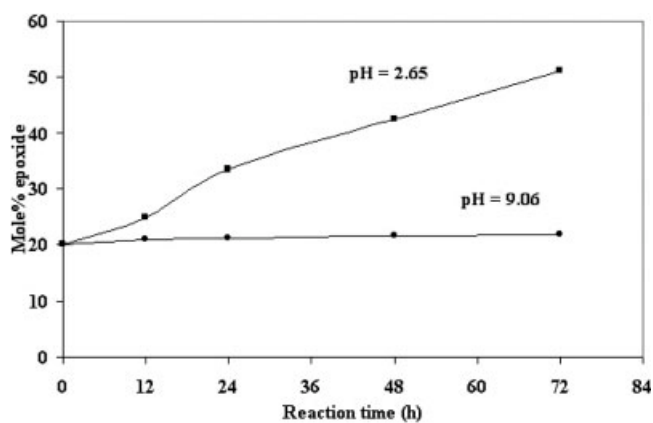


FIG. 4. Influence for pH of ENR latices on levels of epoxide groups.

weight of the ENR in particular in a range concentration of  $\text{NaNO}_2$  of 0–0.75 phr. Increasing concentration of  $\text{NaNO}_2$  also caused increasing reaction time required to prepare the ENRs with levels of epoxide groups of 20 and 30 mole% epoxide. This may be attributed to the existing of a redox reaction between  $\text{NaNO}_2$  and  $\text{H}_2\text{O}_2$  that later resulted in two competitive parallel reactions, viz. the chain-scissoring reaction and epoxidation [4]. Therefore, ENR with various levels of epoxide groups and a given level of an average molecular weight might be prepared by controlling the concentration of  $\text{NaNO}_2$  and reaction time.

Figure 4 shows influence of pH of the latex after incorporating of a reducing agent and reaching a desire epoxide level at a room temperature. pH of the latex was then adjusted to two extreme conditions; one is a basic (i.e., pH = 9.06) and the other is an acidic conditions at a pH of 2.65. It is seen that the reaction in a basic condition gave more or less ENR with a constant level of epoxide groups in the molecules. However, in an acidic condition, increasing trend of the epoxide groups was observed. That is, the epoxide groups of the ENR with 20% mole epoxide marginally increased to a level of epoxide groups of ~50% mole epoxide at a reaction time of 72 h. Therefore, the in situ epoxidation parallel with chain scission were highly favorable in acidic condition. However, the further epoxidation did not proceed in the basic condition. This may be attributed to the  $\text{H}_2\text{O}_2$  presence in the reaction was destroyed in an basic condition. As a consequence, in situ prepared performic acid could not be produced in the reaction pathway of the epoxidation.

#### Effect of a Sulphur Vulcanization System on Properties of Particleboard

The tensile strength and respective cure time of Para rubber wood PB prepared using a sulphur vulcanization system at a ratio of sulphur-to-accelerator = 1.0 using various types of rubber adhesives are shown in Fig. 5. It can

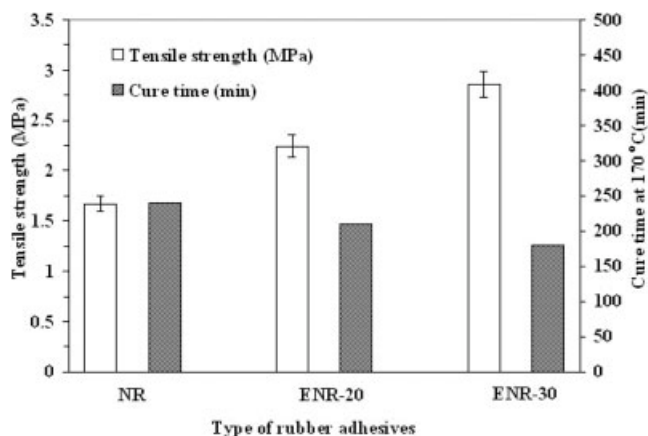


FIG. 5. Tensile strength and cure time of rubber wood particleboard using various types of rubber based adhesives and a sulphur vulcanization system at a sulphur-to-accelerator ratio of 1.0.

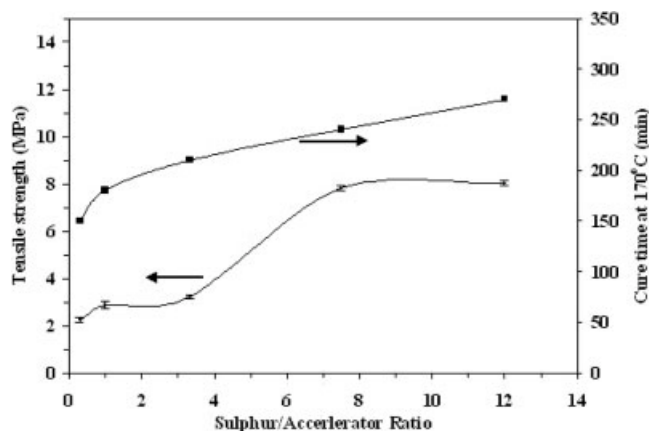


FIG. 6. Tensile strength and cure time of rubber wood particleboard using ENR-30 based adhesive and various sulphur-to-accelerator ratios.

be seen that the ENR-30 bonded-PB exhibited the highest tensile strength, whilst the un-modified NR gave the lowest value. The ENR-20 adhesive provided the PB with the intermediate value of tensile strength and cure time. It was also found that the cure time used to reach a maximum tensile strength of the ENR-30 based adhesive was shorter than those of the ENR-20 and NR, respectively. This may be attributed to the higher polarity of the ENR-30 than that of the ENR-20 and NR, respectively. The Para rubber wood sawdust is also a highly polar material that contains mainly cellulose, hemicellulose, and lignin molecules. Therefore, high polar adhesive material might develop stronger attraction between the sawdust particles [11]. As a result, a shorter time required to approach the maximum tensile strength of the adhesive with higher polarity, i.e., ENR-30 based adhesive. This is because higher degree of chemical interaction between the polar groups in the ENR molecules and wood particles.

Figure 6 shows influence of sulphur-to-accelerator ratios on tensile strength and cure time of the ENR-30 bonded-PB. It can be seen that the tensile strength and cure time increased with increasing a sulphur-to-accelerator ratios. One possible explanation is that the increasing degree of crosslink density of the system with an increasing ratio of sulphur-to-accelerator. However, to reach the maximum tensile strength, a longer time was needed for the system with higher ratio of sulphur-to-accelerator. In sulphur vulcanization system with high ratio of sulphur-to-accelerator, a formation of polysulphidic crosslinks are more favorable [12, 13]. During vulcanization process at 170°C, thermal decomposition of polysulphidic crosslinks might be also occurred. This led to a formation of an acid by-product containing sulphur atoms [14]. It was found that the acid by-product caused a ring-opening reaction of the ENR molecules.

The acid-catalyzed ring-opening reaction of epoxide groups in the ENR took place with a formation of ether crosslinks [15]. As a consequence, a further increasing in a cross-link density was observed [6, 16]. Therefore, the additional crosslinks via the acid-catalyzed ring-opening reaction accounted for the increase in crosslink density and, subsequently, increasing in tensile strength of PB. The

cure time of the system with lower ratio of sulphur-to-accelerator was shorter. This is probably due to the difference in a rate of formation of an active sulphurating agent [6]. That is, the amount of active sulphurating agent increased with an increasing in a ratio of sulphur-to-accelerator. Therefore, a shorter cure time was observed as decreasing trend of a ratio of sulphur-to-accelerator.

#### *Effect of a Multifunctional Amine Curing System on Properties of Particleboard*

Tensile strength and respective cure time of PBs prepared from different types of rubber adhesives using hexamethoxymethylmelamine crosslinking agent are shown in Fig. 7. The results indicate that the ENR-30 bonded-PB provided the highest tensile strength and the shortest cure time, whilst the NR exhibited the lowest tensile strength but the longest cure time. This may be attributed the occurrence of transesterification between methoxymethyl groups of the hexamethoxymethylmelamine and hydroxyl groups in both wood and ENR molecules in an acidic condition. Thus, the ether linkages were formed as a possible reaction scheme in Fig. 8. In the reaction six methoxymethyl groups in the hexamethoxymethylmelamine reacted with OH groups of wood and ENR molecules. This transesterification reaction did not occur in the system with non-polar NR system. Degree for a formation of ether linkage depend on epoxide levels of the ENR. Therefore, ENR-20 exhibited lower tensile strength and longer cure time than that of the ENR-30. Similar observations were made by Imam et al. (1999) who exploited the hexamethoxymethylmelamine as a crosslinking agent to provide a network of crosslink via the hydroxyl groups in wood, starch, and poly(vinyl alcohol) [17, 18]. In addition, the hexamethoxymethylmelamine has also been used to crosslink the epoxy resin via transesterification [19].

Influence of hexamethoxymethylmelamine-to-citric acid ratio on tensile strength of the PB is shown in Fig. 9. It can be seen that the tensile strength decreased progressively with increasing the melamine-to-citric acid ratio or

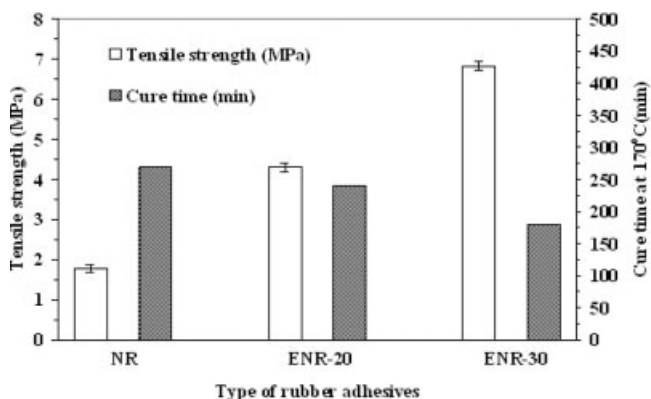


FIG. 7. Tensile strength and cure time of rubber wood particleboard using various types of rubber based adhesives and hexamethoxymethylmelamine curing agent at hexamethoxymethylmelamine-to-citric acid ratio of 2.0.

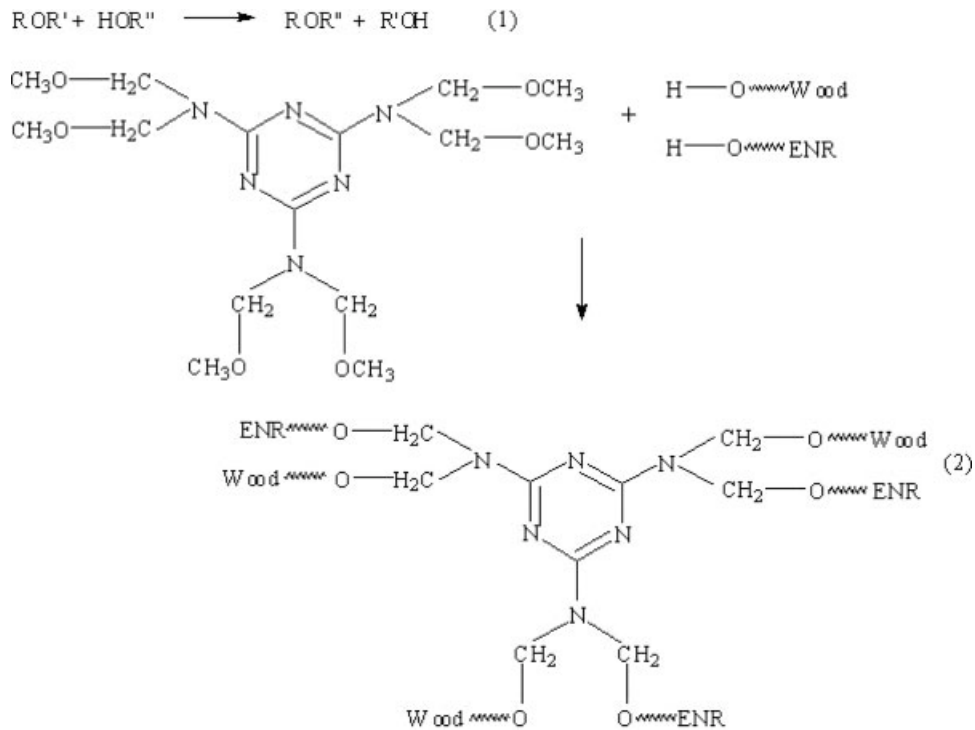


FIG. 8. Schematic representation of chemical crosslinking reaction of ENR and wood molecules via hexamethoxymethylmelamine molecules [17].

decreasing concentration of citric acid in the formulation. This is probably because of the decrease in crosslink density. Therefore, a suitable ratio of melamine-to-citric acid was found at the value less than one.

#### Effect of Molecular Weight of ENR on Tensile Properties of Particleboard

The PB using ENR-30 based adhesive and hexamethoxymethylmelamine vulcanization system exhibited the highest tensile strength, as results indicated in Fig. 5 compared with

Fig. 7. Therefore, the ENR-30 based adhesive with hexamethoxymethylmelamine vulcanization system were selected to perform a further study. Influence of an average molecular weight of ENR-30 on tensile strength of the PB is shown in Fig. 10. The result indicates that by decreasing the number average molecular weight ( $\bar{M}_n$ ) of the ENR-30, the tensile strength of PB was improved. This may be attributed to increasing probability of the adhesive to flow into the porous surface of the sawdust. The lower  $\bar{M}_n$  of ENR-30 caused lower flow resistance. Therefore, the surface of the sawdust was cover with the adhesive compound as indicated

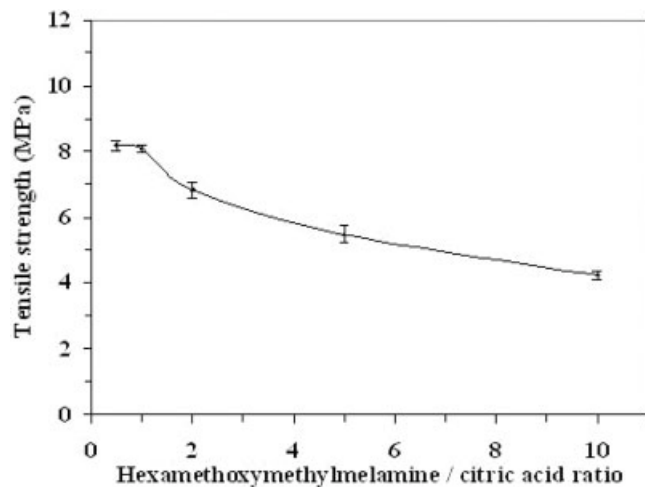


FIG. 9. Tensile strength of ENR-30 ( $\bar{M}_n = 8.9 \times 10^5$ ) bonded-wood particleboard using various hexamethoxymethylmelamine-to-citric acid ratios.

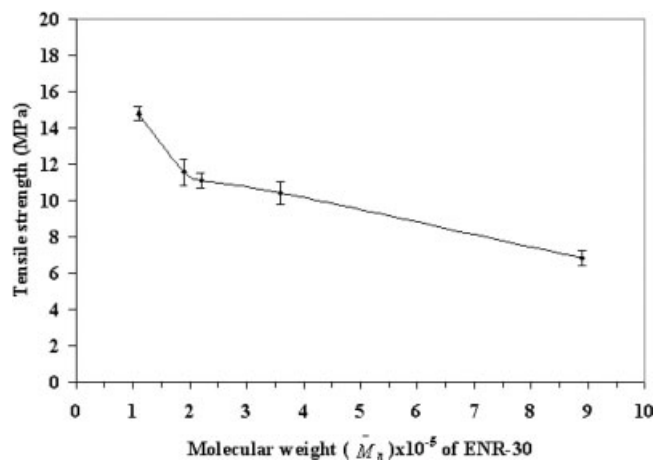


FIG. 10. Influence of  $\bar{M}_n$  of ENR-30 on tensile strength of rubber wood particleboard using hexamethoxymethylmelamine-to-citric acid ratio of 2.0.

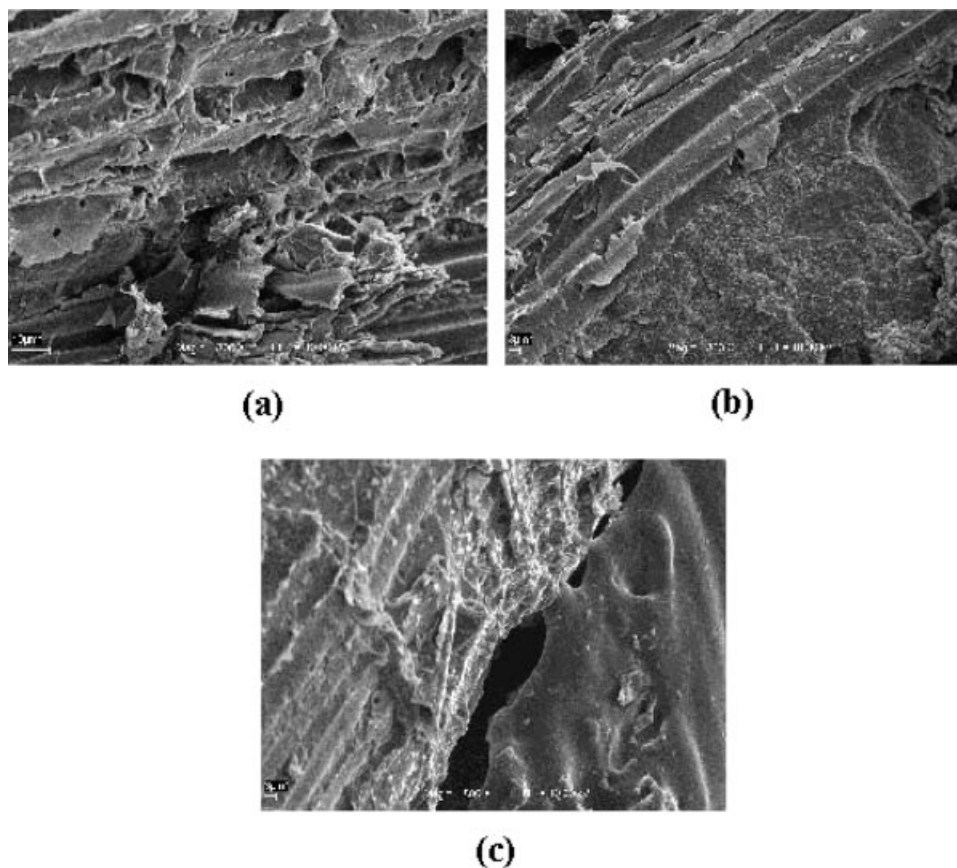


FIG. 11. SEM micrographs of sawdust surface (a); border between ENR-30 ( $\bar{M}_n = 1.10 \times 10^5$ ) based adhesive with sawdust surface (b); border between ENR-30 ( $\bar{M}_n = 8.90 \times 10^5$ ) based adhesive and sawdust surface.

in SEM micrographs in Fig. 11. It is seen that the surface the sawdust was fully covered by the ENR-30 based adhesive with a lower  $\bar{M}_n$  (i.e., in Fig. 11b). However, the ENR-30 based adhesive with a higher  $\bar{M}_n$  (i.e.,  $8.90 \times 10^5$ ) caused a cavitation between adhesive layer and sawdust surfaces (i.e., Fig. 11c). Therefore, decreasing  $\bar{M}_n$  of the ENR caused increasing chemical interaction between the ENR-30 and wood molecules via the multifunctional amine curing agent. Gazeley and Mente also observed that tack properties of ENR adhesive were improved by reducing of the  $\bar{M}_n$  to a range of  $1.50\text{--}4.0 \times 10^5$  [7].

Tensile strength, modulus of rupture and water absorption capacity of the PB prepared using the ENR-30 based adhesive with the  $\bar{M}_n$  of  $\sim 1.10 \times 10^5$  are listed in Table 2. The results were compared with the standard PB according to the American National Standard for basic hardboard

(ANSI A135.4). We found that ENR-30 based PB showed lower water absorption and thickness swelling. Also, it performed similar value of modulus of rupture and tensile strength. Therefore, it is concluded that this type of PB showed some properties equivalent and some properties better than that of the standard PB.

## CONCLUSIONS

Particle board was prepared using Para wood sawdust and natural rubber based adhesives. ENR was first prepared by in situ performic epoxidation followed by adding of reducing agent. The reaction was continued at room temperature. The epoxidation and chains-scission reaction of ENR were took place simultaneously by a chain-scission parallel epoxidation mechanism. It was also found that an average

TABLE 2. Physical properties of particleboard prepared using ENR-30 adhesive ( $\bar{M}_n = 1.1 \times 10^5$ ) using hexamethoxymelamine to citric acid ratio of 2.

Particle board	Nominal thickness (mm)	Water absorption (%)	Thickness swelling (%)	Modulus of rupture (MPa)	Tensile strength (MPa)
Standard <sup>a</sup>	6.4	25	20	31.0	15.2
ENR-30 particleboard <sup>b</sup>	6.39 (0.052)	5.92 (0.316)	8.02 (0.114)	29.64 (0.635)	14.82 (0.383)

<sup>a</sup> American National Standard for Basic Hardboard (ANSI A135.4-2004).

<sup>b</sup> Average values from 5 specimens with standard deviations in parentheses.

molecular weight and level of epoxide groups in the ENR molecules might be controlled by incorporating a suitable concentration of sodium nitrite solution and a given reaction time. The NR and ENR based adhesives were later prepared in a latex state using two types of vulcanization systems, sulphur, and multifunctional amine curing systems. We found that the ENR based adhesives provided PB with superior properties than that of the unmodified NR. Furthermore, ENR with a level of epoxide groups at 30 mole% (i.e., ENR-30) showed superior properties than that of the ENR-20. This may be attributed to the greater chemical interaction between -OH groups in the ENR molecules and -OH in the wood surface. It was also found that the PB prepared using a multifunctional amine curing exhibited superior tensile strength than that of the PB using sulphur vulcanization system. Also, the tensile strength increased with increasing concentration of citric acid in the adhesive formulation. Adhesion of the ENR adhesive was enhanced by reducing its molecular weight. That is, the highest tensile strength of rubber wood PB was observed for the ENR with the lowest  $\bar{M}_n$  at  $1.10 \times 10^5$ . The water absorption and thickness swelling behavior in water of the PB prepared using Parawood sawdust and ENR-30 adhesive were reached the requirement specified by ANSI A135.4. However, the modulus of rupture and tensile strength were slightly lower than that of the specification in the ANSI standard.

## ACKNOWLEDGMENTS

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## REFERENCES

1. D. Gary, *Choose Green Report, October 2001*, **1** (2001).
2. W.C. Wake, *Natural Rubber and Reclaimed Rubber Adhesives*, I. Skeist, editor, Van Nostrand Reinhold, New York, **242** (1977).
3. C. Nakason, A. Kaesaman, P. Klinpituksa, and Songklanakarin, *J. Sci. Technol.*, **23**, 415 (2001).
4. N.V. Bac, L. Terlemezyan, and M. Mihailov, *J. Appl. Polym. Sci.*, **50**, 845 (1993).
5. C.S.L. Baker and I.R. Gelling, *Developments in Rubber Technology-41987*, A. Whelan and K.S. Lee, editors, Elsevier, London, **87** (1987).
6. B.T. Poh and B.K. Tan, *J. Appl. Polym. Sci.*, **42**, 1407 (1991).
7. K.F. Gazeley, P.G. Mente, and G.B. Pat. **2**, 183, 663 A, (1987).
8. A.S. Hashim and S. Kohjiya, *J. Appl. Polym. Sci.*, **32**, 1149 (1994).
9. I.R. Gelling, *Rubber Chem. Technol.*, **87**, 58, (1984).
10. I.R. Gelling and M. Porter, *Chemical Modification of Natural Rubber*, A.D. Roberts, editor, Oxford University Press, New York, **16** (1988).
11. C.R. Scoville, *Characterizing the Durability of PF and PMDI Adhesive Wood Composites Through Fracture Testing*, Department of Wood Science and Forest, Virginia Polytechnic Institute, Virginia (2001).
12. B.T. Poh, C.P. Kwok, and G.H. Lim, *Eur. Polym. J.*, **31**, 223 (1995).
13. H. Ismail and B.T. Poh, *Eur. Polym. J.*, **36**, 2403 (2000).
14. C.S.L. Baker, I.R. Gelling, and B.S. Azemi, *J. Nat. Rubb. Res.*, **2**, 135 (1986).
15. I.R. Gelling and N.J. Morrison, *Rubb. Chem. Technol.*, **58**, 243 (1984).
16. Y. Heping, L. Sidong, and P. Zheng, *J. Ther. Anal. Calor.*, **58**, 295 (1999).
17. S.H. Imam, H. Sherald, L.M. Gordon, and L. Chen, *Polym. Degrad. Stab.*, **73**, 529 (2001).
18. S.H. Imam, L. Mao, L. Chen, and R.V. Greene, *Starch/ Stärke*, **51**, 225 (1999).
19. S.P. Leadley, J.F. Watts, C.J. Blomfield, and B. Tidsch, *The Interface Chemistry of Adhesion: Segregation of A Model Coil Coating Formulation*, Department of Materials Science and Engineering, University of Surrey, UK, **1** (1998).