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Received: 13 January 2022 | Revised: 27 January 2022 | Accepted: 28 January 2022

DOI: 10.1002/pc.26535

RESEARCH ARTICLE

spe INSPIRING PLASTICS PROFESSIONALS | Polymer COMPOSITES | WILEY

Performance of eco-friendly particleboard from agro-industrial residues bonded with formaldehyde-free natural rubber latex adhesive for interior applications

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Funding information

Deputy of Strengthening Research and Development, Ministry of Research and Technology, Grant/Award Number: 12/E1/KP.PTNBH/2021 (PDUPT scheme); Universitas Lampung, Grant/Award Number: 1626/UN26.21/PN/2021

Abstract

In this work, a novel way is proposed to produce an eco-friendly and formaldehyde-free particleboard (PB) panel from agro-industrial residues bonded with natural rubber latex (NRL)-based adhesive. Polyvinyl alcohol (PVOH) was added as an adhesion promoter and polymeric 4,4-methylene diphenyl diisocyanate (pMDI) was used as cross-linker. Different formulations of agro-industrial residues (cassava stem, sengon wood waste, and rice husk) and different contents of NRL-adhesive (10%, 15%, and 20%) were applied to prepare the PB panel. Several techniques were performed to characterize the properties of NRL-based adhesive and to evaluate the performance of PB panels from agro-industrial residues bonded with NRL-based adhesive. The blending of NRL and PVOH resulted in weak hydrogen bonds in the polymer blends. Incorporation of pMDI provided —NCO groups as the reactive site for cross-linking with NRL-PVOH via urethane linkages. The results showed that no remarkable differences in the physical properties of the PB panel, such as density, moisture content, water absorption, and thickness swelling, with different agro-industrial residues formulations and NRL-adhesive content. By contrast, greater NRL-adhesive content affected the mechanical properties of the PB panel. The best mechanical properties of the PB panel were obtained using a formulation of 40% of cassava stem, 30% of sengon wood waste, 30% of rice husk, and bonded with 20% of NRL-adhesive content, which resulted in 4.02 MPa of modulus of rupture (MOR), 441.00 MPa of modulus of elasticity (MOE), and 0.19 MPa of internal bonding (IB) strength. A combination of agro-industrial residues particles and NRL-based adhesive presented a high potential for application as an eco-friendly, formaldehyde-free, and non-structural PB such as interior applications.

KEYWORDS

cassava stem, natural rubber latex, particleboard, physical and mechanical properties, rice husk, sengon wood waste, wood based panels

1 | INTRODUCTION

Wood-based panels are produced about 360 million m^3 annually worldwide. Among them, almost 100 million m^3 of particleboard (PB) is manufactured at the end of 2021 to meet the consumption of PB globally.[1,2] Approximately 45%–47% of PB is manufactured in Asia, followed by Europe with around 27%–29%, and America produces about 25%–26% of PB.[1] In particular, Indonesia is contributing around 0.13% to the global PB's production. However, the increase in production of PB is not supported by the availability of raw materials from the forests owing to deforestation. Over the decade, the net loss in forests globally was around 4.7 million ha per year since 2010. It is estimated that 10.0 million ha of forest were cut down each year.[3] In particular, Indonesia experienced deforestation of around 0.63 million ha in 2016, and the rate of deforestation increased every year.[4]

To overcome that problem, the recycling of wood wastes have been done and resulted in PB panel for ceiling panels, flooring, wall, furniture, cabinets, desktops, and bulletin boards.[5,6] In addition, the utilization of agro-industrial residues particles in the production of PB has improved the growth in the global PB's production from 8.0% in 2016 to 16.0% in 2020 compared with other wood products.[1,7] These agro-industrial residues are rich in lignin and cellulose, also called lignocellulosic materials. Among them, oil palm fiber and rice husk have been the most exploited to produce PB.[8–10] Other agro-industrial wastes such as hemp fiber, sugar cane bagasse, coconut husk, coffee husk, sunflower husk, are also being researched and developed as a potential raw material in the manufacture of PB.[6,11–14]

Cassava (*Manihot esculenta*) is one of the widely agro-industrial plantations in South East Asia. It is harvested for its starchy tuber roots.[15] Indonesia is one of the main cassava-producing countries in the world, with a total production of 21.8 million tons of cassava in 2020 from a total area of nearly 700,000 ha.[16] This makes Indonesia the sixth-largest cassava-producing country in the world, with a productivity of around 23.2 tons/ha/year. The stems and leaves of cassava are usually wasted after the harvesting process. A published work had reported that cassava stem wastes can be used to produce PB bonded with urea-formaldehyde (UF) resins adhesive.[17] PB panel produced using UF resin–cassava stem ratio of 3:1 gave the best results of TS, WA, MOE, and MOR.

However, the presence of UF resin adhesive in the production of PB results in formaldehyde emission (FE) which is classified as hazardous and carcinogenic.[18,19] Strict regulations of FE from wood-based panels have contributed to the resolution of the indoor-air-quality problem.[19] The most acceptable and effective procedure in reducing FE is using chemical additives called formaldehyde scavengers, often referred to as formaldehyde catchers.[20,21] Formaldehyde scavenger is a chemical that can bond free formaldehyde and can be used post-treatment and incorporated into the resin during the mixing process.[22,23] The formaldehyde scavengers can be divided into the following three main types such as synthetic scavengers, bio-based scavengers, and nano-scavengers.[20–26]

In addition, many available bio-resources can be used as raw materials for wood adhesives such as lignin, tannin, and rubber.[27–30] Extensive study on the potential utilization of different types of lignin including lignosulfonates, kraft lignin, organosolv lignin, enzymatic hydrolysis lignin, and soda lignin for wood adhesives have reported comprehensively.[20–26] In addition to lignin, another renewable lignocellulosic feedstock that could be used for the formulation of wood adhesives are tannins.[31,32] Tannins can be classified into two groups, those characterized by having a molecular structure composed of flavonoids, known as condensed tannins, and those consisting of gallic and ellagic acids with a sugar core, known as hydrolysable tannins.[28,33] Among these types, condensed tannins are the most relevant with 90% of wood adhesives market.[34] By contrast to lignin and tannin, natural rubber latex (NRL) is barely used as wood adhesives, thus it is important to develop NRL-based adhesive particularly for PB.

Natural rubber latex is a bio-polymer consisting of repeated units of isoprene. It has been used as adhesives due to its environmental friendliness and sustainability.[35,36] NRL is produced from natural rubber harvested from rubber tree (*Hevea brasiliensis* [Willd. ex A.

Juss]).[35] Incorporating of tackifiers, fillers, viscosity modifiers, adhesion promoters, and cross-linkers can tailor the properties of NRL-based adhesives by.[35,37,38] The advantages of using NRL adhesive can be linked to sustainability, formaldehyde-free, and inexpensive, which was around 1.8–2.1 USD/kg in 2020.[39] Previous works have shown that NRL can be used to produce wood-based panels such as laminated wood,[29] plywood,[40] and PB.[36,41] In particular, the application of NRL adhesive in the production of PB requires low viscosity of adhesive to be sprayed onto the particle during the blending process.

Several modifications have been done to adjust the viscosity of NRL adhesive to a workable range for the production of PB, which is around 50–500 mPa s.[20,22] A simple addition of concentrated ammonia can reduce the viscosity and prevent coagulation of NRL.[41] Moreover, the incorporation of formic acid can further reduce the viscosity and provide an active group in the NRL.[36] The resulting PB panel has a proper bending strength, but not for IB strength. Therefore, polyvinyl alcohol (PVOH) and polymeric methane diphenyl diisocyanate (pMDI) have been added to enhance the adhesion performance of NRL adhesive.[29,40] Addition of PVOH into NRL resulted in greater —OH groups of NRL which could eventually enhance the adhesion of NRL. After addition of pMDI, the —NCO groups of pMDI have reacted with —OH groups of NRL-PVOH and resulted in urethane linkages (R_1—NH—[C═O]—R_2) for strong cohesion strength. The available and free —NCO groups will further react with the —OH of wood strong adhesion.[29,40]

In this work, a low viscosity NRL adhesive was formulated and the adhesion performance was tailored using PVOH as adhesion promoter and pMDI as cross-linker. Further, the NRL adhesive was used to produce PB panels made of agro-industrial wastes such as cassava stem, rice husk, and wood particles. Several analyses have been performed on the properties of NRL adhesive, the agro-industrial wastes, and the PB panel such as basic, thermal, chemical, and mechanical analyses.

2 | MATERIALS AND METHODS

2.1 | Materials

Three types of agro-industrial wastes used in this study were cassava stem (*M. esculenta*), rice husk (*Oryza sativa*), and sengon (*Paraserianthes falcataria*) wood wastes. Those agro-industrial wastes were obtained from a local farmer in Metro, Lampung, Indonesia. The NRL (60% of dry rubber content in ammonium, 128.0 mPa s of viscosity, and 10.2 of pH) was obtained from Garuda Indopacific Company (Bogor, West Java, Indonesia). PVOH (±22.000 g/mole, 387.5 mPa s of viscosity, and 5.9 of pH) was purchased from PT. Bratachem (Bogor, West Java, Indonesia). pMDI (±31% of NCO content, 503 mPa s of viscosity, and 9.8 of pH) was bought from Anugerah Raya Kencana Company (Tangerang, Banten, Indonesia).

2.2 | Preparation of agro-industrial wastes particles

The preparation of particles from agro-industrial residues is illustrated in Figure 1. Briefly, cassava stem, rice husk, and sengon wood wastes were chopped and hammermilled to produce the desirable particles. The obtained particles were screened at 40 mesh sizes. The particles were first air-dried for 4 days until reaching 25% of moisture content (MC), and then were dried at a temperature of 103 ± 2°C until reaching an average MC of 5% All particles were stored in a plastic bag to maintain the MC before the manufacture of PB.

2.3 | Formulation and characterization of NRL adhesive

The PVOH beads were dissolved in distilled water at 80°C to obtain a PVOH solution of 10% wt/vol for adhesion promoter, which resulted in lower viscosity of adhesive.[40] NRL then was mixed with PVOH homogeneously at a temperature of 27 ± 2°C under a stirring rate of 300 rpm with a ratio of 1:1 to promote the adhesion. A general cold-setting adhesive requires about 15% of pMDI. However, as the price of pMDI is expensive, this study tried to reduce the amount of pMDI as cross-linker. Therefore, low addition of pMDI in NRL-based adhesive such as 5% was selected.[40] The mixing process was done at a temperature of 27 ± 2°C under a stirring rate of 300 rpm.

Characterization of NRL-based adhesive was performed according to the published methods.[29,40] The non-volatile solids content of NRL-based adhesive was determined by drying the samples at 105°C ± 2°C for 3 h. The solids content of NRL-based adhesive was calculated by dividing the dry samples with wet samples. The viscosity of NRL-based adhesives was measured using a rotational rheometer (Rheolab QC, Anton Paar, Austria) at a constant speed of 100 rpm, at a temperature of 27 ± 2°C, and using a cylinder concentric No. 27. The gel time of NRL-based adhesive was determined using a gel time meter (Techne GT-6, Coleparmer, USA) at 27 ± 2°C. The pH of NRL-based adhesive was also measured using a digital pH meter (Orion Star A211, ThermoScientific, USA).

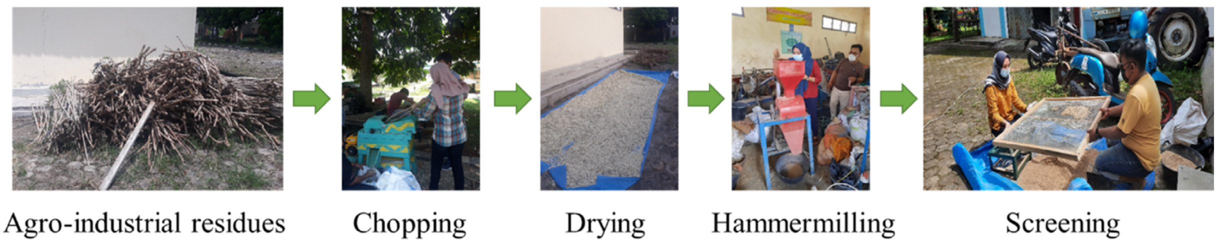

FIGURE 1 Flow chart of the preparation of agro-industrial residues particles

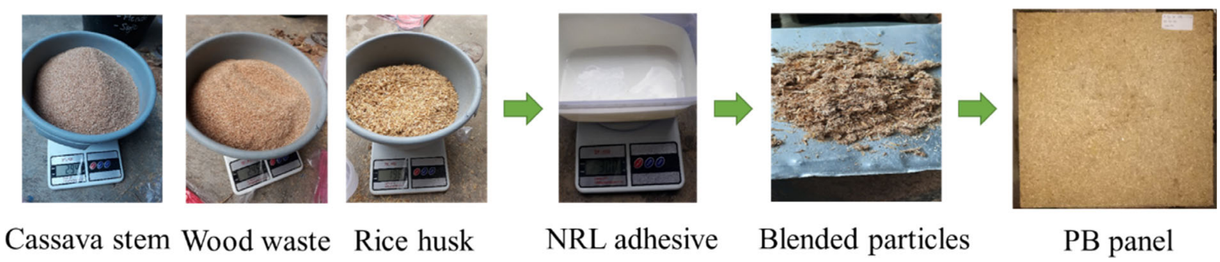

FIGURE 2 Preparation of particleboard (PB) from agro-industrial residues bonded with natural rubber latex (NRL)-based adhesive

TABLE 1 Formulation of a particleboard panel from agro-industrial residues bonded with different contents of natural rubber latex-based adhesive

	Adhesive content (%)								
	10			15			20		
Formulation	Cassava stem (g)	Wood waste (g)	Rice husk (g)	Cassava stem (g)	Wood waste (g)	Rice husk (g)	Cassava stem (g)	Wood waste (g)	Rice husk (g)
A	427.6	320.7	320.7	409.0	306.8	306.8	392.0	294.0	294.0
B	534.5	267.3	267.3	511.3	255.6	255.6	490.0	245.0	245.0
C	641.4	213.8	213.8	613.6	204.5	204.5	588.0	196.0	196.0
D	1069.1	–	–	1022.6	–	–	980.0	–	–

A Fourier transform infrared (FTIR) spectrometer (Spectrum Two, Perkin Elmer, United States) was used to investigate the change in functional groups of NRL-based adhesives in the range of wavenumber of 400–4000 cm^{-1} with a resolution of 2 cm^{-1}. Approximately 16 scans were averaged for the background and sample spectra, respectively. A min-max normalization was performed to normalize the spectrum of NRL-based adhesives using Spectrum Software (Ver. 10.5.3, Perkin Elmer Inc., USA).

Differential scanning calorimetry (DSC 4000, Perkin Elmer, United States) was used to monitore thermal properties of NRL-based adhesives at a scanning range of −50°C until 100°C, and a heating rate of 10°C/min. The nitrogen gas was injected with a flow rate of 20 ml/min was used as a purge gas. The peak temperature (T_p) was calculated using Pyris Software (Ver. 11.1.1.0492, Pyris, USA).

2.4 | Fabrication of the PB bonded with NRL adhesive

The preparation of a PB panel from agro-industrial residues bonded with different contents of NRL-based adhesive is displayed in Figure 2. The NRL adhesive was applied at different levels such as 10%, 15%, and 20%. The density of PB was targeted at 0.7 g/cm^3 with a panel size of 400 × 400 × 10 mm. The particles were formulated according to Table 1. Formula A was a mixture of cassava stem, wood waste, and rice husk at a ratio of 40:30:30

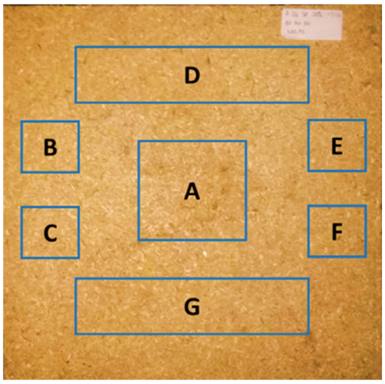

FIGURE 3 Illustration of a cutting pattern for each particleboard (PB) panel. A is density and moisture content (MC) samples, B is water absorption (WA) and thickness swelling (TS) samples, C is internal bonding (IB) strength sample, D is modulus of elasticity (MOE) and modulus of rupture (MOR) samples, E, F, G are the reserve samples

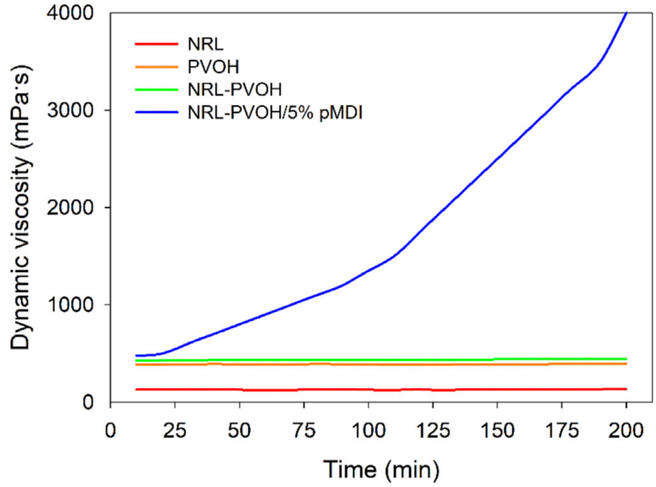

FIGURE 4 Dynamic viscosity of natural rubber latex (NRL)-based adhesive

(% wt/wt), respectively. Formula B was designed as 50:25:25, C was 60:20:20, and D was 100% cassava stem particles. The calculated amount of particles was blended with NRL-based adhesive and the mixture was arranged in a wooden box for the mat-forming process. The mat was hot-pressed at a temperature of 60°C for 30 min under 10 MPa of pressure, and cold-pressed at room temperature for 30 min under 10 MPa of pressure afterward using a compression molding machine (Shinto, Kyoto, Japan). The PB panels were then conditioned at room temperature (25–30°C) with a relative humidity between 65% for 2 weeks until reaching equilibrium MC. Three panels were prepared for each formulation, therefore a total of twelve panels were made in this study.

2.5 | Examination of physical and mechanical properties of PB

The properties of PB panel were evaluated according to the Japanese Industrial Standard (JIS) A 5908 (2003) standard for PB.[42] The parameters of physical properties were density, MC, thickness swelling (TS), and WA. The density and MC were measured with three replications for each formula. The WA and TS of the PB were determined by soaking the sample for 24 h in water and then measured after removing water on the surface. Meanwhile, the parameters of mechanical properties were MOE, MOE, and IB strength. All mechanical properties were measured using a universal testing machine (AGS-I, Shimadzu, Japan) with a load cell of 10 KN at a crosshead speed of 2 mm/min for IB strength test, and 10 mm/min for MOE and MOR test. The board cutting pattern is shown in Figure 3.

2.6 | Statistical analysis

The mean values of PB properties were compared using analysis of variance (ANOVA) and the Duncan multiple range test at $\alpha = 0.05$ was performed to determine optimum NRL-based adhesive content and raw material formulation on PB properties. Statistical analysis was done using SPSS 17 software (SPSS Inc., Chicago, United States).

3 | RESULTS AND DISCUSSION

3.1 | Properties of NRL adhesive

The basic properties of NRL-based adhesive were determined according to the published works.[29,40] The result showed that NRL-PVOH/5% pMDI adhesive has an average viscosity of 457.5 mPa s, a solids content of 31.5%, a gelation time of 112 min at 25°C, and a pH of 10.2. Dynamic viscosity of NRL-based adhesive is displayed in Figure 4. Neat NRL solution had an initial viscosity of around 128.0 mPa s, and the viscosity increased to 135 mPa s after 200 min. This indicated that there was no significant alteration of viscosity owing to molecular stability in neat NRL solution. A similar trend was found for PVOH, where the initial viscosity was around 387.5 mPa s and the viscosity increased to 395.4 mPa s after 200 min. Mixing NRL and PVOH slightly increased the viscosity of the mixture to 425.7 mPa s, and the viscosity increased to 444.5 mPa s after 200 min. However, after incorporation of 5% pMDI into the NRL/PVOH system altered the viscosity remarkably. The initial viscosity was 450.0 mPa s and the viscosity increased drastically to around 3985.0 mPa s. This result showed that incorporation of pMDI greatly increased the viscosity of NRL-based adhesive, meaning that the addition of pMDI resulted in a phase change of adhesive from liquid to viscous liquid and cross-linking of between NRL/PVOH and pMDI.[29,40] Addition of PVOH into NRL resulted in greater —OH groups of NRL which could eventually enhance the adhesion of NRL. After addition of pMDI, the —NCO groups of pMDI have reacted with —OH groups of NRL-PVOH and resulted in urethane linkages (R_1—NH—[C=O]—R_2) for strong cohesion strength.[29,40]

FTIR spectroscopy was conducted to investigate the alteration in the functional group of NRL-based adhesive (Figure 5A–D). The liquid NRL had a typical peak of N—H from the high concentration of ammonium at a wavenumber of 3265 cm^{-1} which acts as a stabilizer.[43] Moreover, the —CH_3 and —CH_2 of cis-polyisoprene were detected at a wavenumber of 2950 cm^{-1} and 2885 cm^{-1} which is the main constituent of NRL.[43,44] The additional peak of vibration of C=C and —CH_2— linkages of NRL were observed at 1685 cm^{-1} and 1050 cm^{-1}, respectively.[44] PVOH as an adhesion promoter has a specific peak of —OH at a wavenumber of 3300 cm^{-1}.[29,40] The addition of PVOH into NRL could result in greater —OH groups of NRL through hydrogen bonding, which could eventually enhance the adhesion of NRL.[29,40] PVOH is known as the most common proton donating polymer for hydrogen bonding.[45] Liquid pMDI has a strong peak of —NCO groups at a wavenumber of 2250 cm^{-1} which is reactive for cross-linking.[46] Incorporation of pMDI into the NRL-PVOH mixture altered the functional groups of NRL-based adhesives. The —NCO groups of pMDI could have reacted with —OH groups of NRL-PVOH and eventually resulted in urethane linkages (R_1—NH—[C=O]—R_2) at a wavenumber of 1645 cm^{-1} for strong cohesion strength as depicted in scheme reaction of NRL, PVA and pMDI (Figure 5C). The available —NCO groups of NRL-based adhesive at 2275 cm^{-1} is expected to have reaction with —OH groups of agro-industrial wastes in PB manufacturing.[29,40] This could result in strong adhesion between NRL-based adhesive and agro-industrial residues particles as well as a good cohesion strength in the adhesive.

Thermal properties of neat NRL, PVOH, and NRL-PVOH/5% pMDI adhesive were investigated using DSC (Figure 6). The differences in thermal properties of NRL, PVOH, and pMDI. Neat NRL and PVOH showed an endothermic peak of melting temperature (T_m) of water in NRL and PVOH at 0.0–0.1°C, which is following the

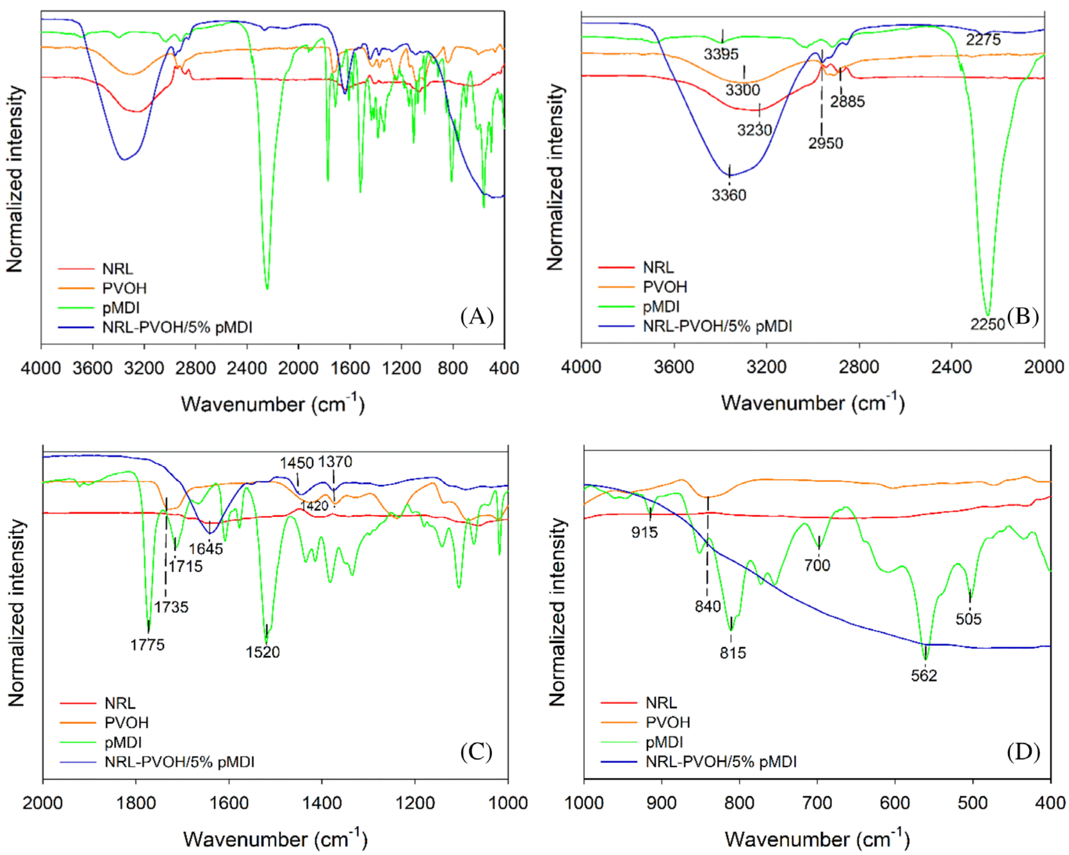

FIGURE 5 Functional group analysis of natural rubber latex (NRL)-based adhesive using Fourier transform infrared spectroscopy: (A) full spectra, (B) wavenumber 4000–2000 cm^{-1}, (C) wavenumber 2000–1000 cm^{-1}, (D) wavenumber 1000–400 cm^{-1}

published work that showed the T_m of NRL and PVOH was around 0.0–1.0°C.[47] The incorporation of NRL and PVOH resulted in higher T_m at 0.9°C and an endothermic peak temperature (T_p) at 55.0°C. This indicated that the NRL-PVOH absorbed energy from the environment for the hardening process at a temperature of 55.0°C. After mixing 5% pMDI to the NRL-PVOH blends uniformly, the value of T_m and T_p decreased to 0.0 and 22.8°C, respectively in NRL-PVOH/5%pMDI. This indicated that the NRL-based adhesive absorbed energy from the environment for the curing process. The result showed that the addition of pMDI in NRL-based adhesives lowered the T_m and T_p values of NRL-based adhesive.

3.2 | Physical properties of PB

Density is the ratio between the weight of the PB panel and its volume. Generally, the higher the average density of a wood-based panel, the greater its strength.[48] The average density of the PB panel was in the range of 0.65–0.74 g/cm^3 (Table 2). The result was in the range of the target density, which was 0.7 g/cm^3. The JIS A 5908:2003 standard requires 0.4–0.9 g/cm^3 of density for the PB panel.[42] The MC is the amount of water contained in the PB in a state of equilibrium with the surrounding environment. The MC of PB manufactured agro-industrial residues bonded with different contents of NRL-based adhesive is presented in Table 2. The results showed that the MC of the PB panel was between 4.51% and 5.52%. The MC of the resulting PB panel is lower than the JIS A 5908:2003 standard, which is 5.0%–13.0%.[42] Based on the results, increasing the NRL

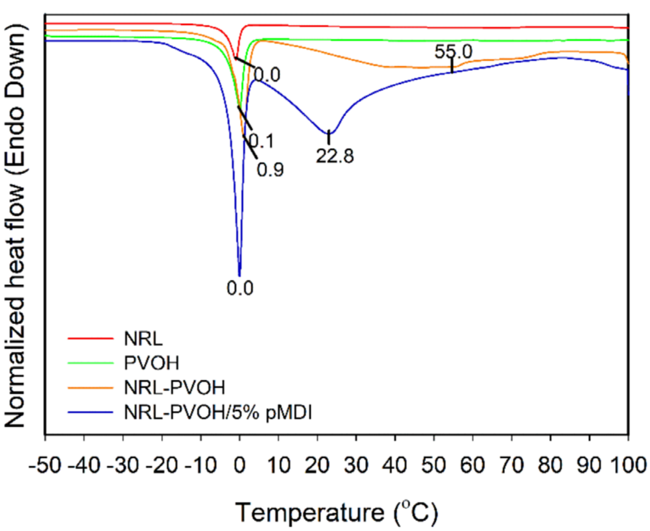

FIGURE 6 Differential scanning calorimetry thermograms of natural rubber latex (NRL)-based adhesive

adhesive content did not remarkably affect the density and MC values of the PB panel. In addition, different agro-industrial residues formulations also did not affect the density and MC values of the PB panel. Table 3 presents the statistical analysis of density and MC of PB. The results showed that raw material formulation and its interaction with adhesive content significantly influenced the density of PB, while adhesives content and its intercaction with raw material formulation significantly affected the MC of PB. According to the results, a combination of formulation A mixed with 20% NRL adhesive content could produce a good PB with 0.70 g/cm^3 denstity and 4.59% MC.

The WA is the ability of PB to absorb water during 24 h immersion.[42] The result showed that the WA of PB manufactured from agro-industrial wastes bonded with different contents of NRL-adhesive was in the range of 62.95%–72.99% (Figure 7A). The highest WA was obtained in type B of PB panel bonded with 10% NRL-adhesive, while the highest WA value was in type A of PB panel bonded with 20% of NRL-adhesive. The results indicated that the formulation of agro-industrial residues and NRL-adhesive content affected the WA of the resulting PB panel. This result is in accordance with the published work that used NRL to produce PB, reporting that higher NRL adhesive content in PB created protection against the water [41]. The increase in dimensional stability of the PB with higher NRL-based adhesive content is probably due to the presence of urethane linkages produced from the reaction between —NCO of pMDI and —OH of agro-industrial residues enhanced the adhesion strength of the panel, which were also reported in the application of NRL-adhesive for plywood and laminated wood.[29,40] There are no remarkable differences in the WA values with different agro-industrial residues formulations, showing that the formulations of agro-industrial residues did not affect the WA values of the PB panel. Based on the statistical analysis, raw materials formulation, adhesive content, and interaction between them were significantly influence the WA of PB (Table 4). According to the results, a combination of formulation A mixed with 20% NRL adhesive content could produce a PB with 62.95% WA, which was lower than other combination.

The TS is an increase in the dimensions of the thickness of the PB due to water filling the cavity in the board after being soaked for 24 h.[42] The TS values of PB panel from agro-industrial residues bonded with different contents of NRL-adhesive are presented in Figure 7B. The TS of the PB panel after 24 h immersion was in the range of 23.67%–29.72%. The lowest TS value was obtained in type A of PB panel bonded with 20% of NRL-adhesive, while the highest TS value was in type C of PB panel bonded

TABLE 2 Density (g/cm^3) and MC (%) of particleboard from agro-industrial residues bonded with different contents of natural rubber latex adhesive

	Adhesive content (%)					
	10		15		20	
Type of panel	Density	MC	Density	MC	Density	MC
A	0.72 ± 0.02	5.23 ± 0.74	0.70 ± 0.01	4.99 ± 0.62	0.70 ± 0.02	4.59 ± 0.57
B	0.69 ± 0.02	5.56 ± 0.73	0.72 ± 0.01	5.33 ± 0.26	0.69 ± 0.01	4.88 ± 0.21
C	0.69 ± 0.01	5.52 ± 0.56	0.71 ± 0.02	5.30 ± 0.72	0.70 ± 0.01	4.93 ± 0.25
D	0.73 ± 0.01	4.58 ± 1.13	0.74 ± 0.01	5.34 ± 0.59	0.65 ± 0.02	4.51 ± 1.20

Abbreviation: MC, moisture content.

TABLE 3 Statistical analysis of density and MC of PB made of agro-industrial residues bonded with different contents of natural rubber latex adhesive

Properties	Variable	Mean square	*F* value	*p* value
Density	Raw materials formulation	0.002	5.082	0.008*
	Adhesive content	0.001	1.026	0.375
	Interaction	0.006	13.555	0.001
MC	Raw materials formulation	0.371	0.849	0.482
	Adhesive content	3.489	7.979	0.002
	Interaction	0.207	0.473	0.821

Abbreviations: MC, moisture content; PB, particleboard.
*The variable with p value lower than $\alpha = 0.05$ is significantly effect the properties of PB.

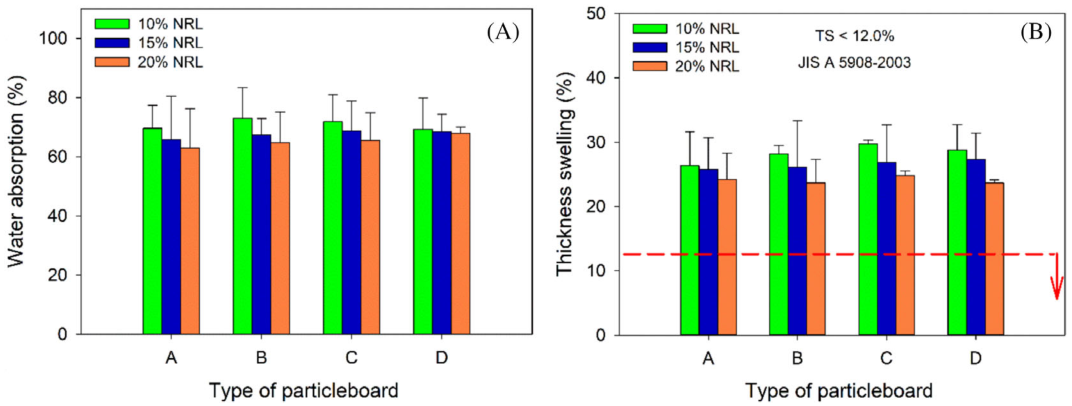

FIGURE 7 Physical properties of particleboard made of agro-industrial residues bonded with different contents of natural rubber latex (NRL) adhesive: (A) water absorption, (B) thickness swelling (TS)

TABLE 4 Statistical analysis of WA and TS of PB made of agro-industrial residues bonded with different contents of natural rubber latex adhesive

Properties	Variable	Mean square	*F* value	*p* value
WA	Raw materials formulation	2981.963	21.212	0.001*
	Adhesive content	1649.373	11.733	0.001
	Interaction	822.208	5.849	0.001
TS	Raw materials formulation	30.156	0.416	0.743
	Adhesive content	227.173	3.136	0.063
	Interaction	49.49	0.683	0.665

Abbreviations: PB, particleboard; TS, thickness swelling; WA, water absorption.
*The variable with p value lower than $\alpha = 0.05$ is significantly effect the properties of PB.

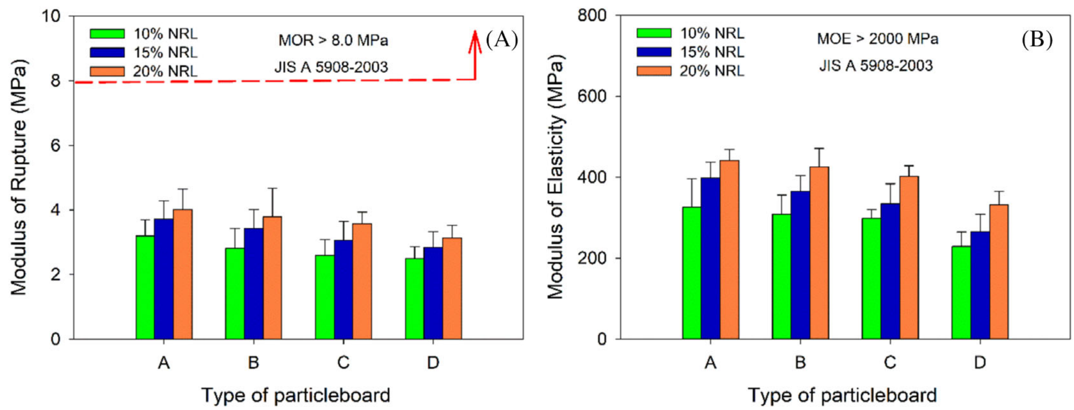

FIGURE 8 Mechanical properties of particleboard made of agricultural residues bonded with different contents of natural rubber latex (NRL) adhesive: (A) modulus of rupture (MOR), (B) modulus of elasticity (MOE)

TABLE 5 Statistical analysis of MOE and MOR of PB made of agro-industrial residues bonded with different contents of natural rubber latex adhesive

Properties	Variable	Mean square	*F* value	*p* value
MOR	Raw materials formulation	1.133	3.444	0.034*
	Adhesive content	2.186	6.643	0.006
	Interaction	0.025	0.075	0.998
MOE	Raw materials formulation	21,628.817	15.699	0.001
	Adhesive content	36,319.6	6.643	0.006
	Interaction	255.235	0.185	0.978

Abbreviations: MOE, modulus of elasticity; MOR, modulus of rupture; PB, particleboard.
*The variable with p value lower than $\alpha = 0.05$ is significantly effect the properties of PB.

with 10% of NRL-adhesive. This result is in agreement with the published work that used NRL to produce PB, reporting that increasing NRL adhesive content could reduce the TS value.[41]

This work revealed that the dimensional stability of the PB increased with higher adhesive content, indicating that the urethane linkages produced from the reaction between —NCO of isocyanate and —OH of agro-industrial residues enhanced the bonding strength of the PB. This result is supported with the published works, which were reported a similar trend in the application of NRL-adhesive for plywood and laminated wood.[29,40] However, the TS value of the resulting PB panel did not meet the JIS A 5908:2003 standard of a maximum of 12.0% TS.[42] This is probably due to the less amount of pMDI added in the NRL-based adhesive. A typical aqueous polymeric isocyanate (API) adhesive usually uses around 15% of pMDI as a cross-linker.[49] However, as the price of pMDI is expensive, this study tried to reduce the amount of pMDI as a cross-linker. Therefore, low addition of pMDI in NRL-based adhesive such as 5% was selected. There were no remarkable differences in the TS values with different agro-industrial residues formulations, showing that the formulations of agro-industrial

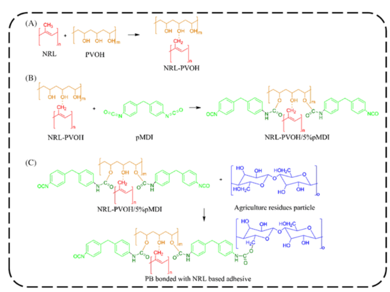

FIGURE 9 Possible scheme reaction of particleboard (PB) bonded with natural rubber latex (NRL)-based adhesive: (A) reaction of NRL and polyvinyl alcohol (PVOH), (B) reaction of NRL, PVOH and 5% polymeric methane diphenyl diisocyanate (pMDI), (C) reaction between agro-industrial residues particle with NRL-based adhesive

wastes did not affect the TS values of the PB panel. Based on the statistical analysis, raw materials formulation, adhesive content, and interaction between them were not significantly influence the TS of PB (Table 4). According to the results, a combination of formulation A mixed with 20% NRL adhesive content could produce a PB with 23.67% TS, which was lower than other combination.

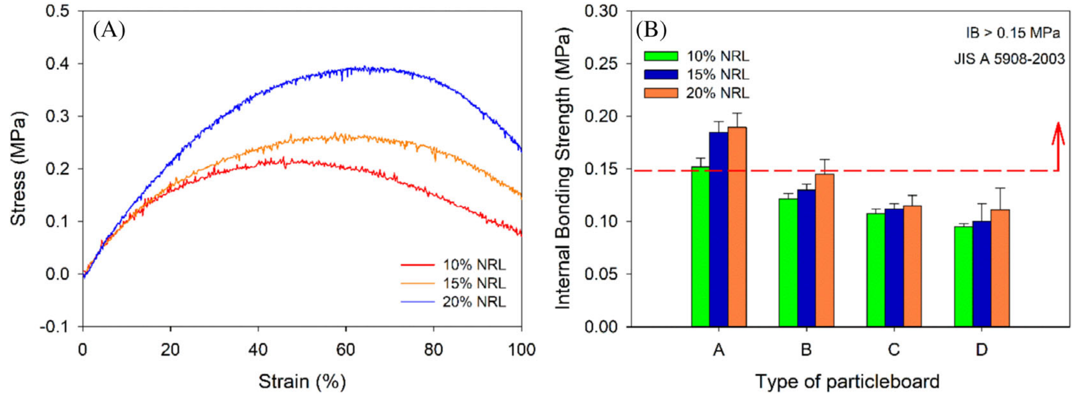

FIGURE 10 The internal bonding (IB) strength of the particleboard panel bonded with different contents of natural rubber latex (NRL) adhesive: (A) typical stress–strain curve, (B) IB strength

TABLE 6 Statistical analysis of IB strength of PB made of agro-industrial residues bonded with different contents of natural rubber latex adhesive

Properties	Variable	Mean square	*F* value	*p* value
IB	Raw materials formulation	0.010	44.998	0.001*
	Adhesive content	0.001	1.374	0.267
	Interaction	0.003	23.991	0.001

Abbreviations: IB, internal bonding; PB, particleboard.
*The variable with *p* value lower than $\alpha = 0.05$ is significantly effect the properties of PB.

This study showed that addition of 5% pMDI limited the enhancement of TS, therefore further study is needed to obtain a high performance NRL-based adhesive.

3.3 | Mechanical properties of PB

The MOR is determined from the maximum load times of the support distance divided by the cross-sectional area.[42] The MOR values of the PB panel are presented in Figure 8A. The MOR of the PB was in the range of 2.49–4.02 MPa. The highest MOR value was obtained in type A of PB panel bonded with 20% NRL-adhesive, while the lowest one was in type D of PB panel bonded with 10% NRL-adhesive content. This result is in-line with the published work that used NRL to produce PB panels.[41] The JIS A 5908: 2003 standard for type 8 of PB requires a minimum MOR value of 8.0 MPa,[42] therefore the MOR values in this study were below the standard. A typical API adhesive usually uses around 15% of pMDI as a cross-linker.[49] However, this study tried to reduce the amount of pMDI as a cross-linker to 5% owing to the price of pMDI is expensive. This work showed that there are no remarkable differences in the MOR values with different agro-industrial residues formulations, showing that the formulations of agro-industrial residues did not affect the MOR values of the PB panel. Based on the statistical analysis, adhesive content and its interaction with raw materials formulation were significantly influence the MOR of PB (Table 5). According to the results, a combination of formulation A mixed with 20% NRL adhesive content could produce a PB with 4.02 MPa MOR, which was higher than other combination. This study showed that addition of 5% pMDI limited the enhancement of MOR, therefore further study is needed to obtain a high performance NRL-based adhesive.

The MOE is a measure of the wood's resistance in maintaining deformation due to loads and in direct contact with wood. The higher the MOE value, the more elastic. The MOE values of the PB from agro-industrial residues bonded with different contents of NRL-adhesive are presented in Figure 8B. The results showed that the MOE value of the PB was between 229.20 and 441.00 MPa. This trend of result is in accordance with the published work that used NRL to produce PB panels.[41] The highest MOR value was obtained in type A of PB panel bonded with 20% NRL-adhesive, while the lowest one was in type D of PB panel bonded with 10% NRL-adhesive content. The main reason for this result is that latex presents elastic behavior, therefore greater NRL-content produced higher MOE value.[41] But, the MOE values of the PB panel did not meet the minimum JIS A 5908:2003 standard for type 8 of PB, which is a minimum of 2000 MPa.[42] A typical API adhesive usually uses around 15% of pMDI as a cross-linker.[49] However, this work reduced the amount of pMDI as a cross-linker to 5% owing to the price of pMDI being expensive.

There were no remarkable differences in the MOE values with different agro-industrial residues formulations, showing that the formulations of agro-industrial residues did not affect the MOE values of the PB panel. Based on the statistical analysis, adhesive content and raw materials formulation were significantly influence the MOE of PB, but interaction of them did not give significant effect to the MOE (Table 5). According to the results, a combination of formulation A mixed with 20% NRL adhesive content could produce a PB with 440.99 MPa MOE, which was higher than other combination.

The increase in mechanical properties of the PB with higher NRL-based adhesive content is also probably due to the cross-linking reaction between NRL-adhesive and agro-industrial residues, which were also reported in the application of NRL-adhesive for plywood and laminated wood.[29,40] As depicted in Figure 9A, the blending of NRL and PVOH resulted in weak hydrogen bonds in the polymer blends. Incorporation of pMDI provided —NCO groups as the reactive site for cross-linking with NRL-PVOH via urethane linkages (Figure 9B). The presence of free —NCO groups in NRL-PVOH/5% pMDI, as detected by FTIR spectroscopy (Figure 5), have further reacted with the —OH groups of agro-industrial wastes to form urethane linkages (R_1—NH—[C═O]—O—R_2) for enhancing the adhesion strength of the PB panel.

The IB strength is a measure of the strength of a material to resist rupturing in the direction perpendicular to the plane of the surface.[42] The IB strength values of the PB panel from agro-industrial residues bonded with different contents of NRL-adhesive are displayed in Figure 10. The typical stress–strain curve of the PB panel showed that increasing the NRL-adhesive content resulted in higher maximum stress. As the result, the IB strength value of the PB panel increased with higher NRL-adhesive content. The IB strength values of the PB from agro-industrial residues bonded with different contents of NRL-adhesive were between 0.10 and 0.19 MPa. This result is in agreement with the published work that used NRL to produce PB panels.[41] The highest IB strength value was obtained in type A of PB panel bonded with 20% NRL-adhesive, while the lowest IB strength was in type D of PB panel bonded with 10% NRL-adhesive content. Only type A of PB panel could meet the minimum standard of IB strength for PB type 8, which is 0.15 MPa.[42] As depicted in Figure 9, the presence of free —NCO groups in NRL-PVOH/5% pMDI have further reacted with the —OH groups of agro-industrial residues to form urethane linkages (R_1—NH—[C═O]—O—R_2) for enhancing the adhesion strength of the PB panel, which was also reported.[29,40,41] Based on the statistical analysis, raw materials formulation and its interaction with adhesive content were significantly influence the IB strength of PB, but adhesive content alone did not give significant effect to the IB strength (Table 6). According to the results, a combination of formulation A mixed with 20% NRL adhesive content could produce a PB with 0.19 MPa IB strength, which was higher than other combination.

4 | CONCLUSION

This work proposed a novel way to produce eco-friendly and formaldehyde-free PB from agro-industrial residues bonded with NRL-based adhesive. The NRL-PVOH/5% pMDI adhesive used in this study has an average viscosity of 457.5 mPa s, a solids content of 31.5%, a gelation time of 112 min at 25°C, and a pH of 10.2. The blending of NRL and PVOH resulted in weak hydrogen bonds in the polymer blends. Incorporation of pMDI provided –NCO groups as the reactive site for cross-linking with NRL-PVOH via urethane linkages. The results showed that no remarkable differences in the physical properties of the PB panel, such as density, MC, WA, and TS, with different agro-industrial residues formulations and NRL-adhesive content. By contrast, greater NRL-adhesive content affected the mechanical properties of the PB panel. The best mechanical properties of the PB panel were obtained at type A of PB bonded with 20% NRL-adhesive, which had 4.02 MPa of MOR, 441.00 MPa of MOE, and 0.19 MPa of IB strength. This work showed that a PB panel could be produced using a formulation of 40% of cassava stem, 30% of sengon wood waste, 30% of rice husk, and bonded with 20% of NRL-adhesive content. This study showed that addition of 5% pMDI limited the enhancement of PB properties, therefore further study is needed to obtain a high performance NRL-based adhesive. Based on the results, NRL-based adhesive presented a high potential for application as an eco-friendly, formaldehyde-free, and non-structural PB such as interior application.

ACKNOWLEDGMENTS

The authors sincerely acknowledge the "*Penelitian Pasca Sarjana*" with Research Grant No. 1626/UN26.21/PN/2021 from the University of Lampung for the financial support. This study was also funded the Japan-ASEAN Science and Technology Innovation Platform (JASTIP) Fiscal Year 2021–2022 with the title "Development of High Performance of Bio-Based Adhesives for Durable Wood-Based Composites". Part of this study was also supported by Deputy of Strengthening Research and Development, Ministry of Research and Technology Fisca Year 2021 with Research Grant No. 22/E1/KP.PTNBH/2021 (PDUPT scheme), March 8, 2021.

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How to cite this article: W. Hidayat, N. Aprilliana, S. Asmara, S. Bakri, S. Hidayati, I. S. Banuwa, M. A. R. Lubis, A. H. Iswanto, *Polym. Compos.* **2022**, 1. https://doi.org/10.1002/pc.26535

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F I G U R E 1 Flow chart of

www.aiditalia.it

F I G U R E 2 Preparation of

oss.ejgo.net

particleboard (PB) from agro-industrial residues bonded with natural rubber latex (...

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panel from agro-industrial residues bonded with

www.infona.pl

F I G U R E 3 Illustration of a

pure.ulster.ac.uk

water absorption (WA) and thickness swelling(TS) samples

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Statistical analysisThe mean values of

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ET AL

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Functional group analysis

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different contents of natural rubber

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The variable with p value lower than α = 0.05 is significantly

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Lee Seng Hua, Lum Wei Chen, Boon Jia Geng, Lubos Kristak et al. "Particleboard from Agricultural Biomass ...

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A) modulus of rupture

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MOE, modulus of elasticity; MOR, modulus of rupture

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The internal

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PropertiesVariableMean squareF valuep value

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