


RESEARCH ARTICLE

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

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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 m³ annually worldwide. Among them, almost 100 million m³ 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 ($\pm 31\%$ 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 $103 \pm 2^\circ\text{C}$ 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 $27 \pm 2^\circ\text{C}$ 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 $27 \pm 2^\circ\text{C}$ 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 $105^\circ\text{C} \pm 2^\circ\text{C}$ 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 $27 \pm 2^\circ\text{C}$, and using a cylinder concentric No. 27. The gel time of NRL-based adhesive was determined using a gel time meter (Techne GT-6, Colparmer, USA) at $27 \pm 2^\circ\text{C}$. The pH of NRL-based adhesive was also measured using a digital pH meter (Orion Star A211, ThermoScientific, USA).

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/cm³ (Table 2). The result was in the range of the target density, which was 0.7 g/cm³. The JIS A 5908:2003 standard requires 0.4–0.9 g/cm³ 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

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 interaction 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/cm³ density 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

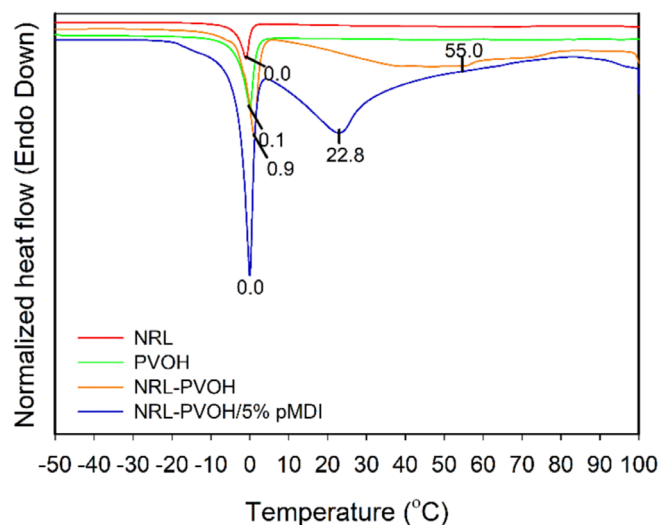


FIGURE 6 Differential scanning calorimetry thermograms of natural rubber latex (NRL)-based adhesive

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.

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