Vol. 13(2): 601-616, May 2025 DOI: https://doi.org/10.23960/jsl.v13i2.1080

P-ISSN: 2339-0913
E-ISSN: 2549-5747

Jurnal Sylva Lestari

Journal homepage: https://sylvalestari.fp.unila.ac.id

Full-Length Research Article

Characteristics of Cold-Setting Adhesive Derived from Waste Styrofoam for Bonding Laminated Gmelina (*Gmelina arborea*) Wood

Tati Karliati[1*], Rudi Dungani[1], Anne Hadiyane[1], Wahyu Hidayat[2], Muhammad Adly Rahandi Lubis[3**]

[1] School of Life Sciences and Technology, Institut Teknologi Bandung, Bandung, Indonesia
[2] Department of Forestry, Faculty of Agriculture, University of Lampung, Bandar Lampung, Indonesia
[3] Research Center for Biomass and Bioproducts, National Research and Innovation Agency, Cibinong, Indonesia
[*] Corresponding author. E-mail address: karliati69@itb.ac.id
[**] Corresponding author. E-mail address: muha142@brin.go.id

ARTICLE HISTORY:

Received: 15 September 2024
Peer review completed: 4 December 2024
Received in revised form: 4 April 2025
Accepted: 30 May 2025

KEYWORDS:

Adhesive
Block shear strength
Cohesion strength
Cold-setting
Gmelina wood

ABSTRACT

Developing cold-setting adhesives derived from waste styrofoam (WS) of expanded polystyrene foam for laminated gmelina wood involves innovative recycling of WS into functional adhesives. As a type of thermoplastic polymer, WS can be used as an adhesive at room temperature. In this study, WS was functionalized into cold-setting adhesives through dissolution and cross-linking reaction with methylene diphenyl diisocyanate (MDI) at a concentration of 40% w/v at 60°C. The mixture was then stirred at 200 rpm for 30 minutes. The adhesive is characterized by its ability to be set at room temperature, offering environmental benefits and practical applications in wood lamination. Solids content, gelation time, viscosity, cohesion strength, functional group analysis, morphological features, and curing temperature analysis were performed to characterize the cold-setting adhesives. The WS-MDI-40% had 62.4% solids content, 906.8 mPa.s viscosity, 182.3 Pa cohesion strength, and 197.8 minutes of gelation time at room temperature. The adhesion performance was evaluated in laminated wood using gmelina wood at glue spread rates of 250 and 300 g/m^2 and cold-pressed at various durations. The block shear strength value of laminated wood bonded with WS-MDI-40% at 300 g/m^2 and cold-pressed for 24 hours was higher than that of other samples, reaching 8.2 MPa, which met the Japanese Agricultural Standard (JAS No. 234) for glued laminated timber, exceeding the minimum requirement of 5.4 MPa. Fourier transform infrared (FTIR) spectroscopy revealed that the cold-setting WS-MDI-40% were cross-linked via urethane linkages (R–NH–COO–R). The free –N=C=O groups could react with the –OH groups of gmelina wood to produce laminated wood during cold-pressing. This study suggested a recycling alternative of WS into a cold-setting wood adhesive for laminated wood, which can be utilized in interior applications.

1. Introduction

Over the past decade, a significant emphasis on the viewpoint of environmental concerns about products manufactured from recycled materials has been raised. The progress of this product recycling method demonstrates that material recycling has become both economically and

environmentally attractive (Capricho et al. 2022; García-Barrera et al. 2022). In developed countries, properly managing waste electrical and electronic equipment, which constitutes a significant amount of discarded material, is a significant concern (Maafa 2021). Although the direct recycling of polystyrene into adhesives is not commonly practiced, several mechanisms exist for repurposing or reusing waste styrofoam (WS).

The increase in WS is not accompanied by an increase in the management of the recycled styrofoam process. Expanded polystyrene (EPS) is a common WS with a wide range of applications, including the manufacture of trays, bowls, plates, boxes, insulation foams, and packaging materials. The global production of WS is around 15 million metric tons per year, and most of the WS ends up in a landfill (Uttaravalli et al. 2021). In the United States (U.S.), over 3 million tons of WS are produced annually, and around 2.3 million tons end up in landfills. It is reported that less than 2% of WS were recycled in the U.S. (Fitriasari and Liu 2024). Around 10 wt.% of the total plastic waste produced annually originates from WS, and burning WS produces harmful gases such as styrene, hydrochlorofluorocarbon, carbon black, carbon monoxide, and polycyclic aromatic hydrocarbons (Aljabri et al. 2017).

WS dissolves readily in various solvents and has been utilized in the preparation of coatings and adhesives. When applied to various materials, such as plastic, wood, and cardboard, the adhesive exhibits good adhesion strength, physical performance, and mechanical properties. Polystyrene is a synthetic aromatic thermoplastic polymer produced from the styrene monomer, and is the world's third most widely used thermoplastic, with a glass transition temperature (*Tg*) of about 100°C (Noori et al. 2023). Below the *Tg*, WS is stiff, hard, and brittle; above *Tg*, it can be melted and bonded to the wood through mechanical interlocking at room temperature.

One way to utilize WS as a wood adhesive is by developing cold-setting adhesives. Cold-setting adhesives have several advantages over other commercial adhesives. This adhesive requires a moderate curing treatment at room temperature (25°C). Thus, this adhesive is eco-friendly, based on renewable resources, and possesses several functionalities, including serving as a plasticizer, emulsifier, and surfactant (Amin et al. 2023; Amin et al. 2024). Cold-setting adhesive also enables in-use, moist bonding processes for laminating wood by applying adhesive to the surface of each wood strip before assembly. Common cold-setting adhesives for laminated gmelina wood include epoxy (Dwianto et al. 2023), polyurethane (Hariz et al. 2023), and phenol-resorcinol-formaldehyde resins (Wibowo and Park 2024). However, conventional cold-setting adhesives are costly, non-renewable, and toxic.

Utilizing WS as raw material for cold-setting adhesive requires novel methods to convert the polystyrene into effective adhesive formulations. The WS-based adhesives possess the property of setting at ambient temperatures, thereby providing environmental advantages and practical utility in the wood laminating process (Noori et al. 2023). To synthesize sticky films, it is dissolved in solvents such as D-limonene, toluene, or mixtures of gasoline and acetone. An adhesive's characteristics, including viscosity, binding strength, and drying time, are influenced by solvent selection (Maulana et al. 2024; Noori et al. 2023; Osemeahon et al. 2024). Cold-setting adhesives, such as those produced from butylphenyl polyester, utilize reactive groups and a hybrid geometry to provide robust adhesion at ambient temperatures (Cao et al. 2024). Butylphenyl polyester and WS have high glass transition temperatures and do not warp or deform at relatively high temperatures. However, the bonding strength decreases with increasing exposure to high relative humidity (Noori et al. 2023).

Incorporating plasticizers, such as glycerol, can enhance the pliability of the adhesive; however, this may increase moisture content. Cross-linking agents containing isocyanate groups and dual-functional polymers can enhance both adhesive bond strength and heat resistance (Osemeahon et al. 2024). The bond strengths of adhesives produced from polystyrene waste vary depending on the formulation. A study by Osemeahon et al. (2022) demonstrated that adhesives containing a combination of toluene and acetone exhibited a strong bonding strength of 7.04 MPa on plywood. The lap shear strength of cold-setting adhesives is significantly high, rendering them well-suited for use in wood applications. Through the recycling of waste materials, these adhesives provide a sustainable solution to mitigate environmental contamination. Developing such adhesives aligns with generating value-added products from waste and promoting environmentally sustainable products (Osemeahon et al., 2022; Younesi-Kordkheili et al., 2024).

To achieve strong bonding and mechanical performance, the process parameters, such as pressure and adhesive content, are carefully adjusted (Budhe et al. 2017). Certain cold-setting adhesives, such as those formulated from butylphenyl polyester, provide repair capabilities, enabling the restoration of wooden constructions (Cao et al. 2024). The optimization of formulas for specific applications of cold-setting adhesives derived from waste styrofoam presents challenges despite the various advantages they offer. Precise control of solvent selection, adhesive composition, and curing conditions is necessary to attain the intended performance features. Optimizing reaction parameters, including temperature, duration, and the quantity of additives, is necessary to form adhesives from waste styrofoam (Montalvo-Romero et al. 2022). The primary requirement for solvent selection is the high solubility of WS, such as toluene and acetone. The selection of solvents affects the adhesive's viscosity, bond strength, and drying time. Optimal adhesive performance on different substrates has been observed when a combination of toluene and acetone is used (Osemeahon et al. 2022).

Recycling WS into adhesives mitigates environmental issues by diminishing pollution and generating sustainable value from waste materials. This method involves dissolving WS in solvents and subsequently reacting it with additives to generate desirable adhesives characterized by high peel strength and a minimal environmental footprint (Beran et al. 2021; Obele et al. 2024). Thermosetting adhesives are produced from WS by cross-linking with agents such as methylene diphenyl diisocyanate (MDI). This process improves the adhesive's thermo-mechanical characteristics and bonding strength, making it suitable for applications such as particleboard production (Karliati et al. 2024). The equilibrium between pliability and resistance to moisture is crucial for maintaining the integrity of adhesive joints in various environmental conditions. These factors underscore the ongoing need for research to develop the promise of cold-setting adhesives.

This work analyses and describes the cold-setting adhesive from WS for laminated wood made from gmelina (*Gmelina arborea*). The evaluation encompassed quantifying the solids content, gelation duration, viscosity, pH, rheological characteristics, functional group analysis, and curing analysis. Moreover, the adhesive strength of cold-setting adhesive obtained from WPS was examined using a block shear strength test on laminated wood.

2. Materials and Methods

2.1. Materials

The primary material was waste styrofoam (WS), collected from Cibinong, Indonesia. The solvents used were toluene (95.0% technical grade), butanol (99.0% technical grade), propanol (99.0% technical grade), methylene chloride (99.0% technical grade), ethyl acetate (99.0% technical grade), and butyl acetate (99.0% technical grade). Methylene diphenyl diisocyanate (MDI) with a ± 31% NCO content and 98.7% solids content was bought from Anugerah Raya Kencana Company, Serang, Indonesia. Aquades were bought from a local chemical store in Bogor, Indonesia. Wood samples of 7-year-old gmelina (*Gmelina arborea*) with a rectangular shape of 200 mm × 100 mm × 20 mm were obtained from a community forest in Bandung, Indonesia.

2.2. Formulation of Cold-Setting Adhesive Derived from Waste Styrofoam

Fig. 1 illustrates the preparation of waste styrofoam (WS)-based adhesive. The WS used in this study was a food container. A comprehensive cleaning was conducted on the WS, followed by its subsequent crushing into smaller fragments. WS particles were dissolved using a previously described design, with certain modifications (Gutierrez-Velasquez et al. 2022; Karliati et al. 2024). About 400 grams (40% w/v) of WS particles were dissolved in a co-solvent mixture. 100 mL of methylene chloride, 100 mL of ethyl acetate, 150 mL of butyl acetate, 90 mL of butanol, 60 mL of propanol, and 500 mL of toluene were used as co-solvents. The dissolution process was carried out at a temperature of 60°C, with continuous stirring at 100 revolutions per minute for 10 minutes. Following the complete dissolution of WS, approximately 40% w/v of MDI was introduced into the liquid WS. The cross-linking reaction was performed at 60°C and maintained at 200 rpm for 30 minutes.

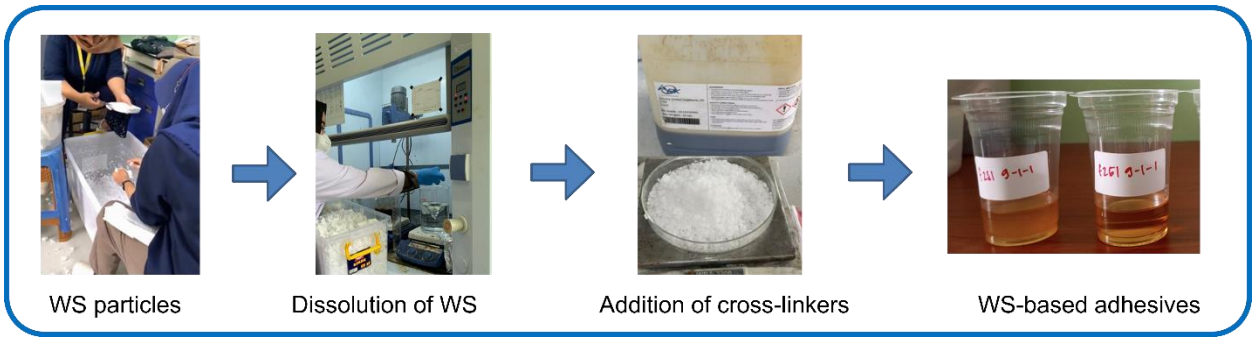

Fig. 1. Preparation of WS-based adhesive.

2.3. Characterization of Cold-Setting Adhesive Derived from Waste Styrofoam

This study evaluated the properties of adhesives derived from WS, including solids content, viscosity, gelation time, and pH. The solids content in WS-modified MDI adhesives at 40% MDI levels was determined by drying a 1 g sample in an oven set at 105°C for 3 hours. Once the drying process was completed, the oven-dried weight was divided by the initial weight to calculate the solids content. The average viscosity of WS-modified MDI adhesives at 40% MDI levels was measured using a rotational rheometer (RheolabQC, AntonPaar, Austria). The measurement was performed at 27°C for 120 seconds with a consistent shear rate of 100/s. The measurement was conducted using a No. 27 spindle apparatus. The gelation time of WS-modified MDI adhesives at

40% MDI levels was assessed by monitoring the duration for the adhesive to become a gel at 30°C and 50 rpm using a gel time meter (GT-6, Techne Inc., USA). Using a digital pH meter (OrionStar A111, Thermo Scientific, USA), the pH of WS-modified MDI adhesives at 40% MDI levels was determined at a temperature of 27°C.

Fourier transform infrared (FTIR) spectroscopy was conducted using the Spectrum Two instrument (Perkin Elmer Inc., MA, USA) to investigate the alterations in the functional groups of WS-modified MDI adhesives at a 40% level of MDI. The adhesive samples were analyzed using the attenuated total reflectance (ATR) technique at a temperature of 25°C. The ATR-FTIR was conducted within the 400 to 4000 cm^{-1}, using a 4 cm^{-1} resolution for each sample. The viscosity, cohesion strength, and rheological behaviour of WS-modified MDI adhesives at 40% levels of MDI were analyzed using a rotational rheometer (RheolabQC, AntonPaar, Austria) at 27°C under a constant shear rate of 200/s.

Dynamic mechanical analysis (DMA 8000, PerkinElmer Inc., USA) was performed to investigate the thermo-mechanical characteristics of WS-modified MDI adhesives at a 40% level of MDI. Each adhesive was used to bind two Whatman filter sheets together, using a glue spread of 200 g/m^2, resulting in a sample measuring 50 mm in length, 8 mm in width, and 0.2 mm in thickness. The DMA was performed at temperatures ranging from 30 to 100˚C. Each specimen's storage modulus (E′) was determined using the dual cantilever mode at a strain level of 0.01%, 1 Hz, and heating rates of 1, 3, and 5˚C/min.

2.4. Preparation and Evaluation of Laminated Gmelina Wood Properties

As shown in **Fig. 2**, the preparation of laminated gmelina wood bonded with WS-modified MDI adhesives at a 40% level of MDI was performed according to the procedure described in a published work (Lubis et al. 2022a). The surface of the gmelina wood was flattened by a sander. The laminated wood was produced at glue spread rates of 250 and 300 g/m^2. The laminated gmelina wood was bonded in parallel fiber orientation using a cold press at 30°C and 10.0 MPa for 2, 6, and 24 hours. Before testing and evaluating the adhesion performance of the laminated gmelina wood, the samples were pre-conditioned at room temperature for one week, following the guidelines of the Japan Agriculture Standard (JAS) for glued laminated timber (JAS 2007). The adhesion performance of laminated gmelina wood bonded with WS-modified MDI adhesives at 40% MDI levels and cold-pressed for different hours was evaluated using block shear strength (BSS). Approximately six samples of each laminated gmelina wood were fabricated, each measuring 30 mm × 25 mm, to assess the BSS. The BSS was measured using a universal testing machine (UTM, AG-IS 50 kN, Shimadzu, Japan) at a cross-head speed of 2 mm/min.

2.5. Data Analysis

Microsoft Excel 2019 (Microsoft, USA) and IBM SPSS Statistics 20 (IBM SPSS Statistics 20, SPSS Inc., USA) were used to statistically analyze the obtained data statistically, employing a completely randomized design with two factors: the glue spread and the cold-pressing times. Each treatment was carried out three times. The analysis of variance (ANOVA) with a 95% confidence level was performed accordingly. Duncan's multiple range test (DMRT) was conducted to identify significant differences between treatments on parameters.

Fig. 2. Preparation and evaluation of laminated gmelina wood bonded with cold-setting WS-MDI adhesive.

3. Results and Discussion

3.1. Basic Properties of WS-MDI Adhesives

Table 1 presents the basic properties of the cold-setting WS-MDI adhesive, including solids content, gelation time, average viscosity, cohesion strength, and pH. The adhesive's solids content measures the amount of particles present in the liquid adhesive. The bonding strength will be improved by augmenting the number of adhesive particles that come into contact with the wood throughout the gluing procedure. The WS had an average solids content of 38.90%, and MDI had a solids content of 98.75%. The mixture of WS-MDI-40% had a solids content of 62.46%. Adding MDI into the WS-based adhesive system could increase the solids content, which is comparable to the standard solids content of commercial cold-setting phenol-resorcinol-formaldehyde (PRF) adhesives, around 60–65% (BSN 1998; Wibowo and Park 2024). As MDI has a solids content of 98.7%, adding MDI to WS-based adhesives increases the solids content. This result was in line with published works that reported that adding MDI increased the solids content of polystyrene-based adhesives (Obele et al. 2024; Uttaravalli et al. 2021).

Table 1. Basic properties of WS, MDI, and cold-setting WS-MDI-40% adhesive

Formula	Solids content (%)	Gelation time (min)	Average viscosity (mPa.s)	Cohesion strength (Pa)	pH
WS	38.90 ± 0.48	nd	31.54 ± 0.52	6.27 ± 0.01	4.8 ± 0.02
MDI	98.75 ± 0.25	359.55 ± 5.70	200.4 ± 0.25	30.05 ± 0.01	5.5 ± 0.03
WS-MDI	62.46 ± 0.37	197.86 ± 2.53	906.81 ± 1.82	182.32 ± 0.10	5.9 ± 0.05

Note: nd= not detected.

Gelation time is the duration needed for an adhesive to change from a liquid to a gel form. This gel time value is essential for establishing the adhesive's working time and setting properties. **Table 1** shows the adhesive's gelation time test at 30°C. The WS did not have a gel time value because the WS did not have reactive functional groups. By contrast, MDI had a gel time of 359.55 minutes due to the presence of reactive isocyanate (-N=C=O) groups. The mixture of WS-MDI-40% had a faster gel time of 197.86 minutes at 30°C. This is possibly because the reaction between the (-N=C=O) groups and the hydrogen (H) atoms in WS to form urethane linkages (R–NH–COO–R) (Zhu et al. 2015). Compared to epoxy and polyurethane resins, the gelation time of WS-MDI adhesive was much longer. Epoxy resin has a gelation time range of 30–50 min at 25°C (Mubarok

et al. 2025), while polyurethane has a gelation time of 80−100 min at a similar temperature (Baskara et al. 2023; Mubarok et al. 2025). A faster gelation time at lower temperatures suggests that a longer pressing time is required when using cold-setting adhesives. However, the adhesive will coagulate rapidly due to the short gelation time, which will reduce the adhesive's shelf life (Wibowo and Park 2024). This is also related to the adhesive's solids content, where the value will be inversely proportional. The higher the solids content of an adhesive, the shorter the gelation time (Fitrianum et al. 2023).

The viscosity of an adhesive significantly impacts its capacity to wet the surfaces, penetrate substrates, and form strong bonds. High-solids-content and high-viscosity adhesives can establish optimal bonding and adequate adhesion (Radabutra 2020). The average viscosity values of WS, MDI, and WS-MDI-40% adhesives tested at 27°C are shown in **Table 1**. The WS had an average viscosity of 31.54 mPa.s, and MDI had an average value of 200.48 mPa.s. The mixture of WS-MDI-40% had an average viscosity of 906.81 mPa.s. MDI acted as a heat stabilizer, effectively inhibiting thermal oxidation by neutralizing the acid catalyst of WS. Additionally, MDI caused a significant increase in the viscosity of neat WS. The comparison of the viscosity enhancement effect of MDI on WS and WS-MDI showed that the viscosity of WS-MDI blends was higher than that of neat WS melted polymer. Cross-linking and the formation of a branched structure in WS-MDI blends contributed to a higher viscosity than that of non-cross-linked WS (Karliati et al. 2024; Uttaravalli et al. 2021). A higher viscosity value can influence the shelf life of the adhesive, where the higher the value, the shorter the shelf life (Ju et al. 2022). This study requires a high-viscosity adhesive for cold-setting adhesive to manufacture laminated gmelina wood. This result was in line with published works, which reported that adding MDI increased the solids content of polystyrene-based adhesives (Obele et al. 2024; Uttaravalli et al. 2021).

Cohesion strength is critical for cold-setting adhesives, specifically formulated to solidify at ambient temperature. This could form a strong adhesion for bonding the laminated wood without the need for hot-pressing. **Table 1** shows the cohesion strength values of WS, MDI, and WS-MDI-40% adhesives tested at 27°C. The WS had a cohesion strength of 6.27 Pa, and MDI had an average value of 30.05 Pa. The mixture of WS-MDI-40% had a cohesion strength value of 182.32 Pa. A higher viscosity value can influence the cohesion strength of the adhesive (Ju et al. 2022). The cohesion strength of adhesives is primarily dependent on the polymer structure as well as on the physical state, which can be characterized by viscosity. Adhesives with low viscosity have low energy for chemical reactions, thus resulting in low values of cohesion strength and fast adhesion failure. Meanwhile, high viscosity results in greater cohesion strength due to the chemical reaction (Gutierrez-Velasquez et al. 2022; Karliati et al. 2024; Uttaravalli et al. 2021). Cold-setting adhesives are designed to cure at ambient temperatures, providing strong bonds without the need for heat. The chemical composition of WS-MDI-40% influences the cohesion strength.

Acidity (pH) can influence the properties of cold-setting adhesives, as it impacts the polymerization process and the surface interaction between the adhesive and wood. An adhesive's pH level can potentially modify its mechanical characteristics and degree of adhesion. As reported in published works, the adhesive bonding strength of wood with a polystyrene-based adhesive was found to be inversely affected by the pH of the adhesive (Uttaravalli et al. 2021). The acidic pH was found to be more harmful to the wood surface than the alkaline pH. As presented in **Table 1**, the liquid WS had a pH of 4.8, while MDI had a pH of 5.5. The mixture of WS-MDI-40% had a higher pH value of 5.9. Generally, the bonding strength was worst in adhesive solutions with the

lowest pH values (Anastasiadis et al. 2018). The addition of MDI increased the pH due to the presence of nitrogen in the isocyanate (–N=C=O) groups.

3.2. Chemical, Thermal, and Curing Behaviour Properties of Cold-Setting WS-MDI Adhesives

Several parameters, including the adhesive composition, curing conditions, and the interaction of its chemical constituents, influence the chemical, thermal, and curing behavior of cold-setting WS-MDI adhesives. The WS-MDI adhesives offer a sustainable approach by upcycling non-biodegradable WS into cold-setting adhesive for laminated wood. **Fig. 3a** reveals the FTIR spectra of WS, MDI, and cold-setting WS-MDI adhesives. The wavenumber around 3050 cm^{-1} was originated from the aromatic C–H stretching vibration of WS. Additionally, the wavenumbers at 1605 cm^{-1} and 1492 cm^{-1} correspond to the aromatic C=C stretching vibration absorption, indicating the presence of benzene rings in WS. The wavenumbers at 2940 cm^{-1} and 2845 cm^{-1} indicated the presence of methylenes (–CH_2–). These findings align with the published work that involves preparing polymer blends consisting of polystyrene (Zhu et al. 2015). Meanwhile, MDI had a typical isocyanate (–N=C=O) group at 2245 cm^{-1} and a carbonyl (C=O) group at 1775 cm^{-1}. Several studies have shown that the –N=C=O and C=O groups of MDI appear at wavenumbers of 2240–2250 cm^{-1} and 1750–1775 cm^{-1}, respectively (Lubis et al. 2020a; Lubis et al. 2020b).

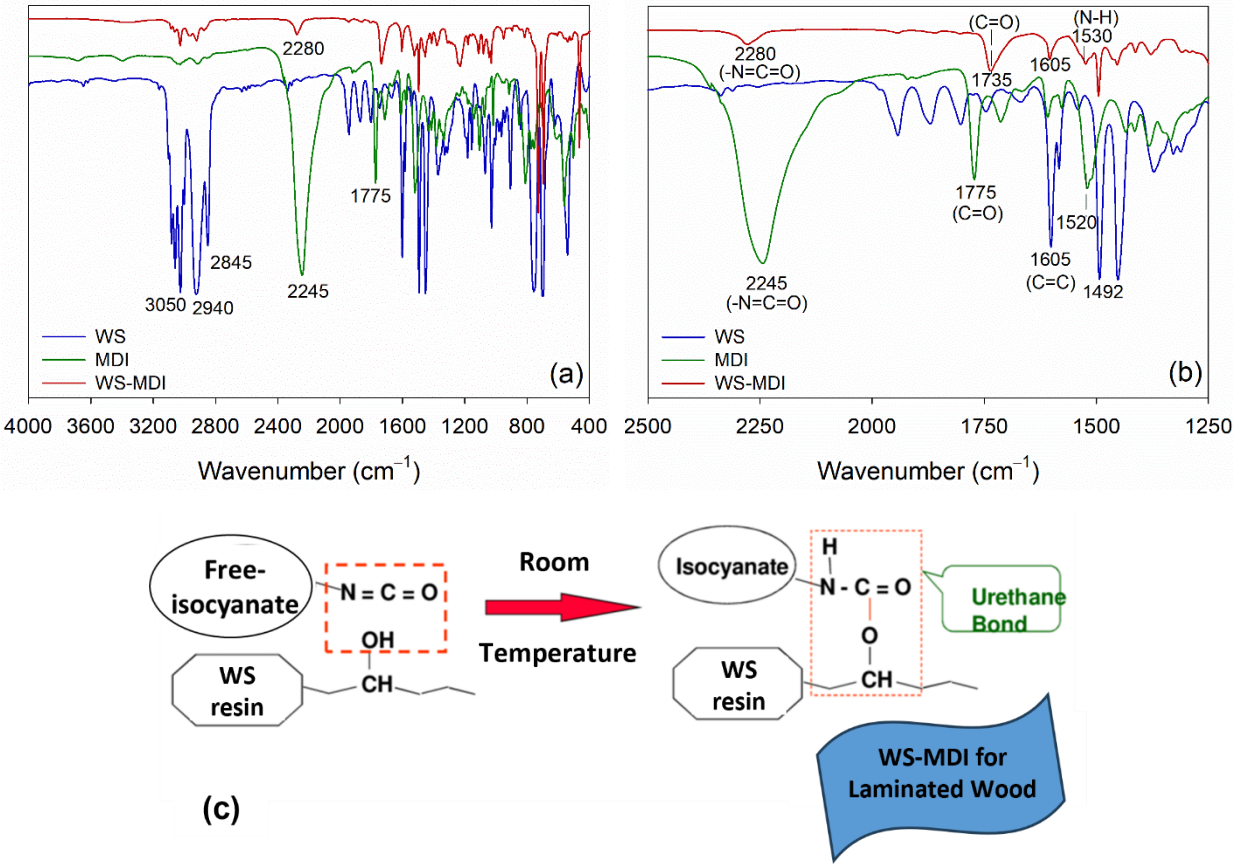

Fig. 3. FTIR spectroscopy of WS, MDI, and cold-setting WS-MDI adhesives. (a) full FTIR spectra, (b) enlarged FTIR spectra, (c) reaction scheme.

The blending of WS with 40% MDI resulted in the formation of several functional groups, as displayed in **Fig. 3b**. The 2280 cm^{-1} wavenumber indicated the presence of free isocyanate

(–N=C=O) groups in the WS-MDI-40% blend. These free isocyanate groups could react with the hydroxyl (–OH) groups on the surface of gmelina wood to manufacture laminated wood. In addition, wavenumbers at 1735 cm^{-1} and 1530 cm^{-1} were assigned to the C=O and N–H stretching vibrations of urethane linkages, respectively. **Fig. 3c** displays the possible reaction scheme of WS and 40% MDI. These findings revealed that the cold-setting WS-MDI-40% were cross-linked via urethane linkages (R–NH–COO–R). The free –N=C=O groups could react with the –OH groups on the surface of gmelina wood to produce laminated wood during cold pressing (Zhu et al. 2015).

Understanding the intricate interactions between WS and MDI is crucial for understanding the rheological behavior, as these interactions can significantly impact the material's flow and deformation. **Fig. 4a** shows the shear stress-shear strain curve of WS-MDI adhesive. The cold-setting WS-MDI was characterized by higher shear stress as a function of shear strain. Shear strain hardening is a prominent characteristic observed in WS-MDI, predominantly at elevated shear stress. Beyond a critical shear stress threshold of approximately 104 Pa, this behavior shifts from conventional stress overshoot to hardening (Liu and Wang 2016). **Fig. 4b** illustrates the relationship between the cohesion strength and viscosity of the WS-MDI adhesive. Higher viscosity resulted in greater cohesion strength of WS-MDI adhesive. The chemical interactions between the constituent polymers affect the viscosity of WS-MDI blends. Specifically, the viscosity increases exponentially when the –N=C=O groups of MDI react with the H atom WS (**Fig. 3c**). Furthermore, blends of WS-MDI demonstrate enhanced cohesion strength compared to the neat WS and MDI (**Table 1**). The reason for this phenomenon is the interplay between different phases, which indirectly impacts the glass transition temperature (*Tg*) and the mechanical properties of the WS-MDI mixture (Li et al. 2015). As displayed in **Fig. 5a**, the neat WS had *Tg* from 90.5 to 134.5°C, with a peak temperature of 107.8°C. Incorporating MDI into WS lowered the temperatures, ranging from 72.4 to 88.7°C. This indicates that the WS-MDI adhesive can be efficiently cured at 72.4°C at a heating rate of 1°C/min. Cold-setting adhesives are specifically formulated to solidify at ambient temperature. The viscosity of the WS-MDI adhesive influences its application procedure, whereas the cohesion strength governs the durability and reliability of the adhesive bond.

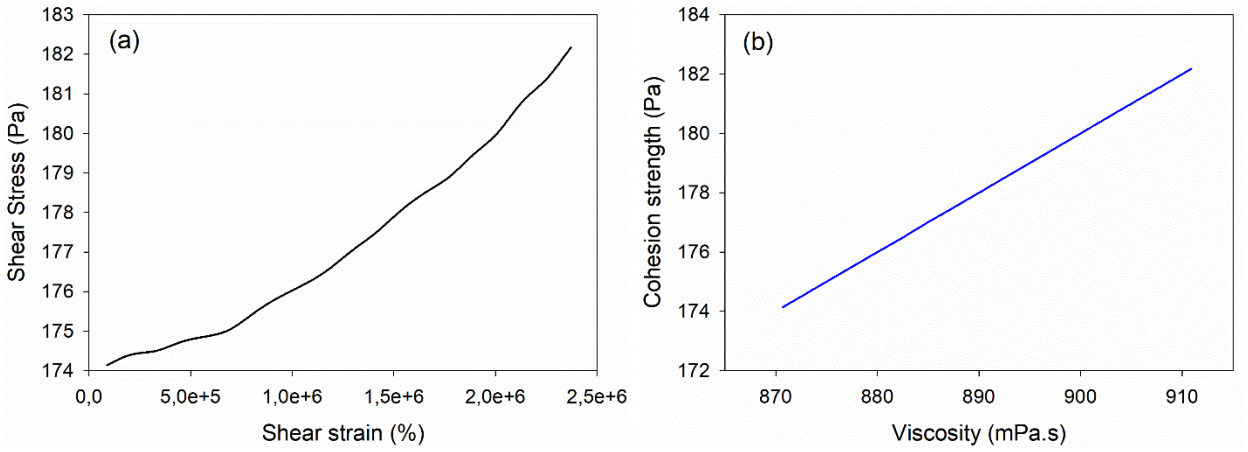

Fig. 4. Rheological behavior of cold-setting WS-MDI adhesive. (a) shear stress-shear strain curve, (b) cohesion strength and viscosity relationship.

DMA efficiently examines the thermal-curing characteristics of cold-setting WS-MDI adhesive, enabling a comprehensive examination of its curing behavior (Dunky 2021). The neat

WS had *Tg* from 90.5 to 134.5°C, with a peak temperature of 107.8°C (**Fig. 5a**). Incorporating MDI into WS increased the storage modulus of the adhesive at lower temperatures, ranging from 72.4 to 88.7°C. This indicates that the WS-MDI adhesive can be efficiently cured from 72.4°C to reach the maximum storage modulus of 3875 MPa at a heating rate of 1°C/min. A lower heating rate of curing, such as 0.10°C/min, can produce cold-setting WS-MDI adhesives at room temperature (30°C). The curing of cold-setting adhesives, such as WS-MDI, is influenced by variables including temperature and catalyst activation. These variables influence the degree of polymerization and the development of bonding strength (Wibowo and Park 2024). Cold-setting WS-MDI adhesives are specifically formulated to cure at room temperature, offering benefits in outdoor applications where maintaining regulated conditions is impossible (Lubis et al. 2023). Nevertheless, lower temperatures can impede the curing process, resulting in inadequate polymerization and decreased mechanical properties, as indicated by the lower storage modulus in **Fig. 5b**.

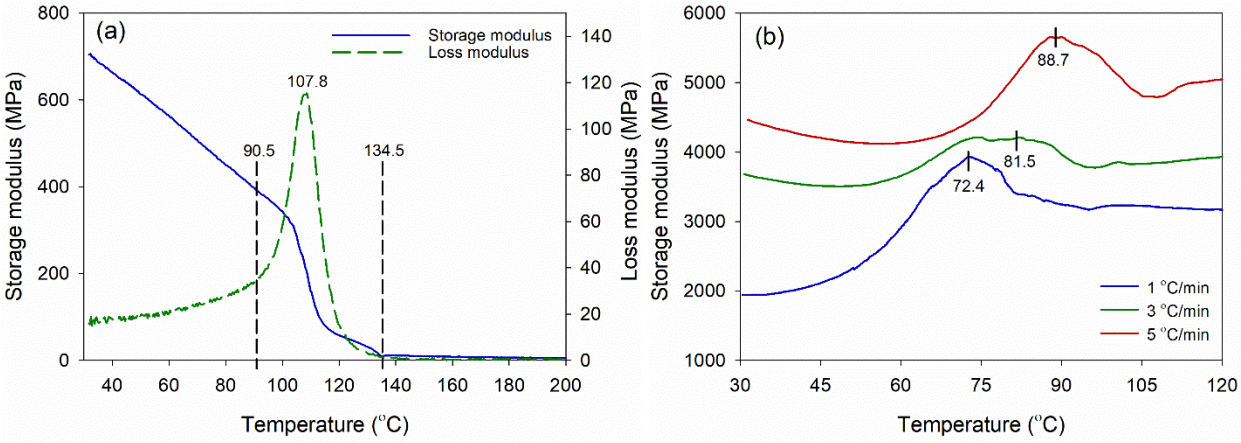

Fig. 5. Curing behavior of cold-setting WS-MDI adhesive: (a) typical DMA thermogram of WS, (b) storage modulus of WS-MDI adhesives at different heating rates.

3.3. Properties of Laminated Wood Bonded with Cold-Setting WS-MDI Adhesives

Fig. 6 displays the stress-strain curves of laminated gmelina wood at 250 and 300 g/m^2 of glue spread and cold-pressed at different times. Higher stress was generally observed at greater glue spread and longer cold-pressing time. Several studies have shown that longer cold-pressing times result in greater stress and bonding strength values, indicating that cold-pressing time has a remarkable influence on the formation of adhesion strength (Hermiati et al. 2019; Lubis et al. 2023). The highest stress, nearly 85.7 MPa, was achieved in laminated wood bonded with WS-MDI at 300 g/m^2 and cold-pressed for 24 hours. The lowest stress, around 20.5 MPa, was obtained in laminated wood bonded with WS-MDI at 250 g/m^2 and cold-pressed for 2 hours. Cold-setting WS-MDI adhesive is specifically formulated to complete the curing process at ambient temperature. However, shorter pressing time can hinder the curing process, resulting in inadequate polymerization and decreased mechanical properties (Kim et al. 2021), as indicated by the lower stress strain in **Fig. 6**. MDI, a prevalent isocyanate employed in cold-setting adhesives, enhances the bonding strength by forming urethane (R–NH–CO–R) linkages (Lubis et al. 2022a).

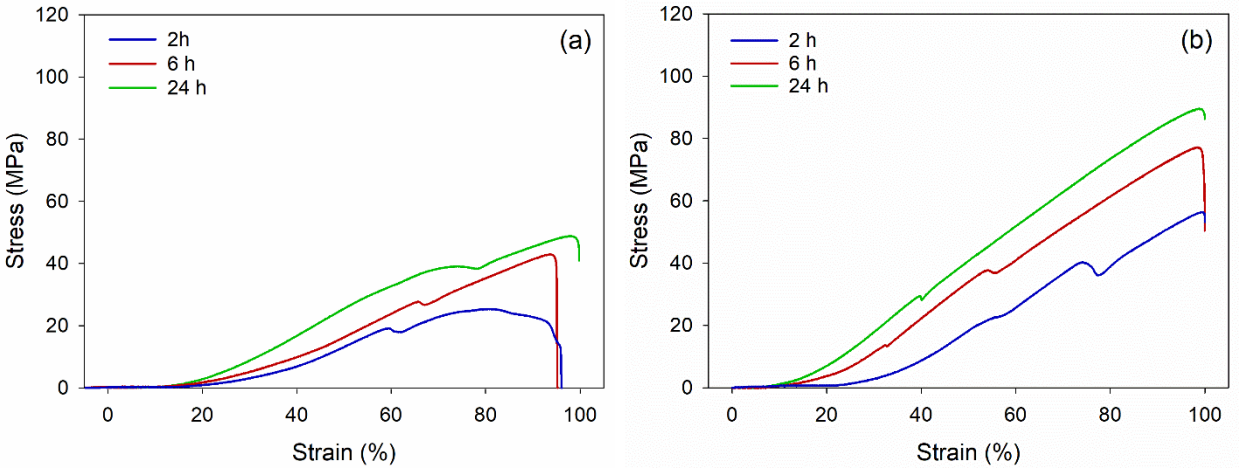

Fig. 6. Stress-strain curves of laminated wood bonded with WS-MDI adhesives at different glue spreads and cold-pressed for different times. (a) 250 g/m^2, (b) 300 g/m^2.

Fig. 7 presents the BSS values of laminated gmelina wood bonded with WS-MDI adhesives at different glue spreads and cold-pressed for different durations. Greater BSS values were generally observed at higher amounts of glue spread (Amin et al. 2023) and longer cold-pressing times (Lubis et al. 2022b). The highest BSS value, nearly 8.2 MPa, was obtained in laminated wood bonded with WS-MDI at 300 g/m^2 and cold-pressed for 24 hours. The lowest BSS value, around 1.7 MPa, was obtained in laminated wood bonded with WS-MDI at 250 g/m^2 and cold-pressed for 2 hours. This study showed that only laminated gmelina wood bonded with WS-MDI at 300 g/m^2 and cold-pressed for 6 and 24 hours achieved a block shear strength exceeding 5.4 MPa, as set by JAS No. 234 for glued laminated timber (JAS 2007). Cold-setting WS-MDI adhesive is specifically formulated to complete the curing process at ambient temperature. Nevertheless, lower temperatures can hinder the curing process, resulting in inadequate polymerization and decreased mechanical properties (Kim et al. 2021).

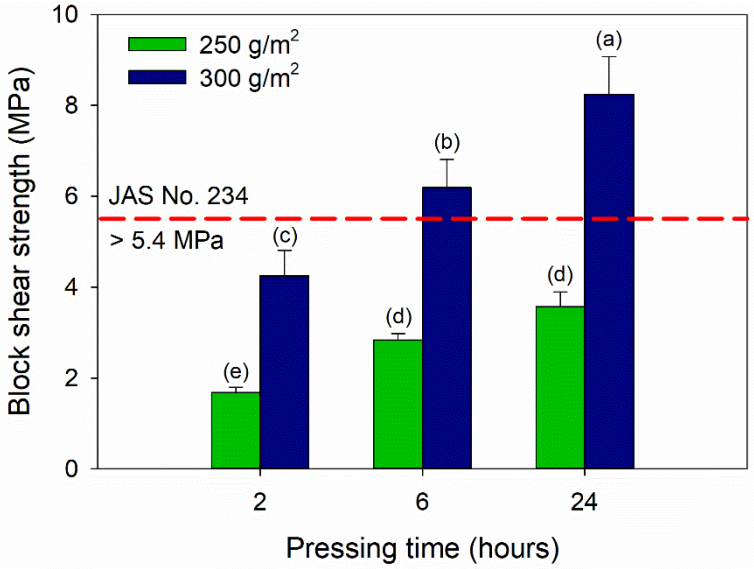

Fig. 7. Block shear strength (BSS) of laminated wood bonded with WS-MDI adhesives at different glue spreads and cold-pressed for different times.

Fig. 8 shows an example of a micrograph of BSS of laminated wood bonded with WS-MDI adhesives at a glue spread of 300 g/m^2 and cold-pressed for 24 hours. The micrograph revealed that not all wood surfaces were wet by the adhesives. This is possibly due to the high viscosity of the WS-MDI adhesive (Amin et al. 2024), which reaches 906.8 mPa.s. Incorporating MDI into WS resulted in a yellow-brown adhesive film. A high addition of MDI (40% w/v) in WS enhanced the cohesion strength of the adhesive (**Table 1**), which eventually increased the bonding strength of laminated gmelina wood.

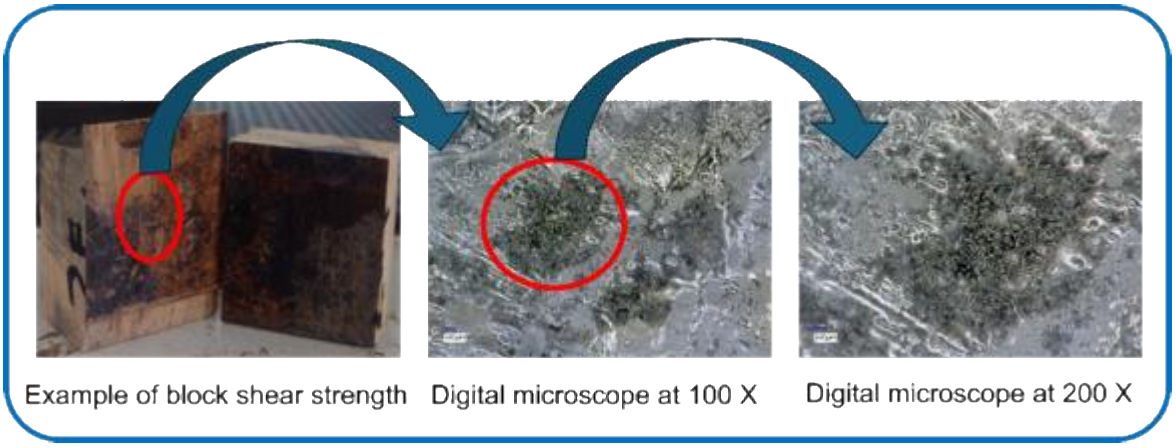

Fig. 8. Micrograph of block shear strength of laminated wood bonded with WS-MDI adhesives at a glue spread of 300 g/m^2 and cold-pressed for 24 hours.

4. Conclusions

A novel approach to recycling waste styrofoam (WS) into cold-setting adhesives is employed for laminated gmelina wood. Cold-setting adhesives were produced by functionalizing the WS by dissolving and cross-linking with methylene diphenyl diisocyanate (MDI) at a solution concentration of 40%. This cold-setting WS-MDI adhesive is distinguished by its ability to solidify at room temperature, providing environmental advantages and practical uses in wood laminating. The WS-MDI-40% had a solids content of 62.4%, a viscosity of 906.8 mPa.s, a cohesion strength of 182.3 Pa, and a gelation time of 197.8 minutes at room temperature. The properties of laminated wood using gmelina were evaluated at different glue spreads of 250 and 300 g/m^2, followed by cold pressing for varying durations. The laminated wood bonded with WS-MDI-40% at a glue spread of 300 g/m^2 and subjected to cold pressing for 24 hours exhibited a block shear strength value of 8.2 MPa, surpassing the minimum requirement of 5.4 MPa set by the JAS No. 234 for glued laminated timber. Fourier transform infrared (FTIR) spectroscopy showed that the cold-setting WS-MDI-40% were interconnected by urethane linkages (R–NH–COO–R). During cold-pressing, the free –N=C=O groups have the potential to undergo a reaction with the –OH groups present on the surface of gmelina wood, resulting in the formation of laminated wood. This work proposes a recycling option for WS as a cold-setting wood adhesive for laminated wood, suitable for interior applications. Further research is expected to enhance the reactivity of cold-setting WS-MDI, allowing it to be cold-pressed within an hour to meet the minimum requirements specified in JAS No. 234.

Acknowledgments

This study was funded by the School of Life Sciences and Technology, Institut Teknologi Bandung, through the Research, Community Service and Innovation (PPMI) Leading Research of Expertise Group Scheme, No. 28A/IT1.C11/SK-TA/2024, titled "*Aplikasi Perekat Bebas Emisi Formaldehida dari Limbah Styrofoam pada Kayu Laminasi* (Laminated Wood) *Berbasis Kayu Hutan Rakyat*". This study was also supported by the Integrated Laboratory of Bioproducts, National Research and Innovation Agency, Republic of Indonesia.

Author Contributions

T.K.: Conceptualization, Methodology, Software, Validation, Writing – Review and Editing, Visualization, Supervision, Project Administration, Funding Acquisition; M.A.R.L.: Conceptualization, Methodology, Software, Validation, Writing – Review and Editing, Visualization, Supervision, Project Administration, Funding Acquisition; R.D.: Formal Analysis, Investigation, Resources, Data Curation, Writing – Original Draft Preparation; A.H.: Formal Analysis, Investigation, Resources, Data Curation, Writing – Original Draft Preparation; W.H.: Formal Analysis, Investigation, Resources, Data Curation, Writing – Original Draft Preparation.

Conflict of Interest

The authors declare no conflict of interest.

Declaration of Generative AI and AI-Assisted Technologies in the Manuscript Preparation

During the preparation of this work, the authors used Grammarly in order to improve the clarity and the English level. After using this tool/service, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

References

Aljabri, N. M., Lai, Z., Hadjichristidis, N., and Huang, K. W. 2017. Renewable Aromatics from the Degradation of Polystyrene under Mild Conditions. *Journal of Saudi Chemical Society*, 21(8): 983–989. DOI: 10.1016/j.jscs.2017.05.005

Amin, Y., Adji, R. P., Lubis, M. A. R., Nugroho, N., Bahtiar, E. T., Dwianto, W., and Karlinasari, L. 2023. Effect of Glue Spread on Bonding Strength, Delamination, and Wood Failure of Jabon Wood-Based Cross-Laminated Timber using Cold-Setting Melamine-Based Adhesive. *Polymers* 15(10): 2349. DOI: 10.3390/polym15102349

Amin, Y., Nugroho, N., Bahtiar, E. T., Dwianto, W., Lubis, M. A. R., Adzkia, U., and Karlinasari, L. 2024. Surface Roughness, Dynamic Wettability, and Interphase of Modified Melamine Formaldehyde-Based Adhesives on Jabon Wood. *Polymers* 16(8): 1084. DOI: 10.3390/polym16081084

Anastasiadis, K., Koulaouzidou, E. A., Palaghias, G., and Eliades, G. 2018. Bonding of Composite to Base Materials: Effects of Adhesive Treatments on Base Surface Properties and Bond Strength. *The Journal of Adhesive Dentistry* 20(2): 151–164. DOI: 10.3290/j.jad.a40302

Baskara, M. I. A., Hadi, Y. S., Lubis, M. A. R., Maulana, M. I., Sari, R. K., Febrianto, F., and Hidayat, W. 2023. Characteristics of Polyurethane Cross-Laminated Timber Made from a Combination of Pine and Coconut. *Jurnal Sylva Lestari* 11(2): 270–293. DOI:10.23960/jsl.v11i2.691

Beran, R., Zárybnická, L., Machová, D., Večeřa, M., and Kalenda, P. 2021. Wood Adhesives from Waste-Free Recycling Depolymerisation of Flexible Polyurethane Foams. *Journal of Cleaner Production* 305: 127142. DOI: 10.1016/j.jclepro.2021.127142

BSN. 1998. *SNI 06-4567-1998 : Testing of Liquid Phenol Formaldehyde for Plywood Adhesive*. Badan Standardisasi Nasional (BSN). Jakarta, Indonesia.

Budhe, S., Banea, M. D., de Barros, S., and Da Silva, L. F. M. 2017. An Updated Review of Adhesively Bonded Joints in Composite Materials. *International Journal of Adhesion and Adhesives* 72: 30–42. DOI: 10.1016/j.ijadhadh.2016.10.010

Cao, W., Cong, R., Pan, W., Duan, X., Ning, C., Xie, T., Zhao, S., Li, Z., Gao, W., Luo, F., and Mao, T. 2024. A Versatile Cold-Curing Wood Adhesive Derived from Butylphenyl Polyester. *Polymer* 290(27): 126546. DOI: 10.1016/j.polymer.2023.126546

Capricho, J.C., Prasad, K., Hameed, N., Nikzad, M., and Salim, N. 2022. Upcycling Polystyrene. *Polymers* 14(22): 5010. DOI: 10.3390/polym14225010

Dunky, M. 2021. Wood Adhesives Based on Natural Resources: A Critical Review: Part IV. Special Topics. *Progress in Adhesion and Adhesives* 6: 761–840. DOI: 10.1002/9781119846703.ch18

Dwianto, W., Darmawan, T., Nugroho, N., Lubis, M. A. R., Bahanawan, A., Adi, D. S., and Triwibowo, D. 2023. *Development of Compressed Bamboo Lamination from Curved Cross-Section Slats*. Springer Nature Singapore, Singapore.

Fitrianum, F., Lubis, M. A. R., Hadi, Y. S., Sari, R. K., Maulana, M. I., Kristak, L., Iswanto, A. H., Mardawati, E., Reh, R., and Sedliacik, J. 2023. Adhesion and Cohesion Strength of Phenol-Formaldehyde Resin Mixed with Different Types and Levels of Catalyst for Wood Composites. *Journal of Composites Science* 7(8): 310. DOI: 10.3390/jcs7080310

Fitriasari, E. I. and Liu, J. J. 2024. Sustainability Assessments of Styrofoam Waste Recycling for Styrene Monomer Recovery: Economic and Environmental Impact. *34 European Symposium on Computer Aided Process Engineering/15 International Symposium on Process Systems Engineering.*

García-Barrera, L. V., Ortega-Solís, D. L., Soriano-Giles, G., Lopez, N., Romero-Romero, F., Reinheimer, E., Varela-Guerrero, V., and Ballesteros-Rivas, M. F. 2022. A Recycling Alternative for Expanded Polystyrene Residues using Natural Esters. *Journal of Polymers and the Environment* 30: 3832–3839. DOI: 10.21203/rs.3.rs-1168206/v1

Gutierrez-Velasquez, E. I., Monteiro, S. N., and Colorado, H. A. 2022. Characterization of Expanded Polystyrene Waste as Binder and Coating Material. *Case Studies in Construction Materials* 16: e00804. DOI: 10.1016/j.cscm.2021.e00804

Hariz, T. M. R., Hadi, Y. S., Lubis, M. A. R., Maulana, M. I., Sari, R. K., and Hidayat, W. 2023. Physical and Mechanical Properties of Cross-Laminated Timber Made of a Combination of Mangium-Puspa Wood and Polyurethane Adhesive. *Jurnal Sylva Lestari* 11(1): 37–65. DOI: 10.23960/jsl.v11i1.645

Hermiati, E., Fatriasari, W., Risanto, L., Darmawan, T., Sari, F. P., Jayadi, Sudarmanto, Laksana, R. P. B., Akbar, F. 2019. Optimization of Application of Natural Rubber Based API Adhesive for the Production of Laminated Wood. *IOP Conference Series: Earth and Environmental Science* 374(1): 012007. DOI: 10.1088/1755-1315/374/1/012007

JAS. 2007. *JAS No. 234 Glued Laminated Timber. Japanese Agricultural Standard*. Ministry of Agriculture, Forestry, and Fisheries, Tokyo, Japan.

Ju, J., Jin, S., Kim, S., Choi, J. H., Lee, H. A., Son, D., Lee, H., and Shin, M. 2022. Addressing The Shortcomings of Polyphenol-Derived Adhesives: Achievement of Long Shelf Life for Effective Hemostasis. *ACS Applied Materials and Interfaces* 14(22): 25115–25125. DOI: 10.1021/acsami.2c03930

Karliati, T., Lubis, M. A. R., Dungani, R., Maulani, R. R., Hadiyane, A., Rumidatul, A., Antov, P., Savov, V., and Lee, S. H. 2024. Performance of Particleboard Made of Agroforestry

Residues Bonded with Thermosetting Adhesive Derived from Waste Styrofoam. *Polymers* 16(4): 543. DOI: 10.3390/polym16040543

Kim, M., Park, B. D., and Hong, S. I. 2021. Effect of Synthesis Parameters of Cold-Setting Melamine-Urea-Formaldehyde Resins on the Adhesion in Wood Bonding. *Journal of Adhesion Science and Technology* 35(21): 2375–2390. DOI: 10.1080/01694243.2021.1885902

Li, X., Kang, H. L., Shen, J. X., Zhang, L. Q., Nishi, T., and Ito, K. 2015. Miscibility, Intramolecular Specific Interactions and Mechanical Properties of a DGEBA Based Epoxy Resin Toughened with a Sliding Graft Copolymer. *Chinese Journal of Polymer Science* 33(3): 433–443. DOI: 10.1007/s10118-015-1596-0

Liu, G., and Wang, S. Q. 2016. Entangled Linear Polymer Solutions at High Shear: From Strain Softening to Hardening. *Macromolecules* 49(24): 9647–9654. DOI: 10.1021/acs.macromol.6b02053

Lubis, M. A. R., Park, B. D., and Lee, S. -M. 2020a. Microencapsulation of Polymeric Isocyanate for the Modification of Urea-Formaldehyde Resins. *International Journal of Adhesion and Adhesives* 100(14): 102599. DOI: 10.1016/j.ijadhadh.2020.102599

Lubis, M. A. R., Park, B. -D., and Hong, M. -K. 2020b. Tuning of Adhesion and Disintegration of Oxidized Starch Adhesives for the Recycling of Medium Density Fiberboard. *BioResources* 15(3): 5156–5178. DOI:10.15376/biores.15.3.5156-5178

Lubis, M. A. R., Falah, F., Harini, D., Sudarmanto, Kharisma, A., Tjahyono, B., Fatriasari, W., Subiyanto, B., Suryanegara, L., and Iswanto, A. H. 2023. Enhancing the Performance of Natural Rubber Latex with Polymeric Isocyanate as Cold-Pressing and Formaldehyde Free Adhesive for Plywood. *Journal of Adhesion* 99(1): 58–73. DOI: 10.3390/jcs7030113

Lubis, M. A. R., Sari, F. P., Laksana, R. P. B., Fatriasari, W., and Hermiati, E. 2022a. Ambient Curable Natural Rubber Latex Adhesive Cross-Linked with Polymeric Isocyanate for Bonding Wood. *Polymer Bulletin* 79(8): 6745–6757. DOI: 10.1007/s00289-021-03845-0

Lubis, M. A. R., Labib, A., Sudarmanto, Akbar, F., Nuryawan, A., Antov, P., Kristak, L., and Papadopoulos, A. N. 2022b. Influence of Lignin Content and Pressing Time on Plywood Properties Bonded with Cold-Setting Adhesive Based on Poly (Vinyl Alcohol), Lignin, and Hexamine. *Polymers* 14(10): 1–20. DOI: 10.3390/polym14102111

Maafa, I. M. 2021. Pyrolysis of Polystyrene Waste: A Review. *Polymers* 13(2): 1–30. DOI:10.3390/polym13020225

Maulana, S., Wibowo, E. S., Mardawati, E., Iswanto, A. H., Papadopoulos, A., and Lubis, M. A. R. 2024. Eco-Friendly and High-Performance Bio-Polyurethane Adhesives from Vegetable Oils: A Review. *Polymers* 16(11): 1613. DOI: 10.3390/polym16111613

Montalvo-Romero, N., Montiel-Rosales, A., Fernández-Lambert, G., and Fernández-Echeverría, E. 2022. Development of an Adhesive Based on Waste Management as a Mechanism Towards Sustainability. *Sustainability* 14(20): 13225. DOI: 10.3390/su142013225

Mubarok, M., Hadi, Y. S., Hermawan, D., Arrasuli, M. H., Ramadhan, V. A., Lubis, M. A. R., Abdillah, I. B., Ilmiawati, A., Darmawan, W., Gérardin, P., and Militz, H. 2025. Adhesion Performance of Furfurylated Plywood Bonded with Polyurethane and Epoxy Adhesives. *Journal of Adhesion Science and Technology* 39(7): 971–992. DOI:10.1080/01694243.2024.2433692

Noori, H., Shalbafan, A., and Kazemi-Najafi, S. 2023. Development of Plywood Bonded with a Novel Adhesive Film Made from Polystyrene Foam Waste Dissolved in D-limonene. *Wood*

Material Science and Engineering 18(6): 2082–2092. DOI: 10.1080/17480272.2023.2221220

Obele, C. M., Ibenta, M. E., and Ogbuagu, J. O. 2024. Production and Characterization of an Ecofriendly Polystyrene Waste Adhesive Made with Toluene–Acetone Solvent. *Polymer Bulletin* 81(11): 10235–10253. DOI: 10.1007/s00289-024-05196-y

Osemeahon, S. A., Akinterinwa, A., Fasina, E., Andrew, F. P., Shagal, M. H., Kareem, S. A., Reuben, U., Onyebuchi, P. U., Adelagun, O. R., and Esenowo, D. 2024. Reduction of Polystyrene/Polyurethane Plastic Wastes from the Environment into Binders for Water-Resistant Emulsion Paints. *Heliyon* 10(19): e27868. DOI: 10.1016/j.heliyon.2024.e27868

Osemeahon, S. A., Reuben, U., and Emmanuel, E. 2022. Development of Adhesive from Polystyrene Waste. *BIOMED Natural and Applied Science* 2(1): 13–24. DOI: 10.53858/bnas02011324

Radabutra, S., Khemthong, P., Saengsuwan, S., and Sangya, S. 2020. Preparation and Characterization of Natural Rubber Bio-Based Wood Adhesive: Effect of Total Solid Content, Viscosity, and Storage Time. *Polymer Bulletin* 77(1): 2737–2747. DOI: 10.1007/s00289-019-02881-1

Uttaravalli, A.N., Dinda, S., Gidla, B.R., Kasturi, G., Kasala, P., and Penta, G. 2021. Studies on Development of Adhesive Material from Post-Consumer (Waste) Expanded Polystyrene: A Two-Edged Sword Approach. *Process Safety and Environmental Protection* 145: 312–320. DOI:10.1016/j.psep.2020.08.026

Wibowo, E. S., and Park, B. D. 2024. Controlling the Stability and Adhesion Performance of Cold-Setting Phenol–Resorcinol–Formaldehyde Resin Adhesives through Methanol Addition. *International Journal of Adhesion and Adhesives* 132: 103717. DOI: 10.1016/j.ijadhadh.2024.103717

Younesi-Kordkheili, H., Pizzi, A., Erfani, S., and Amiri, M. 2024. The Synthesis of Maleated Nanolignin–Polyvinyl Alcohol–Hexamine Resin as a Cold-Setting Wood Adhesive. *European Journal of Wood and Wood Products* 82(1): 159–166. DOI: 10.1007/s00107-023-02003-9

Zhu, S. Y., Li, H., Zhang, X. M., Chen, W. X., and Feng, L. F. 2015. Synthesis, Copolymer Composition, and Rheological Behavior of Functionalized Polystyrene with Isocyanate and Amine Side Groups. *Designed Monomers and Polymers* 18(3): 232–241. DOI: 10.1080/15685551.2014.999462