

Production and Characterization of Non-Pressure and Cast Urea Formaldehyde Composites by Using Hot Air

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Abstract – The human population has increased tremendously over the last century. This increase has caused the rapid destruction of forest resources. This makes it difficult to access raw wood materials. When the forest products industry could not meet its needs for wood, it tried to meet this need using wood composites obtained from wood residues. Urea formaldehyde is one of the most used thermosetting resins in wood composite production worldwide. Urea formaldehyde combines wood particles under temperature and pressure to form a rigid structure. This material is generally used in the production of boards, such as medium-density fiber boards (MDF) and particleboards (PB).

In this study, UF glue was mixed with organic materials (pine flour, pine fiber, MDF powder, and wheat flour) and hardened in hot air without pressure. The samples were characterized physically and mechanically. According to the results, water absorption (WA) of the samples with added MDF powder was lower than that of the other samples. Both WA and thickness swelling (TS) of the WF-added samples were high, whereas the pressure strength and screw withdrawal (SR) strength were low. In this case, it can be said that the addition of WF significantly reduces the UF durability. It was concluded that with the method used in this study, UF composites with organic particle addition could be produced using only hot air without pressure.

Keywords – Urea Formaldehyde, Organic Filler, Wood Material, Thermoset Resin, Without Pressure

I. INTRODUCTION

This Over the past decades, there has been a significant interest in natural fiber-reinforced composites because of their light weight, low cost, less abrasive nature compared to glass fiber, and environmentally friendly nature. These materials have been utilized in a wide range of applications in automobile and construction industries. However, despite their diverse advantages, there are several issues that limit their applications [1]–[6]. In the case of wood composites, the hydrophilic nature of wood fibers makes them unsuitable for composite processing in water-free or high-melting-temperature polymer matrix systems [7]. For natural fiber-reinforced thermoset composites, volatile emissions from wood fibers can cause internal voids and odor issues in the final product. Smoldering can also occur during processing because of the low ignition temperature of wood fibers. This restricts the use of natural fibers in thermoplastic-based

composites and non-flame-retardant thermoset systems. In the case of wood plastic composites, this combination of wood fibers with a thermoplastic matrix has the potential to save energy because no chemical modifications are required, and less complex processing equipment is required.

More recent efforts to improve the mechanical performance, durability, and flame resistance of WPC without the use of highly toxic fire retardants have also led to the development of various wood composite systems [8]–[12]. The industries and individuals involved in wood and wood composite materials are now at the beginning of a significant period of development, clearly evidenced by the abundance of activities that are taking place. The ever-increasing pressure to eliminate environmental damage caused using petrochemical polymers has led to significant research and development of new polymer materials from renewable resources. The large capacity to use fillers (wood and natural fibers) with these materials has also led to great interest in using polymers as a means to improve the durability of wood and as a wood replacement in structural and other sector such as automotive and aviation applications. An example is the utilization of a composite material and design to replace a portion of an existing wooden bridge, not only to reduce maintenance and replacement costs, but also to improve the longevity of the structure [13]–[18].

Urea is the most popular raw material in the manufacture of thermosetting amino resins. It has the advantages of low cost and toxicity. The level of formaldehyde emissions depended on the pH during the curing process. High pH (alkaline conditions) results in the release of free formaldehyde [19]–[21]. Methylene ether groups are formed when formaldehyde reacts with substances containing a hydroxyl group. When urea contains a hydroxyl group, urea resins are produced [22]. Urea resins are commonly used as adhesives for bonding particleboard and MDF. Particleboard or MDF is a substrate for the resin to bond and cure. The resin is cured by the reaction of formaldehyde as well as other aldehydes with free amino groups that result from the decomposition of urea. Lignin and tannin are also used as substrate adhesive resins in the curing of wood composites, and approximately half of the lignin/tannin adhesives are used in global production compared with UF resins [23]–[26].

When urea-formaldehyde resin is applied to a substrate (e.g., a particle board) and cured, it forms a hard and infusible coating. The cured resin is thermosetting, meaning that it softens at high temperatures but does not revert to the liquid form. This is advantageous for many applications. For example, the curing of a powder-form resin on an MDF board is used to prevent the sticking of boards during stacking. Other cured UF resin products have a high resistance to organic solvents and durability when exposed to moisture, making them suitable for exterior applications. The mechanical and physical properties of UF resins depend on the nature of the curing process and the presence of modifiers in the resins. Because of the plastic nature of UF resins, they are usually brittle in their cured state; however, this can be modified for some applications [27], [28].

In the early stages of UF resin development, it was utilized as glue. The initial products were formed by impregnating the veneers with the resin and then piling these veneers and pressing them into a panel. These products do not require the addition of a catalyst or hardener, and UF is considered an acid-cure adhesive. After the Second World War, development work focused on the acid-curing properties of the resin. The alkylated UF resins were cured by reaction with atmospheric formaldehyde at approximately 120-150°C and the product of this reaction was a tough-colored thermosetting film. Several acid-cure applications for UF resins were developed in the 1950s and the 1960s. Forest product laboratory patents detail the use of UF resins as binders in hardwood plywood. The resins are usually applied at 100-150% resin solids based on the weight of the dry veneer, and the assembled veneer layup is pressed at 135-150°C [29]–[33].

In this study, urea formaldehyde glue was filled with pine flour (ÇF), pine fiber (ÇL), MDF powder (MDF), and wheat flour (WF), molded without applying pressure, and hardened in hot air to produce organic particle-filled thermoset composites. Thus, the aim is to produce UF composites without the need for pressure and with low heat energy. The produced samples were characterized physically and mechanically and the differences between them were determined.

II. MATERIALS AND METHOD

In this study, urea formaldehyde (UF) was used as the matrix material. UF was purchased from a local market. The UF solid content was 65%. The viscosity of UF, with a density of 1.281 g/cm³, was 300 cp, and its pH was 8.2. An ammonium sulfate (AS) solution was used as the hardening chemical for UF. The AS hardener was prepared with a concentration of 25 %. AS was purchased from a local market. Pine flour (ÇP), pine fiber (ÇL), MDF powder, and wheat flour (WF) were used as filling materials in the matrix. WP, ÇL, and MDF powders were obtained from timber workshop waste in the Mudurnu region. WF was purchased from a local market. The organic filler ratios and some properties are listed in Table 1.

Table 1: Some properties of the fillers used in composites

Organic Filler type	Sample code	Filler Ratio (g)	UF (g)	Particle Thickness (mm)	Fillers Moisture (%)	Filler Bulk density (kg/m ³)
Pine saw dust	ÇP	100	100	0,9-1,8	10	150
Pine fiber (cut by radial direction)	ÇL	100	100	0,8-1,2	10	100
Medium density fiber board saw dust	MDF	100	100	0,5-1	10	200
Wheat flour	WF	100	100	0,2	10	1200

The UF and AS mixtures were prepared before mixing the glue and fillers. The AS rate was 3% higher than that of the solid UF rate. After the AS hardener was added to the UF in the glass container, the mixture was mechanically mixed with a mixer for 5 min. After the glue solution was prepared, it was mixed with the UF organic filling materials in a plastic container using a mixer for 10 min (Fig.1(a)). The mixture was poured into a 40 × 70 × 70 mm aluminum container (Fig. 1(b)). The containers were then placed in an oven at 150 °C for 15 min. In this manner, WP, ML, MDF, and WF-filled UF composites were cured. The analysis samples were prepared by cutting the samples using a circular saw.

The samples were characterized by screw withdrawal strength (SR) (Fig.1(d)) according to the TS EN 320 standard and compressive strength (PS) (Fig.1(c)) according to TS EN 326. The physical characterization of the samples was carried out by water absorption (WA) according to TS 311, thickness swelling (TS) according to TS EN 325, and density analysis according to TS EN 314. The data obtained from the analyses were statistically analyzed using the SPSS software. Similarities between samples and groups were determined using One-way Anova and Duncan’s analysis.

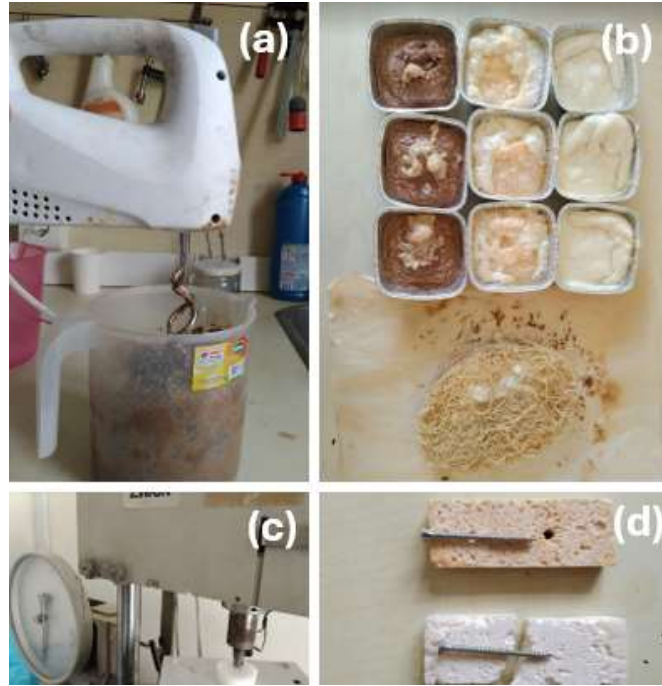


Fig. 1 Preparing composites (a) UF was mixed with filler, (b) molded and hardened, (c) pressure test, (d) screw withdrawal test

III. RESULTS AND DISCUSSION

The results of the analysis are presented in Table 1. When the WA of the samples was compared, it was seen that ÇP and ÇL were in the same group, but WF was 31% more than others. In contrast, MDF added sample was 50% lower than that of the other samples. Here, the densities can be effective in terms of the water absorption ability of the composites. It can be said that when the density increased, WA was negatively affected. This means that a higher density resulted in less WA. When the Fig. 2 was examined, it can be said that the WA24h was 10-15% higher than WA2h. When it was considered that WA24h had 22 h more immersed water time, this increase was not significant.

As shown in Fig. 4, the densities increased from the ÇL direction to the WF, ÇP, and MDF directions. There was a 76% difference in density between the highest and lowest. The difference was statistically significant. Prepared samples had moisture content due to UF. Because UF was 65% concentration that it has 35% water. This water concentration affected the composite's densities because Water boiled at above 100 °C and foamed the matrix of composites. As matrix foamed the densities decreased because of more void in matrix.

Table 1. Analysis results

Samples	WA2h (%)	WA24h (%)	TS2h (%)	TS24h (%)	Density (kg/m ³)	SR (Newton)	PS (Newton)
ÇP	51 (±4) B	56 (±4) B	3,0 (±0,5) A	3,8 (±0,8) A	595 (±28) B	1598 (±106) C	3747 (±595) A
ÇL	52 (±2) B	60 (±2) B	2,4 (±1,2) A	3,6 (±2,1) A	473 (±32) A	730 (±38) B	9630 (±160) C
MDF	23 (±1) A	36 (±2) A	2,7 (±0,8) A	3,8 (±1) A	740 (±8) B	2169 (±147) D	7412 (±510) B
WF	68 (±5) C	71 (±5) C	4,9 (±0,7) B	5,7 (±0,7) A	514 (±44) A	0 (±0) A	3300 (±483) A

MDF filler caused to same thickness swelling according to ÇP and ÇL. There was 12-50% difference between in the TS2h and TS24h. This TS tolerance was acceptable in term of the thickness swelling of board standard. When the TS results were compared according to composites densities, it can be said that the density and TS were not relevance each other. Because although the ÇL was low density (473 kg/m^3), its TS was lower than other samples whose densities were higher than ÇL.

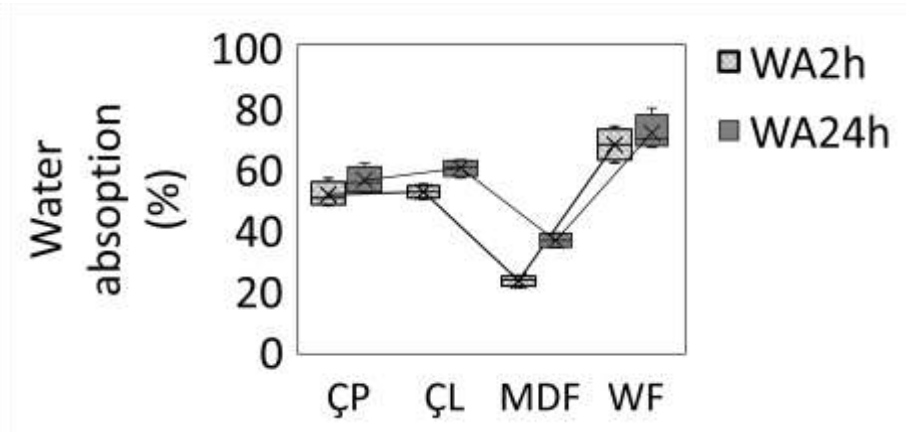


Fig.2 Water absorption (WA2h and WA24h) results of composites

When the Fig. 3 was examined, it can be seen that the ÇL has more TS standard deviation than other (ÇP, MDF and WF) samples. ÇL was fiber filler but others were particle fillers. This situation can be caused by filler morphologies that is ÇL has more standards deviation. WF was the sample which had the most TS and the ÇL was the sample which had the less TS. It can be said that WF had so little particle size (0,2-1 mm) and had more surface area than other fillers. Surface area of the particle increased as the particle size decreased. Thus, WF absorb more water thanks to its surface where larger other sample. Same result obtained from the WA. WF filled samples had more WA.

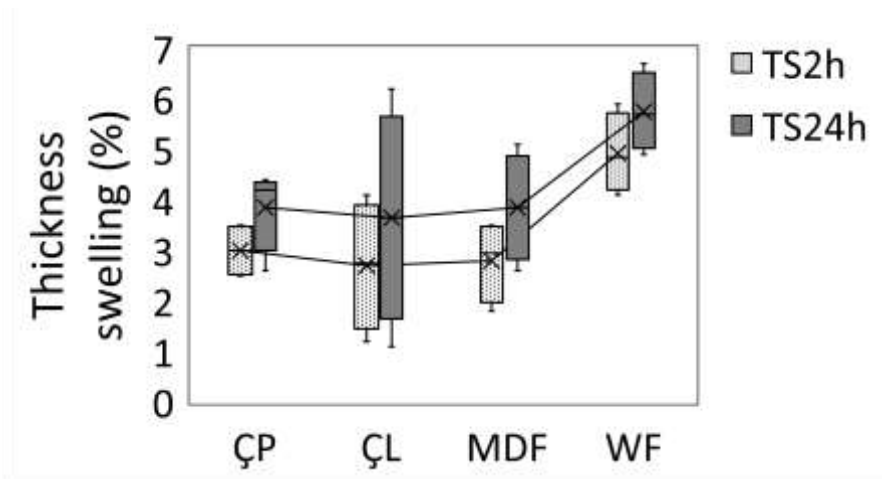


Fig. 3 Thickness swelling (TS2h and TS24h) results of composites

Density is important properties of the composites because of its placed fields. So that researchers studied to obtain most light and most durable composites. Also, we aimed to obtain low density and stronger composites without pressure. Fig. 4 show that the most suitable sample for ÇL because it has low density but more SR and PS strength.

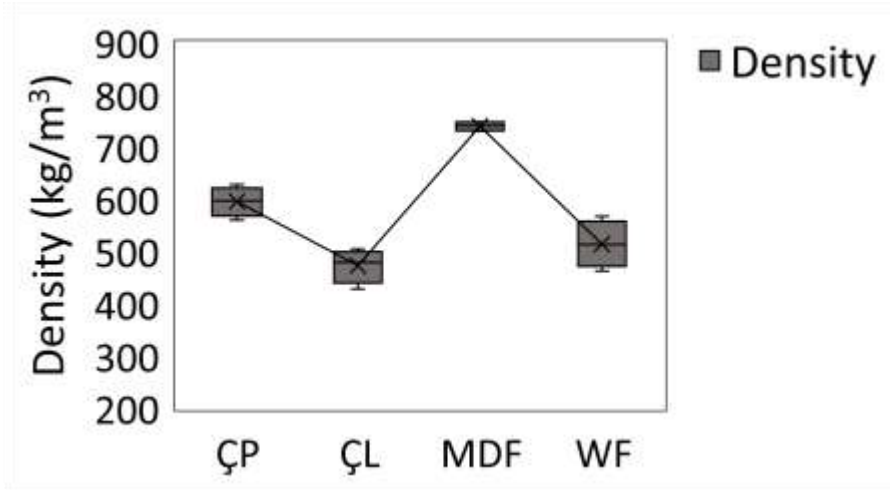


Fig. 4 Density of samples

Consumers prefer the light and strong composites. Thus, wood composites must be low density and durable. Fig. 5 show that ÇL has most PS strength. WF less than others in term of both PS and SR. ÇL fibers were obtained by cutting black pine wood in the radial direction with a band saw. This method is not widely used in timber workshops. This method was cut in the radial direction specifically for this study. As a result, the fibers obtained by chipping the wood in the radial direction using a band saw had very good mechanical resistance because it has low density and high durability. The SR strength of the WF added samples was zero. WF samples were very brittle. The screw split in two as soon as it entered the composite. In this regard, it can be concluded that using high amounts of WF in wood glues will reduce the screw holding strength.

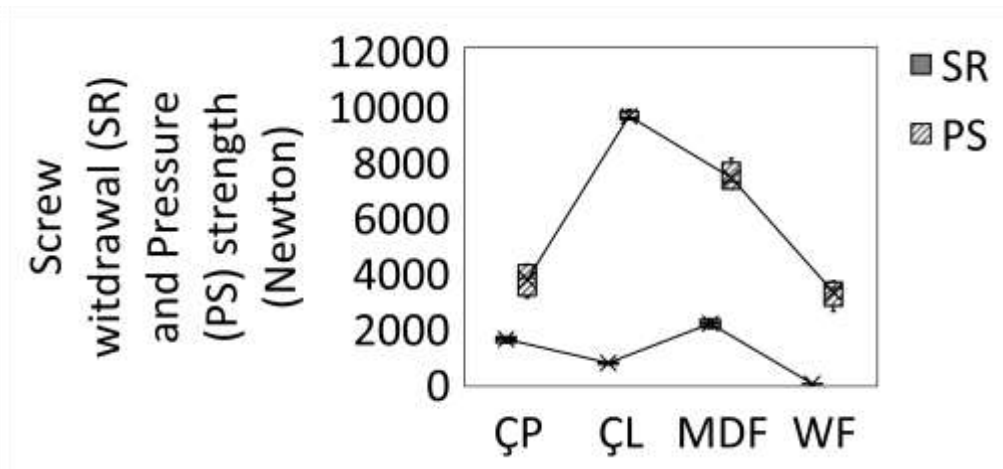


Fig. 5: Pressure (PS) and Screw withdrawal strength (SR) of the samples

IV. CONCLUSION

In the study, urea formaldehyde was cured in hot air without using pressure. Organic fillers (ÇP, ÇL, WF and MDF) were added before curing. In this method, the natural filler filled urea formaldehyde was used in the production of composites without much effort (without using pressure). The composites obtained by this method can be coated on their surfaces using wooden veneers or laminate veneers and used in furniture production. Once the surfaces and edges are coated, formaldehyde emission levels will also decrease. In this form, it will be close to the formaldehyde emission value of MDF and particleboard in the place of use.

It can be said that ÇL has the best mechanical and physical properties among the samples produced in the study. ÇL is a UF composite reinforced with pine wood fiber. Pine fibers were obtained by cutting the wood in a radial direction with a band saw. Normally, there is no such cutting method. This method was

specifically used to obtain pine wood chips in the form of fibers. As a result, it was determined that it had better mechanical strength and physical properties than other particle-filled samples. Wood chips obtained in fiber form can also be used in the production of different wood composites. It is thought that much better boards can be produced, especially by shaping wood fibers treated with UF using heat and pressure.

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REFERENCES

- [1] V. Naik, M. Kumar, and V. Kaup, "A Review on Natural Fiber Composite Material in Automotive Applications," *Engineered Science*, 2021.
- [2] M. Syduzzaman, M. A. Al Faruque, K. Bilisik, and M. Naebe, "Plant-Based Natural Fibre Reinforced Composites: A Review on Fabrication, Properties and Applications," *Coatings*, vol. 10, no. 10, p. 973, Oct. 2020.
- [3] M. Chanes de Souza, I. Moroz, I. Cesarino, A. L. Leão, M. Jawaid, and O. A. Tilton Dias, "A Review of Natural Fibers Reinforced Composites for Railroad Applications," *Applied Science and Engineering Progress*, Mar. 2022.
- [4] Y. K. Kim and V. Chalivendra, "Natural fibre composites (NFCs) for construction and automotive industries," in *Handbook of Natural Fibres*, Elsevier, 2020, pp. 469–498.
- [5] V. Chauhan, T. Kärki, and J. Varis, "Review of natural fiber-reinforced engineering plastic composites, their applications in the transportation sector and processing techniques," *Journal of Thermoplastic Composite Materials*, vol. 35, no. 8, pp. 1169–1209, Aug. 2022.
- [6] R. V. Patel, A. Yadav, and J. Winczek, "Physical, Mechanical, and Thermal Properties of Natural Fiber-Reinforced Epoxy Composites for Construction and Automotive Applications," *Applied Sciences*, vol. 13, no. 8, p. 5126, Apr. 2023.
- [7] S. Avramidis, C. Lazarescu, and S. Rahimi, "Basics of Wood Drying," 2023, pp. 679–706.
- [8] N. M. Nurazzi, M. R. M. Asyraf, M. Rayung, M. N. F. Norrahim, S. S. Shazleen, M. S. A. Rani, A. R. Shafi, H. A. Aisyah, M. H. M. Radzi, F. A. Sabaruddin, R. A. Ilyas, E. S. Zainudin, and K. Abdan, "Thermogravimetric Analysis Properties of Cellulosic Natural Fiber Polymer Composites: A Review on Influence of Chemical Treatments," *Polymers*, vol. 13, no. 16, p. 2710, Aug. 2021.
- [9] M. Asim, M. T. Paridah, M. Chandrasekar, R. M. Shahroze, M. Jawaid, M. Nasir, and R. Siakeng, "Thermal stability of natural fibers and their polymer composites," *Iranian Polymer Journal*, vol. 29, no. 7, pp. 625–648, Jul. 2020.
- [10] A. M. Radzi, S. Zaki, M. Hassan, R. A. Ilyas, K. Jamaludin, M. Daud, and S. Aziz, "Bamboo-Fiber-Reinforced Thermoset and Thermoplastic Polymer Composites: A Review of Properties, Fabrication, and Potential Applications," *Polymers*, vol. 14, no. 7, p. 1387, Mar. 2022.
- [11] J. S. Renner, R. A. Mensah, L. Jiang, Q. Xu, O. Das, and F. Berto, "Fire Behavior of Wood-Based Composite Materials," *Polymers*, vol. 13, no. 24, p. 4352, Dec. 2021.
- [12] P. Samanta, A. Samanta, C. Montanari, Y. Li, L. Maddalena, F. Carosio, and L. A. Berglund, "Fire-retardant and transparent wood biocomposite based on commercial thermoset," *Composites Part A: Applied Science and Manufacturing*, vol. 156, p. 106863, May 2022.
- [13] S. Turayev, X. Tychiyev, T. Sardor, X. Yuldashev, and M. Maxsudov, "The importance of modern composite materials in the development of the automotive industry," *Asian Journal of Multidimensional Research (AJMR)*, vol. 10, no. 3, pp. 398–401, 2021.
- [14] M. Y. Khalid, Z. U. Arif, W. Ahmed, and H. Arshad, "Recent trends in recycling and reusing techniques of different plastic polymers and their composite materials," *Sustainable Materials and Technologies*, vol. 31, p. e00382, Apr. 2022.
- [15] S. M. Rangappa, S. Siengchin, J. Parameswaranpillai, M. Jawaid, and T. Ozbakkaloglu, "Lignocellulosic fiber reinforced composites: Progress, performance, properties, applications, and future perspectives," *Polymer Composites*, vol. 43, no. 2, pp. 645–691, Feb. 2022.
- [16] D. Gay, *Composite Materials*. Boca Raton: CRC Press, 2022.
- [17] M. Z. R. Khan, S. K. Srivastava, and M. K. Gupta, "A state-of-the-art review on particulate wood polymer composites: Processing, properties and applications," *Polymer Testing*, vol. 89, p. 106721, Sep. 2020.
- [18] J. Agarwal, S. Sahoo, S. Mohanty, and S. K. Nayak, "Progress of novel techniques for lightweight automobile applications through innovative eco-friendly composite materials: A review," *Journal of Thermoplastic Composite Materials*, vol. 33, no. 7, pp. 978–1013, Jul. 2020.
- [19] C. N. Njoku, W. Bai, I. O. Arukalam, L. Yang, B. Hou, D. I. Njoku, and Y. Li, "Epoxy-based smart coating with self-repairing polyurea-formaldehyde microcapsules for anticorrosion protection of aluminum alloy AA2024," *Journal of Coatings Technology and Research*, vol. 17, no. 3, pp. 797–813, May 2020.
- [20] R. Tavernier, L. Granado, G. Foyer, G. David, and S. Caillol, "Formaldehyde-Free Polybenzoxazines for High Performance Thermosets," *Macromolecules*, vol. 53, no. 7, pp. 2557–2567, Apr. 2020.
- [21] L. Kristak, P. Antov, P. Bekhta, M. A. R. Lubis, A. H. Iswanto, R. Reh, J. Sedliacik, V. Savov, H. R. Taghiyari, A. N. Papadopoulos, A. Pizzi, and A. Hejna, "Recent progress in ultra-low formaldehyde emitting adhesive systems and

- formaldehyde scavengers in wood-based panels: a review,” *Wood Material Science & Engineering*, vol. 18, no. 2, pp. 763–782, Mar. 2023.
- [22] T. Ren, Y. Wang, N. Wu, Y. Qing, X. Li, Y. Wu, and M. Liu, “Degradation of urea-formaldehyde resin residues by a hydrothermal oxidation method into recyclable small molecular organics,” *Journal of Hazardous Materials*, vol. 426, p. 127783, Mar. 2022.
- [23] C. Rosenfeld, J. Konnerth, W. Sailer-Kronlachner, T. Rosenau, A. Potthast, P. Solt, and H. W. G. van Herwijnen, “Hydroxymethylfurfural and its Derivatives: Potential Key Reactants in Adhesives,” *ChemSusChem*, vol. 13, no. 20, pp. 5408–5422, Oct. 2020.
- [24] M. Hassanpour, “A Review of Four Kinds of Resin Production Technologies Based On Recent Developments,” *International Journal of Industrial Engineering*, vol. 8, no. 2, pp. 1–12, Jun. 2021.
- [25] C. Kumar and W. Leggate, “An overview of bio-adhesives for engineered wood products,” *International Journal of Adhesion and Adhesives*, vol. 118, p. 103187, Oct. 2022.
- [26] M. H. Hussin, N. H. Abd Latif, T. S. Hamidon, N. N. Idris, R. Hashim, J. N. Appaturi, N. Brosse, I. Ziegler-Devin, L. Chrusiel, W. Fatriasari, F. A. Syamani, A. H. Iswanto, L. S. Hua, S. S. A. O. Al Edrus, W. C. Lum, P. Antov, V. Savov, M. A. Rahandi Lubis, L. Kristak, R. Reh, and J. Sedliačik, “Latest advancements in high-performance bio-based wood adhesives: A critical review,” *Journal of Materials Research and Technology*, vol. 21, pp. 3909–3946, Nov. 2022.
- [27] J. Chrobak, J. Howska, and A. Chrobok, “Formaldehyde-Free Resins for the Wood-Based Panel Industry: Alternatives to Formaldehyde and Novel Hardeners,” *Molecules*, vol. 27, no. 15, p. 4862, Jul. 2022.
- [28] A. Pizzi and C. C. Ibeh, “Aminoresins,” in *Handbook of Thermoset Plastics*, Elsevier, 2022, pp. 65–82.
- [29] D. Moutousidis, K. Karidi, E. Athanassiadou, E. Stylianou, N. Giannakis, and A. Koutinas, “Reinforcement of urea formaldehyde resins with pectins derived from orange peel residues for the production of wood-based panels,” *Sustainable Chemistry for the Environment*, vol. 4, p. 100037, Dec. 2023.
- [30] M. Nazerian, M. Akbarzade, P. Ghorbanzad, A. N. Papadopoulos, E. Vatankhah, D. Foti, and M. Koosha, “Optimal Modified Starch Content in UF Resin for Glulam Based on Bonding Strength Using Artificial Neural Network,” *Journal of Composites Science*, vol. 6, no. 10, p. 279, Sep. 2022.
- [31] B. Zhang, S. Jiang, G. Du, M. Cao, X. Zhou, Z. Wu, and T. Li, “Polyurea-formaldehyde resin: a novel wood adhesive with high bonding performance and low formaldehyde emission,” *The Journal of Adhesion*, vol. 97, no. 5, pp. 477–492, Apr. 2021.
- [32] S. Park, B. Jeong, and B.-D. Park, “A Comparison of Adhesion Behavior of Urea-Formaldehyde Resins with Melamine-Urea-Formaldehyde Resins in Bonding Wood,” *Forests*, vol. 12, no. 8, p. 1037, Aug. 2021.
- [33] S. C. Sahoo, A. Sil, and R. T. Solanki, “Effect of adhesive performance of liquid urea formaldehyde (UF)resin when used by mixing with solid UF resin for manufacturing of wood based panels,” *International Journal of Scientific and Research Publications (IJSRP)*, vol. 10, no. 4, p. p10065, Apr. 2020.