Evaluation of Rice Husk Composite Boards Prepared Using Different Adhesives and Processing Methods

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Rice husks, a byproduct of rice milling, were used to develop composite boards. Different processing methods, grinding, and treating with sodium hydroxide (NaOH), were adopted to improve the bonding and mechanical performance. NaOH solution was prepared at 5% (wt/v) concentration. The effect of different adhesives, phenol formaldehyde (PF), and polymeric diphenylmethane diisocyanate (pMDI), was evaluated. Rice husks mixed with resin were hot pressed to the target density of 768 kg/m^3 and thickness of 12.7 mm. Specimens cut from these flat panels were submitted to bending, internal bond, water absorption, and thickness swelling tests. Results revealed that chemical treatment with NaOH significantly improved fiber-to-fiber bonding of rice husks. Internal bond strength of specimens made from chemically treated rice husks increased at least 1000% compared to others. Considering the mechanical properties and water uptake, rice husk boards fabricated with unprocessed rice husks and pMDI showed a better performance.

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INTRODUCTION

Rice, a staple food for nearly half of the world's population, plays a vital role in global dietary habits. Its importance in ensuring human nutrition cannot be overstated, given that it is grown in more than 100 countries and on all continents except for Antarctica (Sanint *et al*. 2004). The Food and Agriculture Organization (FAO) forecast in 2023/24 world rice production will be an astounding 523.7 million tons (milled basis) (Food and Agriculture Organization, n.d.), highlighting its enormous agricultural and economic significance, amounting to a global rice industry estimated current value of $287.45 billion (SGS 2022). The handling of the trash produced during the rice production cycle is one area where this vast output has created several complicated issues.

In the process of cultivating and processing rice, significant quantities of waste are generated, encompassing components like rice straw and rice husk. The rice husk, an outer layer of the rice seed, is discarded during processing, resulting in large volumes of rice husk being produced. Remarkably, despite its prevalence, rice husk is often not effectively repurposed within the industries where it originates. This leads to additional costs for its management and disposal.

One intriguing aspect of rice husk is its peculiarly slow decomposition, which can be attributed to its silica content of approximately 20% (Sun and Gong 2001). This slow decomposition poses challenges for conventional methods of waste management such as composting or manure. The accumulation of rice husk becomes especially problematic due to its low nutritional value, slow decomposition rate, and the environmental challenges associated with its disposal. This issue is further compounded by the fact that rice husks are used as sources of energy, while the byproduct — rice husk ash (RHA)—often remains underutilized, leading to significant environmental threats as it accumulates.

Many nations have put limitations on field-burning operations in response to the environmental and health risks associated with conventional rice farming methods (Mansaray *et al.* 1999). Such open burning has far-reaching effects, generating a variety of pollutants such as carbon dioxide, carbon monoxide, unburned carbon, nitrogen oxides, and other particulates such as PM2.5. Black carbon, a component of PM2.5 or "soot", has a 460 to 1500 stronger impact on global warming than CO_2 (Kaushal 2022). Researchers have been challenged to identify novel answers for these urgent problems as concerns over environmental preservation, energy conservation, and sustainable economic growth continue to grow on a worldwide scale.

A paradigm shift has evolved in response to these worries, moving away from the outmoded practice of disposing of waste and toward a more comprehensive "waste to resource" strategy. By transforming rice straw, rice husk, and rice husk ash into useful resources rather than throwing them away, this process aims to release their hidden potential, leading to a sustainable future. It is feasible to change the way we see and handle these crucial aspects of rice production by combining innovative technology, strategic planning, and diligent environmental stewardship. So far efforts have been made to upcycle rice husk. Some studies have described the incorporation of rice husk in materials, particularly in thermal insulation (Yarbrough *et al.* 2005), plastic composites (Choi *et al.* 2006), lightweight concrete (Chabannes *et al.* 2014), and insulation composite panels (Antunes *et al.* 2019).

Silica makes up the outside of rice husks, resulting in a natural silicon-cellulose membrane (Yoshida *et al.* 1962) that serves as a barrier against termites and bacteria. However, this silica component has been acknowledged as a likely cause of the limited interaction between functional groups on the surface of rice husks and other binding agents. This results in a weak bonding between rice husks and binding substances (Vasishth 1971; Ndazi 2001), which negatively impact the material properties of the final product. Because of this, rice husks have not been utilized as much as other cellulose fibers.

The elimination of silica and other surface pollutants is expected to improve the adhesion qualities between rice husks and binders, which would subsequently improve comprehensive composite features. To effectively remove surface contaminants, scientists have investigated the chemical alteration of rice husks using alkali pre-treatment. A well-proven approach that has been shown to strengthen the binding interface between fibers and matrix is the alkaline treatment of cellulosic fibers using sodium hydroxide (NaOH) (Mohanty *et al.* 2000; Ndazi *et al.* 2007; Syafri *et al.* 2011; Ab Ghani *et al.* 2015; Emdadi *et al.* 2015; Bisht and Gope 2018). The use of diluted KOH solution at 1.0% improved the bond strength of O-Si-O of the silica and reduced the amorphous silica quantity (Emdadi *et al.* 2015).

This research investigated the effect of different processing methods and adhesives on the bonding quality of rice husk material to develop high-performance composite panels. The effects and performance of rice husk composite panels were evaluated through different experiments.

EXPERIMENTAL

Processing

The raw materials, rice husks, were provided by a grain elevator located in the United States inside of Mississippi's Delta region. Chemical treatment with NaOH and mechanical treatment by grinding rice husks were adopted in this study. For alkaline treatment, sodium hydroxide was added to hot water (93 °C) to make a 5 % wt/v NaOH solution. Rice husks were soaked in NaOH solution (4.54 kg (10 lbs) rice husks per 70- L solution) for 1 hour, while the solution was kept at 93 °C (200 °F). To easily submerge and avoid floating, rice husks were placed inside fabric bags before soaking in the solution. After one hour, treated rice husks were taken out and washed with water three to four times to reach a neutral PH, around 7, and then they were dried inside an oven to a target moisture content (MC) of 3 to 10%. Besides alkaline treatment, rice husks were mechanically processed using a grinder with a 3.2-mm (0.125-in.) mesh screen. Therefore, alkaline-treated, ground, and unprocessed (untreated and unground) rice husks as shown in Fig. 1 were the raw materials used in this study.

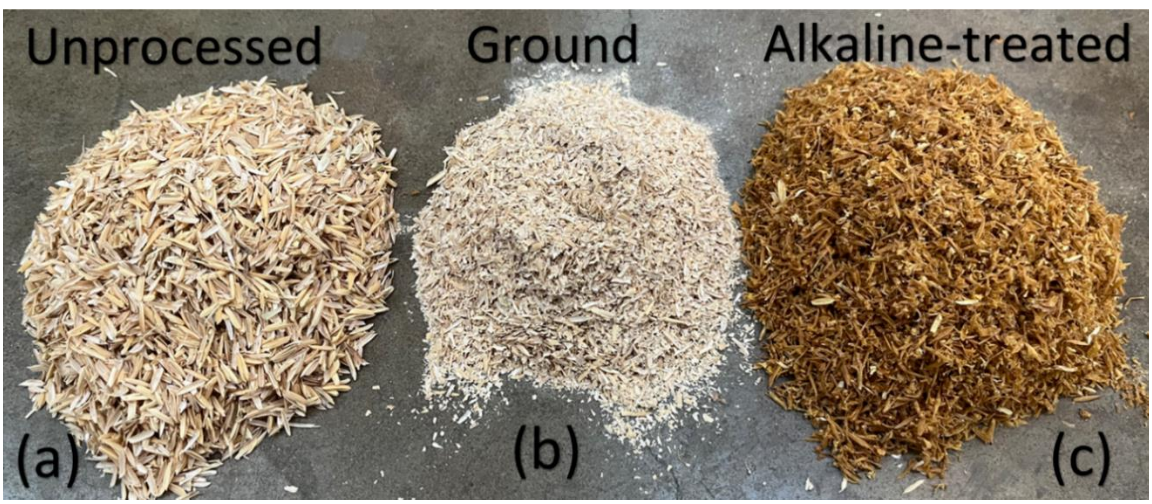

Fig. 1. Raw materials: (a) unprocessed, (b) ground, and (c) alkaline treated rice husks

Panel Fabrication

Phenol formaldehyde (PF) and polymeric methylene diphenyl diisocyanate (pMDI) resins were used as binders. Raw materials were placed inside a drum blender and the resin was sprayed to a target resin content of 7% for PF and 5% for pMDI based on the oven-dry mass of raw materials. Rice husks mixed with resin were placed inside a forming box to make a 787 x 864 mm (31 x 34 in.) pre-pressed mat. The pre-pressed mat was hot-pressed at 160 °C for 5 min to a target thickness of 12.7 mm (0.5 in.) and target density of 767 kg/m^3 (48pcf). Four different panels were made in this study, one per group.

Table 1. Raw Materials and Type of Resins used to Make Different Panels

Panel #	Rice Husks Form	Raw Materials MC	Resin	Resin Content
1	Unprocessed	3-5%	PF	7%
2	Ground	3-5%	PF	7%
3	Alkaline treated	3-5%	PF	7%
4	Unprocessed	8-10%	pMDI	5%

Details regarding MC of raw materials, type of resin, and resin contents are given in Table 1. The first panel, unprocessed rice husks with PF resin, served as control. As shown in Fig. 2a, the processing method could be easily identified by visual inspection. Panels were conditioned to the target MC of 8% before conducting experiments.

Experimental Testing

To evaluate the effect of different processing methods and two different resins on fiber bonding, an internal bond (IB) test was conducted. Since bending is a common load applied on many structures and products and generates different stresses (tensile, compressive, and shear), specimens cut from fabricated boards were submitted to a three-point bending test. The average total length for bending specimens was 356 mm (14 in.) while the span length was 305 mm (12 in.). To evaluate water performance of the fabricated panels, specimens cut from these boards were submitted to water absorption (WA) and thickness swelling (TS) test. ASTM D1037 was followed to conduct all these tests. Dimensions and average density of test specimens are given in Table 2.

Table 2. Average Density of Test Specimens Cut from Each Board

Test	# of Specimens*	Dimensions (mm)	Panel #1	Panel #2	Panel #3	Panel #4
IB	18	51x51	748	768	748	809
Bending	8	76x356	716	733	732	783
WA & TS	8	152x152	751	776	754	808
Average Density (COV%)			741 (3.32%)	762 (4.43%)	746 (2.33%)	802 (4.12%)

Note: Numbers in the parenthesis indicate the coefficient of variation. *This column quantifies the number of specimens cut from each board.

RESULTS AND DISCUSSION

Sieve analysis was conducted on ground rice husks using U.S. standard series no. 10, 14, 18, 20, 25, 30, 35, and 40. The results after 10 min of sieving are given in Fig. 2b.

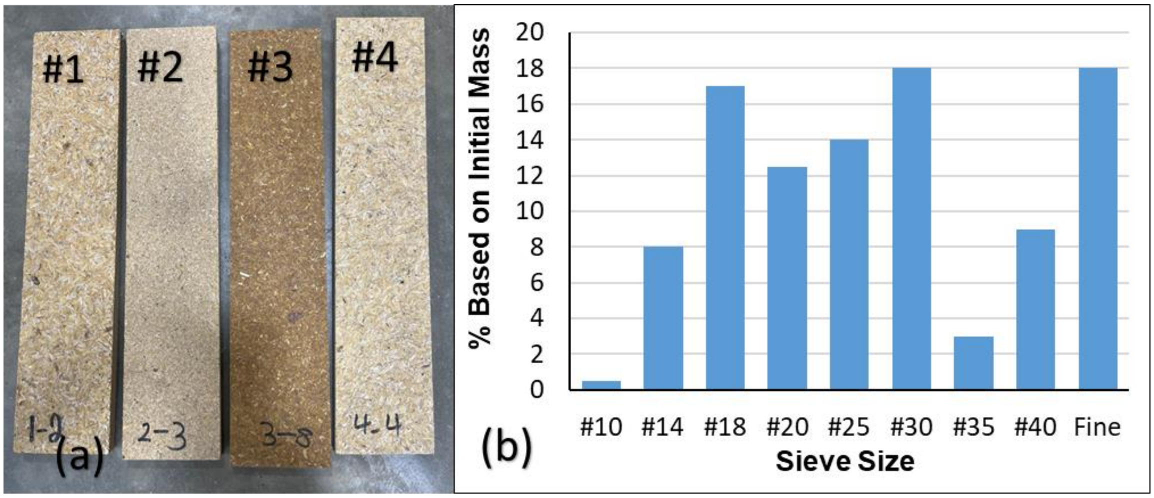

Fig. 2. (a) Fabricated rice husk boards: #1: unprocessed rice husks+PF, #2: ground+PF, #3: treated+PF, and #4: unprocessed +pMDI, and (b) sieve analysis of ground rice husks.

This section is divided into three subsections to report the results of different tests conducted on rice husk composite boards.

Bending

Following ASTM D1037, the span length to thickness ratio of all bending specimens was 24. The results of bending test including bending modulus of elasticity (MOE) and modulus of rupture (MOR) are given in Fig. 3, where the type of raw materials (unprocessed, ground, and treated) and adhesives (PF and pMDI) are also specified. Specimens fabricated with pMDI resin showed the highest MOE compared to those fabricated by PF resin. Such a lower stiffness for specimens fabricated by PF resin shows that PF resin cured in these specimens remains more elastic compared to pMDI (Dziurka *et al*. 2005). Specimens made with ground rice husks and PF resin showed the lowest MOE. This low stiffness can be attributed to the fiber length, as rice husks have been ground to small pieces. Specimens made with alkaline-treated rice husks and PF resin had the highest bending strength (MOR). Such a high MOR is attributed to the fiber length and bonding performance. It shows that treating rice husks with NaOH improves their wettability and bonding performance while the fiber length is not affected. The MOR of specimens made with pMDI resin was about 41% lower than those made by treated rice husks and PF resin. Since the raw materials to make these two panels, unprocessed and treated rice husks, had almost the same fiber length, the difference comes from the strength of fiber-fiber bonds. Treated rice husks developed a better bonding with PF resin compared to unprocessed rice husks with pMDI resin. In contrast, pMDI resulted in a higher MOR, about 68%, compared to PF resin when the same raw materials, unprocessed rice husks, were used to fabricate panels. For raw materials with high silica content, unprocessed rice husks, pMDI results in a better wettability and bonding performance compared to PF resin (Dziurka *et al*. 2005, Pan *et al*. 2010). The low bending strength of specimens made with ground rice husks and PF resin is attributed to both fiber-fiber bonding and mainly fiber length.

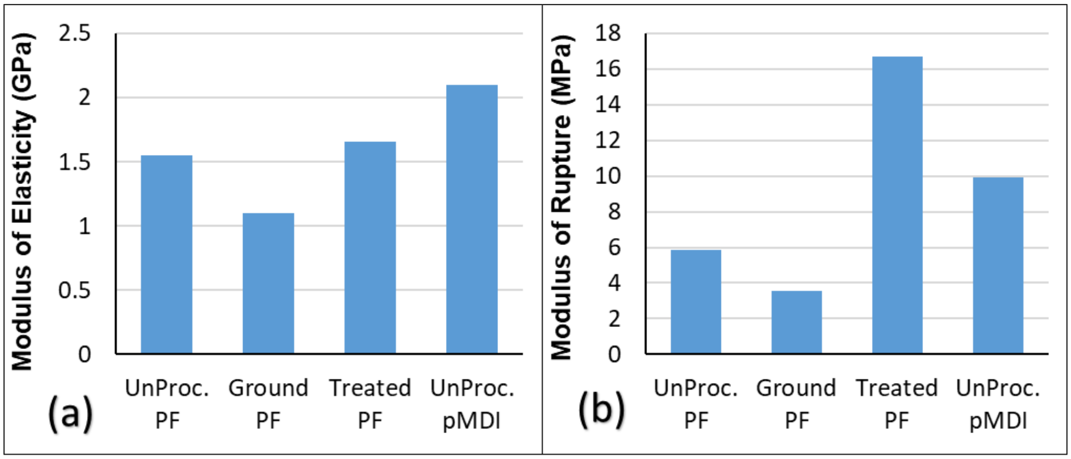

Fig. 3. Bending test results (a) modulus of elasticity and (b) modulus of rupture

Internal Bond

Specimens made by alkaline-treated rice husks and PF resin showed the highest internal bond strength; approximately 1025% and 1255% higher than those of unprocessed rice husks specimens made with PF and pMDI, respectively. This finding was consistent with that obtained from bending strength results shown in Fig. 3b. Such finding demonstrates that alkaline treatment with NaOH was effective to remove the silica and significantly improve the wettability and bonding performance of rice husks. Comparing the unprocessed rice husk boards, those made with PF resin had a higher IB strength compared to those made by pMDI resin. Since pMDI is an oil-based resin while PF is a water-based, a higher IB strength was expected for specimens made by pMDI resin rather than PF. In addition, this finding contradicts that obtained from bending strength results shown in Fig. 3b; pMDI resulted in a better bonding and thus a higher bending strength compared to PF resin for unprocessed rice husks specimens. A question can be asked regarding how to resolve this unexpected result and the inconsistent trend between bending and IB strength of unprocessed rice husks specimens made with PF and pMDI. Failure mode and generated stresses in these specimens could explain this difference. All IB specimens failed around the center, as shown in Fig. 4b.

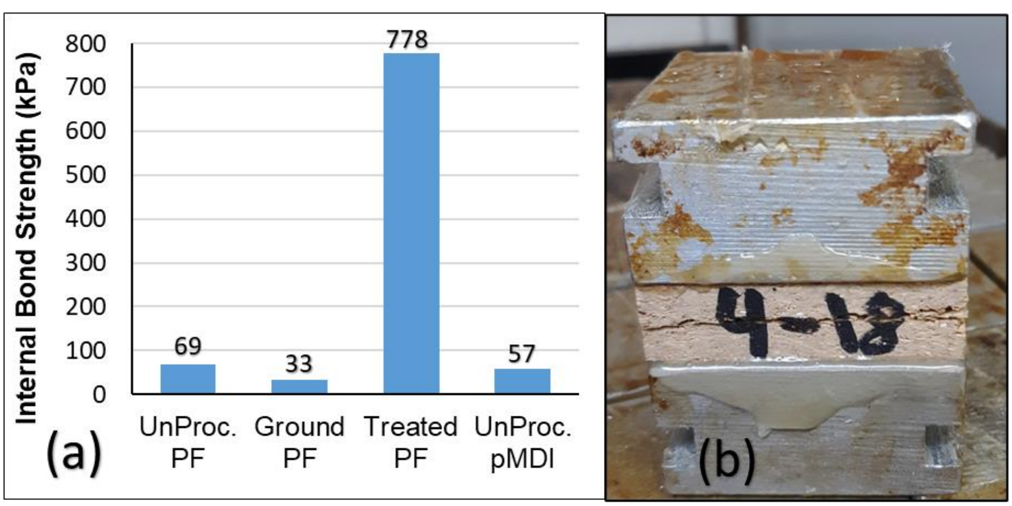

Fig. 4. (a) Internal bond strength of fabricated rice husk composite boards and (b) failure mode of IB specimens

The vertical density profiles of the board specimens showed a lower density and weaker region around the mid-plane, which is a common phenomenon in natural fiber-based composites. In contrast, all bending specimens failed at the bottom surface, where the maximum tensile stress occurs. Since span length to thickness ratio of bending specimens was high, 24, the shear stress generated in the bending specimens was negligible, while the normal stresses control the strength of these specimens. Considering the failure of IB and bending specimens, the present finding should be restated and revised to explain the unexpected result and different trends. For unprocessed rice husk boards, pMDI resulted in a better bonding in the outer surfaces, and in turn a higher bending strength, compared to PF resin, while PF resulted in a better bonding in the mid-plane, and in turn a higher IB strength, compared to pMDI resin. In other words, bonding strength between unprocessed rice husks with PF and pMDI is location-dependent and changes through the thickness of the panel. Thermal resistance of silica (Guo *et al.* 2010), a component with high content in rice husks, can explain this difference. The heat resistance aspect of silica reduces the thermal conductivity of the rice husks mat and delays the temperature rise in the mid layer. This affects the curing process and results in either precuring or incomplete curing around the mid-plane of the rice husks boards. Such an incomplete curing at the mid-layer reduced the IB strength of unprocessed rice husk boards. When the silica is removed through alkaline treatment, a high IB strength is achieved for those alkaline-treated specimens. The lower IB strength of unprocessed rice husk specimens made with pMDI resin compared to that of PF suggests that pMDI is more sensitive to low thermal conductivity of the mat and thus the incomplete curing was intensified. The incomplete curing could be avoided by reducing the thickness and allowing the mid-layer to reach the target temperature much faster. Specimens made with ground rice husks and PF resin showed the lowest IB strength. Besides the incomplete curing due to low thermal conductivity of the mat, small fiber length is another reason for such a low IB strength.

Water Absorption and Thickness Swelling

Results of water absorption (WA) and thickness swelling (TS) test results are given in Fig. 5. The most significant finding of this test is the very low WA and TS of specimens made by pMDI resin compared to those made by PF resin. This shows the effectiveness of pMDI to improve water resistance of the rice husk composite boards. This water resistance was attributed to highly cross-linked polyurea matrix produced because of the reaction between pMDI with both the moisture and hydroxyl groups in the unprocessed rise husks. The difference between WA specimens made by PF resin was not significant. Alkaline treatment that increased bending and IB strength of rice husk boards, had no effect on WA. However, the thickness swelling of alkaline-treated specimens made by PF resin was about 65% and 59% lower than unprocessed and ground rice husk board made with the same PF resin, respectively, after 24 h of immersion. Such a lower thickness swelling can be attributed to strong fiber-fiber bonding due to alkaline treatment and silica removal.

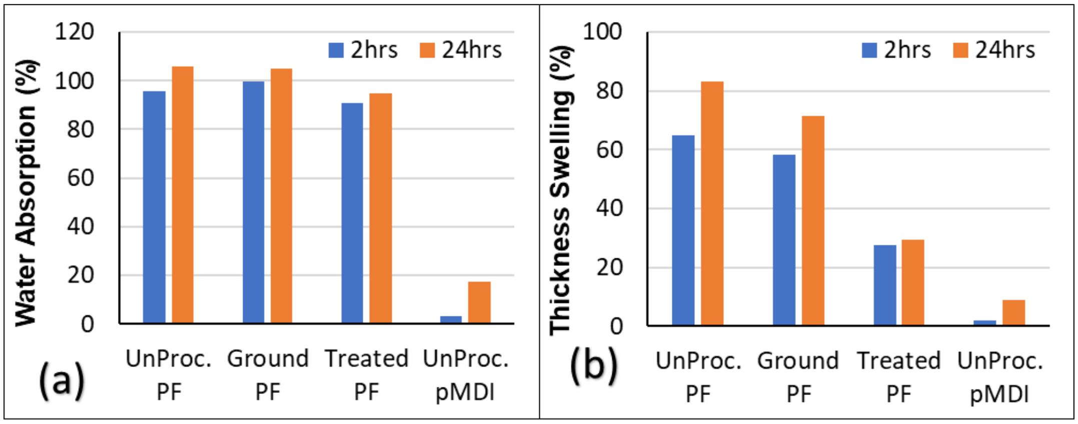

Fig. 5. (a) Water absorption and (b) thickness swelling results of rice-husks board

CONCLUSIONS

Silica content of around 20% in rice husks affects fiber-fiber bonding strength and also the curing process at regions far from the heat source due to its low thermal conductivity. These aspects render it challenging to develop high performance rice husk composite panels. In this study, two different processing methods, grinding and alkaline treatment with NaOH, and two different adhesives, phenol-formaldehyde (PF) and poly-(diphenylmethane diisocyanate) (pMDI), were adopted to find their influence on bonding performance and mechanical properties of rice husk composite boards. Test specimens were submitted to bending, internal bond, water absorption, and thickness swelling tests. Some of the findings are summarized as follows:

1. Specimens made with pMDI were stiffer than those made with PF. The effect of alkaline treatment of rice husks had a negligible effect on bending stiffness. However, grinding resulted in shorter fiber and low stiffness.
2. Fiber length and quality of fiber-fiber bonding controlled the bending strength. Therefore, alkaline treated specimens made by PF resin showed the highest bending strength, while ground rice husks bonded by PF resin showed the lowest bending strength.
3. Internal bond (IB) test results showed that alkaline treatment had a significant positive effect on bonding performance. Alkaline treated specimens had an IB strength over 1000% higher than other specimens.
4. The heat resistance property of silica influenced the curing process and bonding quality. At the surface where the heat can be easily transferred from the platens, pMDI developed a better bonding between unprocessed rice husks compared to PF. At the middle of the panel, where it takes more time to reach target temperature compared to the surfaces, PF developed a better bonding between unprocessed rice husks compared to pMDI. The heat transfer resistance property of silica negatively impacted the curing process of pMDI more than PF at the middle of the unprocessed rice husk boards.
5. The processing methods did not have a significant effect on the water performance of the rice husk panels. However, the effect of pMDI resin was more significant and positive.

Another important point that should not be neglected is the material loss during the alkaline treatment. It was found that around 50% of rice husks were lost during this process. Considering the water, energy, and time required for alkaline treatment, NaOH cost, and negative effect of releasing NaOH solution on the environment, the use of pMDI with no processing method is recommended to produce formaldehyde-free, eco-friendly, and cost-effective rice husk composites. However, alkaline-treated rice husks with pMDI, which was not tried in this study, has the potential to result in high-performance rice husk composites and is suggested as future study.

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