

WATER RESISTANCE AND SOME MECHANICAL PROPERTIES OF RICE STRAW FIBERBOARDS AFFECTED BY THERMAL MODIFICATION

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Rice straw fiberboard was made using 12 wt % urea-formaldehyde (UF) resin as binder and 1.0 wt % polymeric methylene diphenyl diisocyanate (pMDI) and 1.2 wt % wax emulsion as water retardants. The prepared fiberboards were heat-treated at 120, 150, 185, and 210°C in the presence of steam in a high-temperature dry kiln, respectively for 90 min. The effect of water retardants and heat treatment on the water resistant and some mechanical properties of the fiberboards were investigated. It was found that the water resistance of the rice straw fiberboard could not be improved by adding wax emulsion. The use of pMDI to the system significantly increased the interfacial strength and reduced 24-h thickness swelling (TS) compared to the boards with or without wax emulsion. After heat treatment, the TS was significantly decreased due to the decrease in the free reactive hydroxyl group content of rice straw fiber. Some mechanical properties of the fiberboards, such as the internal bonding strength, modulus of elasticity, and modulus of rupture were dramatically reduced with increasing temperature from 120°C to 210°C.

Key words: Rice straw; Fiberboard; Water resistance; Heat treatment; pMDI

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INTRODUCTION

In recent years, great quantities of agricultural (straw) residues, available worldwide, have been produced after harvest. For instance, approximately 0.76 billion tons of straw residues (mainly wheat and rice straw) will be produced in China in 2010 (Zhou and Mei 2000). The applications of straw residues in the production of composite panels (i.e. particleboards, fiberboards) are considered very attractive both from the economical and environmental point of view (Hua 2004). On the other hand, the utilization of agriculture fibers as an alternative to wood fiber for wood based panels can reduce the consumption of wood fiber and thus help protect forests and the environment. However, straw resources possess a high content of silica and wax on their surfaces, making them difficult to bond with urea-formaldehyde (UF) resin. Furthermore, when the straw is bonded with UF resin, the weak interfacial adhesion between the straw and UF resin has a negative influence on dimensional stability and the mechanical properties of the resultant straw panels. Therefore, a number of studies (Sun et al. 1995; Han et al. 2001a, 2001b; Leiva et al. 2007) have been concerned with treatments of the composite systems and their effects on the dimensional stability of straw panels.

A suitable system that includes both resin and water retardant is an attractive method to modify the dimensional stability of straw panels. It is suggested that polymeric methylene diphenyl diisocyanate (pMDI), with highly reactivity towards water and hydroxyl group of straw, is a suitable resin system. However, pMDI is not cost-effective. Han et al. (2001a) prepared the straw particleboards with reed/wheat straw, 12% UF resin, and silane coupling agent. For both reed and wheat boards, the internal bond strength and thickness swelling (TS) were significantly improved at up to 5% silane coupling agent content. Garcia et al. (2005) studied that the dimensional stability of MDF panel influenced by maleated polypropylene wax. This treatment could result in an important reduction in TS and water absorption (WA) after water soaking for panels produced from the treated fibers.

Wax has been used as a traditional water retardant for dimensional stability of wood and straw panels (Pan et al. 2006; Hizioglu et al. 2007; Xu et al. 2009). Two types of wax are utilized in processing of panels, i.e., molten and emulsified wax. Xu et al. (2009) found that wax-sizing affected both the dimensional stability and mechanical properties of bagasse particleboards. The introduction of wax could improve 24-h TS and WA for bagasse particleboards.

Heat treatment of panels is a more effective method to improve dimensional stability, as reported by many authors (Garcia et al. 2005; Boonstra et al. 2006; Paul et al. 2007; Mohebbi et al. 2008; Del Menezzi et al. 2009). It is observed that the thermal degradation of wood is affected by temperature. Between 25°C and 150°C, wood becomes dehydrated and generates water vapor and other noncombustible gases and liquids. From 150°C to 240°C, some components of wood begin to undergo significant pyrolysis. Much of the acetic acid liberated from wood pyrolysis is attributed to deacetylation of hemicelluloses. With the FTIR technique, the cell morphology and components of wood remain stable when exposed at 105-240°C for 0.5-5 hours (Madison 1970; Wu 1990). Moreover, with prolonged exposures at temperature lower than 50°C, the degree of polymerization of wood can become degraded significantly (Lin and Chuai 1999).

After heat treatment, strength properties of wood are considerably reduced (Kamdem et al. 2002; Gosselink et al. 2004). Chow (1971) found that the hydroxyl group content of white spruce was significantly decreased when treated at higher temperature. Furthermore, heat treatment on wood panels possibly results in the changes in the wood-adhesive bond network (Michiel et al. 2006). Some other studies (Inari et al. 2006, 2007a, b) have been reported changes in the chemical composition of wood occurring during heat treatment, using FTIR and XPS technology.

Although the effect of heat treatment on the properties of wood panels has been studied extensively, there is little information available on the effect of heat treatment on the water resistance and mechanical properties of straw panels. Therefore, this study as part of a comprehensive research project will examine the effects of water retardants, and heat treatment in the presence of steam atmosphere on some physical and mechanical properties of rice straw fiberboards.

EXPERIMENTAL

Raw Materials and Preparation

Rice straw was collected from a suburb near Nanjing City, China. The samples were first chipped into short pieces with a length range from 30 to 50 mm, using a chipper mill, and then softened in a pressure tank with a steam pressure of 1.2 bar. Afterwards, the softened rice straw chips were defibrillated by a refiner in the pilot plant of Engineering Research Center of Fast-growing Trees and Agri-fiber Materials, Nanjing. The refiner is equipped with a 300 mm disc powered by a 30 kW motor with a variable speed drive that can reach up to 2700 rpm. For this experiment, the plate gap was adjusted to 1.6 mm. The length of refined fiber ranged from 0.18 mm to 2.91 mm. The width of refined fiber ranged from 31.92 μm to 223.26 μm . The aspect ratio of refined fiber was 16.31. The fiber size distribution was determined by screen analysis as illustrated in Fig. 1.

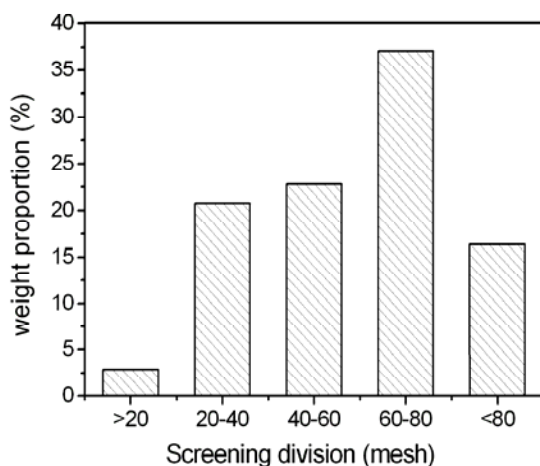


Fig. 1. Screening distribution of the rice straw fiber

The urea-formaldehyde resin (UF) with 64% solid content, and emulsion wax with 50% solid content used were kindly supplied by Anhui Kentier Wanhua Artificialboard CO, Ltd. The pMDI was kindly provided Huntsman.

Board Manufacturing and Heat Treatment

The refined fibers were kiln-dried to about 4.2wt% (dry matter content) before use. An amount of 1.2 wt% (based on UF resin) solid ammonium chloride was mixed into the liquid UF. The rice straw fiberboards were made with 12 wt% UF resin and 1.2 wt% emulsion wax or 1.0 wt% pMDI. To follow the heat treatment on fiberboards, two panels were repeatedly made for each run and, therefore, eighteen 300 \times 300 \times 10 mm³ panels were made in total. The panel characteristics are presented in Table 1.

The UF resin and water retardant was sprayed onto fibers using our laboratory blender. Then, the resinated fibers were manually formed into mats using a wooden frame. The pre-pressed mats were hot-pressed at 180°C for 5 min, including 1 min press

closing time to achieve the target thickness, 2 min polymerization time, and 2 min releasing time. The target density of boards was adjusted to 0.8g/cm³.

All panels were stored at 22°C and 65±1% relative humidity (RH) for one week. The effect of heat treatment on the rice straw fiberboards was carried out by heating the boards at temperatures of 120, 150, 185, and 210°C for 90 min, respectively, and kept in the dry kiln overnight, and then taken away. During this stage superheated steam was used as a sheltering gas to exclude oxygen (reducing fire risks and preventing undesired oxidation reactions). The constant parameters for heat treatment are also presented in Table 1. The weight percent loss (WPL) of the sample as a result of chemical degradation during heat treatment was calculated according to the Equation (1):

$$WPL(\%) = 100 \times \frac{m_0 - m_1}{m_0} \quad (1)$$

where, m_0 is the initial oven-dried mass of the board before heat treatment and m_1 the oven-dried mass of that after heat treatment.

Table 1. The Heat Treatment Conditions for Rice Straw Fiberboard

Run	Sample code	Water retardant	Heat temperature
1	C	no	20°C
2	C-185	no	185°C
3	W	1.2wt% wax emulsion	20°C
4	W-185	1.2wt% wax emulsion	185°C
5	P	1.0wt% pMDI	20°C
6	P-185	1.0wt% pMDI	185°C
7	W-120	1.2wt% wax emulsion	120°C
8	W-150	1.2wt% wax emulsion	150°C
9	W-210	1.2wt% wax emulsion	210°C

Property Evaluation of MDF Panel

After thermal modification, all panels were stored at 22 °C and 65±1% RH for one week again. In accordance with GB/T 11718-1999 (Chinese Standard Association 1999), two static bending (modulus of rupture, MOR and modulus of elasticity, MOE) specimens (250mm×50mm), four internal bonding strength (IB) specimens (50mm×50 mm), four 24-h thickness swelling (TS), and water absorption (WA) specimens (50 mm×50 mm) were cut from each panel and then tested. The density of each specimen for MOR/MOE, TS/WA, and IB were also measured before testing.

RESULTS AND DISCUSSION

Weight Loss

The weight percent loss (WPL) of rice straw fiberboards as a function of temperatures after heat treatment is shown in Fig. 2. The WPL value of rice straw fiberboards without water retardant was 6.33%. With addition of wax emulsion, the WPL after heat treatment at temperature of 185°C for the panel was 6.38%, slightly higher than

that of the pMDI-added panel. With increasing temperature from 150 to 210°C, the WPL was increased from 1.56 to 12.8% due to thermal degradation of the rice straw MDF panels. This means that higher temperature could accelerate thermal degradation of the main polymeric components (lignin, hemicelluloses, and cellulose) of rice straw fiber and UF resin. However the weight percent gained of panels was 0.47% at 120°C, indicating that a small quantity of moisture was absorbed by the panels.

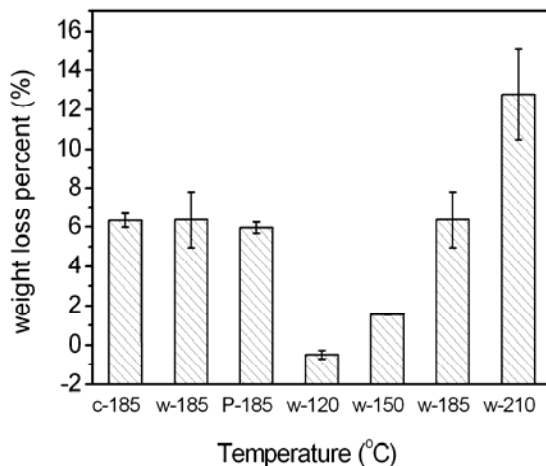


Fig. 2. Weight loss of rice straw fiberboard affected by thermal modification

Twenty-Four-Hour Water Absorption

24-h water absorption (WA) of control fiberboards was as high as 90%, no matter whether they had been heat treated or not heat treated. It was effectively reduced by addition of pMDI into the board, compared with addition of emulsion wax, as shown in Fig. 3. The WA of pMDI-treated panels decreased 60.7% before heat treatment at 185°C, and 54.7% after heat treatment. Significant reduction occurred in the WA of the pMDI-treated panels, due to the reaction between N=C=O functional group for pMDI and hydroxyl groups of rice straw fiber (Liu et al. 2007). By comparing the WA of panels with the same composition, this was not affected by the heat treatment.

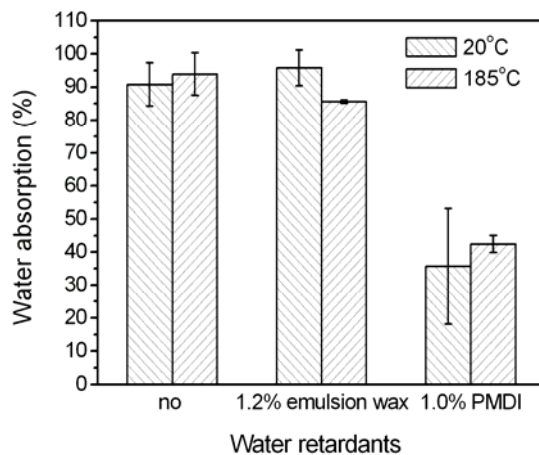


Fig. 3. WA of rice straw fiberboard affected by water retardants with heat treatment at 185°C

Figure 4 shows that the WA of MDF panels produced with addition of 1.2wt% of emulsion wax was affected by the temperature of heating. At temperatures of 120 and 150°C, the WA of the panels was higher than for untreated panels. By increasing the temperature from 185 to 210°C, the WA gradually decreased from 85.6% to 47.8%.

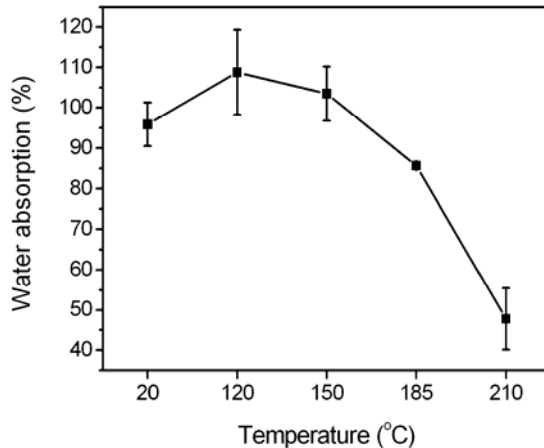


Fig. 4. WA of rice straw fiberboard affected by heat treatment

Twenty-Four-Hour Thickness Swelling

24-h thickness swelling (TS) of rice straw fiberboard, as affected by different water retardant with heat treatment at 185°C, is presented in Fig. 5. It is clearly shown that adding pMDI to the whole system dramatically reduced the TS to 17.44%, which is attributed to good interfacial strength between rice straw fiber and UF resin. This good interfacial strength could result in a reduction of free accessible hydroxyl groups of rice straw fiber. However, the MDF panel with wax emulsion had 31.38% TS, which implies that adding wax emulsion could not decrease TS.

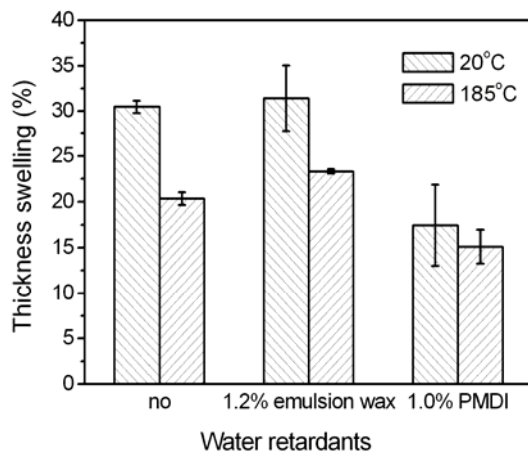


Fig. 5. TS of rice straw fiberboard affected by water retardants with heat treatment at 185°C

Figure 6 shows that the TS of panels was influenced by temperature of heating. The TS of the treated panels decreased linearly with temperature up to 210°C. The value

12.71% implies that the reduction was approximately 50% compared to the untreated panels. The decrease of TS could be related to the thermal degradation of rice straw fiber during heat treatments. As described in previous literature (Pan et al. 2009), degradation of hemicelluloses during heat treatment is generally more severe, compared to cellulose and lignin. Hemicelluloses are complex mixture of polysaccharides, each possessing free accessible hydroxyl groups and exhibiting hygroscopicity. The degradation of hemicelluloses significantly reduces the amount of free accessible hydroxyl groups, and increases the dimensional stability of the panel (Mohebbi et al. 2008).

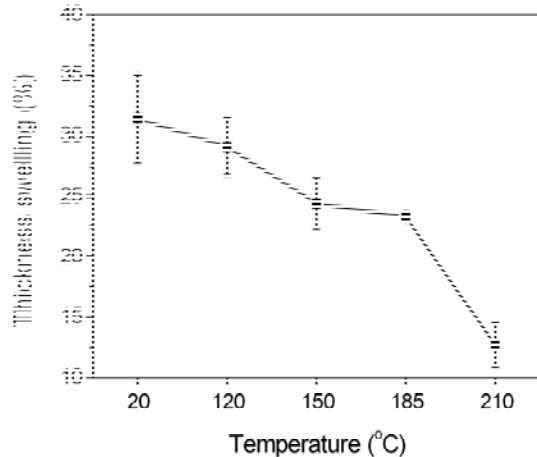


Fig. 6. TS of rice straw fiberboard affected by heat treatment

Internal Bonding Strength

Figure 7 shows that the internal bonding strength (IB) of rice straw fiberboard was affected by different water retardant with heat treatments at 185°C. Adding pMDI to the whole system led to an increase of IB from 0.62 to 1.00 MPa. The improvement of the IB of panels made of pMDI-treated fibers could be explained by a better adhesion in the system. Therefore, it seems that pMDI can act as a coupling agent for rice straw fiber and UF, thus improving the wettability of UF resin on the rice straw fiber surfaces.

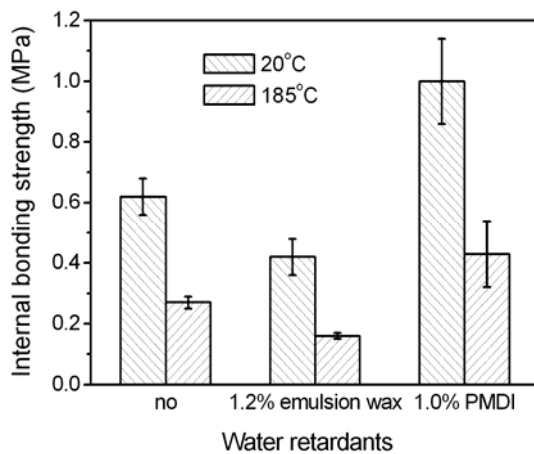


Fig. 7. IB of rice straw fiberboard affected by water retardants with heat treatment at 185°C

Figure 8 shows that the IB of rice straw fiberboard was affected by heating. By increasing the temperature from 120 to 210°C, the IB of panels gradually decreased from 0.31 to 0.09 MPa. Within the range the temperatures of 120-150°C, the IB was decreased by 28.5%, while as the temperature increased to 185°C, the IB reduced dramatically by 61.9%. Thermal degradation of rice straw components and UF resin occurred, and interaction of rice straw fiber with UF resin were mainly responsible for IB reduction. It is noted that heat treatments result in wettability changes associated with a decrease of hygroscopicity of rice straw fiber (Inari et al. 2007). UF resin is a polar adhesive, it initially needs to wet the fiber for bonding, and it is also able to bond materials with the same polarities (Mohebbi et al. 2008). Wettability changes of rice straw fiber lower adhesion strength between fiber and UF resin. Furthermore, the strength of single fiber could be reduced due to the thermal degradation of rice straw fiber. By holding rice straw fiberboard at 210°C, darkly charred boards could be observed after heat treatment. The board surface became darker and this characteristic was correlated with the lower IB (Del Menezzi et al. 2009).

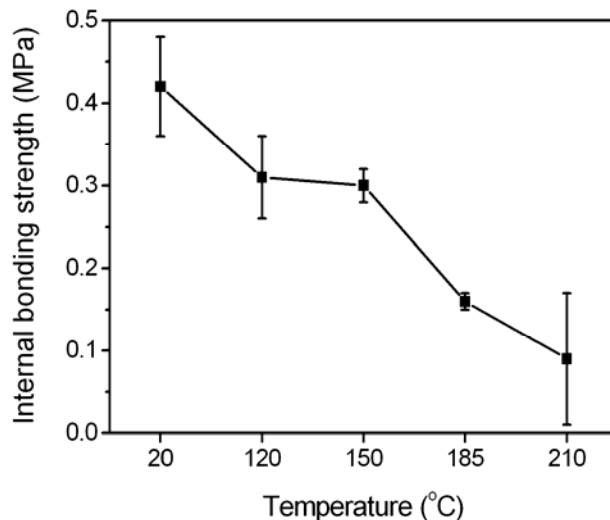


Fig. 8. IB of rice straw fiberboard affected by heat treatment

Bending Strengths

The bending strengths of the rice straw fiberboard with different water retardant levels are shown in Figs. 9 and 10. An increase of modulus of rupture (MOR) and modulus of elasticity (MOE) for panels produced from pMDI-treated rice straw fiber was observed, which might be attributed to a better bonding strength in the system. It was also observed that the wax emulsion could not improve the bending strength of the panels.

The bending strengths of the rice straw fiberboard with heat temperatures are shown in Fig. 11. Increasing temperature from 120°C to 210°C reduced the MOR by 12.1-66.7% in the panels. The results for MOE were mostly similar to those for MOR. The bending strength of panels mainly depends on the strength of single fiber. Between 120°C and 210°C, the natural fiber becomes dehydrated and generates water vapor and other noncombustible gases and liquids including carbon dioxide, formic acid, acetic acid, glyoxal, and water (Browne 1958). It is known that acidic condition at elevated

temperature can degrade the wood by hydrolysis and affected the properties (Tjeerdsma and Militz 2005; Mohebby et al. 2008). A higher temperature could accelerate thermal degradation of rice straw fiber, reducing the strength of single fiber. Furthermore, the interaction of rice straw fiber with UF resin lowers internal bond and, as a result, the strength of the whole panel. The MOE and MOR reduction in thermal treated wood (Kamdem et al. 2002; Sundqvist et al. 2006; Joščák et al. 2007) and composites (Boonstra et al. 2006; Del Menezzi et al. 2009) were also reported by many authors. Table 2 summarizes the properties of rice straw fiberboards with different water retardants and heat treatment.

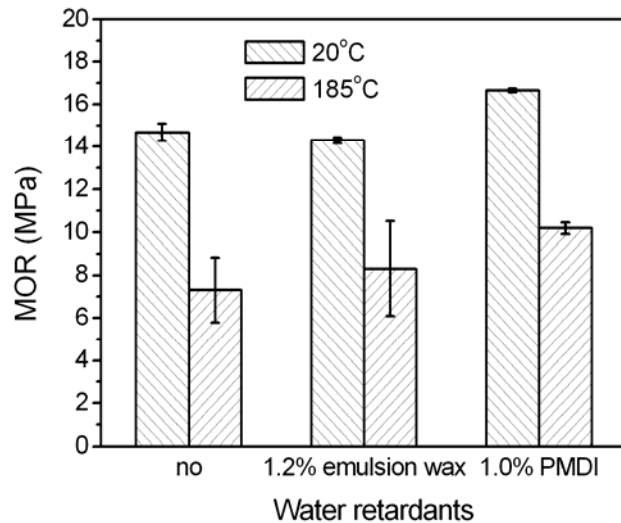


Fig. 9. MOR of rice straw fiberboard affected by water retardants with heat treatment at 185°C

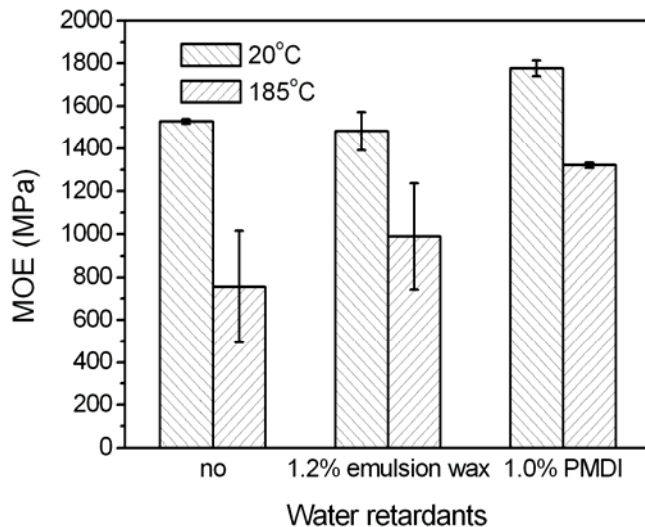


Fig. 10. MOE of rice straw fiberboard affected by water retardants with heat treatment at 185°C

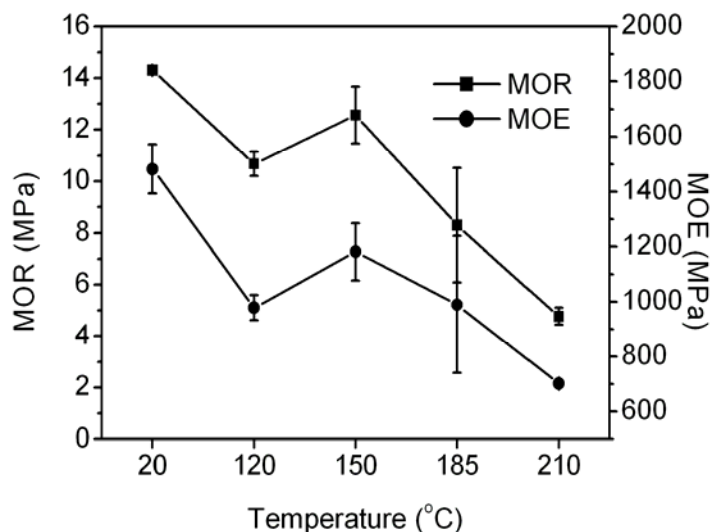


Fig. 11. Bending strengths of rice straw fiberboard affected by heat treatment

Table 2. The Mechanical and Physical Properties of Rice Straw Fiberboard

Run	WA (%)	TS (%)	IB (MPa)	MOR (MPa)	MOE (MPa)
1	90.80 (6.58) ^{a, b}	30.46 (0.69)	0.62 (0.06)	14.68 (0.40)	1528.2 (10.70)
2	93.92 (6.24)	20.35 (0.69)	0.27 (0.02)	7.30 (1.52)	754.5 (260.40)
3	95.83 (5.42)	31.38 (3.65)	0.42 (0.06)	14.30 (0.13)	1482.4 (88.80)
4	85.61 (0.47)	23.32 (0.24)	0.16 (0.01)	8.30 (2.22)	989.6 (248.80)
5	35.71 (17.49)	17.44 (4.43)	1.00 (0.14)	16.66 (0.09)	1777.9 (37.10)
6	42.50 (2.58)	15.11 (1.86)	0.43 (0.11)	10.18 (0.27)	1321.6 (11.00)
7	108.77 (10.56)	29.08 (2.35)	0.31 (0.05)	10.68 (0.47)	978.2 (46.50)
8	103.47 (6.76)	24.39 (2.11)	0.30 (0.02)	12.57 (1.10)	1181.8 (104.20)
9	47.81 (7.67)	12.71 (1.82)	0.09 (0.08)	4.77 (0.33)	700.9 (2.20)

Note: a: The values in parenthesis are standard deviation.

b: Means for each property are not significantly different when the significance at 95%.

CONCLUSIONS

The mechanical properties and water resistance of rice straw fiberboards with different water retardant after heat treatment at different temperatures were investigated. It was found that pMDI could be used as an effective water retardant for rice straw fiber, while wax emulsion did not significantly affect water retardant for rice straw.

With heat treatment on rice straw fiberboard, the 24-h thickness swelling of the fiberboards was significantly decreased due to the decrease in the content of free reactive hydroxyl groups, associated with hemicelluloses degradation. However, with increasing the temperature from 120°C to 210°C, the internal bonding strength was drastically reduced from 0.31 to 0.09 MPa, and the modulus of elasticity and modulus of rupture of the fiberboard also decreased.

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