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Some of the properties of flame retardant medium density fiberboard made from rubberwood and recycled containers containing aluminum trihydroxide

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Abstract

The flame retardancy of medium density fiberboard (MDF) made from mixture of rubberwood fibers and recycled old corrugated containers was studied. Aluminum trihydroxide (ATH) was used as a fire retardant additive and mixed with the fibers to manufacture experimental MDF panels using wet process. Phenol formaldehyde (PF) resin in liquid, 2% based on oven dry weight of fibers, was used along with 0%, 10%, 15% and 20% of ATH. The flame retardant test was done using the limiting oxygen index (LOI) test. The other properties investigated include internal bond strength, thickness swelling and water absorption. The results showed that ATH loading increased as the LOI of MDF increased. This demonstrated that ATH could improved the fire retardant property of MDF at sufficient loading. An increase in concentration of ATH showed an increase in the IB values of MDF made without resin. MDF panels made without resin showed a progressive increase in internal bond as the composition of recycled old corrugated containers fiber increased. Addition of resin improved internal bond strength and reduced thickness swelling, and water absorption. Thickness swelling of panel increased as the composition of recycled old corrugated containers fiber increased. Scanning electron microscopy (SEM) and energy dispersive X-ray analysis (EDAX) showed that there is indication of ATH and resin filling the void space in between fibers.

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1. Introduction

MDF is one of the most widely used wood composites as a substitute to manufacture housing furniture. MDF has drawbacks and limited potential because of its poor fire retardant properties. Thus, delaying or preventing fire spread in MDF would increase the value of MDF.

Rubberwood is currently the main raw material being used by the MDF industries in Malaysia. This is due to

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its availability, ease of use and yields MDF of high quality with acceptable physical properties. Rubberwood is also highly demanded as a raw material for other wood based industries such as furniture, mouldings and particleboard. With the rapid growth of these industries including MDF, there is an ever-increasing demand for rubberwood. Therefore there is a need to utilize other resources for the production of these composites and upgrade its quality.

Several studies have investigated the fire retardant treatment of wood based composites (Draganov, 1968; Myers and Holmes, 1975; LeVan et al., 1996). Several types of fire retardant chemicals were examined including borax-boric acid, chromated zinc chloride,

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ammonium sulphate, and nitrogen-phosphate mixtures (White and Sweet, 1992; Winandy et al., 1988). Fire retardant chemicals can be applied during the processing by the addition of fire retardant chemicals into the furnishing material (Kozlowski et al., 1999).

In this study the flame retardancy of MDF panels made from a mixture of rubberwood fibers and recycled old corrugated containers was investigated. The MDF was incorporated with aluminum trihydroxide (ATH) as fire retardant chemical. The ATH is widely used as fire retardant additives for plastics and elastomers (Brown and Herbert, 1992). Internal bond strength and thickness swelling of the samples were also evaluated.

2. Methods

Rubberwood (Hevea brasiliensis) fibers (R) were obtained from the Merbok MDF Factory, Malaysia, produced using a thermomechanical refining process without any chemical and resin. Recycled old corrugated container (C) fibers were also obtained from a packaging company waste. They were cut into small pieces, soaked in water and further disintegrated using a rotor disintegrator at high consistency for 25 min. Five sets of fiber mixture proportion in percent weight of oven dried weight of fibers were used; R100, R75/C25, R50/C50, R25/C75, and C100 having three replicates for each set. The resin used was 2% phenol formaldehyde (resol) in liquid form along with a control without resin. ATH in a water slurry prepared through a laboratory method and was precipitated on the fibers (How, 2002). Three concentrations of ATH were used; 10%, 15% and 20% based on oven dried weight of fibers along with a control (no ATH). Boards were made with the target density of 700 kg/m³ and with 5 mm thickness.

The fibers were mixed in water with a consistency of 8.0%. Different concentrations of ATH and 2% of phenol formaldehyde was later added to the fibers (based on the over dry weight of composite). The slurry mixture was mixed for 15 min. It was then poured into a $21 \text{ cm} \times 21 \text{ cm}$ stainless steel forming mould with aluminum mesh on both sides of the fiber mat. Wet press was carried out to consolidate and reduce the water content of the fiber mat and stopped at thickness of 10 mm at a pressure below 7 N/mm². After pressing, the weight of the fiber mat was determined. The consolidated mat was oven dried at a temperature of 150 °C without any aluminum mesh to a moisture content of 9-12%. Finally the mat was hot pressed at a temperature of 175 °C, with pressure at 12 N/mm², for 40 min. A total of 24 wet formed panels were made with three from each type of panel. The boards were then conditioned to standard procedure of 65% relative humidity with temperature of 20 °C prior to testing (JIS A 5906, 1983). Flammability of fire retardant MDF was determined by measuring limiting oxygen index (LOI) according to ASTM D 2863 (1997) under control atmosphere with each specimen having 15 replicates.

Internal bond strength, thickness swelling and water absorption of MDF were carried out according to JIS A 5906 (1983). For each test, there were nine replicates and three samples from each panel. The physical differences of fiber networking fracture surface and edges of the boards before and after ATH loading was evaluated using Leica Cambridge S360, SEM. EDAX Falcon System was also employed to confirm the presence of ATH.

3. Results and discussion

Limiting oxygen index (LOI) is the minimum concentration of oxygen, expressed as volume percent, in a mixture of oxygen and nitrogen that will support flaming combustion of a material at room temperature under the conditions specified by ASTM D 2863 (1997). A sample with high flame retardancy needs a higher oxygen concentration to burn based on a previous study (Ashley and Rothon, 1991).

The average LOI value for MDF at different of percent of ATH, with or without resin is shown in Table 1. The results show that the LOI value increases, as the amount of ATH increases. The aluminum oxide surface might promote more effective deposition. The oxide formed may bind with carbonaceous matter resulting in the accumulation of coherent tenacious crust, or ash at the burning surface. This inhibits further combustion by thermally insulating the substrate thereby reducing the rate of volatile evolution and providing a physical barrier to the oxygen diffusion needed to sustain the burning (Wan Hanafi and Hornsby, 1993).

With an increased composition of recycled old corrugated containers in MDF compared with rubberwood, the LOI also tended to increase. This could probably

 Table 1

 Limiting oxygen index (%) of MDF at different concentration of ATH

MDF	ATH content			
	0%	10%	15%	20%
0% PF				
R100	29.97	33.43	37.88	40.38
R75/C25	30.53	35.43	38.48	43.81
R50/C50	31.38	36.31	39.38	44.38
R25/C75	31.31	37.98	41.31	45.31
C100	31.88	37.14	43.65	47.88
2% PF				
R100	29.98	33.69	36.33	41.63
R75/C25	30.69	34.97	37.48	43.14
R50/C50	31.24	35.88	37.88	44.38
R25/C75	31.88	37.38	38.75	45.03
C100	33.63	38.38	42.38	47.62

R—rubberwood; C—recycled old corrugated containers; PF—phenol formaldehyde.

be due to the nature of the fibers (Fig. 1). Insignificant differences were seen in the LOI values for MDF made without resin and with resin.

The internal bond strength (IB) of MDF made with or without resin, at different of concentration of ATH is shown in Table 2. The IB values varied from 0.07 N/mm^2 to 0.85 N/mm^2 . When the IB values were compared with those specified by the Japanese standard for MDF (JIS A 5906, 1983) (Table 3), most MDF panels made without resin from rubberwood and a mixture of rubberwood and recycled old corrugated containers fibers did not meet the minimum requirement of the standard. However, for MDF made from recycled old corrugated containers, as the concentration of the ATH increase, the IB increase and achieved Type 150, Type 200, Type 150, and Type 300 respectively. With addition of PF resin, the IB strength increased at different concentrations of ATH and satisfied the minimum requirement stated in JIS A 5906 (1983).

An increase in concentration of ATH showed a significant increase in the IB values of MDF made without resin. Addition of PF resin improved IB strength properties. The results showed that the IB of MDF made from recycled old corrugated containers fibers is higher than those made from rubberwood fibers. This indicated that the types of raw materials influenced the IB strength of the panels. This effect of fiber mixtures of fibers on IB of MDF was significant for all concentrations of ATH made either with or without resin. One reason MDF made from recycled old corrugated containers fibers exhibited higher IB could be due to more felting

Table 2
nternal bond (N/mm ²) of MDF at different concentration of ATH

MDF	ATH content				
	0%	10%	15%	20%	
0% PF					
R100	0.10	0.07	0.14	0.25	
	$(0.01)^{a}$	(0.02)	(0.06)	(0.06)	
R75/C25	0.18	0.13	0.19	0.32	
	(0.02)	(0.03)	(0.05)	(0.06)	
R50/C50	0.18	0.24	0.37	0.44	
	(0.06)	(0.05)	(0.15)	(0.14)	
R25/C75	0.21	0.28	0.36	0.50	
	(0.07)	(0.07)	(0.11)	(0.17)	
C100	0.31	0.40	0.37	0.51	
	(0.13)	(0.11)	(0.17)	(0.15)	
2% PF					
R100	0.25	0.23	0.23	0.41	
	(0.05)	(0.04)	(0.12)	(0.11)	
R75/C25	0.42	0.25	0.31	0.39	
	(0.06)	(0.09)	(0.06)	(0.03)	
R50/C50	0.34	0.32	0.50	0.45	
	(0.07)	(0.06)	(0.18)	(0.13)	
R25/C75	0.40	0.32	0.49	0.59	
	(0.06)	(0.03)	(0.13)	(0.26)	
C100	0.33	0.33	0.58	0.85	
	(0.07)	(0.02)	(0.18)	(0.36)	

R—rubberwood; C—recycled old corrugated containers; PF—phenol formaldehyde.

^a Numbers in parentheses are standard deviations.

together of the fibers compared to MDF made from rubberwood fibers. This created more fiber to fiber contact and glue line contact (Hunt and Vick, 1999).



Fig. 1. Scanning electron micrographs (×200, 15 kV) of MDF edges made from (a) rubberwood fibers without ATH and without resin, (b) recycled old corrugated containers fibers without ATH and without resin, (c) rubberwood fibers with 20% ATH and 2% PF resin, (d) recycled old corrugated containers fibers with 20% ATH and 2% PF resin.

Table 3 Classification of IB of MDF with reference to JIS A 5906

MDF	ATH content				
	0%	10%	15%	20%	
0% PF					
R100	_	_	_	_	
R75/C25	_	_	_	х	
R50/C50	_	_	х	{ xx }	
R25/C75	_	_	{ x }	{xxx}	
C100	х	XX	{ x }	{xxx}	
2% PF					
R100	_	_	_	XX	
R75/C25	XX	_	х	х	
R50/C50	х	х	XXX	{ xx }	
R25/C75	XX	х	{xxx}	{ xxx }	
C100	х	х	{xxx}	{xxx}	

Requirement for JIS A 5906 (MDF density 400–800 kg/m³); xxx: 300– type MDF with IB ≥ 0.490 N/mm²; xx: 200—type MDF with IB ≥ 0.392 N/mm²; x: 150—type MDF with IB ≥ 0.294 N/mm²; { }: Density > 800 kg/m³; -: below of minimum requirement; R—rubberwood; C—recycled old corrugated containers; PF—phenol formaldehyde.

The thickness swelling at different concentrations of ATH is shown in Table 4. As the concentration of ATH increases, the thickness swelling of the MDF tended to decrease. Lower thickness swelling was observed in MDF made with resin compared to those of without resin. Addition of resin enhanced better bond-

Table 4 Thickness swelling (%) of MDF at different concentration of ATH

MDF	ATH content				
	0%	10%	15%	20%	
0% PF					
R100	26.13	25.29	20.04	15.94	
	$(2.47)^{a}$	(2.43)	(3.89)	(1.57)	
R75/C25	26.14	25.77	20.87	16.29	
	(1.49)	(1.15)	(1.17)	(0.91)	
R50/C50	27.64	26.24	22.12	21.13	
	(1.02)	(1.60)	(0.27)	(0.44)	
R25/C75	30.70	26.65	21.88	20.79	
	(0.82)	(1.08)	(0.71)	(0.59)	
C100	33.00	25.69	23.77	21.84	
	(1.16)	(1.34)	(0.95)	(0.26)	
2% PF					
R100	15.92	20.06	17.13	13.75	
	(0.43)	(0.60)	(1.35)	(0.70)	
R75/C25	17.19	21.06	17.28	15.58	
	(0.59)	(0.91)	(0.25)	(0.37)	
R50/C50	22.21	21.29	20.30	16.00	
	(0.33)	(0.88)	(0.50)	(0.33)	
R25/C75	27.41	23.93	20.81	16.91	
	(1.65)	(0.74)	(0.56)	(0.36)	
C100	25.56	24.91	19.71	16.15	
	(1.71)	(0.54)	(0.42)	(0.48)	

R-rubberwood; C-recycled old corrugated containers; PF-phenol formaldehyde.

^a Numbers in parentheses are standard deviations.

ing of the boards and consequently improved thickness swelling. Generally, it can be seen that MDF made with and without resin showed to have increase thickness swelling with increasing of composition of recycled old corrugated containers, except for panels made from 100% recycled old corrugated containers with resin.

For control MDF made without resin and ATH, as the composition of recycled old corrugated containers increased, the thickness swelling also increased. As a result, MDF made from recycled old corrugated containers had higher thickness swelling than MDF made rubberwood fibers. The recycled old corrugated containers fibers could have been modified during pulping process and became more hydrophilic than the virgin fibers such as rubberwood. They then expanded more when exposed to high relative humidity (Hunt and Vick, 1999). Thickness swelling decreased when the concentration of ATH in the board increased and also with addition of resin. All MDF did not meet the 12% limit specified by JIS standard.

For water absorption as shown in Table 5, MDF made with resin has lower water absorption than MDF panels made without resin. An addition of ATH also generally tended to lower the water absorption.

Representative SEM micrographs taken from the board edges are shown in Fig. 1. The results show that MDF made from rubberwood fibers with 20% ATH and 2% PF resin had more dense fiber network than

Table 5

Water absorption (%) of MDF at different concentration of ATH

MDF	ATH content				
	0%	10%	15%	20%	
0% PF					
R100	103.44	104.02	87.09	73.20	
	$(3.55)^{a}$	(3.25)	(3.00)	(3.16)	
R75/C25	97.12	92.80	77.62	65.25	
	(1.76)	(0.76)	(2.48)	(3.25)	
R50/C50	101.86	90.00	76.55	63.84	
	(0.68)	(1.88)	(2.09)	(2.59)	
R25/C75	99.66	96.77	73.39	65.17	
	(1.81)	(5.52)	(4.10)	(2.55)	
C100	93.60	83.83	77.38	58.95	
	(2.12)	(10.49)	(2.15)	(0.72)	
2% PF					
R100	81.68	72.45	82.24	61.57	
	(1.380)	(2.30)	(2.75)	(0.73)	
R75/C25	81.13	84.70	71.02	60.59	
	(1.21)	(1.54)	(5.26)	(1.87)	
R50/C50	79.60	78.01	60.97	63.34	
	(0.19)	(4.88)	(3.60)	(1.04)	
R25/C75	82.99	82.04	60.59	52.32	
	(1.48)	(10.040)	(2.89)	(4.50)	
C100	80.52	59.44	52.84	50.75	
	(0.09)	(2.60)	(3.48)	(7.21)	

 $R-\!\!\!\!\!\!\!$ rubberwood; $C-\!\!\!\!\!\!$ recycled old corrugated containers; $PF-\!\!\!\!$ phenol formaldehyde.

^a Numbers in parentheses are standard deviation.

control panels made from rubberwood alone. With addition of ATH and resin, inter fiber void were loaded with ATH and resin. ATH acted as filler or attached on the fibers and filled the gap between fibers. Meanwhile, resin enhanced the inter fiber bonding resulting in minimal rubberwood fiber being pulled out when cutting at the edge of board. The cross-section cutting edge of MDF made from recycled old corrugated containers fibers with 20% ATH and 2% PF resin showed a flat surface with less void. MDF made from recycled old corrugated containers alone had smoother edged surfaces compared with the edges of MDF made from recycled old corrugated containers with ATH and resin. This indicated addition of ATH and resin had affected the fiber network of MDF. The densities of the board also influenced the properties to some extent.

Analysis using energy dispersive X-ray analysis (EDAX) is shown in Figs. 2 and 3. Aluminum, potassium and silicon were detected in MDF made from recycled old corrugated containers alone but in a small amount. With addition of ATH, the percentage of aluminum compound detected increase. Meanwhile, sulfate compound was found in both ATH loaded MDF, it may be due to the incomplete discharged of sulfate ions contained in the ATH slurry.



Fig. 2. EDX spectrum of MDF made from (a) rubberwood control without ATH and without resin, (b) rubberwood fibers with 20% ATH and 2% PF resin.



Fig. 3. EDX spectrum of MDF made from (a) recycled old corrugated containers fibers control without ATH and without resin, (b) recycled old corrugated containers fibers with 20% ATH and 2% PF resin.

4. Conclusions

The results showed that as ATH loading increased, the limiting oxygen index of MDF also increased. This demonstrated that at sufficient loading ATH could improve the fire retardant property of MDF. An increase in concentration of ATH showed an increase in the IB values of MDF made without resin. MDF made without resin showed a progressive increase in internal bond as the composition of recycled old corrugated containers fiber increase. Addition of resin improves internal bond strength, reduce thickness swelling and water absorption. Thickness swelling of panel increased as the composition of recycled old corrugated containers fiber increase. The types of raw materials also influenced the property studied.

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