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Production of Low Formaldehyde Emission Particleboard by Using New Formulated Formaldehyde Based Resin

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ABSTRACT

In order to preserve the global market competitiveness, the particleboard industry was affronted with challenges to reduce formaldehyde emission while maintaining the quality strength properties of particleboard. To counter the issue, particleboards with five different surface-to-core ratio were fabricated by applying newly formulated UF and MUF resins which were 30% surface: 70% core (3:7); 40% surface: 60% core (4:6); 50% surface: 50% core (5:5); 60% surface: 40% core (6:4) and 70% surface: 30% core (7:3) based on dry particle weight respectively. Formaldehyde emission and strength properties of the fabricated particleboard were investigated based on Japanese Industrial Standard, which are JIS A 1460 and JIS A 5908, respectively. All the MUF-bonded particleboard complied with the type 18 standard, whereas all the UF-bonded particleboard produced complied with type 13 except thickness swelling of the UF-bonded particleboard. The surface-to-core ratio applied in three layered particleboard for both resins exerted considerable influence on the strength properties and formaldehyde emission of particleboards produced from both resins. MUF-bonded particleboard with 40% surface and 60% core recorded the lowest formaldehyde emission (0.09 mg L^{-1}) and highest strength properties. For UF-bonded particleboard, the ratio of 60% surface and 40% core showed the lowest formaldehyde emission (0.28 mg L^{-1}) with better strength properties. This study highlighted the potential of MUF resin to replace UF resin due to its ability to produce F**** particleboard with better strength properties and lower formaldehyde emission according to JIS A 5908.

Key words: Rubberwood particleboard, japanese industrial standard, strength properties, urea formaldehyde, melamine urea formaldehyde

INTRODUCTION

Due to the reduction of solid wood supply, the popularity of particleboard was increasing gradually, particularly in the furniture manufacturing industry (Ratnasingam and Wagner, 2009). The total export value of particleboard of Malaysia has reached RM 250 million in year 2009 which is three folds of the export value of particleboard in year 2000 (Loh *et al*., 2010). Particleboard industries in Malaysia still rely solely on Urea Formaldehyde (UF) resins as binding agents. The reason behind the successful application of UF resin is due to its economically cheap cost and desired particleboard properties (Nemli and Ozturk, 2006). The utilization of UF resin has been overwhelmed by some undesirable particleboard properties. Significant amount of formaldehyde will also be emitted from particleboard bonded with UF resin. As a consequence, high level of formaldehyde exposure (above 0.1 parts per million of air) can cause watery eyes, burning sensations in the eyes, nose and throat, nausea, coughing, chest tightness, wheezing, skin rashes and allergic reactions. In response to that, formaldehyde has been classified as a known carcinogen by the state of California (prop 65) and the International Agency for Research on Cancer (IARC). The quantity of formaldehyde emission has been regulated by many countries. The Government Of Japan, Australia and United Kingdom has regulated the limit of indoor formaldehyde exposure to 0.1 mg m^{-3}, 0.12 mg m^{-3} and 0.1 mg m^{-3} respectively (Tang *et al.*, 2009).

Over the past decade, a lot of potential efforts have been made to reduce formaldehyde emission from particleboard. Beneficial chemical such as scavenger for pre-treatment and post-treatment is applied on the surface of the board. The manufacturing control is said to have the ability to dramatically reduced formaldehyde emissions of particleboard as much as 80-90% Since the early 1970's (Marutzky and Dix, 2004). However, the product standard was gradually become more demanding, as modern consumers rather emphasized on 'healthy' products and the emergence of resin technology has made the resin more economical while maintaining the performances, particularly MUF (Zanetti and Pizzi, 2004). Fluctuation of particleboard properties often caused problems for the manufacturers (Zaidon, 2007). Hence, to maintain the market competitiveness globally, the industry is affronted with challenges to reduce the formaldehyde emission while maintaining the ideal strength properties of particleboard (Kim and Kim, 2005).

In order to counter this issue, the need to change in resin technology has never being an essential until now. This has led to the introduction of new formaldehyde-based resin by manufacturers. The new Melamine Urea Formaldehyde (MUF) resin and lower mole ratios of formaldehyde to urea F/U seemed to be the best suit for to produce particleboard by using resin technology. The presence of melamine provides a stable triazine ring in MUF resins. This, in terms, provides hydrolytic and thermal stability property of MUF particleboard (Siimer *et al.*, 2008). Whereas, at low F/U ratio resin, the free formaldehyde content and board emission rate are lower. The selection of newly developed MUF and low F/U ratio resin as replacement for urea formaldehyde for particleboard consider practical, because minimum change are important for the process and machinery on current particleboard industry which commonly use UF resin as binder.

Despite this practical progress, great uncertainty still exists as to the precise mechanism by which the reaction of these resin on the properties of particleboard. Moreover, the rate and extent of formaldehyde emission from particleboard is influenced by a large number of parameters with particles surface-to-core ratio is the major contributor. Few researches were carried out on the effect of surface-to-core ratio on the properties of particleboard. Nemli (2003) suggested that increasing the surface-to-core ratio improved the physical and mechanical properties of particleboard. However, the effect of surface-to-core ratio on the formaldehyde emission is still uncertain.

As such, it is important to study the effect of both MUF and low U/F ratio resins on mechanical properties and formaldehyde emission from particleboard produced with different surface-to-core ratio in order to determine which ratio is ideal to be applied in commercial particleboard plant, for ideal particleboard production in terms of formaldehyde emission and strength properties.

MATERIALS AND METHODS

This study was initiated from February 2009. Three layered particleboards were fabricated for the study. This particleboard comprised of surface top and bottom, with core lying in the center of the board. Both surfaces are made of fine particle, while the core is made of coarse particle.

Table 1: The classification of rubberwood particle for surface and core layer

Range of particle dimensions	Surface (mm)	Core (mm)
Length	2-5	10-30
Width	1-2	2-4
Thickness	0.3-0.5	0.2-0.4

Rubberwood (*Hevea brasiliensis*) particles were obtained from a commercial particleboard plant located in Negeri Sembilan, Malaysia. The particles were dried to 2% moisture content prior to the particleboard fabrication. The dried rubberwood particles were screened to obtain required size for surfaces and core materials. All the particles were prepared in Heveaboard Berhad, Negeri Sembilan. The classification of the particles is shown in Table 1.

The binding agents applied consisted of two newly formulated resins supplied by Norsechem (M) Sdn. Bhd. One of which was the MUF resin with the molar ratio of formaldehyde to urea to melamine is 1.0: 0.9: 0.2 and solid content of 63%. The other resin applied was UF with low resin F/U ratio (product identity: RD 100). The urea to formaldehyde ratio was 0.88 and the solid content was ranged from 60-65%.

The effect of surface-to-core ratio (S: L) on formaldehyde emission and strength properties of three layered particleboard bonded by UF resins and MUF resins were studied. Five different surface-to-core ratio which are 30% surface: 70% core (3:7); 40% surface: 60% core (4:6); 50% surface: 50% core (5:5); 60% surface: 40% core (6:4) and 70% surface: 30% core (7:3) based on dry particle weight were selected as the parameter for the particleboard fabrication process.

The dimension of the particleboard produced is 340 mm × 340 mm × 12 mm with targeted density of 750 kg m^{-3} and 680 kg m^{-3} for MUF and UF-bonded particleboard respectively. The resin dosages applied in the MUF-bonded particleboard for core and surface layers was 11 and 14%, respectively based on dry particle weight. Whereas, resin dosage of 7 and 10% based on the dry particle weight were selected for RD 100, for core and surface layers of the particular UF-bonded particleboard respectively. For both UF and MUF-bonded particleboards, ammonium chloride with solid content of 25% was used as hardener and the 0.5% of wax (solid content of 60%) based on dry particle weight is applied during particleboard fabrication. The particleboards with MUF resin were pressed at the temperature and time of 180°C and 630 sec under the pressure of 100 bars. On the other hand, the particleboard using UF with low resin F/U ratio was pressed under the temperature of 180°C for 270 seconds with 100 bars of pressure. After the hot-pressing process, all the particleboards were conditioned 7 days at 20±2°C and relative humidity of 65±5% prior to physical and mechanical properties determination. A total of 30 board panels were manufactured, with three replicates for each treatment.

Strength properties, such as density, bending strength, thickness swelling, wet bending strength and internal bonding strength, were tested for the fabricated particleboard according to JIS A 5908 standard. Desiccator method (Risholm-Sundman *et al.*, 2007) is used to test the formaldehyde emission of particleboard which refers to JIS A 1460. The quality requirements specified in the Japan particleboard standard JIS A 5908 were applied as guide values. Based on the standard, the boards made by UF resin is regarded as type 13 boards while the boards made by MUF resin is regarded as type 18 boards. For the thickness of 12 mm board, the requirements are: the value of MOR is>13.0 N mm^{-2}, IB is>0.2 N mm^{-2} and the value of TS for 24 h is <12% for type 13 boards. For type 18 boards, the standard requirement for MOR is >18.0 N mm^{-2}, IB is

>0.3 N mm^{-2} and the value of TS (24 h) is<12%. The limitation for formaldehyde emission for both UF and MUF-bonded particleboard is similar, that the mean of 9 samples is 0.3 mg L^{-1} or under and the maximum amount is 0.4 mg L^{-1} according to JIS standard. Table 4 showed the concluded JIS A 5908 standard.

All data were statistically analyzed by using one way ANOVA analysis and the mean of each value was compared by using TUKEY test to determine the differences between treatment levels.

RESULTS AND DISCUSSION

MUF resin (Type 18) particleboard: Table 3 shows the result of tested strength properties and formaldehyde emission of MUF particleboard. Unlike MF, the small quantity of melamine addition into UF resin improved the particleboard properties and formaldehyde emission (Hse *et al.*, 2008). MUF-bonded particleboard complied with the entire requirement for the Type 18 particleboard classification according to JIS A 5908 standard. The density of the MUF particleboard ranges from 0.72 g cm^{-3} up to 0.91 g cm^{-3}. According to JIS A 5908, the density of particleboard shall be 0.40 g cm^{-3} or over up to and including 0.90 g cm^{-3} (Table 2).

Particleboard M3:7 failed to meet the targeted density as well as the JIS requirement. The density of M7:3 was significantly different at 0.05 levels as the board contained larger quantity of fine particles on both top and bottom surface of the particleboard. Hence, it caused less space void in the particleboard; hence, the board is practically denser than that particleboard with less fine particles.

All the MUF particleboard fabricated achieved the minimum internal bond (0.3 N mm^{-2}) according to JIS 5908. In general, particleboard M4:6 showed the best performances in terms of strength properties and formaldehyde emission. MUF resin showed better performance than UF resin for water resistance, for all the particleboard bonded with MUF resin achieved the ideal

Table 2: Result of strength properties and formaldehyde emission of MUF particleboard

Treatment	Density g cm^{-3}	TS (%)	*IBN (mm^{-2})	*MORN (mm^{-2})	*Wet bending (N mm^{-2})	**FE (mg L^{-1})
M7:3	0.91±0.13^{b}	11.63±2.29^{a}	1.67±0.28^{a}	23.07±5.94^{ab}	7.66±0.35^{a}	0.12
M6:4	0.77±0.09^{ab}	7.99±0.91^{a}	1.62±0.52^{a}	18.39±5.27^{a}	9.09±1.82^{ab}	0.12
M5:5	0.76±0.10^{ab}	10.65±3.66^{a}	1.88±0.40^{a}	19.82±5.42^{ab}	10.24±0.74^{b}	0.20
M4:6	0.82±0.06^{ab}	10.43±1.81^{a}	2.19±0.28^{a}	29.86±4.34^{c}	12.76±2.02^{c}	0.09
M3:7	0.72±0.09^{a}	9.20±1.23^{a}	1.62±0.14^{a}	28.84±3.62^{bc}	8.36±0.87^{ab}	0.10

*5 samples were tested for Density, Thickness Swelling (TS), Internal Bonding (IB), Modulus Of Rupture (MOR) and Wet Bending. ** 9 samples were tested for Formaldehyde Emission (FE). Least significant difference means followed by same letter in each column are no significantly different at $p<0.05$

Table 3: Result of strength properties and formaldehyde emission of UF particleboard

Treatment	Density (g cm^{-3})	*TS (%)	*IBN (mm^{-2})	*Bending (N mm^{-2})	**FE (mg L^{-1})
L7:3	0.698±0.02^{a}	17.30±1.99^{ab}	1.09±0.54^{ab}	20.51±4.00^{ab}	0.22
L6:4	0.689±0.02^{a}	18.80±2.61^{b}	1.95±0.53^{c}	20.67±2.28^{b}	0.26
L5:5	0.686±0.02^{a}	34.56±3.10^{d}	0.67±0.25^{a}	17.45±2.80^{ab}	0.27
L4:6	0.685±0.01^{a}	14.61±0.68^{a}	1.13±0.46^{ab}	19.63±2.96^{ab}	0.28
L3:7	0.678±0.02^{a}	27.35±2.27^{c}	1.66±0.44^{b}	16.90±2.01^{a}	0.19

*5 samples were tested for Density, Thickness Swelling (TS), Internal Bonding (IB) and Modulus of Rupture (MOR).** 9 samples were tested for Formaldehyde Emission (FE). Least significant difference means followed by same letter in each column are no significantly different at $p<0.05$

percentage of swelling with no significant difference among the ratio. Particleboard M4:6 obtained the lowest formaldehyde emission, while the MOR and wet bending strength is better compared to other particleboard with different ratio. Particleboard M 4:6 was significantly different at 0.05 levels for MOR and wet bending strength. Better wet bending strength is important because MUF resin performed better than UF resin because of the value-added ability for water resistance. All the particleboards with different ratio had no significant difference for IB, as MUF showed better adhesion as the bonding between particles of MUF-bonded particleboard were better than UF resin.

Samples were sent for testing to obtain the average of formaldehyde emission in order to determine if the fabricated particleboard achieve the standard of JIS A5908. A review suggested that production of application of formaldehyde based resin on particleboard production was harmful to human (Moubarik *et al*., 2010). However, the result of this study proved otherwise. The slight addition of melamine content in MUF proved to improve the formaldehyde emission of particleboard (Tohmura, 2001). The formaldehyde emission of all particleboards bonded by special formulated MUF resin ranged from 0.09 to 0.20 mg L^{-1} which is far lower than the requirement for classified as F**** (Super E0) particleboard as stated in the JIS standard.

Low resin F/U ratio (Type 13) particleboard: Average values of static bending strength, thickness swelling and internal bond are shown in Table 4. The fabricated particleboard had achieved the targeted density which is 0.680 g cm^{-3} according to JIS A 5908. All the UF-bonded particleboards were found to comply with the Type 13 particleboard classification as stated in JIS 5908 except for the thickness swelling. The most ideal thickness swelling of UF-bonded particleboard (L 4:6) is significantly different with other group at 0.05 levels. One of the disadvantages of UF-bonded particleboard represented by the thickness swelling of particleboard which was ranged from 14.61% to 34.56% (Table 3) where the maximum thickness swelling allowed is 12% according to JIS 5908. The poor thickness swelling performance by low resin F/U ratio particleboard may due to the existing of amino methylene linkages in urea formaldehyde resins as the resins are not resistant to the water. As the F/U ratio reduced, the resin is more susceptible to moisture and this resin will undergo decomposition with the effect of water absorption of particles (Jackh, 1993). The UF-bonded particleboard with low F/U ratio achieved the minimum requirement for internal bond as stated in JIS 5908 which is 0.2 N mm^{-2}.

In general, the best performances were particleboard of type L6:4 and L7:3, due to the highest IB and MOR. The MOR of particleboard L 6:4 is significantly different with particleboard L 3:7 at 0.05 levels. Hence, it can be concluded that the increment of surface layer of the particleboard would not caused improvement of static bending with UF resin. It can be explained by the fact thathigher amounts of particle usage on the surface layers cause an even tighter structure on the particleboard and consequently enhanced the bending properties (Nemli, 2003).

Table 4: JIS A 5908 standard for density, Thickness Swelling (TS), Internal Bonding (IB), Modulus of Rupture (MOR) and wet bending

	Density	Moisture content	MOR	Wet bending	Internal bonding	Formaldehyde emission
MUF type 18	0.40 g cm^{-3} or over up to and ^{3} including 0.90 g cm^{-1}	5% or over up to and including 13%	Minimum 18.0 N mm^{-2}	Minimum 9.0 N mm^{-2}	Minimum 0.3 N mm^{-2}	Mean 0.3 mg L^{-1} or under maximum of 0.4 mg L^{-1}
UF type 13	0.40 g cm^{-3} or over up to and including 0.90 g cm^{-3}	5% or over up to and including 13%	Minimum 13.0 N mm^{-2}		Minimum 0.2	Mean 0.3 mg L^{-1} or under maximum of 0.4 mg L^{-1}

to 0.28 mg L^{-1} which is still lower than the requirement for classified as F**** (Super E0) particleboard as stated in the particleboard standard.

CONCLUSION

Particleboard produced using MUF resin can be considered as potential solution to replace the particleboard bonded with UF resin in order to brace the challenges of ultra low formaldehyde emission while retaining the required properties. MUF resin contributed better strength properties and formaldehyde emission comparing to UF resin. The surface-to-core ratio exerted considerable influence on the particleboards produced from both resins. For UF-bonded particleboards, board type with higher surface to core ratio shows the best performance in mechanical properties. For MUF particleboards, the best mechanical properties were recorded by board type M 4:6. The effect of surface-to-core ratio of particleboard on formaldehyde emission was not significant.

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