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Natural Rubber Latex (NRL) and rice starch as an alternative binder in wood composite industry.

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ABSTRACT

Medium density fiberboards (MDF) was produced in laboratory using bio-adhesives synthesized using natural rubber latex (NRL) and rice starch (RS) with pressing time of 3 minutes, temperature of 180°C and pressure of 5MPa. This present study is aimed on increasing and improving the physical and mechanical performances of MDF by application of bio-based adhesives which comprises NRL and rice starch solution. The effects of bio-adhesives used on the physical and mechanical properties of boards produced were investigated accordingly and compared with the MDF prepared using urea-formaldehyde according to the specific ASTM standards. In the composite preparation, bio- adhesives were used as manipulated variable, where they were split into 5 further weight proportions as follows: 1) 20g NRL; 2) 15g NRL + 5g RS; 3) 10g NRL + 10g RS; 4) 5g NRL + 15g RS and 5) 20g RS. The samples were also subjected to mechanical testing such as modulus of rupture (MOR), internal bonding (IB) and thickness swelling (TS). Based on the mechanical testing done on the specimens, it is clearly indicated that bioadhesives comprising of 15g of natural rubber latex and 5 g of rice starch blend improves the modulus of rupture (MOR) and internal bonding strength (IB) of MDF significantly, where the mechanical strength value obtained were better and comparable than MDF made with UF alone. Hence, it strongly indicates the promising feasibility of NRL and rice starch to replace conventional UF in wood composite industry, and eventually highlights the success of this research.

INTRODUCTION

Recently, an extensive search and hunt for solutions that would make it feasible to substitute synthetic polymers or at least to limit their application in daily life. The interest in natural products evolved due to challenges in achieving sustainable development as well as because of the escalating level of ecological awareness among human beings. Global wood products manufacturing sums up to about 900 tons per year (FAO, 2013) and the conventionally used synthetic resins in this type of manufacturing is conquered prominently by formaldehyde based resins. Unfortunately, this type of resin is highly volatile and is also classified as a gaseous contaminant as well as a carcinogenic substance. The formaldehyde emission had seriously render human life and ecological balance at risk which led to the banning of this resin by many countries, especially on its usage in wood products manufacturing sector. Therefore, in order to counter this

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critical issue, much attempts and researches had and have been continuously conducted up to date to develop an alternative binder from natural based materials that can fully substitute the usage of UF in wood manufacturing sector. In addition, the transition from synthetic adhesives to bio-based adhesives achieved greater attention throughout the world as everyone believe that only naturally occurring substances can bring no harm to humankind.

Application of natural rubber latex (NRL) as wood adhesive has been on form since many years ago, and many previous researches regarding this raw material has been reported (Weeraratne *et al*., 1972; Hermiati *et al*., 2006). NRL is a milky brown, slightly viscous latex suspension which exhibits good moisture and mold resistance with no toxicity. It has been widely explored and used for the synthesis of wood adhesives especially for non-polar adherends (Pizzi, 1989) with modification done to increase its performance on polar surfaces. Besides NRL, studies on wood adhesives from variety of starch have been conducted since long time ago. Starch is a biopolymer which is renewable, widely available and biodegradable. It has been actively used as binders, glues and pastes even during ancient years (Kennedy Harry, 1989). However, starch has its main limitation of not having strong bonding capacity to glue wood based materials (Imam *et al*., 2001; Imam, Mao, Chen and Greene, 1999). Therefore, blending technique has been considered on starch in order to allow for physical or chemical modifications to take place on the molecules. Recent studies have emphasized on the synthesis of formaldehyde free wood adhesives through the reaction between the blend of starch with other polymers such as starch /PVOH (Imam *et al*., 2001) and starch/ tannin blend (Moubarik, Charrier, Allal, Charrier and Pizzi, 2010).

In the present study, a renewable and eco-friendly NRL-Rice starch blend wood adhesive was synthesized through simple chemical modification done on the natural polymers. The mechanical properties and water resistance of the adhesive synthesized were examined through the application of bio-adhesive in making MDF boards. Besides, the proportions of NRL and rice starch were varied in order to identify the compatibility of the blends as well as the effect of the different weightage of each natural polymer on the adhesive performance. It is highly expected that the findings of this study can help us to understand the adhesion properties found in the nature and further explore their suitability to be incorporated as glue for wood based materials.

MATERIALS AND METHODS

Materials:

Fresh rubber wood (H. Brasiliensis) fibers were used in this study which was obtained from Robin Resources Sdn. Bhd. Natural Rubber Latex of high ammonia concentration was obtained from a rubber estate from Segamat, Johor. Rice starch is used in this experiment which is purchased from Permula Chemicals Sdn. Bhd., Gebeng, Kuantan. The moisture content of the starch is controlled within 8.0-13.0 and its pH is 6.0.

Methods:

Mixing Starch with NRL:

70 wt% of starch was prepared with 30 wt% of 1M HCl by heating both the chemicals at 60° C along with stirring for 15 minutes using a hot plate. After that, high ammonia NRL was added to the suspension at 45° C and stirred until a homogenous mixture can be observed according to the proportion of NRL and rice starch set as Table 1 below. UF binder was set as control (A) and used as the comparison for the bioadhesives formulation throughout the experiment.

Table 1: Formulations of Bioadhesives

Formulation of Adhesive	Wood fibers (gram)	NR Latex (g)	Starch solution (g)
B	200	20	0
C	200	15	5
D	200	10	10
E	200	5	15
F	200	0	20

Preparation of MDF Board:

200 mm x 200 mm x 6 mm MDF boards with target density of 800 kg/m^3 were produced by using 200 grams of wood fibers. The fibers were placed into a rotary drum blender and bioadhesives which were formulated according to Table 1 were sprayed with a spray gun onto the fibers. Then, mixing at 18rpm rotation speed for 5 minutes were carried out so that uniform mixing and tumbling can happen prior to MDF making process. Subsequently, the resin sprayed fibers were molded into a 200 mm x 200 mm panel using the same sized preformed panel and pressed at 5 MPa for 180 s at 180 °C. The prepared boards were then used to find out the physical and mechanical properties so that the suitability of this bio-adhesive to replace UF can be identified. Fiber moisture, pressing time, platen temperature is kept constant throughout the experiments.

Mechanical Testing on MDF Boards:

Mechanical testing were conducted for samples made from UF and also of boards glued together by the different formulations of bio-adhesives such shown in Table 1. Modulus of Rupture (MOR) and internal bonding (IB) were measured according to the standard ASTM D 1037 methods and were done using a Shimadzu UTM AG-X plug series instrument. Data obtained were then analyzed using Trapezium X- Software.

For the final result analysis, a total of 10 samples were taken from each formulation of bio-adhesives, respectively for MOR and IB testing. Internal bonding which defines the tensile strength of the adhesive on the wood fibers is tested perpendicular to the plane of the boards, with the cross head of 1mm/min. MOR testing was carried out by a three-point static bending test which were subjected to a cross head speed of 10mm/min.

Upon analysis of specimens using the universal testing machine, the value of MOR and IB were calculated by using the equations shown below:

$$MOR\ (N/mm^2) = (3 \times P \times L) / (2 \times B \times D^2) \quad (1)$$

P = breaking load

L = distance between knife edges on where the sample was placed or supported

B = average breadth of specimen

D = average depth of specimen

The following is the equation used to calculate IB of the composite samples:

IB (N / mm^2) = F (maximum force calculated at entire area) / A (surface area of specimen)

$$IB = (F/L) \times B \quad (2)$$

Where F = maximum force; L and B = cross section of the sample or specimen.

Thickness swelling test were also conducted on the specimens, in accordance of ASTM D 1037 standard. The length, weight and thickness of each specimen was measured initially. Then, the samples were soaked into water for 24 hours, at room temperature. After that, the specimens were removed, dried and the dimensions were measured again. The values of the thickness swelling of the samples were obtained by following the below listed equation:

$$TS\ (\%) = (T_2 - T_1) * 100 / (T_1); \quad (3)$$

where T_1 is the initial thickness while T_2 is the thickness of the sample soaked in water.

RESULT AND DISCUSSION

The mechanical properties such as Modulus of Rupture (MOR), Internal bonding (IB) and Thickness swelling (TS) for standard UF MDF board and boards made from bioadhesive will be discussed. The compatibility of both NRL and rice starch to give better performance than UF will be investigated through the comparison of mechanical strength between the boards having different type of wood glue in them.

Modulus of Rupture (MOR)*:*

The MOR results for the boards were depicted in the Figure 1 below. It shows that MDF panel board that has 15 g of NRL + 5g of starch exhibited the greatest mechanical strength in terms of MOR value. The board actually surpasses the MOR value of UF board, which clearly explained that presence of NRL and starch at correct proportion can actually be beneficial to play the role of wood adhesive in such a good manner. Besides that, based on Figure 1, we can see that as the composition of rice starch in the bioadhesive formulation increases with the reduction of NRL composition, the MOR value declined gradually. This finding is similar to the results obtained by Akbari *et al*. (2014).

According to Akbari *et al* (2014), the modulus rupture of boards tend to increase with the increase in the composition of NRL latex which can be explained to the improved adhesion between fiber matrix and the NRL adhesive. Besides that, as the concentration of NRL increases, the more sulphur will interact with NRL and more cross-linkage will be formed. Thus, these linkages can definitely help in the increase in the tensile modulus of the MDF boards, as the NRL composition is increased throughout the formulation (Nabil and Hanafi, 2016). In addition to that, the presence of starch above 5 g reduces the tensile strength of the board greatly. As shown in Figure 1, it is visible that as the composition of starch increases in the formulation of bioadhesives, the strength of the MDF produced deteriorated much and make the board weaker. When starch concentration increases, the dispersion of starch in NRL became tough and therefore, starch aggregates were formed easily. This situation is mainly contributed by the strong hydrogen bond that existed among the starch particles which then reduces the modulus tensile of the wood products.

Key Indicator:
A: UF
B: 20g NRL
C:15g NRL + 5g starch
D:10g NRL + 10g starch
E: 5g NRL+ 15g starch
F: 20g starch

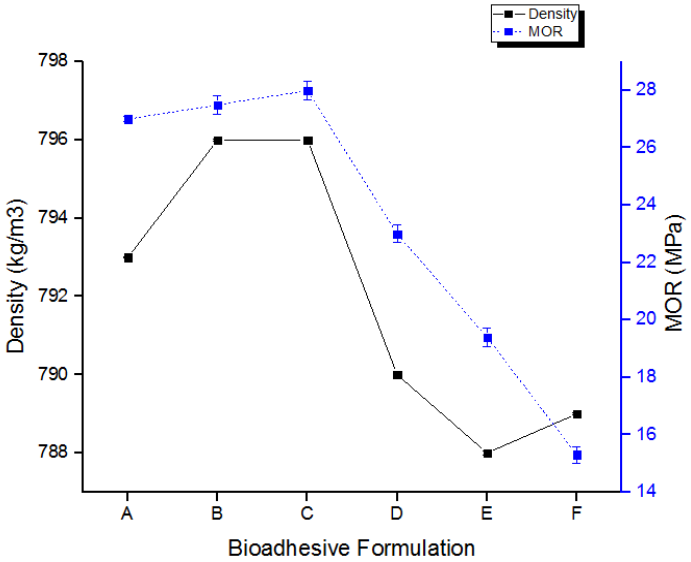

Fig. 1: Comparison of MOR values based on different composition of NRL and rice starch

Internal Bonding (IB):

Figure 2 depicts the intenal bonding values for MDF boards made from UF as well as from the bioadhesives with different composition of NRL and rice starch. The board made of 15g of NRL + 5g rice starch exhibited greater internal bonding value compared to UF and other formulations, which is 0.48 MPa. Besides that, adhesive formulation with only 20g of NRL and in absence of starch also gives comparable adhesion bonding strength like UF with approximately 0.44MPa. The findings of this study is in line with the results discussed by Akbari *et al*. (2014) although the decline in terms of internal bonding value obtained presently happened in gradual manner rather than sharp decline as showed by the referred authors. The better IB value was shown by boards made of 15g NRL+ 5g starch, where it can attributed to the higher level of tension concentration caused by better adhesion between adhesive and wood fibers. Besides that, the adhesion between the blend components of starch and NRL is proven to be great through the higher IB value recorded when starch content is between 5 to 10g range. According to Carvalho *et al*. (2003), this quality arose due to the presence of proteins and lipids at the surface region of the rubber particles in the latex, which played the role of a compatibilizer between starches which is a polar matrix while the rubber is a non-polar material. However, as the amount of starch present in the formulation increases in the bio-adhesive formulations, it can be observed that there is reduction in term of bonding strength on the respective boards.

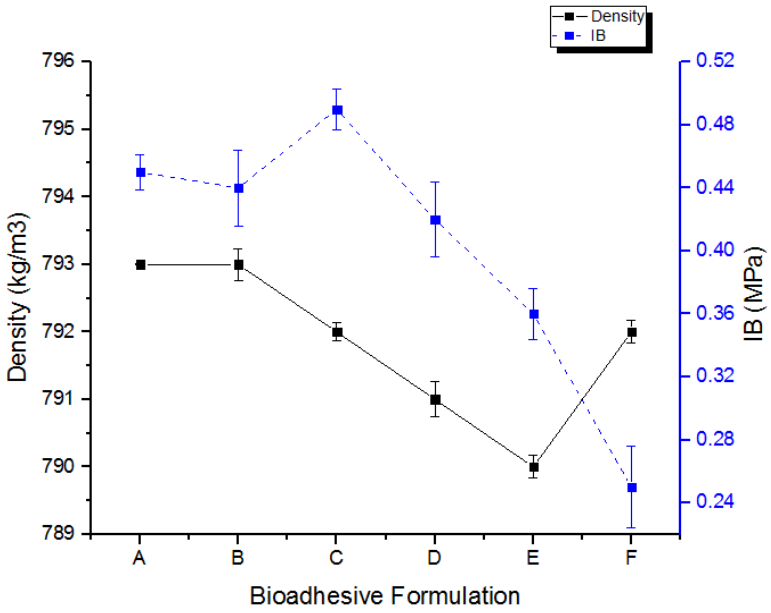

Fig. 2: Internal bonding values for MDF boards with different bio-adhesive formulations

Increase in the concentration of starch present in the bio-adhesive caused the increase in the chance for agglomeration which leads to the region of stress concentrations on the board structure. Thus, less energy is needed to elongate the crack propagation; hence internal bonding gets weaken easily. Therefore, a blend of 15g of NRL and 5g of starch makes up to be the best formulation of bio adhesives that can yield better performance in terms of bonding strength compared to UF in this aspect.

Thickness Swelling (TS):

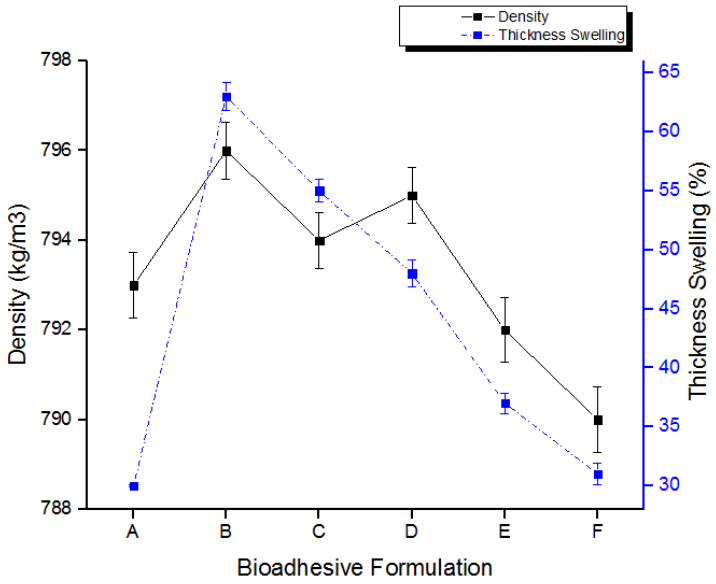

Fig. 3: Thickness Swelling of MDF boards with different bioadhesive compositions

Based on Figure 3, the thickness swelling can be categorized as poor for all the boards with adhesive formulations having higher NRL content. As the concentration of NRL decreases, the thickness swelling is improving as the concentration or proportion of starch is added on. Studies revealed that presence of any starch based materials into wood composites showed lower water uptake capacity (Agnantopoulou *et al*., 2012). Thus, this explains the better performance exhibited by the bio-adhesives with higher content of starch in this study.

Conclusion:

This study investigated the compatibility of NRL and rice starch to be formulated as a green adhesive for wood product manufacturing industry as an alternative to UF. The effect of varying composition of NRL and starch were incorporated throughout this research in order to identify their corresponding effects on the mechanical properties of MDF boards produced. From the results obtained, it is clear that the blend of 15g of NRL and 5g of starch gives the best result for MOR and IB, whereas for thickness swelling, pure starch based adhesive showcased a better performance. The most importantly, stronger interfacial bonding between the fiber matrices and the bioadhesive compositions' contributed to the improved mechanical properties of the boards. Besides that, the role played by the non-rubber constituents of NRL as the blending agents with starch did make a significant impact on the quality of the adhesion in this research. Thus, it is clear that the direct use of natural rubber latex without removing its non-rubber constituents are promising since they did play a very positive role in providing better mechanical properties. In addition to that, starch and NRL are compatible enough to be used as an alternative to UF, provided starch is only used lesser than 10g weight from the 10wt% of adhesive applied to the board, so as to prevent agglomeration from deteriorating the whole adhesion mechanism.

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