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Effect of waste rubber powder as filler for plywood application

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The study investigated the suitability of waste rubber powder (WRP) use as filler in adhesive formulation for plywood application. Melamine Urea Formaldehyde (MUF) was employed as resin for formulating the wood adhesive. To improve chemical properties and bonding quality of adhesive, WRP was treated by different chemicals like 20% nitric acid, 30% hydrogen peroxide and acetone solution. The treated WRP were analysed by XRD and it showed that inorganic compounds were removed and carbon was remained as major component under the treatment of 20% HNO_3. The treatment improved the mechanical properties like shear strength and formaldehyde emission of plywood (high shear strength and low formaldehyde emission). The physico-chemical interaction between the wood, resin and filler was investigated using fourier transform infrared spectroscopic (FTIR) technique and the interactions among N-H of MUF and C=O of wood and WRP were identified. The morphology of wood-adhesive interface was studied by field emission scanning electron microscope (FESEM) and light microscope (LM). It showed that the penetration of adhesives and fillers through the wood pores was responsible for mechanical interlocking. Therefore, chemically treated WRP proved its potential use as filler in MUF based adhesive for making plywood.

Keywords: waste rubber powder, melamine urea formaldehyde resin, filler, wood adhesive, plywood.

INTRODUCTION

Malaysia is one of the member countries of top ten wood suppliers in the world and mainly it exports wood product to Europe, Japan, Taiwan, Singapore and Middle East. The volume of export of plywood by Malaysia was USD 1.57 billion in 2012[1]. At present, most of the plywood adhesives are formaldehyde based product such as phenol formaldehyde (PF), urea formaldehyde (UF) and melamine urea formaldehyde (MUF)[2–4]. The emission of formaldehyde occur mainly from breakdown of formaldehyde based resin in wood panels, which poses a great hazard to human health causing carcinogen[5]. The formaldehyde emission can be lowered by several methods, such as reducing formaldehyde ratio in composition[6] and additional formaldehyde scavengers to the resin[7]. However, the other physical and mechanical properties of wood panels are affected[8, 9]. Resin and filler is the main compound in adhesive formulation. Adhesive mixed with filler to increase the viscosity, control rheology and reduce raw material cost. Wood surface consists of small pores and filler is used to fill up them which help to increase the bonding between the components. Apart from this, filler also used to reduce the penetration of resin into small pores of the wood[10]. Several types of natural fillers were investigated by other researchers, such as wheat flour, corn starch flour, tapioca flour, sorghum flour, soybean meal, palm kernel meal and others[2, 3, 11, 12]. Due to high protein content in these fillers they can enhance the bonding interaction between adhesive and wood. Recently, Hojilla-Evangelista[11] mixed soybean meal (52.8% of protein) and PF to prepare an adhesive and found that the resultant glue has satisfactory bonding performance in plywood. In another study, Hojilla-Evangelista and Bean[12] successfully developed an adhesive system with using sorghum flour (12% of protein) and PF resin for plywood. However, those natural filler has the potential to be used in food sector for human or animal application. Therefore, non-food competing filler is essential for wood industry to substitute natural filler. In the past two decades, the production rate of waste tire rubber dramatically increased due to rapid development of vehicles. The accumulation of huge numbers of waste tire rubber leads to serious environmental issues. The rubber waste can be eliminate or minimize from the environment by reusing or recycling it[13]. In recent years, numbers of researches have been studying on application of waste rubber powder, especially on composite application[14–17]. Al-Tayeb, et al.[16] used waste rubber to partial replace sand and cement in concrete production. They found that, up to 20% of waste rubber loading by volume, given satisfactory properties of concrete. Besides that, Wu and Zhang[14] Wu and Zhang[17] developed new pulp sediment composite by using waste rubber powder and pulp sediment as matrix. They found that, the physical properties of waste rubber-pulp sediment composite were comparable with conventional pulp sediment composite. The new composite reduce the cost of sheet materials and benefit the environment and society. In this context, waste rubber powder was used as filler in MUF adhesive formulation. The aim of the present study is to investigate the influence of the resin WRP-MUF on the mechanical properties of plywood panel, such as shear strength and formaldehyde emission. The physico-chemical interaction between WRP, MUF and plywood was also studied. Chemical properties of WRP were closely observed before and after treatment to elucidate its role as filler in plywood formulation. The interlocking behavior of filler was also studied.

MATERIAL AND METHODS

WRP and Industrial flour (IF) preparation

WRP was collected from local industry. WRP was washed by DI water and dried in oven at 60°C for 16 h. After that, the WRP was grinded with a grinder (RETSCH Ultra Centrifugal Mill ZM 200) and sieved

by using sieving shaker (RETSCH, AS 200 Basic). The 40–50 μm size particles were separated and used as filler. The IF powder was provided by Shin Yang Chemical Sdn. Bhd. The raw IF was sieved by using 40–50 μm sieve shaker to obtain uniform fine powder and then stored.

Modification of WRP

Nitric acid and hydrogen peroxide treatment

The WRP was treated with 20% nitric acid and 30% hydrogen peroxide. A 100 g of WRP was placed in a 500 mL beaker in an ice bath. After that, the oxidizing agent (nitric acid or hydrogen peroxide) was introduced onto the WRP drop wise with continuous stirring until complete immersion of the WRP. The reaction mixture was stirred for 3 h at 100°C and room temperature for nitric acid and hydrogen peroxide, respectively. Both of the reaction mixture were covered and then left for 24 h at room temperature. The treated WRP was washed thoroughly with DI water until a neutral solution was obtained[18]. The WRP was dried in oven at 60°C for 16 h. The treated WRP was grinded and sieved by using 40–50 μm sieve shaker to obtain uniform fine powder and then stored.

Acetone treatment

Meanwhile, the WRP was treated by acetone for oil extraction in a Soxhlet extractor for 10 h. It was repeated three times to ensure the oil was fully removed. The oil free WRP was dried in oven at 60°C for 16 h[13]. The treated WRP was grinded and sieved by using 40–50 μm sieve shaker to obtain uniform fine powder and then stored.

MUF based resin preparation

The method of MUF resin preparation was adopted from Bono, et al.[19]. In brief, 268 mL of 37% formalin was poured in a 500 mL three-necked flask and then 147.50 g of melamine and 26.75 g of urea was added under vigorous stirring. The pH of the mixture was adjusted at 8.5 to 9.0 by adding NaOH (0.1 M) solution and temperature of mixture was maintained at 80°C. After 2 h of reaction the resin was formed and the reaction mixture was cooled down to 60°C. Additional amount of urea (36.75 g) was added into the resin solution with stirring and continued until the temperature fell down to the room temperature. The resin was stored in a closed container.

Adhesive production

The adhesive formulation was conducted by adding 300 g of MUF resin with 12 g of urea in a mixer (KHIND Model SM 210) for 5 min. Required amount of fillers were added to the above mixture and blended well for another 5 min. After that, 13 g of hardener (ammonium chloride) was added into the mixture and mixed for 5 min.

Type II plywood production

The type II plywood was prepared using Red-Meranti 300 mm x 300 mm x 3.3 mm veneer. In order to get consistence result, veneer was maintained at 10% moisture content and required amount of wood adhesive were used to produce plywood. The adhesive was applied on two sides of a core veneer using a glue spreader. The unfinished plywood was left at room temperature for 5 min, following cold pressed 9 kg/cm^2 for 20 min. After cold press, it was left free for 5 min then it was transferred to a hot press. During hot press at a pressure of 9 kg/cm^2, the temperature and the press time were varied in the range of 100–175°C and 50–450 s respectively. After the hot press, the plywood product was stored at ambient condition for further testing.

Fourier transform infrared spectroscopy (FTIR)

Fourier Transform Infrared (FTIR) Spectrophotometer (PerkinElmer) was used to investigate the interaction between WRP, wood and resin. The samples were mixed with KBr and FTIR spectrum was recorded in the range of 400–4000 cm^{-1}.

Shear strength test of plywood

The shear strength test of newly prepared type II plywood was determined by using the bonding test according to the JAS[20]. For each plywood panel, nine specimens (25 mm x 80 mm) were tested. Prior to the test, the specimens were soaked in a hot water bath at 100°C for 4 h. After that, the samples were dried at 60 ±5°C for 20 h and it was again soaked in the hot water bath at 100°C for 4 h. Furthermore, the samples were soaked in cold water bath at room temperature for 1 h. Samples at this stage was used for shear strength testing which was conducted by using of universal testing machine (Shimadzu, AGS-X Series).

Formaldehyde emission test of plywood

The formaldehyde emission test of type II plywood was done according to the JAS[20]. Total ten rectangular specimens with a length of 150 mm and width 50 mm were prepared from each plywood panel. A crystallizing dish with a diameter of 120 mm and a height of 60 mm was placed at the center of (inner volume 9–11 liters) desiccator. The crystallizing dish was filled with 300 mL of distilled water. The specimens were fixed apart from each other and hold by a metallic holder. The metalic holder was placed on the crystallizing dish for 24 h at 20°C. After 24 h, the formaldehyde concentration was measured in the distilled water using acetyl-acetone spectrophotometric analysis method as described by Nash[21]. The solution (25 mL) was put into a conical flask with a co-ground stopper and 25 mL of acetyl-acetone ammonium acetate solution was transfered into conical flask. The conical flask with a co-ground stopper was heated up to 65°C and kept for 10 min at that temperature. After cooling at room temperature, the solution was transferred into a quartz cuvette and the absorbance was measured at a wavelength of 412 nm using an UV-Vis spectrophotometer (Toshiba U1800).

X-Ray Diffraction (XRD)

XRD patterns of the samples were recorded on a Rigaku MiniFlex II operated at accelerating voltage of 30 kV and emission current of 15 mA with graphite-monochromatized Cu Kα radiation and scanning speed 1°/min. The scanning step size 0.02° was over a range of 2θ = 20–80°.

Surface morphology

Morphology of plywood was analyzed by using field emission scanning electron microscope (FESEM) and light microscope (LM) techniques. FESEM samples were prepared with the plywood which was cross-sectionally cut through the wood-adhesive interface using a sliding microtome at a thickness of 90 μm. The sections were then mounted on carbon tapes applied to stubs, coated with gold in a sputter coater and examined with a JEOL JSM-7800F FESEM. The other section was stained with aqueous toluidine blue stain and mounted in glycerol on a glass slide prior to analyze with LM.

RESULTS AND DISCUSSION

FTIR analysis

Figure 1 shows the FTIR spectra of untreated WRP, 20% HNO_3+WRP, 30% H_2O_2+WRP and Acetone-+WRP as i, ii, iii and iv respectively. The spectra of WRP with and without chemical treatment are closely matching at characteristic peaks of O-H and N-H groups at 3500–3300 cm^{-1} [3, 22, 23], the peaks at 2930–2918, 2853–2849 cm^{-1} correspond to the C-H asymmetric and symmetric stretching vibration of CH_2 respectively[3]. The peaks at 1638 cm^{-1} is attributed to C=O stretching and 1565 cm^{-1} vibration of 20% HNO_3 treated WRP is correspond to benzene ring skeleton. Besides that, a shoulder formed at 1424 cm^{-1} is attributed to the deformation of saturated C-H bond. The peak at 1149 cm^{-1} corresponds to the stretching of C-O bond of tertiary alcohol[3]. It is noted that in Figure 1, all spectra are more or less same, only spectrum of WRP treated with 20% HNO_3 exhibited minor shift for N-H and C=O groups from 3441 cm^{-1} and 1633 cm^{-1} to 3447 cm^{-1} and 1638 cm^{-1} respectively. As a result, a certain amount of blue shift observed for both the groups which indicating the functional groups were in freer environment[3]. Meanwhile, the peaks of untreated WRP, 30% H_2O_2 treated, and acetone treated WRP were unchanged. Therefore, 20% HNO_3 treated WRP was chosen for the subsequent inspection.

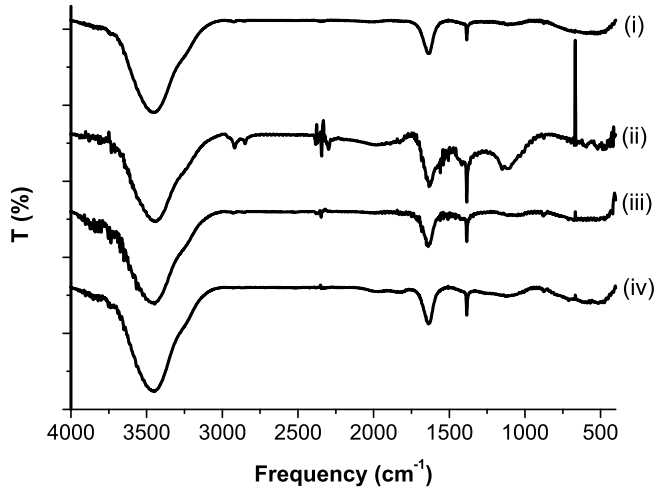

Figure 1. FTIR spectra of WRP with and without chemical treatment [Press conditions: Temperature = 150°C; Time = 250 s; pressure = 9 kg/ cm^2; (i) = WRP, (ii) = 20% HNO_3 WRP, (iii) = 30% H_2O_2, (iv) = Acetone]

The FTIR spectra of 20% HNO_3 treated WRP, Wood and MUF are shown in Figure 2a and the spectra illustrated a number of absorption peaks, representing the multipart nature of the substance. The FTIR spectrum proved that broad bands at 3500–3300 cm^{-1}, correspond to the hydrogen bonded O-H and N-H groups[3, 22, 23]. The bands observed at 2930 cm^{-1} is assigned for stretching frequency of C-H[24]. The peaks at 1638 cm^{-1} is attributed to C=O stretching, which may due to partial oxidation of WRP during the grinding. The peaks of WRP at 1565 cm^{-1} may be due to the benzene ring skeleton vibration and a shoulder formed at 1424 cm^{-1} which lead to the deformation of saturated C-H bond. The peak at 1149 cm^{-1} corresponds to the stretching of C-O bond of tertiary alcohol[3]. The peak observed at 1543–1498 cm^{-1} corresponds to the NH_2 scissoring vibration[23, 25] and which may come from the protein part of wood and amide fraction of MUF[3].

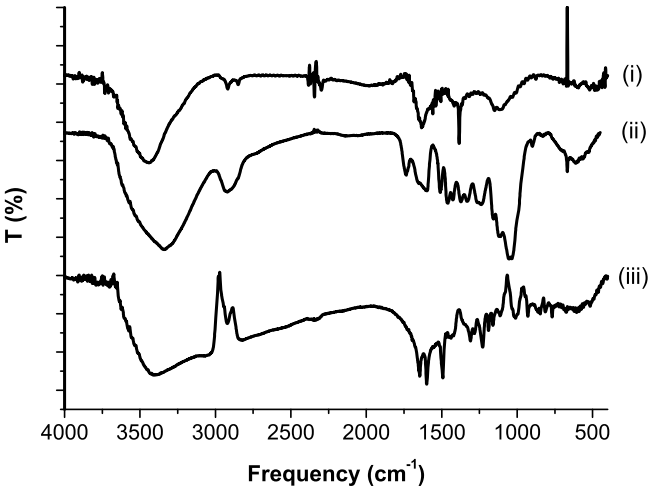

Figure 2a. FTIR spectra of 20% HNO_3 WRP, Wood and MUF [Press conditions: Temperature = 150°C; Time = 250 s; pressure = 9 kg/cm^2; (i) = 20% HNO_3 WRP, (ii) = Wood, (iii) = MUF]

The FTIR spectra of three blends consisting of 20% HNO_3+WRP, wood and MUF are shown in Figure 2b, which exhibit a number of absorption peaks, indicating the complex nature of the materials with the existence of some interactions. The carbonyl C=O group of wood is more reactive compare to the C=O containing functional groups. From spectra of 2a-ii and 2b-i, it is observed that after blend formation, the stretching vibration of C=O (for wood) and C-O (for 20% HNO_3+WRP) groups are shifted from 1740 cm^{-1} and 1149 cm^{-1} to 1637 cm^{-1} and 1083 cm^{-1}, respectively. A significant amount of red shifts were observed for both groups. Therefore, considering the Figures 2a–i,ii and 2b–i, it is possible to summarize that in this blend (20% HNO_3+WRP-wood) the existing interactions are involved between C-O group of 20% HNO_3+WRP and C=O group of wood. Besides that, from 2b-ii it is clear that after blend formation, the stretching vibration of C=O (for 20% HNO_3+WRP) group is shifted from 1633 cm^{-1} to 1638 cm^{-1}. Therefore, a remarkable amount of blue shift is observed for C=O groups. For instance, in MUF, the C=O group experienced some extent of resonance effect because it is flanked by two NH groups like this way –HN-CO-NH-[3]. However, the C=O group in 20% HNO_3+WRP may

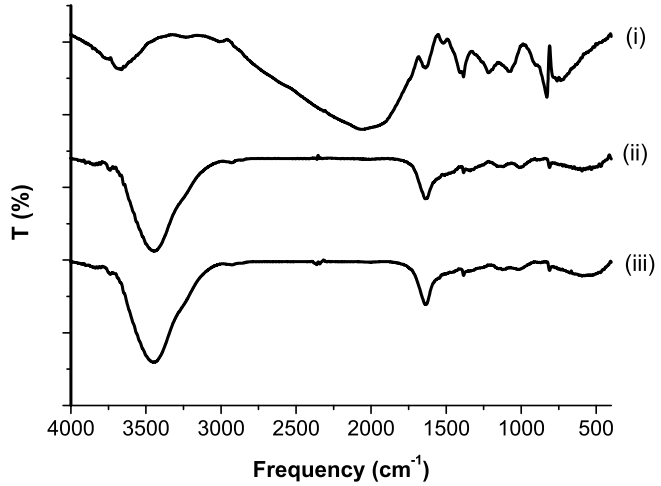

Figure 2b. FTIR spectra of 20% HNO_3 WRP + Wood, 20% HNO_3 WRP+MUF and 20% HNO_3 WRP + Wood + MUF [Press conditions: Temperature = 150°C; Time = 250 s; pressure = 9 kg/ cm^2; (i) = 20% HNO_3 WRP + Wood, (ii) = 20% HNO_3 WRP + MUF, (iii) = 20% HNO_3 WRP + Wood + MUF]

come from oxidation, where the –COOH group residing in axial position with lesser stability. As a result, more reactive C=O group of 20% HNO_3+WRP may involve in an interaction with the N-H of MUF with a greater degree. Although, there is some blue shift observed in 20% HNO_3+WRP, but the red shifts in MUF is higher and it may allow stronger interaction. Summarizing the Figures 2a-i,iii and 2b-ii, it can be concluded that in this blend of 20% HNO_3+WRP-MUF existing interactions are involved between the C=O group of 20% HNO_3+WRP and N-H group of MUF. Furthermore, considering the Figsures 2a–i,ii,iii and 2b–iii, it can be stated that interaction is existed between the blend of 20% HNO_3+WRP-Wood--MUF and that of 20% HNO_3+WRP-MUF. Additionally, the C=O groups of wood are involved in the interaction as in most of the cases red shift is observed.

XRD analysis

Figure 3 presents XRD pattern of WRP for without treatment and treated with 20% HNO_3. In both pattern, the prominent peaks was observed at 2θ values from 15 to 25° correspond to the amorphous carbon[26]. In WRP pattern (Fig. 3i), the peaks at ca. 36.77°, 32.34° and 47.71° correspond to ZnO, CaO and $Ca_2SiO_4 \cdot 3H_2O$, respectively[27, 28], while MgO is identified at 29.66° and 43.41°. As it can be seen that, after treatment the inorganic compound peaks were disminised (Fig. 3ii), carbon amorphous and MgO were remained. The obtained result was in agreement with Alexandre-Franco, et al.[28], which also reported that HNO_3 treatment played good rules to remove inorganic content. They also observed that after HNO_3 treatment the main component remain were carbon and MgO.

Effect of chemically treated WRP

Effect of various chemically treated WRP in wood adhesive on the shear strength and formaldehyde emission of plywood were presented in Fig. 4. The results illustrated the average shear strength values of the WRP and the value for 20% HNO_3 treated WRP was significantly higher than other treated, untreated WRP and IF. This observation matches with the results presented

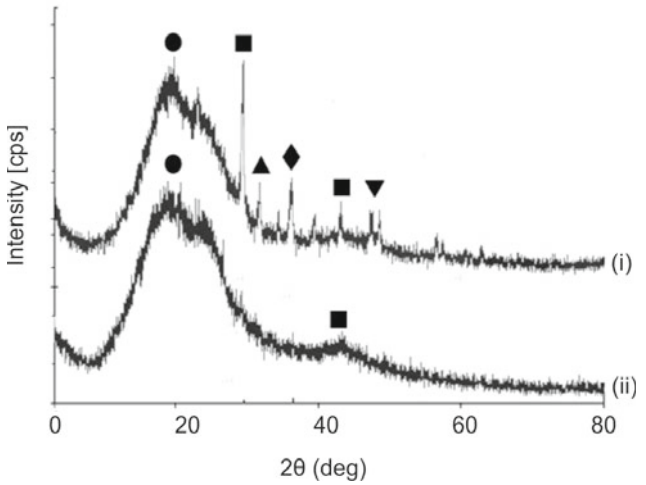

Figure 3. XRD pattern of WRP treated with HNO_3 and without treated [(i) = WRP, (ii) = HNO_3 treated WRP; ● Carbon, ■ MgO, ◆ ZnO, ▲ CaO, ▼ $Ca_2SiO_4 \cdot 3H_2O$]

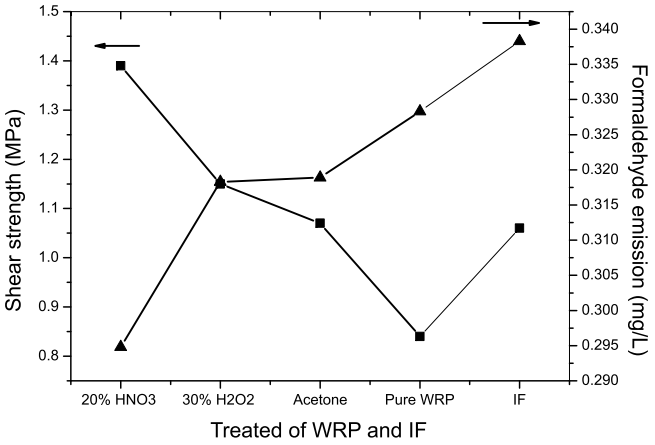

Figure 4. Effect of chemically treated and untreated waste rubber powder (WRP) on shear strength and formaldehyde emission [Press conditions: Temperature = 150°C; Time = 250 s; pressure = 9 kg/cm^2; ■ = shear strength; ▲ = formaldehyde emission]

by Darmawan, et al.[29] Kumar, et al.[30], who reported that additional activated charcoal as filler in wood composite enhanced the bonding strength of wood panel. Mansouri and Pizzi[31] also described that supplement of filler can enhance the properties of plywood significantly.

In addition, the average value of formaldehyde emission of HNO_3 treated sample was relatively low compared to others. It was also observed that, all series of treated and untreated WRP wood panel having low formaldehyde emission compared to IF. These may be due to the presence of carbon phases in WRP, which enable to absorb formaldehyde in wood panel. Similar findings were described by Darmawan et al.[29] Kumar et al.[30] where they reported the use of activated carbon as formaldehyde absorbent that resulted the reduction the formaldehyde emission from wood panel. Besides that, Pizzi[10] mentioned that good bonded wood panel released less formaldehyde because of mechanical interlocking between veneer and core adhesive, which bond is quite high and small pores on surface of the plywood are crammed with filler.

Effects of hot press temperature and time

The effect of hot press conditions on the shear strength and formaldehyde emission of plywood samples is presented in Figures 5a and b. They illustrated that the average shear strength value significantly increased from 0.99 MPa to 1.39 MPa when hot press temperature was raised from 100 to 150°C. Thereafter, the average shear strength value significantly decreased to 1.19 MPa at 175°C. The average formaldehyde emission value was significantly decreased from 100 to 125°C, thereafter it decreased slowly with further increase of temperature. Therefore, based on the obtained results the temperature of 150°C was considered as optimum temperature. According to Figure 5b, with the increase in press time the shear strength increased and reached maximum at 250 s, thereafter it was decreased with further increase of time. The formaldehyde emission was decreased constantly from 50 to 450 s. Therefore, based on the results obtained, the press time was optimized at 250 s. The results obtained were in agreement with the work presented by Ong et al.[2] Bono et al.[32] which reported similar trend of experimental data.

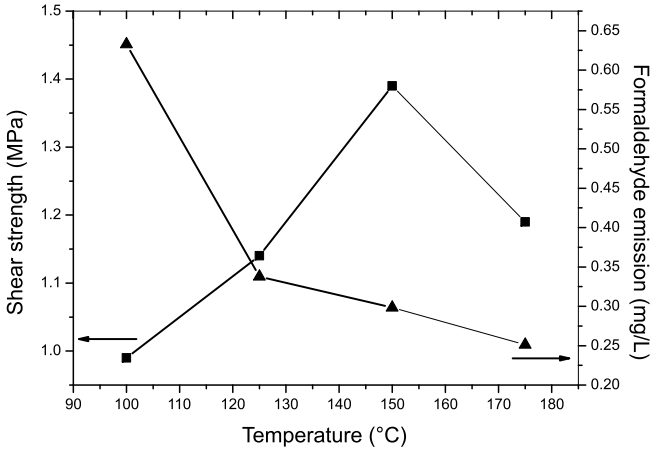

Figure 5a. Effect of hot press temperature on the shear strength and formaldehyde emission [Press conditions: Time = 250 s; pressure = 9 kg/ cm^2; ■ = shear strength; ▲ = formaldehyde emission]

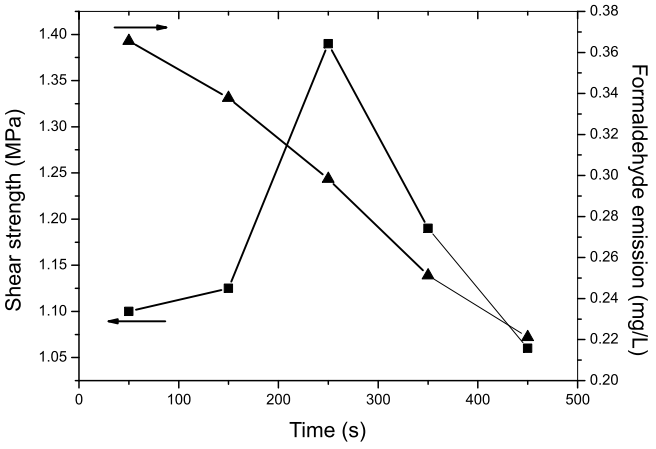

Figure 5b. Effect of hot press time on the shear strength and formaldehyde emission [Press conditions: Temperature = 150°C; pressure = 9 kg/cm^2; ■ = shear strength; ▲ = formaldehyde emission]

Effect of filler concentration

The average shear strength value was significantly increased when the filler concentration increased from 0 to 13% (Fig. 6) thereafter, with further increase of fillers concentration until 23%, the shear strength value was markedly decreased. Furthermore, the average formaldehyde emission was pointedly decreased when fillers concentration increased from 0 to 18%, whereas with further increase of fillers concentration up to 23%, the formaldehyde emission was increased. However, the formaldehyde emission for 13 and 18% were more or less the same. Therefore, 13% concentration of fillers was considered as optimum level.

Figure 6 confirmed that the concentration of filler (in the adhesive formulation) played an important role for adhesion properties. Similar phenomena was reported by Mansouri and Pizzi[31], Babcock and Smith[33], Qiao et al.[34]. A very high concentration of filler caused the blockage of the pores resulting in less penetration of resin in the wood surface and contact area decreased between the wood and adhesive layer. This led to poor interlocking effect. Moreover, due to high loading of fillers (>18%) the adhesive distribution has become highly heterogeneous.

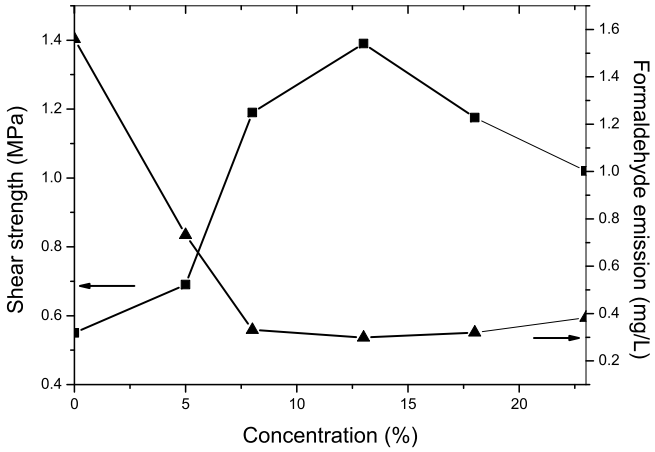

Figure 6. Effects of filler concentration on shear strength and formaldehyde emission of the plywood [Press conditions: Temperature = 150°C; Time = 250 s; pressure = 9 kg/ cm^2; ■ = shear strength; ▲ = formaldehyde emission]

Morphology of plywood cross section

The plywood-adhesive interface was investigated using FESEM and the result is presented in Figure 7a. The pathway of the adhesive penetration (white arrow) into wood tissues of plywood is visible. Though FESEM was able to visualize the adhesive distribution within wood tissues[35, 36], unfortunately, FESEM was unable to identify the presence of WRP. Therefore, LM was employ to further visualize the distribution of the adhesive within the wood tissues (Fig. 7b). In comparison with FESEM, excellent differentiation between wood, resin and WRP was identified with LM image provided different color. It is clearly shown that, the WRP was able to penetrate into wood pores and form blockage. It could be concluded that excellent mechanical interlocking was achieved with the presence of WRP. The results obtained were in agreement with the work presented Kim et al.[37] which

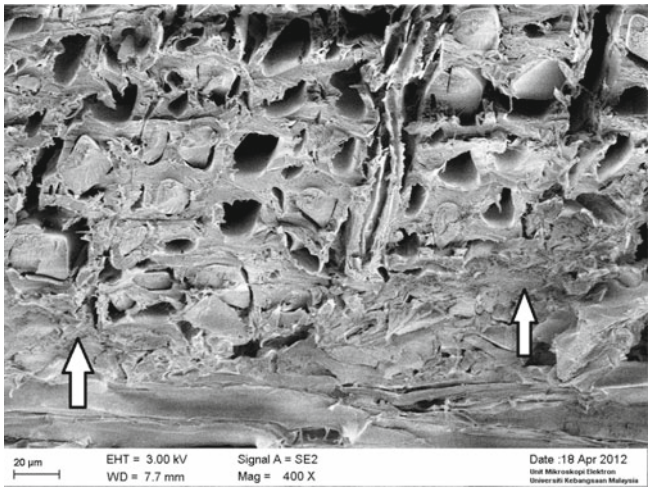

Figure 7a. Field emission scanning electron micrograph (FESEM) of plywood compound with 13% of WRP at 400x [Press conditions: Temperature = 150°C; Time = 250 s; pressure = 9 kg/cm^2]

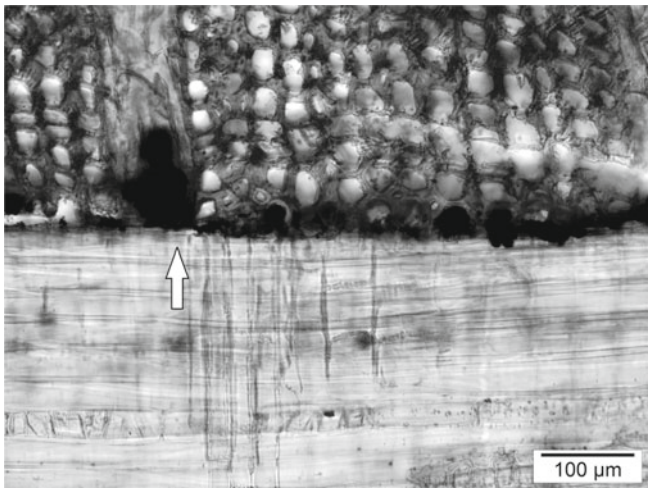

Figure 7b. Light micrograph (LM) (50µm) of plywood compound with 13% of WRP [Press conditions: Temperature = 150°C; Time = 250 s; pressure = 9 kg/cm^2]

observed the glue line between the veneers and the voids in wood near the glue line were filled with adhesive.

CONCLUSIONS

This study demonstrated that chemically treated WRP as filler in MUF based adhesive could be used for making type II plywood. The MUF resin-WRP-Wood blend components were effectively miscible in the amorphous phase among all the blends compositions used. The FTIR analysis revealed strong inter-association hydrogen bonds between the amine groups of the MUF resin, carbonyl groups of WRP and wood. XRD analysis confirmed that, after 20% HNO_3 treatment, inorganic compounds were removed and carbon was the major residue in treated WRP. HNO_3 treated WRP exhibited greater shear strength and lowest formaldehyde emission compared to others treated WRP and IF. The effect of hot press time and temperature were investigated, where 250 s and 150°C were found to be optimum. Under these conditions, filler concentration of 13% showed better result. FESEM and LM images confirmed that mechanical interlocking was established within WRP and wood surface blocking the pores.

ACKNOWLEDGMENT

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Nomenclature

WRP	– Waste rubber powder
MUF	– Melamine urea formaldehyde
PF	– Phenol formaldehyde
UF	– Urea formaldehyde
IF	– Industrial flour
h	– Hour (Time)
μm	– Micro-meter (Length)
g	– Gram (Weight)
mL	– Milliliter (Volume)
M	– Molarities (Concentration)
mm	– Millimeter (Length)
min	– Minute (Time)
Kg/cm^2	– Kilogram/centimeter^2 (Force)
s	– Second (Time)
cm^{-1}	– Centimeter^{-1} (Wavelength)
nm	– Nanometer (Wavelength)
kV	– Kilo-voltage
mA	– Mili-ampere

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