Effect of Sepiolite Filler in Malamine-Urea-Formaldehyde Resin on the Properties of Three-ply Plywood

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A low-cost sepiolite (SEP) was used to replace wheat flour (WF) as a filler applied to melamine-urea-formaldehyde (MUF) resin. Three-ply plywood was fabricated with different SEP/WF formulations, and its wet shear strength and formaldehyde emission were tested. Scanning electron microscopy (SEM) and X-ray diffraction (XRD) were used to explain why the performance of the plywood improved. Results showed that the wet shear strength of the plywood increased with the addition of SEP in the filler. When using SEP to replace 80% of the WF, the wet shear strength was improved by 27.8%, which was attributed to the penetration network with hydrogen bonds formed by SEP and MUF resin. Using SEP in the filler formulation created a smoother, denser, and more regular cross-section to prevent moisture intrusion, which improved the water resistance of the adhesive and thus further increased the wet shear strength of the resultant plywood. The tunnel structure of SEP could accelerate the free formaldehyde emission of the plywood. As a result, using SEP replace 80% WF as a filler, the formaldehyde emission of the plywood was reduced by 7.8% due to the tunnel release effect.

Keywords: Sepiolite; Melamine-urea-formaldehyde (MUF) resin; Wheat flour; Filler

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INTRODUCTION

Sepiolite (SEP) is a clay mineral with the formula $Si_{12}Mg_8O_{30}(OH)_4(H_2O)_4 \cdot 8H_2O$; it belongs to the structural family of 2:1 phyllosilicates (Chen *et al*. 2007; Alver *et al*. 2008). It has three main characteristics. Firstly, it has a unique needle-like fiber structure. When dispersed in water or another polar solvent, the fiber becomes loose rapidly, forming a monomer fiber or small fiber bundles. These fibers disperse irregularly and form a messy framework. Secondly, it has a tunnel-like micro-pore channel structure. SEP shows an alternation of blocks and tunnels that grow in the direction of the fiber. Each structural block is composed of two tetrahedral silica sheets sandwiching a central sheet of magnesium oxide-hydroxide. The discontinuity of the silica sheets gives rise to the presence of silanol groups (Si-OH) at the edges of the tunnels, which are the channels open to the external surface of SEP particles (Bidsorkhi *et al*. 2014; Soheilmoghaddam *et al*. 2014). Thirdly, it has a strong adsorption capability. SEP has a high specific surface area value, not only because of the small size of its particles, but also because of its fibrous morphology and intra-crystalline tunnels. Its particular structure with tunnels increases the specific surface area of SEP up to 320 m^2g^{-1}.

SEP has a wide range of industrial applications based on its physicochemical properties, especially its surface properties. SEP can be used as an adsorbent material

(Ueda and Hamayoshi 1992), catalyst or catalyst carrier (Blanco *et al*. 1994), or thermal insulation material (Shimizu *et al.* 2004; Bokobza and Chauvin 2005; García *et al.* 2009; Samper-Madrigal *et al.* 2015). The clay can also be used to reinforce polymers because of its large interface area, which results in strong interaction between the polymer matrix and the nanofiller (Kader *et al.* 2005; Sangerano *et al.* 2009; Chivrac *et al.* 2010; Chen *et al.* 2011; Idrus *et al.* 2011). However, studies using SEP as a wood adhesive filler are relatively scarce.

Formaldehyde-based adhesives remain dominant in the plywood manufacturing industry. When processing plywood, 25% to 30% of wheat flour (WF) is mixed with the resin. Wheat four contains water-soluble/or swellable pentosans (Ding *et al.* 2013; Vinkx *et al*. 1993), which can improve the viscosity of the resultant glue. In other words adding wheat flour can prevent excessive penetration of adhesive into wood surfaces and improve pre-press strength of the resultant plywood. In the year 2013, WF consumption in the plywood industry was more than one million tons, which is a huge waste of resources. Therefore, it is important to develop new fillers to substitute for WF in the plywood industry. Although SEP doesn't contain pentosans, the needle-like fiber structure and large interface area of it may allow SEP to replace WF as a formaldehyde-based resin filler for plywood fabrication.

In our preliminary research, the performance of a urea formaldehyde resin was improved using SEP as a filler because of the penetration network and cellular structure formation. However, currently MUF resin is widely used in the plywood industry because of its low formaldehyde emissions. Compared with UF resin, MUF resin has a higher degree of crosslinking because of the ring structure of melamine (Gao *et al.* 2012b; Zhang *et al.* 2013; Luo *et al.* 2015). It is meaningful and more practical to study whether SEP can enhance the crosslinking of MUF resin and develop a new and practical method for improving the performance of the MUF resin and reducing its costs.

In this study, the performance of SEP as MUF resin filler in the plywood industry was investigated. The properties of SEP and the resultant adhesives were analyzed by scanning electron microscopy (SEM) and X-ray diffraction (XRD). Three-ply plywood was fabricated and measured to evaluate wet shear strength and formaldehyde emissions.

EXPERIMENTAL

Materials

SEP (2000 RMB per ton) was purchased from the Longtong Sepiolite Factory, Hebei Province, China. Solid urea and melamine were industrial reagents obtained from Lanyi Chemical Co., China. Formaldehyde (aqueous solution, industrial grade, 37%) was purchased from the Guangdong Xilong Chemical Factory, China. Sodium hydroxide, formic acid, and ammonium chloride (NH_4Cl, analytical grade) were obtained from the Beijing Chemical Factory, China. Wheat flour (3400 RMB per ton) was obtained from the Beijing Guchuan Flour Company, China. Poplar veneers with 8% moisture content were purchased from Hebei Province, China.

Methods

Preparation of MUF resin

MUF resin was synthesized using formaldehyde, urea, and melamine at weight ratios of 19.47:12.87:1 in the laboratory following the "alkali-acid-alkali" three reaction

stages (Ding *et al*. 2013). Urea was added three times at weight ratios of 2.64:1.18:1. The melamine was added twice at weight ratios of 1:2 following the first and second additions of urea. In the first stage, formalin (37% solution of formaldehyde) was poured into the reactor, followed by the first portions of urea and melamine. Sodium hydroxide (30% wt. solution) was added to adjust the pH of the mixture to 7.5 to 8.0. In the second stage, the mixture was heated to 90 °C and maintained for 1 h. The acidic reaction was brought about by adding formic acid (20% wt. solution) to obtain a pH of about 5.0, and the condensation reactions were carried out until reaching a target viscosity. Then, the mixture was adjusted to a pH of 7.5 to 8.5. Finally, in stage three of resin synthesis, the second portion of urea and melamine were added. After 0.5 h at 85 °C, final mole ratios 1.08 of MUF resins were adjusted by adding the third portion of urea and further stirring at 70 °C for 0.5 h. Then, the MUF resin was cooled to room temperature, and its pH was later adjusted to 8.0.

MUF resins were prepared using 100 g of MUF resin, 25 g of SEP-based fillers, and 0.6 g of ammonium chloride by mechanical blending. The formulations of SEP-based fillers are shown in Table 1.

Table 1. The Formulations of SEP-based Fillers

No.	SEP (%)	WF (%)
A1	0	100
A2	40	80
A3	80	40
A4	100	0

Preparation of three-ply plywood

Poplar veneers (8% moisture content) with dimensions of 400 × 400 × 1.5 mm were used to prepare three-ply plywood panels. The resins were applied to both sides of each veneer at a spread rate of 350 gm^{-2}. The adhesive-coated veneer was then stacked between two uncoated veneers with the grain directions of two adjacent veneers perpendicular to each other. The stacked veneers were hot-pressed at 1.0 MPa and 120 °C for 6 min. The samples were maintained at room temperature for 24 h before the evaluation of wet shear strength and formaldehyde emissions.

Scanning electron microscopy (SEM)

Samples were placed into a piece of aluminum foil and dried in an oven at 120 ± 2 °C until reaching a constant weight, then sputter-coated with gold using an E-1010 Hitachi Ion Sputter (Japan). A Hitachi (Hitachi Science System, Ibaraki, Japan) scanning electron microscope was then used to observe the samples.

X-ray diffraction (XRD)

Adhesives were cured in an oven at 120 ± 2 °C until reaching a constant weight, then ground into powder. XRD patterns were recorded on an XRD diffractometer (XRD-6000, Shimadzu, Kyoto, Japan) using a cobalt source and 0.2-theta scan ranging from 5° to 60° at 45 kV and 30 mA. The relative crystallinity index (Cr_I) was directly calculated by the measurement instrument using Eq. 1,

$$Crl=A_c\times(A_c+A_a)^{-1} \quad (1)$$

where A_c is the area of the crystalline region and A_a is the area of the amorphous region. Three replicates were gathered for each resin composition.

Water resistance measurement

The water resistance of each adhesive was measured by testing its resultant plywood in accordance with the National Standard (GB/T 17657-2013). Plywood specimens (25 × 100 mm) were soaked in water at 63 ± 2 ºC for 3 h, then cooled to room temperature for 10 min before tension testing. Six specimens were tested for each sample, and the results were averaged.

Formaldehyde emission measurement

The formaldehyde emissions of plywood were determined using the desiccator method in accordance with the procedure described in the China National Standard (GB/T 17657-2013). After the completed three-ply plywood was stored for 24 h, the plywood was cut into specimens with dimensions of 10 × 50 × 150 mm. Ten specimens per panel were put into a sealed desiccator with a diameter of 240 mm at 20 ± 2 °C for 24 h. The formaldehyde emitted was absorbed by 300 mL of deionized water in a container, and formaldehyde concentration in sample solutions was determined using acetyl acetone-ammonium acetate solution and the acetyl-acetone method, with colorimetric detection at 412 nm.

RESULTS AND DISCUSSION

SEM Analysis

Adhesive filler is widely used in the plywood industry for reducing adhesive cost and preventing adhesive penetration into the wood. It also fills up tiny holes in the wood surface to avoid bonding failure. SEM images of WF and SEP are shown in Fig. 1.

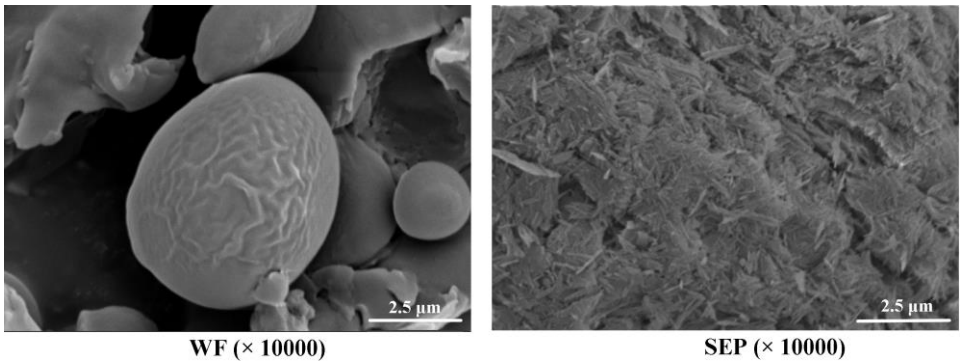

Fig. 1. SEM images of the WF and SEP

WF was composed of ellipsoid starch granules with a smooth surface. SEP was composed of irregular needle-like aggregates. The needle-like particle size of SEP increased the specific surface area and was easily dispersed in the water or other polar solvent to form a messy framework, as was shown earlier in the work Huang and Kavas

and their co-authors (Huang *et al.* 2012; Kavas *et al.* 2004). Therefore, the MUF resin molecule might fill in the framework and the needle-like aggregates may act as a bridge joint between SEP and the MUF resin. As a result, the wet shear strength of the resultant plywood could be improved (Li *et al.* 2014).

XRD Analysis

XRD patterns of the SEP and MUF resins are shown in Fig. 2. The strong peaks appearing at 8.44, 26.63, and 30.95° correspond to the (110) plane of SEP (Suárez and García-Romero 2012). These peaks also appeared in the MUF resin with 80% SEP + 20% WF, which indicated that the crystalline morphology of SEP was not destroyed (Lei *et al.* 2008). The crystallinity of the cured MUF resin with 80% SEP + 20% WF was 52.4%, which was 10% higher than that of the cured MUF resin, with 100% WF (42.6%). Park and Causin (2013) showed that formaldehyde emission and crystallinity of the resin had an inverse relation. So the higher crystallinity of MUF resin with SEP maybe led to lower formaldehyde emission of the resultant plywood. Also, no new crystallization peak was observed in the X-ray diffraction patterns, which indicated that no new crystal structure was formed as a results of adding SEP to MUF resin.

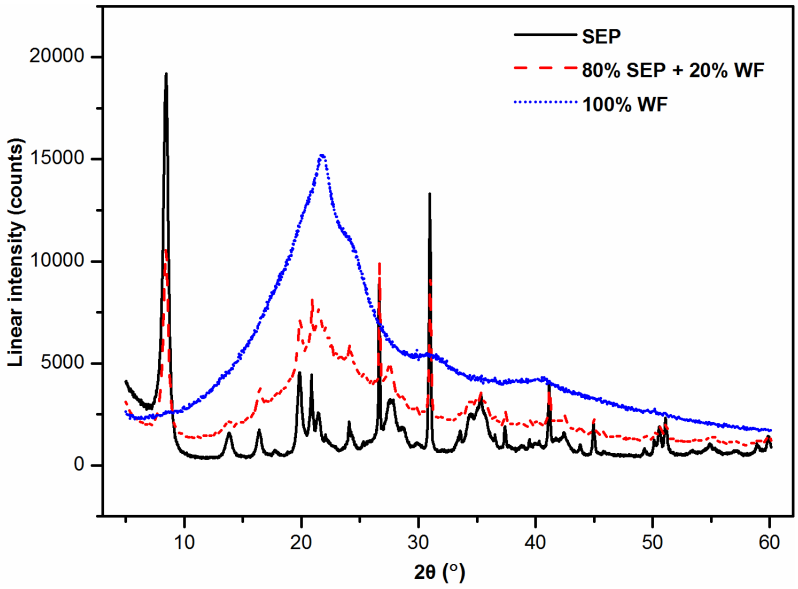

Fig. 2. XRD patterns of the SEP and MUF resin with various filler formulations: (A1) 100% WF and (A3) 80% SEP + 20% WF

Wet Shear Strength Measurement

Plywood was prepared using the MUF resin with SEP-based fillers. The influences of various filler formulations on the wet shear strength of the resultant plywood are shown in Fig. 3. All the wet shear strengths were above 0.7 MPa and increased with SEP addition. When using 80% SEP to substitute WF, the wet shear strength of the resulting plywood was increased by 27.8% compared with that of 100% WF. This could be attributed to the micro-fibrous morphology of the SEP, which caused the MUF resin molecule to fill in the SEP framework during the curing process (Kavas *et al.* 2004). Therefore, the MUF resin molecule was distributed more homogeneously, which could reduce the interior stress of the adhesive layer. Another possible reason was

that the SEP with fibrous morphology could form an interpenetrated network with the MUF resin system by hydrogen bonds during the curing process, which further improved the wet shear strength of the produced plywood. These results were consistent with the discussion in the above inferences of SEM and XRD analyses. However, when adding 100% SEP, the wet shear strength decreased to 0.92 MPa. The main reason was that WF could absorb water and swell in the MUF resin, then become viscous, which could prevent the MUF resin molecule from penetrating into the wood surface (Vinkx *et al*. 1993; Ding *et al.* 2013). However, the SEP did not possess those properties. Thus, using 100% SEP filler led to an excessive penetration of the resin into the veneer. As a result, the wet shear strength of plywood bonded by MUF resin with 100% SEP decreased.

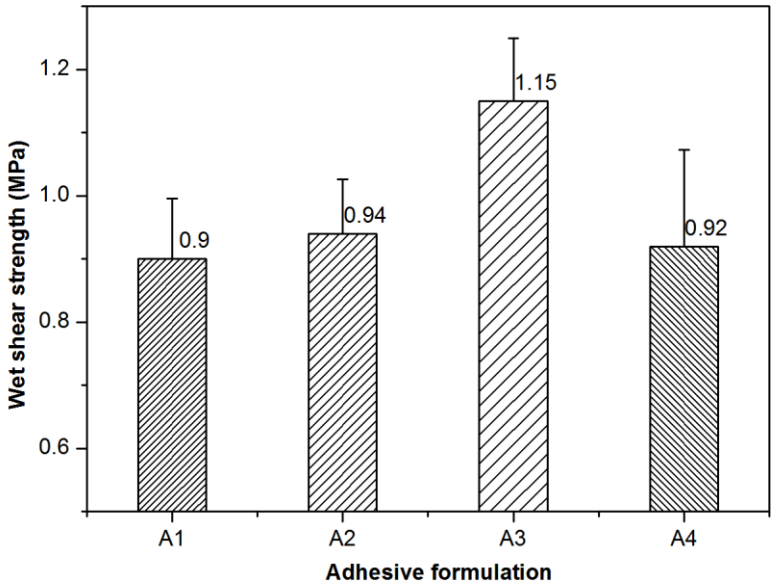

Fig. 3. The wet shear strength of plywood bonded by MUF resin with various filler formulations: (A1) 100% WF, (A2) 40% SEP + 60% WF, (A3) 80% SEP + 20% WF, and (A4) 100% SEP

Formaldehyde Emissions

The formaldehyde emissions of the plywood bonded by MUF resin with various SEP-based fillers are shown in Fig. 4. The formaldehyde emissions increased with the addition of SEP. Especially, the formaldehyde emission of the plywood bonded by MUF resin with 80% SEP (0.84 mg/L) was 12% higher than that with 100% WF (0.75 mg/L). This results didn't correspond to the deduction of the XRD analysis. The results of XRD analysis showed that higher crystallinity of MUF resin with SEP should lead to lower formaldehyde emission of resultant plywood. This opposite phenomenon suggests that the higher formaldehyde emission was mainly due to the tunnel release effect of SEP. Because SEP possesses a tunnel structure, which was beneficial for the release of free formaldehyde in the cured resin layer through these micro channels. Interestingly, although the tunnel release effect of SEP may play a main role in the formaldehyde emission process, the formaldehyde emission of the plywood bonded by MUF resin with 100% SEP was up to 1.21 mg/L. The high formaldehyde emission can be mainly attributed to the fact that the viscosity of the resin with 100% SEP was very low, making it very easy to penetrate into wood gaps, which increases the formaldehyde emission of the plywood. In theory, the SEP channel structure could accelerate the formaldehyde

emissions of the resultant plywood over a long period of time. With an increase of storage time, the formaldehyde emissions of the adhesive layer with a tunnel structure should be faster than that without a tunnel structure. Therefore, the formaldehyde emissions of the resulting plywood specimens were retested after 30 days in this research (Fig. 5).

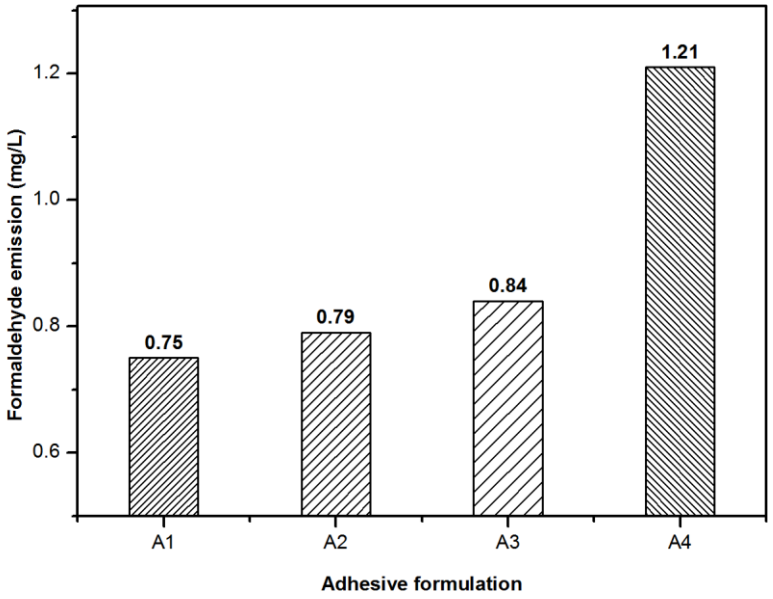

Fig. 4. The formaldehyde emissions of the plywood bonded by MUF resin with various filler formulations: (A1) 100% WF, (A2) 40% SEP + 60% WF, (A3) 80% SEP + 20% WF, and (A4) 100% SEP

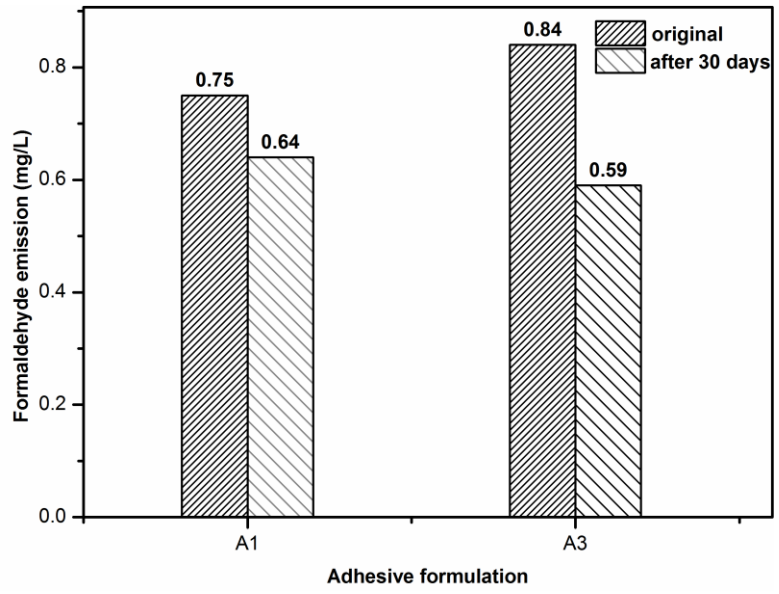

Fig. 5. Comparison of the initial formaldehyde emissions and the values after 30 days for plywood bonded by MUF resin with various filler formulations: (A1) 100% WF and (A3) 80% SEP + 20% WF

The formaldehyde emissions of the plywood after 30 days were evaluated and are shown in Fig. 5. When using 80% SEP to substitute WF, the formaldehyde emissions of the resultant plywood (0.84 mg/L) were 12% higher than that of the MUF resin with 100% WF (0.75 mg/L). After 30 days, the formaldehyde emissions of the plywood bonded by the MUF resin with 100% WF declined by 14.7% to 0.64 mg/L, and that of the MUF resin with 80% SEP + 20% WF declined by 29.8% to 0.59 mg/L. As a result, the final formaldehyde emissions of the plywood bonded by the MUF resin with 80% SEP + 20% WF were 7.8% lower than that with 100% WF. In the plywood fabrication industry, the resulting plywood undergoes a 3- to 4-week hot stacking process to cool down, which is beneficial for releasing formaldehyde, especially for the plywood bonded by the MUF resin with SEP.

SEM Analysis

Figure 6 shows the cross-section of the cured MUF resin with various formulations of fillers. The cross-section of the MUF resin with 100% WF presented a large number of holes. In addition, the whole fracture section appeared rough and irregular. These holes were caused by water evaporation in the adhesive during hot-pressing. Thus, the moisture could intrude into the adhesive layer, which led to low water resistance and low wet shear strength (Gao *et al.* 2012).

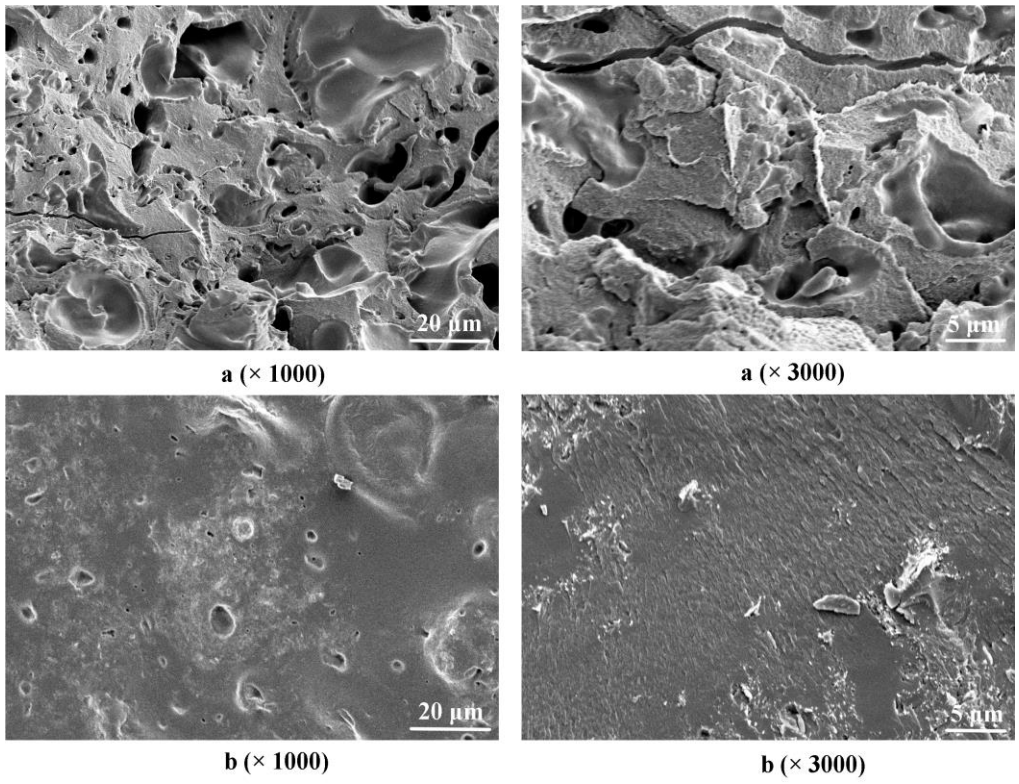

Fig. 6. MUF resin: a) formulation A1 b) formulation A3

After using 80% SEP to replace WF, fewer holes were observed and the cross-section became smoother, denser, and more regular. The denser cross-section could prevent the moisture from intruding into the adhesive layer, which improved the water resistance of the resin. Further, the wet shear strength increased the plywood bonded by

resultant resin. The SEM analysis was in agreement with the results of the part Wet Shear Strength Measurement, indicating that the wet shear strengths increased with SEP addition. Especially, when using 80% SEP to substitute WF, the wet shear strength of the resulting plywood increased by 27.8% compared with that of 100% WF. Also, the denser, regular and smoother cross-section of the resin with SEP implied that MUF resin molecule filled in the SEP framework and was distributed more homogeneously. Therefore, SEP improved the wet shear strength of the MUF resin.

CONCLUSIONS

1. Sepiolite (SEP) is able to form a framework structure to reduce evaporation of water in MUF resin and create a smooth cross-section of the cured adhesive to prevent moisture intrusion, thus improving the wet shear strength of the resulting plywood. SEP can also combine with the MUF resin by hydrogen bonds to form a penetration network, which would further improve the wet shear strength of the resultant plywood. When using 80% SEP to substitute WF, the wet shear strength of the resulting plywood increased by 27.8% compared with that of 100% WF.
2. The tunnel release effect of the SEP accelerated the formaldehyde emissions from the resulting plywood. The final formaldehyde emissions of the plywood bonded by the MUF resin with 80% SEP + 20% WF were 7.8% lower than that with 100% WF.

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