Journal of Applied Polymer Science

# Effects of diatomite inorganic fillers on the properties of a melamine-urea-formaldehyde resin

**Xiaona Li, Jing Luo, Jianzhang Li, Qiang Gao**

Key Laboratory of Wooden Material Science and Application (Ministry of Education), Engineering Research Center of Forestry Biomass Materials and Bioenergy (Ministry of Education), Beijing Key Laboratory of Lignocellulosic Chemistry, Beijing Forestry University, Beijing 100083, China

Correspondence to: J. Li (E-mail: lijzh@bjfu.edu.cn) and Q. Gao (E-mail: gaoqiang@bjfu.edu.cn)

**ABSTRACT:** In this study, a low-cost diatomite was used to partly substitute wheat flour as one type of melamine–urea–formaldehyde (MUF) resin filler. Five-ply plywood was fabricated, and its performance was measured. The crystallinity, fracture surface, and functional groups were tested to determine the effects of diatomite on the performance of the MUF resin. The results show that diatomite was well distributed in the MUF resin system and formed an embedding structure; this improved the wet shear strength of the resulting plywood by 33% to 1.36 MPa. Diatomite captured the free formaldehyde in the resin and the microporous structure formed in the resin accelerate formaldehyde release of the plywood. Consequently, the formaldehyde emission of the plywood was reduced. The diatomite partly replaced wheat flour as an MUF resin filler and could be applied in the plywood industry. © 2016 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 44095.

**KEYWORDS:** adhesives; applications; clay; resins

Received 28 June 2015; accepted 19 June 2016
**DOI: 10.1002/app.44095**

## INTRODUCTION

Melamine–urea–formaldehyde (MUF) resins are commonly used in the production of building and furniture materials, such as medium-density fiberboard, particleboard, and plywood,[1–5] and are favored for their improved hydrolytic and thermal stability over urea–formaldehyde resin and lower cost than phenol–formaldehyde resin.[6,7]

During the plywood fabrication process, fillers are usually used to decrease the cost of plywood production and improve the performance of the resulting adhesive. Fillers can reduce the interior forces of the cured adhesive, improve the prepress strength, and prevent the resin from dissipating into the wood.[8] Fillers used in the plywood industry contain starches, proteins, and cellulose. However, considering the cost and performance, wheat flour is the most common filler used in plywood manufacturing. Wheat flour can prevent the resin from penetrating into the veneer and reduce production costs. However, the use of wheat flour at concentrations than 30% will decrease water absorption resistance and, consequently, lead to declining physical and mechanical properties in plywood.[9] Wheat flour is quite expensive, and its use is considerably wasteful because wheat is, of course, an edible crop. It is, thus, highly necessary to develop resin fillers that can substitute effectively for wheat flour in the plywood industry and improve the performance of the resulting adhesives.

In this study, a low-cost clay, diatomite, was used to replace part of wheat flour as a filler. Diatomite is a type of mineral, the main component of which is amorphous $SiO_2$. Diatomite possesses unique properties, including a low density, high porosity, large surface area, and high absorptive capacity.[10] Its reserves are abundant, and it is relatively low in cost.[11] Diatomite is a sedimentary rock composed of the fossilized skeletons of dead microscopic single-cell algae (diatoms) from the ocean and fresh water floors.[10] Diatomic skeletons have honeycomb-shaped silica structures that lend materials useful characteristics, such as a high absorptive capacity and surface area, chemical stability, and low bulk density.[12] These properties make diatomite a promising candidate for a broad spectrum of applications, such as in filter aids, fillers,[13] adsorbents, and catalyst supports.[14] As a filler, diatomite is already used in the coatings, plastic, and rubber industries. Its unique properties and low cost makes it practical as a filler to substitute for wheat flour and for use as a filler for the MUF resin; it can hopefully improve the physical and mechanical properties of plywood and decrease the formaldehyde emission of plywood.

In this study, different proportions of diatomite were used to partially substitute for wheat flour as an MUF resin filler during

**Table I.** Different Formulations of the Diatomite-Based Filler

| No. | Diatomite (%) | Wheat flour (%) | Wheat flour (g) | Diatomite (g) |
|---|---|---|---|---|
| A1 | 0 | 100 | 25 | 0 |
| A2 | 40 | 60 | 15 | 10 |
| A3 | 60 | 40 | 10 | 15 |
| A4 | 100 | 0 | 0 | 25 |

the plywood preparation process. Five-ply plywood was fabricated, and we measured its wet shear strength and formaldehyde emission. The primary objective was to investigate the effects of diatomite addition on the MUF resin's physicochemical properties.

## EXPERIMENTAL

### Materials

Solid urea and a 37% formaldehyde solution were purchased from China National Petroleum Corp. and Guangdong Xilong Chemical Factory, respectively. Sodium hydroxide, melamine, formic acid, and ammonium chloride ($NH_4Cl$; analytical grade) were obtained from the Beijing Chemical Factory (China). Wheat flour was obtained from the Beijing Guchuan Flour Co. (China). Eucalyptus veneers with 8% moisture contents were purchased from Wen'an (Hebei Province, China). Diatomite powder, with an average particle size of 20 μm, was purchased from Qingdao Wall Material Co., Ltd. (Shandong Province, China).

### Adhesive Preparation and Apparent Viscosity Measurements

An MUF resin with a molar ratio [Formaldehyde/(Urea + Melamine)] of 1.12 was prepared in the laboratory according to the traditional alkali–acid–alkali method. Urea was added three times at a molar ratios of 2.2, 1.4, and 1.12, respectively, and melamine (10 wt % of the entire resin) was added along with the first and second addition of urea at 2 and 8%, respectively. Synthesis was conducted according to a procedure described in previous research.[15] Different adhesive formulations were prepared by the blending of the MUF resin with diatomite and wheat flour as fillers. The formulations of the diatomite-based filler samples are shown in Table I.

The viscosity of the adhesive was measured with a Brookfield DV-II viscometer with a 61# rotor with a spinning rate of 100 rpm. An average of three replications was used for each measurement.

### Plywood Preparation and Evaluation

Eucalyptus veneers (8% moisture content) with dimensions of $400 \times 400 \times 1.5\,mm^3$ were used to prepare five-ply plywood panels. The resins were prepared with 100 parts MUF resin, 0.8 parts $NH_4Cl$, and 25 parts filler and then applied to both sides of each veneer at a spread rate of $350\,g/m^2$. The stacked veneers were hot-pressed at 1.0 MPa and 120 °C for 7 min. Two plywood panels were prepared for each formulation of the resin.

The shear strength of plywood was determined according to the China National Standard for type II plywood (GB/T 17657-2013).[16] The five-ply plywood was cut into $25 \times 100\,mm^2$ specimens, with a $25 \times 25\,mm^2$ notch in each piece (Figure 1). The specimens were soaked in water at 63 ± 2 °C for 3 h and then cooled to room temperature for 10 min before measurement. Then, the wet shear strength of the specimen was evaluated with a universal tensile testing machine at a speed of 10 mm/min. The force required to break the glued specimen was recorded. The shear strength was calculated as the ratio of the force to the glue area. Eight specimens from two panels were tested for each sample, and the results were averaged.

### Water Uptake

The resin samples were cured in an oven at 120 ± 2 °C until a constant weight was reached. The initial mass ($m_i$) was determined with an analytical balance (±0.0001 g). Then, the resin samples were put in an oven with a constant temperature of 60 °C and a constant relative humidity of 75–80% for 12 h. Then, the final weight ($m_f$) was determined. The water intrusion was calculated as follows:

$$\text{Water intrusion}(\%) = (m_f - m_i) \times m_i^{-1} \times 100 \quad (1)$$

Five replicates were performed for each sample.

### Formaldehyde Emission Measurement

The formaldehyde emission of the plywood was determined with the desiccator method in accordance with the procedure described in China National Standard GB/T 17657-2013.[16] Twenty specimens were obtained from two panels with the same adhesive formulation and were cut into specimens with dimensions of $50 \times 150\,mm^2$. The specimens were then placed into a sealed desiccator with a diameter of 240 mm at 20 ± 2 °C for 24 h. The emitted formaldehyde was absorbed by 300 mL of deionized water in a container. The water was measured by a visible spectrophotometer to obtain the formaldehyde emission value. Ten pieces of the specimen were placed indoors for 15 days before testing. For the other 10 pieces, we first measured their formaldehyde emission and then placed them into an oven at 120 °C for 3 h. After heat treatment, these specimens were retested for formaldehyde emission. The formaldehyde emission reduction was calculated by the following equation.

$$\text{Decline rate}(\%) = (F_0 - F_t) \times F_0^{-1} \times 10 \quad (2)$$

where $F_0$ is the formaldehyde emission of the specimens tested directly and $F_t$ is the formaldehyde emission of the specimens after the heat treatment.

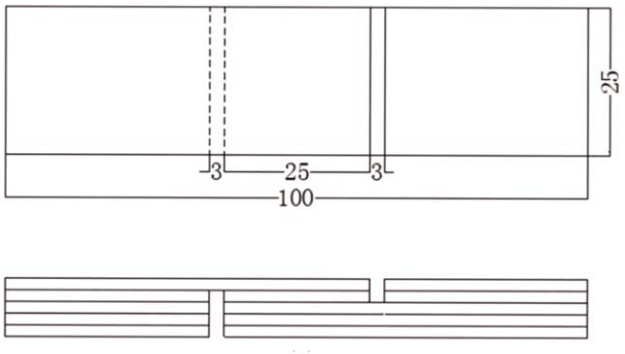


**Figure 1.** Size of the specimen for the water-resistance measurement.

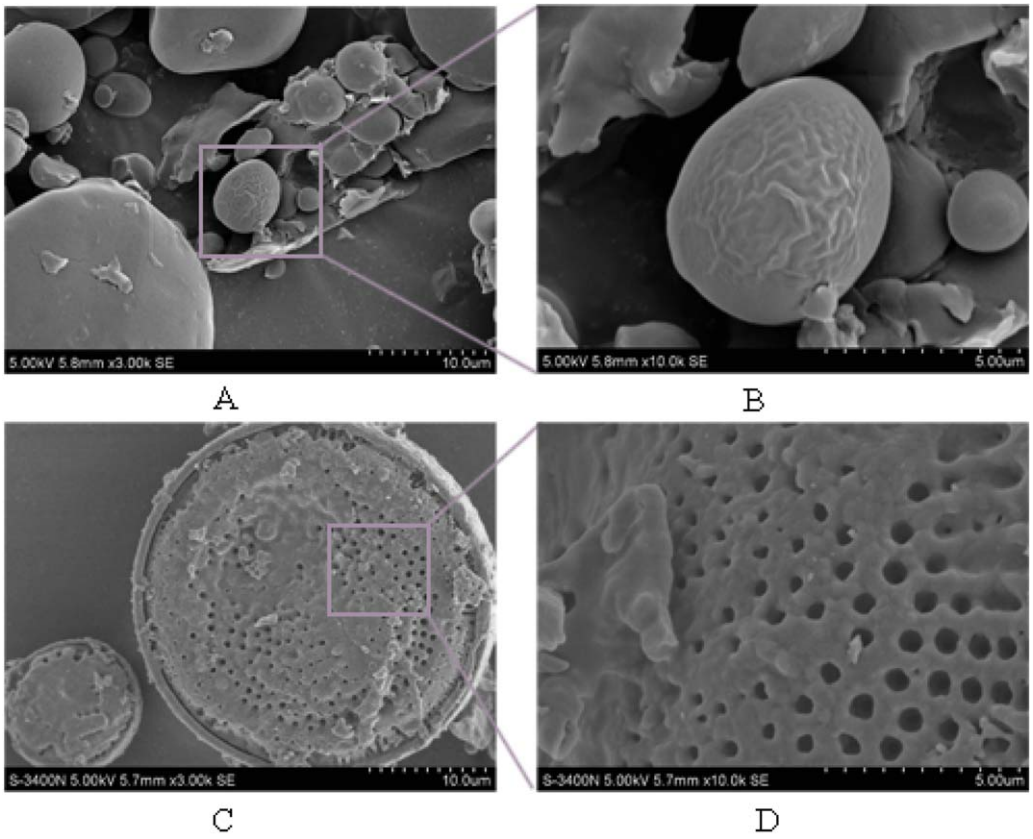


**Figure 2.** SEM images of wheat flour and diatomite: (A) wheat flour (3000×), (B) wheat flour (10,000×), (C) diatomite (3000×), and (D) diatomite (10,000×). [Color figure can be viewed in the online issue, which is available at wiley onlinelibrary.com.]

### Fourier Transform Infrared (FTIR) Spectroscopy, Scanning Electron Microscopy (SEM), and X-ray Diffraction (XRD) Measurements

The resin samples were placed in an oven at 120 ± 2 °C until they reached a constant weight and were then ground into a fine powder. The powder was mixed with KBr crystals at a mass ratio of 1:70 and pressed into a specialized mold to form a sample folium. The FTIR spectra were recorded with a Thermo Nicolet 6700 FTIR apparatus over a range of 400–4000 $cm^{-1}$ with a 4-$cm^{-1}$ resolution in 32 scans.

Each sample was placed into a piece of aluminum foil and dried in an oven at 120 ± 2 °C until it reached a constant weight; it was then sputter-coated with gold with an E-1010 Hitachi ion sputterer (Japan). A Hitachi S-3400N scanning electron microscope (Hitachi Science System, Ibaraki, Japan) was then used to observe the samples.

The resin samples were cured in an oven at 120 ± 2 °C until they reached a constant weight and were then ground into powder. XRD patterns were recorded on an XRD diffractometer (XRD-6000, Shimadzu, Kyoto, Japan) with a cobalt source in a 0.2 θ scan ranging from 5 to 60 ° at 45 kV and 30 mA. The relative crystallinity index was directly calculated by the measurement instrument with eq. (3):

$$\text{Relative crystallinity index}(\%) = A_c \times (A_c + A_a)^{-1} \times 100 \quad (3)$$

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.

## RESULTS AND DISCUSSION

### Wheat Flour and Diatomite Morphologies

Volume shrinkage generally occurs during the resin curing process; this results in internal stress formation and concentration.[8] This may crack or destroy glue lines and directly affect the bonding performance. The addition of fillers helps ease shrinkage during the curing process; this decreases the difference in the thermal expansion coefficient between the adhesive and veneer. In addition, fillers can prevent glue-line fractures from extending. Both of them improve the performance of the adhesive. As shown in Figure 2, wheat flour is characterized by smooth ellipsoid starch granules with a particle size of 38–53 μm. Diatomite presents a round sieve morphology with an average particle size of 20 μm. Its honeycomb micropore structures, as mentioned previously, provide the diatomite with several useful characteristics, including a high absorptive capacity and large surface area; this increased the contact area with the MUF resin. The compact contact between the diatomite and MUF resin was conducive to the dispersal of stress due to volume shrinkage or external impact.

### Wet Shear Strength and Viscosity Measurements

The effects of different filler formulations on the wet shear strength of the plywood samples are illustrated in Figure 3. The

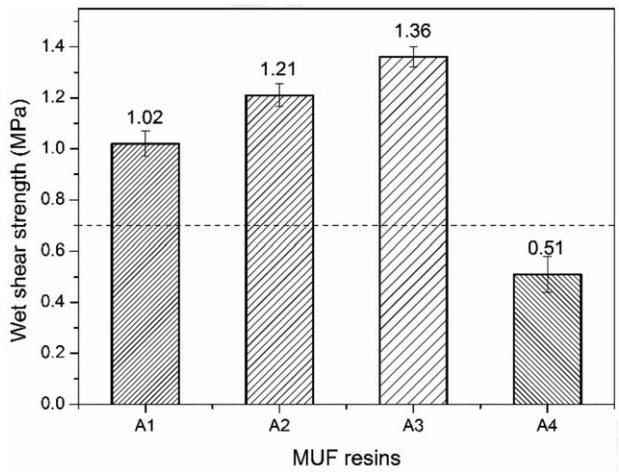


**Figure 3.** Wet shear strength of plywood bonded by the MUF resin with different fillers: (A1) 100% wheat flour, (A2) 40% diatomite plus 60% wheat flour, (A3) 60% diatomite plus 40% wheat flour, and (A4) 100% diatomite.

wet shear strength of the plywood bonded by the MUF resins with 100% wheat flour (resin A1) was measured at 1.02 MPa. When diatomite was added, the wet shear strength of the resulting plywood increased as the diatomite addition increased. When 40% diatomite (resin A2) was incorporated, the wet shear strength of the resulting plywood was 1.21 MPa; this was an 18.6% improvement compared to that of resin A1. The addition of 60% of diatomite (resin A3) was particularly effective, and the wet shear strength of the resulting plywood was improved to 1.36 MPa; this was a 33% improvement compared to resin A1. The elevated wet shear strength could be explained by the fact that the MUF resin molecules permeated into the diatomite pores and formed an embedding structure after curing. The confinement of polymer chains within the diatomite pores affected the local chain dynamics to a certain extent,[17] and this reduced the volume shrinkage in the cured glue line and improved the wet shear strength of the resulting plywood. The embedding structure also effectively prevented glue-line swelling during the 3 h of the soaking process at 63 °C. The wet shear strength of the plywood bonded by the MUF resin with 100% diatomite resin, however, dropped to 0.51 MPa; this was much lower than the interior plywood requirement (0.7 MPa, the dashed line in Figure 3) of China National Standard GB/T 17657–2013.[16] Wheat flour could absorb water and become viscous,[18] and this prevented the MUF resin molecules from penetrating the wood surface; diatomite was not able to function in the same way, and this resulted in a low viscosity. Generally, the viscosity of the liquid phase of the adhesive will influence penetration. Penetration happens with most types of resins at a low viscosity.[8] For further analysis of the penetration properties, the viscosity of the resin sample was tested (Table II). Compared with resin A1, the viscosity of resin A4 was only 356 cP; this resulted in an excessive amount of resin penetrating into the veneer. As a result, the wet shear strength of the plywood bonded by resin A4 was the lowest.

**Table II.** Viscosities of the Resin Samples

| Sample | A1 | A2 | A3 | A4 |
|---|---|---|---|---|
| Viscosity (cP) | 15297 ± 4 | 2447 ± 3 | 1820 ± 2 | 356 ± 5 |

### Formaldehyde Emission Measurements

The formaldehyde emissions of the plywood after 15 days are shown in Figure 4. The formaldehyde emission of the plywood bonded by the MUF resin with 100% wheat flour (resin A1) was 0.51 mg/L (the initial formaldehyde emission of the plywood bonded by resin A1 was 0.69 mg/L). When diatomite was added, the formaldehyde emission of plywood decreased as the diatomite addition increased in the filler formulation, except for the plywood bonded by the MUF resin with 100% diatomite (resin A4). The formaldehyde emission of the plywood bonded by resin A4 was 0.64 mg/L; this was higher than that of resin A1. With 100% diatomite as the MUF filler, the resulting adhesive had a low viscosity (356 cP; Table II), and this caused the resin excessive to penetrate into the veneer so that the free formaldehyde in the adhesive was easily emitted and the formaldehyde emission increased (the initial formaldehyde emission of the plywood bonded by resin A4 was 1.16 mg/L). The formaldehyde emission bonded by the MUF resin with 40% diatomite (resin A2) was 8% lower than that with 100% wheat flour (resin A1). In particular, when 60% diatomite (resin A3) was added, the formaldehyde emission of the resulting plywood was 24% lower than that of 100% wheat flour (resin A1). The formaldehyde emission reduction of the plywood bonded by the resin with diatomite could be explained by two reasons. One reason was the absorptive capacity of the diatomite. Diatomite contains many siloxane groups or —Si—O—Si— bridges with oxygen atoms, and the surface is terminated by —OH groups and oxygen bridges; this makes diatomite well suited to applications requiring sorbent and filling compounds.[19,20] Consequently, the free formaldehyde in the adhesive was captured by diatomite. In addition, the addition of diatomite created a micropore structure in the cured resin layer,[21] which contributed to the fast release of free formaldehyde. To prove the second

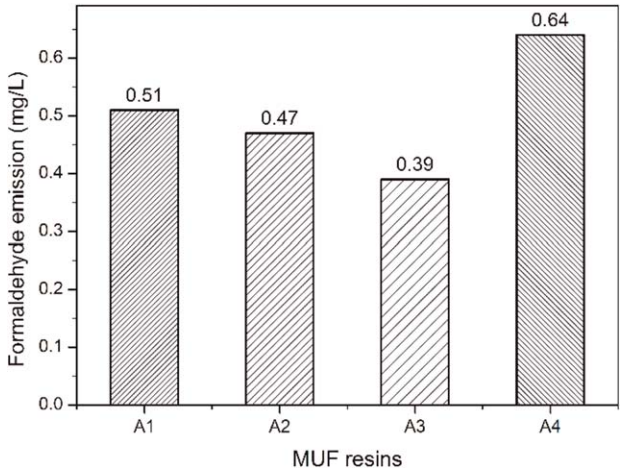


**Figure 4.** Formaldehyde emission of plywood bonded by the MUF resin with different fillers after 15 days: (A1) 100% wheat flour, (A2) 40% diatomite plus 60% wheat flour, (A3) 60% diatomite plus 40% wheat flour, and (A4)100% diatomite.

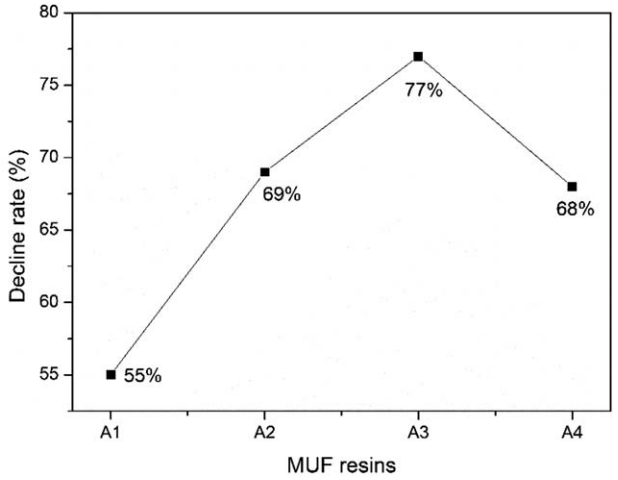


**Figure 5.** Decline rate of the plywood formaldehyde emission after heat treatment: (A1) 100% wheat flour, (A2) 40% diatomite plus 60% wheat flour, (A3) 60% diatomite plus 40% wheat flour, and (A4) 100% diatomite.

reason, a secondary experiment was conducted to further investigate the formaldehyde emissions of the plywood specimens after an acceleration release process–heat treatment.

An oven heat treatment can accelerate the formaldehyde emission of plywood and simulate the formaldehyde emission of the plywood over a long period of time. Figure 5 shows the formaldehyde emission decline rate of the plywood after heat treatment. After treatment, the specimens with diatomite presented a higher formaldehyde emission reduction than those without diatomite. The formaldehyde emission decline rate of the plywood bonded by the MUF resin with 100% wheat flour (resin A1) was 55%. When diatomite replaced 40% of the white flour (resin A2), the decline rate was 69%; this was 14% higher than that of resin A1. In particular, the formaldehyde emission decline rate of the plywood bonded by resin A3 was 77%; this was 22% higher than resin A1. This secondary experiment further verified that the diatomite-created micropore structure accelerated the formaldehyde emissions of the plywood. Diatomite supplementation of the MUF resin would be highly beneficial for the release of excess formaldehyde during the 3- to 4-week hot stacking process typically applied to plywood before use.

The results indicate that the filler formulation of A3 (60% diatomite with 40% wheat flour) showed good performance with the MUF resin. Therefore, the A3 formulation was chosen as an optimum formulation and was tested further to determine why the performance of the resin increased with diatomite.

### FTIR Spectroscopy, SEM, and XRD Analysis

FTIR measurements are provided in Figure 6. The peaks at 3350, 2961, 1636, and 814 $cm^{-1}$ were the characteristic absorption peaks of —OH or —NH, —CH, —CO, and triazine rings, respectively. The four primary peaks indicated the successful formation of the MUF resin.[22,23] The positions of the absorption bands in the curves of MUF resin containing diatomite were nearly the same as those of the pure MUF resin; the only difference was that the MUF resin with diatomite naturally contained peaks characteristic of diatomite; this indicated that the diatomite was well distributed in the adhesive system. The wide bands centered at 1090 $cm^{-1}$ were attributed to Si—O—Si in-plane vibrations. Similar observations at 792 and 470 $cm^{-1}$ were likewise characteristic of silica. The weak absorption peak at 618 $cm^{-1}$ was most likely due to Si—O deformation.[24,25] Besides these, no other new peaks appeared in the IR absorption spectrum of the MUF resin with diatomite. Therefore, the FTIR results confirm that no chemical reaction occurred between the diatomite and MUF resin but that the diatomite had merely blended physically with the MUF resin.

The fracture surfaces of the cured MUF resin with different fillers were observed and are shown in Figure 7. The fracture surface of the cured resin with 100% wheat four showed a compact and smooth surface. After diatomite [Figure 7(b)] was incorporated, micropores were observed on the surface of the cured resin; these were caused by water evaporation during the hot-pressing process. The well-distributed diatomite, acting as zeolite, accelerated the water evaporation around the diatomite; this caused holes in the fracture section.[26] At the same time, the micropores were beneficial for formaldehyde emission, which was in accordance with the analysis of formaldehyde emissions. Through the higher resolution, Figure 7(c) shows that the MUF resin was intercalated in the diatomite micropores (marked with arrows) to form an embedded structure. As shown in Figure 3, the wet shear strength of the plywood bonded by resin A3 was improved to 1.36 MPa; this indicated that the embedding structure played a major role in the strength. The embedding structure could reduce the volume shrinkage of the cured resin and, thus, improve the resulting plywood's wet shear strength. However, this effect was not observed in the SEM image. At the same time, we believe the formed embedded structure prevented moisture intrusion, so water uptake measurement was further conducted. The results of the water uptake measurement are shown in Table III. The water uptake of resin A3 was 20.5%;

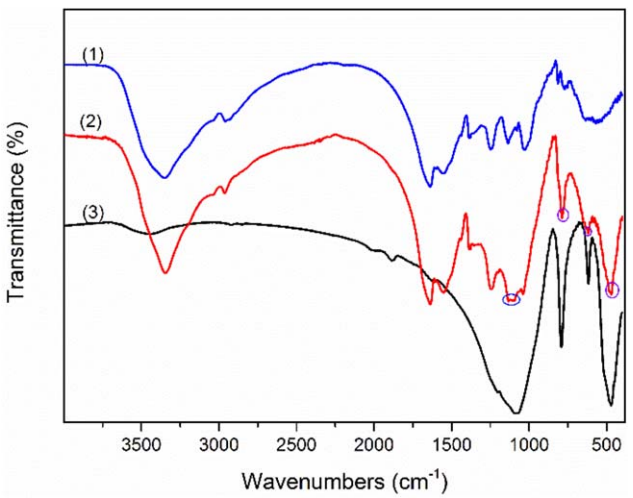


**Figure 6.** FTIR spectra: (1) MUF resin plus A1 (100% wheat flour), (2) MUF resin plus A3 (60% diatomite plus 40% wheat flour), and (3) diatomite. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

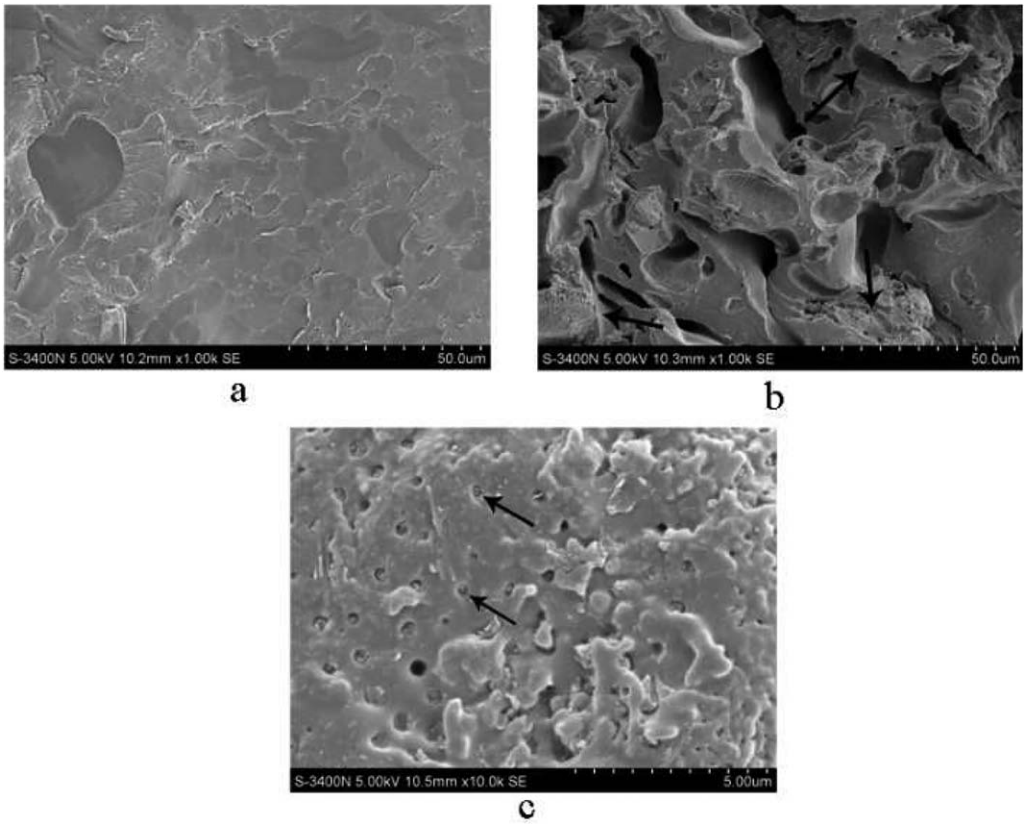


**Figure 7.** SEM images of the fracture surfaces of the cured MUF resin with different fillers: (a) MUF resin and 100% wheat flour (1000×); (b) MUF resin, 60% diatomite, and 40% wheat flour (1000×); and (c) MUF resin, 60% diatomite, and 40% wheat flour (10,000×).

this was 20% lower than that of resin A1. This proved that the embedded structure prevented moisture intrusion and, thus, improved the water resistance of the resins. From the other perspective, the fracture surface of the MUF resin with wheat flour was smooth, which is a typical brittle fracture. After the incorporation of diatomite, the fracture surface showed a lot of irregular folds and bumps and presented a ductile fracture. This was attributed to the fact that the embedded structure of diatomite prevented the extension of the fracture surface caused by outside forces; this improved the damage resistance of water intrusion and, thus, improved the water resistance of the resulting adhesive.

Figure 8 shows the XRD patterns of the diatomite and MUF resin with different fillers. In the pattern of diatomite, two strong characteristic peaks observed at 2θ values near 21.8 and 36.0° were attributed to amorphous silica (diatomite).[27–30] Compare to the pattern of the MUF resin with 100% wheat flour, after diatomite was added to the filler formulation, the pattern of the resulting resin also showed these characteristic peaks of diatomite. This indicated that the diatomite was distributed well in the MUF resin.

**Table III.** Water Uptake of the Resin Samples

| Sample | A1 | A3 |
|---|---|---|
| Water intrusion (%) | 41.2 ± 2.4 | 20.5 ± 1.8 |

The crystallinity of the samples was calculated and is shown in Table IV. The diatomite was a high-crystallinity material (76.6%) compared with wheat flour (29.3%). The crystallinity of the MUF resin with wheat flour was 41.3%; after the incorporation of diatomite, the crystallinity decreased to 34.7%. This was attributed to the embedded structure formation. During the

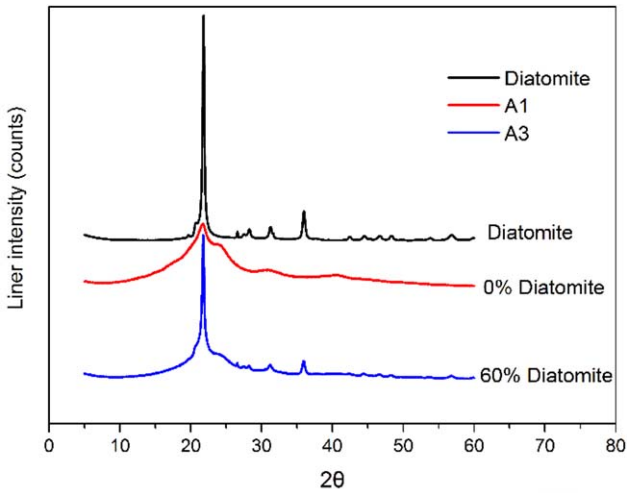


**Figure 8.** XRD patterns of diatomite and the MUF resin with different fillers: (A1) 100% wheat flour and (A3) 60% diatomite plus 40% wheat flour. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

| Samples | Crystallinity (*CrI*) (%) |
|---|---|
| Diatomite | 76.6% |
| Wheat flour | 29.3% |
| MUF+A1 | 41.3% |
| MUF+A3 | 34.7% |

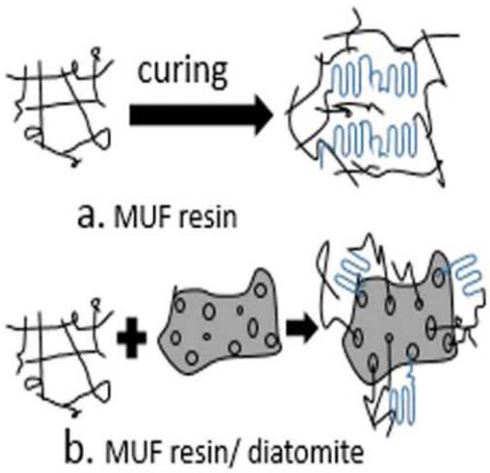


**Figure 9.** Schematic of the effect of diatomite on the crystallinity reduction of the resulting resin. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

curing process, some of the MUF resin molecules arranged in an orderly manner and formed a crystalline zone, which could be detected by the X-rays. As the diatomite was incorporated, the resin formed an embedded structure and, thus, broke the crystalline zone of the MUF resin; this reduced the crystallinity of the resulting resin. This process is illustrated in Figure 9.

## CONCLUSIONS

The most notable conclusions of this study are summarized as follows.

With diatomite used to replace 60% wheat flour as an MUF resin filler, the wet shear strength of the resulting plywood was increased by 33% to 1.36 MPa compared to the 100% wheat flour filler. This improvement was attributed to the embedded structure formed by diatomite and the MUF resin. The embedded structure of diatomite prevented the extension of the fracture surface caused by outside forces, improved the damage resistance of water intrusion, and thus improved the water resistance of the resulting adhesive.

**Table IV.** Crystallinity of the Samples

| Sample | Relative crystallinity index (%) |
|---|---|
| Diatomite | 76.6 |
| Wheat flour | 29.3 |
| MUF + A1 | 41.3 |
| MUF + A3 | 34.7 |

After diatomite was added to replace 60% wheat flour in the MUF resin filler, the formaldehyde emission of resulting plywood decreased by 24% compared to that of the 100% wheat flour filler. This reduction was due to the following two reasons. First, diatomite captured the free formaldehyde in the resin. Second, the microporous structure formed in the resin accelerate formaldehyde release in the plywood.

The FTIR and XRD results confirm that no chemical reaction occurred between the diatomite and MUF resin and that the diatomite was distributed well in the MUF resin.

On the basis of its performance and low cost, diatomite could be used to partly replace wheat flour in the development of a low-cost MUF resin filler to be applied in the plywood industry.

## ACKNOWLEDGMENTS

The authors are grateful for the financial support of the National Natural Science Foundation of China (contract grant numbers 31400501 and 30972310) and the Beijing Natural Science Foundation (contract grant number 2151003).

## REFERENCES

1. Park, B.-D.; Lee, S.-M.; Roh, J.-K. *Eur. J. Wood Wood Prod.* **2009**, *67*, 121.
2. Silva, D. A. L.; Lahr, F. A. R.; Varanda, L. D.; Christoforo, A. L.; Ometto, A. R. *J. Clean Prod.* **2015**, *96*, 299.
3. Dunky, M. *Macromol. Symp.* **2004**, *217*, 417.
4. Kim, S. *Constr. Build. Mater.* **2009**, *23*, 2319.
5. Ayrilmis, N.; Kaymakci, A. *Holzforschung* **2012**, *66*, 443.
6. Gao, Q.; Shi, S. Q.; Zhang, S.; Li, J.; Wang, X.; Ding, W.; Liang, K.; Wang, J. *J. Appl. Polym. Sci.* **2012**, *125*, 3676.
7. Stockel, F.; Konnerth, J.; Kantner, W.; Moser, J.; Gindl, W. *Holzforschung* **2010**, *64*, 337.
8. Kamke, F. A.; Lee, J. N. *Wood Fiber Sci.* **2007**, *39*, 205.
9. Doosthoseini, K.; Zarea-Hosseinabadi, H. *J. Indian Acad. Wood Sci.* **2011**, *7*, 58.
10. Janicijevic, J.; Krajisnik, D.; Calija, B.; Dobricic, V.; Dakovic, A.; Krstic, J.; Markovic, M.; Milic, J. *Mater. Sci. Eng. C* **2014**, *42*, 412.
11. Guo, Q. L.; Li, L. L.; Cheng, Y. C.; Jiao, Y. B.; Xu, C. *Mater. Des.* **2015**, *66*, 51.
12. Gomez, J.; Gil, M. L. A.; de la Rosa-Fox, N.; Alguacil, M. *Food Chem.* **2014**, *159*, 381.
13. Ibrahim, S. S.; Selim, A. Q. *Physicochem. Probl. Miner. Process.* **2011**, *47*, 113.
14. Cong, P.; Chen, S.; Chen, H. *Constr. Build. Mater.* **2012**, *30*, 495.
15. Ding, W.; Li, W.; Gao, Q.; Han, C.; Zhang, S.; Li, J. *Bioresources* **2013**, *8*, 2568.

16. Test Methods of Evaluating the Poperties of Wood-Based Panels and Surface Decorated Wood-Based Panels; GB/T 17657. Standardization Administration of The People's Republic of China: Beijing, **2013**.
17. Moya, R.; Rodríguez-Zúñiga, A.; Vega-Baudrit, J.; Álvarez, V. *Int. J. Adhes. Adhes.* **2015**, *59*, 62.
18. Ding, R.; Su, C.; Yang, Y.; Li, C.; Liu, J. *Int. J. Adhes. Adhes.* **2013**, *41*, 1.
19. Zhang, M.; Xu, Y.; Wang, S.-L.; Shi, J.-Y.; Liu, C.-Y.; Wang, C.-Y. J. *Forestry Res.* **2013**, *24*, 741.
20. Khraisheh, M. A. M.; Al-Ghouti, M. A.; Allen, S. J.; Ahmad, M. N. *Water Res.* **2005**, *39*, 922.
21. Yuan, W.; Yuan, P.; Liu, D.; Yu, W.; Deng, L.; Chen, F. *Micropor. Mesopor. Mater.* **2015**, *206*, 184.
22. Tong, X. M.; Zhang, M.; Wang, M. S.; Fu, Y. *J. Appl. Polym. Sci.* **2013**, *127*, 3954.
23. Kandelbauer, A.; Despres, A.; Pizzi, A.; Taudes, I. *J. Appl. Polym. Sci.* **2007**, *106*, 2192.
24. Tsai, W.-T.; Lai, C.-W.; Hsien, K.-J. *J. Colloid Interface Sci.* **2006**, *297*, 749.
25. Gomez, J.; Gil, M. L. A.; de la Rosa-Fox, N.; Alguacil, M. *Food Chem.* **2015**, *170*, 84.
26. Tao, Y.; Kanoh, H.; Abrams, L.; Kaneko, K. *Chem. Rev.* **2006**, *106*, 896.
27. Alyosef, H. A.; Ibrahim, S.; Welscher, J.; Inayat, A.; Eilert, A.; Denecke, R.; Schwieger, W.; Munster, T.; Kloess, G.; Einicke, W. D.; Enke, D. *Int. J. Miner. Process.* **2014**, *132*, 17.
28. Ibrahim, S. S.; Selim, A. Q. *Physicochem. Probl. Miner. Process.* **2012**, *48*, 413.
29. Ha, J. H.; Oh, E.; Bae, B.; Song, I. H. *Ceram. Int.* **2013**, *39*, 8955.
30. Jia, Y. X.; Han, W.; Xiong, G. X.; Yang, W. S. *Sci. Technol. Adv. Mater.* **2007**, *8*, 106.