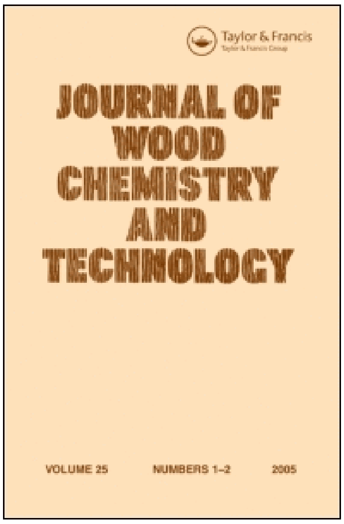

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Abstract: It is important to utilize reinforcement by fibers in wood plastic composites (WPC). However, when a thermoplastic polymer is mixed with fibrous wood material, the melt fluidity of the mixture usually decreases and its fabrication becomes difficult. This tendency is pronounced for the composite with a high cellulose content. In this study, the effects of added fly ash, which is a spherical silica, on the fluidity of melt mixture and injection molded composite are reported. Increase in fluidity of compound and fall of molding viscosity were observed by the addition of fly ash. The spherical shape of fly ash was maintained after molding. The increase in water-resistance of the injected mold was also confirmed. Addition of fly ash improved the molding of cellulose/plastic composites with high cellulose content and also improved their water-resistance.

Keywords: Cellulose, composite, fly ash, fluidity, polypropylene

INTRODUCTION

In recent years, wood plastic composite (WPC), which uses wood flour as a cellulose material and polyolefin resin, and has the features of both wood and plastic, has seen expanding demand for use in exterior construction materials such as decks, since cellulose has a reinforcing effect as a fiber.[1–7] However, the effects of cellulose in WPC have been focused on the function of cellulose

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as a filler for increasing volume, and few reports have given attention to its reinforcing effect as a fiber. In processes such as extrusion and injection molding, the flow of compound is important, and molding becomes difficult with a high fiber content, so fiber is not currently used widely in the industry, especially for WPC with high cellulose content.[8] Although there are many reports on cellulose/plastic composite using fibrous fillers, most of them examine composites with a thermosetting polymer that does not need a high flow during molding.[9–14]

There are also some reports on WPC composites using inorganic fillers,[15] although few of them have studied the flow characteristic of compound with high filler content. In this study, the effect of spherical silica on the flow characteristics of cellulose/plastic compound with high cellulose content for injection molding was evaluated.

EXPERIMENTAL

Materials

The following raw materials were used: cellulose powder, Arbocel BC200 (fiber length 300 micrometer, fiber diameter 20 micrometer) supplied by J. Rettenmaeyer and Soene (Rosenbreg, Germany); polypropylene (PP), PM930V (random, MFR30, Sun Alomar (Tokyo, Japan)). As a compatibilizing agent, PP resin modified with maleic anhydride (Umex 1010, Sanyo Kasei (Kyoto, Japan)) was used to improve interface adhesion between the cellulose powder and PP resin. Fly ash (Chuden Techno, Nagoya, Japan) was used. Densities of raw materials were: BC200, 1.40; PM930V, 0.90; Umex1010, 0.90; Fly ash, 2.16g/cm^3.

Compounding Process

Formulations of compound are shown in Table 1. Compounding (preparation of compound) was carried out using a conical twin screw extruder (Taitan 80;

Table 1. Formulation (by weight)

Cellulose	Fly ash	PP-resin	MaPP
70	0	28	2
69	1	28	2
67	3	28	2
65	5	28	2
60	10	28	2
50	20	28	2

Cincinnati Extrusion, Vienna, Austria) to give pellets of 4 mm diameter × about 5 mm length at a molding temperature of 230°C and screw rotation speed of 15 rpm.

Melt Flow Rate (MFR)

MFR was measured using a melt indexer (Nihon Dynisco, Yokohama, Japan) using a die with a diameter of 7 mm under a 10 kg load at 220°C. Measurement was started after loading the compound and preheating for 5 min.

Viscosity of Mold

Viscosity of molten compound was measured using a small size conical twin screw extruder (Mini Lab; Eiko Seiki, Tokyo, Japan) at a molding temperature of 180°C and screw rotating speed of 100 rpm. The viscosity was determined from the difference in the two bypass part (screw-top and screw-end) pressures.

Injection Molding

Injection molding was carried out using a pelletized compound by NS20-2A molder (Nissei Plastic, Tokyo, Japan) and a molding temperature was adjusted to 180°C to give a dumbbell-shaped composite (length, 120 mm; width, 9 mm; thickness, 5 mm).

Water Absorption

Water absorption was tested in water for 0 to 2000 hours under atmospheric pressure at room temperature.

Scanning Electron Microscope (SEM)

SEM (Miniscorp TM1000; Hitachi, Tokyo, Japan) was used for analyzing fracture surface of injection molding at 15 kV.

RESULT AND DISCUSSION

Effect of Fly Ash Addition on Flow Characteristics of Compound

Correlation between flow characteristics of compound measured by MFR and fly ash content is shown in Figure 1. MFR increased with the fly ash content. Although there was a possibility of breakdown of fly ash into small fragments during the compounding process, the particles of fly ash were observable in the compound containing 10% fly ash by electron microscope (Figure 2b). This result supports that fly ash maintains its spherical shape in the compounding

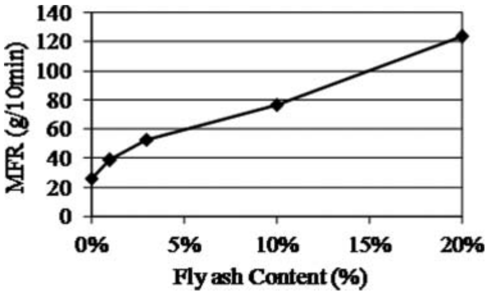


Figure 1. Effect of fly ash content on molten fluidity (MFR).

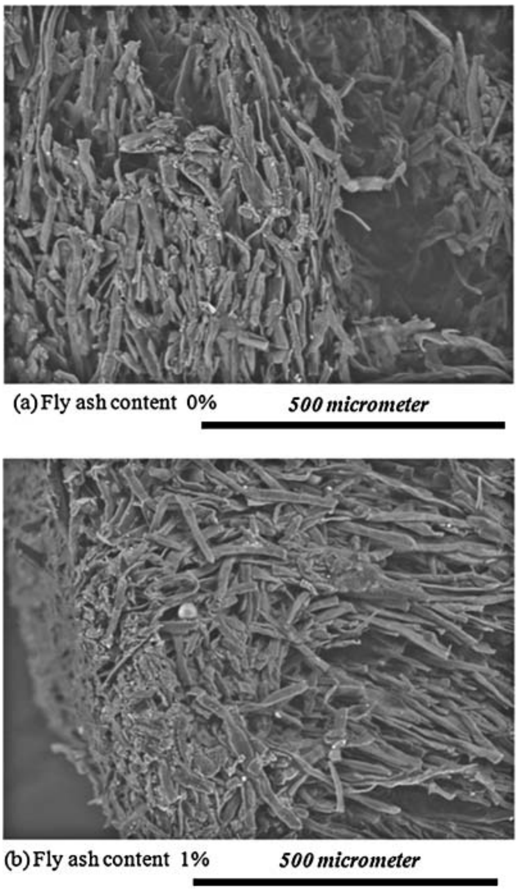


Figure 2. SEM photograph of compound (pellet). (a) Fly ash content 0%. (b) Fly ash content 1%.

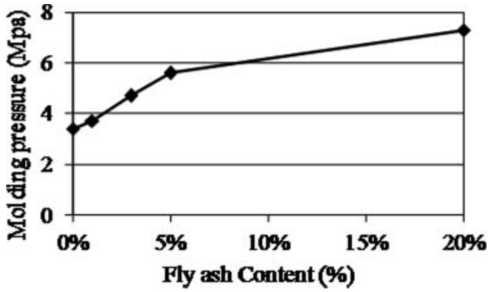


Figure 3. Effect of fly ash content on compounding pressure.

process and has an effect of improving the flowability of the compound. The effect may be called the bearing effect of fly ash.

Effect of Fly Ash Addition on Compounding Pressure (3.1⇔3.2)

Figure 3 shows the correlation between molding pressure and fly ash content in compounding raw material by extruder at the same screw rotating speed of 10 rpm. The instrument for measuring pressure was set just before the compression zone of the pelletizing die. There was a tendency for molding pressure to increase with fly ash content. This result is interpreted that firstly, the weight of material that passed the pressure measuring zone in a unit time increased with the fly ash content and/or secondly, the compound was already compressed at the site of measuring pressure before the compression zone, because the flowability of compound was increased by fly ash addition, as mentioned earlier.

Effect of Fly Ash Addition on Viscosity in Compounding Process

The correlation between compounding time and molten viscosity is shown in Figure 4 for compounds containing 0, 1, and 10% of fly ash prepared with a small size conical twin screw extruder. An almost linear time-dependent decrease of viscosity was observed by compounding regardless of the fly ash content. This result shows that dispersion of fiber was improved and tangles of fibers decreased due to break down of fibers by shear of compounding. This discussion is confirmed by comparison of the SEM photograph of the fracture surface of the injection mold (Figure 5) with that of compound (Figure 2), which shows that the cellulose fibers of the former are shorter than the latter.

The effect of fly ash content on viscosity (Figure 4) was not remarkable with 1% fly ash content, but with 10% fly ash the viscosity was low. SEM photographs of fracture surfaces of the injection mold for each fly ash content

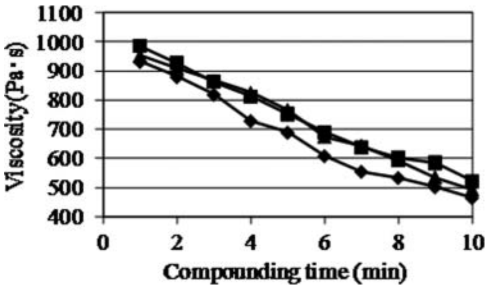


Figure 4. Effect of fly ash content on molten-viscosity at 180° of compounding temperature. Symbols: solid square, fly ash content 0%; solid triangle, fly ash content 1%; solid diamond, fly ash content 10%.

are shown in Figure 5. Although spherical fly ash particles were slightly detectable with 1% fly ash content, particles were observed with increasing fly ash content, indicating that fly ash retained its spherical shape after molding.

Effect of Fly Ash Addition on Properties of Injection Molded Composite

The mechanical properties of injection molded composite with several fly ash contents are shown in Table 2. Density increased with fly ash content, because of the high density of fly ash. Bending strength decreased with the fly ash content. In our experiments total content of fillers consisting of fly ash and cellulose fiber was kept constant because molding process was unstable under varying filler/resin ratio. As the result, the ratio of cellulose to PP varied from 2.5 to 1.8 corresponding to the change of fly ash content from 0 to 20%, respectively. Decrease in the cellulose content usually gives composite of weaker bending strength, but the effect is small compared with the present results of fly ash addition.[8] Under the conditions where more than a minimum amount of PP necessary for covering cellulose fiber is used, the ratio of cellulose/PP does not show significant influence on bending strength of composite and also on

Table 2. Mechanical properties of injection molding

Fly ash content (%)	Density (g/cm^3)	Bending strength (MPa)	Elastic modulus (GPa)
0	1.22	92.09	5.25
1	1.22	94.08	5.18
3	1.22	85.87	5.18
5	1.23	84.77	4.98
20	1.30	82.82	5.23

water absorption.[8] Thus the decrease of bending strength in Table 2 can be accepted mainly due to the effect of fly ash addition. In the literature, there are reports on wood flour/plastic composite showing improved strength by fly ash addition.[15]

In other experiments, we have results of cellulose/PP injection molded composite of high cellulose content that decreased bending strength of composite was observed for a compound of low fluidity when fibers flocculated.[16] The results of the present experiment with fly ash are another case since the

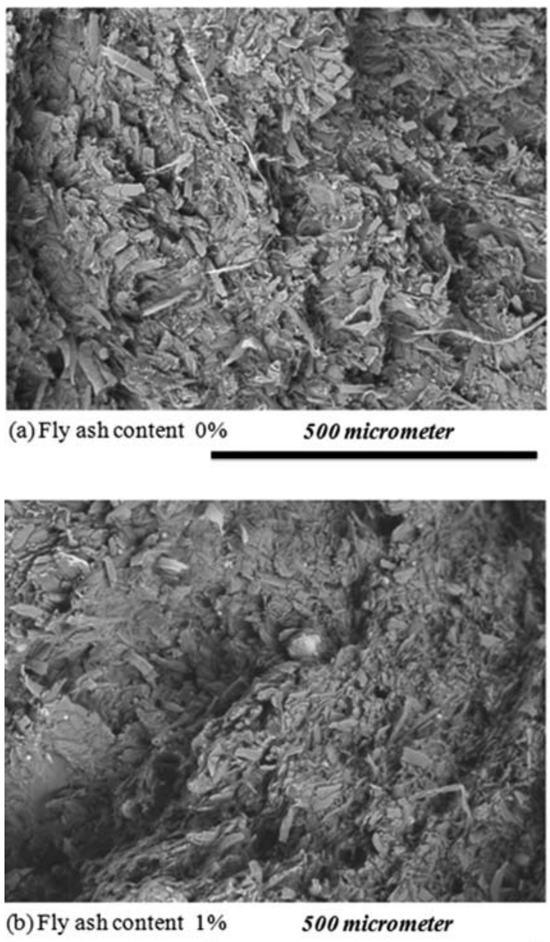
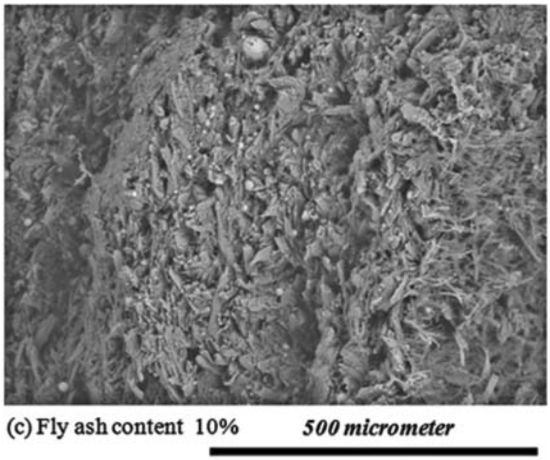


Figure 5. SEM photographs of fracture surface of injection molded composites. (a) Fly ash content 0%. (b) Fly ash content 1%. (c) Fly ash content 10%. (d) Fly ash content 20%.



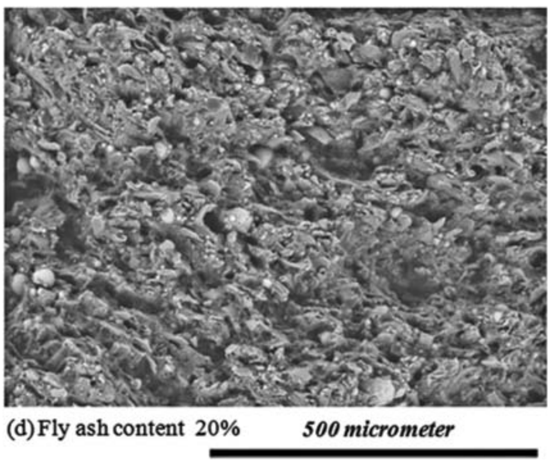


Figure 5. (Continued)

compound showed high fluidity and flocculation of fibers was not detectable in photographs of the fracture surface (Figure 5).

The effect of water absorption on injection molded composites is shown in Figure 6. Water absorption decreased with increasing fly ash content. Usually water absorption of WPC has some correlation with the void space of composite, which can be estimated by the comparison of hypothetical density of composite calculated from densities of each component and their composition, with the measured density of composite.[16,17] Since there was no difference in the void spaces of the composites judging from the results of density in Table 2 and in general, the effect of change in the net cellulose content of less than 10% on water absorption is small, as described earlier.[8] The observed decrease of

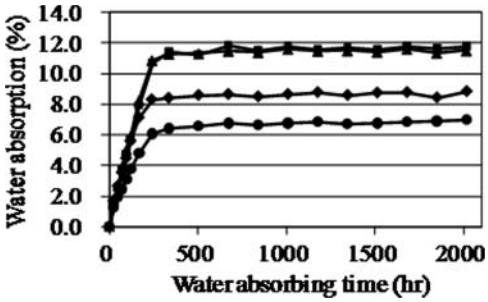


Figure 6. Effect of fly ash content on water absorption for 0 to 2000 hours under atmospheric pressure at room temperature of composite. Symbols: solid square, fly ash content 0%; solid triangle, fly ash content 1%; solid diamond, fly ash content 5%; solid circle, fly ash content 20%.

water absorption can be ascribed to the effect of fly ash. The swelling in the length-wise direction in Figure 7 decreased with increasing fly ash content, similar to water absorption.

In Figure 8 bending strength of composites after 168 hours of water absorption is shown. Although bending strength of dry composite decreased by fly ash addition, there was no difference in bending strength after sufficient water absorption (Figure 6). These results show that some bonding force that can be weakened by absorbed water such as hydrogen bond and/or tangling of fibers may contribute for the dry strength of high filler content cellulose composite. Mechanism of the effect of fly ash addition on bonding force and tangle[18] remained to be clarified.

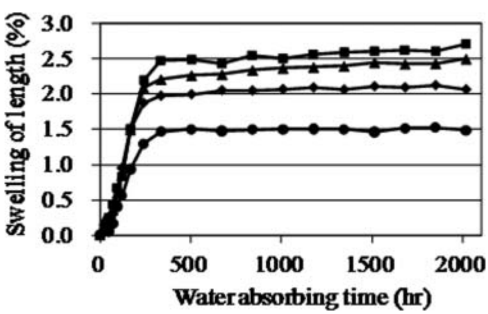


Figure 7. Effect of fly ash content on swelling by water absorption in length-wise direction (water absorption conditions were the same as in Figure 6). Symbols: solid square, fly ash content 0%; solid triangle, fly ash content 1%; solid diamond, fly ash content 5%; solid circle, fly ash content 20%.

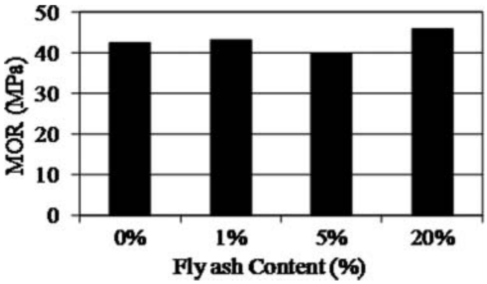


Figure 8. Effect of fly ash content on bending strength of composite after water absorption for 168 hours.

CONCLUSION

For the purpose of improving fluidity of cellulose/plastic compound with high cellulose content, the effect of added fly ash was examined. Fluidity of compound (MFR and viscosity) was improved by the addition of fly ash, probably through the bearing effect of spherical fly ash. Observed effect of fly ash on properties of the injection molded composite was negative on flexural strength and modulus of elasticity, but water-resistance was improved. Although we have discussed the contribution of tangle of fiber to the mechanical properties,[18] fluidity remained an issue in processing cellulose/plastic composites with high cellulose content. This work represents a new strategy for improving fluidity during molding, which will contribute to promoting use of wood fiber for composites. In addition, fly ash is cheap spherical silica and it also contributes to improve water-resistance, which is an important property for cellulose/plastic composites with high cellulose content.

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