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ORIGINAL ARTICLE

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Fundamental studies on wood/cellulose–plastic composites: effects of composition and cellulose dimension on the properties of cellulose/PP composite

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Abstract Although wood/cellulose–plastic composites (WPC) of low wood/cellulose content have been more accepted worldwide and are promoted as low-maintenance, high-durability building products, composites containing high wood/cellulose content are not yet developed on an industrial scale. In this study, flow properties, mechanical properties, and water absorption properties of the compounds of cellulose microfiber/polypropylene (PP) and maleic anhydride-grafted polypropylene (MAPP) were investigated to understand effects of the high cellulose content and the dimensions of the cellulose microfiber. The molding processes studied included compression, injection, and extrusion. It was found that fluidity is not only dependent on resin content but also on the dimension of the filler; fluidity of the compound declined with increased fiber length with the same resin content. Dispersion of the composite was monitored by charge-coupled device (CCD) microscope. Increasing the plastic content in the cellulose–plastic formulation improved the strength of mold in addition to the bond development between resin and filler, and the tangle of fibers. The processing mode affected the physicomechanical properties of the cellulosic plastic. Compression-molded samples exhibited the lowest modulus of rupture (MOR) and modulus of elasticity (MOE) and the highest water absorption, while samples that were injection-molded exhibited the highest MOR (70 MPa) and MOE (7 GPa) and low water absorption (2%).

Key words Cellulose–plastic composite · Compression · Extrusion · Injection molding · High wood content

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Introduction

In recent years, interest in composites based on renewable materials has grown tremendously because of social requests for low environmental stress, low-maintenance and high-durability products, and ultraviolet (UV) durability.[1–3] Construction, transportation, industrial, and consumer applications for wood/cellulose–plastic composites (WPC) are all on the rise. WPC have been primarily produced with a low and medium percentage of wood/cellulose. Products typically contain approximately 50% (by weight) wood/cellulose, although some composites contain very little wood/cellulose and others as much as 60%.[1,4–7] Wood/cellulose content may range from 70% to 90% and the interfacial adhesion between wood/cellulose particles and polymers can be improved by adding a compatibilizer. One of the major challenges for WPC is to optimize the content of the wood/cellulose in WPC to reduce costs and compete with solid wood and engineered wood composite products.

Cellulose, one of the basic structural components of wood fibers, has great potential for the preparation of novel composite materials with thermoplastic resins. However, the highly polar nature of the cellulose surface does not lend itself to strong interactions with the nonpolar polymer, such as polypropylene (PP).[8,9] Various fiber surface compatibilizers have been reported, such as silane, alkali, combination of alkali and silane, monomer grafting under UV radiation, maleic anhydride-grafted polypropylene, and others.[8–11] Among these treatments, coupling agent maleic anhydride-grafted polypropylene (MAPP) has been found to be the most efficient in improving the mechanical properties of cellulose composite materials.

However, extruded WPC are a relatively new product for which market growth has been rapid. The engineering design of extrusion operations requires proper knowledge of the flow mechanism of these highly filled melts. The WPC formulations also require reliable evaluation of flow performance of the composite melts. Rheological characterization affords knowledge about both the fundamental flow behav-

Table 1. Basic properties of all cellulose powders

Cellulose	Color	Structure	Fiber length (μm)	Fiber diameter (μm)	Bulk density (g/l)
BE00	White	Short fiber	120	20	150–180
BC200	White	Medium fiber	300	20	60–80
B400	White	Long fiber	900	20	20–40

ior of the highly filled plastics and practical methods for evaluating the flow performance for process development. The technical challenges are to overcome flow problems, the low bulk density of the wood/cellulose as well as separation problems of the composite mixture. Therefore, advancing this area of knowledge for this new material class is important for further developments of the science and industry.[4,12–14]

In this article, a study of the properties of composites made of cellulose microfibers and random PP is reported. Composites prepared with different fiber lengths of cellulose and with the different content (70%, 80% and 90%) were studied. A study on the mechanical and water absorption of compression-, extrusion-, and injection-molded composites is presented here. Melt flow characteristics such as melt index, have been studied with special reference to the effect of blend ratio, cellulose fiber length, temperature, and diameter of die hole. The effect of the different fiber length and content of cellulose on the morphology and water absorption of the composites is discussed. Finally, our ultimate aim is to make clear the possibility of the preparation of composites with very high wood/cellulose content.

Experimental

Materials

The materials used in this study were cellulose, a thermoplastic polymer, polypropylene (PP), and the coupling agent MAPP, which is polypropylene modified with maleic acid. The thermoplastic polymer was a random PP (PM930V, MFI 30 g/10 min, density 0.9 g/cm^3) supplied by SunAllomer (Tokyo, Japan). Three different shapes and sizes of fibrous cellulose (BE00, BC200, B400) were products of Rettenmaier and Söhne (Rosenberg, Germany), produced from softwood. The basic properties of all the cellulose powders are listed in Table 1. The coupling agent was MAPP (umex1010) obtained from Sanyo Kasei (Tokyo, Japan).

Blending procedure

The cellulose fibers, PP, and MAPP were sequentially fed into a conical twin-screw extruder (Model Taitan 80, Cincinnati). The temperature profile of the extruder from cylinder 1 through cylinder 4 was kept between 230° and 180°C and the extruder was operated at a screw speed of 7 rpm. After compounding, the composite was then crushed into a cutter mill (wood grinder) or hammer mill and used for compression molding. Extruded strands were also pelletized and the temperatures from cylinder 1 to cylinder 4 were decreased from 230° to 180°C. Pellets were used for extrusion-molding and injection-molding composites. Crushed pellets used for injection samples were also prepared with the addition of 2% (w/w) calcium stearate as a lubricant. The compositions of cellulose powder, PP and MAPP are listed in Table 2.

Table 2. Composition of cellulose powder, polypropylene (PP), and maleic anhydride-grafted polypropylene (MAPP)

No.	Composition (%, w/w)				
	Cellulose			PP	MAPP
	BE00	BC200	B400	PM930V	UMEX1010
1	80			18	2
2		70		28	2
3		80		18	2
4		90		8	2
5			80	18	2

Molding process

Three molding processes commonly used for thermoplastics were used to make test specimens of cellulose/PP/MAPP for studying the effect of molding on the physicomechanical properties of the corresponding cellulosic plastics.

Compression molding

The specimens for mechanical tests were prepared by compression molding with an oil hydraulic press HP-1B-P (method A) at 200°C. Other press conditions were set to provide the best opportunity to reach the theoretical density. The theoretical density was calculated on the basis of weight percent and the density of components.

Specimens were cut from the sheets of the cutter mill compound molded by method A (AC) and the hammer mill compound molded by method A (AH). The blends were also compression-molded using a steel mold (mold dimensions $1 \times 8 \times 10$ cm) by a Shinto hot press model HCC-BSN-2 (method B) at 220°C and 11 MPa for 5 min, before being cooled to room temperature. Teflon films were used to avoid the adhesion of MAPP to the stainless surface of the mold. Twelve standard specimens (dimensions $1 \times 2.5 \times 10$ cm^3) were cut from the sheets of the cutter mill sample molded by method B (BC).

Table 3. Molding conditions of injection mold

No.[a]		Lubricant (%)	Injection time (s)	Nozzle temperature (°C)	Peak pressure (MPa)
1	IX	0	0.29	210	29.6
	IO	2	0.30	195	29.4
2	IX	0	0.57	185	22.7
	IO	2	0.60	185	23.9
3	IX	0	0.29	210	29.5
	IO	2	0.31	195	29.5

IX, injection mold without lubricant; IO, injection mold with lubricant

[a] Same as in Table 2

Extrusion molding

Pellets were extruded as thick long sheets in this process. Pellets were fed into a conical twin-screw extruder (Titan 68, Taiyo Gosei, Kagawa, Japan). The molding conditions were as follows: temperatures on cylinder 1 to cylinder 4 were held between 215° and 170°C and the temperature of exit die from heating zone to cooling zone declined from 175° to 45°C from the heating zone to the cooling zone. The extruder was operated at a screw speed of 5 rpm. The resulting sheets were cut lengthwise (EL) and widthwise (EW) to make the test specimens.

Injection molding

The injection molder used was a Roboshot a-100C (Fanac). The molding conditions are listed in Table 3. The resulting dumbbell-shaped mold without lubricant (IX) and mold with lubricant (IO) were used appropriately to test mechanical properties. A regular injection nozzle was used, with a nozzle diameter of 2 mm.

Melt flow index

Melt flow index (MFI, weight of polymer in grams extruded in 10 min through a capillary), was determined using a Dynisco melt flow indexer LMI 4000 series in accordance with Japan Industrial Standards (JIS) K 7210. The applied loads and dice diameters were 5 and 10 kg, and 6 and 7 mm, respectively. The measurement was carried out at 230°C.

Mechanical test

The small rectangular specimens were tested on a Shinto Model TCM-500 and the load–deflection curve was determined. The support span depended on the length of the mold and the crosshead speed was 5.0 mm/min. At least 12 specimens were tested for each composite in accordance with JIS A5908. All specimens showed a yielding fracture mode. The modulus of rupture (MOR) and modulus of elasticity (MOE) were calculated from the load–deflection curve. The MOR was defined as the first point on the load–deflection curve to show a slope of zero. The MOE was determined from the slope in the initial elastic region of the load–deflection curve.

Water absorption test

The samples based on increasing fiber length, diameter, and cellulose content of compression molding were tested for water absorption for 0 to 24 h under atmospheric pressure at room temperature. Water absorption in weight gain (Wt), and thickness gain (Th) were calculated by Eqs. 1 and 2, respectively:

$$Wt = (W_1 - W_0)/W_0 \tag{1}$$
$$Th = (t_1 - t_0)/t_0 \tag{2}$$

where W_1, W_0, t_1, and t_0 are the weight of the composite containing water, weight of the dried composite, thickness of wet composite, and thickness of dry composite, respectively.

Morphology of composites

Microscope images of cellulose fiber of fiber length 120, 300, and 900 μm and the fractured surface after mechanical testing were taken using CCD Moritex Inf 500 DA at different magnifications.

Results and discussion

Possibility of compression, injection, and extrusion molding

Table 4 illustrates the feasibility of compression, injection, and extrusion molding. It is noted that the feasibility is highly dependent on both the fiber content and length of fiber. Unfortunately, feasibility studies on extrusion molding and injection molding of the highly loaded wood/cellulose plastics are scarce in the open literature. Two extrusion-molded composites and three injection-molded composites have been developed.

Melt flow rate

Flowability of compound is one of the most important factors to be considered when establishing injection molding and extrusion molding.[4,15] Figures 1 and 2 show the experimental results obtained for the melt flow index of pellets. The melt flow rate provides valuable information about the

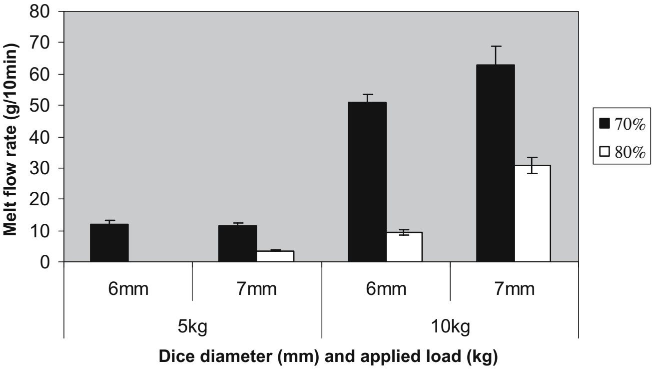


Fig. 1. Melt flow indexes of pellets containing 300-µm-long cellulose with different cellulose/plastic compositions. *Error bars* indicate standard deviations. *Filled bars*, 70/28/2 cellulose/polypropylene (PP)/maleic anhydride-grafted polypropylene (MAPP); *open bars*, 80/18/2 cellulose/PP/MAPP

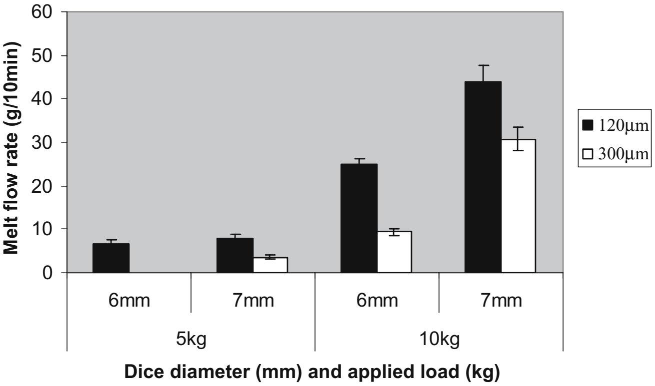


Fig. 2. Melt flow indexes of pellets with 80%/20% (w/w) cellulose/plastic composition and different fiber lengths. *Error bars* indicate standard deviations

Table 4. Feasibility of compression, injection, and extrusion molding of compositions of cellulose fiber/PP/MAPP

No.	Molding process				
	Compression		Extrusion	Injection	
	Hammer	Cutter		IO	IX
1	✓	✓	✓	✓	✓
2	✓	✓	×	✓	✓
3	✓	✓	✓	✓	✓
4	✓	✓	×	×	×
5	✓	✓	×	×	×

Ticks indicate that molding is feasible; crosses indicate that molding is not feasible
[a] Same as in Table 2

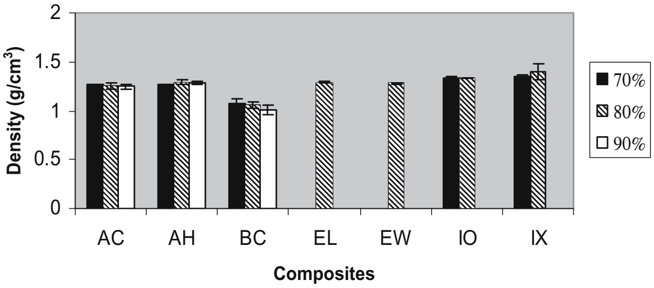


Fig. 3. Density of composites of different fiber content of 300-µm cellulose. *Filled bars*, 70/28/2 cellulose/PP/MAPP; *shaded bars*, 80/18/2 cellulose/PP/MAPP; *open bars*, 90/8/2 cellulose/PP/MAPP. *AC*, cutter mill sample molded by method A; *AH*, hammer mill sample molded by method A; *BC*, cutter mill sample molded by method B; *EL*, extrusion mold sample in lengthwise direction; *EW*, extrusion mold sample in widthwise direction; *IO*, injection mold with lubricant; *IX*, injection mold without lubricant

flow behavior of materials. MFI experiments were done at 230°C. It was found that the MFI value decreased with an increase of cellulose content because the incorporation of rigid material to the polymeric matrixes limited their free mobility and increased the material apparent viscosity. Formulations with 15% and 10% resin loadings did not flow and MFI values were unable to be measured. Caraschi and Leão[16] obtained similar results in which the increase of fiber amount decreased the MFI value. On the other hand, 300-µm fiber length cellulose pellets showed lower values than 120-µm cellulose pellets. For each pellet, an increase in dice diameter increased the melt flow, especially with 10-kg load. With 5-kg load, the 20% resin content compound could not be measured with 6-mm diameter. However, it could be measured with 7-mm dice. With 10-kg load, compound with 20% resin content could be measured with both dice of 6- and 7-mm diameter. The compound with 30% (w/w) resin content had remarkably low fluidity relative to a general plastic material. Therefore, it was concluded that desirable conditions for measurement were 5-kg load/7-mm dice and 10-kg load/6-mm dice. A good extrusion-molded and injection-molded composite could be obtained at this temperature (230°C). However, degradation of cellulose fibers starts at 180°C.[17]

Composite density

Effect of resin content

Method A. Figure 3 presents the variations in the composite density for formulations with different plastic content in compression, injection, and extrusion molding. When the resin content was 20% or less, the densities of hammer mill compounds were higher than those of cutter mill compounds, probably because the relative composition of component reinforcements, their aspect ratio, and also the extent of chemical modification influences the overall rheological behavior. Fluidity was low when the resin content was low. There was a tendency for the particle sizes of hammer mill compounds to be smaller than those of cutter mill compounds. As a result, the aspect ratio became larger for cutter mill fiber. For this reason, the compound with the same resin content may have shown low fluidity and caused the density difference of the composites AH and AC. Because the fluidity was high for 30% resin content, there was little influence of the pulverization method on the densities of the composites. No remarkable relationship was observed between the resin content and product density. Method A composite was prepared under a press condition setup to attain a density close to the theoretical density. Therefore, all press pressures and press times were different. When the resin content was high, the theoretical density was attained at low pressure and with a short press time. However, high pressure was needed when the resin content was low. In this case, a compound of low fluidity would be crushed during the molding process. If the fiber shape changed during the molding process, the resin content may have affected various performances. On the other hand, press molding has merit that can be applied for a wide range of molding conditions. Therefore, a wide range of resin contents was selected to make composites by compression molding.

Method B. The densities of composites also decreased with increased cellulose content. Hence, the void ratio of these composites increased with increasing fiber content. A possible explanation for the voids is the movement of air from within the cell lumens to the cell wall–PP interface during compaction.[18] Again, the fluidities of the compounds were high when the resin content was high. Because method B molding was carried out under the constant press conditions, compounds of high fluidity were expected to give more compact composites. The melt flow characteristic was peculiar for each formulation and could be estimated because the technique used in this experiment exerted little influence, except on flow characteristics, on the various product performances.

Effect of fiber length of cellulose

Figure 4 presents the density of composites from compression, injection, and extrusion molding with different fiber lengths and 80% fiber content. A hammer mill produces finer powder than a cutter mill. Therefore, when the fiber length was long, the hammer mill compound was more finely powdered, ensuring high melt fluidity, and so the composite density was high. When fiber length was short, this influence was not clear because the fiber was originally small and fine. In addition, packing may have been influenced by fiber length. When fibers were long, maximum packing may have occurred with optimum particle size distribution leading to high density, but such a conclusion can be only tentative with a density difference of this magnitude. Composite density decreased when the fiber length was increased.

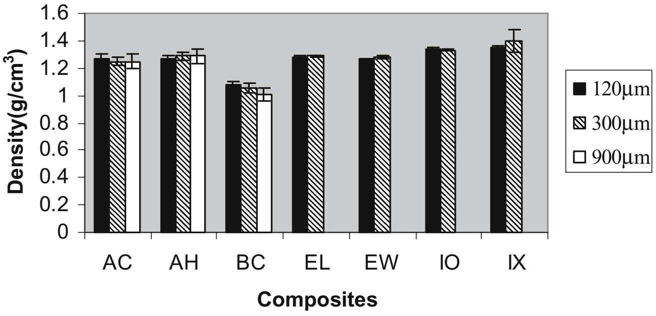


Fig. 4. Density of composites of different fiber lengths of 80% fiber content and 20% plastic content. *Error bars* indicate standard deviations

Injection molding and extrusion molding

In the case of injection-molded composite without processing aids, the density decreased with increasing plastic content. The density was reduced with lubricant at a fiber content of 80%; hence, the processing aid developed some voids. Similar observations were made in the case of comparison of fiber length.

Mechanical test

Compression molding

Method A. The effects of PP content on MOE and MOR of the cellulose/PP/MAPP composites are shown in Figs. 5 and 6, respectively. Having the compound crushed by a hammer mill tended to decrease the MOR with the increase of resin content from 20% to 30%. If the extent of phase separation was very severe, the strength of the materials

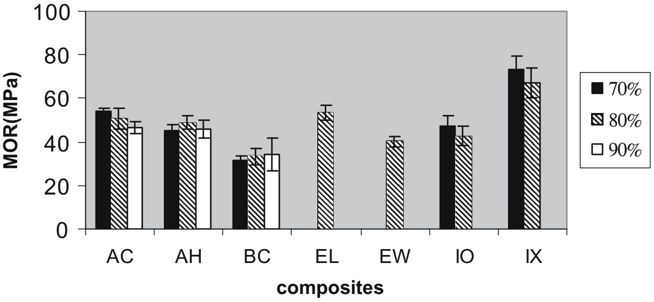


Fig. 5. Effects of fiber content on modulus of rupture (MOR) of composites of 300-μm cellulose. *Error bars* indicate standard deviations. *Filled bars*, 70/28/2 cellulose/PP/MAPP; *shaded bars*, 80/18/2 cellulose/PP/MAPP; *open bars*, 90/8/2 cellulose/PP/MAPP

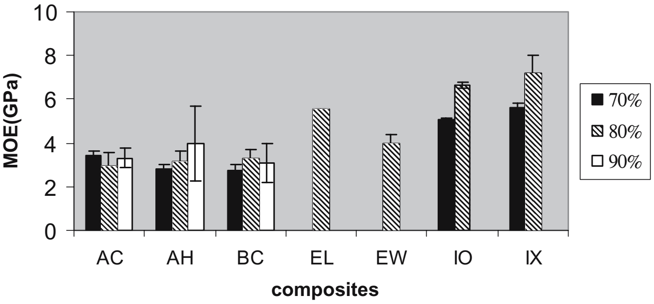


Fig. 6. Effects of fiber content on modulus of elasticity (MOE) of composites of 300-μm cellulose. *Error bars* indicate standard deviations. *Filled bars*, 70/28/2 cellulose/PP/MAPP; *shaded bars*, 80/18/2 cellulose/PP/MAPP; *open bars*, 90/8/2 cellulose/PP/MAPP

decreased.[19] The MOR of cutter mill composite simultaneously increased with an increase in PP content, suggesting that a relatively strong interaction exists between cellulose and PP.[19] The cutter mill compound contained fibers with a high average aspect ratio, and so they oriented easily under conditions of high fluidity. Because the press pressure of method A was high, it was easy for a cutter mill compound to orient under high fluidity press conditions with 30% resin content. The bending strength and the elastic modulus were high as expected. Because a compound of 20% resin content had low fluidity, the resulting composite did not differ much from the composite of hammer mill compound. In addition, the MOE of the composite AC decreased as the fiber length of cellulose increased.

Method B. There was a tendency for the bending strength and the elastic modulus to decrease with the resin content, although the tendency was not remarkable.

Effect of fiber length of cellulose

In Figs. 7 and 8, the enhancing effect of the fiber length of cellulose in the mechanical properties is clearly shown. While 300- and 900-µm fibers did not have much effect on MOE, the MOE of the composite BC increased as the fiber length increased. However, the mechanical properties of composites with 900-µm fiber length showed high variation (large standard deviation; SD). Because the fibers were long, the resin did not disperse uniformly. There was also a suggestion of a lack of miscibility between cellulose and PP/MAPP.

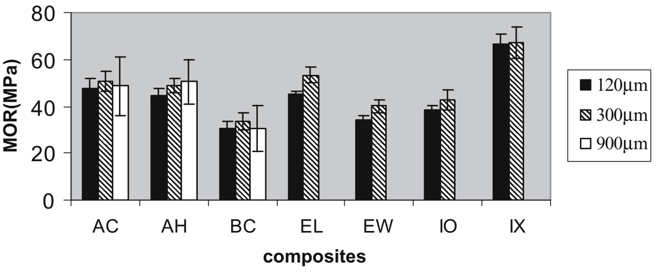


Fig. 7. Effects of fiber length on MOR of composites of 80% fiber content and 20% plastic content. *Error bars* indicate standard deviations

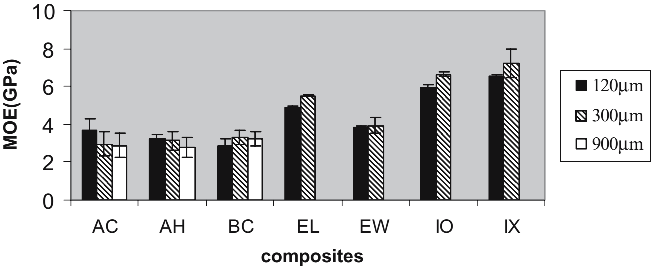


Fig. 8. Effects of fiber length on MOE of composites of 80% fiber content and 20% plastic content. *Error bars* indicate standard deviations

Effect of method A, method B, injection and extrusion molding

Widthwise extrusion composite (EW) had lower MOR and MOE than those of the lengthwise extrusion composite (EL). The injection molding without lubricant (IX) had higher MOR and MOE than those of the injection molding with lubricant (IO). Composite BC had lower MOR and MOE than those of composite AH and composite AC because of low density. The volume of BC was high with plenty of voids, and as a result, bonding of the resin and fibers was decreased. However, the injection molding without lubricant of 80% PP gave the highest MOR and MOE.

Water absorption

Effect of resin content

Method A. As shown in Figs. 9 and 10, water absorption increased as the resin content decreased. It is considered that water absorption took place initially by permeating voids and by binding with cellulose. Therefore, molded composites with densities significantly different from theoretical densities may have undergone high water absorption. Water absorption is also considered in the molded composite of near-theoretical density. This is explained in terms of water permeating through the part that has not

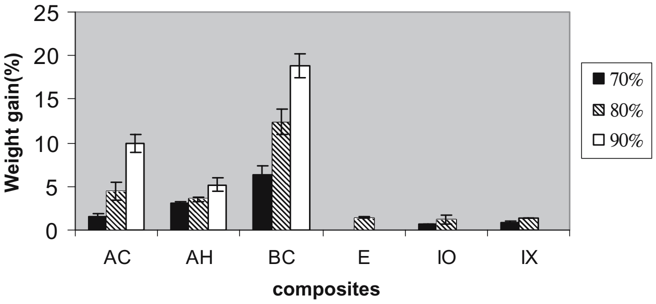


Fig. 9. Effects of fiber content on weight gain of composites of 300-µm cellulose. *Filled bars*, 70/28/2 cellulose/PP/MAPP; *shaded bars*, 80/18/2 cellulose/PP/MAPP; *open bars*, 90/8/2 cellulose/PP/MAPP. *E*, Extrusion molding. *Error bars* indicate standard deviations

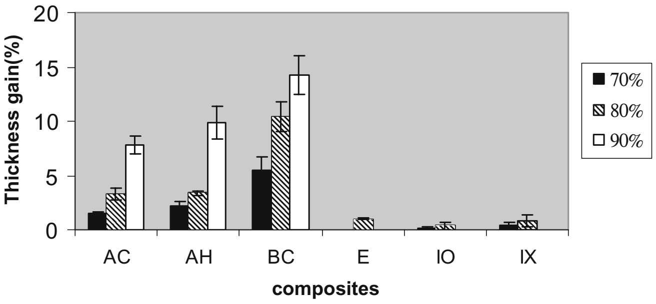


Fig. 10. Effects of fiber content on thickness gain of composites of 300-µm cellulose. *Filled bars*, 70/28/2 cellulose/PP/MAPP; *shaded bars*, 80/18/2 cellulose/PP/MAPP; *open bars*, 90/8/2 cellulose/PP/MAPP. *E*, Extrusion molding. *Error bars* indicate standard deviations

been combined with resin, and remains as cellulose-binding water and induces swelling of the composite. In addition to expansion due to the cellulose-binding water, a tangle of fiber was destroyed with the water that permeated into the composite. Therefore, expansion of the molded composite was large for composites with high water absorption. The thickness swelling increased as the resin content decreased.

Method B. Water absorption increased as the resin content decreased. This composite was porous and contained many void spaces; hence, much water was absorbed. The ratio of water absorption to the theoretical void (measured density/theoretical density) showed a parallel tendency. Because the composite did not absorb water completely when immersed in water for 24 h, it was expected to be similar the method A molded composite in that water had permeated between fibers, and so on as above. The thickness swelling increased as the decrease of resin content. The factor controlling thickness swelling was the same as for method A molded composite. Therefore, a composite with high water absorption shows high expansion, and so on.

Effect of method A, method B, injection and extrusion molding

Method A molded composite had little water absorption and expansion compared with method B molded composite. The difference is explained on the basis of density difference and void ratio. For the composites of 30% resin content, thickness swelling per 1% of water absorption was larger for method A composite than for method B. Because the composite with 30% resin was high in resin, there were few fibers that were not covered with resin. Therefore, when there are many voids inside the composite, cellulose/PP/MAPP composite of high density tends to expand more by water absorption. Injection-molded samples showed the least water absorption. The values obtained for water absorption in injection-molded composite with lubricant and without lubricant were similar. Although micrographs did not show a clear difference in this study, more coverage of fiber by resin is expected for injection-molded composites.

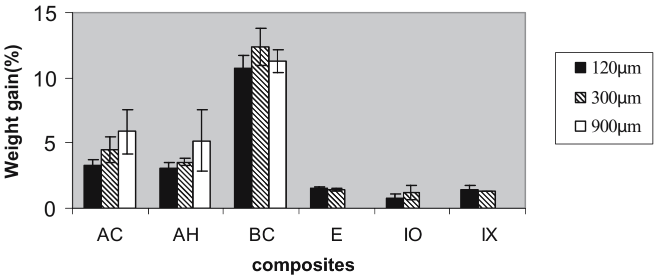


Fig. 11. Effects of fiber length on weight gain of composites of 80% fiber content and 20% plastic content. *Error bars* indicate standard deviations

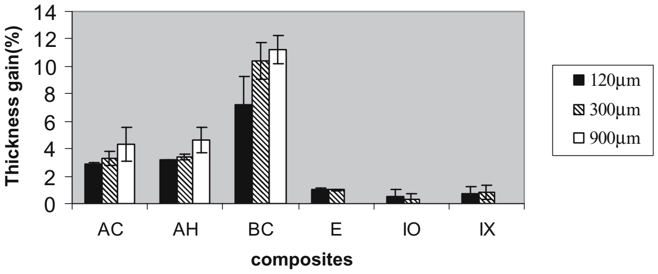


Fig. 12. Effects of fiber length on thickness gain of composites of 80% fiber content and 20% plastic content. *Error bars* indicate standard deviations

Effect of fiber length of cellulose

Figure 11 shows the trend in weight gain of increasing fiber length of cellulose, while Fig. 12 represents the thickness gain of the increasing fiber length of cellulose. Composite with fiber length of 120 μm had low swelling per 1% of water absorption. Because fiber length was short, tangles between fibers were less developed. Water absorption was near the theoretical ratio of void, but because water absorption for 24 h in water did not reach the saturation level throughout the composite, it is thought that, like method A molded composite, water permeated into the space between fibers. It was also observed that water absorption increased with an increase in fiber length. Therefore, as the entangled part of fibers increased, inducing more clearance in the composite, the bonded part of fibers with resin may have decreased. Method A molded composite had little void. Therefore, it is expected that water permeated and was absorbed by the fiber that was not bonded with resin. With longer fibers, it is possible that the amount of fiber not bonded with resin increased, which led to increased water absorption. The thickness swelling increased as the fiber length become long; tangles of fiber may have been destroyed with the permeated water, the water bound to cellulose may have increased, and the composite expanded. Therefore, longer fibers with many tangles between fibers may show high expansion.

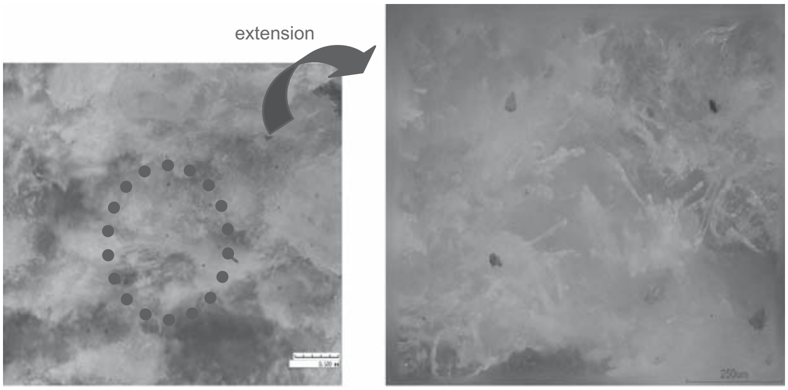
Morphology of composites

Effect of resin content on the morphology of the fracture surface

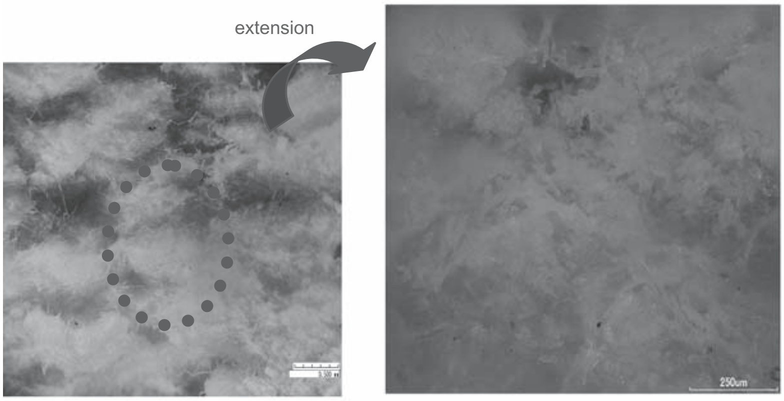
CCD micrographs of fractured surfaces of compression-molded composite containing 30%, 20%, and 10% PP are shown in Fig. 13. As the resin content decreased, more fluff of fiber was detected. The occurrence of fluff is considered to be a starting point of fracture. This observation is more pronounced with composites of low resin content, so it is assumed that strength of the composite is maintained by tangles between fibers. As the resin content decreased, more fiber ball-like material was observed on the fracture surface. In composite with 30% PP, the fiber was dispersed into the matrix and coated with PP. For the composite

Fig. 13. Micrographs of fractured surfaces of compression-molding composites containing 30%, 20%, and 10% PP/MAPP

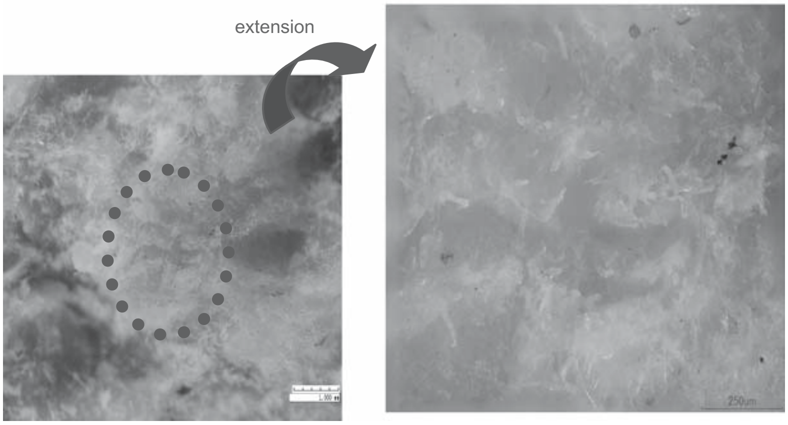
PP = 30%



PP = 20%



PP = 10%



containing 10% PP, a fibrous particle of cellulose was partially covered by the PP, with part of the surface still not covered. This proves that a fully developed interfacial interaction was not developed in the 10% PP composite.

Effect of cellulose fiber length on the morphology of the fracture surface

Microscope images of cellulose fiber and fractured surfaces of compression-molded composite with increasing fiber length of cellulose are shown in Fig. 14. When long fibers were present, they were conspicuous on the fracture surface. A stratified pattern was conspicuous on fracture surfaces when fibers were long. Fracture of composites took place at weak parts of the composite where fibers were not been well bonded with resin. Fluff was more conspicuous for composites with long fibers. On other hand, lamination also occurred when fibers were long. This phenomenon should have some influence on the strength. The cellulose fibers with fiber lengths of 120, 300, and 900 μm partially changed into microparticles during melt-extrusion processing. In the case of 120-μm fibers, homogeneity of the blended mixture was observed, indicating that it should serve as a good polymer matrix.

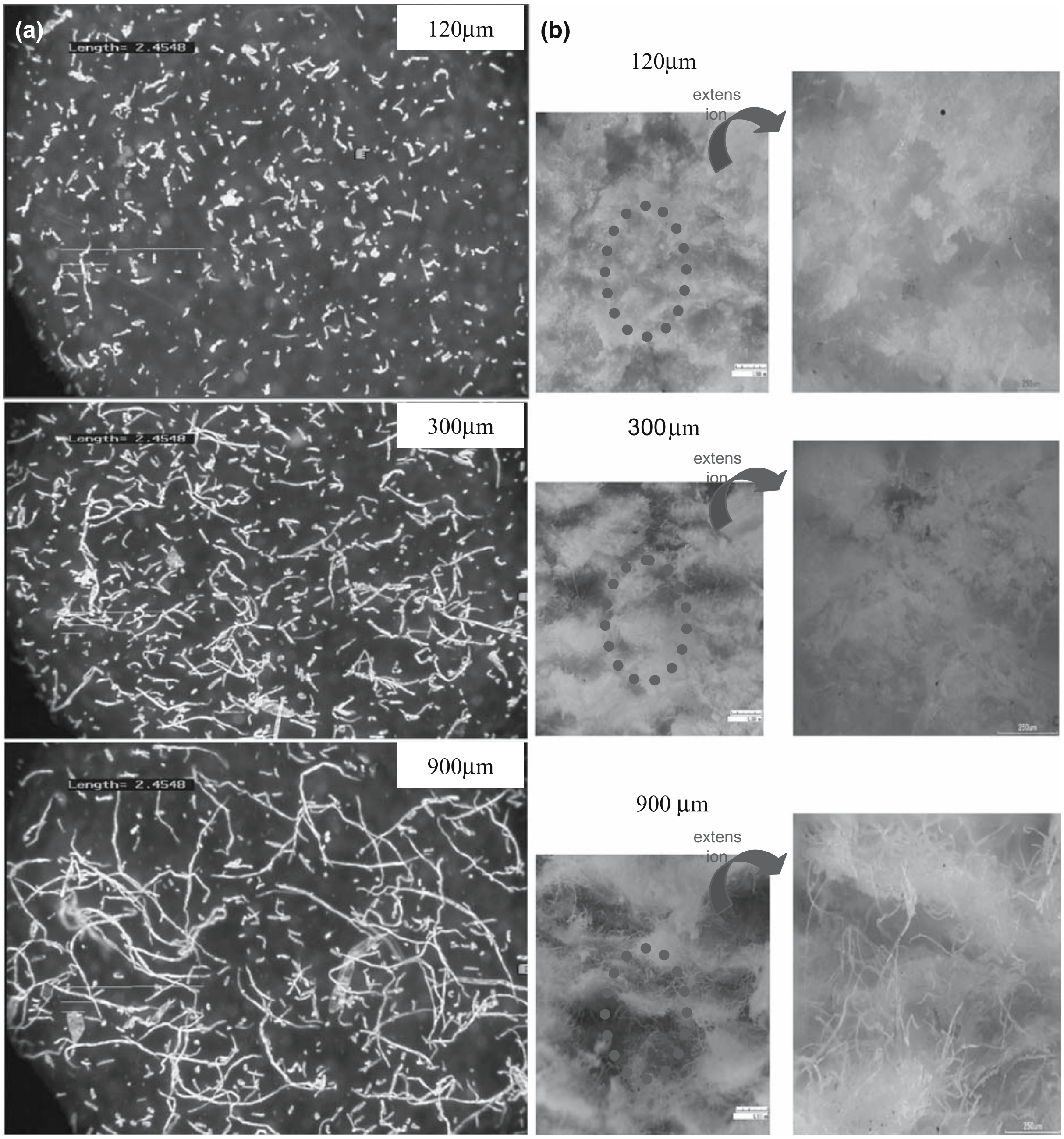


Fig. 14a,b. Micrographs of **a** cellulose fibers of 120μm (BE00), 300μm (BC200), and 900μm length (B400), and **b** fractured surfaces of compression-molded composites containing cellulose fibers of 120μm (BE00), 300μm (BC200), and 900μm length (B400)

Effect of compression, injection, and extrusion molding

Micrographs of fractured surfaces of extrusion moldings and injection moldings are shown in Fig. 15. The interfacial interactions were limited in the compression-molding composites, probably because of a smaller number of ester bonds between highly fibrous cellulose and the MAPP/PP matrix.[8,9] The micrographs of extrusion-molded composites EW no. 1 composition (120μm fiber length, 80% cellulose) clearly show gaps and voids in the PP matrices as well as along the cellulose fibers, the surfaces of which are smooth and practically intact, and that there exist some aggregates of cellulose fibers. Much improved distribution of particles of cellulose was found in the injection molding with lubricant IO and without lubricant IX with 120-μm fibers and 80% cellulose. The micrographs also reveal a marked improvement in interfacial adhesion between cellulose particles and the MAPP/PP matrix, which was brought about by increased area of surface contact. In this stage, the fiber was not uniformly dispersed in the melted PP matrix.

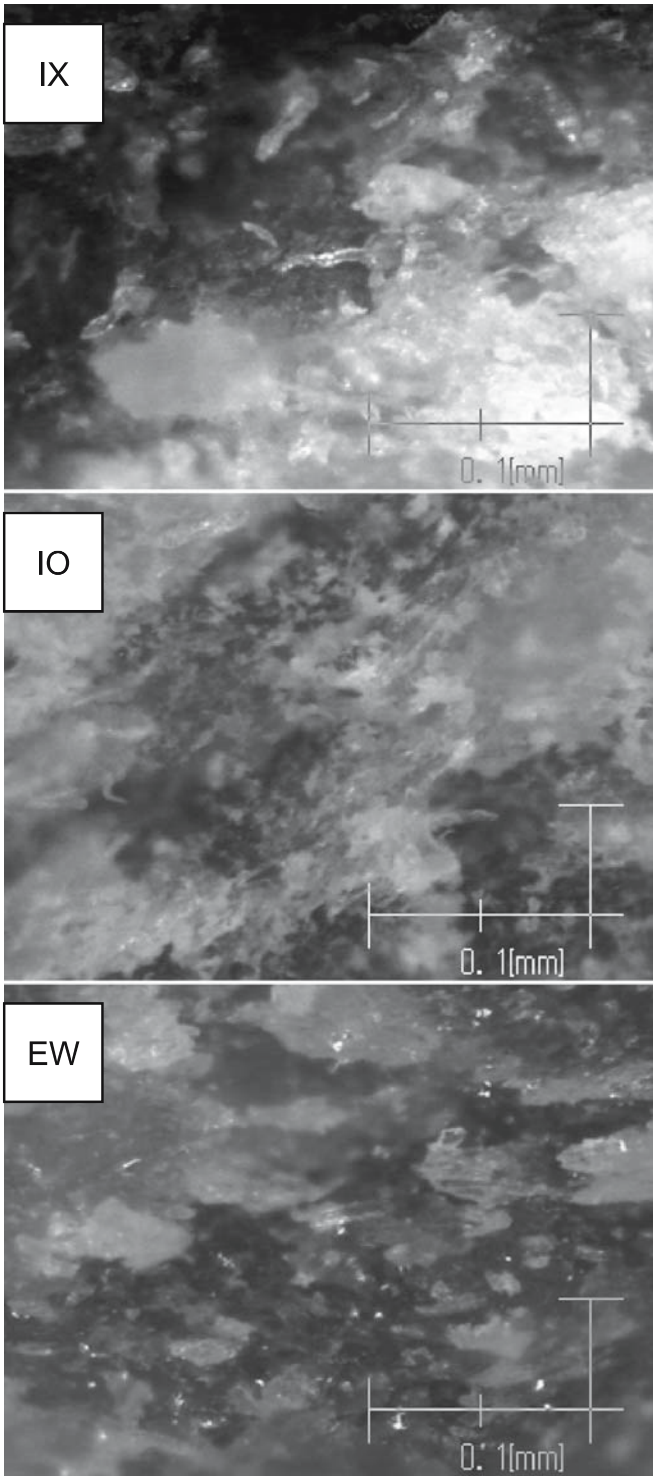


Fig. 15. Micrographs of fractured surfaces of extrusion molding and injection molding of composition of 80% fiber content and 20% plastic content. *IX*, Injection molding without lubricant; *IO*, injection molding with lubricant; *EW*, extrusion mold sample in widthwise direction

Conclusions

Cellulose/PP/MAPP, a wood–plastic material based on a renewable resource, has excellent potential to prepare wood based material with high content of cellulose as "green," eco-friendly composites. Better flowability of pellets was achieved with higher levels of cellulose and PP/MAPP at 230°C. It is encouraging to see that with suitable selection and content of plastic, the wood/cellulose–plastic composites can achieve stiffness and toughness properties comparable with other wood composites. Extrusion allows better mixing of cellulose, thermoplastic, and additives because of the high shear of extrusion processing, as shown by CCD micrographs. The mechanical and physical properties of composites vary considerably depending on the plastic content, shape and size of cellulose fiber, and molding methods. The mechanical properties of composites are influenced mainly by the adhesion between the matrix and tangle of fibers. The results for the mechanical properties also supported the existence of a certain degree of miscibility in the composite plastics. Because of loosely arranged fibers, composites with 900-μm fibers and 90% plastic content had the highest water swelling among the composites. The trend of water resistance and mechanical properties was injection molding > extrusion molding > compression molding by method A > compression molding by method B. We conclude that injection- and extrusion-molded composites of cellulose in PP with MAPP give the best mechanical and physical properties.

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