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Structure and properties of composites of highly crystalline cellulose with polypropylene: Effects of polypropylene molecular weight

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Abstract

Composite of highly crystalline fibrous cellulose (CE) and polypropylene (PP) of different molecular weights ($\overline{M_w}$) was prepared via melting–mixing, maleic anhydride grafted polypropylene (MAPP) was used as a compatibilizer. And the effects of molecular weight of PP on the properties of the composites were investigated. Through the studying of mechanical properties, dynamic mechanical properties, melting and crystallization behaviors, thermo-oxidative properties, water absorption behaviors, and the morphology of the composites, it was found that PP with higher molecular weight revealed stronger interfacial interaction with cellulose in the composites. Compared with the lower molecular weight, the composites derived from higher molecular weight of PP exhibited stronger tensile strength at the same cellulose content.

Keywords: Polypropylene (PP); Cellulose; Molecular weight; Composites; Mechanical properties

1. Introduction

Polypropylene as one of the most popular versatile thermo-plastic polymers, provides many advantages with regard to its low cost, recyclability, and high thermal stability, and has yielded many kinds of composites. The reinforcing materials have been extensively used in plastics industry are mineral fillers and fibers. Considering the serious environmental problems caused by the growing consumption of synthetic polymers, it is potentiated to minimize the use of the synthetic polymers derived from petroleum and also to maximize the use of renewable resources. So the considerable attention has been picked up momentum to the natural fibers both in the literature and in industry in recent years [1–8]. Advantages of natural fibers over conventional reinforcement such as glass or carbon fibers are lower cost, low density, biodegradability, as well as high specific properties. The natural fibers can be grouped into bast (jute, banana, flax, hemp, kenaf, mesta), leaf (pineapple, sisal, screw pine), and seed or fruit fibers (coir, cotton, oil palm) [9]. For some

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natural fibers of plants origin, cellulose is the main component. Cellulose-reinforced polymer composites are characterized by low cost, low density, high stiffness, biodegradability, and good mechanical properties as well as availability of renewable natural resources. The main disadvantage of cellulose-reinforced polymer composites is the inherent incompatibility between the hydrophilic cellulose fibers and the hydrophobic thermo-plastic matrix, particularly, the polyolefin such as polypropylene (PP). As a result, lack of interfacial adhesion between cellulose and polypropylene results in the unsatisfactory properties of the resultant composites. To overcome the disadvantage, many efforts have been carried out to improve the interfacial adhesion between fibers and matrix [1,2,9–24]. Among these methods, maleic anhydride grafted polypropylene (MAPP) was proved to be very efficient in improving interfacial adhesion of fibers and polypropylene matrix.

A lot of works have been done in the cellulosic composites, such as making use of various cellulosic materials and different matrixes, surface treatment of cellulosic materials, modification of matrixes, processing conditions, characterization of the composites, and so on [1]. We reported elsewhere [25,26] that MAPP is a good compatibilizer for cellulose and PP, and the effects of MAPP content as well as the cellulose content on the properties of the composites were investigated. Recently, we found that the enhanced interfacial esterification between MAPP and activated cellulose had much effect on the properties of the cellulose composites, the resultant composites show a greater improvement in tensile strength, and the evidence of the esterification were observed directly in the melt–mixed composites [27–29]. The un-grafted free MA monomers remained in the matrix gives a slight positive effect to PP composite while a negative effect to MAPP composite [30]. The graft degree of MAPP on the properties of cellulose composites was studied, too, results showed that even an amount of grafted MA as small as 0.2 wt.% is sufficient to impart satisfactory tensile strength to the resulting composites [29,30]. However, the relationship between molecular weight of PP matrix and properties of the composites is not clear yet. In this work, we concentrated upon the effect of molecular weight of polypropylene on the properties of composites of PP and the highly crystalline fibrous cellulose compatibilized by MAPP.

2. Experimental

2.1. Materials

Three kinds of isotactic polypropylene pellets were obtained commercially from Aldrich (Milwaukee, WI). They were notated as PP1, PP2, and PP3, of which the molecular weights given by the supplier are $\overline{M_w} \approx 250,000$ [melt flow index (MFI) 12 g/10 min, density 0.900 g/cm^3], $\overline{M_w} \approx 340,000$ (MFI 4 g/10 min, density 0.900 g/cm^3), and $\overline{M_w} \approx 580,000$ (MFI 0.5 g/10 min, density 0.900 g/cm^3), respectively. Polypropylene-graft-maleic anhydride (MAPP pellets, ≈0.6 wt.% MA; melt index 115; density 0.950 g/cm^3, as reported by the supplier) was also obtained from Aldrich. Fibrous cellulose {CF11, 50–350 μm in length, about 20 μm in diameter; crystallinity 93% (cellulose I type) [31]} was supplied by Whatman Int. Ltd. (Maidstone, England).

2.2. Preparation of composites and samples

PP and CF11 were dried in vacuo at 70 °C for 3 days, and MAPP was pretreated in vacuo at 120 °C for 15 h before preparing composites. All composites with a 30 wt.% cellulose content were prepared by a melt-mixing method with a Toyoseiki Labo Plastomill 30C150 Rheomix (the volume of the mixing chamber: 60 cm^3). The mixing was carried out in air at 190 °C at 55 rpm for 20 min.

Sheets of 1 mm thickness were obtained by using the resulting composites and compression molding in a Shinto press (Shinto Metal Ind. Ltd.) at 200 °C, under a pressure of 3 MPa for 6 min, followed by cooling under a pressure of 8 MPa to room temperature at 2 °C/min. Teflon films were used to avoid the adhesion of MAPP to the stainless surface of the mold. The sheet sample was then placed between two stainless steel plates, maintained in vacuo at 60 °C for 24 h in an oven, then switched off the power of the oven, and continued the vacuo at room temperature over night. Standard specimens were cut from the sheets for the measuring of properties.

For comparing, pure PP as well as pure MAPP sample was melt–mixed and thermal pressed under the same conditions, as described above. And all the samples for the property measurements were obtained in the same conditions.

2.3. Tensile tests

Tensile properties were measured with a Shimadzu AGS-5kNG universal testing machine in accordance with the testing method for tensile properties of plastics (at a strain speed of 5 mm/min) [32]. Samples were stored in a room with a constant temperature of 20 °C and a constant humidity of 65%, over 24 h before the measurement. The results were obtained from six parallel tests.

2.4. Dynamic mechanical analysis (DMA)

Dynamic mechanical analysis (DMA) of composite was performed with a Perkin–Elmer DMA 7e using a three point bending-rectangular measuring system at a frequency of 1 Hz. The sample of $15 \times 3\ \mathrm{mm}^2$ area and 1 mm thickness was cooled to −30 °C, maintained at the temperature for 3 min, and then heated to 130 °C at 5 °C/min in nitrogen atmosphere. The static and dynamic forces were 330 mN and 300 mN, respectively. The visco-elastic parameters, i.e., the mechanical loss factor $\tan\delta$ and storage modulus E' were recorded as a function of temperature.

2.5. Scanning electron microscopy (SEM)

The morphology of composite was examined using a Hitachi S-2 460N electron microscope at 25 kV. The compression molded sheet was cryogenically fractured in liquid nitrogen and the fractured surface was sputter-coated with gold–palladium alloy before viewing.

2.6. Differential scanning calorimetry (DSC)

DSC measurements were performed with a Perkin–Elmer DSC Pyris 1 instrument in a nitrogen atmosphere. Sample was held at 230 °C for 3 min to erase the thermo-mechanical prehistory, cooled to 50 °C at a rate of 10 °C/min, maintained at 50 °C for 3 min, and then reheated to 230 °C at a rate of 10 °C/min. The heat-flow rates as a function of temperature during cooling and the second heating runs were recorded. The degree of crystallinity was calculated with the heat of fusion, and took the $\Delta H_\mathrm{m} = 190$ J/g for 100% crystalline PP [22]. Three samples were measured, and the data in Tables 1 and 2 were an average value.

2.7. Thermo-oxidative property

Thermo-oxidative stability of composite was examined with a MAC Science TG-DTA 2000 instrument under a flow of air, heating from 50 °C to 500 °C at a rate of 10 °C/min.

2.8. Water absorption tests

Water absorption of composite was examined by immersion of sheet sample of 1 mm thickness in distilled water at room temperature for 1500 h, periodically measuring the increase in weight of the sample.

3. Results and discussion

Previous works demonstrated that MAPP had good compatibility for cellulose and PP composites [25,26], and the esterifications between MAPP and ball-milling activated cellulose were observed directly with FT-IR in the melt–mixed composites [27–29]. For the highly crystalline cellulose, the ester bonds between MAPP and cellulose are too few to be recorded by FT-IR [26–29], however, the interfacial chemical bonds were confirmed with XPS from the melt–mixed samples [27–29]. We assume that the compatibility is due to the chemical reactions between MAPP and cellulose. In this paper, the effect of molecular weight of PP on the structure and properties of the MAPP compatibilized PP/cellulose composites was studied, by comparing the tensile properties, dynamic mechanical properties, morphology, crystallization and melting behavior, thermo-oxidative property, and water absorption of composites.

3.1. Tensile properties

The tensile strength/strain behavior of PP is shown in Fig. 1. The tensile strength of PP1 and PP2 was higher than that of PP3. PP1 broke during the tensile testing, however, PP2 and PP3 exhibited necking behavior and did not break in the experimental conditions (till the elongation of 200%). The young's modulus decreased with the increase of molecular weight of PP.

Fig. 2 reveals the strength/strain behavior of PP3 and its composites. Due to the high cellulose content (30 wt.%) and high stiffness of the microfibers cellulose, the elongation dropped markedly for all

Table 1
DSC characterization of composites about MAPP content

PP/MAPP/CE (wt.%)	T_m (°C)	ΔH_m (J/g)	T_c (°C)	T_{onset} (°C)	$-\Delta H_c$ (J/g)	$T_m - T_c$ (°C)	X_c (%)
100/0/0	159.3[a] 163.3	83.4	111.1	117.0	90.5	48.2[b]	47.6
70/0/30	161.8	60.9	123.8	127.7	65.7	38.0	49.4
68/2/30	161.8	58.1	124.7	128.3	69.0	37.1	51.9
65/5/30	161.8	58.3	125.7	129.2	66.8	36.1	50.2
60/10/30	162.2	59.5	125.8	129.6	65.7	36.4	49.4
50/20/30	161.8	62.0	125.8	129.5	65.4	36.0	49.2

(PP pellets, $\overline{M_w} \approx 580,000$).

T_m: melting peak temperature.

T_c: crystallization peak temperature; T_{onset}: onset temperature of dynamical crystallization.

ΔH_m and ΔH_c denote the enthalpies of melting and crystallization.

X_c: crystallinity, based on total specimen weight and $-\Delta H_0 = 190$ J/g for 100% crystalline PP[22].

[a] Main peak.

[b] The value calculated from main melting peak temperature.

Table 2
DSC characterization of composites about PP molecular weight[*]

PP/MAPP/CE (wt.%)	T_m (°C)	ΔH_m (J/g)	T_c (°C)	T_{onset} (°C)	$-\Delta H_c$ (J/g)	$T_m - T_c$ (°C)	X_c (%)
$\overline{M_w}$ 250,000							
100/0/0	159.5[a] 164.6	86.9	113.2	118.7	96.2	46.3[b]	50.6
70/0/30	162.8	64.8	125.5	129.6	71.2	37.3	53.5
60/10/30	163.1	65.6	126.4	130.3	69.7	36.7	52.4
$\overline{M_w}$ 340,000							
100/0/0	159.8[a] 165.5	86.5	112.5	118.2	93.4	47.3[b]	49.2
70/0/30	162.8	67.3	125.1	129.3	68.9	37.7	51.8
60/10/30	163.2	63.4	126.8	130.7	68.9	36.4	51.8
$\overline{M_w}$ 580,000							
100/0/0	159.3[a] 163.3	83.4	111.1	117.0	90.5	48.2[b]	47.6
70/0/30	161.8	60.9	123.8	127.7	65.7	38.0	49.4
60/10/30	162.2	59.5	125.8	129.6	65.7	36.4	49.4

[*] The same notations as in Table 1.

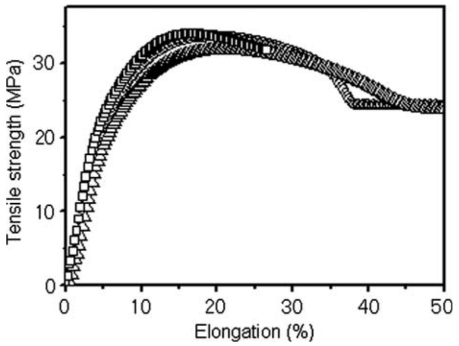

Fig. 1. Tensile strength/strain behavior of neat PP with different molecular weight: PP1 (□), PP2 (○), and PP3 (△).

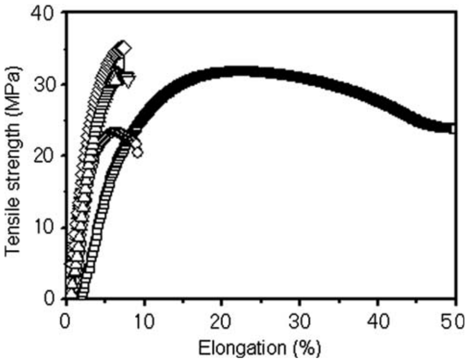

Fig. 2. Tensile strength/strain behavior of PP3 (□) and its composites with a MAPP content of: 0 (○), 2 (△), 5 (▽), 10 (◇), and 20 wt.% (◁).

composites. Without the compatibilization of MAPP, although the young's modulus of PP and cellulose binary composite got larger, the tensile strength of the composite decreased sharply by comparing with the neat PP. However, in the MAPP compatibilized composites, the tensile strength of composite was enhanced and much higher than that of non-compatibilized one, and the young's modulus was improved slightly, too. Same behavior was seen in Fig. 3, furthermore, the composite of which the matrix with high molecular weight exhibited larger elongation at break due to the higher inherent elongation.

The effects of molecular weight of PP as well as the content of the compatibilizer (MAPP) on the tensile properties of the composites are illustrated in Fig. 4. Compared with the non-compatibilized composites, even very little compatibilizer being used, the tensile strength of the composite was enhanced markedly. The tensile strength of the composite increased greatly with an increase in MAPP content, showing a maximum around a MAPP content of 10 wt.%. This suggests that there is a critical content of the compatibilizer at which MAPP exhibits the optimum interactions with both cellulose fibers and the PP matrix. Thus, beyond a MAPP content of 10 wt.%, the tensile strength of the ternary composite decreases due to the poor compatibility between MAPP and PP matrix [33], as well as to the lower tensile strength of MAPP [25]. The molecular weight of PP has very slight effect on tensile strength both in the non-compatibilized composites and in the compatibilized composites at lower MAPP content. In non-compatibilized composites, due to no interfacial interaction between cellulose and PP matrix, the tensile strength is mainly controlled by cellulose content [25]. In composites compatibilized with slight amount of MAPP, only a small part of cellulose were interacted with MAPP, thus the interfacial adhesion between cellulose and the matrix of the composites is not so strong. However, at high MAPP content, the composites with higher molecular weight of PP revealed larger tensile strength. It is proposed that in the MAPP compatibilized composite, the entanglements between PP chains and MAPP chains was enhanced and more physical cross-linking sites between PP chains and MAPP chains were generated at higher molecular weight of PP. Furthermore, the chain movements get difficult with the increase of molecular weight of PP matrix, therefore, the phase separation between PP and MAPP gets more difficult with the increases of molecular weight of PP matrix in the composite. The composite with higher molecular weight of PP yielded a little smaller young's modulus. Compared with the non-compatibilized composites, the young's modulus shown a very slight increase in MAPP compatibilized composites.

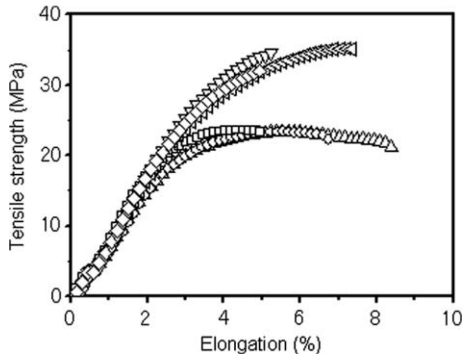

Fig. 3. Tensile strength/strain behavior: PP1/CE (70/30) (□), PP2/CE (70/30) (○), PP3/CE (70/30) (△), PP1/MAPP/CE (60/10/30) (◊), PP2/MAPP/CE (60/10/30) (▽), PP3/MAPP/CE (60/10/30) (◁).

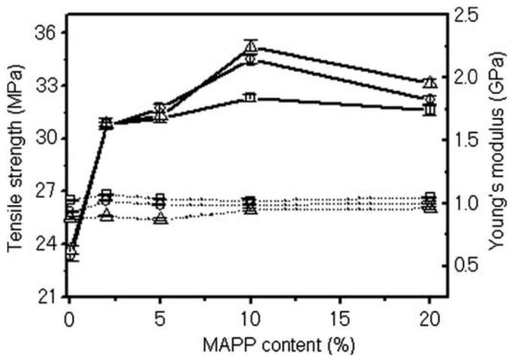

Fig. 4. Effect of molecular weight of PP and MAPP content on the tensile strength (—) and young's modulus (- - -) of composites: PP1 (□), PP2 (○), PP3 (△).

3.2. Dynamic mechanical properties

Dynamic mechanical spectra of neat PP are shown in Fig. 5. The peaks around −8 °C on tan δ

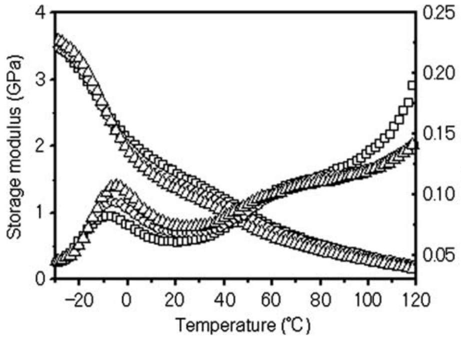

Fig. 5. DMA spectra of neat PP: PP1 (□), PP2 (○), PP3 (△).

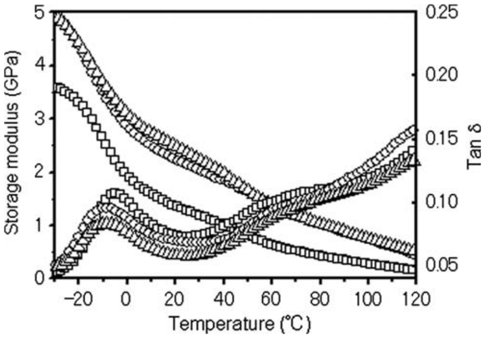

Fig. 6. DMA spectra of neat PP3 (□), PP3/CE = 70/30 (○), PP3/MAPP/CE = 60/10/30 (△).

curve corresponded to the glass transition of amorphous portions in PP, and the temperature of the peak maximum was assigned to glass temperature (T_g). As expected, PP with higher molecular weight exhibited higher T_g. The peak intensity and magnitude decreased with the decrease of molecular weight of PP. At room temperature, PP with higher molecular weight revealed higher tan δ values. The storage modulus (E') showed that E'-PP3 < E'-PP2 < E'-PP1 in room temperature, indicating that PP with higher molecular weight is more ready to deform.

Figs. 6 and 7 show that a decrease in tan δ values as well as an increase in E' in the PP and cellulose composites, compared with neat PP (see Fig. 5). This indicates that the composite had enhanced stiffness due to the adding of stiffer cellulose. The decrease in tan δ values as well as the peak intensity may be caused by the decreased amount of PP in the samples, and the lowered mobility of the PP chains due to the addition of the rigid cellulose phase. The compatibilization of MAPP enhanced these effects both in tan δ and in storage modulus, particularly in the PP3 composite (see Fig. 7). For PP1 and PP2 composites, the effect was slight. The results imply that MAPP has much more compatibilization effect on PP3/cellulose composite than on PP1/cellulose as well as PP2/cellulose composites.

3.3. Morphology of composites

A typical SEM micrograph of fractured surfaces of the composites is shown in Fig. 8. The micrograph of the PP and cellulose composite show

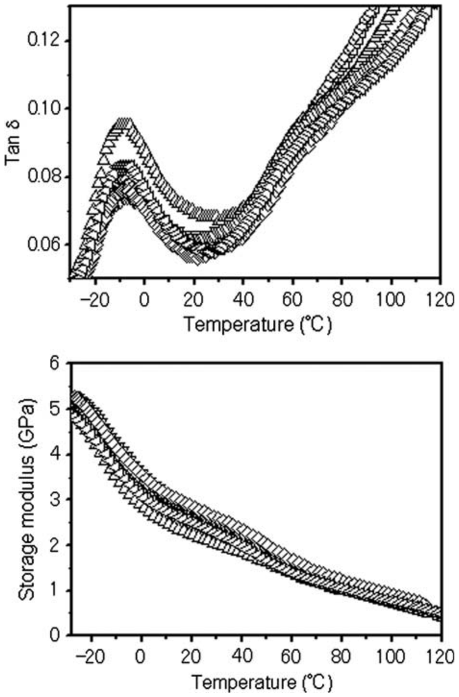

Fig. 7. Tan δ and storage modulus of composites: PP1/CE (70/30) (□), PP2/CE (70/30) (○), PP3/CE (70/30) (△), PP1/MAPP/CE (60/10/30) (▽), PP2/MAPP/CE (60/10/30) (◇), PP3/MAPP/CE (60/10/30) (◁).

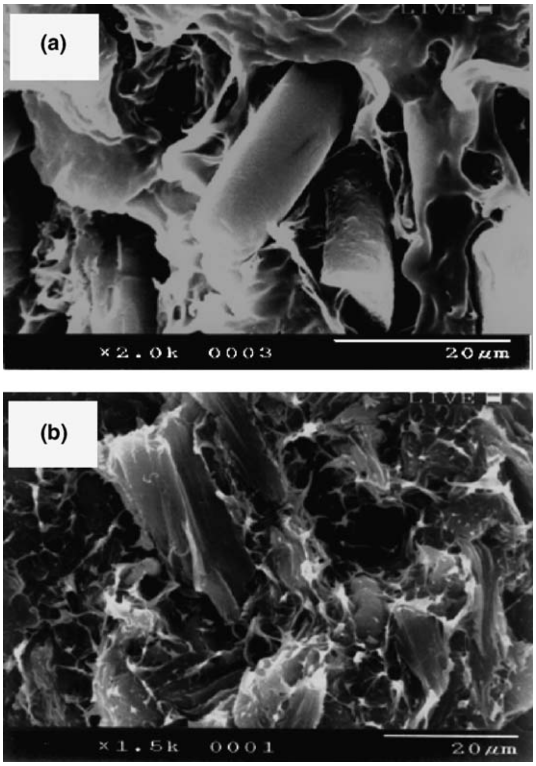

Fig. 8. SEM of cryogenically fractured surface of composites: PP3/CE = 70/30 (a), and PP3/MAPP/CE = 50/20/30 (b).

clearly that there were large gaps and voids between PP matrices and cellulose fibers, and the surfaces of cellulose fibers were smooth. This indicates that the interfacial adhesion between cellulose and the PP matrix is very poor, which is consistent with the poor tensile strength. The morphology of MAPP compatibilized composites revealed strong interfacial adhesion between MAPP and cellulose fibers: cellulose fibers were tightly connected with the matrix, and the fibers were broken and/or torn up. The improved interfacial adhesion in MAPP compatibilized composites is in agreement with the enhanced tensile strength.

3.4. Crystallization and melting behavior

Fig. 9 shows a typical melt behavior of some samples. The thermal parameters of melting and crystallization of the samples are summarized in Tables 1 and 2. The neat PP revealed a melting endothermic (T_m) at about 160 °C as well as a shoulder in higher temperature, while the composites exhibited a single melting peak, and PP3 in composites gave a little lower T_m by comparing with PP1 and PP2. Table 1 shows that the addition of cellulose caused a remarkable increase in crystallization temperature (T_c) and onset temperature (T_{onset}) of the PP3 matrix. The increasing values of T_c and T_{onset}, accordingly, the decreasing in supercooling ($T_m - T_c$), indicate that PP3 was more apt to crystallize in cellulose composite. The crystallinity of PP3 increased a little in composite. This is due to the cellulose fibers acting as an efficient nucleating agent for the crystallization of PP3, consequently increasing the crystallization rate of PP3 [8,34], and making the crystallization faster. The nucleating effect increased with the presence of MAPP. This is likely due to the stronger interaction between cellulose fibers and the PP matrix compatibilized by MAPP, which improved the nucleating activity of cellulose fibers for PP. We can also find that the improved nucleating effect induced by MAPP maintained constantly after MAPP reached to a critical amount. The same phenomena can be found in PP1 and PP2 composites. As expected, the crystallinity of neat PP decreased a little with the increase of its molecular weight (Table 2). From

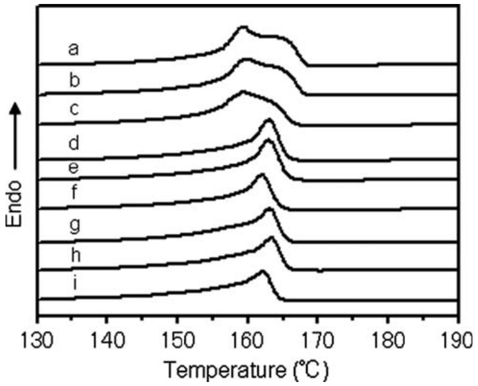

Fig. 9. DSC curves of PP1 (a), PP2 (b), PP3 (c), PP1/CE (70/30) (d), PP2/CE (70/30) (e), PP3/CE (70/30) (f), PP1/MAPP/CE (60/10/30) (g), PP2/MAPP/CE (60/10/30) (h), and PP3/MAPP/CE (60/10/30) (i).

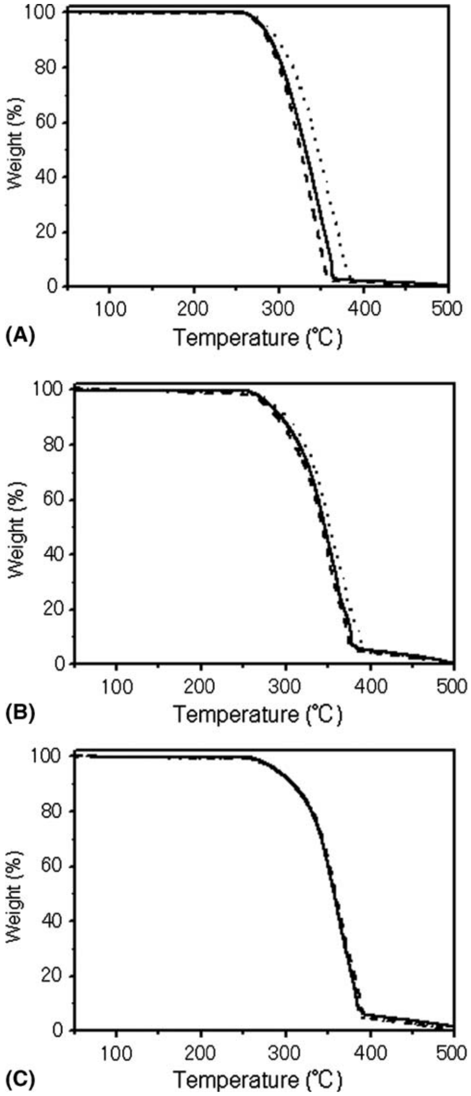

Fig. 10. TG curves of PP (A), PP/CE (70/30) (B), and PP/MAPP/CE (60/10/30) (C): PP1 (—), PP2 (–––), and PP3 (···).

Table 2, besides the same conclusions obtained above, we found that the decrease in supercooling caused by MAPP in PP3 composite was larger compared with PP1 and PP2 composites. This means that cellulose influences the PP3 matrix much more than the other two neat PPs.

3.5. Thermo-oxidative property

Fig. 10 shows TG curves of neat PP and some composites. The neat PP revealed the inherent thermo-oxidative stability as the order of PP2 < PP1 < PP3 (Fig. 10A). The same thermo-oxidative stability of PP and cellulose composites can be found in Fig. 10B, due to no compatibility between PP and cellulose. Unlike the results in Fig. 10A and B, Fig. 10C shows that in MAPP compatibilized systems, the composites yielded almost the same thermo-oxidative stability regardless of the PP used. Fig. 11 shows that the thermo-oxidative stability was in the order CF11 > PP3/MAPP/CE > PP3/CE > PP3 > MAPP. The thermo-oxidative stability of PP/cellulose composite was enhanced with the addition of MAPP. The reason may be attributed to the strong interfacial interaction between cellulose and the PP matrix caused by MAPP. And this stronger interfacial interaction also caused the improvement of thermo-oxidative stability of MAPP.

3.6. Water absorption

We measured the moisture content of neat cellulose as follows: CF11 sample was stored in a chamber at room temperature for 72 h, the atmosphere of the chamber was controlled to a relative humidity of 65%. And the measured moisture content of CF11 was 6.8–7.2 wt.%. Although we do not have the water holding capacity (water retention value) of CF11, however, it is reasonable to assume that the saturated water holding capacity of CF11 is higher than the measured moisture content, i.e., 7 wt.%.

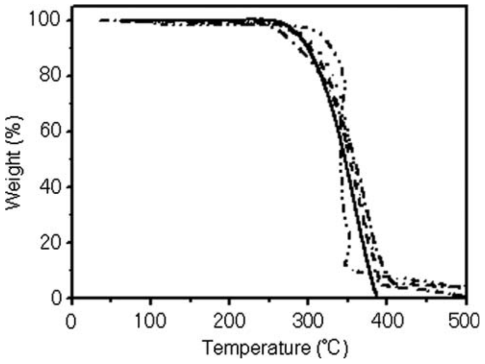

Fig. 11. TG curves of: PP3 (—), PP3/CE = 70/30 (– – –), PP3/MAPP/CE = 60/10/30 (· · ·), MAPP (–·–·–), and CF11 (–··–··–).

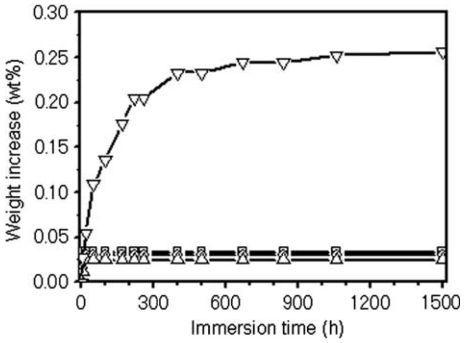

Fig. 12. Water absorption of neat MAPP (▽), PP1 (□), PP2 (○), and PP3 (△).

The water absorption behavior of neat matrixes and composites are shown in Figs. 12 and 13, respectively, which are given as a percent weight increase as a function of immersion time. Fig. 12 shows that the water absorption of PPs was quite small, less than 0.03 wt.% even after 1500 h, although MAPP showed a little higher water absorption than PP, it was still quite small, about 0.25 wt.% after immersed in water for 1500 h. Compared with cellulose, the absorbed water of both MAPP and PP in the composites is negligible. Fig. 13 shows that the water absorption of composites increased with time, quickly at the beginning several hundreds hours, then increased slowly, however, the composites still did not reach to saturation absorption even after immersion of 1500 h. The

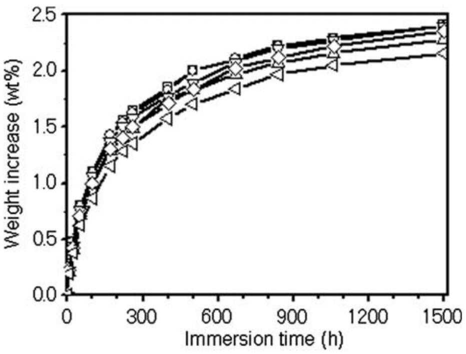

Fig. 13. Water absorption of composites: PP1/CE (70/30) (□), PP2/CE (70/30) (○), PP3/CE (70/30) (△), PP1/MAPP/CE (60/10/30) (▽), PP2/MAPP/CE (60/10/30) (◇), PP3/MAPP/CE (60/10/30) (◁).

sorption is a diffusion control process, which depends on the thickness of sample, Fig. 13 revealed 1500 h immersion was not enough time for saturation composites. The composite derived from lower molecular weight showed higher water absorption, and the MAPP compatibilized composites yielded lower water absorption than the corresponding non-compatibilized one. After immersed 20 h, the water absorption was 0.481, 0.480, and 0.410 wt.% for composite from PP1, PP2 and PP3, while 0.443, 0.421, and 0.378 wt.% for the corresponding MAPP compatibilized composite. After immersed for 1500 h, the water absorption increased to 2.406, 2.401, and 2.272 wt.% for composite from PP1, PP2 and PP3, while 2.402, 2.353, and 2.157 wt.% for the corresponding MAPP compatibilized composite. Part of water may entrapped and accumulated in the voids at the interface between fibers and matrix, due to poor adhesion between them, and this part of water in composite also need be considered besides the cellulose absorbed (the matrix absorbed water is minor, as described above). Obviously, the interfacial property has an effect on the water absorption of the composites. It is expected that the water absorption will be lessened if there is good interfacial adhesion between cellulose and the matrix. The PP and cellulose composites exhibited higher rates of water absorption as well as higher water absorption than those of MAPP compatibilized composites. Due to the strong adhesion between cellulose fibers and the matrix in MAPP compatibilized composites, the cellulose fibers were encapsulated within the matrix, which prevented water molecules from penetrating into the cellulose, meanwhile, the voids at the interface were lessened, too. The composites with higher molecular weight of PP exhibited lower water absorption compared with the lower molecular weight PP, and the drop in water absorption caused by MAPP was larger in the composites with higher molecular weight, for example, by using of MAPP, the water absorption dropped from 2.406 wt.% to 2.402 wt.% for PP1 composite, while 2.272 wt.% to 2.157 wt.% for PP3 composite, after 1500 h immersion in water. This also means that the interfacial interaction is greater in the composite with higher molecular weight of PP.

4. Conclusions

MAPP may be used as a compatibilizer in preparing PP and cellulosic composites. The properties, especially tensile strength, of the composite were influenced markedly by the MAPP content and the PP molecular weight. For all the PPs used and at a cellulose content of 30 wt.%, the maximum tensile strength of the composite may be obtained at a MAPP content about 10 wt.%. PP with higher molecular weight will be more readily affected by both MAPP and cellulose than the other two PPs. Compared with PP of lower molecular weight, PP with higher molecular weight shows a stronger interfacial interaction with cellulose in MAPP compatibilized composite, exhibits stronger tensile strength, and yields much more influence on other properties of the composites.

References

[1] Bledzki AK, Gassan J. Composites reinforced with cellulose based fibres. Prog Polym Sci 1999;24:221–74.
[2] Gassan J, Bledzki AK. The influence of fiber-surface treatment on the mechanical properties of jute–polypropylene composites. Composites Part A 1997;28A:1001–5.
[3] Bataille P, Ricard L, Sapieha S. Effects of cellulose fibers in polypropylene composites. Polym Compos 1989;10:103–8.
[4] Felix JM, Gatenholm P. The nature of adhesion in composites of modified cellulose fibers and polypropylene. J Appl Polym Sci 1991;42:609–20.
[5] Felix JM, Gatenholm P, Schreiber HP. Controlled interactions in cellulose–polymer composites. 1. Effect on mechanical properties. Polym Compos 1993;14:449–57.
[6] Sain MM, Kokta BV. Toughened thermoplastic composite. I. Cross-linkable phenol formaldehyde and epoxy resins-coated cellulosic-filled polypropylene composites. J Appl Polym Sci 1993;48:2181–96.
[7] Collier JR, Lu M, Fahrurrozi M, Collier BJ. Cellulosic reinforcement in reactive composite systems. J Appl Polym Sci 1996;61:1423–30.
[8] Amash A, Zugenmaier P. Study on cellulose and xylan filled polypropylene composites. Polym Bull 1998;40:251–8.
[9] George J, Sreekala MS, Thomas S. A review on interface modification and characterization of natural fiber reinforced plastic composites. Polym Eng Sci 2001;41:1471–85.
[10] Harris B, Beaumont PWR, Moncunill de Ferran E. Strength and fracture toughness of carbon fibre polyester composites. J Mater Sci 1971;6:238–51.
[11] Sanadi AR, Subramanion RV, Manoranjan VS. The interphasial regions in interlayer fiber composites. Polym Compos 1991;12:377–83.
[12] Dong S, Sapieha S, Schreiber HP. Mechanical properties of corona-modified cellulose/polyethylene composites. Polym Eng Sci 1993;33:343–6.
[13] Belgacem MN, Bataille P, Sapieha S. Effect of corona modification on the mechanical properties of polypropylene/cellulose composites. J Appl Polym Sci 1994;53:379–85.
[14] Bledzki AK, Reihname S, Gassan J. Properties and modification methods for vegetable fibers for natural fiber composites. J Appl Polym Sci 1996;59:1329–36.
[15] Coutinho FMB, Costa THS, Carvalho DL. Polypropylene–wood fiber composites: effect of treatment and mixing

conditions on mechanical properties. J Appl Polym Sci 1997; 65:1227–35.

[16] Raj RG, Kokta BV, Maldas D, Daneault C. Use of wood fibers in thermoplastics. VII. The effect of coupling agents in polyethylene–wood fiber composites. J Apply Polym Sci 1989;37:1089–103.

[17] Maldas D, Kokta BV. Influence of maleic anhydride as a coupling agent on the performance of wood fiber–polystyrene composites. Polym Eng Sci 1991;31:1351–7.

[18] Chiang W, Yang W. Polypropylene composites. III. Chemical modification of the interphase and its influence on the properties of PP/mica composites. Polym Eng Sci 1994;34: 485–92.

[19] Kazayawoko M, Balatinecz JJ, Woodhams RT. Diffuse reflectance Fourier transform infrared spectra of wood fibers treated with maleated polypropylenes. J Appl Polym Sci 1997;66:1163–73.

[20] Oksman K, Clemons C. Mechanical properties and morphology of impact modified polypropylene–wood flour composites. J Appl Polym Sci 1998;67:1503–13.

[21] Anglès MN, Salvadó J, Dufresne A. Steam-exploded residual softwood-filled polypropylene composites. J Appl Polym Sci 1999;74:1962–77.

[22] Amash A, Zugenmaier P. Morphology and properties of isotropic and oriented samples of cellulose fibre–polypropylene composites. Polymer 2000;41:1589–96.

[23] Qiu WL, Zhang FR, Endo T, Hirotsu T. Isocyanate as a compatibilizing agent on the properties of highly crystalline cellulose/polypropylene composites. J Mater Sci 2005;40: 3607–14.

[24] Qiu WL, Mai KC, Zeng HM. Effect of macromolecular coupling agent on the property of PP/GF composites. J Appl Polym Sci 1999;71:1537–42.

[25] Qiu WL, Zhang FR, Endo T, Hirotsu T. Preparation and characteristics of composites of high-crystalline cellulose with polypropylene: effects of maleated polypropylene and cellulose content. J Appl Polym Sci 2003;87:337–45.

[26] Qiu WL, Zhang FR, Endo T, Hirotsu T. Effect of maleated polypropylene on the performance of cellulose/polypropylene composite. Polym Compos 2005;26:448–53.

[27] Qiu WL, Zhang FR, Endo T, Hirotsu T. Milling-induced esterification between cellulose and maleated polypropylene. J Appl Polym Sci 2004;91:1703–9.

[28] Qiu WL, Endo T, Hirotsu T. Interfacial interactions of a novel mechanochemical composite of cellulose with maleated polypropylene. J Appl Polym Sci 2004;94:1326–35.

[29] Qiu WL, Endo T, Hirotsu T. Effect of degree of interfacial chemical bonds on tensile strength of cellulose/polypropylene composites. Trans Mater Res Soc Jpn 2004;29:2475–8.

[30] Qiu WL, Endo T, Hirotsu T. Interfacial interaction, morphology and tensile properties of a composite of highly crystalline cellulose and maleated polypropylene. J Appl Polym Sci, submitted for publication.

[31] Endo T, Kitagawa R, Hirotsu H, Hosokawa J. Fine-powdering of fibrous cellulose by mechanical milling. Kobunshi Ronbunshu 1999;56:166–73 [in Japanese].

[32] Japan Standard Society. JIS K7113-1995 Testing method for tensile properties of plastics. Japan Standard Society: Tokyo; 1995.

[33] Li SC, Järvelä PK, Järvelä PA. Melt rheological properties of polypropylene-maleated polypropylene blends. II. Dynamic viscolelastic properties. J Appl Polym Sci 1999; 71:1649–56.

[34] Mi Y, Chen X, Guo Q. Bamboo fiber-reinforced polypropylene composites: crystallization and interfacial morphology. J Appl Polym Sci 1997;64:1267–73.