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ORIGINAL PAPER

Development of continuous process enabling nanofibrillation of pulp and melt compounding

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Abstract Microfibrillated cellulose (MFC), a mechanically fibrillated pulp mostly consisting of nanofibrils, is a very attractive material because of its high elastic modulus and strength. Although much research has been done on composites of MFC and polypropylene (PP), it has been difficult to produce such composites at an industrial level because of the difficulties in using MFC in such composites are not only connected to the polarity (that can be improved with compatibilizers), but also with the challenge to make a homogeneous blend of the components, and also the low temperature stability of cellulose that could cause problems during processing. We developed a new processing method which enables continuous microfibrillation of pulp and its melt compounding with PP. Never-dried kraft pulp and powdered PP were used as raw materials to obtain MFC by kneading via a twin-screw extruder. Scanning electron microscopy showed nano to submicron wide fibers entangled in the powdered PP. MFC did not aggregate during the melt compounding process, during which the water content was evaporated. Maleic anhydride polypropylene (MAPP) was used as a compatibilizer to reinforce interfacial adhesion between the polar hydroxyl groups of MFC and non-polar PP. We investigated the effect of MAPP content on the mechanical properties of the composite, which were drastically improved by MAPP addition. Needle-leaf unbleached kraft pulp (NUKP)-derived MFC composites had better mechanical properties than needle-leaf bleached kraft pulp (NBKP)-derived MFC composites. Injection molded NUKP-derived MFC composites had good mechanical and thermal properties. The tensile modulus of 50 wt% MFC composite was two times, and the tensile strength 1.5 times higher than that of neat PP. The heat distortion temperature of 50 wt% MFC content composite under 1.82 MPa flexural load was increased by 53 °C, from 69 to 122 °C. This newly developed continuous process using powder resin has the potential for application at an industrial level.

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Introduction

Fillers have been used in polymer systems to improve mechanical properties. Natural fiber-reinforced composites in particular have been developed over the last few decades because of the renewable and environmentally friendly nature of natural fibers.

Cellulose microfibrils, which being 3–4 nm wide could be considered nanofibrils, constitute the main structural elements of wood cell walls. They show high elastic modulus and strength along the longitudinal direction (Sakurada et al. 1962; Page and El-Hosseiny 1983), and their coefficient of thermal expansion (CTE) is as low as that of quartz (Nishino et al. 2004). As a result, cellulose microfibrils, 10–20 nm wide bundles of cellulose microfibrils, and MFC, a mechanically fibrillated pulp consisting of fibrils and fiber fragments with various dimensions in which fibrils in nanoscale is a major fraction (Chinga-Carrasco 2011), have been extracted from plant fibers and their reinforcing effects studied (Nakagaito and Yano 2004; Nakagaito and Yano 2005; Eichhorn et al. 2010; Siró and Plackett 2010; Klemm et al. 2011).

Many studies of cellulose morphologies such as those of MFC and cellulose nano whisker-reinforced thermoplastic resin have been done in the last few decades. Samir et al. (2004) studied composites based on sugar beet cellulose microfibrils and poly-(styrene-*co*-butyl acrylate) latex, and found that the resulting entanglement of cellulose microfibrils in the latex drastically affected the mechanical behavior of the composites. Zimmermann et al. (2004) reported that a MFC-reinforced hydroxypropyl cellulose (HPC) prepared by film casting had three times higher tensile modulus and five times higher tensile strength than neat HPC (fiber content 20 wt%). Chakraborty et al. (2006) investigated the reinforcing potential of microfibrils and microcrystalline cellulose (MCC, cellulose nanowhisker aggregates) in polyvinyl alcohol (PVA), and showed that the tensile properties of composites with microfibrils were better than those with MCC because the minimum aspect ratio of a fiber is more critical than its crystallinity. In addition, many studies have been done on the compounding of cellulose nanoelements and water insoluble thermoplastic resins such as polylactic acid (PLA). Many researchers have used PLA dissolved in organic solvent to produce well dispersed cellulose nanoelement composites (Iwatake et al. 2008; Suryanegara et al. 2009, 2010, 2011; Jonoobi et al. 2010).

Iwatake et al. (2008) investigated the effects of pulp microfibrillation on the mechanical properties of PLA composites using three types of fiber; NBKP (needle-leaf bleached kraft pulp), refiner-treated NBKP, and MFC (microfibrillated cellulose by high-pressure homogenizing process). Fibrillation increased in the order NBKP < refiner-treated NBKP < MFC. Tensile modulus and strength increased with degree of fibrillation. Moreover, Mathew et al. (2005) showed that compounding of MCC and PLA increases the Young's modulus of the resulting PLA composite but reduces the yield strain, decreasing its tensile strength compared with that of neat PLA.

PP has been used in many applications including automobiles, buildings, and many other products, so a large number of studies on environmentally friendly natural fiber and cellulose nanofiber reinforced PP have been carried out. One of the major problems in the fabrication of cellulose nanofiber-reinforced PP is improvement of interfacial adhesion between hydrophilic cellulose and apolar PP, and many investigations have been done to improve their compatibility (Qiu et al. 2004, 2005, 2006; Lee et al. 2009; Osman et al. 2010; Haydaruzzaman et al. 2010; Sykacek et al. 2011; Nakatani et al. 2011) to achieve uniform dispersion of cellulose nanoelements in the non-polar PP matrix.

Ljungberg et al. made PP composite films by mixing a toluene suspension of cellulose whiskers with solubilized isotactic polypropylene (iPP). The cellulose whiskers, which had no surface modification, aggregated in the toluene suspension and decreased the tensile strength of the composites compared with that of neat iPP. However, the tensile strength of composites made with surfactant-modified cellulose whiskers increased significantly compared with that of iPP, and the modified whiskers were well dispersed in the iPP matrix. Nevertheless this method was considered too complicated and costly to be applied as an industrial process, so a more practical method was desired.

In this study, continuous nanofibrillation and melt compounding of PP was developed by processing water slurry containing refiner treated kraft pulp and

powdered PP resin. Scanning electron micrographs showed that the pulp had been microfibrillated to nanometer to submicron widths and was well dispersed in the PP matrix. The tensile modulus of the obtained 50 wt% MFC composite was two times higher than that of neat PP, and the tensile strength was 1.5 times higher. In addition, the heat distortion temperature (HDT) (1.82 MPa) of the 50 wt% MFC composite was increased by 53 °C, from 69 °C to 122 °C.

Experimental

Materials

Kraft pulps (NUKP(Needle-leaf unbleached kraft pulp) and NBKP(Needle-leaf bleached kraft pulp)), were supplied by Oji Holdings Corporation (Tokyo, Japan) as never dried kraft pulp with solid content of 20–25 wt% pulp. The pulps were fibrillated until Canadian Standard Freeness (CSF) as per TAPPI standard T227om-09 was not more than 10 mL. The degree of NUKP polymerization was 3.1×10^3, while that of NBKP was 2.2×10^3, calculated as the value of relative viscosities as per TAPPI standard T230om-99. The lignin contents of NUKP and NBKP were 5, and 0 wt%, respectively, as per the Klason lignin method, TAPPI standard T230om-02. Isotactic polypropylene (PP), NOVATEC^{TM}PP MA4AHB, was provided by Japan Polypropylene Corporation (Tokyo, Japan), and had an average molecular weight of 200,000–230,000. The average molecular weight was characterized by gel permeation chromatography in 1,2-dichlorobenzene using polystyrene standards. We assigned Seishin Enterprise Co., Ltd. to make powdered PP. They dissolved PP in organic solvent and spray dried to obtain powdered PP. The particle size varied from a few to several tens of μm. Maleic anhydride grafted polypropylene (TOYOTAC, PMA-H1000P, weight average molecular weight of 72,000 according to GPC) was purchased from TOYOBO CO., LTD (Osaka, Japan). According to the supplier, the amount of grafted maleic anhydride was 4 wt%.

Preparation of PP composite

A schematic illustration of the composite preparation method is shown in Fig. 1. Refiner-treated never dried kraft pulp was mixed with powdered PP and MAPP using an agitator (Ken Mix Aicoh PRO, Aikosya, Saitama, Japan), and the wet mixture was kneaded by a twin-screw extruder (Technovel Corp., TZW15-TW, Osaka, Japan). The temperature of the barrels was set to 0 °C, the screw speed 400 rpm, and the compound output was approximately 400 g/h.

In Experiment 1, the kneaded MFC/PP mixtures were melt extruded by a twin rotary roller mixer (Labo Plastomill Model 30C 150, Toyo Seiki Seisaku-sho, Tokyo, Japan). Compounding was carried out for 10 min at a rotary speed of 40 rpm and 180 °C, and water was removed at this stage. The composite was crushed into small pieces and compressed at 180 °C and 2 MPa for 10 min.

To verify if melt extrusion process, which is used at industrial level, could be used to our compound, we did continuous melt extrusion at laboratory-scale. The fibrillated MFC/PP mixtures were melt extruded by a twin-screw extruder in Experiment 2. The diameter of the extruder screws was 15 mm, and the L/D was 45. The temperature of the barrels was set at between 110 and 180 °C, the screw speed at 200 rpm, and the compound output was approximately 300 g/h. Water and volatiles were vented out from two barrels (Fig. 2). Standard test pieces were prepared from the obtained extruded compounds using an injection molding machine (TM30, Toyo Machinery & Metal Co., Hyogo, Japan) operating at an injection temperature of 200 °C, pressure of 100 MPa, speed of 80 mm/s, and mold temperature of 40 °C.

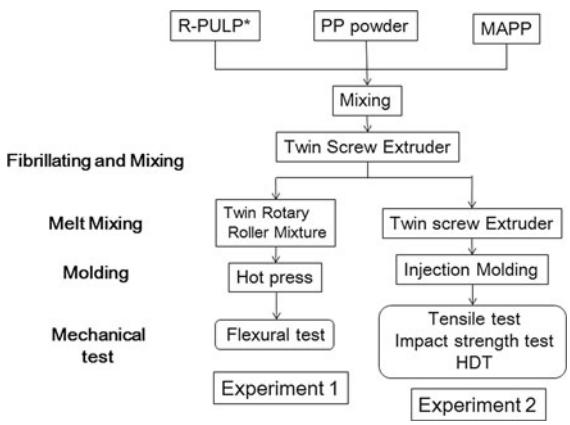

Fig. 1 Composite preparation procedure. *Asterisk* Refiner-treated pulp

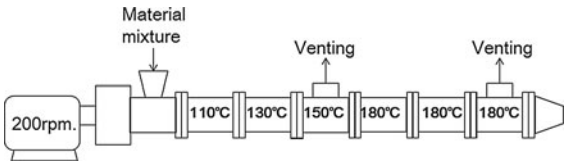

Fig. 2 Processing parameters for melt compounding

Testing

Flexural test

Flexural strength and modulus of the composites were measured by three-point flexural tests using a universal testing machine (model 3365; Instron Corp., Canton, MA, USA). Specimens were about 1 mm thick, 50 mm long, and 10 mm wide. Test span was set to 30 mm and crosshead speed to 5 mm/min. Five replicates were tested for each sample and the average data and standard deviations were calculated.

Tensile test

The tensile strength and modulus of the composites were measured according to ISO527/1-1993 using a universal testing machine (model 3365; Instron Corp., Canton, MA, USA). The test specimens were type 1A dumb-bell-shaped with a width of 10 mm (narrow portion) and thickness of 4 mm. Three replicates were tested for each sample and average data were calculated.

Notched Izod impact strength test

Notched Izod impact strength of the composites was determined using a No. 258 Universal Impact Tester (Yasuda Seiki Seisakusho, Hyogo, Japan) according to ISO180-1993. The dimensions of the specimens were approximately 80 × 10 × 4 mm, and a notch (0.25 mm radius, 2 mm depth) was made in each specimen. Three replicates were tested for each sample and the average data were calculated.

Heat distortion temperature (HDT)

HDT was measured under 1.82 MPa flexural load using a No. 533 Heat Distortion Tester (Toyo Seiki Seisakusho, Tokyo, Japan) according to JIS K7191-1. Samples with dimensions of 80 × 10 × 4 mm were tested in the edgewise position. Average data were calculated for three replicates.

Scanning electron microscopy (SEM)

The morphology of the fibers was observed using a field emission scanning electron microscope (FE-SEM, JSM-6700F; JEOL, Tokyo, Japan). The samples were coated with Pt (ca. 2 nm) before imaging using an auto-fine-coater (JFC-1600; JEOL, Tokyo, Japan).

Results and discussion

Process of making MFC mixed PP compounds

Morphological changes observed for refiner-treated kraft pulp during kneading by twin-screw extruder and melt compounding by twin rotary roller mixer in Experiment 1 (Fig. 1) are shown in Fig. 3. The contents of NUKP, powdered PP, and MAPP were 50 wt%, 44.4, and 5.6 %, respectively. SEM images of NUKP and refiner-treated NUKP are shown in Fig. 3a, b, respectively, and reveal that the refiner-treated NUKP was partially fibrillated. Images of powdered PP and MAPP are shown in Fig. 3c, d. Their particle sizes varied from a few to several tens of μm, and were considerably smaller than commercially-used PP pellets. Fibrillation process load was reduced by using refiner-treated NUKP and powdered PP, and the partially fibrillated NUKP was entwined with the powdered PP and MAPP. Figure 3e shows the mixture of refiner-treated NUKP, powdered PP, and MAPP after kneading by the twin-screw extruder. We used never-dried kraft pulp to avoid degradation of the crystallinity and degree of polymerization of the cellulose (Ljungberg et al. 2006). Figure 3(e) shows that NUKP was fibrillated into nanometer to sub-micron wide sizes, and were well entwined around the powdered PP and MAPP particles. Thus, fibrillation was promoted using refiner-treated never-dried kraft pulp as the raw material (Iwamoto et al. 2007; Abe et al. 2007) which mixed effectively with the powdered PP and MAPP.

To investigate the morphology of MFC inside the composite, the composite was immersed in boiling p-xylene after melt compounding to remove the resin. Water is evaporated during the melt compounding process, and it is known that cellulose nanofibers irreversibly bind to each other by hydrogen bonding during this drying process (Abe et al. 2007; Iwamoto et al. 2008; Hult et al. 2001; Duchesne et al. 2001). However, Fig. 3f shows that the MFC did not

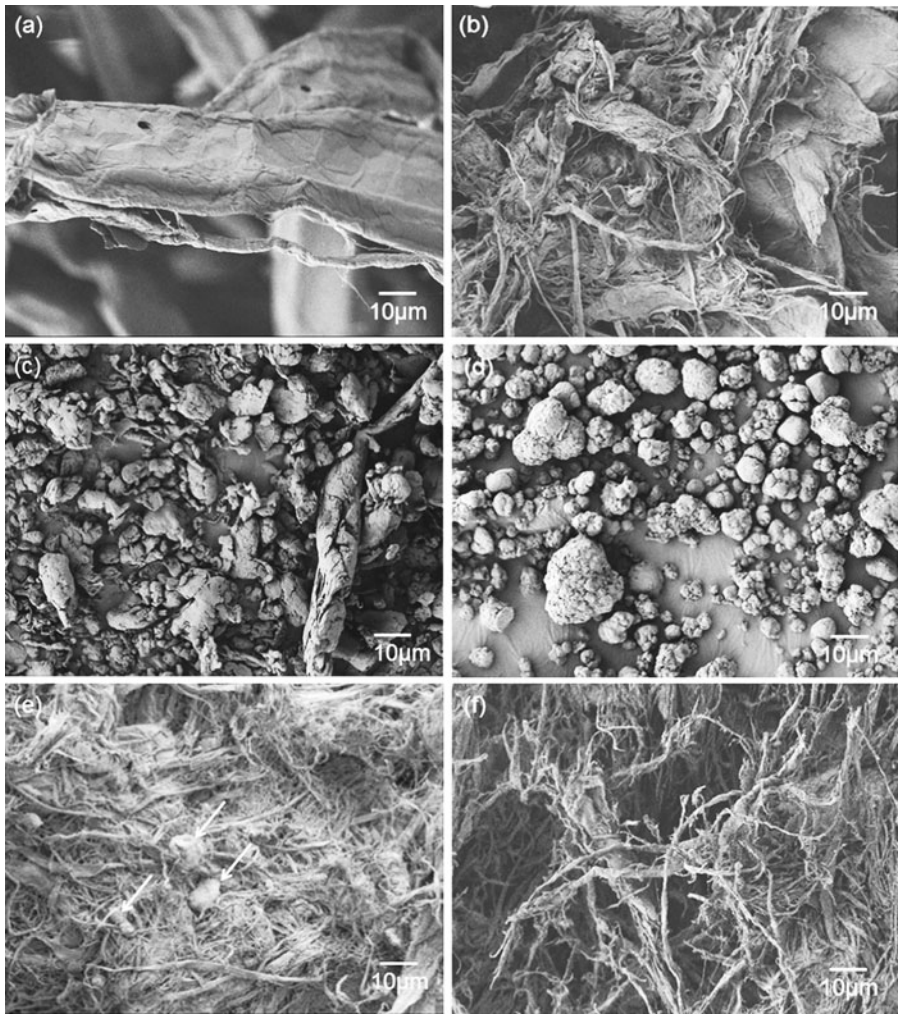

Fig. 3 Filler morphologies during composite processing: **a** NUKP, **b** refiner-treated NUKP, **c** powdered PP, **d** MAPP, **e** mixture of refiner-treated NUKP, powdered PP, and MAPP after kneading (*arrows* indicate powdered PP), **f** after matrix removal by p-xylene washing. *Scale bar*: 10 μm

aggregate after melt extrusion. It was assumed that powdered PP and MAPP had plasticized, changed shape, and melted to fill the space between the fibers during melt compounding, thereby preventing MFC from aggregating. Thus, the present process makes it possible to continuously prepare MFC-reinforced PP using water containing refiner-treated kraft pulp and powdered PP.

Effect of MAPP on the mechanical properties of MFC composites

As shown in section "Process of making MFC mixed PP compounds", the kraft pulp was microfibrillated and the resulting MFC composites had large interface between cellulose and PP. We then investigated the effect of the coupling agent, MAPP, by following the scheme of Experiment 1 (Fig. 1). MAPP strongly interacts with cellulose OH groups through covalent bonds (ester bonds) and/or acid/base interactions (Qiu et al. 2004; Felix and Gatenholm 1991), and thus the amount of coupling agent was expected to be a very important factor in the fabrication of composites of cellulose nanofibers and non-polar PP. The effect of MAPP content on the MFC composites is shown in Table 1. The flexural modulus and strength of NUKP-derived composites were obviously higher than those of NBKP-derived composites, as will be discussed later.

Table 1 Flexural properties of Pulp/PP/MAPP composites as functions of MAPP content

Sample	MAPP (%)	Flexural modulus (GPa)	Strength (MPa)	Strain at break (%)
Neat PP	0	2.3 (0.0)	68.6 (1.0)	>100
NUKP/PP-p/MAPP	0	3.5 (0.2)	63.0 (1.5)	5.4 (0.5)
	2	3.7 (0.1)	88.8 (2.7)	4.7 (0.4)
	3	3.5 (0.2)	90.5 (2.8)	5.4 (0.3)
	4	3.7 (0.3)	96.8 (2.3)	5.1 (0.5)
	5	3.5 (0.1)	90.9 (3.5)	5.2 (0.2)
	6	3.7 (0.2)	88.9 (1.0)	5.5 (0.6)
NBKP/PP-p/MAPP	0	3.3 (0.1)	63.7 (0.9)	5.7 (0.3)
	2	3.2 (0.0)	78.4 (2.8)	4.8 (0.5)
	3	3.4 (0.2)	80.2 (2.3)	4.9 (0.2)
	4	3.1 (0.2)	80.3 (2.7)	5.5 (0.1)
	5	3.2 (0.1)	83.8 (2.2)	5.2 (0.1)
	6	3.4 (0.1)	81.0 (2.2)	4.6 (0.3)

Values in parentheses indicate standard deviations

The flexural modulus of composites containing 30 wt% NUKP-derived MFC increased from 2.3 to 3.5 GPa, compared with that of neat PP, while the flexural strength decreased from 68.6 to 63.0 MPa. However, flexural strength increased considerably to 88.8–96.8 MPa when MAPP was added to the composite even though the flexural modulus and strain at break were almost the same. Furthermore, flexural strength increased with the amount of MAPP added until 4 wt%, after which it decreased gradually with further addition. In the case of composite containing 30 wt% NBKP-derived MFC, the flexural modulus increased from 2.3 to 3.3 GPa compared with that of neat PP, while flexural strength decreased from 68.6 to 63.7 MPa, in the same manner as that observed for the composite containing 30 wt% NUKP-derived MFC. Flexural strength appeared to increase with amount of MAPP added up to 5 wt%, but the differences were not obvious. These results indicate that when too much MAPP, which has a lower MW than PP, is added to the composite, the strength of the composite decreases because MAPP is not long enough to make entanglements with the polymer matrix. However, our results also show that the interface between cellulose and PP was strengthened by adding an adequate amount of MAPP in the composite because of the strong covalent bonds (ester bonds) and/or acid–base interactions between MAPP and the cellulose OH-groups.

Effects of MFC content on the mechanical properties of MFC composites

Scanning electron micrographs displayed in Fig. 3f in section "Process of making MFC mixed PP compounds" show that there was a large degree of MFC entanglement in the composite after melt compounding. This result indicated that not only chemical interactions occurred between cellulose and PP through MAPP, but also entanglement of MFC and hydrogen bonding between MFC surfaces, in agreement with the observed effect of MFC content on composite mechanical properties.

Subsequently, we investigated the relationship between MFC content and flexural properties of PP compounds prepared in accordance with Experiment 1.

The results of flexural tests are shown in Table 2 as a function of MFC content. The MAPP/MFC weight ratio was around 0.11. The relationships between MFC content and flexural modulus and flexural strength are shown in Fig. 4. It was observed that flexural modulus increased linearly with MFC content. There was no significant difference in flexural modulus between NUKP-derived MFC and NBKP-derived MFC, and the flexural modulus at 50 wt% NUKP- and NBKP-derived MFC content was almost twice that of neat PP. Flexural strength also increased with MFC content, but was higher for composites containing NUKP-derived MFC than those containing NBKP-derived MFC. This was probably because NUKP is not subjected to bleaching treatment, so it was not damaged compared to NBKP (the degree of polymerization of NUKP and NBKP are 3.1×10^3 and 2.2×10^3, respectively). In addition, NUKP contains 5 % lignin, which is basically of non-polar character. Moreover, as shown in the strain stress curves displayed in Fig. 5, the flexural energy absorption of NUKP-derived MFC composite was larger than that of NBKP because of the larger degree of polymerization and non-polar lignin. As a result, NUKP-derived MFC composites had higher flexural strength than NBKP-derived MFC composites.

Overall, these results indicate that the method of kraft pulp preparation affects the mechanical properties of the resulting cellulose nanofiber-reinforced composite.

Table 2 Flexural properties of MFC/PP/MAPP composites

MFC	MFC/PP/MAPP (wt%)	Flexural modulus (GPa)	Strength (MPa)	Strain at break (%)
Neat PP	0/100/0	2.3 (0.0)	68.6 (1.0)	>100
NUKP derived fiber	10/88.9/1.1	2.8 (0.1)	71.4 (0.9)	>100
	20/77.8/2.2	3.0 (0.2)	82.0 (1.2)	6.2 (0.7)
	30/66.7/3.3	3.6 (0.1)	87.7 (2.1)	5.7 (0.2)
	40/55.5/4.5	3.8 (0.2)	95.3 (3.8)	4.9 (0.2)
	50/44.4/5.6	4.3 (0.2)	97.6 (3.2)	3.8 (0.1)
NBKP derived fiber	10/88.9/1.1	2.4 (0.1)	76.8 (1.6)	5.7 (0.5)
	20/77.8/2.2	2.9 (0.2)	77.5 (1.8)	6.3 (1.2)
	30/66.7/3.3	3.4 (0.2)	80.2 (2.3)	4.9 (0.2)
	40/55.5/4.5	3.8 (0.1)	83.8 (2.0)	4.4 (0.2)
	50/44.4/5.6	4.2 (0.2)	88.8 (2.7)	4.0 (0.3)

Values in parentheses indicate standard deviations

Mechanical and thermal properties of injection molded MFC-reinforced PP composites

To verify the present continuous microfibrillation of pulp and PP melt compounding process for industrial applications, we injection-molded MFC composites following the scheme of Experiment 2 in Fig. 1. The tensile properties of the obtained injection molded samples are shown in Table 3. NUKP was used as the MFC raw material. Although the melt viscosity of the composite increased with MFC content, injection molding of samples with up to 60 wt% MFC content was possible.

The mechanical properties of MFC composites made by injection molding were significantly increased as effectively as samples made in Experiment 1. The flexural modulus of 30 wt% NUKP-derived MFC composite made by compression molding in Experiment 1 was 1.6 times higher than that of neat PP, while the tensile modulus of the injection-molded 30 wt% NUKP-derived MFC composite in Experiment 2, was 1.4 times higher than that of neat PP. In general, fiber orientation is induced by polymer flow during the injection molding process, and this causes anisotropic modulus. However, there was little difference between the flexural modulus of the NUKP-derived MFC composite obtained in Experiment 1 and the tensile modulus of the NUKP-derived MFC composite obtained in Experiment 2. It was supposed that fiber orientation was suppressed to some extent by entanglement of MFC.

The relationships between MFC content and tensile modulus and tensile strength are shown in Fig. 6. The

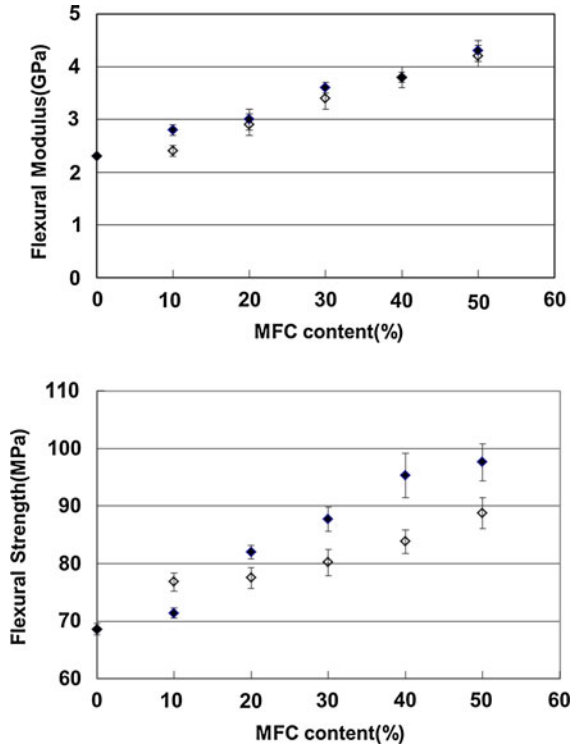

Fig. 4 Flexural modulus and flexural strength as functions of MFC content for (*filled diamond*) NUKP-derived MFC composite and (*diamond*) NBKP-derived MFC composite (*error bars* represent standard deviation)

tensile modulus of the MFC composites linearly increased with MFC content. Tensile strength linearly increased until 50 wt% MFC content, but decreased at 60 wt% content. This was probably due to MFC agglomerates appearing at high fiber content becoming fracture origins during mechanical testing.

The results of notched Izod impact strength and HDT under 1.82 MPa flexural load are shown in Table 4. Figure 7 shows notched Izod impact strength as a function of fiber content. Notched impact strength at 40 wt% MFC content was almost 2 times that of neat PP, but decreased at 50 wt%. This was probably because crack propagation stopped at the MFC during notched Izod impact testing, but fiber pull-outs from the matrix were easily induced by the weak interface, especially in high MFC content composites which had large interfacial area. This will be improved by strengthening the interface and also by blending the impact-modified PP, although the modulus will be decreased to some extent as a result (Oksman and Clemons. 1998).

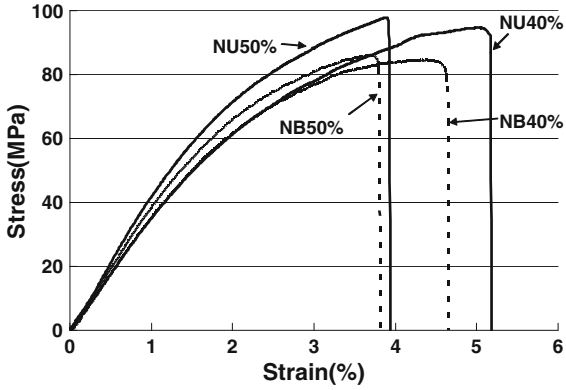

Fig. 5 Comparison of typical stress–strain curves for NUKP-derived MFC and NBKP-derived MFC composites

Results of HDT at 1.82 MPa as a function of MFC content are shown in Fig. 8. HDT exponentially increased with MFC content, reaching 122 °C at 50 wt% MFC content, 53 °C higher than for neat PP. This was probably because the entangled MFC matrix and hydrogen bonds between MFC molecules restricted the mobility of polymer chains. In addition, interfacial strength enhancement by MAPP contributed to increased HDT (Chattopadhyay et al. 2011).

Conclusion

A new process for continuous microfibrillation of pulp and its melt compounding with polypropylene was developed. Well dispersed MFC-reinforced PP composites was obtained using powdered PP and never-dried refiner-treated kraft pulp. The kraft pulp was fibrillated to nano and submicron size using a twin-screw extruder and was well mixed with powdered PP at the kneading stage. MFC did not agglomerate during melt compounding, and compounded pellets of

Table 3 Tensile properties of NUKP-derived MFC/PP/MAPP composites

MFC/PP/MAPP (wt%)	Tensile modulus (GPa)	Strength (MPa)	Elongation at break (%)
0/100/0	2.7	44.2	79.1
30/66/4	3.9	56.3	5.3
40/54.6/5.4	4.5	61.1	4.2
50/43.3/6.7	5.3	65.5	3.2
60/32/8	6.3	62.6	2.5

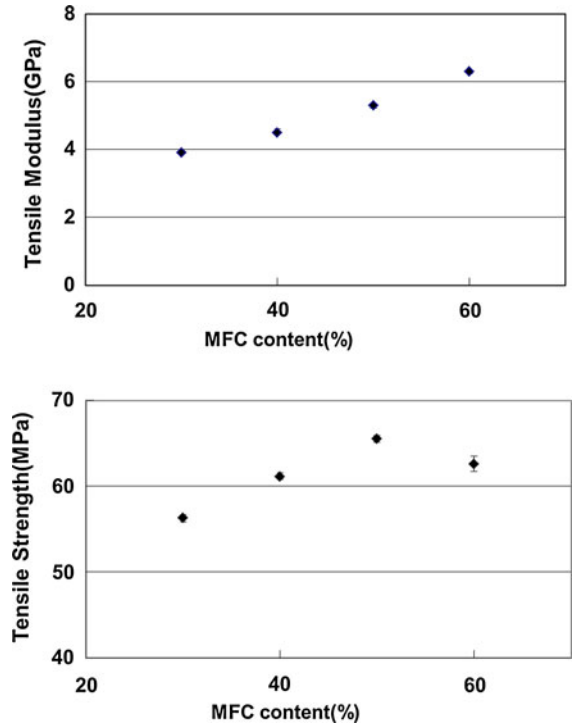

Fig. 6 Tensile modulus and tensile strength as functions of MFC content (*error bars* represent max and minimum values)

Table 4 Impact strength and HDT of NUKP-derived MFC/PP/MAPP composites

MFC/PP/MAPP (wt%)	MAPP content (%)	Izod impact strength (kJ/m^2)	HDT 1.8 MPa (°C)
0/100/0	0	1.9	69
30/66/4	4	3.3	86
40/54.6/5.4	5.4	3.9	97
50/43.3/6.7	6.7	3.2	122

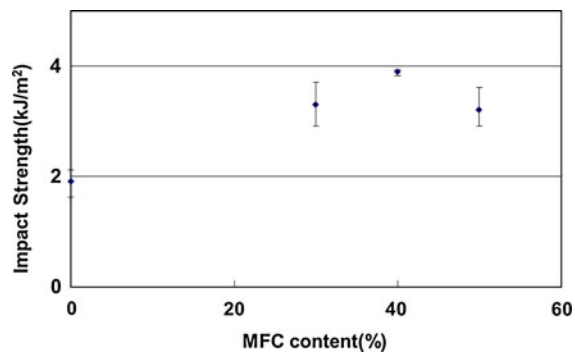

Fig. 7 Impact strength as a function of MFC content (*error bars* represent max and minimum values)

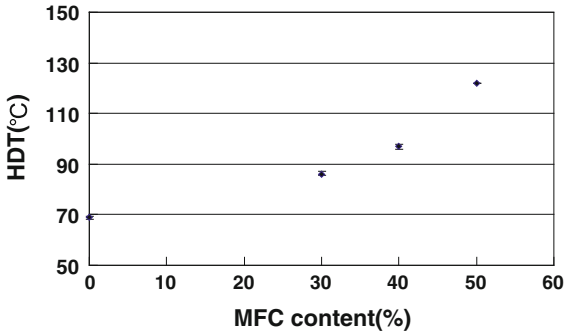

Fig. 8 HDT as a function of MFC content (*error bars* represent max and minimum values)

up to 60 wt% MFC content were successfully injection-molded.

For composites made by compression molding, both flexural modulus and strength increased with MFC content, and the tensile strength of NUKP-derived MFC composites was higher than that of NBKP-derived MFC composites. This is probably because NUKP is less damaged than NBKP during kraft pulping, and NUKP contains non-polar lignin which increases the compatibility between cellulose and PP.

We investigated the effect of MAPP content on the mechanical properties of the resulting composites. While flexural modulus was not affected, flexural strength was drastically increased by addition of MAPP.

Injection molded MFC composites produced in Experiment 2 showed good mechanical and thermal properties. Tensile strength, modulus, notched Izod impact strength, and HDT (under 1.82 MPa flexural load) at 50 wt% MFC content were 65.5 MPa, 3.2 kJ/m^2, and 122 °C, respectively, while neat PP had values of 44.2 MPa, 1.9 kJ/m^2, and 69 °C, respectively.

The present newly developed process is expected to enable the manufacture of high strength cellulose nanofiber-reinforced PP composite for industrial applications. In this study, MFC was well dispersed even at high MFC content, so this compound could be used as a master batch to make many products.

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