DETERMINING THE MECHANICAL PROPERTIES OF MICROCRYSTALLINE CELLULOSE (MCC)-FILLED PET–PTT BLEND COMPOSITES

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Abstract. Polymer composite materials consisting of poly(ethylene terephthalate) (PET)–poly(trimethylene terephthalate) (PTT) blends and microcrystalline cellulose (MCC) were prepared by injection molding. The composites were analyzed for tensile, flexural, and impact strength as well as density determinations. There was no statistical difference in terms of mechanical properties between the control PET–PTT blend and 2.5 wt% MCC-filled composites. Because of better compatibility as well as better stress-transfer properties, the tensile strength of the composites was larger (reaching values from 24.8-36.3 MPa with the addition of 20 wt% MCC). Elongation at break of the composites was greater (reaching values from 2.3-3.3% with the addition of 20 wt% MCC). The tensile modulus of MCC-filled composites systemically increased with increasing MCC loading (reaching values from 1.11-1.68 GPa with the addition of 30 wt% MCC). The flexural modulus of composites was higher than the control PET–PTT blend. The modulus also increased with increasing MCC loading (reaching values from 2.10-3.37 GPa with the addition of 30 wt% MCC). The Izod impact strength of the composites decreased as the MCC loading increased and this observation was in good agreement with commonly observed filled polymer systems.

Keywords: Microcrystalline cellulose (MCC), PET–PTT blends, mechanical properties, engineering thermoplastic composites, strength.

INTRODUCTION

Recently, there has been increased interest in the use of cellulose as mechanical reinforcing phases in polymer matrices because of its lower density compared with inorganic fillers (glass and carbon fibers), renewable character, biodegradability, lower price, lack of abrasion to the processing equipment, and relatively reactive surface (Helbert et al 1996; Azizi Samir et al 2004a, 2004b; Bondeson et al 2006; Ljungberg

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et al 2006; Panaitescu et al 2007a, 2007b). Wood is a primary resource for cellulose and, in addition, contains hemicelluloses and lignin (Seydibeyoglu and Oksman 2008). The thermogravimetric stability of wood component polymers shows that cellulose is more thermally stable than noncellulosic components such as lignin and hemicelluloses (Caulfield et al 2001a; Xu 2008). Engineering thermoplastics such as nylon (nylon 6 and 66) and polyesters (poly[ethylene terephthalate] [PET] and poly [trimethylene terephthalate] [PTT]) have greater mechanical properties and service temperatures than commodity thermoplastics. The automobile and construction industries desire low-density and inexpensive reinforcement for engineering thermoplastics to obtain better mechanical and thermal properties as well as lower density. Therefore, cellulose is a good candidate as reinforcement for engineering thermoplastics (Caulfield et al 2001a; Xu 2008).

Microcrystalline cellulose (MCC) is naturally occurring, crystalline cellulose obtained from high-quality wood pulp (Mathew et al 2004; Azizi Samir et al 2005). MCC has the advantage of high specific surface area in comparison with other conventional cellulose fibers and can be used as a cellulosic reinforcement for polymer matrices (Mathew et al 2004). MCC has been incorporated into a number of different matrices such as poly(lactic acid) (Mathew et al 2004; Petersson and Oksman 2006), polystyrene (Laka and Chernyavskaya 1996), and polyethylene (Maskavs et al 1999, 2001; Panaitescu et al 2007a, 2007b). Both tensile strength and elongation to break are lower in MCC-filled poly(lactic acid) composites than they are in pure poly(lactic acid) polymers. Conversely, increased MCC content has a positive effect on composite modulus compared with unfilled poly(lactic acid) (Mathew et al 2004). Tensile strength at break for a composite containing 40 wt% MCC was two times greater than the tensile strength for an unfilled polystyrene matrix (Laka and Chernyavskaya 1996). Addition of 5 wt% MCC in polyethylene contributes to a small increase in tensile strength, but higher MCC loadings show no significant tensile strength effect. However, tensile modulus increases with an increase of MCC loading up to 2.4 times that of unfilled polyethylene for 20 wt% MCC loading (Panaitescu et al 2007b).

It is thought that engineering thermoplastics that have higher melting temperatures cannot be effectively reinforced with cellulosic fibers because of thermal degradation of cellulose (Caulfield et al 2001a). Recent patent applications and three recent papers have demonstrated that this common belief is not true (Caulfield et al 2001a; Sears et al 2001a, 2001b, 2004; Jacobson et al 2001, Jacobson and Caulfield 2003). Despite technological interest in engineering thermoplastics, only a few studies of engineering thermoplastic/cellulose polymer composites have been published (Caulfield et al 2001a; Jacobson et al 2001; Sears et al 2001a, 2001b, 2004; Jacabson and Caulfield, 2003; McHenry and Stachurski 2003; Chen and Gardner 2008; Xu 2008). However, there is lack of information about MCC-filled engineering thermoplastics in the peer-reviewed literature. Aromatic polyesters, including PET and PTT, are important polyesters and high-performance engineering plastics and are widely used in many industries, including textiles, electronics, automotives, etc (Son et al 2003; Supaphol et al 2004; Chen et al 2007; Ge et al 2007; Liang et al 2007, 2008). Polymer blending is an attractive alternative to produce a new material with desirable properties such as versatility, simplicity, and low cost without having to synthesize a totally new material (Supaphol et al 2004). In recent years, many researchers have been interested in blends or copolymers to increase the crystallization rate of PET or to improve the mechanical and thermal properties of PTT (Son et al 2003; Supaphol et al 2004; Chen et al 2007; Ge et al 2007; Liang et al 2007, 2008; Wei et al 2005). In this study, PTT was used to reduce the processing temperature and increase the crystallization rate of PET (Liang et al 2007). There is also lack of information of cellulose fiber-filled polyester composites.

The objective of this study was to investigate the influence of MCC filler loading on the mechanical properties of MCC-filled PET–PTT composites. Microcrystalline cellulose was incorporated over a content range of 0-30 wt%. Analyses were conducted using tensile, flexural, and impact tests; composite densities were also determined.

MATERIALS AND METHODS

Matrix Polymer

The PET and PTT were supplied as polymer pellets by Shell Co and Azom Co, respectively. The PET and PTT had a density of 1300-1400 and 1350 kg/m^3 and intrinsic viscosities of 0.065 and 0.092 mL/kg, respectively. The lubricant (Struktol TPW 113) processing aid, being a blend of a complex, modified fatty acid ester and having a specific gravity of 1.005 and dropping point of 67-77°C, respectively, was supplied by Struktol Company of America.

Reinforcing Fillers

The MCC used as the reinforcement was a powder with particle sizes ranging 26-96 μm with an average of 50 μm. The MCC supplied by Sigma Aldrich Co was highly crystalline. It was stored in sealed containers after being oven-dried.

Sample Preparation

The MCC and PET–PTT blends were dried to moisture contents below 1% in an oven at 105°C for 16 h. The matrix polymer PET–PTT blend was mixed with the MCC. The compounding was done with a Brabender Prep-mixer equipped with a bowl mixer, and the process temperature and torque changes were measured in real time. Melt temperature and torque changes for every run were recorded to determine optimum processability for the PET–PTT blend–MCC composites. The basic processing parameters are listed in Table 1. The temperature was set to 270°C and rotor speed at 60 rpm. MCC was added to the mixer when the polymer melt appeared well mixed. After addition of the MCC, the melt temperature dropped sharply to 200°C and increased again as the mixing progressed. The melt mixture was released from the mixture immediately after the temperature reached 230°C. These temperature ranges and MCC residence times were recognized as a relatively safe temperature range to prevent severe thermal degradation with a guarantee of composite processability. Mixing was done for 10 min until the torque stabilized. The PET–PTT blend–MCC compounds were granulated using a laboratory scale grinder. The ground particles were dried in an oven at 105°C for 16 h before being injection-molded into ASTM test samples. All materials were injection-molded using a barrel temperature of 270°C, a mold temperature of 270°C, and an injection pressure of 17 MPa. The compositions of composites are shown in Table 2.

Statistical Analysis

The tensile strength, tensile modulus, elongation at break, flexural modulus, impact strength, and density were compared using a one-way analysis of variance followed by Tukey-Kramer honestly significant differences (HSD) test with JMP statistical analysis program (JMP Statistical Discovery Software Version 8 2008).

Scanning Electron Microscopy

Studies on the morphology of the tensile fracture surfaces of the composites were carried out using an AMR 1000 (AMRay Co) scanning electron microscope (SEM). Images were taken

Table 1. Basic operating parameters of the brabender rheomixer for PET–PTT blends.

	Operating parameters[a]				
Stage level	Set temperature	Melt temperature	RPM	Mix melt temperature	Reaction time (min)
First stage (PET–PTT)	270	260	60-70	—	7
Second stage (with MCC)	230	200-230	50	Under 230	3

[a] Temperatures are °C.

Table 2. Composition of MCC-filled PET–PTT composites.

	Composition[a]			
Sample code	MCC contents	PET	PTT	Lubricant
Neat PET–PTT	0	63	32	5
PET–PTT–MCC2.5	2.5	61.5	31	5
PET–PTT–MCC10	10	56.5	28.5	5
PET–PTT–MCC20	20	50	25	5
PET–PTT–MCC30	30	43	22	5

[a] Values are percentage by weight (wt%).

at 10 kV with 200 × and 2000 × SEM micrograph magnifications. All samples were sputtered with gold before the microscopic observations were obtained.

MECHANICAL PROPERTIES

Tensile Tests

All the tension tests were conducted according to the American Society of Testing and Materials (ASTM) standard D 638-03, "Standard Test Method for Tensile Properties of Plastics." The tensile behaviors of composites were measured using an Instron 8801 with a 10-kN load cell. All the tension tests were run at a rate of 5 mm/min. An extensometer was used for elongation determinations. At least six samples were tested for each composition, and the results are presented as an average for tested samples. Tensile modulus of the PET–PTT blend and MCC-filled composites was obtained from the slope of the stress–strain curve.

Flexural Tests

The flexure tests were conducted according to ASTM D 790-03, "Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials," Test Method 1, Procedure A, ie three-point loading system using center loading using an Instron 8801 with a 10-kN load cell. The support span was 50 mm. Tests were run at a rate of 1.25 mm/min. At least six samples were tested for each composition and the results are presented as an average for tested samples.

Impact Tests

The impact tests were conducted according to ASTM D 256-06, "Standard Test Methods for Determining the Izod Pendulum Impact Resistance of Plastics." The notches were added using a NotchVIS machine manufactured by Ceast. The samples were tested on a Resil 50 B impact test machine manufactured by Ceast. The sample was clamped in the bottom of the test fixture and the hammer was then released from a specified height. The depth under the notch and the sample width were entered, and the machine then recorded the energy taken to break the sample. All breaks must be completed breaks to count as a data point. A 2.75-J hammer was used to impact the samples. At least seven samples were tested for each composition.

Density Tests

The density profile of the PET–PTT blend and MCC-filled composite samples was measured using an X-ray densitometer (QMS, Model QDP-01). At least five samples were tested for each composition. The dimension of density samples was 63.5 × 12.5 × 3.2 mm. Density determination by the QDP scanning system is based on the relationship between X-ray attenuation and density as expressed in the following equation (QMS 2001; Jeong 2005).

$$I/I_0 = e^{\mu\rho t}$$

where I = intensity of radiation beam after passing through the sample, I_0 = intensity of radiation beam without passing through the sample, μ = material mass attenuation coefficient (m^2/kg), ρ = material density (kg/m^3), and t = material thickness (m).

RESULTS AND DISCUSSION

Tensile Properties

The tensile behavior of the neat PET–PTT blend as well as the MCC-filled composites reinforced from 2.5-30 wt% was performed by tensile testing at room temperature. Figures 1 and 2 show

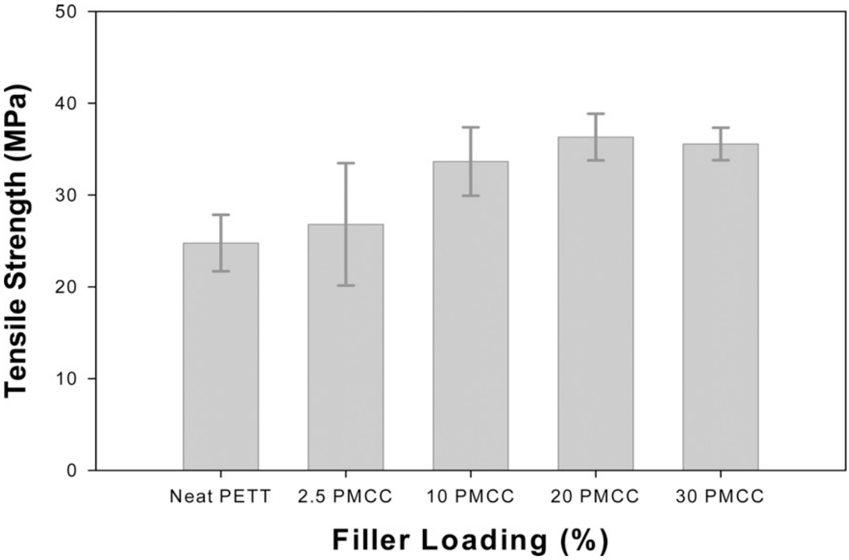


Figure 1. Effect of MCC loading on tensile strength of MCC-filled PET–PTT composites.

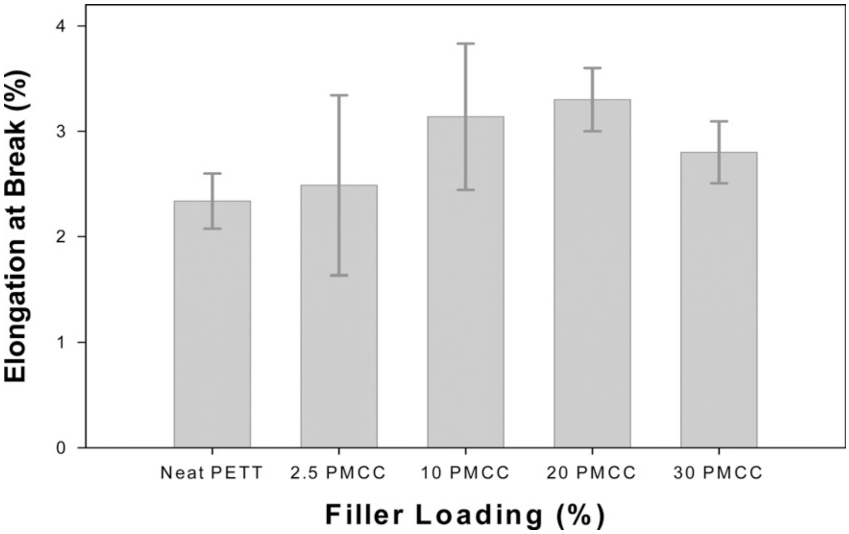


Figure 2. Elongation at break as function of MCC loading for MCC-filled PET–PTT composites.

the tensile strength and elongation at break of the neat PET–PTT blend and MCC-filled composites, respectively. The tensile strength and elongation at break were determined from the stress and strain curves. Incorporation of filler into a polymer matrix may increase or decrease the tensile strength of the composite and this phenomenon depends on filler type (Zaini et al 1996). It was observed that neat PET–PTT exhibits a nonlinear elastic behavior with a tensile strength of 24.75 MPa and an elongation at break of 2.34%. None of the composites including the neat PET–PTT showed signs of stress yielding. This lack of stress yielding suggests that the mechanism behind the elongation and rupture of the composites was quite similar compared with the neat PET–PTT blend. The composite reinforced with MCC displayed enhanced tensile properties in comparison with the neat PET–PTT blend. Because of better stress–transfer properties, the tensile strength of the composites was greater (reaching values from 24.8-36.3 MPa with the addition of 20 wt% MCC) (Ljungberg et al 2006). After 20 wt% MCC addition, the tensile strength decreased, but it was greater compared with the neat PET–PTT blend. The reason why tensile strength decreased after 20 wt% MCC addition was attributed to very little or no stress–transfer properties in the higher weight percent MCC-filled composite (Ljungberg et al 2006). In general, almost all filled polymer systems display smaller elongation at break compared with a neat polymer matrix because of deformability of a rigid interphase between the filler and matrix material (Zaini et al 1996). Figure 2 seems to be contradictory to the general observation. In Fig 2, elongation at break of composites was greater (reaching values from 2.3-3.3% with the addition of 20 wt% MCC). The reason for this was a better dispersion below 20 wt% MCC addition. With high MCC contents, the degree of MCC–MCC interaction became more prominent and, as a consequence, a reduction in elongation at break was observed (Dikobe and Luyta 2007). Figure 3 shows the tensile modulus of elasticity of PET–PTT blend and MCC-filled composites. The tensile modulus of elasticity of a polymeric material has been shown to be significantly improved when nanomicrocomposites are formed with cellulose (Mathew et al 2004; Petersson and Oksman 2006; Seydibeyoglu and Oksman 2008). The modulus of elasticity of MCC-filled composites systemically increased with increasing MCC loading (reaching values from 1.11-1.68 GPa with the addition of 30 wt% MCC). Tensile measurements show that the effect of MCC is more pronounced on the tensile modulus of elasticity and MCC acts as a mechanical reinforcement of the polymer chains. Mathew et al prepared polylactic acid–MCC composites and they observed a similar reinforcement effect with MCC (Mathew et al 2004).

Flexural Properties

The flexural behavior of the neat PET–PTT blend as well as the MCC-filled composites reinforced from 2.5-30 wt% was determined by flexure testing at room temperature. In Fig 4, the average values of the flexural modulus of elasticity as a function of MCC are shown. As can be seen in the figure, the flexural modulus of elasticity of composites was higher than that of the PET–PTT blend. The modulus of elasticity also increased

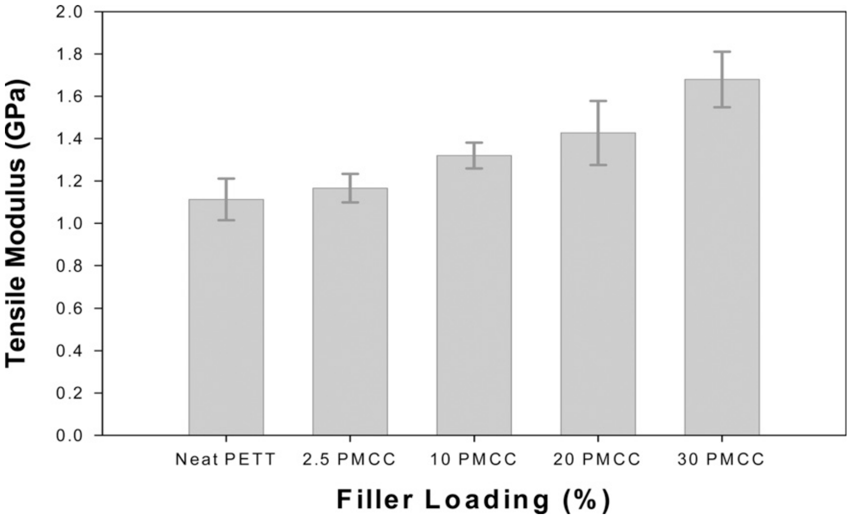


Figure 3. Tensile modulus of elasticity of MCC-filled PET–PTT composites.

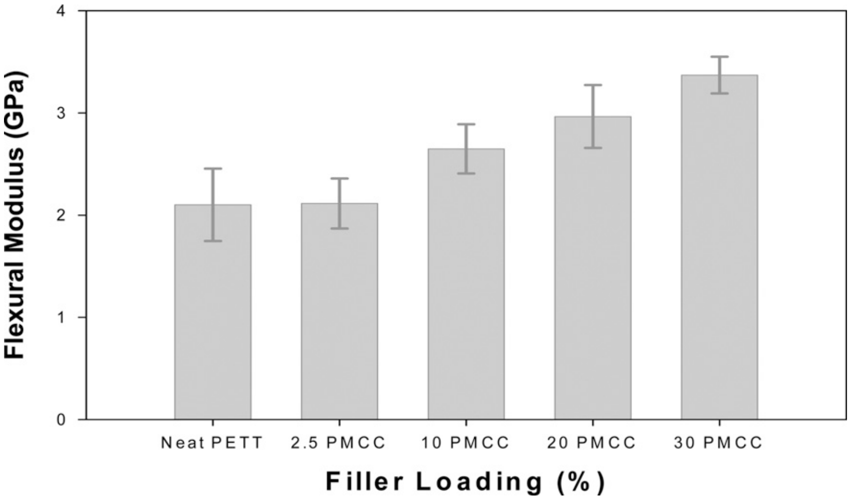


Figure 4. Flexural modulus of elasticity of MCC-filled PET–PTT composites.

with increasing MCC loading (reaching values from 2.10-3.37 GPa with the addition of 30 wt% MCC). The flexural modulus of elasticity is strongly affected by MCC loading and MCC orientation and less by the PET–PTT blend–MCC interaction (Bengtsson and Oksman 2006). Caulfield et al prepared aliphatic polyketone and polyester–cellulose composites and observed similar reinforcements with 33 wt% cellulose pulp fiber content (Caulfield et al 2001b). Also, Sears et al used cellulose wood pulps as reinforcement for nylon 6 composites and found that cellulose wood pulps increased flexural modulus compared with neat nylon 6 polymer (Sears et al 2001b).

Impact Properties

The impact strength of the composites based on the amount of filler loading and type of testing is dependent on whether the samples were notched or unnotched (Sanadi et al 1997). The impact behavior of the neat PET–PTT blend as well as the composites reinforced from 2.5-30 wt% MCC was performed by notched Izod impact testing at room temperature. Figure 5 shows the Izod impact strength of PET–PTT and MCC-filled composites. The Izod impact strength of composites decreased as the MCC loading increased and this observation is quite expected for filled polymer systems and has been commonly observed (Zaini et al 1996; Sanadi et al 1997; Liang 2002; Bengtsson and Oksman 2006; Cui et al 2008). Because of the possibility of poor wetting of the MCC particles by the PET–PTT blend, poor interfacial adhesion may have been created between the MCC and the PET–PTT blend and it contributed to weak interfacial regions in Fig 6, which presents the tensile fracture surfaces of the composites (Zaini et al 1996). Figure 6(a) as a low resolution shows debonded traces of MCC particles and Fig 6(b) as a high resolution shows the microcrack between a MCC particle and PET–PTT matrix. This poor wetting phenomena might cause lower impact strength with the addition of MCC loading. Another reason for this observation was the untreated filler surface, which caused poor dispersion of the MCC in the PET–PTT matrix. In this case, the impact strength decreased quickly although at low filler addition (Zaini et al 1996; Bengtsson and Oksman 2006). On the other hand, a relatively small stress concentration might be formed in the PET–PTT blend because of the smooth rod-like shape of the MCC. This phenomenon causes weak sensitivity of impact strength to filler content (Liang 2002). During the impact tests, cracks travel through the polymer as well as along the weak

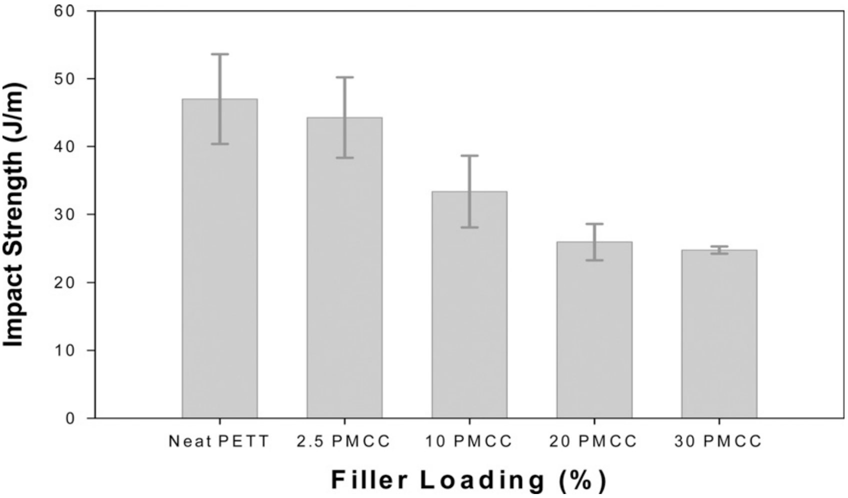
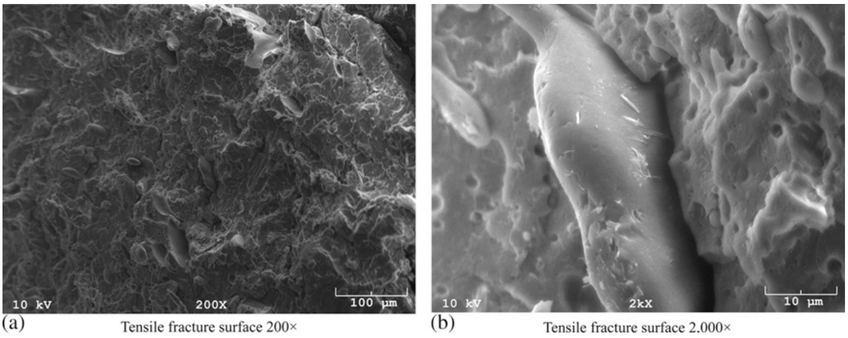


Figure 5. Effect of MCC loading on Izod notched impact strength of PET–PTT blend and MCC-filled composites.



(a) Tensile fracture surface 200× (b) Tensile fracture surface 2.000×

Figure 6. Scanning electron micrographs of 20 wt% MCC filled PET–PTT blend composites.

interfacial regions, which might be caused from poor wetting of the particles by the PET–PTT blend. As a result of crack travel, the latter cannot resist crack propagation as effectively as the polymer region (Zaini et al 1996). Increasing MCC content only increases the interfacial regions, which cause crack propagation. Also, addition of MCC might cause polymer immobility, and that can contribute to lower impact strength. Figure 5 also shows that the Izod impact strength of the composites decreased from 47 for the control to 25 J/m for 30 wt% MCC-filled composite. The loss of impact strength of the composite can be improved by providing flexible and strong interfacial bonding in the composite or by using coupling agents or impact modifiers (Zaini et al 1996; Sanadi et al 1997; Liang 2002; Bengtsson and Oksman 2006; Cui et al 2008).

Density Properties

Figure 7 shows the density of PET–PTT and MCC-filled composites. The density of the composites increased slightly as a function of MCC loading. This was expected because MCC has a greater density compared with the PET–PTT blend. Generally, there is a certain correlation between density and mechanical properties such as flexural strength and modulus of elasticity (Klyosov 2007). There was not a strong correlation between density and

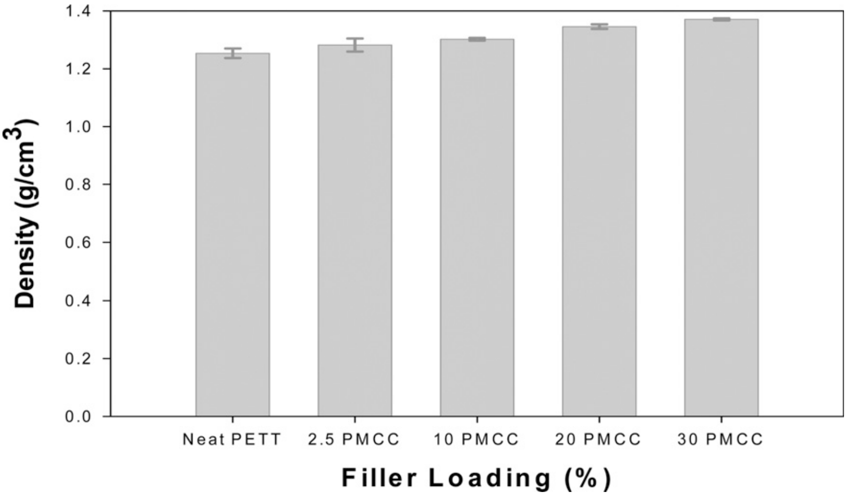


Figure 7. Density as function of MCC loading for MCC-filled PET–PTT composites.

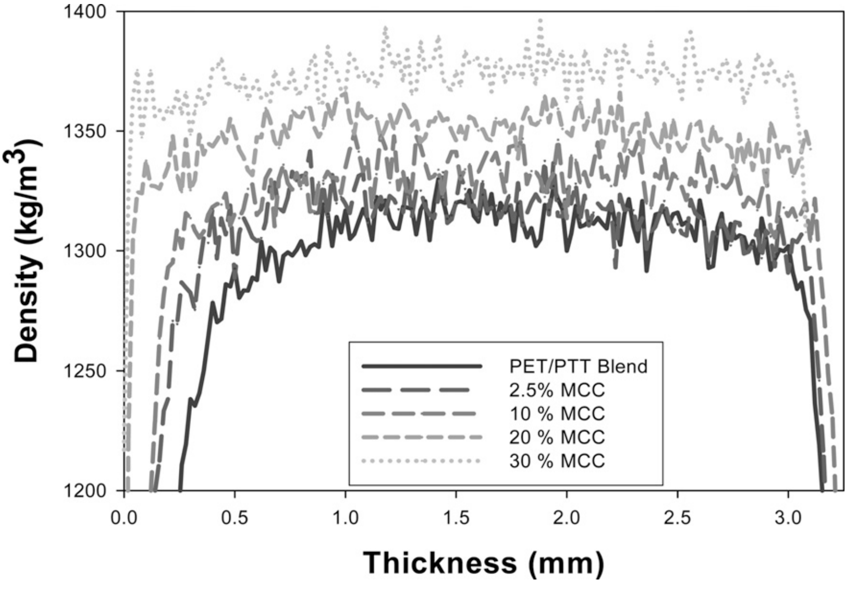


Figure 8. Average density profile of PET–PTT blend and MCC-filled composites.

tensile modulus of elasticity ($R^2 = 0.74$) or density and flexural modulus of elasticity ($R^2 = 0.80$) (in this study). It was assumed that these large improvements in the mechanical properties would come from the MCC itself. The strong hydrogen bonding between the MCC molecules, which leads to strong interactions between fibers and fibrils, might contribute to better mechanical properties in comparison with the PET–PTT blend (Seydibeyoglu and Oksman 2008). As a result of the density study, it can be said that MCC provided the reinforcement (flexural and tensile modulus) with a smaller increase in density, which could be important for low weight applications, especially in the automobile industry (Caulfield et al 2001b). As expected, the

Table 3. Results of Tukey-Kramer HSD comparison at $\alpha = 0.05$ for PET–PTT blend and MCC-filled composites.

	Tensile properties		Flexural properties		Impact properties	
Sample code	Strength (MPa)	Modulus (GPa)	Elongation (%)	Modulus (GPa)	Izod Notched (J/m)	Density g/cm^3
Neat PET–PTT	B	C	B	C	A	C
PET–PTT–MCC2.5	B (NC)	C (NC)	AB (NC)	C (NC)	A (NC)	BC (NC)
PET–PTT–MCC10	A (+36%)	B (+19%)	A (+34%)	B (+26%)	B (−29%)	B (+4%)
PET–PTT–MCC20	A (+47%)	B (+28%)	A (+41%)	AB (+41%)	BC (−45%)	A (+7%)
PET–PTT–MCC30	A (+44%)	A (+51%)	AB (NC)	A (+60%)	C (−47%)	A (+9%)

The same letters indicates no statistical difference between properties of composites and those around it. NC is no significant change on the addition of MCC ($\alpha = 0.05$) and parentheses show that effect of MCC loading on the mechanical properties of composites in comparison with the PET–PTT blend.

PET–PTT blend and MCC-filled composites manufactured by injection molding had highly uniform density distribution through their thickness as shown in Fig 8. Table 3 shows a summary of the effect of MCC loading on the mechanical properties and density of composites in terms of statistical analysis.

CONCLUSIONS

MCC-filled PET–PTT blend composites were prepared by melt compounding followed by injection molding. The following conclusions are reported.

1. There was no statistical difference in terms of mechanical properties between the PET–PTT blend and 2.5 wt% MCC-filled composites. The composite reinforced with high filler loading MCC displayed enhanced tensile properties in comparison with the neat PET–PTT blend. Because of better stress–transfer properties, the tensile strength of MCC-filled composites, compared with that of the control, ranged from 36-47%. Tensile modulus of elasticity under tensile load increased from 19-51% when MCC was added to the PET–PTT blend. Elongation at break of composites was greater (reaching values from 2.3-3.3% with the addition of 20 wt% MCC). Elongation at break increased compared with the controls from 34-41% when MCC was added to the PET–PTT blend.
2. The flexural modulus of elasticity increased with increasing MCC loading (reaching values from 2.10-3.37 GPa with the addition of 30 wt% MCC). Flexural modulus of the MCC-filled composites compared with that of the control increased from 36-47%.
3. Izod impact strength decreased compared with the control from 29-47% when MCC was added to the PET–PTT blend.
4. The density of the composites increased slightly as a function of MCC loading. This was expected because MCC has a greater density compared with the PET–PTT blend. There was not a strong correlation between density and tensile modulus of elasticity ($R^2 = 0.74$) or density and flexural modulus of elasticity ($R^2 = 0.80$). It was assumed that large improvements in the mechanical properties arose from the MCC itself. MCC-filled composites manufactured by injection molding had a highly uniform density distribution through their thickness.

Overall, MCC-filled composites showed comparable or better mechanical properties compared with the PET–PTT blend without adding any compatibilizer or coupling agent. MCC could be good reinforcing filler for engineering thermoplastic applications such as in automobiles. It is believed that the formulation containing only PET–PTT blend and MCC (without lubricant) will show better mechanical properties in comparison with a lubricant-added PET–PTT blend and MCC formulation. In this study, lubricant was used only as an additive to improve processing conditions.

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