

Compatibilization of Recycled Poly(ethylene terephthalate) and Polypropylene Blends: Effect of Polypropylene Molecular Weight on Homogeneity and Compatibility

Hiroyuki Inoya,^{1,2} Yew Wei Leong,¹ Warunee Klinklai,³ Supaporn Thumsorn,¹
Yuki Makata,¹ Hiroyuki Hamada¹

¹Advanced Fibro-Science, Kyoto Institute of Technology, Matsugasaki, Sakyo-ku, Kyoto 606-8585, Japan

²Yasuda Sangyo Co. Ltd., Minaminekoyacho, Fushimi-ku, Kyoto 612-8379, Japan

³Faculty of Engineering, Rajamanagala University of Technology Thanyaburi, Klong 6, Thanyaburi, Patumthani 12110, Thailand

Received 24 December 2008; accepted 21 October 2009

DOI 10.1002/app.34405

Published online 29 November 2011 in Wiley Online Library (wileyonlinelibrary.com).

ABSTRACT: The effectiveness of compatibilizers in enhancing the dispersion of polypropylene (PP) at various molecular weights in recycled polyethylene terephthalate (RPET) was elucidated. The idea of incorporating PP of different molecular weights evolved from the intention of simultaneously recycling the PET bottles together with the PP-based bottle caps, which are often of low molecular weight (M_w). Three grades of PP with known molecular weights were blended with RPET at various loadings of compatibilizers. Morphological analyses suggest that the dispersion of the PP particles was more homogeneous, and the average particle size was smaller when low M_w PP was incorporated. This indicates that the interaction between

the compatibilizer and PP particles was more intense with the presence of a large number but shorter PP molecular chains. Moreover, specimens containing low M_w PP were found to remain homogeneous regardless of compatibilizer and PP content in the RPET/PP blends. The homogeneity of the blends significantly affected their mechanical performance as well. Higher stiffness, yield strength, deformability, and toughness were observed when low M_w PP was incorporated, regardless of PP and compatibilizer loadings. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 124: 3947–3955, 2012

Key words: polyethylene terephthalate; polypropylene; recycling; molecular weight; compatibilization

INTRODUCTION

Polyethylene terephthalate (PET)-based waste is abundant because of its popularity in the disposable food packaging industry. According to statistics, textile and soft drink bottles were the main sources of PET waste, and the amount increases year after year.^{1–4} In Japan alone, the vast usage of PET bottles for mineral water, soy sauce, edible oil, and all soft drinks has contributed to a total of about 554,104 tons of waste, which has prompted the Japanese government to impose a law to ban PET bottles from being sent to the landfills but instead systematically collected for recycling to save landfill space and also to promote energy conservation.^{4,5}

During the recycling process, the PET bottles are first crushed into flakes together with the attached labels and caps. The caps and labels, which are typically made from polypropylene (PP) or polyethylene

(PE), are separated from the PET flakes by passing them through a floatation bath where PP and PE will float because of their low densities. The resulting PET flakes are then washed and dried before being used for other processes, while the remaining PP and PE wastes are either discarded or sold as scrap. Although the caps constitutes only between 5 and 10 wt % of the total bottle weight, the gross weight is estimated to be more than 55,000 tons. Therefore, in our previous work, some efforts were devoted to investigate the possibilities of recycling the PET bottles together with the caps.⁶ Because of the incompatibility between PET and PP, a two-phase morphology is expected, whereby the dispersed phase takes the form of microspheres, which suggest very weak interaction between the matrix and dispersed phase. The morphology and interfacial characteristics between the various phases are predominant factors that influence the ultimate properties of these immiscible blends. Therefore, our previous study involved the optimization of compatibilizer contents with respect to the amount of PP present in the recycled PET (RPET)/PP blends.⁶ A higher degree of homogeneity in the blends was

Correspondence to: Y. Wei Leong (leong@kit.ac.jp).

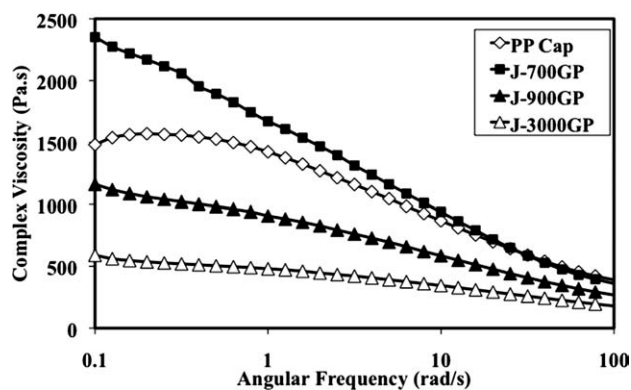


Figure 1 Comparison of complex viscosities between neat PP of various grades and PP from waste bottle caps.

found with increasing compatibilizer content. However, the incorporation of more compatibilizers would almost certainly incur higher cost; thus, it is necessary to find other means of improving the homogeneity and mechanical performance of the blend with minimal compatibilizer contents.

In this work, blending of RPET with different molecular weight of PP was performed, with and without the presence of compatibilizers. The main goal of this study is to obtain a high degree of compatibilization with the least amount of compatibilizers. A high degree of compatibilization refers to significant improvements in mechanical properties of the blend, with special attention to the blend deformability and toughness, since brittleness is typically related to an inhomogeneous or incompatible polymer blend.

EXPERIMENTAL

Materials

The RPET used in this study was obtained in the form of flakes through the crushing of postconsumer PET bottles that were collected by Yasuda Sangyo Co., Ltd., Japan. The flakes were prewashed in water to minimize impurities prior to usage.

Since the caps were separated from the bottles and have all been discarded, the source of PP from the caps was not available. Therefore, instead of recycled PP, neat PP of various grades, i.e., PP J700GP ($MI = 8$, $M_w = 3.1 \times 10^5$ g/mol), PP J900GP ($MI = 13$, $M_w = 2.3 \times 10^5$ g/mol), and PP J3000GP ($MI = 30$, $M_w = 1.9 \times 10^5$ g/mol), which were obtained from Idemitsu Petrochemical Co., Ltd. (Japan), were incorporated into RPET. Specimens incorporated with J700GP, J900GP, and J3000GP will be represented with a designation of H, M, and L that represent high, medium, and low molecular weight PP, respectively. The rheological properties of the neat PP as compared with the waste PP from bottle

caps at 200°C are presented in Figure 1. It can be observed that the complex viscosity of the PP caps are within the range of the three grades of neat PP chosen for this study, irrespective of the angular frequencies used.

The RPET/PP ratio was varied at 95/5 and 90/10 based on the fact that these compositions are representative of the actual PET/PP content of a drinking bottle with a cap (based on 500 and 1250 mL bottles). Styrene-ethylene-butadiene-styrene-based terpolymer (SEBS) (Dynaron 8630P; JSR Co., Japan) was used as a compatibilizer with loadings of 3 and 5 phr. Blends without compatibilizers were also prepared as control. A comprehensive list of specimen designations according to their compositions is compiled in Table I.

Prior to blending, RPET was dried in a dehumidifying dryer for 5 h at 120°C. The RPET and PP were dry-blended prior to compounding in a single screw extruder (SRV-P500, Nihon Yuki Co., Ltd., Japan) set at a barrel temperature between 265 and 290°C and screw rotation speed of 430 rpm. The extrudate was air cooled through a conveyor belt lined with cooling fans before being pelletized. The resulting pellets were then dried using special dehumidifying drier for RPET (Piccolo), which was supplied by ITSWA Co., Ltd., Japan for 5 h at 80°C. Dog bone shaped specimens were prepared using a 50-ton injection molding machine (PO YUEN (TO's) Machine FTY Ltd., Hong Kong). The mold used in this study had a dog bone shaped cavity that is 175 mm in length, and the midsection is 10-mm wide and 3-mm thick. The gauge length is 115 mm. The injection molding barrel temperature, mold temperature, and injection speed was set at 270°C, 30°C, and 100 mm/s, respectively.

Thermal property characterizations

The effects of compatibilizer content and PP M_w on the crystallization behavior of the blends were characterized by differential scanning calorimetry (DSC) (TA Instruments DSC2920). The samples were initially heated to 300°C at 100°C/min to erase the thermal history of the blends, followed by cooling at 5°C/min to 30°C to obtain the crystallization temperature of the RPET phase. Fresh samples were heated to 200°C at 100°C/min and cooled at 5°C/min to 30°C to obtain the crystallization temperature of the PP phase. All analyses were conducted under nitrogen atmosphere to prevent oxidation of the specimens.

Morphological characterizations

Morphological observations were performed by using a scanning electron microscope (SEM), JEOL JSM-5200, Japan. The cross sections of tensile-

TABLE I
Specimen Designations, Impact Performance, and Thermal Characteristics of Various RPET/PP Blends

PP grade	Specimen designation	Material composition			Un-notched Izod impact strength (kJ/m ²)	Notched Izod impact strength (kJ/m ²)	Crystallization temperature of PP (°C)
		RPET (wt %)	PP (wt %)	Compatibilizer (phr)			
J700GP (high M_w)	H-95/5/0	95	5	0	55.9	1.8	123.22
	H-95/5/3	95	5	3	NB	2.1	114.86
	H-95/5/5	95	5	5	NB	1.8	106.88
	H-90/10/0	90	10	0	33.9	1.8	123.97
	H-90/10/3	90	10	3	NB	1.8	116.16
	H-90/10/5	90	10	5	NB	1.7	109.55
J900GP (medium M_w)	M-95/5/0	95	5	0	32.8	1.9	124.33
	M-95/5/3	95	5	3	NB	1.8	117.88
	M-95/5/5	95	5	5	NB	1.6	108.94
	M-90/10/0	90	10	0	42.9	1.6	124.56
	M-90/10/3	90	10	3	NB	1.6	116.95
	M-90/10/5	90	10	5	NB	1.5	115.82
J3000GP (low M_w)	L-95/5/0	95	5	0	62.7	1.8	123.50
	L-95/5/3	95	5	3	NB	1.9	116.60
	L-95/5/5	95	5	5	NB	1.8	109.09
	L-90/10/0	90	10	0	35.3	1.9	124.00
	L-90/10/3	90	10	3	NB	1.7	115.91
	L-90/10/5	90	10	5	NB	1.7	110.96

fractured surface dumbbell specimens were coated with a thin layer of gold prior to observation. An image analysis software (Image J) was used to measure the size of the Polypropylene phase from the pictures taken by SEM. A total of 200 particles were randomly selected from different locations of the specimens for measurements to ensure statistical validity. The homogeneity of the blends could be evaluated from the polydispersity of the dispersed phase particles^{7–11}:

$$\text{Polydispersity} = d_v/d_n, \quad (1)$$

where d_v is the volume average particle diameter, and d_n is the number average particle diameter; both these values can be calculated from equations given in the literature.^{7–11} The measurements of particle size diameter were found to be reproducible with an error of $\pm 15\%$.

Mechanical testing

Tensile tests of blends were performed on the dumbbell test specimens at a cross head speed of 50 mm/min by using a universal testing machine (Instron 4466, INSTRON USA), in accordance to ASTM D638.

An Izod impact tester (Toyoseiki, Japan) was used to measure the Izod impact strength with notched and un-notched specimens. The samples with dimensions of 10 mm \times 6 mm \times 3 mm were cut from dumbbell specimens. Notches were 2-mm deep and angled at 45°. For all mechanical property characterizations, at least five samples were tested at 27°C and their results averaged.

RESULTS AND DISCUSSION

Thermal properties of RPET/PP blends: Effect of compatibilizer content and PP M_w

The introduction of compatibilizers into the RPET/PP blends resulted in a shift in crystallization temperature (T_c) of the PP phase toward a lower temperature, as can be observed from Figure 2 and in Table I. This shift is more evident with increasing compatibilizer content. By varying PP M_w , the T_c of PP in the uncompatibilized blend remained constant. However, when compatibilizers were incorporated, the shift in T_c was more evident in specimens containing high M_w PP. Meanwhile, the thermal properties of the RPET phase remained unchanged. This indicates that the compatibilizers have a preferential interaction and affinity toward the PP phase, which delays the formation of PP spherulites during cooling. Interestingly, the melt enthalpy of the PP phase was independent of the presence of compatibilizers, which means that the crystallinity of the PP phase remained unchanged. Therefore, it is suggested that only the size of the PP spherulites or crystallization behavior might have been affected by the M_w of PP. Furthermore, the crystallization peaks of the PP phase in compatibilized blends appeared to be broader than those in uncompatibilized blends, which suggest that a reduction in PP crystallization rate is possible. Considering that the cooling rate during DSC measurements was set at 5°C/min, the crystallization rate of PP could be suppressed during actual injection molding process given the much higher cooling rate. The suppression of PP crystallization rate could lead to significant changes in

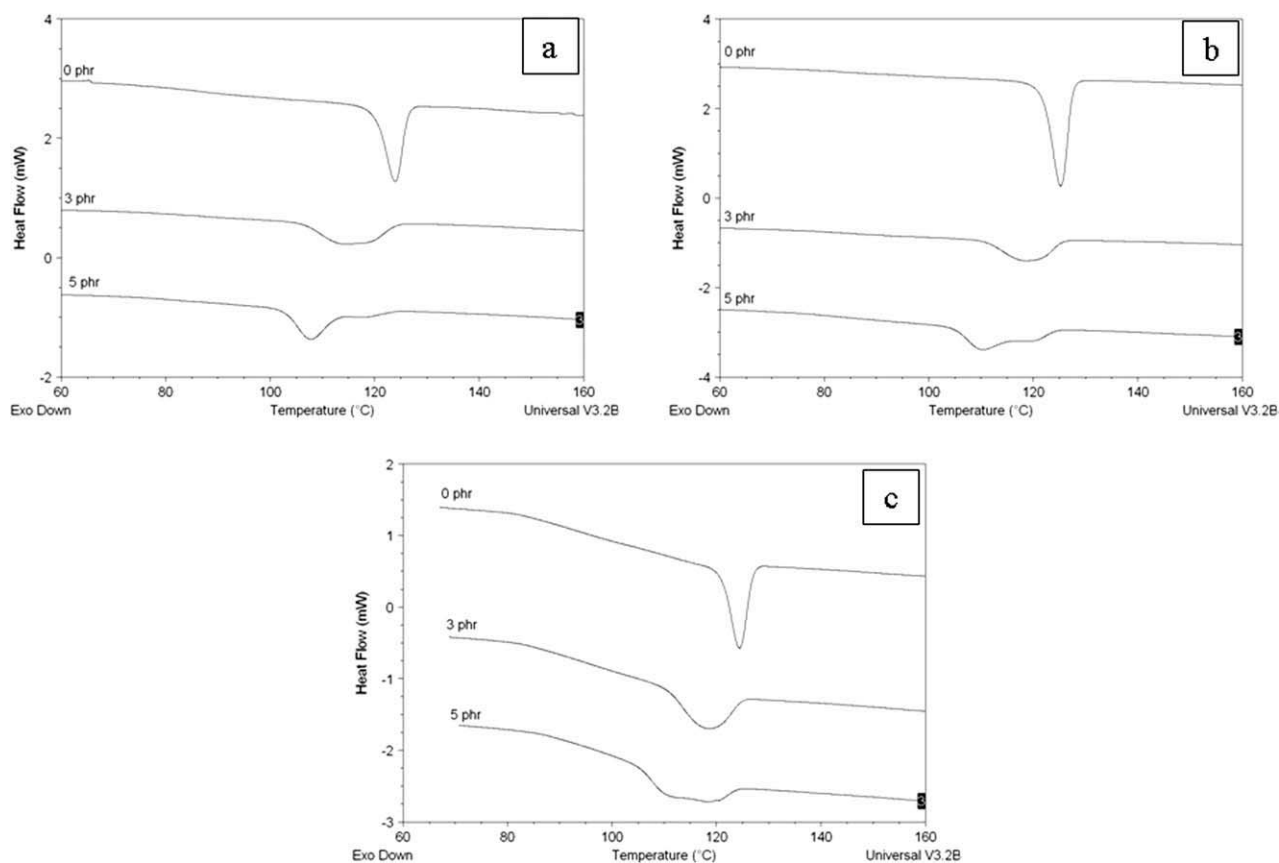


Figure 2 Exothermic DSC thermograms of the PP phase in RPET/PP (95/5) blends at various compatibilizer contents: (a) H-95/5; (b) M-95/5; and (c) L-95/5.

morphological development as well as mechanical performances of the blends.

Mechanical properties of RPET/PP blends: Effect of PP content and M_w

The notched and un-notched Izod impact performance of the blends can be elucidated from Table I. It is clear that with the incorporation of compatibilizers, the un-notched specimens did not fracture upon impact. The enhanced toughness of the blends could be attributed to the significant reduction in dispersed phase size, which will be discussed later, as well as the suppression of PP crystal formation in the presence of compatibilizers. It is also noteworthy that the L-95/5/0 specimens recorded the highest impact strength among all un-notched specimens even without the presence of compatibilizers, which indicates that the incorporation of low M_w PP into the blend can also induce toughening. Nevertheless, there was no improvement in the notch sensitivity of RPET, judging from the consistently low impact strength of the notched specimens regardless of PP M_w and compatibilizer content.

The effect of PP M_w on the stiffness of RPET/PP blends at various compatibilizer loadings can be

seen in Figure 3. Generally, the presence of compatibilizers would cause a slight decline in the stiffness of the blend because of the inherent property of the compatibilizers. However, it was observed that the change in stiffness is even more distinct when the M_w of PP was varied, irrespective of compatibilizer content. An increase in Young's modulus of about 8–13% could be observed when the blends were incorporated with low M_w PP (J3000GP) as opposed to high M_w PP (J700GP), especially when higher PP content was present, as depicted in Figure 3. It is generally accepted that a higher M_w polymer would exhibit higher stiffness. However, in this case, it is obvious that the increments in modulus of the blends are despite the decreasing stiffness of the low M_w PP phase. Hence, it is thought that the interaction between the RPET and PP phases plays a more significant role in determining the stiffness as opposed to the intrinsic properties of the individual components.

The yield strengths of the blends are compared in Figure 4 for RPET/PP blend ratios of 95/5 and 90/10. It could be seen that the yield strengths of the 95/5 specimens were more dependent on the PP M_w than the compatibilizer content. However, in 90/10 blends, a more drastic increase in yield strength

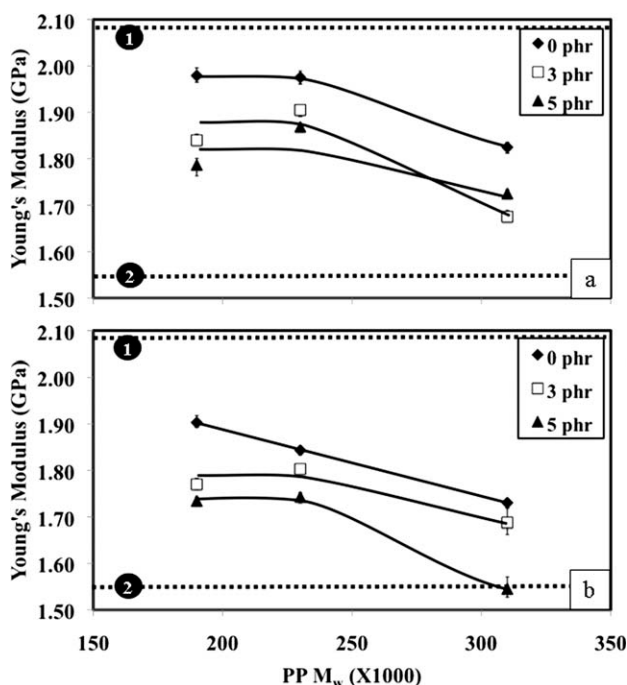


Figure 3 Effect of PP M_w on Young's moduli of (a) RPET/PP (95/5) blends and (b) RPET/PP (90/10) blends at various compatibilizer contents. Dotted lines represent the Young's moduli of (1) neat RPET and (2) neat PP J-3000GP.

could be observed with decreasing PP M_w . The incorporation of compatibilizers would further improve the yield strength, as have also been

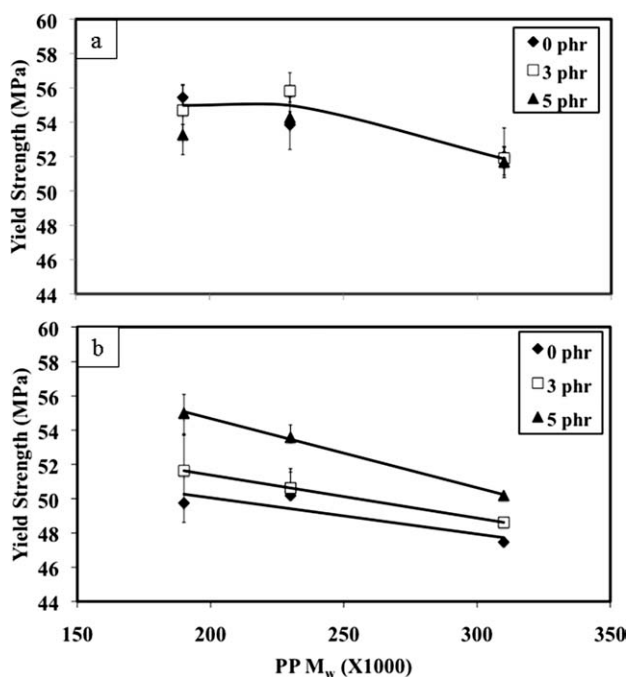


Figure 4 Effect of PP M_w on yield strengths of (a) RPET/PP (95/5) blends and (b) RPET/PP (90/10) blends at various compatibilizer contents. The yield strengths of neat RPET and PP J-3000GP are 61.5 MPa and 38 MPa, respectively.

reported in our previous work.⁶ These increments in yield strengths suggest that blends with lower M_w PP exhibit higher resistance to plastic deformation. It should be noted that the 5 phr compatibilized 90/10 blends could achieve similar yield strengths as that of compatibilized 95/5 blends with low M_w PP. This indicates the possibility of incorporating a higher loading of PP into RPET while maintaining the mechanical properties should a lower M_w PP be used.

The most obvious effects of varying PP M_w in the blends could be seen from the deformability of the blends, as depicted in Figure 5. Typically, polymers with high M_w would exhibit higher ductility and toughness as opposed to low M_w polymers. This is true in uncompatibilized 95/5 and 90/10 blends incorporated with high M_w PP whereby high elongations at break of up to 200% were recorded despite their low stiffness and yield strengths. However, with the incorporation of compatibilizers, significant increases in deformability were observed especially in blends containing low M_w PP. A sevenfold increase in elongation at break was observed when the 95/5 blends with low M_w PP were compatibilized. Similar compatibilization was not observed when the 95/5 blends were incorporated with high M_w PP. Similar tendencies could be observed for 90/10 blends, as depicted in Figure 5(b), which suggest that compatibilization is most effective with the presence of low M_w PP. Similar observations of enhanced ductility and toughness of compatibilized

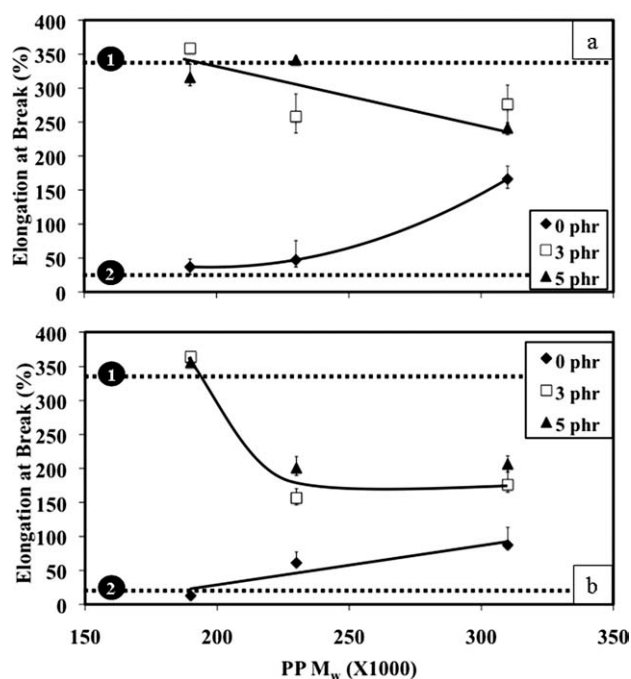


Figure 5 Effect of PP M_w on elongations at break of (a) RPET/PP (95/5) blends and (b) RPET/PP (90/10) blends at various compatibilizer contents. Dotted lines represent the elongations at break of (1) neat RPET and (2) neat PP J-3000GP.

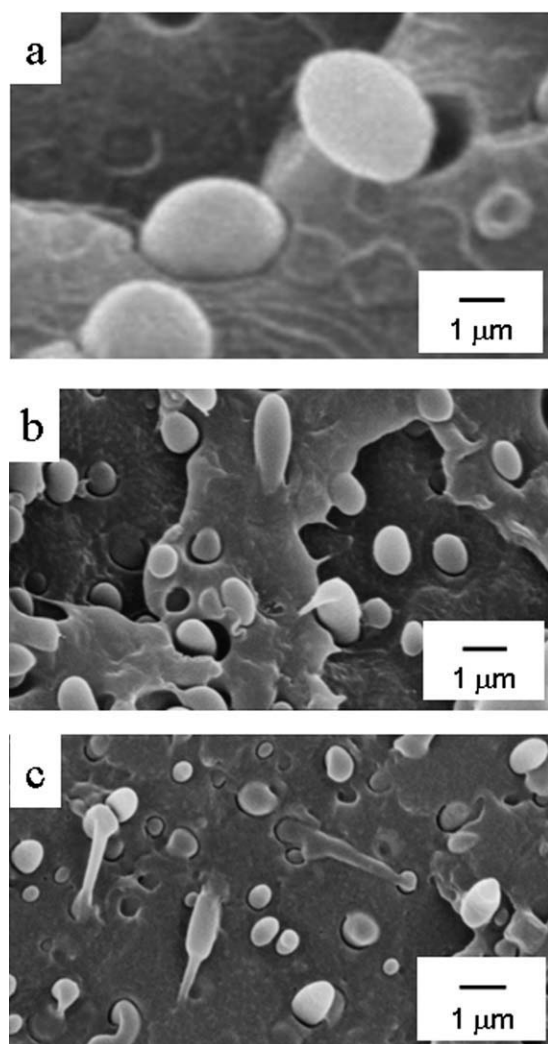


Figure 6 SEM micrographs depicting the size and appearance of PP particles in L-95/5 blends at various compatibilizer contents: (a) L-95/5/0; (b) L-95/5/3; and (c) L-95/5/5.

blends could be found in the literature, although the toughening mechanisms vary depending on the dispersed phase material.^{12–14} In this case, the toughening of the blend is possible due to the prevention of PP particle coalescence with the presence of compatibilizers. Kormendy et al.¹³ have observed enhanced elongation at break in PP/PET (PET being the dispersed phase) blend fibers when using low PET molecular weight.

Dispersed phase particle size and distribution in RPET: Effect of PP molecular weight

The SEM micrographs depicting the appearance and sizes of the PP dispersed phase particles at various compatibilizer contents for L-95/5 specimens are compared in Figure 6. The PP particle size distribution was obtained from these photographs through

image analysis, and the results are depicted in Figure 7 for 95/5 blends and Figure 8 for 90/10 blends. In general, it could be observed that the PP particle size distribution is extremely broad without the presence of compatibilizers in the blends. Large PP particles of more than $3 \mu\text{m}^2$ in area could be observed especially in the uncompatibilized 90/10 blends, as depicted in Figure 8(a–c).

With the incorporation of compatibilizers into the blends, the distribution drastically shifted toward the left, indicating a significant reduction in PP particle size. The extent of shifting was observed to be dependent on the amount of compatibilizers, PP content, as well PP M_w . It is not surprising to find that blends with higher PP contents would require larger amounts of compatibilizers to obtain a more homogeneous blend with finer particle sizes. However, the content of compatibilizers has little effect on the particle size distribution. Instead, it was observed that a highly homogeneous blend with excellent dispersion of small PP particles ($<0.5 \mu\text{m}^2$) could be obtained when low M_w PP was present in the blend, regardless of compatibilizer and PP contents. This enhanced homogeneity of the blends suggests that the effectiveness of the compatibilizers could be enhanced by just altering the molecular weight of the dispersed phase.

Theoretically, the lower melt viscosity that is characteristic of low M_w PP would result in a decrease in viscosity ratio, $\eta_r = \eta_d/\eta_m$, and the capillary number, Ca , both of which contribute toward finer PP droplet size^{15,16}:

$$Ca \equiv \dot{\gamma}\eta_m D / (2\Gamma) \quad (2)$$

where $\dot{\gamma}$ is the shear rate, η_m is the matrix phase viscosity, η_d is the disperse phase viscosity, D is the diameter of the dispersed phase particle, and Γ is the interfacial tension. Wu¹⁷ has given a correlation between capillary number and viscosity ratio in polymer blends that have undergone both steady state shear and elongational shear flows:

$$D = \frac{4\Gamma\eta_r^{\pm 0.84}}{\dot{\gamma}\eta_m} \quad (3)$$

where the plus (+) sign in the exponent is applicable if $\eta_r > 1$, whereas the minus (–) sign applies for $\eta_r < 1$. The tendency for particle coalescence of low M_w PP is diminished because of lower difference in viscosity between the PP phase and RPET matrix. The more active interaction between the compatibilizer and low M_w PP would also prevent particle coalescence through steric stabilization.^{18–21} Although the interfacial tension (Γ) values for the materials were not measured in this study, it is expected that the interfacial tension between RPET and PP would

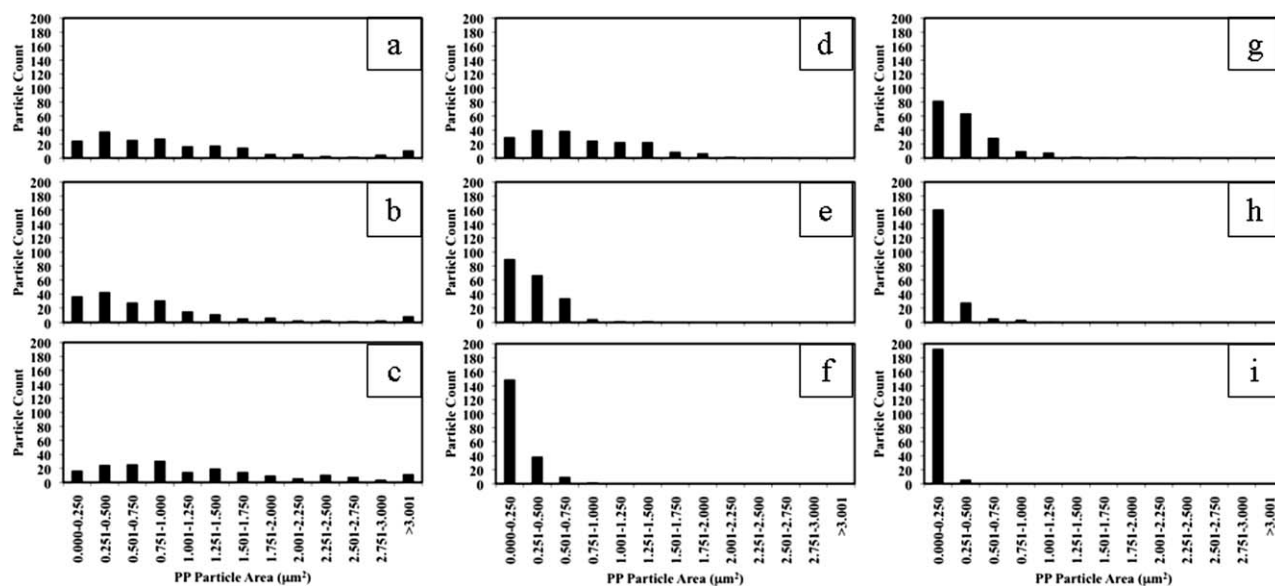


Figure 7 Dispersed phase (PP) particle distribution in RPET/PP (95/5) blends at various compatibilizer contents and PP M_w : (a) H-95/5/0; (b) M-95/5/0; (c) L-95/5/0; (d) H-95/5/3; (e) M-95/5/3; (f) L-95/5/3; (g) H-95/5/5; (h) M-95/5/5; and (i) L-95/5/5.

decrease with decreasing M_w , as has been shown in several other blends in the literature.^{22–24}

Obvious differences in homogeneity of the blends incorporated with various PP grades could be seen in Figure 9. Blends with low M_w PP were substantially more homogeneous regardless of PP and compatibilizer contents. However, with increasing PP M_w , the consistency of the PP phase distribution is compromised, whereby large discrepancies in polydispersity could be observed especially when the blends contain higher PP loadings. One possible

reason that causes inhomogeneity in the blends is that the dispersed phase particles of high M_w PP would require significantly more energy to break up because of higher intermolecular entanglements, thus large particles remain in the blend even after high-shear compounding. Interparticle collision happens much easier when large particles are present, and the probability for coalescence is also higher.²⁵ While the presence of compatibilizers would prevent particle coalescence, it is not an effective tool for promoting particle breakup. Therefore,

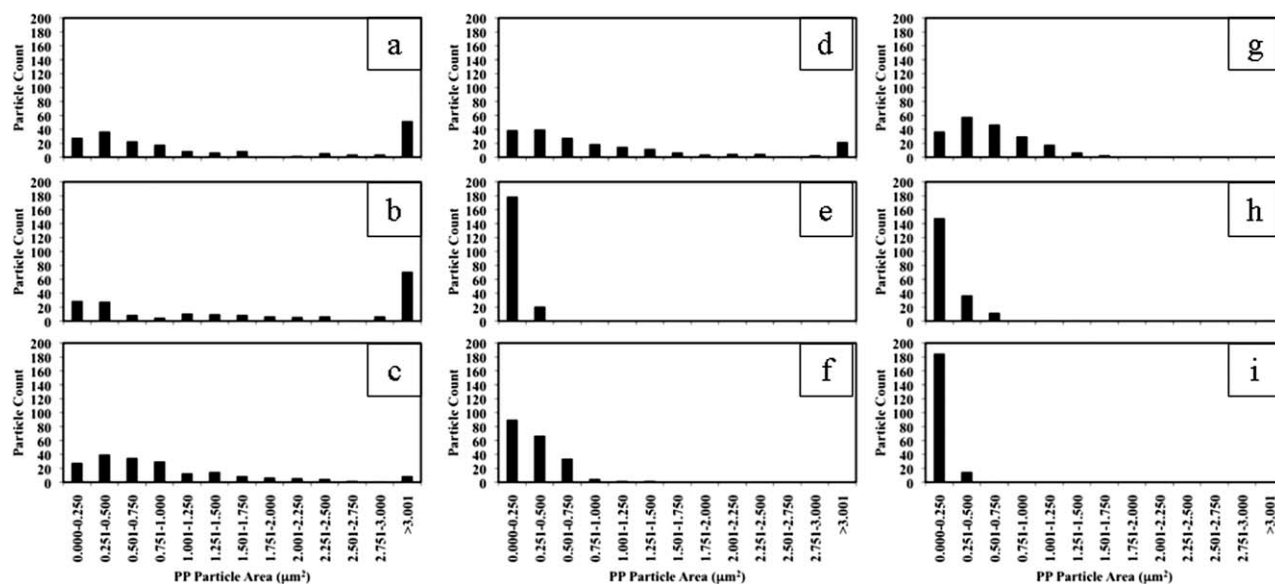


Figure 8 Dispersed phase (PP) particle distribution in RPET/PP (90/10) blends at various compatibilizer contents and PP M_w : (a) H-90/10/0; (b) M-90/10/0; (c) L-90/10/0; (d) H-90/10/3; (e) M-90/10/3; (f) L-90/10/3; (g) H-90/10/5; (h) M-90/10/5; and (i) L-90/10/5.

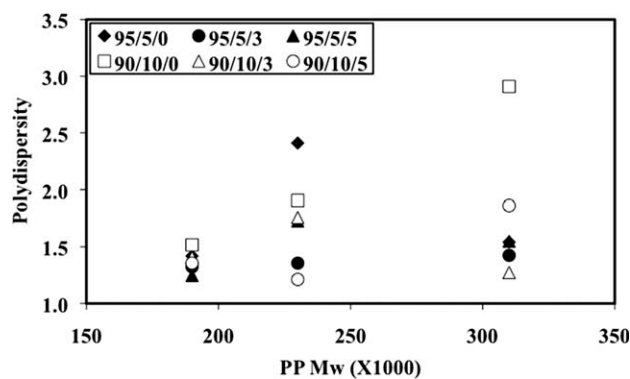


Figure 9 Dependence of polydispersity of the dispersed phase particles on PP M_w in RPET/PP blends at various PP and compatibilizer contents.

uncompatibilized blends would exhibit large and inhomogeneous distribution of dispersed phase particles because of the massive coalescence, as could be seen in Figures 10 and 11.

Heino et al.²⁶ have suggested that SEBS copolymers have an affinity toward the PP phase. Therefore, it is thought that the compatibilizers are able to diffuse toward the interface more easily during blending when low M_w PP is present, thus reducing interfacial tension and preventing particle coalescence. On the other hand, it would be more difficult for the compatibilizers to diffuse through high M_w PP toward the interface although the compatibilizers are thermodynamically more stable at the interface.^{27–31} Lepers et al.,¹⁴ in their study on PET/PP blend films, were able to confirm the saturation of compatibilizers at the interfacial regions. Compatibilizers that are able to diffuse toward the interface would surround the dispersed phase particles to form a core-shell like structure whereby the compatibilizers would constitute as the shell to prevent direct interaction and coalescence between the dispersed phase (core) particles. A lower M_w dispersed phase would promote the formation of a thicker

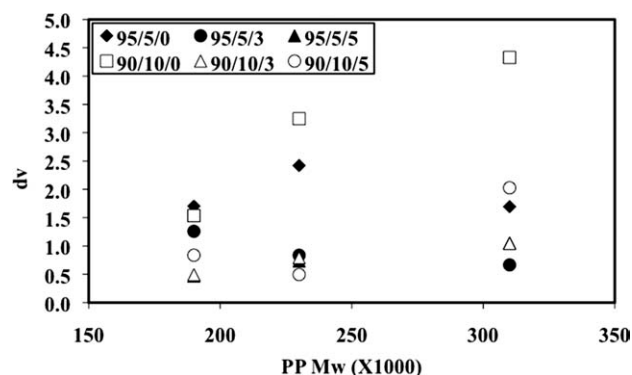


Figure 10 Dependence of volume average PP particle diameter, d_v , on PP M_w in RPET/PP blends at various PP and compatibilizer contents.

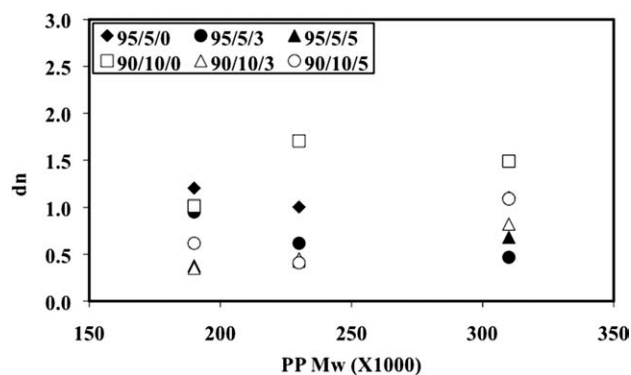


Figure 11 Dependence of number average PP particle diameter, d_n , on PP M_w in RPET/PP blends at various PP and compatibilizer contents.

shell that is more effective in preventing particle coalescence through elastic recoil should the particles come into contact with each other. In other words, this layer of compatibilizers or “interphase” has to be overcome in order for the cores of particles to be exposed to each other before any particle coalescence is possible. This result is important since it suggests that the incorporation of recycled PP, which is usually of lower M_w , will be able to provide better homogeneity and enhance the mechanical properties of the RPET/PP blends.

CONCLUSIONS

The contributions by the PP M_w and compatibilizer content toward improving the homogeneity and mechanical properties of RPET/PP blends were evaluated. This is important since an excellent dispersion of small droplets throughout the matrix would enable the blend to effectively resist crack propagation and fracture. The two main factors that are thought to be essential for obtaining a highly homogeneous blend are the deformation of the dispersed phase and prevention of coalescence during blending. A dispersed phase of low M_w PP would require less energy to cause particle deformation and break up as opposed to that of high M_w PP. Furthermore, effective diffusion of the compatibilizer through low M_w PP ensures high saturation of the compatibilizer at the PP particle surface that prevents particle coalescence.

References

- Imai, Y.; Nishimura, S.; Abe, E.; Tateyama, H.; Abiko, A.; Yamauchi, A.; Aoyama, T.; Taguchi, H. *Chem Mater* 2002, 14, 477.
- Lin, C. C. *Macromol Symp* 1998, 135, 129.
- Gupta, V. B.; Bashir, Z. *Handbook of Thermoplastic Polyester*, Wiley-VCH: Weinheim, Germany, 2002; Vol. 1, p 317.
- The Council for PET bottle Recycling. Available at: <http://www.petbottle-rec.gr.jp/english/index.html>. Accessed: September 2007.

5. Awaja, F.; Pavel, D. *Eur Polym Mater* 2005, 41, 1453.
6. Inoya, H.; Klinklai, W.; Takai, Y.; Leong, Y. W.; Hamada, H. *J Appl Polym Sci*. Accepted on 31 August, 2009.
7. Marino, P.; Lorenze, R.; Donatella, C.; Andrzej, G. *Macromol Chem Phys* 2002, 203, 1473.
8. George, S.; Neelakantan, N. R.; Varughese, K. T.; Thomas, S. *J Polym Sci Part B Polym Phys* 1997, 35, 2309.
9. Willis, J. M.; Favis, B. D. *Polym Eng Sci* 1990, 30, 1073.
10. Lepers, J. C.; Favis, B. D.; Tabar, R. J. *J Polym Sci Polym Phys* 1997, 35, 2271.
11. Cigana, P.; Favis, B. D. *Polymer* 1998, 39, 3373.
12. Maciel, A.; Salas, V.; Manero, O. *Adv Polym Tech* 2005, 24, 241.
13. Kormendy, E.; Marcincin, A.; Hricova, M.; Kovacic, V. *Fibres Textil Eastern Eur* 2005, 13, 20.
14. Lepers, J.-C.; Favis, B. D.; Kent, S. L. *Polymer* 2000, 41, 1937.
15. Taylor, G. I. *Proc R Soc London* 1932, A138, 41.
16. Taylor, G. I. *Proc R Soc London* 1934, A146, 501.
17. Wu, S. *Polym Eng Sci* 1987, 27, 335.
18. Chesters, A. K.; Hofman, G. *Appl Sci Res* 1982, 38, 353.
19. Chesters, A. K. *Chem Eng Res Des* 1991, 69, 259.
20. Abid, S.; Chesters, A. K. *Znt J Multiphase Flow* 1994, 20, 613.
21. Molau, G. E. *J Polym Sci Part A* 1965, 3, 4235.
22. Kamal, M. R.; Lai-Fook, R.; Demarquette, N. R. *Polym Eng Sci* 1994, 34, 1834.
23. Arashiro, E. Y.; Demarquette, N. R. *J Appl Polym Sci* 1999, 74, 2423.
24. Bailey, A. I.; Salem, B. K.; Walsh, D. J.; Zeytountsain, A. *Colloid Polym Sci* 1979, 257, 948.
25. Sundararaj, U.; Macosko, C. W. *Macromolecules* 1995, 28, 2647.
26. Heino, M.; Kirjava, J.; Hietaoja, P.; Seppala, J. *J Appl Polym Sci* 1997, 65, 241.
27. Leibler, L. *Makromol Chem Macromol Symp* 1988, 16, 1.
28. Vilgis, T. A.; Noolandi, J. *Macromolecules* 1990, 23, 2941.
29. Noolandi, J. *Makromol Chem Rapid Commun* 1991, 12, 517.
30. Wang, Z. G.; Safran, S. A. *J Phys Fr* 1990, 51, 185.
31. Dan, N.; Tirrell, M. *Macromolecules* 1993, 26, 637.