

Compatibilization of Recycled Poly(ethylene terephthalate) and Polypropylene Blends: Effect of Compatibilization on Blend Toughness, Dispersion of Minor Phase, and Thermal Stability

Hiroyuki Inoya,^{1,2} Yew Wei Leong,¹ Warunee Klinklai,³ Yoshihiro Takai,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 28 November 2008; accepted 31 August 2009

DOI 10.1002/app.34385

Published online 15 December 2011 in Wiley Online Library (wileyonlinelibrary.com).

ABSTRACT: Blending of recycled polyethylene terephthalate (RPET) from waste bottles with polypropylene (PP) was performed in an attempt to enhance the processability of RPET. The idea of blending RPET with PP sprouted from the intention of recycling PET bottles together with their PP-based caps. Therefore, preliminary blending of RPET with neat PP (RPET/PP) was performed at various PP and compatibilizer contents. Morphological analyses on the extruded pellets of uncompatibilized blends indicate that the PP particle size and state of dispersion at skin and core regions were vastly different. The particles at the skin were at least 10 times smaller than that at the core although the size distribution was very wide. With the incorporation of just 5 phr of compatibilizer, the particles at the core region

became significantly smaller and appeared to emulate that of the skin region. Furthermore, the overall homogeneity of the blends was vastly improved irrespective of PP content in the blend. The reduction in particle size and improved homogeneity inherently reduced stress concentration points and enhanced the mechanical performance of the blends. More importantly, the incorporation of PP into RPET significantly increased the degradation temperature of the blends, provided the dispersion of PP phase in RPET was excellent. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 124: 5260–5269, 2012

Keywords: polyethylene terephthalate; polypropylene; recycling; polymer blends; compatibilization

INTRODUCTION

Public awareness of environmental and energy problems from plastics wastes has posed a challenge to many research groups to find ways to convert plastic waste into useful materials and product instead of ending up in landfills. Polyethylene terephthalate (PET), which is recognized to be one of the most versatile engineering plastics that are mainly used for textile and soft-drink bottles,^{1–3} is also one of the most discarded packaging materials. The usage of PET bottles, that is, mineral water, soy sauce, edible oil, and all soft drinks, has been increasing year by year in Japan, which contributed to a total about 554,104 tons of waste PET bottles that were collected for recycling in 2006.⁴ The recycling of PET lies not only serves as a partial solution to the solid waste problem such as landfill shortages, but also contrib-

utes to the conservation of raw petrochemical substance and energy. Typically, a recycled PET costs ~10% less than virgin PET while 50–60% in energy savings can be expected when RPET is used instead of virgin PET resin to make a similar product.⁵

In previous works, some efforts were devoted to investigate the possibilities in manufacturing products such as household equipments, structural parts of vehicles, containers, bottles, raw materials for polyurethane, composite materials, strapping, cloths, and fiberfill for pillow from RPET through injection, extrusion, and thermoforming processes.⁶ However, the amount of RPET usage is still very minimal compared to the quantity of discarded PET (mainly from soft drink bottles) because of a difficulty in collecting and sorting of postconsumer PET, especially when it comes to separating the PET bottle from the caps and labels before recycling. Thus, it is necessary to establish an efficient and cost-effective recovery method that can convert postconsumer PET bottles (including the caps) into useful products in packaging, commodity, and engineering filed. To do this, we recognized the importance to blend RPET with PP, because most caps are made from the latter material.

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

Contract grant sponsors: Venture Laboratory, Kyoto Institute of Technology, Japan.

Journal of Applied Polymer Science, Vol. 124, 5260–5269 (2012)
© 2011 Wiley Periodicals, Inc.

The recycling of PET has been carried out in four main approaches, that is, primary, mechanical, chemical, and energy recycling.⁷⁻¹⁰ Mechanical recycling of postconsumer PET by blending it with other plastics under suitable conditions is recognized to be an effective means to formulate new materials quickly and efficiently from the existing ones. However, most polymer pairs are immiscible and results in inhomogeneous materials. In particular, PET and PP are incompatible due to differences in chemical structure, whereby the former is known to be a polar polymer while the latter is nonpolar. Therefore, their blends would exhibit a two-phase morphology, whereby the dispersed phase (usually PP) 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. Typically, the strength and stiffness of PET blends are proportional to the PET content, but owing to the phase separation, the blend exhibits very poor impact strength and elongation at break. Therefore, one possible method to obtain an effective blend is to gain control of the phase morphology and tailor the interfacial properties to improve their compatibility. It is well known that the incorporation of copolymers or compatibilizers is effective in enhancing the properties of immiscible blends. It has been suggested that the compatibilizers would concentrate at the interfacial regions and modify them during melt blending through the reduction of interfacial tension and prevention of coalescence, which would result in the reduction in the dispersed phase particle size and improved interfacial adhesion and mechanical performance.^{11,12} Hence, optimum compatibilization is needed to enhance the compatibility between these two immiscible phases to overcome the brittleness of PET.

In the present work, blending of RPET with various contents of PP and compatibilizer was performed to obtain the optimum blend formulation with the best possible mechanical performance. In particular, the effect of compatibilization on the morphological characteristics was elucidated. Because the processing temperatures usually employed for PET and PP are very different, the thermal degradation characteristics of the individual components and the blends were also evaluated to confirm the viability of blending.

EXPERIMENTAL

Materials

The recycled polyethylene terephthalate (RPET) used in this study was obtained in the form of flakes from

TABLE I
Specimen Designation and Izod Impact Strength of RPET/PP Blends at Various PP and Compatibilizer Contents

| Sample | Composition | | | Izod impact strength | |
|---------|-------------|-----|----------------------|----------------------|---------|
| | RPET | PP | Compatibilizer (phr) | Un-Notch | Notched |
| 100/0 | 100 | 0 | 0 | NB ^a | 1.395 |
| 95/5 | 95 | 5 | 0 | 36.992 | 1.692 |
| 90/10 | 90 | 10 | 0 | 32.780 | 1.933 |
| 70/30 | 70 | 30 | 0 | 18.188 | 2.299 |
| 0/100 | 0 | 100 | 0 | 52.068 | 1.006 |
| 95/5/1 | 95 | 5 | 1 | 100.809 | 1.882 |
| 95/5/3 | 5 | 5 | 3 | NB | 1.766 |
| 95/5/5 | 5 | 5 | 5 | NB | 1.611 |
| 95/5/7 | 5 | 5 | 7 | NB | 1.689 |
| 90/10/1 | 90 | 10 | 1 | 42.880 | 1.590 |
| 90/10/3 | 90 | 10 | 3 | NB | 1.629 |
| 90/10/5 | 90 | 10 | 5 | NB | 1.507 |
| 90/10/7 | 90 | 10 | 7 | NB | 1.669 |

^aNB, No Break.

crushed postconsumer PET bottles courtesy of Yasuda Sangyo Co., Ltd., Japan. The flakes were pre-washed in water to minimize impurities before usage.

Because the caps were separated from the bottles and have all been discarded even before the bottles were crushed, the source of polypropylene (PP) from the caps were not available. Therefore, instead of recycled PP, neat PP J900GP (MI = 13, $M_w = 2.3 \times 10^5$) obtained from Idemitsu Petrochemical Co., Japan, was incorporated into RPET. Styrene-ethylene-butadiene-styrene-based terpolymer was used as a compatibilizer at loadings of between 1 and 7 phr. The RPET/PP ratio was varied at 95/5 and 90/10 for compatibilized blends while uncompatibilized blends of 100/0, 0/100, 95/5, 90/10, and 70/30 were prepared for comparison purposes. Specimen compositions are listed in Table I. Only RPET/PP blend compositions of 95/5 and 90/10 (after referred to as 95/5 and 90/10 blends) were chosen 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).

Before blending, RPET was dried in a dehumidifying dryer for 5 h at 120°C. The RPET and PP were dry-blended before 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 dried again by using a dehumidifying drier (Piccolo, Itswa Co., Ltd., Japan) for at least 5 h at 80°C. Dumbbell specimens were prepared by using a 50-ton injection-molding machine (UM50, PO YUEN (TO's) Machine FTY Ltd., Hong Kong). The mold used in this study had a dumbbell-shaped

cavity that was 175 mm in length and the mid-section was 10-mm wide and 3 mm thick. The gauge length was 115 mm. The maximum barrel temperature, mold temperature, and injection speed was set at 285°C, 30°C, and 100 mm/s, respectively.

Morphological characterizations

Morphological observations were performed on the extruded pellets of RPET/PP/compatibilizer blends by using a scanning electron microscope (SEM), JSM-5200 (JEOL Ltd., Japan). The cross sections of cryogenically fractured pellets were coated with a thin layer of gold before observation. From the high-resolution SEM images taken at high magnification, the size (area) of the PP particles near the surface (skin) and core of the pellets was measured by using an image analysis software (Image J). From these measurements, the number average diameter, d_n , and volume average diameter, d_v , for at least 200 dispersed phase particles in each material composition were calculated through the following equation:

$$d_n = \frac{\sum N_i d_i}{\sum N_i} \quad (1)$$

where N_i is number of particles having a diameter d_i . Because RPET and PP are immiscible, the resulting dispersed phase particles are mostly spherical in shape. Therefore, calculation of the volume average diameter, d_v , is more representative of the particle size in three-dimensional space and shall be reported in this study.

$$d_v = \frac{\sum N_i d_i^3}{\sum N_i d_i^3} \quad (2)$$

Tensile fractured surfaces of injection-molded samples were also subjected to microscopic examination, and the particle size distribution was found to resemble that of the pellets.

Mechanical testing

Tensile tests were performed on the dumbbell test specimens at a cross head speed of 50 mm/min 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 unnotched specimens. The samples with dimensions of 10 × 6 × 3 mm were cut and notched from dumbbell specimens. Notches were 2 mm deep and angled at 45°. At least five samples were tested, and their results were averaged.

Flexural test was conducted with specimens cut from the dumbbell specimens by using a similar Instron universal testing machine at a cross head speed of 3 mm/min. Span length was set at 48 mm.

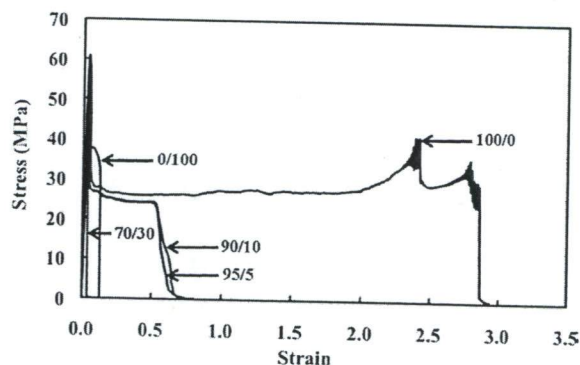


Figure 1 Stress-strain diagrams of uncompatibilized RPET/PP blend compositions at varying PP contents.

Thermogravimetry analysis

Because the components in the blend are commonly processed at very different temperatures, the thermal degradation characteristics of the individual components as well as the blends were evaluated by using a TA Instrument TGA2950 high-resolution-modulated thermogravimetric analyzer. About 10 mg of RPET, PP, compatibilizer, and their blends in pellet form were heated from room temperature to 600°C by using a high-resolution temperature ramp feature. By using this feature, the heating rate is automatically adjusted according to the rate of sample degradation, so that very sharp derivative peaks can be obtained to accurately pinpoint the onset degradation temperatures. All samples were analyzed in dry air.

RESULTS AND DISCUSSION

Mechanical properties of RPET/PP blends

Effect of PP content

The RPET monotonic specimens were extremely ductile and able to deform more than three times their original lengths, as can be seen from Figure 1. Furthermore, at high extensions, an increase in stress was apparent due to strain hardening, which is characteristic of PET. The strain recorded for neat PP was, however, quite low due to the severe degradation when exposed to the high-barrel temperature during the injection-molding process. When PP was incorporated into RPET, the primary effect observed was the significant deterioration in deformability, as shown in Figure 1. Even during the compounding and extrusion process, blends with high-PP content, that is, 30 wt % PP, would tend to lump and extruded profile often broke easily before pelletizing due to the weak melt tension and posed a tremendous challenge during processing. The significant alteration in mechanical properties due to the incorporation of PP is not only limited to deformability

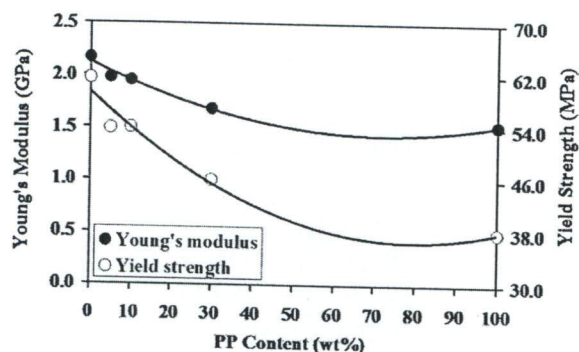


Figure 2 Effect of PP content on the Young's modulus and yield strengths of RPET/PP blends.

but also the stiffness and yield strength of the blend, as could be observed from Figure 2. A gradual decrease in both the Young's modulus and yield strength could be seen while the necking of specimens was observed to be more rapid with increasing PP loading. The RPET/PP (70/30) specimens were even more brittle than neat PP and fractured with little necking, which indicate that a very unstable morphology has developed in this blend. The tendency for phase separation between PP and RPET due to their immiscibility allows for the coalescence of the PP phase to form large anisotropic particles that could act as stress concentration regions, thus deteriorating structural performance.

Effect of compatibilizer content

The effects of compatibilizers on the mechanical performance of the blends containing 5 and 10 wt % of PP are clearly illustrated in Figures 3–6. Because of the elastomeric properties of the compatibilizer, the stiffness of the blends would generally deteriorate with increasing compatibilizer content, as depicted in Figure 3. This deterioration is more obvious in 90/10 blends, which suggests insufficient compatibilization between RPET and PP phases. The effects of compatibilizer addition on the yield strength are

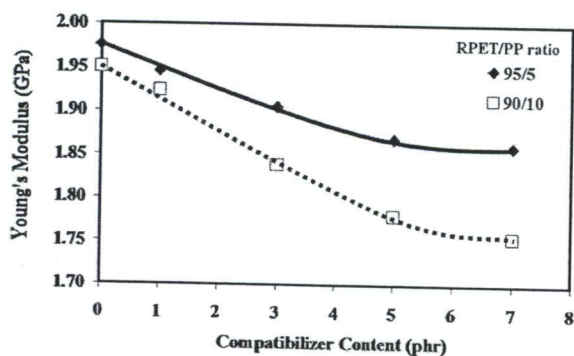


Figure 3 Effect of compatibilizer content on the Young's modulus of 95/5 and 90/10 blends.

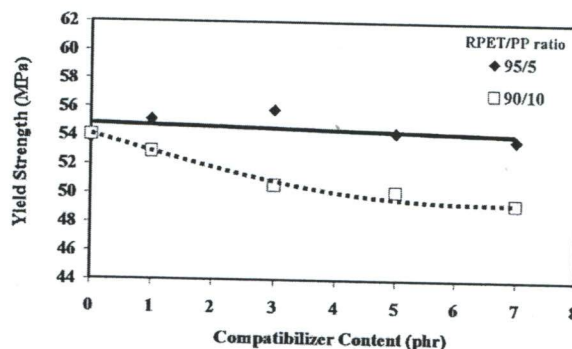


Figure 4 Effect of compatibilizer content on the yield strengths of 95/5 and 90/10 blends.

also apparent especially when the blends contain more PP, as shown in Figure 4. The decreasing yield strength could be an indication of weakening adhesion between the RPET and PP phases.

A strain of more than 3.6 ± 0.5 could be achieved with the incorporation of just 3 phr of compatibilizer when compared with a strain of just 0.7 ± 0.2 for uncompatibilized specimens, as depicted in Figure 5 for 95/5 blends. The amount of PP phase in the RPET/PP blends could also affect the effectiveness of the compatibilizers, as noted when comparing Figures 5 and 6. The 95/5 blends were able to maintain higher stiffness even with the presence of high compatibilizer contents. Another interesting observation from Figure 5 was that most of the compatibilized 95/5 blend specimens exhibited strain hardening, as indicated by the increasing stresses at high strains before failure while similar deformation characteristics could not be found in Figure 6 for 90/10 blends. Strain hardening in RPET is induced by stretching and orientation of the molecular networks, and the lack of strain hardening in 90/10 blends suggest that the matrix ruptured even before any significant molecular orientation could occur. This could be due to the presence of a higher volume of PP particles in the blend, which obstructed the mobility and therefore prevented the alignment of RPET molecules.

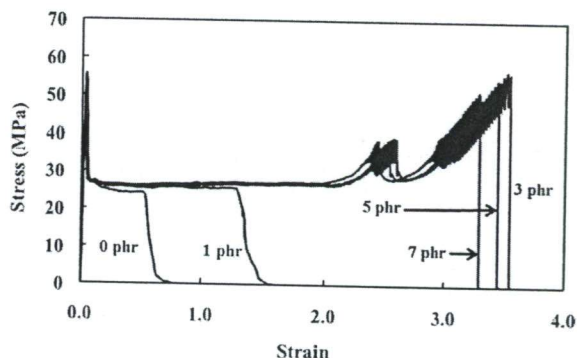


Figure 5 Effect of compatibilizer content on the appearance of the stress-strain curves of 95/5 blends.

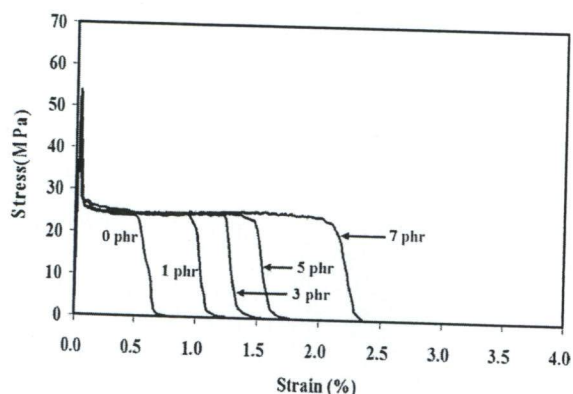


Figure 6 Effect of compatibilizer content on the appearance of the stress-strain curves of 90/10 blends.

The effect of compatibilizer on the notched and unnotched impact strength of RPET/PP blends was also investigated. As shown in Table I, the unnotched impact strength of the RPET/PP blends with 95/5 and 90/10 weight ratios increased about 2.7 times and 1.3 times, respectively, with the incorporation of only 1 phr compatibilizer. Furthermore, no fracture upon impact (NB) was observed when samples were incorporated with at least 3 phr of compatibilizer, which implies that the compatibilizer effectively improves the overall toughness of the blends. Nevertheless, the incorporation of compatibilizers was found to have no profound influence on the notched impact strength of the specimens, which underscores the notch sensitiveness of PET as reported elsewhere.¹³

PP particle size and distribution in RPET

Effect of PP content

The pellet fracture surface morphologies of uncompatibilized RPET/PP blends at various PP contents are shown in Figure 7. The blend exhibits a typical morphology of an immiscible blend with poor adhesion between the matrix and dispersed phases. The PP phase appeared as spherical particles of varying sizes, which is an indication of a very unstable morphology that will nonetheless affect the performance of the blends. Particle size of the dispersed phase was mainly dependent upon PP content; that is, particle size increased with increasing PP content from 5 to 30 wt %. The PP particles appear to be loosely bound to the matrix resin, and cavities left behind by pulled-out particles were abound. Furthermore, the interparticle distance appeared to be narrower when the blends contain high-PP contents, which could promote particle coalescence should these pellets be used for further processing such as injection molding.

Effect of compatibilizer content

It should be noted that for uncompatibilized blends, the PP particles at the core of the pellets appeared to be larger by an order of magnitude than those at the skin, as can be observed in Figure 8. Because the skin regions were the first to solidify during the extrusion process and considering that the shear stresses near the die wall is relatively low, the size of the PP particles in the skin region is representative of the initial

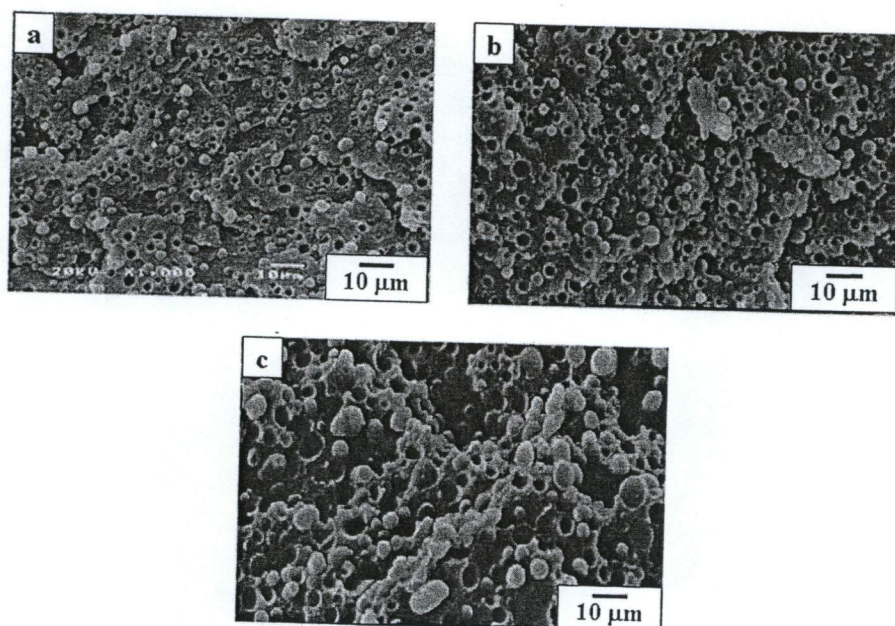


Figure 7 Comparison of SEM photographs showing the size of the dispersed PP phase in RPET/PP blends when the PP content was (a) 5; (b) 10; and (c) 30 wt %.

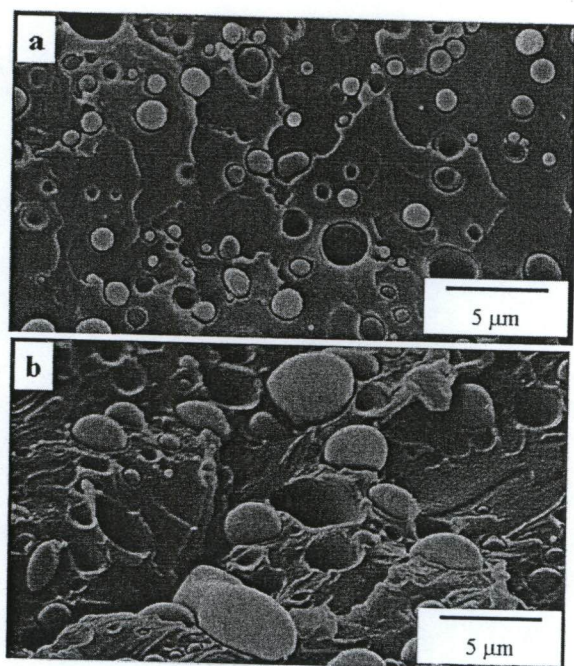


Figure 8 SEM photographs showing a vast difference in PP dispersed phase size at the (a) skin and (b) core regions of the uncompatibilized 95/5 blend pellets.

state of dispersion in the barrel of the extruder. The existence of larger particles at the core therefore suggests that coalescence of the dispersed phase have occurred due to the lower cooling rate at this region. This coalescence of dispersed phase particles is promoted by the high interfacial tension between RPET and PP phases. Therefore, a suitable compatibilizer is needed to improve the state of dispersion of PP in RPET.

With the introduction of compatibilizers, however, the difference in PP particle size between the skin and core regions was very small, and an overall improvement in homogeneity could be noted in Figure 9. It is thought that the compatibilizer was effective in promoting fragmentation of the PP particles by reducing interfacial tension while at the same time preventing recoalescence of the fragmented particles. It should be expected that the variation in mechanical properties of RPET/PP blends come from structural changes of the dispersed PP phase in RPET.

Figure 10 depicts the morphological characteristics of RPET/PP (95/5) blends upon the incorporation of compatibilizers at varying concentrations. Significant changes to the size and distribution of the PP phase could be observed as the content of compatibilizer was increased from 1 to 7 phr. The smaller dispersed phase particle and cavity sizes could have made the crack propagation path more complicated and promoted crazing during tensile and impact loading.

The PP size distributions at both the skin and core regions of the pellets for the 95/5 and 90/10 blends are shown in Figures 11 and 12, respectively. Meanwhile, the volume average particle diameter, d_v , that was calculated from eq. (2) is plotted in Figure 13. In the uncompatibilized blend, a broad distribution of large PP particles can be observed in both the skin and core regions due to the instability and coalescence of the PP phase during melt mixing as stated previously. However, the number of large particles was far more prominent in the core region when compared with the skin irrespective of PP content in the blend, as could be observed in both Figures 11 and 12. When 1 phr of compatibilizer was added to the 95/5 blends, the PP particle size distribution became significantly narrower while the d_v values experienced a significant reduction of about 50% at the skin and 22% at the core. By increasing the compatibilizer content up to 7 phr, the particle size distribution at the skin became much narrower although the d_v values remained similar. This is an indication that the particles at the skin have already attained the smallest possible size with a minimum d_v value of about 0.65 μm . Meanwhile, the particle size at the core regions gradually decreased toward a similar minimum d_v value at the skin with increasing compatibilizer content. At compatibilizer contents of 5 phr or higher, very similar d_v values were recorded at the skin and core regions. This indicates that at the optimum compatibilizer content of 5 phr, a stabilized

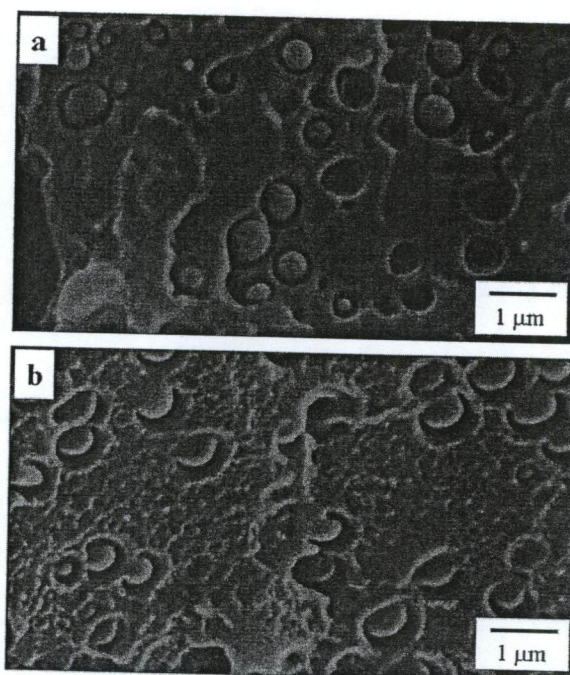


Figure 9 SEM photographs showing very similar PP dispersed phase size at the (a) skin and (b) core regions of the compatibilized 95/5/7 blend pellets.

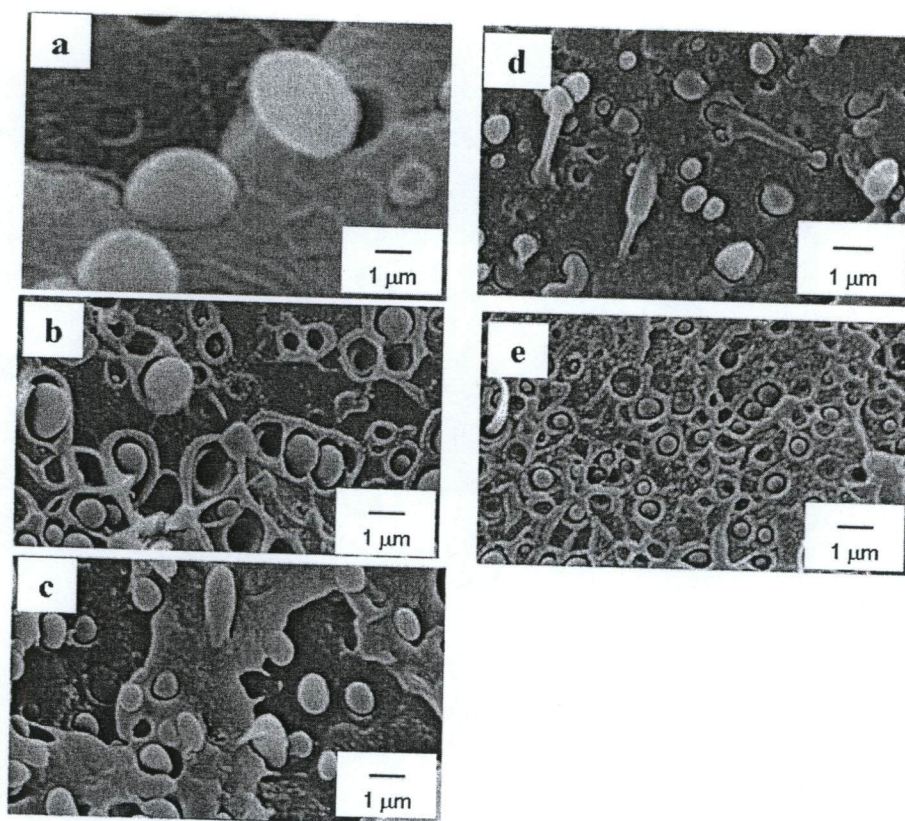


Figure 10 SEM photographs showing PP dispersed phase size in 95/5 blends at various compatibilizer contents: (a) 0 phr; (b) 1 phr; (c) 3 phr; (d) 5 phr; (e) 7 phr.

morphology was achieved where the coalescence of PP phase at the core regions of the moldings was effectively suppressed. This led to an overall reduction in PP particle size and improved homogeneity,

which are critical for obtaining good mechanical performance. Similar compatibilization effects have also been reported elsewhere for other polymer blend systems.¹⁴⁻¹⁸

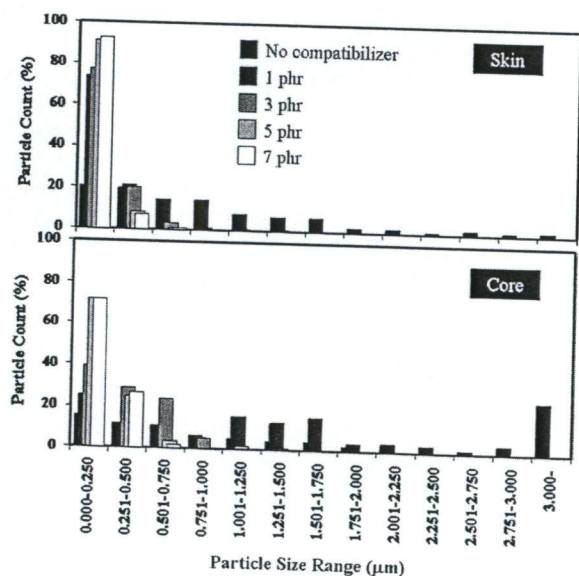


Figure 11 Effect of compatibilizer content on particle size distribution determined from SEM micrographs of 95/5 blends at the skin and core regions of pellets.

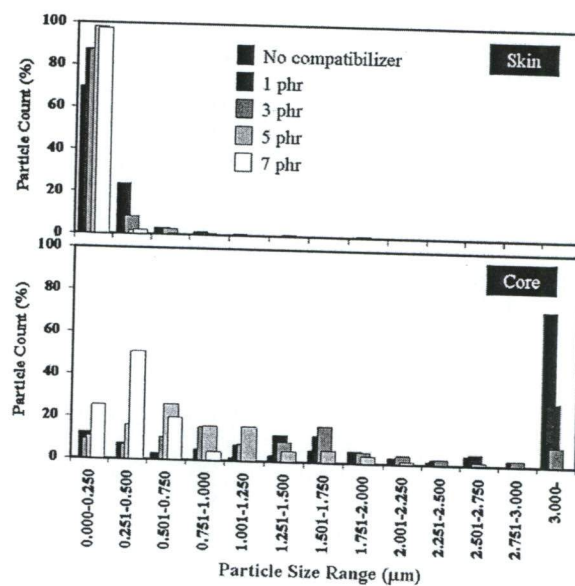


Figure 12 Effect of compatibilizer content on particle size distribution determined from SEM micrographs of 90/10 blends at the skin and core regions of pellets.

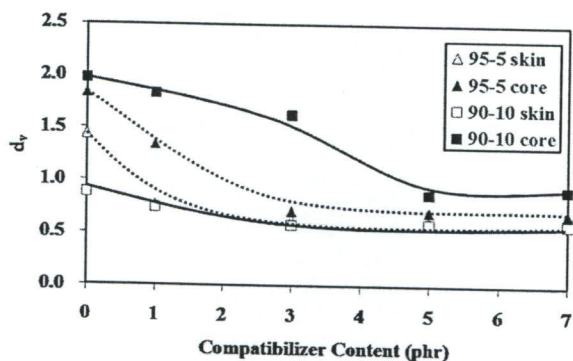


Figure 13 Volume average PP particle diameter (d_v) of 95/5 and 90/10 blends at various compatibilizer contents.

The PP particle size polydispersity (P_d), which quantitatively measures the broadness of particle size distribution, was calculated by obtaining the ratio between d_v and d_n . The P_d values at the skin and core of the pellets at various compatibilizer contents are given in Figure 14. Generally, high- P_d values could be observed at both the skin and core regions when the blends were uncompatibilized, which indicates very broad particle distribution. The particles at the core region of 90/10 blends were mostly larger than 3 μm in diameter and therefore were not included into the predetermined particle size range, as can be seen from Figure 12 presented previously. Hence, the low- P_d value recorded at the core region of 90/10 blends is not statistically viable and does not represent a narrow size distribution. With increasing compatibilizer content especially at 5 phr and above, however, the P_d values significantly decreased to approach unity at the skin and core regions for both the 95/5 and 90/10 blends, thus suggesting very narrow and homogeneous particle size distribution. It is therefore confirmed that at the optimum compatibilizer content of 5 phr, a stable morphology of the RPET/PP blends could be obtained, which would enhance reproducibility in terms of mechanical performance of the blends.

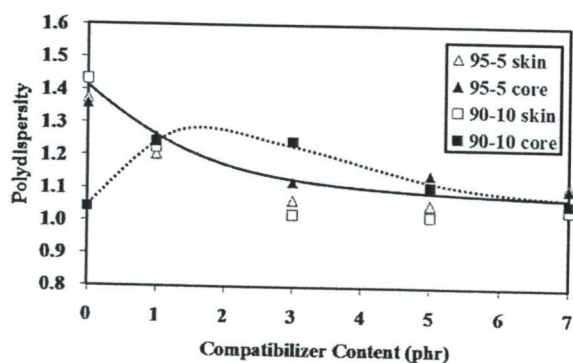


Figure 14 PP particle size polydispersity (P_d) of 95/5 and 90/10 blends at various compatibilizer contents.

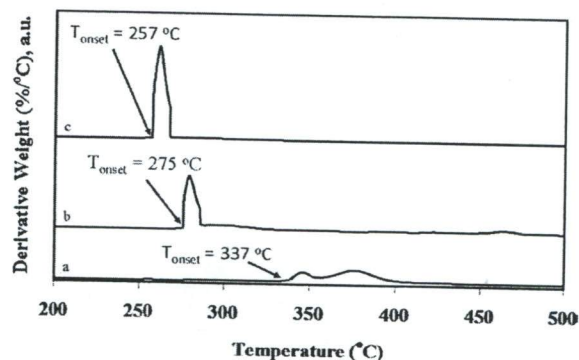


Figure 15 Derivative weight loss peaks of monotonic components showing the onset degradation temperatures (T_{onset}) during thermogravimetric analysis in air: (a) RPET; (b) compatibilizer; (c) neat PP.

Thermal stability of RPET, PP, compatibilizer, and their blends

From thermogravimetric analysis (TGA) results depicted in Figure 15, the onset degradation temperature (T_{onset}) of RPET pellets in air was recorded at 340°C , and the broad derivative peaks indicate a gradual degradation of RPET, which is mainly due to hydrolysis. Therefore, RPET is not expected to degrade at the designated processing temperature of 285°C during injection molding as well as compounding. However, the respective T_{onset} recorded for neat PP and compatibilizers were 257 and 275°C . The sharp derivative peaks following T_{onset} for these components suggest that both materials would degrade at a very high rate mainly due to oxidative reactions. Hence, these components would be extremely unstable and volatile if they were to be processed individually at 285°C .

However, when these components were heated under nitrogen atmosphere, the T_{onset} for all components increased significantly, as can be seen from

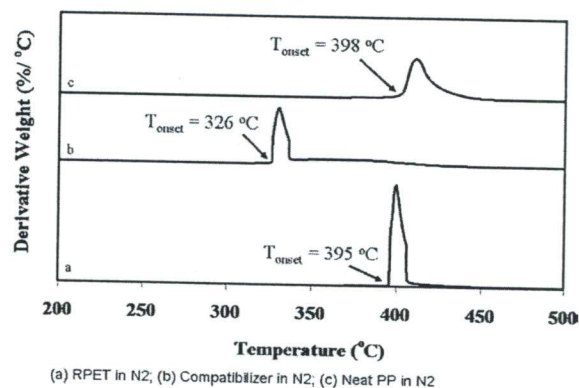
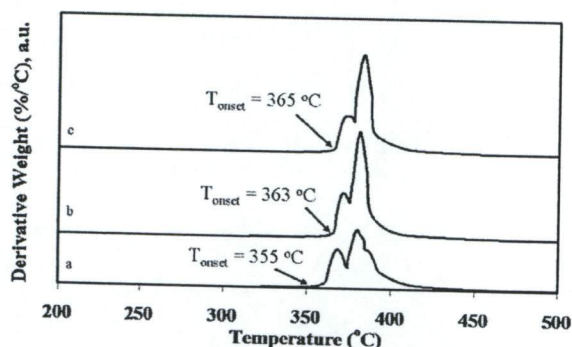


Figure 16 Derivative weight loss peaks of monotonic components showing the onset degradation temperatures (T_{onset}) during thermogravimetric analysis in N_2 atmosphere: (a) RPET; (b) compatibilizer; (c) neat PP.

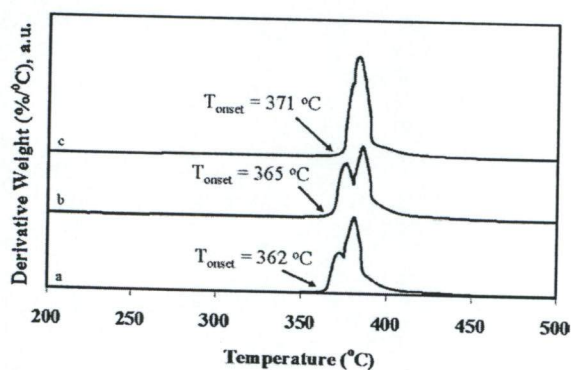


RPET/PP=95/5; Compatibilizer content: (a) 0 phr; (b) 3 phr; (c) 5 phr

Figure 17 Effect of compatibilizer content in 95/5 blends on the T_{onset} during thermogravimetric analysis in air. Compatibilizer content = (a) 0 phr; (b) 3 phr; (c) 5 phr.

Figure 16. The most notable improvement in thermal degradation resistance was in PP where a 140°C increment from a T_{onset} of 258°C could be observed. Even the T_{onset} of the compatibilizer was increased to 326°C from 275°C with the absence of oxygen. This is a strong indication that both PP and compatibilizer are extremely sensitive to oxidative degradation, but they can nevertheless be processed at high temperatures should their exposure to oxygen be limited.

Interestingly, when TGA was performed on RPET/PP/compatibilizer blends in air, the T_{onset} of PP and compatibilizer was absent, as could be observed from Figure 17. Furthermore, the T_{onset} of the blends recorded at above 355°C was even higher than that of monotonic RPET. It is also interesting to note in Figure 18 that higher PP content in the blend could also result in an increase in T_{onset} of the blends despite the apparent immiscibility and phase separation between PP and RPET. The apparent increment in T_{onset} of the blends is attributed to the excellent barrier properties of RPET, which limited the exposure of PP and compatibilizer to oxygen and therefore



RPET/PP=90/10; Compatibilizer content: (a) 0 phr; (b) 3 phr; (c) 5 phr

Figure 18 Effect of compatibilizer content in 90/10 blends on the T_{onset} during thermogravimetric analysis in air. Compatibilizer content = (a) 0 phr; (b) 3 phr; (c) 5 phr.

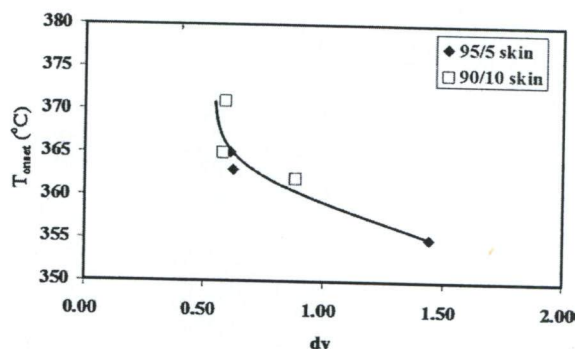


Figure 19 Influence of PP particle size (d_v) on T_{onset} of both 95/5 and 90/10 blend pellets.

preventing extensive degradation of these phases. The dispersed phases, on the other hand, were able to enhance the overall thermal degradation resistance of the blend, provided they are homogeneously dispersed.

The effects of compatibilization on the thermal degradation properties could also be seen from Figures 17 and 18. The incorporation of compatibilizers into the blends resulted in further shifting of the T_{onset} toward higher temperatures, although a high degradation rate was noted from the sharp derivative peaks once degradation has begun. When the compatibilizer content was gradually increased to 5 phr, the degradation initiated at a higher temperature, regardless of PP content in the blends. A particularly interesting observation from Figure 18 was that the derivative twin peaks, characteristic of RPET during degradation, appeared to be consolidated into a single peak at 5 phr compatibilizer content. The onset temperature at 371°C was 6°C higher than that recorded for blends with 3 phr compatibilizer, although the peak temperature of the former was similar. This could be due to the excellent dispersion and improved homogeneity of the PP phase in the blend with the presence of higher compatibilizer concentrations.

The effect of PP particle size (d_v) on the T_{onset} was elucidated and presented in Figure 19. Because the onset of degradation would most likely occur from the surface of the pellets, the d_v values at the skin region of the pellets are presented. From the results, it could be deduced that smaller PP particle size at the skin region was effective in suppressing thermal degradation of the blends. Therefore, it should be noted that good thermal stability of the blends can be achieved, provided excellent homogeneity and small PP particle size (preferably in the submicron range) can be obtained in the pellets.

CONCLUSIONS

In this investigation, the PP minor phase particle dispersion was found to be vastly different at the skin and core regions of the pellets as well as injection-

molded specimens, which is largely due to the variation in cooling rates as well as shear stresses between these regions. The PP particle size and homogeneity of the RPET/PP blends could be modified by the use of compatibilizers, and this would profoundly influence the mechanical properties as well as thermal stability of the blends. At an optimum compatibilizer content of 5 phr, the PP particle size in the blends would be mostly in the submicron range while excellent overall homogeneity could be achieved. This level of particle dispersion and homogeneity was the key toward obtaining blends with excellent mechanical performance and enhanced thermal resistance. This information will be very useful for the PET waste bottle recycling industry, because it provides the possibility of recycling the bottles even without first separating the PP caps and yet able to obtain blends with enhanced properties.

References

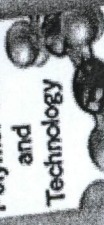
1. Imai, Y.; Nishimura, S.; Abe, E.; Tateyama, H.; Abiko, A.; Yamauchi, A.; Aoyama, T.; Taguchi, H. *Chem Mater* 2002, 14, 477.
2. Lin, C. C. *Macromol Symp* 1998, 135, 129.
3. Gupta, V. B.; Bashir, Z. In *Handbook of Thermoplastic Poly Esters*, Vol.1; Fakirov, S., Ed.; Wiley-VCH: Weinheim, Germany, 2002; p 317-388.
4. The Council for PET bottle Recycling, Available from <http://www.petbottle-rec.gr.jp/english/index.html>, date of access September 2007.
5. Awaja, F.; Pavel, D. *Eur Polym J* 2005, 41, 1453.
6. Birch, S.; Yamaguchi, J.; Demmler, A.; Jost, K. *Automot Eng* 1993, 101, 109.
7. Avila, A. F.; Duarte, M. V. *Polym Degrad Stab* 2003, 80, 373.
8. Daniel, P.; Tadeusz, S. *Ind Eng Chem Res* 1997, 36, 1373.
9. Scheirs, J. *Polymer Recycling*; Wiley: New York, 1998; Chapter 4.
10. Shunkla, S. R.; Kulkarni, K. S. *J Appl Polym Sci* 2002, 85, 1765.
11. Creton, C.; Brown, H. R.; Deline, V. R. *Macromolecules* 1994, 27, 1774.
12. Matos, M.; Farvis, B. D. *Polymer* 1995, 36, 3899.
13. Pawlak, A.; Morawiec, J.; Pazzagli, F.; Pracella, M.; Galeski, A. *J Appl Polym Sci* 2002, 86, 1473.
14. Marino, P.; Lorenze, R.; Donatella, C.; Andrzej, G. *Macromol Chem Phys* 2002, 203, 1473.
15. George, S.; Neelakantan, N. R.; Varughese, K. T.; Thomas, S. *J Polym Sci Part B: Polym Phys* 1997, 35, 2309.
16. Willis, J. M.; Favis, B. D. *Polym Eng Sci* 1990, 30, 1073.
17. Lepers, J. C.; Favis, B. D.; Tabar, R. J. *J Polym Sci Polym Phys* 1997, 35, 2271.
18. Cigana, P.; Favis, B. D. *Polymer* 1998, 39, 3373.

The Essential Resource in Polymer Science...
 Click for further information

ENCYCLOPEDIA OF Polymer Science and Technology

Now Available Online!

ENCYCLOPEDIA OF Polymer Science and Technology
 NEW AND UPDATED ARTICLES



Journal of Applied Polymer Science

Copyright © 2012 Wiley Periodicals, Inc., A Wiley Company



15 June 2012

Volume 124, Issue 6

Pages 4409–5292

1. [Miscibility and crystallization behavior of biodegradable poly\(\$\epsilon\$ -caprolactone\)/tannic acid blends \(pages 4409–4415\)](#)

Yonggang Liang, Fang Yang and Zhaobin Qiu

Article first published online: 1 DEC 2011 | DOI: 10.1002/app.35430

- [Abstract](#)
- [Full Article \(HTML\)](#)

- [Request Permissions](#)
- 103. [Compatibilization of recycled poly\(ethylene terephthalate\) and polypropylene blends: Effect of compatibilization on blend toughness, dispersion of minor phase, and thermal stability \(pages 5260–5269\)](#)

Hiroyuki Inoya, Yew Wei Leong, Waranee Klinklai, Yoshihiro Takai and Hiroyuki Hamada

Article first published online: 15 DEC 2011 | DOI: 10.1002/app.34385

- [Abstract](#)
- [Full Article \(HTML\)](#)
- [PDF\(1557K\)](#)
- [References](#)
- [Request Permissions](#)

- 104. [Characterizations on the amidized multiwalled carbon nanotubes grafted with polyaniline via *in situ* polymerization \(pages 5270–5278\)](#)

Yi-Jhen Wu, Liang Chao, Ko-Shan Ho, Ying-Jie Huang, Yu-Lan Huang, Chi-Shiang Yang and Bo-Hao Tseng

Article first published online: 15 DEC 2011 | DOI: 10.1002/app.34607

- [Abstract](#)
- [Full Article \(HTML\)](#)
- [PDF\(1453K\)](#)
- [References](#)
- [Request Permissions](#)

- 105. [Engineering properties of compatibilized polypropylene/liquid crystalline polymer blends \(pages 5279–5285\)](#)

P. K. Mandal, S. K. Siddhanta and D. Chakraborty

Article first published online: 15 DEC 2011 | DOI: 10.1002/app.34277

- [Abstract](#)
- [Full Article \(HTML\)](#)
- [PDF\(1077K\)](#)
- [References](#)
- [Request Permissions](#)

- 106. [Surface organic modification of titanium dioxide fine particles through photo polymerization \(pages 5286–5292\)](#)