

Compatibilization by Homopolymer: Significant Improvements in the Modulus and Tensile Strength of PPC/PMMA Blends by the Addition of a Small Amount of PVAc

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ABSTRACT Poly(propylene carbonate) (PPC), a polymer produced from CO₂, has been melt-mixed with 30 wt % poly(methyl methacrylate) (PMMA) with the aim of enhancing the physical properties of PPC for practical use but keeping a relatively high CO₂ fixing rate in the compound. The observation of a coarse phase structure with a large PMMA domain size and a large size distribution in the blend indicates the immiscibility between PPC and PMMA. The addition of a small amount of poly(vinyl acetate) (PVAc) not only shifts the glass transition temperatures (T_g 's) of both PPC and PMMA markedly but also significantly increases the modulus and tensile strength of the blend. The prepared compound with 5 per hundred parts of resin PVAc shows a 26 times higher elastic modulus and an approximately 3.8 times higher tensile strength than pure PPC at room temperature. The morphological investigation indicates that the incorporation to PVAc not only induces the finer dispersion of PMMA in the PPC matrix but also results in the phase transformation from a sea-island to a co-continuous structure.

KEYWORDS: poly(propylene carbonate) • polymer blending • compatibilizer • interfacial tension • co-continuous structure • mechanical properties

INTRODUCTION

A synthetic scheme in which polymers are produced from CO₂ was first reported 40 years ago by Inoue et al. (1, 2). Recently, the synthetic strategy and synthesized polymers have received renewed attention as a technology for fixing and utilizing CO₂. The use of CO₂ as one of the monomers in the preparation of polymers can not only reduce the dependence on petroleum but also contribute to reducing the massive emission of CO₂, which has been considered to be the main factor causing the greenhouse effect in the world. Poly(propylene carbonate) (PPC) has attracted the most attention among all polymers from CO₂ owing to its relatively low cost and well-established polymerization method. However, PPC as an amorphous polymer suffers many drawbacks, such as a low tensile strength and a very low heat resistance. Therefore, the produced PPC is still far from practical use. Some efforts have been made to use PPC as a plasticizer/additive for rubber, thermoplastics, or surfactants (3–6). Investigations have also been carried out to modify the physical properties of PPC by melt compounding. Most of the studies focused on PPC blends with other biodegradable polymers to fabricate completely biodegradable materials (7–14), such as poly(L-lactide) (7, 8), poly(butylene succinate) (9, 10), and poly(hydroxybutyrate) (11, 12). The nanocomposites based

on PPC and organoclay have also been reported (15–17). However, there is very limited improvement for the investigated compounds using PPC as the dominant component. On the other hand, few works focused on PPC blends with a nonbiodegradable polymer (18) or low-molecular-weight compound (19). Poly(methyl methacrylate) (PMMA) shows a high glass transition temperature and excellent mechanical properties, especially a very high elastic modulus and a very high tensile strength. It is therefore considered that PMMA can be used to improve both the mechanical and thermal properties of PPC.

Compatibilization strategies for immiscible polymer blends have extensively been investigated over the past 4 decades. The most general method involves the use of block or graft copolymers, whose one block is identical or miscible with one component and whose other block is identical or miscible with the other component. Reactive blending by the in situ formation of a graft copolymer is also an effective technique of fabricating compatibilized polymer blends. In both cases, the compatibilizer should be located at the interface between the constituent components. Very limited success was achieved for strategies using a homopolymer or a random copolymer as the compatibilizer for immiscible polymer blends (20–24). Jerome et al. reported the finer morphology and improved physical properties of a polycarbonate (PC)/poly(vinylidene fluoride) (PVDF) blend, which were obtained by the incorporation of PMMA (20, 21). As they indicated, the compatibilization effect of PMMA was obtained by the migration of PMMA from the PVDF phase to the interface between PVDF and PC with increasing

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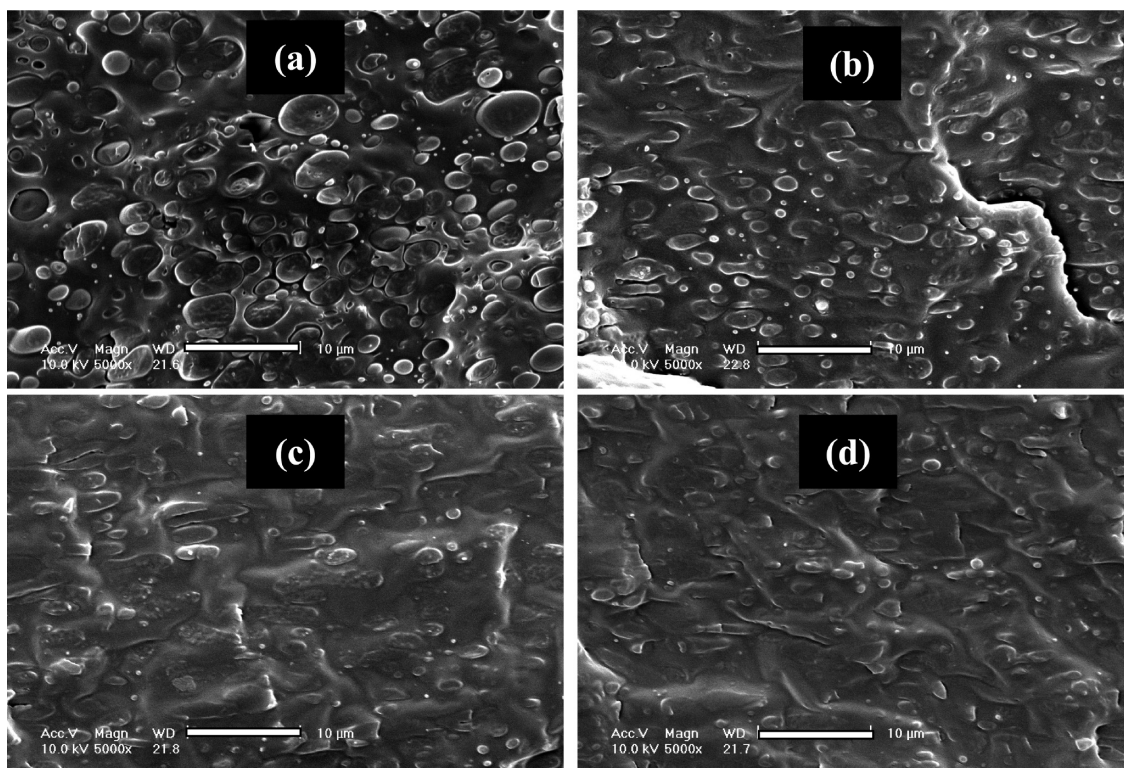


FIGURE 1. SEM images of (a) PPC/PMMA = 70/30, (b) PPC/PMMA = 70/30 with 2.5 phr PVAc, (c) PPC/PMMA = 70/30 with 5 phr PVAc, and (d) PPC/PMMA = 70/30 with 10 phr PVAc.

PMMA loading content. In other words, PMMA is exclusively located in the PVDF phase and forms a homogeneous PVDF/PMMA phase because it has a higher affinity with PVDF than with PC. With a large amount of PMMA added (more than 20 wt %), a small interfacial tension can be achieved between the PVDF/PMMA and PC phases. Therefore, PMMA is not the real compatibilizer for the PVDF/PC blend.

In this work, the morphology and properties of PPC/PMMA blends were studied by the incorporation of a small amount of a homopolymer, poly(vinyl acetate) (PVAc). It was found that PVAc not only enhances the physical and thermal properties of PPC/PMMA blends but also leads to the phase transformation from a sea-island to a co-continuous structure. It was therefore concluded that PVAc is a good compatibilizer for the PPC/PMMA blend.

EXPERIMENTAL SECTION

Materials and Sample Preparation. The PPC sample with a weight-average molecular weight (M_w) of 447 000 was kindly provided by Changchun Institute of Applied Chemistry, Chinese Academy of Sciences. PMMA was purchased from Sumitomo Chemical (Tokyo, Japan) with the trade name SUMIPEX. PVAc with a molecular weight of approximately 260 000 was purchased from Sowa Science Co. (Tokyo, Japan).

The PPC/PMMA = 70/30 blends with various amounts of PVAc were prepared using a Brabender-type plastic mixer (KF70 V; Toyo Seiki Co., Hyogo, Japan) with two rotors at a rotation speed of 100 rpm at 155 °C for 5 min. After blending, all of the samples were then hot pressed at 155 °C for 5 min into films with a thickness of 500 μm , followed by quenching in ice water. The obtained films were used for further characterization.

Characterization. The phase structure of the blends was observed by field-emission scanning electron microscopy (FE-

SEM). A Philips XL-20 scanning electron microscope was used for measurements at an acceleration voltage of 10 kV. All of the samples were fractured by immersion in liquid nitrogen for about 10 min. The fractured surface was then coated with a thin layer of gold before the observation. Transmission electron microscopy (TEM) was performed using a JEM 1230 instrument (JEOL, Tokyo, Japan) operating at an acceleration voltage of 120 kV. The blend sample was ultramicrotomed at -120 °C to obtain a section with a thickness of about 70 nm. The section was then stained with ruthenium tetroxide (RuO_4) for 20 min.

Dynamic mechanical analysis (DMA) was carried out using a Rheovibron DDV-25FP (Orientec Co., Ltd., El Monte, CA) in a tensile mode. The dynamic storage and loss moduli were determined at a frequency of 1 Hz and a heating rate of 3 °C/min as a function of the temperature from -150 to $+100$ °C. Differential scanning calorimetry (DSC) was carried out under a nitrogen flow at a heating or cooling rate of 10 K/min with a Perkin-Elmer DSC-7 differential scanning calorimeter calibrated with the melting temperatures of indium and zinc.

Tensile tests were carried out according to the ASTM D 412-80 test method using the dumbbell-shaped samples punched out from the molded sheets. The tests were performed using a tensile testing machine, Tensilon UMT-300 (Orientec Co., Ltd., El Monte, CA), at a crosshead speed of 10 mm/min at 23 °C and 50% relative humidity.

RESULTS

Morphologies. Phase morphologies of PPC/PMMA blends with various amounts of PVAc are shown in Figure 1. The PPC/PMMA blend without PVAc shows a typical sea-island morphology, where PMMA domains are dispersed in the PPC matrix. The domain size ranges from 0.5 to 4 μm . The observation of coarse structures with large domain sizes and large size distribution indicates that PMMA and PPC are thermodynamically immiscible. However, the PMMA do-

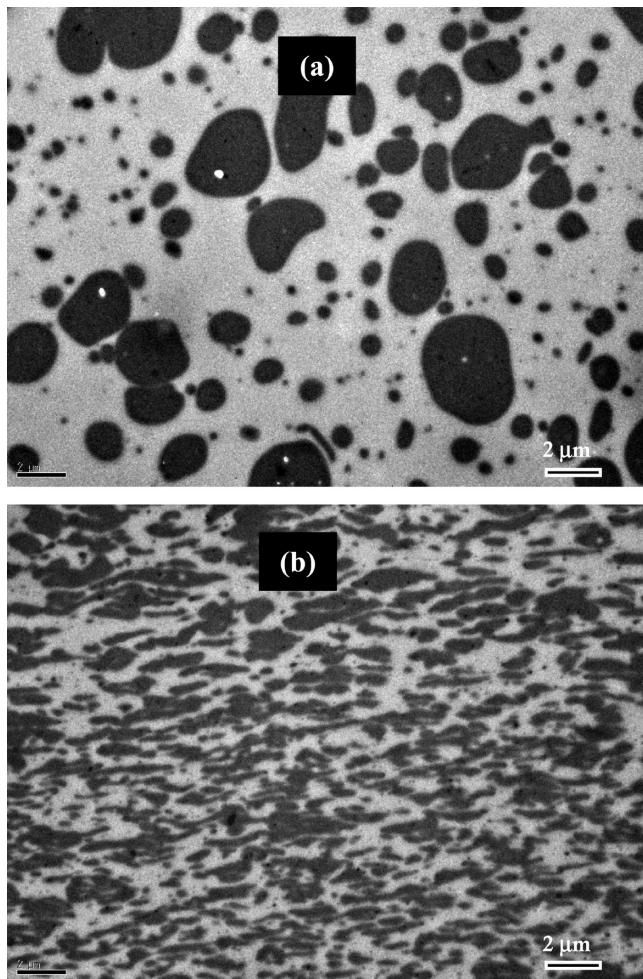


FIGURE 2. TEM images for (a) PPC/PMMA = 70/30 and (b) PPC/PMMA = 70/30 with 5 phr PVAc.

main size is reduced to about 2 μm by the addition of only 2.5 per hundred parts of resin (phr) PVAc. Moreover, the PMMA particles are also uniformly dispersed in the PPC matrix. With a further increase in the PVAc content, the blend morphology shows a significant change. For blends with more than 5 phr PVAc, no discrete spherical particles are observed and PMMA phases tend to connect with each other, indicating the formation of a co-continuous structure.

TEM provides more detailed morphological information for PPC/PMMA blends without and with 5 phr PVAc, as shown in Figure 2. The white part corresponds to the PPC phase, and the black part corresponds to the PMMA phase. It can be observed that the phase size and morphology of the blend observed from the TEM micrographs are in agreement with those obtained from the SEM image. PMMA disperses in the PPC matrix with a size ranging from 0.5 to 3 μm for the sample without PVAc. The particle size distribution is very large, indicating the lack of miscibility between PPC and PMMA. The addition of 5 phr PVAc results in the phase transition from a sea-island to a co-continuous structure. The phase width is estimated to be as small as about 300 nm for the PPC/PMMA blend with 5 phr PVAc.

Thermal Analysis. Figure 3 shows the DSC thermograms for pure PPC, pure PMMA, and PPC/PMMA blends

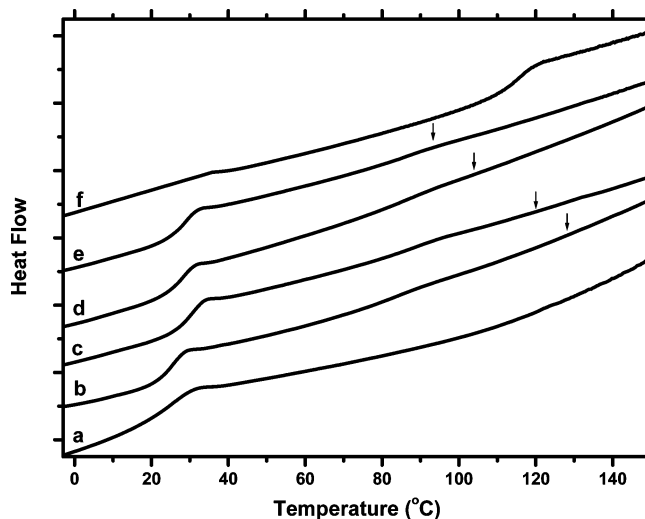


FIGURE 3. DSC thermograms of (a) PPC, (b) PPC/PMMA = 70/30, (c) PPC/PMMA = 70/30 with 2.5 phr PVAc, (d) PPC/PMMA = 70/30 with 5 phr PVAc, (e) PPC/PMMA = 70/30 with 10 phr PVAc, and (f) PMMA.

with various amounts of PVAc. The glass transition temperatures (T_g 's) obtained by DSC of pure PPC and pure PMMA are 24 and 108 $^{\circ}\text{C}$, respectively. The blend without PVAc exhibits two T_g 's of about 25 and 106 $^{\circ}\text{C}$, corresponding to the T_g 's of PPC and PMMA, respectively. However, it is apparent from Figure 3 that the glass transition temperatures of both PPC and PMMA shift toward each other with the addition of PVAc, indicating that PVAc improves the miscibility between PPC and PMMA.

The DMA properties were analyzed for pure PPC and PPC/PMMA blends containing various amounts of PVAc. The storage modulus, loss modulus, and $\tan \delta$ are shown in Figure 4 as a function of the temperature. The storage modulus of pure PPC decreases significantly at about 30 $^{\circ}\text{C}$. PPC becomes very soft when the temperature is above 30 $^{\circ}\text{C}$, and it starts to flow when the temperature is further increased. Therefore, pure PPC does not have any strength when the temperature is higher than 35 $^{\circ}\text{C}$. The PPC/PMMA blend without the addition of PVAc has a higher relaxation peak temperature at about 33 $^{\circ}\text{C}$, indicating an improved heat resistance compared with pure PPC. However, such an improvement is very limited because the storage modulus becomes zero above 54 $^{\circ}\text{C}$. A significant difference for the DMA curve was observed for the PPC/PMMA blends upon the addition of a small amount of PVAc. The relaxation peak temperature of the PPC component significantly increases, and the samples with more than 5 phr PVAc could be self-supported in the high-temperature region. For these samples, they start to flow at temperatures up to about 95 $^{\circ}\text{C}$, which is near the T_g of PMMA. Therefore, the heat resistance of PPC compounds was significantly increased by the incorporation of 30 wt % PMMA and a small amount of PVAc.

Tensile Properties. Figure 5 shows the strain–stress curves for pure PPC and PPC/PMMA blends with various amounts of PVAc measured at 23 $^{\circ}\text{C}$. Pure PPC exhibits rubberlike features including an extremely low elastic modulus and a very low tensile strength but a high elongation at break. The low physical performance characteristics render

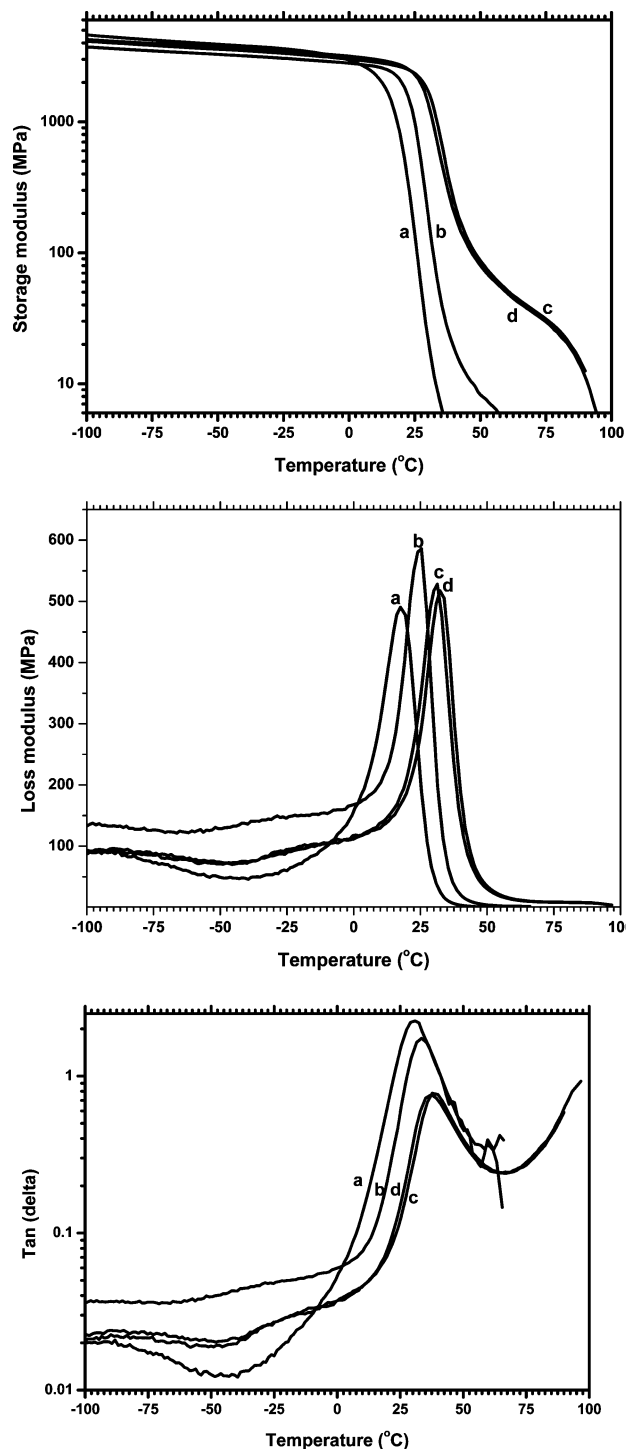


FIGURE 4. Storage modulus, loss modulus, and loss $\tan \delta$ of (a) pure PPC, (b) PPC/PMMA = 70/30, (c) PPC/PMMA = 70/30 with 5 phr PVAc, and (d) PPC/PMMA = 70/30 with 10 phr PVAc.

pure PPC far from practical use without modification. To maintain the high CO_2 fixation rate in the final compound, PPC is used as the dominant component during melt mixing with PMMA. The blend prepared by mixing PPC and PMMA at a weight ratio of 70/30 shows an increased elastic modulus and an increased tensile strength but a low elongation at break, as shown in Figure 5b, which can be attributed to the much higher modulus and tensile strength of PMMA. Note that the elongation at break for the PC/PMMA blend

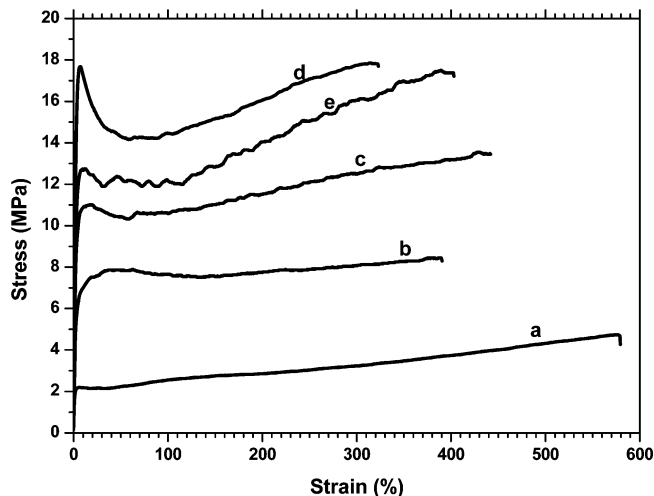


FIGURE 5. Strain–stress curves for (a) PPC, (b) PPC/PMMA = 70/30, (c) PPC/PMMA = 70/30 with 2.5 phr PVAc, (d) PPC/PMMA = 70/30 with 5 phr PVAc, and (e) PPC/PMMA = 70/30 with 10 phr PVAc.

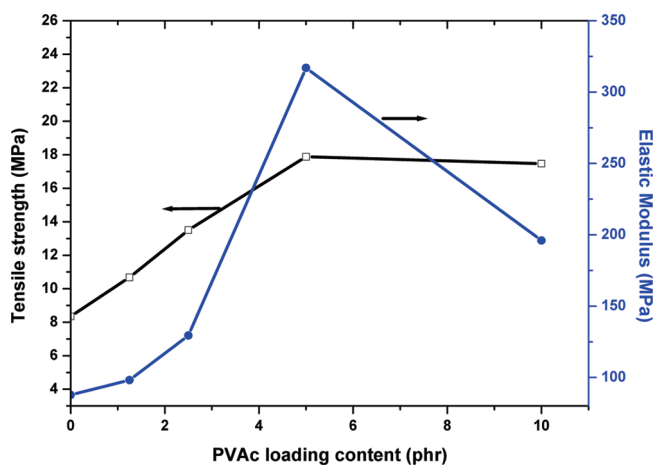


FIGURE 6. Tensile strength and elastic modulus for PPC/PMMA = 70/30 blends with various amounts of PVAc.

still remains as high as about 400%, indicating the compatibility between PPC and PMMA. On the other hand, the addition of a small amount of PVAc in the PPC/PMMA blend markedly changes its tensile behavior upon tensile deformation. Both the elastic modulus and the tensile strength increase significantly with the incorporation of a small amount of PVAc, while the elongation at break is similar to or even longer than that of the blend without PVAc. The elastic modulus and tensile strength of the PPC/PMMA blend with 5 phr PVAc are 317 and 17.9 MPa, while those for pure PPC are 12.1 and 4.8 MPa, respectively. However, it seems that the improvements in the mechanical properties by PVAc saturate at the level of 5 phr addition; thus, excessive PVAc addition deteriorates the physical behavior of the compounds, as shown in Figure 6. The blend with 10 phr PVAc exhibits a lower tensile strength and a lower modulus than that with 5 phr PVAc. The physical properties of the blends with and without PVAc, including tensile strength, elastic modulus, storage modulus, and elongation at break, are shown in Table 1. Those of low-density polyethylene (LDPE) and polypropylene (PP), which are general-purpose plastics, are also listed for comparison. It can be observed that the

Table 1. Basic Mechanical Properties of PPC and PPC/PMMA Blends with and without PVAc

sample	tensile strength (MPa)	elastic modulus ^a (MPa)	storage modulus ^b (MPa)	elongation at break (%)
pure PPC	4.8	12.1	101	578
PPC/PMMA = 70/30	8.4	87.8	1564	390.6
PPC/PMMA = 70/30 + 5 phr PVAc	17.9	317	2431	322
LDPE	16.6	286	142	616
PP	26.1	519	979	1071

^a Calculated from the strain–stress curves. ^b Obtained by DMA measurements.

physical properties of PPC compounds are significantly enhanced by the incorporation of both PVAc and PMMA, and the elastic moduli of the compounds are comparable to those of LDPE and PP.

DISCUSSION

The results of both the thermal analysis and the morphological observation indicate the good compatibilization effects of PVAc for PPC/PMMA blends, as shown by the shifted T_g 's and the decreased domain size. These compatibilization effects can be attributed to the high miscibility between PVAc with PPC and PMMA. PVAc has been reported to be thermodynamically miscible with PMMA (25, 26). For PVAc/PPC blends, only one T_g depending on the composition ratio was observed (see Figure S1 in the Supporting Information), indicating the miscible states for PVAc and PPC in their melt blends. When polymer 3 is added to an immiscible blend of 1 and 2, the location of polymer 3 can be predicted using a spreading coefficient λ_{ij} (27):

$$\lambda_{31} = \gamma_{12} - (\gamma_{31} + \gamma_{23}); \quad \lambda_{13} = \gamma_{23} - (\gamma_{31} + \gamma_{12})$$

where γ_{ij} is the interfacial tension between phases i and j . If $\lambda_{31} > 0$ and $\lambda_{13} < 0$, encapsulation of 1 by 3 will occur. In other words, polymer 3 will locate at the interface between 1 and 2. Although we lack interfacial tension data, the value is definitely positive owing to the immiscibility of the PPC/PMMA blend. The interfacial tensions of PVAc/PMMA and PVAc/PPC are effectively zero because both blends are miscible. According to the above-mentioned spreading theory, PVAc will locate at the interface between PPC and PMMA with thermodynamic drive. The high-magnification TEM image of the PPC/PMMA blend with 5 phr PVAc is shown in Figure 7. A broad interface between the PPC and PMMA phases can be observed, and it is rational to attribute this layer to the PVAc-rich layer. A small amount of PVAc added not only reduces the interfacial tension but also suppresses the PMMA domain coalescence. Therefore, a finer phase structure is observed for the PPC/PMMA blend with 2.5 phr PVAc.

On the other hand, the marked improvements in the mechanical and thermal properties of PPC were achieved by combining the minor PMMA component (30 wt %) and

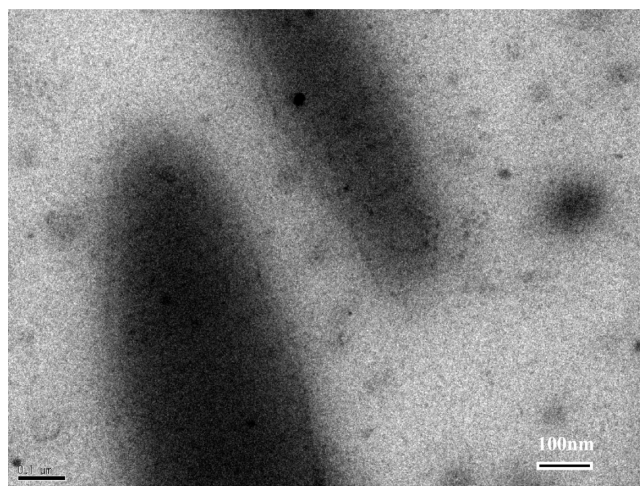


FIGURE 7. High-magnification TEM image for PPC/PMMA = 70/30 with 5 phr PVAc.

a small amount of PVAc, which is attributed to not only the compatibilization effects of PVAc but also the formation of the co-continuous structure. Numerous investigations indicated that the co-continuous structure could provide a better combination of component properties than the sea-island structure (28–31). It is generally accepted that the co-continuous structure of a binary polymer blend can be formed within a composition region near the phase inversion, which is basically governed by the relative melt viscosities of the two components (32, 33). Although the viscosity of PMMA decreases upon incorporation of a small amount of PVAc, the viscosity of PMMA/PVAc is still much higher than that of pure PPC (see Figure S2 in the Supporting Information). Considering the minor component of PMMA in the blend (30 wt %), it is difficult to explain the formation of co-continuous morphology by only variations in the viscosity ratio of PMMA and PPC. On the other hand, the GPC measurement (see Figure S3 in the Supporting Information) indicates that no chemical reaction occurs during the melt mixing. We therefore attributed the formation of the co-continuous structure to the decreased interfacial tension between PPC and PMMA by PVAc. The minimum volume fraction ϕ of the minor component at which co-continuity exists was investigated by Willemse et al. (34–36) and defined as

$$1/\phi = 1.38 + 0.0213 \left(\frac{\eta_m \gamma_p R_0}{\sigma} \right)^{4.2}$$

where η_m is the viscosity of the matrix, γ_p is the shear rate during mixing, σ is the interfacial tension, and R_0 is the equivalent sphere diameter. The equation indicates that a lower interfacial tension shifts the limiting composition to lower values and broadens the co-continuous range. The addition of PVAc improved the miscibility between PPC and PMMA and reduced the interfacial tension. Therefore, a co-continuous PPC/PMMA blend was observed even with only a 30 wt % minor component. At a low temperature below the T_g of PPC, both PMMA and PPC

phases contributed to the mechanical behavior. However, the PMMA phase provided the mechanical properties independently when the temperature was higher than the flow temperature of PPC. Therefore, the co-continuous PPC/PMMA blends exhibited not only improved mechanical properties but also an increased thermal resistance compared with the same blends with the sea-island structure. Note that the elastic modulus and tensile strength of pure PVAc were measured to be approximately 60 and 4.6 MPa, respectively, at 23 °C, indicating that PVAc itself is soft and cannot increase the modulus and strength of the PPC/PMMA blends. Therefore, excessive PVAc addition deteriorated both the mechanical properties and the thermal resistance, as shown in Figure 6.

CONCLUSION

We have succeeded in fabricating a high-performance material based on PPC as a dominant component by the incorporation of PMMA and a small amount of PVAc. The compounds demonstrated not only improved mechanical properties but also increased heat resistance, and they are expected to be a viable alternative to petroleum-based general-purpose plastics, such as polyethylene, PP, and polystyrene. Moreover, it was found that the homopolymer PVAc is a good compatibilizer for the PPC/PMMA blend. Sea-island PPC/PMMA blends transformed into co-continuous structures upon the addition of more than 5 phr PVAc, which accounted for the marked improvements in the physical properties of the blend.

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Supporting Information Available: DMA measurements for PPC/PVAc blends, melt viscosities, GPC curves of PPC, PVAc, and selected blends. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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