



Fabrication of polycarbonate blends with poly(methyl methacrylate-co-phenyl methacrylate) copolymer: Miscibility and scratch resistance properties



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ABSTRACT

Poly(methyl methacrylate-co-phenyl methacrylate) (PMPA) was blended with polycarbonate (PC) to ameliorate the poor scratch resistance of pure PC. The miscibility of the PC/PMPA blends was analyzed by dynamic-mechanical thermal analysis (DMTA), differential scanning calorimetry (DSC) and optical measurements. These results indicated that PC/PMPA blends are miscible at all of the compositions used in this study. Also, quantitative analysis of their miscibility was performed by using classical glass-transition temperature (T_g)-composition models (Fox equation and Gordon-Taylor equation). The PMPA effect on the tensile properties and scratch resistance of PC/PMPA blends were investigated. With increasing PMPA content, the scratch resistance of the blends was found to increase, whereas other tensile properties such as tensile strength and tensile modulus did not change due to the good miscibility between PC and PMPA.

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Introduction

For many years, there has been much interest in the research of polymer blends because they can help to obtain high-performance polymers instead of synthesis of newly designed polymer when the desirous properties of each polymer are united synergistically. In other words, one may achieve useful combinations by blending the materials into a single product. Furthermore, one should infer a continuous range of the properties simply by varying the blend composition. Generally, polymers having high glass-transition temperature and transparency are attractive for industrial polymer engineering due to strong economic benefits that should come from their utilitarian applications.

Polycarbonate (PC) is an amorphous engineering thermoplastic with an excellent combination of properties such as good processability, low density, high transparency, and good strength. Transparent PC has been used to a various industrial applications such as flat-display panels for portable electronic devices, eyewear lenses, compact disks, and safety windows [1]. In spite of its great physical and optical properties, the PC has limited applications because of its poor scratch and abrasion resistance characteristics

[2–5]. Therefore, PC has been modified and tailored in various ways, particularly by blending with other polymers for use in demanding applications [6].

PC/Poly(methyl methacrylate) (PMMA) blends have received many research attention [7–9] because PMMA is also one of the transparent polymers that possesses many usable properties, such as low weight, high transmittance, chemical resistance, resistance to weathering corrosion, scratch resistance, and good insulating properties [10]. Importantly, though PMMA and PC are transparent polymers, their resulting blends show heterogeneous phase morphology having bad interfacial adhesion and therefore, their final properties are poor. It has been demonstrated that various blending strategies using *in-situ* or *out-situ* modification of the blends can increase the compatibility between PC and PMMA.

To enlarge PC's property (durability) window and thereby its application field, PC needs to be scratch resistant. In this study, we tackled the poor scratch resistance problem of PC using PMMA copolymers containing compatible comonomer. We showed that PC blends with this PMMA copolymer could dramatically improve PC's scratch resistance without reducing their tensile properties.

Experimental

Materials. PC (Lexan 143, melt flow index \approx 10.5 g/10 min at 300 °C and 1.2 kg load) was obtained from SABIC. Poly(methyl

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methacrylate-co-phenyl methacrylate) (PMPA) was supplied from Stahnek Co., Ltd., Incheon, Korea. The number-average molecular weight (M_n), weight-average molecular weight (M_w) and polydispersity index (PDI) were 18,800 g/mol, 36,500 g/mol, and 1.95, respectively. The content of phenyl methacrylate monomer in the copolymer was 29 mol%.

Preparation of blends. The melt blending was carried out in a Thermo Haake batch mixer (PolyLab system) at 230 °C for 10 min and a rotor speed was 60 rpm. After melt blending, 1.5 mm thick sheets of the blends were prepared by using a hot-press (Carver laboratory Inc.) at 230 °C for 3 min.

Measurements. The thermal behavior of the phase transitions was studied using a differential scanning calorimetry (DSC; TA DSC Q1000). The blend samples were analyzed under nitrogen gas with 20 °C/min heating rate. Transmittance measurement was conducted by ultraviolet-visible (SHIMAZU, UV-2501PC) spectrometer. The dynamic thermomechanical measurements of the blends were conducted with a Seiko Exstar 6000 under the tension mode. The loss tangent and storage modulus of blends were measured at 1 Hz with a static load at 1% strain and the dynamic load at 0.5% strain. The temperature ramp was 3 °C/min and the scanning range from 0 to 180 °C. Color-difference of the blends were analyzed by SA 4000 color-difference meter (Nippon Denshoku). The tensile properties of the blends were determined using a tensile tester (Lloyd LSK100 K) at room temperature. A crosshead speed was 5 mm/min in these tests. Scratch tests were carried out using a CSM Micro-Scratch Tester (MST) equipped with a conical diamond indenter (50 μm diameter and 120° cone angle). Measurements of Vickers hardness number (VHN) were made with a microhardness indenter machine (Micromet 2100; Buehler, Lake Bluff, IL) at a 100 gr load for 30 s.

Results and discussion

Generally, a single glass-transition temperature (T_g) is a reasonable evidence for homogeneous phase in polymeric blend systems. Thus, thermal behavior measurement was conducted to ascertain the glass transition behavior for PC/PMPA blends. Fig. 1 shows DSC thermograms for PC/PMPA blends with one T_g in all compositions. As shown in Fig. 1, only one T_g for each composition was appeared and this T_g shifted systematically with the blend composition. These results strongly indicate that the PC/PMPA blend was miscible. However, the DSC thermograms obtained in this study showed some T_g broadening in the blend compositions. This phenomenon indicates that there are heterogeneous phases under micro-scale in blend compositions. These micro-sized

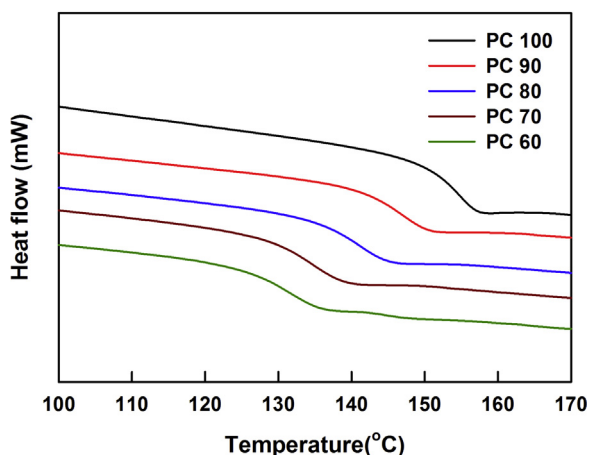


Fig. 1. DSC thermograms of pure PC and PC/PMPA blends.

heterogeneous phases are generally difficult to detect by using DSC instrument. These broadening phenomena can also occur in many miscible blends that show miscibility without any strong specific interactions between them. The little T_g broadening phenomenon observed in this study may suggest that some aggregation under micro-scale should come out in the blends.

Because the T_g -composition relationship for polymer blends can serve another clue to investigate the scale of blend homogeneity, we performed quantitative analysis by classical T_g -composition models for miscible polymer blend systems. Fig. 2 shows the quantitative tendency of T_g variation for the PMPA content in the blend, with the initial T_g for each blend plotted as a function of the PMPA content. Curve fitting of the T_g data using two well-known equations for miscible polymer blend systems was investigated. First of all, curve fitting was compared to the common Fox equation [11] as shown in Fig. 2.

$$\frac{1}{T_g} = \frac{w_1}{T_{g1}} + \frac{w_2}{T_{g2}}$$

As shown in Fig. 2, the observed T_g values were not much deviant from the prediction curve calculated from Fox equation. To evaluate the T_g -composition relationships of the PC/PMPA blends, the Gordon-Taylor equation was used for curve fitting the observed T_g values [12]:

$$T_g = \frac{(w_1 T_{g1} + k w_2 T_{g2})}{(w_1 + k w_2)}$$

Where, w_i is the weight fraction of the i th polymer, and $k = \Delta C_{p2} / C_{p1}$, i.e. the ratio of heat capacity change for the PMPA copolymer ($T_{g1} = 115.6$ °C) to that of PC (high- T_g polymer, $T_{g2} = 155.2$ °C) at T_g . A suitable curve fitting with the Gordon-Taylor equation was calculated and the well-fitted parameter value was obtained to be quite high at about $k = 0.45$. The parameter, k , in the Gordon-Taylor equation is correlated with the intensity of intermolecular interactions or the degree of phase homogeneity in polymer blends [13]. The relatively high k -value obtained in this study indicated that the intermolecular interactions between PC and PMPA were particularly strong and that the blends were homogenous.

Further evidence for the above observation can be drawn from the $\tan \delta$ curves of the PC/PMPA blends. Fig. 3(b) shows loss tangent peaks corresponding to the T_g s of the PC/PMPA blends. These single loss tangent peaks moved to lower temperatures with increasing PMPA content, which was in good agreement with the

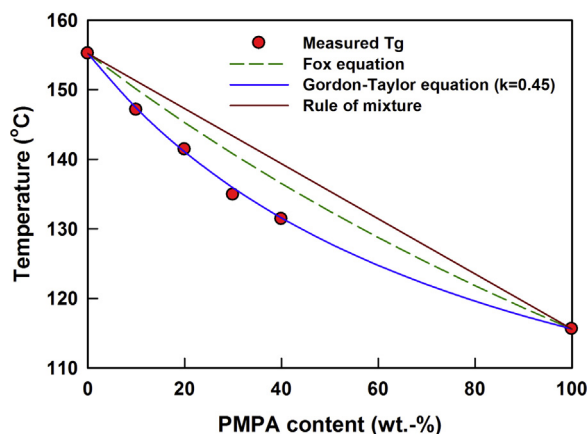


Fig. 2. Plot of T_g versus composition of PC/PMPA blends.

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