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polymer communications

Toughened blend of polycarbonate and epoxidized ethylene propylene diene rubber

Xue-hui Wang[a], Zhi-gang Wang[b,*], Wei Jiang[b], Chang-hai Liu[b], Hai-dong Yang[a], Hui-xuan Zhang[a] and Bing-zheng Jiang[b]
[a] *Department of Chemical Engineering, Jilin Institute of Technology, 130012 Changchun, People's Republic of China*
[b] *Polymer Physics Laboratory, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, 130022 Changchun, People's Republic of China*

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A super-tough polycarbonate (PC) blend was obtained by using epoxidized ethylene propylene diene (eEPDM) rubber as modifier. The notched Izod impact strength of PC/eEPDM (96/4) blend shows a great improvement, with a value about 25 times of that of pure PC. Finely and homogeneously dispersed rubber particles (0.2–0.8 μm) in the PC matrix indicated good adhesion between the eEPDM rubber phase and the PC matrix. © 1997 Elsevier Science Ltd.

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Introduction

As the use of polymers in structural applications increases, the fracture toughness of the polymer may become a decisive factor in material selection. Therefore much work has been done to develop effective toughening methods for polymers. One method which has been successful in toughening brittle or notch sensitive polymers is rubber toughening[1-3].

Owing to the low entropy of mixing, most polymer blend systems are found to be immiscible[4-6]. Such multiphase blends can exhibit poor mechanical properties for their low interfacial adhesion, leading to a dewetting of the phases during mechanical loading. This can result in an early fracture of the blend systems, i.e. at low strain and low stress, which clearly limits their utility[7]. This problem can be resolved by improving the adhesion between the phases, either by the addition of a third component, acting as a compatibilizing agent, or by chemical reaction at the phase boundaries[8-10].

Our recent work involves a novel blend of polycarbonate (PC) and epoxidized ethylene propylene diene rubber (eEPDM). Although many polymer blends have already been studied to improve the toughness of PC, such as PC/MBS/SAN, PC/ABS, PC/core-shell rubber and PC/EPDM (containing 0.2–2wt.% maleic anhydride) blends[11-16], to our knowledge, there are no papers dealing with the PC/eEPDM blend system. In this study, EPDM was first epoxidized with in situ formed performic acid, which induced functional epoxy groups into the EPDM macromolecular backbone[10-17], then blended with PC resin, resulting in a new kind of super-tough PC blend. The notched impact property and the morphology of PC/eEPDM blends were studied.

Experimental details

Material and specimen preparation. The PC (bisphenol A type} used was PG-2y made in Changzhou Organic Chemical Plant, China, and was of a standard grade of medium melt viscosity with $M_w = 30\,000$. To avoid hydrolysis of this polymer, the material was dried at 120°C for 12 h in a vacuum oven to remove absorbed water before melt processing. EPDM was also a commercial polymer, EPT 4045 (made in Japan), having diene component 5-ethylidene-2-norbornene (ENB), propylene content (C_3) of 35.9 mol%, and a glass transition temperature (T_g) of about −42°C mesured by dynamic mechanical analysis.

The eEPDM used in this work was synthesized in our own laboratory. The EPDM was first dissolved in toluene, then epoxidized with performic acid which was formed in situ by formic acid and hydrogen peroxide. FTIR spectra were recorded on a BIO-RAD FTS-7 FTIR spectrophotometer. The eEPDM and EPDM were dissolved in toluene to produce 5 wt.% polymer solutions, respectively. The thin films were cast onto potassium bromide plates and laid in a dry atmosphere for 5 days, then dried in a vacuum oven at 40°C for 3 days. Sixteen scans at a resolution of 4 cm^{-1} were signal averaged.

The PC/eEPDM blends with different rubber contents were mixed in a Brabender-like apparatus (Rheocoder XSS-300, made in Shanghai, China) at 245°C for 5 min at a roller speed of 40 rev min^{-1}.

Notched impact tests. The notched Izod impact strength of PC/eEPDM blends was measured by an XJU-22 Izod impact tester (made in Chengde, China) at 9°C. The samples for impact testing were obtained by compression-moulding the PC/eEPDM blends at 240°C, then cutting them into rectangular specimens which were sharply notched with a fresh razor blade. The size of the rectangular specimens was 63.5 mm × 12.7 mm × 3.2 mm.

Morphology. The morphology of PC/eEPDM blends was characterized by scanning electron microscopy (SEM) (model Japan JXA-840). The test specimens were first cold fractured in liquid nitrogen, then coated with a thin layer of

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[*] To whom correspondence should be addressed

gold–palladium alloy to avoid charging under the electron beam.

Results and discussion

Characterization of eEPDM rubber. *Figure 1* shows the typical FTIR spectrum for eEPDM used in this paper, meanwhile that of unepoxidized EPDM is also provided. From *Figure 1*, an apparent difference between the FTIR spectrum of eEPDM and that of EPDM can be seen. In the FTIR spectrum of eEPDM there appears a new peak at 871 cm^{-1} which is the typical peak of epoxy groups, and at the same time the peak area at 808 cm^{-1}, the specific peak of double bonds in the diene component (ENB) in EPDM[18], decreased. Therefore the double bonds in EPDM were converted into epoxy groups by epoxidation. Using the methyl deformation band at 1377 cm^{-1} as internal standard, the conversion ratio of the double bonds into epoxy groups can be obtained as about 70%[17,19].

Impact testing analysis. The mechanical properties of a series of PC/eEPDM blends containing 0–20 wt.% eEPDM were determined. *Figure 2* shows the variation of notched Izod impact strength for specimens with increasing rubber content. The sharp notch causes a catastrophic reduction of impact strength for pure PC[20]. However, it is shown that even a relatively small amount of eEPDM (less than 5 wt.%) leads to significant toughening for the blends. When the rubber content is only 4 wt.%, the notched Izod impact strength of PC/eEPDM blend shows a great improvement, with a strength value being about 25 times of that of pure PC, which means that the eEPDM rubber is a very effective modifier. It is supposed that when EPDM is epoxidized, its

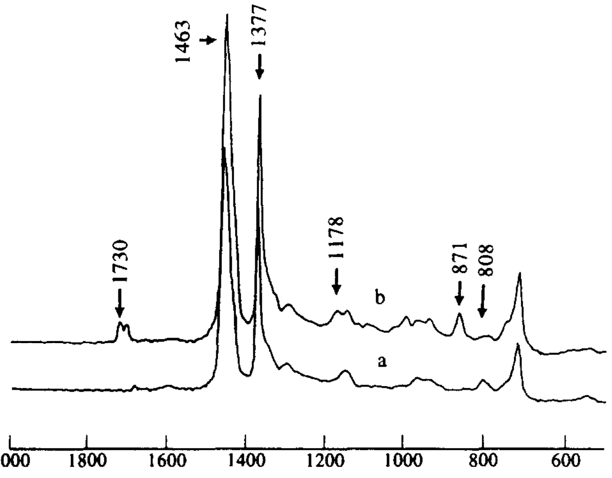

Figure 1 FTIR spectra of EPDM (a) and eEPDM (b)

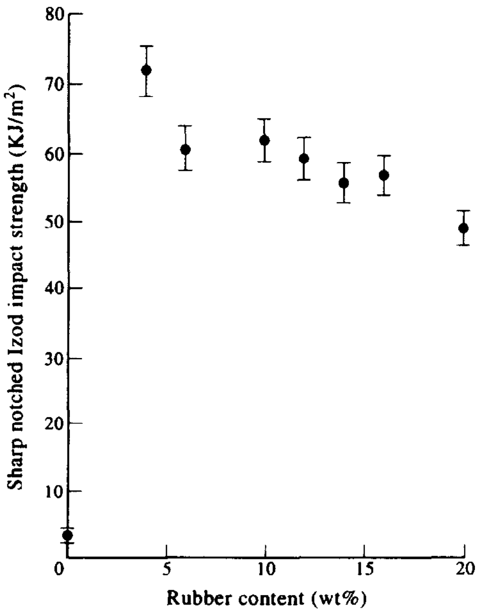

Figure 2 Sharp notched Izod impact strength *versus* rubber content for PC/eEPDM blends at 9°C

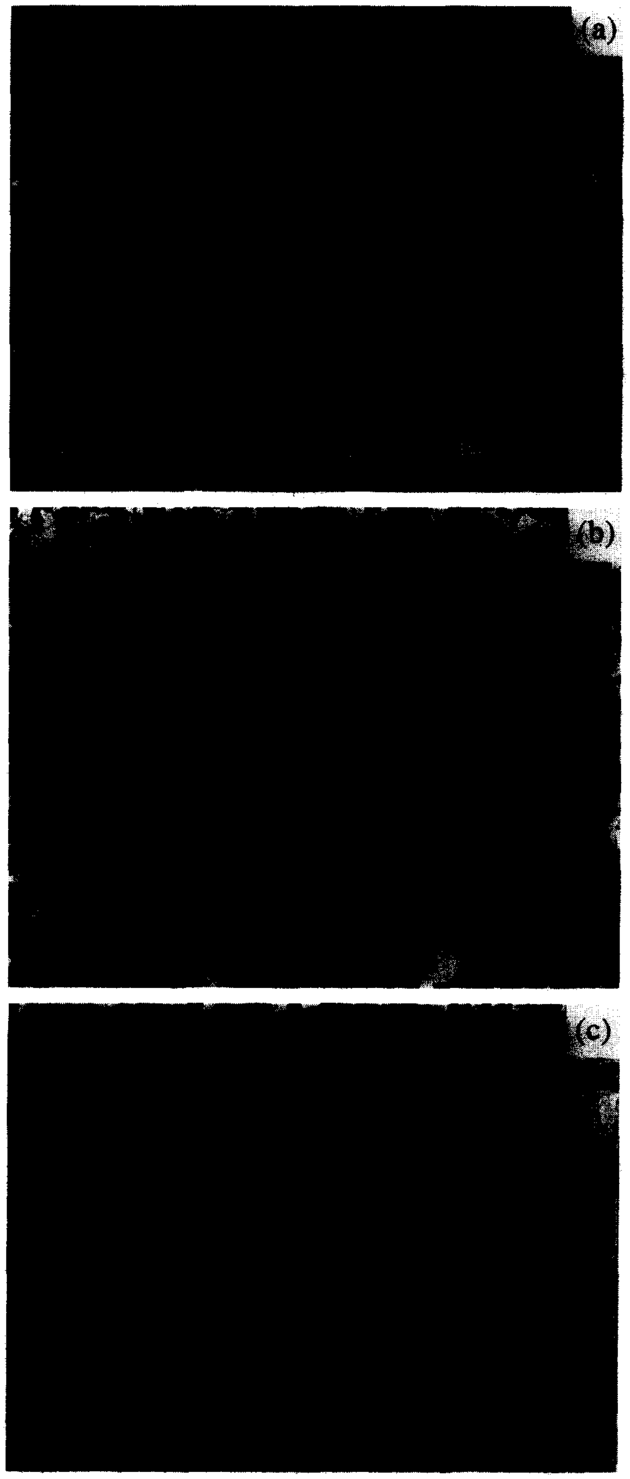

Figure 3 SEM images of cold-fractured surfaces of pure PC (a) and PC/eEPDM (96/4) blend (b,c)

epoxy groups make eEPDM a more polar rubber, providing a certain degree of adhesion with the PC matrix and in the end greatly improving the toughness of the blends. From *Figure 2*, it can also be seen that when the content of eEPDM increases beyond 4%, the toughness decreases because a sufficient amount of modifier (4%) is enough to enhance local shear yielding of the blends. This result is similar to that of ref.[20].

Morphology. The pure PC and PC/eEPDM blend samples were cryogenically fractured and observed by SEM. *Figure 3*(a) shows the SEM image of pure PC. As expected, only a few smooth lines can be seen and a large area of the fracture surface is smooth and featureless indicating the absence of a significant energy-absorption-related deformation mechanism. Compared to the featureless fracture surface of pure PC, that of PC/eEPDM (96/4) blends (shown in *Figure 3*(b)) is no longer smooth. The eEPDM rubber particles are clearly seen, have sizes of 0.2–0.8 μm, and are dispersed finely and homogenously in the PC matrix. This phenomenon implies that relatively small interfacial tension and good adhesion exist between the eEPDM rubber phase and the PC matrix.

Furthermore, it can be found that between eEPDM rubber particles and the PC matrix, there exist cavities (*Figure 3*(c), indicated by the arrow). These cavities at the boundaries between rubber particles and the matrix, which is beneficial to relieve triaxial tension, may occur first and then be followed by enhanced shear yielding of the matrix[21], which can be confirmed by the stress-whitening phenomenon at the impact-fractured surface. Therefore, when the PC/eEPDM blends are impacted the cavities at the boundaries of rubber particles and the following shear yielding of the matrix will relieve the plane strain constraint, dissipate the impact energy and reduce the notch and rate sensitivity of this material. This is thought to be the toughening mechanism for PC/eEPDM blends.

Conclusions

Epoxidized EPDM rubber was used to toughen PC resin and a super-tough PC blend was obtained. The notched Izod impact strength increased greatly when the rubber content was only 4 wt.%. The morphology of PC/eEPDM (96/4) blends showed finely and homogeneously dispersed rubber particles in the PC matrix with sizes of 0.2–0.8 μm, which indicated good adhesion between the eEPDM rubber phase and the PC matrix. The great increase in toughness of PC/eEPDM blends may be due to the cavities at the boundaries of rubber particles and the following shear yielding of the matrix.

Acknowledgements

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