Miscibility and Interchange Reactions in Blends of Bisphenol-A-Type Epoxy Resin and Poly(ethylene terephthalate)

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ABSTRACT: Blends of diglycidyl ether of bisphenol A (DGEBA) and poly(ethylene terephthalate) (PET) were prepared by solution casting from 1,1,2,2-tetrachloroethane. The miscibility and interchange reactions in DGEBA–PET blends were studied by differential scanning calorimetry (DSC) and optical microscopy. PET was found to be miscible with DGEBA, as revealed by the existence of a single composition-dependence glass transition temperature (T_g). Interchange reactions between DGEBA and PET components in the blends at elevated temperatures were proven by appearance of the enhanced glass transition temperatures and the marked decrease in the crystallinity of PET. These results are attributed to the formation of copolymers based on the blend components due to interchange reactions. The morphological observations confirmed that there existed interchange reactions between DGEBA and PET. There also existed a self-crosslinking reaction among the DGEBA molecules. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 73: 639–647, 1999

Key words: epoxy resin; poly(ethylene terephthalate); polymer blends; polymer miscibility; interchange reactions

INTRODUCTION

Miscibility in polymer blends has been extensively studied during the past decades.^{1–3} Because of the very small entropic contribution in high-molecular-weight polymer blend systems, polymers are normally immiscible unless they are very similar chemically or there is a specific interaction between them. Specific intermolecular interactions, such as dipole–dipole force, acid–

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base attraction, ion—ion interactions, and hydrogen bonding interaction, are usually considered to be the driving force for miscibility, and they are responsible for the exothermic heat of mixing that is the thermodynamic basis of miscibility in polymer blends. Additionally, if there exists reaction between blend components, it is also in favor of the blend miscibility improvements.

A fascinating feature for blends in the polyester family is the potential for interchange reaction, also called transreaction or transesterification. As polyesters readily transesterify, interchange reactions commonly occur between constituents when the temperature is near or above their melting points.^{4–16} Particularly, some authors have reported that ester interchange is rapid in the melt and also takes place, but somewhat slowly, at

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 15° C below the melting temperature.^{13,14} Furthermore, an interesting type of interchange reactions take place in blends composed of a polymer with ester or carbonate groups in the main chain and a second component with lateral hydroxyl groups. In these blends, interchange reactions may give rise to grafting and crosslinking processes.^{17–24}

Epoxy resins are a class of important polymeric materials that have been extensively investigated for many years. However, the most parts of studies focused on physical or macroscopic mechanical behavior of hardener-cured epoxy blends, and few works were involved in the true phase behavior between epoxy resin and other polymers, which was easily masked by the presence of the hardener. Epoxy resins, in the monomeric, or oligomeric forms before being crosslinked with a hardener, are quite good in dissolving or dispersing many linear polymers. Many precured mixtures of linear polymers with epoxy resins form homogeneous blends before curing, but most of these homogeneous liquid blends would turn into multiphase systems after they undergo crosslinking reactions with curing agents or hardeners.^{25–27}

In our previous work, we have found that uncured bisphenol-A-type epoxy resin, that is, diglycidyl ether of bisphenol A (DGEBA), is miscible with several linear polymers, including poly(ethylene oxide),^{28,29} phenolphthalein poly(ether ether ketone),^{30,31} phenolphthalein poly(ether ether sulphone),³² poly(hydroxyether of bisphenol A),³³ poly(vinyl acetate),³⁴ and poly(styrene-coacrylonitrile).³⁵ However, the morphology and phase behavior of the cured blends were found to be greatly dependent on the choice of curing agent. Reactions or strong specific interactions between epoxy resin and the linear polymer, if taking place at appropriate temperature, might prevent or lessen the extent of phase separation at the fully cured state. Recently, Nichols and Robertson investigated the blends of DGEBA with poly(butylene terephthalate) and found that the thermoreversible gelation occurred.^{36,37} Woo and Wu studied the chemical interactions and miscibility in hardener-free DGEBA-polycarbonate blends by differential scanning calorimetry (DSC) and Fourier transform infrared (FTIR) spectroscopy. They found that chemical exchange reactions occurred, and a crosslinked structure formed in the blends after heat treatment.³⁸

In this work, we have carried out a systematic study of the miscibility and interchange reactions in blends of DGEBA and poly(ethylene terephtha-

late) (PET). We choose PET because it is a polyester widely developed as engineering plastics. The effect of interchange reactions on the blend miscibility, as well as on the crystallization and melting behavior of PET, was investigated. In order to directly observe the effect of thermal treatment on the binary blend system, no hardeners of any type were used. Therefore, this study can focus on the relationships between the chemical reactions and phase behavior of the hardenerfree blend system. By understanding the role of interchange reactions, the phase behavior of hardener-cured blends of polymer-epoxy networks can be easier to be predicted and controlled.

EXPERIMENTAL

Materials and Preparation of Samples

The bisphenol-A-type epoxy resin E-51 with epoxide equivalent weight 185–210, that is, diglycidyl ether of bisphenol A (DGEBA), was obtained from Wuxi Resin Factory, Wuxi, People's Republic of China. The PET was supplied by Changzheng Chemical Industrial Factory, Beijing, People's Republic of China. It had an intrinsic viscosity $[\eta]$ = 0.74 dL/g in the solution of phenol/1,1,2,2-tetrachloroethane (3/5, v/v) at 30°C. To calculate the molecular weight, the Mark–Houwink equation was used, as follows:

$$[\eta] = KM_{\eta}^{\alpha} \tag{1}$$

where M_{η} is the viscosity-average molecular weight, and the values of K and α for PET are 22.9 $\times 10^{-3}$ mL/g and 0.73, respectively.³⁹ The viscosity-average molecular weight M_{η} was calculated to be 6.42×10^4 .

PET was first dissolved in 1,1,2,2-tetrachloroethane with continuous stirring at 120°C. Then, DGEBA was added to the mixture, and a homogeneous ternary mixture was obtained. Most of the solvent was evaporated slowly at 50°C. The residual solvent was removed under vacuum at 50°C for 2 weeks.

Differential Scanning Calorimetry

Calorimetric measurements were performed on a Perkin–Elmer DSC-7 thermal analysis apparatus in a dry nitrogen atmosphere. The instrument was calibrated with an Indium standard. All sam-



Figure 1 DSC curves of the as-cast DGEBA–PET blends.

ples (about 10 mg in weight) were scanned from -60 to 280°C. A heating rate of 20°C/min was used. The glass transition temperature (T_g) was taken as the midpoint of the capacity change. The crystallization temperature (T_c) was taken to be the maximum of the exothermic peak and the melting point (T_m) that of the endothermic melting peak. The heats of crystallization (ΔH_c) and fusion (ΔH_f) were measured from the areas under the crystallization and melting peaks, respectively.

Morphological Observations

The morphology of the DGEBA–PET blends was investigated by optical microscopy using a Jiangnan XPR-2 optical microscope. The samples were maintained at 280°C for different periods of time and then rapidly cooled to the crystallization temperature, selected to be 210°C, and held for 20 min.

RESULTS AND DISCUSSION

The As-Cast DGEBA-PET Blends

The DSC curves of the as-cast blends are presented in Figure 1. It is clear that all the blends display a single transition temperature (T_g) , intermediate between those of the 2 pure components and changing with the blend composition. Figure 2 summarizes the T_g values as a function of blend composition. The overall variation of T_g with composition suggests that DGEBA-PET blends are completely miscible in the amorphous phase.

Several theoretical and empirical equations have been used to describe the T_g -composition dependence of miscible polymer blends. One of these, the Gordon–Taylor equation,⁴⁰ is written as

$$T_g = (W_1 T_{g_1} + k W_2 T_{g_2}) / (W_1 + k W_2)$$
(2)

where T_g is glass transition temperature of the blend, T_{g_1} and T_{g_2} are the glass transition temperatures of the components 1 and 2, respectively, W is the weight fraction, and k is a constant. The curve in Figure 2 is drawn using the Gordon–Taylor equation with a k value of 0.10, fitting the experimental data well.

Belorgey and coworkers^{41,42} have suggested that the k value can be taken as a semiquantitative measure of the strength of interaction be-



Figure 2 Composition dependence of glass transition temperature of as-cast DGEBA–PET blends. The curve is as predicted by the Gordon–Taylor equation using a k value of 0.10.



Figure 3 DSC curves of the DGEBA-PET blends treated at 280°C for 10 s.

tween the components of the blend. For instance, in blends of poly(ε -caprolactone) with chlorinated polyethylene, poly(vinyl chloride) (PVC) and chlorinated PVC, k increases from 0.26 to 1.0. When such an approach is used for the DGEBA–PET blends, we note that the k value (0.10) is much smaller, suggesting that the interaction between DGEBA and PET is very weak. The miscibility of the DGEBA–PET blends can be considered to be due mainly to the nonnegligible entropy contribution as the molecular weight of DGEBA is rather low.

It can also be seen from Figure 1 that the T_m of PET decreases with increasing DGEBA content. The T_m depression is a common phenomenon in miscible crystallizable-amorphous polymer blends.^{43,44}

Thermally Treated DGEBA-PET Blends

Interchange reactions in blends composed of a crystallizable and an amorphous polymer may be studied by means of the effects of reactions on the thermal behavior of the blends, and mainly following their effects on the crystallization-melting behavior of the crystallizable polymer. Such effects arise from the formation of copolymers as a consequence of reactions. Therefore, the crystallization-melting behavior of the PET in the DGE-BA-PET system was investigated.

Figures 3 and 4 show the DSC curves of the DGEBA–PET blends treated at 280°C for 10 s and 1 min, respectively. It is interesting to see that the 20/80 DGEBA–PET blend treated at 280°C for 1 min shows a crystallization peak (Fig. 4), while that treated for 10 s has no crystallization peak (Fig. 3), which suggests that crystallization rate of PET in the blends was decreased after thermal treatment. It can also be seen that the T_m of PET remarkably decreases with increasing DGEBA content.

The fraction crystallinity X_{c} can be calculated from

$X_c(\text{PET}) = (\Delta H_f(\text{PET}) - \Delta H_c(\text{PET})) / \Delta H_f^0(\text{PET}) \quad (3)$

where $\Delta H_f^0(PET) = 27.8$ kJ/mol is the heat of fusion for 100% crystalline PET,⁴⁵ and $\Delta H_c(PET)$ and ΔH_f (PET) are the heats of crystallization and fusion for PET in the blend. Figure 5 shows the plots of X_c as functions of blend composition for the as-cast blends and the blends treated at 280°C for 10 s and 1 min. The X_c (PET) values of



Figure 4 DSC curves of the DGEBA-PET blends treated at 280°C for 1 min.



Figure 5 Fractional crystallinity X_c as functions of PET content for the as-cast DGEBA–PET blends (\blacksquare) and the blends treated at 280°C for 10 s (\blacktriangle) and 1 min (\triangle).

the treated blends are lower than those of the as-cast blends, which implies that the thermal treatment had a remarkable effect on the crystallization-melting behaviors of PET in the blends. In addition, we can see that the X_c (PET) values first increase with DGEBA content and come to their maximum when DGEBA content is 50 or 60 wt %. This result seems to indicate that the presence of DGEBA favors the crystallization of PET. A similar behavior has also been found in other polymer blends containing polyesters.^{19,46,47}

Glass Transitions of the DGEBA-PET Blends Treated with Various Times at 280°C

Figure 6 shows the plots of T_g as functions of blend composition for the DGEBA–PET blends treated at 280°C for various treatment time ranging from 10 s to 60 min. The T_g values of the treated blend samples significantly increased as the thermal treatment time increasing.

The values of Gordon-Taylor parameter k for the blends treated at 280°C with treatment times of 10 s and 1, 3, 5, and 10 min are indicated in Figure 6. It should be pointed out that for the blends treated for more than 10 min, the GordonTaylor equation cannot describe the T_g -composition relations. It can be seen in Figure 6 that the k value increases with the thermal treatment time. For the DGEBA-PET blends treated at 280°C for 10 s and 1 min, k = 0.12 and 0.15, respectively. These values approaches that of the as-cast DGEBA-PET blends (k = 0.10) suggests that no obvious reaction occurs between the DGEBA and PET components. However, for the blends treated at 280°C for 3 min, k was 0.32. This increase of k value implies that there was considerable change in the blends after treated at 280°C for 3 min.

The similar effects have been observed for the epoxy–polycarbonate systems,^{38,48} in which interchange reactions were found to occur between the 2 components. Therefore, in the present case, a similar reaction possibly occurred between the hydroxyl group (—OH) of the DGEBA with the ester group in PET. The transreaction led initially to the formation of a graft copolymer, and the grafted copolymers were then transformed into crosslinked copolymers upon further reaction. These grafted and crosslinked copolymers were responsible for the tendency to the increase of T_g at high reaction times. It should be pointed out that when thermal treatment at 280°C was



Figure 6 T_g s of DGEBA-PET blends treated at 280°C for treatment time of 10 s (\blacktriangle) and 1 (\triangle), 3 (\bigcirc), 5 (\bigcirc), 10 (\blacksquare), 20 (\Box), 30 (\diamondsuit), and 60 min (\diamondsuit).

conducted for longer times (but less than 10 min), the changes in the k value for the treated blend samples became increasingly larger, suggesting an even greater interaction between the components. For the same blend, thermal treatment at 280°C for even longer time (30 min), the T_g -composition curve began to exhibit a sigmoidal shape. The appearance of the sigmoidal shape in the T_g curves can be attributed to the fact that the extent of crosslinking and network structure varied with the PET content in the blends.

However, it also can be observed from Figure 6 that the T_g of the pure DGEBA dramatically increased as the thermal treatment time is longer than 1 min. When treated at 280°C for 60 min, the T_g of the pure DGEBA was even higher than that of pure PET, indicating that the self-crosslinking reactions of DGEBA occurred after the high temperature treatment for a long time. The self-crosslinking reaction of DGEBA was responsible for the enhancement of T_g values of the blends as the thermal treatment time increasing.

The solubility of the blends was examined to assess the effects of thermal treatment on the extent of reactions or network forming in the blends. For the 50/50 DGEBA-PET blend treated at 280°C for 5 min, the blend can be dissolved in the 1,1,2,2-tetrachloroethane at 120°C. However, the blend treated for 10 min did not dissolve at all, clearly displaying that the crosslinking reaction took place during the thermal treatment of the blends.

Effect of Thermal Treatment Time on Thermal Properties of the Blends

The DSC curves of the 50/50 DGEBA-PET blend treated with various times at 280°C are shown in Figure 7. It can be seen from Figure 7 that, with the increase of treatment time, the extent of reaction increased. When the thermal treatment time was not more than 3 min, the extent of reaction was low, and crystallization of PET in the blend was so rapid that it could be completed during the cooling process, which led to no crystallization peak occurring on heating. When the treatment time at 280°C was increased to 5 min or longer, a crystallization peak appeared in the DSC curve, as shown in Figure 7. With the increase of treatment time, the peak area gradually increases to a maximum, then decreases. The peak position is observed to shift to higher temperature, which indicates a decrease in the crystallization rate of PET in the blend. Furthermore,



Figure 7 DSC curves of the 50/50 DGEBA–PET blends treated at 280°C for various treatment times.

the blends treated at 280°C for 30 min or longer do not show any crystallization exotherm. The T_c values of 80/20, 50/50, and 20/80 DGEBA–PET blends are plotted in Figure 8 as functions of treatment time at 280°C. With an increase of the extent of reaction, some DGEBA molecules were incorporated into the PET chains, and, hence, the crystallization rate of PET decreased. The decreasing crystallization rate of PET in the blends is presumably due to a great decrease of the crystallizable segment length as a consequence of interchange reactions and restricted segmental motions.

It can also be seen from Figure 7 that with the increase of treatment time at 280°C, the melting peak of PET in the blends gradually shifts to lower temperature, and the area of the melting peak decreased. The T_m values of 80/20, 50/50, and 20/80 DGEBA–PET blends are also shown in Figure 8 as functions of treatment time at 280°C.

It is evident that both crystallinity and crystallization ability of PET in the blends decreased with the increase of thermal treatment time. This can be considered as a result of the loss of perfection of the PET crystallites due to the decrease of the crystallizable segment length of this polymer.



Figure 8 Melting temperatures and crystallization temperatures of DGEBA-PET blends as functions of treatment time at 280°C.

In the initial stages of reaction, the increase in the area of the crystallization peak in Figure 7 can be attributable to a decrease of crystallization ability of PET in the blends with the introduction of the DGEBA into the PET chains. The crystallization behavior of PET in the blends did not change during the thermal treatment. In the final stages of reaction, the decrease in the area of the crystallization peak is due to the decrease of crystallinity of PET in the blends as the recrystallization process was hindered by the DGEBA molecules incorporated in the PET chains. When the reaction extent reached a critical value, both the melting peak and crystallization peak disappeared, which indicates that the crystalline regions of PET in the blends were completely destroyed. Figure 9 shows fractional crystallinity X_c of DGEBA-PET blends as functions of treatment time at 280°C. The same variation of the crystallinity with treatment time is also observed for the 80/20 and 20/80 compositions. The fraction crystallinity X_c decreased greatly when treated at 280°C for 10 min. Similar results were also found in other blends containing a crystallizable condensation polymer in which an interchange reaction takes place at high temperatures, $^{16,1\overline{7},19}$ and such an effect could not be attributed only to the

self-crosslinking reactions among the DGEBA molecules. Therefore, it can be concluded that an interchange reaction has occurred between the DGEBA and PET components when treated at 280°C for a long enough time.

Optical microscopy can also provide a clear evidence of the effect of interchange reactions on the crystallization behavior of PET in DGEBA-PET blends. Figures 10(a)-(d) show optical photomicrographs with crossed polar for the 50/50 DGE-BA-PET blend first thermally treated for 1, 3, 5, and 10 min at 280°C, respectively, followed by rapid cooling to 210°C to crystallize at this temperature for 20 min. In Figure 10(a) it can be seen that the blend treated for 1 min has completely crystallized after 20 min at 210°C. However, with increase of the treatment time, the crystallinity was markedly decreased, and the crystallization of the blend became increasingly difficult [Fig. 10(b)–(d)]. The longer the treatment time, the less the extent of crystallization. Moreover, there was no crystallization observed for the blend heated at 280°C for 15 min and longer. These results further support the idea that there exist an interchange reaction between DGEBA and PET.

CONCLUSIONS

PET was found to be miscible with DGEBA as revealed by the existence of single composition



Figure 9 Fractional crystallinity X_c of DGEBA–PET blends as functions of treatment time at 280°C.



Figure 10 Optical photomicrographs for 50/50 DGEBA-PET blends. The samples were first treated at 280°C for (a) 1, (b) 3, (c) 5, and (d) 10 min and subsequently crystallized at 210°C for 20 min.

dependent T_g . The interchange reaction took place during thermal treatment of DGEBA–PET blends in the molten state. The reaction of the hydroxyl groups of DGEBA with the ester groups of PET gave rise to grafted and crosslinked copolymers. Interchange reactions resulted in an increase in T_g , a decrease in the crystallinity, and in the perfection of the crystalline structures of PET. These results were confirmed by the morphological observations.

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