Evidence for Mechanical Coupling Effects in Binary Polymer Blends: Relationships with Morphology

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ABSTRACT: The viscoelastic properties of binary thermoset and thermoplastic polymer blends were investigated in connection with blend morphologies. Christensen and Lo's model was used to predict mechanical coupling effects in such binary multiphased systems by accounting for the actual morphology of samples. Thus, it was shown that the magnitude of mechanical coupling effects between phases in polymer blends, as in composite materials, depends not only on mechanical properties and relative content of each phase but also on the geometric arrangement of the polymeric phases. Furthermore, based on both theory and experiment, a well-suited probe of blend morphology was also proposed. © 2000 John Wiley & Sons, Inc. J Appl Polym Sci 76: 530–541, 2000

Key words: thermoset/thermoplastic blend; self-consistent scheme; mechanical coupling effects; probe of morphology

INTRODUCTION

Dynamic mechanical spectrometry has been extensively used for the characterization of heterogeneous polymeric systems such as block- and graft-copolymers, as well as blends. The key feature of the heterogeneity of a two-phase system appears to be the existence of two maxima on temperature dependence of mechanical losses. Most of changes on mechanical spectra of polymer blends are usually interpreted[1] with a physico-chemical approach, including considerations about the compatibility between polymeric species. Subsequently, Lipatov detailed the concept of "mutual influence between the polymeric components" and explained that "in most polymer–polymer systems, the flexible polymer becomes stiffer while the stiffer softens."[2] Bohn[3] also indicated that, in some multiphased systems, the apparent glass transition temperature T_α of dispersed polymeric phase tends to be shifted somewhat. The shifts were shown to be especially significant[3] for polymers of high glass transition temperature T_g dispersed in polymer of low T_g: the high-temperature loss tangent maximum is sometimes shifted toward higher temperatures than might be expected. Furthermore, the effects appear less pronounced, but still significant, for polymer blends with low T_g polymer dispersed in high T_g polymer matrix.

However, it is important to recall that mechanical spectra reflect the overall viscoelastic behavior of multiphased materials. As a matter of fact, in particular for polymers reinforced by rigid mineral fillers, it is well known that the viscoelastic response of samples is governed by two major factors: (1) the nature of bonds between fillers and polymer matrix[5–11] and (2) mechanical coupling effects between phases.[5,11–15] Accordingly, as for composite materials, mechanical coupling effects between phases also must be expected on mechanical spectra of heterogeneous polymeric systems, such as polymer blends. Thus, before interpreting changes on the viscoelastic behavior of each polymeric component, it is first necessary to predict the sole effects resulting from mechanical coupling between phases.

This paper focuses on such an approach derived from that proposed by Alberola et al.,[16,17] who recently developed an improved mechanical

model to predict mechanical coupling effects between phases by accounting for the morphology of composite materials. It was shown that the magnitude of the reinforcement effect, so-called mechanical coupling between phases, depends not only on mechanical properties and the relative content of each phase but also on the geometric arrangement of the dispersed phase within the polymer matrix. In this way, the aim of the present work is to investigate the magnitude of mechanical coupling effects in binary thermoset and thermoplastic blends by accounting for the geometric arrangement of the different polymeric phases. Subsequently, based on both theory and experiment, a probe of the blend morphology is proposed.

EXPERIMENTAL SECTION

Materials

The thermoset and thermoplastic blends used were prepared by Girard-Reydet et al.[19] They are made up of the diglycidyl ether of Bisphenol A (DGEBA; Dow Chemical (Midland, USA); DER332, M_n = 348.5 g/mol) and 4,4′-methylene bis[3-chloro-2,6-diethylaniline] (MCDEA; Lonza (Basel, Switzerland), M_n = 380 g/mol). Components were mixed at the stoichiometric ratio amino-hydrogen-to-epoxy equal to 1. The high-T_g thermoplastic used was an amorphous nonfunctionalized polyetherimide supplied by General Electric Company (Pittsfield, MA) (PEI Ultem 100, M_n = 26 kg/mol, M_w = 50 kg/mol). The mixture with PEI being miscible at 80°C, the phase separation occurred during curing. The kinetic study of such modified epoxy systems was reported elsewhere by Girard-Reydet et al.[19] Precure time (120 h at 80°C) was chosen to be longer than vitrification times to be sure that most of the microstructure developed isothermally. In order to ensure a complete network formation without any degradation, all samples were postcured at 185°C for 2 h.

Here, two thermoset and thermoplastic blends are investigated and denoted as B10 and B20, respectively. For example, B10 denotes the system DGEBA-MCDEA modified with 10 wt % of PEI. Their morphologies, which were generated through the reaction-induced phase-separation procedure, were also investigated.[20] Thus, for the B10 sample, phase separation produced a particulate morphology, with a thermoplastic-rich phase dispersed in a thermoset-rich continuous

Table I   Glass Temperatures of Components from Differential Scanning Calorimetry

DGEBA-MCDEA Network	Glass Transition Temperature, T_g (°C)	
	Thermoset Phase	Thermoplastic Phase
	175 ± 2	/
B10	166 ± 2	—
B20	169 ± 2	180 ± 2
PEI	/	214 ± 2

—, Too weak change in the heat capacity baseline to determine.

phase. On the opposite, with 20 wt % of PEI, phase separation led to the formation of an inverted structure with a continuous phase that is mainly rich in thermoplastic.

Sample Characterization

Differential scanning calorimetry (DSC) thermograms were recorded using a DSC 7 Perkin-Elmer device with a heating rate of 10°C/min under nitrogen atmosphere. The T_g was determined from the slope change of the baseline.

The torsion pendulum micromechanalyser (Metravib Co., Lyon, France) was used under isochronal conditions at 1 Hz to measure the temperature dependence of the complex shear modulus G^* from 120°C to 260°C with a heating rate of ∼ 1°C/min. The samples were approximately 34 mm long, 5 mm wide, and 0.8 mm thick.

RESULTS AND DISCUSSION

Experimental Results

DSC Measurements

The T_gs of both thermoset and thermoplastic phases in B10 and B20 samples are given in Table I as well as T_g values of pure components (PEI and DGEBA-MCDEA network).

First, it can be seen that T_g of thermoset phase in both B10 and B20 is located at about 168°C, that is, at a lower temperature than T_g of pure epoxy network. Such an observation probably results from a decrease in the crosslinking degree of the epoxy network in the blend. This could be induced by the phase separation of the thermoplastic during curing.
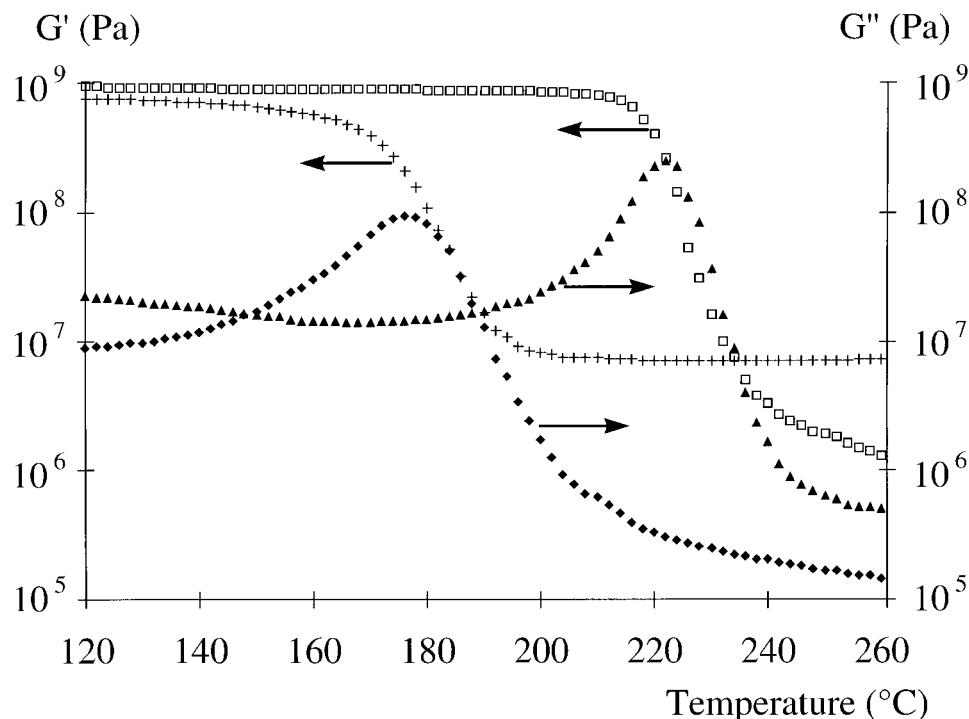
Figure 1 Viscoelastic characteristics (storage modulus G' and loss modulus G'') *versus* temperature at 1 Hz for DGEBA-MCDEA network (+, ◆) and PEI (□, ▲).

It can be also seen that T_g of thermoplastic phase is located at a lower temperature in B20 than in pure PEI ($\Delta T \sim 30°C$). This strong shift toward lower temperatures could be attributed to the remaining low molar masses epoxy-amine species dissolved in the thermoplastic phase, which act therefore as plasticizers.[20] This is consistent with a lower crosslinking degree of the epoxy network.

Mechanical Spectrometry

The experimental real (G') and imaginary (G'') parts of the complex moduli for pure components (PEI and DGEBA-MCDEA network) are given in Figure 1. Figure 2(a) shows the experimental values of G' for B10 and B20 samples, and Figure 2(b) gives the experimental values of the loss factor (tan δ) for all the materials. In Table II are listed values of the following viscoelastic characteristics: (1) the temperature location of the main relaxation related to T_g for both thermoset and thermoplastic phases (denoted as T_α^{TD} and T_α^{TP}, respectively) and (2) the height of the corresponding peak of tan δ (denoted as tan δ_{max}), displayed by all the materials.

The analysis of data leads to the following features:

1. T_α^{TD} displayed by B10 sample is very close to that of pure epoxy network. In contrast, a significant shift toward the lower temperatures is observed for T_α^{TD} in B20 sample.
2. T_α^{TP} displayed by both B10 and B20 samples remain constant at about 209°C, that is, at a lower temperature than that of pure PEI ($\Delta T \sim 20°C$).

Experimental Conclusion

The changes in the mechanical properties of epoxy network induced by thermoplastic, that is, the slight shift of T_α^{TD} toward the lower temperatures displayed by B10 and B20 samples, could indicate an increase in the molecular mobility of the thermoset matrix near T_g. Therefore, a change in the microstructure of the DGEBA-MCDEA network induced by PEI can be invoked. Such an evolution agrees with conclusions derived from DSC analysis. The same tendency also is found for the modification of T_α of the thermoplastic phase. However, it is relevant to underline that the shifts toward the lower temperatures of both T_α^{TD} (mainly) and T_α^{TP} are significantly lower than T_g shifts found from DSC analysis. To accurately interpret such a result, it is required to recall that the overall mechanical behavior of
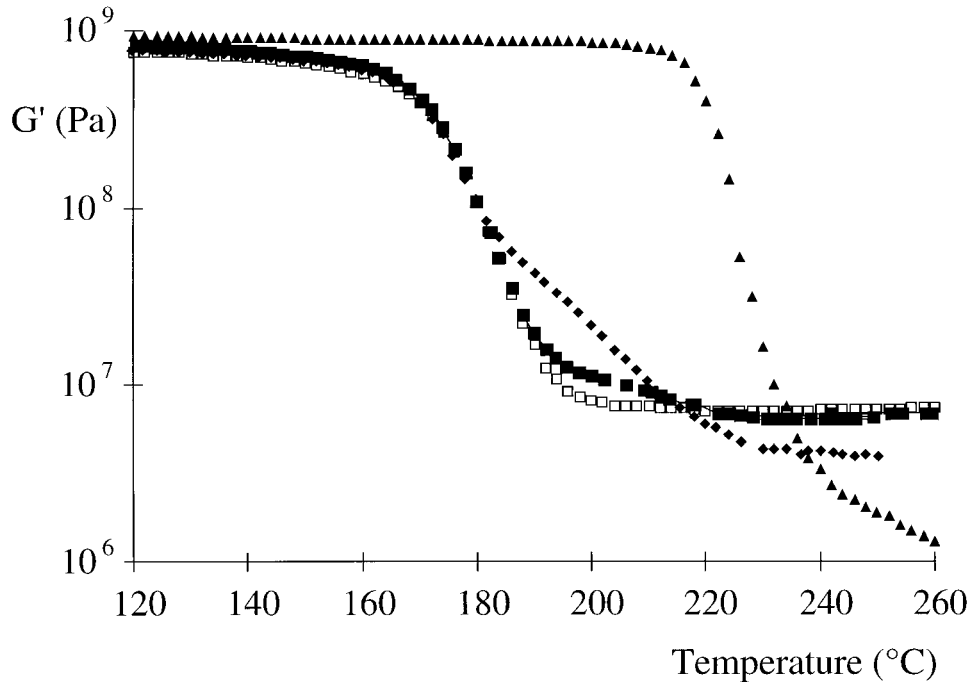
(a)
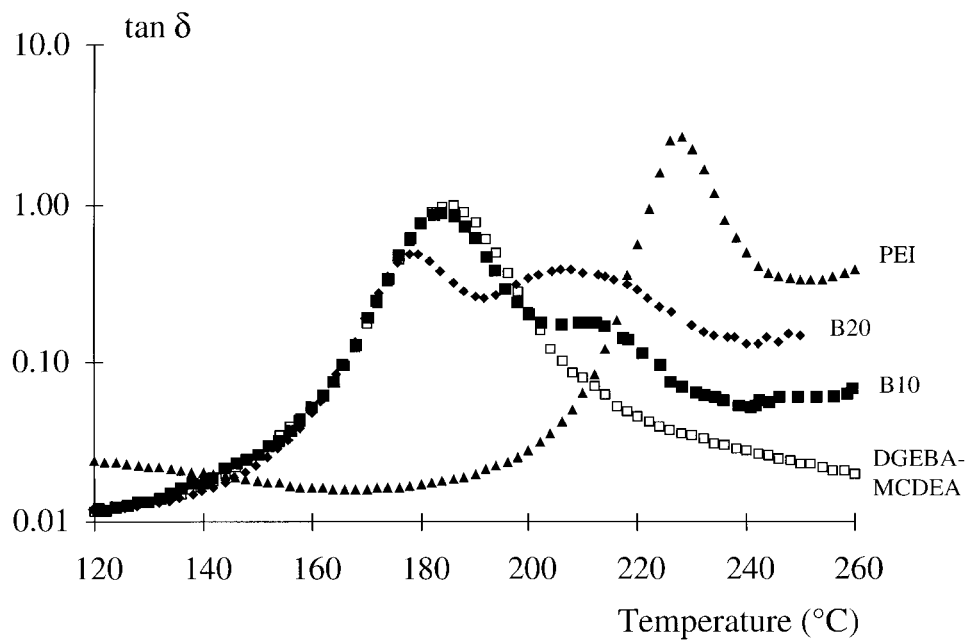
(b)

Figure 2 Viscoelastic behavior at 1 Hz for B10 (■) and B20 (◆) blends. (a) Storage modulus G' *versus* temperature; (b) loss factor tan δ *versus* temperature. The experimental viscoelastic behavior at 1 Hz for DGEBA-MCDEA (□) and PEI (▲) are recalled for comparison.

multiphased systems is also governed by morphology. In other terms, mechanical coupling effects have to be predicted by accounting for the morphology of samples.

The morphology analysis of B10 and B20 samples was previously investigated[20] and revealed that the continuous phase (i.e., matrix) for the B10 sample is the thermoset phase, whereas in

Table II Characteristic Issues from Dynamic Mechanical Analysis at 1 Hz

DGEBA-MCDEA Network	Thermoset Phase		Thermoplastic Phase	
	T_α^{TD} (°C)	tan $\delta_{max\ TD}$	T_α^{TP} (°C)	tan $\delta_{max\ TP}$
	186 ± 1	0.94	/	/
B10	184 ± 1	0.70	208 ± 2	0.33
B20	179 ± 1	0.39	210 ± 2	0.32
PEI	/	/	228 ± 2	2.51

B20 sample it is the thermoplastic phase. Such a change in the macroscopic phase arrangement of both thermoset and thermoplastic phases could lead to a modification of the effects resulting from mechanical coupling between phases. Accordingly, it is of interest to investigate the influence of the geometric arrangement of phases on the contribution of mechanical coupling in mechanical spectra.

Mechanical Modeling Section

Presentation

The prediction of the mechanical behavior of such two-phase materials can be performed by the well-known Christensen and Lo's model,[18] extended to describe the viscoelastic behavior through the correspondence principle.[21] The representative volume element (RVE) is constituted by two concentric spheres embedded in an equivalent homogeneous medium. Accounting for the different morphologies exhibited by B10 and B20 samples, two RVE must be defined. They are denoted as RVE-10 and RVE-20, respectively, and are depicted in Figures 3(a,b). In RVE-10 [Fig. 3(a)], a thermoplastic inclusion, acting as the central core with the radius r_1 is embedded by a shell of thermoset matrix that is limited by the spheres with the radii r_1 and r_2. The ratios of radii are expressed as function of the volume fractions of the dispersed phase (V_{p10}) and matrix (V_{m10}) as follows:

$$V_{p10} = r_1^3/r_2^3 \quad \text{and} \quad V_{m10} = 1 - (r_1^3/r_2^3)$$

In RVE-20 [Fig. 3(b)], the central core is constituted by a thermoset DGEBA-MCDEA inclusion with the radius R_1, which is surrounded by a shell of thermoplastic PEI matrix (limited by the spheres with the radii R_1 and R_2). The ratios of radii also are expressed as function of the volume

fractions of the dispersed (V_{p20}) and continuous phases (V_{m20}) as follows:

$$V_{p20} = R_1^3/R_2^3 \quad \text{and} \quad V_{m20} = 1 - (R_1^3/R_2^3)$$

Then, prediction of elastic and linear viscoelastic properties of the binary polymer blends is based on the derivation of the elastic strain or stress field in an infinite medium constituted by a two-layer inclusion embedded in a matrix submitted to uniform stress or strain at the infinity.

Numerical simulations are performed by accounting for the following assumptions:

1. Viscoelastic properties of both thermoset and thermoplastic phases are taken to be identical to those displayed by pure DGEBA-MCDEA and pure PEI, respectively. This is to predict the sole theoretical mechanical coupling effects governed by the geometric arrangement of the phases.
2. Volume fractions are first chosen in agreement with blend composition, that is, 0.1 (or 0.2) for PEI and 0.9 (or 0.8) for DGEBA-MCDEA in B10 (or B20) sample. To supply discussion, other theoretical volume fractions from 0.10 to 0.90 for both PEI and DGEBA-MCDEA, respectively, are also considered in simulations.
3. To account for the glassy-rubbery transition undergone by the polymers through their respective glass transition, an S-shape variation of the Poisson's ratio of both components is assumed from 0.33 to 0.50 at $T > T_g$.[5]

To better illustrate the influence of the geometric arrangement of phases on the mechanical coupling effects, it is relevant to establish the relationship between the radii of concentric spheres, which are considered in RVE-10 and RVE-20. As
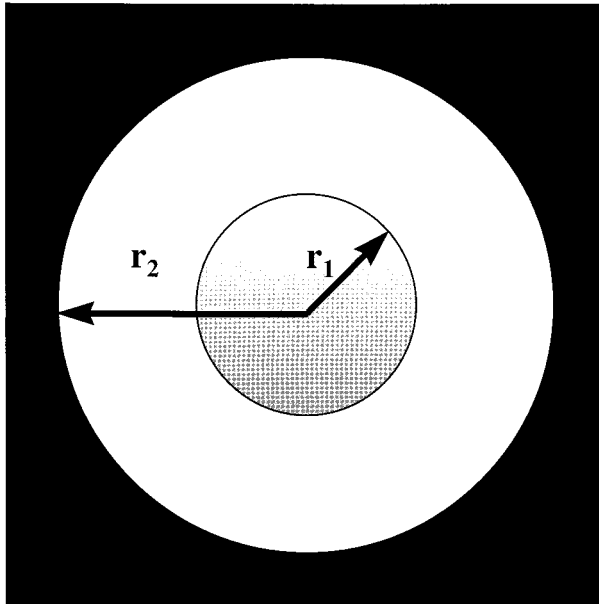
(a)
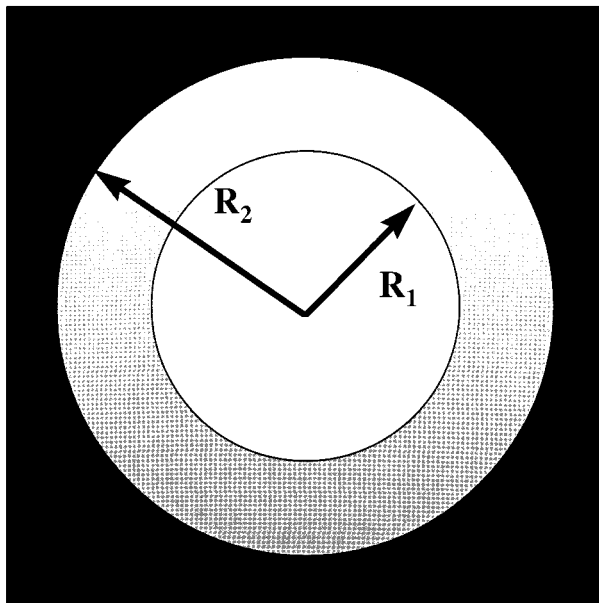
(b)

Figure 3 Illustrations of the representative elemental volumes (RVE) considered in this work. (a) RVE-10: DGEBA-MCDEA considered as continuous phase; (b) RVE-20: DGEBA-MCDEA considered as dispersed phase. Black area, equivalent homogeneous medium; white area, thermoset DGEBA-MCDEA; shaded area, thermoplastic PEI.

a matter of fact, RVE-10 and RVE-20 can be used for a given blend composition. This leads to $V_{p10} = V_{m20}$ and $V_{m10} = V_{p20}$. Accordingly, the radii r_1

and r_2 in RVE-10 can be connected to the radii R_1 and R_2 in RVE-20 by the following expression:

$$R_1 = R_2(1 - r_1^3/r_2^3)^{1/3}$$

Numerical Results

Figure 4(a,b) shows the theoretical evolution of the storage modulus G' at 1 Hz versus temperature for binary blends containing 10, 20, 50, and 90 vol % of PEI, respectively. In Figure 4(a), predictions are based on RVE-10, that is, DGEBA-MCDEA network is first taken as the continuous phase. In Figure 4(b), theoretical issues are based on RVE-20, that is, PEI is considered as the continuous phase. The experimental evolutions of G' for pure DGEBA-MCDEA network and PEI are also given for comparison. Whatever the binary blend composition can be, it is observed that the overall prediction of G' versus temperature of blends is mainly governed by that shown by the chosen continuous phase. The main influence of the continuous phase is also clearly shown for the blend containing 50 vol % of each phase. This result is in agreement with Bohn's conclusion,[4] which indicated that the temperature dependence for polymer blends is very often similar to that of the continuous phase in the temperature range between the two main relaxations. It is also nice to notice that all predictions of G' of blends versus temperature [see Fig. 4(a,b)] have a point in common at about 235°C. The presence of such a so-called isosbestic point confirms the self-consistency of the used mechanical modeling.

Figure 5 reports theoretical evolution of the loss factor tan δ at 1 Hz versus temperature for binary blends containing 10 or 20 vol % of PEI. As for Figure 4(a,b), modeling was performed by considering either RVE-10 or RVE-20. T_α^{TD} and T_α^{TP} (given in Table II) for pure components (DGEBA-MCDEA network and PEI, respectively) are indicated with arrows for comparison. Figure 5 appears very interesting for investigating mechanical coupling effects between phases in polymer blends. As a matter of fact, it can be seen that

1. For a same composition, the magnitude of the main relaxations exhibited by both thermoset and thermoplastic phases are the highest when the respective component is chosen as the continuous phase in modeling.
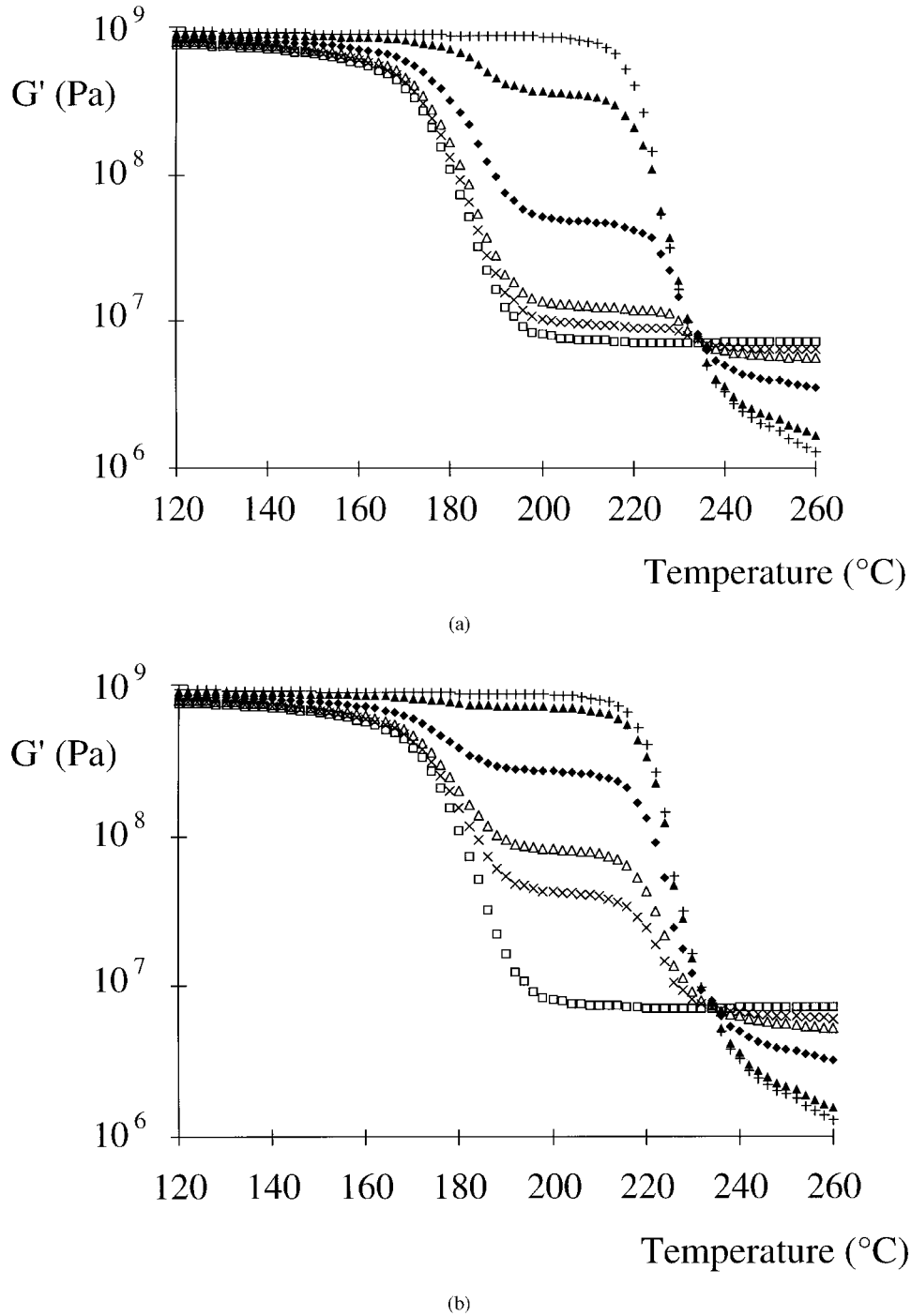2. The temperature location of the main relaxation related to T_g (denoted as T_α^{TD} and

(a)

(b)

Figure 4 Theoretical evolution of the storage modulus G' *versus* temperature for binary blends containing 10 ($\times$), 20 ($\triangle$), 50 ($\blacklozenge$), and 90 vol % ($\blacktriangle$) of PEI as a function of the morphology considered in RVE. (a) DGEBA-MCDEA considered as continuous phase (RVE-10); (b) DGEBA-MCDEA considered as dispersed phase (RVE-20). The experimental viscoelastic behavior at 1 Hz for DGEBA-MCDEA ($\square$) and PEI ($+$) are recalled for comparison.

T_{α}^{TP} in Table II) is governed by the accounted morphology in the modeling. As a matter of fact, by chosing the thermoset phase as the continuous phase (RVE-10), it can be observed a shift of both T_{α}^{TD} and T_{α}^{TP} toward higher temperatures compared to
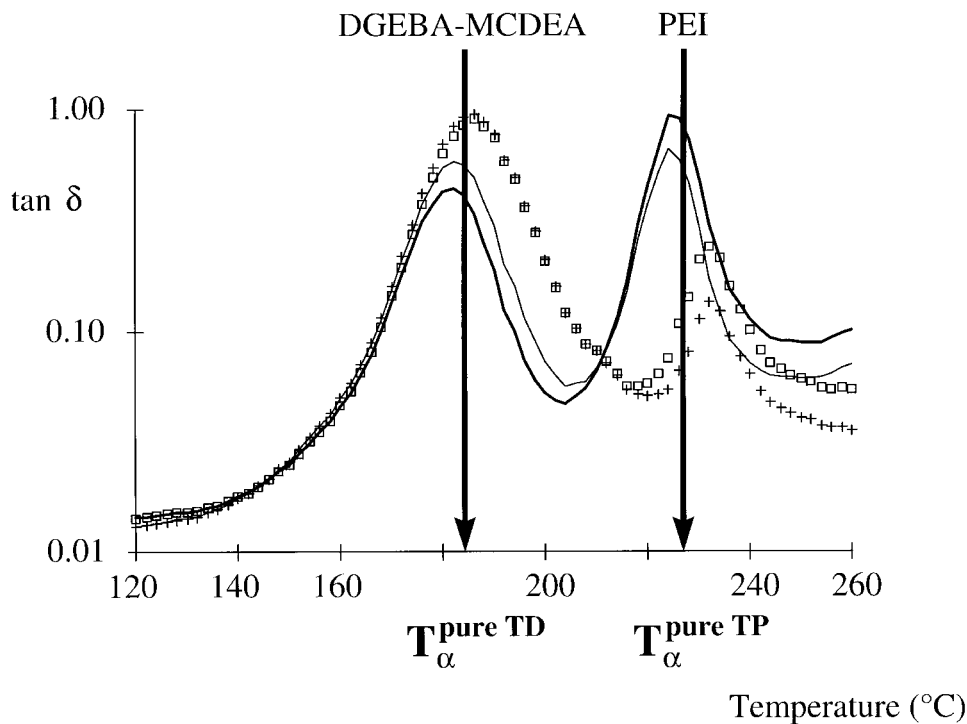
Figure 5 Theoretical evolution of the loss factor tan δ *versus* temperature at 1 Hz for binary blends containing 90 (+, —) or 80 ($\square$, —) vol % of DGEBA-MCDEA, which is considered in RVE as continuous phase (symbols, RVE-10) or dispersed phase (full lines, RVE-20).

T_α values of pure DGEBA-MCDEA network and pure PEI, respectively. In contrast, when modeling is carried out by considering the thermoplastic phase as the continuous phase (RVE-20), a significant shift of both T_α^{TD} and T_α^{TP} of blends toward the lower temperatures compared to the ones of pure components can be observed.

Figure 6(a) gives the evolution of $\Delta T_{\mathrm{DGEBA\text{-}MCDEA}} = T_\alpha^{\mathrm{TD}} - T_\alpha^{\mathrm{pure\ DGEBA\text{-}MCDEA}}$ as a function of the amount of DGEBA-MCDEA network in the blend for the continuous phase (RVE-10) or the dispersed phase (RVE-20) in mechanical modeling. By considering PEI either as the continuous phase (RVE-20) or the dispersed phase (RVE-10), Figure 6(b) gives the evolution of $\Delta T_{\mathrm{PEI}} = T_\alpha^{\mathrm{TP}} - T_\alpha^{\mathrm{pure\ PEI}}$ as a function of the amount of PEI in the blend. From Figure 6(a,b), it can be observed that evolution of ΔT *versus* the amount of phases follows opposite ways according to the phase chosen as the matrix in the modeling. Furthermore, it also can be seen that the change in the temperature location T_α of each phase is more significant when the considered component is chosen as the

dispersed phase. Thus, the magnitude of mechanical coupling effects between phases in polymer blends is predicted to depend on the geometric arrangement of the phases.

DISCUSSION

Experimental and Theoretical Analysis

Now, it is of interest to compare theory and experiment.

For the B10 sample, in which the thermoset component acts as the continuous phase, the theoretical approach predicts a shift of T_α^{TD} toward the higher temperatures. However, from experimental viscoelastic analysis, it was observed that T_α^{TD} in B10 sample is very close to T_α displayed by the pure DGEBA-MCDEA network. Recalling also that DSC measurements revealed a shift of T_g toward the lower temperatures for the thermoset phase in the blend (which is related to a decrease in the crosslinking degree of the epoxy network), it can be concluded that changes in microstructure and mechanical coupling effects
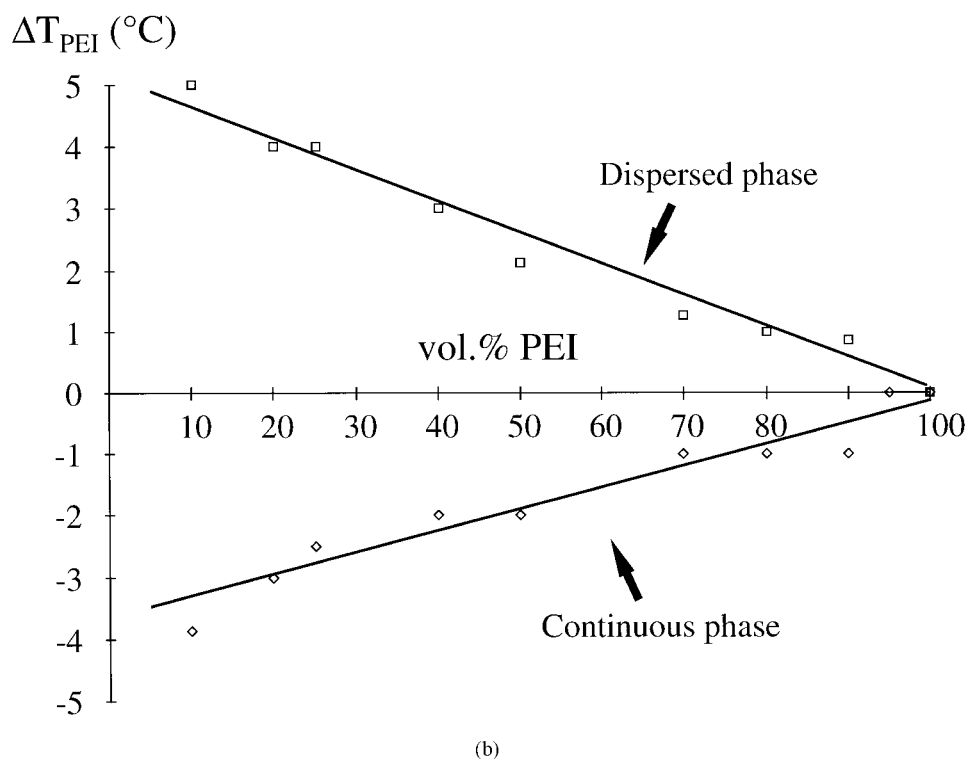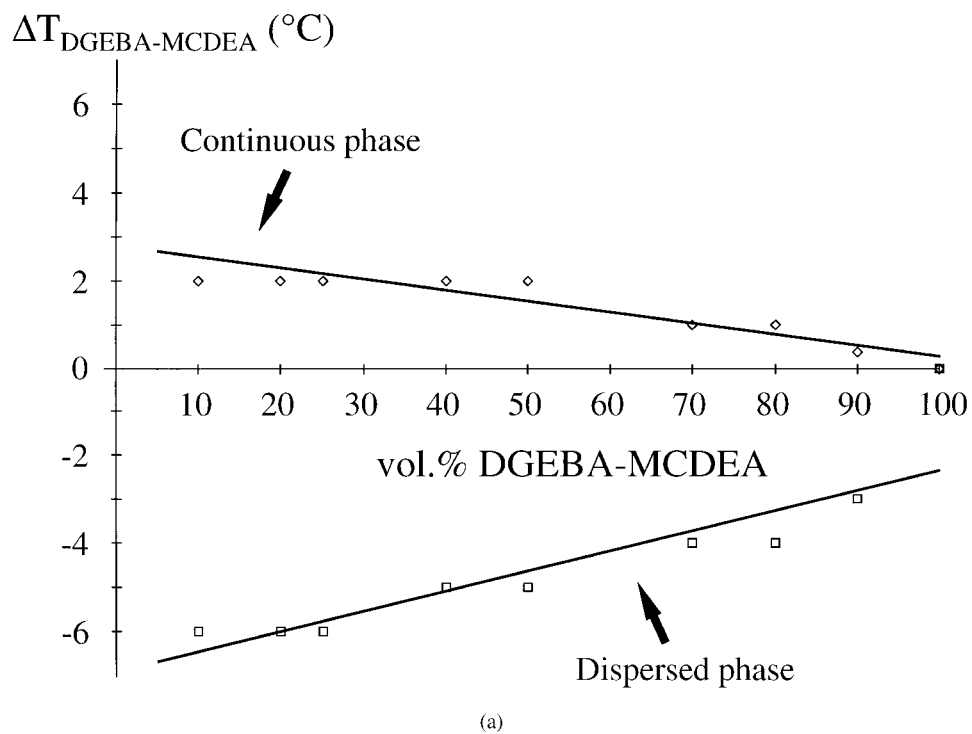
(a)

(b)

Figure 6 $\Delta T = T_\alpha$ (theoretical, blended component) $- T_\alpha$ (experimental, pure component) as a function of the amount of the component, which is considered in RVE as continuous ($\Diamond$) or dispersed phase ($\square$). (a) component is DGEBA-MCDEA network; (b) component is PEI.

act in opposite ways, leading to the weak change in the T_{α}^{TD} location in mechanical spectra.

For B20 sample, the thermoplastic phase acts as the continuous phase. In this case, mechanical modeling predicts a shift of T_{α}^{TD} toward the lower temperatures. Thus, changes in microstructure (revealed by DSC measurements) and mechanical coupling act here in similar ways, which leads to the significant shift toward lower temperatures observed in the T_{α}^{TD} location in mechanical spectra.

From the comparison between theoretical and experimental viscoelastic behavior displayed by binary polymer blends, it would be interesting to give the relative contribution of mechanical coupling effects and that of microstructural changes in the actual properties of the blends. Thus, by considering the PEI phase unchanged, the use of mechanical modeling in a "reverse" mode could allow access to the actual viscoelastic behavior of the DGEBA-MCDEA network in the blends. This type of reverse approach was first proposed[17] to investigate the actual viscoelastic behavior of an epoxy-anhydride network acting as the matrix in unidirectional fiber-reinforced polymers. Then, "reverse" mechanical modeling also was applied to predict the actual viscoelastic properties of the interphase component in a ternary compatibilized thermoset or thermoplastic blend.[22] However, such a reverse approach cannot be used for B10 and B20 samples because the nonevolution (after blending) of the viscoelastic properties of one of the components is required.

It is also of interest to show that both (1) experimental viscoelastic data of polymer blends and pure components and (2) mechanical modeling can be combined to probe the morphology of the polymer blends. Recently, Girard-Reydet et al.[20] have shown that the experimental ratio of the heights of loss peaks in binary polymer blends may be an appropriate empirical parameter to predict qualitatively the kind of blend morphology. Furthermore, numerous examples in literature[23–27] also reported that the knowledge of both evolution of dynamic shear modulus G_{d} (defined as $G^*(T, f) = G_{\mathrm{d}}(T, f) \times e^{i\delta(T,f)}$) and phase angle δ over a wide range of temperatures (T) and frequencies (f) can lead to the evaluation of morphological variation in a number of polymer blends. Thus, Eklind and Maurer[28] plotted δ versus G_{d} in order to explore the possibilities and limitations of determining the morphology of heterogeneous polymer blends by melt-state dynamic mechanical spectroscopy. They also compared experimental results with theoretical issues from

mechanical modeling to discuss morphology of blends. Such an approach also can be used for polymer blends at the solid state. In this way, we propose to compare the experimental ratio ($\Delta W/W$) of the dissipated energy (ΔW) and the total energy (W) on one cycle of strain versus G_{d} to the one issued by considering either the thermoset or the thermoplastic as the continuous phase in the blends. In Figure 7(a), the experimental evolution of $\Delta W/W$ [shown in ref. 29 as $\propto$ tan $\delta/(1 + \tan^2\delta)^{1/2}$] versus G_{d} for the B10 sample is compared to the ones theoretically determined by using mechanical modeling for the DGEBA-MCDEA network or PEI as the continuous phase. A good agreement between experiment and theory when the thermoset phase is chosen as the continuous phase can be observed. In contrast, for the B20 sample, a good agreement is found between experimental evolution and theoretical data when considering PEI as the continuous phase. Such results agree with the previous investigation of morphology by transmission electron microscopy,[20] and clearly shows that our approach can be an useful tool for describing the actual morphology of polymer blends.

CONCLUSIONS

In this paper, the viscoelastic properties of binary thermoset and thermoplastic polymer blends were investigated in connection with blend morphologies.

By accounting for the geometric arrangement of the polymeric phases in mechanical modeling, it was shown that

1. In the temperature range between the two main relaxations, whatever the binary blend composition can be, the overall prediction of the storage modulus of blend versus temperature follows mainly the one of the chosen continuous phase.

2. The magnitude of mechanical coupling effects on mechanical spectra of binary polymer blends is different when one component is considered in mechanical modeling either as the continuous phase or the dispersed phase. Thus, as for composite materials, the magnitude of mechanical coupling effects between phases in polymer blends is here predicted to depend not only on mechanical properties and relative content of each phase but also on the geomet-
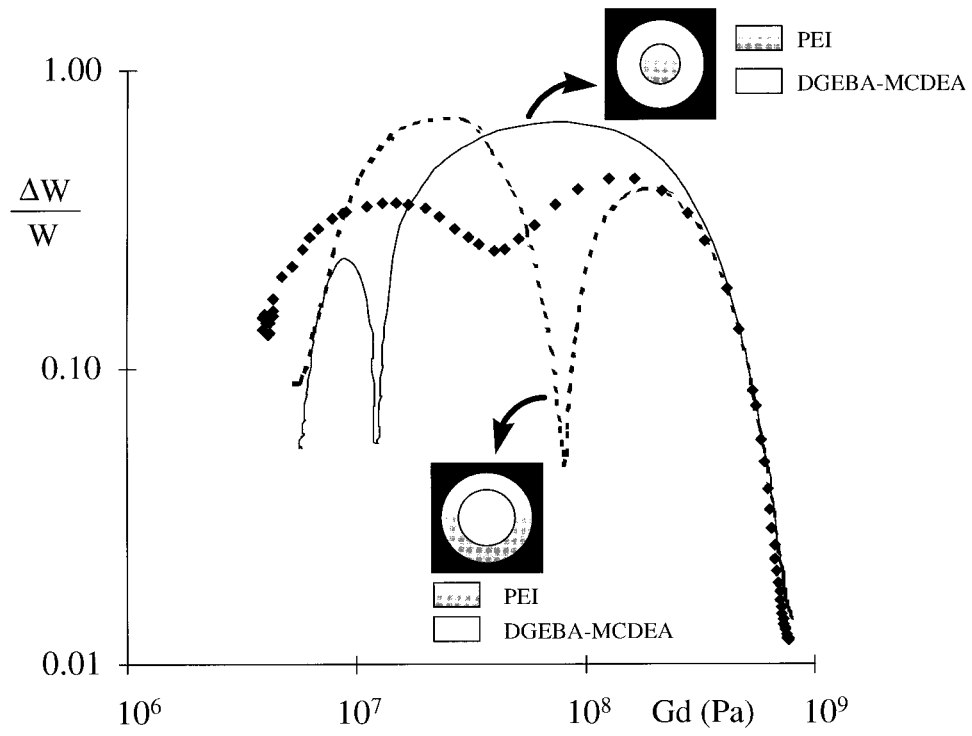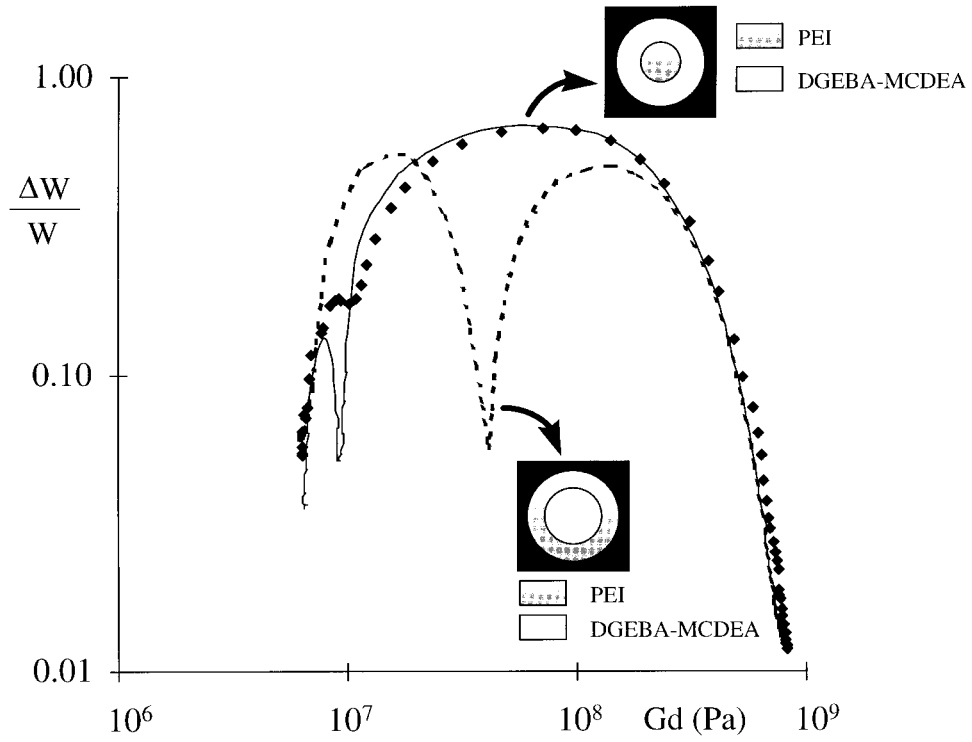
Figure 7 $\Delta W/W$ *versus* G_d plots from experimental (◆) and theoretical data given by mechanical modeling performed either with RVE-10 (full line) or with RVE-20 (dotted line). (a) B10 sample; (b) B20 sample.

ric arrangement of the phases accounted for in modeling.

Thus, both experimental results and the accurate knowledge of mechanical coupling effects predicted by using mechanical modeling led to the unequivocal conclusion of a decrease in the crosslinking degree of the epoxy network.

Lastly, the use of new graphs, that is, $\Delta W/W$ versus G_d, combining both theoretical and experimental data, can be a qualitative, well-suited probe of the actual blend morphology.

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