Rheological Study of the Miscibility of LLDPE/LDPE Blends and the Influence of T_{mix}

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The miscibility of LLDPE and LDPE blends and the influence of mixing temperature (T_{mix}) are discussed. Adequate amounts of antioxidants were added during melt blending. Dynamic and steady shear measurements were carried out at 190°C in a Rheometrics Mechanical Spectrometer 800. The dependence of rheological properties on blend composition indicated that blends of LLDPE (butene) and LDPE mixed at 190°C and 220°C are only partially miscible; immiscibilty is likely to occur around the 50/50 composition and in the LDPE-rich blends. Blends at 190°C are likely miscible in the LLDPE-rich range. Increasing T_{mix} did not improve the miscibility of LLDPE/LDPE blends at 190°C or influence the dynamic shear properties (η' or G') of the "pure" resins or blends. It is suggested that the molecular order [see Hussein and Williams, *J. Non-Newt. Fluid Mech.*, **86**, 105 (1999); Hussein and Williams, *Macromol. Rapid Commun.*, **19**, 323 (1998)] and mismatch of the molecular conformations of different polyethylene structures provide an explanation for the immiscibility of polyethylenes. Agreement was observed between the measured dynamic properties and theoretical predictions of Palierne and Bousmina-Kerner emulsion models.

INTRODUCTION

Blends of LLDPE and LDPE combine the favorable mechanical properties of the LLDPE with the ease of processing of LDPE. Yet, the melt rheological consequences of blending LLDPE and LDPE have received far less attention than have linear/linear or linear/branched blends (1). Previous reports showed that blends of LLDPE/LDPE were either immiscible or partially miscible (2–5).

Considering the components of these blends separately, melts of "pure" LLDPE are likely to phase separate as detected or suggested by different investigators (6–8). On the other hand, melts of LDPE possess a substantial amount of ordered phase as revealed by NMR studies of Bremner and Rudin (9). The existence of liquid-state molecular order and high-temperature transitions (208° and 227°C) in linear and lightly branched polyethylenes and their absence in highly branched LLDPEs was discussed in detail elsewhere (10–14). The existence of these high-temperature transitions prompted the extension of the miscibility study to cover the influence of the mixing temperature (T_{mix}) in the vicinity of those transitions.

We are not aware of any previous study that discussed the influence of T_{mix} on the miscibility of LLDPE/LDPE blends. However, Hill's group (15) presented experimental work with PE blends that indicated that a melt of 20% Linear PE and 80% LDPE is a single phase at 220°C and is biphasic at 170°C. Furthermore, DSC thermal analysis of a 50/50(w) blend of UHMWPE and normal MWPE showed one melting peak for blends mixed at 220°C or above (up to 280°C), suggesting miscibility, while two peaks (suggesting immiscibility) and melt segregation were obtained for blends mixed at 190°C (16).

For immiscible blend systems, the state of dispersion and specifically the shape of the dispersed phase (i.e. droplets) greatly influence the rheological responses of immiscible polymer blends. As a basic rule, phase separation causes G' and G'' to increase as a result of "emulsion morphology" present in phase separated systems. The emulsion morphology with consequent surface tension (α) gives rise to a non-zero storage modulus (G') even in a mixture of two Newtonian liquids.

Scholz *et al.* (17) presented a constitutive equation for dilute emulsions of spherical, monodisperse

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droplets of Newtonian liquids. The two liquids are assumed to be incompressible, and totally immiscible. In the linear viscoelastic range of deformation the dynamic viscosity (and loss modulus) are given by:

$$\frac{G''(\omega)}{\omega} = \eta' = \eta_m \left[1 + \phi \left(\frac{2.5k + 1}{k + 1} \right) \right] \quad (1)$$

where, η_m = the viscosity of the matrix liquid, $k = \frac{\eta_d}{\eta_m}$, η_d = the viscosity of the dispersed droplets, and ϕ = volume fraction of the dispersed phase.

Experimentally, $\eta'(\omega)$ obtained at low ω for immiscible polymer blends, other than PE/PE systems, is found to be higher than those of the components (17–21), as predicted by *Eq 1*. This principle was applied to differentiate between miscible and phase-separated polymeric blends. The high-ω data ($\omega > 10$ rad/s) is not generally useful for a similar interpretation of the miscibility of a blend (17).

Palierne (22) developed a general expression for the complex shear modulus of an emulsion of viscoelastic fluids, $G^*(\omega)$, in terms of the ratio α/R where α is surface tension. Expressions for G' and G'' are given elsewhere (23). Recently, Bousmina (24) extended Kerner's model for the modulus of composite elastic media (25) to an emulsion of viscoelastic phases undergoing deformations of small amplitude. Bousmina obtained a simple expression for $G^*(\omega)$ that gave predictions similar to those of Palierne. Expressions for G' and G'' for the Bousmina-Kerner model were obtained from Bousmina through a personal communication.

$$G' = \frac{[G'_m(AC + BD) - G''_m(BC - AD)]}{(C^2 + D^2)} \quad (2)$$

$$G'' = \frac{[G'_m(BC - AD) + G''_m(AC + BD)]}{(C^2 + D^2)} \quad (3)$$

where

$$A = (2 + 3\phi)\left(G'_d + \frac{\alpha}{R} \right) + 3(1 - \phi)G'_m \quad (4)$$

$$B = (2 + 3\phi)\, G''_d + 3(1 - \phi)\, G''_m \quad (5)$$

$$C = 2(1 - \phi)\left(G'_d + \frac{\alpha}{R} \right) + (3 + 2\phi)G'_m \quad (6)$$

$$D = 2(1 - \phi)G''_d + (3 + 2\phi)G''_m \quad (7)$$

and R = radius of the dispersed domains.

EXPERIMENTAL

The two polyethylene resins used in this study were supplied by Nova Chemicals Corporation. S229 (butene LLDPE) was a Ziegler-Natta product with M_w = 105 kg/mol and a density (ρ) of 918 kg/m³, while S216 (LDPE) had M_w = 100 kg/mol and ρ = 0.919 kg/m³. Branch content was the same in both resins (22 CH_3/1000 carbon atoms). More characterization

data including TREF (temperature rising elution fractionation) profiles are given elsewhere (10, 26). A commercial sample of atactic polystyrene (PS) was obtained from Dow Chemicals.

The LDPE and LLDPE resins used in this study were conditioned (or blended) in a Haake Rheocord 90 melt blender for 10 minutes at 50 rpm and either 190°C or 220°C. Blends of S229 with S216 as well as "pure" polymers were conditioned in the presence of at least 1000 ppm of additional antioxidant (AO). Adequate amounts of AO (up to 3000 ppm) were added to LLDPE conditioned at 220°C to prevent degradation (27). High performance liquid chromatography was used to confirm the presence of considerable active AO remaining at the end of the conditioning process. The AO was a 50/50 mixture of Irganox 1010 and Irgafos 168 made by Ciba-Geigy.

A Rheometrics Mechanical Spectrometer RMS 800 was employed for small-strain ω-sweep and *T*-sweep measurements of dynamic properties. A shear strain amplitude of 10% was used, after a strain sweep showed that this was in the linear viscoelastic range. The ω-sweep testing was performed at 190°C over the range $\omega = 10^{-2}$ to 10^2 rad/s. The *T*-sweep measurements were carried out in the range 260° to 160°C with all samples being heated to 260°C and then cooled to 160°C in steps of $\Delta T = 10°$ with a thermal soak of one minute. The ω-sweep tests were carried out using a cone-and-plate (CP) geometry with a cone angle of 0.1 radian and a 53 μm truncated apex. On the other hand, the *T*-sweeps were performed in parallel plates (PP) geometry with a gap spacing of 1.5 mm; a gap correction of 3 μm/°C was used to correct for the thermal expansion/contraction of the tools. For both geometries, 25-mm diameter platens were used. All measurements were conducted using nitrogen as the convection-heating medium to alleviate possible oxidative sample degradation during the sweep tests. The stability of the blends was established from time sweep measurements, $G'(t)$, at different temperatures and repeat measurements, as given in *Fig. 1a* for the 10% S229 blend with S216. Sample preparation and test procedure are explained elsewhere (11). *Figure 1a* verifies the reproducibility of our measurements and also the absence of sample degradation.

RESULTS AND DISCUSSION

While *Fig. 1a* represents testing at 190°C, properties were measured for LDPE and PS (used as reference for typical amorphous behavior) at different temperatures in the range 260°–160°C. Results for $\eta^*(T)$ are given in *Fig. 1b*. As expected for amorphous melts, the PS (dashed line) shows Arrhenius behavior over the whole range ($r^2 = 0.999$) with calculated activation energy of 17.4 kcal/mol (72.8 kJ/mol) in excellent agreement with previous literature reports (28, 29). On the other hand, the LDPE shows an obvious break in the curve near $T = 206°C$ ($1000/T \cong 2.09 K^{-1}$). The plot shows linear behavior above and below the break
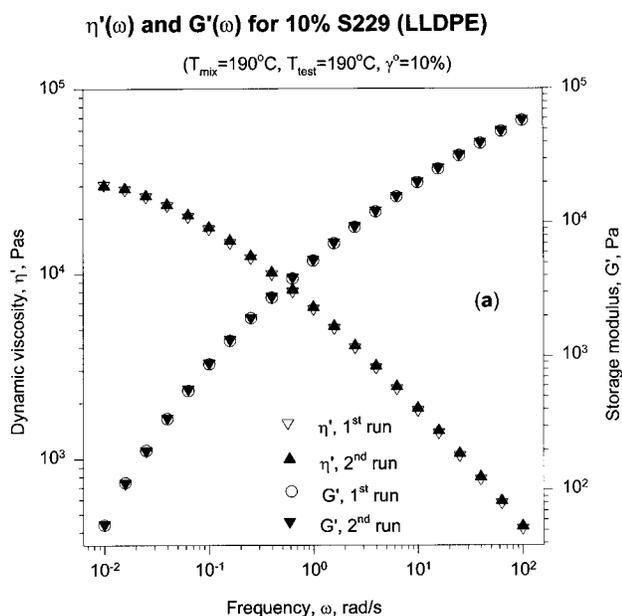
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η'(ω) and G'(ω) for 10% S229 (LLDPE)

(T$_{mix}$=190°C, T$_{test}$=190°C, γ°=10%)

Fig. 1a. *η'(ω) and G'(ω) for 10% S229 (LLDPE) blend (T$_{mix}$ = 190°C, T$_{test}$ = 190°C, γ° = 10%).*

Arrhenius plot of η*(1/T) for S216 (LDPE)

(T$_{mix}$=190°C, ω=1rad/s, γ°=10%)
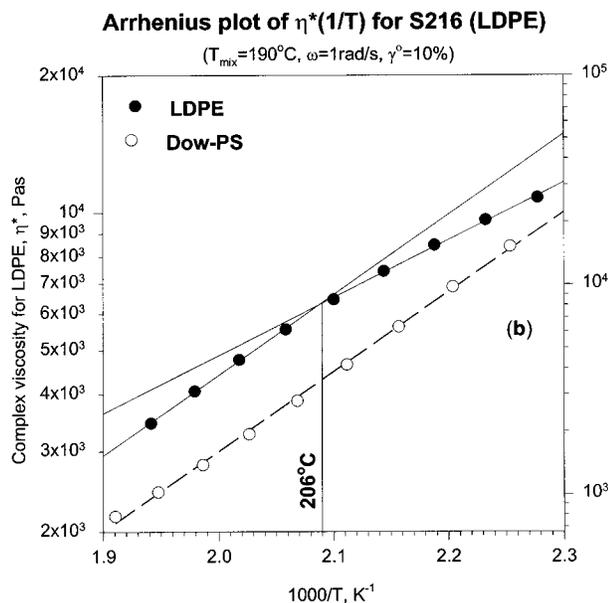
Fig. 1b. *Arrhenius plot of η*(1/T) for S216 (LDPE) and Dow-PS (T$_{test}$ = 260° to 160°C, ω = 1 rad/s, strain amplitude γ° = 10% for LDPE and 15% for PS).*

(transition point). Activation energies below, E_B, (160° < T < 206°C) and above, E_A, (206° < T < 260°C) were found to be E_A = 8.1 kcal/mol (r² = 0.997) and E_B = 5.8 kcal/mol (r² = 0.999). Fitting the whole collection of data above and below the transition by a straight line (as would happen for widely scattered data) resulted in an activation energy of 6.7 kcal/mol (r² =

0.999) in excellent agreement with earlier reports (30, 31) for LDPE. Furthermore, the transition in S216 was also detected by DSC (13). This raised the question about whether LDPE blends prepared above and below 206°C have different properties and miscibility.

In *Fig. 2a*, η'(ω) data are given for blends of LLDPE and LDPE mixed at 220°C. From this, we extract data from the low-ω regime to obtain the function η'(φ) shown in *Fig. 2b*. There, it is clear that η' for the LDPE-rich blends (low φ) exceeded the viscosity of the more viscous component (LDPE) suggesting the immiscibility of these blends and identifying emulsion-like rheology. Similar results were obtained for blends mixed at 190°C. Comparisons between η'(φ) for T_{mix} = 190°C and 220°C are given in *Fig. 2b* (ω = 0.1 rad/s) and a similar comparison for G' (φ) in *Fig. 2c* (ω = 1 rad/s). The measurements show a strong positive deviation behavior (PDB) from linear-additivity and log-additivity (not shown) in the LDPE-rich blends and around φ = 50%. Similar behavior was obtained when η' was plotted against stress (τ) with τ defined as τ = η* × ω (1). However, for the high-φ (LLDPE-rich) blends total (or partial) miscibility was observed, defined as near-linear η(φ) behavior. For example, results displayed in *Figs. 2b* and *2c* suggest the immiscibility of the 10% S229 blend mixed at 190° and 220°C and the miscibility or partial miscibility of the 90% S229 blend. The viscometric data cannot distinguish in a chemical sense between complete miscibility (single-phase behavior) and "partial miscibility" near φ = 1.0, where a very small concentration of tiny dispersed particles may exist.

The dilute emulsion model (17) was utilized to predict the viscosity of the 10% S229 and, hence, to explain the observed increase in the blend viscosity. The likelihood of the 10% S229 blend being a two-phase system is supported by the accurate predictions of the dilute emulsion model (shown as a line in *Fig. 2b*) at φ = 10%.

At high mixing temperature (220°C), the miscibility of blends was expected to improve; however, the η(φ) rheology of LLDPE/LDPE blends was very similar when mixed at 190° and 220°C (*Figs. 2b* and *2c*). These results are consistent with previous dynamic and steady shear measurements of Micic *et al.* (32) which showed PDB in LDPE-rich blends with LLDPE (for LDPE of similar LCB) tested at 190° and 220°C (T_{mix} was 230°C in their study).

To investigate further the miscibility at the two ends of the composition spectrum, steady shear measurements were performed. Utracki and Schlund (33) suggested that η' (ω) ≅ η(γ̇) for ω = γ̇ for miscible blends. The steady shear measurements were obtained in the CP geometry in the range γ̇ = 0.01 to 1.0 s⁻¹; 3 minutes of steady shearing were allowed after a γ̇-change to achieve steady state followed by 30 seconds of data acquisition, or measurement time.

To prepare for η' and η comparisons, and also to propose our view of viscosity as a function of stress, we display η'(τ) in *Fig. 3a*, covering both pure components and φ = 10%, 90%, and also both T_{mix}.

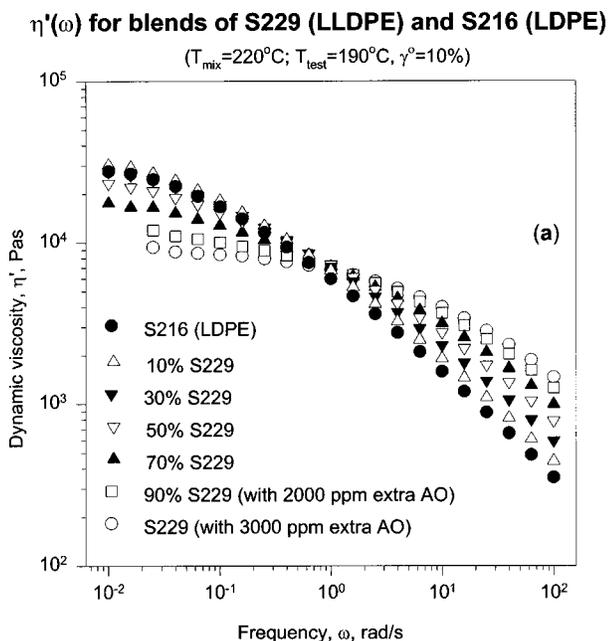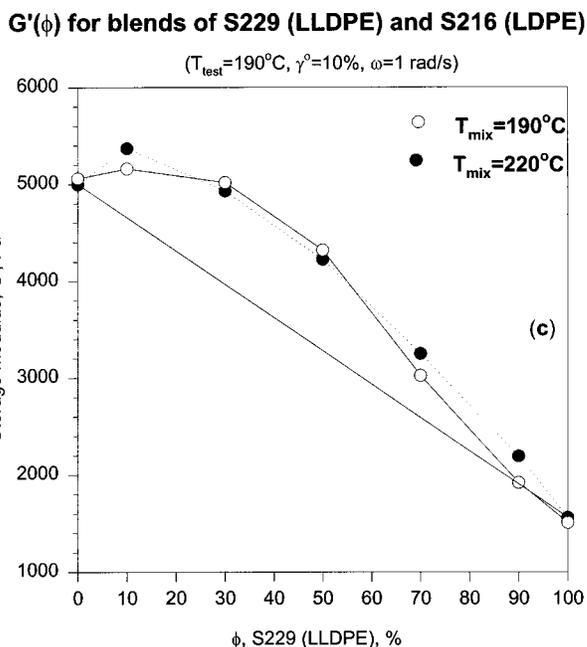## η'(ω) for blends of S229 (LLDPE) and S216 (LDPE)

(T_{mix}=220°C; T_{test}=190°C, γ°=10%)

- ● S216 (LDPE)
- △ 10% S229
- ▼ 30% S229
- ▽ 50% S229
- ▲ 70% S229
- □ 90% S229 (with 2000 ppm extra AO)
- ○ S229 (with 3000 ppm extra AO)

Fig. 2a. η'(ω) *for blends of S229 (LLDPE) and S216 (LDPE)* (T_{mix} = 220°C, T_{test} = 190°C, γ° = 10%).

η'(φ) for blends of S229 (LLDPE) and S216 (LDPE)

(T_{test}=190°C, γ°=10%, ω=0.1 rad/s)

dilute emulsion model (Eq. 1)

- ○ T_{mix}=190°C
- ● T_{mix}=220°C
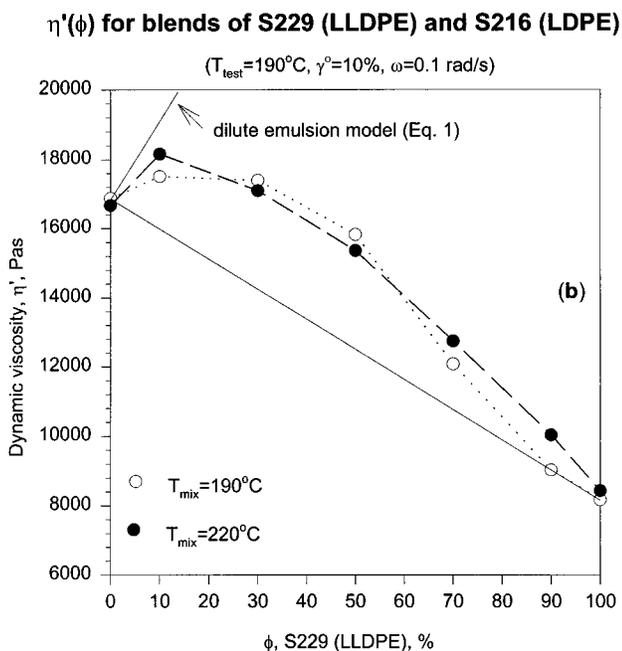
Fig. 2b. η'(φ) *for blends of S229 (LLDPE) and S216 (LDPE)* (T_{test} = 190°C, γ° = 10%, ω = 0.1 rad/s).

From *Fig. 3a*, we extract the dynamic shear measurements for the 10% and 90% S229 blends and display them in *Fig. 3b* as η'(τ) along with steady shear measurements η(τ). The 90% blend (LLDPE-rich blend, suggested above to be miscible) shows a very good degree of superposition of η'(τ) and η(τ) over the whole range of shear stress confirming the previous

G'(φ) for blends of S229 (LLDPE) and S216 (LDPE)

(T_{test}=190°C, γ°=10%, ω=1 rad/s)

- ○ T_{mix}=190°C
- ● T_{mix}=220°C

Fig. 2c. G'(φ) *for blends of S229 (LLDPE) and S216 (LDPE)* (T_{test} = 190°C, γ° = 10%, ω = 1 rad/s).

interpretations. On the other hand, the steady and dynamic shear results for the 10% S229 (LDPE-rich) blend, (suggested above to be immiscible) lack superposition. This comparison of steady and dynamic shear viscosities on selected compositions that represent the LLDPE-rich and LDPE-rich blends reinforce the findings of the previous methods of data treatment. Similar behavior was obtained (26) when the shear stress, τ, was replaced by the rate variables (ω or γ̇).

The above results seem consistent with theoretical findings of Fredrickson and co-workers (34–36) that acknowledge the important influence of the conformational and architectural "mismatch" between components of polyolefin blends on the miscibility of polyolefins. They found that adding a small amount of a highly branched component [usually contained in ZN-LLDPE (37)] to a lightly branched melt is more likely to cause liquid-liquid phase separation than vice versa (36). These theoretical findings explain why immiscibility is always observed when the highly heterogeneous ZN-LLDPE (19) is added to the less heterogeneous LDPE (see TREF profiles of S229 and S216 given in reference 2). Furthermore, our previous findings (11) showed the persistence of molecular order in polyethylene even at temperatures as high as 250°C, and this order should promote immiscibility in blends.

Predictions of *G'* and *G''* over a wide range of frequency, using the Palierne and Bousmina-Kerner emulsion models are given in *Fig. 4*, displaying the best fitting of T_{mix} = 220°C G* (ω) data, obtained with α/R = 10^3 N/m². The data at φ = 10% are deemed

Influence of T_{mix} on η' for blends of S229 (LLDPE) and S216 (LDPE)
(T_{test}=190°C, $\gamma°$=10%, ω=0.01-100 rad/s)
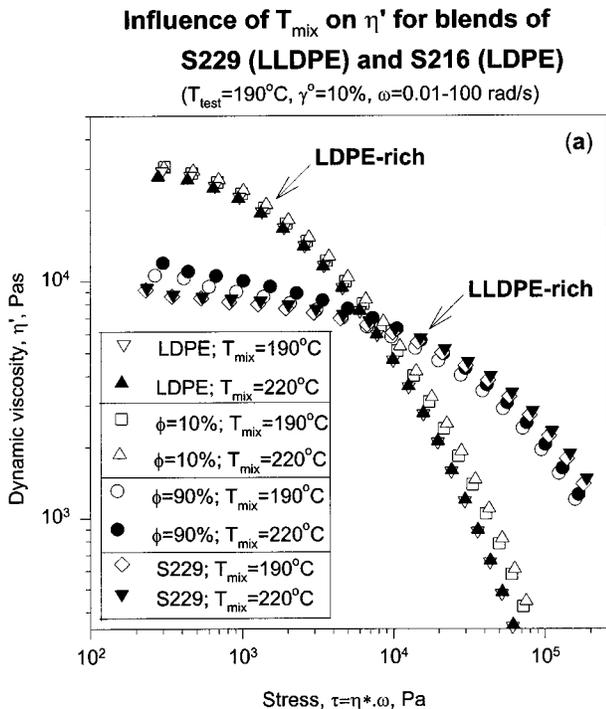
Fig. 3a. Influence of T_{mix} on η' for blends of LLDPE/LDPE (T_{test} = 190°C, $\gamma°$ = 10%, ω = 0.01–100 rad/s).

Comparison of $\eta(\tau)$ and $\eta'(\tau)$ for blends of S229 and S216
($\gamma°$=10%, ω=0.01-100 rad/s; $\dot{\gamma}$=0.01-1 s^{-1}, T_{mix}=190°C)
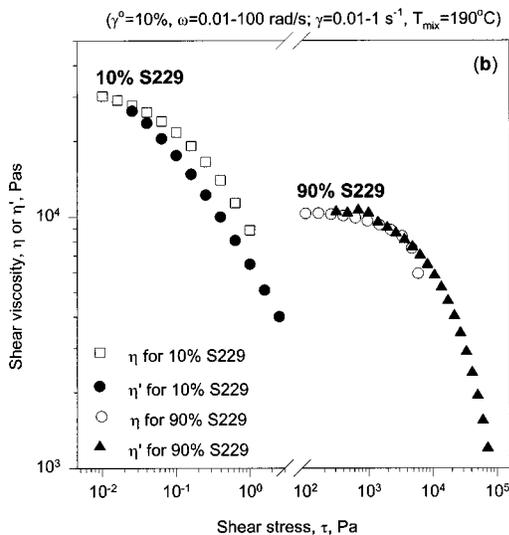
Fig. 3b. Comparison of $\eta(\tau)$ and $\eta'(\tau)$ for blends of S229 and S216 (T_{mix} = 190°C, T_{test} = 190°C, $\gamma°$ = 10%, ω = 0.01–100 rad/s, $\dot{\gamma}$ = 0.01 – 1 s^{-1}).

sufficient to represent the dilute condition for practical purposes. The two models give almost the same predictions of G' and G'' and show good agreement with experimental data over the whole frequency range. The value $\alpha/R = 10^3$ N/m^2 obtained here for LLDPE/LDPE blends is comparable to that reported

Predictions of Palierne and Bousmina models for the 10% S229 (LLDPE)
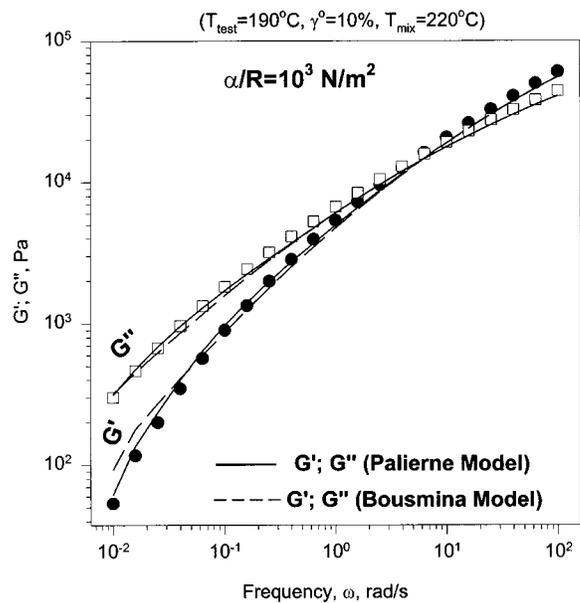(T_{test}=190°C, $\gamma°$=10%, T_{mix}=220°C)

Fig. 4. Predictions of Palierne and Bousmina-Kerner models for the 10% S229 blend (T_{mix} = 220°C, T_{test} = 190°C, $\gamma°$ = 10%, α/R = 10^3 N/m^2).

for known immiscible but compatibilized polymer blends (38). If the droplet phase has a size of $R \cong 1$ micron, then the order of magnitude of α is about 1 mN/m (1 dyne/cm) -very low, but consistent with an interface between two phases so chemically similar.

IMPLICATIONS FOR INDUSTRIAL PRACTICE

For immiscible blends, the nonlinear additivity of viscosity as a function of composition is accompanied by a similar behavior in elasticity-related capillary exit-pressure and normal stress, which is more pronounced at high shear stresses (39). For dynamic measurements, the $G'(\phi)$ displays similar behavior for blends of LLDPE/LDPE. The G' data obtained at T_{mix} = 190° and 220°C reinforced the previous findings of the immiscibility of the LDPE-rich blends and the insensitivity of miscibility/immiscibility to T_{mix}. In fact, $G'(\phi)$ plots are almost identical in shape to their counterparts of $\eta'(\phi)$ shown in *Fig. 2b* above.

Furthermore, the influence of T_{mix} on the miscibility of blends is examined by comparing the whole set of $\eta'(\tau)$ data obtained for "pure" resins and selected blends representative of LDPE-rich and LLDPE-rich blends. Results are shown in *Fig. 3a*. Again, $\eta'(\tau)$ for the 10% S229 blends exceeds that of "pure" resins up to $\tau = 7 \times 10^3$ Pa, while that of the 90% blend followed the linear additivity rule. For all blends, the high T_{mix} (220°C) did not force the immiscible (LDPE-rich) blends to become miscible. However, previous melt strength measurements of Micic *et al.* (32) showed that a high temperature together with a

strong flow field can resultin the miscibility of LLDPE in some LDPEs. In this study, high T_{mix} (220°C) accelerated the degradation of LLDPE (27), and higher amounts of AO were added at high T_{mix} to prevent this.

CONCLUSIONS

The miscibility of LLDPE (butene)/LDPE blends and the influence of mixing temperature were investigated. Dynamic and steady shear measurements and the different methods of data treatments suggest that:

1. Blends of LLDPE (butene) and LDPE mixed at 190°and 220°C are partially miscible. Immiscibilty is likely to occur around the 50/50 composition and in the LDPE-rich blends. Blends are likely miscible in the LLDPE-rich range.

2. The immiscibility of the blends leads to $\eta(\phi)$ that can be explained by emulsion models (e.g., dilute emulsion model, Palierne model, Bousmina-Kerner model). Although we have no direct morphological studies to document the presence of a multiphase structure in these melts, transmission electron microscopy by Hill and co-workers has detected such morphologies in a number of polyethylene blends (7, 15). Our rheological evidence and their work are supportive of each other. Other rheological signatures of phase-separated samples (e.g., failure of the Cox-Merz "rule") found in our studies are reported elsewhere (10, 26).

3. The complexity of the molecular mixture constituting ZN-LLDPE and the "mismatch" in the molecular conformation of LLDPE and LDPE are likely responsible for their immiscibility.

4. The mixing temperatures of 190° and 220°C did not influence the dynamic shear properties at 190°C and the miscibility/immiscibility of blends. However, degradation of LLDPE is accelerated by the high temperature (220°C) and extra AO is needed to prevent degradation.

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