**Summary:** The influences of short chain branching (SBC) on the melt miscibility of low-density polyethylene (LDPE) with linear high-density polyethylene (HDPE) were investigated by rheological methods. Two LDPE resins with different branch contents were blended with the same linear HDPE. Dynamic and steady shear measurements were carried out using a Rheometrics ARES rheometer at 190 °C. The rheology of the low-SCB LDPE (9 CH$_3$/1 000 C) blends with HDPE can be predicted by the linear additivity rule. Hence, blends were suggested to be completely miscible at all compositions. However, blends of the high branch content LDPE (SCB = 19 CH$_3$/1 000 C) were completely immiscible. Also, the different viscous and elastic properties of all the immiscible blends were much higher than the corresponding values for the more viscous and elastic blend component. The ratio of interfacial tension to droplet radius was estimated from Scholz et al. model as $\approx 1\,500$ N $\cdot$ m^{-2}. The level of SCB in LDPE was found to have a strong influence on its miscibility with linear HDPE.
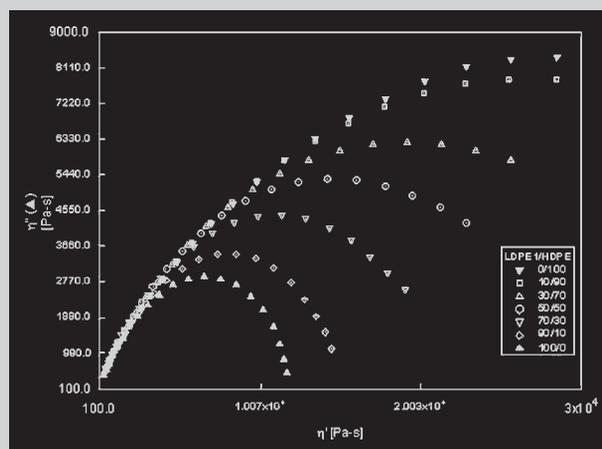
Cole-Cole plot for blends of LDPE1 with HDPE.

Effect of Short Chain Branching of LDPE on its Miscibility with Linear HDPE

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Introduction

High-density polyethylene (HDPE) has a linear structure with very little or no branching and possesses a high degree of crystallinity, but it is difficult to process. Low-density polyethylene (LDPE) however has a chaotic structure with short and long branches and is easy to process. Blending of polyethylenes (PEs) is a common industrial practice. The principal reasons behind the blending of PEs are property modification and improvement of processability.[1] The miscibility of different PE blends has been a subject of great interest for the past few decades[2−16] due to its direct impact on the processing and final properties of blends. PE blends were reported to be miscible, partially miscible or immiscible depending on molecular parameters such as: molecular weight ($\bar{M}_w$), molecular weight distribution (MWD), branch content (BC) and distribution and composition distribution (CD).[16−21]

Martinez-Salazar and co-workers[18,19] used differential scanning calorimetry (DSC) and correlated thermal properties of HDPE/LDPE systems to the branch content of LDPE. In addition, miscibility was suggested for blend components with BC less than 20 CH$_3$/1 000 C. Also, Alamo and co-workers[7,8] used small-angle neutron scattering (SANS) and found LDPE/HDPE blends to be completely miscible in the melt in the whole composition range. The LDPEs used in the SANS study had SCB and long chain branches (LCB) in the range 13−16 and 2−3 CH$_3$/1 000 C, respectively. However, the MWDs of the LDPEs were broader than those of the HDPEs. On the other hand, Hill and co-workers[2,6] used Transmission Electron Microscopy (TEM) and DSC and reported immiscibility of HDPE and LDPE blends in the melt state. The

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LDPE used in that study had 16 SCB and 10 LCB; however, this level of LCB is not common for commercial LDPE.

Further to this, Dobrescu[22] studied blends of HDPE and LDPE of different $\bar{M}_w$ using a capillary rheometer. The shear viscosity as a function of composition plot, $\eta(\phi)$ showed positive deviation behavior (PDB) relative to various linear mixing rules, such as the arithmetic "rule of mixtures", $\eta = \eta_A + \phi\eta_B$, and the log-additivity rule ($\log \eta = \Sigma \phi_i \log \eta_i$). The stronger the mismatch between the viscosities of the 'pure' polymers, the larger was the PDB. Deviations from the linear mixing rules for $\eta(\phi)$ have generally been taken to imply immiscibility. Also, Garcia-Rejon and Alvarez[23] reported the incompatibility (immiscibility) of HDPE/LDPE blends. They observed that low concentrations of HDPE (10%) had increased the elastic modulus, G', of LDPE by 50% (strong PDB). No details about the branching of LDPE were given.

Curto et al.[24] indicated good superposition of reduced viscosity as functions of shear rate, $\eta(\dot{\gamma})$, capillary flow data for a series of HDPE/LDPE blends at $T = 160\,^{\circ}\mathrm{C}$ to $200\,^{\circ}\mathrm{C}$, which was interpreted as support for likely miscibility (or stable morphology). Lee and Denn[14] and Cho et al.[20] reported miscibility of LDPE/HDPE blends in the melt state and immiscibility of the same blends in the solid state. The number of SCB of LDPE used in both studies was $32-34$ $CH_3/1\,000$ C; however, the $\bar{M}_w$ and MWD of the HDPE and LDPE were very different. Also, Fan et al.[21] used molecular dynamic (MD) simulations and immiscibility of LDPE/HDPE blends was suggested when the level of SCB of LDPE was higher than 30 branches/1 000 C.

The above literature review shows the basic disagreement regarding the miscibility of LDPE/HDPE blends as revealed by TEM, SANS, DSC, and rheometry. Even the findings from capillary rheometry[22,24] are contradictory. So far, the effect of $\bar{M}_w$, MWD, and SCB on the miscibility of LDPE/HDPE is not very clear since most of the previous studies used resins of high MWD and had large $\bar{M}_w$ variations among the blend components.[2−8,14,20] The interactions between these molecular parameters could be behind the disagreement between the different techniques. Also, little was published on the effect of SCB of LDPE and its influence on miscibility.[18,19,21] Still, the DSC and MD simulations hold opposing views.

Here, rheology was used to investigate the melt miscibility of HDPE/LDPE blends. The effect of SCB on miscibility was examined by blending two LDPEs with the same linear HDPE. The LDPEs had very different SCB, but were of the same LCB and $\bar{M}_w$ and of similar MWD. The two LDPEs represent the lower and upper range of the density of commercial LDPE. The aim of this investigation is to study the influence of SCB on the miscibility of LDPE with linear HDPE.

Experimental Part

Two LDPEs and one HDPE were used in this study. The resins were commercial polymers supplied as pellets by ExxonMobil Chemicals, Belgium. Table 1 provides characterization data such as density at room temperature, melt index (MI) at $190\,^{\circ}\mathrm{C}$ as provided by ExxonMobil. The molecular parameters of materials such as $\bar{M}_n$, $\bar{M}_w$ and MWD were measured by a WATERS GPC2000 instrument, using 1,2,4 tricholorobenzene (TCB) as a solvent at $150\,^{\circ}\mathrm{C}$. The GPC was calibrated with polystyrene standards. The degree of branching for the LDPE samples was evaluated from ^{13}C NMR. 15% w/w solutions of polymer in TCB were prepared in NMR tubes and an adequate amount of deuterated benzene was added. The spectra were recorded at 500 MHz and $135\,^{\circ}\mathrm{C}$. NMR data analysis followed the procedure outlined by Hansen et al.[25]

The two LDPE resins were blended with the same linear polyethylene in a Haake PolyDrive melt blender in the presence of an adequate amount of antioxidant.[13,26] The antioxidant was supplied by Ciba Specialty Chemicals and was a 50/50 mixture of Irganox 1010 and Irgafos 168. The LDPE with low SCB was designated as LDPE1 and the one with high SCB as LDPE2. Blends of 10, 30, 50, 70 and 90% by weight in addition to pure resins were prepared. Conditions of melt blending, the sample preparation procedure, and rheological testing are discussed elsewhere.[13] Results of a typical check for degradation are shown in Figure 1. Both dynamic viscosity, η', and elastic modulus, G', are given for the as-received and conditioned resin. It is clear that the presence of antioxidant has preserved the viscous and elastic nature of the resin.

Dynamic measurements and a few steady shear measurements were carried out using a Rheometrics ARES rheometer. In all tests, a cone and plate geometry with a cone angle of 0.1 rad and a plate diameter of 25 mm was used. The earlier work of Hussein et al.[26] suggested that a reproducibility test of rheological measurements should use samples prepared from different batches. In this study, a 50/50 blend of LDPE2 with HDPE was taken as an example. Results for η' and G' on samples obtained from two different batches are given in Figure 2. Excellent reproducibility of data was obtained.

Table 1. Characterization of resins.

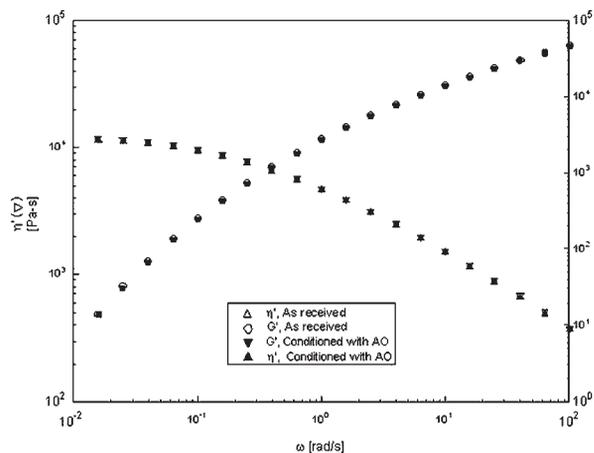| Resin	Density	MI	$\bar{M}_w \times 10^{-3}$	$\bar{M}_w/\bar{M}_n$	SCB/1 000 C	LCB/1 000 C
LDPE1	0.923	1.2	100	4.1	9	3
LDPE2	0.900	1.2	101	6.7	19	3
HDPE	0.961	1.2	116	6.50	0	0

Figure 1. Comparison of the rheology of as-received and conditioned LDPE1 ($T_{mix} = 190\,^\circ C$, $T_{test} = 190\,^\circ C$, $\gamma^\circ = 15\%$).

Figure 3. $\eta'(\omega)$ and $G'(\omega)$ for blends of LDPE1 and HDPE ($T_{mix} = 190\,^\circ C$, $T_{test} = 190\,^\circ C$, $\gamma^\circ = 15\%$).

Results and Discussion

Both dynamic and steady-shear measurements were performed on LDPE1 and LDPE2 blends with HDPE. In the dynamic shear measurements part, the blends were characterized by η' (or G'') and G' (or η'') as functions of composition, ϕ, and frequency, ω. For the steady shear mode, first normal stress difference, $N_1(\dot{\gamma}, \phi)$, was obtained.

For blends of LDPE1 (SCB = 9 CH$_3$/1 000 C) with HDPE results for $\eta'(\omega)$ and $G'(\omega)$ are given in Figure 3. Data are shown for the 10, 30, 50, 70, and 90% LDPE1 blends with HDPE as well as for the pure polymers. Filled triangles were used for pure polymers, while open symbols represent blends. In each Figure, the same symbol was used for the same composition, whenever applicable. The LDPE1 resin showed the lowest values for both $\eta'(\omega)$ and $G'(\omega)$ over the whole ω range. For all other blend compositions, both η' and G' were found to lie between the pure

resins. The increase in both η' (or G') for all blend compositions was found to follow the increase in the fraction of the more viscous (or elastic) component as shown in Figure 3. Hence, the rheology of the low-SCB LDPE1 blend with the linear HDPE suggests the miscibility of the blends.

For blends of LDPE2 (SCB = 19 CH$_3$/1 000 C) with the same linear HDPE, results of $\eta'(\omega)$ and $G'(\omega)$ are displayed in Figure 4. Over more than a decade, $\eta'(\omega)$ of the two blend components were very close and for all other blend compositions values of η' are higher than the more viscous component (HDPE). The same behavior was observed for $G'(\omega)$ over more than two decades of the low-ω data. The fact that G' at all blend compositions is much higher than any of the blend components is a result of emulsion rheology. Also, the increase in η' and G' of HDPE as a result of the addition of a low viscosity component (LDPE2) is a clear indication of the presence of multiphase systems. Similar results were obtained at low-ω for immiscible
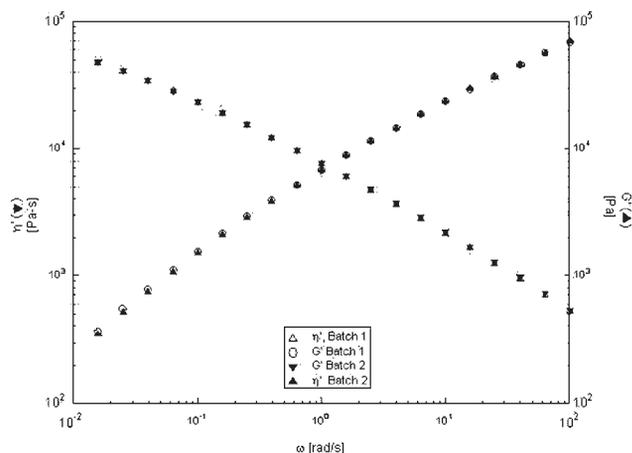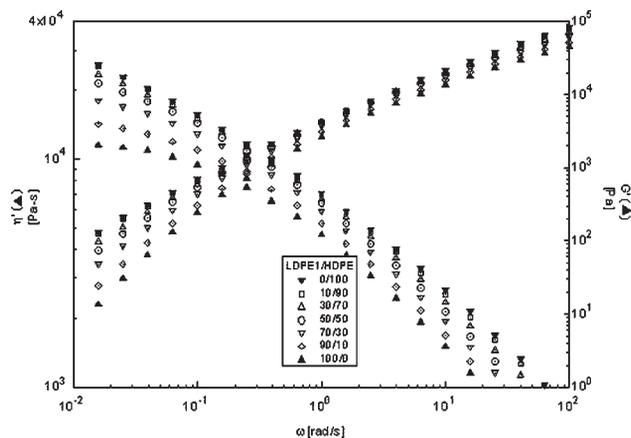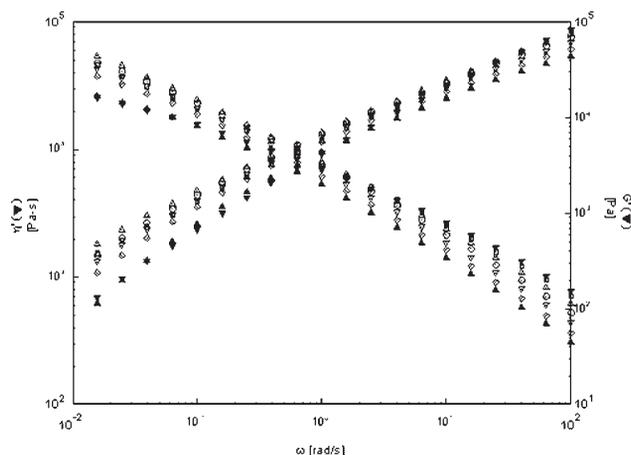

Figure 2. Reproducibility test: $\eta'(\omega)$ and $G'(\omega)$ for 50% LDPE2 blend with HDPE ($T_{mix} = 190\,^\circ C$, $T_{test} = 190\,^\circ C$, $\gamma^\circ = 15\%$).

Figure 4. $\eta'(\omega)$ and $G'(\omega)$ for blends of LDPE2 with HDPE ($T_{mix} = 190\,^\circ C$, $T_{test} = 190\,^\circ C$, $\gamma^\circ = 15\%$).

polymer blends, other than PE/PE systems.[27−31] At high ω ($\omega > 50$ rad $\cdot$ s^{-1}), the values of G' were intermediate between those of LDPE2 and HDPE, similar to previous observations by Scholz et al.[31] on the immiscible blend of polypropylene/polyamide 6. Hence, only the low-ω data were used for the interpretation of the miscibility of blends due to its sensitivity to changes in morphology.[30−32] These results suggest the immiscibility of LDPE2 blends with linear HDPE at all compositions.

Further, ARES software was used to find the zero shear viscosity, η_o, by fitting the $\eta'(\omega)$ data to the Cross model:

$$\eta = \frac{\eta_o}{[1 + (c_1 \dot{\gamma})^{c_2}]} \qquad (1)$$

where c_1 and c_2 are constants. Figure 5 shows $\eta_o(\phi)$ for blends of LDPE1 and LDPE2 with linear HDPE. For the low-SCB pair, $\eta_o(\phi)$ followed the linear additivity rule, while the high-SCB pair showed very strong PDB over the whole composition range with a peak at $\phi = 0.3$. The results of $\eta_o(\phi)$ for both pairs support the $\eta'(\omega)$ and $G'(\omega)$ data. Hence, the low-SCB LDPE1 blends are suggested to be completely miscible; however, the high-SCB LDPE2 blends with linear HDPE are deemed to be completely immiscible.

Likewise, $G'(\phi)$ at low ω ($\omega = 0.04$ rad $\cdot$ s^{-1}) were plotted in Figure 6. The results of $G'(\phi)$ data for the high-SCB LDPE2 blends with HDPE were almost identical to the previous results of $\eta_o(\phi)$. Again, a strong PDB was observed for blends of LDPE2 with HDPE with a maximum at $\phi = 0.3$. Also, blends of LDPE1 with HDPE followed linear additivity rule. In addition, the Scholz et al.[31] model for

dilute emulsions of noninteracting, spherical and monodisperse droplets of Newtonian liquids was used. $G'(\omega)$ data for the 10% and the 90% LDPE blends with HDPE were analyzed. In the linear viscoelastic range of deformation, $G'(\omega)$ for the emulsion was given by:

$$G'(\omega) = \frac{\eta_m^2 \phi}{80(\alpha/R)} \left(\frac{19k + 16}{k + 1} \right)^2 \omega^2 \qquad (2)$$

where η_m is the Newtonian viscosity of the matrix liquid, η_d is the Newtonian viscosity of the dispersed droplets, $k = \frac{\eta_d}{\eta_m}$, R is the radius of the dispersed domains, α is the surface tension between the two liquids and ϕ is volume fraction of the dispersed phase. The data in Figure 6 were used to predict α/R for the immiscible blend of LDPE2 with HDPE using the Scholz et al. model. The value of α/R for the 10% and the 90% blends of LDPE2 with HDPE were calculated as 1312 and 1545 N $\cdot$ m^{-2}, respectively. The value of α/R was similar to previous values reported for LLDPE/LDPE systems.[9,13] Also, the value of α/R obtained here was comparable with similar values reported for compatibilized polymer blends.[33] The presence of an interface is another support for phase separation of blends of the high-SCB LDPE2 with HDPE.

Furthermore, the dynamic shear data are presented in Cole-Cole plots, which are often used to analyze the miscibility of polymer blends.[20,30,31,34−36] Smooth, semicircular shaped Cole-Cole plots imply miscibility.[20,35] Plots of η'' vs. η' for the LDPE1 and LDPE2 blends with HDPE are shown in Figure 7a and 7b, respectively. The blends of LDPE1 with HDPE form semicircles of varying diameters and all the blends did lie between the pure resins.
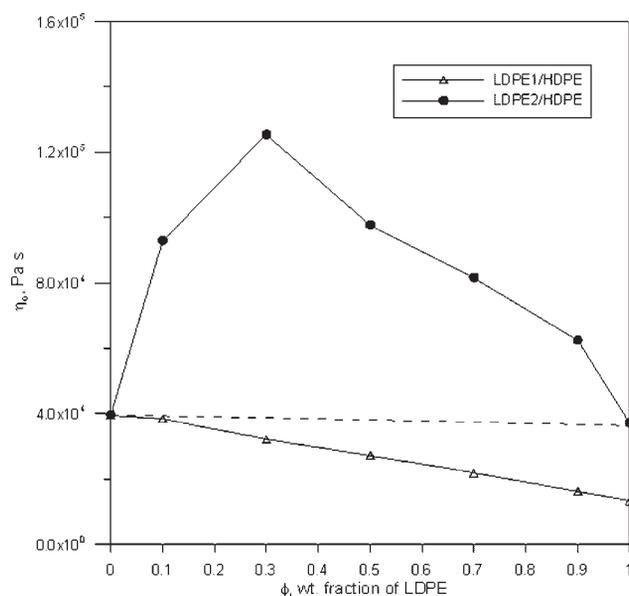

Figure 5. $\eta_o(\phi)$ For blends of LDPE1 and LDPE2 with HDPE ($T_{mix} = 190\,°C$, $T_{test} = 190\,°C$, $\gamma° = 15\%$).
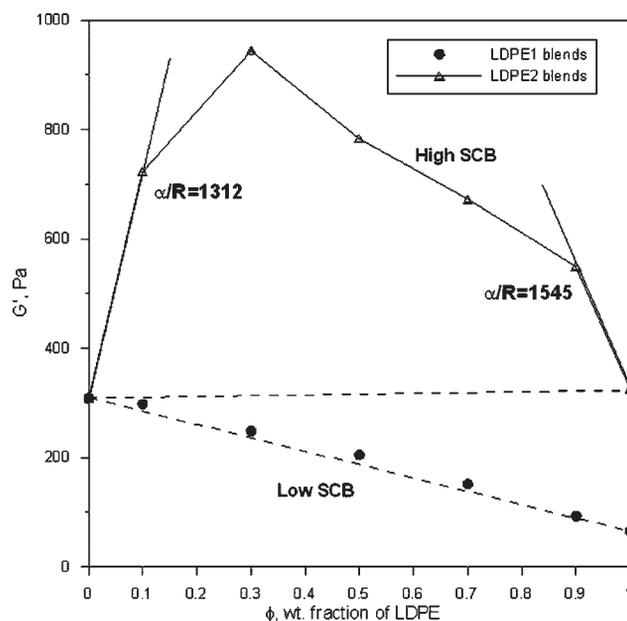
Figure 6. $G'(\phi)$ for blends of LDPE1 and LDPE2 with HDPE ($T_{mix} = 190\,°C$, $T_{test} = 190\,°C$, $\gamma° = 15\%$, $\omega = 0.04$ rad $\cdot$ s^{-1}).

a)

b)
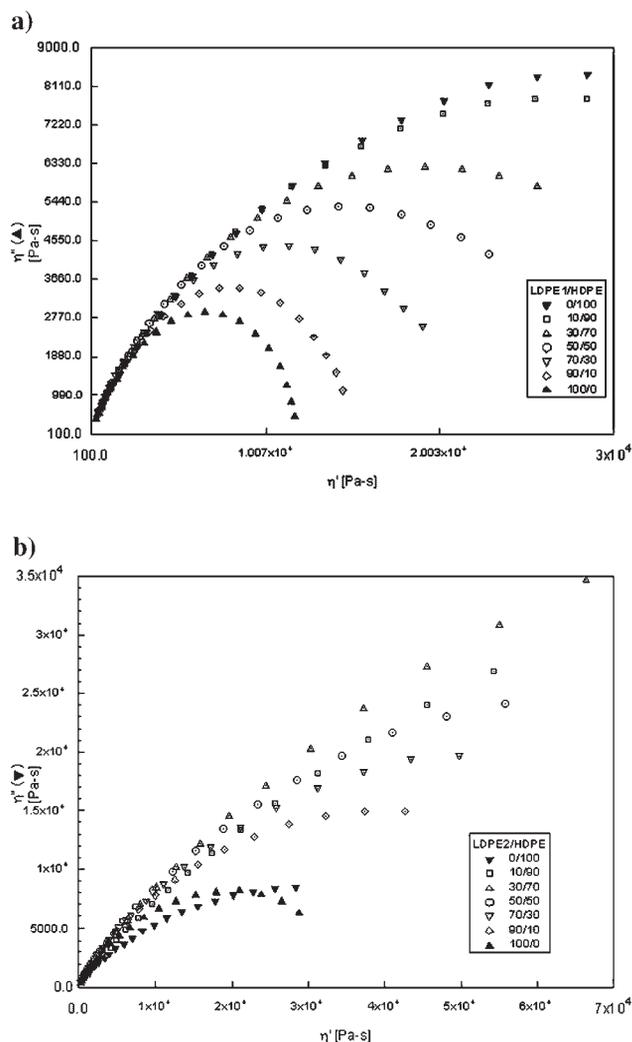
Figure 7.   a) Cole-Cole plot for blends of LDPE1 with HDPE ($T_{mix} = 190\,°C$, $T_{test} = 190\,°C$, $\gamma° = 15\%$). b) Cole-Cole plot for blends of LDPE2 with HDPE ($T_{mix} = 190\,°C$, $T_{test} = 190\,°C$, $\gamma° = 15\%$).

Also, plots of η'' were independent of composition at low η'. However, the results were quite different for blends of LDPE2 with HDPE. For all blends, η'' was higher than the pure resins. In addition, plots of η'' were found to be composition dependent. The Cole-Cole plots suggest the miscibility of the LDPE1 pair and the immiscibility of the LDPE2 pair.

In addition, steady shear measurements were performed on all blends using a cone-and-plate geometry. Transient measurements preceded the steady shear measurements for the determination of the delay time. For steady shear measurements, each data point was collected over a period of 30 s and then an average was reported. $N_1(\dot{\gamma})$ were measured and $N_1(\phi)$ data at low-$\dot{\gamma}$ were extracted and plotted in Figure 8. For the determination of the morphology of the blend, the low-$\dot{\gamma}$ is always preferred to avoid shear induced mixing.[37,38] The $N_1(\phi)$ results for LDPE1/HDPE pair are almost linear. On the other hand, the LDPE2/HDPE
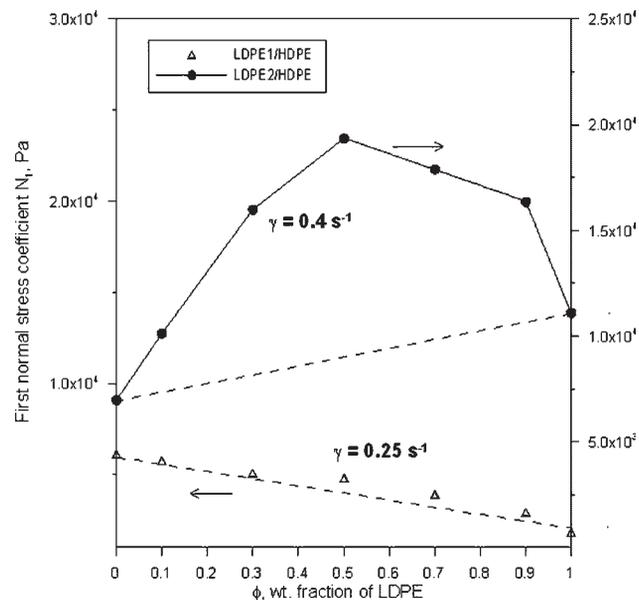

Figure 8. $N_1(\phi)$ for blends of LDPE1 and LDPE2 with HDPE ($T_{mix} = 190\,°C$, $T_{test} = 190\,°C$, delay time = 200 s).

blends showed a strong PDB at all compositions with a maximum around the 50/50 blend. The trend of the steady-shear data was similar to that of the dynamic shear results.

Thus, the different methods of presenting the dynamic and steady-shear data suggest the complete miscibility of the low-SCB LDPE (9 CH_3/1 000 C) in linear HDPE at all compositions. Also, all rheological data presented here imply the immiscibility of the high-SCB LDPE2 (19 CH_3/ 1 000 C) in HDPE at all compositions. The immiscibility of the blends can be predicted by rheological models based on the assumption of two-phase blends. This study suggests a significant role for SCB on miscibility. With regard to the level of SCB of LDPE required for phase separation, this study suggests that immiscibility of LDPE/HDPE blends is likely to take place if the level of SCB is high. Previously, different techniques have shown that the miscibility of linear HDPE and branched polyethylenes decreases with increasing branching of the branched component.[2−11,14,15,17−19,21] Therefore, the effect of branching of LDPE on its miscibility with HDPE is in general agreement with the general trend of previous reports.

However, the critical SCB or cut-off value for phase separation in the melt for LDPE/HDPE blends is suggested to lie between 9 and 19 SCB/1 000 C. This value of critical SCB is less than that suggested by SANS results of Alamo et al.[7,8] and the MD simulations of Fan et al.[21] The current results are however in agreement with the DSC findings of Martinez and co-workers.[19] It is important to note that the only major difference between the two LDPEs used in this investigation was the number of short chain branches (LCB, $\overline{M}_w$ and MWD were very close). The comparison of the current results with previous SANS and MD work is somewhat difficult since in the previous studies no

attempt has been made to isolate the interaction of the different molecular parameters. Hill and co-workers[2−6] used polymers with high MWD and wide variations of $\overline{M}_w$ among the blend components ($\overline{M}_w = 98\,000$; $\overline{M}_w/\overline{M}_n = 3.47$, LDPE: $\overline{M}_w = 208\,000$; $\overline{M}_w/\overline{M}_n = 8.22$). Also, the LDPE used had an exceptionally high degree of long chain branching (LCB) i.e. 10 LCB/1 000 C while the typical LCB in LDPE has been reported to be 2−3 LCB/1 000 C.[7,8,21] This high degree of LCB could be the reason for the partial miscibility in the HDPE-rich phase as observed by Hill and co-workers.[2,3] Similarly, Lee and Denn[14] used polymers with wide variations of $\overline{M}_w$ and MWD (HDPE: $\overline{M}_w = 40\,000$; $\overline{M}_w/\overline{M}_n = 3.1$, LDPE: $\overline{M}_w = 338\,000$; $\overline{M}_w/\overline{M}_n = 10$). The reasons for the miscibility/immiscibility of the PE/PE blend have been discussed in previous publications.[9,12,13,39,40] Recently, one of the authors studied the effect of SCB of metallocene LLDPE (m-LLDPE) on its miscibility with linear HDPE[9] and the results were qualitatively similar; however, the critical branch content for phase separation was different. The authors have also found that SCB has influenced the miscibility of m-LLDPE/LDPE blends.[40]

Conclusions

The low-branch content LDPE1 (SCB = 9 CH$_3$/1 000 C) was found to be miscible with the linear HDPE of the same molecular weight at 190 °C across the whole composition range. However, blends of the high-branch content LDPE2 (SCB = 19 CH$_3$/1 000 C) with the same HDPE were found to be immiscible at all compositions. For the miscible blend all rheological properties were predictable by the simple rule of linear additivity. However, the immiscible blends showed a strong PDB in the different rheological properties of blends as functions of composition. These rheological properties include: $\eta_o(\phi)$, $\eta'(\phi)$, $G'(\phi)$ $G^*(\omega)$, and N_1. Also, these viscous and elastic properties of the immiscible blends were much higher than the corresponding values for the more viscous and elastic blend component. Immiscibility can be predicted by dilute emulsion models such as the Scholz et. al. model. The ratio of interfacial tension to droplet radius (α/R) was $\approx 1\,500\,\mathrm{N\cdot m^{-2}}$ as computed from rheological model. The value of α/R is consistent with an interface between chemically similar polyethylenes.

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