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Miscibility of hexene-LLDPE and LDPE blends: influence of branch content and composition distribution

Ibnelwaleed A. Hussein[a],*, Tayyab Hameed[a], Basel F. Abu Sharkh[a], Khaled Mezghani[b]

[a]*Department of Chemical Engineering, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia*
[b]*Department of Mechanical Engineering, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia*

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Abstract

The influences of branch content (BC) and composition distribution (CD) of hexene linear low-density polyethylene (LLDPE) on its miscibility with low-density polyethylene (LDPE) were investigated. Ziegler–Natta (ZN) and metallocene-LLDPE (m-LLDPE) were used to study the melt miscibility using rheological tools. Dynamic, steady shear and transient measurements were carried out in an ARES rheometer at 190 °C. The miscibility was revealed by the dependence of their η_o, $\eta'(\omega)$, G', and $N_1(\dot{\gamma})$ on blend composition. The CD of LLDPE has influenced its miscibility with LDPE. The ZN–LLDPE blend with LDPE was found to be more miscible than an m-LLDPE of the same M_w and similar BC. On the other hand, a high-BC m-LLDPE (32.2 CH_3/1000 C) was found to be more miscible with LDPE than a low-BC m-LLDPE (14.4 CH_3/1000 C) of the same M_w and MWD. The high-BC m-LLDPE blends with LDPE were partially miscible and immiscibility is likely to develop in LDPE-rich blends. Agreement was observed between the measured rheology and theoretical predictions of Einstein, Scholz et al., Palierne, and Bousmina emulsion models.

Keywords: Polyethylene blend miscibility; Branch content; Composition distribution

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1. Introduction

Low-density polyethylene (LDPE) is usually blended with linear low-density polyethylene (LLDPE) to improve the processing of LLDPE. Their blends are widely used in film applications. So far, the blend miscibility of LLDPE and LDPE has received far less attention than blends of high-density polyethylene (HDPE) with LLDPE and LDPE [1–7].

Utracki and Schlund [8] found that a blend of LLDPE/LDPE was immiscible; however, other blends of LLDPE/LDPE were reported to be partially miscible [7,9–13]. Molecular parameters such as: molecular weight (M_w) and molecular weight distribution (MWD) were reported to have strong influence on miscibility of PE/PE blends [1–5, 11–15]. In addition, the details of molecular structure like branch content (BC) and composition distribution (CD) are important given that 'pure' Ziegler–Natta (ZN) LLDPEs can phase separate [16–18]. This raises questions about the use of ZN–LLDPE in most of the previous blend miscibility studies. It should be noted that ZN catalysis produces both linear and branched chains with more branches on smaller molecules [4,5,13,16–19]. The interactions of molecular parameters such as heterogeneity of structure and branch distribution make it very difficult to isolate the effects of BC and CD. The use of metallocene-LLDPE (m-LLDPE) is expected to isolate these interactions due to its intermolecular chain homogeneity [16,17]. Also, m-LLDPE has almost uniform distribution of branches [18].

The effect of BC on the miscibility of blends of branched (LLDPE and LDPE) and HDPE was the subject of many investigations [2–5,11,15,20–26]. The previous work has mainly used transmission electron microscopy (TEM) and small angle neutron scattering (SANS). Although, there exists a basic disagreement over the issue of miscibility of HDPE with LLDPE or LDPE, it was agreed that BC of branched component was the major molecular factor that controls miscibility of HDPE/LLDPE and HDPE/LDPE blends [2–5,22–26]. Hill and coworkers [2,20–22] used DSC and TEM and reported that 60 branches/1000 C are required for HDPE/LLDPE blends to phase separate. While

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* Corresponding author. Tel.: +966-3-860-2235; fax: +966-3-860-4234.
E-mail address: ihussein@kfupm.edu.sa (I.A. Hussein).

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Graesseley and coworkers [3,23] suggested a critical value of 40 branches/1000 C. Molecular dynamics simulations of Choi [24] also reported a critical value of 40 branches/1000 C. The effect of BC on the miscibility of LDPE/LLDPE blends is yet to be studied.

Also, the effect of CD (i.e. ZN–LLDPE vs. m-LLDPE) was found to have significant effect on the critical BC for liquid–liquid phase separation and co-crystallization of linear and branched polyethylenes [4,5,26,27] with ZN–LLDPE being more miscible in HDPE. However, the influence of CD on the miscibility of LLDPE/LDPE was not studied.

Blends of HDPE/HDPE and LDPE/LDPE of different molecular weight, M_w, fractions were reported to be miscible [6–8] and the viscosity vs. composition relationship followed the log-additivity rule. Plochocki [28] surveyed the rheology of various kinds of polyolefin blends, and Utracki [29] reviewed that of polyethylenes in particular.

In a recent study, the authors have investigated the effect of M_w and branch type of m-LLDPE on its miscibility with LDPE [14]. In this paper, the investigation is extended to study the effect of BC of LLDPE and CD (ZN vs. m-LLDPE) on the miscibility of LLDPE blends with LDPE using rheological tools.

As illustrated by Chuang and Han [30], rheological behavior of immiscible blends is strongly affected by the type of applied shear. While shear-induced mixing is observed in some steady shear experiments [31,32], no such effects are reported for small amplitude dynamic shear measurements [30,33]. The storage and loss moduli G' and G'' exceed values for the matrix phase, due to the presence of droplets of the dispersed phase [34,35].

Choi and Schowalter [36] derived expressions for G' and G'' as functions of frequency, ω, for an emulsion of two Newtonian liquids, which can also be applied to a phase separated polymeric system in the low-ω Newtonian regime [37,38], where the G' had increased due to the presence of droplets. Scholz et al. [37] derived a constitutive equation for dilute emulsions of non-interacting, spherical and monodisperse droplets of Newtonian liquids.

At low ω, $G'(\omega)$ for immiscible polymer blends was reported to be higher than those of the pure components as obtained experimentally for blends other than PE/PE systems [30,37–40]. Ajji and Choplin [41] found that effects of phase separation on G' were more pronounced than on G''. Scholz et al. [37] reported that the values of the dynamic moduli at high ω ($\omega > 10$ rad/s) for the immiscible blend of PP/Polyamide 6 were intermediate between those of PA6 and PP. Hence, the high-ω data were *not* used for the interpretation of the miscibility of blends.

The matrix of resins used as blend components was designed to study one variable at a time. In this second paper, only the effect of BC and catalyst type will be discussed. Two m-LLDPE resins and one LDPE were selected to study the effect of BC. A high-BC m-LLDPE (32.2 CH_3/1000 C) and a low-BC m-LLDPE (14.4 CH_3/1000 C) were blended with the same LDPE. The effect of the m-LLDPE BC was investigated by comparing blends of two hexene-based m-LLDPEs of the same M_w and MWD with the same LDPE. Similarly, the effect of catalyst type was examined while keeping M_w, MWD and average BC of LLDPE constant.

2. Experimental

Two commercial samples of hexene m-LLDPE, one ZN–LLDPE, and one LDPE were obtained from ExxonMobil. Characterization data such as density at room temperature, melt index (MI) at 190 °C as provided by ExxonMobil was given in Table 1. The number-average and weight-average molecular weights as well as MWD were obtained by a gel permeation chromatography (GPC). GPC data was collected using 1,2,4-trichlorobenzene as solvent at 150 °C in a WATERS GPC2000 instrument. Polystyrene standards were used for calibration. The label 1 denotes the m-LLDPE with low BC as m-EH1 and the resin with high BC was labeled as m-EH2. The same LDPE was used in all of these blends. Blends of 10, 30, 50, 70, and 90% by weight LLDPE were studied. The effect of LLDPE BC on its miscibility with LDPE was investigated by studying blends of m-EH1 and m-EH2 with LDPE. Samples m-EH1 and m-EH2 were chosen in a way that BC is the *only* variable in this comparison. As shown in Table 1, m-EH1 and m-EH2 have the same branch type (hexene), M_w, and both LLDPEs are metallocene resins. Hence, comparison of m-EH1 and m-EH2 blends with LDPE will reveal the effect of BC on the miscibility of m-LLDPE/LDPE systems. On the other hand, comparison of m-EH1 and ZN–EH blends with LDPE will disclose the influence of catalyst type or CD on the miscibility of LLDPE/LDPE blends.

The LDPE, all the LLDPE resins, and their blends were conditioned (or blended) in a Haake PolyDrive melt blender in the presence of 1000 ppm of additional antioxidant to avoid polymer degradation during melt blending [42]. Test for degradation during melt blending was performed for all pure resins. Excellent agreement of dynamic viscosity, η', and elastic modulus, G', data (results are not shown here) for conditioned and as-received resin indicated absence of degradation during melt blending. Conditions of melt blending, sample preparation, and rheological testing in ARES rheometer were reported in previous publications [12,14]

3. Results and discussion

Results (not shown here) for η' and G' on samples obtained from two different batches showed excellent reproducibility of the rheological measurements. Steady-shear testing was always conducted with a fresh sample. In

Table 1
Characterization of resins

Resin	Density (g/cm^3)	MI (g/10 min)	M_w (kg/mol)	PD = M_w/M_n	Branch content CH$_3$/1000 C
m-EH1	0.912	1.2	102	2.14	14.4
m-EH2	0.883	2.20	97	2.02	32.2
ZN–EH	0.917	2.80	107	4.01	16.7
LDPE	0.923	1.2	100	4.14	11[a]

[a] Total number of short and long branches.

cone-and-plate tests, data on $\eta(\dot{\gamma})$ and $N_1(\dot{\gamma})$ were obtained. Testing procedures were the same as that given in a recent publication [14].

3.1. Influence of branch content

In Fig. 1, results of $\eta'(\omega)$ are given for blends of m-EH1 (BC = 14.4) with LDPE. Data were shown for the 10, 30, 50, 70, and 90% blends of m-EH1 with LDPE as well as for the pure resins. A Newtonian plateau that extended over a period of about two decades was observed for m-EH1. Also, the m-EH1 resin showed the lowest values for both $\eta'(\omega)$ and $G'(\omega)$ in comparison with LDPE and other blends.

As mentioned earlier, the low-ω data is especially sensitive to changes in the morphology and hence will be used for the interpretations of the blend miscibility. At low-ω ($\omega < 10$ rad/s), the increase in η' for the 70 and the 90% blends of m-EH1 with LDPE are found to lie between those of the pure resins. However, $\eta'(\omega)$ for all other blends are either equal to or greater than that of the more viscous component (LDPE). Also, for the $G'(\omega)$ data (not shown) the elastic modulus of the 70 and 90% blends of EH1 with LDPE are located between the pure resins. For the 10, 30, and 50% blends of EH1 with LDPE, $G'(\omega)$ data were either slightly lower or slightly higher than the more elastic component (LDPE). This shows that the addition of a small amount of the less viscous component (m-EH1) to the more viscous component has resulted in a slight increase in viscosity rather than a decrease. As discussed earlier, this observation is an indication of emulsion morphology (or immiscibility). The same observation about viscosity could be extended to the elasticity data, $G'(\omega)$. Miscibility is expected to increase both viscosity and elasticity of the blend due to the presence of the interface.

Likewise, $\eta'(\omega)$ data for the high-BC blend of m-EH2 (32 CH$_3$/1000 C) with LDPE were displayed in Fig. 2 The high-BC pair showed a Newtonian plateau over more than two decades, which is a result of low MWD. In the low-ω range, the viscosities of the pure components bound the viscosities of all blends as shown in Fig. 2. Here, the addition of the less viscous component (m-EH2) to the more viscous component (LDPE) has resulted in a decrease in the viscosity of the blend. Also, $G'(\omega)$ data of all blends followed a similar fashion. However, the 10 and 30% m-EH2 blends with LDPE were slightly higher than expected. These results suggest a high degree of miscibility of m-EH2 blends with LDPE.

Further, the $\eta'(\omega)$ data for blends of m-EH1 and m-EH2 with LDPE were fitted to cross model using ARES Orchestrator software to obtain zero shear viscosity, η_o.

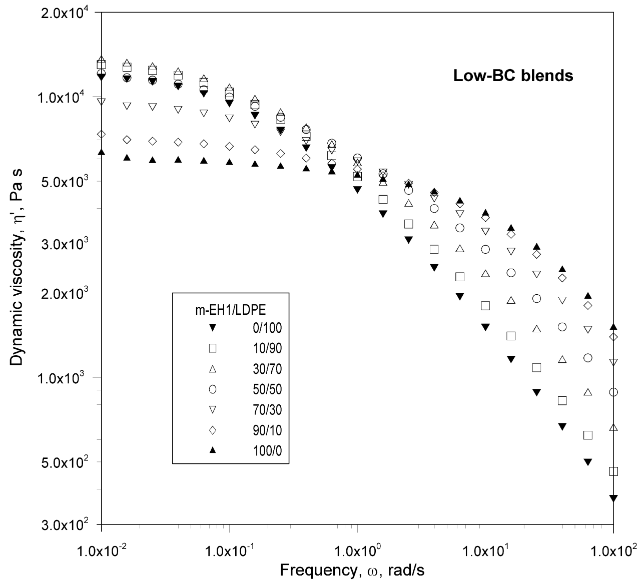

Fig. 1. $\eta'(\omega)$, for blends of m-EH1 and LDPE ($T_{mix} = 190$ °C, $T_{test} = 190$ °C, $\gamma^o = 15\%$).

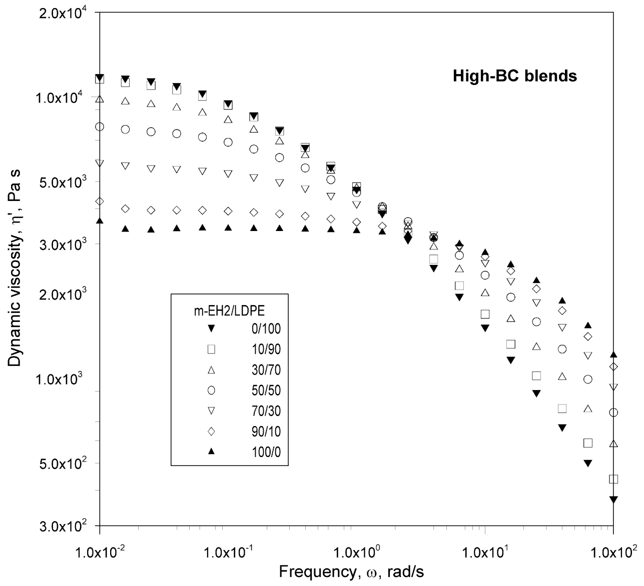

Fig. 2. $\eta'(\omega)$, for blends of m-EH2 with LDPE ($T_{mix} = 190$ °C, × $T_{test} = 190$ °C, $\gamma^o = 15\%$).

Results for m-EH1 and m-EH2 blends with LDPE are presented in Fig. 3(a) and (b), respectively. The plot of $\eta_o(\phi)$ for the low-BC blends, given in Fig. 3(a), shows a strong positive deviation behavior (PDB) from log-additivity for most of the blends specially in the LDPE-rich blends range. Hence, these blends were suggested to be immiscible. To determine the degree of immiscibility, Einstein model $[\eta_b = \eta_m(1 + 2.5\phi)]$ was used, where η_b and η_m were viscosity of the blend and matrix, respectively. This model represents an upper limit for emulsion systems. Model predictions for the 10% blend, shown as a solid line, suggested a very high degree of immiscibility for the 10% m-EH1 blend with LDPE. However, the degree of immiscibility was low in m-EH1-rich blends. The findings from $\eta_o((\phi)$ data support the previous results of $\eta'(\omega)$ and $G'(\omega)$ (see Fig. 1).

On the other side, $\eta_o((\phi)$ for the high-BC blends are given in Fig. 3(b). The results are different than that of the low-BC blends. The plot showed weak PDB from log-additivity rule. Also, the deviation of the 10% m-EH2 blend with LDPE from linear additivity rule is much lower than the predictions of the Einstein model. This suggests a better of miscibility of the high-BC m-EH2 blends with LDPE. The results of $\eta_o((\phi)$ data did support the previous findings from $\eta'(\omega)$ and $G'(\omega)$ (see Fig. 2).

Also, $\eta'(\phi)$data for blends of m-EH1 and m-EH2 with LDPE at low ω near-Newtonian regime ($\omega = 0.1$ rad/s) are plotted in Fig. 3(c) and (d), respectively. Data in Fig. 3(c)

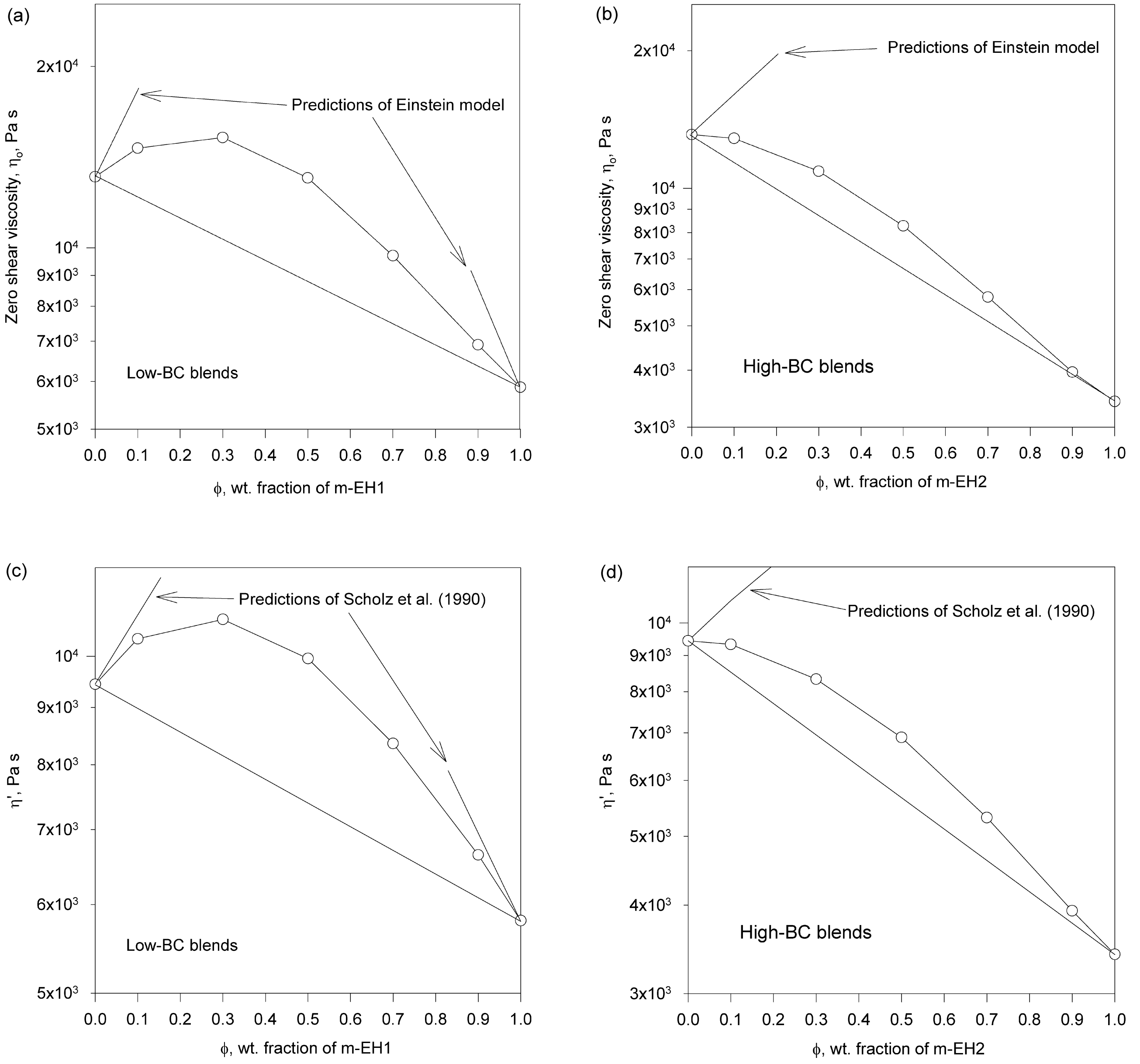

Fig. 3. (a) $\eta_o(\phi)$ for blends of m-EH1 with LDPE computed from cross model ($T_{mix} = 190$ °C, $T_{test} = 190$ °C, $\gamma^o = 15\%$); (b) $\eta_o(\phi)$ for blends of m-EH2 with LDPE computed from cross model ($T_{mix} = 190$ °C, $T_{test} = 190$ °C, $\gamma^o = 15\%$); (c) $\eta'(\phi)$ for blends of m-EH1 with LDPE ($T_{mix} = 190$ °C, $T_{test} = 190 \times$ °C, $\omega = 0.1$ rad/s); (d) $\eta'(\phi)$ for blends of m-EH2 with LDPE ($T_{mix} = 190$ °C, $T_{test} = 190$ °C, $\omega = 0.1$ rad/s).

support the previous results and suggest a high degree of immiscibility of the low-BC m-EH1 blends with LDPE. Likewise, the $\eta'(\phi)$ data (Fig. 3(d)) obtained at the same frequency followed the previous trends of $\eta_o(\phi)$ data shown in Fig. 3(b). Here, the predictions of Scholz et al. emulsion model [37] of η' (Eq. (1)) are displayed for the 10% m-EH2 blend with LDPE in Fig. 3(c).

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

where, η_m = the viscosity (Newtonian) of the matrix liquid; η_d = the viscosity (Newtonian) of the dispersed droplets; $k = \eta_d/\eta_m$.

It is clear that the deviations from model predictions were large. All of these observations suggest a high degree of miscibility of the high-BC m-EH2 blends with LDPE.

Further, the $G'(\phi)$ data obtained at low ω ($\omega = 0.04$ × rad/s) were examined. Results for the low-BC blends were given in Fig. 4. The data suggest the immiscibility of the 50/50 and the LDPE-rich blends. The m-LLDPE-rich blends showed more miscibility. The 90% EH2 blend with LDPE was suggested to be miscible. For the 10% blend of the low-BC pair, predictions of Scholz et al. model (Eq. (2)) were shown as a solid line in Fig. 4.

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

where R = radius of the dispersed domains and α = surface tension between the two liquids. The experimental data and model predictions were in excellent agreement and suggest the complete immiscibility of the 10% m-EH1 blend with LDPE. Hence, Eq. (2) was used to calculate the ratio α/R from Scholz et al. model [37]. The value of (α/R) was calculated as ~ 470 N/m^2. Palierne and Bousmina models [43,44] provided similar predictions.

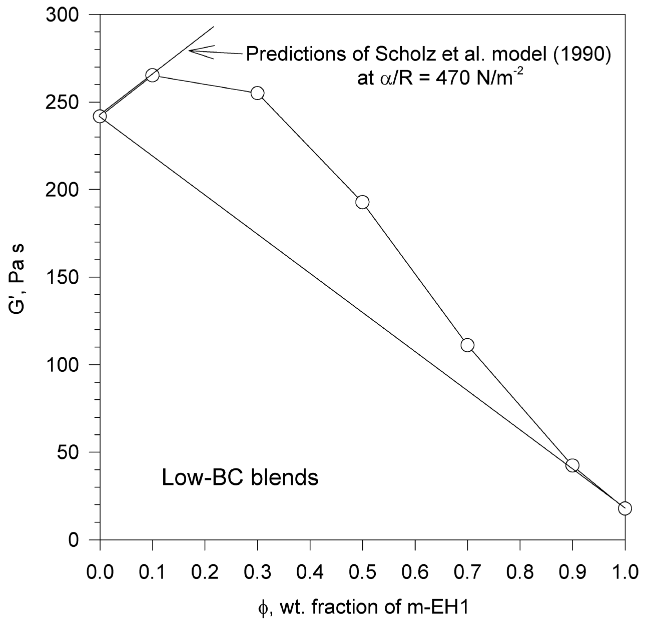

Fig. 4. $G'(\phi)$ for blends of m-EH1 with LDPE ($T_{mix} = 190$ °C, × $T_{test} = 190$ °C, $\gamma^\circ = 15\%$, $\omega = 0.1$ rad/s).

The LDPE-rich blends were likely to be immiscible, while the m-EH1-rich blends were likely miscible. At high-ω, the linear additivity was followed. For $\omega > 10$ rad/s, dynamic shear data is insensitive to morphology similar to the previous observations of Scholz et al. [37] and Hussein [45].

In addition, steady shear measurements were carried out on m-EH1 and m-EH2 blends with LDPE using the same cone-and-plate geometry. Results of $N_1(\dot\gamma)$ for the low-BC (m-EH1) and the high-BC (m-EH2) blends were shown in Fig. 5(a) and (b), respectively. Data are given for m-EH1 blends with LDPE in the range $\dot\gamma = 0.4$–4 s^{-1}, and for m-

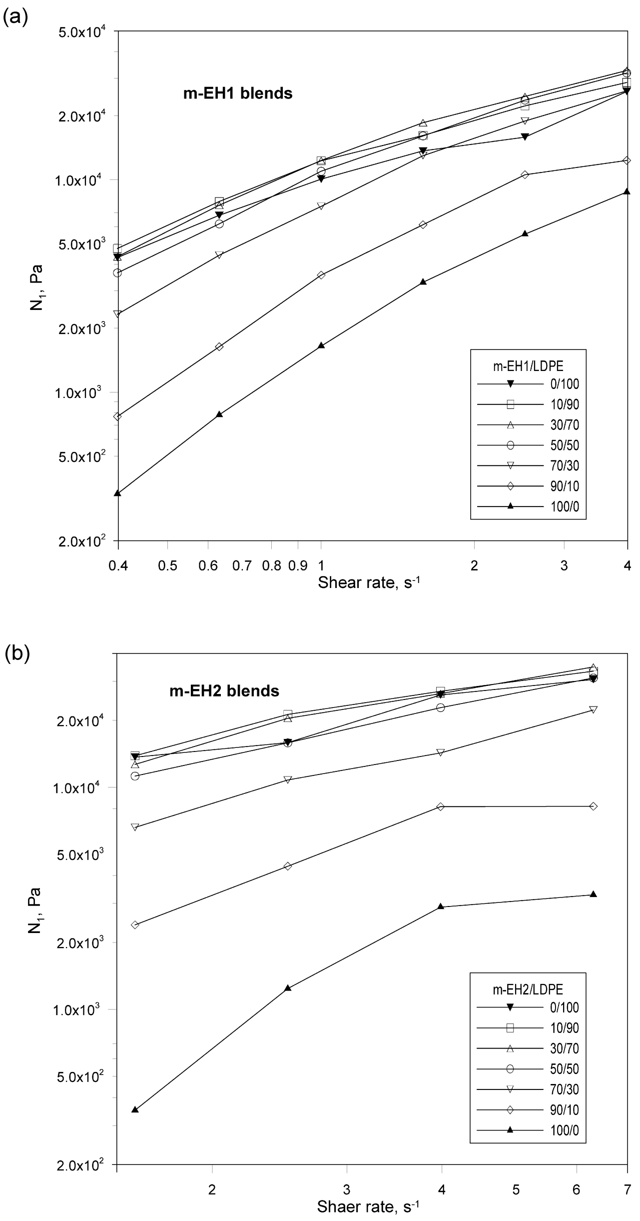

Fig. 5. (a) $N_1(\dot\gamma)$ for blends of m-EH1 with LDPE ($T_{mix} = 190$ °C, × $T_{test} = 190$ °C, tbm = 200 s, mt = 30 s); (b) $N_1(\dot\gamma)$ for blends of m-EH2 with LDPE ($T_{mix} = 190$ °C, $T_{test} = 190$ °C, tbm = 200 s, mt = 30 s).

EH2 blends with LDPE for $\dot{\gamma} = 1.5-6\ \mathrm{s}^{-1}$. For the determination of the morphology of the blend, low-$\dot{\gamma}$ is preferred; however, for the blends of m-EH2 with LDPE the choice of the low range was limited by the sensitivity of the rheometer transducer. In the low-$\dot{\gamma}$ range, $N_1(\dot{\gamma})$ for the 50, 70, and the 90% blends of m-EH1 with LDPE were found to lie between the $N_1(\dot{\gamma})$ values of the pure polymers. For the 50/50 blend, N_1 is slightly lower than the more elastic component (LDPE). The LDPE-rich blends showed N_1 values that were higher than LDPE. The increase in the elasticity of these blends is a clear indication of a multiphase morphology and is likely a result of the interfacial tension. The steady-shear rheology of blends of m-EH1 with LDPE suggested the immiscibility of the 50/50 and LDPE-rich blends of EH1 with LDPE; the miscibility of EH1-rich blends.

On the other hand, results obtained for blends of m-EH2 with LDPE (Fig. 5(b)) showed that $N_1(\dot{\gamma})$ values for all blends were bounded by the $N_1(\dot{\gamma})$ values of the pure resins. In general, $N_1(\dot{\gamma})$ increased with the increase in the proportion of the more elastic component. Again, the 10% m-EH2 blend with LDPE showed N_1 values that were of the same magnitude as the pure LDPE, which suggests some degree of immiscibility supporting the previous dynamic shear data. Here, a high degree of miscibility of m-EH1 blends with LDPE was suggested by both the steady-shear and dynamic shear data.

Moreover, another steady-shear test was performed. This time stress relaxations of blends were explored. The relaxation experiment followed a period of 500 s of steady shearing at 1 s^{-1} The test was carried out on all blends of m-EH1 and m-EH2 with LDPE as well as the pure resins. Results of $N_1(t)$ for the two systems, not shown here, followed the same pattern of the $N_1(\dot{\gamma})$ data shown in Fig. 5(a) and (b) above.

All of these predictions suggest the presence of an interface in the LDPE-rich blends and its weakness or absence in the m-EH2-rich blends. For the high-BC m-LLDPE blends with LDPE, all models suggest the weakness of the interface, if any, and hence a high degree of miscibility. The value of α/R obtained for the LDPE-rich blends m-EH1 with LDPE were comparable with similar values reported for compatibilized polymer blends [46].

Thus, the different rheological measurements suggest a high degree of miscibility of the high-BC m-LLDPE blends with LDPE. For the low-BC m-LLDPE blends with LDPE, high degree of miscibility is likely to occur in the m-LLDPE-rich blends range ($\phi \geq 70\%$ m-LLDPE); and immiscibility is likely to be observed for the 50/50 and LDPE-rich blends ($\phi \leq 50\%$ LLDPE). Immiscibility of the blends was predicted by the Einstein and Scholz et al. models. Most of the previous rheological investigations of multiphase systems have concentrated on dynamic-shear rheology [36–41], but it seems that the steady-shear rheology was very useful.

3.2. Influence of composition distribution (catalyst type)

To study the influence of the catalyst type or CD another hexene ZN–LLDPE (ZN–EH) was blended with LDPE. The ZN–EH resin was chosen to have about the similar M_w, BC ($M_w = 107k$; BC = 16.7 CH_3/1000 C) as the corresponding hexene m-EH1 ($M_w = 102k$; BC = 14.4 CH_3/1000 C). Plots of $\eta'(\omega)$ for blends of ZN–EH with LDPE are shown in Fig. 6. The results were almost identical to those obtained for the m-EH1 blends and given in Fig. 1. This was a direct consequence of matching the M_w (sensitive to viscosity) and BC (sensitive to elasticity). Here, the hexene-based blends showed a Newtonian plateau at low-ω with the Newtonian plateau much clearer in blends with higher weight fractions of m-EH1. For all blends, both η' and G' [not shown] were bounded by the corresponding pure components.

The $\eta'(\omega)$ data for ZN–EH blends with LDPE were fitted to cross model. Computed η_o was obtained from the fitting and plotted in Fig. 7. The plot showed weak PDB for the LDPE-rich and the 50/50 blends, while η_o almost followed the log-additivity for the 90% m-EB blend with LDPE. The behavior of $\eta_o((\phi)$ data for the ZN–EH blends is different than that of m-EH1 blends as shown in Fig. 7. For m-EH1 blends with LDPE, the LDPE-rich blends were likely more immiscible. However, ZN–EH blends with LDPE showed less PDB at all compositions and high degree of miscibility. This suggests that CD plays an important role in the miscibility of LLDPE/LDPE blends especially in the LDPE-rich blends range with more miscibility for ZN–LLDPE blends with LDPE in comparison with m-LLDPE blends with LDPE.

The present study suggests the influence of both BC and CD of LLDPE (ZN vs. metallocene) on the miscibility of

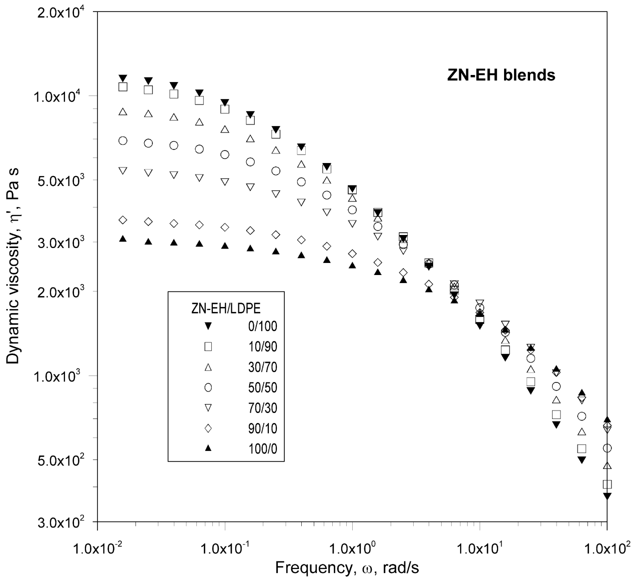

Fig. 6. $\eta'(\omega)$ for blends of ZN–EH with LDPE ($T_{\mathrm{mix}} = 190$ °C, × $T_{\mathrm{test}} = 190$ °C, $\gamma^\circ = 15\%$, $\omega = 0.1$ rad/s).

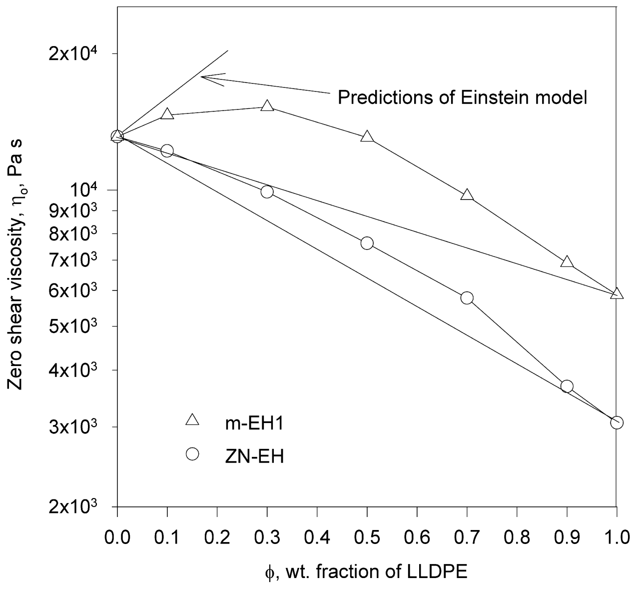

Fig. 7. $\eta_o(\phi)$ for blends of m-EH1 and ZN–EH with LDPE computed from cross model ($T_{\text{mix}} = 190$ °C, $T_{\text{test}} = 190$ °C, $\gamma^o = 15\%$).

LLDPE with LDPE. The current observations with regard to the effect of CD were similar to the previous observations on blends of linear and LLDPE systems obtained from different techniques [4,5,26,27,45]. For the effect of BC, higher BCs are expected to lead to better miscibility, which is contrary to previous observations of SANS and TEM results on LLDPE/HDPE systems. This suggests that the effect of BC on the miscibility of m-LLDPE/LDPE and m-LLDPE/HDPE blends is different. The reason for the current observation is likely to be the chaotic structure of LDPE (free radical polymerization) which is expected to match the conformations of highly branched m-LLDPE [45,47]. Details about tentative explanations for immiscibility of PE/PE blends were discussed by one of the authors in previous publications [12,14,45,47]

4. Conclusions

In conclusion, the effects of BC and branch distribution on the miscibility of LLDPE/LDPE blends were investigated using metallocene and ZN LLDPE resins. The BC of the shortly branched m-LLDPE has influenced its miscibility with LDPE. Blends of high-BC m-LLDPE (hexene, BC = 32 CH_3/1000 C) and LDPE were almost *miscible* in the whole composition range. However, the low-BC m-LLDPE was found to be partially miscible. Immiscibility is likely to develop in the LDPE-rich blends range, while miscibility is likely to develop in the LLDPE-rich range. For the effect of CD, ZN–LLDPE was found to be more miscible with LDPE than m-LLDPE of the same M_w and BC.

Several rheological measurements and data-treatment approaches were used in this study. First, plots of $\eta_o(\phi)$, × $\eta'(\omega)$, $\eta'(\phi)|_{\omega=\text{const}}$, $G'(\omega)$, $G'(\phi)|_{\omega=\text{const}}$ and $N_1(\dot{\gamma})$ were shown to be helpful. These techniques and approaches made it possible to identify the miscibility/immiscibility of blends. Secondly, the three-parameter cross model was used to model $\eta'(\omega)$ data, allowing the assessment of miscibility through $\eta_o(\phi)$.

The immiscibility of the blends leads to increase in $\eta_o(\phi)$, $\eta'(\phi)$, $G'(\phi)$ *and* $G^*(\omega)$ that *can* be explained by *emulsion* models (e.g. Einstein and Scholz et al. dilute emulsion models). The ratio of interfacial tension to droplet radius (α/R) was found to be ~500 N/m^2 as computed from rheological models. The value of α/R is consistent with an interface between chemically similar polyethylenes.

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