

# Implications of melt compatibility/incompatibility on thermal and mechanical properties of metallocene and Ziegler–Natta linear low density polyethylene (LLDPE) blends with high density polyethylene (HDPE): influence of composition distribution and branch content of LLDPE

Ibnelwaleed A Hussein\*

Department of Chemical Engineering, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia

**Abstract:** In this paper, the implications of melt compatibility on thermal and solid-state properties of linear low density polyethylene/high density polyethylene (LLDPE/HDPE) blends were assessed with respect to the effect of composition distribution (CD) and branch content (BC). The effect of CD was studied by melt blending a metallocene (m-LLDPE) and a Ziegler–Natta (ZN) LLDPE with the same HDPE at 190 °C. Similarly, the effect of BC was examined. In both cases, resins were paired to study one molecular variable at a time. Thermal and solid-state properties were measured in a differential scanning calorimeter and in an Instron mechanical testing instrument, respectively. The low-BC m-LLDPE (BC = 14.5 CH<sub>3</sub>/1000 C) blends with HDPE were compatible at all compositions: rheological, thermal and some mechanical properties followed additivity rules. For incompatible high-BC (42.0 CH<sub>3</sub>/1000 C) m-LLDPE-rich blends, elongation at break and work of rupture showed synergistic effects, while modulus was lower than predictions of linear additivity. The CD of LLDPE showed no significant effect on thermal properties, elongation at break or work of rupture; however, it resulted in low moduli for ZN-LLDPE blends with HDPE. For miscible blends, no effect for BC or CD of LLDPE was observed. The BC of LLDPE has, in general, a stronger influence on melt and solid-state properties of blends than the CD.

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**Keywords:** LLDPE; HDPE; mechanical properties; DSC; branch content; composition distribution

## INTRODUCTION

High density polyethylene (HDPE) is used for applications that require stiff products and linear low density polyethylene (LLDPE) is usually blended to improve flexibility<sup>1</sup> or to reduce extruder backpressure and increase output.<sup>2</sup> The compatibility (the word miscibility is usually used rather than compatibility) of LLDPE and HDPE blends has received wide attention in the literature.<sup>3–10</sup> LLDPE is produced either by Ziegler–Natta (ZN-LLDPE) or more recently by metallocene catalysis (m-LLDPE). Most of the previous studies used ZN-LLDPE, which is known for its inter- and intra-molecular heterogeneity.<sup>11–13</sup> The multi-site nature of ZN catalyst results in a product with a diverse composition distribution (CD):

weight-average molecular weight ( $M_w$ ), molecular weight distribution (MWD) and branch content (BC). These molecular parameters have a strong influence on the compatibility of PE/PE blends.<sup>3–7,14–17</sup> This explains the suspected or suggested phase separation in ‘pure’ ZN-LLDPE,<sup>18–22</sup> which is in fact a blend of high- $M_w$  mostly linear molecules with low- $M_w$  highly branched molecules.<sup>11</sup>

The crystallization behavior of blends of linear and branched PE was reported to be a critical function of BC.<sup>7,23–27</sup> The critical BC for separate crystallization in m-LLDPE/HDPE blends was observed to be lower than that of ZN-LLDPE/HDPE systems.<sup>11,24</sup> Also, CD is another important parameter that determines the crystallization behavior of PE blends. Recently,

\* Correspondence to: Ibnelwaleed A Hussein, Department of Chemical Engineering, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia

E-mail: ihussein@kfupm.edu.sa

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ZN-LLDPE/LDPE blends were suggested to have a higher degree of co-crystallization when compared to m-LLDPE/LDPE blends for the same BC and comonomer type.<sup>28</sup> However, other researchers have suggested no effect for CD (usually referred to in these publications as branch distribution).<sup>25,27,29</sup> The effect of the CD of LLDPE on the crystallization of LLDPE/HDPE blends is yet to be studied.

Mechanical properties of polyolefin blends have received wide attention in the literature.<sup>30–39</sup> Acierno and co-workers<sup>31,32</sup> investigated the effect of comonomer type and  $M_w$  on the rheology and mechanical characteristics of ZN-LLDPE/LDPE blends. Comonomer type was found to influence the mechanical properties only slightly. In contrast, mechanical properties were a strong function of  $M_w$ . Cho *et al.*<sup>33</sup> observed that ZN-LLDPE/LDPE blends show synergistic effects on the yield strength and elongation at break while the strength at break varies linearly with composition. The mechanical properties of blends of different LLDPE/LLDPE resins of uniform branch distribution were found to be a function of total crystallinity rather than of the type of crystallinity.<sup>39</sup>

The polymers used in this study were paired in order to study one molecular variable at a time. To study the effect of CD, two butene LLDPE resins of comparable BC and  $M_w$  were selected and blended with the same HDPE. The CD was the only molecular parameter that was varied. The effect of BC was studied by using two butene m-LLDPEs of the same  $M_w$  and MWD but of different densities (BC). All LLDPEs were blended with the same HDPE. This approach of isolating the effect of other molecular parameters was not followed in most of the previous studies. In recent studies, the author and co-workers showed that both BC and CD of LLDPE influence its melt compatibility with HDPE<sup>3</sup> and LDPE.<sup>17,40</sup> Here, the implications of melt compatibility (or incompatibility) on thermal and mechanical properties of LLDPE/HDPE blends were examined on the same blend systems.<sup>3</sup>

## EXPERIMENTAL

### Materials and sample preparation

Three commercial samples of butene LLDPE and one linear HDPE were used in this study. Two of the LLDPE ethylene-butene copolymers were metallocene resins (m-EB1 and m-EB2) of different BC and the third was an ethylene-butene ZN-LLDPE copolymer (ZN-EB). All samples were ExxonMobil products. Table 1 provides characterization data such

**Table 1.** Characterization of resins

Resin	Density (g cm <sup>-3</sup> )	MI (g/10 min)	$M_w$ (kg mol <sup>-1</sup> )	$M_w/M_n$	BC (CH <sub>3</sub> / 1000 C)
ZN-EB	0.918	1.0	118	3.1	13.2
m-EB1	0.9100	1.2	108	2.0	14.5
m-EB2	0.8800	0.8	125	1.8	42.0
HDPE	0.9610	0.7	102	6.7	0

as density at room temperature and melt index (MI) at 190 °C as provided by ExxonMobil. In addition, information about  $M_w$  and BC as revealed by gel permeation chromatography (GPC) and <sup>13</sup>C NMR is included. Details about the GPC and NMR characterizations were given in a previous publication.<sup>3</sup> The rheology of all the LLDPEs does not suggest the presence of any long chain branches. The resins with low and high BC were labeled as m-EB1 and m-EB2, respectively. The effect of BC of LLDPE was investigated by examining m-EB1/HDPE and m-EB2/HDPE blend systems. Resins m-EB1 and m-EB2 were selected so that BC would be the only primary molecular variable in this comparison (see Table 1).

Similarly, to study the effect of CD, ZN-EB was chosen to have about the same  $M_w$  and BC as m-EB1. The HDPE used in all of these blends has a density higher than 0.960 g cm<sup>-3</sup> and hence should have no branches,<sup>41</sup> which is confirmed by our own <sup>13</sup>C NMR measurements. The HDPE and LLDPE resins used in this study were conditioned (or melt blended) in a Haake PolyDrive melt blender at 190 °C for 10 minutes at 50 rpm in the presence of an adequate amount of antioxidant as discussed in previous publications.<sup>3,40,42</sup> Sample preparation in a Carver press for thermal and mechanical testing was the same as that discussed earlier in the rheological study.<sup>3</sup>

### Differential scanning calorimetry (DSC)

Thermal analysis was performed with a TA Instruments DSC 2910 equipped with Thermal Analyst 2200 software, and nitrogen gas flow was maintained from a liquid nitrogen vessel. The samples obtained from the Carver press were used in the DSC analysis. Samples of 5–10 mg were sliced and then compressed into aluminium pans for testing. The samples were heated from 25 to 200 °C at a heating rate of 10 °C min<sup>-1</sup> to eliminate the thermal history, held at 200 °C for 1 min and then cooled to 25 °C at 10 °C min<sup>-1</sup>. All samples were subjected to a second heating cycle identical to the first one. Calculations of crystallinity were based on a heat of fusion of 290 J g<sup>-1</sup> for polyethylene crystal.<sup>43</sup>

### Mechanical testing

Samples obtained from the Carver press were cut according to ASTM D638 (type V). The tensile tests were performed on the Instron 5567 tensile testing machine at room temperature. The gauge length was kept at 25 mm with a crosshead speed of 100 mm s<sup>-1</sup>. The results reported in this study are based on an average of a minimum of five samples.

## RESULTS AND DISCUSSION

A previous rheological study<sup>3</sup> on the melt compatibility of these blends suggested the following conclusions:

- (1) Blends of m-EB1 and HDPE are miscible at all compositions.
- (2) Blends of ZN-EB and HDPE are miscible at all compositions and ZN-EB blends are more miscible than their m-EB counterparts.
- (3) Blends of the high-BC m-EB2 and HDPE showed that HDPE-rich blends are miscible while m-EB2-rich blends are immiscible. The rheology of miscible systems followed additivity rules while immiscible blends displayed negative deviation from additivity. Layered morphology was proposed as an explanation for the observed rheology.

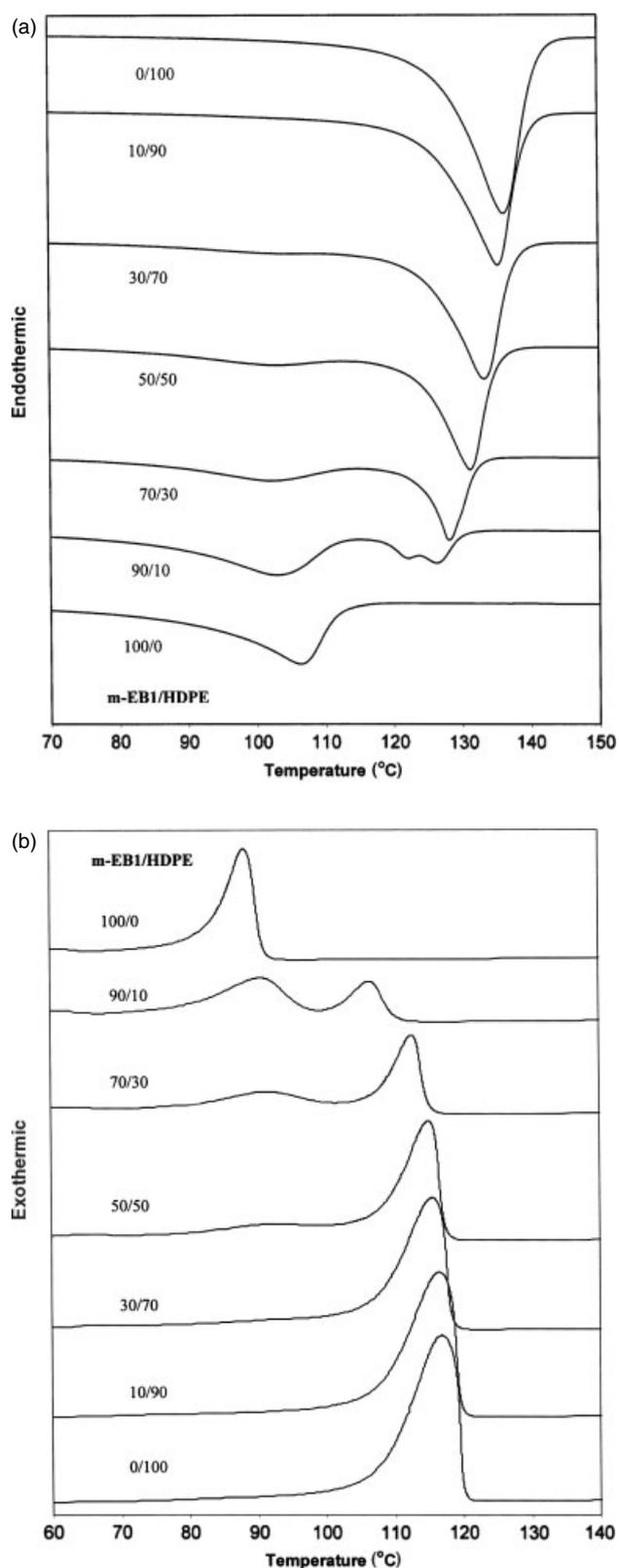
In the following sections, the implications of this compatibility/incompatibility on the thermal and mechanical properties are discussed.

### Thermal analysis

DSC melting and crystallization thermograms for m-EB1 (the low-BC component) blends with HDPE together with pure resins are given in Fig 1. The values of the melting ( $T_m$ ) and crystallization ( $T_c$ ) peaks are shown on all DSC thermograms. In all endotherms, the second heating curves were used to remove previous history.<sup>21,35,40</sup> Similarly, in Fig 2 the DSC results for m-EB2 (the high-BC component) blends with HDPE are displayed. In all DSC curves, HDPE showed the highest  $T_m$  (or  $T_c$ ), which shifts to a lower value as the amount of LLDPE increases in the blend. Also, m-EB2 showed the lowest  $T_m$ , followed by m-EB1. These observations are due to the occurrence of shorter ethylene sequences between short chain branches in the high-BC m-LLDPE.<sup>13,19,24</sup> The depression in the melting point for the three blend systems is given in Table 2.

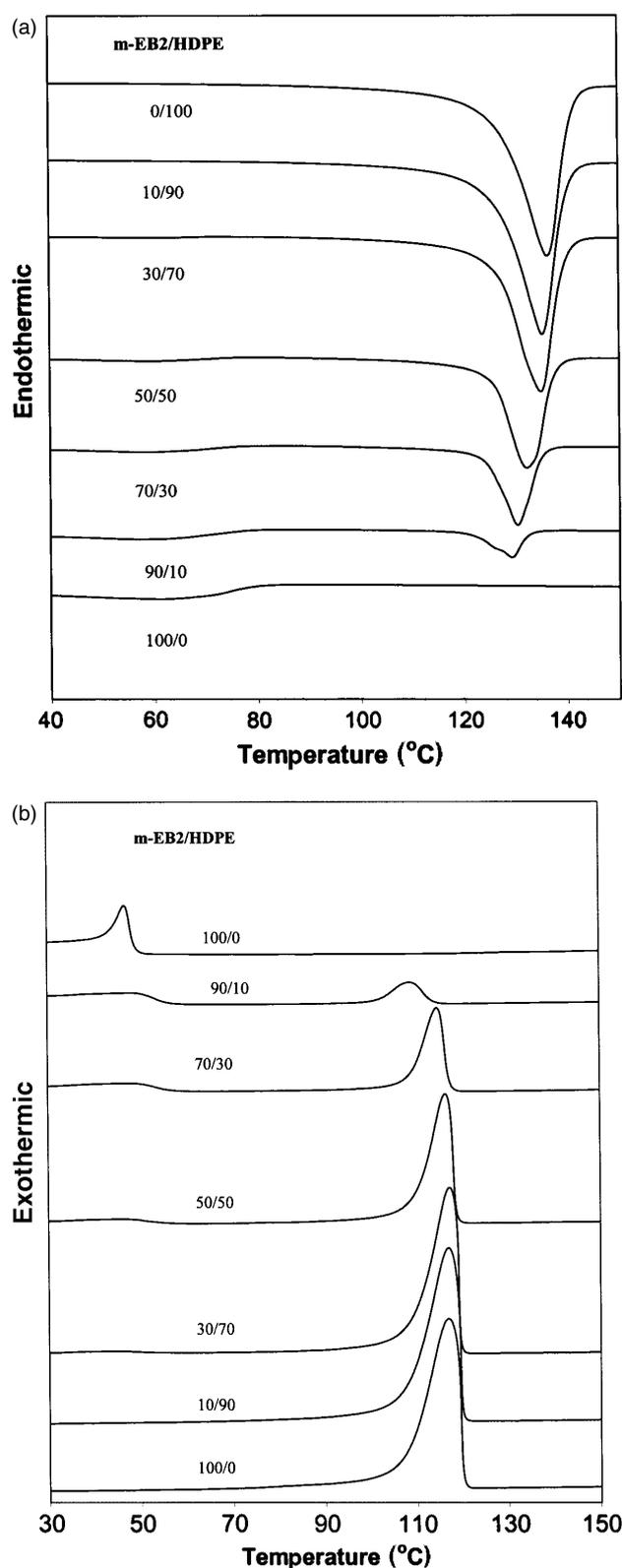
Both HDPE and m-EB1 showed a single peak; however, the m-EB1 peak is very broad. Furthermore, all HDPE-rich blends with either m-EB1 or m-EB2 showed a single sharp melting peak. This suggests the existence of one type of crystal population, hence, the complete homogeneity of these blends in the solid state. However, the m-EB1-rich blends had at least two melting peaks. The 90% m-EB1 blend showed three melting peaks, with two major peaks representing the pure components and a third one that is probably due to co-crystallization.<sup>24</sup> In m-EB1-rich blends, the sharpness of the lower melting peak decreases as the fraction of the linear component is increased, which further supports co-crystallization.

The crystallization mechanism of m-EB1 blends with HDPE was examined, and results are displayed in Fig 1(b). Here, only single peaks were observed for HDPE-rich blends and a maximum of two peaks were obtained for m-EB1-rich blends. The 90% m-EB1 blend showed two distinct peaks that are located between the two peaks representing the pure components. This shift in  $T_c$  supports co-crystallization and partial compatibility of the m-EB1-rich blends. Again, the sharpness of the m-EB1 peak



**Figure 1.** DSC scans for m-EB1/HDPE blends: (a) heating; (b) cooling.

decreased as the fraction of the linear component was increased. So, for low-BC pair, blends of m-LLDPE and HDPE are compatible in the solid state in the HDPE-rich range, and partially compatible in the m-LLDPE-rich range. In addition, single crystals in HDPE-rich blends and multi-crystals in



**Figure 2.** DSC scans for m-EB2/HDPE blends: (a) heating; (b) cooling.

the m-LLDPE-rich blends were also observed. The depression in the peak melting temperature,  $\Delta T_m$ , for m-EB1 blends followed the same trend. Plots of  $\Delta T_m(\phi)$  showed strong depression in m-EB1-rich blends (results not shown here).

**Table 2.** Melting point depression, in °C, for blends of different LLDPEs with HDPE

Composition	ZN-EB blends	m-EB1 blends	m-EB2 blends
0 (HDPE)	—	—	—
0.1	0.80	0.84	1.05
0.3	2.39	2.92	1.32
0.5	3.77	5.04	4.13
0.7	6.81	8.12	6.17
0.9	9.74	9.53	7.00
1.0 (LLDPE)	12.47	29.88	73.67

For blends of m-EB2 with HDPE, the results of heating and cooling are shown in Fig 2. The highly branched m-EB2 shows a very broad and weak transition in the range 60–80 °C. The depression in  $T_m$  for increasing comonomer incorporation is explained by the occurrence of shorter ethylene sequences between short chain branches in the sample containing high amounts of comonomer.<sup>24</sup> Addition of HDPE to m-EB2 resulted in a peak at  $\approx 130$  °C, which represents the HDPE crystals. The sharpness of the peak increases with the increase of the HDPE fraction in the blend. For HDPE-rich blends, the major melting peak of HDPE shifts with the increase of m-EB2 fraction and single peaks were observed, suggesting solid-state compatibility in this composition range. However, for m-EB2-rich blends, a shoulder was observed in the 90 % m-EB2 blend and a weak and broad melting point representing the m-EB2 was detected. This observation suggests the existence of separate crystals of the pure components. The third crystal population in the 90 % blend is probably due to the formation of co-crystals of HDPE with the linear part of the highly branched m-EB2. Highly branched m-LLDPEs were recently suggested to have intra-molecular heterogeneity.<sup>24</sup> Also, temperature rising elution fractionation analysis on blends of LLDPE and LDPE suggested that fractions containing similar branches were more likely to co-crystallize than fractions with different BCs.<sup>44,45</sup> A previous rheological study<sup>3</sup> suggested the existence of a layered morphology for m-EB2-rich blends in the melt state, which was supported by molecular dynamics investigation.<sup>46</sup> The layered morphology is likely to promote co-crystallization and explain the observed third crystal population in the 90 % m-EB2 blend. These results are in agreement with previous reports that suggest separate crystallization with some amount of co-crystals in blends of high-BC m-LLDPE and HDPE.<sup>47</sup>

The crystallization of m-EB2 blends with HDPE is given in Fig 2(b). The results are similar to those of the low-BC pair. Again, only single peaks were observed for HDPE-rich blends and a maximum of two peaks were observed for m-EB2-rich blends. The 70 % and the 90 % m-EB1 blends with HDPE showed two peaks that are located between the two peaks representing the pure components. This shift in  $T_c$  supports the co-crystallization and compatibility of

the m-EB2-rich blends. In addition, the sharpness of the m-EB2 peak decreased as the fraction of the linear component was increased. However, the broad and shallow melting peak of pure m-EB2 shown in Fig 2(a) has turned into a sharp peak in crystallization. So, for high-BC blends of m-LLDPE and HDPE, solid-state compatibility is suggested by DSC at all compositions; however, a multi-crystal population is likely to develop in m-EB2-rich blends. The comparison of the melting and crystallization of low- and high-BC pairs suggests that BC has little or no influence on melting and crystallization of HDPE-rich blends, with blends showing single peaks. However, m-LLDPE-rich blends are likely to be partially compatible with multi types of crystals. Hence, the melting and crystallization of low- and high-BC m-LLDPE/HDPE blends are similar, but composition-dependent.

DSC melting and crystallization peaks for ZN-EB blends with HDPE are shown in Fig 3. HDPE and ZN-EB as well as all blends exhibit sharp melting and crystallization peaks over the range of temperature examined. Both  $T_m$  and  $T_c$  increased with the increase in the fraction of the linear component. The results are thus similar to those of m-EB1 blends shown in Fig 1. No multiple peaks were observed. Comparison of m-EB1 and ZN-EB (with almost the same BC) shows that ZN-EB has higher  $T_m$  and  $T_c$ , which is mainly due to the presence of a linear fraction in ZN-LLDPE.<sup>11,12</sup> The linear fractions of ZN-EB can form thicker lamellae, melting at higher temperature. Comparison of ZN-EB and m-EB1 blends with the same HDPE reveals that all ZN-EB blends showed a single peak that shifts with composition, which suggests the complete compatibility of ZN-EB blends. Hence, ZN-EB is compatible with HDPE over the whole composition range, with blends showing single melting and crystallization peaks. These observations are in agreement with previous DSC and X-ray measurements that suggest the formation of perfect co-crystallites between ZN-LLDPE and HDPE<sup>48–51</sup> when the BC of ZN-LLDPE is lower than 42 CH<sub>3</sub>/1000 C (8.2 mol %). However, the thermal compatibility and melt compatibility of ZN-EB blends with HDPE may not lead to mechanical compatibility, as will be discussed in the next section.

HDPE-rich blends with m-EB1 are suggested to be compatible; however, m-EB1-rich blends are likely to be partially compatible in the solid state. Hence, the influence of CD (ZN-EB *versus* m-EB1) on thermal properties of LLDPE/HDPE blends is evident and ZN-LLDPE is more compatible with HDPE than m-LLDPE with the same average BC and  $M_w$ . It is worth noting that the rheological study resulted in the same suggestion with regard to melt compatibility.<sup>3</sup>

The crystallinity of m-EB1, m-EB2 and ZN-EB blends with HDPE were calculated from Figs 1(a), 2(a) and 3(a), respectively, and the results are given in Fig 4. DSC cooling curves for ZN-EB

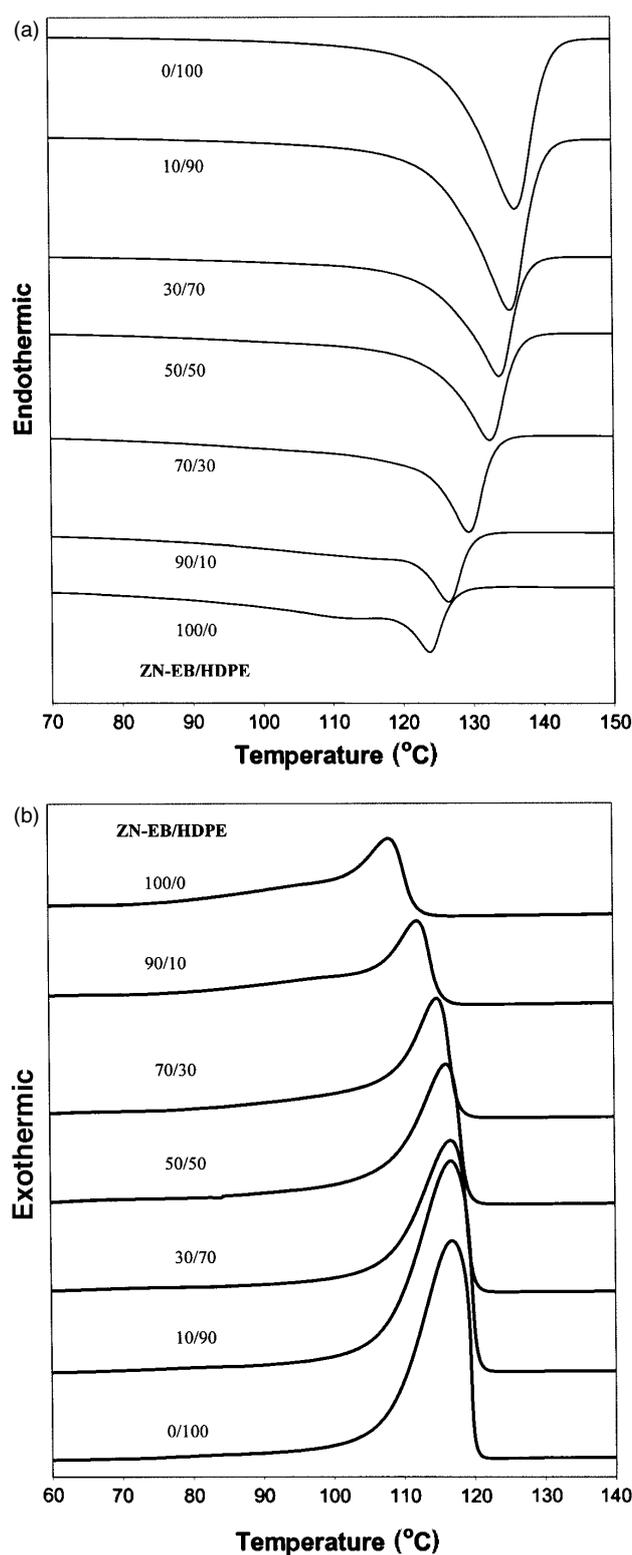


Figure 3. DSC scans for ZN-EB/HDPE blends: (a) heating; (b) cooling.

blends are shown in Fig 3(b) as a function of composition. The lines represent the calculated crystallinity obtained by assuming linear additivity. A comparison of Figs 4(a), 4(b) and 4(c) suggests that neither BC nor CD influenced the total crystallinity. For ZN-EB blends, the results shown in Fig 4(a) are consistent with those given in Fig 3(a). Also, for HDPE-rich

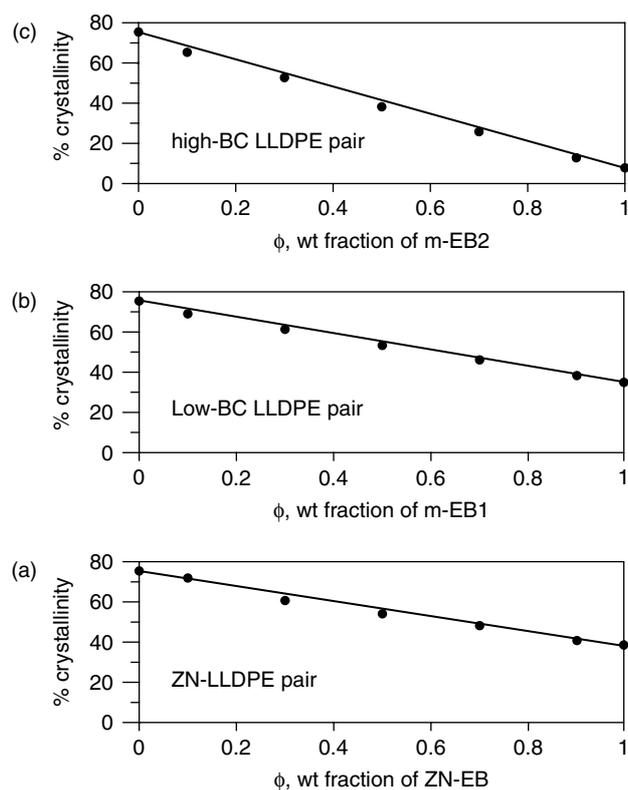


Figure 4. Percent crystallinity as a function of composition.

blends with either m-EB1 or EB2, the results are in agreement with the DSC observations shown in Figs 1 and 2. However, for m-LLDPE (both m-EB1 and m-EB2) rich blends, co-crystallization was suggested, yet the total crystallinity followed a linear additivity rule. This observation requires further X-ray investigation on the effect of blending on lamella thickness of m-LLDPE/HDPE blends.

In summary, both BC and CD of m-LLDPE were found to influence the melting and crystallization of LLDPE/HDPE blends. HDPE-rich blends with either low-BC or high-BC pairs showed single melting and crystallization peaks. However, the ZN-LLDPE blends behaved quite differently with single melting and crystallization peaks at all compositions. The heterogeneity of ZN-LLDPE, especially the presence of linear PE, has probably helped in the co-crystallization with the linear HDPE. On the other hand, m-LLDPE-rich blends (regardless of their BC) showed multiple peaks. Total crystallinity is only composition dependent, with no influence of BC or CD.

### Mechanical properties

The tensile moduli as a function of composition for blends of m-EB1, m-EB2 and ZN-EB with HDPE are shown in Fig 5. In figures the displayed mechanical properties represent the average of at least five measurements. The error bars indicate the range of these results.

In Fig 5(b), the modulus for HDPE is  $\approx 1100$  MPa, while that of m-EB1 is  $\approx 200$  MPa. In general, the moduli for the blends follow the linear rule of

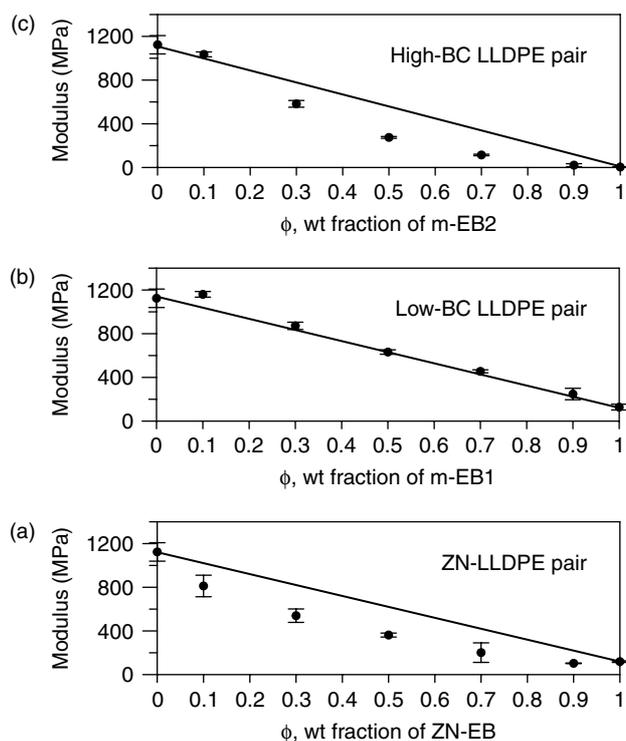


Figure 5. Tensile modulus as a function of composition.

mixtures. However, the moduli for blends of ZN-EB and HDPE, as shown in Fig 5(a), are lower than the predictions of the linear rule of mixtures. Comparison of Figs 5(a) and 5(b) suggests that the CD of ZN-EB has influenced the moduli of its blends with HDPE at all compositions. However, the moduli for blends of m-EB1 (characterized by more uniformly distributed short chain branches) and HDPE are predicted by the simple rule of mixtures.

For the effect of BC on tensile modulus, Figs 5(b) and 5(c) are compared. The moduli for all m-EB2 blends, except the 10 % m-EB2, are lower than the simple rule of mixtures. The m-EB2-rich blends show the stronger deviations. The addition of 10 % m-EB2 to HDPE produced a proportional decrease in the modulus. On the other hand, addition of up to 30 % HDPE (the component with the highest modulus) to the highly branched m-EB2 did not result in any significant improvement in the tensile modulus. It is interesting to note that similar composition dependency of the melt dynamic viscosity and elastic modulus were observed for m-EB2-rich blends showing negative deviation behavior.<sup>3</sup> The proposed interlayer morphology<sup>3,43</sup> could explain the current observations. These results suggest that BC has a strong influence on the tensile modulus, especially for m-LLDPE-rich blends.

The effect of BC and CD on the strength at yield is shown in Fig 6. Again, comparison of Figs 6(a) and 6(b) is expected to show the effect of CD, while that of Figs 6(b) and 6(c) will indicate the effect of BC. The strength at yield, for ZN-EB blends with HDPE, is in agreement with previous measurements of Cho *et al.*<sup>33</sup> HDPE showed the highest yield strength. It is

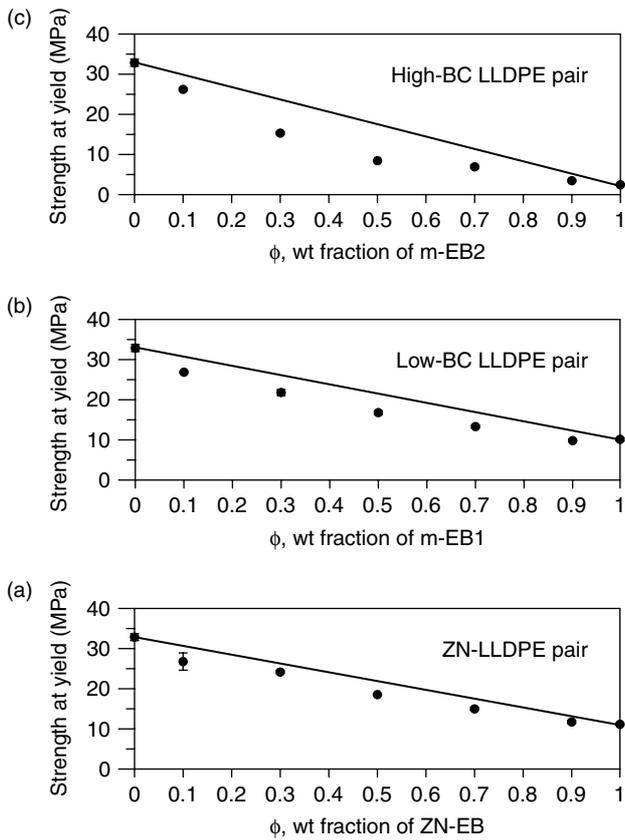


Figure 6. Strength at yield as a function of composition.

clear that CD has little or no influence on strength at yield. Hence, the behavior of yield strength for ZN-LLDPE/HDPE blends is different from that of ZN-LLDPE/LDPE blends.<sup>33</sup> For the effect of BC, the results of the yield strength are similar to those of the tensile modulus. Negative deviations from linear additivity were observed. So, strength at yield is strongly influenced by BC, but not by CD.

The influence of CD and BC on the ultimate tensile strength is shown in Fig 7. The current findings for strength at break for the ZN-EB blends are similar to previous reports of Cho *et al.*<sup>33</sup> In general, the ultimate tensile strength for blends of m-EB1 and ZN-EB with HDPE is lower than the predictions of linear additivity. Nonetheless, CD showed little or no influence on ultimate tensile strength. It should be noted that the ultimate tensile strength (or strength at break) for ZN-LLDPE/HDPE blends showed a behavior that is contrary to that reported for ZN-LLDPE/LDPE systems. In Fig 7(c) the results for the high-BC pair are given. Both negative (HDPE-rich blends) and positive deviations (m-EB2-rich blends) are observed. The behavior of HDPE-rich blends is similar for both m-EB1 and m-EB2 blends. The effect of BC is observed in m-LLDPE-rich blends only. So, CD has little or no influence on ultimate tensile strength, and BC influences m-LLDPE-rich blends only.

The elongation at break for the different blend systems is shown in Fig 8. The elongation at break for all LLDPEs was found to be higher than that of HDPE, irrespective of BC or synthesis mechanism.

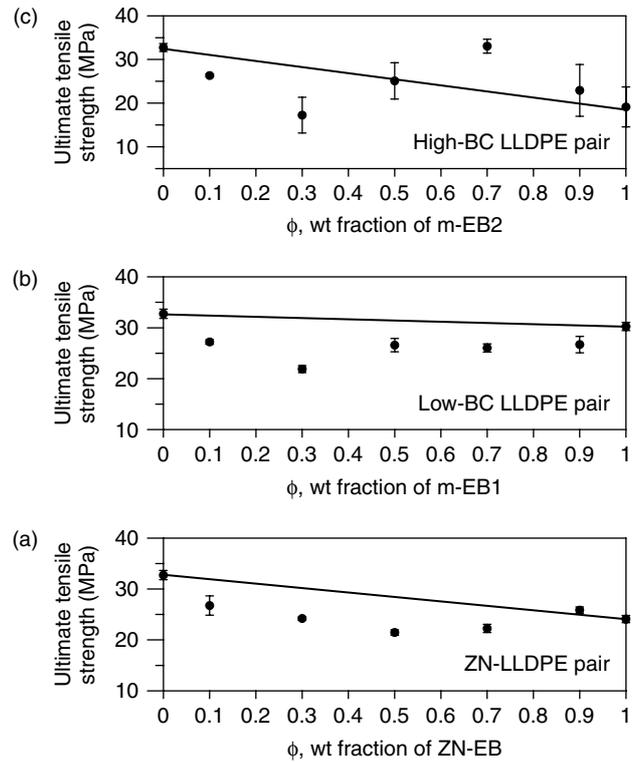


Figure 7. Ultimate tensile strength as a function of composition.

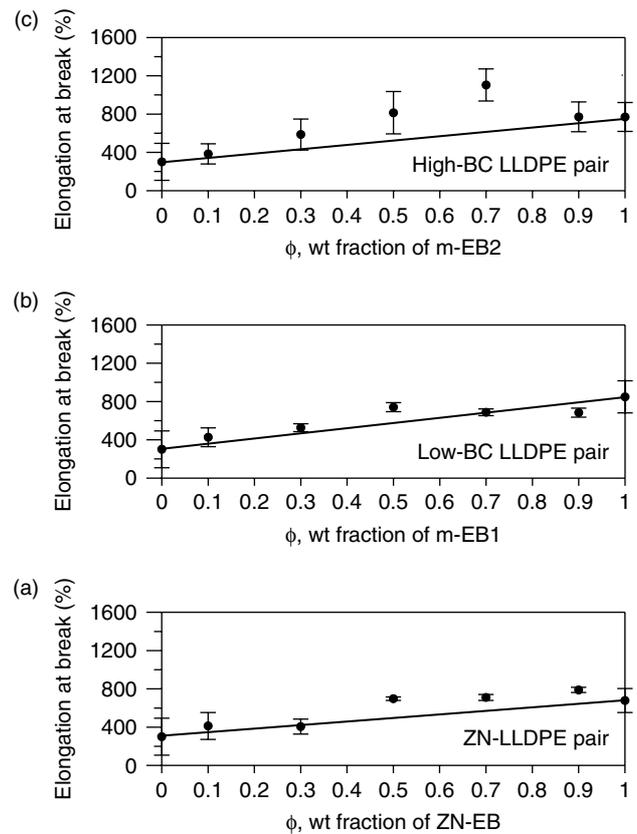


Figure 8. Elongation at break as a function of composition.

The elongation at break of m-EB1 is higher than that of ZN-EB, and this is mainly due to the absence of linear molecules in m-EB1. As seen in Figs 8(a) and 8(b), ZN-EB and m-EB1 blends with HDPE display similar

trends, which suggests no influence for CD. For Figs 8(b) and 8(c), the behavior of HDPE-rich blends is similar, but m-EB2-rich blends show synergistic effects. The elongation at break obtained for the 50 % and 70 % m-EB2 blends with HDPE was much higher than that of m-EB1. This suggests that adding up to 30 % HDPE to highly branched m-LLDPE will improve the elongation at break. Hence, CD has no effect on elongation at break, and BC influence is observed for m-LLDPE-rich blends only. The area under the stress–strain curve, which is proportional to the toughness of material, is given in Fig 9. The effects of BC and CD on the work of rupture are similar to those of elongation at break.

In summary, for the same  $M_w$ , the three LLDPEs, m-EB1, m-EB2 and ZN-EB, exhibited different mechanical properties as shown in Figs 5 to 9. In the linear elastic range, the solid-state tensile modulus and yield strength for the low- and high-BC pairs are mirror images of the melt state. However, ZN-LLDPE was suggested to be more miscible with HDPE than m-LLDPE, but it showed poorer moduli and yield strength. This is contrary to what was expected. This suggests that ZN-LLDPE/HDPE crystals are likely to have smaller thickness in comparison to those of m-LLDPE/HDPE. This observation requires further exploration by X-ray. The incompatibility of the high-BC pair resulted in poor tensile modulus and strength. Also, CD was observed to influence both the moduli and the tensile strength of blends at all compositions, with ZN-LLDPE blends showing poorer properties. For the ultimate tensile strength, elongation at break and work of rupture, CD has little or no influence, and BC influences m-LLDPE-rich blends only. In general,

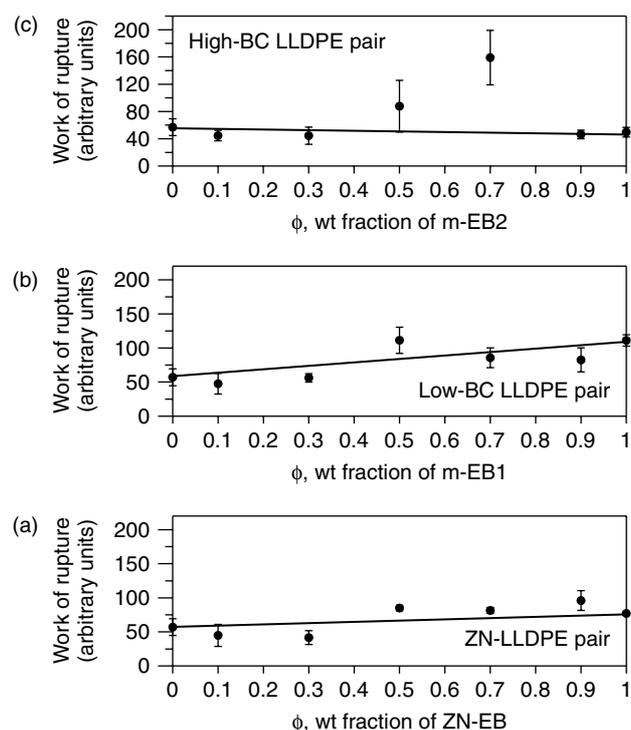


Figure 9. Work of rupture as a function of composition.

BC has a stronger influence on mechanical properties than CD.

## CONCLUSIONS

Blends of three different butene LLDPEs (two metallocene-based and one Ziegler–Natta-based) with similar  $M_w$  were melt blended with the same HDPE. The LLDPEs were selected in order to study the effect of branch content and composition distribution of LLDPE on thermal and solid-state properties of LLDPE/HDPE blends. Resins were paired to study one molecular parameter at a time while keeping other parameters very close to each other. The total crystallinity of all blends studied was found to follow a linear additivity rule and no effect for BC or CD was observed. A correlation between melt compatibility (or incompatibility) and mechanical properties of the low- and high-BC m-LLDPE blends with HDPE was observed. Miscible blends showed rheological<sup>3</sup> and many mechanical properties that can be estimated from additivity rules. High branch content of m-LLDPE was observed to influence the rheological and mechanical properties of its blends with HDPE. The effect is pronounced in the m-LLDPE-rich range. Elongation at break and work of rupture showed synergistic effects, while modulus was lower than predictions by linear additivity. Composition distribution showed no significant effect on thermal properties or on the work of rupture; however, it did result in low moduli for ZN-LLDPE blends with HDPE. Mechanical properties of LLDPE/HDPE blends are not a function of total crystallinity like LLDPE/LDPE blends<sup>39</sup>. It seems that the type and thickness of crystals formed in each case are different but this requires confirmation by scattering methods. Comparison of the DSC results and mechanical testing suggests that thermal analysis is inconclusive with regard to the compatibility of blends. In addition, total crystallinity does not correlate to melt or solid-state properties of LLDPE/HDPE blends. In conclusion, a direct correlation exists between melt compatibility (or incompatibility) and thermal and mechanical properties of LLDPE blends with HDPE, and branch content, in general, has a stronger influence on these properties than composition distribution.

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