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Liquid–liquid phase separation in ternary blends of linear polyethylene with two ethylene–butene copolymers

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A commercial ethylene–butene linear low density polyethylene has been fractionated by preparative temperature rising elution fractionation. Each fraction was characterized by Fourier transform infra-red spectrometry to determine the branch content and by gel permeation chromatography to determine the molecular weight. Two fractions were then blended, in various proportions, with each other and with a linear polyethylene. Using differential scanning calorimetry and transmission electron microscopy to examine quenched blends, the phase behaviour of this ternary system was investigated at temperatures above the melting point. The phase behaviour is very similar to that previously found for a ternary system of a linear polyethylene with two ethylene–octene copolymers with branch contents close to those of the ethylene–butene copolymers used in the present work. We deduce that the phase behaviour of blends containing ethylene–butene copolymers is essentially the same as that of blends containing ethylene–octene copolymers. We further deduce that the length of the branches must be of secondary importance in determining phase behaviour. (Previously we have shown that the variation of molecular weight has a secondary effect on phase behaviour). Thus we conclude that in blends of linear polyethylene with lightly branched ethylene copolymers it is the number of branches that is the most important factor influencing the extent of phase separation in the melt. © 1997 Elsevier Science Ltd. All rights reserved.

(Keywords: ethylene–butene copolymers; ternary blends; liquid–liquid phase separation)

INTRODUCTION

The phase behaviour of blends of linear with lightly branched polyethylenes has recently attracted considerable interest. It is difficult to detect any phase separation directly in the melt because of the similarity between the components (for example, the usual light scattering techniques are not sufficiently sensitive owing to the similarity of the refractive indices of the components). However, we have developed indirect techniques with which we have been able to deduce the phase behaviour of these blends in the melt (e.g. refs 1–11).

Our indirect methods for determining phase behaviour involve the examination of very rapidly quenched melts by differential scanning calorimetry (d.s.c.) and transmission electron microscopy (TEM). We have shown that after very rapid quenching the phase structure of the crystalline polymer closely resembles that of the melt. We have measured diffusion rates and shown that where, in the solid state, we observe well separated domains containing crystals of two distinct types, there must have been phase separation in the melt. We know this because the polymer would take some minutes to separate on the scale observed, whereas the time of the quench is less than a second[6].

Some workers have argued that the phase separation that we see must all take place on crystallization[12]. However, we do not believe this to be the case for three reasons. Firstly, as stated above, the diffusion rates are too slow to allow phase separation, on the scale observed, to take place during the time of the quench. Secondly, we observe that single uniform morphologies can be obtained by quenching some blends from higher temperatures, whereas biphasic morphologies are obtained on quenching the same blends from lower temperatures[5]; if all melts were mixed, and separation took place only on crystallization, this observation would be hard to explain. Finally, our experiments show that the average size of the dispersed phase increases with time in the melt, while the overall amount of the dispersed phase remains constant[11]. No such ripening process would take place if the blends were not separated in the melt. (A similar ripening process has been seen, by other workers using TEM, in blends of polyethylene with polypropylene and linear polyethylenes with highly branched copolymers[13–17].)

The only published attempts to search for melt phase separation in polyethylene blends using direct methods concern neutron scattering using blend systems where one component is deuterated[12,18–22]. However, unless experiments are carried out at very low angles the large scale phase separation that we believe to be present

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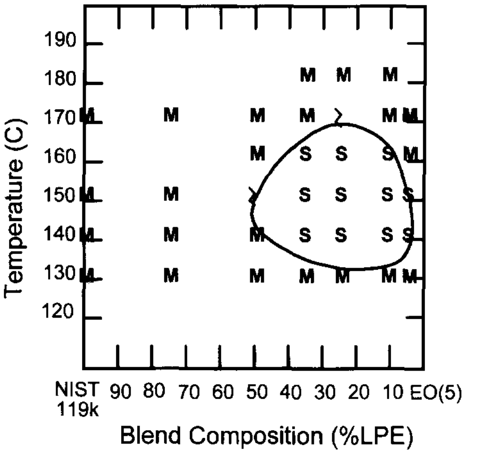

Figure 1 Binary phase diagram for blends of the linear, sharp linear polyethylene fraction NIST119K with the ethylene–octene copolymer EO(5). The characterization of these materials is as follows–NIST119K M_w 119 × 10^3, M_w/M_n 1.2; EO(5) M_w 37 × 10^3, M_w/M_n 2 , 5 mol% octene comonomer content. The LPE fraction was supplied by NIST, USA. The phase diagram was built up by sampling blends of various compositions, quenched from various temperatures, to see if the melt was mixed or separated. In this figure 'M' indicates that the melt was found to be mixed, and 'S' that it was found to be separated. ≷ indicates a borderline situation

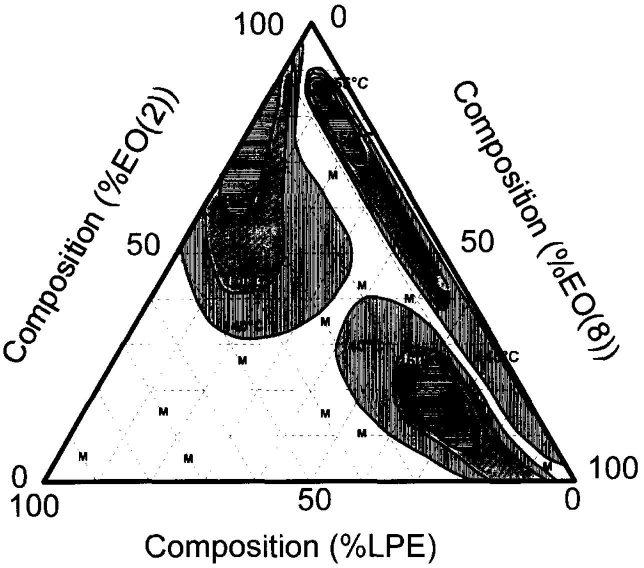

Figure 2 Ternary diagram for blends of a linear polyethylene with two ethylene–octene copolymers of differing branch content, 2 mol% and 8 mol%. This diagram is taken from ref. 9. The blends quenched from within shaded regions show two clear morphologies, and the temperature contour lines indicate the extent of these regions at the temperatures shown. The points marked 'M' are representative experimental points where a single crystal population was obtained on quenching from 140°C and 160°C. Hence, the melt from which the blend was quenched was judged to be mixed at all temperatures

cannot be detected by this technique[22]. Hence we believe that our indirect methods remain the most satisfactory for this work, and the only methods where neither of the blend components is deuterated.

Using our indirect techniques we have found phase separation in blends of linear polyethylenes (a number of linear polyethylenes of different molecular weights have been used), with a variety of branched polyethylenes. We have worked with a low density polyethylene containing both 'short' and 'long' branches[1–6], with well characterized, near random, ethylene octene copolymers[7,9] and with a number of linear low density polyethylenes (LLDPEs)[10]. Phase separation has also been found in binary blends of lightly branched polyethylenes[9–11]. In every case, where the molecular weight of the linear polyethylene was more than about 10^4, we have found melt phase separation of characteristic type–a loop of liquid–liquid phase separation (LLPS) at low linear polyethylene content. *Figure 1* shows a typical (but previously unpublished) binary phase diagram–that of the low polydispersity National Institute of Science Technology (USA) (NIST) 119K standard–blended with a near random ethylene–octene copolymer with 5 mol% branching. We have found that the extent of the phase separated loop depends only weakly on the molecular weight of the linear polyethylene[3,4]. However, at least for ethylene–octene copolymers blended with linear polyethylene, the extent of the phase separated loop depends strongly on the degree of branching of the copolymer[7,10]. The LLPS region was found to be wider when the copolymer contained less branches. This result was surprising at first sight, but can be understood in terms of a simple scheme based on energy considerations, if an extra, asymmetric free energy term is added to the usual Flory–Huggins model[8].

Having found that the closed loop of phase separation was general for binary blends, we began to investigate ternary blends. We studied a ternary system consisting of a linear polyethylene and two ethylene–octene copolymers of different branching[9]. The results, shown in *Figure 2*, were consistent with the results from all binary systems, and with the same simple explanatory scheme[8,9]. We went on to study phase behaviour when copolymers with bimodal branch distribution were blended with homogeneous polyethylenes (either linear polyethylenes or near random copolymers)[10]. We found quite complex phase behaviour which could, in each case, be simply regarded as a 'cloud point curve', i.e. a section across a ternary diagram. It was particularly interesting that the ternary diagram of *Figure 2*, measured for a linear/ethylene–octene system, could, equally well, be used to interpret the behaviour of linear/ethylene–butene and ethylene–octene/ethylene–butene cloud point curves. This suggested that ethylene–butene copolymers behave in a way very similar to ethylene–octene copolymers. At that time we did not have well characterized fractions of ethylene–butene copolymers. We have now obtained such ethylene–butene copolymers by preparative temperature rising elution fractionation (TREF), characterized them and determined a ternary phase diagram for linear/ethylene–butene polyethylene blends. In this paper we report our results. We have already shown that, for ethylene–octene copolymers blended with linear polyethylene[7], and for a range of linear polyethylenes blended with the same low density polyethylene[3,4], the molecular weights of the constituent materials affect the phase behaviour much less than the branch content. We now investigate the effect of decreasing the branch length from 6 to 2 carbons.

EXPERIMENTAL

The materials blended were the linear polyethylene Sclair 2907 (LPE) and fractions of a commercial ethylene–butene LLDPE labelled as EB(3.7), because it contained 3.7 mol% butene copolymer overall. The details of these polymers, and the fractions used for the research, are

Table 1 Details of the polymers used in this work

Polymer	M_w	M_w/M_n	Branches, mol%	Melting peak after quenching from 140°C
Sclair 2907	10^5	4.3	0	129.0
EB(3.7) whole	1.1×10^5	3.6	3.7	122/117[a]
EBb(2.1)	0.92×10^5	4.2	2.1	119.2
EBb(8.0)	1.1×10^5	2.5	8.0	101.6
EBn(1.7)	10^5	3	1.7	116.2
EBn(6.0)	10^5	3	6.0	93

[a] Two melting peaks are seen in quenched EB (3.7)

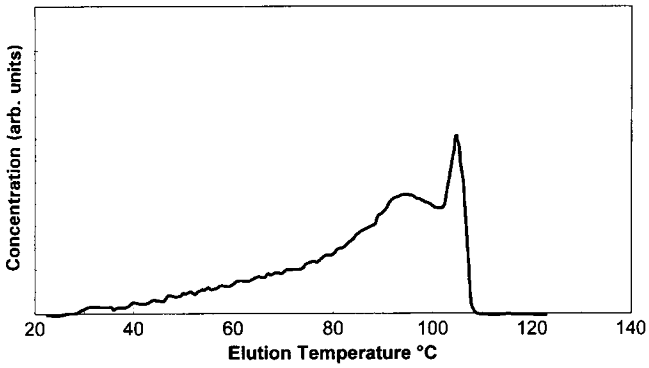

Figure 3 Analytical TREF data for EB(3.7). Higher elution temperatures correspond to materials of increasingly lower branch content

given in *Table 1*. *Figure 3* shows the analytical TREF curve of EB(3.7).

EB(3.7) was fractionated, by branch content, using preparative TREF. During this process the material is fractionated by selective dissolution of molecules deposited in a column filled with glass beads of diameter ~ 300 μm. Two fractionations were carried out, under different conditions. In the first, the LLDPE was divided into three fractions of relatively broad branch content. Reasonably large quantities of material were obtained in this process, as required to investigate ternary phase behaviour. The column was loaded with (450 ml of a 0.4% w/v) solution of EB(3.7) in xylene. The solvent contained 200 ppm Santanox R antioxidant. The column was cooled from 120°C to below 30°C at 5° h^{-1}. A stepwise temperature profile was used during elution, to obtain three fractions, and a thermocouple probe was inserted into the base of the column to keep a check on the temperature. The column was flushed for 1 h at 75°C, $\frac{3}{4}$ h at 90°C and 1 h at 120°C, with a solvent input rate of 15 ml min^{-1}. We also wished to produce small quantities of sharper fractions. Nine such fractions were obtained. The temperature profile given to the oven that contained the column was again stepwise. However, measurements with the thermocouple probe showed that due to thermal lag between the column and the oven, a continuous ramp in temperature was actually applied to the column. A solvent input rate of 15 ml min^{-1} was again used and each fraction was collected over a time of 30 min. Both sets of fractions were recovered from solution by the addition of excess non-solvent, acetone, followed by filtration of the precipitate. All samples were dried under vacuum for 15 h at 70°C.

The fractions were characterized by gel permeation chromatography (g.p.c.) for molecular weight, and by Fourier transform infra-red spectrometry for branch content. Spectra were obtained from melt pressed samples of the fractions, each film being approximately 150 μm thick. The branch content calculation was based on the absorbance at 1379 wavenumbers, which was measured in each case after the subtraction of the spectrum of a high molecular weight linear polyethylene. Five ethylene–butene copolymers of known branch content (previously measured by ^{13}C nuclear magnetic resonance) were used for calibration and an end chain correction, based on number-average molecular weight, was performed for each sample.

We chose to investigate the phase behaviour of ternary blends of the linear polyethylene Sclair 2907, with two of the broader fractions of EB(3.7) We labelled the fractions 'EBb' for broad fractions and 'EBn' for narrow fractions, with the branch content in parentheses. Thus the two materials chosen for the ternary study were EBb(2.1), the broad fraction with 2.1 mol% butene comonomer content, and EBb(8.0), the broad fraction with 8.0 mol% butene comonomer content. We had reasonable quantities of these two fractions, their molecular weights were similar to that of the linear polyethylene, Sclair 2907, and the branch contents were identical, within experimental error, to those of the ethylene–octene copolymers used to plot *Figure 2*, the linear/ethylene–octene ternary phase diagram (2.1 and 8.0 mol% for the ethylene–butenes and for the ethylene–octenes[9]).

Between 6 and 12 blends were made for each binary pair (LPE/EBb(2.1), LPE/EBb(8.0) and EBb(2.1)/EBb(8.0)) and 40 further blends were needed to examine the interior of the ternary phase diagram. Of each blend, 10–50 mg was made. Our method of blend preparation has been discussed before (see refs 1–4, especially ref. 4). Briefly, the materials, in the correct proportions, are dissolved together in xylene to make a 0.3% solution. They are then precipitated by quenching into cold acetone. We have found this method of blending to be very satisfactory[1–6].

Our TEM techniques are described in detail in refs 1 and 2, and our d.s.c. method is discussed in ref. 23; the methods are only summarized here. We prepare surface replicas of quenched blends and examine them using TEM. We choose surface replicas because the surface is the part of the sample experiencing the fastest quench, and so nearest in phase structure to the melt. If we see two distinct crystal types, usually groups of thicker crystals well separated in a matrix of thinner crystals, we judge the blend to have been separated in the melt. If

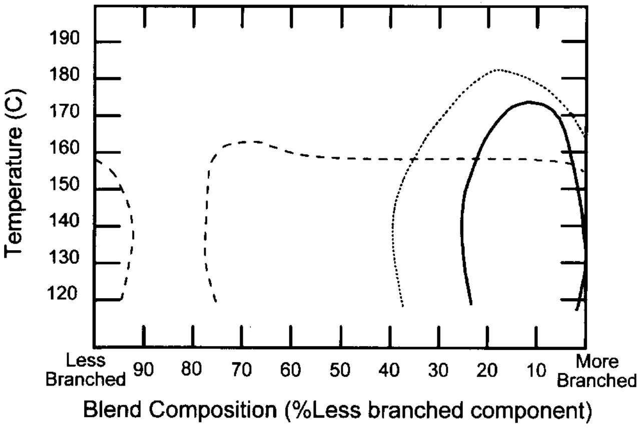

Figure 4 Binary phase diagram for the three blend systems forming the binary sides of the LPE/EBb(2.1)/EBb(8.0) ternary system. Sclair 2907 with EBb(2.1), dotted line; Sclair 2907 with EBb(8.0), solid line; and EBb(2.1)/EBb(8.0), dashed line. Note that this last binary system has two phase separated regions

Figure 5 TEM micrograph of a replica of the fraction EBb(2.1), after isothermal crystallization at 120°C for 60 h. No further crystallization is observed in samples that have been held isothermally at 120°C for 1 week; all possible crystallization has taken place after 60 h. The polymer which has crystallized isothermally is found in large spherical domains; this is clear evidence of phase separation in the melt from which the crystallization took place[2,6]. The scale bar represents 1 μm

only one crystal type is present then we judge the blend to have been mixed before quenching. On heating quenched samples in the d.s.c. we look at the melting peaks; in these samples the presence of two melting peaks indicates two crystal populations and one melting peak indicates one crystal population. (We are aware of the existence of annealing effects, which can give rise to two peaks on heating[24–26]; however, we are satisfied that in our samples the two peaks, when observed, are not a consequence of annealing[23,27]. We have found our TEM and d.s.c. methods to be in good agreement in all but a few special (and well understood) cases (see refs 1–11 and especially ref. 23).

We studied samples of each of the 72 blends by TEM, to see whether they were mixed or separated prior to quenching from 140°C and 160°C. We also looked at a few blends quenched from higher temperatures in order to map out the tops of the LLPS regions. We looked at each blend by d.s.c. heating at 10°C min^{-1} under nitrogen, after quenching from 140°C.

Some of our results (see below) led us to believe that EBb(2.1), one of the broad fractions used for this initial work, was itself somewhat bimodal, so that we would not be able to obtain the complete ternary phase diagram. To check this supposition (which is in line with results in the literature[28]) some blends were made using two of our limited supplies of narrow TREF fractions. The fractions chosen were EBn(1.7) (near to EBb(2.1) in branching) and EBn(6.0) (fairly near to EBb(8.0) in branching). Details of these materials are shown in *Table 1*. Only the LPE/EBn(1.7) and EBn(1.7)/EBn(6.0) binaries were explored because we had very limited supplies of these narrow fractions.

RESULTS

We found that the d.s.c. results were generally in agreement with the TEM results. However, phase separation was difficult to detect by d.s.c. in the LPE/

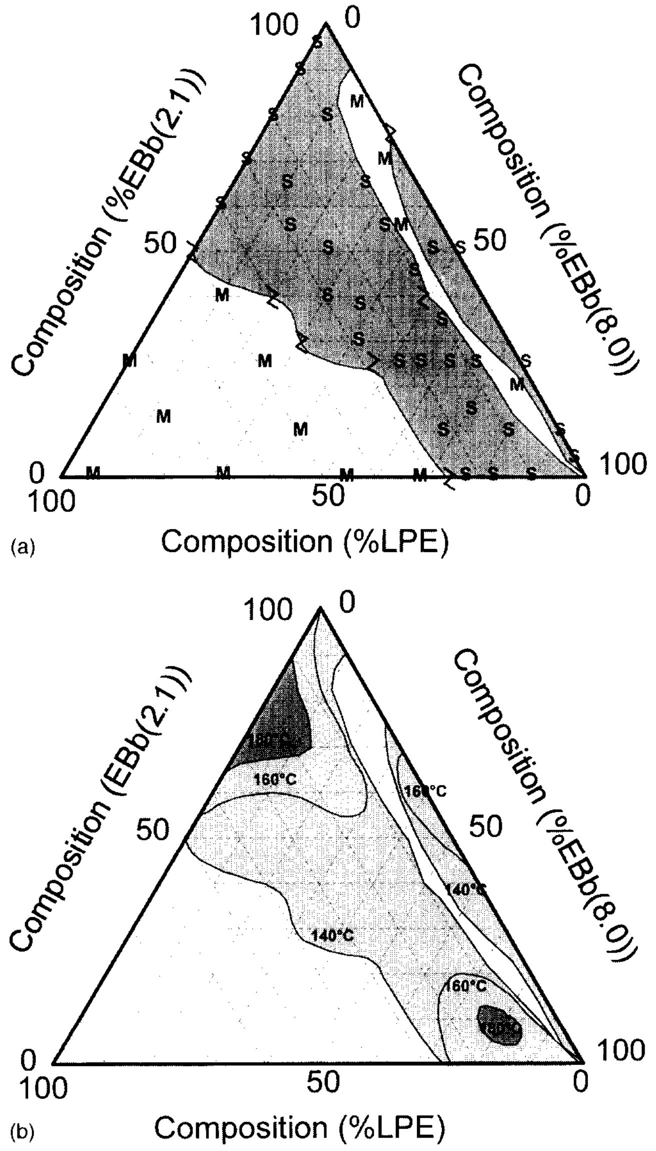

Figure 6 Contours through the LPE/EBb(2.1)/EBb(8.0) ternary system. (a) 140°C. Shaded regions are found to be phase separated in the melt at 140°C. (b) High temperatures. The unshaded region is not found to be phase separated at 140°C and above, the lowest contour shows separation at 140°C, the middle contour separation at 160°C and the highest contour (not present in the finger region) phase separation at 180°C

EBb(2.1) binary system because of the closeness of the melting points of the two materials and the rather wide endotherm obtained from melting EBb(2.1) alone. Nevertheless, phase separation could be clearly detected from the micrographs, showing that TEM is more sensitive for the study of these LPE/EBb(2.1) blends (as it is for other blends where the comonomer content is low[27]). For the other two binary systems, and for all ternary blends examined, d.s.c. was clear and in agreement with TEM.

The phase behaviour, along each of the three 'binary sides' is shown in *Figure 4*. The behaviour along the LPE/EBb(2.1) and LPE/EBb(8.0) sides is as we have found before—in each case a loop, at low LPE content, and a wider loop where the copolymer is less branched. The phase separation along the EBb(2.1)/EBb(8.0) side is unexpected, and more complex. There are two LLPS loops, a very broad loop, extending from pure EBb(8.0) to a blend with 75% EBb(2.1), and an unexpected narrow region around EBb(2.1). It appears that EBb(2.1) phase separates in itself at certain temperatures. *Figure 5* shows this very clearly–EBb(2.1) was crystallized isothermally for 60 h at 120°C before quenching. The resulting morphology has groups of thicker crystals, crystallized from phase separated droplets, in a matrix which crystallized into small, thin crystals on quenching. This morphology is a clear indication of phase separation at the crystallization temperature[1–6].

The full ternary phase diagram for the LPE/EBb(2.1)/EBb(8.0) system is shown in *Figure 6*. *Figure 6a* shows the phase separated regions at 140°C, *Figure 6b* the separation contours at 140°C and above.

The binary phase diagrams for the LPE/EBn(1.7) and EBn(1.7)/EBn(6.0) systems are shown in *Figure 7*. Note that, unlike EBb(2.1), EBn(1.7) does not itself phase separate at any temperature, and that the EBn(1.7)/EBn(6.0) system only shows phase separation at low EBn(1.7) content. These two binary phase diagrams are very like those found for the equivalent linear/ethylene–octene and ethylene–octene/ethylene–octene systems.

DISCUSSION

A comparison of *Figures 6b* and *2* immediately shows that the linear/ethylene–butene ternary phase diagram (*Figure 6b*) is similar to the linear/ethylene–octene ternary phase diagrams (*Figure 2*). In both cases there is mixing for blends with high linear polyethylene content and extensive separation for blends with low linear polyethylene content. All the phase separated regions are upper critical, and the loops are wider where the copolymer contains fewer branches. In both these ternary diagrams (i.e. both the linear/ethylene–butene and the linear/ethylene–octene), the molecular weights of the three components are nearly equal–this should minimize any differences between the two systems with regard to molecular weight[7]. However, note that the molecular weights of the materials used for the ternary linear/ethylene–butene phase diagram are notably higher than those used for the linear/ethylene–octene ternary, and that all the polymers in the linear/ethylene–butene system have higher polydispersities than those in linear/ethylene–octene system. The widths of the LLPS regions along the linear/ethylene–butene binary sides are, roughly, as would be predicted from the linear/ethylene–octene system[7–10].

There are, however, two obvious differences between the phase behaviour of the two systems. Firstly, in the linear/ethylene–butene ternary phase diagram the two loops of LLPS extending from the linear/copolymer binary sides join up, but in the ethylene–octene system they do not. Secondly, the phase separation along the

Table 2 Molecular weights of the polymers used to determine the phase diagrams in *Figures 2* and *6–8*

Polymer	M_w	M_w/M_n
Sclair 2907, the linear polyethylene used to determine the linear/ethylene–butene ternary	10^5	4.3
EBb copolymers used to determine the linear/ethylene–butene ternary	10^5	4
The linear polyethylene used to determine the linear/ethylene–octene ternary	5×10^4	2.8
Ethylene–octene copolymers used to determine the linear/ethylene–octene ternary	5×10^4	2

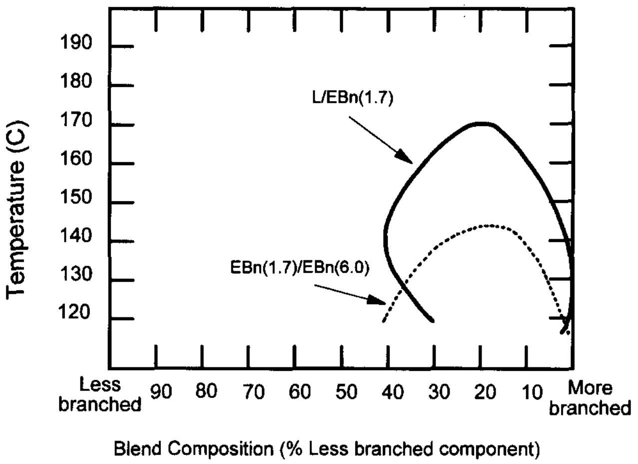

Figure 7 Phase behaviour of the Sclair 2907/EBn(1.7) and EBn(1.7)/EBn(6.0) binary systems. The regions of melt within the loops are judged to be phase separated

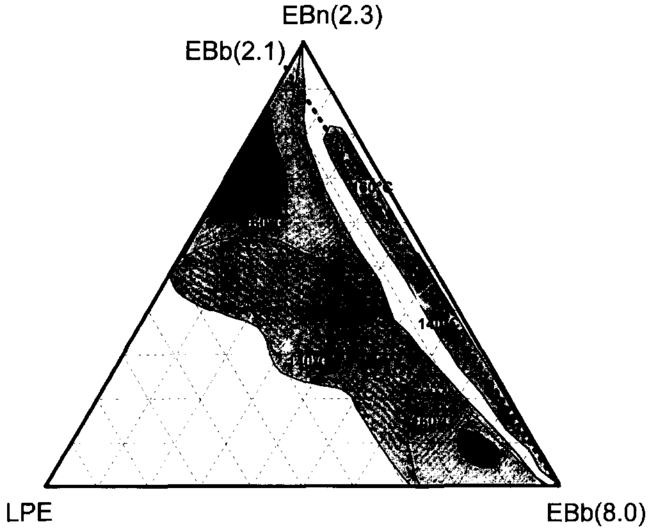

Figure 8 The full ternary phase diagram for a LPE/EBn(2.3)/EBn(8.0) ternary system. The part that was explored using EBb(2.1) and EBb(8.0) is on the linear polyethylene side of the thick dotted line between EBb(8.0) and EBb(2.1)

copolymer/copolymer binary side of the linear/ethylene–butene ternary phase diagram is much more extensive than that along the copolymer/copolymer binary side of the linear/ethylene–octene ternary diagram.

The first feature, the joining up of the two LLPS loops from the linear/copolymer binary sides, should be considered in the light of our simple model[8–10]. We have shown previously[9,10] that such behaviour is to be expected, on the basis of our simple model, when the components of a ternary system have similar copolymer contents. To a first approximation, if there is no change in phase behaviour as a result of copolymer type, we would expect the predicted phase diagrams to be the same for the linear/ethylene–octene and the linear/ethylene–butene ternary systems, because the branch contents are very similar. In fact, the two large LLPS regions are just separate in the linear/ethylene–octene ternary, but are joined in the linear/ethylene–butene ternary system. Although the branch contents of the components in the two systems are very similar, the systems are different in several other respects: branch type; breadth of branch distribution (the EBb copolymers have wider distributions than the ethylene–octenes); weight-average molecular weight and molecular weight distribution. All the components of the linear/ethylene–butene system have higher weight-average molecular weights and broader distributions than those in the linear/ethylene–octene system, see *Table 2* for details. We have previously shown that the phase separation is slightly less widespread where the linear polyethylene has a lower weight-average molecular weight[3,4]. We have no quantitative data concerning the effects of the other three factors, although it is clear from our past work[3,4,7,10] that none of them affects the phase behaviour strongly. We can safely say that the two larger LLPS regions in the linear/ethylene–octene and linear/ethylene–butene ternary phase diagrams of *Figures 2* and *6c* are similar, but not the same. There are four possible physical factors that may cause this small change, and we are not at present in a position to tell which factor, or which combination of factors, is most important. However, the evidence is that the variation in phase behaviour that we see here is small in comparison with changes observed as a result of changes in branch content.

We believe that the second difference arises from the imperfect nature of our fraction EBb(2.1) (compared with the two ethylene–octene copolymers). EBb(2.1), as noted above, is shown to be separated when crystallized isothermally at 120°C (*Figure 5*) and when quenched from temperatures up to 150°C. We believe that this fraction is best regarded as a binary blend itself. The overall branch content is 2.1 mol% so one of the components of EBb(2.1) must have a lower branch content than 2.1 mol%; it will be lower in branch content than either of the two branched components of the resultant ternary phase system, i.e. near to zero branch content (linear polyethylene). (In confirmation, the d.s.c. melting trace of isothermally crystallized EBb(2.1) shows a high melting peak very close to that of linear polyethylene.)

Work with the sharper fractions, EBn(1.7) and EBn(6.0) confirms that the imperfection of EBb(2.1) influences the ternary phase diagram, *Figure 6b*. EBn(1.7) did not crystallize at all when held isothermally at 120°C for 60 h. Unfortunately, we did not have enough of the fractions EBn(1.7) and EBn(6.0) to plot a complete ternary phase diagram, so binary phase behaviour was investigated. We looked at LPE/EBn(1.7) and EBn(1.7)/EBn(6.0) binary systems (the linear material being Sclair 2907, as before). The results, shown in *Figure 7*, show single regions of LLPS at low contents of the less branched materials. These binary phase diagrams are as would be expected from our previous work using near random ethylene–octene copolymers[7,9,10].

If EBb(2.1) is bimodal, and acting as a blend containing some linear material, then *Figure 6b* is not a complete ternary phase diagram for three well defined, unimodal materials. D.s.c. of isothermally crystallized EBb(2.1) indicates that the linear content is about 7% by weight, so we can place EBb(2.1) appropriately along an LPE/EB binary line, as in *Figure 8*–the branch content of the constituent EB will be rather more than 2.1 mol%, we estimate that it will be about 2.3 mol%. On the basis of our considerable experience with phase diagrams of this type, we believe that the full phase diagram for a LPE/2.3/8.0 mol% ethylene–butene system, with unimodal, random copolymers is as shown in *Figure 8*. Because of our imperfect EBb(2.1) fraction, we have access to most, but not all, of the full ternary phase diagram for pure components–only the region of the LPE side of the line between EBb(2.1) and EBb(8.0) in *Figure 8* can be reached. The LPE/EBn(1.7) and EBn(1.7)/EBn(6.0) binaries confirm that for unimodally branched ethylene–butene copolymers the binary sides are similar to those for ethylene–octene copolymers and in line with the prediction of *Figure 8*. Hence we are confident that the full phase diagram for a linear/ethylene–butene ternary system, with copolymers of about 2.3 and 8.0 mol% branching, would be as shown in *Figure 8*.

Thus it appears that the phase behaviour of the linear/ethylene–butene ternary system is very much as would be predicted for a linear/ethylene–octene ternary system of similar branch content. Previously, we have shown that the variation of molecular weight has a secondary effect on phase behaviour[3]. Now we have shown that (for alkane branches of between 2 and 6 carbons) the length of the branches is also of secondary importance in determining phase behaviour. At least in this branch length range, it appears to be the actual number of branches that is the key variable in determining the phase behaviour. Previously we worked with a low density polyethylene with both 'long' and 'short' branches; the phase behaviour of this material, when blended with any linear polyethylene that we tried, was close to that of our ethylene–octene copolymer with similar branch content. This leads us to believe that branch content is also the critical factor outside the range of branches of 2–6 carbons in length. We conclude that it is *the density of branches along the polyethylene chain* that is the dominant factor giving rise to the phase separation that we see in systems where linear polyethylenes are blended with lightly branched alkane copolymers. As yet we are not yet clear if the number of actual branch points or the number of chain ends is the key feature giving rise to phase separation.

CONCLUSIONS

- The phase behaviour of blends containing ethylene–butene copolymers is essentially the same as that of blends containing ethylene–octene copolymers.

- The length of the branches is of secondary importance in determining phase behaviour. (Previously we have shown that the variation of molecular weight has a secondary effect on phase behaviour[3,4].)
- In blends of linear polyethylenes with lightly branched copolymers it is the number of branches that is the most important factor influencing the extent of phase separation.

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