

## Stress-Strain Behavior of Polyolefin Blends

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### Synopsis

As part of a study on reuse of plastics as blends, the yield tensile strength, elongation at break, and the modulus of melt blends of low-density polyethylene, high-density polyethylene, and polypropylene have been studied over the entire ternary composition range. The modulus and strength are nearly monotonic functions of blend composition. The contribution of the pure components to these properties is roughly additive. The elongation at break is a more complex function of composition in that minima are observed near the center of the triangular composition diagram and on each of the three binary legs. The response is nearly symmetrical along two of the binary legs but is skewed toward high-density polyethylene for blends of high- and low-density polyethylene. The deterioration of elongation produced by blending is much less severe for polyolefins than observed for other blend systems. This, combined with the observed additivity of strength, make polyolefin blends mechanically superior to blends of other plastics found in wastes. This fact is interpreted in terms of compatibility and amorphous phase interactions which are likely for polyolefins. Modification of polyolefin blends by addition of a rubbery copolymer of ethylene and propylene produced large improvements in elongation at break for some compositions.

### INTRODUCTION

Previous publications<sup>1-4</sup> have examined certain aspects of the reuse of waste plastics by reprocessing mixed generic types to form a polymer blend which may have some value as a material. This route of dealing with waste plastics makes a trade-off between the difficulties of generic segregation<sup>5</sup> and a sacrifice in mechanical properties owing to the incompatibility of the various plastics in the blend.<sup>1</sup> The composition of reclaimed plastics for reuse will depend on where they are generated, municipal, commercial, or industrial, and how they are isolated. Generally, the low cost materials such as polyolefins, polystyrene, and poly(vinyl chloride) comprise the major volume of waste, and a reprocessed blend may contain significant portions of each type.<sup>1</sup> Certain instances, however, may produce a more refined mixture in that certain types are excluded. The object of this paper is to explore the case where the predominate ingredients of such a blend are the polyolefins: low-density polyethylene (LDPE), high-density polyethylene (HDPE), and polypropylene (PP), since some wastes consist almost entirely of these ingredients.<sup>3</sup>

Various aspects of a number of polyolefin blends have been dealt with previously in the literature. A brief summary of this literature as it relates

to the current interest will be given in the next section. Very little information is available on the ultimate mechanical properties of these blends. Knowledge of the ultimate behavior is of particular importance in surveying the potential use of blends since often this is their most deficient characteristic. A blend of two incompatible polymers such as polyethylene and polystyrene may have such poor ultimate mechanical properties that the material is of very little value.<sup>1</sup> Polyolefin blends, on the other hand, should have properties quite superior to this example owing to the more similar chemical structures of the blend ingredients. It is the purpose of this paper to present such data for ternary blends of HDPE-LDPE-PP over the entire composition map so that quantitative evaluation of these materials is possible. In general, it is found that these blends have quite good properties in comparison to other systems; however, certain compositions suffer a considerable loss in toughness. The use of a rubbery ethylene-propylene copolymer, EPR, as a blend additive was explored as a way to improve this deficiency.

### PREVIOUS WORK ON POLYOLEFIN BLENDS

Blends that include some combination of LDPE, HDPE, and PP plus other polyolefins have been the subject of commercial interests<sup>6,7</sup>; however, the extent to which such blends have been commercially implemented is difficult to document. One measure of this interest is the patents issued on polyolefins blends which describe specific benefits to be accrued by blending a particular composition. Examples of some of the claimed benefits include improved impact strength,<sup>8-11</sup> shear strength,<sup>12</sup> environmental stress-cracking resistance,<sup>13,14</sup> clarity,<sup>15</sup> gloss,<sup>16</sup> resistance to thermal embrittlement<sup>12</sup> as well as to lowering the brittle point,<sup>17,18</sup> increase the crystallization rate,<sup>19</sup> and to make better extrusion coating resins.<sup>20</sup>

A number of fundamental studies of polyolefin blends have been reported in the literature,<sup>21-40</sup> and a few important features of this work will be summarized. Bohn,<sup>41</sup> in a comprehensive review, has listed all the possible binary combinations of HDPE-LDPE-PP as incompatible and suggests that this is to be expected for blends composed of crystalline components. However, determination of miscibility of polymer pairs in blends is not a simple matter, especially when there are crystalline components, and a simple designation of compatible or incompatible is probably not an adequate description in many instances.

Most of the evidence for designating these systems as incompatible stems from observation of the individual melting points of the components in the blend<sup>21-26</sup> by techniques such as differential thermal analysis. Such studies have shown the individual melting endotherms in PP and polyethylene blends<sup>25</sup> and even in HDPE and LDPE blends.<sup>21,22</sup> There is a suggestion of shifts in the temperature at which these peaks occur<sup>21</sup> in the latter system which might be interpreted as evidence for some crystalline mixing or cocrystallization, or alternately it might be owing to the difficulty

in resolving two peaks which are close together. Clampitt<sup>22</sup> suggests the presence of a cocrystal on the basis of an intermediate third peak he observed in HDPE-LDPE blends; however, it is likely that this peak is a morphologic artifact of an annealing procedure used in that study. Another study<sup>26</sup> reveals that per cent crystallinity of HDPE-LDPE blends as judged by density, IR, and NMR is an additive function of the crystallinity of the individual components. This provides no detailed insight except to say that blending does not decrease or increase the amount of material that crystallizes.

From the above it is clear that probably HDPE and LDPE in blends crystallize independently with only minimal interaction. However, what is not clear from any work in the literature is what happens in the amorphous phase. This phase is more ambiguous in HDPE than LDPE due to the more complex physical picture required for highly crystalline polymers. Any type of interaction in noncrystalline regions could be very important to the mechanical behavior of blends. Unfortunately, the pragmatic mechanical property data reported here do not shed any definitive light on such interactions; however, these fundamental questions do underlie the current observations and thus attention is called to them.

The literature also contains some studies of dynamic mechanical properties.<sup>24,27-32</sup> This approach observes more subtle solid-state transitions than just melting, and often valuable clues about compatibility can be gained by observing how these transitions behave in blends as compared to pure components. The results of these limited studies substantiate the thermal analysis conclusions of incompatibility in polyolefin blends. However, the range of compositions studied by this technique is not extensive nor have the analyses of current experiments been as refined as might be required to reach alternate conclusions.

Melt rheology of blends potentially contains valuable information about miscibility in the molten state which may then be used to infer something about amorphous-phase interactions in the solid state. Some information is available about the melt rheology of polyolefin blends.<sup>29,33-36,42</sup> However, as will be discussed later, any conclusions derived regarding miscibility should be viewed with caution owing to both insufficient data and inadequate techniques for analyzing it.

Reports on the stress-strain behavior of polyolefin blends is quite limited, and what data are available<sup>24,28,29,37,38,42</sup> do not cover adequate composition ranges or employ comparable conditions to allow comparisons or specific definitive statements. In general, it is noted that the mechanical properties of polyolefin blends are quite good, especially in comparison to grossly incompatible systems.

The fact that polyethylene and polypropylene do not cocrystallize is easily understood from the differences in their molecular structure. A lack of gross miscibility would also be expected for the same reason.<sup>41</sup> In the case of HDPE and LDPE, the structural differences are less dramatic. For a simple model, branched polyethylene may be viewed as an ethylene

copolymer with  $\alpha$ -hexene, whereas HDPE is an ethylene homopolymer. A typical LDPE with two branches per 100 carbon atoms<sup>43</sup> would then correspond to a copolymer with 4% (mole)  $\alpha$ -hexene. This amount of copolymerization will dramatically alter the fraction of the polymer that can crystallize and the melting point as is well known for polyethylene. It is generally regarded for LDPE, however, that only ethylene sequences of the chain crystallize. Consequently, the crystallizable parts of LDPE are structurally identical to HDPE, and that these two do not cocrystallize in blends is apparently for reasons of morphology or unfavorable surface effects. Aside from crystallizability, it is not at all clear as to whether a 4% difference in copolymer content is grounds for immiscibility. Molau<sup>44</sup> has reported that styrene-acrylonitrile copolymers are compatible, provided the comonomer contents of two polymers do not differ by more than about 4%. The critical composition difference in the methyl methacrylate-*n*-butylacrylate system is reported at 10% to 25%.<sup>45</sup> Of course, in the solid state, the branch points of LDPE are concentrated in the amorphous phase, so the difference may be larger than 4%. From this information it is hard to make a clear-cut case for amorphous-phase miscibility in the LDPE-HDPE system; however, it does appear rather certain that there will be very strong interactions which can provide mechanisms for good blend properties. Evidence of this is provided by the adhesion between LDPE and HDPE.<sup>1</sup>

## EXPERIMENTAL

Virgin commercial resins were used to generate melt blends for property evaluations. Table I provides a brief description of these materials which were all supplied in pellet form. Subsequent references to these materials will employ their commercial designations. The low-density polyethylenes DYNH and DFD are equivalent in terms of mechanical properties and will be used interchangeably in various tests.

Melt blending was accomplished in a Brabender Plasticorder using procedures similar to those described previously,<sup>1</sup> except that a temperature of 190°C was used to facilitate the melting of polypropylene. Dogbone specimens were cut from sheets compression molded from such blends and

TABLE I  
Polymers

Generic type	Designation	Source	Description
Low-density polyethylene	DYNH	Union Carbide	Den = 0.917 g/cc MI = 1.2
Low-density polyethylene	DFD	Union Carbide	Den = 0.917 MI = 2.0
High-density polyethylene	DGDA	Union Carbide	Den = 0.964 MI = 0.2
Polypropylene	PP E-115	Enjay	standard molding grade MI = 4.4

were tested by an Instron at a cross-head speed of 0.2 in./min as before.<sup>1</sup> Yield tensile strength, elongation at break, and initial modulus were computed from the Instron chart. For each property, an average from at least four samples (and often many more) was computed. In subsequent graphic presentations, the range of these determinations are shown to illustrate the variation observed except in cases where the range is too small to illustrate in this way.

### RESULTS FOR UNMODIFIED BLENDS

Figure 1 provides a summary of the stress-strain properties of melt blends of HDPE-LDPE-PP on a triangular diagram. The position of the data boxes indicates the composition of each blend. The boxes contain average values of the tensile yield stress, the elongation at break, and the initial modulus taken from the Instron record. A more detailed examination of performance along the binary legs will be provided by graphical presentations.

The strength of the blends varies rather uniformly across this diagram between the limits set by the pure components located at the apices of the triangle. No dramatic minimum is apparent, which is in contrast to similar diagrams for ternary blends of PE-polystyrene-PVC reported earlier.<sup>1</sup> In the latter case, a minimum was located near the center of the

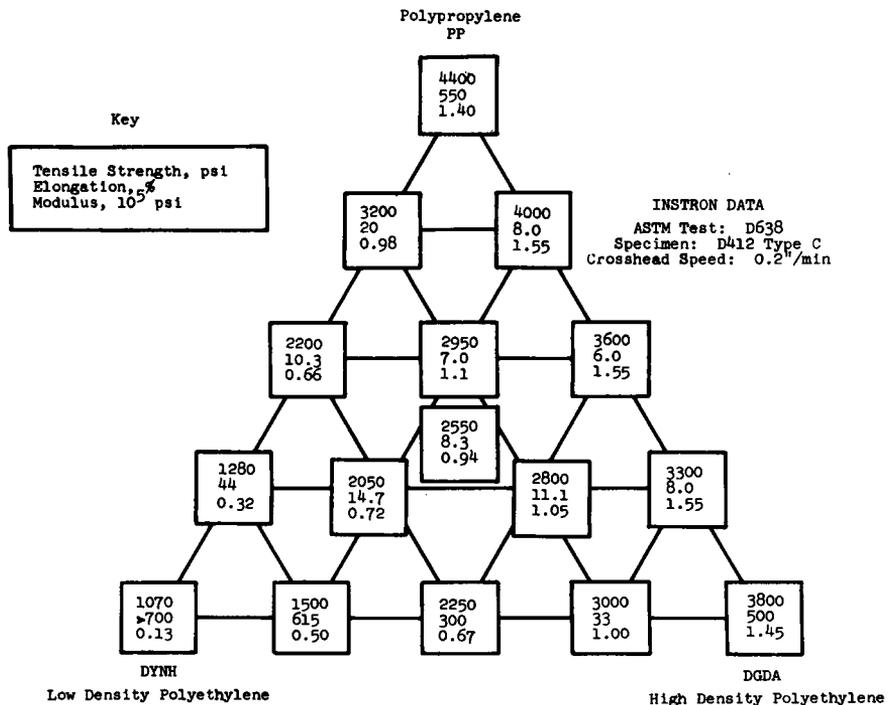


Fig. 1. Mechanical properties of unmodified polyolefin blends.

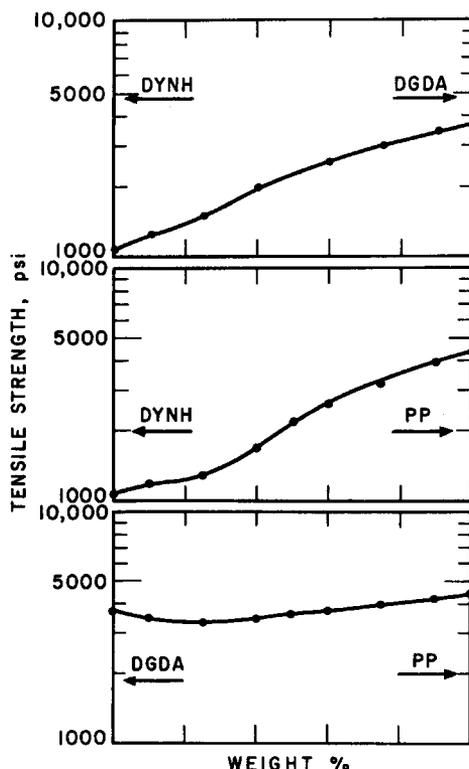


Fig. 2. Yield tensile strength for binary polyolefin blends. DYNH is a low-density polyethylene, DGDA is a high-density polyethylene and PP refers to polypropylene.

triangle which dipped to strength values well below that of the weakest component. The more nearly additive strength behavior of the current polyolefin blends is illustrative of the superior properties of these blends.

Figure 2 shows further evidence of this near additivity of strength along the three binary legs. The high-density polyethylene, DGDA, and polypropylene (PP) are, of course, considerably stronger than the low-density polyethylene, DYNH. There is a slight minimum noted for the HDPE-PP binary, but the strengths for the two binaries with LDPE depart only slightly from a straight line connecting the pure component properties.

The behavior of the elongation at break as seen in Figure 1 is more complex. All points away from the apices have a lower elongation at break than any of the pure components and dip to a minimum near the center of the triangle. Even this central point is considerably better, however, than the same property at this location for PE-polystyrene-PVC blends.<sup>1</sup> This is further evidence of the superiority of polyolefin blends.

Figures 3-5 show how the elongation at break depends on blend composition along the three binary legs. In each case a pronounced minimum is observed, which is considerably lower in magnitude than observed for the pure materials. Comparison of this property of the blends with the pure

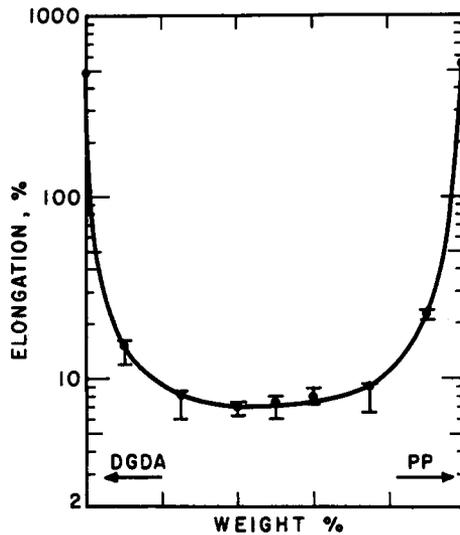


Fig. 3. Elongation at break for high-density polyethylene (DGDA) and polypropylene (PP) binary blends.

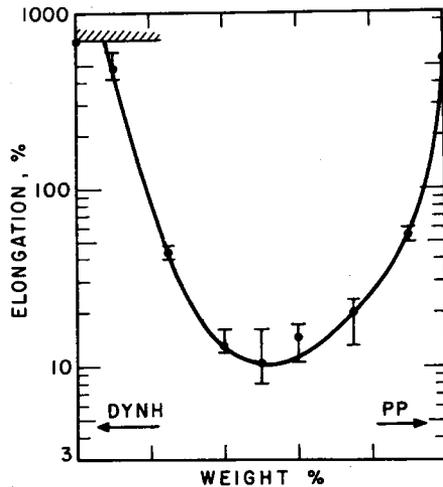


Fig. 4. Elongation at break for low-density polyethylene (DYNH) and polypropylene (PP) binary blends.

components as shown here is somewhat misleading and deserves further comment. The indicated elongation for DGDA and PP is about 500% and in excess of 700% (maximum cross-head travel) for DYNH. These values exist because the sample necks and cold-draws. Most of the blends do not neck but break somewhere in the vicinity of the yield point, generally where the stress goes through a maximum. Necking and cold-drawing is not ordinarily a usable property of an end product, so it may not reflect a serious material limitation unless cold-drawing is a required

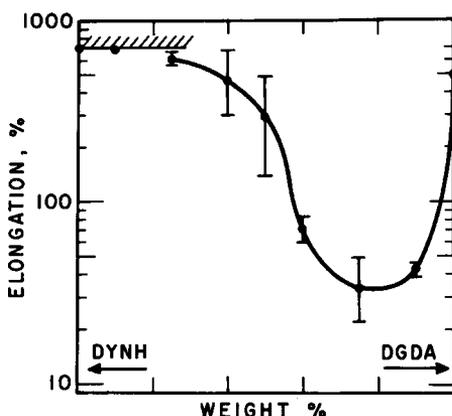


Fig. 5. Elongation at break for low-density polyethylene (DYNH) and high-density polyethylene (DGDA) binary blends.

part of fabrication. Therefore, while numerically the reduction in ultimate elongation produced by blending is large, it represents more the loss of a phenomenon rather than the continual diminution of a property. The ability to neck and draw is evidently very defect or morphology sensitive. In fact, this ability can be absent in both pure DGDA and PP. It was observed that among a large number of DGDA or PP specimens prepared in what should have been a comparable manner, there was always a significant fraction which would not draw after yielding. Instead, they broke in the vicinity of the yield point at an elongation of about 20% to 50%. It is surmised that a rather small morphologic difference accruing from slightly different thermal histories or defects caused this. Such defects seem to be assured in blends. It would perhaps be more realistic to compare blends with samples of the pure components that did not draw, in which case the detrimental effect of blending would not seem very large at all. DYNH, however, does not seem to suffer this problem because all samples could be drawn to the full cross-head travel of the Instron ( $\sim 700\%$ ).

The minimum of Figure 3 is quite broad, but the diagram is more or less symmetrical about the 50:50 composition. Figure 4 is less symmetrical, but the minimum is rather sharply located at the center composition. Figure 5 is not at all symmetrical, and the minimum composition is shifted to roughly 75:25 DGDA:DYNH.

It is believed that these diagrams for elongation at break are very sensitive indicators of compatibility, interactions, and the morphology they produce. The minima in Figures 3 and 4 may be attributed to the incompatibility of polypropylene and polyethylene. There may be some interaction between these components due to their polyolefin nature. Evidence of this is seen in the fact that the degradation of this property in PE-PP blends is much less severe than what is observed in other systems that do not have such similar structures and hence possibilities for interaction.<sup>1</sup> The possibilities for amorphous-phase interaction or even miscibility is

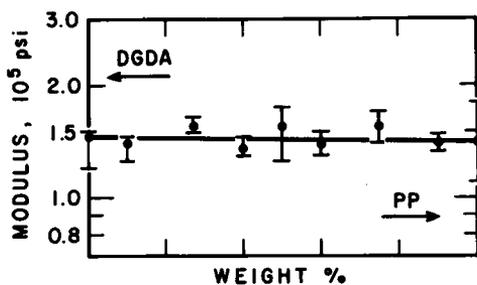


Fig. 6. Modulus of HDPE-PP binary blends.

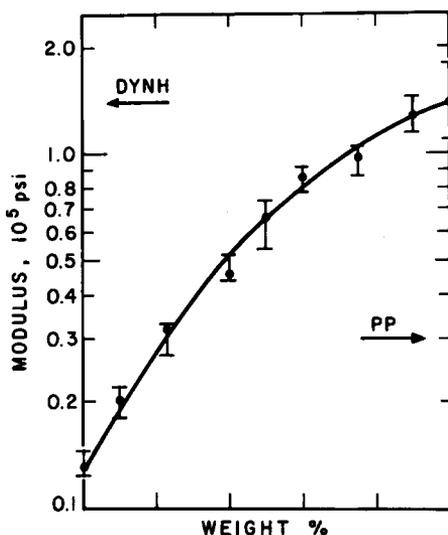


Fig. 7. Modulus of LDPE-PP binary blends.

much greater for the LDPE-HDPE system. These possibilities must be operative in Figure 5, and its unusual nature confirms that this system is not just an ordinary case of incompatibility, as we might say for Figures 3 and 4. Figure 5 shows that blends rich in LDPE can neck and draw even up to the 50:50 composition. One might suggest that this is simply owing to the very ductile nature of LDPE. Figure 4 refutes this simplistic explanation. This figure is skewed to some extent so that blends very rich in LDPE are still fairly ductile; however, the effect is quite minimal compared to Figure 5. We feel that the response for this pair is due to a strong amorphous phase interaction or possibly even miscibility. The slight minimum at high HDPE content is likely a consequence that in this region it is the responsibility of the HDPE to form the continuous phase, and even amorphous phase compatibility cannot prevent some loss of ductility since it is so highly crystalline. The morphology of such blends is very complex.

The initial modulus of these blends varies rather uniformly across Figure 1 between the limits set by the pure components. Figures 6-8 show

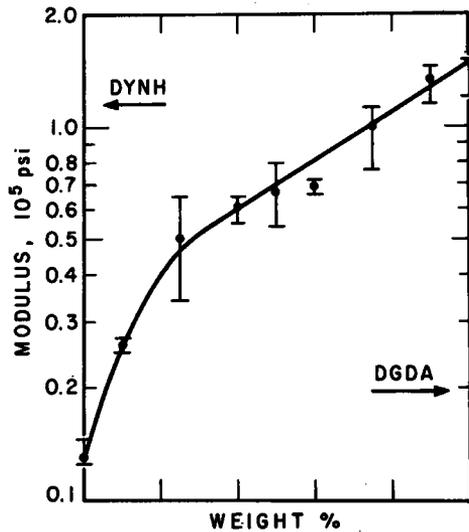


Fig. 8. Modulus of LDPE-HDPE binary blends.

graphic evidence of this along the binary legs. The HDPE and PP have essentially the same modulus, and within experimental error all binary blends of these two have this modulus. The LDPE has a considerably lower modulus, of course, and so the modulus of its binary blends with PP (Fig. 7) and HDPE (Fig. 8) varies with composition. This variation is monotonic with no minimum or maximum, which is to be expected for this property.<sup>1</sup> It is significant, however, that the two curves in Figures 7 and 8 are different in shape even though the values for the pure components are identical in both cases. These shapes are believed to be due to the different interactions between HDPE and PP with LDPE and the resulting difference in blend morphology as discussed above.

Most studies of polymer blends have been concerned with the solid state and virtually all knowledge of compatibility is restricted to this state since most indicators of compatibility involve observations of solid-state transitions. Information on compatibility in the melt state would be of considerable interest and easier to interpret since the complication of crystallization is no longer involved. Melt rheology appears to offer a promising route by which inferences on miscibility can be made, although methods of interpretation are not well worked out. A recent paper by Prest and Porter<sup>46</sup> is one of the most fundamental approaches to this problem available. One method of interpretation which abounds in the literature (see, e.g., ref. 47) states that for compatible melts semilogarithmic plots of viscosity versus blend composition should be linear whereas nonlinearity is an indicator of incompatibility. There is no published justification for this rule, and apparently its use stems from the fact that this mixing law often holds very closely for two polymers of the same chemical structure but different molecular weights. The limited data available

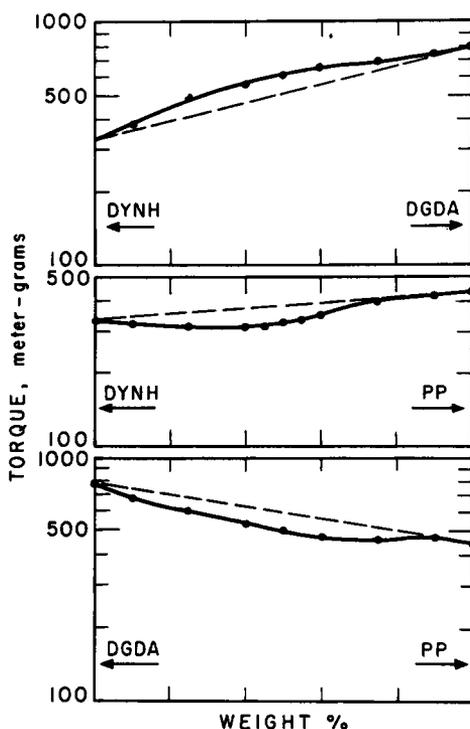


Fig. 9. Steady-state torque required to mix binary polyolefin blends at 190°C.

for blends suggest there may be some value to this rule. Most information available on polyolefin blends conform rather closely to this mixing rule.

During the course of preparing these blends, some related information was generated, viz., the steady-state torque required to turn the Brabender during blend mixing. These torque data along the binary legs are shown in Figure 9. This torque is an indicator of melt viscosity although the relation is somewhat complex.<sup>48,49</sup> In no case here do the blend data simply connect the pure-component endpoints by straight lines, although the departure from such a line (see dotted line in Fig. 9) is not large. A closer interpretation is unwarranted in view of the complex flow situation under which this measurement is made. However, it does appear significant that all departures are *below* the dotted line except for the HDPE-LDPE binary which departs *above*. This, too, is likely a result of the nature of the different interaction between these systems. It might be added that visual inspection revealed all of these blends were rather transparent in the molten state.

### RESULTS FOR EPR-MODIFIED BLENDS

The above results show that, in general, polyolefin melt blends have quite good properties in comparison to melt blends of grossly incompatible polymers.<sup>1</sup> However, there is a loss of toughness in some composition

regions that is undesirable. The use of a blend additive to improve toughness was explored. In related work, such additives have proved very effective if properly selected.<sup>50</sup> Block or graft copolymers have been especially valuable for this purpose since they can provide adhesion at domain interfaces in incompatible systems. For polyolefins, however, there is ample evidence that toughening can be obtained by blending with certain rubbery hydrocarbon polymers without graft or block structures. For example, both HDPE and PP can be impact modified by blending in small amounts of polyisobutylene or rubbery ethylene-propylene copolymers, EPR.<sup>6,7,28,32</sup> There is one report of using EPR to improve the impact strength of HDPE-PP blends.<sup>10</sup>

Based on these leads, it was reasoned that EPR should be an effective blend additive to improve the toughness of the polyolefin blends. Experiments to confirm this were performed using a copolymer containing approximately 43% ethylene and 57% propylene which is commercially available from Enjay and is known as Vistalon 404. A principal objective was to improve the poor elongation at break observed for certain blends, and so the compositions at the minima of Figures 3-5 were selected for these tests. Blends were prepared as before with varying amounts of EPR added and tested mechanically. The LDPE designated as DFD was used in these experiments rather than DYNH.

The response of the elongation to EPR addition at these three compositions is shown in Figure 10. In all cases, the EPR produces some increase above that for the unmodified blend. The effect is most dramatic for the 3-to-1 HDPE-LDPE blend. The elongation is increased from about 30% to about 400% by EPR addition. It is significant that this increase requires only 5% EPR and does not appear to increase with further addition. A similar but less dramatic change is seen for the 1-to-1 LDPE-PP blend. Here, the elongation increases from 10% to 50% with only 5% EPR. The rapid rise of these curves followed by a plateau has an important economic consequence in that the optimum EPR content is of the order of 5%, which is quite low.

The third system, 1-to-1 HDPE-PP blends, are not improved as much by EPR addition as were the other two. The improvement observed is roughly proportional to the amount of EPR added, but the elongation is not even doubled by 20% EPR.

The improvements in elongation at break gained by EPR addition are accompanied by changes in other properties. The yield strength and modulus both decline. Figure 11 shows the effect of % EPR on the strength. The decline is gradual. At 20% EPR, all blends are still stronger than LDPE. At 5% EPR, the loss in this property is not at all severe.

The effect on the modulus is shown in Figures 12-14. It, too, steadily declines with EPR content. This feature may be a liability or an asset depending on the modulus desired. The modulus of the unmodified blends varies over about one decade depending on the composition.

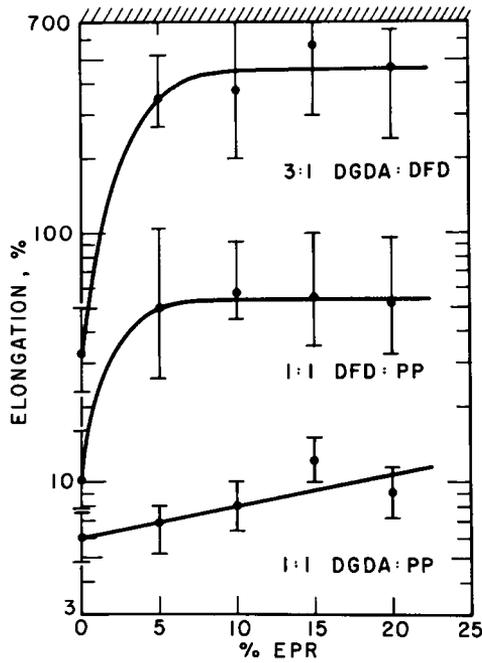


Fig. 10. Effect of ethylene-propylene copolymer addition on elongation at break of binary blends.

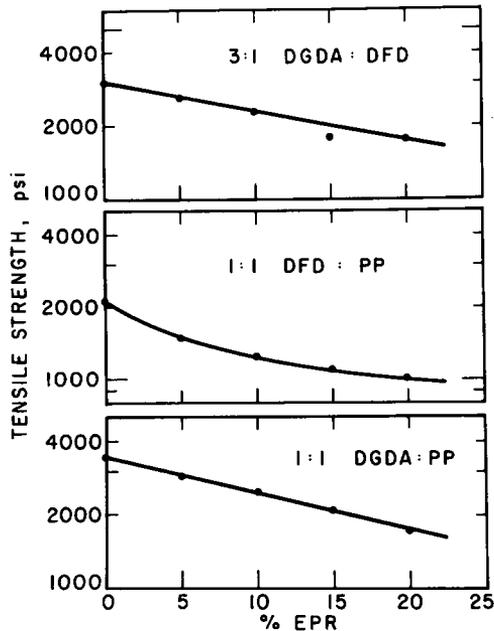


Fig. 11. Effect of ethylene-propylene copolymer addition on yield tensile strength of binary blends.

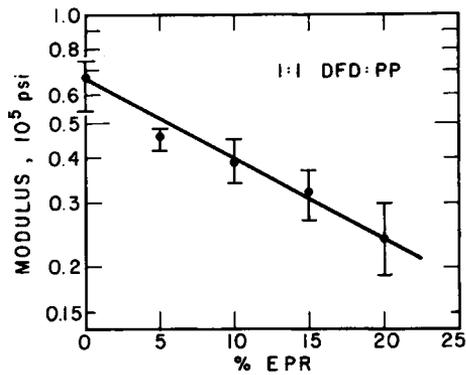


Fig. 12. Effect of EPR addition on the modulus of a low-density polyethylene (DFD)-polypropylene blend.

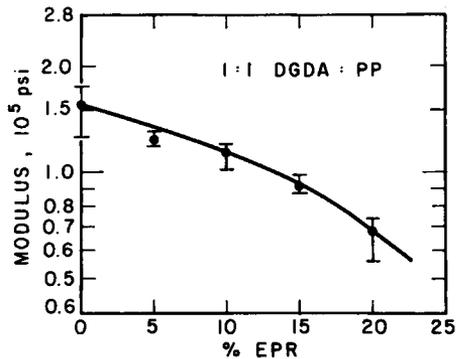


Fig. 13. Effect of EPR addition on the modulus of a HDPE-PP blend.

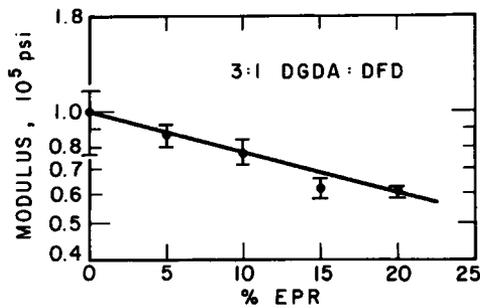


Fig. 14. Effect of EPR addition on the modulus of a HDPE-low-density polyethylene (DFD) blend.

Figure 15 shows a triangular diagram where every point is modified by 20% EPR. This may be compared with Figure 1 to see how EPR addition changes properties across the entire ternary composition spectrum. It is seen that the strength and modulus of the pure components are reduced by EPR, with LDPE being affected least. The elongation at break for the

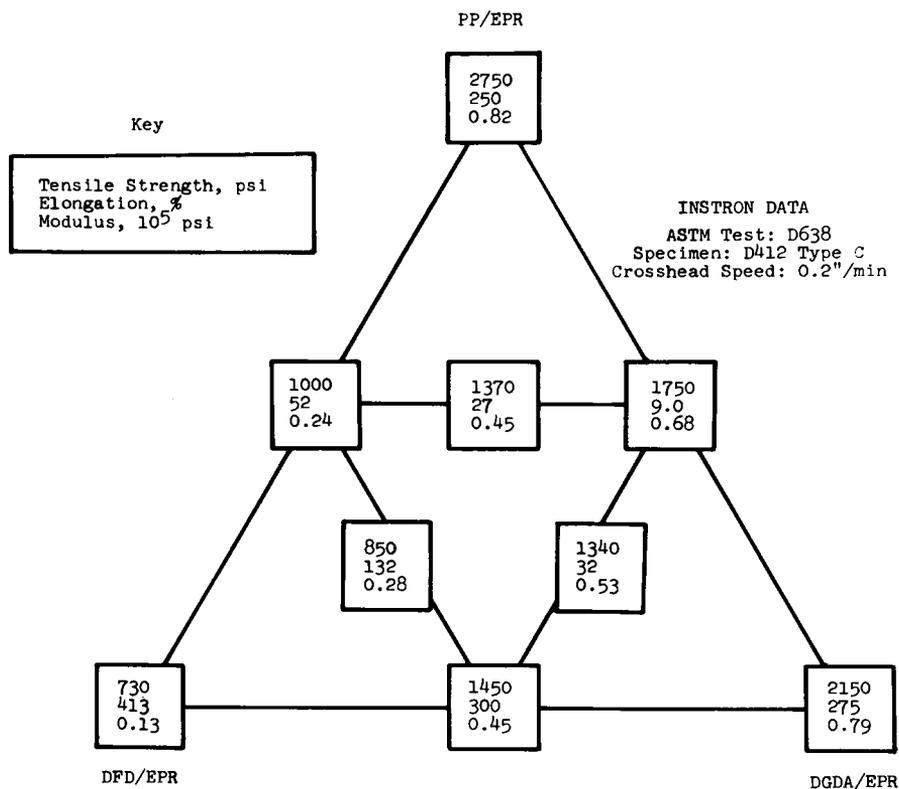


Fig. 15. Mechanical properties of polyolefin blends and pure components modified with 20% EPR.

pure components is also decreased somewhat by EPR addition. All pure components blended with EPR did draw and neck. Other than at the apices, then, EPR addition produces some improvement in elongation across the entire triangular diagram. The improvement along the PP-HDPE binary leg, however, is apparently rather minimal. The effect of EPR addition along the HDPE-LDPE leg is to eliminate the minimum and to raise the elongation to roughly 300% for this entire side of the triangle. From the data available, it appears that the minimum in elongation that occurs at the center of the triangle for the unmodified blends (Fig. 1) is shifted to the midpoint of the PP-HDPE edge.

In summary, EPR addition is very effective for improving elongation within certain regions of the ternary composition diagram. It is not needed if the composition is near any apex and especially the LDPE apex. Its use is least effective in the region near the HDPE-PP edge.

The mechanism by which EPR acts to modify these blends cannot be deduced from these data, and there is very little information in the literature on such effects. An increase in the amorphous content of semicrystalline systems such as polyethylene, polypropylene, and perhaps their blends

may be expected to decrease the yield strength and modulus while increasing the elongation at break. EPR is an amorphous polyolefin, and we might view its addition as simply increasing the amorphous fraction of the sample. However, for this to work there must be a certain level of interaction or compatibility since addition of any rubbery polymers will not have this same effect. In fact, some rubbers make the blend worse rather than better. Another facet to consider is the shape of the upper two curves in Figure 10, which show that the increase in ductility rises rapidly with EPR addition and does not change thereafter. This suggests that a small amount of this material can mend "flaws" in the blend and shows that the function of the EPR is not simply to alter the amorphous fraction of the blend. This concept also requires a great deal of interaction or at least adhesion between the additive and the blend components. All evidence, therefore, points to a strong role of amorphous-phase interactions in such systems.

### SUMMARY

The current data show that the modulus and tensile yield strength of polyolefin blends are nearly additive functions of blend composition and pure-component properties. The elongation at break is a more complex function of composition. This property is always diminished to some degree by blending. In other systems it is common to find that blending results in losses in yield strength plus losses in elongation which are much more severe than that observed here. The superior mechanical behavior of polyolefin blends is attributed to the greater interaction between components possible owing to similarity in chemical structure.

It must be concluded that the loss in toughness that results when many polymers are blended together is not a severe limitation for HDPE-LDPE-PP blends.

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