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Effect of EPDM on the mechanical properties of blends of high and low density polyethylene

Roberto Olayo[1,]*, Angel Manzur, J. Héctor Llamas, Angel Escobar[2]

[1] Departamento de Física, [2] Departamento de Ingeniería de Procesos e Hidráulica,
Universidad Autómoma Metropolitana-Iztapalapa, Apartado Postal 55-534, 09340 México,
D.F., México

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Summary

We studied the effect of adding ethylene-propylene-diene rubber (EPDM) to blends of high (HDPE) and low (LDPE) density polyethylene. The extrusion torque of the blend without EPDM shows a deviation from the linear addition rule, but blends with rubber follow the addition rule. Two composition regions that are compatible with the torque behavior are present in the Young's modulus and extension at break curves. The EPDM content improves the extension at break of LDPE rich blends. This improvement extends to higher compositions of HDPE as the EPDM content is increased.

Introduction

The production of polyethylenes with a wide range of molecular weights and degrees of branching has given polymers with a large variety of properties (1). Different types of polyethylene blends have been studied in order to promote the understanding of their properties and technological usefulness (2). A large amount of work has been performed on binary blends of polyethylenes. Among these studies, some focused on the rheology (3-5), the degree of branching, density and molecular weight (6-9), and using deuterated samples to avoid the chemical resemblance of the components (10). The use of elastomers especially the ethylene propylene diene terpolymer (EPDM) as a third component in the blend has been shown to improve properties (11,12). In this work, the effect of adding EPDM to blends of HDPE/LDPE was studied. The manner in which EPDM changes the mechanical behavior of the blend and the evolution of these changes with the EPDM content is discussed.

Experimental

The polymers used were high density polyethylene (PADMEX 60120, Pemex, México), low density polyethylene (PX, 17070, Pemex, México) and EPDM rubber (Nordel 1040, Dupont). Materials were powdered with a Brabender type mill. Blends were made using powders in the right weight proportion and extruded through a single-screw Brabender extruder with D=19.1 mm and L=20D. The compression ratio of the screw was 3:1; the extrusion conditions were 180 °C in the barrel and die, and an extrusion velocity of 25 rpm. Extrusion torque was measured in the stationary state.

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* Corresponding author

Extrusion gave 0.5-0.7 mm thick sheets. Dumbbells (22 mm long and 7 mm wide in the central part) were submitted to tensile test. Stress-strain (σ-ε) curves were obtained by uniaxial deformation using an Instron tensile tester (model 4502). All the experiments were carried out at room temperature to the rupture point, with an elongation rate of 20 mm/min. The reported values are averages of seven specimens.

Samples were grouped in sets of two types. In the sets of the first type, the EPDM content was held constant while the polyethylene composition was changed. In the set of the second type, the ratio of the two polyethylenes was held constant (1:1) and the percentage of EPDM was changed. Three sets of the first type were made, with constant rubber contents of 0, 5, and 10%. One set of the second type was made with the rubber content varying from 0 to 24%.

Results and Discussion

Figure 1 shows the extrusion torque as a function of HDPE composition for 0, 5 and 10% of EPDM. The straight line between the ends of the curves shows the linear addition rule. In the case of viscosity, deviations from this line can be interpreted as a manifestation of compatibility or incompatibility of the blend. Some authors extrapolate this criterion to the extrusion torque since it should be proportional to the viscosity (13). It also has been established that the shape of the viscosity-composition curve is highly dependent on the shear stress (14) and since this is rather high during extrusion, it is difficult to discuss compatibility based on this assumption. Nevertheless, the presence of deviations will be emphasized since they correlate well with different tendencies of the blends mechanical properties.

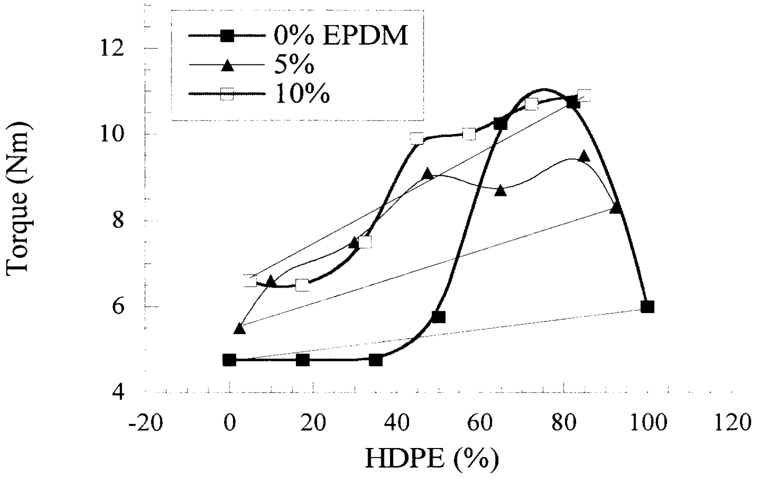

Fig. 1. Extrusion torque of blends as a function of HDPE and EPDM contents.

The curve for the blend of the two polyethylenes shows a large deviation from the linear addition rule at high HDPE contents (Figure 1), starting around 40% HDPE. This may be interpreted as a synergistic interaction and in this region other mechanical properties could be affected. Distinct behavior is observed when 5% of EPDM are added to the blends. The curve is completely different since its deviations are positive for all compositions and smaller than those for the blends without EPDM. Taking into account that the torque is uncertain (~15%), the interaction may have been reduced. With the addition of 10% of EPDM, there are negative and positive deviations. However, taking into account the uncertainty, the points are close to the linear relation, meaning that the synergism has disappeared. From the behavior of the curves, it is possible to conclude that the EPDM has the ability to disperse the components in the blends. This means that it reduces the interaction between the two polyethylenes and in some cases may change the mechanical properties.

The stress-strain (σ-ε) curves of HDPE and LDPE are characteristic of these types of materials. Figure 2 shows the σ-ε curves for blends of these polymers. The behavior goes from a polymer that yields almost with no maximum and does not present strain hardening, to a polymer that nearly does not yield and breaks at small deformations giving a large stress. Figure 2 does not show the breaking point for the high LDPE composition curves (2.5 and 10% HDPE). It is important to note that even for the higher composition that shows a yield point (47.5% of HDPE), after this point it does not show stress hardening. This indicates that even in the zone where the torque shows positive deviations from the addition rule, the interaction is not large enough to produce entanglements and generate stress hardening. This behavior implies that the HDPE domains are separated and that it is in these domains where failure takes place. The σ-ε curves for blends containing EPDM exhibit the same tendency but the strains at break are different.

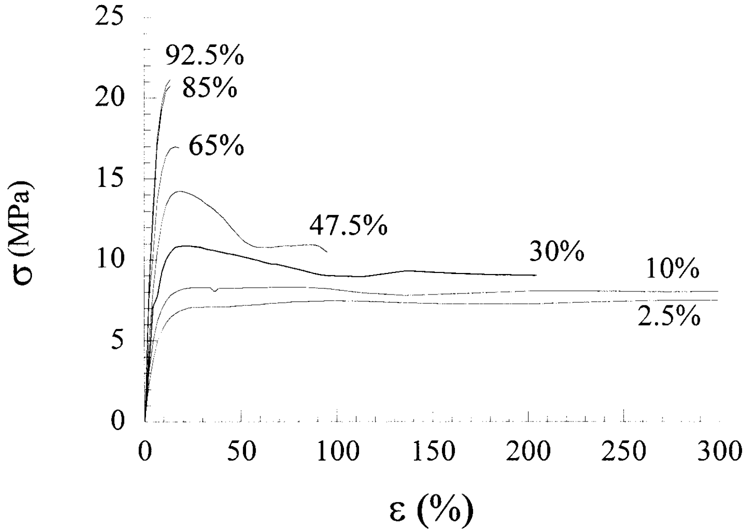

Fig. 2. Stress-strain curves for HDPE/LDPE blends at different HDPE contents

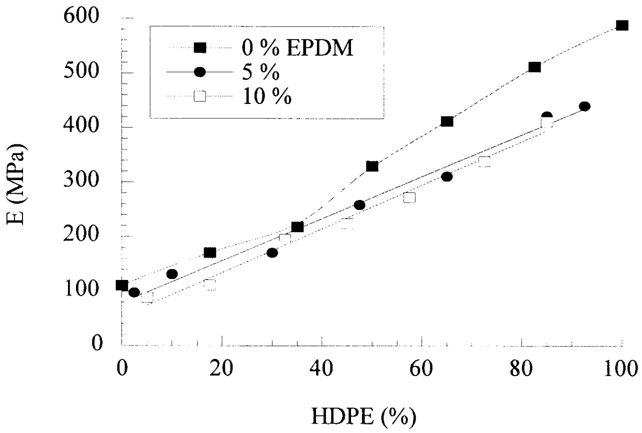

Fig. 3. Young's modulus as a function of HDPE content for blends with different EPDM content.

One of the most significant initial properties is the Young's modulus (E). Figure 3 shows the modulus as a function of HDPE composition. The blends with constant EPDM contents are displayed. Without rubber in the blend, the behavior can be described by two straight lines of different slopes whose interception at approximately 40% defines the limit of two regions. This same limit was established in the torque curve. The torque suggested a region with interactions between the phases; in this region, the slope shows a large increase of the modulus with HDPE content. However, the sets with 5 and 10% of EPDM behave almost as a linear function of the HDPE contents. For these two sets, the two regions observed become only one, giving the same slope for all compositions. The moduli are also smaller than those of blends without EPDM, but the difference is not large, especially below ~40% HDPE. In a similar way as with torque, the rubber disperses the polyethylenes and decreases the interaction between the PE domains.

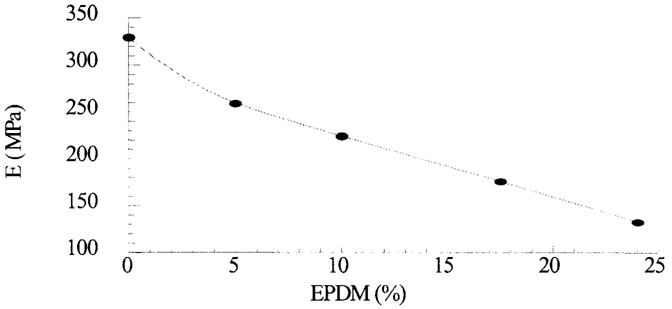

Fig. 4. Young's modulus as a function of EPDM content for blends with HDPE/LDPE = 1:1.

The Young's modulus as a function of EPDM content is shown in Figure 4 for blends with equal proportions of the two polyethylenes where it was observed that the modulus decreases as the percentage of EPDM increases. The behavior is linear except for the first point since the blend without EPDM is in the second region where interaction exists between the two phases. Once the rubber is present, the interaction disappears. The first point therefore, has a grater value than that predicted by the linear fitting of the points with rubber.

The effect of EPDM on the strain at break (ε_b) is illustrated in Figure 5. The plot of ε_b for the blends without EPDM changes significantly beyond 20% HDPE. From 35 to 100% HDPE the data follow a straight line with a small slope, indicating a gradual decrease. This behavior is dominated by the presence of HDPE and again, the behavior in the range from 40 to 100% is different. The shape of the curve is completely different with only 5% of EPDM, where for low HDPE contents the rubber improves the property by almost 20%. The sample with 2.5% of HDPE has a higher ε_b value than that for the blend without rubber. From 10% to 100% HDPE, the ε_b stays below that for the blend without rubber. The behavior of the curve for the set with 10% EPDM is similar to the one without EPDM, but the ε_b is larger for compositions below 20% HDPE, and after this point it behaves like the 5% EPDM curve. The rubber improves the ε_b at low HDPE contents and as the amount of EPDM increases the improvement moves to higher HDPE contents. This improvement has been reported for polypropylene-HDPE blends containing EPDM (16). It was interpreted as an indication that the EPDM increases the compatibility between the amorphous phases. In the present case the effect is similar, the rubbery phase reduces the interaction between the polyethylenes, and helps to release high stress concentrations that appear at large strains. The EPDM may also affect the crystal growth of the polymer with

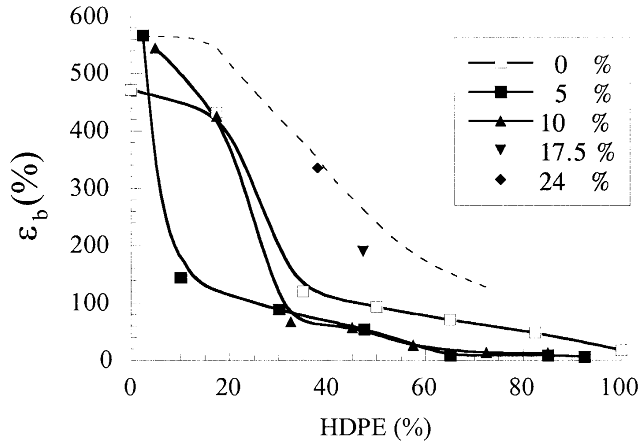

Fig. 5. Strain at break as a function of HDPE content for blends with different EPDM content. The broken line is discussed in the text.

higher crystallinity (HDPE). Therefore, as the EPDM content increases the ε_b increases and the Young's modulus decreases (17).

Figure 5 also shows the change in the strain at break as a function of the EPDM content for equal amounts of both polyethylenes. The onset of improvement may be observed beyond 10% EPDM, from this point the ε_b is higher than that of the blend without EPDM. The samples with 5 and 10% of EPDM have lower values than the 0% blend, but above 10% the ε_b increases. As it was discussed above, the content of EPDM improves the ε_b, but this improvement only affects blends with a low amount of HDPE. Of the blends containing 5% EPDM, only that with 2.5% HDPE shows an improvement relative to the 0% EPDM curve, but as the amount of EPDM increases, blends with higher HDPE contents exhibit improvement. Thus, for the 10% EPDM blends the threshold composition is 20% HDPE, which means that all blends with compositions below 20% HDPE will have larger ε_b than the corresponding blend without EPDM. Figure 5 shows the ε_b for 17.5% and 24% EPDM with equal amounts of polyethylenes. Both values are larger than that of the blend without EPDM. Therefore they should have a curve that keeps the larger ε_b to a composition higher than 50% HDPE. This behavior is hypothetically illustrated in Figure 5 by the thin broken line.

Conclusions

The extrusion torque for HDPE/LDPE blends shows two regions, one of which is a synergistic range between 40 and 100% HDPE. The mechanical properties show differences in the same regions. The addition of EPDM homogenizes the blends and disperses the polyethylenes, giving a more homogeneous variation on the extrusion torque and Young's modulus. The strain at break shows an evolution of its behavior as the content of EPDM increases, changing the threshold of HDPE content where the ε_b is larger than that for the blends without EPDM.

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