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Brittle–tough transition in PP/EPDM blends: effects of interparticle distance and tensile deformation speed

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Abstract

The toughness of polypropylene (PP)/ethylene–propylene–diene monomer rubber (EPDM) blends containing various EPDM contents as a function of the tensile speed was studied. The toughness of the blends was determined from the tensile fracture energy of the side-edge notched samples. A sharp brittle–tough transition was observed in the fracture energy versus interparticle distance (ID) curves when the crosshead speed <102.4 mm/min. It was observed that the brittle–ductile transition of PP/EPDM blend occurred either by reducing ID or by decreasing the tensile speed. The correlation between the critical interparticle distance and tensile deformation rate was compared with that between the critical interparticle distance and temperature for PP/EPDM blends. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: PP/EPDM blend; Toughness; Brittle–tough transition

1. Introduction

Toughness is a key parameter that influences the performance of polymers in various engineering applications. It is generally known that several polymers tend to become brittle under severe condition of low temperature or high tensile speed deformation. Consequently, they can undergo a sharp transition from ductile to brittle mode. The brittle–ductile transition of polymers has been the subject of interest for the polymer scientists. The effect of temperature or deformation speed on the brittle–tough transition (BTT) of the polymers can be seen clearly from the schematic diagram as shown in Fig. 1. The regions 1, 2 and 3 in this diagram correspond to brittle, transition and tough regimes, respectively. The parameter X denotes the temperature, deformation speed an so on. X_c is a critical value of X at which BTT occurs. For the polymer blends, Wu [1] have proposed that the interparticle distance (ID, also called matrix ligament thickness) is a key factor that dominates the brittle–tough transition of polymer blend. If ID is smaller than a critical value ($\mathrm{ID_c}$), the blend is tough, otherwise the blend is brittle. Several workers have used this concept to study the toughening behavior of polymers [2–23]. Generally, it is found that the $\mathrm{ID_c}$ not only depends on the matrix materials [6–8], but also depends on the properties of the dispersed phase [9–11]. In addition, Gaymans [23] et al. found that $\mathrm{ID_c}$ is also temperature dependent for the nylon/ethylene–propylene–diene monomer (EPDM) rubber blends. Recently, similar results were reported by Jiang et al. [24] for the polypropylene (PP)/EPDM blends. Moreover, it was also found that the curves for different blend systems reduced down to a single master curve if the $\mathrm{ID_c}$ was plotted versus $T_{\mathrm{BT}}^{\mathrm{m}} - T$, where $T_{\mathrm{BT}}^{\mathrm{m}}$ is defined as the brittle–tough transition temperature of the matrix polymer [24]. However, the correlation between the $\mathrm{ID_c}$ and tensile speed has not been studied in the past few years. In this article, we investigate the toughening behavior of PP/EPDM blends with a wide ranges of composition and tensile speed by means of an Instron tensile tester. The aim is to study the effects of interparticle distance and tensile speed on the brittle–tough transition of PP/EPDM blends.

2. Experimental

2.1. Materials

The PP was a commercial product of Himont (Profax 6331) with a melt flow index of 12 g/10 min. EPDM

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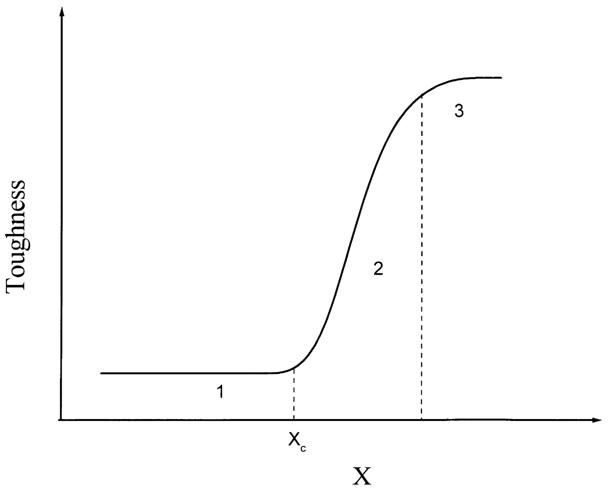

Fig. 1. Schematic diagram showing the brittle–tough transition of a polymer.

(4045) used was purchased from Mitsui Petrochemical Industries Ltd., Japan.

2.2. Sample preparation

PP blends containing various EPDM contents were prepared in a twin-screw Brabender Plasticorder at 200°C and 45 rpm. The extrudates exiting from Brabender were cut into pellets by a pelletizer. The plaques with dimensions of 200 × 80 × 3.2 mm were injection molded. The barrel zone temperatures of injection molder were set at 200°C for all the blend specimens. These plaques were cut into rectangular bars with dimensions of 80 × 13 × 3.2 mm. A single-edge notch (SEN) was cut in these bars. The radius of curvature for the V-shaped notch was 0.025 mm.

2.3. SEN tensile test

Single-edge notch (SEN) tensile specimens were used to study the fracture behavior of PP/EPDM blends [25–28]. The tests were carried out using an Instron tester (model 4206) at 22°C, and at various crosshead speeds. The tensile fracture energy was used to characterize the toughness of the blends. The grip distance remained 52.0 mm for all samples. The initial strain rates were corresponding to the constant crosshead speeds.

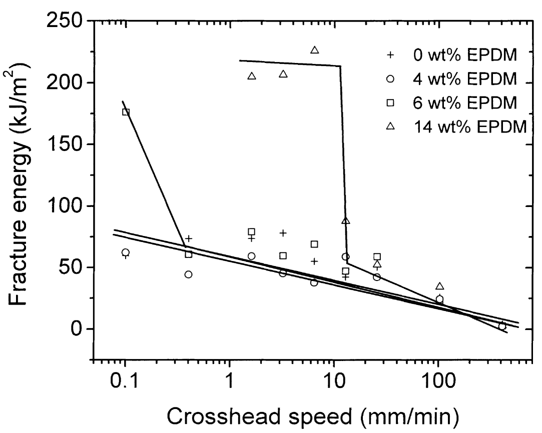

Fig. 2. Variation of fracture energy with tensile speed for PP/EPDM blends.

2.4. EPDM rubber particle size analysis

Specimens for particle size analysis were initially cryo-fractured in liquid nitrogen. The morphologies of the surfaces were observed in a scanning electron microscope (SEM, Jeol JSM 820). All the specimens were coated with a thin layer of gold prior to SEM observations. The weight average diameter d_w of rubber particles was defined as [23,24]

$$d_{\mathrm{w}} = \frac{\sum_i n_i d_i^2}{\sum_i n_i d_i}. \tag{1}$$

3. Results and discussion

Fig. 2 shows the variation of the fracture energy with crosshead speed for the PP/EPDM blends. For pure PP specimen, the fracture energy decreases linearly with increasing the crosshead speed, and there is no brittle–tough transition in the crosshead speed regime from 0.1 to 409.6 mm/min. However, BTT takes place in PP/EPDM blends containing rubber particles >4 wt%, and the fracture energy drops sharply with increasing tensile speed. The higher the EPDM content, the higher the tensile speed at which BTT occurs is.

In order to study the effect of interparticle distance (ID) on BTT, it is necessary to determine the ID values of PP/EPDM blends. ID can be determined from [5,23]

$$\mathrm{ID} = d\left[\left(\frac{k\pi}{6V_{\mathrm{r}}}\right)^{1/3} - 1\right] \tag{2}$$

where V_r is the rubber volume fraction, $k = 1$ for cubic packing, d is the rubber particle diameter. The volume fraction V_r of the EPDM can be given by

$$V_{\mathrm{r}} = \frac{W_{\mathrm{f}}\rho_{\mathrm{m}}}{W_{\mathrm{f}}\rho_{\mathrm{m}} + (1 - W_{\mathrm{f}})\rho_{\mathrm{f}}} \tag{3}$$

where W_f, ρ_f and ρ_m refer to the weight fraction of the EPDM rubber, density of the EPDM and density of the matrix, respectively. For PP/EPDM blends, $\rho_f = 0.85\ (\mathrm{g/cm^3})$, $\rho_m = 0.90\ (\mathrm{g/cm^3})$. On the basis of SEM micrographs and Eqs. (1)–(3), the average diameter of rubber particles, volume fraction V_r of the EPDM, and ID value for PP/EPDM blends are shown in Table 1.

The fracture energy versus ID at different tensile deformation speeds is shown in Fig. 3a–b. The results indicate that the transition from notch brittle to notch tough behavior in PP/EPDM blends can be achieved either by decreasing ID or by decreasing tensile speed, i.e. decreasing tensile speed

Table 1
Average EPDM rubber particle diameter and ID value of PP/EPDM blends

No.	Weight fraction of EPDM (%)	Volume fraction of EPDM (%)	Average EPDM particle diameter (μm)	ID value (μm)
1	0	0	/	∞
2	1.0	1.1	0.87	2.3
3	2.0	2.1	0.87	1.7
4	4.0	4.2	0.89	1.2
5	6.0	6.3	0.87	0.89
6	8.0	8.4	0.87	0.73
7	10.0	10.5	0.87	0.62
8	12.0	12.6	0.88	0.53
9	14.0	14.7	0.88	0.46
10	16.0	16.8	0.89	0.41
11	20.0	20.9	0.88	0.32
12	24.0	25.1	0.88	0.24

and decreasing interparticle distance (ID) have equivalent effect on the brittle–trough transition of PP/EPDM blends.

Fig. 4 shows the plot of the critical interparticle distance (ID_c) versus tensile speed for PP/EPDM blends. Apparently, ID_c decreases continuously with decreasing tensile speed. To the best of our knowledge, this is the first report in the literature showing the correlation between the critical interparticle distance and tensile speed. For the purpose of comparison, the experimental data of Jiang et al. [24] showing the effect of temperature on the BTT of PP/EPDM blends is also plotted in this figure. It can be seen that ID_c increases considerably with temperature, especially near to the brittle–tough transition temperature of PP polymer (T_{BT}^{m}). Finally, Fig. 4 also implies that decreasing the deformation rate and increasing temperature have a similar effects on the brittle–tough transition of polymer blends.

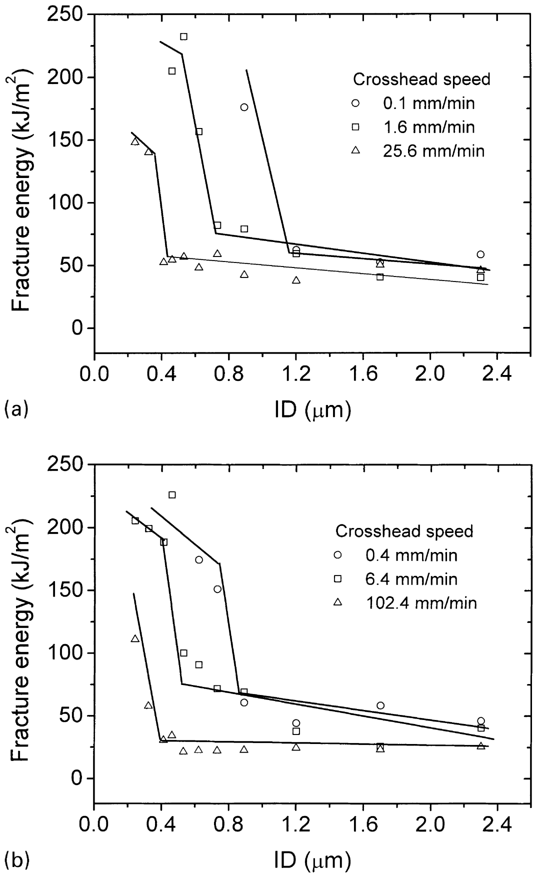

Fig. 3. Variation of fracture energy with ID for PP/EPDM blends.

4. Conclusions

The effects of interparticle distance ID_c and tensile speed on the brittle–ductile transition of PP/EPDM blends containing various EPDM contents were studied. It was found that decreasing tensile speed and the interparticle distance (ID) had a similar effect on the brittle–trough transition (BTT) of PP/EPDM blends. The brittle–ductile transition of the PP/EPDM blends occurred either by reducing ID or by decreasing the tensile speed. The correlation between the tensile speed and critical interparticle distance

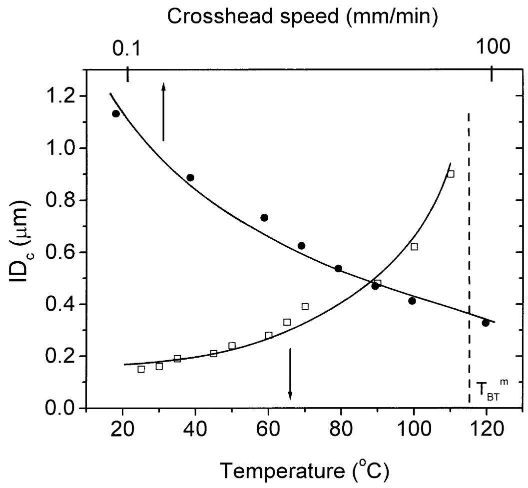

Fig. 4. Variation of critical interparticle distance (ID_c) with tensile speed for PP/EPDM blends. The critical interparticle distance (ID_c) versus temperature [24] is also shown for the purposes of comparison.

was compared with that between the temperature and critical interparticle distance for PP/EPDM blends.

References

[1] Wu S. Polymer 1985;26:1855.
[2] Margolina A, Wu S. Polymer 1988;29:2170.
[3] Wu S. Polymer 1990;31:972.
[4] Li Q, Zheng W, Qi Z, Zhu X, Choy CL. Science in China B 1992;22:236.
[5] Jiang W, Liang HJ, Jiang BZ. Polymer 1998;39:4437.
[6] Wu S. J Appl Polym Sci 1988;35:549.
[7] Dijkstra K, Gaymans RJ. Polymer 1994;35:332.
[8] Gaymans RJ, Borggreve RJM, Spoelstra AB. J Appl Polym Sci 1989;37:479.
[9] Jiang W, Wang ZG, Liu CH, Liang HJ, Jiang BZ, Wang XH, Zhang HX. Polymer 1997;38:4275.
[10] Borggreve RJM, Gaymans RJ, Schuijer J. Polymer 1989;30:71.
[11] van der Sanden MCM, de Kok JMM, Meijer HEH. Polymer 1994;35:2995.
[12] Liu ZH, Zhang XD, Zhu XG, Li RKY, Qi ZN, Wang FS, Choy CL. Polymer 1998;39:5035.
[13] Liu ZH, Zhu XG, Li Q, Qi ZN, Wang FS. Polymer 1998;39:1863.
[14] Bartczak Z, Argon AS, Cohen RE, Weinberg M. Polymer 1999;40:2331.
[15] Borggreve RJM, Gaymans RJ. Polymer 1989;30:63.
[16] Jiang W, Liang H, Jiang BZ, et al. J Appl Polym Sci 1995;58:537.
[17] Wang XH, Zhang HX, Jiang W, Wang ZG, Liu CH, Liang HJ, Jiang BZ. Polymer 1998;39:2697.
[18] Bartczak Z, Argon AS, Cohen RE, Weinberg M. Polymer 1999;40:2347.
[19] Liu ZH, Li RKY, Tjong SC, Qi ZN, Wang FS, Choy CL. Polymer 1998;39:4433.
[20] Liu ZH, Zhang XD, Zhu XG, Li RKY, Qi ZN, Wang FS, Choy CL. Polymer 1998;39:5019.
[21] Liang JZ, Li RKY. Polymer 1999;40:3191.
[22] Fu Q, Wang G. J Appl Polym Sci 1993;49:1985.
[23] Borggreve RJM, Gaymans RJ, Schuijer J, Housz JI. Polymer 1987;28:1489.
[24] Jiang W, Liu CH, Wang ZG, An LJ, Liang HJ, Jiang BZ, Wang XH, Zhang HX. Polymer 1998;39:3285.
[25] van der Wal A, Mulder JJ, Thijs HA, Gaymans RJ. Polymer 1998;39:5467.
[26] van der Wal A, Mulder JJ, Gaymans RJ. Polymer 1998;39:5477.
[27] Dijkstra K, ter Laak J, Gaymans RJ. Polymer 1994;35:315.
[28] van der Wal A, Gaymans RJ. Polymer 1999;40:6045.