

## Brittle–tough transition in PP/EPDM blends: effects of interparticle distance and temperature

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The toughness of PP/EPDM blends was measured over a wide range of temperature (25–132°C) and composition (0–26 wt% EPDM). It was found that increasing temperature and decreasing interparticle distance have equivalent effects on the brittle–tough transition of toughening PP with EPDM, and the shift factor increases with increasing temperature. A correlation was also found between temperature and critical interparticle distance. When critical interparticle distance was plotted *versus*  $T_g - T$ , where  $T_g$  is defined as the brittle–tough transition temperature of the matrix itself, the curves for different blend systems converge to a single master curve. © 1998 Elsevier Science Ltd. All rights reserved.

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### Introduction

Figure 1 shows a typical brittle–tough transition (BTT) in thermoplastic, in which regions 1, 2 and 3 refer to brittle, brittle–tough transition and tough regions. Regions 1 and 3 correspond to matrix crazing and matrix shear yielding, respectively. The variable  $X$  may be temperature, rubber content, strain rate and so on, and  $X_c$  corresponds to temperature, rubber content, strain rate and so on at BTT, defined as brittle–tough transition of the blends. Great effort has been devoted to study the correlation among these factors at BTT<sup>1–6</sup>. For example, Borggreve *et al.*<sup>2</sup> found a correlation between the brittle–tough temperature and the interparticle distance for nylon/rubber blends. But there is little experimental work in which the equivalent effects of temperature and interparticle distance (ID) on the brittle–tough transition and the shift factor are studied.

### Experimental

**Materials and specimen preparation.** Polypropylene (PP) used in this paper was a commercial polymer PP5004 and was manufactured by Liaoyang Petrochemical Industries Ltd, China. The elastomer was EPDM4045, which was also a commercial polymer and was manufactured by Mitsui Petrochemical Industries Ltd, Japan.

The PP/EPDM blends with different rubber contents were mixed in a Brabender-like apparatus (Rheocoder XSS-300, made in Shanghai, China) at 200°C for 4 min at roller speed of 40 rpm. The temperature, mixing time and roller speed remain unchanged in the experiment for all the blends.

**Notched impact tests.** The samples for impact testing were obtained by compression moulding the PP/EPDM blends at 200°C, then cutting them into rectangular specimens which were sharply notched with a fresh razor blade. The size of the rectangular specimens was 63.5 mm × 12.7 mm ×

3.2 mm. The notched Izod impact strength of PP/EPDM blends with different rubber content were measured by XJ-40A Izod impact tester (made in Wuzhong, China) at different temperatures.

**Scanning electron microscopy.** The test specimens were first cold fractured in liquid nitrogen, then coated with a thin layer of gold palladium alloy to avoid charging under electron beam. The average particle diameter was obtained by scanning electron microscope (SEM) (model Japan JXA-840). In this study the so-called weight average diameter is used:

$$d_w = \sum_i n_i d_i^2 / \sum_i n_i d_i \quad (1)$$

### Results and discussion

Figure 2 illustrates the influence of rubber concentration on the notched impact strength of PP/EPDM blends over a wide range of temperatures. The notched impact strength of pure PP increases dramatically when the temperature exceeds its brittle–tough transition temperature  $T_g = 115^\circ\text{C}$ . Here we would like to note that this  $T_g$  is different from the glass transition temperature of the matrix, so we call it the brittle–tough transition temperature of the matrix itself.

For the blends with different rubber content, the average diameters are nearly constant ( $d = 0.47 \mu\text{m}$ ). This is in agreement with Borggreve's<sup>2</sup> and Tang's<sup>7</sup> results showing that coalescence during blending of two polymer melts is prevented when there is sufficient interfacial adhesion between the dispersion and the matrix phase. The values of ID can be calculated by Wu's equation<sup>8</sup>:

$$\text{ID} = d \left[ \left( \frac{k\pi}{6V_r} \right)^{\frac{1}{3}} - 1 \right] \quad (2)$$

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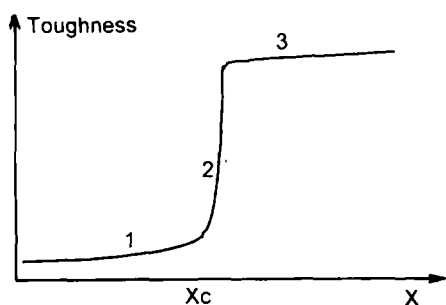


Figure 1 Schematics for brittle-tough transition of a thermoplastic

where  $d$  is the rubber particle diameter,  $V_r$  is the rubber volume fraction, and  $k = 1$  for the cubic lattice.

The notched Izod impact strength versus ID at different temperatures is shown in Figure 3, in which it is clear that the distance between two isotherm curves with same temperature difference ( $T = 20$  K), namely, the shift factor, increases with increasing temperature, especially at higher

temperature regions (close to  $T_g$ ). These results show that increasing temperature and decreasing interparticle distance have equivalent effects on the brittle-tough transition of toughening PP with EPDM, and the shift factor increases with increasing temperature.

Figure 4 shows the critical interparticle distance ( $ID_c$ ) versus temperature in PP/EPDM blends. These results are similar to that of Borggreve *et al.*<sup>2</sup> for nylon/EPDM blends, while the values of  $ID_c$  at same temperature are much different, although both of the dispersed elastomers are EPDM rubber for these two blend systems. For nylon/EPDM blends  $ID_c = 0.30 \mu\text{m}$  at  $25^\circ\text{C}$ , but  $ID_c = 0.15 \mu\text{m}$  at  $25^\circ\text{C}$  for PP/EPDM blends. This difference must result from the properties of the polymer matrix because Wu<sup>8</sup> and Borggreve *et al.*<sup>2</sup> have pointed out that a van der Waals adhesion between the two phase is enough for the toughening effect and the adhesion has no influence on BTT. From equation (2), it is known that as the rubber volume fraction moves toward zero, namely, for pure matrix without rubber phase, ID approaches infinity. That is to say, when temperature is increased to  $T_g$  of the matrix,  $ID_c$  will

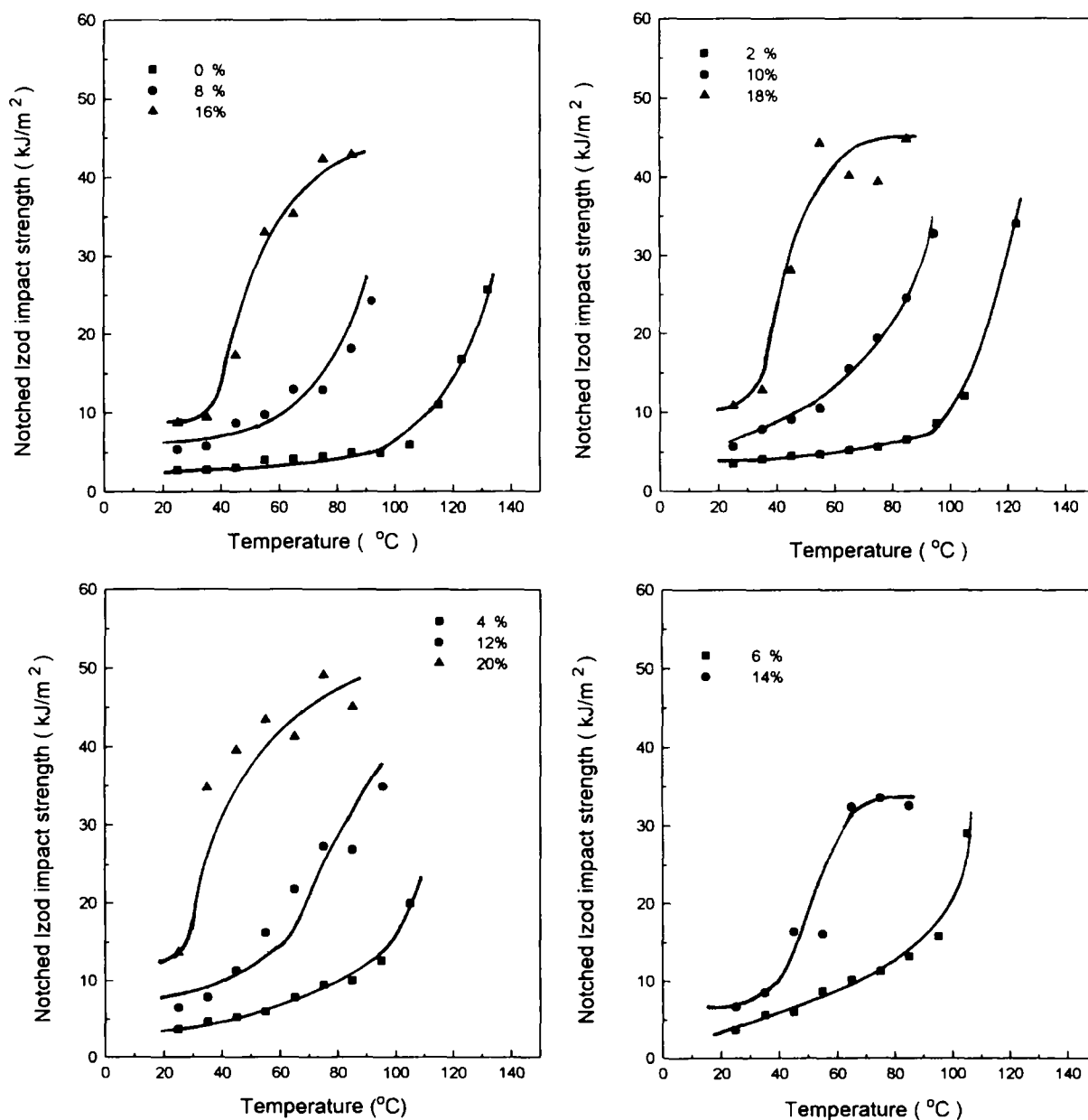


Figure 2 Notched Izod impact strengths versus temperature for different rubber content

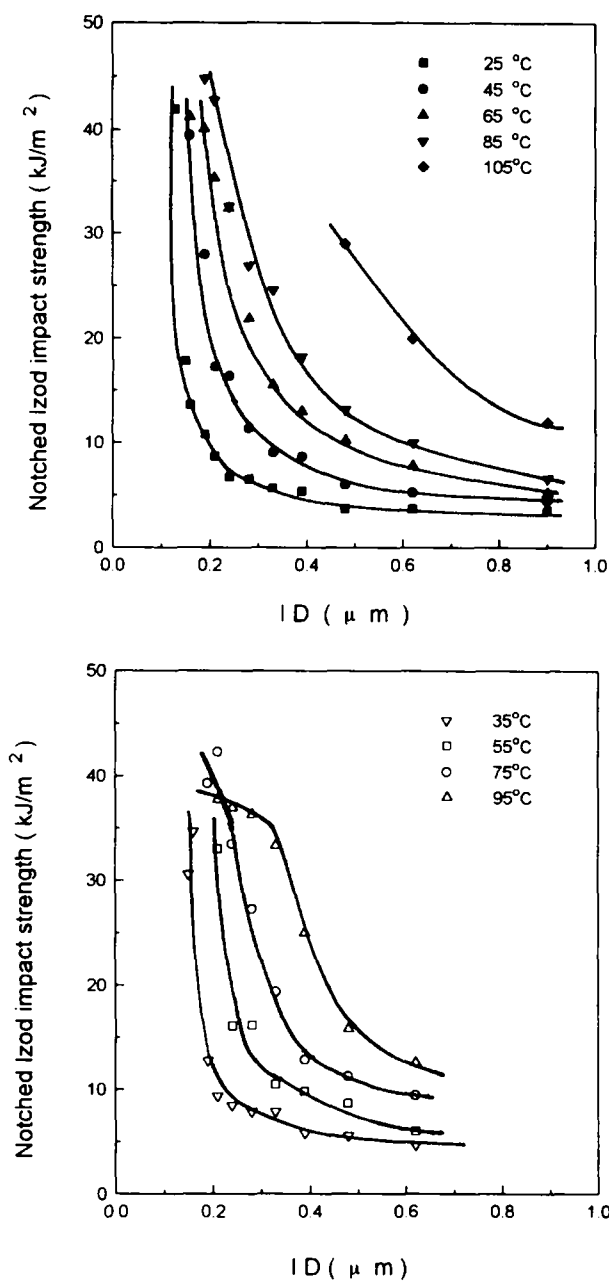


Figure 3 Notched Izod impact strengths versus ID for different temperature

absolutely approach infinity, so in the temperature range close to  $T_g$ ,  $ID_c$  increases rapidly with increasing temperature. Most interestingly, the two curves of  $ID_c$  versus temperature for nylon/EPDM blends<sup>2</sup> and PP/EPDM blends converge to a single master curve when plotted versus  $T_g - T$ , which is shown in Figure 5. Maybe this is the master curve of  $ID_c$  for toughening crystalline thermoplastic with EPDM using the same test method.

#### Conclusions

The notched Izod impact strength of PP can be improved by increasing temperature or adding EPDM rubber, namely, by decreasing interparticle distance if the particle size remains unchanged. Increasing temperature and decreasing interparticle distance have equivalent effects on the brittle-tough transition of toughening PP with EPDM, and the shift

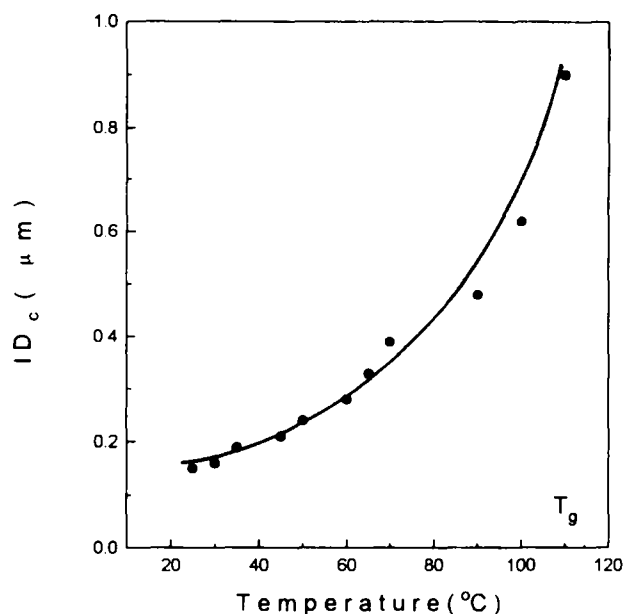


Figure 4 Critical interparticle distance  $ID_c$  versus temperature

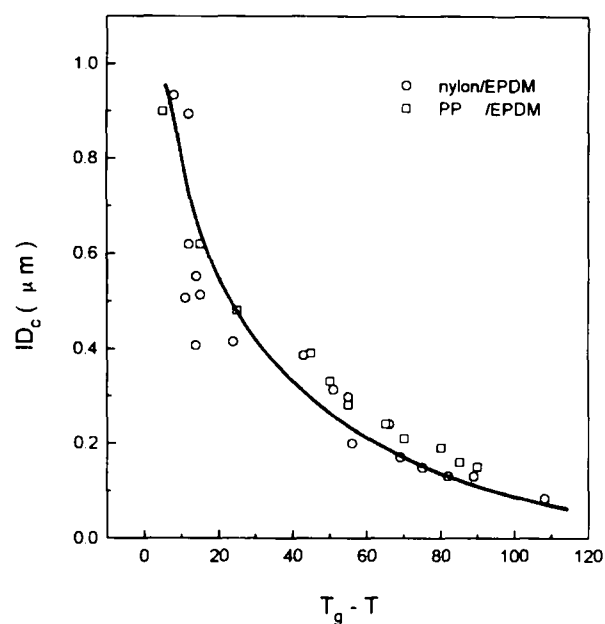


Figure 5 Critical interparticle distance  $ID_c$  versus  $T_g - T$  for nylon/EPDM and PP/EPDM blends, in which  $T_g = 74^\circ\text{C}$  for nylon and  $T_g = 115^\circ\text{C}$  for PP

factor increases with increasing temperature. Critical interparticle distance increases with increasing temperature and increases dramatically when the temperature is close to  $T_g$ . The curve of  $ID_c$  versus  $T_g - T$  is independent of the matrix material, which suggests that there is a single master curve of  $ID_c$  for toughening crystalline thermoplastic with EPDM using the same test method.

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