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Brittle–ductile transition in PP/EPDM blends: effect of notch radius

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Abstract

The toughness of polypropylene (PP)/ethylene–propylene–diene monomer (EPDM) blends was studied over wide ranges of EPDM content and temperature. In order to study the effect of notch radius (R), the toughness of the samples with different notch radii was determined from Izod impact test. The results showed that both toughness and brittle–ductile transition (BDT) of the blends were a function of R, respectively. At test temperatures, the toughness tended to decrease with increasing $1/R$ for various PP/EPDM blends. Moreover, the brittle–ductile transition temperature (T_{BT}) increased with increasing $1/R$, whereas the critical interparticle distance (ID_c) reduced with increasing $1/R$. Finally, it was found that the different curves of ID_c versus test temperature (T) for different notches reduced down to a master curve if plotting ID_c versus $T_{BT}^m - T$, where T_{BT}^m was the T_{BT} of PP itself for a given notch, indicating that $T_{BT}^m - T$ was a more universal parameter that determined the BDT of polymers. This conclusion was well in agreement with the theoretical prediction.

Keywords: Polypropylene/EPDM blend; Notch radius; Brittle-tough transition

1. Introduction

Since 1985 Wu [1] first proposed the interparticle distance (ID) model for nylon/ethylene–propylene–diene monomer (EPDM) blends, the brittle–ductile transition (BDT) of polymers has been extensively studied from this model [2–20]. Initially, it is found that the critical interparticle distance (ID_c) is a function of temperature for nylon/EPDM blends. The ID_c increases nonlinearly with increasing temperature [2]. Similar results were observed in polypropylene (PP)/EPDM blends [3]. In addition, it is reported that the two curves of ID_c versus test temperature (T) for nylon/EPDM and PP/EPDM blends are obviously separated each other. However, these two curves reduce down a master curve if plotting ID_c versus $T_{BT}^m - T$, where T_{BT}^m is the brittle–ductile transition temperature (T_{BT}) of the matrix polymer itself [3]. On the other hand, the experimental results from Meijer and Gaymans' research groups [4–5] show that ID_c decreases with increasing the modulus of dispersed elastomer particle for nylon blend system and polycarbonate (PC) blend system, respectively. Furthermore, Gaymans' results [6] indicate that ID_c drops with decreasing the molecular weight of nylon for nylon/rubber blends. With regard to the effect of strain rate, Gaymans's and Jiang's [7–8] results suggested that ID_c tends to reduce nonlinearly with increasing strain rate. Recently, the ID model was successfully applied to the polymer/rigid particle blends. The results show that ID is still a key parameter that determines the BDT of polymer/rigid particle blends [9–13], and ID_c increases nonlinearly with increasing temperature [9].

On the other hand, it is generally known that the toughness for some polymers such as nylon, PP, PC is notch sensitivity. The notched toughness of these polymer samples is much lower than that of unnotched samples. The effect of notch radius (R) on the toughness of the polymers and their blends was widely studied by Fraser, Pitman and Ward [14–15], Dekkers and Hobbs [16], Kinloch, Shaw and Hunston [17], Havriliak, Cruz and Slavin [18]. Comparing with these, the effect of R on the

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BDT of polymers and their blends has been seldom studied. More recently, Inberg and Gaymans [19] reported the effect of R on the toughness of PC/acrylonitrile–butadiene–styrene (ABS) blends. It is found that the T_{BT} of both PC and PC/ABS blend tend to decrease with increasing R from 0.004 to 1 mm.

The PP/EPDM blends samples with various notches are employed in this study. The toughness of the blends was studied over wide ranges of EPDM content, temperature and R. The purpose is to study the effect of R on the BDT of the blends, and to reveal the correlation between ID_c and R, as well as the correlation between T_{BT} and R for PP/EPDM blends.

2. Experimental

2.1. Materials

The PP (type 5004) was a commercial product of Liaoyang Petrochemical Fiber Ltd, China. EPDM (4045) was purchased from Mitsui Petrochemical Industries Ltd, Japan.

2.2. Sample preparation

PP blends containing various EPDM contents were prepared in a Brabender-like apparatus (Rheocoder XSS-300, made in Shanghai, China) at 180 °C for 4 min at roller speed of 40 rpm. The temperature, mixing time and roller speed remained unchanged in the experiment for all the blends. The samples for impact test were obtained by compression moulding the PP/EPDM blends at 180 °C, then cutting them into rectangular specimens. The size of the rectangular specimens was 63.5 × 12.7 × 3.2 mm. A notch was cut in these specimens. In order to study the effect of R, three kinds of notch named A, B and C were prepared by three knives, respectively. Fig. 1 is the SEM photographs showing the three notch tips. Notch A is a 45° V-shaped notch with the tip radius (R) of 0.25 mm, notch B is a 45° V-shaped notch with the R of 1.0 mm, and notch C is a rectangular notch.

2.3. Notched Izod impact test

The notched Izod impact strength of PP/EPDM blends with different EPDM contents were measured by a XJu Izod impact tester (made in Chengde, China) at various temperatures.

2.4. EPDM particle size analysis

Specimens for particle size analysis were initially cryo-fractured in liquid nitrogen. The morphologies of the surface were observed in a scanning electron microscope (SEM, JSM-5600LV, Japan). All the specimens were coated with a thin layer of gold prior to SEM observation. The weight average diameter d was defined as [1–2]:

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

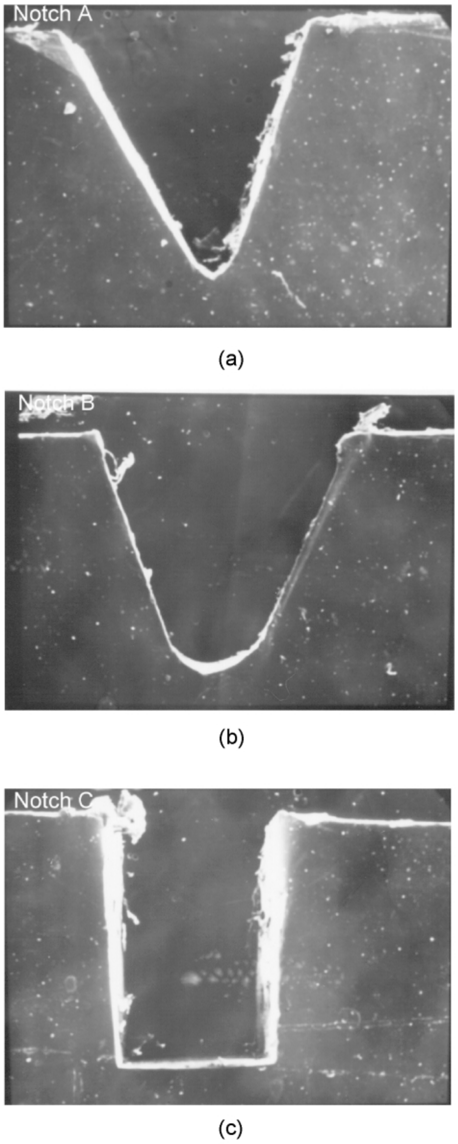

Fig. 1. SEM photographs showing the three notch tips.

3. Results and discussion

In general, the toughness (G) of a polymer or its blends is a function of R. Increasing R up to approaching infinity, the toughness of the polymer or its blends reach to a limit value G_{lim}. It can be expressed as:

$$G_{lim} = \lim_{\substack{R \to \infty \\ 1/R \to 0}} G_{(R)} \quad (2)$$

In present study, it was mentioned in the previous section that notch C is a rectangular notch, thereby its radius (R) approaches infinity. In this case, the toughness for notch C (G_C) is the limit value that the toughness can reach to with increasing R, and Eq. (2) can be rewritten as:

$$G_{\lim} = \lim_{\substack{R \to \infty \\ 1/R \to 0}} G_{(R)} = G_C \tag{3}$$

Fig. 2 shows the variation of the toughness of PP/EPDM blends with $1/R$ at various temperatures. The results reveal that the toughness of the PP/EPDM blends decreases with increasing $1/R$ at various temperatures. Moreover, increasing R from 1 mm to approaching infinity leads to a limited increase of the notched impact strength of the blends, indicating that the toughness of the PP/EPDM blends is less sensitivity to R if $R > 1$ mm.

Fig. 3(a)–(d) show the variation of Izod impact strength with temperature for various PP/EPDM blends and notches. The results indicate that the impact strength increases with increasing temperature. Moreover, a sharp transition of the toughness induced by temperature can be obviously observed for various blends and notches. The variation of T_{BD} with $1/R$ for various PP/EPDM blends is shown in Fig. 4. It is seen that the T_{BD} increases slowly with increasing $1/R$ from 0 to 4 mm^{-1}. Similar results were observed by Inberg and Gaymans [19] in PC/ABS blends. They found that the T_{BD} of the PC/ABS blend decreased slowly with increasing R from 0.25 to 1 mm.

In order to further study the effect of R on BDT, ID model proposed by Wu [1] was introduced into this study. The mathematical definition of ID was given as:

$$\mathrm{ID} = d_w\left[\left(\frac{k\pi}{6V_f}\right)^{1/3} - 1\right] \tag{4}$$

where d is the average diameter of dispersed particles, V_f is the volume fraction of dispersed particles, and $k = 1$ for the cubic lattice. In present study, V_f can be obtained form the weight fraction of EPDM rubber, the densities of PP and EPDM ($\rho_{EPDM} = 0.85$ g/cm^3, $\rho_{PP} = 0.90$ g/cm^3). The average EPDM particle diameters (d) for various EPDM contents can be obtained from SEM photographs and Eq. (1). The values of ID calculated from Eq. (4) are listed in Table 1.

Fig. 5(a)–(d) show variation of Izod impact strength with ID for various notches and temperatures. The results indicate that the impact strength increases with decreasing ID. A sharp transition of the toughness induced by ID can be observed for various temperatures and notches. The variation of $\mathrm{ID_c}$ with $1/R$ for various temperatures is shown in Fig. 6. The results reveal that $\mathrm{ID_c}$ not only depends on temperature, strain rate, properties of matrix and dispersed particle, et al. [2–8] as previous introduced, but also depends on R. The $\mathrm{ID_c}$ tends to decrease with increasing $1/R$ from 0 to 4 mm^{-1} for PP/EPDM blends at various temperatures. To the best of our

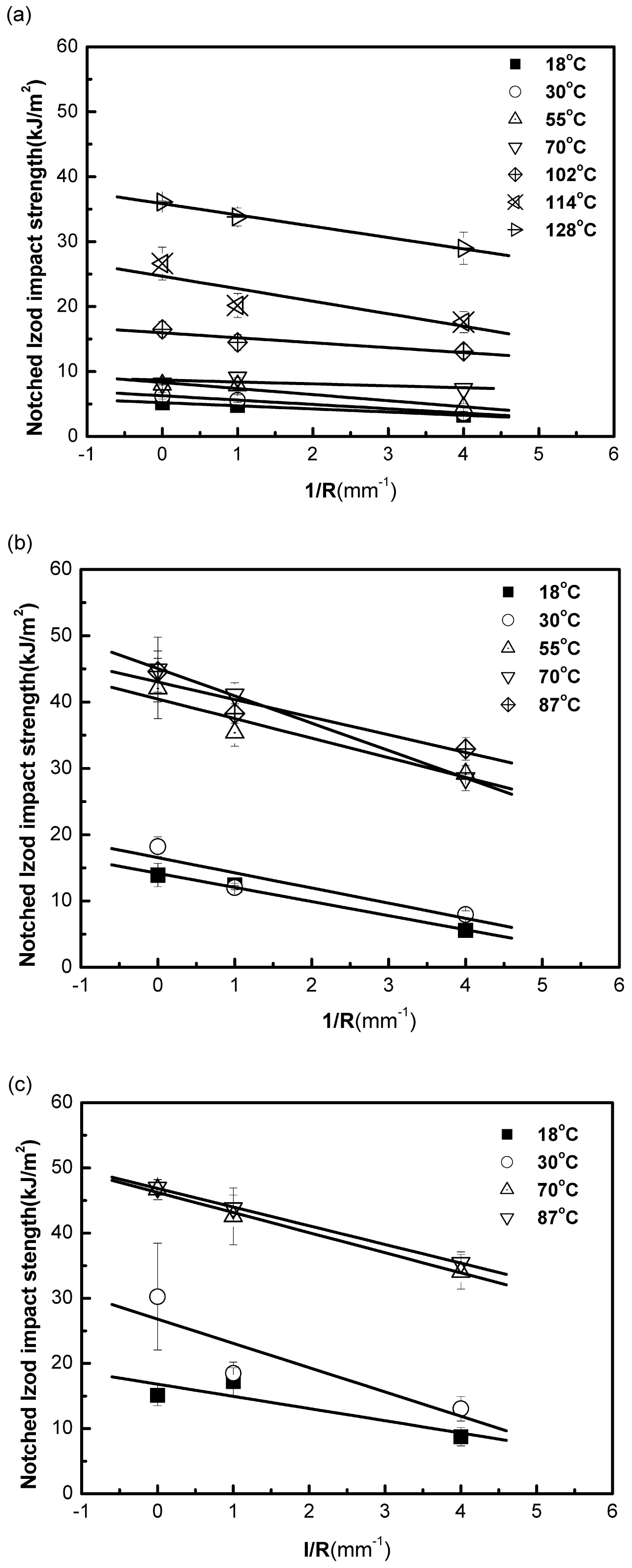

Fig. 2. Variation of notched Izod impact strength with $1/R$ at various temperatures. (a) 4.0 wt% EPDM; (b) 16 wt% EPDM; (c) 24 wt% EPDM.

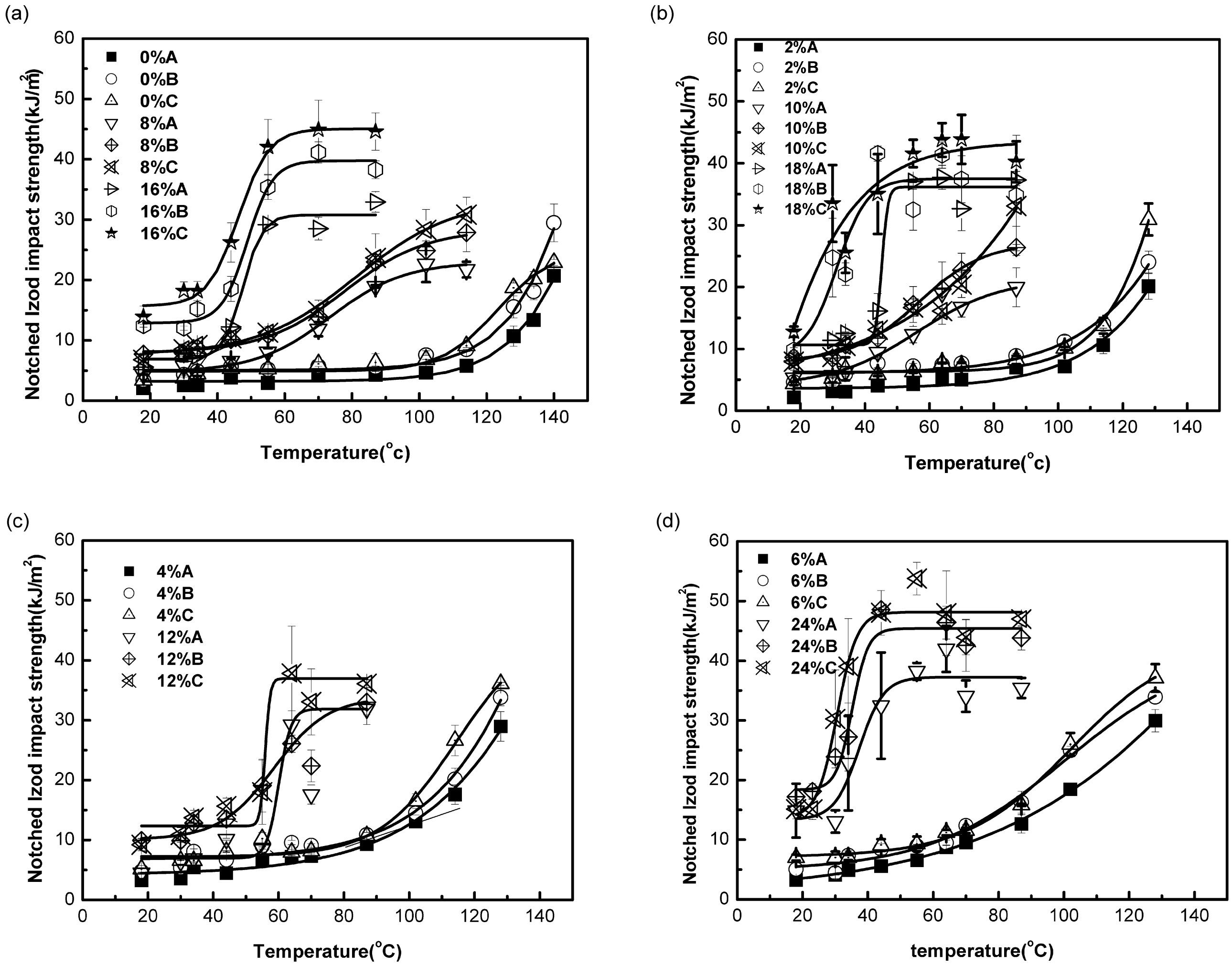

Fig. 3. Variation of notched Izod impact strength with temperature for various EPDM contents (weigh fraction) and notches (A, B, C).

knowledge this is the first time to report the correlation between ID_c and R for polymer blends.

Fig. 7 shows the variation of ID_c with temperature for notch A, B and C, respectively. It is seen that ID_c increases slowly with in creasing temperature in lower temperature region, whereas it increases sharply with increasing temperature in higher temperature region for various notches. Moreover, the results of Fig. 7 indicate that the ID_c-T curve shifts to higher temperature with changing notch from notch C to A (i.e. from higher R to lower R). Furthermore, it is found that these curves for different notches reduce down to a master curve (Fig. 8) if plotting ID_c versus $T_{BT}^{m}-T$, where T_{BT}^{m} is the T_{BT} of PP itself for a given notch. In this study, the values of T_{BT}^{m} for notches A, B, C are 114, 109, and 105 °C, respectively. Similar result was observed in nylon/EPDM and PP/EPDM blends. The two curves of ID_c versus T for PP/EPDM and nylon/EPDM blends reduce down a master curve if plotting ID_c versus $T_{BT}^{m}-T$ [3]. Present results further confirm that $T_{BT}^{m}-T$ is a more universal parameter that determines the BDT of polymers.

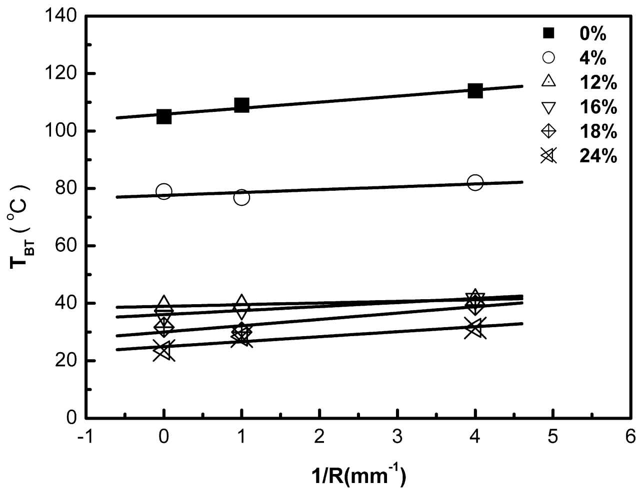

Fig. 4. Variation of brittle–ductile transition temperature with $1/R$ for various PP/EPDM blends.

4. Theoretical

Based on the elastic viscous theory of polymer and

Table 1
Values of weight average EPDM particle diameter (d_w), number average EPDM particle diameter (d_n), d_w/d_n and ID for various PP/EPDM blends

No.	Weight fraction of EPDM (%)	Volume fraction of EPDM (%)	d_w (μm)	d_n	d_w/d_n	ID value (μm)
1	0	0	–	–	–	∞
2	2.0	2.1	0.49	0.42	1.2	0.94
3	4.0	4.2	0.38	0.34	1.1	0.50
4	6.0	6.3	0.48	0.42	1.1	0.49
5	8.0	8.4	0.34	0.30	1.1	0.29
6	10.0	10.5	0.34	0.31	1.1	0.24
7	12.0	12.6	0.33	0.26	1.3	0.21
8	16.0	16.8	0.33	0.27	1.2	0.15
9	18.0	18.9	0.34	0.30	1.1	0.14
10	20.0	20.9	0.33	0.27	1.2	0.12
11	24.0	25.1	0.34	0.30	1.1	0.10

percolation model, the BDT equation for particle toughening polymer is given as [20]:

$$ID_c = \left[\frac{QE}{(T_{BT}^m - T)^2} + d^3\right]^{\frac{1}{3}} - d \qquad (5)$$

The definitions of d, ID_c, T, T_{BT}^m are same as those in the experiment, i.e. the diameter of dispersed phase, ID_c, test temperature and the T_{BT} of matrix polymer itself, respectively. From Eq. (5), it is known that ID_c is a function of T_{BT}^m and test temperature. ID_c increases slowly with increasing test temperature, whereas it increases sharply

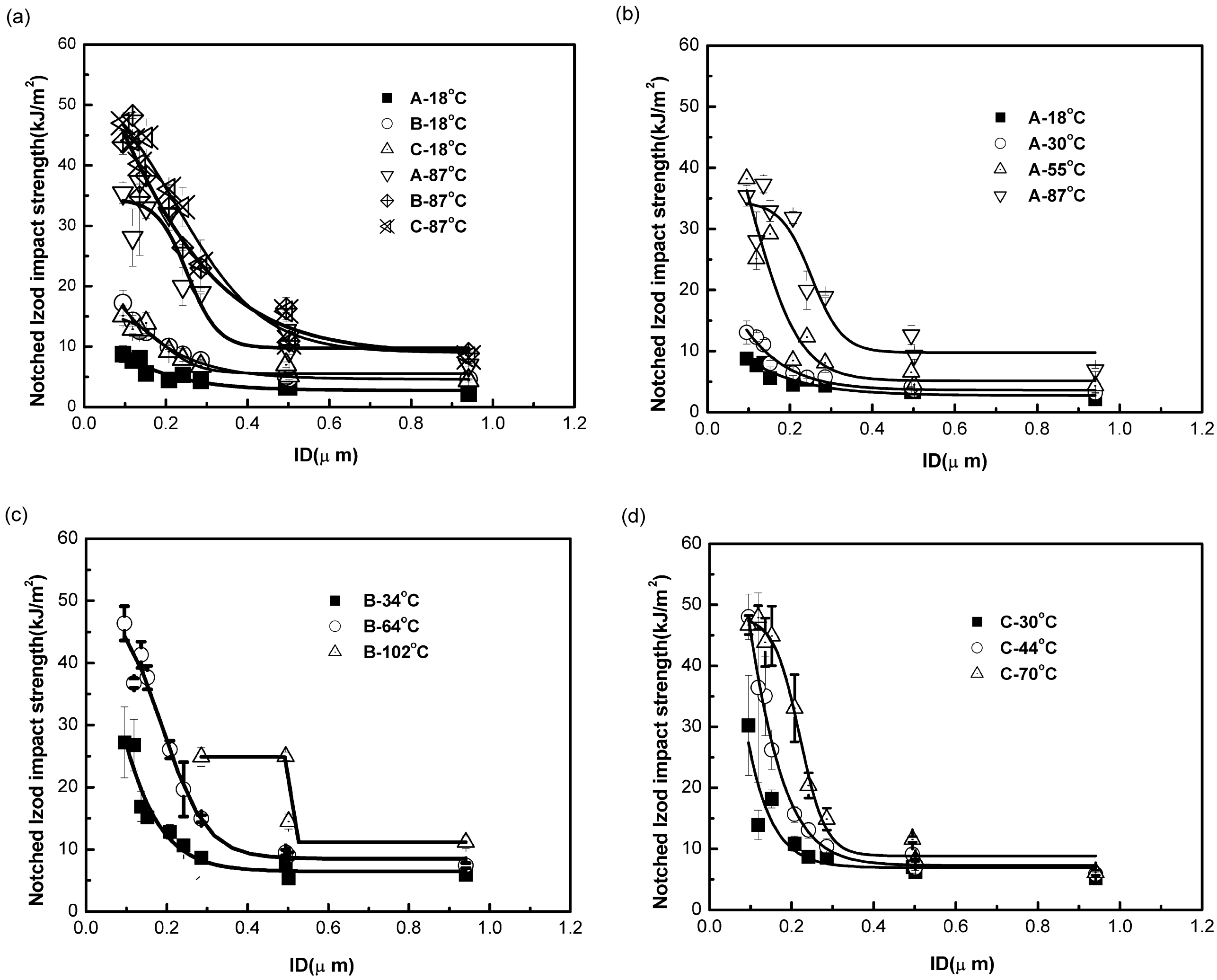

Fig. 5. Variation of notched Izod impact strength with interparticle distance (ID) for various notches and temperatures.

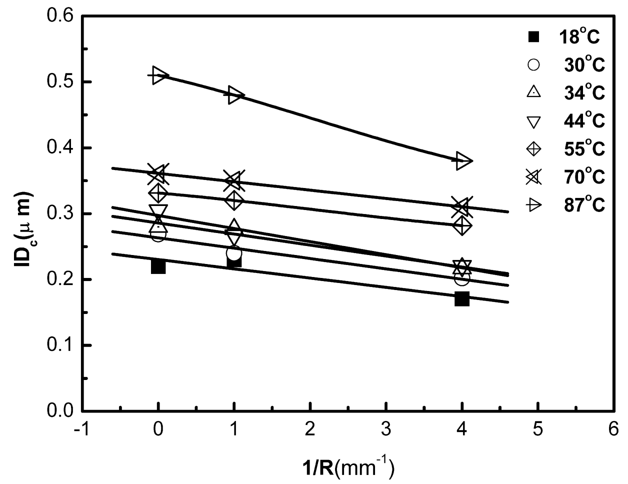

Fig. 6. Variation of critical interparticle distance (ID_c) with $1/R$ at various temperatures.

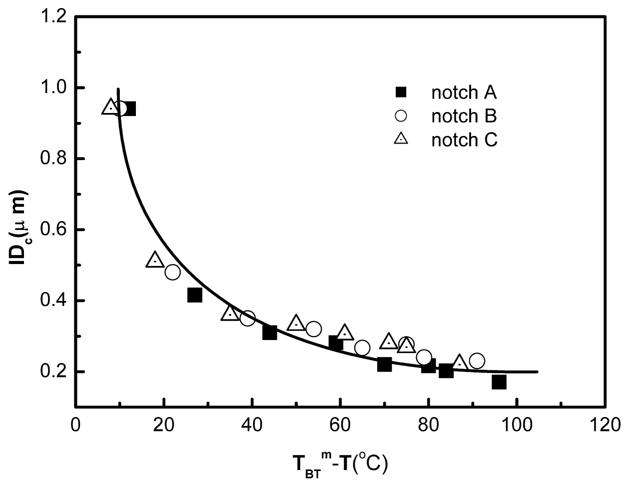

Fig. 8. Variation of critical interparticle distance (ID_c) with $T_{BT}^{m}-T$ for various notches.

with increasing temperature in the temperature region closing to T_{BT}^{m}. Moreover, from this equation it is known that ID_c reduces with increasing T_{BT}^{m}. However, comparing with T and T_{BT}^{m}, Eq. (5) indicates that $T_{BT}^{m}-T$ is a more important parameter that determines the brittle ductile transition of polymers. ID_c decreases nonlinearly with increasing $T_{BT}^{m}-T$, and the variation of ID_c with $T_{BT}^{m}-T$ is independent of T_{BT}^{m} and T, respectively. That is to say $T_{BT}^{m}-T$ is more important parameter to determine the BDT of polymers. For the purpose of comparison with the experiment, Eq. (5) was used to calculate the relation between ID_c and $T_{BT}^{m}-T$. In present study, we let $d = 0.34$ (μm) and $QE = 550$ (μm^3 K^2) for PP/EPDM blends. The calculated result for ID_c versus $T_{BT}^{m}-T$ from Eq. (5) is shown in Fig. 9. It is found that the experimental results are well in agreement with the theoretical prediction.

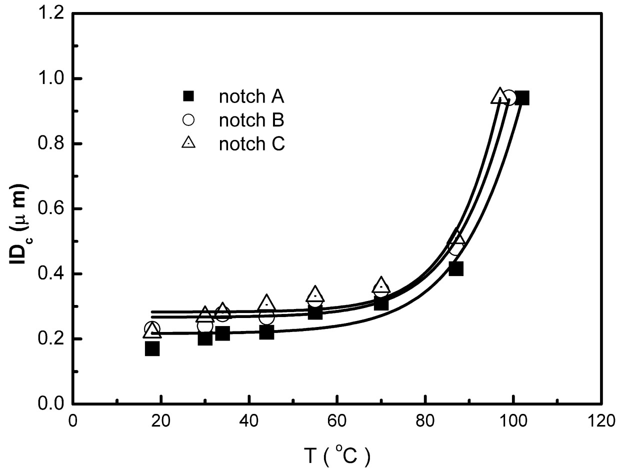

Fig. 7. Variation of critical interparticle distance (ID_c) with temperature (T) for various notches.

5. Conclusions

The results in present study revealed that both toughness and BDT of PP/EPDM blends depended on the R. The toughness tended to decrease with increasing $1/R$ for various PP/EPDM blends. Moreover, the T_{BT} increased with increasing $1/R$, whereas the ID_c reduced with increasing $1/R$. Finally, it revealed that the different curves of ID_c versus T for different notches reduced down to a master curve if plotting ID_c versus $T_{BT}^{m}-T$, where T_{BT}^{m} was the T_{BT} of PP itself for a given notch, indicating that $T_{BT}^{m}-T$ was a more universal parameter that determined the BDT of polymers. This conclusion was well in agreement with the theoretical prediction.

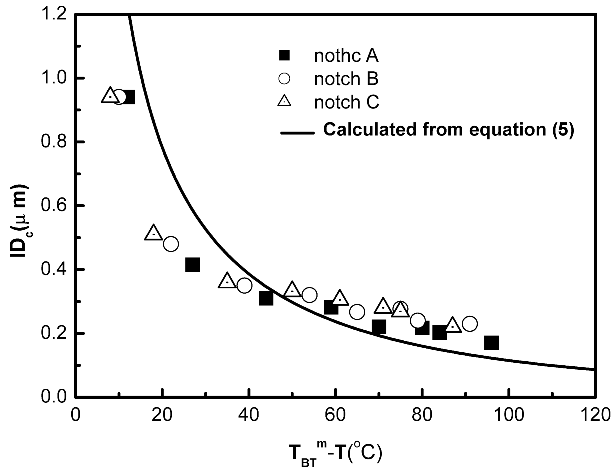

Fig. 9. Calculation results for variation of critical interparticle distance (ID_c) with $T_{BT}^{m}-T$ from Eq. (4), in which $d = 0.34$ (μm) and $QE = 550$ (μm^3 K^2). For the purpose of comparison, the experimental points are also plotted in this figure.

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