

STUDY ON HIGH WELD STRENGTH OF IMPACT PROPYLENE COPOLYMER/HIGH DENSITY POLYETHYLENE LAMINATES*

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Abstract The impact propylene copolymer (IPC) and isotactic polypropylene (*i*PP) were separately selected to prepare laminates with high density polyethylene (HDPE) by hot press. The peel forces of IPC/HDPE and *i*PP/HDPE laminates were examined, and it was found that the welded joint strength in IPC/HDPE laminate was dramatically higher than that of *i*PP/HDPE laminate. According to the special microstructure of IPC, the co-crystallization of the ethylene segments in ethylene-propylene block copolymer (EbP) component of IPC and the PE chain in HDPE was proposed to explain the high-strength welding. The DSC results indicated that there indeed existed some interaction between IPC and HDPE, and the crystallizable PE component in IPC could affect the crystallization of HDPE. The scanning electron microscope (SEM) observations of IPC/HDPE blends demonstrated that HDPE tended to stay with the PE-rich EbP chains to form the dispersed phase, indicating the good miscibility between HDPE and EbP components of IPC. According to the above results, the effect of co-crystallization of the PE components of the IPC and HDPE on the high weld strength of IPC/HDPE laminate was confirmed.

Keywords: Impact propylene copolymer; Polyethylene; CocrySTALLIZATION; High weld strength.

INTRODUCTION

For the composites prepared from two or more polymer materials, the interfacial adhesion between the components is one of the most concerned aspects because it strongly affects the resulting mechanical performances. Since the weld strength between different components for the polymer-polymer composites is mainly determined by interdiffusion, the relationship between interdiffusion and adhesive strength has attracted great researcher's interest^[1–8]. It was found that for the polymer-polymer laminates prepared by hot press, an interface layer would form, and in this region, polymer chains entangled if their molecular weights were higher than the critical entanglement molecular weight (M_c). As far as the laminates prepared with amorphous polymers are concerned, when they are cooled down from the molten state, the entanglement structure would be frozen in place, providing physical links. In this way, even immiscible polymers can adhere well^[1]. On the other hand, for

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the laminates prepared with semicrystalline polymers, the case is rather complex, because the entanglements may be eliminated or partly disengaged during the crystallization process^[8]. For example, the Ziegler-Natta catalyzed PE and isotactic polypropylene (*i*PP) can only generate low-strength welds unless some special thermal treatments are conducted^[6].

Since 1990s, a polypropylene in-reactor alloy, also called impact polypropylene copolymer (IPC), has attracted great academic and industrial attention. In general, IPC is prepared by two-step polymerization, *i.e.* bulk polymerization of propylene and then gas-phase polymerization of ethylene and propylene with spherical Ziegler-Natta catalysts^[9, 10], and researches demonstrate that IPC mainly contains three compositions, *i.e.* ethylene-propylene random copolymer (EPR), a series of ethylene-propylene block copolymers with different sequence lengths (EbP) and propylene homopolymer (HPP)^[11–14]. Due to its good impact toughness, IPC is widely applied in many fields, ranging from exquisitely engineered limousine bumpers to cheap crates. In recent decade, many studies on composition, microstructure, morphology, crystallization and melting behavior of IPC have been conducted^[12, 13, 15–23]. However, up to now, there exist few reports about IPC composites. Recently, we prepared the IPC/HDPE laminate and surprisingly found that it possessed rather high weld strength. In the present article, we try to study this phenomenon in detail and interpret the cause of high weld strength in IPC/HDPE laminates through probing the thermal behavior and morphology of IPC/HDPE blends.

EXPERIMENTAL

Materials and Sample Preparation

Materials

The commercial IPC (SP179) was purchased from SINOPEC Qilu petrochemical Co., Ltd. The commercial *i*PP (T300) was purchased from SINOPEC Shanghai petrochemical Co., Ltd. The commercial HDPE (5000S) was purchased from SINOPEC Yanshan petrochemical Co., Ltd. The molecular weights and polydispersities of the three polymers are listed in Table 1.

Table 1. Molecular weights and polydispersity of the raw materials

Polymer	$M_n \times 10^{-4}$	$M_w \times 10^{-4}$	Polydispersity
IPC	4.4	17.3	3.9
<i>i</i> PP	5.7	26.8	4.7
HDPE	4.8	28.3	5.9

Preparation of IPC/HDPE and *i*PP/HDPE laminates

The IPC, *i*PP and HDPE sheets (155 mm × 120 mm) with the thickness of 0.9 mm were prepared by hot press. The IPC/HDPE and *i*PP/HDPE laminates with the thickness of 1.2 mm were made by hot-pressing these sheets at 180°C for 15 min. Subsequently, the laminates were cooled down to room temperature. At last, the specimens for peeling test were prepared by cutting the laminates into strips with length of 150 mm and width of 30 mm.

Preparation of IPC and HDPE blends

The IPC/HDPE blends with different compositions were prepared by HAAKE miniaturized internal mixer rheomix 600 at 180°C with a roll speed of 40 r/min. The mixing time was 10 min.

Peeling Test

The peel tests of IPC/PE and *i*PP/PE laminates were conducted on a CMT 4204 universal test machine (Shenzhen Sans Co., Ltd., China). The sheets were peeled at 90° angle with a tensile speed of 100 mm/min at ambient conditions of temperature being 20°C and air humidity being 50%. The applied force was recorded as a function of displacement.

SEM Observation

The surface of IPC and HDPE sheets before hot press and after peeling was observed through SEM (Hitachi S-4800) after coating gold-palladium. Fracture surfaces of specimens of IPC and IPC/HDPE blends obtained at

liquid N₂ were etched in xylene for 15 h at room temperature, and then were observed through SEM (SIRION, FEI, Netherland) after coating gold-palladium.

DSC Analysis

The thermal behavior was examined by using a Q100 MDSC (TA Instruments Corporation) with N₂ as purge gas. The samples were heated to 200°C and held for 10 min to eliminate previous thermal history. The cooling and heating rate adopted in this study was 10 K/min.

Successive self-nucleating and annealing (SSA)^[24] was performed according to the following procedure: First the samples were preheated to 200°C for 10 min and then cooled down to 30°C at a cooling rate of 10 K/min; subsequently, the SSA treatment was started. As introduced in literatures^[19, 25, 26], the first self-seeding temperature (T_s) in SSA course should be the temperature at which almost all the crystals have been molten and only small crystal fragments acting as self-nuclei were left. After the self-nucleation experiments have been done^[26], 169°C was chosen as the first T_s for blends of IPC and HDPE. For comparison, 169°C was chosen as the first T_s for HDPE. The fractionation window adopted here was 5°C and the annealing time was 10 min. The scanning rate during the thermal conditioning steps was 10 K/min. The temperatures of thermal fractionation ranged from 84°C to 169°C. After the SSA treatment, the melting traces were recorded at a heating rate of 10 K/min.

RESULTS AND DISCUSSION

High Strength-welds in IPC/HDPE Laminate

Figure 1 gives the plots of peel force versus displacement for IPC/HDPE and *i*PP/HDPE laminates. The adhesion between *i*PP and HDPE is so weak that the welds can not even support a 90° angle during the peeling process, while the peel force of IPC/HDPE laminate is much greater than that of *i*PP/HDPE laminate. In addition, the peeled surface of *i*PP/HDPE laminate appears smooth while that of IPC/HDPE laminate is rather coarse. These results imply that the IPC/HDPE laminate possesses higher weld strength compared with the *i*PP/HDPE laminate. Figure 2 gives the SEM images for the surface of IPC sheets before hot press (a) and after peeling (b), respectively. It can be seen that before hot press, the IPC sheet is smooth and flat, but after peeling process, holes and cracks can be observed on the IPC sheet. It seems that some materials were pulling out of the sheet during the peeling process. Figure 3 gives the SEM images for the surface of HDPE sheet before hot press (a) and after peeling (b). It can be seen that some materials are attached on the surface of the HDPE sheet after peeling. The morphology of the peeled surface of the IPC and HDPE can well demonstrate the high peeling force for the IPC/HDPE laminate.

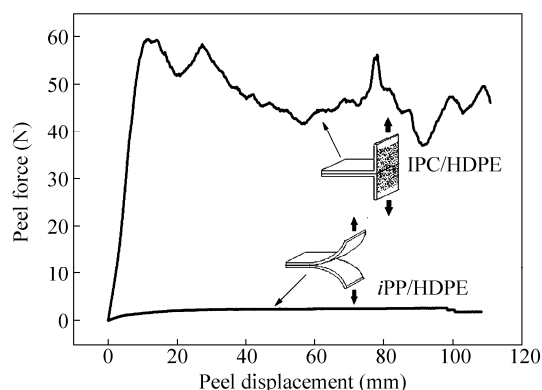


Fig. 1 Peel force versus displacement for IPC/HDPE and *i*PP/HDPE laminates

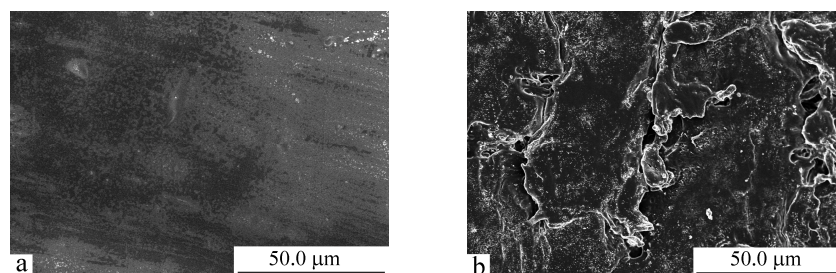


Fig. 2 SEM micrographs for the surface of IPC sheets before hot press (a) and after peeling (b)

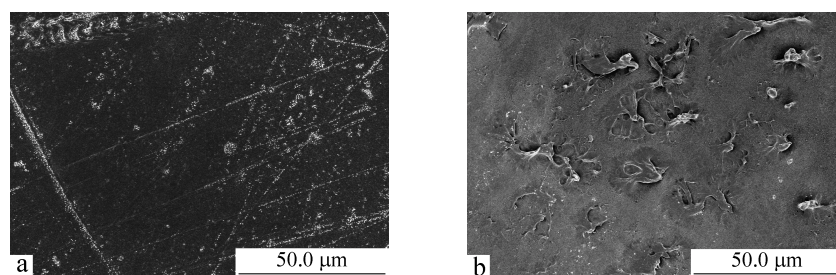


Fig. 3 SEM micrographs for the surface of HDPE sheets before hot press (a) and after peeling (b)

It is reasonable to believe that during hot-press process an interface between components will develop in *i*PP/HDPE and IPC/HDPE laminates due to the motions of polymer chains. As the HDPE and *i*PP or IPC molecular chains move across the molten phase boundary, numerous topological entanglements will form. For the *i*PP/HDPE laminate, the polymer chains separately fold into crystalline lamellae during cooling course because there is no tendency to form co-crystals^[8, 25]. Hence, most of entanglements formed by *i*PP and HDPE chains will disengage due to the accumulation of amorphous PP or PE materials at the interface layer. This result has been confirmed by transmission electron microscopy (TEM) observation and it was believed that the amorphous materials were swept to the interface layer through a process analogous to zone refining during the crystallization^[8]. As a result, the interfacial strength of *i*PP/HDPE laminate is extremely weak because the weld is composed of rubbery materials. It is noted that the main difference between *i*PP and IPC is that IPC contains a certain amount of ethylene-propylene copolymers. Therefore, it is suggested that these copolymers may play a key role in enhancing the joint strength of IPC/HDPE laminate. It is believed that even the content of *EbP* is lower than that of EPR in this IPC^[14], the good welding property of IPC/HDPE laminate should be contributed by the *EbP* component, not EPR, because EPR hardly crystallizes and is in high-elastic state at room temperature. Therefore, despite there is good miscibility between EPR and PE, and lots of entanglements formed between the molecular chains of two polymers can be preserved during the crystallization process, the soft and rubbery EPR could hardly support the high peeling force imposed on the laminate. However, the case for *EbP* may be different totally. During the cooling process the crystallizable PE segment in *EbP* chains can form co-crystals with PE chains in HDPE. In addition, the crystallizable PP segment in *EbP* chains can entangle with the HPP chains in IPC, and PP co-crystals can form during the cooling process^[14], and as a result, IPC and HDPE components are anchored by these *EbP* chains.

The co-crystallization between IPC and HDPE can also give a rational explanation for the morphology of the peeled surface of the IPC and HDPE. It is well known that the dispersed phase of IPC has a core-shell structure in which the core is mainly composed of PE crystals and the shell consists of EPR^[18, 20, 27]. Therefore, for IPC, the PE component is warped by EPR which has a relatively low strength. During the peeling, the PE cores in IPC were easy to be pulled out together with some EPR shells, even with some HPP component due to the connection between some of the PE cores and the HPP matrix^[27], thus holes and cracks formed on the surface of IPC sheet and some materials from the IPC were attached on the surface of HDPE sheet.

Crystallization of IPC/HDPE Blends

Since the interface layer of the IPC/HDPE laminate is too thin to be separated, the melting blends of IPC and HDPE were prepared to seek the possibility of formation of PE co-crystals between IPC and HDPE. Figure 4 gives a comparison of the DSC melting traces between the unmixed and melting-mixed IPC/HDPE samples with various compositions nonisothermally crystallized from 200°C to 30°C at a cooling rate of 10 K/min. Figure 4(a) gives the melting traces of pure IPC, HDPE and their ‘unmixed samples’. The so called ‘unmixed sample’ is the one in which two components never contact with each other inside DSC crucible. It can be found that HDPE presents a melting peak at 132.2°C, corresponding to the melting of PE crystals. Meanwhile, IPC exhibits two distinct melting peaks. Moreover, the endothermic peak at about 163.2°C should be attributed to the melting of propylene homopolymer and the another peak at about 115°C may result from the melting of polyethylene crystal. Different from crystals formed by linear PE chains in HDPE, PE crystals in IPC should result from ethylene-propylene copolymer, especially the PE-rich *EbP* chains. Furthermore, due to the existence of polypropylene units in copolymer chains, the lamellar thickness of these PE crystals in IPC is lower than that in HDPE. Figure 4(b) gives the melting traces of melting-mixed blends of IPC and HDPE. Different from the unmixed samples exhibiting two PE melting peaks, the melting-mixed blends only exhibit a single PE melting peak whose temperature is higher than the melting temperature of PE in IPC, but lower than that of HDPE. In addition, the temperatures of PE melting peaks for the blends shift to high temperature with the increase of HDPE content. These results indicate that the PE co-crystals form between two polymers so that only a single PE melting peak can be observed for these blends. At the same time, for the mixed samples the melting peak temperatures of PP are also a bit lower than those of the unmixed samples, implying that HDPE has a limited dilution effect for the IPC. Hence, to understand the co-crystallization of PE better, the SSA treatment is adopted to investigate their thermal behavior.

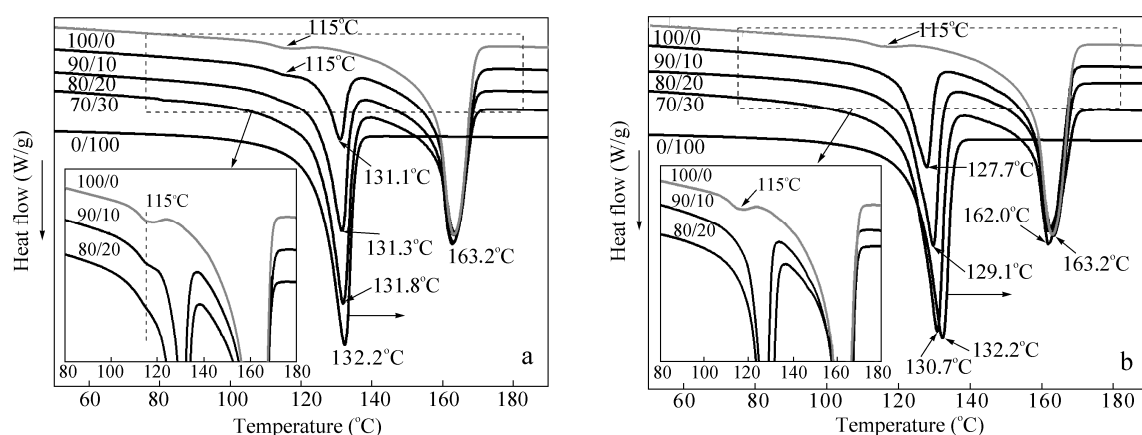


Fig. 4 DSC melting traces for IPC/HDPE blends at a heating rate of 10 K/min for (a) unmixed samples and (b) solution-mixed samples

Figure 5 gives a comparison of the melting traces between unmixed and melting-mixed IPC/HDPE samples after experiencing an SSA treatment from 169°C to 84°C. Figure 5(a) gives the melting traces of pure IPC, HDPE and their unmixed samples. For pure HDPE, the thermal fractionation effect of SSA on the main crystallization temperature range is limited, indicating that the crystals in HDPE are relatively uniform. This phenomenon could be easily understood because the fractionation effect of SSA mainly depends on the heterogeneity or defects content of the polymer chains, and HDPE is mainly composed of long and linear PE chains. In contrast, the fractionation effect on PE crystals in IPC is much strong, indicating that these crystals result from the PE chain segments with various lengths. In total, the melting traces of the mixed blends present the effect of the proportional addition of the heat flow for the two polymers. Figure 5(b) gives the melting traces

of mixed blends after being subjected to SSA treatment. Compared with the unmixed samples, the melting-mixed samples present different melting characteristics. To demonstrate the differences clearly, the melting peaks are labeled with numbers and the peaks with the same number represent the melting of the crystals crystallized at the same temperature. It can be seen that the width of peak 1 for the mixed blend with composition of 70/30 is smaller than that for the unmixed sample with the same composition. When the composition varies to 80/20, the difference between the unmixed and mixed sample in peak 1 is more obvious. For the composition of 90/10, the peak 1 of the melting-mixed blend disappears while the corresponding peak of the unmixed blend is strong and obvious; in contrast, the peak 2 of the melting-mixed blends is stronger than that of unmixed samples with same composition. On the other hand, the difference between the unmixed and mixed samples in peak 2 becomes more obvious with the increase of the IPC content. These results confirm the existence of PE co-crystallization resulting from PE chains in HDPE and EbP chains in IPC, since the peaks 1 and 2 should be ascribed to the melting of long linear PE chains from HDPE. Considering that the melting temperature of polymeric crystals is determined by the lamellar thickness, peak 1 should be attributed to the linear long PE chains which can form crystals with relative thick and uniform lamellae. When HDPE is blended with IPC, the co-crystallization of HDPE chains and PE-rich EbP chains results in a decrease in the lamellae thickness of the PE crystals with highest melting temperature because the PP or EPR segments contained in the EbP chains will act as the defects in the crystallization process. Furthermore, with the increase of IPC content, more and more EbP chains are involved in the co-crystallization course and the lamellar thickness decreases further. As a result, for the melting-mixed blend with composition of 90/10, the peak 1 disappears because there is no PE crystal whose lamellar thickness is equivalent or close to the crystals in HDPE.

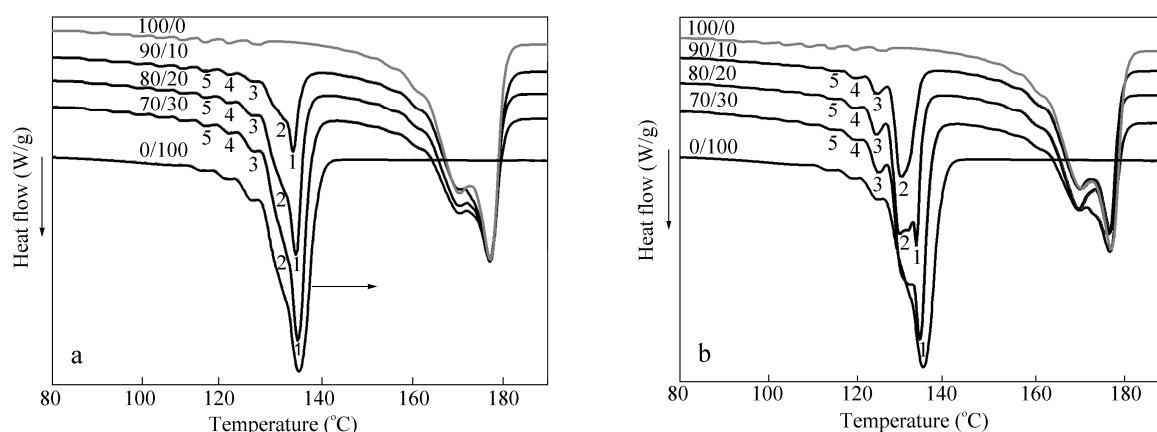


Fig. 5 DSC melting traces at a heating rate of 10 K/min after SSA treatment for (a) unmixed samples and (b) solution-mixed samples

Morphology Study of IPC/HDPE Blends

To further investigate the co-crystals formed by IPC and HDPE, it is necessary to observe the morphology of the melting-mixed IPC/HDPE blends. Figure 6(a) gives the SEM images of the fracture surface for virgin IPC after being etched by xylene. As reported previously^[27], a two-phase structure with good dispersion can be observed and the dispersed phase presents three different cases: (i) isolated holes marked as 'A'; (ii) holes but containing a smaller granule marked as 'B'; (iii) aggregate granules marked as 'C'.

As a phase-separated system, the matrix and the main dispersed phase in IPC are considered to be HPP and EPR, respectively^[18, 20, 21]. Based on the chain structure analysis and morphology observation, we have proposed a phase structure model of IPC, as shown in Fig. 6(b)^[27]. The dispersed phase is composed of a PE-rich EbP core and two layers, and the outer layer is PP-rich EbP and the inner one is EPR. In particular, some EbPs pass through the EPR layer and form some bridges connecting the core and EbP layer. When the IPC sample is etched by xylene and the EPR shells are solved, the EbP cores completely wrapped by EPR layer are removed

and the EbP cores with weaker EbP bridges are also removed. Consequently, the holes form on the fracture surface, as indicated by arrows 'A' in Fig. 6(a). However, for the dispersed phase where the PE-rich cores are not totally wrapped by the EPR shell, the cores may still remain and form the holes with a smaller granule, being indicated by arrows 'B' in Fig. 6(a). In addition, when the dispersed particles get together and the outer EbP layers result in a invalid etch of xylene, the aggregation of dispersed particles can be observed on the fracture surface, as shown by arrows 'C' in Fig. 6(a).

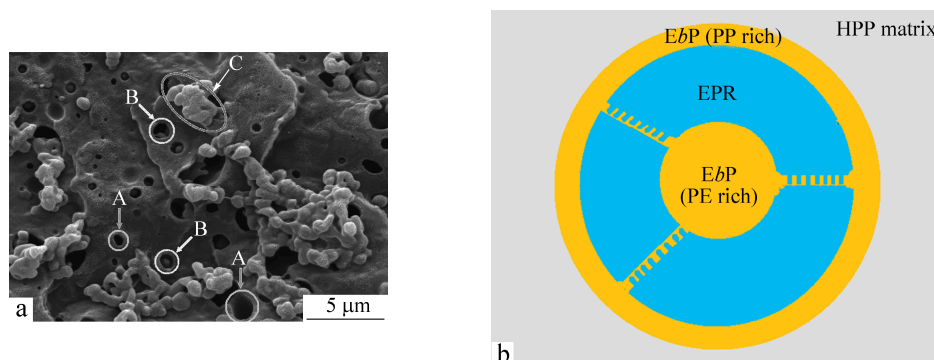


Fig. 6 SEM micrographs of fracture surface (a) and phase structure diagram (b) for IPC

Figure 7 presents the SEM image of the fracture surface for IPC/HDPE blends with different compositions. Figures 7(a) and 7(b) show the images of IPC/HDPE blends with composition of 100/0 and 95/5 respectively. Compared with the virgin IPC, the 95/5 blend presents different morphological characteristics. First, the size of the dispersed phase is slightly larger. Secondly, nearly all the holes produced by the etching of xylene are not empty, as if some materials are filled into them. Thirdly, the aggregation of dispersed particles as shown in the SEM images of virgin IPC could hardly be found here. In other words, for the 95/5 blend sample, most of the dispersed particles present the characteristics of type 'B'. Figures 7(c)–7(f) give the SEM images for IPC/HDPE blends with compositions of 90/10, 80/20, 70/30 and 60/40, respectively. It can be found that the morphologies of these blends are similar to that of the 95/5 blend, but the sizes of the dispersed particles increase with the increase of HDPE content. Figure 7(g) shows the image of IPC/HDPE blends with compositions of 50/50. It can be found that phase inversion is occurred and the HPP component becomes the dispersed phase.

The morphological differences between virgin IPC and IPC/HDPE blend may lie in the reasons as follows: When IPC and HDPE are melt-mixed, both of them are broken into small liquid drops and the core-shell structure in IPC is also destructed due to the shear effect. As a result, HDPE has chances to touch with every component of IPC during the mixing process. Since the PE has a better miscibility with ethylene-propylene copolymer than HPP, the HDPE components tend to stay away from the HPP component, but stay together with the copolymer component in IPC, especially the EbP component comprising long PE segments. When the mixing process ceases, a core-shell structure of the dispersed phase can re-form, and the core is composed of PE from HDPE and PE-rich EbP from IPC and the shell is composed of EPR from the IPC. Therefore, with the increase of HDPE content, the size of the cores becomes larger so that the cores can not be totally wrapped by the EPR shell. Hence, when the blends are etched by xylene, only the EPR shells can be removed, and the cores could remain in the holes due to their bonding to the HPP matrix, as shown in Fig. 7.

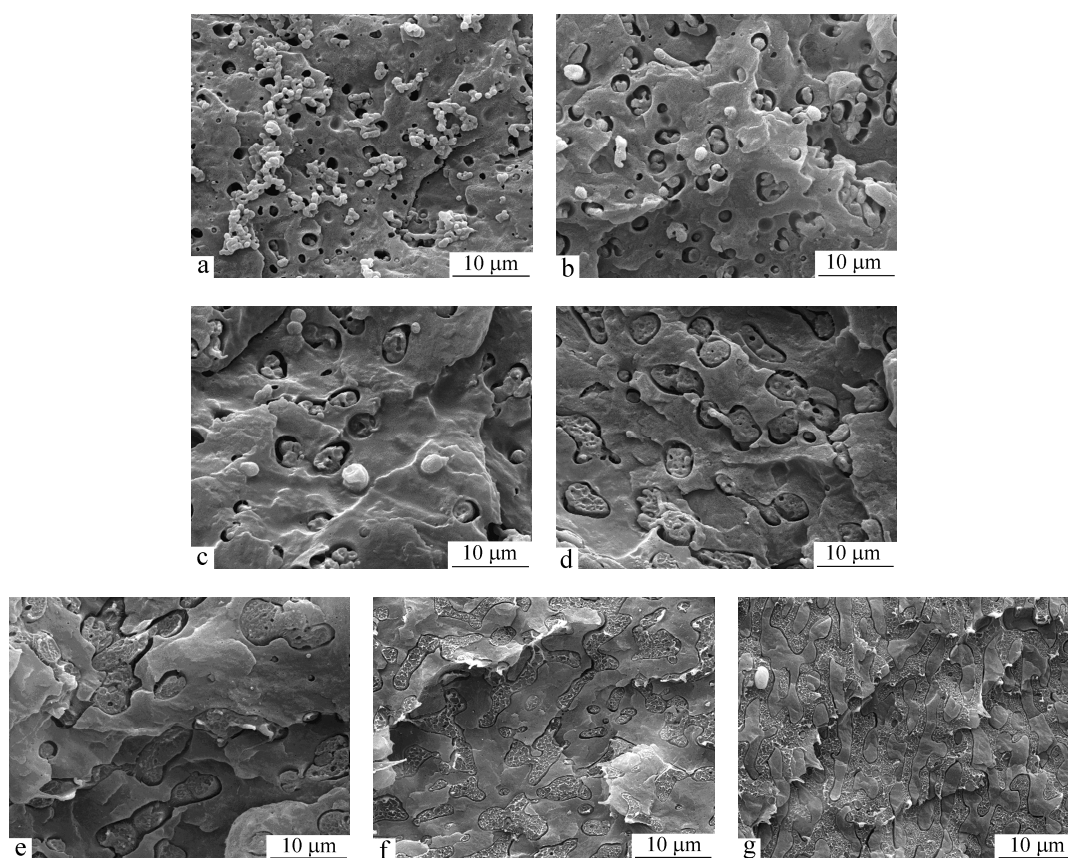


Fig. 7 SEM micrographs for fracture surface of the IPC/HDPE blends with different compositions: (a) 100/0, (b) 95/5, (c) 90/10, (d) 80/20, (e) 70/30, (f) 60/40 and (g) 50/50

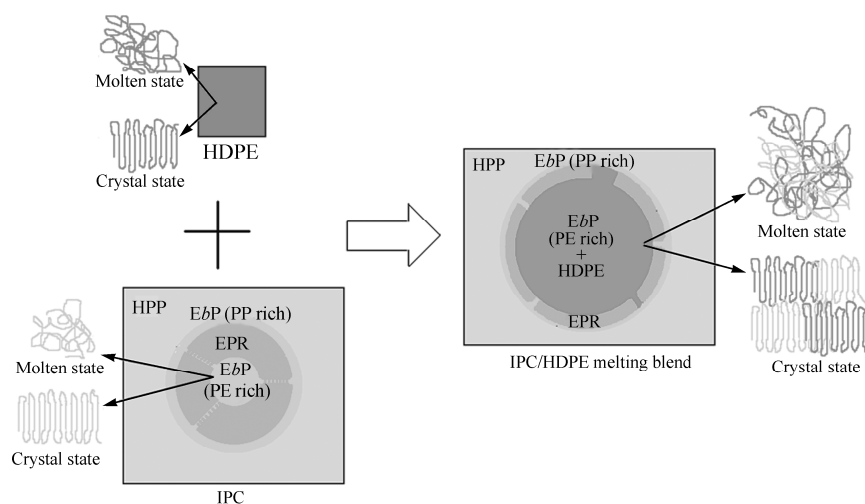


Fig. 8 Schematic diagrams for the formation and crystallization of IPC/HDPE melting blend

Taking above morphological results into consideration, we believe that the compatibility between the HDPE and PE-rich EbP component of IPC is good, which could facilitate the co-crystallization between the PE-rich EbP chains in IPC and PE chains in HDPE. Figure 8 gives the schematic of re-form of dispersed phase and the formation of the co-crystals. During the melting mixing, the PE-rich EbP chains in the IPC and the PE chains

in the HDPE will mix and entangle with each other, and during cooling their chains fold together and form PE co-crystals.

SUMMARY

The peel force tests of IPC/HDPE and *i*PP/HDPE laminates show that the welded joint strength of IPC/HDPE laminate is much higher than that of the *i*PP/HDPE one. The DSC results indicate that there exist some interaction between IPC and HDPE, and the crystallizable PE component in IPC could affect the crystallization of HDPE. Considering the special two-phase structure of IPC, it is proposed that the co-crystallization of polyethylene chain segments in ethylene-propylene block copolymer component of IPC and the PE chain in HDPE is responsible for this high-strength welding property. On the basis of the morphological results of IPC/HDPE blends, it is suggested that HDPE tends to stay with the PE-rich *EbP* chains in IPC to form the dispersed phase, indicating the good compatibility between HDPE and *EbP* components of IPC. According to the above results, the effect of co-crystallization between the PE components of the IPC and HDPE on the high strength weld of IPC/HDPE laminate is demonstrated clearly.

REFERENCES

- 1 Cole, P.J., Cook, R.F. and Macosko, C.W., *Macromolecules*, 2003, 36: 2808
- 2 Schnell, R., Stamm, M. and Creton, C., *Macromolecules*, 1998, 31: 2284
- 3 Schnell, R., Stamm, M. and Creton, C., *Macromolecules*, 1999, 32: 3420
- 4 Willett, J.L. and Wool, R.P., *Macromolecules*, 1993, 26: 5336
- 5 Kim, H.J., Lee, K.J. and Seo, Y., *Macromolecules*, 2002, 35: 1267
- 6 Lo, C.T., Laabs, F.C. and Narasimhan, B., *J. Polym. Sci. Part B: Polym. Phys.*, 2004, 42: 2667
- 7 Chaffin, K.A., Bates, F.S., Brant, P. and Brown, G.M., *J. Polym. Sci. Part B-Polym. Phys.*, 2000, 38: 108
- 8 Chaffin, K.A., Knutsen, J.S., Brant, P. and Bates, F.S., *Science*, 2000, 288: 2187
- 9 Galli, P. and Haylock, J.C., *Prog. Polym. Sci.*, 1991, 16: 443
- 10 Simonazzi, T., Cecchin, G. and Mazzullo, S., *Prog. Polym. Sci.*, 1991, 16: 303
- 11 Xu, J., Feng, L., Yang, S., Wu, Y., Yang, Y. and Kong, X., *Polymer*, 1997, 38: 4381
- 12 Fan, Z., Zhang, Y., Xu, J., Wang, H. and Feng, L., *Polymer*, 2001, 42: 5559
- 13 Tan, H., Li, L., Chen, Z., Song, Y. and Zheng, Q., *Polymer*, 2005, 46: 3522
- 14 Zhang, C., Shangguan, Y., Chen, R. and Zheng, Q., *J. Appl. Polym. Sci.*, 2011, 119: 1560
- 15 Zheng, Q., Shangguan, Y., Yan, S., Song, Y., Peng, M. and Zhang, Q., *Polymer*, 2005, 46: 3163
- 16 Shangguan, Y., Song, Y. and Zheng, Q., *Polymer*, 2007, 48: 4567
- 17 Shangguan, Y., Tao, L. and Zheng, Q., *J. Appl. Polym. Sci.*, 2007, 106: 448
- 18 Tan, H., Xie, K., Liu, W., Hou, B., Shangguan, Y. and Zheng, Q., *Acta Polymerica Sinica*(in Chinese), 2006, (9): 1106
- 19 Zhu, H., Monrabal, B., Han, C.C. and Wang, D., *Macromolecules*, 2007, 41: 826
- 20 Chen, Y., Chen, W. and Yang, D., *J. Appl. Polym. Sci.*, 2008, 108: 2379
- 21 Song, S., Feng, J., Wu, P. and Yang, Y., *Macromolecules*, 2009, 42: 7067
- 22 Nitta, K.H., Shin, Y.W., Hashiguchi, H., Tanimoto, S. and Terano, M., *Polymer*, 2005, 46: 965
- 23 Wang, S. and Yang, D., *Polymer*, 2004, 45: 7711
- 24 Arnal, M.L., Sánchez, J.J. and Müller, A.J., *Polymer*, 2001, 42: 6877
- 25 Arnal, M.L., Balsamo, V., Ronca, G., Sanchez, A. and Muller, A.J., Canizales, E., Navarro, C.U.D., *J. Therm. Anal. Calorim.*, 2000, 59: 451
- 26 Muller, A.J. and Arnal, M.L., *Prog. Polym. Sci.*, 2005, 30: 559
- 27 Zhang, C., Shangguan, Y., Chen, R., Wu, Y. and Zheng, Q., *Polymer*, 2010, 51(21): 4969