

# Mechanical Behaviors of Ethylene/Styrene Interpolymer Compatibilized Polystyrene/Polyethylene Blends

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**ABSTRACT:** In this study, ethylene/styrene interpolymer was used as a compatibilizer for the blends of polystyrene (PS) and high-density polyethylene (HDPE). The mechanical properties including tensile and impact properties and morphology of the blends were investigated by means of uniaxial tension, instrumented falling-weight impact measurements, and scanning electron microscopy. Tensile tests showed that the yield strength of the PS/HDPE/ESI blends decreases considerably with increasing HDPE content. However, the elongation at break of the blends tended to increase significantly with increasing HDPE content. The excellent tensile ductility of the HDPE-rich blends resulted from shield yielding of the matrix. Izod and Charpy impact

measurements indicated that the impact strength of the blends increases slowly with HDPE content up to 40 wt %; thereafter, it increases sharply with increasing HDPE content. The impact energy of the HDPE-rich blends exceeded that of pure HDPE, implying that the HDPE polymer can be further toughened by the incorporation of brittle PS minor phase in the presence of ESI compatibilizer. The correlation between the impact property and morphology of the blends is discussed. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 104: 4001–4007, 2007

**Key words:** impact; compatibilization; ethylene/styrene interpolymer; polystyrene; polyethylene; blends; morphology

## INTRODUCTION

Polymer blending has proved to be an economically viable and versatile way to control the performance of polymeric materials with readily available polymers.<sup>1,2</sup> However, the great challenge confronting the development of polymer blends has been related to the thermodynamical immiscibility of most polymers. Compatibilization of the immiscible component polymers is therefore a must for preparing polymer blends with fine-tuned properties of the component polymers. Therefore, interfacially active compatibilizers are often introduced in polymer blends by the use of either additive compatibilization or reactive compatibilization.<sup>3–14</sup> Although reactive compatibilization is now widespread,<sup>4–11</sup> additive compatibilization, that is, mixture with appropriate block copolymers (BCs), appears to be the most effective procedure and was reviewed in detail.<sup>12–14</sup> BCs with block miscible or at least compatible with the corresponding blend component effectively reduce the interfacial tension between the constituent homopolymers, thus leading to finer dispersion as well as improved interfacial adhesion of two phases.

It is generally known that PS is incompatible with PE.<sup>15</sup> Their blends normally form either dispersed or

continuous phase structures, depending on the chemical composition of two component polymers. Typically, these blends exhibit weak adhesion (i.e., poor stress transfer) between the PS and PE phases, which manifest as inferior mechanical properties in the final blends.<sup>16</sup> Considerable efforts have been made to improve the compatibilization of PS and PE to either increase the toughness of PS or recycle PE waste. Over the past three decades, both reactive compatibilization using graft copolymers<sup>7–11</sup> and additive compatibilization using BCs<sup>12–30</sup> as compatibilizers have been introduced for PS-PE blends. The effect of compatibilization on the morphology of the blends<sup>1,15–18</sup> and its mechanical properties<sup>19–30</sup> has been intensively studied. Representative compatibilizers including styrene-butadiene (SB),<sup>28–30</sup> hydrogenated butadiene-PS diblock copolymers (HPB-*b*-PS),<sup>18–20</sup> hydrogenated butadiene-polystyrene copolymers (SEB),<sup>17,22</sup> polystyrene-hydrogenated butadiene-polystyrene triblock copolymers (SEBS)<sup>17,22,25,26</sup> have been reported. It follows from some studies that diblock copolymers are the most efficient compatibilizers.<sup>17,22,28–30</sup> On the other hand, some authors refer to triblocks or multiblocks as more effective than diblocks.<sup>29</sup> However, all compatibilizers were found to trigger finer and more homogenous dispersion of dispersed particles in continuous matrix. Compatibilization promotes the formation of an interlocking structure in PS/PE blends, which allows more equal sharing of imposed stresses and might therefore improve the mechanical properties of the blends.

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Recently, Dow Chemical Company developed ethylene/styrene interpolymers (ESIs), which contain styrene up to 80 wt % and encompass materials ranging from semicrystalline elastomers to amorphous thermoplastics, dependent upon the copolymer styrene content.<sup>31,32</sup> Due to the inherent combination of olefinic and styrenic functionality in the backbone of molecular chains, ESIs show excellent compatibility with styrenic polymers, polyolefins, and a wide variety of other thermoplastics.<sup>23,33,34</sup> However, little information is available on the compatibilization efficiency of ESI on PS-PE blends. This article studies the effect of ESI addition on the mechanical properties and morphology of PS-HDPE polymers blends.

## EXPERIMENTAL

### Materials

PS (code 666D) was obtained from Yanshan Petrochemical, China, and had a density of 1.05 g/cm<sup>3</sup> ( $M_w = 310,000$ ;  $M_n = 87,000$ ;  $M_w/M_n = 3.6$ ). High-density polyethylene (HDPE, code GF7750) was supplied by Liaoyang Petrochemical, China, which had a melt flow rate of 0.3 g/10 min (99.5 mol % ethylene and 57.2% crystallinity). ESI (ESI24) was provided by Phil Island, USA, and had a density of 0.945 g/cm<sup>3</sup> ( $M_w = 240,000$ ; PS% = 29.3 wt %).

### Blends preparation

All polymers were received as pellets and were dried at 80°C overnight in a vacuum oven. The melt blending process was carried out with a twin-screw extruder operated at 210–220°C and 90 rpm. The chemical composition of PS/HDPE blends prepared were 90/10, 80/20, 60/40, 50/50, 40/60, and 20/80. The ternary PS/HDPE/ESI blends were prepared by adding 10 wt % ESI as compatibilizer, with the chemical composition being 80/10/10, 70/20/10, 50/40/10, 40/50/10, 30/60/10, and 10/80/10.

The extrudates exiting from the extruder were palletized and dried at 100°C for 6 h. The dog-bone-shaped tensile bars (ASTM D 638) and notched specimens for Charpy impact tests (ASTM D 256) were obtained on an injection-molding machine (SZ-160/80 NB, China). The barrel temperature profiles were set at 200-210-220°C.

### Measurement of mechanical properties

Tensile tests were carried out on an Instron tensile tester (model 3211) at a crosshead speed of 10 mm/min.

Izod and Charpy impact behaviors were determined with a pendulum impact testing machine (XJ-40A; Wuzhong Material Testing Machine Company, Hebei, China). During impact testing, a load cell in

the tup recorded the force generated in the deformed sample.

All mechanical property tests were performed at (23 ± 1)°C. At least five specimens of each composition were tested and the average value was reported in results.

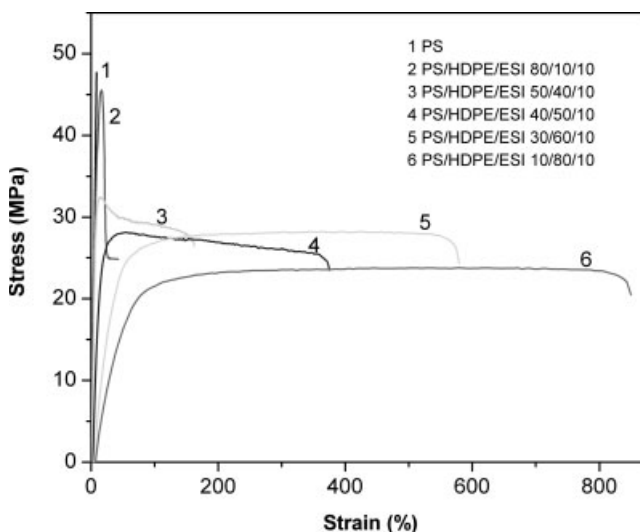
### Morphological observations

To evaluate the fracture mechanism, the fracture surfaces of PS/HDPE/ESI blends were observed with a Cambridge-5250 scanning electron microscope (SEM). Samples were freeze-fractured in liquid nitrogen. The cryogenically fractured surfaces were coated with a thin layer of gold to increase the contrast between the matrix and the dispersed phase in morphology study.

## RESULTS AND DISCUSSION

### Tensile behavior

Figure 1 shows the stress–strain curves of the pure PS and PS/HDPE/ESI blends. Apparently, as a typical brittle polymer, pure PS exhibits only elastic behavior followed by a brittle fracture at low elongation (ca ~ 2.6%). However, with the addition of only 10 wt % ESI, PS/HDPE/ESI blends obtained dramatic improvement in tensile ductility. The tensile behavior of this blend is characterized by the presence of yield point and necking, followed by homogeneous drawing. As illustrated in Figure 1, the stress–strain curve of PS/HDPE/ESI blends with the HDPE content above 60 wt % exhibited the typical behavior of toughened plastics, which presented extensive plastic deformation (exceeding 500% strain) before breaking. It is noted that 10 wt % addition of ESI is quite high.



**Figure 1** Stress–strain curves of pure PS and PS/HDPE/ESI blends with 10 wt % addition of ESI.

**TABLE I**  
**Mechanical Properties of the PS/HDPE Binary Blends and PS/HDPE/ESI Ternary Blends**

PS/HDPE/ESI	Tensile strength (MPa)	Elongation at break ratio (%)	Tensile modulus (MPa)	Izod impact strength (J/m)
100/0/0	37.7 ± 1.02	2.56 ± 0.36	952 ± 32	7.5 ± 0.14
0/100/0	24.5 ± 0.41	500 ± 15	443 ± 8	220.2 ± 3.2
0/0/100 <sup>a</sup>	33.3 ± 2.4	517 ± 28	26.3 ± 1.3	n/a
90/10/0	43.3 ± 1.32	11 ± 1	872.9 ± 28	21.1 ± 0.4
80/20/0	42.8 ± 1.25	17 ± 2	853.5 ± 22	22.4 ± 0.4
60/40/0	41.6 ± 1.16	62 ± 3	810.6 ± 19	23.6 ± 0.3
50/50/0	40.4 ± 1.18	160 ± 6	774.6 ± 20	24.3 ± 0.5
40/60/0	37.5 ± 1.07	286 ± 10	690.9 ± 26	34.6 ± 0.7
20/80/0	35.1 ± 1.06	484 ± 12	640.8 ± 23	45.2 ± 1.1
80/10/10	45.5 ± 1.26	13 ± 1	825.6 ± 28	23.5 ± 0.8
70/20/10	43.1 ± 1.29	62 ± 6	800.2 ± 21	31.3 ± 1.2
50/40/10	35.9 ± 1.06	162 ± 5	683.7 ± 15	123.6 ± 2.9
40/50/10	32.4 ± 1.12	378 ± 11	600.3 ± 16	210.5 ± 5.7
30/60/10	28.2 ± 0.96	588 ± 14	542.1 ± 12	341.8 ± 9.6
10/80/10	23.8 ± 0.88	860 ± 18	472.5 ± 10	705.6 ± 13.1

n/a, not available.

<sup>a</sup> Data are obtained from Ref. 32.

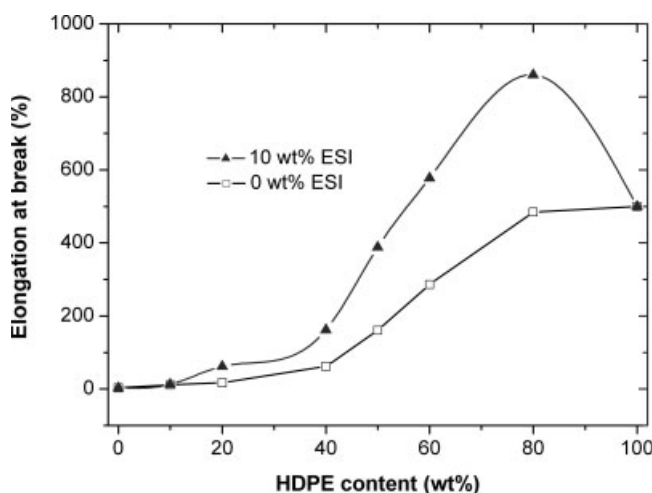
Therefore, only a part of the ESI acted as emulsifier; the other part was dispersed in PS or PE, acting as a rubber toughening agent. Because the ESI content in the PS/HDPE/ESI blends is fixed at 10 wt %, it is considered that the role played by ESI in other blends studied is similar to that of PS/HDPE/ESI 80/10/10 blend. Like PS/HDPE blends toughened by other rubbers (ca SEBS),<sup>17,22,25,26</sup> increasing the HDPE content results in a considerable decrease of yield strength and elastic modulus but an increase in tensile ductility as well as toughness.

The results of tensile strength, elongation, and tensile modulus for PS/HDPE and PS/HDPE/ESI blends are summarized in Table I. Although PS and HDPE are immiscible and incompatible over whole composition ranges, the PS/HDPE binary blends exhibited modified mechanical properties intermediate between PS and HDPE, which present higher toughness but lower modulus in comparison with pure PS. As shown in Table I, with the addition of 10 wt % ESI, the tensile modulus of both PS/HDPE and PS/HDPE/ESI blends tends to decrease continuously with increasing HDPE content. However, the decrease in tensile strength with the addition of HDPE is more obviously observed for PS/HDPE/ESI blends. For instance, with the addition of 80 wt % HDPE, the PS/HDPE only presented 26% loss in tensile strength, whereas the PS/HDPE/ESI lost nearly half tensile strength in comparison with pure PS.

The variation of tensile modulus with HDPE content is similar to that of tensile strength. As shown in Table I, the tensile modulus of PS/HDPE binary blends decreased slowly with HDPE content (ca 19% loss with 50 wt % HDPE), whereas the PS/HDPE/ESI ternary blends displayed a sharp decrease in tensile modulus with increasing HDPE content (ca 37% loss

with 50 wt % HDPE). However, the elongation at break of the blends behaves differently. As illustrated in Figure 2, the elongation of both PS/HDPE and PS/HDPE/ESI blends tends to increase gradually with increasing HDPE content up to 40 wt %, followed by a sharp increase with HDPE content. It is noted that the addition of 10 wt % ESI to the PS/HDPE blends resulted in overall improvement in the elongation at break. Furthermore, the HDPE-rich blends containing HDPE content above 60 wt % exhibited a higher elongation than the HDPE homopolymer.

Based on the results shown in Table I, it is evident that the tensile strength and ductility of PS/HDPE blends exhibit a minimum value smaller than that of either pure component, which is constant with the previous observation.<sup>20</sup> However, the addition of ESI



**Figure 2** Variation of elongation at break with HDPE content for PS/HDPE binary blends (□) and PS/HDPE/ESI ternary blends (▲).

significantly enhances the elongation at break of PS/HDPE blends. This may be due to this interpolymer acting as an efficient emulsifier, leading to strong adhesion between the PS and HDPE phases. Similar observations have also reported for the compatibilizers such as HPB-*b*-PS<sup>18–20</sup> and SEBS,<sup>25,26,35</sup> which effectively enhance the adhesion between the PS and HDPE phases in HDPE-rich blends.

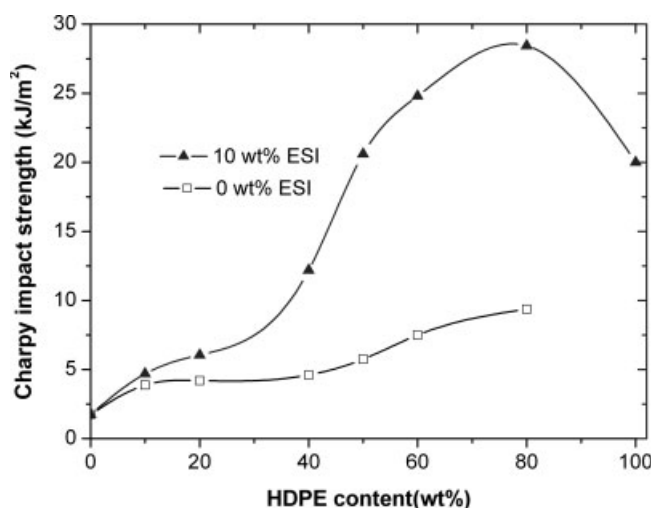
### Impact behavior

The variation of notched Izod impact with HDPE content is shown in Table I, whereas the variation of Charpy impact strength for PS/HDPE binary blends and PS/HDPE/ESI ternary blends is shown in Figure 3. Although PS and HDPE are immiscible and incompatible over the composition ranges studied here, the PS/HDPE binary blends still exhibited some improvement in impact toughness with increasing the content of ductile HDPE. For PS/HDPE blends, their impact strength falls between those of PS and HDPE. As shown in Figure 3, to achieve a fourfold increase in the impact strength of pure PS, HDPE should be added as high as 80 wt %. With the addition of 10 wt % ESI, the PS/HDPE/ESI ternary blends generally presented higher impact properties than the uncompatibilized PS/HDPE blends. Such improvement was even more obvious at HDPE-rich blends. The impact strength of PS/HDPE/ESI can be easily improved by varying HDPE content. In general, the impact strength of the ternary blends increases slowly with HDPE content up to 40 wt %; thereafter, it increases sharply with increasing HDPE content. It is noted that the addition of 40 wt % of HDPE resulted in a sixfold improvement in the Charpy impact strength of pure PS. Moreover, a 15-fold increase in impact strength

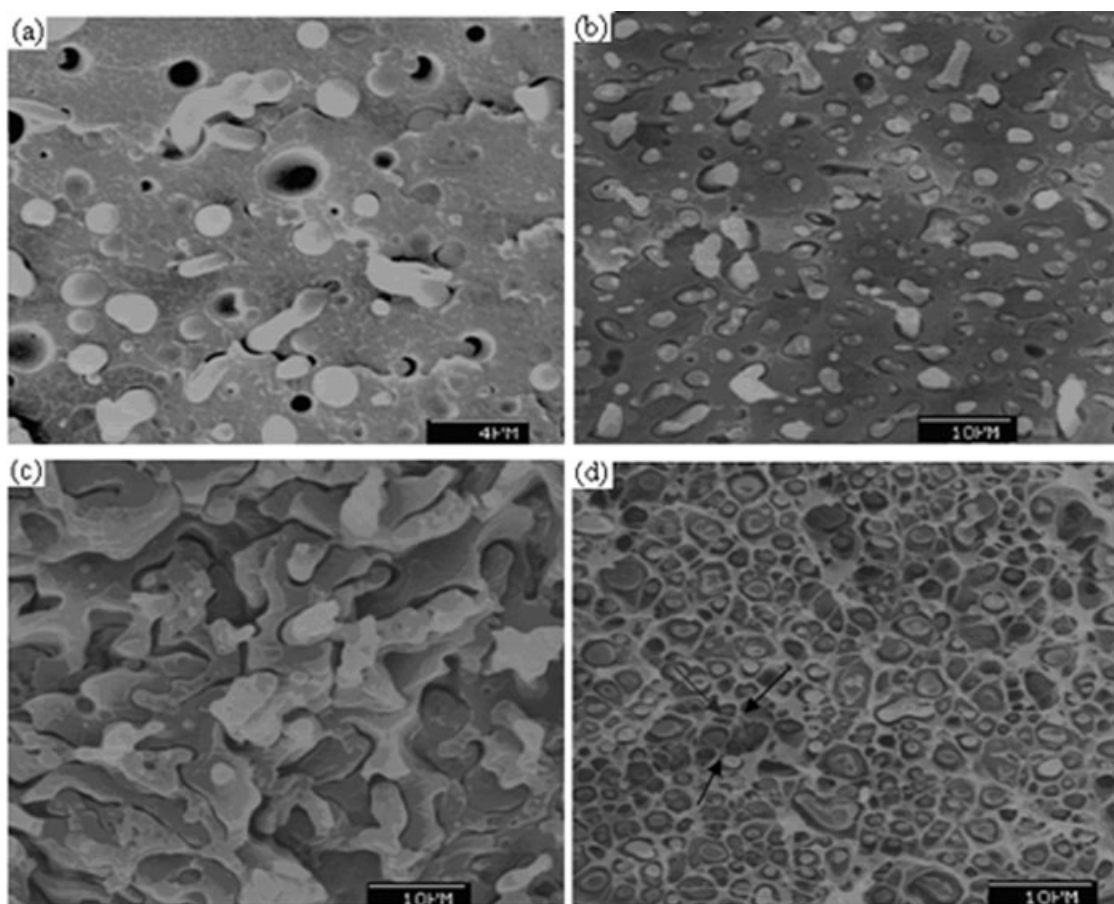
can be obtained with 80 wt % of HDPE, i.e., the impact strength increases from 1.8 to 8.4 kJ/m<sup>2</sup>.

A close examination at the variation of Charpy impact strength with HDPE content, we can find HDPE-rich blends containing 60–80 wt % HDPE presented impact strength higher than that of HDPE homopolymer. This implies that the HDPE was toughened by brittle PS polymer in the presence of ESI compatibilizer. In this case, the HDPE-rich blend is composed of rigid PS particles in highly ductile HDPE matrix. Such behavior has also found in HDPE-rich PS/HDPE/SEBS blends compatibilized by SEBS<sup>26,35</sup> and ductile polymers like polycarbonate toughened by brittle glassy polymers such as poly(methyl methacrylate) and styrene-acrylonitrile.<sup>36–38</sup> The increase in ductility and toughness can be related to the adequate differences in Young's modulus and Poisson's ratio between the brittle particles and polymer matrix, thereby inducing compressive stress acting on the dispersed particles. Consequently, the deformation mechanism in dispersed particles may change from crazing to cold drawing, due to the compressive stress.<sup>36</sup> It is worth noting that the ESI interpolymer played an important role in improving the tensile ductility of HDPE-rich blends. The presence of ESI leads to an increase of the adhesion between the rigid PS particles and HDPE matrix but a decrease in the dispersed phase size. In the latter case, it is known that the size of dispersed particles has a dramatic effect on deformation behavior in ductile matrix/brittle dispersed phase systems. The ductility of such systems generally increases with finer phase dispersions due to the lower resultant stress concentration when a small particle cracks, which was readily proved in the following morphology study.

Interestingly, the impact behavior of ternary blends is similar to that of elongation at break, as shown in Figure 2. However, the PS-rich blends display only small improvement in impact strength (e.g., sixfold increase with 40 wt % HDPE), whereas these blends exhibit a significant higher elongation at break than pure PS (25-fold increase). The low impact strength in the PS-rich blends may be due to the high strain rate associated with the impact test and to the plane-strain condition existing near to notch of impact specimens. However, in the tensile test, the plane-strain condition prevails, therefore producing higher ductility values. Tjong and Xu<sup>26</sup> reported the similar impact properties for the PS/HDPE blends compatibilized by 10 wt % SEBS, where impact strength increased significantly after a slow improvement up to 50 wt % HDPE and elongation at break exhibited a higher increase than impact energy when increasing the content of HDPE. Since the ESI used contained 70.7 wt % of ethylene, it shows higher compatibilization toward HDPE than PS. It is evident that the interfacial adhesion between PS and HDPE can be significantly improved with the



**Figure 3** Variation of Charpy impact strength with HDPE content for PS/HDPE binary blends (□) and PS/HDPE/ESI ternary blends (▲).



**Figure 4** SEM micrographs showing the cryogenically fractured surfaces of (a) PS/ESI 90/10 blend; (b) PS/HDPE/ESI 80/10/10 blend; (c) PS/HDPE/ESI 60/30/10 blend; and (d) PS/HDPE/ESI 50/40/10 blend.

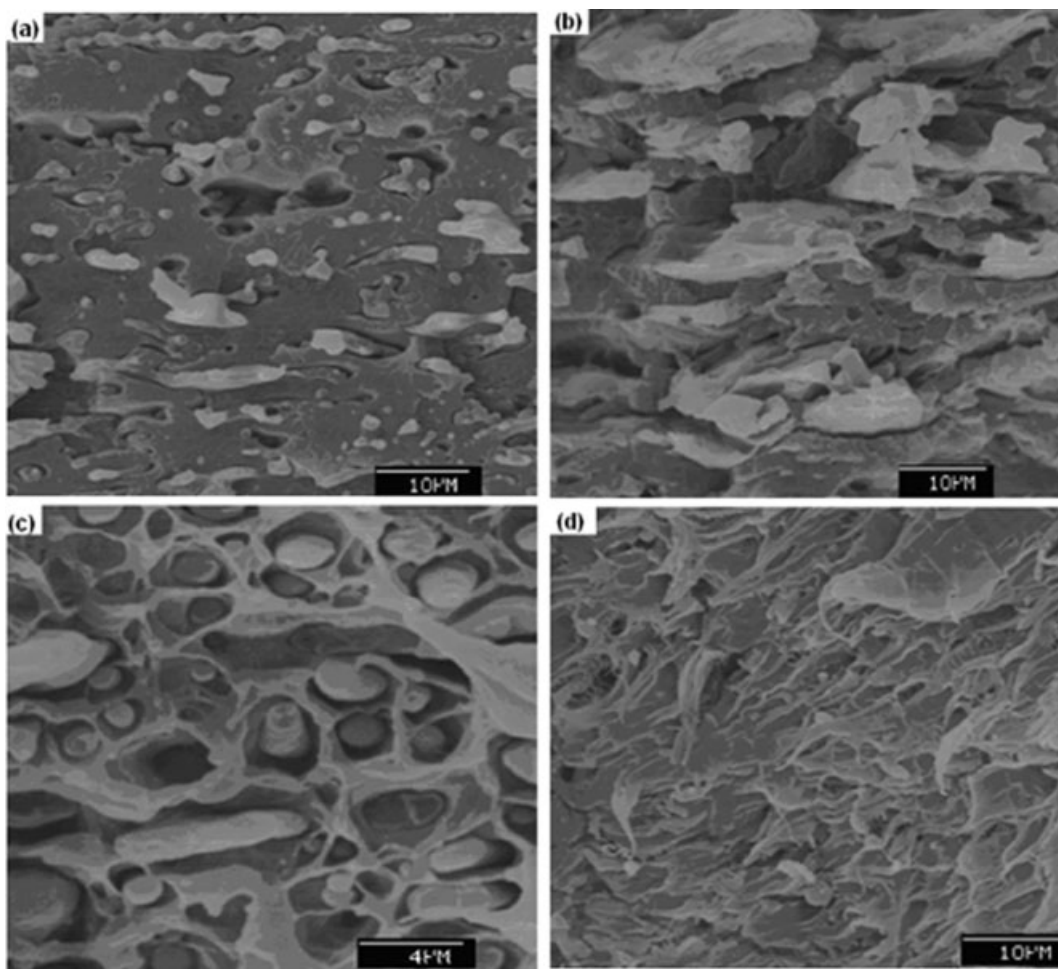
addition of ESI in HDPE-rich blends, compared with PS-rich ones. Therefore, the impact toughness is greatly improved in HDPE-rich blends.

### Morphology

In the SEM image of the cryogenically fractured surfaces of PS/ESI (90/10) blend shown in Figure 4(a), a rough surface with typical matrix-droplet structure could be observed. PS was the matrix whereas ESI is droplet. From the obscure interface between the phases, it can conclude the PS is partially compatible with ESI. A large amount of ESI is dispersed as particles in PS matrix, with a size distribution ranging from 0.5 to 1.5  $\mu\text{m}$ . While some ESI dispersed in PS act as a rubber toughening agent, which can be justified by the presence of ESI fibrillar structure. As the HDPE content increased from 10 to 40 wt %, the SEM micrograph changed from typical matrix-droplet structure [Fig. 4(b)] to cocontinuous or interlocking structure, with PS as the continuous phase [Fig. 4(c,d)]. In these cases, brittle fracture occurred when subject to impact, which can be indicated by the smooth-fractured surfaces. As shown in Figure 4(c), the PS/HDPE/ESI 50/40/10 fractured surface presented much finer disper-

sion. Numerous HDPE particles were observed with dimension of 0.5–5  $\mu\text{m}$  and were covered by a relatively thin ESI layer. In addition, tiny ESI particles were observed incorporated in the HDPE particles. Besides, part of the ESI acted as emulsifier, which exists in the interface of PS and HDPE and reduced the interfacial tension of the phases, which can be justified by the obscure interface between PS and HDPE.

Figure 5(a) shows the SEM fractograph of PS/HDPE/ESI 80/10/10 bend at slow crack-growth region. The fracture appears quite smooth after tensile tests. HDPE particles dispersed in matrix with wide-size distribution (0.5–10  $\mu\text{m}$ ) and some underwent some plastic deformation. In addition, tiny ESI particles were observed incorporated in the PS matrix. As the HDPE content is increased to 30 or 40 wt %, the SEM fractograph shows a much rougher surface morphology [Fig. 5(b,c)]. HDPE particles are adhered strongly to the matrix and underwent large plastic deformation. It is likely that the dispersed HDPE particles act as stress concentrators during the tensile process, thereby initiating crazes in the PS matrix. Further increasing HDPE content to 60 wt % and above results in extensive fibrillation of the matrix phase; it is evident that drawing of the cocontinuous structure

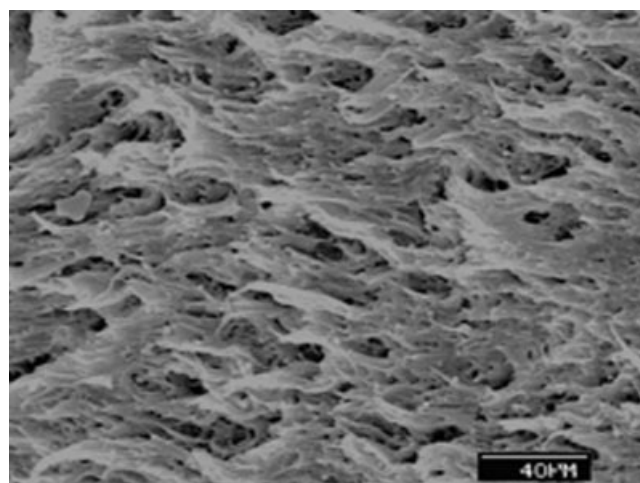


**Figure 5** SEM micrographs showing the fractured surface features of the slow crack-growth region of (a) PS/ESI 80/10/10 blend; (b) PS/HDPE/ESI 60/30/10 blend; (c) PS/HDPE/ESI 50/40/10 blend; and (d) PS/HDPE/ESI 30/60/10 blend.

begins to occur. In this respect, the impact strength appears to increase sharply [Fig. 5(d)]. As shear deformation dissipates a large amount of energy, the elongation at break of the PS/HDPE/SEBS blends begins to rise sharply when the HDPE content reaches 50 wt %. According to percolation model proposed by Wu,<sup>39</sup> the tough-brittle transition of our current PS/HDPE/ESI system occurs when the interparticle distance  $d_c$  reaches  $\sim 2 \mu\text{m}$  [Fig. 5(c)]. For PS/HDPE/SEBS systems, it was previously reported that crazing of the PS matrix is the main toughening mechanism for the PS-rich blends, whereas shear yielding of the matrix is the main deformation mode for the HDPE-rich blends.<sup>40,41</sup> The SEM fractography of the PS/HDPE/ESI blends in the present work is similar to that of the PS/HDPE/SEBS blends.

Figure 6 shows the SEM fractograph of the slow crack-growth region of the HDPE-rich blend containing 80 wt % HDPE. It can be seen that extensive fibrillation of the HDPE matrix occurs in the slow crack-growth region. Moreover, a few small cavities (dimensions of 1–20  $\mu\text{m}$ ) can also be observed in the fractograph. Such cavitation originates from debonding of

the ESI-encapsulated PS particles from the HDPE matrix during impact deformation. As the size of dispersed PS particles is 1  $\mu\text{m}$ , the larger voids observed in this fractograph can be related to the coalescence of



**Figure 6** SEM micrographs showing the fractured surfaces of PS/HDPE/ESI 10/80/10 blend.

individual voids initiated by the dispersed PS particles. Furthermore, ductile tearing of the ligaments between particles can lead to the formation of fibrils once the voids are initiated. As shear yielding of the ligaments between particles dissipates a large amount of the energy the PS/HDPE/ESI 10/80/10 blend exhibits excellent impact toughness. This phenomenon is analogous to the shear yielding of thermoplastic matrix induced by fine rubber particles during impact deformation.<sup>25,26</sup>

### CONCLUSIONS

PS/HDPE blends, and PS/HDPE blends compatibilized with ESI interpolymer were prepared by extrusion. The tensile and impact properties of both PS/HDPE blends and ESI-compatibilized PS/HDPE blends were investigated. Tensile tests showed that the yield stress of the PS/HDPE/ESI decreases substantially with increasing HDPE content but the elongation at break appeared to increase dramatically with increasing HDPE content. The improvement in tensile ductility in the PS-rich blends was caused by the crazing of the PS matrix, whereas the increase in elongation in the HDPE-rich blends resulted from the shear yielding of the HDPE matrix. SEM observations revealed that ESI addition leads to a finer dispersion of either PS or HDPE particles, and to a stronger adhesion between these two phases. Impact measurements indicated that the impact strength of the blends increases slowly with HDPE content up to 40 wt %, followed by a significant increase with increasing HDPE content. The impact strength of the HDPE-rich blends exceeded that of the HDPE polymer. This implies that ductile HDPE can be further toughened by the incorporation of a minor PS phase in the presence of ESI interpolymer. Such behavior can be related to extensive fibrillation of the HDPE matrix associated with debonding of the PS particles from the matrix.

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