Interfacial Design of the Nonpolar Polyolefin Ternary Composite with High Strength, High Toughness, and High Modulus

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SYNPOSIS

An interfacial model was proposed for the ternary thermoplastics (matrix)/elastomer/rigid-particle filler composite with high strength, high toughness, and high modulus. A dispersed phase of rigid particle as a core and elastomer as a shell that has a good interfacial adhesion with the matrix is the key point of the model. A composite with high strength, high toughness, and high modulus was obtained in the styrene (ST) and maleic anhydride (MAH) modified high-density polyethylene (HDPE)/ethylene-propylene-diene monomer (EPDM) rubber/carbon black (CB) with ditertiary butyl peroxide (DTBP) as the initiator through the reactive extrusion. The electrical resistivity measurement showed that CB of the unmodified composites distributed at the interface of the HDPE and EPDM, while that of the modified composites distributed mainly in the EPDM phase. The morphology of the ternary composite was consistent with the wetting coefficient analysis. That the mechanical properties of the γ-ray–irradiated unmodified composites were not as good as those of the modified composites further indicated that the mechanical properties of the composite could not be improved significantly purely by introducing the interfacial adhesion and matrix crosslinking without forming the proposed dispersed phase structure. SEM observation supported the conclusion that the different phase structures are the major reason that leads to the different toughness. © 1996 John Wiley & Sons, Inc.

INTRODUCTION

Recently, rigid-particle fillers have been extensively used in creating a polymer composite with high strength, high modulus, and low shrinkage.[1,2] However, the decreased toughness generally results for the composite.[3] On the other hand, elastomers are generally employed in increasing the polymer toughness, but generally result in the deteriorated stiffness.[4-10] In a word, two-phase composites exhibit only a partial improvement over the matrix material alone, because some selected properties are enhanced at the expense of others.

In our earlier work, a kaolin-toughened polypropylene[11] and polyamide 6[12] composite was obtained. In the case of kaolin-toughened polypropylene, an elastic interlayer that had a good interfacial adhesion with the kaolin was inserted between the filler and matrix; therefore, the toughness of the composite was increased significantly. But due to the lack of interfacial adhesion between the interlayer and the matrix, the strength of the composite was not improved. In the case of the kaolin-toughened polyamide 6, the interfacial adhesion both between the filler and interlayer and between the interlayer and matrix was good due to the presence of functional group in both the filler and the matrix. A polyamide 6 composite with high strength, high toughness, and high modulus was obtained. The functionalization of the matrix was not present in the nonpolar polypropylene. As a result, the above interfacial

modification method could not improve both the strength and toughness for the nonpolar polyolefin composite.

On the other hand, the nonpolar polyolefin thermoplastics (matrix)/elastomer/filler systems have been investigated recently in expectation of obtaining a composite with high strength, high toughness, and high modulus.[13-26] But the possible presence of matrix/filler, matrix/rubber, and rubber/filler interfaces in the ternary composite results in the complexity of phase structure[16] and corresponding variation of the composite properties. There exist two major kinds of dispersed phase structure, i.e., with filler, elastomer separately dispersed in the matrix, and with filler as a core, elastomer as a shell. In three of the articles concerning the mechanical properties of the ternary systems[13,17,26] opposite opinions were held with regards to whether the separate dispersed phase or the core/shell dispersed phase was beneficial to the composite toughness. Theoretical and experimental studies[27-32] have indicated that in filler core/elastomer shell polymer composites the properties strongly depend on three factors: (a) the thickness of the elastomer shell, (b) the properties of the material in this phase, (c) the interfacial adhesion of the various phases. According to the theoretical analysis of Matonis et al.,[28] it is doubtful that a mixture of a separately dispersed phase of filler and elastomer, exhibiting two different and distinct responses to the applied load (or deformation), could result in a composite with desirable properties. On the contrary, the encapsulation method is suggested. The proposed model by Matonis et al. is a polyblend of spherical high modulus inclusions encapsulated within a uniform layer of low modulus elastomer and dispersed throughout a suitable matrix. This hypothetical three-phase ordered composite will permit design of a new material both stiffer and tougher than the matrix phase alone. The contribution of the rigid phase inclusion to the overall modulus will depend on the ability of the soft phase shell to transmit load from matrix to inclusion, which is further dependent on the interfacial adhesion of the matrix/elastomer and elastomer/filler.

Based on the above theoretical analysis, a filler core/elastomer shell-dispersed phase structure is essential for the improvement of composite toughness; an excellent interfacial adhesion is essential for the improvement of composite strength and modulus. This is the key point of the interfacial design of a composite with high strength, high toughness, and high modulus. The schematic representation of the proposed model is shown in Figure 1.
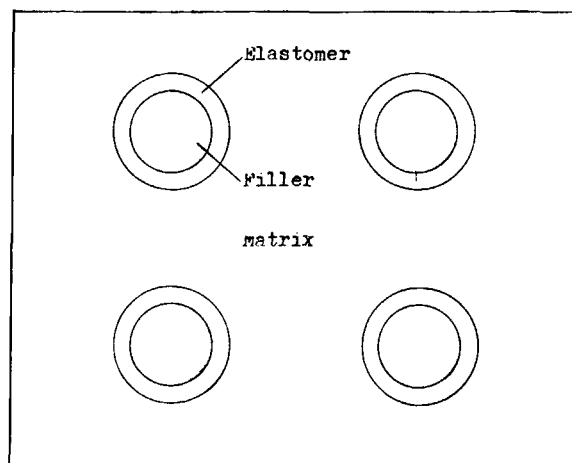
Figure 1 Schematic representation of the proposed model.

Due to the lack of functional group in the nonpolar polyolefin material, commonly used coupling agents could not assure an excellent interfacial adhesion between the matrix and other phases. Moreover, if nonpolar EPDM is used as the elastomer, still more problems result. Although the functionalized material of MAH grafted EPDM and HDPE[13,26] were used in the ternary systems, the effect was not significant. In our opinion, recently developed reactive extrusion technique[33-36] may be the answer to the improved interfacial adhesion.

In this article, an interfacial modifier (IM) ST and MAH-modified ternary composite of HDPE/EPDM/CB with high strength, high toughness, and high modulus was obtained through the reactive extrusion. Electrical resistivity and surface tension measurements were used to characterize the composites. The γ-ray–irradiated unmodified composites were used to compare with the modified composites in order to clarify the function of pure crosslinking and interfacial adhesion. SEM observation is used to explain the toughening mechanism of the modified ternary systems.

EXPERIMENTAL

Materials

High-density polyethylene (HDPE, MI = 4.8), ethylene-propylene-diene monomer (EPDM) rubber were used in this investigation. The filler used was carbon black, with specific surface area 74.7 m^2/g. The interfacial modifier used was styrene (ST) and maleic anhydride (MAH). The initiator used in the

Table I Tensile Strengths of the HDPE/CB and HDPE/EPDM/CB Composites (MPa)

CB wt.	Unmodified HDPE/CB	Modified HDPE/CB	Unmodified HDPE/EPDM/CB	Modified HDPE/EPDM/CB
0	28.6 (±0.4)	30.3 (±0.0)	19.6 (±0.5)	25.1 (±0.1)
5	29.3 (±0.6)	32.5 (±0.3)	24.6 (±0.3)	27.0 (±0.2)
10	31.8 (±0.2)	35.1 (±0.6)	24.9 (±0.1)	28.3 (±0.5)
15	33.8 (±0.7)	38.8 (±0.3)	26.9 (±0.5)	33.3 (±0.3)
20	35.6 (±0.4)	42.9 (±0.5)	28.1 (±0.4)	34.8 (±0.2)
30	37.0 (±0.5)	48.8 (±0.8)	30.9 (±0.9)	43.1 (±0.6)

For binary systems, HDPE = 100 wt.; for ternary systems, HDPE = 90 wt., EPDM = 10 wt.

reactive extrusion was ditertiary butyl peroxide (DTBP).

Preparation of Composites

The MAH and DTBP were first dissolved in the ST. Then the CB was pretreated with the solution. The binary HDPE and ternary HDPE/EPDM composites filled with untreated and treated CB were all extruded and granulated on a Brabender twin-screw extruder (35 mm Φ) at 30 rpm. The barrel temperatures were 210–220°C.

Mechanical Property Testing

The tensile strengths and flexural moduli of the composites were measured on an Instron 1122 at a crosshead rate of 50 mm/min and 2 mm/min, respectively. Izod impact strengths were measured on an XJ-40A Impact Tester.

Electrical Resistivity Measurement

The electrical resistivity was measured through the vertical thickness of the cylindrical composite specimen (diameter = 100 mm, thickness = 2 mm). Silver paste was employed to ensure good contact between the electrodes of the resistance tester and the specimen surface. High resistivity specimens were measured using a RP2680 High Resistance Tester. Low resistivity specimens were measured using a QJ23 DC Wheatstone Bridge Tester.

Surface Tension Measurement

The surface tensions were measured on a FACE Contact Angle Tester at 25°C. The surface tensions of the HDPE and EPDM were measured by single-liquid method with the reference liquids water and ethylene glycol.[37,38] The surface tensions of the un-

treated and treated CB were measured by double-liquid method[39] according to the Young–Duprc equation and Fowkes theory[40] through the measurement of contact angle of water drop on the filler in the n-hexane, n-heptane, n-octane, and n-decane. The surface tensions were all extrapolated to the processing temperature 215°C from the room temperature.[41,42]

γ-Ray Irradiation

The unmodified polymer composites were irradiated with γ-ray using a ^{60}Co source. The irradiation was completed until the gel contents of the unmodified composites were equal to those of the modified ones.

SEM Observation of the Toughening Mechanism

The impact fractured surfaces of the composites were characterized by the scanning electron microscopy (SEM). All observations were carried out on a Hitachi S-530 SEM.

RESULTS AND DISCUSSION

Mechanical Properties

The mechanical properties of the HDPE/CB and HDPE/EPDM/CB composites are listed in Tables I, II and III. From the tables it could be seen that although the tensile strengths and flexural moduli of the composites are increased by filling the matrix with CB, a marked decrease is also observed for the impact strengths. Although the mechanical properties of the modified HDPE/CB are generally better than those of the unmodified ones, the impact strengths are still less than that of the matrix. On the other hand, although incorporation of EPDM into the matrix results in a higher impact strength, there is a significant decrease in the tensile strength.

Table II   Impact Strengths of the HDPE/CB and HDPE/EPDM/CB Composites (J/m)

CB wt.	Unmodified HDPE/CB	Modified HDPE/CB	Unmodified HDPE/EPDM/CB	Modified HDPE/EPDM/CB
0	115 (±8.2)	163 (±5.6)	570 (±12.3)	603 (±7.2)
5	85 (±4.3)	95 (±2.6)	293 (±6.5)	345 (±11.3)
10	55 (±2.9)	73 (±5.1)	188 (±4.8)	500 (±13.5)
15	30 (±1.4)	73 (±8.2)	133 (±0.0)	360 (±7.4)
20	20 (±5.2)	70 (±5.3)	110 (±5.8)	395 (±12.5)
30	18 (±3.6)	40 (±4.9)	50 (±0.0)	338 (±5.4)

For binary systems, HDPE = 100 wt.; for ternary systems, HDPE = 90 wt., EPDM = 10 wt.

Introduction of interfacial modifier leads to a slight increase of the mechanical properties, but its tensile strength is still less than that of the matrix. The above results are in agreement with much experimental work for the binary systems in the literatures, which indicates that a composite with both high strength and high toughness could not be produced simply by introduction of filler or elastomer. For the unmodified ternary HDPE/EPDM/CB composites, when compared with the matrix, the tensile strength decreases slightly and the impact strength increases moderately at lower CB content, while the tensile strength increases slightly and the impact strength decreases significantly at higher CB content. The above results show that a composite with both high strength and high toughness is not obtained as expected of the ternary system. For the modified ternary systems, at lower CB content, the tensile strength is comparable with that of the matrix and the impact strength is much higher than those of the matrix and the unmodified composites. At higher CB content, a significant increase is observed for the tensile strength while the impact strength still remains comparatively high. Comparing the mechanical properties of the modified ternary composite of 30 wt CB with those of the matrix, it could be clearly seen that a composite with high

strength, high toughness, and high modulus is successfully prepared. In our earlier work[43] it has been shown that the crystallization behavior of the matrix is only slightly changed and the DMA results indicate a different phase structure for the unmodified and modified composites. A separate dispersion of the filler, elastomer, and a filler core/elastomer shell dispersed phase structure are expected for the unmodified and modified composites, respectively. The lower flexural moduli of the modified ternary systems compared with those of the unmodified composites is an indication of the structural difference.

Electrical Resistivity

Carbon black has always been a kind of conductive filler to be introduced into the insulating polymers in expectation of obtaining a conductive composite.[44-51] It is known that the electrical conductivity of insulating polymers filled with conductive particles such as metal and carbon black, discontinuously increases at a certain content of the filler named percolation threshold.[50,52-57] In one article by Sumita et al.,[51] dispersion of carbon black and electrical conductivity of polymer blends were discussed. There are two types of heterogeneous distribution of carbon black in filled polymer blends. One is pre-

Table III   Flexural Moduli of the HDPE/CB and HDPE/EPDM/CB Composites (MPa)

CB wt.	Unmodified HDPE	Modified HDPE/CB	Unmodified HDPE/EPDM/CB	Modified HDPE/EPDM/CB
0	576 (±22)	—	599 (±19)	—
5	805 (±35)	974 (±19)	852 (±21)	727 (±12)
10	986 (±37)	990 (±25)	882 (±48)	732 (±25)
15	1086 (±15)	1085 (±28)	936 (±32)	799 (±15)
20	1196 (±26)	1256 (±18)	1047 (±15)	828 (±31)
30	1162 (±39)	1542 (±51)	1121 (±42)	969 (±38)

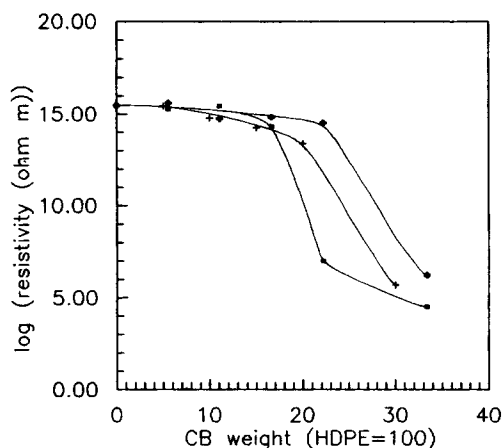For binary systems, HDPE = 100 wt.; for ternary systems, HDPE = 90 wt., EPDM = 10 wt.

Figure 2 Electrical resistivity of HDPE/CB, unmodified and modified HDPE/EPDM/CB composites as a function of CB weight in HDPE phase (HDPE = 100 wt). + HDPE/CB; ● unmodified HDPE/EPDM/CB; □ modified HDPE/EPDM/CB.

dominantly distributed in one phase of the blend, and the other is distributed concentratedly at the interface of two polymers. If carbon black is distributed at the interface, the envelope formation of CB particles around the dispersed phase makes the conductive path more effective than the single matrix. On the other hand, if carbon black is distributed in one phase of the blend, the electrical conductivity of CB filled polymer blends is determined by two factors. One is the concentration of CB in the filler rich phase, and the other is the structural continuity of this phase. The conclusion was confirmed by the CB filled PMMA/PP, HDPE/PMMA, and HDPE/PP blends. The electrical resistivity of the binary and ternary composites vs CB weight is plotted in Figure 2, where the CB weight is calculated in HDPE phase. Figure 2 shows that the percolation threshold of the unmodified HDPE/EPDM/CB composites is less than that of the HDPE/CB composites, which means that the conductive path of the HDPE/ EPDM/CB composites is more effective than that of the HDPE/CB composites. This indicates that CB particles distribute mainly at the interface of the EPDM and HDPE phases. Thus, the electrical resistivity characterization further clarifies the concrete pictures of the separate dispersion of filler and elastomer that are not revealed by the DMA results.[43] On the other hand, the percolation threshold of the modified HDPE/EPDM/CB composites occurs at a larger filler content than that of the HDPE/ CB composites. If CB particles distribute mainly in the HDPE phase, its percolation behavior will be close to that of the HDPE/CB composites. If CB

particles distribute mainly at the interface, its percolation behavior will approach that of the unmodified HDPE/EPDM/CB composites. The only reasonable explanation for the retardation of the percolation threshold of the modified HDPE/EPDM/ CB composites is that CB particles distribute mainly in the EPDM phase. This is consistent with the DMA results.[43] Therefore, the percolation threshold of modified HDPE/EPDM/CB composites depends on the CB concentration in the EPDM and the structural continuity of EPDM. The occurrence of the percolation threshold in the modified ternary systems may be explained by a nonequilibrium thermodynamical model proposed by Wessling.[57-59] In this case, the adsorbed layers are EPDM.

Surface Tension Characterization of the Morphology of the Composites

Interfacial forces have been found to be one of the most important factors in determining the morphology of the polymer blends and composites if the viscosities of the polymers are comparable.[41,51] In order to further confirm the morphology of the ternary composites characterized by the DMA and electrical resistivity, surface tension is measured for the various material at 25°C, which is listed in Table IV. In the table the γ_s^d and γ_s^p are dispersion and polar components, respectively. From the Table IV it could be seen that the surface tensions of untreated CB and HDPE measured from the present methods are comparable with those from references 42 and 51, which demonstrates the validity of the characterization methods. Because the processing temperature is 215°C, variation of the polymer surface tension with the temperature cannot be neglected. The values of γ at 215°C are obtained by using the relation: $-d\gamma/dT = 0.057$, which is a general value for PE and PP.[42] The calculated values are listed in Table V. The harmonic mean average equation is employed in calculating the interfacial tension.[38,41,42,51]

Table IV Surface Tension at 25°C (dyne/cm)

	γ_s	γ_s^d	γ_s^p
CB (Untreated)	57.0	51.0	6.0
CB (Treated)	38.9	26.3	12.6
HDPE	36.2	36.2	0.0
EPDM	32.6	29.5	3.1

Table V Surface Tension at 215°C (dyne/cm)

	γ_s	γ_s^d	γ_s^p
CB (Untreated)	57.0	51.0	6.0
CB (Treated)	38.9	26.3	12.6
HDPE	25.4	25.4	0.0
EPDM	21.8	19.7	2.1

$$\gamma_{12} = \gamma_1 + \gamma_2 - 4\left[\frac{\gamma_1^d\gamma_2^d}{\gamma_1^d + \gamma_2^d} + \frac{\gamma_1^p\gamma_2^p}{\gamma_1^p + \gamma_2^p}\right] \quad (1)$$

The calculated interfacial tensions are listed in Table VI. The spreading coefficient[41] and wetting coefficient[51] have been successfully correlated with the morphology of polymer blends and composites. In fact, they are equivalent. The wetting coefficient is defined as follows:[51]

$$\omega_A = \frac{\gamma_{CB-B} - \gamma_{CB-A}}{\gamma_{A-B}} \quad (2)$$

when $\omega_A > 1$, CB particles distribute within the A phase; When $\omega_A < -1$, CB particles distribute within the B phase; when $-1 < \omega_A < 1$, CB particles distribute at the interface.

The calculated value are listed in Table VII. It could be seen that CB of the unmodified HDPE/EPDM/CB composites should distribute at the interface between the HDPE and EPDM. And from the wetting coefficient, it could be concluded that most part of a CB particle distributes in the HDPE phase, while CB of the modified composites should distribute in the EPDM phase. The conclusion is in agreement with that from the electrical resistivity analysis.

Mechanical Properties of the Unmodified Composites after γ-Ray Irradiation

Since the introduction of a little peroxide initiator may lead to the crosslinking of the matrix and

Table VI Interfacial Tension at 215°C (dyne/cm)

	γ_{12}
CB (Untreated) – HDPE	14.6
CB (Untreated) – EPDM	15.7
HDPE – EPDM	2.8
CB (Treated) – HDPE	12.6
CB (Treated) – EPDM	8.4
HDPE – EPDM	2.8

Table VII Wetting Coefficient at 215°C (A phase is HDPE, B phase is EPDM)

	ω_A	CB Distribution
CB (Untreated)	0.39	Interface
CB (Treated)	-1.5	EPDM Phase

EPDM. At the same time, the interfacial adhesion between the various phases is better compared with that of the unmodified composites. Thus, it should be worth investigating the pure effect of crosslinking of the matrix, EPDM, and improved interfacial adhesion between the various phases in order to clarify the contribution of the morphological difference between the unmodified and modified composites. The γ-ray irradiation is completed until the gel contents, which are measured by the insoluble part of the composites from the toluene reflux in a Soxhlet's extractor, between the irradiated and modified composites are equal. The measured mechanical properties of the irradiated composites are listed in Tables VIII and IX. For both the binary and ternary composites, at lower CB content, the tensile strengths of the γ-ray–irradiated composites are higher than those of the modified composites, while at higher CB content, the opposite trend is observed. The impact strength is a more reliable parameter of the morphology of the composites because the deterioration effect of the CB particles are significant when they have direct contact with the thermoplastics matrix. From Table IX it could be seen that the crosslinking of matrix, EPDM, and interfacial adhesion could not offset the deterioration effect of the CB particles, especially when the CB effect is significant; that is, when the CB content is high. This indicates that the morphological structure is one of the most important factors in determining the mechanical properties of the end products. The above results again support the conclusion

Table VIII The Tensile Strengths of the γ-Ray Irradiated Unmodified HDPE/CB and HDPE/EPDM/CB Composities (MPa)

CB wt.	HDPE/CB	HDPE/EPDM/CB
0	32.5 (±0.5)	25.1 (±0.3)
10	38.8 (±0.1)	31.5 (±0.4)
20	40.0 (±0.8)	32.5 (±1.0)
30	39.4 (±0.2)	37.8 (±0.2)

For binary systems, HDPE = 100 wt.; for ternary systems, HDPE = 90 wt., EPDM = 10 wt.

Table IX   The Impact Strengths of the γ-Ray Irradiated Unmodified HDPE/CB and HDPE/EPDM/CB Composites (J/m)

CB wt.	HDPE/CB	HDPE/EPDM/CM
0	175 (±5.1)	700 (±10.5)
10	70 (±3.5)	413 (±8.2)
20	25 (±2.2)	250 (±7.9)
30	25 (±2.8)	130 (±2.5)

For binary systems, HDPE = 100 wt.; for ternary systems, HDPE = 90 wt., EPDM = 10 wt.

that the dispersed phase structure of the unmodified composites has a lot of CB particles to contact with the thermoplastics matrix. This is in agreement with several other characterization results.

Toughening and Strengthening Mechanism of the Modified Ternary Composites

SEM micrographs of impact fractured surface of HDPE/EPDM/CB composites were shown in Figure 3. From Figure 3 it can be seen that although there are all existing shear yielding bands in the unmodified and modified ternary composites, the cavitation is obvious in the unmodified ones.

The shear yielding band is formed in both the unmodified and modified composites due to the presence of elastomer in the thermoplastics matrix. But in the unmodified composites, due to the direct contact of particulate filler and matrix, the cavitation is significant and becomes an important way of dissipating the energy. This can account for the low impact strengths of the unmodified composites. On the contrary, due to no direct contact of particulate filler and matrix, the shear yielding band is the major way of dissipating the energy and, consequently, the impact strength increases significantly. For the unmodified composites, due to the lack of interfacial adhesion between the various phases and the deterioration effect of elastomer, their tensile strengths are low. For the modified composites, the interfacial adhesion between the CB, EPDM, and HDPE is easily improved by the peroxide initiator because of their easy production of free radicals. Due to the core/shell dispersed phase structure and excellent interfacial adhesion between various phases introduced by the reactive extrusion, the stress can be easily transmitted from the matrix to the filler through the elastomer phase. Consequently, the tensile strengths of the modified composites are increased.

CONCLUSIONS

An interfacial model has been proposed for the nonpolar polyolefin ternary composite with high strength, high toughness, and high modulus. The key point of the model is a filler core/elastomer shell-dispersed phase structure and an excellent interfacial adhesion between the various phases.

Based on this, a ternary HDPE/EPDM/CB composite with high strength, high toughness, and high modulus is successfully prepared.

Various characterization techniques have indicated that the different phase structures and differ-
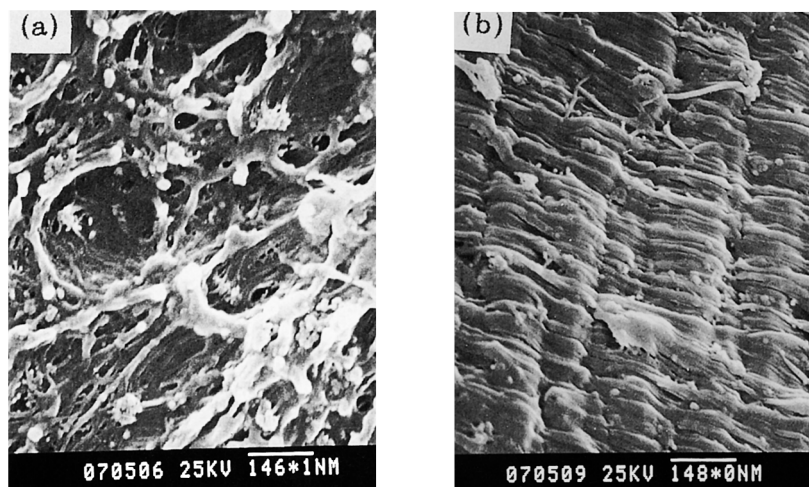

Figure 3   SEM micrographs of the impact fractured surfaces of (a) unmodified and (b) modified HDPE/EPDM/CB composites.

ent interfacial adhesions are the two major reasons that lead to the different mechanical properties. This is in agreement with the proposed model.

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