

Mechanical Properties of High-Density Polyethylene/Scrap Rubber Powder Composites Modified with Ethylene–Propylene–Diene Terpolymer, Dicumyl Peroxide, and Silicone Oil

Yan Li, Yong Zhang, Yinxi Zhang

Research Institute of Polymer Materials, Shanghai Jiao Tong University, Shanghai 200240, China

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ABSTRACT: The effect of ethylene–propylene–diene terpolymer (EPDM), dicumyl peroxide (DCP), and dimethyl silicone oil on the mechanical properties of high-density polyethylene (HDPE) composites filled with 60 mesh cryogenically scrap rubber powder (SRP) was studied. The addition of 10 wt % EPDM, 0.2 wt % DCP, and 4 wt % dimethyl silicone oil significantly increased both the impact strength and elongation at break of the HDPE/SRP composites. After the modification, the impact strength increased by 160%, and the elongation at break increased by 150% for the composites containing 40 wt % SRP. The impact load–time

curves showed that the increase of impact energy for the modified composites was attributed to the increase of the maximum force at yield point and the ductile deformation after yielding. The rheological behavior, dynamic mechanical properties, and morphology observation suggested that an enhanced adhesion between SRP and polymer matrix formed in the modified HDPE/SRP composites. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 88: 2020–2027, 2003

Key words: scrap rubber powder; high-density polyethylene; modification; mechanical properties; composites

INTRODUCTION

The increasing number of used tires is becoming a disturbing problem in most countries. It was reported that 240 million used tires in the United States, 40 million in the UK, and 10 million in Australia were occurring annually.¹ Worn rubber tires and other rubber goods such as rubber tubes can no longer be easily reclaimed because they have already been vulcanized and are not biodegradable. In recent years a number of approaches have been proposed to use such a large resource: combustion for fuel (often in cement kilns or paper mills); pyrolysis to recover gas, oil, char, and other chemical components; landfills or monofills; incorporating in asphalt to produce road surfaces; and preparation of athletic/recreational surfaces.² However, some of the approaches to use waste rubber offer limited consumptions compared with sufficient quantities of waste tires generated every year and bring about serious environmental and economic problems. For instance, tires discarded in landfills tend to float on top, causing mosquito breeding and damaging the surface layers of clay.² Most municipalities in the United States no longer permit the inclusion of tires in regular landfills.³ An application that has potential

both to be economical and to utilize large volumes of tires is to shred and grind the tire to scrap rubber powder (SRP) and then to compound SRP with polymer composites.

SRP to be used as a kind of filler in thermoplastic polymer composites has been under consideration for some time.⁴ The use of SRP in thermoplastic polymer matrices retains processability and end-use properties. Among the various thermoplastic polymer matrices in SRP composites, polyethylene (PE) is the most common.^{5,6} Other polymer matrices for making composites include polypropylene (PP), poly(vinyl chloride) (PVC), polystyrene (PS), ethylene–vinyl acetate copolymer (EVA), and thermoplastic elastomers.^{7–10} Deanin and Hashmiolya¹⁰ found that the low polarity and/or the low crystallinity of a polymer matrix appeared to favor better compatibility with SRP.

Previous studies suggested that the addition of SRP generally resulted in significant deterioration in the mechanical properties of these composites. It was reported that linear low-density polyethylene (LLDPE) filled with SRP caused a drop in impact resistance by 50–70%, depending on the SRP type and average mesh size of SRP particles.¹¹ Poor mechanical properties had also been observed for blends of SRP with PP.⁷ In these SRP composites, the poor adhesion between matrix and SRP and the large particle size of SRP were believed to be major factors deteriorating the mechanical properties of the composites. The large, essentially unbonded SRP particles generally

Correspondence to: Y. Zhang (yong_zhang@sjtu.edu.cn).

cause the principal stress maxima occurring at the equatorial interface, which led to crack propagation and eventual failure of the composites.¹² Because there was little or no reduction in the size of SRP particles during normal melt-mixing operations and at the same time the particle size was limited by the cost of the grinding process, this led to a focus on developing methods of improving the interfacial adhesion or compatibility between SRP and the host polymer, in the hope of improving the properties of composites.^{13–18}

Several investigations have been reported to improve the adhesion of SRP with various thermoplastic polymers. One approach for this improvement involves the surface modification of SRP, either by chemical means or by physical means. Photografting of acrylamide (AAM) onto SRP was carried out using ultraviolet rays to modify the surface characteristics of SRP.¹³ The surface-modified SRP was melt-blended with high-density polyethylene (HDPE). Meanwhile, maleic anhydride (MAH)-modified PP was added to improve interfacial adhesion through a reaction between MAH and AAM-grafted SRP. The results showed that the AAM-grafted rubber powder/HDPE/compatibilizer composites had better mechanical properties than those of the HDPE/unmodified SRP composites. It also was reported that high energy treatments, including plasma, corona, and electron beam radiation, could increase the oxygen concentration on the SRP surface and bring about better adhesion of SRP with the matrix.¹¹ Of these high energy treatments, electron beam radiation led to a higher increase in impact energy of the SRP composites than that treated by corona and plasma. Tuchman et al.^{9,14} reported that grafting of styrene onto SRP particles using an aqueous slurry process provided better mechanical properties than those of straight mechanical blends of SRP with PS, although the properties were still inferior to those of the PS without SRP.

Another approach for improving the compatibility of SRP and matrix polymer is to use compatibilizer(s). The addition of a silane coupling agent improved the mechanical properties of the SRP-polymer composites.¹⁵ Phadke et al.⁷ found that the addition of natural rubber as a compatibilizer increased the adhesion between SRP and PP matrix. The reactive/interfacial coupling mechanism also was introduced to solve the interfacial incompatibility between the filler and the polymer matrix. Among the various functional compatibilizers used, ethylene-*co*-glycidyl methacrylate copolymer was considered to effectively improve the impact energy of such composites.¹⁶ Investigations were also undertaken to employ two-component reactive compatibilizing systems to induce the *in situ* reactive compatibilization of SRP in a thermoplastic polymer matrix. The use of two-component compatibilizers like epoxydized natural rubber/ethylene-*co*-acrylic acid copolymer and amine-terminated poly-

butadiene/ethylene-*co*-acrylic acid copolymer had limited success in improving the impact strength, depending on the composition of the composites.^{17,18}

In this work, a novel method was introduced to improve the adhesion of SRP and HDPE matrix. EPDM was chosen as the compatibilizer, which has some structural similarity with the HDPE phase and was rubbery in nature. Dicumyl peroxide (DCP) and dimethyl silicone oil were also used to enhance the compatibilization. Here, the HDPE/SRP composites were defined as unmodified composites, whereas the HDPE/SRP composites containing the three additives were defined as modified composites. The contents of the additives of EPDM, DCP, and dimethyl silicone oil were 10, 0.2, and 4 wt % based on the HDPE loading, respectively. Mechanical properties, dynamic mechanical properties, melt flow index, and microstructure were measured to evaluate the effect of compatibilization.

EXPERIMENTAL

Materials

Cryogenically made SRP was provided by Hubei Dawei Co. (China), with an average particle size of 60 mesh by sieving. Analysis of SRP by thermogravimetry yielded 54.9 wt % rubber hydrocarbon content, 6.1 wt % oil, 35.9 wt % carbon black, and 3 wt % ash. The SRP sample was dried at 80°C for 24 h before blending. Commercial HDPE 5000s [melt flow index (MFI), 0.923 g/10 min; density, 0.954 g/cm³; Yangtze Petroleum Chemical Co., Ltd., China] was used as the matrix polymer.

EPDM (EP35) had 43 wt % propylene content; termonomer 5-ethylidene-2-norbornene (ENB); iodine number, 26; ML (1+4) 83 (100°C), and was supplied by Japan Synthetic Rubber Co. (Tokyo). Dimethyl silicone oil, 201-100, was made by the Shanghai Special Resin Research Institute, China. DCP was an industrial-grade product.

Compounding

Melt blending was carried out in the mixing chamber of a Haake rheometer RC90 (Germany) at 180°C at a rotor speed of 50 rpm for 13 min. A blend of SRP with HDPE and EPDM was mixed for 2 min and then silicone oil was added to the mixture; after 5 min, DCP was added. The melt-mixing lasted for 8 min to obtain HDPE/SRP/EPDM/silicone oil/DCP composites.

Specimen preparation

Samples of 1 and 3 mm in thickness for morphology observation and properties testing were prepared by

compression molding at 180°C for 8 min and at room temperature for 10 min.

Mechanical properties

Notched Charpy impact load–time curves were measured on an Instron Charpy impact tester (USA) according to ISO 179. Dumbbell-shape specimens for tensile test were cut from the 1-mm-thick sheets and tested by using an Instron 4465 electron tensile tester according to ASTM D638 at a crosshead speed of 50 mm/min. Hardness was measured in a Zwick Shore D hardness tester (Germany) according to ASTM D 2240.

Rheological behavior

Melt flow measurements of the composites were carried out using an Instron 4467 capillary rheometer ($L/D = 40$) at 180°C.

Dynamic mechanical properties

The dynamic mechanical properties of composites were measured using an RSI Orchestrator (USA) at 0.01% strain and 1 Hz frequency. Molded samples with dimensions $6 \times 4 \times 0.8$ mm were used for testing. The samples were tested in a temperature range of -80 to 80°C at a heating rate of $3^\circ\text{C}/\text{min}$.

Morphology observation

Impact fracture surfaces for morphology observation were obtained by fracturing the notched samples in the impact tester at 23°C . The fracture surfaces were then gold-coated and observed using a Hitachi S-2150 scanning electron microscope (SEM; Hitachi, Tokyo, Japan).

RESULTS AND DISCUSSION

Mechanical properties of HDPE filled with SRP

There was no difficulty in incorporating SRP, even at high loading into polymer matrices. Only the composites containing 10–60 wt % SRP (60 mesh) were studied in this investigation. The addition of SRP led to a dramatic deterioration of all the mechanical properties, especially the impact strength and elongation at break, as shown in Figures 1–4. The poor adhesion between SRP and polymer matrix and the large particles facilitates the propagation of cracks and leads to a significant decline in the mechanical properties.¹²

Figure 1 shows the impact strength of the composites containing different SRP loadings with respect to the pure HDPE matrix. For the unmodified composites, the impact strength increased slightly with increasing the SRP loading, which can be attributed to

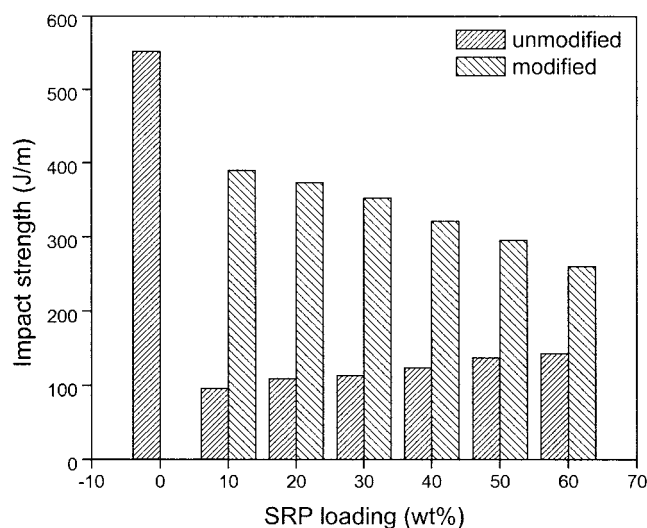


Figure 1 Effect of SRP loading on the impact strength of the HDPE/SRP composites.

the increased amount of elastic (or bending) deformation before the onset of ductile deformation with increasing the SRP loading in the simple mixing systems.¹⁶ After the 10 wt % EPDM, 0.2 wt % DCP, and 4 wt % dimethyl silicone oil were added into the HDPE/SRP composites, the impact strength was improved significantly; that is, the impact strength increased by 160% for the composites containing 40 wt % SRP, as shown in Figure 1. With increasing the SRP loading, the improvement in the impact strength became less apparent for the modified composites.

To improve the adhesion between SRP particles and polymer matrix, some interactions must form at the interface. The surface modification of SRP and the use of compatibilizers have been adapted by many researchers as discussed above. In this study, the improvement in the impact strength could be explained as the consequence of a good adhesion between SRP and HDPE related to the addition of the additives of EPDM, DCP, and silicone oil.

The effect of individual additives on the mechanical properties of the composites containing 40 wt % SRP is shown in Table I. The impact strength of the SRP composites containing the three additives was higher than that of the composites containing only one or two of the additives. The three additives played a role in the “synergistic toughness” of the HDPE/SRP composites.

It is well known that the toughness of polyolefins such as PP and PE at low temperature can be improved by blending a small amount of rubbery materials, including ethylene–propylene copolymer, EPDM, and the like.¹⁹ EPDM has good compatibility with polyolefin. In addition, EPDM can encapsulate the surface of the dispersed SRP particles and form a co-continuous phase, which has been identified by

TABLE I
Mechanical Properties of the HDPE/SRP Composites with the Additive(s)^a

Composition	Impact strength (J/m)	Tensile strength (MPa)	Elongation at break (%)
0	124	14.3	45.7
10 wt % EPDM	210	12.1	58.8
0.2 wt % DCP	110	13.1	45.4
4 wt % silicone oil	127	14.6	35.6
10 wt % EPDM + 0.2 wt % DCP + 4 wt % silicone oil	320	11.6	113

^a Basic formulation: HDPE 100, SRP 40.

TEM morphology observation.²⁰ Therefore, the addition of EPDM improved the combination of SRP/HDPE matrix.

Infrared analysis showed that there were no other functional groups but double bonds on the surface of SRP particles.²¹ The addition of DCP probably initiated the double-bond reactions on the surface of SRP particles and the EPDM molecules in the modified composites. Therefore, the interaction between SRP and HDPE matrix was further enhanced by the reaction of the double bonds.

It was reported in our earlier studies that, after the addition of silicone oil into highly filled HDPE composites, the impact strength and elongation at break could be improved if the fillers were modified with a suitable coupling agent.^{22,23} In this study, silicone oil might encapsulate the SRP particles and form a soft, thick layer between SRP and the polymer matrix. Kalinski et al.²⁴ reported that the liquid layer in the polymer and filler could inhibit fracture phenomena (i.e., crack formation and propagation). Under this consideration, the encapsulation of silicone oil should have released the stress concentration around the SRP

particles and hence increased the mechanical properties of the modified composites.

However, the addition of only one additive such as silicone oil could not increase the mechanical properties of the HDPE/SRP composites, as shown in Table I. This can be attributed to the poor adhesion that still exists between SRP and HDPE after the addition of silicone oil. That is to say, the suitable adhesion between SRP and HDPE is necessary to realize the role of the soft layer of silicone oil.

The SRP particles also have detrimental effects on the tensile strength and elongation at break of the composites, as shown in Figures 2 and 3, respectively. With increasing the SRP loading, the tensile strength decreased for all the composites. The elongation at break of the composites was significantly enhanced after the modification. For instance, the elongation at break of the modified composites containing 40 wt % SRP increased by 150% compared to that of the unmodified composites. The increases of the impact strength and elongation at break after the modification imply a good adhesion formed by synergistic effects of the additives. The hardness decreased with increasing

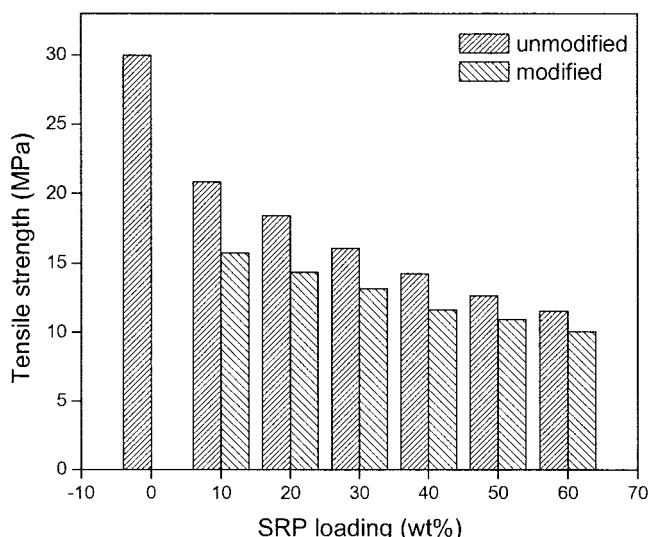


Figure 2 Effect of SRP loading on the tensile strength of the HDPE/SRP composites.

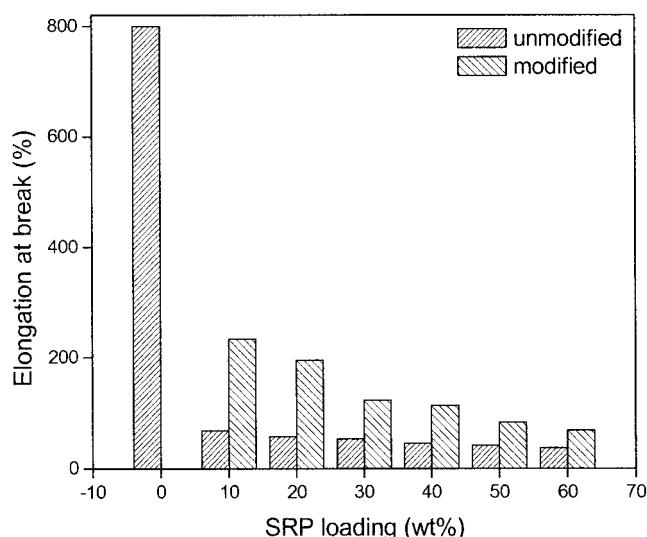


Figure 3 Effect of SRP loading on the elongation at break of the HDPE/SRP composites.

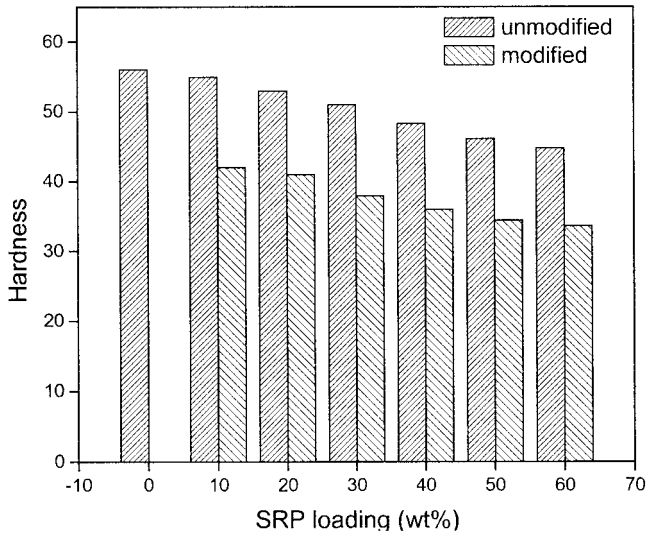


Figure 4 Effect of SRP loading on the hardness of the HDPE/SRP composites.

the SRP loading. Moreover, the addition of additives further decreased the hardness, as shown in Figure 4.

Impact process

To clearly understand the impact failure process of the HDPE/SRP composites, the impact load–time curves were recorded by the notched Charpy tester and shown in Figure 5. This figure presents the load induced at the tip of the falling dart plotted against its time through the specimen. The impact energy was given by the area under a curve. The changes of the load and ductile deformation (represented by the time beyond the yield point) during the impact fracture process can be seen in Figure 5. The impact strength was defined as the impact energy absorbed by material per unit thickness. The initial slope of the load–time curve gives the relative stiffness of the composites corresponding to a high-speed load. As expected, the addition of relatively softer SRP particles led to a decrease in the stiffness of the composites. With increasing the SRP loading, the stiffness decreased for all the composites with or without the additives. Simultaneously, the maximum force, which corresponded to the load at the yield point in the failure process, decreased after the addition of SRP into HDPE matrix.²⁵

The comparison of the impact processes of the unmodified and modified composites is also shown in Figure 5. The modified composites had a low initial slope, which should be attributed to the addition of the EPDM elastomeric phase. The maximum force for the composites containing the additives was higher than that of the corresponding composites without the additives. At the meantime, the modified composites had more ductile deformation than that of the unmod-

ified SRP composites. These experimental results might be accounted for by the improved adhesion between SRP and HDPE, which favored the increase in stress at the yield point and in the predominance of ductile deformation.

The impact energy of the composites was in the sequence $a > b > d > e > c$, as shown in Figure 5. The addition of the additives improved the impact resistance for the composites containing 10 or 40 wt % SRP. However, the extent of improvement was higher for the composites containing 10 wt % SRP than for the composites containing 40 wt % SRP at a given loading of the additives, compared to that of the impact energy.

Good adhesion between SRP and the HDPE matrix in the composites containing the additives can also be seen through SEM morphology observation. Although good adhesion results in noticeably better properties, especially the impact strength and elongation at break, the properties of the composites are still inferior to those of the pure HDPE.

Effects of SRP and additives on the rheological behavior

Figure 6 shows the apparent viscosity versus shear rate on a log–log scale for the HDPE/SRP composites at 180°C. The shear rate was limited below 60 s^{-1} ; otherwise, the HDPE/SRP composite melts would exhibit unsteady flow. The rheological behavior of the composites depended greatly on the SRP loading. When the SRP loading varied from 0 to 40 wt % in the HDPE/SRP composites, the apparent viscosity increased by 44% at a shear rate of 6 s^{-1} . The addition of

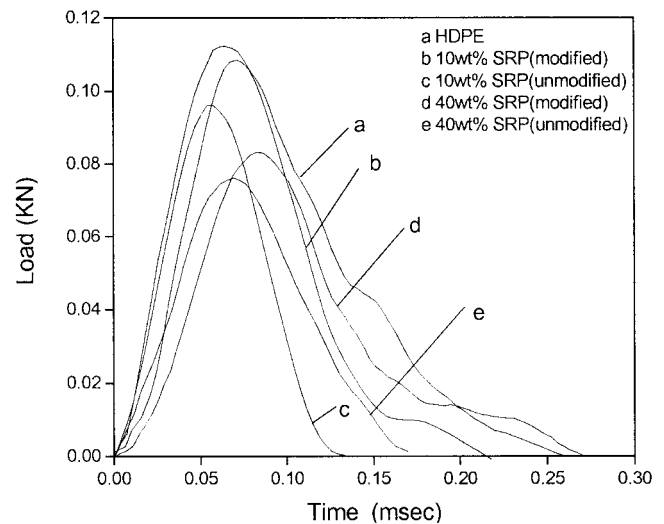


Figure 5 Load–time curves of the HDPE/SRP composites. Formulations: (a) HDPE; (b) HDPE 100, SRP 10, EPDM 10, DCP 0.2, silicone oil 4; (c) HDPE 100, SRP 10; (d) HDPE 100, SRP 40, EPDM 10, DCP 0.2, silicone oil 4; (e) HDPE 100, SRP 40.

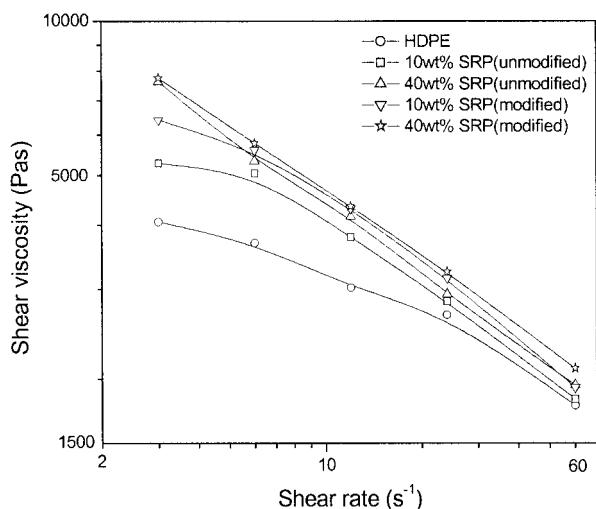


Figure 6 Log-log plot of apparent viscosity versus shear rate of the HDPE/SRP composites.

additives significantly increased the apparent viscosity of the composites.

The apparent viscosity can be used to assess the interaction between the phases. Petrovic et al.²⁶ studied the properties of PP/ethylene-propylene rubber/LDPE composites. They found that in the presence of interaction, the effective particle size, and the volume fraction increased because of an interfacial layer that formed around the particle, and then relative viscosity would become higher than that predicted by Einstein's equation. Choudhury et al.⁶ found that the decrease in the MFI or the increase in the apparent viscosity with the addition of chlorinated polyethylene (CPE) in the SPR/LDPE composites was attributed to the improved adhesion of CPE with the component phases. Therefore, the increase in apparent viscosity reported in this study should be ascribed to the improvement of adhesion between SRP and HDPE because of the addition of additives, as shown in Figure 6.

Dynamic mechanical properties

The dynamic mechanical properties [i.e., storage modulus and loss tangent ($\tan \delta$)] are given in Figures 7 and 8, respectively. Dynamic mechanical analysis was used to further understand the effect of surface modification on the properties of the composites.

Figure 7 shows the storage modulus as a function of temperature at a frequency of 1 Hz. The addition of SRP led to a decrease in the storage modulus of HDPE, and the addition of additives led to a further decrease in the storage modulus of the HDPE/SRP composites. The decreases in the storage modulus can be attributed to the existence of SRP and EPDM. The elastomeric phases impart their properties to the modified

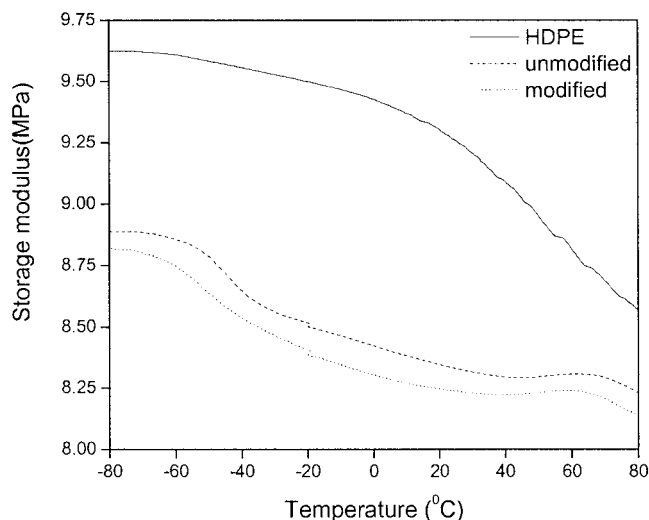


Figure 7 Effect of temperature on storage modulus of the composites containing 40 wt % SRP.

composites, and hence increase the impact strengths of the modified composites.⁷

Figure 8 presents a typical loss tangent as a function of temperature for the composites containing 40 wt % SRP. In the experimental temperature range, the $\tan \delta$ peak of HDPE was not observed, whereas the SRP-filled composites showed the $\tan \delta$ peaks at -44°C for the unmodified composites and -53°C for the modified composites. The two peaks should indicate the glass-transition temperature (T_g) of SRP and SRP/additives.

It may be seen that the composites containing 40 wt % SRP modified with the additives had a lower T_g value for the rubber phase than that of the unmodified composites. Bucknall et al.²⁷ reported that the energy absorbed during the propagation of cracks was par-

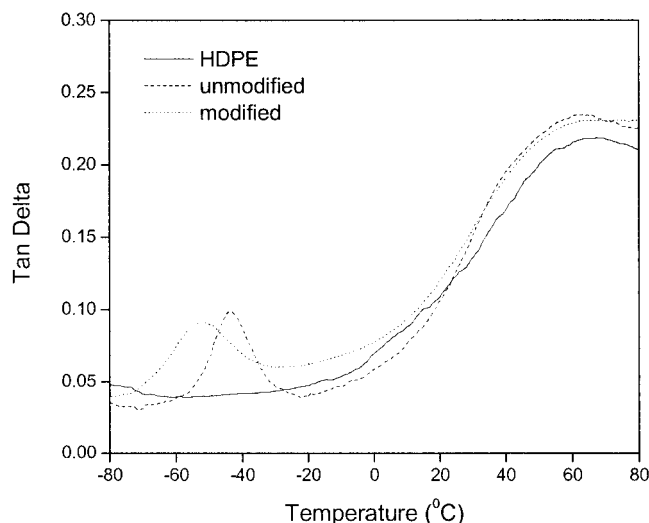


Figure 8 Effect of temperature on loss tangent of the composites containing 40 wt % SRP.

tially dependent on the relaxation behavior of the rubber phase in high impact polystyrene (HIPS). The rubber phase of polybutadiene in HIPS with low T_g had higher notched Izod impact strength than that of the rubber phase of butadiene–styrene copolymer with high T_g . Here, the T_g of the rubber phase (SRP/additives) might have some advantageous effects on the impact property of the modified composites.

Moreover, the modified composites had lower values of $\tan \delta$ than that of the unmodified composites. Nielsen et al.²⁸ found that powder–polymer friction, where there was essentially no adhesion at the interface and excess damping in the polymer near the interface because of induced thermal stresses or changes in polymer conformation, resulted in a high value of $\tan \delta$. In other words, good interactions between the powders and polymer matrix and the good dispersion of powder in the matrix should result in the reduction of the damping factor. Therefore, the dynamic mechanical test results indicated that a good adhesion had formed in the modified HDPE/SRP composites, and, at the same time, the addition of the additives improved the dispersion uniformity of the SRP.

SEM studies

Surfaces of the impact fracture specimens were observed using SEM and the results are shown in Figure 9, where different states of adhesion can be observed. Although a small amount of plastic deformation formed in the unmodified composites, there were substantial vacuole and debonding regions, indicating the poor adhesion between the SRP particles and the polymer matrix, as well as the stress concentration around them, as shown in Figure 9(a). Addition of the additives into the HDPE/SRP composites caused the changes in the impact fracture surface, as shown in Figure 9(b). It may be observed that a lot of the rubber powders still remained on the impact fractured surfaces of the modified composites, whereas few rubber powders are found in the unmodified composites. A better adhesion and shear deformation lines, which started at the interface, probably were beneficial in absorbing the impact energy, thus leading to an improvement in impact properties and elongation at break.

Oliphant et al.¹⁶ reported that the HDPE/SRP and LLDPE/SRP composites had different failure profiles. The failure of the SRP/LLDPE composites was a ductile yielding process, but the failure mechanism of the HDPE/SRP composites was mainly catastrophic propagation of a crack through the impact zone. They found that only moderate adhesion was necessary for the SRP/LLDPE composites with a large particle size, but high levels of adhesion or much smaller particles were essential for the HDPE/SRP composites to ob-

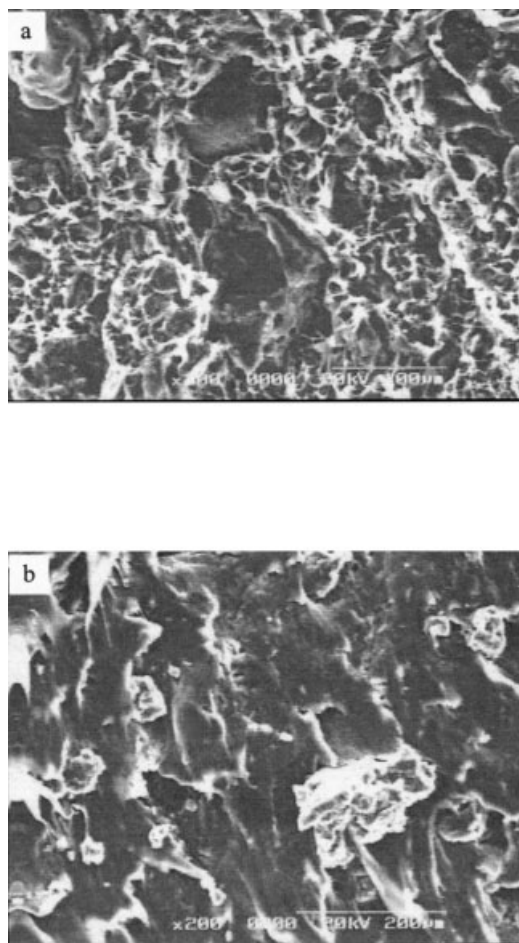


Figure 9 SEM photomicrographs of impact fracture surfaces of the composites. Formulations: (a) HDPE 100, SRP 40; (b) HDPE 100, SRP 40, EPDM 10, DCP 0.2, silicone oil 4.

tain useful mechanical properties. In our work, the additives EPDM, DCP, and silicone oil might play an important role in modifying adhesion and hence inhibit the crack propagation of the pure HDPE.

CONCLUSIONS

The addition of SRP into HDPE led to notable deterioration in the mechanical properties, especially the impact strength and elongation at break. The additives of 10 wt % EPDM, 0.2 wt % DCP, and 4 wt % silicone oil could increase the impact strength and elongation at break of the HDPE/SRP composites significantly. The composites modified by the additives had higher impact energy than that of the unmodified composites, which resulted from the increased force at yield point and the ductile deformation. The presence of SRP in the HDPE led to an increase in the apparent viscosity. The SRP and the additives had significant effects on the storage modulus and $\tan \delta$ values of HDPE, and the additives had an effect on the mor-

phology of the fracture surface of the HDPE/SRP composites.

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