Studies on the Dynamically Vulcanized Polypropylene (PP)/Butadiene Styrene Block Copolymer (SBS) Blends: Mechanical Properties

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ABSTRACT: Unvulcanized and dynamically vulcanized blends of isotactic polypropylene (PP) and butadiene styrene block copolymer (SBS) in the composition range of 10–40 wt % SBS were prepared by melt mixing in an internal mixer and evaluated for impact and tensile properties. Dynamic vulcanization of blends gave superior mechanical properties. Systematic changes with varying blend composition were found in stress–strain behavior in both the blend systems. The effect of blend composition on the state of dispersion and morphology of the dispersed phase droplets were studied by scanning electron microscopy. Analysis of the yield stress data in terms of various theoretical models revealed the variation of stress concentration effect with blend composition and higher interphase adhesion in dynamically vulcanized blends. © 1997 John Wiley & Sons, Inc. J Appl Polym Sci **65:** 2691–2701, 1997

Key words: polypropylene; butadiene styrene block copolymer; dynamic vulcanization; yield stress; impact strength

INTRODUCTION

Isotactic polypropylene (PP), despite having some excellent properties, suffers from poor impact strength. Melt blending of PP with different elastomers and rubbers such as ethylene propylene copolymers (EPM), ethylene–propylene diene terpolymers (EPDM), polyisobutylenes (PiB), butadiene–styrene block copolymers (SBS), etc., is widely used for increasing the impact strength of PP, especially at low temperatures.[1] Incorporation of an elastomer, although increases the impact strength, results in deterioration of tensile properties. The mechanical properties of a thermoplastic elastomer blend can be further improved by dynamic vulcanization.[2] The dynamically cured thermoplastic elastomer blends have been widely used in plastic industry[3–5] and have important technical advantages in processing because the blends can be fabricated by such methods as extrusion and molding, although they possess a crosslinked elastomer as one component. In the present work a comparative study of impact and tensile properties of unvulcanized and dynamically vulcanized PP/SBS blends has been undertaken. Scanning electron microscopy was used to study the state of dispersion and change in morphology of dispersed phase droplets with blend composition. Analysis of tensile data in terms of various theoretical models is presented to reveal the variation of interfacial adhesion and the stress concentration effect with blend composition in both the unvulcanized and dynamically vulcanized blend systems.

EXPERIMENTAL

Materials

Isotactic Polypropylene (PP), Koylene S-3030 (MFI—3 g/10 min at 230°C and 2.16 kg/cm^2) was
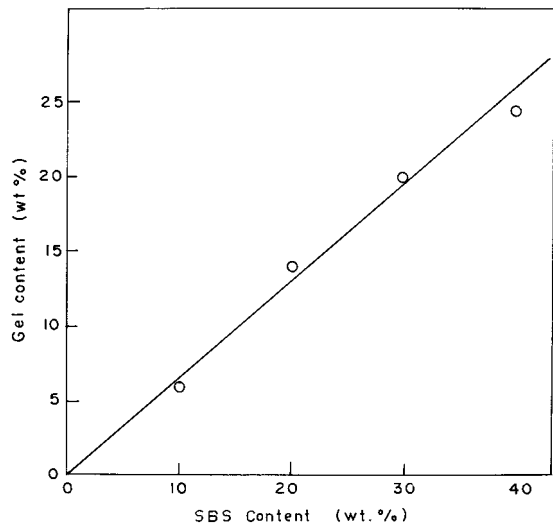
Figure 1 Variation of gel content with blend composition in dynamically vulcanized PP/SBS blends.

obtained from Indian Petrochemicals Corpn., Ltd. Butadiene styrene block copolymer (SBS), Cariflex-1101 (30% styrene content) was procured from Shell Chemical Co. A modified phenolic resole resin, Hylak HR-6415 from Bakelite Hylam, India, and zinc oxide (LR) from Qualigens were used as a curing agent and accelerator, respectively, for vulcanization.

Preparation of Blends and Test Samples

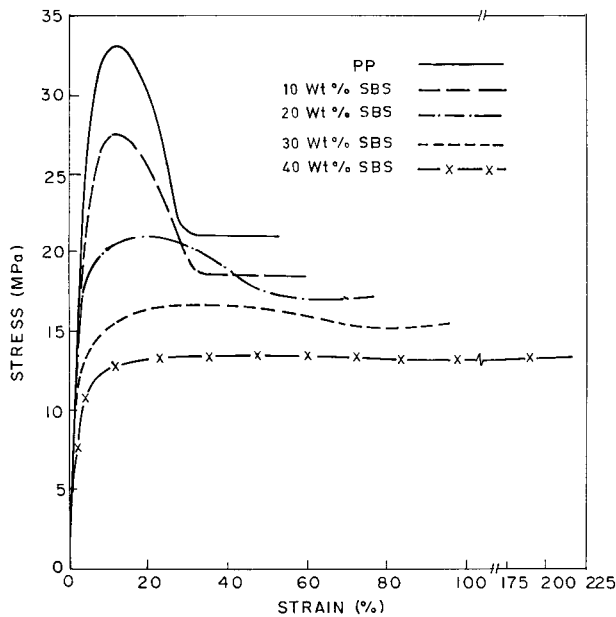The unvulcanized PP/SBS blends of compositions 10, 20, 30, and 40 wt % SBS were prepared by

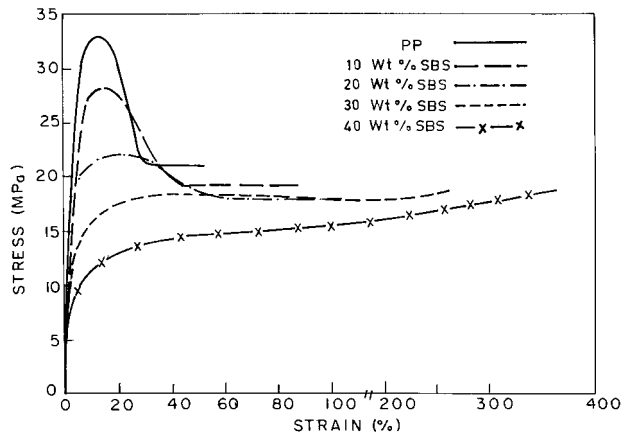**Figure 2** Stress–strain behavior of unvulcanized PP/SBS blends.

Figure 3 Stress–strain behavior of dynamically vulcanized PP/SBS blends.

melt mixing at 180°C for 10 min in the internal mixer of a torque rheometer, HAAKE RHEO-CORD RC-90 using roller rotors. The charge weight taken was 200 g. The corresponding dynamically vulcanized blends were also prepared
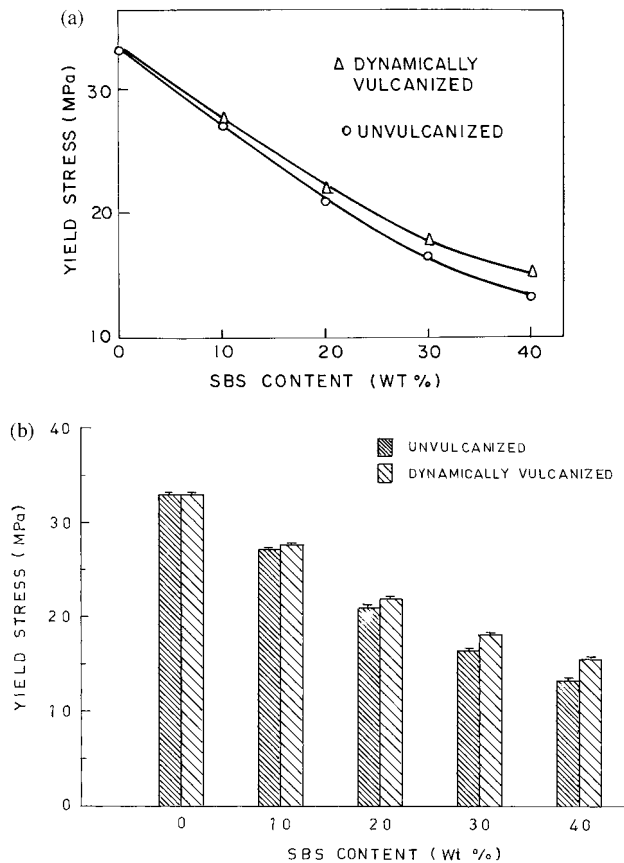

Figure 4 (a) Variation of yield stress with blend composition for PP/SBS blends. (b) Error bar diagram of yield stress data for PP/SBS blends.
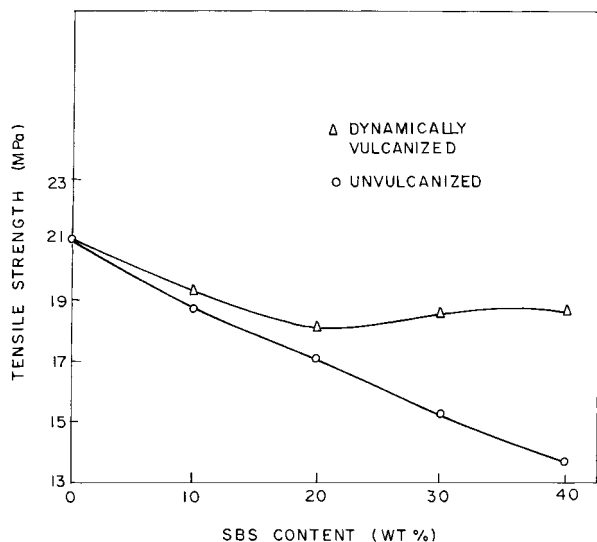
Figure 5 Variation of tensile strength with blend composition for PP/SBS blends.

under identical conditions except that after initial mixing for 5 min, a mixture of curing agent and accelerator (10 wt % and 1 wt %, respectively, on the basis of SBS content in the blend) was introduced in the cavity of the mixer while mixing, which was further continued for 5 min until an equilibrium torque was reached after curing.

The molten mass from the mixer was cooled and cut into fine granules. The granules were compression molded in a laboratory press at 190°C and 30 MPa pressure. The mold was specially designed in such a way that it could be cooled immediately after molding, keeping the samples still under compression. The samples for tensile test, impact test, and SEM were cut out from the molded sheet.
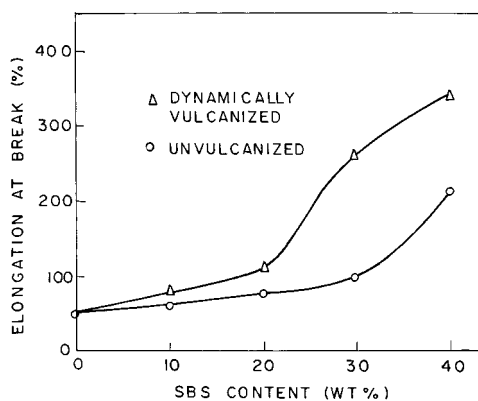
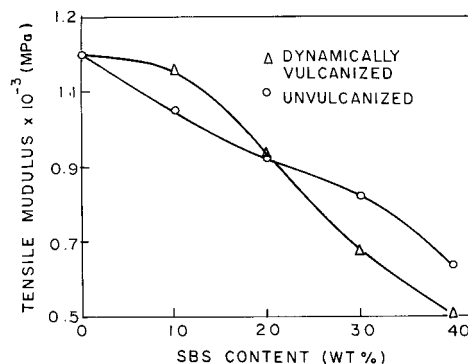The fine granules of the blend samples were

Figure 7 Variation of tensile modulus with blend composition for PP/SBS blends.

further powdered in a wiley mill for determining the gel content.

Gel Content

The gel content in dynamically vulcanized blends were determined by extraction of powdered samples in boiling xylene in a R.B. flask for 72 h.

Tensile Properties

Tensile testing of the blend samples was carried out at 25 ± 2°C on an Instron Universal Testing Machine (Model-4302) on dumbell-shaped samples according to ASTM D-638 at a strain rate of 50 mm/min with an initial gauge length of 2.5 cm.

Impact Properties

Izod impact strength was measured on notched specimen on an FIE (Model 0.42) impact tester ac-
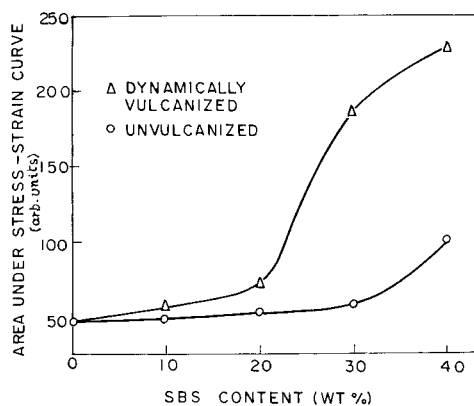

Figure 6 Variation of elongation at break with blend composition for PP/SBS blends.

Figure 8 Variation of the area under the stress–strain curve with blend composition for PP/SBS blends.
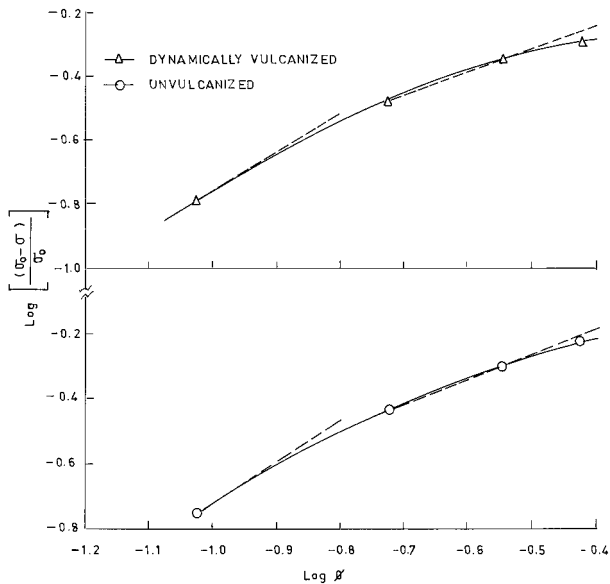
Figure 9   Variation of $\log[(\sigma_O - \sigma)/\sigma_O]$ with $\log \phi$ for PP/SBS blends.

Figure 11   Yield stress data as a function of volume fraction of SBS for the dynamically vulcanized PP/SBS blend and the theoretical curves according to eq. (3).

cording to ASTM-D-256. Rectangular specimens of dimensions 5×1 cm and thickness 0.3 cm with 2 mm deep triangular notches of 45° were used.

Cambridge instrument Scanning Electron Microscope (Model S4−10).

RESULTS AND DISCUSSION

Gel Content

The variation of gel content with blend composition in dynamically vulcanized PP/SBS blends is shown in Figure 1. The gel content increases linearly with an increase in the SBS content. Dynamic vulcanization causes crosslinking of SBS particles, which is marked by a decrease in extractables or an increase in the gel content.

Morphology Studies

Molded samples of blends were broken after freezing the samples in liquid nitrogen. The microtoned edges of the samples were kept immersed in cyclohexanone for about 48 h at ambient temperature to dissolve out the SBS phase. Scanning electron micrographs (SEMs) of cryogenically fractured and etched samples were recorded on a
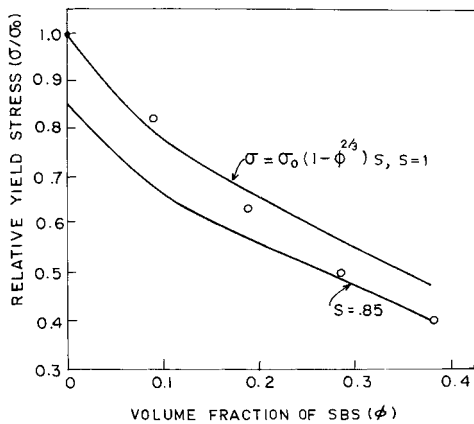

Figure 10   Yield stress data as a function of volume fraction of SBS for the unvulcanized PP/SBS blend and the theoretical curves according to eq. (3).

Figure 12   Yield stress data as a function of volume fraction of SBS for the unvulcanized PP/SBS blend and the theoretical curves according to eq. (4).
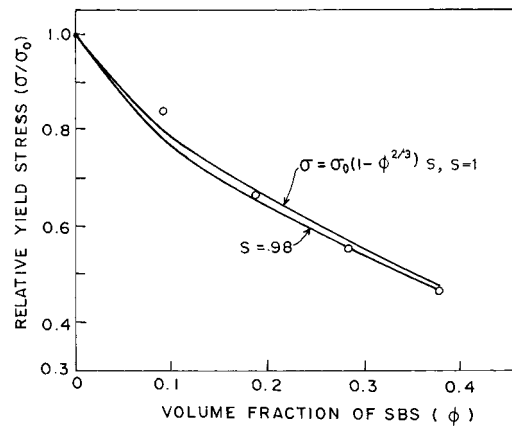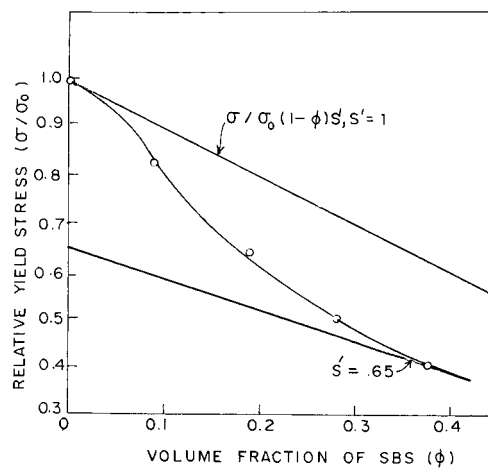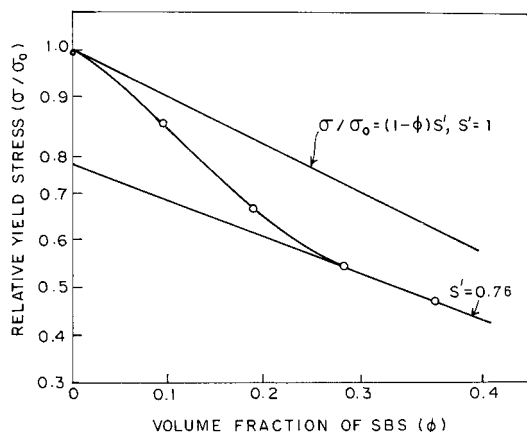
Figure 13 Yield stress data as a function of volume fraction of SBS for the dynamically vulcanized PP/SBS blend and the theoretical curves according to eq. (4).

Mechanical Properties

Tensile Properties

The stress–strain curves of unvulcanized and dynamically vulcanized PP/SBS blends at various compositions are shown in Figures 2 and 3, respectively. The differences in deformation characteristics of PP and its blends under an applied load are evident from the stress–strain curve. PP shows a well-defined yield peak and finally fails by a necking type of rupture. Blending of PP with SBS reduces the yield stress in general over the entire studied range of blend composition with an increase in the yield peak width, indicating an increase in the elastomeric nature. The yield peak disappears and the stress–strain curve flattens to a shape similar to that of an elastomer at higher SBS content. The disappearance of the yield peak takes place at a lower composition range, i.e., at 20 to 30 wt % SBS content in the dynamically vulcanized blends than in the unvulcanized blends, i.e., at 30–40 wt % SBS content. In these cases, the yield stress was defined as the stress at which the slope of the stress–strain curve equals zero. The tensile properties thus determined are shown in Figures 4 to 8 as a function of blend composition. Blending of PP with SBS, while decreasing yield stress, tensile strength, and tensile modulus, increases elongation at break and the area under the stress–strain curve in general. This behavior of the PP blend with an elastomer is well documented in the literature.[6,7] The total area under the stress–strain curve is a measure of the energy required to break the sample and can be correlated with impact strength.[8] The simultaneous occurrence of lowering of the yield stress and increase in the area under the stress–strain indicates that although blending causes yielding to occur at a lower stress, the process of yielding requires greater energy.

In dynamically vulcanized blends, there is a network structure and higher chain entanglements, which results in yielding to occur at a higher stress than in the unvulcanized system [Fig. 4(a,b)]. The dynamically vulcanized blends are having higher elongation at break as the craze fibrils are stabilized by molecular entanglements and can sustain high stress for long periods (Fig. 6). Similar improvement in properties in the dynamically vulcanized blends is documented in the literature.[2,3] However, the dynamically vulcanized blends have higher tensile modulus than the unvulcanized blends only at compositions having up to 20 wt % SBS content (Fig. 7). This may be because at higher SBS content, the increase in yield strain on vulcanization is manyfold larger than the increase in yield stress, resulting in lower modulus in the dynamically vulcanized blends at these compositions.

Analysis of Blend Composition Dependence of Mechanical Properties

Analysis of the variation of mechanical properties, for example, the yield stress with blend composition in terms of some existing theoretical models provides knowledge about discontinuities in the stress transfer in two-phase blends and reveals the difference in the unvulcanized and dynamically vulcanized blend systems.

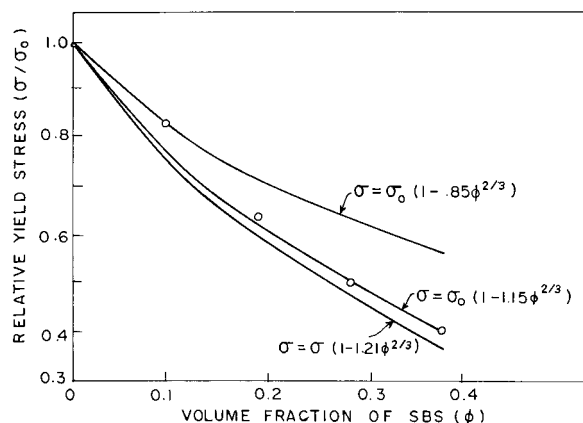The two most common expressions for composition dependence of mechanical properties of two-

Figure 14 Theoretical curves according to eq. (5) with weightage factors 0.85, 1.15, and 1.21 and the experimental data points for unvulcanized PP/SBS blends.
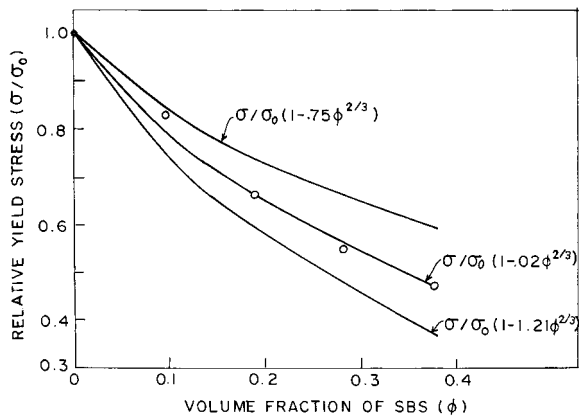
Figure 15 Theoretical curves according to eq. (5) with weightage factors 0.75, 1.02, and 1.21 and the experimental data points for the dynamically vulcanized PP/SBS blends.

phase blends (or composites) are based on the "first power law" [eq. (1)] and "two-third power law" [eq. (2)].

$$\sigma = \sigma_o(1 - \phi) \qquad (1)$$

$$\sigma = \sigma_o(1 - \phi^{2/3}) \qquad (2)$$

where σ and σ_o denote the given mechanical property (yield stress, tensile strength, etc.) of the blend and the matrix, respectively. ϕ is the volume fraction of the dispersed phase (i.e., the inclusions). These power laws originate from the considerations of the effect of area fraction and volume fraction of the inclusions in the modification of the property.[9,10] The first power relationship of area fraction to volume fraction in any randomly chosen plane of fracture is derived for a completely random distribution of the dispersed phase, while the two-third power law with appropriate weightage factor is derived for spherical inclusions. Validity of both the first power and two-thirds power relationships of area fraction and volume fraction for the same blend, depending upon the composition or shape of inclusions, have been shown by Kunori and Geil,[9] based on the analysis of a SEM picture of fractured surfaces of two-phase blends. Hence, both the power laws have been used for the present analysis of data.

Validity of the first power law or two-thirds power law at different compositions for the present blend systems has been explored by considering the plots of $\log[(\sigma_o - \sigma)/\sigma_o]$ versus $\log \phi$ from the yield stress data for both the blend systems and are shown in Figure 9. The slope of the plot

is equal to the exponent ϕ of eqs. (1) and (2). For unvulcanized blend systems, the value of slope is 0.62 at low volume fraction (i.e., $\phi \leq 0.095$) and changes to 0.4 at higher volume fraction (i.e., $\phi > 0.095$), while for dynamically vulcanized blend systems the slope value is 0.60 at lower volume fraction ($\phi \leq 0.095$) and 0.38 at high volume fraction. This shows that for both the blend systems the power law exponent is closer to two-third than unity at all the studied range of compositions.

Stress Concentration Parameters

To take into account the effects of stress concentrations at the inclusion–matrix interface, modifications of the power laws have been done by various authors.

Nielsen[8] suggested the use of a parameter S in two-third power law, given by eq. (3), where S can acquire a maximum value of unity when the stress transfer is continuous or there is no stress concentration effect, and the lower the value of S, the greater is the stress concentration effect. Comparison of theoretical curves according to eq. (3) incorporating appropriate S values with the experimental data points are shown in Figures 10 and 11 for unvulcanized and dynamically vulcanized blend systems, respectively.

$$\sigma = \sigma_o(1 - \phi^{2/3})S \qquad (3)$$

The modified two-thirds power law [eq. (3)] with S less than 1 includes the additional effect of stress concentration and shifts σ/σ_o versus the ϕ curve downward than the curve with $S = 1$.

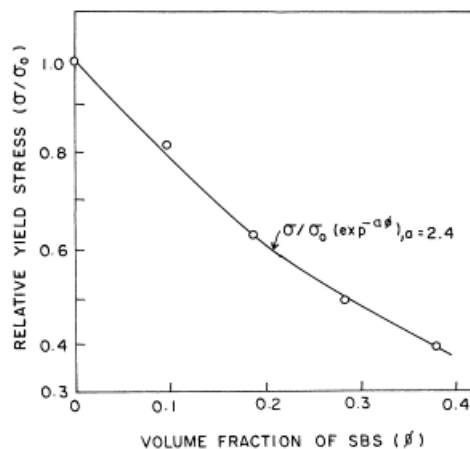The experimental data points of yield stress for

Figure 16 Theoretical curve according to eq. (7) for $a = 2.4$ with experimental data points for unvulcanized PP/SBS blends.
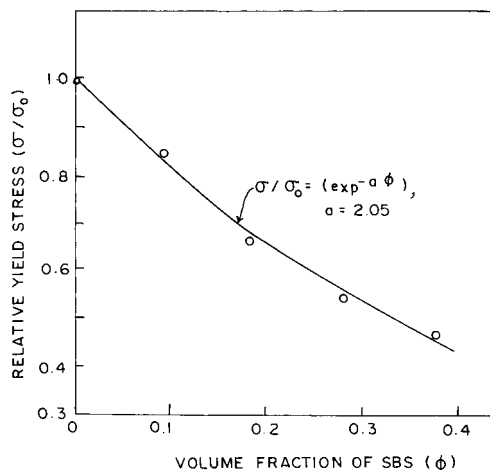
Figure 17 Theoretical curve according to eq. (7) for $a = 2.05$ with experimental data points for dynamically vulcanized PP/SBS blends.

the unvulcanized blend system at a higher volume fraction ($\phi > 2.5$) of SBS are in perfect agreement with the theoretical curve according to eq. (3), with $S = 0.85$ (Fig. 10), and deviates upward systematically towards the curve with $S = 1$ (no stress concentration) as the volume fraction of inclusion decreases. Similar behavior is observed in dynamically vulcanized blends (Fig. 11). However, here the experimental data points at a higher volume fraction ($\phi > 0.1$) lie very close to the theoretical curve [eq. (3)] with $S = 0.98$. This indicates that in both systems the stress concentration effect is predominant at a higher volume fraction of SBS, and comparison of the stress concentration parameter values indicates that the stress concentration effect is less in the case of the dynamically vulcanized blend system ($S = 0.98$) than in the unvulcanized blend system ($S = 0.85$).

A similar modification of the first power law has been proposed by Gupta and Purwar[11] by incorporating a stress concentration factor S', where in the case of no stress concentration effect $S' = 1$ and $S' < 1$, where there is occurrence of stress concentration and is given by eq. (4).

$$\sigma = \sigma_o(1 - \phi)S' \qquad (4)$$

Comparison of theoretical curves according to eq. (4), incorporating appropriate S' values with experimental data points, are shown in Figures 12 and 13 for the unvulcanized and dynamically vulcanized blend systems, respectively. The experimental data points fall between the two extremes of theoretical curves according to eq. (4),

with $S' = 1$ and $S' = 0.65$, with a sigmoidal variation and lie close to the theoretical curve, with $S' = 1$ at low volume fraction and $S' = 0.65$ at higher volume fraction of SBS. Similar behavior is observed in the dynamically vulcanized blends with experimental data points falling between the two extremes of the theoretical curves according to eq. (4), with $S' = 1$ and 0.76 with a sigmoidal variation. The above observations indicate that in both blend systems there is a sigmoidal transition from a state of low stress concentration to a state of high stress concentration with increasing volume fraction of SBS. Comparison of stress concentration parameter S' values of the blend systems here also indicates that the stress concentration effect is less in case of the dynamically vulcanized blends than in the corresponding unvulcanized blends.

Nicolais and Narkis[12] modified the two-thirds power law by incorporating a weightage factor 1.21 [eq. (5)], which takes into account the stress concentration effect and has been shown by Piggot and Leidner[10] to be the outcome of spherical inclusions

$$\sigma = \sigma_o(1 - 1.21\phi^{2/3}) \qquad (5)$$

To determine the weightage factor, theoretical curves representing eq. (5) with the weightage factors 1.21, 1.15, and 0.85 and experimental data points of the unvulcanized blend system are shown in Figure 14, and with weightage factors
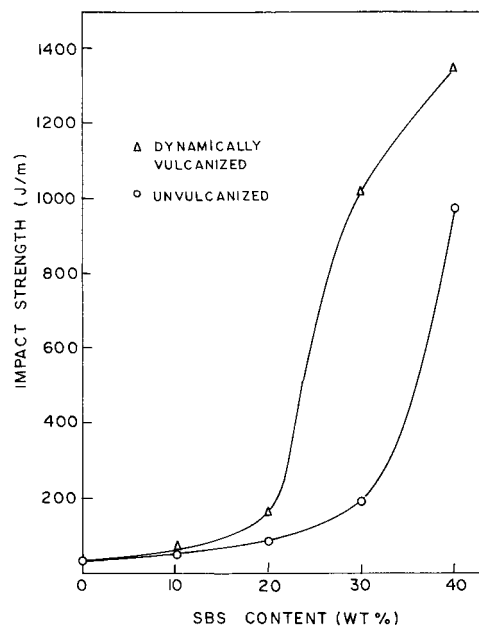
Figure 18 Variation of impact strength with blend composition in PP/SBS blends.

Figure 19   Scanning electron micrographs of the unvulcanized PP/SBS blend at different blend compositions: (a) 0 wt % SBS, (b) 10 wt % SBS, (c) 20 wt % SBS, (d) 30 wt % SBS, and (e) 40 wt % SBS.

1.21, 1.02, and 0.75 and experimental data points for the dynamically vulcanized blends are shown in Figure 15. At a low volume fraction of SBS, i.e.,

($\phi \leq 0.1$), the experimental data points fall closer to the theoretical curves with weightage factor 0.85 in the unvulcanized blends and with 0.75 in
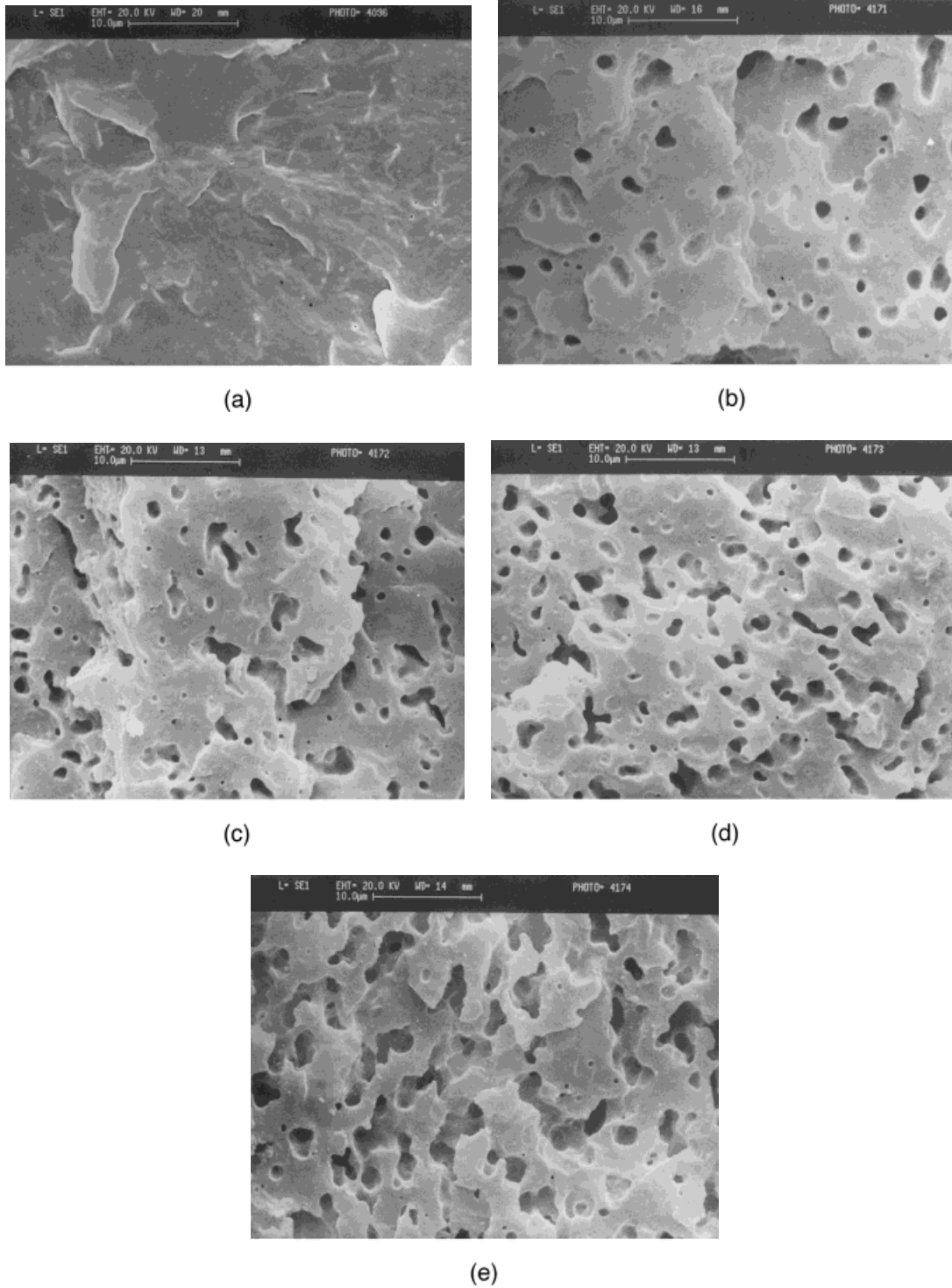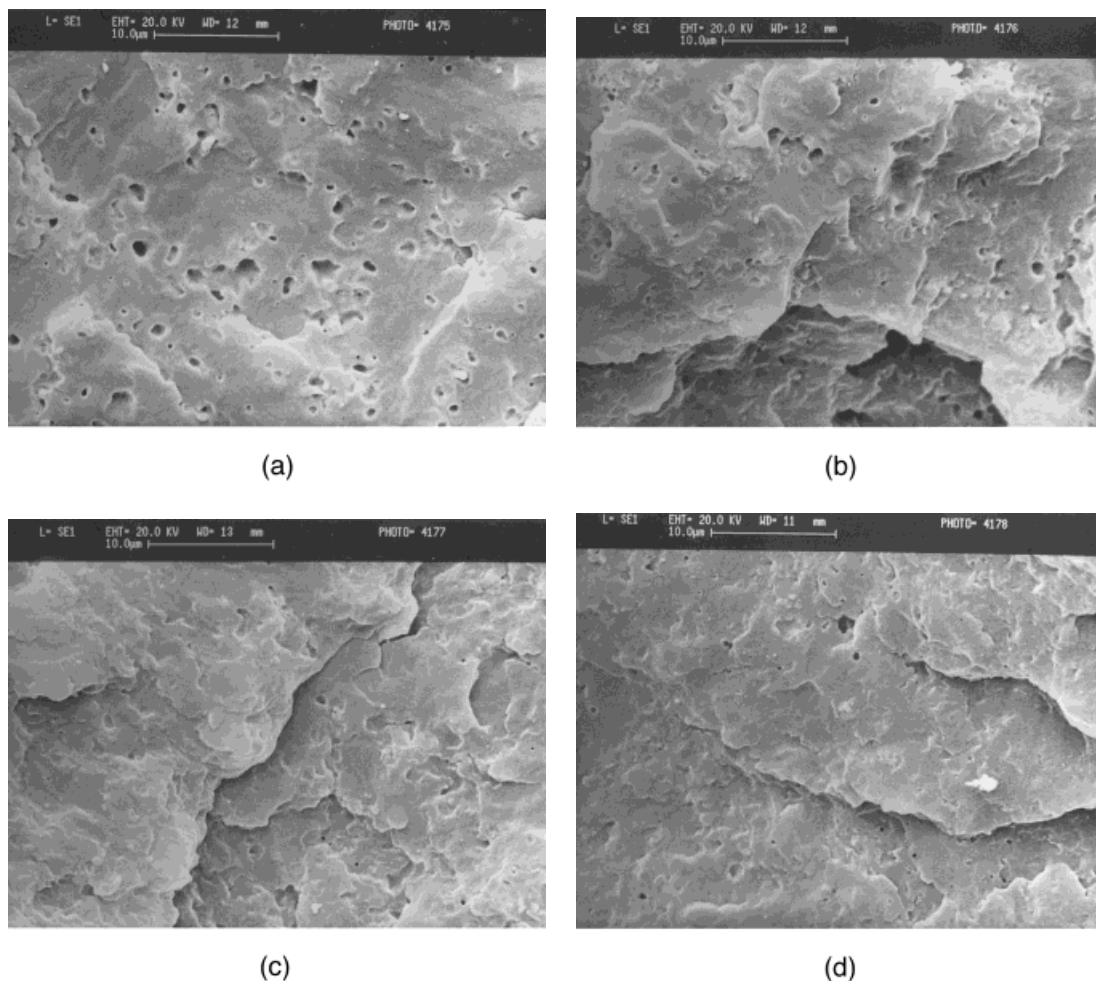
Figure 20 Scanning electron micrographs of the dynamically vulcanized PP/SBS blend at different blend compositions: (a) 10 wt % SBS, (b) 20 wt % SBS, (c) 30 wt % SBS, and (d) 40 wt % SBS.

the dynamically vulcanized blends. At a higher volume fraction of SBS ($\phi = 0.175$) the experimental data points fall close to the theoretical curves with weightage factor 1.15 in the unvulcanized blends and with weightage factor 1.02 in the dynamically vulcanized blend. Analysis of the above trends suggest that in both the blend systems there is a lesser stress concentration effect than what is accommodated by Nicolais and Narkis for spherical droplets of the dispersed phase.

Porosity Model

The porosity model, which considers a two-phase system showing discontinuity of stress transfer as a matrix with pores or voids, has been suggested by Nielsen[13] to be applicable to polymer matrix with voids or holes. This porosity concept has been

used for a two-phase blend with poor adhesion by Kunori and Geil,[9] and has also been used for the analysis of these results. According to the porosity theory, the specific change $d\ \sigma/\sigma$ in a property is directly proportional to porosity P or

$$d\sigma/\sigma = aP \tag{6}$$

where a is a proportionality constant, and a negative sign implies the mutually opposite sense of variation of P and the mechanical property σ. Replacing the total porosity by volume fraction of the inclusion ϕ in eq. (6) one obtains

$$\sigma = \sigma_o \exp.(^-a\phi) \tag{7}$$

It has been suggested[9] that the parameter a is related to the stress concentration effect in a

manner such that higher the value of a, the higher is the stress concentration and poorer is the interfacial adhesion. The expression of the porosity model [eq. (7)] fits quite satisfactorily with the experimental yield stress data with a single value of the parameter $a = 2.4$ for the unvulcanized blend system (Fig. 16) and $a = 2.05$ for the dynamically vulcanized blend system (Fig. 17) over the entire range of composition studied. This value of a is rather low compared to the value obtained for PP/polybutadiene blend (viz., 3.1),[14] implying the superiority of SBS over polybutadiene for blending with PP. A lower value of a in the dynamically vulcanized blend system indicates a higher interfacial adhesion or lower stress concentration effect than in the unvulcanized blend system and is consistent with that obtained from the stress concentration parameters of eqs. (3) and (4).

The increased adhesion in dynamically vulcanized systems can be explained on the basis of a smaller size of the dispersed rubber particles, as the mixing torque for vulcanized blends are higher than that of unvulcanized blends. Further, as has been suggested in the literature,[15] there can be a chemical graft formation or interlinking between SBS and PP during melt mixing and vulcanization that increases the interfacial adhesion and results in improvement in mechanical properties. The analysis based on the various theoretical models leads to identical conclusions and strengthens their validity.

Impact Properties

Variation of Izod impact strength of notched samples of unvulcanized and dynamically vulcanized PP/SBS blends with SBS content are shown in Figure 18. The impact strength increases gradually with increasing SBS content until 30 wt %, and much more rapidly beyond that in unvulcanized blend system. As the SBS content increases from 30 to 40 wt % in the blend, a nearly 50 times increase in impact strength of parent PP is observed. In the dynamically vulcanized blend system, the gradual increase in impact strength with increasing SBS content takes place until 20 wt % SBS content, and a steep rise is observed in the composition range of 20 to 30 wt % SBS content. This behavior can be explained by morphological studies discussed in a subsequent section.

The dynamically vulcanized blends have higher impact strength than the corresponding unvulcanized blends at all the studied compositions. At 20 wt % SBS content a threefold increase in impact strength of parent PP is observed in unvulcanized blends, while a sixfold increase is observed in the dynamically vulcanized blends. Interfacial adhesion, particle size, and volume fraction of the elastomeric component are the critical factors that affect the mechanical properties of a rubber-toughened plastic. The overall higher impact strength of dynamically vulcanized blends can be explained on the basis of its higher interfacial adhesion, as discussed in the previous section. Further, the number density of the SBS domains is more in the dynamically vulcanized blends, as is evidenced in the SEM studies, which provides larger number of potential craze initiating sites at a constant volume fraction of rubber. However, the effect of vulcanization in improvement in mechanical properties is predominant at higher SBS content due to the higher gel content.

Morphological Studies

The scanning electron micrographs of the unvulcanized blends are shown in Figure 19. The morphologies obtained are noneqilibrium morphologies and depend strongly on blend preparation. The black voids visible in these micrographs are of SBS droplets that have been dissolved out on etching with cyclohexanone. There is a considerable distribution of droplet size and shape. Small spherical droplets are depicted that are dispersed in continuous PP matrix at composition having 10 wt % SBS content. The droplets are somewhat nonspherical, and larger at blend compositions having 20 and 30 wt % SBS, where some smaller droplets retaining their shape and size can also coexist. The increase in stress concentration with increasing SBS content is in total agreement with increase in size and nonspherical shape of the rubber domains. The average droplet size in these compositions vary from 0.5 to 1.5 μm, and is well within the range of critical rubber size recognized by Jang et al.[16] However, as the SBS content increases from 30 to 40 wt %, the droplets become large and elongated, apparently formed by coalescence of several small droplets. Occurrence of coalescence at higher concentrations has been reported by many authors.[17–20] PP is clearly the continuous phase here, but the voids also seem to be interconnected, or in other words, there is a change in morphology from the dispersed phase to the interconnected phase as the SBS content increases from 30 to 40 wt % in the blend. The interconnected elastomeric phase at 40 wt % SBS content imparts high elastomeric nature to the blend and is depicted in flattened yield stress versus a yield strain curve and a remarkably high increase in impact strength at

this blend composition. The micrographs of the dynamically vulcanized blends do not depict the black voids clearly, as no extraction of the rubber phase was possible owing to its gelled nature, except in blend compositions having lower SBS and gel content (i.e., 10 and 20 wt % SBS), where spherical SBS domains in continous PP matrix are observed (Fig. 20). The number density of the domains is more, while the average size is less, in the dynamically vulcanized blend than in the corresponding unvulcanized blend, because of its higher mixing torque. The disappearance of yield peak and remarkably high impact strength of the dynamically vulcanized blend at 30 wt % SBS suggest an interconnected elastomeric phase at that composition and a change in morphology from the dispersed to the interconnected phase in the region of 20 to 30 wt % SBS content. The covalent crosslinks in the dynamically vulcanized blends, although providing distinct and stable structure to SBS particles, might get labile through heat activation or during the thermoplastic processing step and can undergo exchanges at higher concentration, resulting in coalescence and a continuous rubber phase.[21]

CONCLUSION

PP/SBS blends form a two-phase morphology with discrete spherical SBS droplets dispersed in continous PP matrix at low concentration, with an interconnected phase morphology at higher SBS concentration. There is a significant enhancement in impact strength of PP upon incorporation of SBS. Blending with SBS lowers the yield stress, increases yield strain, widens the yield peak, and increases the work of the yield of PP. The dynamically vulcanized PP/SBS blends show higher impact strength, yield stress, and percent elongation than the corresponding unvulcanized blends.

Theoretical analysis of yield stress data reveals that discontinuities in stress transfer due to stress concentration become appreciable when the SBS content exceeds 20 wt % in the blend and stress concentration effect is less in the dynamically vulcanized blends than in the corresponding unvulcanized blends. Increase in the stress concentration effect is accompanied by an increase in the size of SBS domains, as is indicated by SEM.

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