

# Gas Permeability and Mechanical Properties of Polystyrene–Polypropylene Blends

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**ABSTRACT:** Permeability to water vapor and oxygen, elastic modulus, tensile strength, and impact strength of polystyrene–polypropylene and high-impact polystyrene–polypropylene blends were determined as functions of blend composition and morphology. Three types of styrene–butadiene block copolymers were tested as compatibilizers and found to improve mechanical properties of blends. The experimental data on permeability and modulus were compared with the predictions for the studied binary and ternary blends. The predictive scheme employs a two-parameter equivalent box model and the data on phase continuity of constituents calculated using general equations derived from percolation theory. Blends with decreased permeability and plausible mechanical properties were proposed with regard to intended applications in food packaging. © 1998 John Wiley & Sons, Inc. *J Appl Polym Sci* 69: 2615–2623, 1998

**Key words:** polystyrene; polypropylene; blends; permeability; modulus; strength; equivalent model

## INTRODUCTION

A great number of articles that are devoted annually to problems of polymer blends, as well as the ever increasing quantity of various blends commercially produced, give evidence about their great importance in the field of polymer materials.<sup>1–3</sup> Because of immiscibility of most polymers, it is usually necessary to use compatibilizers in the blend preparation. Incorporation of compatibilizers in a blended system is a process that makes it possible to improve interfacial adhesion and to achieve finer phase

structures. Compatibilization is a unique procedure that allows us to prepare multiphase polymeric materials with desirable combination of physical properties.

Polystyrene–polypropylene (PP–PS) and high-impact polystyrene–polypropylene (HIPS–PP) blends seem to be promising materials applicable to various purposes in packaging and for parts with improved resistance to oils in automotive industry. An attractive feature of these blends is low permeability to water vapor in comparison with neat polystyrene and, also, higher resistance of such blends to environmental stress cracking in the presence of fats. At the same time, of course, the material requires a certain level of mechanical properties, such as stiffness and strength. These blends were studied to a smaller extent than, for example, polystyrene–polyethylene blends; there are only a few articles on the rheological behavior<sup>4–9</sup> and mechanical properties<sup>10–17</sup> of these materials, while gas trans-

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**Table I** Composition of PS–PP Blends

Sample No.	Polymer Components (%)		Compatibilizer (%)		
	PS	PP	SB	SBS	SBSBS
1	100	0	0	0	0
2	80	20	0	0	0
3	60	40	0	0	0
4	40	60	0	0	0
5	20	80	0	0	0
6	0	100	0	0	0
7	76	19	5	0	0
8	76	19	0	5	0
9	76	19	0	0	5
10	72	18	0	10	0
11	77.6	19.4	0	3	0
12	79.2	19.8	0	1	0

port in these blends has not been described in the literature so far.

Our study of PS–PP and HIPS–PP blends comprises measurement of permeability of oxygen and water vapor, measurements of basic mechanical properties, and selection of effective compatibilizers (which improve mechanical properties but do not enhance permeabilities).

Experimental data on permeability and elastic modulus are compared with the prediction of a new model and, moreover, interpreted with regard to the blend morphology, as determined by electron microscopy.

## EXPERIMENTAL

### Materials

PS: polystyrene, Krasten 151 (Kaučuk a.s., Kralupy, Czech Republic);  $M_w = 330,000$ ;  $M_n = 125,000$ .

HIPS: high-impact polystyrene, Krasten 562E (Kaučuk a.s., Kralupy, Czech Republic);  $M_w = 190,000$ ;  $M_n = 93,000$ ; 7% polybutadiene (particle size 1–10  $\mu\text{m}$ ) is dispersed in the polystyrene matrix.

PP: polypropylene, Mosten 52 492 (Chemopetrol a.s., Litvínov, Czech Republic);  $M_w = 320,000$ ;  $M_n = 51,000$ .

SB: block copolymer styrene–butadiene, Buna BL 6 533 (Bayer);  $M_w = 205,000$ ;  $M_n = 180,000$ ; 30 wt % PS content.

SBS: block copolymer styrene–butadiene–sty-

rene, Cariflex TR 1102 (Shell);  $M_w = 100,000$ ;  $M_n = 88,000$ ; 28 wt % PS content.

SBSBS: block copolymer styrene–butadiene–styrene–butadiene–styrene, TRP 752, a developmental product of Kaučuk a.s., Kralupy, Czech Republic

### Preparation of Blends and Determination of Mechanical Properties

The blends were obtained by melt mixing of components in a single-screw laboratory Goettfert extruder at 80–100 rpm. The temperatures in 3 sections of the extruder were 160, 190, and 200°C. Test specimens for mechanical testing were prepared in accordance with the ISO 294 standards by using a Battenfeld injection-molding machine (Type BA 500/200 CD). The temperature of the injected material was 200°C, the mold temperature was 60°C, the injection time was 3 s, and the total molding cycle was 1 min. Specimens were conditioned for 24 h at 23°C and 50% relative humidity. Elastic modulus and tensile strength were determined by standard methods using an Instron Tester. The composition of the prepared polymer blends is given in Tables I and II.

### Membrane Preparation and Permeability Measurements

The membranes used for the evaluation of gas permeability were prepared by the following two-step procedure: (1) a 2-mm plate was compression-molded from granules in a distance frame at 200°C; (2) after cooling, a piece of about 0.5 g was cut off the plate and put between aluminium foils

**Table II** Composition of HIPS-PP Blends

Sample No.	Polymer Components (%)		Compatibilizer (%)		
	HIPS	PP	SB	SBS	SBSBS
13	100	0	0	0	0
14	80	20	0	0	0
15	60	40	0	0	0
16	40	60	0	0	0
17	20	80	0	0	0
18	0	100	0	0	0
19	76	19	5	0	0
20	76	19	0	5	0
21	76	19	0	0	5
22	47.5	47.5	0	5	0

(with silicone-rubber-modified surface) into a Fontijne table press (200°C, 150 kN load). After 5 min, the membranes were transferred from the hot press into a cold press and slowly cooled to laboratory temperature. Under these conditions, a membrane thickness of 0.05–0.10 mm was achieved; the thickness variation with the position was negligible.

The apparatus using a modified differential method<sup>16–18</sup> consists of a cell partitioned with a flat membrane: one side of the membrane is flowed round by a carrier gas (at a constant rate), while the second side by a gas (or vapor), whose permeability is measured. The sample gas penetrated through membrane is mixed with carrier gas, and the composition of the gaseous mixture is determined on the basis of the change in thermal conductivity of the system. The change in thermal conductivity was indicated by a pair of thermistors built into the Wheastone resistance bridge as a voltage signal. The time dependence of the voltage signal is connected with the permeation flux density of the gas through the membrane, which is directly related to the transport parameters of the gas used. The apparatus was connected online with a computer to increase the productivity and reliability of measurement. The resulting value of the permeability is an arithmetic mean of three measurements.

### Electron Microscopy

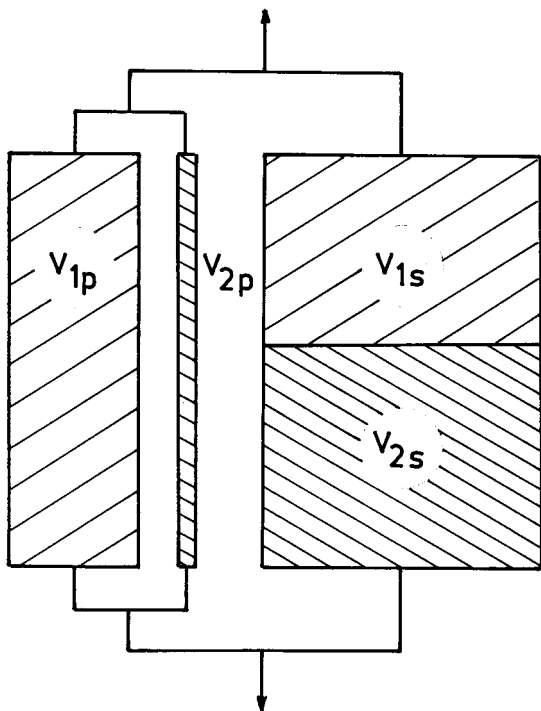
Scanning electron microscopy (SEM) observations were carried out on fracture surfaces using a JEOL JSM 6 400. Specimens were coated with a 10-nm thick layer of gold; a Balzer Sputter Coater SCD 050 was used for the purpose. The same device with an STEM adapter was used for trans-

mission electron microscopy (TEM). Samples cut from test pieces (using cryo-ultramicrotomy technique with LKB Ultratome III) were stained with osmium tetroxide vapor.

### MODEL CONSIDERATIONS

As preparation of polymer blends ranks among the cost-effective ways of upgrading existing polymers, it is desirable to anticipate the values of physical properties of intended blends, for example, modulus  $E_b$ , tensile strength  $S_{ub}$ , and permeability  $P_b$  to various gasses or vapors. Recently, a new predictive scheme has been proposed<sup>19</sup> for  $E_b$  along with  $S_{ub}$ <sup>19–21</sup> and for permeability  $P_b$ ,<sup>22</sup> which allows for (1) the respective property of both parent components, (2) a wide interval of co-continuity of phases, and (3) strength of interfacial adhesion. The prediction of physical properties is implemented in the following two steps: the equations for properties under consideration are derived using a two-parameter equivalent box model (EBM) (Fig. 1); the volume fractions  $v_{ij}$  occurring in Figure 1 are calculated with the aid of modified equations proposed by the percolation theory for the modulus of binary blends with a negligible contribution of one component.<sup>23</sup> The objective of this part of article is to correlate experimental data with simultaneous prediction of (1) the modulus and (2) permeability to oxygen or water vapor of binary blends, PS-PP, and ternary polymer blends, HIPS-PP. A significant feature of the employed predictive scheme is that all predicted properties are related to an identical phase structure (for a given blend composition).

The EBM in Figure 1 is a two-parameter model as of four volume fractions  $v_{ij}$ ; only two are inde-



**Figure 1** Equivalent box model for a binary blend, 60/40.

pendent. The fractions of components 1 and 2 coupled in parallel (subscript  $p$ ) or in series (subscript  $s$ ) are interrelated as follows:

$$v_p = v_{1p} + v_{2p}; v_s = v_{1s} + v_{2s}; v_1 = v_{1p} + v_{1s};$$

$$v_2 = v_{2p} + v_{2s}; v_1 + v_2 = v_p + v_s = 1 \quad (1)$$

The modulus of two-component blends is given as the sum of the contributions of the parallel and series branches, as follows<sup>19</sup>:

$$E_b = (E_1 v_{1p} + E_2 v_{2p}) + v_s^2 / [(v_{1s}/E_1) + (v_{2s}/E_2)] \quad (2)$$

The permeability of two-component blends is given by a formally analogous equation, as follows<sup>19–22,24</sup>:

$$P_b = (P_1 v_{1p} + P_2 v_{2p}) + v_s^2 / [(v_{1s}/P_1) + (v_{2s}/P_2)] \quad (3)$$

The following formulae for  $v_{1p}$  and  $v_{2p}$  as functions of the blend composition were derived in our previous articles<sup>19,20</sup>:

$$v_{1p} = [(v_1 - v_{1cr}) / (1 - v_{1cr})]^{t_1} \quad (4a)$$

$$v_{2p} = [(v_2 - v_{2cr}) / (1 - v_{2cr})]^{t_2} \quad (4b)$$

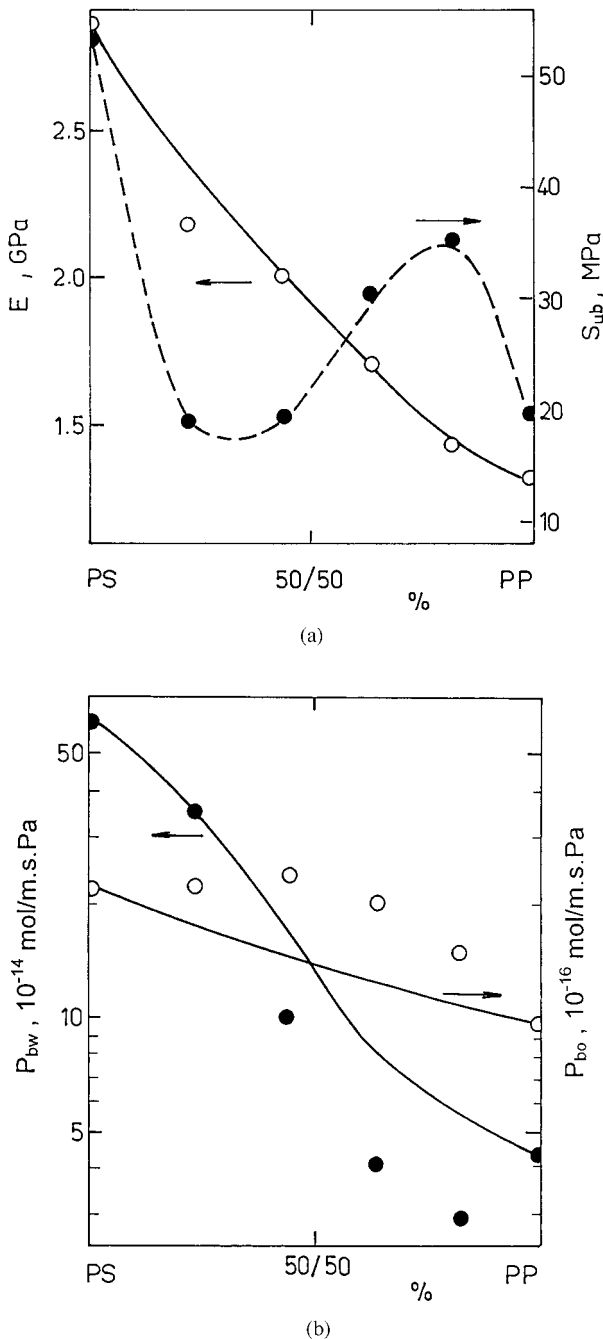
where  $v_{1cr}$  and  $v_{2cr}$  are the critical volume fractions (percolation thresholds) and  $t_1$  and  $t_2$  are the critical universal exponents. For discrete domains of spherical form,  $v_{cr} = 0.156$  was calculated.<sup>25–27</sup> An average value of  $v_{cr} = 0.19 \pm 0.09$  was reported<sup>25</sup> for several binary blends. Most experimental values of  $t$  are within the interval 1.7–1.9, which complies well with the theoretical prediction of 1.8.<sup>23</sup> In addition to  $v_{1p}$  and  $v_{2p}$ , the remaining  $v_{1s}$  and  $v_{2s}$  are evaluated using eq. (1). On the other hand, by fitting experimental data with dependences calculated over the whole composition range, it is possible to evaluate  $v_{1cr}$  and  $v_{2cr}$  for studied blends. However, experimental  $v_{1cr}$  frequently differs from  $v_{2cr}$  in binary blends so that the dependences of  $v_{1p}$  and  $v_{2p}$  on composition may not be symmetrical. Obviously,  $v_{1cr} < v_1 < (1 - v_{2cr})$  is the interval of the phase duality (co-continuity) in which the phase inversion takes place. In the marginal zone,  $0 < v_1 < v_{1cr}$  (or  $0 < v_2 < v_{2cr}$ ), where only component 2 (or 1) is continuous, a simplified relation holds for the minority component, that is,  $v_{1p} = 0$ ,  $v_{1s} = v_1$  (or  $v_{2p} = 0$ ,  $v_{2s} = v_2$ ).

## RESULTS AND DISCUSSION

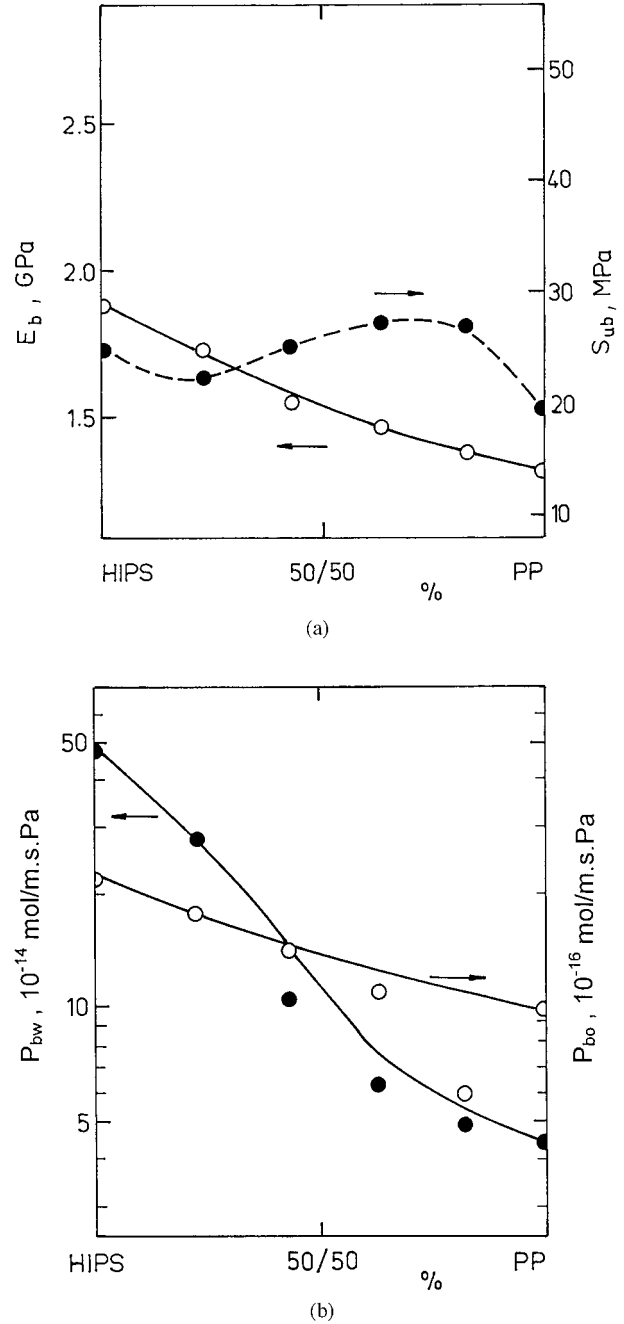
Predicted values of the modulus and permeability of the studied blends are confronted with experimental data in Figures 2 and 3. [In Figs. 2(b) and 3(b), a semilogarithmic plot of permeabilities is used because it is more instructive than the rectilinear one.] It is worth noting that complete sets of experimental data, including  $E_b$ ,  $P_b$ , and  $S_{ub}$ , which can be used for verification of the predictive scheme over the whole interval of blend compositions, are rather rare in the available literature.<sup>28,29</sup> Unfortunately, the values of  $S_{ub}$  in both series of our blends show nonmonotonic dependences on blend composition, which also holds for impact strength (Tables III and IV). Probable reasons for these irregularities can be seen in the changes of fracture mechanism. Thus,  $S_{ub}$  were ignored in the process of adjusting  $v_{1cr}$  and  $v_{2cr}$ ; consequently, no conclusions can be drawn concerning the interfacial adhesion in the studied blends.

Blends HIPS–PP should be regarded as three-component systems. As can be seen from the analysis of Figures 2 and 3, the presence of 7% of PB in PS matrix accounts for a significant decrease in the both mechanical characteristics, while changes in permeabilities are very small. This difference can be attributed to the well-known

fact that PB particles contain islands of PS so that the effective volume fraction of core-shell particles amounts to 20% (mechanical properties of



**Figure 2** Effect of composition (in vol %) of polystyrene-polypropylene blends on their (a) tensile modulus (○) and tensile strength (●), and (b) permeability for oxygen (○) and water vapor (●). Full lines correspond to eqs. (2) and (3) for  $v_{1cr} = 0.30$ ;  $v_{2cr} = 0.20$ ;  $t_1 = t_2 = 1.8$ . The dashed line indicates the experimental data on tensile strength (not correlated with the theory).



**Figure 3** Effect of composition (in vol %) of high-impact polystyrene-polypropylene blends on their (a) tensile modulus (○) and tensile strength (●), and (b) permeability for oxygen (○) and water vapor (●). Full lines correspond to eqs. (2) and (3) for  $v_{1cr} \times 0.30$ ;  $v_{2cr} \times 0.20$ ;  $t_1 = t_2 = 1.8$ . The dashed line indicates the experimental data on tensile strength (not correlated with the theory).

such particles are predetermined by those of the outer shell,<sup>30</sup> while the permeability of the particles corresponds to their actual composition). In the model calculations, HIPS is approximated by

**Table III Permeability and Mechanical Properties of PS-PP Blends**

Sample No.	Permeability (mol m <sup>-1</sup> s <sup>-1</sup> Pa <sup>-1</sup> )		Elastic Modulus (MPa)	Tensile Strength (MPa)	Impact Strength (kJ/m <sup>2</sup> )
	$P_o \times 10^{15}$	$P_w \times 10^{13}$			
1	2.22	6.18	2882	52.9	22.7
2	2.28	3.56	2174	18.9	10.9
3	2.43	0.99	2013	19.4	14.7
4	2.04	0.41	1700	30.3	20.1
5	1.50	0.29	1432	35.5	37.3
6	0.97	0.44	1330	20.0	N <sup>a</sup>
7	2.72	3.70	2033	23.3	12.5
8	2.40	3.57	1903	34.1	35.9
9	2.56	2.68	1947	41.6	22.3
10	2.56	3.75	1664	28.7	59.3
11	1.80	2.63	2062	40.1	23.5
12	2.31	2.24	2145	27.6	17.8

<sup>a</sup> N indicates that the sample was unbreakable under test conditions.

a single phase with regard to the facts that the ratio of PS to PB is constant and the phase structure of HIPS is stabilized by PB crosslinking.

In both series of blends, identical parameters  $v_{1cr} = 0.30$ ,  $v_{2cr} = 0.20$ , and  $t_1 = t_2 = 1.8$  are appropriate to fit experimental data for  $E_b$  and  $P_b$ . The difference between  $v_{1cr}$  for PS (or HIPS) and  $v_{2cr}$  for PP can be related to the well-known fact that the component with lower relative viscosity (in the blend melt) shows stronger tendency to form a co-continuous phase than the other component,<sup>25,31</sup> which is manifested in a lower  $v_{2cr}$ . (The theoretical "basic" constants  $v_{1cr} = v_{2cr} = 0.156$  do not allow eqs. (2) and (3) to fit well our experimental data.) Nevertheless, the adjusted

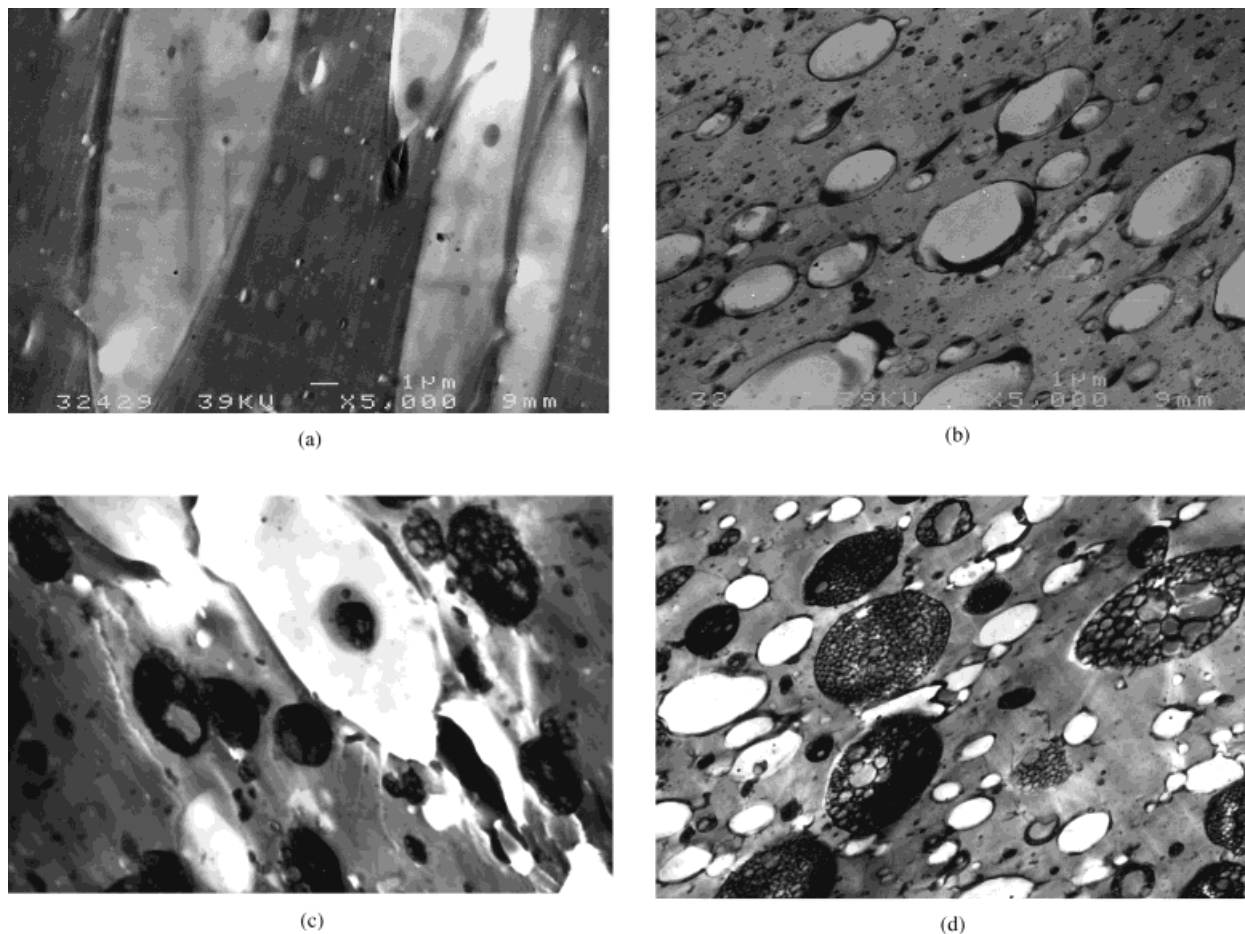
$v_{1cr}$  and  $v_{2cr}$ , which specify the interval of phase co-continuity, are only approximative because of a relatively large scatter of the experimental data on  $E_b$  and  $P_b$ .

Figures 2(a) and 3(a) concurrently show a good agreement between predicted and experimental values of moduli, which evidences regular changes in phase structure of blends with their composition. On the other hand, calculated dependences of permeabilities in Figure 2(b) do not fit well the experimental data. The decrease in  $P_b$  for water vapor with increasing PP fraction seems to be the only more profound than the predicted trend. In contrast, the observed maximum of  $P_b$  for oxygen can hardly be explained in terms of the

**Table IV Permeability and Mechanical Properties of HIPS-PP Blends**

Sample No.	Permeability (mol m <sup>-1</sup> s <sup>-1</sup> Pa <sup>-1</sup> )		Elastic Modulus (MPa)	Tensile Strength (MPa)	Impact Strength (kJ/m <sup>2</sup> )
	$P_o \times 10^{15}$	$P_w \times 10^{13}$			
13	2.19	4.84	1877	25.0	92.0
14	1.76	2.76	1730	22.5	15.9
15	1.42	1.05	1557	25.5	20.7
16	1.13	0.62	1483	27.5	25.6
17	0.59	0.49	1380	27.1	50.4
18	0.97	0.44	1300	19.7	N <sup>a</sup>
19	2.47	4.32	1505	22.9	19.8
20	1.64	3.73	1367	22.2	73.7
21	1.99	5.16	1442	24.3	53.5
22	0.89	0.61	1166	25.9	93.7

<sup>a</sup> N indicates that the sample was unbreakable under test conditions.



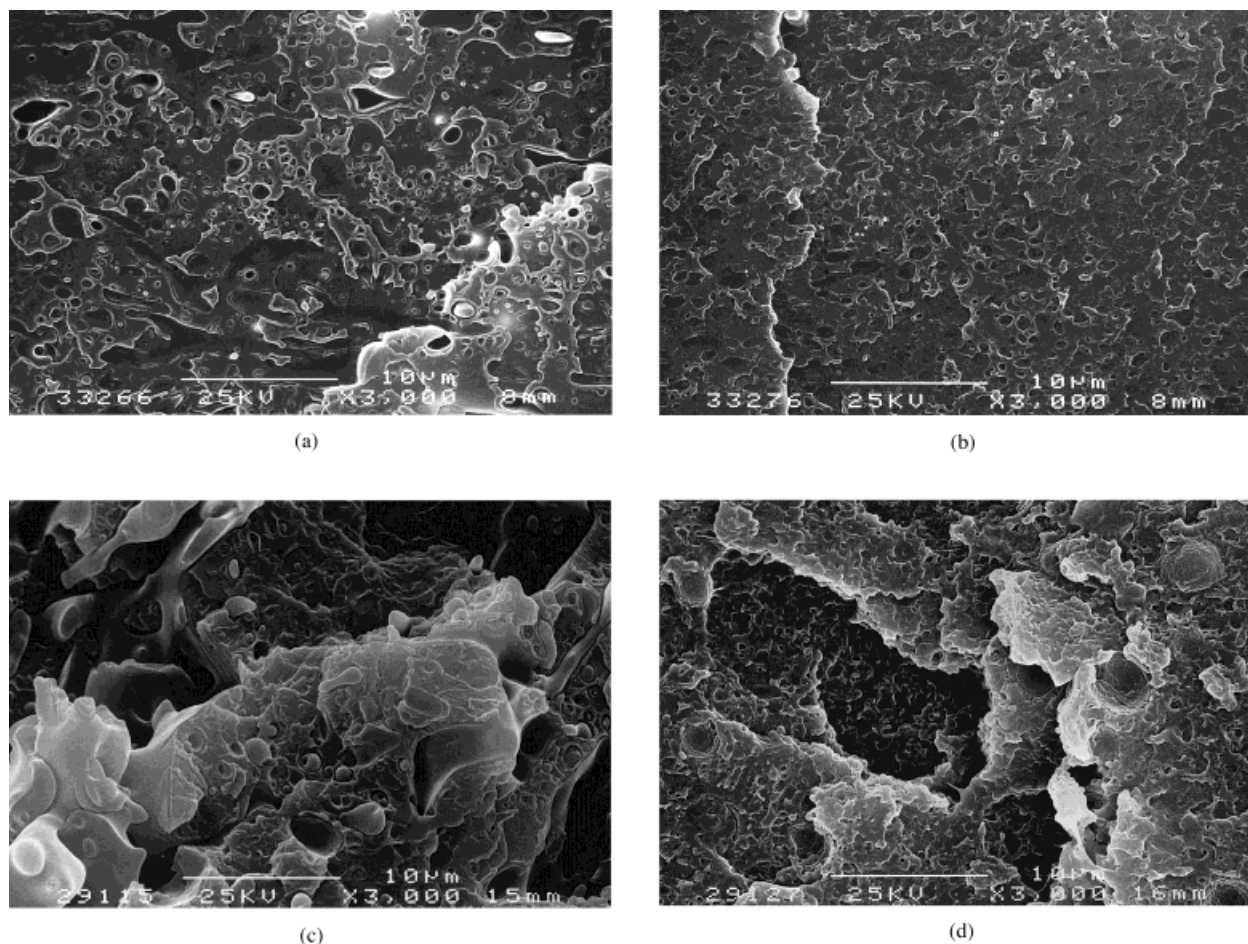
**Figure 4** TEM micrographs of polystyrene-polypropylene (4/1) blends: (a) PS-PP without compatibilizer; (b) PS-PP with triblock; (c) HIPS-PP without compatibilizer; (d) HIPS-PP with pentablock.

used model. On the other hand, in Figure 3(b), relatively good accord can be observed between all experimental and calculated values, despite the fact that ternary systems are more complicated than binary ones. Thus, Figures 3(a) and (b) document that the predictive scheme is suitable for simultaneous prediction of the modulus and permeability of heterogeneous polymer blends.

Permeabilities to oxygen and water vapor, elastic modulus, tensile strength, and impact strength of PS-PP blends containing compatibilizers are summarized in the lower part of Table III. As can be seen, the permeability of both parent polymers to oxygen is very similar and relatively low; in contrast, the permeabilities of PS and PP to water vapor are about 100 and 10 times higher, respectively, than those for oxygen. The difference in permeabilities of PS and PP makes it possible to control transport of water vapor by adjusting an appropriate ratio of PS-PP in the

blends. Thus, for instance, the difference of order in the water vapor permeability of net PS and PS-PP (40/60) blend represents a substantial improvement in the barrier characteristic of the latter material. However, the blends prepared from immiscible polymers by mixing of molten components without any compatibilizer show strong a tendency to phase structure coarsening, which may impair the end-use properties of blends. Therefore, several types of styrene-butadiene block copolymers were used for compatibilization of these polymers in order to achieve an acceptable set of mechanical and transport properties required for membranes.

The blends PS-PP (80/20), which show poor strength characteristics (in comparison with both polymer components), were compatibilized by linear styrene-butadiene block copolymers. Selected characteristics sensitive to compatibilization efficiency indicate (Table III) that addition of the SB diblock



**Figure 5** SEM micrographs of polystyrene-polypropylene (4/1) blends: (a) PS-PP without compatibilizer; (b) PS-PP with triblock; (c) HIPS-PP without compatibilizer; (d) HIPS-PP with pentablock.

affects only slightly the mentioned properties, while the multiblocks (SBS and SBSBS) significantly improve both impact strength and tensile strength. This essential fact was already pointed out in our former articles.<sup>14,15</sup> The addition of compatibilizers leads to a small decrease in modulus (as a result of increased elastomer concentration) but has no observable effect on permeability of measured gases. The change in mechanical properties is brought about by morphology change, as documented in Figure 4, showing TEM micrographs. The improvements of fineness and the uniformity of the compatibilized sample are quite evident. The relation between morphology and mechanical characteristics is seen also from SEM observation of fracture surfaces (Fig. 5) of the same samples. In the case of uncompatibilized blends, the fracture path follows the phase boundaries, while the compatibilized blends show transformed zones and fractures, predominantly in the PS matrix.

Due to a higher impact strength and elongation of HIPS in comparison with PS, the HIPS-PP system shows comparatively better properties than PS-PP (Table IV). The permeabilities to oxygen and water vapor are similar to those of PS-PP blends, despite the fact that the morphology of HIPS-PP blend is different; however, mechanical properties are strongly affected by the presence of PB particles. As a result, the basic uncompatibilized blends containing HIPS have higher impact strength, lower modulus, and tensile strength than analogous blends with PS. Incorporation of SB multiblock copolymers has effects similar to those in the previous case: impact strength is increased and modulus decreased, while tensile strength remains practically unchanged. In our opinion, blend 22 (Table IV) can serve as a sample with properly balanced properties. In comparison with HIPS, this blend shows 8 times lower permeability to water vapor and al-



most 3 times lower permeability to oxygen, while its strength characteristics correspond to those of HIPS. An inevitable decrease in elastic modulus is small and should not be a limiting factor in packaging applications.

## CONCLUSIONS

1. Compatibilization of polystyrene-polypropylene and, in particular, high-impact polystyrene-polypropylene blends with styrene-butadiene linear multiblock copolymers, makes it possible to obtain materials with (1) a higher barrier for water vapor and oxygen than original polystyrene and with (2) balanced mechanical properties required for various application in packaging and/or in selected automotive parts.
2. The modulus and permeability of binary blends PS-PP and ternary blends HIPS-PP without compatibilizers are in relatively good accordance with the dependences calculated using the predictive scheme proposed earlier,<sup>19,22</sup> which takes into account (1) respective properties of both constituents, (2) the interval of co-continuity of components (phase duality), and (3) interfacial adhesion.

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