

Ternary Polymer Blends: Prediction of Mechanical Properties for Various Phase Structures

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ABSTRACT

A predictive scheme is proposed for the simultaneous calculation of the modulus and yield (or tensile) strength of ternary polymer systems. According to the continuity or discontinuity of constituting phases, the scheme combines in two steps the models for binary systems: (i) in the interval of phase duality (co-continuity), a two-parameter equivalent box model is used along with the data on the phase continuity rendered by modified equations of the percolation theory; and (ii) the effects of a dispersed phase on the mechanical properties of a continuous phase are treated by using the approach developed earlier for particulate systems. Simultaneously predicted values of the modulus and yield (or tensile) strength of ternary systems are interrelated because they are calculated by using an identical set of input parameters characterizing a specific phase structure. The predictive scheme will allow the experimentalists: (i) to anticipate selected mechanical properties of envisaged blends (for presumed phase structures); (ii) by comparing experimental and theoretical data, to assess to which percentage the potential of a material has been exploited; (iii) to analyze the phase structure of prepared ternary blends; and (iv) to evaluate interfacial adhesion or the extent of interfacial debonding. The versatility of the predictive scheme is demonstrated on three examples of

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KEYWORDS: ternary blends; predictive models; elastic modulus; yield strength; tensile strength

INTRODUCTION

As preparation of polymer blends ranks among the cost-effective ways of upgrading existing polymers, it is very desirable to anticipate the values of physical properties of intended blends, such as modulus E_b , yield strength S_{yb} , tensile strength S_{ub} and permeability P_b to gases or vapors. So far various models predicting mechanical or other physical properties of particulate composites have also been frequently used for heterogeneous polymer blends. However, such models [1–3] are not suitable for polymer blends because (i) they do not allow for an interval of the co-continuity (duality) of constituting phases and (ii) they cannot simultaneously predict several mechanical (physical) properties of a blend, thus relating them to a specific phase structure. We have succeeded in overcoming these deficiencies by proposing a new predictive scheme [4–7] based on (i) a two-parameter equivalent box model (EBM) (Fig. 1) and (ii) the equations rendered by the percolation theory [8, 9]. As the EBMs are not “self-consistent” models, their adjustable parameters (volume frac-

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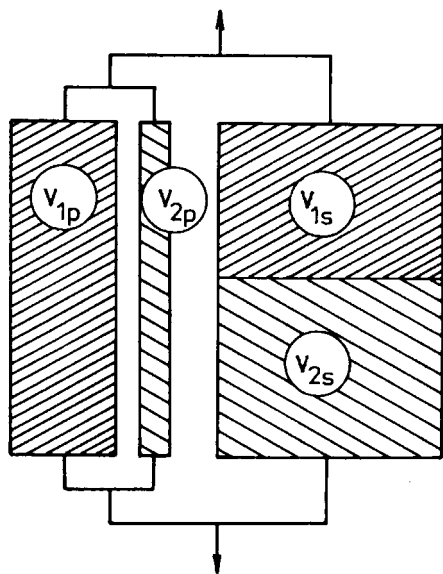


FIGURE 1. Equivalent box model for binary blends.

tions) have so far been determined *a posteriori* by fitting experimental data [3, 10]. However, the EBMs can be used for the prediction of selected physical properties of polymer blends if the input parameters are determined *a priori* with the aid of a suitable theory or model. The percolation theory [8] renders a universal equation for the modulus of two-component heterogeneous isotropic systems where the contribution of the second component (phase) is negligible. It has been shown experimentally [6, 9] that the proposed equation plausibly fits experimental data for glassy plastics/elastomer blends not only in the vicinity of the percolation threshold v_{cr} (the critical volume fraction) at which the glassy polymer assumes phase continuity, but also over the region $v_{cr} < v < 1$. So far the predictive scheme has been successfully used for (i) prediction of the yield strength of binary polymer blends [4, 7], (ii) evaluation of the extent of interfacial debonding in binary polymer blends [5], and (iii) simultaneous prediction of the modulus, yield (or tensile) strength and permeability of binary polymer blends [7, 11].

By using "theoretical" values of the input parameters (calculated for single-size spheres in a three-dimensional lattice [8, 12–15]) we can obtain "universal" dependences of physical properties on blend composition. Alternatively, if experimental data are available (e.g. on the modulus and/or yield strength and/or permeability), v_{cr} of constituents can be adjusted (by a fitting procedure), which may be rather different from the theoretical value being affected by relative melt viscosities of components, interface energy, mixing equipment and conditions, phase structure coarsening, etc. In this way, the proposed scheme becomes a powerful tool in obtaining quantitative information on the phase structure of studied blends. As all predicted properties are interrelated through an identical set of input parameters, the identification of a phase

structure becomes more trustworthy the more measured properties of blends can be compared with their simultaneous prediction, e.g. modulus and yield (or tensile) strength or modulus and permeability.

The objective of this paper is to show how the predictive scheme can be adapted for the prediction of selected mechanical properties of various types of heterogeneous ternary polymer systems (consisting of three immiscible or partly miscible components). A continuing search for cost-effective materials has stimulated studies of three-component blends which provide broader options of tailoring for a variety of specific purposes. Also, we have found [16, 17] that mechanical—in particular, ultimate—properties of ternary blends are superior to those of binary counterparts. Prediction of the physical properties of ternary blends is clearly more difficult than for binary blends for at least two reasons: (i) phase structure of ternary blends is more complex and variable; and (ii) the EBM applied to binary blends has to be extended and/or combined with other models in a suitable manner. We will show that the predictive procedure can be implemented in two steps: (i) properties of a two-component blend (or composite) can be calculated where one component is continuous, while the second component is either continuous or discontinuous; and (ii) this binary system can be then formally "mixed" with the third (co-continuous) component to form a three-component heterogeneous system. We believe that the proposed predictive scheme will allow the experimentalists (i) to anticipate selected mechanical properties of envisaged blends (for presumed phase structures); by comparing experimental and theoretical data (ii) to assess to which percentage the potential of a material has been exploited; (iii) to analyze the phase structure of prepared ternary blends; and (iv) to evaluate interfacial adhesion or the extent of interfacial debonding. The versatility of the predictive scheme is demonstrated on three examples of various types of ternary systems.

MODEL APPROACH

At present, there exist various models predicting individual physical properties of systems consisting of a continuous matrix and one or more dispersed components, e.g. modulus [2, 10, 18–20], yield or tensile strength [21–25] and permeability [26–28]. We can employ such models for polymer blends in marginal composition intervals in which the minority components are fully discontinuous. However, we need models that allow for the interval of phase duality (co-continuity) delimited by the critical volume fractions v_{1cr} and v_{2cr} of the constituents and take into account their interfacial adhesion. Our predictive scheme for the modulus and yield (or tensile) strength of such systems requires implementation of two steps: (i) derivation of an equation for a considered property in terms of the EBM; and (ii) calculation of the input para-

meters (which are identical for all considered properties) of the EBM by using the percolation approach introduced earlier [8, 9]. Obviously, the EBM approach is not applicable if the blend mixing process accounts for a significant change in the structure, e.g. in the degree of crystallinity, of a component or gives rise to a new mechanism, e.g. multiple crazing of a matrix containing elastomeric inclusions (thus, impact strength cannot be predicted by the scheme).

Equivalent Box Model for Systems with Co-continuous Phases

The well-known parallel and series models [2, 18] are sometimes incorrectly used as first approximations of the upper and lower bounds, respectively, of physical properties of isotropic heterogeneous materials (cf. ref. 7), such as modulus, permeability, yield strength and tensile strength. In principle, phase structure of these materials does not correspond to parallel or series coupling of components, but more complex models combining both couplings have to be introduced. The EBM in Fig. 1 is a two-parameter model since of the four volume fractions v_{ij} , only two are independent. The dimensions of blocks indicate which volume fractions of each constituent can be regarded as coupled in parallel or in series so that the EBM response to loading can be equivalent to that of the modeled system. The fractions of components 1 and 2 coupled in parallel (subscript p) or in series (subscript s) are interrelated as follows:

$$\left. \begin{aligned} v_p &= v_{1p} + v_{2p} \\ v_s &= v_{1s} + v_{2s} \\ v_1 &= v_{1p} + v_{1s} \\ v_2 &= v_{2p} + v_{2s} \\ v_1 + v_2 &= v_p + v_s = 1 \end{aligned} \right\} \quad (1)$$

The following equations hold [4-6] for the parallel and series branches of the EBM:

$$E_p v_p = E_1 v_{1p} + E_2 v_{2p} \quad (2a)$$

$$\frac{v_s}{E_s} = \frac{v_{1s}}{E_1} + \frac{v_{2s}}{E_2} \quad (2b)$$

The resulting modulus of two-component blends is then given as the sum ($E_p v_p + E_s v_s$):

$$E_b = E_1 v_{1p} + E_2 v_{2p} + \frac{v_s^2}{(v_{1s}/E_1 + v_{2s}/E_2)} \quad (3)$$

As we have shown earlier [6, 29], the blend modulus is always a monotonic function of the blend composition because no interfacial debonding is presumed at small strains at which the blend moduli are routinely measured. It is worth mentioning that the permeability of two-component blends is given [7, 30] by a formally analogous equation.

A linear stress-strain relationship indispensable

for modulus measurements can be granted for glassy polymers only at very low strains, typically below 1%, where perhaps all blends show interfacial adhesion sufficient for the transmission of the acting (very low) stress. At higher strains (usually 4-6%), the applied tensile stress is likely to exceed the linearity limit and attain the value of yield strength S_y , thus inducing yielding and plastic deformation. In our previous papers [4-7], we have derived the following equation for the yield strength of polymer blends in terms of the EBM visualized in Fig. 1:

$$S_{yb} = S_{yp} v_p + S_{ys} v_s =$$

$$(S_{y1} v_{1p} + S_{y2} v_{2p}) + A S_{y1} v_s \quad (4)$$

where S_{y1} and S_{y2} characterize the parent polymers and A is inversely proportional to the extent of interfacial debonding [5]. Two limiting values of S_{yb} , identified with the lower or upper bound, can be distinguished by means of eq. (4): (i) Interfacial adhesion is so poor that complete debonding occurs between the fractions of constituents coupled in series ($A=0$ at the yield stress); consequently, the series branch does not contribute to the resulting yield strength and the lower bound of S_{yb} is therefore equal to the sum of the contributions of two parallel elements. (ii) Interfacial adhesion is strong enough to transmit the achieved stress between constituents so that no debonding appears ($A=1$); then the contribution of the series branch is added to that of the parallel branch (the effect on S_{y1} and S_{y2} of slightly different strain rates in the parallel and series branches is neglected). However, as two components differing in the yield strength are coupled in series, the branch shows yielding at S_{y1} or S_{y2} , whichever is lower ($S_{y1} < S_{y2}$ is assumed in eq. (4)). For $A=1$, the EBM predicts a monotonic dependence of S_{yb} in the interval between S_{y1} and S_{y2} ; however, as soon as $A < 1$, S_{yb} passes through a minimum as a function of blend composition [4, 5, 30]. With regard to general experience [2, 23] that formally identical equations can be used for the yield as well as tensile strength of isotropic polymer materials, we have also successfully used [4-7] eq. (4) for S_{ub} by replacing the yield strengths S_{y1} and S_{y2} by the tensile strengths S_{u1} and S_{u2} , respectively.

Calculation of v_{ij} for the EBM employs a universal percolation formula derived [8, 31] for the modulus of binary systems with negligible contribution of one component:

$$E = E_0 (v - v_{cr})^t \quad (5)$$

where E_0 is a constant, v_{cr} is the critical volume fraction and t is the critical universal exponent. As eq. (5) has been shown [6, 9] to plausibly fit the modulus of model blends with $E_1 \gg E_2$ in the range $v_{1cr} < v_1 < 1$, it can be modified to the following form:

$$E_{1b} = E_1 \left(\frac{v_1 - v_{1cr}}{1 - v_{1cr}} \right)^{t_1} \quad (6)$$

where $E_1 = E_0(1 - v_{1cr})^{t_1}$ is the modulus of the neat component 1 and E_{1b} expresses the modulus of a "single-component" blend in which the second component occupies space corresponding to its volume fraction, but its contribution to the blend modulus is negligible. As long as $E_1 \gg E_2$, the contribution $E_2 v_{2p}$ of that part of component 2 which is coupled in parallel and the contribution of the whole series branch (Fig. 1) to the modulus of the EBM (eq. (3)) are negligible in comparison with the contribution $E_1 v_{1p}$ of component 1.

Consequently, $E_1 v_{1p}$ (or $E_2 v_{2p}$ for $E_2 \gg E_1$) can be set equal to the apparent modulus E_{1b} (or E_{2b}):

$$E_{1b} = E_1 v_{1p} \quad (7a)$$

$$E_{2b} = E_2 v_{2p} \quad (7b)$$

To obtain v_{1p} and v_{2p} as functions of the blend composition, we will combine eqs (6) and (7):

$$v_{1p} = \left(\frac{v_1 - v_{1cr}}{1 - v_{1cr}} \right)^{t_1} \quad (8a)$$

$$v_{2p} = \left(\frac{v_2 - v_{2cr}}{1 - v_{2cr}} \right)^{t_2} \quad (8b)$$

The remaining v_{1s} and v_{2s} can be evaluated by using eqs (1). Experimental values of t are usually located between 1.7 and 1.9, which complies well with the theoretical prediction [8] $t = 1.8$; the latter value will be considered in our calculations as a constant. For discrete domains of spherical form, the percolation threshold $v_{cr} = 0.156$ was calculated [12, 31, 32]. The values of $v_{cr} = 0.19 \pm 0.09$ were reported [19] for components in binary blends.

Models for Systems with One Continuous and One Discontinuous Phase

The effect of particulate fillers on the modulus of isotropic composites with glassy matrix can be evaluated by using the Kerner-Nielsen equation [2, 10, 33]:

$$E_c = E_m \left(\frac{1 + A_f B_f v_f}{1 - P_f B_f v_f} \right) \quad (9)$$

where E_c is the modulus of the composite, E_m is the modulus of the matrix and v_f is the volume fraction of the filler. The constants A_f , B_f , P_f are defined [2] as follows:

$A_f = (7 - 5\nu_m)/(8 - 10\nu_m)$, where ν_m is the Poisson ratio of the (glassy) matrix;

$B_f = (E_f/E_m - 1)/(E_f/E_m + A_f)$, where E_f is the modulus of the incorporated filler;

$P_f = 1 + [(1 - v_{fmax})/v_{fmax}] (v_f)^2$, where v_{fmax} is the maximum packing fraction of the filler (the

value of $v_{fmax} = 0.63$ for random close packing of monodisperse spheres is frequently used [2]).

If the modulus E_i of the dispersed component is lower than E_m , e.g. in rubber-toughened plastics, then inverted relations should be used [2]:

$$E_s = E_m \left(\frac{1 - P_i B_i v_i}{1 + A_i B_i v_i} \right) \quad (10)$$

where E_s is the modulus of the system and v_i is the volume fraction of the inclusions. The constants A_i , B_i , P_i are defined [2] as follows:

$$A_i = (8 - 10\nu_m)/(7 - 5\nu_m);$$

$$B_i = (E_m/E_i - 1)/(E_m/E_i + A_i);$$

the formulae for P_i and P_f are analogous.

As generally known, the yield strength of particulate composites S_{yc} is approximately equal to that of the matrix S_{ym} if the interfacial adhesion is strong enough. (Detailed calculations based on the method of final elements show [34] that S_{yc} is slightly higher than S_{ym} mainly because of reduced molecular mobility in the interphase adhering to the filler surface.) In the case of "zero" adhesion, S_{yc} drops with the filler volume fraction following approximately the formula derived by Smith [21] and modified by Nicolais and Narkis [23]:

$$S_{yc} = S_{ym} \left[1 - \left(\frac{v_f}{v_{fmax}} \right)^{2/3} \right] \quad (11)$$

The same formula holds for glassy matrices with rubber-like inclusions [10] regardless the adhesion between components.

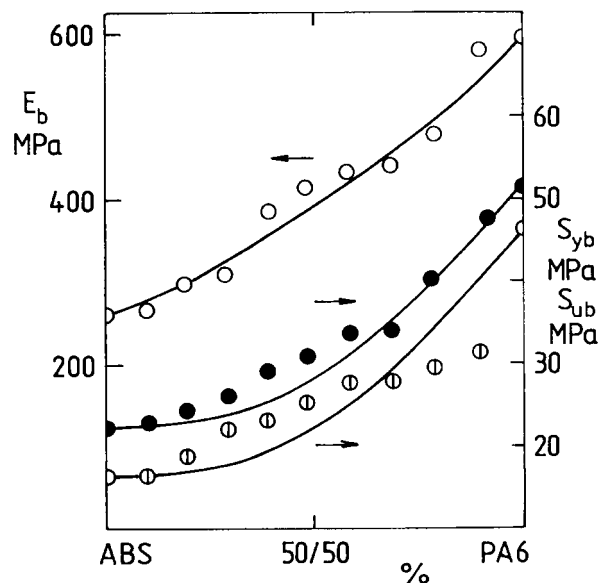


FIGURE 2. Effect of the composition of ABS/polyamide 6 (PA6) blends on \circ tensile modulus E_b , \bullet yield strength S_{yb} and \odot tensile strength S_{ub} (data from ref. 35). Lines calculated by using "universal" input parameters

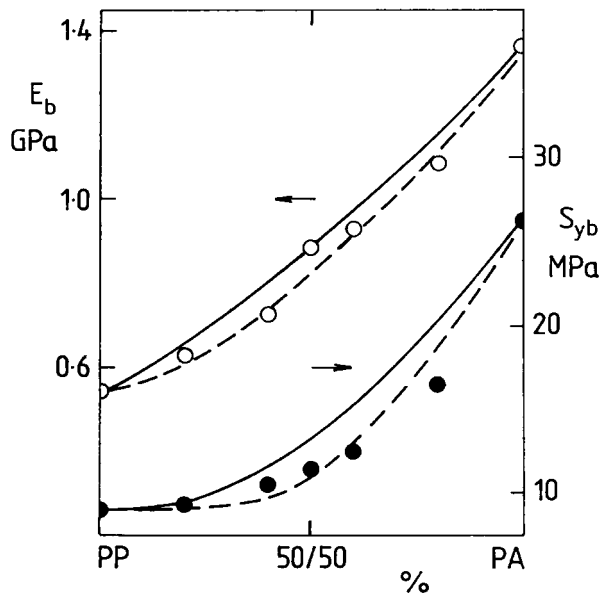


FIGURE 3. Effect of the composition of polypropylene (PP)/polyamide 66 (PA)/glass beads (GB) blends on tensile modulus E_b and yield strength S_{yb} (data from ref. 36). Solid lines calculated by using "universal" input parameters $v_{1cr} = v_{2cr} = 0.16$; $t_1 = t_2 = 1.8$; dashed lines calculated with $v_{2cr} = 0.30$ (the other parameters unchanged).

RESULTS AND DISCUSSION

In this paper, we will compare simultaneously predicted E_b and S_{yb} (and/or S_{ub}) with experimental data for various types of ternary systems in order to demonstrate the versatility of the proposed predictive scheme. In Fig. 2, experimental data [35] on E_b , S_{yb} and S_{ub} are presented for the blends of ABS and polyamide 6 (PA6). ABS is a two-phase system (blend) consisting of continuous poly(styrene-co-acrylonitrile) matrix (PSAN) and rubber-like particles of butadiene. It would be possible to evaluate the properties of ABS by using eqs (10) and (11) if the corresponding properties of the two constituents were given. However, as the volume fractions of the components as well as the ABS phase structure are fixed and do not change during the blend mixing, experimental values of E_b , S_{yb} and S_{ub} found [35] for ABS can be used directly, thus eliminating the first step in the predictive scheme. The second step in predicting the properties of ABS/PA6 blends is analogous to that used for binary blends [4, 7, 11]. The curves of E_b and S_{yb} calculated by using theoretical values $v_{1cr} = v_{2cr} = 0.16$ fit experimental data with a plausible accuracy. S_{ub} of blends with 30–60% of PA6 is somewhat higher than that predicted, while the inverse relation holds for the blends with 80 and 90% of PA6. Nevertheless, the monotonic dependence of S_{ub} on the blend composition evidences—according to the EBM prediction [30]—sufficient interfacial adhesion between ABS and PA6 up to the blend fracture. Thus, in this case, the predictive

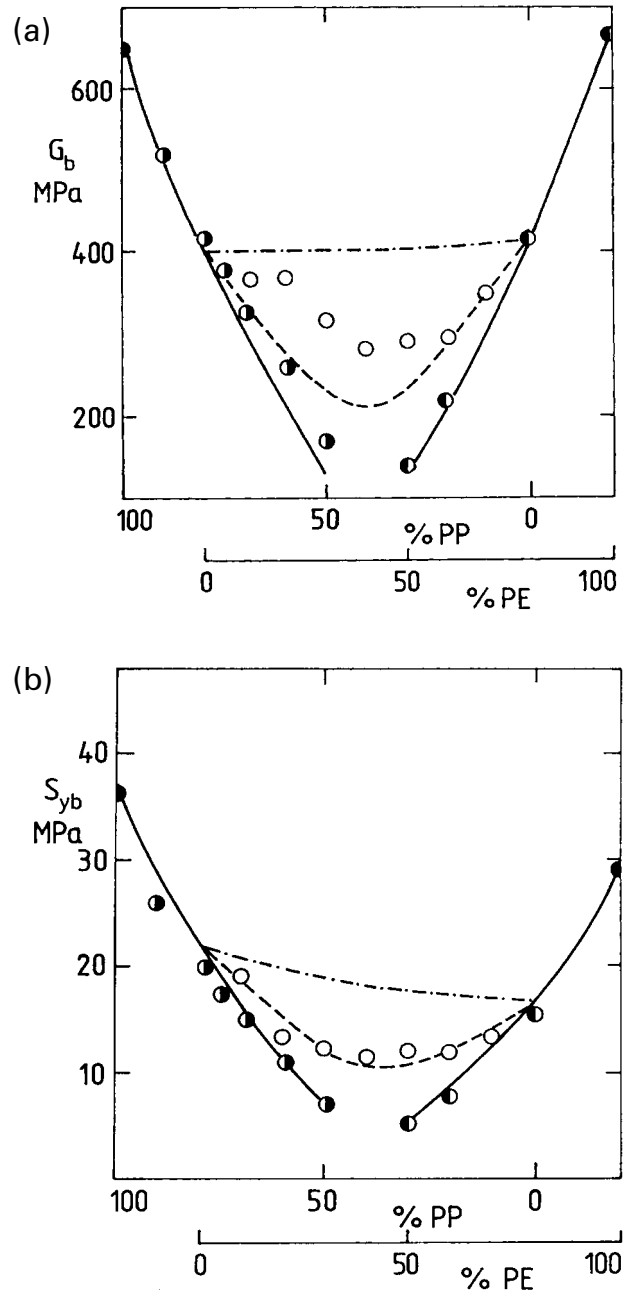


FIGURE 4. Effect of the composition of polypropylene (PP)/polyethylene (PE)/ethylene-propylene rubber (EPR) blends on (a) shear modulus G_b and (b) yield strength S_{yb} . Experimental data (from ref. 33): ● PP/EPR; ● PE/EPR; ○ PP/PE/EPR. — dependences for binary blends calculated by using "universal" input parameters $v_{1cr} = v_{2cr} = 0.16$; $t_1 = t_2 = 1.8$; - - - EPR is presumed to form an interlayer between PP and PE in ternary blends (the input parameters as above); - · - EPR is presumed to be evenly distributed in PP and PE in ternary blends (the input parameters as above).

scheme anticipates quite well the experimental results.

The second example (Fig. 3) presents a ternary system [36] where a constant volume fraction (10%) of glass beads (GB) was added to a fused binary matrix consisting of varying fractions of polyamide 66 (PA) and polypropylene (PP). Microscopic analysis [36] has shown that GBs were always

embedded in the PA component of the blends despite the fact that PP contained a small percentage of maleic anhydride, which simultaneously contributed to the adhesion between PP and PA. The prediction of mechanical properties of the PP/PA/GB systems requires implementation of two steps: (i) calculation of respective properties of the PA (present in a specific blend) reinforced by added GB; (ii) calculation of properties of the systems consisting of PP and reinforced PA.

The effect of GBs on the modulus of PA can be evaluated by using eq. (9) and the following values for the components [2, 36]: $E_m = 1.10$ GPa; $E_f = 70$ GPa; $\nu_m = 0.35$; $\nu_{fmax} = 0.63$. As far as the yield strength is concerned, we can assume [34] $S_{yc} = S_m$ (owing to sufficient interfacial adhesion) if the volume fraction of PA in the matrix PA/PP is higher than 0.1. In the second step, the mixing of the composite PA/GB with PP is regarded as analogous to the mixing of polymers. As can be seen in Fig. 3, the curves of E_b and S_{yb} calculated under the assumption $v_{1cr} = v_{2cr} = 0.16$ do not fit the experimental data well; however, an optimum description has been found for $v_{1cr} = 0.16$ and $v_{2cr} = 0.35$. A high value of v_{2cr} can be explained by the fact that GBs markedly increase the melt viscosity of PA, which then shows a decreased tendency to phase continuity in the blended system [1, 37, 38]. Anyway, Fig. 3 documents that E_b and S_{yb} calculated for the PP/PA/GB systems by using the outlined procedure can be viewed as a valuable preliminary information.

The third example (Fig. 4) is concerned with ternary blends PP/PE/EPR containing a constant volume fraction $v_3 = 0.2$ of ethylene-propylene rubber (EPR). It has been shown [39–42] for ternary systems that the components with the highest and the lowest surface energy do not go into contact because their interface would enormously increase the Gibbs energy of the system related to the interface. As the surface energy of EPR is higher than that of PP, but lower than that of PE [33], ternary blends tend to assume the following phase structures: if volume fraction v_1 of PP or v_2 of PE is small, say lower than 0.2, then particles of the minority component are surrounded by an EPR shell and embedded in the majority component; if v_1 and v_2 are comparable, then PP and PE are co-continuous, which, however, means that also EPR forms a co-continuous phase (as an interlayer). Microscopic analysis [33] has corroborated the fact that EPR shows a very strong tendency to separate PP and PE regardless of their ratio in ternary blends.

The phase structure of such ternary blends will change with the blend composition in a rather complex manner, i.e. various types of phase structures are likely to coexist. Model calculations implemented for several presumed phase structures may be helpful for the phase structure analysis because the model phase structures can be expected to correspond to the real ones in the composition intervals where the calculated curves fit the experimental data. An extended EBM

suitable for modeling the PP/PE/EPR blends can be obtained by adding the third element (corresponding to the EPR component) to both parallel and series branches of the EBM in Fig. 1. Equations (2) then assume the following form (tensile modulus E is replaced by shear modulus G):

$$G_p v_p = G_1 v_{1p} + G_2 v_{2p} + G_3 v_{3p} \quad (12a)$$

$$v_s/G_s = v_{1s}/G_1 + v_{2s}/G_2 + v_{3s}/G_3 \quad (12b)$$

As the values $G_3 = 1$ MPa and $S_{y3} = 0.18$ MPa are much lower than $G_1 = 650$ MPa and $S_{y1} = 36.3$ MPa or $G_2 = 670$ MPa and $S_{y2} = 28.8$ MPa, respectively, it is quite evident that the contributions $G_3 v_{3p}$ and $S_{y3} v_{3p}$ of the EPR fraction v_{3p} to G_b and S_{yb} , respectively, are negligible. Moreover, any series combination of components encompassing an EPR fraction will display G_s and S_{ys} close to G_3 and S_{y3} , respectively, so that the contribution to the resulting G_b and S_{yb} will be negligible (regardless of the adhesion at the formed interfaces and the exact value of v_{3s}). Thus, the mechanical properties of the PP/PE/EPR blends are mainly controlled by the fractions v_{1p} and v_{2p} coupled in parallel.

In Fig. 4, the data are given for the ternary blends along with those for binary blends PP/EPR and PE/EPR. It is shown that G_b and S_{yb} of the binary blends are well described by eqs (3) and (4), respectively, if theoretical value 0.16 is used for all critical volume fractions. Model calculations of G_b and S_{yb} for ternary blends have been attempted under the following assumptions: (i) PP and PE are mixed as a binary system and EPR is considered as an "interlayer" which does not affect v_{1p} and v_{2p} (such a phase structure is in accord with previous discussion); and (ii) EPR is evenly distributed throughout the blend, i.e. both PP and PE contain 20% of EPR; binary blends PP/EPR and PE/EPR are then mixed in various proportions to form ternary blends. As can be seen in Fig. 4, the second assumption leads to unrealistic (high) values of G_b and S_{yb} ; as G_1 and G_2 or S_{y1} and S_{y2} are rather similar, the calculated dependences are very flat. On the other hand, the values of S_{yb} calculated under the first assumption fit experimental data very well (Fig. 4b). As far as the blend modulus is concerned, the experimental data are somewhat higher than the model ones, but the dependences are parallel (Fig. 4a). Obviously, EPR entrapped in PP or PE, i.e. not located in the interlayer, will account for an increase in G_b or S_{yb} . In this way, we can explain the experimental data located between the curves corresponding to the model phase structures. Nonetheless we can conclude that the model with an EPR interlayer between PP and PE fits available experimental data quite well, which is in conformity with microscopic observations.

CONCLUSIONS

Modulus and yield (or tensile) strength of ternary polymer blends have been simultaneously predicted over the whole composition range with the

aid of the proposed predictive scheme which takes into account (i) respective properties of constituents, (ii) phase continuity of each component and (iii) interfacial adhesion. According to the continuity or discontinuity of constituting phases in ternary systems, the scheme combines in two steps the models for binary systems: (i) in the interval of phase duality (co-continuity), a two-parameter equivalent box model is used along with the data on the phase continuity rendered by modified equations of the percolation theory; and (ii) the effect of a dispersed phase on the mechanical properties of a continuous phase is treated by using an approach developed earlier for particulate systems. Simultaneously predicted values of the modulus and yield (or tensile) strength of a blend are interrelated since they are calculated for a presumed phase structure by using an identical set of input parameters. Predicted patterns for E_b (or G_b) and S_{yb} are in a fairly good accord with experimental data for three different types of ternary systems characterized by sufficient interfacial adhesion. Besides, the fitting of experimental data with the dependences calculated under various structural assumptions is an efficient tool for identification and quantitative analysis of formed phase structures.

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