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SIMULTANEOUS PREDICTION OF THE MODULUS, TENSILE STRENGTH AND GAS PERMEABILITY OF BINARY POLYMER BLENDS

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Abstract—An original scheme is used for simultaneous prediction of the modulus, yield (or tensile) strength and permeability of binary polymer blends. The scheme employs a two-parameter equivalent box model and the data on the phase continuity of components rendered by general equations of the percolation theory. The scheme takes into account (i) respective properties of constituents, (ii) interval of phase duality (co-continuity) and (iii) interfacial adhesion. The upper and lower bounds of the yield or tensile strength are distinguished which are related, respectively, to the interfacial adhesion sufficient and insufficient for the transmission of the tensile stress up to yielding or fracture. Simultaneously predicted values of the modulus, tensile strength and permeability of a blend are related to the same phase structure through an identical set of input parameters. Calculated dependences are in a reasonable accord with two sets of experimental data from literature. © 1998 Elsevier Science Ltd. All rights reserved

INTRODUCTION

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, e.g. modulus E_b, yield strength S_{yb}, tensile strength S_{ub} and permeability P_b for gases or vapours. At present, there exist various models predicting individual properties of composite systems consisting of a continuous matrix and one or more dispersed (discontinuous) components, e.g. modulus [1–6], yield or tensile strength [7–12] and permeability [13–18]. Some of these models are also applicable to polymer blends, but only in the marginal composition intervals in which the minority component is discontinuous [4, 5]. However, recent studies [19, 20] have shown that the minority component in two-component heterogeneous blends may become continuous at volume fractions as low as $0.1 < v < 0.2$. Moreover, a dispersed polymer component showing yielding and plastic deformation is very different from an inorganic filler having "infinite" modulus and "zero" permeability or plastic deformation. Thus, it is evident that models predicting physical properties of polymer blends should allow for (i) the respective property of both components, (ii) a wide interval of co-continuity of phases (in which phase inversion takes place) and (iii) the strength of interfacial adhesion.

Recently we have proposed [21–23] a predictive scheme for (i) E_b along with S_{yb} (or S_{ub}) [24] and for (ii) permeability P_b [25] where all these requirements are taken into consideration. The scheme is based on the combination of a two-parameter equivalent box model (EBM, Fig. 1) and the concept of phase continuity. This concept was used with regard to the fact that there is much experimental evidence [1, 24–28] that elastic, yield, ultimate and transport properties of blends are profoundly affected by the degree of continuity of constituents, although these properties are related to the phase structure in diverse ways. So far, a single-parameter EBM has been generally viewed as a convenient framework for systematic phenomenological description of elastic behaviour of various particulate systems [29–32]. A two-parameter model analogous to that in Fig. 1 was later used [33] for discussing elastic properties of phase continuity in polyethylene/polycarbonate blends. Parameters, i.e. volume fractions, of the EBMs were adjusted by fitting experimental data because the EBMs are not self-consistent models.

Thus, if the EBM is intended for the *prediction* of physical properties of blends, the procedure requires two steps: (1) to derive the equations for the properties under consideration; (2) to calculate the volume fractions v_{ij} (Fig. 1) with the aid of an appropriate model or theory. In our previous papers [22–25] we have shown that v_{ij} can be evaluated with the aid of modified equations proposed by the percolation theory [34, 35] for the modulus of binary systems with a negligible contribution of one component. Our approach seems to be useful for at least two reasons: (i) the equations for v_{ij} are simple and independent of each other; (ii) the interval of the co-continuity of phases is delimited by the critical volume fractions v_{1cr} and v_{2cr} of the constituents. The objective of this paper is to show that the two-parameter equivalent box model combined with the percolation data on phase continuity can be used for simultaneous prediction of (i) the mod-

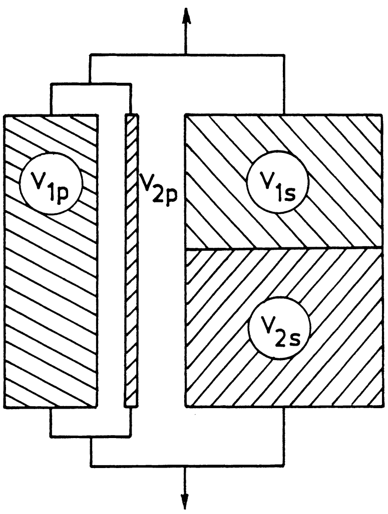

Fig. 1. Equivalent box model for a model binary blend 60/40.

ulus, (ii) permeability and (iii) the upper and/or lower bounds of yield (or tensile) strength of binary polymer blends. An essential feature of the proposed predictive scheme is that all considered properties of a blend are related to the same phase structure through an identical set of input parameters.

MODEL CONSIDERATIONS

Equivalent box model: equations for modulus, permeability and yield (or tensile) strength

The concept of the "phase continuity" used in this paper is related to the well-known parallel and series models [1, 2]. These simple models are sometimes incorrectly used as first approximations of the upper and lower bounds, respectively, of physical properties of isotropic heterogeneous materials, such as modulus [26, 28, 36, 37], permeability [38–40], yield strength [36, 37, 41, 42] and tensile strength [1, 37]. As generally known, phase structure of these materials cannot be modelled by a simple parallel or series coupling of components, but more complex models combining both couplings are required (Fig. 1). The blocks are presumed to have mechanical (physical) properties of the corresponding components; 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 modelled system. The EBM can be used for the calculation of physical properties of polymer blends under the assumption that the structure and properties of constituents are identical with those of parent polymers. The EBM is difficult to employ for the prediction of blend properties if the mixing process produces a significant change in the properties of a constituent (e.g. due to changes in crystallinity) or new mechanisms (e.g. enhancement of toughness due to the matrix multiple crazing induced by elastomer inclusions). The EBM in Fig. 1 is a two-parameter model as of four volume fractions v_{ij} only two are independent. The fractions of either component coupled in parallel (subscript p) or in series (subscript s) are interrelated as follows:

$$v_{1p} = v_1 C_1; \quad v_{1s} = v_1(1 - C_1); \tag{1a}$$

$$v_{2p} = v_2 C_2; \quad v_{2s} = v_2(1 - C_2), \tag{1b}$$

where C_1 and C_2 are the phase continuity parameters. (It should be noted that "phase continuity" or "phase connectivity" may have different meaning in other papers, e.g. in [19, 20, 35, 43].) Besides, the following relations hold:

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

The moduli of the parallel and series branches of the EBM are expressed [22–24] by following equations:

$$E_p = (E_1 v_p + E_2 v_{2p})/v_p; \tag{3a}$$

$$E_s = v_s/[(v_{1s}/E_1) + (v_{2s}/E_2)]. \tag{3b}$$

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

$$E_b = (E_1 v_{1p} + E_2 v_{2p}) + (v_s^2/[(v_{1s}/E_1) + (v_{2s}/E_2)]). \tag{4}$$

The permeability of two-component blends is given [25] by a formally analogous equation:

$$P_b = (P_1 v_{1p} + P_2 v_{2p}) + v_s^2/[(v_{1s}/P_1) + (v_{2s}/P_2)]. \tag{5}$$

"Perfect" adhesion between constituents and a linear stress–strain relationship indispensable for modulus measurements can be granted for glassy polymers only at very low strains, typically below 1%, where practically all blends show interfacial adhesion sufficient for the transmission of the acting (very low) stress. At higher strains (usually 4–6%), applied tensile stress may exceed the linearity limit and attain the value of yield strength, thus inducing yielding and plastic deformation of constituents. In our previous papers [22–24] we have derived the following equation for S_{yb} (or S_{ub}) of the EBM visualized in Fig. 1:

$$S_{yb} = (S_{y1} v_{1p} + S_{y2} v_{2p}) + A S_{y1} v_s, \tag{6}$$

where S_{y1} and S_{y2} characterize the parent polymers and A the extent of interfacial debonding. Two limiting values of S_{yb}, identified with the lower or upper bound, can be distinguished by means of Equation (6): (i) Interfacial adhesion is so weak 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 contributions of two parallel elements. (ii) Interfacial adhesion is strong enough to transmit the acting 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 of slightly different strain rates in the parallel and series branches on S_{y1} and S_{y2} is neglected). However, if two components differing in the yield strength are coupled in series, then the branch will yield at S_{y1} or S_{y2}, whichever is lower [$S_{y1} < S_{y2}$ is assumed in Equation (6)].

As demonstrated for various types of binary blends [24], the blend modulus is always a monotonic function of the blend composition because no interfacial debonding occurs at small strains at which the blend moduli are routinely measured. For $A = 1$, the EBM also 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 [22, 23]. With regard to previous experience [1, 9] that formally identical equations of various types have been used for the evaluation of yield as well as tensile strength of particulate systems, also we have successfully used [22–24] Equation (6) for S_{ub} by replacing the yield strengths S_{y1} and S_{y2} by the tensile strengths S_{u1} and S_{u2}, respectively. The applicability of the EBM for the estimate of S_{ub} implies that fracture mechanisms in individual components in blends are the same as in parent polymers.

Calculation of volume fractions in the EBM

The second step of the predictive scheme is the evaluation of v_{ij} defined in Fig. 1. Percolation theory [34, 44] provides a universal formula for the elastic modulus of binary systems where the contribution of one component is negligible:

$$E = E_o(v - v_{cr})^t, \tag{7}$$

where E_o is a constant, v_{cr} is the critical volume fraction (the percolation threshold), and t is the critical universal exponent [34]. Equation (7) was shown [35] to plausibly fit the modulus of model blends with $E_1 >> E_2$ in the range $v_{1cr} < v_1 < 1$. Thus, Equation (7) can be modified to the following form:

$$E_{1b} = E_1[(v_1 - v_{1cr})/(1 - v_{1cr})]^{t_1}, \tag{8}$$

where $E_1 = E_o(1 - v_{1cr})^{t_1}$ is the modulus of the neat component 1 and E_{1b} expresses the modulus of a "single-component" blend in which the component 1 assumes the same phase structure as in the blend with another polymer.

The assumption of the percolation theory that the second component occupies space corresponding to its volume fraction but has a negligible modulus is also convenient for the evaluation of v_{ij}. (Obviously, the phase structure of a solid blend is independent of the ratio E_1/E_2 which varies with temperature.) If $E_1 >> E_2$, the contribution E_2v_{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 [equation. (4)] are negligible in comparison to the contribution E_1v_{1p} of component 1. Consequently, E_1v_{1p} (or E_2v_{2p} for $E_2 >> E_1$) can be set equal to the apparent modulus E_{1b} (or E_{2b}):

$$E_{1b} = E_1v_{1p}; \tag{9a}$$

$$E_{2b} = E_2v_{2p}. \tag{9b}$$

To obtain v_{1p} and v_{2p} as functions of the blend composition, we will combine Equations (8) and (9):

$$v_{1p} = [(v_1 - v_{1cr})/(1 - v_{1cr})]^{t_1}; \tag{10a}$$

$$v_{2p} = [(v_2 - v_{2cr})/(1 - v_{2cr})]^{t_2}. \tag{10b}$$

The remaining v_{1s} and v_{2s} are evaluated using Equation (2).

For discrete domains of spherical form, the percolation threshold $v_{cr} = 0.156$ was calculated [19, 44]. The average value of $v_{cr} = 0.19 +/- 0.09$ was reported [19] for components in binary blends; however, v_{1cr} frequently differs from v_{2cr}. Most experimental values of t are located in a narrow interval of 1.7–1.9, which complies with the theoretical prediction [34] $t = 1.8$ used also in our calculations as the universal value. Obviously, $v_{1cr} < v_1 < (1\text{-}v_{2cr})$ 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, simplified relations hold for the minority component, i.e. $v_{1p} = 0$, $v_{1s} = v_1$ (or $v_{2p} = 0$, $v_{2s} = v_2$). In general, predicted patterns may not be in a good accord with experimental data if "universal" values $v_{1cr} = v_{2cr} = 0.16$ are used. On the other hand, Equations (4)–(6) were found [22–25] to fit experimental data with remarkable accuracy if v_{1cr} and v_{2cr} were properly adjusted. In this way we can obtain realistic values of v_{1cr} and v_{2cr} characterizing the studied blend.

RESULTS AND DISCUSSION

Recently we have successfully applied the outlined scheme to simultaneous prediction of (i) E_b and S_{yb} or S_{ub} [24, 45] and (ii) E_b and P_b [46]. In this paper, simultaneous prediction of E_b, S_{ub} and P_b of binary blends is confronted with experimental data in Figs 2 and 3. It is to be noted that complete sets of experimental results (over the whole composition range) suitable for the verification of the predictive scheme are very rare in the available literature. Thus, we used the data from [47] and [48] in this paper, although they are not best suited for our purpose.

Experimental data [47] on E_b and S_{ub} [Fig. 2(a)] of the blends of polyamide 6 (PA6) and polypropylene (PP) are somewhat higher in the interval 20 < % PP < 70 than those predicted. However, functionalized PP (containing 0.4 wt % of maleic anhydride) was admixed to PP in a percentage proportional to the volume fraction of PA6 in blends; thus, in the blends PP/PA6 = 90/10 or 20/80, the PP phase contained 1.1 or 40%, respectively, of functionalized PP. As the modulus of PP rises with the content of carboxyl groups [49, 50], it is understandable that experimental values of E_b are located above the dependence (4) calculated with E_1 for neat PP. The observed positive deviation cannot be ascribed to mere strengthening of interfacial ad-

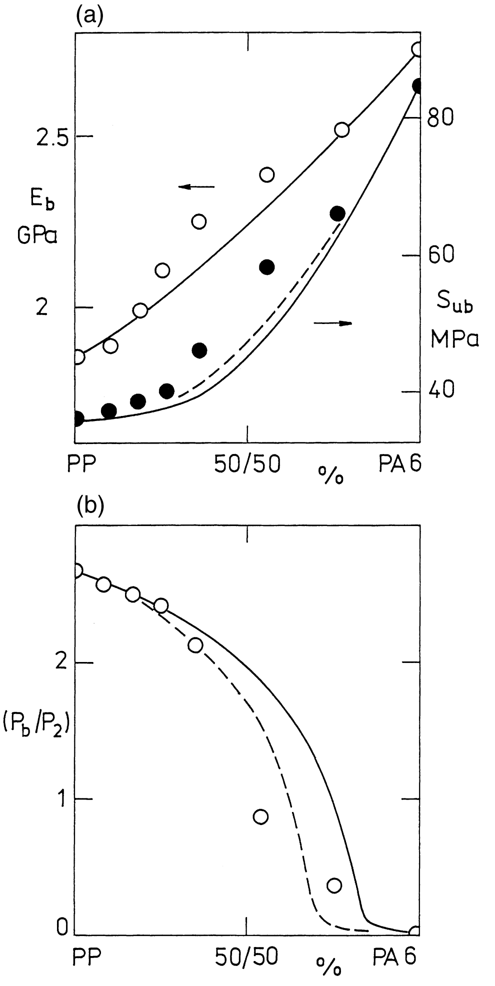

Fig. 2. Effect of composition of polypropylene (PP)/polyamide 6 (PA6) blends on their (a) tensile modulus E_b and tensile strength S_{ub} and (b) relative permeability P_b/P_2 to oxygen (data from [47]). Prediction of Equations (4), (6) and (5): full lines correspond to "universal" input parameters $v_{1cr} = v_{2cr} = 0.16$; $t_1 = t_2 = 1.8$; dashed lines calculated with an adjusted value of v_{1cr} = 0.30 (the other parameters unchanged).

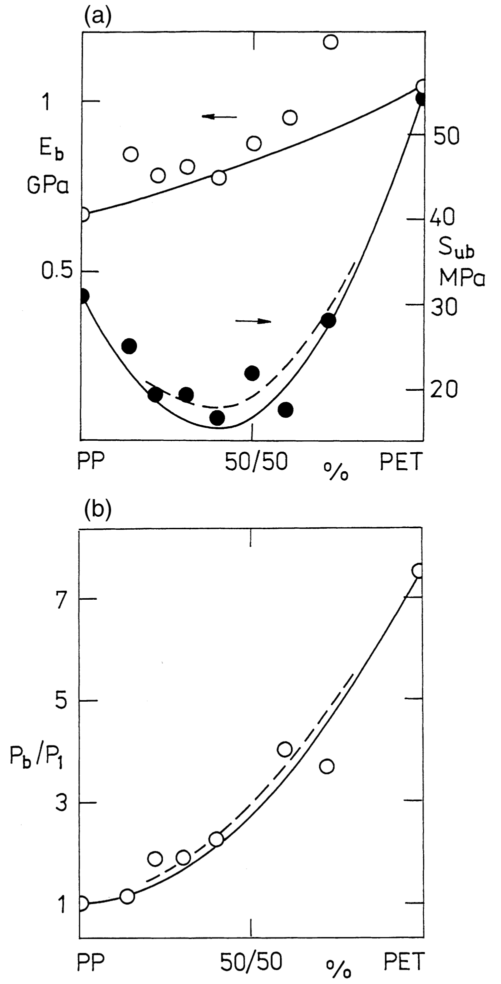

Fig. 3. Effect of composition of polypropylene/poly(ethylene terephthalate) (PET) blends on their (a) tensile modulus E_b and tensile strength S_{ub} and (b) relative permeability P_b/P_1 to water vapour (data from [48]). Prediction of Equations (4), (6) and (5): full line corresponds to "universal" input parameters $v_{1cr} = v_{2cr} = 0.16$; $t_1 = t_2 = 1.8$; dashed lines tentatively calculated with an adjusted value of $v_{1cr} = 0.08$ (the other parameters unchanged).

hesion because the modulus of blends is routinely measured at very small strains where no interfacial debonding occurs. The dependence of S_{ub} on blend composition approximately follows the upper bound given by Equation (6) for A = 1, which means that the interfacial adhesion is strong enough to transmit the stress up to the break. Higher experimental values of S_{ub} for PP/PA6 = 65/35 and 45/55 are obviously related to the increased percentage of carboxyl groups in the PP phase (*cf.* ref. [50]).

While PP has lower modulus and tensile strength than PA6 [Fig. 2(a)], an opposite relation holds for the permeability to oxygen [Fig. 2(b)]. As PP and PA6 are polymers of very different permeabilities [47], the semilogarithmic plot of relative permeability of their blends [Fig. 2(b)] is more instructive than the rectilinear one. Equations (4)–(6) show that v_{1cr} affects the considered property more in the interval $v_2 > v_1$ than in $v_1 > v_2$ (analogous relations are valid for v_{2cr}). This also means that the adjustment of v_{cr} by fitting experimental data is less accurate for the component with a lower value of the considered physical property. Thus, v_{1cr} of PP can be better adjusted with the aid of P_b [Fig. 2(b)] than of E_b or S_{ub} [Fig. 2(a)]. Relative permeability predicted for $v_{1cr} = 0.16$ is too high in the interval 50 > % PP > 0; much better accord between experiment and theory is achieved under the assumption that $v_{1cr} = 0.30$ (further refinement of v_{1cr} is difficult because of insufficient number of data in the mentioned interval). As expected, this rise in v_{1cr} does not perceptibly affect predicted dependence of E_b, while S_{ub} is only slightly enhanced [Fig. 2(a)]. Thus, we can conclude that the critical volume fractions of PP and PA6 in their blends $v_{1cr} = 0.30$ and $v_{2cr} = 0.16$, respectively, allow us to reasonably describe experimental data on E_b, S_{ub} and P_b by Equations (4)–(6).

The experimental data published [48] for blends PP/poly(ethylene terephthalate) (PET) are replotted against volume fractions in Fig. 3. Unfortunately, the values of modulus are so inconsistent that they are irrelevant in the confrontation of experiment and theory. In contrast to Fig. 2, S_{ub} passes through a deep minimum at PP/PET = 60/40 and relative permeability P_b/P_1 to water vapour rises with decreasing percentage of PP in blends. The adjustment of v_{1cr} for PP is rather difficult because PP has lower values of all considered properties than PET, which means that predicted dependences are more sensitive to the adjustment of v_{2cr} of PET. Experimental data on S_{ub} and P_b/P_1 are very well approximated by the curves calculated by using "universal" values of the input parameters, i.e. $v_{1cr} = v_{2cr} = 0.16$ and $t_1 = t_2 = 1.8$. Only a slightly better accord between experimental and calculated dependences of S_{ub} is achieved with $v_{1cr} = 0.16$ and $v_{2cr} = 0.08$; however, this difference seems to be in contradiction with the original paper [48] claiming that PP had a lower relative melt viscosity than PET. As generally known, the component with a higher relative viscosity in the melt shows a weaker tendency to the formation of a (co)continuous phase, which is manifested by a higher v_{cr} [29, 43]. As $A = 0$ is to be used in fitting experimental data for S_{ub}, it can be inferred that complete interfacial debonding occurs before the rupture. Except for enormously scattered data on modulus, the predictive scheme conforms fairly well with the experimental data.

CONCLUSIONS

The modulus, permeability and tensile strength of binary polymer blends have been for the first time simultaneously predicted over the whole composition range with the aid of the proposed predictive scheme. The procedure employs (i) the equations derived for considered properties in terms of the two-parameter equivalent box model and (ii) the phase continuity parameters of constituents derived from a general formula rendered by the percolation theory [34, 35]. The scheme takes into account (i) respective properties of both constituents, (ii) the interval of cocontinuity of components (phase duality), (iii) interfacial adhesion sufficient or insufficient for the transmission of the stress corresponding to yield or tensile strength. Predicted patterns for the modulus, permeability and tensile strength are in a fairly good accord with experimental data (selected from literature) for two types of binary blends characterized by either strong or poor interfacial adhesion. In the first approximation, physical properties of blends can be predicted by using the "universal" parameters proposed by the percolation theory. Conversely, by fitting experimental data with calculated dependences it is possible to ascertain more realistic critical volume fractions of components specifying the interval of phase duality. It should be noted that all considered physical properties of a series of blends are calculated by using the same set of input parameters; consequently, once v_{1cr} and v_{2cr} have been adjusted, e.g, by fitting experimental data on modulus, other physical properties of the blends can be predicted with a better accuracy than in the case of using the universal values $v_{1cr} = v_{2cr} = 0.16$.

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