A model for the yield strength of binary blends of thermoplastics

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A model procedure has been proposed to predict the yield strength of binary blends of thermoplastics on the basis of the information on the continuity of phases acquired from the Hill model for the elastic properties of two-component systems. Utilizing this information in an equivalent box model and assuming either 'perfect' or 'zero' interfacial adhesion, the upper and lower bounds of the yield strength can be calculated. The upper bound may be close to (but lower than) the dependence corresponding to the additivity (rule of mixtures). The lower bound passes through a minimum at a composition of about 50/50 (by volume), which corresponds to the phase-inversion point in the Hill model. The minimum yield strength is linked to the minimum of the sum of the continuity parameters of the components in the blends. Predicted dependences of the yield strength on the blend composition are in reasonable accord with the experimental data for blends with good or poor interfacial adhesion. If the model is to be employed for the appraisal of the interfacial adhesion in polymer blends, other factors affecting the phase structure should be taken into account, e.g. relative melt viscosities, particle size, orientation during processing, cavitation, etc.

(Keywords: polymer blends; yield strength; box models)

INTRODUCTION

Predictions of mechanical properties of multicomponent materials are very valuable because unnecessary experiments can be avoided, or at least their number can be reduced. At present, the modulus of various multiphase systems can be predicted[1-4] with reasonable accuracy owing to the fact that there exist numerous models relating the modulus to particular phase structures so that a suitable model can usually be found. (Conversely, if the prediction based on a specific model fits the experimental data, one can expect that the structure of the studied material is similar to that assumed in the model that is being used[5,6].) The yield strength of particulate composites is frequently predicted by using relatively simple formulae[2,7-9], which take into account the reduction of the effective cross-section of the matrix due to the presence of poorly adhering particles of dispersed phase(s). Pukanszky and coworkers[8,9] have shown that various degrees of interfacial adhesion can be explicitly encompassed by means of an exponential term. These models have also been used for blends[2,5,8-11], but none of them can estimate the yield strength in the region of the inversion or the co-continuity of the phases which is frequently encountered in blends of thermoplastics. Moreover, it is not clear how the yield strength of blends will depend on their composition in the case of 'perfect' interfacial adhesion. As is generally known, it is difficult to anticipate the impact properties of polymer blends because of the complexity of the processes underlying the fracture phenomena (associated, e.g. with multiple crazing or shear yielding[2,12,13]) and because of their sensitivity to particular aspects of the various structures.

There is considerable experimental evidence[2,12-15] to show that the phase structure of polymer blends simultaneously affects their elastic, yield and ultimate properties, although these properties are related to the phase structure in very different ways. For example, matrix phase continuity markedly affects all of the mechanical properties of polycarbonate (PC) blends[10,16-21] with poly(styrene-*co*-acrylonitrile) (PSAN) or poly(methyl methacryl(ate) (PMMA). This is because the PC matrix is clearly superior to that of the PSAN or PMMA as far as tensile and impact strengths are concerned. The ultimate properties of these blends decrease with decreasing matrix volume fraction and deteriorate at compositions where the PC phase loses its continuity, thus confirming the fact that the degree of the matrix phase continuity is of primary importance.

The continuity of the components (which are frequently identical with the phases) in these blends is routinely but only qualitatively documented by scanning or transmission electron microscopy. Therefore, other ways are sought which can supplement the microscopic evidence and provide at least semiquantitative data on phase structures. For this purpose, the existing models of the elastic properties of particulate systems can be exploited. In our previous paper[6] on polyethylene (PE)/elastomer/polypropylene (PP) ternary blends, comparison of the model modulus calculations with the experimental data supported the opinion that the ethylene–propylene (EPR) elastomer separated the PP and PE phases, regardless of their ratio in the blends. (The phase structure of ternary

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systems is dictated by the surface energies of the components[22–26].) Using similar models and some simplifying assumptions, we were able to approximately evaluate the percentage of filler particles encapsulated by the elastomer in polypropylene/elastomer/filler ternary composites[26–28]. In our more recent work[21], we tried to quantify the differences in the matrix phase continuity, C_m, between (i) two-phase blends (where phase inversion takes place at around the 50/50 volume ratio) and (ii) three-phase blends (containing two minority phases separately dispersed in a matrix) with the aid of the Hill[12,29] and Kerner–Nielsen[2,4] equations, respectively, for the shear moduli of these systems. The very limited number of available experimental data indicated some correlation between C_m and the tensile energy to break or strain at break, which is in conformity with earlier observations[10,16–18].

As polymer blends represent one of the most cost-effective ways of upgrading existing polymers, it is desirable to foresee the limits of their mechanical properties, such as modulus, yield strength, tensile strength, tensile energy to break, etc., which can be achieved for the systems under consideration. The objective of this paper is to utilize the data on the continuity of phases (cf. ref. 21) implicitly comprised in the Hill model[12,29] (predicting the modulus of two-component systems) for the calculation of the upper and lower bounds of the yield strength of binary blends of thermoplastics over the whole range of compositions. These bounds are tentatively related to the (i) 'perfect' or (ii) 'zero' interfacial adhesion between components in the 'equivalent box model' employed in this paper.

MODEL CONSIDERATIONS

Polymer blends consisting of immiscible (or partly miscible) components are typical isotropic heterogeneous materials. Their mechanical properties depend not only on the volume fractions and the respective properties of the matrix and other components, but also on phase structure, continuity of phases, interfacial adhesion, and other factors. To illustrate the term 'phase continuity', we can refer to the well known parallel and series models (*Figure 1*), which are sometimes used as first approximations of the upper and lower bounds of various mechanical properties of isotropic heterogeneous materials, e.g. modulus[3,11,12,30,31], yield strength[10,11,18,31,32], tensile strength[2,31], etc. The parallel coupling of components implies that (i) the strain of all of the phases is equal (the isostrain model) and (ii) the contribution of each phase to the final value of a certain mechanical property is given by the rule of mixtures. The constituent phases are continuous in the direction of the acting force and respond to loading in the same way as the 'neat' parent materials. Therefore, the phase continuity[21], c_p, of each phase can be considered as being equal to 1. Any line of force passes through only one phase and does not cross any interface; in this direction, therefore, the mechanical properties of the system are independent of the interfacial adhesion.

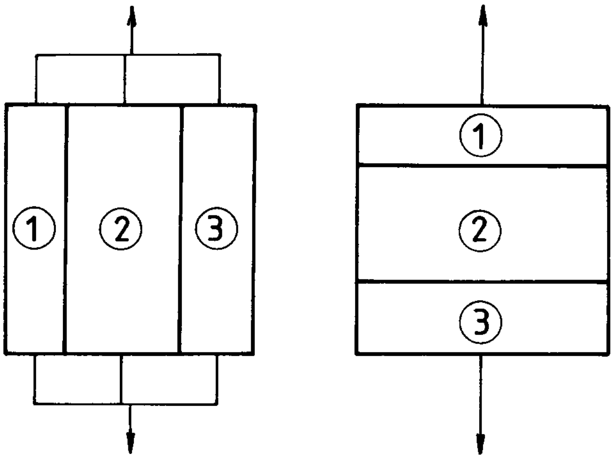

Figure 1 Parallel (left) and series (right) models for the coupling of components in binary blends

The lower bound of the above mentioned mechanical properties is given by the series coupling of the components (*Figure 1*, right) where all lines of force pass through all phases which are discontinuous in the direction of the acting force (isostress model). Therefore, the continuity of any phase in the direction of the acting force can be regarded as being equal to 0. The contributions of the individual phases to a system property are given by the inverted rule of mixtures. As all stress (load) is transmitted via the interfaces, interfacial adhesion between the constituents is of primary importance. Obviously, if a mechanical property of one of the components, or the adhesion at any interface approaches a 'zero' value, the resulting property of the series system drops to zero.

Equivalent box model

The models visualized in *Figure 1* are adequate for, e.g. orthotropic laminae, multilayer materials, etc., some of the mechanical properties of which are habitually described (in a certain direction) by the rule of mixtures[1,2]. The mechanical properties of isotropic heterogeneous materials cannot be accurately represented by such simple models, and somewhat more complex models are needed which combine the parallel and series coupling of the various components[2,3,30,31,33–35]. Although such a combined model does not correspond to the actual structure of a blend, its mechanical response to loading is equivalent to that of the modelled blend. For two-component blends, we can tentatively introduce an 'equivalent box model' (EBM), as visualized in *Figure 2*. In this model, each block has the mechanical properties of one of the blend constituents. The blocks indicate which fractions of each component can be regarded as being coupled in parallel or in series in relation to the acting force. Thus, an EBM consists of both a parallel branch and a series branch, which are themselves coupled in parallel.

Equivalent mechanical models furnish a convenient framework for systematic phenomenological descriptions of blend behaviour but they do not provide any prediction of mechanical properties which is based on the knowledge of the volume fractions and the respective properties of the constituents. Their adjustable parameters have to be determined by means of a fitting procedure, employing data from other sources, i.e. experiments, model calculations, etc. To evaluate all of the volume fractions encountered in the EBM, we will use values for the elastic moduli for the two-phase blends provided by the 'self-consistent' model, as proposed by Hill[29]. The reason for this procedure is that the EBM can be viewed as a more versatile model as it allows us

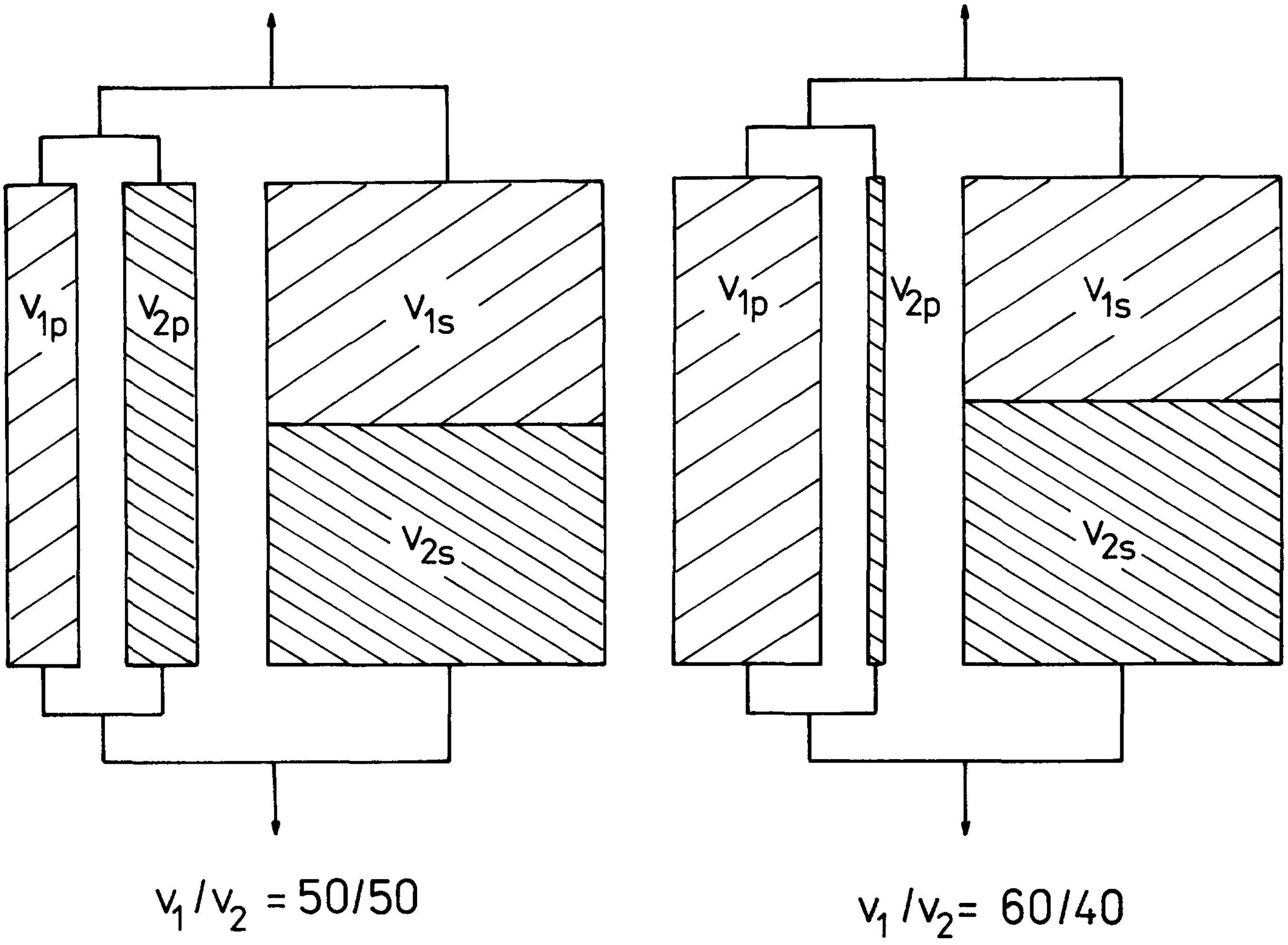

Figure 2 Equivalent box models for binary blends with compositions (by volume) of 50/50 (a) and 60/40 (b)

to distinguish between good and poor adhesion in the course of yielding. Thus, we will tentatively employ the EBM as a 'mixing rule' for the calculations of yield strength because this model expresses, in a similar way to the above cited models[2,7–9,11] for yield strength, the load bearing fraction of the components and their contribution to stress transmission. It is to be noted that, in this way, the validity of the EBM is empirically extended beyond the linearity limit. Although the yield point is situated on the stress–strain curves just beyond the linearity limit, yielding is associated with the initiation of plastic deformation.

As can be seen in *Figure 2*, four volume fractions have to be known for the EBM, of which, however, only two are independent; thus the EBM is a two-parameter model. The fractions of either phase coupled in parallel (subscript p) or in series (subscript s) are given as follows:

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

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

where the phase continuity parameter, C_1 (or C_2), introduced in a previous paper[21], is identified with that fraction of the component 1 or 2 (present in a volume unit of the blend) which can be considered as being coupled in parallel to the acting force. The following retlationships hold for the volume fractions in the EBM:

$$v_p = v_{1p} + v_{2p} \qquad v_s = v_{1s} + v_{2s}$$

$$v_1 = v_{1p} + v_{1s} \qquad v_2 = v_{2p} + v_{2s}$$

$$v_1 + v_2 = v_p + v_s = 1 \tag{2}$$

The modulus of the parallel or series branch is expressed by the following equations:

$$G_p = (G_1 v_{1p} + G_2 v_{2p})/v_p \tag{3a}$$

$$G_s = v_s/[(v_{1s}/G_1) + (v_{2s}/G_2)] \tag{3b}$$

The modulus of the two-component blends is then given as follows:

$$G_b = (G_1 v_{1p} + G_2 v_{2p}) + \{v_s/[(v_{1s}/G_1) + (v_{2s}/G_2)]\}v_s \tag{4}$$

In order to make the calculation of C_1 (or C_2) simpler, we can assume that the shear modulus G_1 of component 1 is much higher than the shear modulus G_2 of component 2 (or vice versa). Obviously, the phase structure and the continuity of the components in a solid blend are independent of the ratio G_1/G_2, which can be controlled (over a certain range) by changes in temperature. The contribution $G_2 v_{2p}$ of that part of component 2 which is coupled in parallel, plus the contribution of the whole series branch (see *Figure 2*), to the modulus of the EBM are then negligible, in comparison to the contribution $G_1 v_{1p}$ of component 1. Therefore, $G_1 v_{1p}$ (or $G_2 v_{2p}$) can be set equal to the calculated modulus G_{1b} (or G_{2b}), which characterizes the stiffness of the component 1 (or 2) contained in a volume unit of the blend:

$$G_{1b}=G_1v_{1p}=G_1v_1C_1 \tag{5a}$$

and in an analogous fashion:

$$G_{2b}=G_2v_{2p}=G_2v_2C_2 \tag{5b}$$

To complete the calculation of the volume fractions, the values of G_{1b} or G_{2b} need to be determined.

Modulus of two-component blends

The modulus G_b of two-component blends can be calculated by means of the equation derived by Hill[29]:

$$v_1K_1/(K_1+4G_b/3)+v_2K_2/(K_2+4G_b/3)$$
$$+5v_1G_2/(G_b-G_2)+5v_2G_1/(G_b-G_1)+2=0 \tag{6}$$

where the indices 1 and 2 stand for the individual components and K is the bulk modulus. It is to be noted that this equation encompasses the entire range of blend compositions, including the phase inversion. Thus equation (6) implicitly provides unique information on the continuity of the phases in binary blends. However, the phase inversion is assumed to occur at around the 50/50 (volume) ratio of the components and cannot be adjusted. In real binary blends, the phase inversion and co-continuity may occur at somewhat different compositions of the components[36,37].

Assuming $G_1 \gg G_2$, the calculated values of G_b can be identified with G_{1b}, which allows us to calculate C_1 by means of equation (5a) and v_{1p} and v_{1s} by means of the relationships given in equation (1a). As the components are interchangeable in the Hill model, the dependences of v_{1p} (or v_{1s}) and v_{2p} (or v_{2s}) on blend composition will be symmetrical. Thus, the calculated dependences given in *Figure 3* are quite versatile for blends undergoing a phase inversion at the 50/50 (volume) ratio. For these model calculations, we used values for G_1 and G_2 of 1 GPa and 2 MPa, respectively; assuming values for the Poisson ratios of $\nu_1=0.3$ and $\nu_2=0.4995$, the bulk modulus $K=(2/3)G(1+\nu)/(1-2\nu)$ assumes values of $K_1=2.17$ and $K_2=2$ GPa, respectively (cf. ref. 38). Although we are concerned with model calculations, we have selected these representable values, which were found or calculated for PC/PMMA blends[10] at 125°C, in order to achieve reasonable interrelationships between the constants of the material.

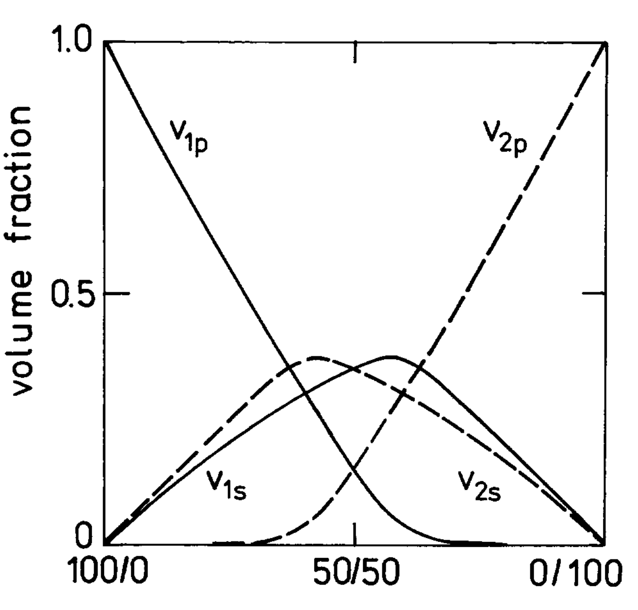

Figure 3 Volume fractions of each phase, coupled in parallel or in series, as functions of composition in two-component blends

Model calculations of yield strength

In general, the models predicting the elastic properties of multicomponent particulate systems assume perfect adhesion between the phases. As elastic properties are measured at very low strains, typically below 1%, so as to remain in the region of a linear stress–strain relationship, low stresses are applied so that almost all systems are apparently characterized by a good interfacial adhesion. However, the situation may change profoundly at higher deformations (3–5%), when a tensile stress inducing yielding and plastic deformation is achieved. Two limiting cases, corresponding to the upper and lower bounds, can be distinguished: (i) the interfacial adhesion is strong enough to transmit an acting stress between the constituents so that the series branch of the EBM also contributes, comparably with the parallel branch, towards the final value of the yield strength; (ii) the interfacial adhesion is so weak that debonding (dewetting) occurs between the fractions of the constituents coupled in series. In this case, the series branch does not contribute to the resulting yield strength, which is therefore determined only by the parallel branch.

With a knowledge of the volume fractions in the EBM, we can employ the model for making tentative calculations of the yield strength, S_{yb}, of two-component blends under the conditions as explained above. As the ratio G_1/G_2, used in the previous calculations, is much higher than the ratios S_{y1}/S_{y2} of the yield strength values encountered in any pair of common thermoplastics, the accuracy of the volume fractions given in *Figure 3* is fully sufficient for these yield strength calculations. Since, by definition, each block (ideally) has the mechanical properties of one of the constituents, the EBM may be attempted for the calculation of blend properties only in such cases in which the properties of the constituents are not affected by mixing, i.e. they are identical with the properties of the parent polymers. If the mixing accounts for a relevant change in the properties of a component (e.g. due to changes in crystallinity), or for a 'synergistic' effect, then the EBM is not applicable. To obtain formulae for the lower and upper bounds of the yield strength for characterizing binary blends of thermoplastics, we can employ equation (4), where the yield strengths of the components, S_{y1} and S_{y2}, can be formally substituted for G_1 and G_2, respectively:

$$S_{yb-}=S_{y1}v_{1p}+S_{y2}v_{2p} \tag{7}$$

$$S_{yb+}=(S_{y1}v_{1p}+S_{y2}v_{2p})+\{v_s/[(v_{1s}/S_{y1})+(v_{2s}/S_{y2})]\}v_s \tag{8a}$$

However, when two components differing in yield strength are coupled in series, it may be expected that the system will yield at strengths of S_{y1} or S_{y2}, whichever is the lower. Thus, if $S_{y1}<S_{y2}$, equation (8a) will assume the following form:

$$S_{yb+}=(S_{y1}v_{1p}+S_{y2}v_{2p})+S_{y1}v_s \tag{8b}$$

which seems to be more realistic.

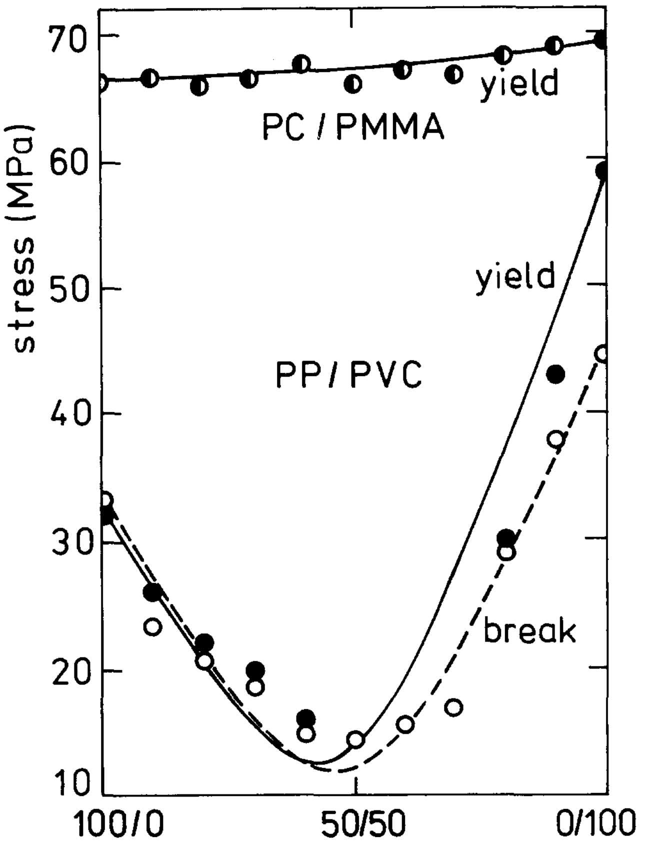

Figure 4 Comparison of experimental and calculated dependences of yield and tensile strength on composition in a two-component blend; upper bound of yield strength of polycarbonate/poly(methyl methacrylate) blends is calculated by means of equations (8a) and (8b), while lower bounds of yield or tensile strength of polypropylene/poly(vinyl chloride) blends are calculated from equation (7). Experimental data points are taken from refs 10, 39 and 40

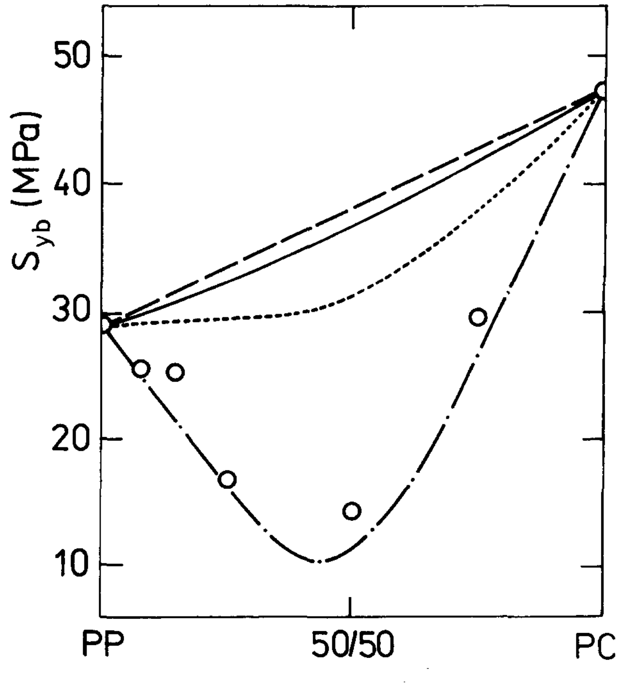

Figure 5 Comparison of calculated upper and lower bounds of yield strength of polypropylene/polycarbonate blends with experimental data[41]: dashed, continuous, dotted, and dash-and-dot lines represent the results obtained by using equations (9), (8a), (8b), and (7), respectively

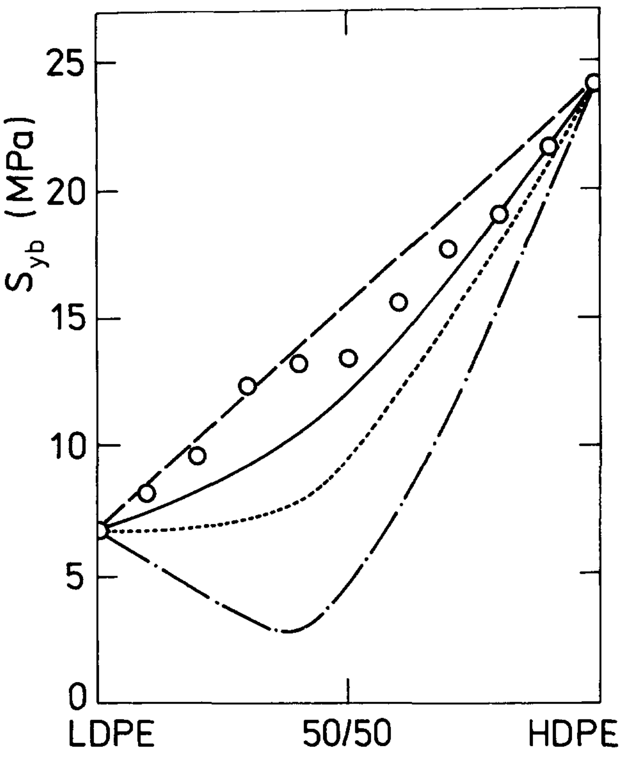

Figure 6 Comparison of calculated upper and lower bounds of yield strength of low-density polyethylene/high-density polyethylene blends with experimental data[40,42]: dashed, continuous, dotted, and dash-and-dot lines represent the results obtained by using equations (9), (8a), (8b), and (7), respectively

RESULTS AND DISCUSSION

The values of the yield strength provided by the EBM are compared with the corresponding experimental data[10,39–42] in *Figures 4–6*. As can be seen in *Figure 4*, the values obtained for the yield strengths of the polycarbonate and poly(methyl methacrylate) components are almost identical, although the mechanisms of yielding are different[13]. When S_{y1} and S_{y2} are similar to each other, the upper bounds, as derived from equations (8a) and (8b), coincide and, moreover, cannot be distinguished from the additivity (linear interpolation) represented by the rule of mixtures:

$$S_{\text{ybrm}} = S_{y1}v_1 + S_{y2}v_2 \qquad (9)$$

Therefore, we can explain the simplified empirical assumption made earlier[10,11,18,32] that equation (9) is valid for the yield strength of binary blends in the case of good interfacial adhesion. (In principle, the rule of mixtures holds for the structures visualized in *Figure 1* and cannot be valid for isotropic heterogeneous materials.) With an increasing difference between S_{y1} and S_{y2}, resulting differences between equations (9), (8a), and (8b) are bound to arise (see *Figures 5* and *6*). Nevertheless, the upper bound fits the experimental data for PC/PMMA blends very well (see *Figure 4*), which clearly proves that a strong interfacial adhesion has been achieved, as a result of partial miscibility of the constituents[10].

The lower bound is expected to be adequate for systems with very poor interfacial adhesion. *Figures 4* and *5* show that the yield strength in poly(vinyl chloride) (PVC)/polypropylene and polycarbonate/polypropylene blends follows quite well the calculated dependence. It is evident that the incompatibility (total immiscibility) between the pairs of dissimilar polymers actually prevents any interfacial adhesion. It is interesting to note that the predicted lower bound also reasonably fits the experimental data obtained for the tensile strength of PP/PVC blends. Thus, it seems that the EBM might also be applicable for the tensile strength of binary blends with poor interfacial adhesion. Similarly enough, equations based on the reduction of load-bearing cross-sections have also been tentatively used for the evaluation of tensile strength[2,39,40].

Figure 6 summarizes data[40,42] on the yield strength of the blends consisting of low-density and high-density polyethylenes. As can be seen, the experimental data points are situated between the curves corresponding to equations (8a) and (9), which undoubtedly gives evidence of strong interfacial adhesion. However, relatively high values of the yield strength, clearly exceeding those obtainable from equation (8b), require analysis of possible factors which can affect the blend structure and cause the observed values to lie between the bounds (thus apparently indicating 'intermediate' interfacial adhesion) or above the upper bound.

Obviously, all factors controlling the continuity of the individual phases are of primary importance. It is well known[36,37,43] that the component with the lower (relative) viscosity in the melt shows a higher tendency to form the continuous or co-continuous phase than the component with the higher viscosity. If the component with the higher yield strength has a higher phase continuity than that calculated from the Hill model, then the experimental data will be situated above the calculated dependence, which holds for blends with both good or poor adhesion. Nevertheless, the yield strengths of binary blends which exceed the values rendered by the rule of mixtures[11] can hardly be understood without assuming some structural changes in the components as a result of blending. It should also be noted that the predicted minimum value of $S_{\mathrm{yb}-}$ is shifted on the composition scale towards the component with a lower S_y, even though the phase inversion point corresponds to a 50/50 (volume) ratio.

Another interfering factor is the phase structure orientation during preparation of a test specimen, e.g. by injection moulding, which may cause the samples to no longer remain isotropic. A higher continuity of both phases in the direction of orientation will necessarily result in an increase in the yield strength in that direction, irrespective of the interfacial adhesion. Furthermore, as the stress necessary for the dewetting of the spherical particles embedded in a matrix is inversely proportional to the square root of the particle radius[44], a particular situation may occur in which the stress acting during the yielding process will cause dewetting of a certain fraction of the larger particles, while the smaller particles will remain bound to the matrix. In an analogous way, an increase in diameter of the particles due to the increase in volume fraction of the dispersed phase[39,40,45–47] in the blend melt, or phase structure coarsening (the detrimental effect of which on mechanical properties has been previously pointed out[24,48]), etc., might lead to an increasing volume fraction of debonded particles in the system and thus cause a shift of the yield strength towards the lower bound. Another reason for a drop in yield strength may be cavitation of one of the phases. However, the detection and quantitative evaluation of these effects, as outlined above, which could be misinterpreted as changes in the interfacial adhesion, would be rather complex.

CONCLUSIONS

Information on the continuity of phases in two-component systems, which was obtained from the Hill model for elastic behaviour, was utilized in an equivalent box model in order to calculate the yield strength of the systems under the assumption of either (i) 'perfect' or (ii) 'zero' interfacial adhesion. These limits are tentatively identified with the upper and lower bounds of the yield strength of binary blends of thermoplastics. The degree of continuity of each constituent decreases with its decreasing fraction in the blends; the sum of these quantities passes through a minimum at the 50/50 (volume) ratio composition, which is implicitly assumed in the Hill model as the phase inversion point. If the adhesion between the constituents is strong enough to transmit the yield stress, the resulting upper bound may be rather close to (but always lower than) the dependence calculated from the 'rule of mixtures (additivity). In the case of poor adhesion, the fractions of both constituents formally coupled in series are ineffective in stress transmission as a result of interfacial debonding. Therefore, the lower bound shows a minimum close to the 50/50 composition. However, if the values of the yield strengths of the components are rather different, then the minimum is shifted on the composition scale towards the component with the lower strength. The predicted dependences are in good accord with the experimental data obtained for binary blends characterized by either strong or poor adhesion.

When analysing the experimental data on the yield strengths with the aid of the EBM, possible interfering factors should be taken into account which may cause an increase in the measured values; namely (i) a higher phase continuity (than that calculated from the Hill model) of the component with the higher yield strength, due to its lower relative viscosity in the melt, or (ii) orientation of the phase structure in the processing stage, e.g. injection moulding. Moreover, as the stress necessary for dewetting of the spherical particles is inversely proportional to the square root of the particle radius[42], the measured yield strength may be affected by the particle size and its distribution, the increase in particle size with the increasing volume fraction of the dispersed component, coarsening of the phase structure due to annealing or reprocessing, etc. Obviously, the resulting shifts in the yield strength might be erroneously attributed to changes in the 'mean' adhesion.

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