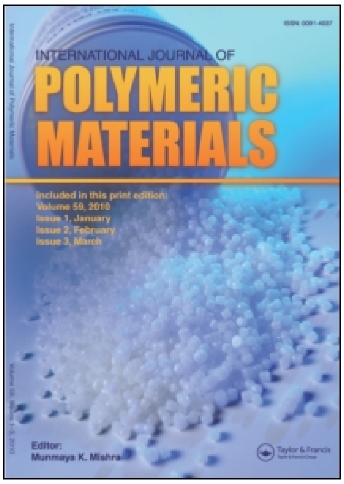

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Generalized Characteristics of Polymer Blend Melt Viscosity

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Generalized Characteristics of Polymer Blend Melt Viscosity

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A very simple reduction procedure is suggested for the blend viscosities of different polymer pairs. This procedure is based on the comparison of the blend viscosity, normalized either to the matrix or to the disperse phase viscosity, with the viscosities ratio of the initial polymers (η_m/η_d). We have obtained, for 13 different pairs containing 30% of the second component, the universal linear dependencies, mutual analysis of which allows connection of their special points with the stream morphology. The fibrillous morphology takes place in the range of $\eta_m/\eta_d = 0, 1$–5. Simultaneously, the thin skin consisting of the disperse phase polymers is formed. These results confirm the predominant role of the viscosities ratio in fibrillar composite material formation in comparison with the interphase tension phenomena.

KEY WORDS Polymer blends, melt viscosity.

RESULTS AND DISCUSSION

In recent years, a great deal of attention has been given to the problem of investigating and practically employing immiscible polymer blends. This activity has increased substantially with the use of liquid crystalline (LC) thermoplastic materials such as the disperse phase, which considerably decreases the blend viscosity and simultaneously improves the mechanical characteristics of final articles.[1–4]

These effects are especially distinctive in the extrusion scheme, inherent in which is the convergent flow accompanying the extension strain, leading to the elongation of the disperse phase drops and the formation of continuous fibers.[5] Besides, this scheme allows formation of extrudate envelopes from the disperse phase polymer, thus providing additional effects for decreasing the viscosity and modifying the properties of the solidified extrudates.[6] The process of extrusion of immiscible polymer blend melts is regarded as a complex process in which we can observe simultaneously: the phase and relaxation transitions, the heat and mass transfers, and the superficial and rheological phenomena.[7] In this connection, it is extremely difficult to choose the rheological criteria of the specific fiber formation which would describe the viscous properties of polymer blends.

There are a number of works[3,8–10] which indicate that at similar viscosity values of the initial components, the fiber formation process is most intensive, but the reasons for this behavior are not yet clear. In particular, the conditions for transmission of the stresses from the matrix to the disperse phase are not discussed.

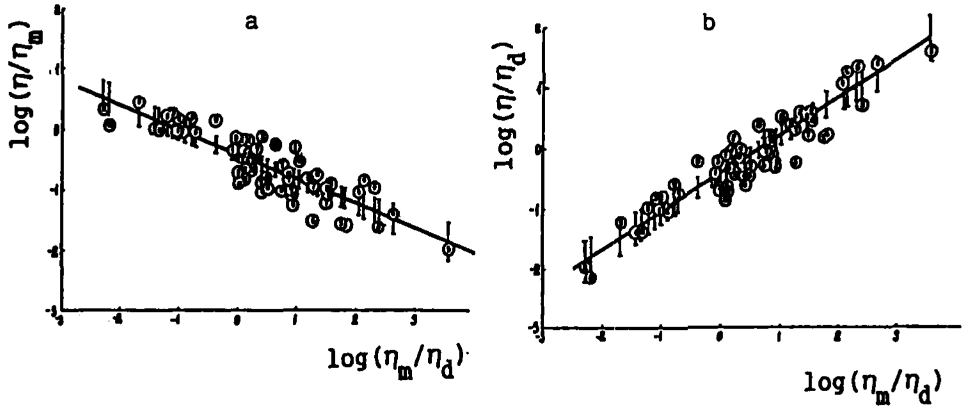

FIGURE 1 The dependence of the polymer blend viscosity, normalized to the matrix viscosity (a) or to the disperse phase viscosity (b) on the viscosities ratio of the initial polymers. The figures inside the experimental points indicate the blend numer given in the table.

Furthermore, in these works the conclusions are made with respect to specific polymer pairs only and refer mainly to qualitative behavior.

Taking into consideration the generality of rheological behavior and structure of different immiscible polymer blends, we formulated the problem of finding the means for the invariant presentation of the rheological data and for establishing their relation to the stream structure.

Analyzing the original experimental results and the literature data devoted to viscosity vs. composition dependences, we tried to present this information in the form of unified curves. It appears that using logarithmic scales, the dependences of the blends viscosity at definite contents of disperse phase, normalized to the matrix viscosity (η/η_m) or to the disperse phase viscosity (η/η_d), on the ratio of the initial components viscosities (η_m/η_d), are linear and universal for different polymer pairs. This fact indicates that the reduction parameters are related by the power function. For blends containing 30% of the second component with increasing ratio η_m/η_d, the blend viscosity normalized to the matrix viscosity linearly decreases (Figure 1(a)), while the blend viscosity normalized to the disperse phase viscosity increases (Figure 1(b)).

These dependences are described by the empirical Equations (1) and (2):

$$\log(\eta/\eta_d) = -0.41 + 0.63\log(\eta/\eta_m) \tag{1}$$

$$\log(\eta/\eta_d) = -0.39 - 0.40\log(\eta_m/\eta_d) \tag{2}$$

The correlation coefficient in the former case equals 0.85, while in the latter case it equals 0.9.

Equations (1) and (2) are not independent functions. The connection between them can be revealed by the next simple reasoning:

$$\log(\eta/\eta_d) = a + b\log(\eta_m/\eta_d)$$

$$\log(\eta/\eta_d) + \log(\eta_d/\eta_m) = a + b\log(\eta_m/\eta_d) + \log(\eta_d/\eta_m)$$

$$\log(\eta/\eta_m) = a + (1 - b)\log(\eta_m/\eta_d)$$

For both cases $a = -0.4$, $b = 0.6$ and $(1 - b) = 0.4$, i.e. coincidence between experimental and calculative data is practically total.

The above-indicated dependencies are plotted for 13 different polymer pairs (see Table I). Both the ordinary thermoplastic polymers[8,9,11] and the liquid crystalline-[2,3,13-16] and mesophase polymers[6] are used as the disperse phase. The change in the ratio η_m/η_d in the blends under consideration was attained either by changing the molecular weights or by varying the viscosities of the initial polymers by manipulating the shear rate and temperature.

Of special interest is the consideration of the obtained dependences upon their superposition (Figure 2).

TABLE I

The analyzed polymer blends

NN	Pair of Polymers		Literature
	Matrix	Disperse phase	
	I. Blends of commercial thermoplastics.		
1.	Copoly(ethylvinylacetate)	Polyoxymethylene	[8]
2.	Polystyrene	Polyethylene (PE)	[9]
3.	Polypropylene	PE	[11]
	II.Blends with mesophase components		
4.	Poly-4-methylpentene-1	Poly(bis-trifluoroethoxyphosphazene(PPh)	[×]
5.	PE	PPh	[6]
	III. Blends with LC-polyesters		
6.	Polysulfone(PSF)	Ultrax KR-4002	[12]
7.	PSF	Copoly(ethyleneterephthalate-p-oxybenzoate) (30:70) (PES)	[×]
8.	Polycarbonate	PES	[3]
9.	Polyvinylchloride	PES	[13]
10.	Nylon 6	Vectra A-900	[2]
11	PE	Copoly-(hexa-decamethylenefumaroyl-bis-4-oxybenzoate)	[×]
12.	Poly(butyleneterephthalate)	Vectra B-950	[14]
13.	Poly(etherimide) "Ultem"	Copolyester (Bayer Co)	[15]

× Unpublished data

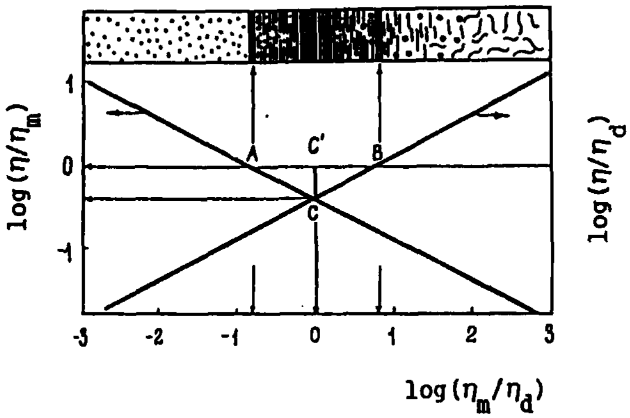

FIGURE 2 The schematic dependences of the normalized blend viscosities and stream morphologies on the initial components viscosities ratio (see explanation in text).

The analysis of the triangle with apices *A*, *B*, and *C* has shown that points *A* and *B* are formed by the intersection of the above mentioned dependencies with the "isoviscous line" where $\eta = \eta_m = \eta_d$, while point *C* corresponds to the equality condition $\eta_m = \eta_d$. In this case, the viscosity-composition dependence passes through a minimum, i.e. the blend viscosity is less than the viscosities of the two components. So, we can expect the minima on concentration dependences of blend viscosity at any content of disperse phase, if the ratio of initial viscosities will be convenient.

Worthy of attention is the fact that the specific points separate different morphological patterns of the flow schematically illustrated in Figure 2. The natural pictures of the morphology are given in the cited literature. Let us analyze the stream morphology with a rise in η_m/η_d for the dependence $\lg(\eta/\eta_m) = f[\lg(\eta_m/\eta_d)]$. At very low η_m/η_d ratios, the disperse phase particles appeared to be stiff to such an extent that the low-viscous matrix could deform them in the flow. Such a system behaves as being filled in relation to the matrix, and its viscosity is always higher than η_m, and is located between η_m and η_d.

At point *A*, $\eta = \eta_m$. In other words for condition $\eta_m/\eta_d \approx 0.1$ the matrix is responsible for the flow. During the transition from *A* to *B*, the drop strain and the formation of continuous threads created from them and penetrating the matrix proved to be essential. Point *C* corresponds to the most perfected fibrillous structure. Such a self-reinforced system possesses the best mechanical properties. A further increase in η_m/η_d leads to decreasing η, resulting not only from the preservation of the liquid threads of the disperse phase, but also from the formation of an envelope consisting of a low-viscous component at the periphery of the flow. The formation of such an envelope is completed at $\eta_m/\eta_d \approx 0.1$–5. This corresponds to the condition $\eta = \eta_d$ (the formation of the envelope proceeds especially effectively for liquid crystalline disperse phases). At $\eta_m/\eta_d > 5$, the fibers transform into layers and the blend viscosity continues to decrease due to the macrostructural plastification of the system.

The universality of the suggested approach shows the decisive influence of the rheological effects on the flow morphology in comparison, for instance, with the surface effects. One should expect this, since the tangential and normal stresses at the interphase boundary inducing the deformation of the disperse phase particles depend largely on the viscosity ratio of the components. The surface phenomena and in a more general form, the interaction of the blend components, play an important role in the vicinity of the hypothetical triangle *ABC*, where most of the data scatter is observed.

On the basis of the above, we may conclude that the suggested generalization has a predictive virtue. Depending on the formulated task (structural, rheological, mechanical) and knowing the viscous properties of the initial components, it is possible to estimate beforehand the blend viscosity of the given composition by using the above-mentioned empirical expressions, and in this way choose the appropriate processing regime.

The suggested generalization refers to 30% content of the disperse phase, but it can also be realized at lesser concentrations, only in this case, point *C* is shifted upward along the line $\lg(\eta_m/\eta_d) = 0$, while the slopes of lines "*a*" and "*b*" increase.

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