

# Structure, Mechanical, and Rheological Properties of Polyethylene–Poly(oxymethylene) Blends

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## INTRODUCTION

Obtaining polymeric mixtures and alloys is an important means of modifying physical properties of polymeric compositions. Addition of small amounts of one polymer to the other may result in the improvement of structural homogeneity of the polymer and an increase in its strength.<sup>1-4</sup> Variation of strength for small additives was explained by the fact that the additives act as a damper in redistribution of the internal stresses or fill some defects in microstructure of the bulk polymer. However, a detailed study of the changes in properties of the polymers due to small additions of other polymers has not been reported so far, and the mechanism of their effect remains unclear.

The purpose of the present work is to establish general regularities in the change of properties of a polymer when others of various compositions are added to it.

## EXPERIMENTAL

High-density polyethylene (PE) and poly(oxymethylene) (POM) were chosen as subjects for our investigation. Polymeric mixtures were prepared by extrusion of the powder mixture at 180°C. For rheological study of these mixtures we took cylindrical samples. Concentration of one polymer in the other was 0.5% to 40.0% by weight both in the case when POM was added to PE and vice versa.

Dynamic mechanical properties were determined using the frequency relaxometer.<sup>5</sup> Temperature and frequency dependences of the real part of the complex shear modulus  $E'$ , the tangent of the angle of mechanical losses  $\tan \delta$  in the temperature range of 20°–140°C, and deformation frequencies from 0.004 to 0.4 Hz were studied. Frequency dependences had been processed using the method of Williams-Landell-Ferry<sup>6</sup> which enabled us to obtain generalized viscoelastic functions and determine activation energies of the deformation process. Viscosity of the melts was determined in a nitrogen atmosphere with the capillary viscosimeter MV-2 at 140°–200°C at constant shear stress  $\tau = 3.6$  dynes/cm<sup>2</sup>.

Contact angles were determined for the mixtures, and surface tension of the mixtures was calculated according to the Elton eq. (7). A flat plate holder camera and the URS-55 diffractometer were used for x-ray studies. During the experiment we used emission of the copper anode with nickel filter in the primary beam. Calorimetry data and radiothermoluminescence curves were obtained by the techniques described in references 4 and 13, respectively.

## RESULTS AND DISCUSSION

Concentration dependencies of the real part of the complex shear modulus for various samples are shown in Figure 1. It is obvious that, for small concentrations of each of the components,  $\log G'$  varies considerably with concentration. However, when the temperature rises, the effect decreases and, at 100°C, in the case when POM is added to PE, it disappears completely. It is quite clear from the temperature dependences that  $\log G'$  for pure components and mixtures (Fig. 2), comprising additives in amounts corresponding to the extreme values. The curves for the mixtures are much higher than for the pure components, and the intensity of their variation increases with temperature. However, POM additive

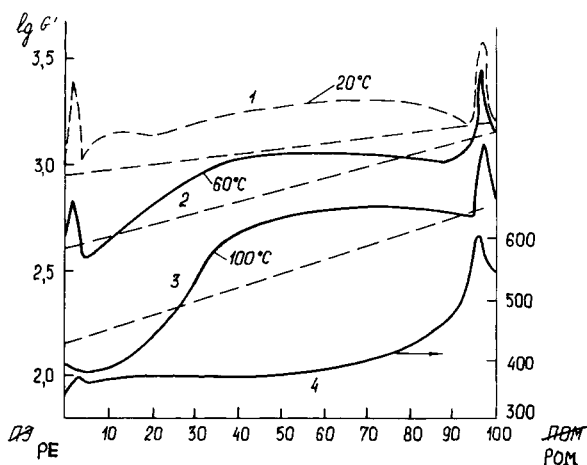


Fig. 1. Concentration dependence of the real part of complex shear modulus  $G'$  for PE-POM mixture at various temperatures: (1) 20°; (2) 60°; (3) 100°; (4) tensile strength at 20°; dotted lines are additive values.

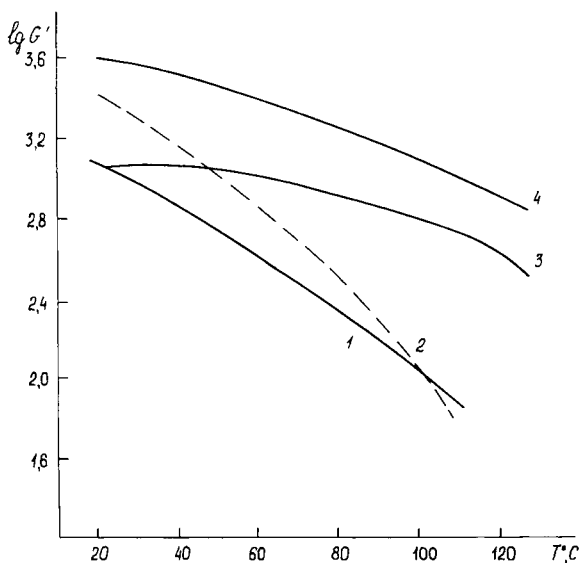


Fig. 2. Temperature dependence of real part of complex shear modulus: (1) pure PE; (2) PE + 1% POM; (3) POM; (4) POM + 3% PE.

does not influence the properties of the mixture very much, which may presumably be related to the changes in the PE structure at temperatures in the proximity of its melting point.

On the basis of the results obtained using the WLF method, the generalized viscoelastic functions were calculated (Fig. 3). The relation  $\log G' = f(\log \omega a_T)$  is a straight line, typical of crystalline polymers,<sup>6</sup> and shows that the degree of crystallinity within the temperature range studied is constant. At the same time, difference in dependences  $\log G'$  for the initial components and their mixtures with transition into the low-frequency range grows, being indicative of the essential change of the quasi-static characteristics of the composition.

On the basis of the generalized viscoelastic functions, the temperature dependence of the shift factor  $a_T$  was calculated. The mixture containing 3% POM has the highest activation energy (Fig. 4). It is in good agreement with the results of the strength test of the same samples (Fig. 1, curve 4).

The most important results of the rheological studies are shown in Figure 5 as  $\log \eta$  versus the reciprocal temperature. When the POM content in the mixture is as high as 20%, at 170°C a sharp change in the viscosity temperature coefficient occurs and the slope of the curve increases with POM concentration. Change in the character of the viscosity-temperature dependence at 170° may be related to the melting of POM, as this temperature nearly equals the POM melting temperature. Thus, when POM content is 1%–20% in the temperature range of 140°–170°, dispersion of the POM solid particles in the high molecular weight (PE) is observed. Of particular interest is the variation of the slope of the curve  $\log \eta = f(1/T)$ , i.e., the dependence of the activation energy on the POM content (Fig. 6).

Figure 7 shows dependence of the viscosity on the POM weight concentration at various temperatures. Calculation of the experimental data in terms of the Mooney eq. (10),

$$\eta_{\text{rel}} = \exp [2.5\varphi/(1 - K\varphi)]$$

has shown that for the system studied  $\eta_{\text{rel}}$  increases more intensively than expected. Addition of small amounts of POM to PE melt causes a decrease in

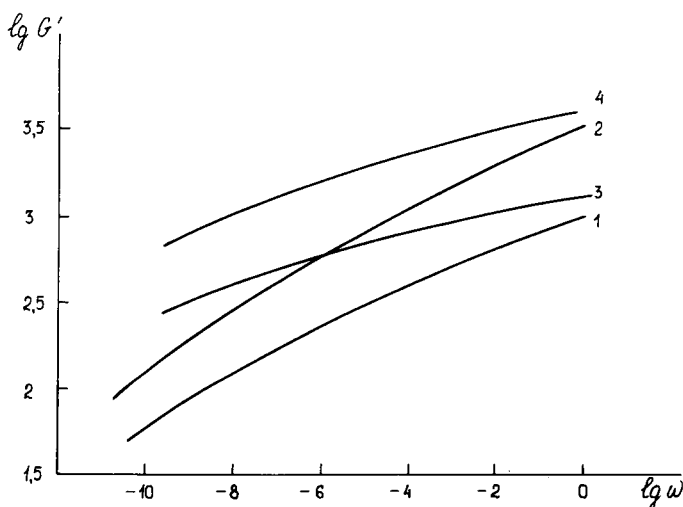


Fig. 3. Generalized dependence of  $G'$  (for designations see Fig. 2).

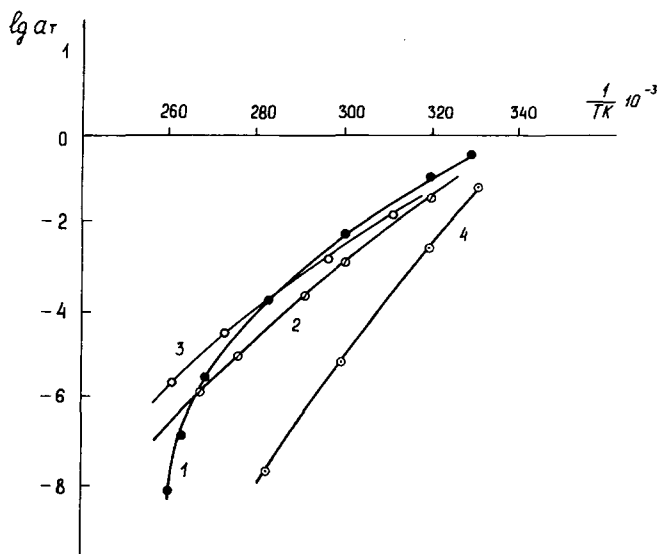


Fig. 4. Dependence of reduction factor  $\log a_T$  on  $1/T$  (for designations see Fig. 2).

viscosity of the melt. Only at concentrations higher than 5% does viscosity of the melt increase regularly (Fig. 7). Temperature rise does not decrease viscosity. Transition of POM from the solid to the liquid state at temperatures higher than the melting point does not affect its ability to decrease the viscosity of the mixture. Figure 8 represents dependence of the viscosity of the PE and POM melts on the composition over the concentration range studied at  $170^\circ$ – $200^\circ$ . When small amounts of PE were added to POM, the viscosity of the system did not drop sharply. Viscous flow activation energy (Fig. 9) changes with composition in a complicated way.

We also evaluated density and heat of melting (using a calorimeter) in relation to composition (Fig. 10). Density and heat of melting dependences on mixture composition are also nonmonotonic.<sup>4</sup> Within the range of small concentrations of POM additives, the density of the mixture is much greater than the additive value for the mixture. PE heat of melting is also greater than that of the initial PE. With further increase in POM content, the density of the mixture becomes lower, that follows from additivity rule.

Let us now consider some results of the x-ray study. Both components in the mixture are capable of crystallization. X-Ray wide-angle studies show that diffraction patterns correspond to superposition from components, i.e., the angle position of the specific reflections remains unchanged at various ratios. This indicates the absence of cocrystallization. POM crystallizes in a hexagonal and PE in an orthorhombic lattice.<sup>10</sup> To discover the interaction between the two components, we have chosen a method based on the transfer of the system into the oriented state.<sup>11</sup> Orientation was carried out by drawing the mixture at a rate of 0.6 mm/min at  $100^\circ$  to ultimate elongation. Figure 11 represents the most typical x-ray patterns of the mixtures studied within the entire concentration range. Figures 11(a) and 11(g) demonstrate that pure PE and POM turn to the highly oriented state after elongation. This can be seen from the equatorial character of reflections 110 and 200 PE and 100 POM and from splitting the 105 reflection POM into four arcs. When comparatively small amounts of POM—up

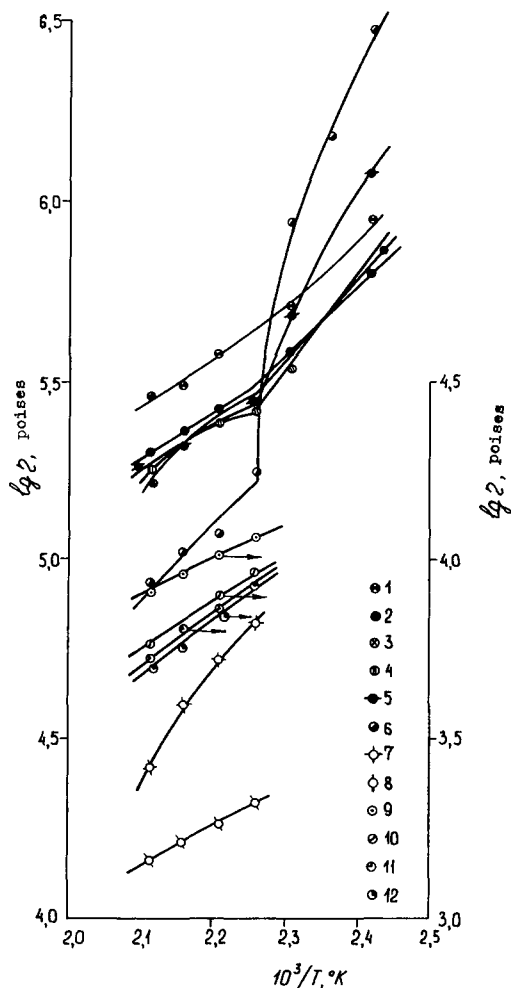


Fig. 5. Temperature dependence of viscosity of PE, POM, and mixtures at constant shear stress. Content of POM (% by weight): (1) 0; (2) 1; (3) 2; (4) 5; (5) 10; (6) 20; (7) 40; (8) 60; (9) 80; (10) 90; (11) 95; (12) 98, 99, 100.

to 20%—were added to PE, elongation resulted in formation of a highly oriented PE texture. Simultaneously, POM inclusions at these concentrations retained their isotropic nature [Fig. 11(b)]. With a POM concentration range of 40%–80%, corresponding x-ray patterns [Figs. 11(c) and 11(e)] showed orientation effects for both components. If POM content is higher than 60%, its texture becomes more appreciable, and if the content is lower, the texture is less appreciable. For PE, the contrary is the case. If POM content is more than 90% [Fig. 11(f)], x-ray patterns exhibit highly oriented texture. Yet PE inclusions in this case remain isotropic. Thus, x-ray data show that texture effects, which manifest themselves as a result of ultimate elongation of the mixture, are dependent on the ratio of the components.

There is a relationship between the deformability degree of each component and the adhesion between components. In the case of more or less intensive interactions in the mixtures, one may expect that orientation of inclusion during elongation will occur. But this does not take place up to 20% POM. Apparently,

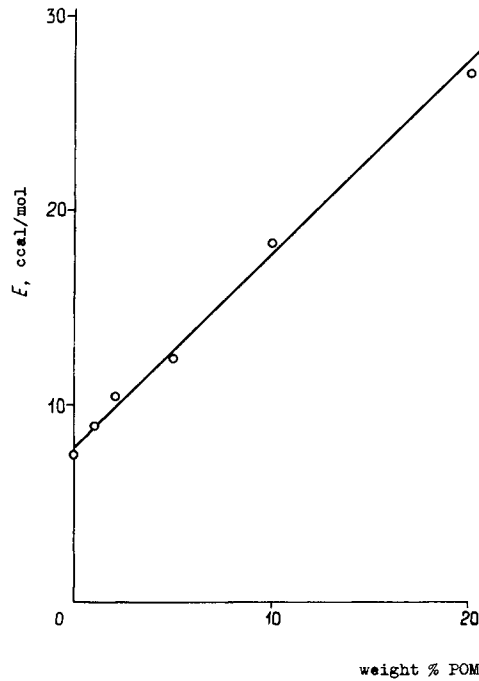


Fig. 6. Concentration dependence of activation energy for POM particle dispersion in PE melt.

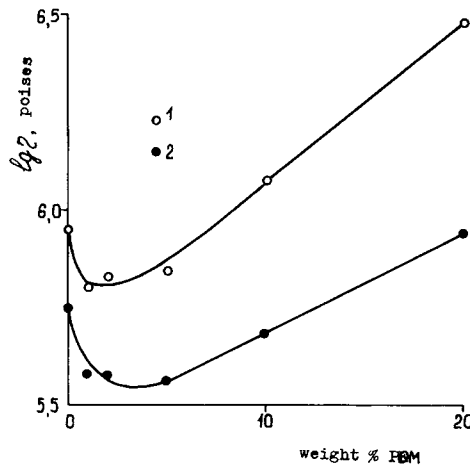


Fig. 7. Dependence of viscosity of PE melt on POM particle content at 140° (1) and 160° (2).

adhesion between the components is poor due to their thermodynamic incompatibility. Absence of PE orientation during elongation of the samples containing more than 90% POM proves that adhesion interaction between the components of the mixture is weak. Essential orientation effects within the concentration range of 40%–80% for both components proves to be inconsistent with the idea of dispersion of one component in the other. Most probably in this case there exist two continuous phases, formed by the two components. These polymers are thermodynamically incompatible and, within concentration ranges

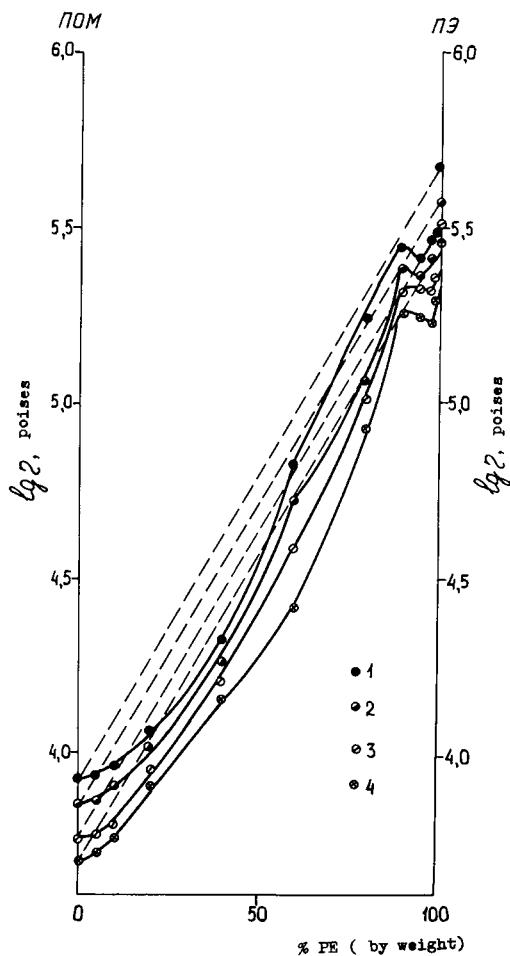


Fig. 8. Dependence of melt viscosity of PE-POM mixtures on composition at 170° (1), 180° (2), 190° (3), and 200° (4).

of up to 3% POM in PE and up to 2% PE in POM, they can be distinguished by electron microscopy<sup>4</sup> as a dispersion phase and dispersion medium.

Thus, the thesis of a two-phase heterogeneous structure is a starting point in the discussion. Considering the influence of the additive with a high melting temperature (POM) on mechanical properties of the mixture, the effects observed might be explained on the basis of possible influence of the additives, curing, or filling defects of the initial polymer structure. As a result, the system becomes structurally more homogeneous, regardless of coexistence of two phases, and its mechanical properties improve.<sup>4</sup> On the other hand, one can expect that the additives will also influence structure formation<sup>12</sup> that follows from an increase in density and the melting heat of PE.

The process, however, may proceed without change in crystallinity degree, since x-ray studies<sup>4</sup> have shown that crystallinity within the entire range of compositions is close to its additive values. Influence of the components on each other manifests itself appreciably within the range of small additives.<sup>13</sup>

Radiothermoluminescence (RTL) maxima at -49° for PE and at -72° for POM were chosen as typical luminiscence maxima. Positions of the maxima

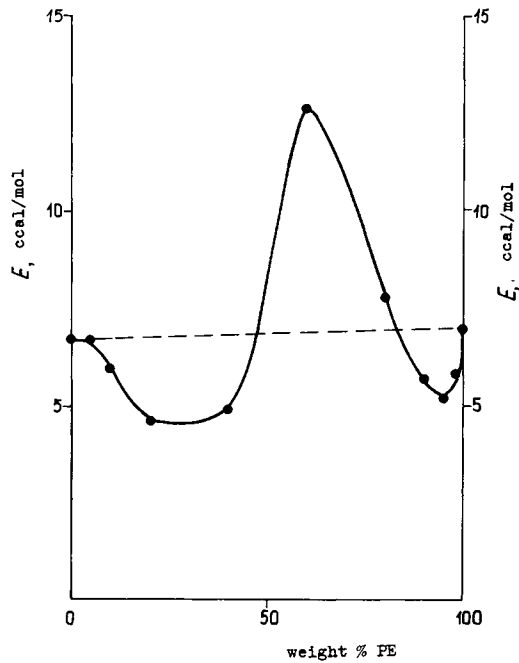


Fig. 9. Dependence of activation energy on composition for melt of POM-PE mixtures.

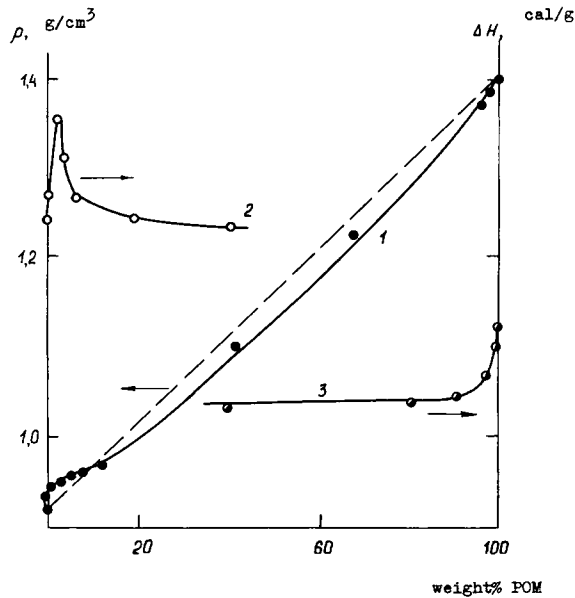


Fig. 10. Dependence of density (1) and heat of melting for PE (2) and POM (3) as a function of composition.

are determined by mobility in the amorphous phase and coincide with the values of the glass transition points.<sup>13</sup> When POM additives are small (less than 2%) the maximum corresponding to the PE glass transition temperature is shifted to lower temperatures (down to  $-55^\circ$ ). Starting with 5% POM additive, the position of the maximum remains constant up to 40% [Figs. 12(a) and 12(b)]. A



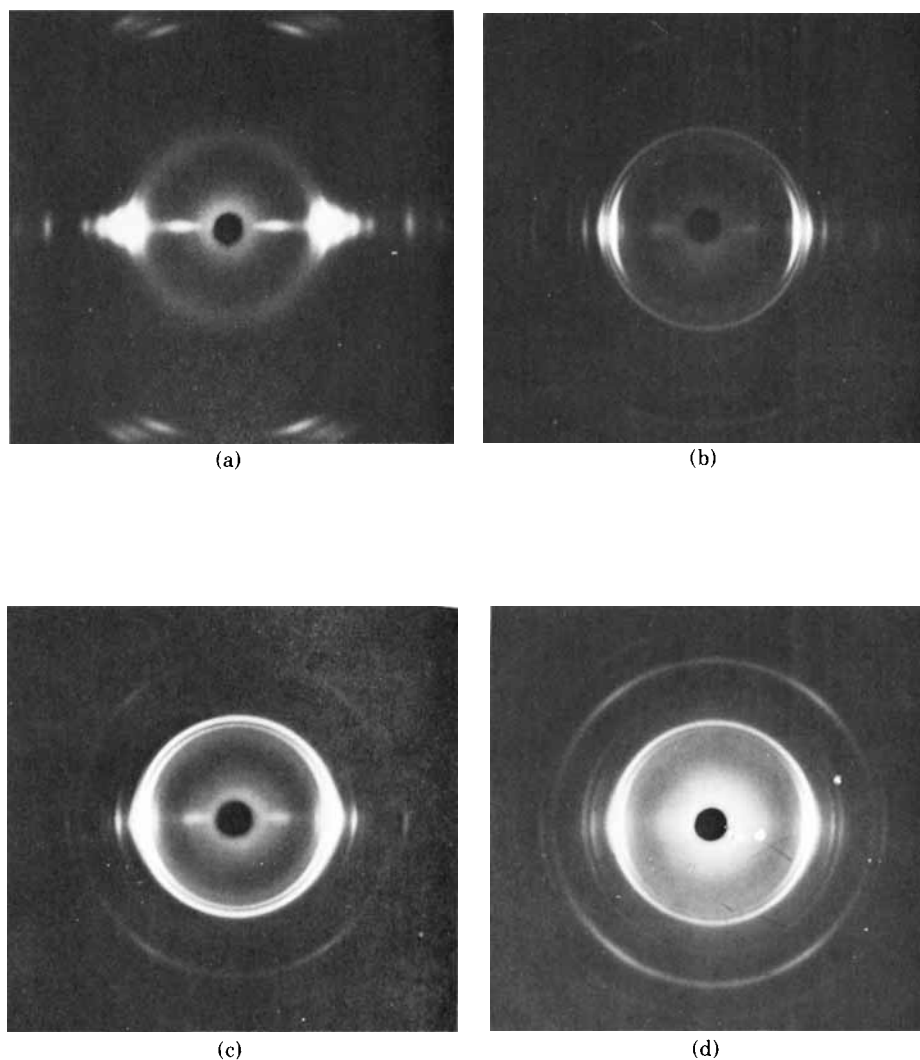
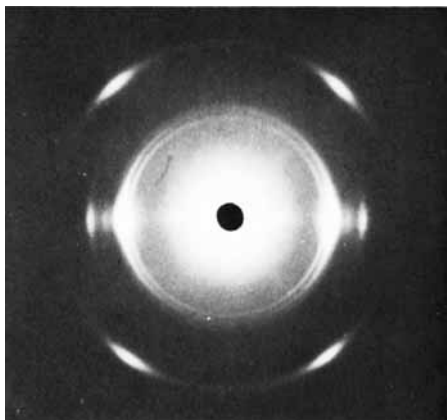


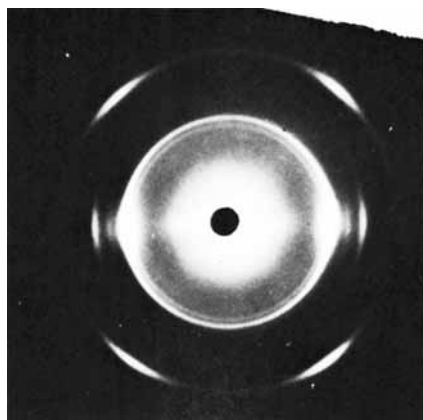
Fig. 11. X-Ray diffraction pattern for POM-PE blends.

10% addition of POM causes appearance of another maximum, which is clearly seen on the RTL curve of the mixture with 40% POM; it is located at  $-67^\circ$ . In the case of small additions of PE to POM (1%), maximum luminescence is shifted to  $-67^\circ$  [Fig. 12(b)]. With further increase in PE content, the maximum remains at the same place, and, starting with 5% PE additive, a new maximum appears in the proximity of the shifted PE maximum with small POM additives. Variations of RTL maximum temperature corresponding to glass transition points of PE and POM depending on concentration of the components in the mixture are seen in Figure 12(c).

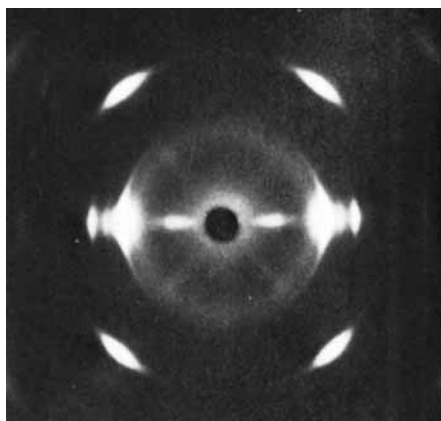
The data obtained demonstrate changes in the amorphous regions of PE and POM when mixed. In the case of small additions of POM to PE (less than 2%), and of PE to POM (less than 1%), there may occur local compatibility of the components and, as a result, one shifted luminescence peak is observed for PE and POM. When PE content in POM and POM content in PE increase further,



(e)



(f)



(g)

Fig. 11 (Continued from previous page.)

the amorphous phase will consist of two regions, and two shifted glass transition temperatures exist. There is a possibility of formation of a transition layer between the two components in the absence of thermodynamic compatibility between them.<sup>14,15</sup> The structure of such transition layers has been discussed in detail before.<sup>16</sup> Existence of the transition layer, found by independent techniques and foremost by electron microscopy, allowed us to explain some anomalies in the range of small additives. Mechanical characteristics are connected with interpenetration of the additive component into the surface defects of the bulk component. The two-phase transition layer formed in this way controls rather strong contact interaction between the two components and makes the system more homogeneous.<sup>14</sup> Structural changes cause partial inhibition of the structure formation in the contact zone, influenced by the surface of the other component<sup>16</sup>; this may affect destruction of the structure and lead to the formation of a less ordered layer, having decreased internal stresses.

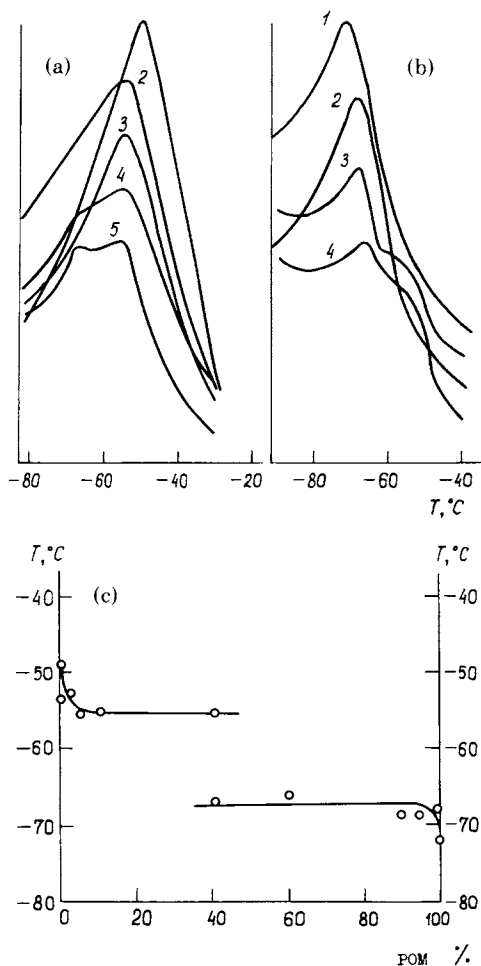


Fig. 12. RTL curves for PE-POM mixtures at different amounts of (a) POM in PE: (1) 0%; (2) 0.5%; (3) 1.0%; (4) 10.0%; (5) 40.0%. (b) PE in POM: (1) 0%; (2) 1%; (3) 5%; (4) 40%. (c) Glass transition temperatures determined from RTL curves as a function of composition.

Existence of the transition layer also explains anomalies in the rheological behavior of the system. In the temperature region up to  $170^\circ$ , i.e., before melting of the particles in the disperse phase (POM), a definite dependence of the activation energy of the viscous flow on the content of the dispersed phase is observed. When the transition layer is absent, the mechanism of the melt flow would not have been dependent on the presence of the dispersed particles in the system.<sup>17</sup> Existence of such dependence indicates that within a certain temperature range a transition layer is adsorptionally connected with the surface of the particles in the dispersion media and is formed by the two components. The transition layer is compatible with the particle of the dispersed phase, as has been established from the rheological properties of filled polymers.<sup>18</sup> The layer increases local viscosity and effective content of the particles in the border regions; this is the reason for activation energy dependence on the content of the dispersed phase, formed by the second component.<sup>19</sup>

Absence of compatibility between the components causes anomalies in their

rheological behavior at temperatures higher than the POM melting point. Since the system is incompatible, an excess free volume develops in it,<sup>14</sup> facilitating the process of viscous flow. Appearance of the excess free volume in the case of small additions may result in a decrease in the viscosity of the system due to microseparation.<sup>20</sup> If the additive is large enough, viscosity of the melt will increase due to a hydrodynamic effect caused by particles of the second component (independent of their phase state). The role of the effects changes nonmonotonously with composition, causing anomalies in the rheological behavior of the mixture. Activation energy varies (Fig. 9) nonmonotonously and coincides with activation energy values calculated according to the additive rule up to a PE content in the mixture of 5%. Increase in the incompatibility of the mixture results in a decrease in activation energy in comparison with the activation energy of each of the components when PE concentration equals 10%–45% or 85%–100%. The sharp increase in activation energy is somewhat unexpected if compared with the additive value when PE content equals 50%–85%; this is probably connected with the change in the mechanism of viscous flow in this composition range.

In fact, x-ray data show that irrespective of the lack of strong adsorption (which cannot take place because the surface tension values for both components are very close) in the POM composition range of 40%–80%, noticeable orientation effects can be detected for both components. In this composition range the system is not a traditional colloid, i.e., consisting of the dispersion medium and the dispersed phase. It is obvious that microheterogeneity in the structure of the mixture is determined by the existing two interpenetrating media, causing the change in the flow mechanism in this composition range.

The defect in the density of the mixtures (Fig. 10) when  $\text{POM} > 10\%$  can be related to the surplus free volume, located at the interphase border because of incompatibility of the components. It is worth noting that the dependence of the free volume in the system is based on the inapplicability of the concept of the isofree-volume state to the glass transition in heterogeneous polymeric systems and, particularly, in the mixtures.<sup>21</sup>

To treat the experimental findings from the colloidochemical point of view, let us discuss some data on the wetting of the surfaces of binary mixtures. It is known<sup>8</sup> that from contact angle measurements the so-called "critical surface tension" can be calculated. This value is not true surface tension. From a thorough investigation of the problem<sup>9</sup> it was concluded that a more suitable characteristic of the surface is the value calculated according to the Elton equation.<sup>7</sup> Taking into account that the values are tightly connected by true surface tension (they coincide in many cases<sup>9</sup>), we have used these values to evaluate the surface characteristics of binary mixtures and will refer to it later as surface tension for convenience.

Considering the fact that changes mainly occur in the range of small additions (Fig. 13), the following concentration ( $c$ ) was used: up to 50% PE,  $\log c/\log 2$ ; and after 50%,  $\log(100 - c)/\log 2$  were plotted. Comparison of these results with the data of Figure 1 reveals that extreme points in both cases can be found in the same composition range. The maximum value  $\gamma$  is higher than values of each constituent. It is concluded that when the mixture is being formed, low molecular weight polymer homologues and surface-active impurities migrate into the interphase area of the disperse system. As such migration proceeds along

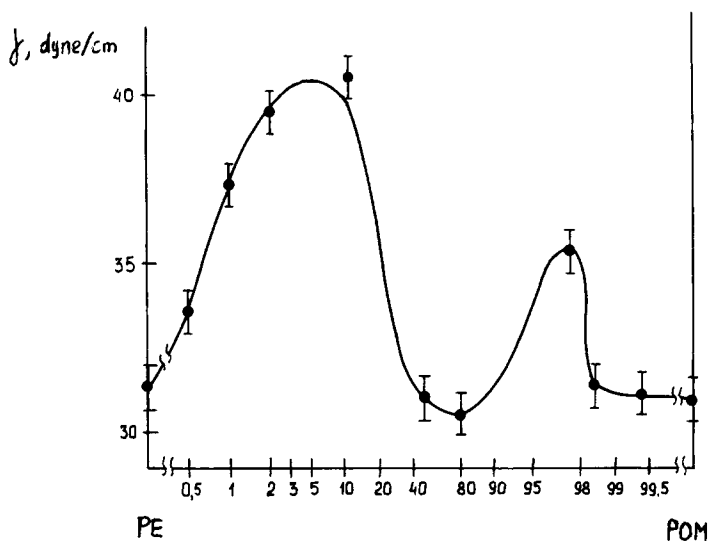


Fig. 13. Surface tension of POM-PE blends.

with decrease of the interphase tension, the process becomes thermodynamically advantageous. An increase occurs in surface tension, which changes with molecular weight according to the equation<sup>22</sup>

$$\gamma = \gamma_{\infty} - K/M^{2/3}$$

where  $\gamma_{\infty}$  is the surface tension of the polymer with infinitely long chains and  $M$  is the molecular weight. Increase in surface tension of the disperse phase can be a thermodynamic reason for the increase in its strength, since strength properties are determined to a large degree by the cohesion energy of the disperse medium. It is known that,  $W_c$  equals twice the value of the surface tension. This is only a qualitative explanation, since direct comparison of the strength characteristics, determined under nonequilibrium conditions with the cohesion energy, may merely prove the existence of certain correlations. Likewise, interphase phenomena in the melts may cause anomalies in mechanical and rheological behavior. Both components have very close values of surface tension, and it may be the reason for the weak interphase tension.<sup>23</sup> The interphase border becomes unstable when the tension reaches several tenths of dynes/cm.<sup>24,25</sup> Under these conditions, when components are being mixed at temperatures higher than their melting points, a process of induced emulgation of one component into the other is possible.<sup>30</sup>

The idea of spontaneous emulgation as a result of microseparation of the incompatible mixture of two polymers was successfully developed by Kuleznev.<sup>20</sup> He has found, that after microseparation when two incompatible polymers form an emulsion, due to absence of interaction at the interphase (when the total border surface is very large), there occurs an anomalous decrease in viscosity. The effects observed are clearly in agreement with these ideas. Our concept of the induced emulgation, in accordance with the direct electron microscopy data,<sup>16</sup> explains formation of the transition layer at the phase border between a disperse particle (additive component) and disperse medium (main component). The transition layer may be described as an emulsion of the type described, formed

as a result of instability of the interphase border; composition of such emulsion transition layer is changed with transition from the phase of one component to the phase of the other. Thus, for the case of small additives of one component to the other, we can discuss an "emulsion" transition layer between two components, the mixture as a whole consisting of emulsion as well, but with particles of greater sizes. The scheme of the transition layer may be presented as follows (Fig. 14). The dispersed particle is surrounded by a transition layer, which is a microemulsion, with particles much smaller than those of the dispersed phase. In principle, reaching the limit equilibrium state, the system should have been a microemulsion, but it did not occur because of high viscosity and lengthy transition. In other cases, the uniform distribution of emulsion particles in the entire volume is not possible. The phenomena discovered for PE-POM systems were found for other blends as well. Anomalies in rheological behavior were observed for polypropylene glycol-polyethylene glycol mixtures of different molecular weights.<sup>26</sup> For mixtures of PE with polyamide and polystyrene, very similar changes of the surface tension were observed.<sup>27</sup> Changes in molecular mobility of both components in the interphase region<sup>28</sup> also contributed to the general behavior of the binary system.

Viscoelastic properties are much influenced by small additives since the glass transition point in the amorphous regions fluctuates. This phenomenon was reported for other systems such as PE-polyamide and PE-PS.<sup>29</sup> For PE, extreme dependence of  $T_g$  on the composition of the mixture with polyamide, which was dependent on the distribution of the components, was found. In the case of small additives restriction of the molecular mobility at the interphase may prevail, whereas in the region of intermediate compositions the main effects are connected with the development of excess free volume, and as a result mobility of macromolecules of both phases in the interphase is increased.

### CONCLUSIONS

The data obtained demonstrate peculiarities in the behavior of the binary mixture with close values of the surface tension of both components. These results allow the following conclusions for determining complex dependence of mechanical and rheological properties of the mixtures on their composition:

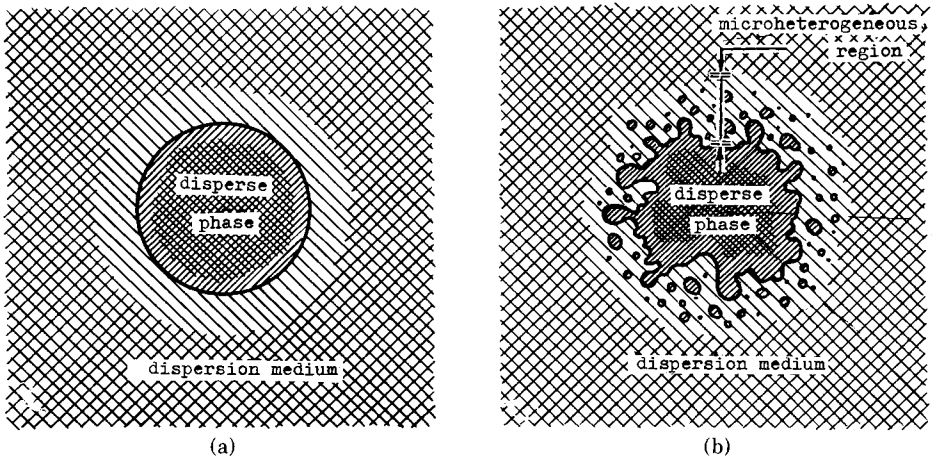


Fig. 14. Scheme of intermediate "emulsion" layer: (a) initial state immediately after mixing; (b) after induced emulgation.

- (1) Thermodynamic incompatibility of the components causes weakening of the adsorption interaction between them or a weakening of the adhesion.
- (2) Development of the excess free volume is localized in the interphase region.<sup>15</sup>
- (3) Formation of the transition layer with different structural organization of the components (ordering degree and packing density) is in the contact area).
- (4) Close values of the surface tension of the components makes the induced emulgation in the interphase zone possible when components are mixed and low molecular weight homologues migrate into the interphase region.

The components of the blend influence molecular mobility and transition temperatures at the interphase border. The sum of these factors and their contributions to the properties of the system as a function of the mixture composition may be the reasons for the anomalies discussed above. At the same time these factors provide certain possibilities to regulate properties of the compositions and conditions for processing.

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