Physicomechanical properties of polybutylene terephthalate modified with polyazomethine

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INTRODUCTION

Along with dispersed and fibrous mineral fillers, wide use is made of polymeric additives for the modification of polybutylene terephthalate (PBT) [1, 2]. The use of such additives has certain advantages. PBT-based materials modified with polymer additives are characterised by increased impact strength, lower density, and increased water resistance [3, 4]. Furthermore, the coefficients of thermal expansion of polymeric additives and of polymer matrices themselves are very similar, which creates the additional effect of strengthening of composites as a result of a reduction in thermal stresses [5]. However, the production of high-quality composite materials based on different polymers, in particular based on PBT and different polymeric additives, is connected with solution of the problem of developing the optimum ratios of components.

EXPERIMENTAL

In the present work an examination is made of the results of physicomechanical investigations of PBT modified with polymeric additives. Previously synthesised polyazomethines (PAMs) of different chemical structure [6] were used as the polymeric additives (Structure A).

Polymeric azomethines were introduced into PBT by the procedure given in [7]. The content of modifiers $C_{PAM}$ amounted to 0.1–1.0 wt.%. It must be pointed out that the selection of these concentrations of PAM is not random, but rather based on the results of investigations in [7].

Impact tests conducted by the procedure described in [8] showed that the addition of PAMs to PBT leads to a significant improvement in the mechanical properties of the initial polymer. Here it was found that unnotched PBT + PAM composites (where $C_{PAM} = 0.1$–$0.3$ wt.%) do not fail, i.e. their impact strength can be estimated at $A_f > 140$ kJ/m$^2$. In turn, the initial PBT fails without a notch. To assess the deformation and strength properties of PBT + PAM composites, the latter were tested with a notch. The results are given in Table 1.

An analysis of the data in Table 1 indicates that a PBT + 0.3%PAM composite has the highest value of $A_f$. This is due to an increase in the plasticity of a composite of the given composition, i.e. the introduction of the more elastic PAM phase into the PBT leads to plastic failure of the material. To

\[ \text{Structure A. Where} \ R = \text{H, CH}_3; \ R' = (\text{CH}_2)_{\text{n}}; \ n = 4, 6, 8. \]
establish in more detail the mechanism of increase in the impact strength of PBT when it is modified with PAM, the fracture surfaces of initial PBT and of a PBT + 0.3%PAM composite under high-velocity loading were studied.

Micrographs were obtained by means of optical microscopy at a magnification of ×100.

RESULTS

An analysis of the fracture surfaces of the initial PBT and of the PBT + 0.3%PAM composite, given in Figure 1, indicates that the nature of failure of the initial polymer differs considerably from that of the composite. As can be seen from Figure 1a, failure of the initial PBT is nucleated on the entire boundary of the surface layer, and then a zone of reduced roughness is observed which is replaced by a zone of relief fractures. On the basis of such a fracture surface, it can be concluded that the initial PBT has brittle failure. In contrast to the initial specimen, the fracture surface of the PBT + 0.3%PAM surface has a different nature. Thus, from Figure 1b it can be seen that the process of failure is nucleated in the region of an artificial defect (notch). However, failure began not over the entire length of the notch but from a certain individual defect. Characteristic elements of the fracture surface are a smooth zone and lines (roughnesses) fanning out from the smooth zone. Such a nature of the fracture surface of the PBT + 0.3%PAM surface is evidently due to the emergence of shear strains on account of the PAM macromolecules. Consequently, shear strain makes a considerable contribution to the higher Af of the PBT + 0.3%PAM composite in impact tests by dissipating the impact energy and by preventing crack growth.

In turn, these results are confirmed by a reduction in the elastic modulus with an increase, accordingly, in the strain εf (Table 1). In this case the introduction of PAMs into PBT leads to an increase in the mobility of macromolecules in relation to each other which is expressed in a drop in viscosity of the melt (increase in the melt flow index values, determined under standard conditions), in the melting temperature, and in the density of the composites (Table 2). Consequently, the addition of PAMs to PBT promotes its plasticisation, which also leads to a change in the mechanical properties of the initial polymer.

Along with an increase in Af and εf of PBT when PAMs are introduced into it in a quantity of 0.3 wt.%, as can be seen from Table 1, there is also an increase in the stress causing failure σf. Thus, σf of the PBT + 0.3%PAM composite is 2.5 times higher than that of the initial polymer. Such an increase in σf can be attributed to the fact that the introduction of PAMs into PBT leads to an increase in impact energy dissipation. This, in turn, signifies a certain increase in the level of molecular mobility of the composite, which confirms earlier reasoning concerning the plasticising role of PAMs.

### Table 1. Deformation and strength properties of PBT modified with PAM, determined in impact tests by Charpy procedure [1, 2]

<table>
<thead>
<tr>
<th>Specimens</th>
<th>Af, kJ/m²</th>
<th>σf, MPa</th>
<th>E, GPa</th>
<th>εf, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>PBT (V-110) TU2226.001-453722 59–98</td>
<td>18</td>
<td>25</td>
<td>1.1</td>
<td>2.4</td>
</tr>
<tr>
<td>PBT + 0.1% PAM</td>
<td>22</td>
<td>35</td>
<td>1.3</td>
<td>3.0</td>
</tr>
<tr>
<td>PBT + 0.3% PAM</td>
<td>29</td>
<td>56</td>
<td>0.9</td>
<td>7</td>
</tr>
<tr>
<td>PBT + 1.0% PAM</td>
<td>21</td>
<td>38</td>
<td>1.2</td>
<td>3.5</td>
</tr>
</tbody>
</table>

Notes
1. Specimens were tested with notch of 0.2 mm.
2. PAM:

![polymeric structure]

Figure 1. Fracture surfaces of specimens based on initial PBT (a) and PBT + 0.3 wt.%PAM composite (b)
This argument is borne out by the results of dielectric investigations of PBT–PAM composites (Figure 2). Dielectric investigations were carried out by the procedure described in [8]. From Figure 2 it can be seen that the molecular mobility of the PBT + PAM composite is slightly higher than in PBT in the region of lower temperatures – below the glass transition temperature. Furthermore, the temperature of the start of through conductivity is also slightly higher in the case of the PBT + 0.3 wt. % PAM composite.

CONCLUSIONS

Thus, the results obtained make it possible to draw the following conclusions.

The introduction of small additions of PAM ($C_{PAM} = 0.1–0.3$ wt.%) into PBT has a modifying effect on the main physicomechanical properties of the initial polymer.

A change in the main physicochemical properties of PBT as a result of the plasticising effect of PAM has been found.

The results obtained make it possible to create materials with a combination of improved deformation and strength characteristics.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>$\text{MFI}_{203}$, g/10 min</th>
<th>$T_{\text{melt}, \text{K}}$</th>
<th>$\rho$, g/cm$^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Industrial PBT</td>
<td>36</td>
<td>487</td>
<td>1.245</td>
</tr>
<tr>
<td>PBT + 0.1%PAM</td>
<td>45</td>
<td>483</td>
<td>1.240</td>
</tr>
<tr>
<td>PBT + 0.3%PAM</td>
<td>65</td>
<td>470</td>
<td>1.230</td>
</tr>
</tbody>
</table>

**Table 2. Certain properties of PBT and PBT + PAM composites**

![Figure 2](No date given)

**Figure 2.** Temperature dependence of tangent of angle of dielectric losses $\tan \delta$ for initial PBT (1) and PBT + 0.3 wt. % PAM composite (2)

REFERENCES


(No date given)