Properties of talc-filled polypropylene

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Composites of talc-filled polypropylene (PP) are widely used in the motor industry for the injection moulding of vehicle interior elements — panels, stanchions, insertion pieces, etc. The introduction of a dispersed filler improves the processing properties, increases the service indices, and lowers the cost of articles (ref. 1). However, the achievement of maximum technical and economic effectiveness in the processing and use of polypropylene is possible only when a composite of optimum composition is used. Experience with filled PP indicates that the combination of components differing significantly in physicochemical and thermophysical properties has a considerable effect both on melt rheology and on the process of structure formation of the matrix phase, and consequently on the physicomechanical properties of injection moulded articles.

In spite of the large number of studies of filled PP, the laws governing melt flow and the formation of strength properties, associated with the ratio of components, the filler particle shape, and the processing parameters, remain little studied, which makes it difficult to optimise injection moulding. The aim of the present work was to investigate the melt rheology and strength properties of talc-filled PP with different processing schedules.

The investigation was carried out on Balen-grade PP (TU 2211-020-00203521-96) with a melt index of 10.5 g/10 min, a yield point in elongation of 34.8 MPa, and a breaking elongation of 9.0%, filled with 4.4, 9.8, 19.6, and 39.5% talc TPM-V (TU 27-003-10733471-2000) and with 4.4, 9.8, 19.6, and 26% talc MT-2 (TU 5727-002-21250238-95). As shown by sedimentation analysis, the specific surface of talc TPM-V was 0.54 m²/g. Here, 69% of the particles have an effective diameter of less than 20 mm, and 6% an effective diameter of over 90 mm. Talc MT-2 has a specific surface of 0.97 m²/g and contains 86% particles with a size of less than 20 mm. To facilitate processing and prevent degradation, 0.25% heat stabiliser and lubricant was introduced into the composition.

Rheological investigations were carried out on a constant-pressure viscometer of the IIRT-AM type with a capillary of 2.095 mm diameter at a temperature of 230°C under loads of 3.75, 12.0, 21.6, 38.0, and 50.0 N. To assess the entry losses, use was made of a die of 1.18 mm diameter. The entry pressure losses during flow through the capillary of 2.095 mm diameter depended on the degree of filling of PP and ranged from 61 to 63% for a load of 3.25 N, from 50 to 52% for a load of 12 N, from 28 to 33% for a load of 21.6 N, from 19 to 26% for a load of 38 N, and from 13 to 22% for a load of 50 N. The pressure losses were taken into account when determining the flow index and the effective viscosity.

For mechanical tests, use was made of at least eight type I specimens to GOST 11262-85 with a width and thickness of the working part of 8 and 2 mm respectively, injection moulded on a vertical machine at a unit pressure of 60 MPa and a temperature of the melt and mould of 230 and 70°C respectively.

An analysis of rheological curves (Fig. 1) indicates that, in the region of shear stresses above 5300 Pa, PP flows like a pseudoplastic liquid — the melt viscosity decreases and, with increase in the shear stress (rate) to 38 332 N, reaches a minimum value of 370 Pa s.

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PP melt flow of this nature is due to strain of statistical coils of macromolecules, and to their extension and orientation in the direction of action of the force. With increase in the shear stress, the strain of the coils increases, while the number of entanglements of macromolecules decreases, which leads to a reduction in internal friction and effective viscosity. The flow index, comprising the slope tangent of the logarithmic stress–shear rate relationship, which characterises the viscosity anomaly, amounts to 0.89 for PP.

The appearance in PP of talc agglomerates readily breaking down during flow is accompanied with a sharp increase in melt/suspension flow. Thus, under a stress of 1082 Pa, the effective viscosity of PP amounts to 659 Pa s, while that of a composite containing 4.4% talc TPM-V and MT-2 is 142 and 712 Pa s respectively. Although, as the talc concentration increases to 19.6%, the viscosity of the composites increases, its magnitude does not exceed the value for the base grade. For example, with a shear stress of 9300 Pa, the effective viscosity for the base grade and the composite amounts to 501, 455, and 590 Pa s. With higher degrees of filling, the effective viscosity of the composite continues to decrease, and the effective viscosity exceeds that of the base grade. Thus, with a shear stress of 31 416 Pa, the viscosity of PP composites containing 39.5% TPM-V and 26% MT-2 amounts to 374, 551 and 476 Pa s respectively.

The flow index of composites (see Table 1) with a talc content of up to 4.4% increases to 0.96 by comparison with 0.89 for unfilled PP, which indicates a reduction in the viscosity anomaly and flow of the composite containing a small amount of plastic filler, irrespective of the type of talc, like a Newtonian fluid. With a higher talc content, a viscous flow anomaly again appears and is gradually intensified. Thus, with a filler content of 9.8%, the flow indices of unfilled and filled PP practically coincide (0.89, 0.85, 0.84), and with a content of 39.5% they are 0.82.

Analytically, the ratio between the stress and shear rate of the studied composites and unfilled PP is approximated by a power-law relationship:

$$\tau = \eta_{\text{eff}} \gamma^n$$

where $\tau$ is the shear stress (Pa), $\eta_{\text{eff}}$ is the effective viscosity, $\gamma$ is the shear rate (s$^{-1}$), and $n$ is the flow index.

The correlation coefficient of the logarithmic stress–shear rate relationship ranged from 0.963 to 0.996, which corresponds to a 99% level of significance.

The obtained laws governing the change in rheological properties as a function of the content and size of talc particles can be explained from positions of the physicochemical mechanics of dispersed systems (ref. 2), according to which the powder fillers produced by the crushing of natural minerals are characterised by a particular degree of aggregation of particles. Talc is a very soft mineral that has the lowest hardness among the fillers of plastics – hardness 1 according to the Mohs scale (ref. 3).

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**Table 1  Rheological properties of PP composites**

<table>
<thead>
<tr>
<th>Filler content, %</th>
<th>0</th>
<th>4.4</th>
<th>9.8</th>
<th>19.6</th>
<th>26, 39.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow index</td>
<td>0.89</td>
<td>0.96/0.96</td>
<td>0.85/0.84</td>
<td>0.82/0.84</td>
<td>None/0.87/0.82/none</td>
</tr>
<tr>
<td>Viscosity $\eta_{\text{eff}}$, Pa·s, with shear rate of 1</td>
<td>636/642</td>
<td>538/560</td>
<td>742/746</td>
<td>835/851</td>
<td>None/875/1136/none</td>
</tr>
</tbody>
</table>

_Note: Numerators – TPM-V; denominators – MT-2_
Talc particles readily form aggregates, the size of which depends on the degree of dispersion of the fillers — large associates in the case of particles of less than 7–8 µm and smaller sizes for particles with a size of over 20 µm, as indicated, in particular, by the bulk density of the filler, amounting to 857 kg/ m³ for talc TPM-V and to 786 kg/ m³ for talc MT-2. Asymmetric particles and associates of particles in the melt suspension are oriented and broken down during viscous flow, facilitating slip of segments of macromolecules, and therefore, with a low talc content, the melt viscosity amounts to 130–175 Pa s and remains constant for all processing rates, which corresponds to the rheological behaviour of a Newtonian fluid.

Aggregates of talc particles possess different resistance to the action of shear stresses, and therefore it is far from always possible, during the preparation of a composite in a screw press, even with prolonged and intense mixing of the components, entirely to break down all aggregates and wet the surface of the individual particles. The presence in the melt of filler aggregates is equivalent to an increase in the volume concentration, and therefore the viscosity of the composite should increase more strongly when particles of small size, prone to aggregation, are introduced, even with an unchanged weight content of talc. In the process of determining the rheological properties under the action of shear stress, breakdown of some of the aggregates occurs, as a result of which the melt viscosity decreases, this decrease being more intensive the higher the shear stress. Aggregates of larger particles break down more readily, and therefore the viscosity of a composite with talc MT-2 is higher than that of a composite with TPM-V (see Table 1).

Rheological investigations can be the basis of description of the laws governing the formation of the physico-mechanical properties of talc-filled PP. By comparing the rheological properties of the melt with the strength properties of moulded composites, it is possible to substantiate the optimum injection moulding parameters. Figure 2 gives the shrinkage and yield point of composites with different filler contents.

As can be seen from Fig. 2, the strength of a composite with an increased content of talc TPM-V initially decreases slightly to 32.5 MPa, then increases, and with a content of 9.8–19.6% exceeds the yield point of the base grade, reaching 34.9 MPa. With a higher concentration, the strength of the composite gradually decreases to 32 MPa at 39.5%. In the case of filling of PP with talc MT-2, a monotonic decrease in strength is observed to 25.2–26.0 MPa.

As the talc content increases (Fig. 2), the shrinkage of mouldings decreases from 3.27% for PP to 0.875 and 0.881% for a composite with 39.5% TPM-V and 26% MT-2 respectively. A sharp reduction in shrinkage to 1.4% occurs at a filler concentration of 4.4%, which is due to the presence of a large number of crystallisation centres promoting the formation of a denser structure. Increase in the filler content to 9.8% causes a much smaller change in shrinkage — to 1.2% for PP filled with talc TPM-V and to 1.35% for PP filled with MT-2. With increase in the talc concentration, the differences in shrinkage, connected with the particle size, become more evident — with the same talc content, a composite based on finer MT-2 particles shrinks more strongly than a composite filled with larger TPM-V particles, for example, with a content of 19.6% the shrinkage is 1.02 and 1.26% respectively.

As the talc content increases, the water absorption by PP within 24 h increases from 0.017 to 0.046–0.040% for a 39.5% concentration of coarsely and finely dispersed filler respectively.

Unfilled PP has an amorphous–crystalline structure with a degree of crystallinity ranging from 0.5 to 0.75 depending on the cooling rate of the moulded article (ref. 4). With a cooling rate of less than 0.5 K/min, a coarse spherulite structure is formed with a size of the formations of 55–65 µm. With cooling rates of 0.5–1.2 K/min, a crystalline structure is formed with a spherulite size of 23–12 µm. The introduction of a dispersed filler increases the number of crystallisation centres with increase in the degree of crystallinity to 0.75 and reduction in the spherulite size to 8 mm. The morphology of the structure has a considerable influence on the strength properties of articles of PP. In a field of mechanical forces, coarse spherulites readily crack around the periphery, and also over the contact surface with neighbouring macrostructural formations, resulting in a reduction in the yield point.

Owing to the lamellar shape of the particles and the high characteristic ratio of their geometric characteristics – the thickness is 0.1–0.18 of the length (width) of the lamellae – talc is potentially a reinforcing filler (ref. 1).
However, the mechanical properties of PP composites depend not only on the content but also on the distribution and size of the talc particles.

The results obtained can be explained by the fact that the strength of composite material filled with particles of characteristic shape depends on the competing influence of two effects: increase in the strength of the composite on account of 'constriction' of the binder on the one hand and breakdown of the talc agglomerates on the other. With degrees of filling of 20 and 30% for MT-2 and TPM-V particles respectively, with good impregnation and in the absence of agglomeration, a strengthening effect of the filler is realised. With a higher filler content, aggregation of particles and incomplete wetting lead to weakening of the composite.

Thus, with filling of up to 9.8%, the strength properties of talc-filled composites are determined by the conditions and rate of crystallisation of PP in the presence of talc, while with higher filler contents they are determined by the melt viscosity. The agglomeration of comminuted talc, increasing with increase in the degree of dispersion and affecting the flow, the strength properties, and the qualitative indices of appearance (colour, dullness, the appearance of stains, and so on), is a considerable obstacle to the production of highly filled composites. A promising solution of the given problem is to use additives preventing agglomeration — surfactants that facilitate the wetting of talc particles by the PP melt.

The results of investigations were used in the creation of composites of talc-filled polypropylene Novokom, which is successfully used for the injection moulding of articles for the VAZ Motor Works.

REFERENCES

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(No date given)