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Time-dependence of the tensile stiffness and anisotropy of a reinforced thermoplastic

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The stiffening in tension of injection-moulded polypropylene with the addition of glass fibres or spheres was found to depend on strain, time and the direction relative to that of the melt flow, as well as on the percentage and aspect ratio of the glass inclusions. Under loading of long duration this stiffening, expressed in terms of the stiffness factor, increased with time for fibre-filled material but varied little for the sphere-filled. In the latter case, the tensile creep could be predicted with some accuracy by a simple model. The anisotropy of the fibre-filled polypropylene was not time-dependent and conformed to the theory of orthotropic materials.

1. Introduction

Following the successful use of glass fibre-plastics composites made with thermosetting resins, there has been increasing interest in materials in which glass fibres are combined with thermoplastics. Because of the viscoelasticity of the matrix and orientation of the fibres, the properties of these materials are, in general, both time dependent and anisotropic. Usually, the glass is included in the thermoplastic in the form of short, chopped fibres for convenience of processing by, for example, extrusion or injection moulding. It has been shown that considerable improvements in the stiffness of thermoplastics can be achieved by the addition of glass fibres in this way [1]. More recently, the inclusion of small glass spheres in thermoplastics has been considered as a means of producing a stiffer material [2].

The improvement obtainable in the stiffness of a thermoplastic by reinforcing it with glass has been shown to depend on a number of factors in addition to the glass content. Amongst these are the quality of the coupling between the glass and the thermoplastic matrix [3, 4], the strain imposed on the material [5, 6] and the aspect ratio and degree of alignment of the glass inclusions [6].

The possible influence of several other factors remains, however, to be elucidated. One of the most important of these is the effect of time under load, as it might be expected that the viscoelasticity of the thermoplastic matrix would have a considerable influence on the stiffness of the composite. A further problem to be clarified concerns the anisotropy of the composite material arising out of the orientation of the fibres [7, 8] and, possibly, of the matrix material [9] during processing, and, in particular, the dependence of such anisotropy on time under load.

In this paper measurements are described of the uniaxial tensile creep of polypropylene containing glass in the form of either fibres or spheres, and its variation with glass content, filler geometry and direction relative to that of the melt flow.

2. Experimental procedure

2.1. Materials and specimen preparation

The materials used in this study were based on a polypropylene homopolymer (ICI Propathene GW 522 M) containing a coupling agent. This was filled with different amounts of either $\frac{1}{4}$ in. (6.35 mm) Owens-Corning 801 K-filament glass fibres (13 to 14 μm diameter) or Ballotini

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3000/CP03 glass spheres (mean diameter about 25 μm). The behaviour of these materials at short loading times was the subject of an earlier investigation by the authors [6] in which the results were compared with different theories of reinforcement. Polypropylene was chosen because in both its unfilled and glass-filled forms it is a material of considerable industrial interest and because it was known to be more sensitive to the quality of the glass to polymer bond than some other thermoplastics [3, 10].

The filled and unfilled polypropylene were direct-injection moulded (i.e. without preblending the polymer and the glass) in the form of ⅛ in. thick (approximately 3 mm) ASTM type I tensile specimens [11] in end-gated moulds. These moulds ensured a high degree of alignment of the glass fibres parallel to the axis of the specimens, which was desirable to highlight differences between the effect of the fibres with their relatively high aspect ratio and that of the unit aspect ratio spheres [6]. Smaller specimens were subsequently machined from the centre of the ASTM tensile specimens to suit either ICI-type [12] or GKN-type [13] tensile creep testing machines. The long axis of the machined specimens lay on that of the moulded ASTM specimens and was, therefore, parallel to the direction of alignment of the fibres and to any orientation in the polymeric matrix [9]. The filled polypropylene contained three different nominal amounts of either glass fibres or spheres, namely 10, 20 and 30 % by weight. These corresponded to volume fractions of 3.7, 8.1 and 13.2 % respectively.

In addition to the above, the 20 % by weight fibre-filled material and the unfilled polypropylene were direct-injection moulded in edge-gated moulds in the form of ⅛ in. thick, square plaques measuring 165 × 165 mm^2. These moulds were used in an earlier investigation into the tensile properties and anisotropy of glass filled nylon [14]. It was inferred that these moulds produced a somewhat lower degree of fibre alignment in the plaques than was obtained in the ASTM specimen mouldings since specimens cut in the melt flow direction from the plaques had a tensile creep modulus under corresponding conditions which was approximately 10 % lower than that of specimens from the ASTM-type moulds (although such a difference could also reflect the effect of variations in the moulding conditions on the polypropylene matrix [9]). Tensile specimens of the ICI-type or GKN-type were cut from the plaques at different angles to the nominal direction of melt flow in order to study the anisotropy of the material.

2.2. Test method

The tensile creep measurements were performed on ICI-type [12] or GKN-type [13] tensile creep testing machines. Both of these are dead-weight lever-loading machines operating with a nominal lever arm ratio of 5 to 1. Strains are measured using modified Lamb's optical extensometers [15]. The test procedure, from the machining of the specimens to the calibration of the machines and extensometers, is designed to produce results within an overall accuracy of 1 %. This corresponds to class A of BS 4618 [16].

Specimens were loaded in these machines either using an interrupted step loading technique to obtain 100 sec isochronous tensile stress-strain curves [15], or using constant loads for periods up to 3×10^6 sec (about 4½ weeks) to obtain constant load tensile creep curves. All the tests were performed in a controlled environment at a temperature of $20 \pm \frac{1}{2}$°C and a relative humidity of 60 ± 5 %. The test materials had been stored for several weeks in the same environment before being tested.

3. Experimental results

3.1. Strain effects and stiffness factors

The 100 sec isochronous tensile stress-strain curves for the glass fibre-filled and unfilled polypropylene are shown in Fig. 1. As might be expected, the slopes of the stress-strain curves for the fibre-filled materials are considerably greater than that for the unfilled material, and the slopes increase with increasing fibre content.

The slope of all the curves, however, decrease with increasing strain, indicating that glass-filled as well as unfilled polypropylene behaves as a non-linear viscoelastic material [17]. What the curves do not bring out is the fact that, although the stiffness of the fibre-filled polypropylene decreases with increasing strain, the relative effectiveness of the fibres in stiffening the matrix material does not.

This relative effectiveness of fillers in stiffening a material is best expressed in terms of a stiffness factor (SF). This has also been called a reinforcement factor [4] or a (modulus) enhancement factor [10]. It may be redefined as the ratio of the stresses required to produce equal strains, ϵ, in the filled and unfilled materials. If these stresses are denoted by σ_F and σ_U, respectively, then

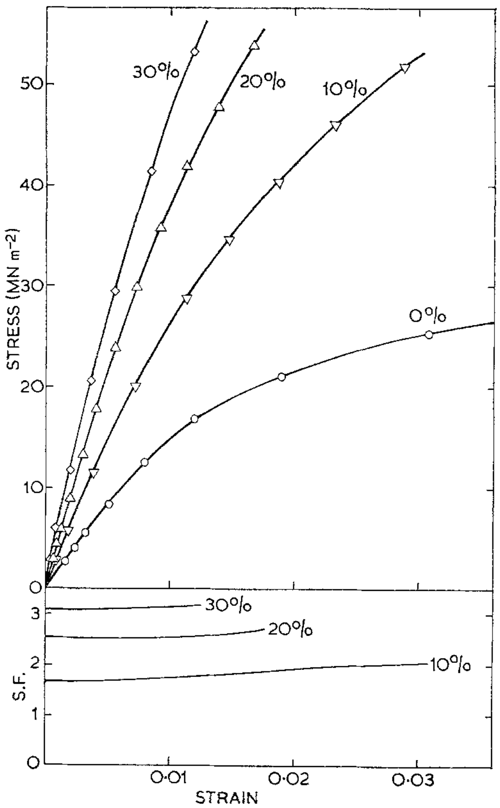

Figure 1 100 sec isochronous tensile stress-strain curves for polypropylene with 0, 10, 20 and 30% of glass fibres by weight, and, below, the corresponding stiffness factor-strain curves.

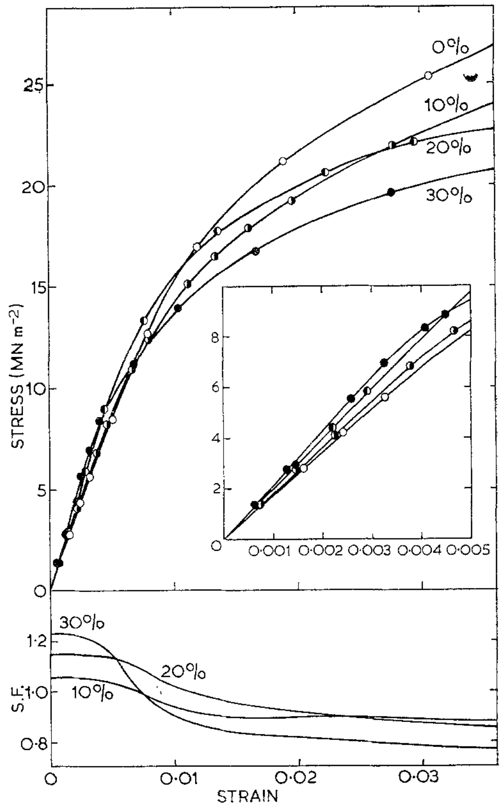

Figure 2 100 sec isochronous tensile stress-strain curves for polypropylene with 0, 10, 20 and 30% of glass spheres, by weight, and, below, the corresponding stiffness factor-strain curves.

$$SF = \left(\frac{\sigma_F}{\sigma_U}\right)_\epsilon \cdot \qquad (1)$$

Values of SF for the three fibre-filled materials are shown in the lower part of Fig. 1 in which it can be seen that, following the pattern of the stress-strain curves, the SF values increase with increasing fibre content. What is more significant in the context of this paper is that these values increase with increasing strain, demonstrating that the relative effectiveness of the fibres is an increasing function of strain in the range of strains considered here.

The results for the glass sphere-filled polypropylene present quite a different picture. Fig. 2 shows the 100 sec isochronous tensile stress-strain curves for the three sphere-filled materials and for the unfilled polypropylene, together with the corresponding curves of SF. The values of SF are lower than those for the fibre-filled materials at all strains, but, more significantly, after a small region of constant SF at low strains, the values of SF decrease with strain, falling below unity at strains above about 1%. This indicates that the spheres, in addition to the expected result of being less effective than the fibres in stiffening the polypropylene, actually have the effect of making the material less stiff than the unfilled material at strains greater than about 1%.

3.2. Time effects

The behaviour of the different forms of glass-filled polypropylene under long term loading was investigated by means of constant load creep tests, and a typical set of creep curves, in this case for the unfilled and the 20% by weight glass-filled materials, are shown in Fig. 3 for two different values of starting strain. It is apparent that the rate of creep of the fibre-filled material is considerably lower than that of the material filled with spheres despite the differences in the stress levels applied to the two materials. It can

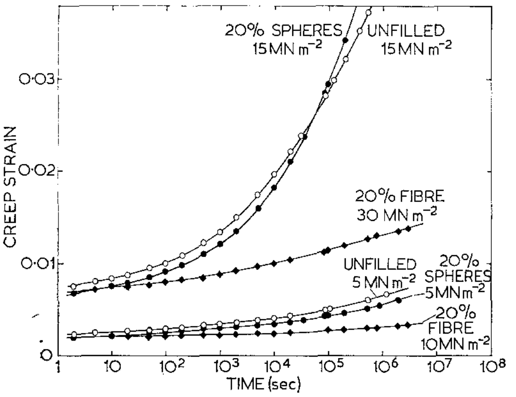

Figure 3 Creep curves for polypropylene with 20% by weight of glass fibres or glass spheres and for unfilled polypropylene.

also be seen that, whilst at the lower starting strain there is little to choose between the sphere-filled and the unfilled materials, at higher strains the creep rate of the sphere-filled polypropylene is greater than that of the unfilled.

The relative effectiveness of the fillers is again best brought out in terms of the SF. In Fig. 4 values of SF at two different strains are shown as a function of time for the three fibre-filled materials. The values of the stresses at different times at the two strain levels were determined by interpolation of the experimental creep data plotted on double logarithmic axes to obtain the strains at 100 sec and, hence, the stresses from the 100 sec isochronous results. The use of this procedure increases the possible errors in the absolute values of SF, but any such errors are still much smaller than the variations of SF with time shown in Fig. 4. The increasing values of SF with time show that the fibres become relatively more effective as stiffeners when the time-dependent softening of the polypropylene matrix increases. This effect might be regarded as analogous to that found in glass fibre-filled nylon under short-term loading conditions in which the SF increases as the matrix softens due either to increasing temperature [5] or to increasing humidity [18].

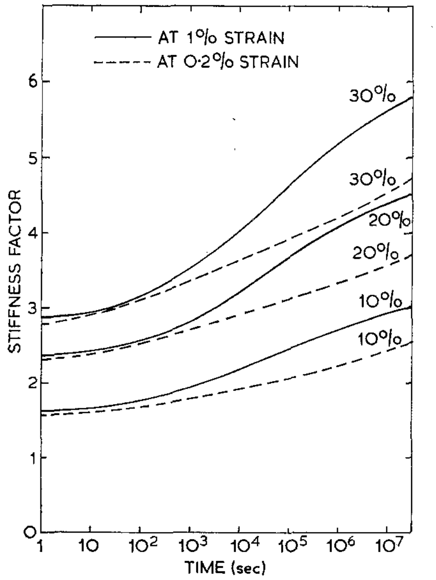

Figure 4 Stiffness factor versus time at 0.2 and 1.0% strain for polypropylene with 10, 20 and 30% by weight of glass fibres.

The pattern of variation of SF with time for the glass sphere-filled material, which is shown in Fig. 5, is more complex. At 0.2% strain the SF appears virtually independent of time for all three sphere contents. At 1% strain, however, the values of SF apparently increase with time initially and then pass through a maximum. Although the values at 100 sec are consistent with those derived from the 100 sec isochronous results shown in Fig. 2, it is difficult to reconcile the variation of SF with time at these higher strains with the variation with strain found earlier. It should be borne in mind, however, that the very much greater creep rate of the sphere-filled materials compared with the fibre-filled at the higher strains made the interpolation procedure subject to much larger errors.

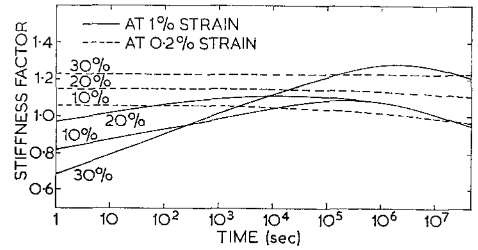

Figure 5 Stiffness factor versus time at 0.2 and 1.0% strain for polypropylene with 10, 20 and 30% by weight of glass spheres.

3.3. Creep model

As in the case of behaviour at short times, it is desirable to be able to predict the creep of reinforced thermoplastics from a knowledge of the properties of their constituents and the proportions in which they are combined. An attempt was made, therefore, to apply the simple models of the stiffness of composites which the authors had used with some success to predict 100 sec isochronous tensile stress-strain curves [6]. In particular, the solution for the tensile secant modulus, E_C, of a two-phase composite based on the "cube-within-cube" model and the assumption that stresses do not vary in the direction of the applied tensile stress, was applied to the sphere-filled material. This solution is given by

$$E_C = E_P \frac{\{E_G(1 - n^{1/3} + n) + E_P(n^{1/3} - n)\}}{E_G(1 - n^{1/3}) + E_P\, n^{1/3}} \qquad (2)$$

where E_P = tensile creep (secant) modulus of the plastic; E_G = tensile modulus of glass; n = volume fraction of glass.

The results are illustrated in Fig. 6, which indicates a reasonably close correlation between the curves based on Equation 2 and the experimental results, except in the case of the material containing the greatest amount of glass (30% by weight) at the higher stresses. This is in keeping with this material's greater decrease in SF with increasing strain under loads of short duration (Fig. 2). The increasing discrepancy between the theoretical and experimental results with increasing strain and increasing glass sphere content in Fig. 6, together with the reduction of SF with strain in Fig. 2, might be ascribed to an increasing debonding between the glass spheres and the polypropylene matrix [6].

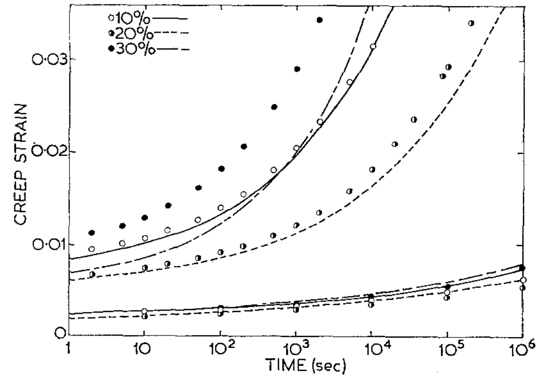

Figure 6 Creep of polypropylene with 10, 20 and 30% by weight of glass spheres compared with predicted creep curves based on Equation 2.

No comparably close correlation between the theoretical models and the experimental results was obtained in the case of the fibre-filled materials. The solutions derived earlier by the authors for their short-term secant moduli were based on a "prism-within-prism" model, and required a knowledge of the aspect ratio of the fibres. Although their nominal aspect ratio was known, from the dimensions of the fibres, to be 470 (the ratio of their length to their diameter), there are several factors which act to reduce the effective aspect ratio of the fibres from the nominal value. Amongst these are reductions in the lengths of the fibres due to processing damage (although direct-injection moulding, as opposed to pre-blending and then injection moulding, decreases the damage to the fibres [3, 19]), clumping of the fibres instead of their being evenly dispersed through the material, and imperfect alignment of the fibres in the direction of melt flow. Thus the values of effective aspect ratio inferred by fitting the solution based on the "prism-within-prism" model to the observed short-term results for these materials ranged from 50 to 150 [6].

Using these effective aspect ratios in an attempt to predict the tensile creep results did not lead to creep curves which correlated with the observed creep strains. Correlation could only be achieved by allowing the aspect ratios in the solution to vary with time. Some variation in the effective aspect ratio of the fibres might certainly have occurred during creep, but as such a variation could not be determined independently of the theoretical solutions this question was not pursued further.

3.4. Anisotropy

Any orientation of the fibres in the specimen mouldings was bound to produce anisotropy in stiffness in the plane of the orientation. The presence of this anisotropy was confirmed by performing 100 sec interrupted step loading tests [15] on 20% by weight fibre-filled specimens cut at different angles to the nominal direction of the melt flow into the edge-gated 165 × 165 mm² square moulds. Values of the tensile creep modulus at 0.2% strain were obtained from the 100 sec isochronous stress-strain curves, and these values are shown plotted as a function of angle to the direction of melt flow in Fig. 7. Preliminary tests had shown that the values of modulus measured parallel to the direction of melt flow varied across the plaque by about 8% of the mean value, with maximum values at the centre and edges of the plaque. Thus, in order to minimize effects due to this variation, all specimens were cut so that their centres lay on the

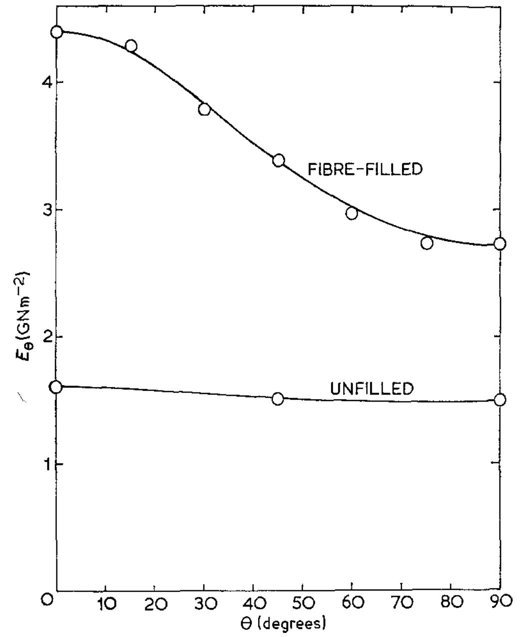

Figure 7 100 sec tensile creep modulus at 0.2% strain at different angles to the direction of melt flow for polypropylene with 20%, by weight of glass fibres and for unfilled polypropylene compared with curves derived from Equation 3.

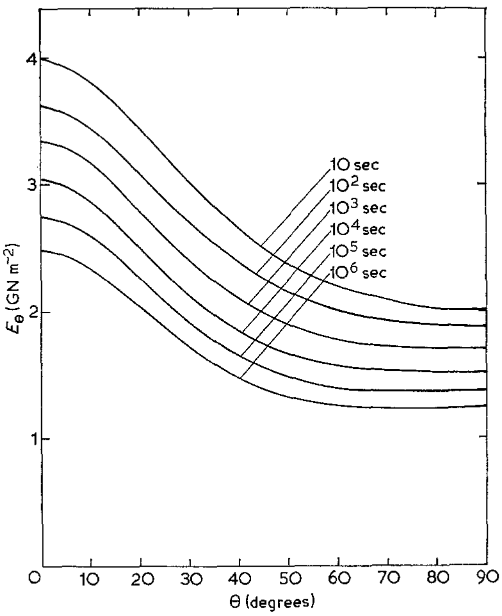

Figure 8 Curves of tensile creep modulus at 1% strain versus angle to direction of melt flow at different times for polypropylene with 20% by weight of glass fibres.

centre-line of the plaque which was parallel to the melt flow direction.

Also shown in Fig. 7 is the variation of the 100 sec tensile creep modulus with angle to the melt flow direction of the unfilled polypropylene. It is apparent that there is virtually no anisotropy present in the unfilled material and this was confirmed by the lack of any detectable crystallite anisotropy in X-ray diffraction photographs. A similar lack of crystallite anisotropy was found for the polypropylene matrix of the fibre-filled material.

The pattern of anisotropy was found to fit the theoretical solution for the variation of modulus of an orthotropic material [20], which the authors had earlier found to describe the anisotropy of oriented but unfilled polypropylene [9] as well as that of a glass fibre-epoxy resin system [21]. In particular, the tensile creep modulus E_θ at any angle θ to the direction of melt flow could be determined from the values of modulus at 0, 45 and 90° and the relation

$$\frac{1}{E_\theta} = \frac{\cos^4\theta}{E_0} + \frac{\sin^4\theta}{E_{90}} \times \left\{\frac{4}{E_{45}} - \frac{1}{E_0} - \frac{1}{E_{90}}\right\} \sin^2\theta \cos^2\theta \cdot \qquad (3)$$

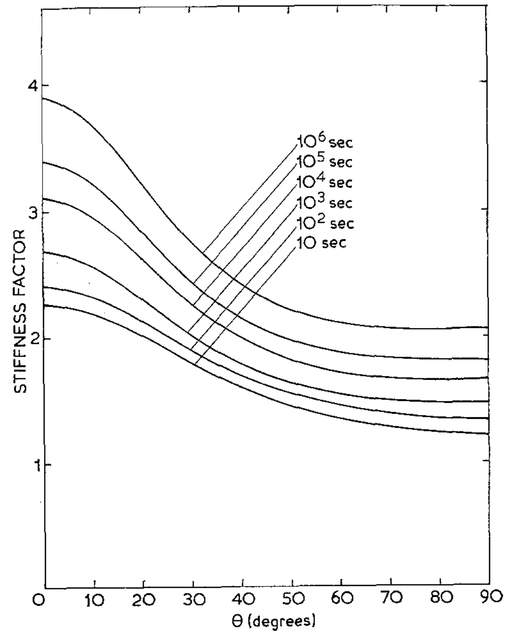

Figure 9 Stiffness factor at 1% strain versus angle to the direction of melt flow for polypropylene with 20% by weight of glass fibres at different times from the commencement of loading.

The close correlation between the experimental points and the curves based on Equation 3 is shown in Fig. 7. This enabled creep tests to be confined to specimens cut at 0, 45 and 90° to the direction of melt flow. From the results of these tests, and following a similar interpolation procedure to that previously adopted, values of the tensile creep modulus at different times at 1% strain were determined. This higher strain level was chosen since it was thought that any change in anisotropy with time was more likely to be brought out at higher strains. Fig. 8, which shows the pattern of anisotropy at various times, was constructed using these values of modulus and Equation 3. There is no indication that the pattern of anisotropy in this glass fibre-filled thermoplastic changes with time for loading periods up to 10^6 sec (approximately two weeks).

The values of the SF at 1% strain show a similar pattern of anisotropy to those of modulus (Fig. 9). However, they increase with time at all angles to the direction of melt flow, indicating that the increasing relative effectiveness with time of the fibres in stiffening the material occurs at all angles, although the effect is greatest in the direction of melt flow.

4. Conclusions

The improvement in the tensile stiffness of injection moulded polypropylene by the addition of glass fibres or spheres was found to decrease with strain and time under load and, in the case of the fibre-filled material, with direction away from the direction of melt flow during the moulding of the material. The improvement is best expressed in terms of a stiffness factor, which was found to be independent of strain but to increase with time for the fibre-filled material; for the sphere-filled material the stiffness factor decreased with strain and varied little with time, which shows that low aspect ratio fillers such as spheres are far less effective in stiffening a plastic than fibres.

The creep of the sphere-filled material was found to correlate with that predicted by a simple "cube-within-cube" model of a composite material and the anisotropy of the fibre-filled material was found to conform to the pattern given by the theory of orthotropic materials and to remain independent of time for loading periods of up to 10^6 sec.

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