JOURNAL OF MATERIALS SCIENCE 33 (1998) 3101–3108

Residual stresses in short-glass-fibre-reinforced thermoset injection mouldings

S. B. WILKINSON and J. R. WHITE
Materials Division, Department of Mechanical, Materials and Manufacturing Engineering, University of Newcastle upon Tyne, Newcastle upon Tyne NE1 7RU, UK

Residual stress distributions have been measured in injection mouldings made from glass-fibre-reinforced phenolic and polyester (dough moulding compound (DMC)) thermosets. The results were inconsistent and this is believed to correspond to variability within the materials rather than to measurement error. The phenolic composite mouldings tended to have compressive residual stress near to the surface and tensile stress in the interior but examples were found in which this sense was reversed. Post-curing the phenolic composite mouldings caused the residual stress magnitudes to increase and reduced the variability in the observed stress distributions. Two DMCs were investigated. The stresses were generally lower in magnitude than those observed in the phenolic composite mouldings; examples in which the residual stress was compressive near the surface and those in which it was tensile were found with almost equal probability.

1. Introduction

Short-fibre-reinforced polyester and phenolic thermosetting resins have an excellent combination of properties with better stiffness, strength and operating temperature range than most reinforced thermoplastics. Grades have been developed that can be injection moulded but their properties can be highly variable even for mouldings made in the same batch and under the same conditions, and this limits their use [1–6]. The variability in properties is probably the result of fibre clumping and variations in the fibre orientation distribution. A study of variations in Young's modulus and density has been presented elsewhere [7, 8].

Another source of property variation in injection mouldings is the residual stress distribution. Residual stresses cause warping when asymmetrical about the midplane in the wall of a moulding and may have an influence over the fracture behaviour. Considerable attention has been paid to residual stresses in injection-moulded thermoplastics [9–28] but very little literature exists regarding residual stresses in thermoset mouldings. Some studies have been made of thermosetting coatings but there are few reports concerning residual stresses in thick sections (say 2 mm or greater). Miyano *et al.* [29] studied rapidly quenched samples and their measurements indicated that the residual stresses were thermally produced, the result of differential cooling. Srivastava and White [30] examined sheets 5 mm thick made from an epoxy resin using different curing temperatures and showed that the stresses were compressive near the surface and tensile in the interior. Neither of these studies included injection-moulded samples and it is expected that they will show significant differences in residual stress distribution as well as other properties. Furthermore, the materials of most interest are those with fibre reinforcement, another departure from the investigations by Miyano *et al.* [29] and Srivastava and White [30].

Differential thermal contraction under the temperature gradients that prevail during solidification is the principal source of residual stresses in thermoplastics. Although the same is indicated to be true in the thermosets studied by Miyano *et al.* [29] and Srivastava and White [30], their samples were cured under fairly benign conditions. In the case of injection moulding, the material is heated rapidly and very non-uniformly. The material is prepared in the barrel at a relatively low temperature, high enough to promote easy flow but too low to cause rapid reaction. When injected, the material that is adjacent to the hot mould wall heats up rapidly and starts to cure. The material in the interior heats up more slowly but because of the exothermic nature of the curing reaction it may rise to above the mould temperature and the temperature gradient in the material may become reversed. At the curing temperatures used in injection moulding, the reaction is rapid and difficult to control. Hot spots may develop. Although the presence of non-reactive fillers should help to control the exotherm, they may even enhance the likelihood of localized temperature variations if matrix–filler segregation occurs or if the filler is in the form of fibres which become oriented, causing anisotropic thermal conduction to occur. Thus the thermoelastic residual stresses that form in reinforced thermosets may not bear any resemblance to those in thermoplastics.

There are other differences between thermoplastics and thermosets that may cause differences in the residual stress distributions in injection mouldings made from the two classes of material. Firstly, the curing

process can be expected to change the intermolecular packing and is generally accompanied by shrinkage; in the case of injection moulding, with curing proceeding at different rates at different depths, this will result in constrained shrinkage in the parts of the moulding that cure last and will result in stress development. Secondly, stress relaxation will be far less likely to occur in a cured thermoset than in a thermoplastic so that the post-moulding relaxation (ageing) of residual stresses [19] will be much less in thermosets. Thirdly, in thermoplastics there are flow-related stresses [18] that are partly related to the orientation and subsequent recoil of the long molecular chains whereas the thermoset molecules are much shorter during the flow phase of the process and are not expected to behave in the same way.

The purpose of the study reported here was to determine the size and distribution of residual stresses in short-fibre-reinforced polyester and phenolic thermosetting injection mouldings and to investigate whether they can be controlled.

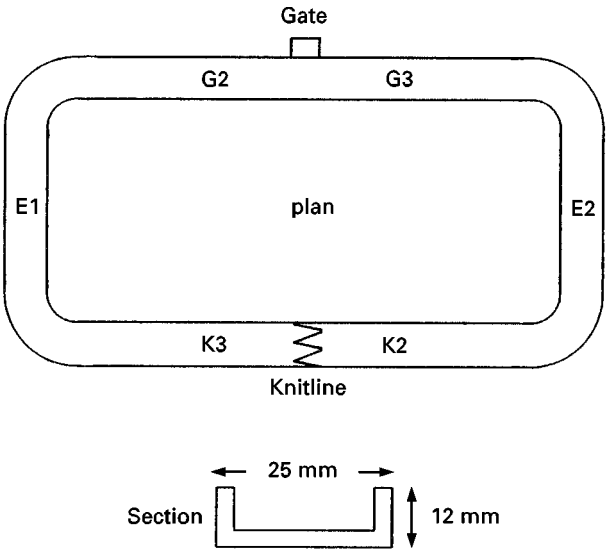

Figure 1 Plan and section of the mouldings (not to scale). The wall thickness was about 3 mm. Bars with axis parallel to the flow direction were cut from positions E1, E2, G2, G3, K2 and K3. In a limited number of experiments bars were cut spanning the gate (G1) or spanning the knitline (K1) but the studies of these samples are not reported here.

2. Experimental procedure

2.1. Materials and moulding conditions

Studies have been made of a reinforced phenolic material, Vynckier RX613, and two experimental polyester dough moulding compounds (code named DMC1 and DMC2). For the Vynckier RX613 phenolic compound, the volume composition was resin 52%, calcium carbonate filler 33% and glass (fibres plus some spheres) 15%. DMC1 was described as a *low-shrink* grade with volumetric composition given by the supplier as 44% resin plus "low profile additive" (LPA), 44% mineral filler and 12% glass fibre. The volumetric composition of DMC2 (*nil shrink*) was given as 53% resin plus LPA, 36% mineral filler and 11% glass fibre. The mouldings were in the form of picture frames with a deep rim (Fig. 1). The mould temperature used for the phenolic mouldings was 158–160 °C, the back pressure was 5 bar and the hold pressure was 25 bar (for 15 s) or 45 bar (for 5 s). Some of the phenolic mouldings were post-cured in an air circulating oven at 170 °C for 7 h. The mould temperature for the DMC materials was 145 °C, the back pressure was 27 bar and the hold pressure was either 96 bar (for 10 s) or 192 bar (for 19.8 s). Test bars 10 mm wide, 3 mm thick and 90 mm or 110 mm long were extracted from (i) the gate side, from 10 mm downstream of the gate and not spanning the gate region (identified below and in Fig. 1 as G samples), (ii) the knitline side (not spanning the knitline itself) (K samples) and (iii) the short connecting ends (E samples).

2.2. Residual stress measurement

The residual stress distribution was measured using the layer removal procedure [10–13]. This method requires that thin layers (about 0.1 mm thick) are removed from one surface of the bar-shaped sample, causing it to bend in response to the resulting imbalance of the residual stresses. The curvature is measured at the beginning and after each layer removal; then a plot is made of the curvature, ρ, versus the total depth $z_0 - z_1$, of material removed, where $2z_0$ is the initial bar thickness and z_1 is the distance from the machined surface to the plane that was located at the bar centre before layer removals commenced. Data taken from this plot are then used to determine the through-thickness residual stress distribution in the x (bar axis) direction using the equation provided by Treuting and Read [31]:

$$\sigma_{i,x} = -\frac{E}{6(1-\nu^2)}\left[(z_0 - z_1)^2\left(\frac{d\rho_x(z_1)}{dz_1} + \frac{\nu d\rho_y(z_1)}{dz_1}\right) + 4(z_0 + z_1)[\rho_x(z_1) + \nu\rho_y(z_1)] - 2\int_{z_1}^{z_0}[\rho_x(z) + \nu\rho_y(z)]\,dz\right] \quad (1)$$

where ρ_x and ρ_y are the curvatures in the x and y directions, respectively, E is Young's modulus and ν is Poisson's ratio. It is difficult to measure the curvature, ρ_y, across the bar width and the assumption was made that the stresses in the plane of the mouldings were equi-biaxial so that $\rho_x = \rho_y = \rho$, giving

$$\sigma_{i,x}(z_1) = -\frac{E}{6(1-\nu)}\left((z_0 + z_1)^2\frac{d\rho}{dz_1} + 4(z_0 + z_1)\rho - 2\int_{z_1}^{z_0}\rho\,dz\right) \quad (2)$$

Layers were removed by high-speed milling using a single-point cutter and fly-cutting action. Whereas this has proved to be most satisfactory in many studies of residual stresses in thermoplastics, the tool tip became damaged much more rapidly with the reinforced thermosets than with thermoplastics or even reinforced thermoplastics. As a result the tool was changed much more frequently than was the case with

the analysis of thermoplastic bars. The bar was fastened to the milling machine bed using a vacuum chuck, permitting swift stress-free removal of the sample at the end of a machining pass. The plots of curvature versus depth removed were found to have much more scatter than is the case with thermoplastics samples [10–13, 19, 26], an observation which is discussed later. Improvements were made by increasing the frequency of milling tool change but the scatter was never reduced to the level routinely achieved with thermoplastics mouldings.

The curvature was measured using the optical lever principle with a laser as light source [10, 32]. A profilometer was also used in some cases for comparison. The thermosets tested here have greater stiffness than the thermoplastics samples examined in previous studies, and the chance that the force applied by the profilometer to the sample bar causes an error is much less with the reinforced thermosets. Nevertheless the method proved to be less satisfactory than the optical lever method for practical reasons [8].

The Treuting–Read expressions (Equations 1 and 2 are based on an analysis for samples with uniform Young's modulus. In another phase of the investigation the distribution of Young's modulus through the depth of the mouldings was determined [7, 8] and was found to vary quite significantly. The measurements provided the data required to perform a refined residual stress analysis introduced by Paterson and White [33] in which the variation in Young's modulus is taken into account. This analysis is very laborious and was performed only for a representative selection of samples.

3. Results

The scatter in the curvature measurements was unacceptably large in a significant fraction of the analyses conducted for this investigation. In some cases it was possible to attribute this to the low level of stress in the sample, resulting in very small curvatures and, consequently, a large fractional measurement error. This was not the only problem and large scatter was often found even in samples which produced significant curvatures when layers were removed. The results presented below are selected to illustrate these difficulties and to provide a representative sample of those analyses which seemed most reliable because of their relative freedom from scatter

3.1. Phenolic composites

3.1.1. Standard mouldings (no post-cure)

Fig. 2 shows the curvature measurements for a sample extracted from the knitline side (K2) of a standard moulding (made using a hold pressure of 25 bar for 15 s). The scatter is large and it is not possible to construct with confidence a suitable line upon which to base a Treuting–Read analysis. This is an example where it can be deduced that the residual stresses are small. To demonstrate this an analysis is carried out using the best-fit straight line (shown in Fig. 2). When the curvature plot is represented by a straight line, the

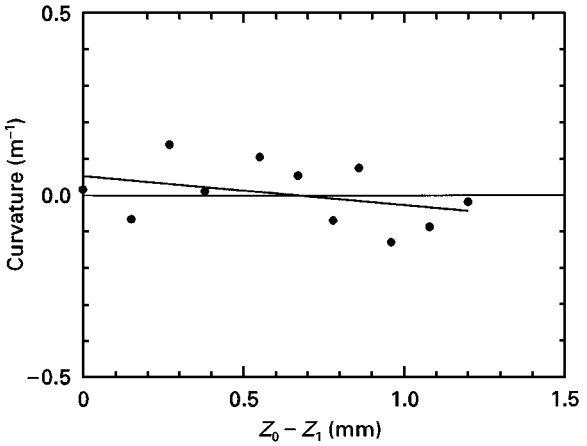

Figure 2 Curvature versus depth removed for a sample extracted at position K2 from a standard phenolic moulding. The least-squares best-fit straight line is shown.

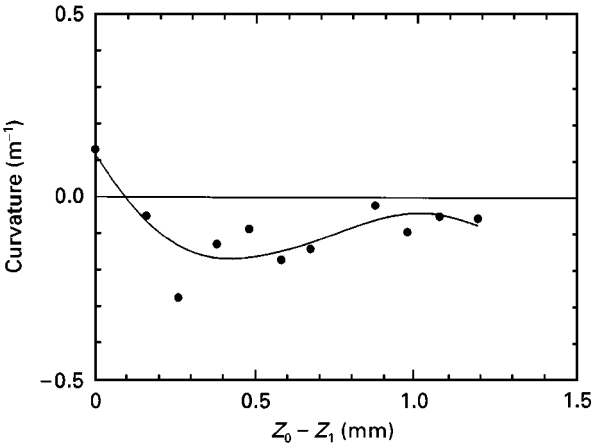

Figure 3 Curvature versus depth removed for a sample extracted at position E2 from the phenolic moulding that also provided the sample used for Fig. 2. The line represents a cubic fit and is included simply for guidance.

Treuting–Read analysis gives a parabolic residual stress distribution with the maximum or minimum at the centre of the moulding and with stress magnitudes everywhere proportional to the gradient of the curvature plot line [11, 12]. In the example given in Fig. 2 the gradient is negative and this corresponds to tensile stress at the surface and a compressive minimum at the centre. The gradient is very small, and the stress magnitudes are correspondingly small: 1.7 $MN\,m^{-2}$ (tensile) at the surface and 0.85 $MN\,m^{-2}$ (compressive) at the moulding centre. It is rare to find a tensile stress near the surface of an as-moulded thermoplastic injection moulding but the observation of a tensile stress at the surface of a reinforced phenolic moulding was not unique, as shown by the data obtained with a sample taken from one of the connecting ends (E2) of the same moulding (Fig. 3). It is quite clear that the gradient of the curvature plot is negative near the surface ($z_0 - z_1 = 0$) and this means that the residual stress at the surface is tensile. A full analysis is not given here because of the lack of precision in locating the best line to represent the curvature plot in the presence of the scatter. A sample extracted from the gate side (G3) of the same moulding gave a curvature

plot with gradients of the opposite sign, indicating that compressive stress was present near the surface (Fig. 4).

Samples made with a larger hold pressure (45 bar) but shorter hold time (5 s) were found to have stresses of the same sense but lower magnitudes in the K2 and G2 positions but in the E2 position there were compressive stresses near the surface (compared with tensile stresses in the standard moulding) [8].

3.1.2. Post-cured samples

Post-cured samples tended to contain larger stresses and always had compressive stresses near the surface and tensile stresses in the interior. Thus the post-curing did not relax the residual stresses but actively promoted a change.

Fig. 5 is the curvature plot for a connecting end (E1) bar extracted from a post-cured standard phenolic moulding. Although there is some scatter after $z_0 - z_1 \approx 0.5$, the early trend is easily located and shows that the stress near the surface is compressive and substantial (Fig. 6). Fig. 7 shows the curvature plot for the G2 sample from the same moulding. The best-fit straight line was used as the basis of the residual stress analysis (Fig. 8). The K2 sample also contained significant residual stresses, raising to about 10 MN m^{-2} compressive near the surface and 7 MN m^{-2} tensile near the centre [8].

After the post-cure treatment the samples moulded with a larger hold pressure (45 bar) but shorter hold time (5 s) were also found to have compressive stresses near the surface and tensile stresses near the centre. As with the post-cured standard mouldings, the curvature plots were steeper than for the as-moulded state, giving larger stress magnitudes. Fig. 9 shows the curvature plot for a G3 sample and, although there is some doubt regarding the exact location of the best line to represent the data, the analysis indicates strong compressive residual stresses near the surface and correspondingly strong tensile stresses near the centre (Fig. 10). In addition to the residual stress distribution obtained using the Treuting–Read analysis, Fig. 10 also gives the results of the Paterson–White analysis that takes into account variations in Young's modulus with depth, based on the same curvature plot (Fig. 9). The Paterson–White analysis shows the same general features as the Treuting–Read analysis, confirming that the use of the simpler Treuting–Read method is justifiable in the studies described here because of the

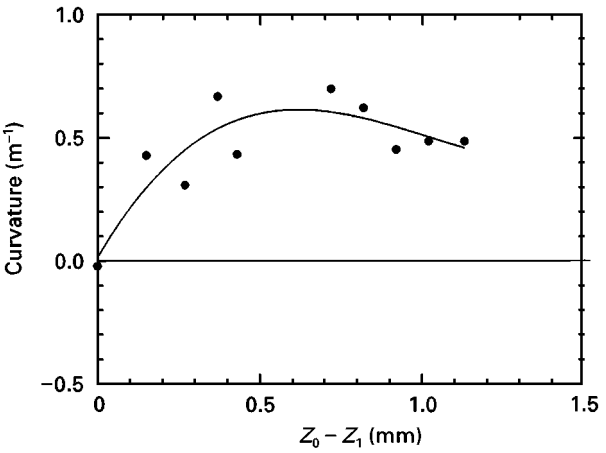

Figure 4 Curvature versus depth removed for a sample extracted at position G3 from the phenolic moulding that also provided the sample used for Fig. 2.

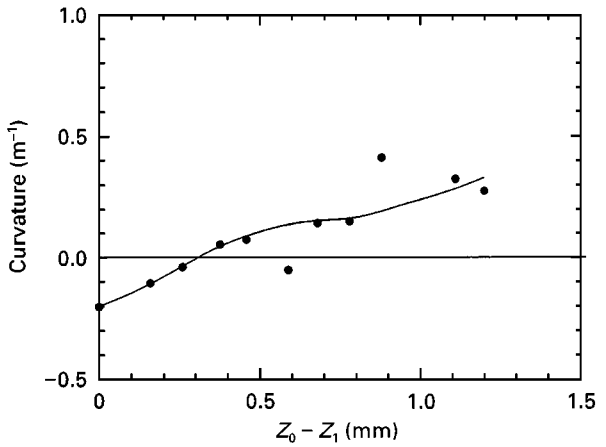

Figure 5 Curvature versus depth removed for a sample extracted at position E1 from a post-cured phenolic moulding.

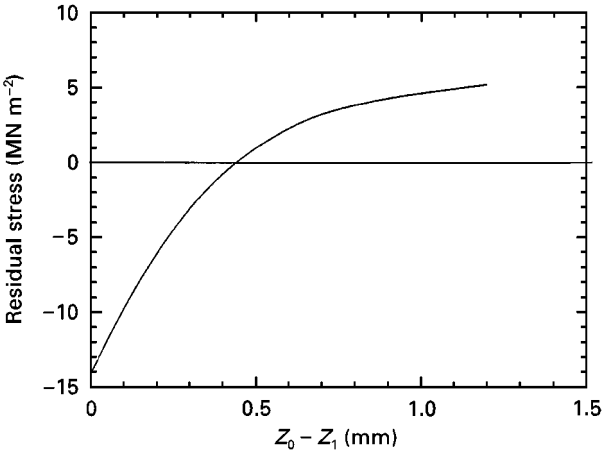

Figure 6 Residual stress distribution derived from the curvature plot shown in Fig. 5.

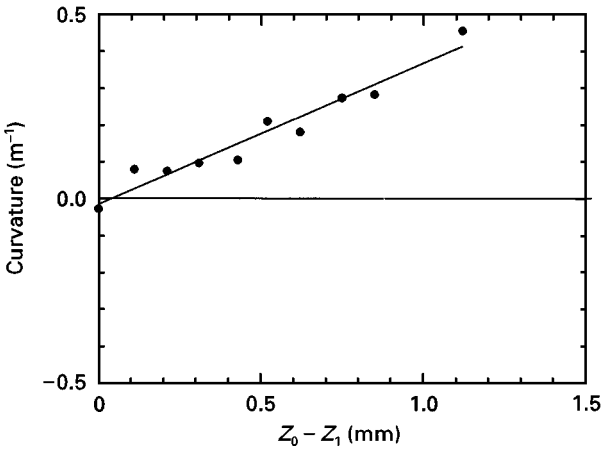

Figure 7 Curvature versus depth removed for a sample extracted at position G2 from a post-cured phenolic moulding. The least-squares best-fit straight line is shown.

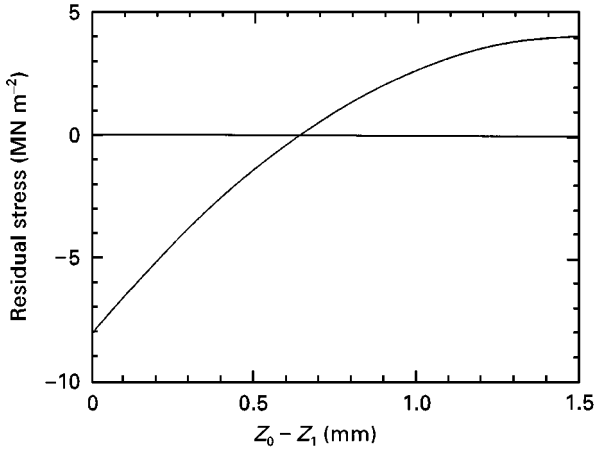

Figure 8 Residual stress distribution derived from the curvature plot shown in Fig. 7.

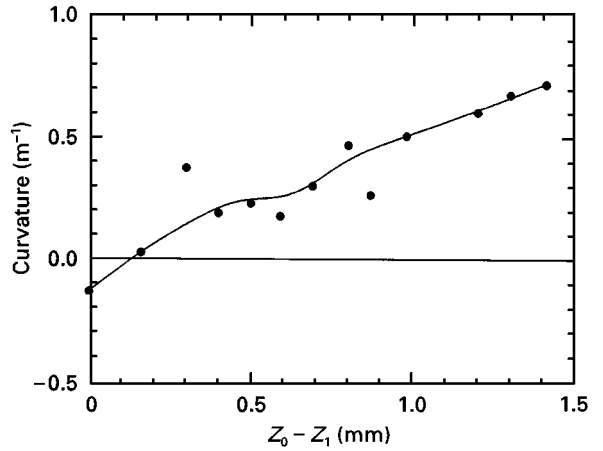

Figure 9 Curvature versus depth removed for a sample extracted at position G3 from a post-cured phenolic moulding that was originally moulded using 45 bar hold pressure for 5 s.

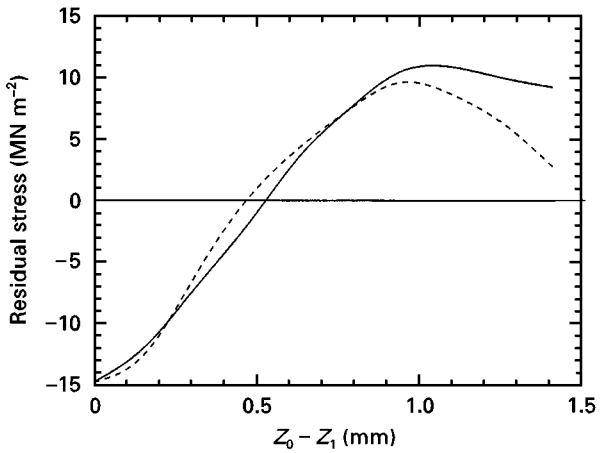

Figure 10 Residual stress distributions derived from the curvature plot shown in Fig. 9 using the Treuting–Read method (——) and the Paterson–White procedure (- - - -), respectively.

large time penalty incurred when using the more exact method due to Paterson and White. This is true even though Young's modulus varied quite considerably in the example given above (Fig. 11). A similar result was obtained with a bar extracted from the K2 position of the same moulding (Figs 12 and 13).

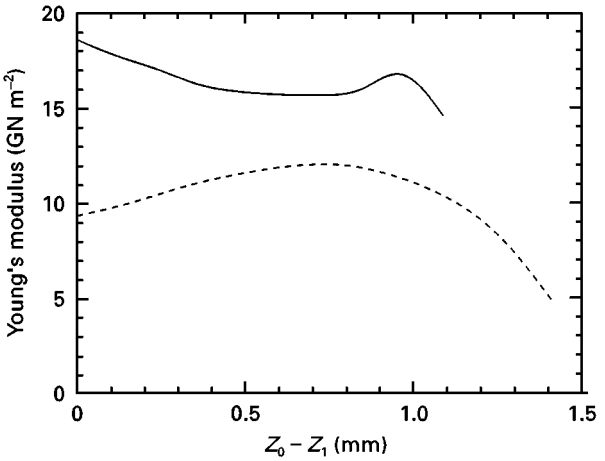

Figure 11 Young's modulus distributions for samples extracted at the K2 (——) and G3 (- - - -) positions from the post-cured phenolic moulding used for Figs 9 and 10.

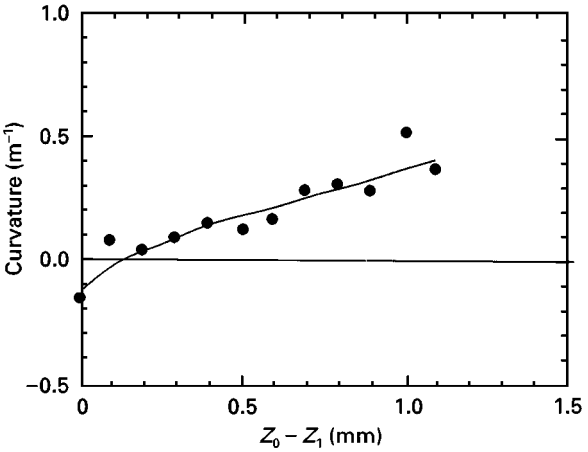

Figure 12 Curvature versus depth removed for a sample extracted at position K2 from the post-cured phenolic moulding used for Figs 9–11.

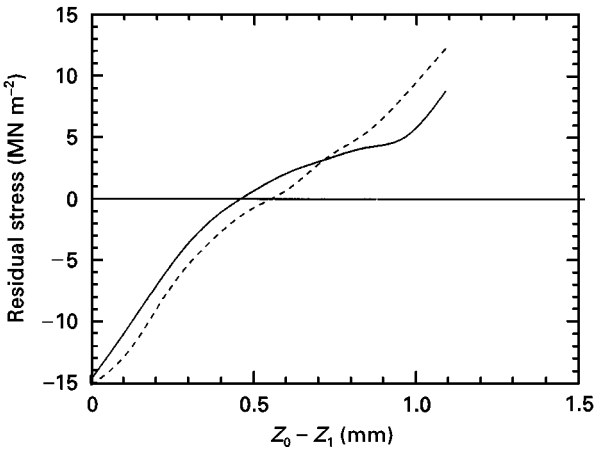

Figure 13 Residual stress distributions derived from the curvature plot shown in Fig. 12 using the Treuting–Read method (——) and the Paterson–White procedure (- - - -), respectively.

3.2. DMC1

The curvature plots for DMC1 were rather shallow. There was a tendency for the curvature to fall near the left-hand axis, indicating tensile stresses near the

surface. An example is shown in Fig. 14 for a sample extracted from the E2 position of a moulding that was made using a high hold pressure (192 bar) and high hold time (19.8 s). The corresponding residual stress distribution is shown in Fig. 15. The K2 sample from the same moulding also appeared to have a (weak) tensile stress near the surface [8] but the curvature plot for the K3 sample showed a positive gradient, indicating compressive stress near the surface (Fig. 16). The best-fit straight line was used for the residual stress analysis of Fig. 16 and the result is shown in Fig. 17. Samples extracted from DMC1 mouldings made with the lower hold pressure (96 bar) and shorter hold time (10 s) tended to produce curvature plots with greater scatter; the curvatures were generally quite small and there were indications that the residual stresses were tensile near the surface in some samples and compressive in others [8].

3.3. DMC2

The curvatures obtained when layers were removed from samples made from DMC2 were generally small, indicating fairly small residual stress magnitudes. An example is shown in Fig. 18 in which the best-fit straight line is chosen for analysis in the presence

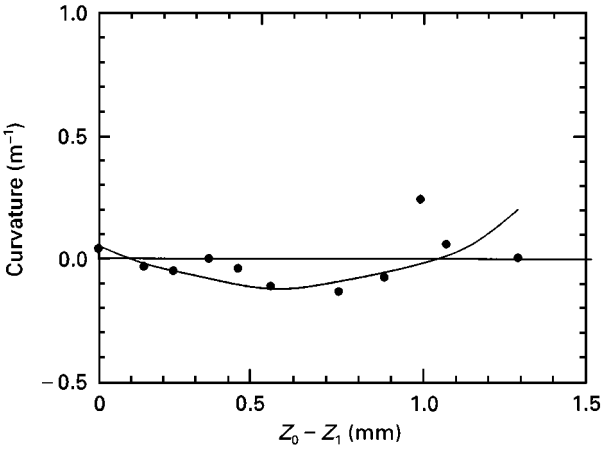

Figure 14 Curvature versus depth removed for a sample extracted at position E2 from a DMC1 moulding.

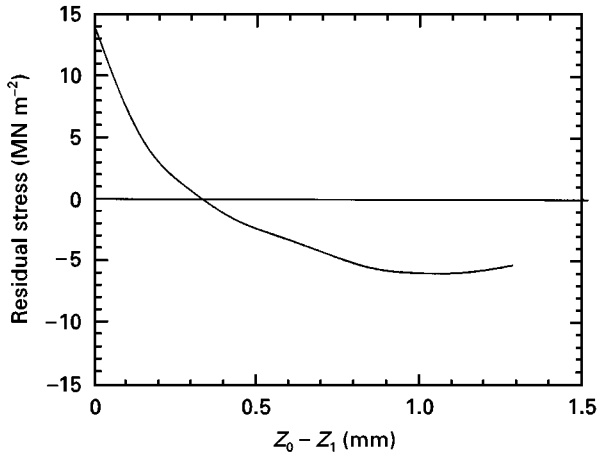

Figure 15 Residual stress distribution derived from the curvature plot shown in Fig. 14.

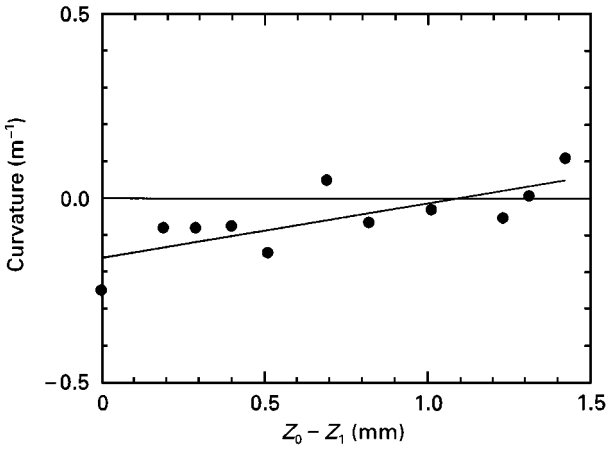

Figure 16 Curvature versus depth removed for a sample extracted at position K3 from a DMC1 moulding. The least-squares best-fit straight line is shown.

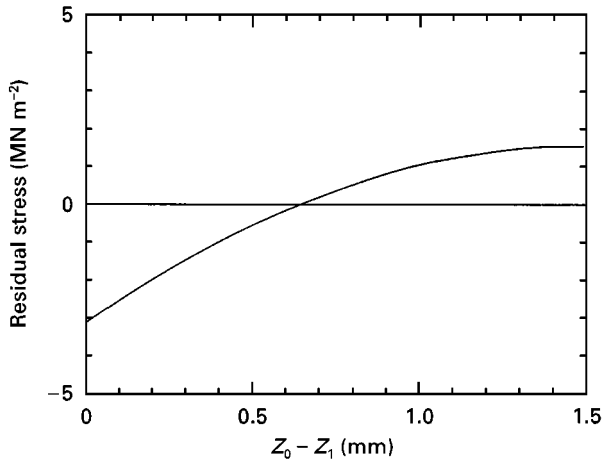

Figure 17 Residual stress distribution derived from the straight line in the curvature plot shown in Fig. 16.

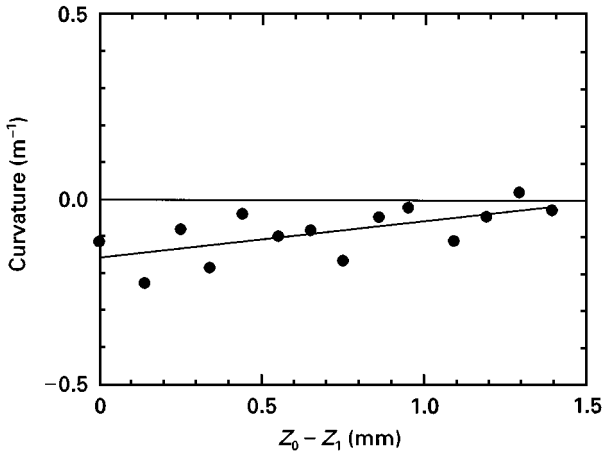

Figure 18 Curvature versus depth removed for a sample extracted at position G3 from a DMC2 moulding. The least-squares best-fit straight line is shown.

of significant scatter. The gradient is small and positive and corresponds to a parabolic stress distribution with a maximum compressive stress of about 1.5 MN m^{-2} at the surface and a maximum tensile stress of less than 0.8 MN m^{-2} at the centre.

4. Discussion

We believe that this is the first study in which significant attention has been paid to the determination of residual stresses in short-fibre-reinforced thermosets. The testpieces were extracted from injection mouldings that were made in batches using controlled conditions and taking all reasonable precautions to produce consistent parts. It is shown that the stress levels were often significant, with magnitudes of around 15 $MN\,m^{-2}$ common. In many cases the scatter in the curvature data was so large that no sensible line could be chosen to represent them and no residual stress analysis could be performed. Nevertheless many satisfactory analyses were obtained that gave considerable insight into the nature of the residual stresses present in these mouldings.

The residual stresses varied in an unpredictable manner and repeat measurements were rarely in close agreement. Residual stress distributions were usually different at different locations within the same moulding and were usually different at equivalent locations in different mouldings from the same batch. Examples were found in which the senses of the stress distribution were different at different locations within the same moulding, with compressive stresses at the surface at some locations and tensile stresses at the surface at other locations. Compressive stresses form at the surface of thermoplastic mouldings as the result of the temperature gradient that is present during solidification. The opposite temperature gradient is present at the beginning of the moulding cycle with injection-moulded thermosets because warm material is injected into a hotter mould. If this temperature gradient were to be maintained during curing, then residual stresses of opposite sense to those in thermoplastic mouldings would be expected to form when a uniform temperature is finally re-established. The curing process is exothermic, however, and, if the reaction is rapid enough, the temperature at positions remote from the stabilizing influence of the mould wall may rise above that of the mould, so reversing the temperature gradient. If this situation prevails during the curing process, then the residual stress is predicted to be compressive near the surface and tensile in the interior when the part has cooled to a uniform temperature. The exotherm is difficult to control and may produce different effects at different locations. In regions with high fibre concentration the effect will be quelled somewhat by the heat absorption of the non-reacting filler. If the fibres are aligned, the thermal conductivity will be anisotropic and this may lead to further differences if the fibre orientation distribution is non-uniform throughout the moulding. Light optical examination of sections of these mouldings confirmed the presence of fibre bunching and of non-uniform fibre orientation distribution [7, 8].

The majority of analyses performed on as-moulded phenolic samples showed that the residual stress was compressive near the surface and tensile in the interior. There was a significant number of examples in which the opposite sense was obtained, however, and it is evident that control of residual stress is extremely difficult with this type of material. In the examples given above the use of a higher hold pressure for a shorter time appeared to produce smaller residual stress magnitudes, but the apparent change is much smaller than the range of values obtained from samples from a single batch of mouldings and cannot be taken as a reliable guide to a means of control.

The post-cured phenolic mouldings appeared to have a more consistent residual stress distribution, with compressive stresses near the surface and tensile stresses in the interior. The stress magnitudes were generally higher than those measured in as-moulded samples, indicating that some active change was promoted by the post-cure. It might have been expected that post-curing would cause stress relaxation but instead it seems that the effect of the completion of the chemical reactions involved in curing dominated. The changes in stress distribution are consistent with shrinkage in the interior and it is speculated that the state of cure was less developed there at the end of the original cure so that, during the post-cure treatment, more shrinkage took place in the interior than near the surface, producing or enhancing tensile stresses there and causing the setting up of opposing (compressive) stresses near the surface.

The residual stress magnitudes measured in the DMC samples were generally smaller than those in the phenolic mouldings, with the nil-shrink grade (DMC2) tending to have smaller stresses than the low-shrink grade (DMC1). In all cases there was a wide spread of behaviour and the inconsistency is the most important characteristic. It was found that tensile residual stresses and compressive residual stresses were found near the surface in the DMC mouldings with almost equal probability. Thus there was more tendency to have tensile residual stresses near the surface with DMC samples than with phenolic mouldings.

Although the results presented here are the most positive confirmation yet published that residual stresses can form in injection-moulded reinforced thermosets, this should be no surprise. Mouldings made from these materials are known to be prone to warping and are frequently placed in jigs immediately after ejection from the mould to ensure that they retain the target dimensions as they complete curing. The importance of this phase of the forming process is confirmed in the study of post-curing of phenolics in which it was demonstrated that the residual stresses change markedly after ejection from the mould. If the changes are unbalanced, this will lead to warping and distortion. The current study did not examine imbalances in residual stresses across the section; this would be almost impossible to achieve since the preferred method would be to conduct layer removal measurements from opposite sides of two nominally identical samples [11, 12, 34–36]. In the presence of the inconsistency between samples tested in this study it would not be possible to select identical samples for such an investigation.

5. Conclusions

Significant residual stresses may form in injection mouldings made from fibre-reinforced thermosets.

Measurements of the order of 15 $MN\,m^{-2}$ were common; this is a significant fraction of the tensile breaking stress for this class of material. The magnitude of the stresses and even their sense varied unpredictably from moulding to moulding and even from one location to another within the same moulding. This is believed to be at least in part due to fibre bunching and variations in fibre orientation distribution, possibly through the effect on the temperature rise associated with the curing exotherm. Modifications in the residual stress distribution are promoted by post-curing in phenolic mouldings; stress magnitudes generally increased and the variability reduced. The effect of changes in processing conditions on residual stresses was difficult to determine in the presence of such large variations when no changes in processing conditions were applied.

Acknowledgements

S.B.W. held a Science and Engineering Research Council CASE scholarship. The studies formed part of a programme coordinated by the National Physical Laboratory Working Group on Injection Moulding of Thermosetting Plastics and we are particularly indebted to K. Thomas for discussion and advice.

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Received 26 February 1997
and accepted 18 March 1998