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Computer modelling of the origin of defects in ceramic injection moulding

Part I *Measurement of thermal properties*

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The acquisition of thermal property data for the subsequent computer modelling of solidification-induced defects in the injection moulding of ceramic suspensions is described. Thermal diffusivity of a polystyrene–zirconia suspension was measured between 80 and 160 °C. Volume thermal expansion and specific heat were also recorded as a function of temperature and the equation of state was derived. All the experiments use standard laboratory equipment making the procedure widely applicable. In subsequent work, these data will be used to predict the origin of voids and cracks in ceramic moulded bodies.

1. Introduction

The problems associated with the assembly of fine ceramic particles prior to firing are gradually attracting more serious attention as technical ceramics move closer to mass production. Injection moulding receives considerable attention [1, 2] because of the complexity of shape that can be reproduced. Previous work has identified two major problems which beset the manufacture of large injection-moulded ceramic components: the difficulty of removing the organic vehicle [3] and the problem of cracks or voids appearing at the solidification stage [4, 5]. In the work of which this forms a part, the solidification of simple shapes is modelled in order to predict the influence of material and machine parameters on (a) the incidence of voids and (b) the residual stress distribution and hence the incidence of cracks. During solidification in the cavity, pressure and specific volume of the suspension vary as the temperature decreases and in order to execute this work several material properties of the suspension are required; specific heat, volume thermal expansion, the equation of state and thermal diffusivity.

The heat capacity per unit volume at constant stress of a two-phase composite is given by [6]

$$C = \bar{C} + 9T\left(\frac{\alpha_2 - \alpha_1}{1/K_2 - 1/K_1}\right)^2\left[\frac{1}{K^*} - \overline{\left(\frac{1}{K}\right)}\right] \tag{1}$$

where α is the coefficient of expansion, K the bulk modulus, subscripts 1 and 2 refer to the ceramic and matrix, respectively, K^* the bulk modulus of the composite, $\overline{1/K}$ is the volume average of the reciprocals of bulk modulus and T temperature.

If the matrix bulk modulus is low compared with the bulk modulus of the filler then the specific heat of the composite is given by the law of mixtures [7].

In the present work, using a fine high surface area powder and a diluent in the polymer, the specific heat, C_p was measured as a function of temperature for the suspension.

The cubical thermal expansivity of the polymer can be found from mercury displacement [8] and from this information the equation of state of the composite can be derived. This avoids the need to record volume dilations using high pressure equipment.

Several equations of state for polymers have been derived both empirically [9–13] and from theory [14–17]. For example, Griskey and Whitaker [11] gave the following empirical equation of state for polymers

$$V = \left(\frac{0.0797}{\rho_o^{0.9421}}\right)\left(\frac{P}{10^5}\right)^{n-1}\left(\frac{T}{T_g}\right)^{m+1} R \tag{2a}$$

where P is the pressure in Pa, ρ_o the density at 25 °C and at 1 atmosphere in kg m^{-3}, R the gas constant, T the temperature in K and V specific volume. m and n are functions of pressure such that

$$n = 1.000 - 3 \times 10^{-11} P \tag{2b}$$

$$m = -0.91 - 3 \times 10^{-10} P \tag{2c}$$

in the pressure range 0 to 100 MPa.

Spencer and Gilmore [12, 13] show how the van der Waals equation can be applied to polymers

$$(P + \pi)(V - \omega) = \frac{R}{M}T \tag{3}$$

where M is the molecular weight of the repeat unit and π and ω constants to be determined by experiment. ω can be found by extrapolation of thermal expansion data to 0 K whereupon $V(0) = \omega$. The slope of the volume expansion curve dV/dT at nominally zero pressure then gives a value for π

$$\pi = \frac{R}{M}\left(\frac{\partial V}{\partial T}\right)^{-1}_{P=0} \tag{4}$$

In order to calculate temperature gradients in an injection moulded ceramic body during solidification and cooling, thermal diffusivity of the suspension is needed as a function of temperature from the injection temperature to room temperature. Hands and Horsfall [18] present a method suitable for rubbers and for polymers in the solid and liquid states. In this method two discs of the material are sandwiched between brass plates with thermocouples at the surfaces and at the centre between the samples. The assembly is then heated at a controlled linear rate and the thermal diffusivity at any temperature may be found from

$$\alpha = \frac{l^2(T_1' + 5T_o')}{12(T_1 - T_o)} \tag{5}$$

where l is the thickness of each disc, T_1 the average of the two surface temperatures, T_o the centre temperature and the prime indicates the rate of temperature rise.

The thermal conductivity of a particle-filled composite can be expressed as a function of volume loading of filler by a number of functions [7, 19]. Of particular interest are the upper and lower bounds due to Hashin and Shtrikman [19].

$$k^* = k_2 + \frac{V_1}{[1/(k_1 - k_2)] + V_2/3k_2} \tag{6}$$

$$k^* = k_1 + \frac{V_2}{[1/(k_2 - k_1)] + V_1/3k_1} \tag{7}$$

where k^* is the thermal conductivity of the composite, V the volume fraction and subscripts 1 and 2 refer to the ceramic and polymer, respectively.

2. Experimental details

2.1. Materials

The zirconia used was grade HSY 3.0 manufactured by Daiichi-kigenso. Grade HF555 polystyrene from BP Chemicals and dibutyl phthalate were used as the organic components. The composition prepared is shown in Table I. The exact weight percent of zirconia based on the results of three ashing experiments was 82.4 wt % and the volume percent calculated therefrom was 45.2 vol % assuming the organic species were present in the proportions in which they were weighed.

2.2. Mixing

The procedure has been described previously [20]. The components were first dry blended in a Henschel high speed non-refluxing mixer at 3000 r.p.m. for 2 min. The mixture was then processed by twin screw extrusion using a Betol TS40 co-rotating intermeshing extruder with barrel temperatures 200, 210, 220, 210 °C feed to exit. The extrudate was water cooled, dried and granulated.

TABLE I Composition of zirconia–polystyrene suspension

Material	wt %	vol %
ZrO_2	82.31	45.0
polystyrene	14.75	45.8
dibutyl phthalate	2.95	9.2

2.3. Determination of specific heat

The specific heat of the composite was determined using a Perkin Elmer DSC 2 differential scanning calorimeter with output to a chart recorder and following the method recommended by the manufacturer [21].

The temperature range 20 to 220 °C was explored in four smaller ranges, an empty aluminium pan and lid being placed in each holder in order to obtain horizontal straight baselines within each temperature range. Pans and lids were selected to be similar in weight on each occasion so that no correction for aluminium was necessary. Power as a function of time was recorded for samples of the composite and for a sapphire standard over each range of temperature at a fixed heating rate of 10 K min^{-1}. The specific heat of sapphire was taken from the work of Ginnings and Furukawa [22].

The value of C_p was calculated from

$$C_p = \frac{W_A}{W}\frac{D}{D_A}C_{pA} \tag{8}$$

where W and W_A are the weights of the sample and sapphire standard, respectively, D and D_A are the displacements in power over a fixed temperature interval for the sample and sapphire standard, respectively, and C_{pA} is the specific heat of sapphire.

2.4. Determination of volume thermal expansion

The standard procedure [8] was followed except that the sample bar measured 60 × 10 × 10 mm^3 for ease of fabrication. The edges of the bar were rounded on SiC paper. The sample was annealed *in vacuo* for 5 h at 70 °C and furnace cooled to relieve residual stresses. The weight of the bar and the weight of mercury were recorded. The apparatus was allowed to equilibrate for 5 to 15 min at each temperature, the oil bath being vigorously stirred. The change in height of the mercury meniscus was followed with a vernier travelling microscope.

2.5. Determination of thermal diffusivity

In the method of Hands and Horsfall [18] two discs each 2 mm thick by 48 mm diameter were used for the determination of thermal diffusivity of polymers and rubbers. In the present work, where suspensions have a higher thermal diffusivity, the apparatus was scaled up so that samples 6 mm thick and 180 mm diameter could be employed. In order to scale up the apparatus the following formula was used which was derived by

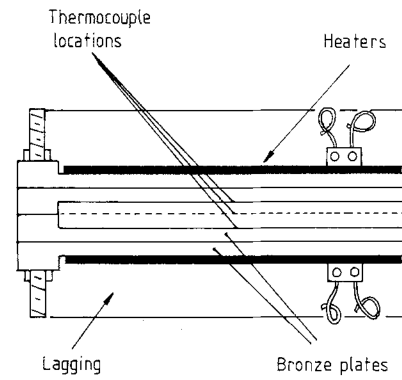

Figure 1 The thermal diffusivity apparatus.

Shoulberg [23] for a similar experiment:

$$\alpha = \frac{kl^2}{2\Delta T} \qquad (9)$$

where k is the rate of temperature rise, α the diffusivity, l the semi-thickness and T the temperature difference across the semi-thickness of the disc. Thus increasing l allows an increase in α to be measured for the same ΔT. Some estimate of the unknown diffusivity is needed for scaling up the apparatus and this can be found from the Hashin and Shtrikman bounds [19].

The discs were incorporated between brass plates which were cast from 5% tin bronze and machined after stress relieving. Heating was achieved using two 1.8 kW electrical resistance heaters controlled by a Eurotherm 818P programmable ramp generator with adaptive tune facility for the PID control parameters. The apparatus is shown in Fig. 1. The discs were individually compression moulded *in situ* at 220 °C and at 13.7 MPa. 36 swg type K thermocouples were used. Those at the outside were set in small grooves in the bronze plates. A heating rate of 15 °C min^{-1} was used for the composite and 12.5 °C min^{-1} was used for the unfilled polystyrene. The thermocouple wire was calibrated against a thermometer with a BSI certificate in the temperature range 25 to 100 °C. The individual thermocouples were not calibrated because a new set of thermocouples was used for each experiment.

3. Results and discussion

3.1. Specific Heat

In order for the procedures used in this ceramic injection moulding study to be applied to diverse suspensions it is important that the material parameters needed for predictive use of the model be easily obtainable using standard laboratory facilities. The measurement of specific heat as a function of temperature by DSC is a simple procedure and the results are shown in Fig. 2. Above the glass transition temperature of 58 °C the suspension can be considered to be comprised of rigid particles in a low modulus matrix and the specific heat is given by the rule of mixtures based on mass fractions. Below the glass transition temperature, the correction term in Equation 1 becomes significant if the inverse of the bulk modulus of the composite deviates from the average of the inverses of bulk modulus of the two phases. For subsequent computing purposes the curve in Fig. 2 was divided by nodes joined by best straight lines. T_g was found from a change in slope of the DSC trace used for measuring specific heat.

3.2. Volume thermal expansion

The change in height of the mercury meniscus X is related to the three volume changes by

$$A\Delta x = \Delta V_s + \Delta V_m - \Delta V_g \qquad (10)$$

where A is the area of the capillary bore, ΔV_s, ΔV_m and ΔV_g are the changes in volume of the sample, mercury and borosilicate glass bulb, respectively.

Equation 10 may be rewritten as

$$A\Delta x = \Delta V_s + \beta_m V_m \Delta T - (V_m + V_s)\rho_g \Delta T \qquad (11)$$

where β_m and β_g are the cubical thermal expansivities of mercury and borosilicate glass, respectively, taken as $18.2 \times 10^{-5}\,K^{-1}$ and $1.0 \times 10^{-5}\,K^{-1}$ [8]. V_m and V_s are the volumes of mercury immersed in the heating bath and the sample, respectively, at the start temperature and ΔT is the temperature change. The test piece, with a density of 3260 kg m^{-3} had a volume of $6.34 \times 10^{-6}\,m^3$ at the start temperature of 18 °C while the mercury occupied $9.65 \times 10^{-6}\,m^3$. The thermal expansion curve from three repeated experiments is shown in Fig. 3.

3.3. Equation of state

Injection moulding operations typically involve maximum injection pressures of 150 MPa and the bulk compressibility, being the reciprocal of bulk modulus, of zirconia, is $0.12 \times 10^{-10}\,Pa^{-1}$ [24] compared with $2.18 \times 10^{-10}\,Pa^{-1}$ for polystyrene [25]. The error in the change of specific volume of the suspension in undergoing a compression from 0 to 150 MPa at 298 K introduced by neglecting the compressibility of the ceramic would be 4.3%. The corresponding change in specific volume of the suspension would be 1.9%.

In order to simplify the calculation and make it generally applicable to ceramic powders, the specific volumes of the two components were treated separately in deriving the equation of state. This has the advantage that once an organic binder system has been characterized, the equation of state for suspensions based upon it, consisting of any ceramic at any volume loading, can be obtained without further experiment provided the influence of the adsorbed layer at the polymer–ceramic interface can be neglected.

From quantitative X-ray diffraction the zirconia has been shown to consist of 88.5 wt % tetragonal and the density at room temperature was calculated to be 5960 kg m^{-3} [20]. The coefficient of linear expansion for tetragonal zirconia is 10.9 to $11.0 \times 10^{-6}\,K^{-1}$ [26] and taking the lower value, using the approximation that cubical expansion coefficient equals three

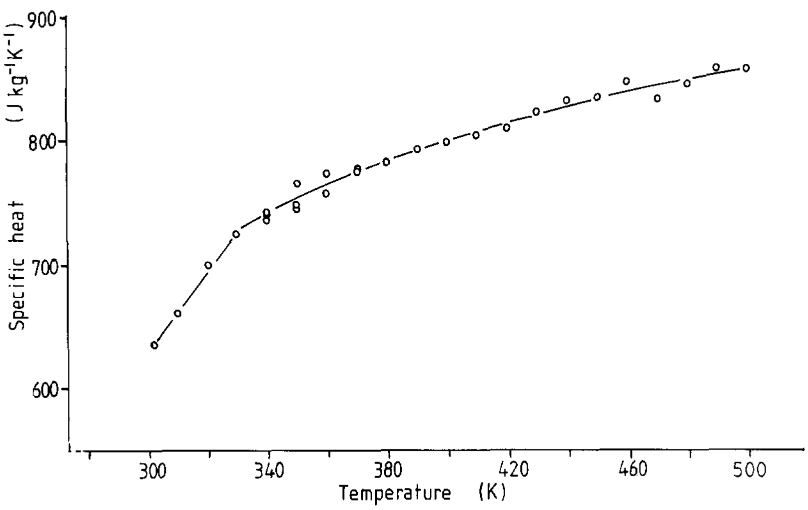

Figure 2 Specific heat of the zirconia injection moulding composition as a function of temperature.

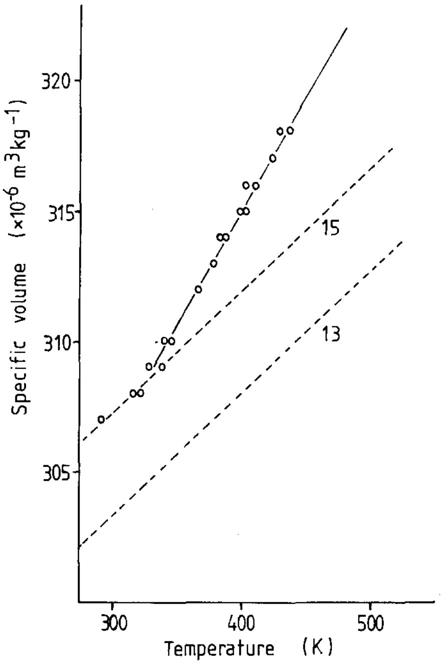

Figure 3 Specific volume of the zirconia injection moulding suspension as a function of temperature: Experimental data points (O), ——— Spencer and Gilmore equation, ----- Griskey and Whitaker Equations 13 and 15.

times the linear coefficient, a value of $32.7 \times 10^{-6}\,\mathrm{K}^{-1}$ was obtained for the zirconia. The specific volume of the ceramic then varies as

$$V_c = \frac{1}{5960}[1 + 32.7 \times 10^{-6}(T - 293)] \quad (12)$$

The density of the polymer was $1050\,\mathrm{kg\,m}^{-3}$ and substituting this value into Griskey and Whitaker's equation of state for the polymer fraction alone, Equation 2 becomes

$$V_p = 9.44 \times 10^{-4}\left(\frac{P}{10^5}\right)^{n-1}\left(\frac{T}{T_g}\right)^{m+1} \quad (13)$$

The specific volume of the composite as a function of temperature and pressure can then be found by combining Equations 12 and 13 by using the rule of mixtures

$$V = m_c V_c + m_p V_p \quad (14)$$

where V represents specific volume, m mass fraction and subscripts c and p represent ceramic and polymer, respectively.

Inspection of Equation 13 shows that when $P = 1$ atmosphere and $T = T_g$ the coefficient should be the specific volume at T_g. This quantity was found experimentally to be $9.65 \times 10^{-3}\,\mathrm{m}^3\,\mathrm{kg}^{-1}$ giving

$$V_p = 9.65 \times 10^{-4}\left(\frac{P}{10^5}\right)^{n-1}\left(\frac{T}{T_g}\right)^{m+1} \quad (15)$$

This modification of Griskey and Whitaker's method was also used to obtain an equation of state for the composite by combining it with Equations 12 and 14.

The data can also be used to obtain the constants in the Spencer and Gilmore modification of the van der Waals Equation 3. The molecular weight of the repeat unit of the polymer is $0.1042\,\mathrm{kg\,mol}^{-1}$ and so $R/M = 79.83\,\mathrm{J\,K}^{-1}\,\mathrm{kg}^{-1}$. The curve for the expansion of the polystyrene–phthalate blend was obtained from the experimental expansion curve for the composite and extrapolated to 0 K to give $\omega = 8.06 \times 10^{-4}\,\mathrm{m}^3\,\mathrm{kg}^{-1}$. The slope of the experimental curve $(\mathrm{d}V/\mathrm{d}T)_{p=0}$ was taken as the slope at 1 atmosphere giving $\pi = 1.68 \times 10^8$ Pa.

The Spencer and Gilmore equation for the polymer–plasticizer blend was obtained as

$$(P + 1.68 \times 10^8)(V_p - 8.06 \times 10^{-4}) = 79.83T \quad (16)$$

The equation of state for the composite can then be obtained by combining Equations 12, 14 and 16.

It is interesting to compare the equation of state for the polystyrene–plasticizer fraction with the corresponding equation obtained by Spencer and Gilmore for polystyrene [12]. They obtained

$$(P + 1.86 \times 10^8)(V - 8.22 \times 10^{-4}) = 79.98T \quad (17)$$

The thermal volume expansivity

$$\beta = \frac{1}{V}\left(\frac{\partial V}{\partial T}\right)_P \quad (18)$$

and the isothermal compressibility

$$\kappa = -\frac{1}{V}\left(\frac{\partial V}{\partial P}\right)_T \quad (19)$$

can be obtained from both equations and compared. For polystyrene of density 1050 kg m^{-3} at room temperature and ambient pressure, $\beta = 4.5 \times 10^{-4}$ K^{-1} and $\kappa = 7.1 \times 10^{-10}$ Pa^{-1} while the experimental values for the polystyrene–plasticizer blend give $\beta = 5.0 \times 10^{-4}$ K^{-1} and $\kappa = 8.7 \times 10^{-10}$ Pa^{-1}. Clearly the larger value of compressibility may be accounted for by the presence of 16.6% dibutyl phthalate based on the polystyrene–phthalate content of the suspension.

Fig. 3 shows the specific volume–temperature data obtained experimentally together with curves based on the combination of Equation 12 with the Spencer and Gilmore Equation 16 and with both variations of the Griskey and Whitaker Equations 13 and 15. The reason that the Spencer and Gilmore equation offers the best fit to the data is simply because the slope and intercept of the experimental data were used to obtain the constants ω and π. The Spencer and Gilmore equation of state was used above T_g but experimental data were obtained down to room temperature. The change in slope of the line at 58 °C corresponds to T_g and thermal expansion in the solid state was obtained from separate dilatometric experiments and will be used for the analysis of residual stresses in Part III.

Figs 4 and 5 show the predicted specific volume for the suspension based on the three procedures at 100 °C and at 200 °C. Without high pressure experiments it is not possible to check the accuracy of these predictions.

The nature of this work places two restraints on the choice of an equation of state. Firstly, since a wide range of organic blends may be used for ceramic injection moulding the equation of state must be easily obtained by experiment at atmospheric pressure. The use of minor binders and plasticizers rules out the use of literature data. Secondly, the equation must be suitable for incorporation in the computer programs developed. It should be possible to calculate any one of the P–V–T parameters from the other two directly. The Griskey and Whitaker equations, for example, involve a pressure term raised to the power of a function of pressure and so can only be solved iteratively for pressure.

The comparison of the equations of state used here highlight the variability of the diverse proposed equations of state for polymers. The Spencer and Gilmore procedure was used throughout subsequent work because it has found general acceptance [27, 28] and because it fits both the above constraints.

3.4. Thermal diffusivity

Fig. 6 shows the thermal diffusivity of the as-received polystyrene as a function of temperature. The data collected below 80 °C were not reported because the rates of temperature rise at the outside and centre of the discs had not become linear. Hands and Horsfall [18] overcame this problem and obtained measurements down to room temperature by storing their apparatus in a refrigerator at −20 °C before use. With the present larger apparatus this was not practicable.

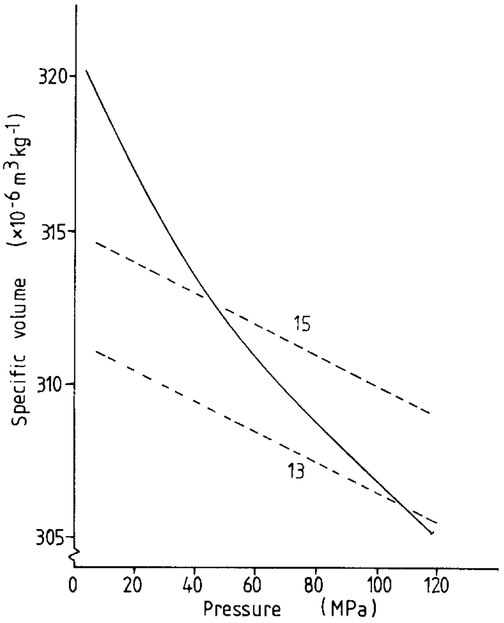

Figure 4 Predicted specific volume as a function of pressure at 100 °C from ——— Spencer and Gilmore and ----- Griskey and Whitaker Equations 13 and 15.

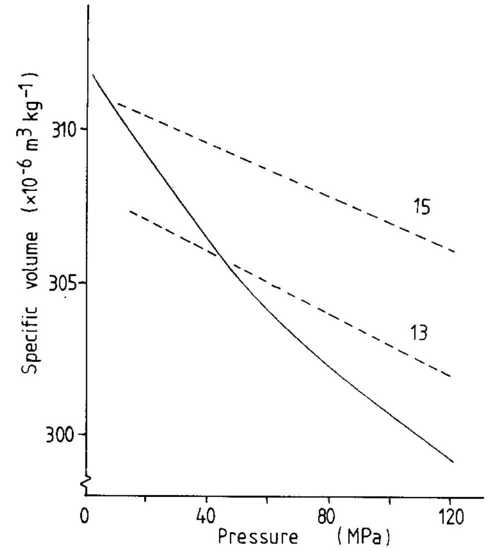

Figure 5 Predicted specific volume as a function of pressure at 200 °C from ——— Spencer and Gilmore and ----- Griskey and Whitaker Equations 13 and 15.

Fig. 6 can be compared with the results of others [23, 29–31] which show variations in both the value and the shape of the curve. This could be explained partly by differences in the properties of the various polystyrene grades, but since the form of the curves varies it is more likely to be due to the experimental difficulties of obtaining precise measurements. The results are similar to those of Ueberreiter [31] but are slightly higher. Ueberreiter narrowed the molecular weight distribution of his sample by fractionation.

For the ceramic suspension (Fig. 7) the lower glass transition temperature of 58 °C effected by the dibutyl phthalate content means that the apparent transition in thermal diffusivity noted for the unfilled polystyrene at T_g is displaced and since data below 80 °C were set aside it does not appear in the results. The scatter in the results is comparable to the experimental errors noted for polystyrene. In order to increase ΔT a heating rate of 15 °C min^{-1} was used for the suspension whereas 12.5 °C per minute was used for polystyrene. The value of ΔT was above 10 °C for the composite and above 20 °C for the polystyrene; Hands and Horsfall [18] recommend that this temperature difference should be between 5 and 10 °C. They used nichrome–constantan thermocouples for which the calibration uncertainty is reported to be ± 0.5 °C [32]. The accuracy of 5% claimed for the method is, therefore, optimistic. A difference in temperature between the two outer thermocouples was also observed on some experiments and the data were discarded when this difference exceeded 4 °C. This probably arises from imperfect contact between the bronze plates and the sample and this is one of the problems inherent in the method. Hands and Horsfall [18] used a silicone fluid to eliminate air and improve thermal contact. In the present study the samples were compression moulded *in situ* and clamped by six bolts equally spaced on the flange.

Other sources of error include the conduction of heat along thermocouple wire. This was minimized by employing 0.2 mm diameter wire. The error in positioning of the thermocouple was minimal since the sample half thickness was 6 mm. The error due to lateral heat flow has been calculated to be less than 0.002% [18].

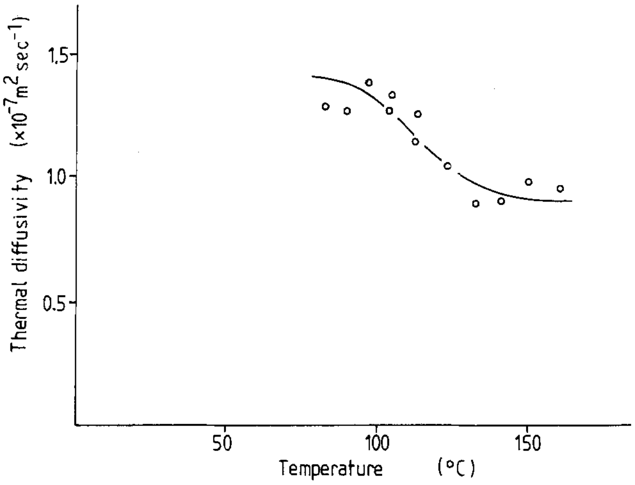

Figure 6 Thermal diffusivity as a function of temperature for the polystyrene.

Equation 5 used for calculating α was derived by assuming a power series variation of temperature with distance from the centre line and a linear variation of temperature with time [18]. These assumptions are obviously not valid for materials in which α varies rapidly with temperature. This is, therefore, a further possible source of error in the transition region of amorphous polymers such as polystyrene and in the melting region of crystalline polymers.

The straight line in Fig. 7 was used for subsequent calculation

$$\alpha = -2.5 \times 10^{-10} T + 3.5 \times 10^{-7} \quad (20)$$

where T is the temperature in K. The position of the thermal conductivity of the composite in respect of the Hashin and Shtrikman upper and lower bounds [19] can only be estimated because precise values of the conductivity of tetragonal zirconia were not available. From Fig. 6 the diffusivity of polystyrene at 150 °C was 0.925×10^{-7} m^2 sec^{-1}. The specific heat of polystyrene at 150 °C was taken to be 2010 J kg^{-1} [33] and the density was 993 kg m^{-3} from the experimentally obtained Spencer and Gilmore equation. A value of 0.185 W m^{-1} K^{-1} was thus obtained for the conductivity of polystyrene. The conductivity of cubic zirconia was taken to be 2.09 W m^{-1} K^{-1} [34] and estimates for the lower and upper bounds for the conductivity of the composite were calculated from Equations 6 and 7 to be 0.48 and 0.88 W m^{-1} K^{-1}, respectively. At 150 °C the measured specific heat of the composite (Fig. 2) was 810 J kg^{-1} K^{-1} and the density (Fig. 3) was 3153 kg m^{-3}. Fig. 7 gives the diffusivity as 2.5×10^{-7} m^2 sec^{-1} at 150 °C and so thermal conductivity is 0.64 W m^{-1} K^{-1} intermediate between the upper and lower bounds.

3.5. The effect of pressure

The computer modelling of the complex process of solidification in the cavity involves a compromise between the widespread applicability of the model as an aid to manufacturing processes and the complexity needed to account for the interplay of parameters.

Thus measurements of the effect of pressure on the thermal properties of the composite were not made in order that the experiments may be easily and quickly

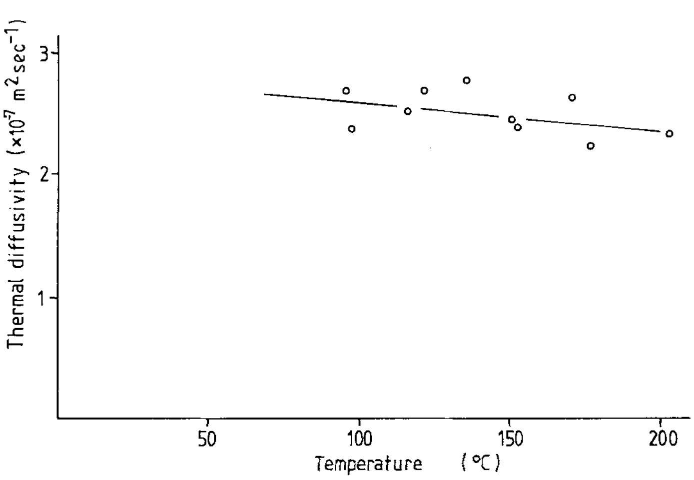

Figure 7 Thermal diffusivity as a function of temperature for the ceramic suspension.

reproduced without the need for specialized high pressure equipment.

Nevertheless, pressure may have a considerable effect on the glass transition temperature of the polymer. For example, the glass transition point of unplasticized polystyrene may increase by as much as 31 °C for an increase in pressure of 100 MPa [35]. The influence that this has on the properties of mouldings will be discussed in subsequent sections.

Few experimental data have been reported in the literature for the effect of pressure on the thermal properties of polymers. A study of the effect of pressure on the specific heat, thermal diffusivity and thermal conductivity of high and low density polyethylene has shown that the effect of pressure on the properties in the range 0 to 100 MPa is small [36]. Any effect that pressure has on the polymer becomes insignificant as far as the properties of the composite are concerned when the influence of the filler is taken into account.

The pressure may, however, also have an effect on the composite insofar as the thermal properties are related to the volume filler loading which itself increases as the polymer is compressed. For the composite under consideration at 220 °C where the differential thermal expansion between polymer and ceramic has reduced the volume fraction of ceramic to 43 vol %, an increase to 45 vol % occurs when the pressure is increased from 0 to 100 MPa. For suspensions of higher volume fraction of ceramic, this may influence thermal conductivity, notably where the conductivity of the filler is high. Clearly, the effect can be obtained by measuring thermal properties at atmospheric pressure and at higher volume loadings. The influence on other properties, notably viscosity, may also be significant but is not dealt with here.

4. Conclusions

Routine laboratory procedures have been adapted to the acquisition of thermal property data essential to the computer modelling of defects introduced in ceramic injection moulded bodies during solidification in the cavity and these procedures can be applied to diverse suspensions. Differential scanning calorimetry permits the measurement of specific heat over the full range of injection moulding temperature. The specific volume of the suspension can be measured by mercury displacement in a borosilicate glass phial. These data can be used to obtain a general equation of state which predicts the specific volume of the suspension over the temperature and pressure ranges experienced in injection moulding. The thermal diffusivity of the suspension can be measured as a function of temperature by adapting an existing procedure to accommodate the higher thermal diffusivity of a ceramic suspension.

Acknowledgements

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