

**NPL REPORT
DEPC-MPR 041**

**The Effect of Pressure on the
Thermal Conductivity of
Polymer Melts**

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JANUARY 2006



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The Effect of Pressure on the Thermal Conductivity of Polymer Melts

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ABSTRACT

The effect of pressures typically experienced in polymer processing on the thermal conductivity of polymer melts, measured using the line source probe technique, is described. The thermal conductivity of both amorphous and semi-crystalline polymers was found to increase with increasing applied pressure, and generally with increasing temperature. However, for HDPE and PP the crystallisation transition, on cooling, was clearly indicated by a step increase in the measured thermal conductivity values. A step decrease in specific volume on cooling, also indicative of the crystallisation transition, occurred at lower temperatures presumably due to differences in the effective cooling rates of the two methods. The measurement of thermal conductivity at near-ambient conditions would result in a lower bound to values for amorphous polymers, whereas a mid-range value would be obtained for semi-crystalline materials that exhibited a step-change in values on crystallisation. This has significant implications for modelling if a single-point value for thermal conductivity is used. Relationships to model the thermal conductivity and specific volume behaviours of amorphous polymers are presented.

Keywords: thermal conductivity, injection moulding, processing, pressure dependence, semi-crystalline polymer, amorphous polymer

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ISSN 1744-0270

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1 INTRODUCTION

There is increasing demand for accurate and relevant data for the behaviour of polymers at elevated pressure for use in design simulations. Previous studies [1] have reported that thermal conductivity is the least accurately measured property of polymers, and yet it is one of the most important material properties for polymer processing. The thermal conductivity of polymers has a major influence on the cooling of mouldings, which is critical in determining cycle times. Since polymers have low thermal conductivities and relatively high specific heat capacities, heat transfer in the polymer is the rate determining part of the process.

Until recently [2] most of the experimental thermal conductivity data in the literature were obtained at atmospheric pressure conditions, far from the actual conditions experienced during polymer processing. The thermal conductivity behaviour of liquids near ambient conditions are well known in comparison with the thermal conductivity behaviour of molten materials encountered in the polymer processing industry.

Improved measurement, prediction and understanding of heat transfer in production processes could lead, for example, to improved processing equipment, materials design and reduced cycle times, so increasing productivity. Scrap rates could also be reduced through, for example, the elimination of hot spots that could cause material degradation, or excessive temperature gradients leading to internal stresses and unacceptable warpage of product. Accurate process simulation is only possible when good quality credible data relevant to industrial processing conditions are made available to software users.

The measurement of thermal conductivity at high pressures (up to 120 MPa) and melt temperatures (up to 250 °C), as carried out in this work, brings thermal conductivity testing into the range of industrially relevant polymer processing conditions.

2 EXPERIMENTAL

2.1 MATERIALS

The polymers studied were commercial grades of glass filled nylon (GFN), polycarbonate (PC), polystyrene (PS), acrylonitrile-butadiene-styrene (ABS), polypropylene (PP), high density polyethylene (HDPE), and polyethylene (terephthalate) (PET).

2.2 METHOD FOR THERMAL CONDUCTIVITY

Thermal conductivity measurements were made using a commercial apparatus, the pvT 100 (SWO Polymertechnik GmbH) with a thermal conductivity cell attached. A diagram of the instrument is shown in Figure 1.

The sample (diameter 9.8 mm, length approximately 50 mm) is contained within a heated chamber, held at the test temperature. The thermal conductivity probe, containing both a heating element and a thermocouple, is inserted into the sample. The heating element and the thermocouple are at a horizontal distance of 0.5 mm apart and

are vertically parallel to each other within the probe. The sample cell is sealed at the top and bottom by PTFE seals. Atmospheric pressure and pressures in the range of 15 MPa to 160 MPa can be applied to the sample at temperatures from 21 °C to 320 °C.

The measurement consists of applying a known voltage to the heater element in the probe for a set time. The thermocouple within the probe records the temperature transient during this process. The thermal conductivity of the material is obtained from a plot of temperature versus $\ln(\text{time})$, which shows three distinct regions, Figure 2.

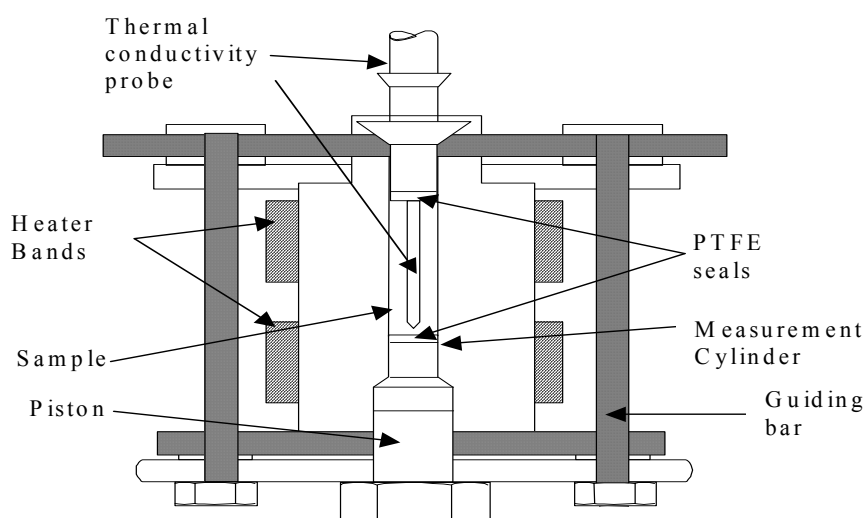


Figure 1 Schematic of the thermal conductivity apparatus

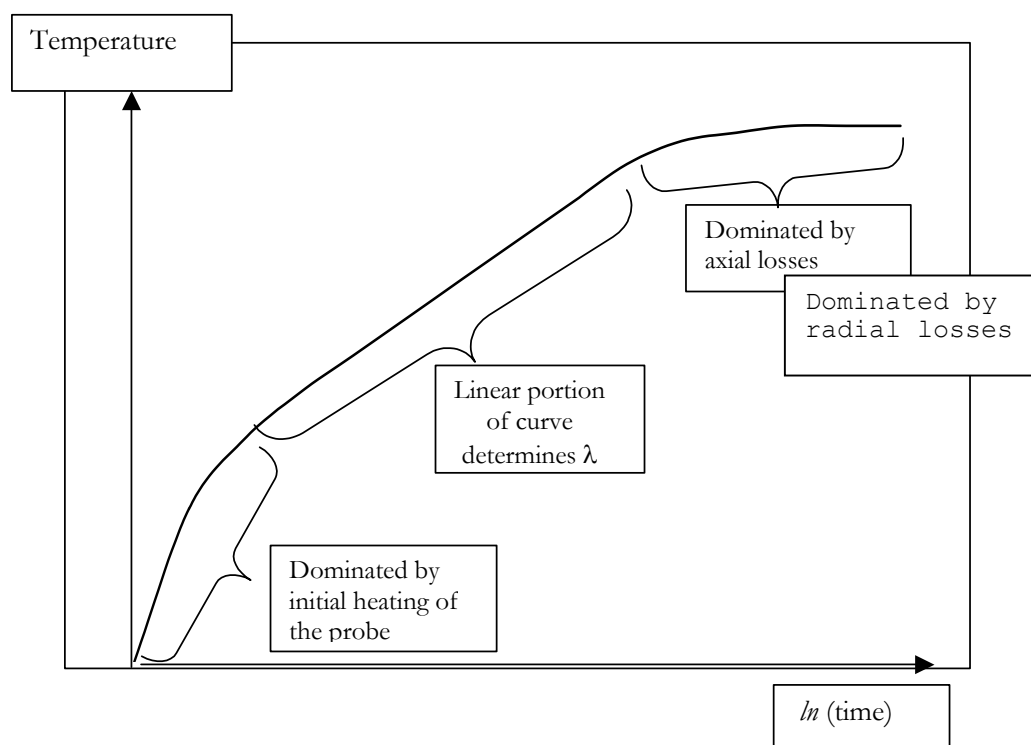


Figure 2 A schematic temperature-time profile for thermal conductivity measurements

At short times the temperature rise is dominated by the initial heating of the probe. At long times the probe temperature is affected by radial heat losses from the sample through the wall of the sample cell. Between these two extremes there is a linear region that is characteristic of the sample's properties.

It can be shown [3] that the temperature rise ΔT from temperature T_1 to T_2 produced within the probe, over the time interval from t_1 to t_2 is given by

$$\Delta T = T_2 - T_1 = \frac{Q}{4\pi\lambda} \ln\left(\frac{t_2}{t_1}\right) \cdot B \quad (1)$$

where Q is the heat energy supplied to the probe, λ is the thermal conductivity of the sample, and B is a correction factor that accounts for deviations from the theoretical model. The correction factor B is determined by calibration of the instrument with a glycerol sample of known thermal conductivity. The gradient of the linear portion of the curve corresponds to $QB/4\pi\lambda$. Hence, a value for thermal conductivity can be obtained for the material providing the applied energy is known.

An analysis of the uncertainty budget showed that the repeatability contribution for a liquid polymer, PDMS, under isobaric conditions at atmospheric pressure was $\pm 1.4\%$ [4]. The repeatability for a semi-crystalline polymer, HDPE, measured at atmospheric pressure over a range of temperatures from the melt state to the solid state was determined to be $\pm 15.6\%$ [4]. This contributes towards a final uncertainty budget value of $\pm 20.7\%$. Other sources of uncertainty that contribute to the uncertainty budget include reproducibility and non-uniformity of heat output.

It has been demonstrated that at higher pressures the repeatability contribution to the uncertainty budget of thermal conductivity measurements for polymers measured under isobaric conditions from the melt state to solid state is likely to be less than the $\pm 15.6\%$ that has been measured at atmospheric pressure. This is considered to be due to better thermal contact between the test probe and the sample due to the elimination, or reduction, of the effect of trapped air at higher pressures. Therefore at higher pressures the final uncertainty budget value would be expected to be less than the $\pm 20.7\%$ value at atmospheric pressure.

2.3 METHOD FOR SPECIFIC VOLUME

The specific volume of the polymers as functions of pressure and temperature were measured using a pressure-volume-temperature (pvT) instrument. The SWO pvT 100 instrument is a direct measurement machine (Figure 3).

The polymer sample of known mass is introduced into the cylindrical test cell, and the top piston inserted. Pressure is applied hydraulically to the sample, via the piston at the top of the cell, which is fitted with a PTFE sealing disc. An identical piston and sealing system is located at the bottom of the cell. After pre-compression, the sample is heated to the initial test temperature and the chosen pressure is applied. The volume is determined from the cell diameter and the piston position that is measured to ± 0.001 mm, the zero position of the piston having been established prior to testing. The

sample is then cooled at the required rate whilst being held at constant pressure and the changing sample length is measured. If the instrument's cooling fluid (air) is passed through liquid nitrogen, cooling rates of up to 250 °C/min can be achieved. Using the final sample mass, the cell diameter and the measured sample lengths, the specific volume at each temperature and pressure can be determined. Once the specific volume vs. temperature curve at a given pressure is obtained, the sample is reheated and the shrinkage behaviour at a different pressure measured.

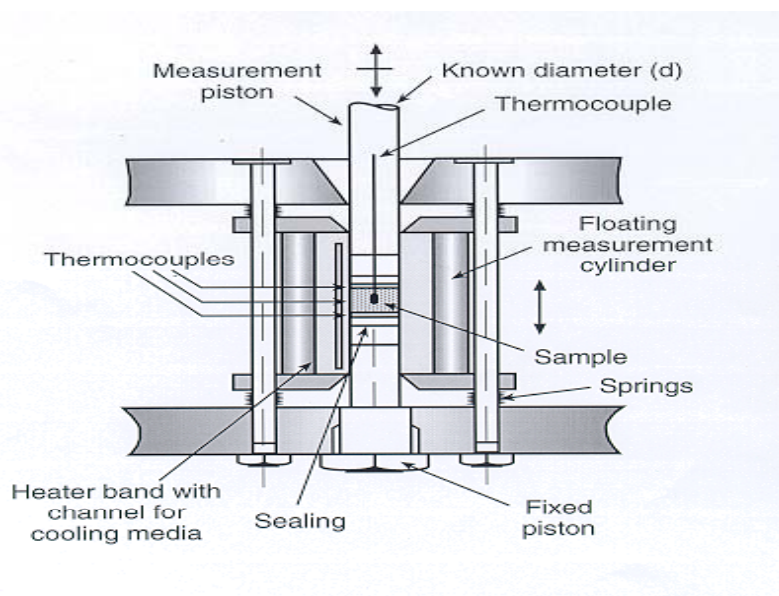


Figure 3 Schematic diagram of pvT apparatus.

The uncertainty in specific volume values for polymers measured isothermally has been estimated to be approximately 0.01 cc/g. This figure does not take into account issues concerning high cooling rate testing for which temperature gradients within the sample will exist. The values of cooling rate quoted are the set rates, corresponding to the temperatures of the barrel wall.

3 RESULTS AND DISCUSSION

Thermal conductivity and specific volume measurements were made at 20 MPa, 80 MPa and 120 MPa pressures over a temperature range of 250 °C to 50 °C.

For each material tested, five separate thermal conductivity readings were taken for each temperature and pressure and the reported value is the average of the five results.

The specific volume behaviour of two polymers, polypropylene, a semi-crystalline polymer, and polystyrene, an amorphous polymer, was measured.

The specific volume and thermal conductivity were then modelled as functions of pressure and temperature using exponential equations of the same form.

3.1 SEMI-CRYSTALLINE POLYMERS

(i) Thermal conductivity

The thermal conductivity behaviour of PP and HDPE at pressures of 20 MPa, 80 MPa and 120 MPa over the temperature range from 250 °C to 50 °C is shown in Figures 4 and 5. Although measurements were performed under isothermal conditions, a decreasing temperature profile was used, i.e. testing was commenced at 250 °C and subsequent measurements made at lower temperatures.

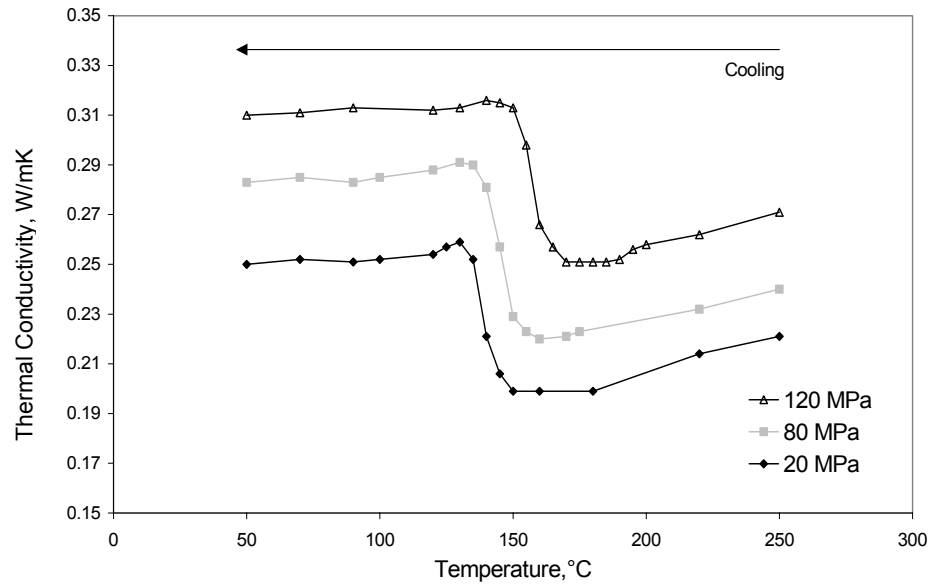


Figure 4 Thermal conductivity of polypropylene from 250 °C to 50 °C at pressures of 20 MPa, 80 MPa and 120 MPa

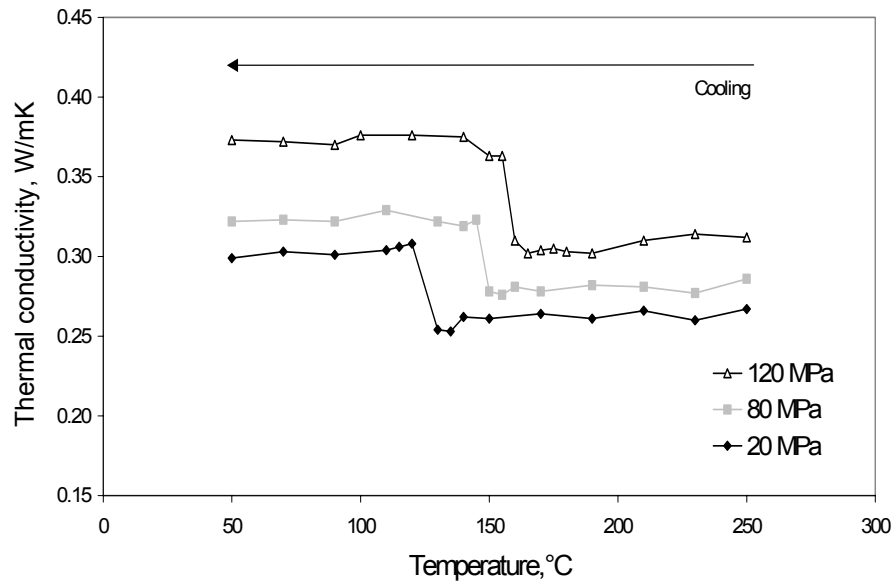


Figure 5 Thermal conductivity of high density polyethylene from 250 °C to 50 °C at pressures of 20 MPa, 80 MPa and 120 MPa

The isobaric ‘cooling’ plots for the polypropylene and high density polyethylene show a typical “Z” shape for each of the pressures indicating a phase transition, apparent from the sharp increase in thermal conductivity on cooling due to crystallisation. Such semi-crystalline polymers have percentage crystallinity values typically greater than 60 % [5, 6].

Increasing the pressure from 20 MPa to 120 MPa increased the onset temperature of crystallisation by approximately 25 °C (Table 1).

Furthermore, the effect of increasing pressure from 20 MPa to 120 MPa increased the thermal conductivity values by up to approximately 0.7 W/(m.K), or 20% (Table 2). The lower change in thermal conductivity values for the glass filled nylon with increase in pressure and the higher overall thermal conductivity values are, presumably, due to the glass fibre content of the material (glass has a thermal conductivity of approximately 1 W/(m.K)).

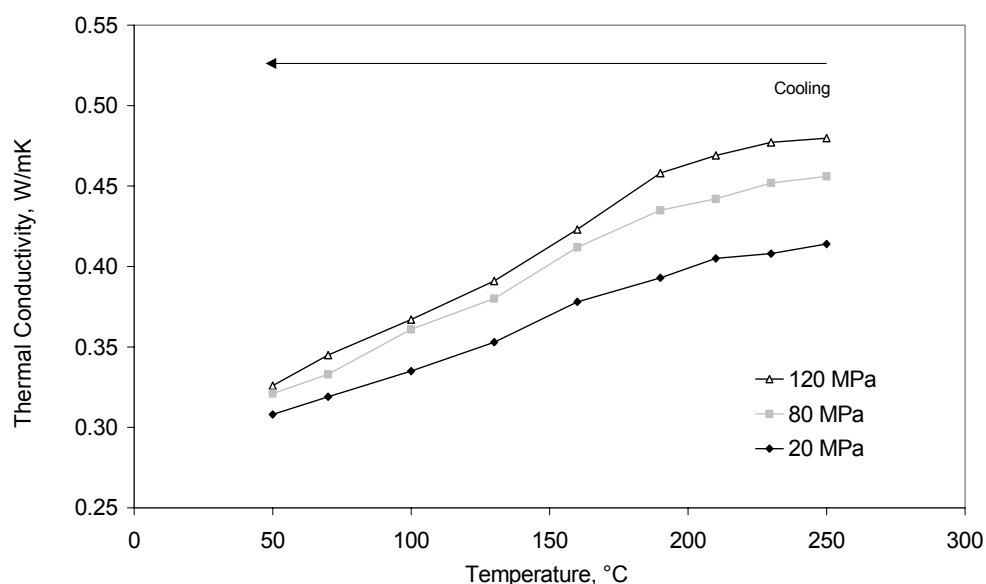


Figure 6 Thermal conductivity of polyethylene (terephthalate) from 250 °C to 50 °C at pressures of 20 MPa, 80 MPa and 120 MPa

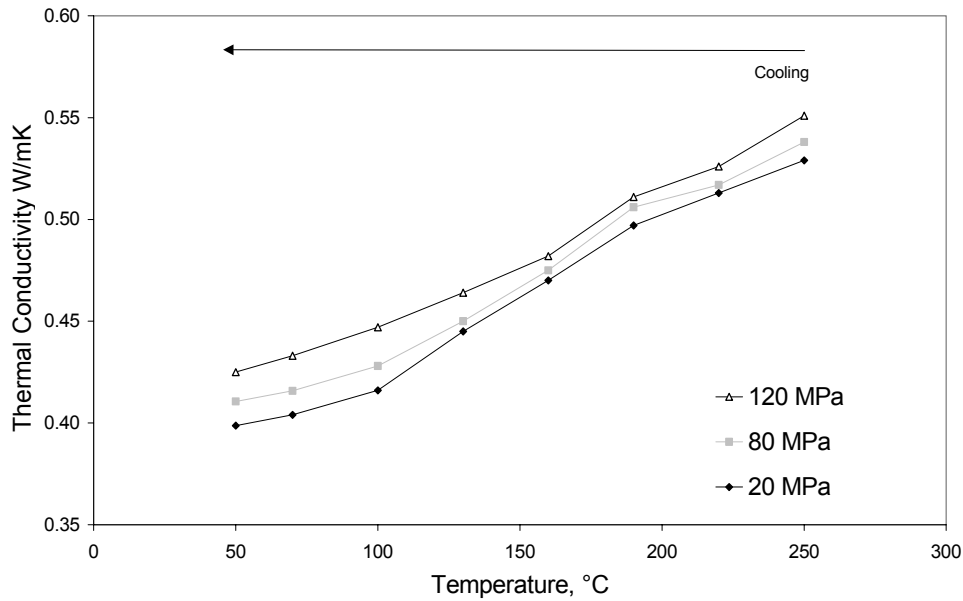


Figure 7 Thermal conductivity of glass filled nylon on cooling from 250 °C to 50 °C at pressures of 20 MPa, 80 MPa and 120 MPa

The thermal conductivity behaviour of a polyethylene (terephthalate) and a glass filled nylon at pressures of 20 MPa, 80 MPa and 120 MPa over the temperature range from 250 °C to 50 °C are shown in Figures 6 and 7.

The isobaric cooling plots for polyethylene (terephthalate) and glass filled nylon show a decrease in thermal conductivity with decreasing temperature at all pressures. For these semi-crystalline materials, which typically have percentage crystallinity values lower than 60 % [7, 8], there was not the distinct “Z” shaped step increase in thermal conductivity that was observed for polyethylene and polypropylene.

(ii) Specific volume

The specific volume behaviour of polypropylene at pressures of 20 MPa, 80 MPa and 120 MPa over a temperature range from 250 °C to 50 °C, in cooling at 5 °C/min, is shown in Figure 8.

The isobaric specific volume cooling plots show a typical “S” shaped step decrease in specific volume, indicating a phase transition region. At the phase transition, on cooling, there is an increase in density of the polymer, which corresponds to a decrease in specific volume.

Increasing the pressure applied to the polymer increased the onset temperature of crystallisation during cooling (Table 1). However, the onset temperatures determined from the thermal conductivity measurements were approximately 20 °C higher than those determined from specific volume measurements. The difference in the observed onset of crystallisation temperature by the thermal conductivity and specific volume

methods is considered to be due to the different, effective cooling rates employed in the measurements.

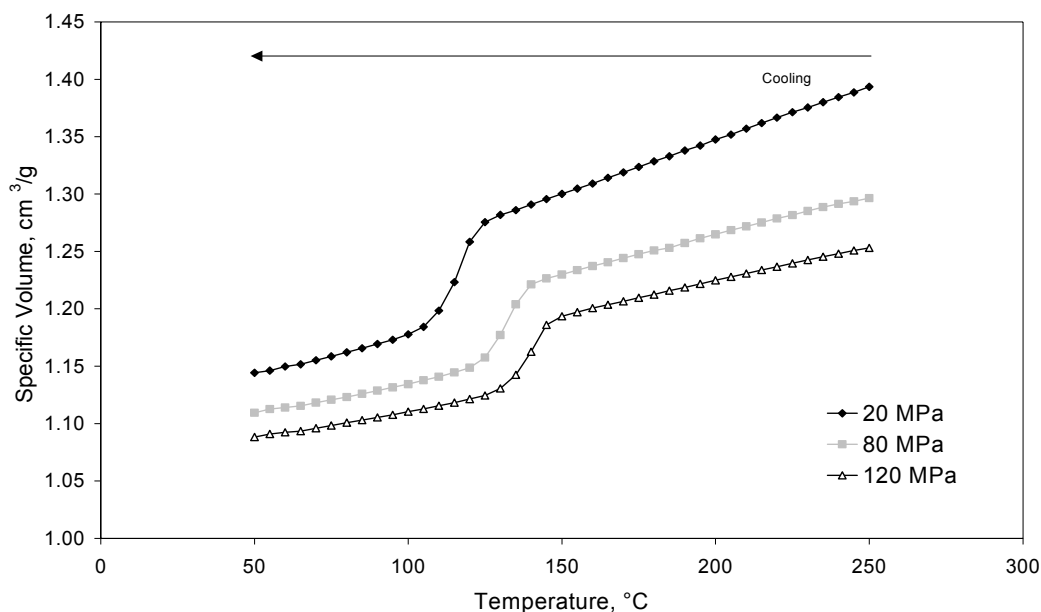


Figure 8 Specific volume of a polypropylene on cooling from 250 °C to 50 °C at pressures of 20 MPa, 80 MPa and 120 MPa

Table 1: Onset of crystallisation temperature at different pressures for polypropylene and polyethylene

Material	Onset of crystallisation temperature, °C, at		
	20 MPa	80 MPa	120 MPa
Polypropylene (onset measured by thermal conductivity)	150	160	170
Polypropylene (onset measured by specific volume)	125	145	150
High Density Polyethylene	130	150	165

3.2 AMORPHOUS POLYMERS

(i) Thermal conductivity

The thermal conductivity behaviour of acrylonitrile-butadiene-styrene, polystyrene, and polycarbonate at pressures of 20 MPa, 80 MPa and 120 MPa over the temperature range from 250 °C to 50 °C are shown in Figures 9 to 11. The isobaric ‘cooling’ plots for acrylonitrile-butadiene-styrene, polystyrene, and polycarbonate show a decrease in thermal conductivity with decreasing temperature at all pressures.

Percentage changes in thermal conductivity values, on increasing pressure from 20 MPa to 80 MPa and 120 MPa, are presented in Table 2 for both the semi-crystalline and amorphous materials. They average 7% for a pressure increase from 20 MPa to 80 MPa, and 13% for a pressure increase from 20 MPa to 120 MPa.

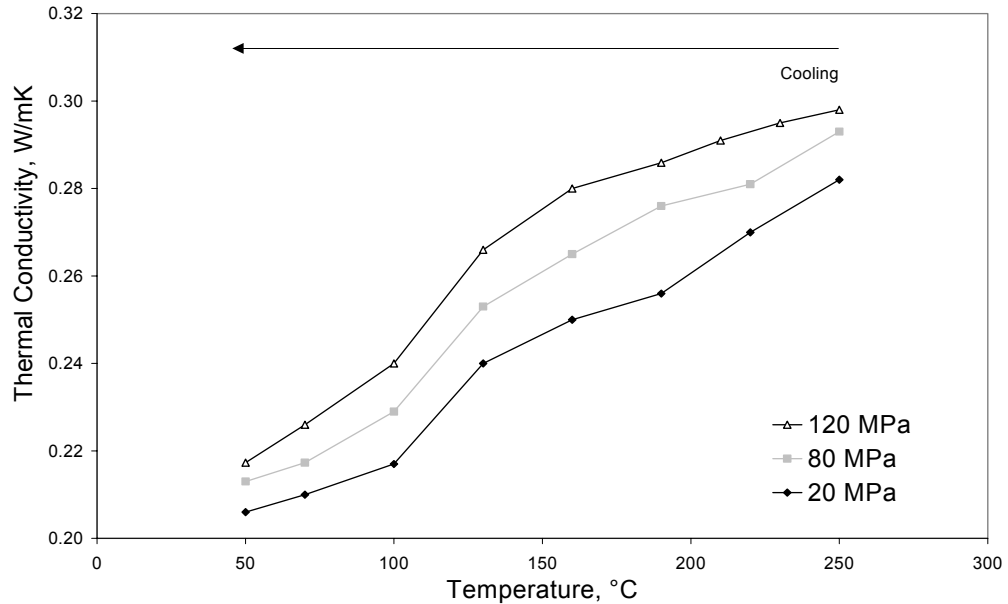


Figure 9 Thermal conductivity of on cooling of acrylonitrile-butadiene-styrene from 250 °C to 50 °C at pressures of 20 MPa, 80 MPa and 120 MPa

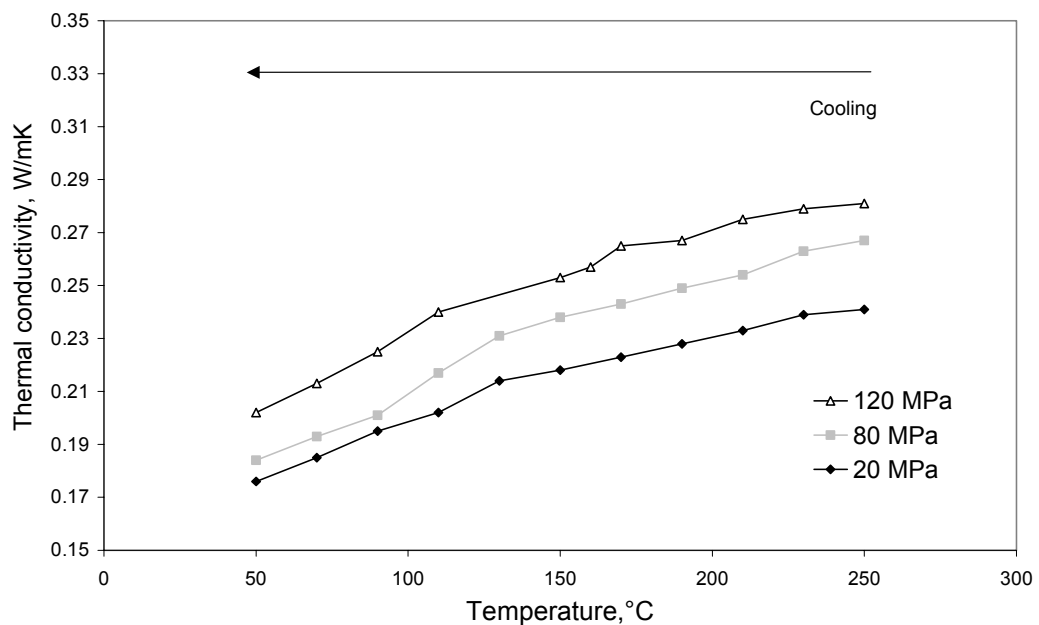


Figure 10 Thermal conductivity of polystyrene on cooling from 250 °C to 50 °C at pressures of 20 MPa, 80 MPa and 120 MPa

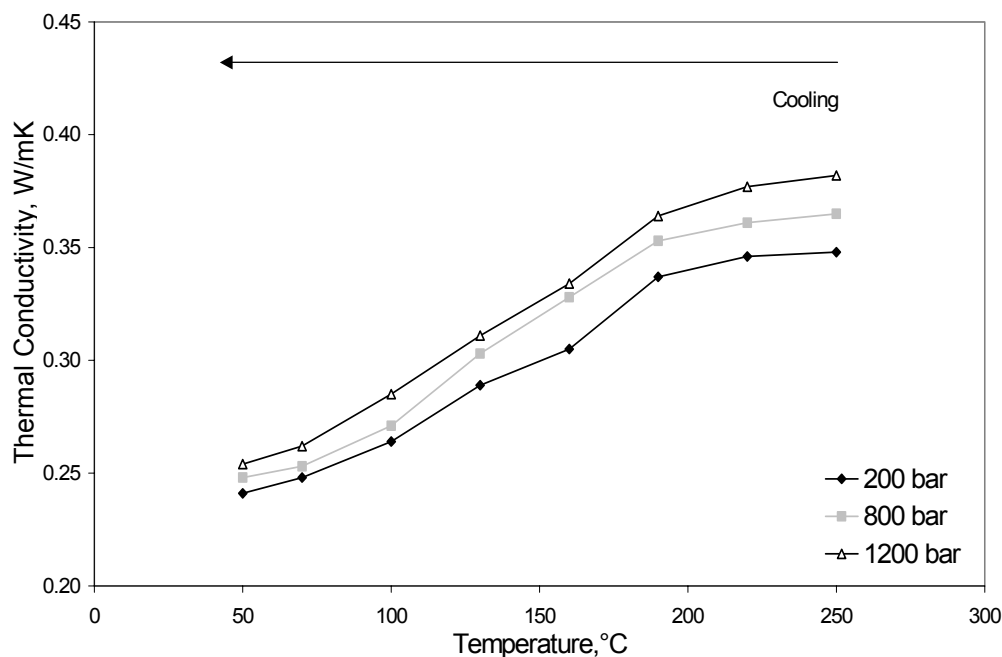


Figure 11 Thermal conductivity of polycarbonate on cooling from 250 °C to 50 °C at pressures of 20 MPa, 80 MPa and 120 MPa

Table 2: Percentage increase in thermal conductivity with increase in pressure of semi-crystalline and amorphous polymers at 250 °C

Material	Approximate % increase in thermal conductivity	
	Pressure increase from 20 MPa to 80 MPa	Pressure increase from 20 MPa to 120 MPa
Polypropylene	8	23
High Density Polyethylene	7	17
Polyethylene (terephthalate)	10	16
Glass filled nylon	2	4
Polystyrene	11	17
Acrylonitrile-butadiene-styrene	4	6
Polycarbonate	5	10

(ii) Specific volume

The specific volume behaviour of polystyrene at pressures of 20 MPa, 80 MP and 120 MPa over the temperature range from 250 °C to 50 °C, in cooling, is shown in Figure 12. The isobaric cooling plots show a decrease in specific volume with decreasing temperature at all pressures, corresponding to an increase in density. The change in gradient of the specific volume versus temperature plot around 100 °C at all pressures (Figure 12) may be due to the polystyrene reaching the glass transition temperature and therefore undergoing a transition to the glassy state. It is noted that this transition temperature increases with increasing pressure.

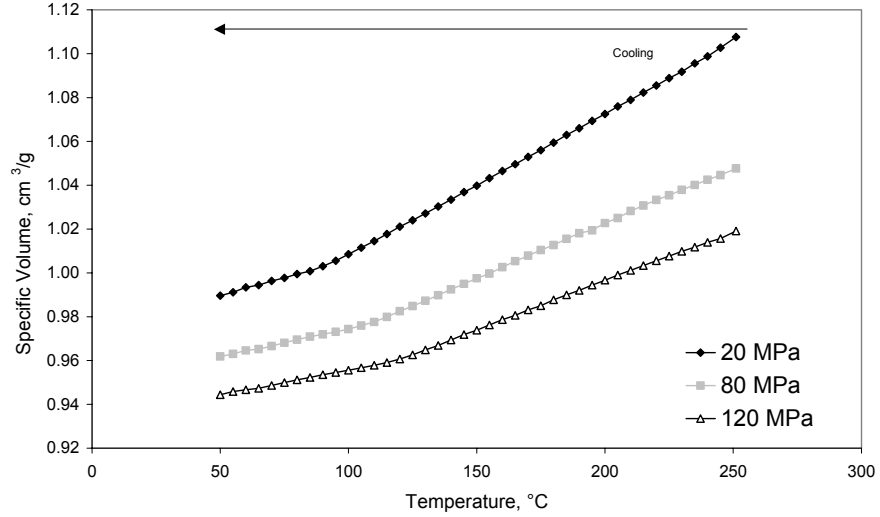


Figure 12 Specific volume of polystyrene on cooling from 250 °C to 50 °C at pressures of 20 MPa, 80 MPa and 120 MPa

3.3 MODELLING OF THERMAL CONDUCTIVITY AND SPECIFIC VOLUME DATA

The specific volume (ν) and thermal conductivity (λ) were modelled as functions of pressure and temperature using the same exponential form, equations 1 and 2 respectively, where ν_0 (cm^3/g), k and ℓ are constants of the specific volume equation, λ_0 ($\text{Wm}^{-1}\text{K}^{-1}$), k' and ℓ' are constants of the thermal conductivity equation, θ is temperature (K), p is pressure (MPa) and θ_0 (K) and p_0 (MPa) are a reference temperature and reference pressure respectively. The values of the constants ν_0 and λ_0 are the specific volume and thermal conductivity values, respectively, at temperature θ_0 and pressure p_0 . Both the temperature dependence and pressure dependence have been modelled using an exponential form, which has been used for modelling the dependence of shear viscosity on temperature and pressure [9,10]. However, an additional exponential temperature term has been included to give a temperature dependence to the pressure dependence factor to account for the divergence of the curves with increasing temperature.

$$\nu = \nu_0 [\exp(k'(\theta - \theta_0))] [\exp(\ell'\theta(p - p_0))] \quad (1)$$

$$\lambda = \lambda_0 [\exp(k'(\theta - \theta_0))] [\exp(\ell'\theta(p - p_0))] \quad (2)$$

The ratio of these expressions can be used to model the relationship between thermal conductivity and specific volume, equations 3 to 5

$$\frac{\nu}{\lambda} = \frac{\nu_0}{\lambda_0} [\exp((k - k')(\theta - \theta_0))] [\exp(\theta(\ell - \ell')(p - p_0))] \quad (3)$$

$$\nu = \lambda \frac{\nu_0}{\lambda_o} [\exp((k - k')(\theta - \theta_o))] [\exp(\theta(\ell - \ell')(p - p_o))] \quad (4)$$

$$\lambda = \nu \frac{\lambda_o}{\nu_0} [\exp((k - k')(\theta - \theta_o))]^{-1} [\exp(\theta(\ell - \ell')(p - p_o))]^{-1} \quad (5)$$

Equations 1 and 2 have been used to fit the data for the polystyrene, Figures 13 and 14. The models do not describe the slight change in gradients at approximately 100 °C, possibly due to the glass transition temperature (T_g) of polystyrene ($T_g = 100$ °C [11]). The specific volume shows a slightly higher gradient and the thermal conductivity a lower gradient above the T_g compared with below the T_g . The specific volume plot, Figure 12, clearly indicates that the change in gradient occurs at higher temperature as the pressure is increased (85 °C at 20 MPa to 110 °C at 120 MPa) suggesting that T_g increases as pressure increases. The effect is not clearly seen in the thermal conductivity plot, Figure 10, primarily due to the fewer data points in the critical temperature range.

It is accepted that Equations 1 - 5 are not suitable for describing the full behaviour of polymers exhibiting a clearly defined melting - crystallisation transition typical of semi-crystalline polymers. However, the model is likely to fit reasonably well to data either side of the transition.

For the polystyrene, good fits of the models to the data have been obtained, with the discrepancy for thermal conductivity being less than 7 % and for specific volume less than 1 %. The correlation of thermal conductivity with specific volume is shown in Figure 15, where data for the temperature range from 250 °C to 50 °C, obtained in cooling, are presented. The correlation is clearly dependant on the pressure. The coefficients of equations 1 and 2, presented in Table 3, have been used to fit the experimental data and provide a reasonable fit. The transition at the T_g is more clearly shown, the effects in thermal conductivity and specific volume being additive in this plot.

Table 3: Coefficient values for the specific volume and thermal conductivity models (equations 1 and 2) for polystyrene

Polystyrene					
Thermal conductivity, λ Wm⁻¹K⁻¹	λ_o Wm ⁻¹ K ⁻¹	k'	ℓ'	θ_o °C	p_o MPa
	0.274	0.00165	3.43E-06	250	80
Specific volume, ν cm³g⁻¹	ν_o cm ³ g ⁻¹	k	ℓ	θ_o °C	p_o MPa
	1.047	0.000427	-1.54E-06	251.1	80

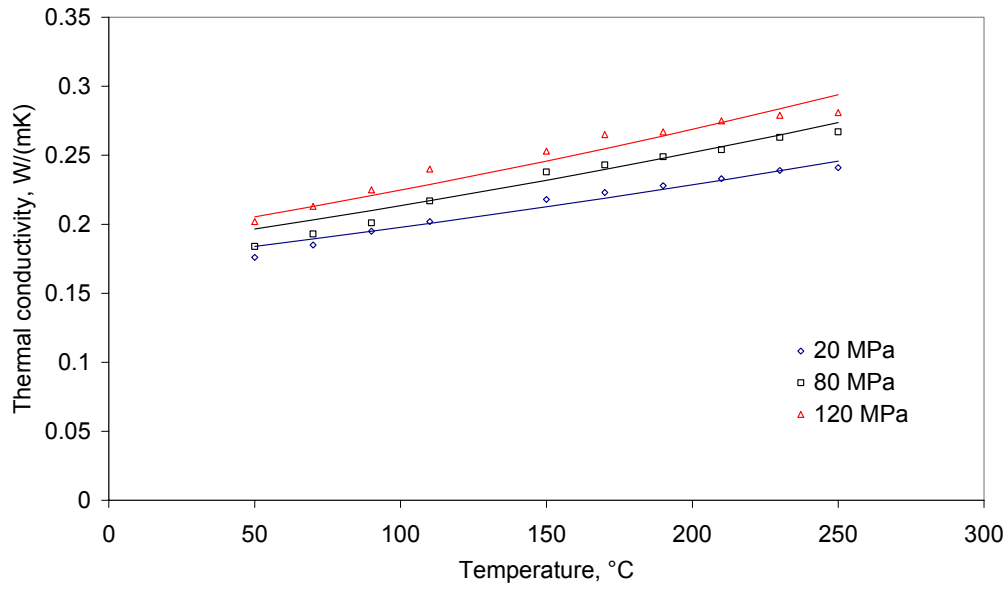


Figure 13 Thermal conductivity data for polystyrene, fitted using equation 1

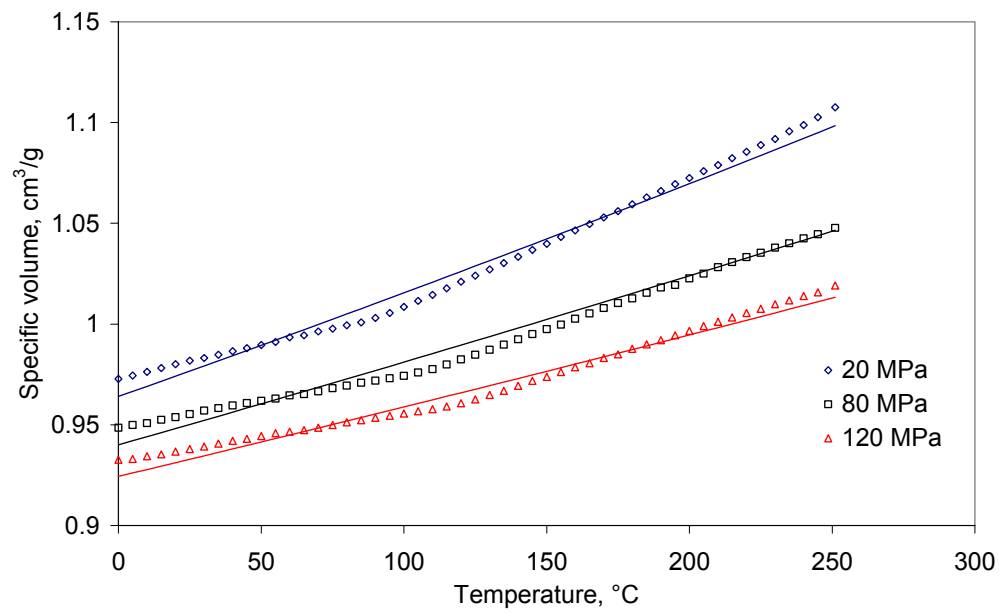


Figure 14 Specific volume data for polystyrene, fitted using equation 2

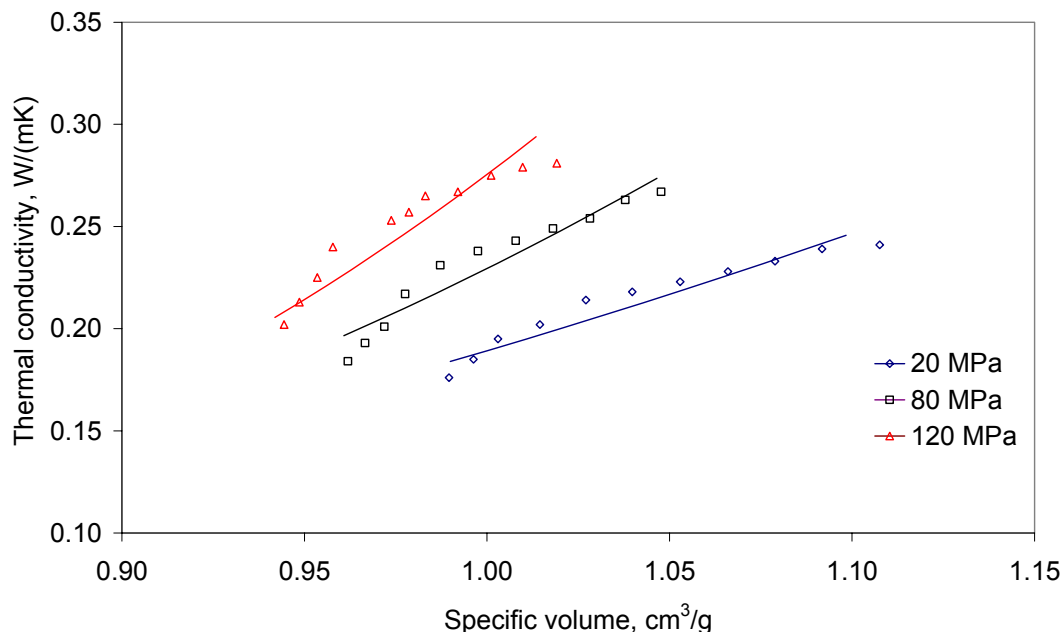


Figure 15 Correlation of thermal conductivity with specific volume data for polystyrene, fitted using equation 5

4 DISCUSSION AND CONCLUSIONS

Measurements of thermal conductivity have been carried out under conditions of pressure and temperature typical of those experienced in industrial polymer processing.

For all the polymers tested, increasing the pressure significantly increased the thermal conductivity of the polymer. Similarly, there was a significant effect of temperature on the thermal conductivity of the material, although the trends were complicated by the occurrence of crystallisation. In general, there was an decrease in thermal conductivity with decreasing temperature. However, for the HDPE and PP materials the values of thermal conductivity either side of the crystallisation transition were relatively independent of temperature. At the crystallisation transition, observed for PP and HDPE, there was a step increase, on cooling, in the measured thermal conductivity values. For PP and HDPE a step decrease in specific volume on cooling, also indicative of the crystallisation transition, was observed. The difference in crystallisation onset temperatures between the thermal conductivity and specific volume methods was considered to be due to differences in the effective cooling rates of the two methods.

In using simulation software for process modelling a single value for thermal conductivity is often used. If the thermal conductivity of the material obtained at ambient conditions is used, then for semi-crystalline materials exhibiting a step change in values at crystallisation, the variation from the near-ambient thermal conductivity value would be relatively small, e.g. for HDPE it would be $0.25 \text{ W/(m.K)} \pm 0.06 \text{ W/(m.K)}$, and for PP it would be $0.30 \text{ W/(m.K)} \pm 0.08 \text{ W/(m.K)}$. However, for PET the near-ambient value was the lowest measured value, being 0.3 W/(m.K) , with values at higher temperatures and pressures being up to 0.48 W/(m.K) . For PS the near-ambient value was 0.17 W/(m.K) with values at higher temperatures and pressures up to 0.28

W/(m.K). These equate to variations in values from the ambient conditions values of up to approximately $\pm 25\%$ for HDPE and PP materials, and an increase above the ambient conditions value by up to 60% for PET, 37% for the glass filled nylon, 38% for ABS, 65% for PS, and 65% for PC. Such unaccounted for variations in thermal conductivity are likely to have a significant effect on modelling predictions.

The increase in thermal conductivity with pressure could, potentially, lead to a reduction in cycle times and so greater processing efficiency. The pressure during an injection moulding cycle can be controlled by configuring the settings of the moulding machine. Increasing the pressure settings on the injection moulding machine may lead to an increase in thermal conductivity of the polymer being moulded and so an increase in cooling rate of the moulded part. However, the shot weight may increase as a result of the increase in pressure specified, energy consumption may be greater and warpage/distortion problems are likely to be worse. Therefore any economic benefit brought about by a reduction in cycle times would have to be considered against these additional costs and potentially other factors.

In conclusion, engineers and designers in the mould design and polymer processing sectors would benefit through improved accuracy of processing predictions through the incorporation and use of temperature and pressure dependent thermal conductivity data in commercially available software packages. The equations presented could be used to model thermal conductivity and specific volume behaviours for amorphous polymers. It is suggested, however, that improvements to the modelling should be pursued to improve the fit to data, particularly across transition temperatures. Through improved processing predictions there is potential for improvements in processing efficiency, and improved product performance through, for example, avoidance of material degradation due to hot spots.

5 ACKNOWLEDGEMENTS

This work was carried out as part of a project on heat transfer during polymer processing (MPP 7.1), which is part of a programme of underpinning research financed by the National Measurement System Directorate (NMSD) of the Department of Trade and Industry. A section of the work was also carried out and funded as part of EUREKA project 2846-Advances In Injection Moulding Technology (AIMTECH).

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