Heat Transfer in Polymers - summary

- Introduction
- Heat Transfer Coefficient
- Thermal Conductivity
- Thermal Imaging
- Industrial Demonstrations
- Standards for Thermal Properties
- Summary of current heat transfer project
- Outline of heat transfer project 2005-08
- Future Needs
Aim of the project

• To help companies measure and model heat transfer in polymer processing

• This should lead to:
  – Right first time design
  – Higher productivity (faster processing)
  – Energy saving
  – Fewer failures in service

Resulting in reduced costs and improved quality
Tasks in the DTI Project

- **Heat Transfer Coefficient**
  - New facility

- **Thermal Conductivity**
  - Uncertainty analysis
  - Extension of method to new materials

- **Simulation**
  - To identify the important data
  - To help design equipment
  - Moldflow & NPL’s own software

- **Industrial Demonstrations**
  - Zotefoams
  - Corus

- **Dissemination**
  - Web site, IAGs, PAA Newsletter articles, trade press articles, measurement notes, scientific paper
An associated Eureka project (AIMTECH) is progressing. Its aim is to improve productivity of injection moulding, with a main focus on reducing cycle times by using copper alloy moulds in injection moulding.

NPL Role:
- Measurement of the thermal conductivity of polymer melts (T,P)
- Understanding the role of the mould/melt interface: Modelling heat transfer and the effect of uncertainties

Six UK companies involved with a £25k co-funding contribution, close fit with the DTI project.
Heat transfer coefficient
Heat Transfer Coefficient

- It is the heat flux per unit area (q) across an interface from one material of temperature $T_1$ to another material of temperature $T_2$:

$$h = \frac{q}{T_1 - T_2}$$

units: $\text{Wm}^{-2}\text{K}^{-1}$

- Boundary condition for process simulation

- In injection moulding & compression moulding
  - Polymer to metal
  - Polymer-air-metal (GASM, …)

- In extrusion & film blowing
  - Polymer to fluid (eg air or water)

- This project has built apparatus to measure heat transfer coefficient and will investigate the significance of different interfaces to commercial processing
Features of apparatus

- Room temperature to 275 °C, pressure to at least 500 bar
- Polymer samples 2 mm to 25 mm thick
- Interchangeable top plate to investigate
  - Different surface finishes
  - Effect of mould release agents
- Option to introduce a gap between polymer & top plate
  - Shrinkage, sink marks
- Instrumented with temperature measurement devices and heat flux sensors
Heat transfer apparatus

Side view

cold plate

hot plate

sample
Heat transfer apparatus

- Displacement transducer
- Cooling pipes
- Thermocouples
- Heat flux sensors
- Pressure transducer port
- Mould face
- Heater element
- PTFE seal
- Thermo-couple ports
- Fibre optic thermocouple port
- Air gap
- Sample
- Outer guard ring
- Pressure transducer port

NPL
National Physical Laboratory
Heat transfer coefficient
Modelling of key features

• Effect of an air gap

• Effect of vertical thermocouple on distort the temperature field
Air Gap

Mould at 50 °C with air gap of 0, 0.5 & 1 mm

Polymer at 250 °C
Effect of a thermocouple

Mould at 50 °C

Polymer at 250 °C
Simulation of Heat Transfer with Fibre Optic (left) & Thermocouple (right)
Comparison of thermocouple & fibre optic

TherMOL prediction of the temperature difference between the sensor (centre) and the edge (PP) for PP & a thermocouple and PP & a optical fibre after 400s
Heat transfer coefficient effect of uncertainties
Pipe ‘T’ piece and 80 mm diameter disc models
The Change in Temperature Over Time For A Given Location (T1149) on the T-Pipe For Analyses With Different Heat Transfer Coefficient
Effect of uncertainties in HTC

The Effect Of Mould-Melt Heat Transfer Coefficient Upon Time To Freeze Part
For Discs Of Different Thickness

Percentage Variation In Time To Freeze Part
From The Standard Simulation Result
For The Given Disc Thickness, %

- Default Mould-Melt Heat Transfer Coefficient
- Minimum Mould-Melt Heat Transfer Coefficient
- 1/10 Of Default Mould-Melt Heat Transfer Coefficient
- *10 Default Mould-Melt Heat Transfer Coefficient

Disc Thickness, mm
The Effect of Uncertainty in Heat Transfer Data on The Simulation of Polymer Processing

J. M. Urquhart and C. S. Brown

http://libsvr.npl.co.uk/npl_web/search.htm
• Initial testing commenced using HTC equipment

• To investigate effect of:
  – Different surface finishes/mould materials
  – Mould release agents
  – Air gap between polymer & top plate
    (simulating shrinkage and sink marks)
Thermal conductivity measurements
Thermal Conductivity Measurements Under Industrial Processing Conditions:

• More accurate data for modelling software

• Reduce warpage and hot spots during injection moulding process – reduce waste

• Reduce cycle times and improve processing efficiency
Plan of Action:

• Measured thermal conductivity of amorphous and semi-crystalline polymers at injection moulding pressures

• Used experimental techniques to attribute uncertainty to thermal conductivity measurements

• Compared thermal conductivity measurements with known pvT technique
Line source probe apparatus

\[ T_2 - T_1 = \frac{C Q^*}{4 \pi \lambda} \ln(t_2/t_1) \]

- \( C = \text{"Probe" \cdot Constant} \)
- \( Q^* = \text{Heat input per length} \)
- \( \lambda = \text{Thermal conductivity} \)

Measures thermal conductivity at industrial processing pressures
Thermal conductivity repeatability measurements and uncertainty
Thermal Conductivity of HDPE (Atmospheric and 1000 bar Pressures)

Repeatability (95% confidence level) of thermal conductivity test data for one operator testing HDPE HCE000 from 170°C to 50°C at 1000 bar pressure (green) and at ambient pressure (blue)

1000 bar repeatability 8%
ambient pressure repeatability 16%
## Uncertainty Budget For NPL Line-Source Thermal Conductivity Probe (Atmospheric Pressure)

<table>
<thead>
<tr>
<th>Type</th>
<th>Value ± %</th>
<th>Probability Distribution</th>
<th>Divisor</th>
<th>C_i</th>
<th>Uncertainty Contribution ± %</th>
<th>Uncertainty Squared ± %</th>
<th>V_i or V_{eff}</th>
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</thead>
<tbody>
<tr>
<td><strong>Type A</strong>&lt;br&gt;Repeatability</td>
<td>15.6 @ 2 std devs</td>
<td>Normal</td>
<td>2</td>
<td>1</td>
<td>7.815 @ 1 std dev</td>
<td>61.07</td>
<td>89</td>
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<tr>
<td>Reproducibility</td>
<td>13.6 @ 2 std devs</td>
<td>Normal</td>
<td>2</td>
<td>1</td>
<td>6.801 @ 1 std dev</td>
<td>46.25</td>
<td>89</td>
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<tr>
<td><strong>Type B</strong>&lt;br&gt;Non-uniformity of heat input</td>
<td>0.002</td>
<td>Rectangular</td>
<td>1.73</td>
<td>1</td>
<td>0.00116</td>
<td>1.34E-06</td>
<td>∞</td>
</tr>
<tr>
<td>Non-uniformity of temperature</td>
<td>0.0</td>
<td>Rectangular</td>
<td>1.73</td>
<td>1</td>
<td>0.0000</td>
<td>0.000</td>
<td>∞</td>
</tr>
<tr>
<td>Sample height</td>
<td>0.0</td>
<td>Rectangular</td>
<td>1.73</td>
<td>1</td>
<td>0.0000</td>
<td>0.000</td>
<td>∞</td>
</tr>
<tr>
<td>Time</td>
<td>0.0</td>
<td>Normal</td>
<td>1</td>
<td>1</td>
<td>0.0000</td>
<td>0.000</td>
<td>∞</td>
</tr>
</tbody>
</table>

### Calculation of Uncertainty

- Sum of squares
- Square root of sum of squares
- Multiplication by $k = 2$ for 95% confidence level
- Final uncertainty value

```
107.3 %
10.4 %
±20.7 %
```

**Final Uncertainty Value**
Thermal conductivity measurements under pressure
Materials tested:

Amorphous:

- Acrylonitrile-butadiene-styrene
- Polystyrene
- Polycarbonate

Semi-crystalline:

- Polypropylene
- Polystyrene
- Polyethylene(terephthalate)
- Glass filled nylon
Thermal Conductivity Behaviour of Typical Amorphous Material (PS) Under Pressure

Thermal conductivity of polystyrene (AAATK002) on cooling from 250°C to 50°C at pressures of 200, 800 and 1200 bar.
Thermal Conductivity Behaviour of Typical Semi-crystalline Material (PP) Under Pressure

Thermal conductivity of polypropylene (AAATK004) on cooling from 250°C to 50°C at pressures of 200, 800 and 1200 bar.
pvT measurements under pressure
Schematic of pvT Instrument

Measurement piston

Known diameter (d)

Thermocouple

Floating measurement cylinder

Sample

Springs

Heater band with channel for cooling media

Sealing

Fixed piston
pvT Behaviour of a Typical Amorphous Polymer (PS)
pvT Behaviour of a Typical Semi-crystalline Polymer (PP)
Models for specific volume and thermal conductivity

\[ v = v_0 \left[ \exp(k(\theta - \theta_0)) \right] \left[ \exp(\ell(p - p_o)(\theta + 273.15)) \right] \]

\[ \lambda = \lambda_0 \left[ \exp(k'(\theta - \theta_0)) \right] \left[ \exp(\ell'(p - p_o)(\theta + 273.15)) \right] \]

<table>
<thead>
<tr>
<th>PS</th>
<th>( \lambda_0 )</th>
<th>( k' )</th>
<th>( \ell' )</th>
<th>( \theta_0 )</th>
<th>( p_o )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal conductivity, ( \lambda )</td>
<td>0.274</td>
<td>0.00165</td>
<td>3.43E-06</td>
<td>250</td>
<td>80</td>
</tr>
<tr>
<td>Specific volume, ( \nu )</td>
<td>1.047</td>
<td>0.000427</td>
<td>-1.54E-06</td>
<td>251.1</td>
<td>80</td>
</tr>
</tbody>
</table>

\[ \lambda = \nu \frac{\lambda_0}{\nu_0} \left[ \exp((k - k')(\theta - \theta_0)) \right]^{-1} \left[ \exp((\ell - \ell')(p - p_o)(\theta + 273.15)) \right]^{-1} \]
Thermal conductivity data for polystyrene

Thermal conductivity, $W\, m^{-1}\, K^{-1}$

Temperature, °C

20 MPa
80 MPa
120 MPa
Specific volume data for polystyrene

<table>
<thead>
<tr>
<th>Temperature, °C</th>
<th>Specific volume, cm³/g</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.9</td>
</tr>
<tr>
<td>50</td>
<td>0.95</td>
</tr>
<tr>
<td>100</td>
<td>1.0</td>
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<tr>
<td>150</td>
<td>1.05</td>
</tr>
<tr>
<td>200</td>
<td>1.1</td>
</tr>
<tr>
<td>250</td>
<td>1.15</td>
</tr>
<tr>
<td>300</td>
<td>1.2</td>
</tr>
</tbody>
</table>

- **20 MPa**
- **80 MPa**
- **120 MPa**
Correlation of thermal conductivity with specific volume data for PS

Thermal conductivity, W m\(^{-1}\)K\(^{-1}\)

Specific volume, cm\(^3\)/g

- 20 MPa
- 80 MPa
- 120 MPa
Implications of Results

- Increase in pressure gives increase in thermal conductivity - reduction in cycle times – possible cost benefits

- Increase in crystallisation temperature for semi-crystalline polymers with increase in pressure – may reduce time to freeze parts - possible cost benefits

- More accurate data based on industrial processing conditions - improvements in commercial modelling packages - cut scrap rates by improving warpage and hot-spot prediction – possible cost benefits

- Crystallisation temperature for PP occurred over a similar temperature range for thermal conductivity and specific volume results confirming validity of TC tests

- Correlation of specific volume and thermal conductivity values
Thermal Imaging
DEPC IR Camera
Schematic Diagram of IR Camera Operation

Infrared camera

Image grabber/ recorder

Image Analysis
(manual or automated)

Sample
Cooling of Hot Melt Adhesive Study Using IR Camera

Time after extrusion

37 seconds 152 seconds
Heating of Hot Melt Adhesive Study Using IR Camera

Time after start of heating

0 seconds 130 seconds
Infra Red Camera

- Non contact method
- Produces visual record of thermal changes during heating and cooling of sample
- Visual record can be analysed in quantitative way to produce a time vs. temperature plot of thermal changes
- Can be customised to an individual system
- Easy to operate once it has been set up correctly
- Samples to be tested have to be of similar weight and geometry for comparisons to be made
INDUSTRIAL TRIALS
Corus & Zotefoams
Industrial Demonstrations

• Aim is to demonstrate practical benefits of heat transfer measurements and modelling

• Corus
  – Thermal conductivity of plastisol coated steel before and after solidification

• Zotefoams
  – Heat transfer during cooling of polyolefin foam
• Use DSC method to measure thermal conductivity of bilayer Plastisol/steel

• Measure before and after solidification

• Data useful in predicting optimum line speeds

  – Earlier work had shown that the polymer layer was significant in terms of heat transfer
DSC method for thermal conductivity

DSC pan

Indium or eutectic

Multilayer sample
Heat Transfer for sapphire

\[ K_x = K_i \left( \frac{m_x}{m_i} \right)^2 \frac{h_x}{h_i} \] (Khanna et al. (1988))

DSC method for thermal conductivity
Problem: waviness in foams – thermal issue
• Model heat transfer
• Measure (T, heat flux) over time
• Model/measure shrinkage
• Calculate internal stresses
• Use bending theory to predict curvature
Standards in Thermal Properties Measurement
ISO 11357 Plastics - Differential scanning calorimetry (DSC)

ISO 11357-1: 1997 Part 1: General principles (now due for revision)

ISO 11357-2: 1999 Part 2: Determination of glass transition temperature

ISO 11357-3: 1999 Part 3: Determination of temperature and enthalpy of melting and crystallization

ISO/FDIS 11357-4 Part 4: Determination of specific heat capacity

ISO 11357-5: 1999 Part 5: Determination of characteristic reaction-curve temperatures and times, enthalpy of reaction and degree of conversion

ISO 11357-6: 2002 Part 6: Determination of oxidation induction time

ISO 11357-7: 2002 Part 7: Determination of crystallization kinetics

*Potential proposal for thermal conductivity measurement by temperature modulated DSC*
Thermal conductivity standards

ISO/AWI 22007 Plastics - Determination of thermal conductivity and thermal diffusivity

ISO/AWI 22007-1 Part 1: General principles

ISO/AWI 22007-2 Part 2: Gustafsson hot-disc method

ISO/AWI 22007-3 Part 3: Temperature wave analysis method

ISO/CD 22007-4 Part 4: Laser flash method
**Thermal conductivity standards**

**Hot Wire**

**Line Source**
- ASTM D 5930-01, Test Method for Thermal Conductivity of Plastics by Means of a Transient Line-Source Technique

**Laser Flash**
- ISO 18755: 2005 Fine ceramics (advanced ceramics, advanced technical ceramics) - Determination of thermal diffusivity of monolithic ceramics by laser flash method

**Guarded Hot Plate**

**Guarded Heat Flux**
Heat transfer project concluding summary
Summary – Heat Transfer

• Heat transfer coefficient apparatus now being used
  – Design assisted by numerical modelling studies
  – Effect of uncertainties investigated (report available)

• Melt thermal conductivity
  – Nano-filled materials
  – Powders/granules
  – Effect of pressure
  – Effect of uncertainties investigated (report available)

• ISO Standards being developed

• New IAG members facility on website
  http://www.npl.co.uk/npl/cmmt/polyproc
The next 6 months

• Complete commissioning and trials on heat transfer coefficient equipment

• Industrial demonstrations (Corus / Zotefoams) to be completed

• Dissemination of thermal conductivity measurement work
  – scientific and conference paper, articles
Objectives:

• Development of the method for the measurement of heat transfer properties across surfaces (particular interest has been expressed in the effect of the solid/air interface)
• Industrial case study to demonstrate the value of reliable heat transfer data
• Support development of standards for measurement of thermal properties of plastics, including an intercomparison of thermal conductivity methods that are being proposed for standardisation
• Assessment of uncertainties in heat transfer data and effect on modelling predictions
• Development of a new user-friendly web-enabled modelling facility, to facilitate industrial adoption of the above
Your:
Ideas,
comments,
suggestions,
participation,
contributions, ...

to steer the project to maximise the benefits to you.
Heat Transfer
Future Needs
Heat transfer is:

• key to polymer processing
• still inadequately understood
• key to increasing throughput - process times dominated by the cooling phase
• significant in affecting product properties, e.g. warpage, inadequate melting, thermal degradation
Improved heat transfer could:

- Contribute significantly to reduction in UK energy bill
- Bring indirect benefit to quality of life
- Save money for UK industry
Areas where future work to increase understanding of heat transfer required:

- Water assisted injection moulding (WAIM)
- Gas assisted injection moulding (GAIM)
- Effect of air gaps, mould materials, supercritical CO₂, helium
- Micro-moulding
- Additives, fillers effect on decreasing thermal conductivity of insulators
- Developing techniques for measuring heat transfer properties of foam
- Curing of fibre/matrix composites and cross-linking of rubbers
Further areas where future work to increase understanding of heat transfer required:

• Effect of nanoparticles on heating and cooling of polymer nanocomposites during processing
• Effect of dispersion of nanoparticles on thermal conductivity and heat transfer coefficient of nanofluids
• Measurement of heat transfer within microfluidic systems to improve data available for modelling
• Investigation of heat transfer during processing of foods for packagers and processors
• Development of techniques for increasing heating/cooling rates for food
• Measurement of surface heat transfer coefficient and external heat transfer medium (water, air) for range of foods
Your suggestions/comments?
AOB: