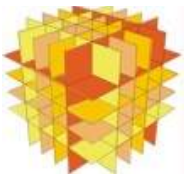


Flow Induced Crystallisation of PE and PP melts within a Multi-Pass Rheometer

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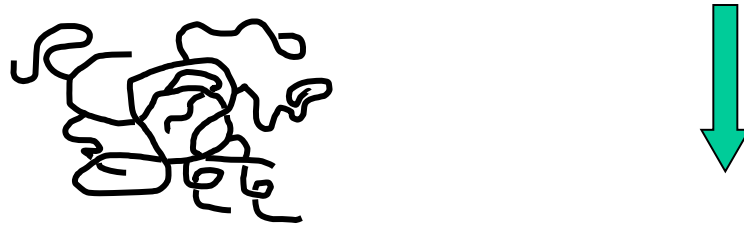
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Knowledge Based Materials
Estremoz, 20-29 August 2007

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What is Flow Induced Crystallisation (FIC) of polymers ?

Upon cooling from the molten state the polymer chains tend to pack together in an organized way

Polymers → long, entangled chains → limited mobility



➤ **Imperfect** crystallisation

➤ **Non-equilibrium, Kinetics-dominated process**

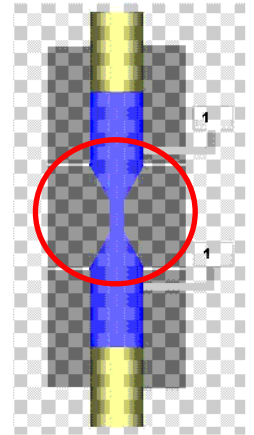


Great influence of Processing Conditions, and thus of **Flow**, on the final structure

Experimental method

- The polymer melt was cooled down to the desired T_c
- Crystallisation was induced by moving the pistons of the **multipass rheometer** (MPR) and forcing the material to flow within a narrow well defined test section geometry.

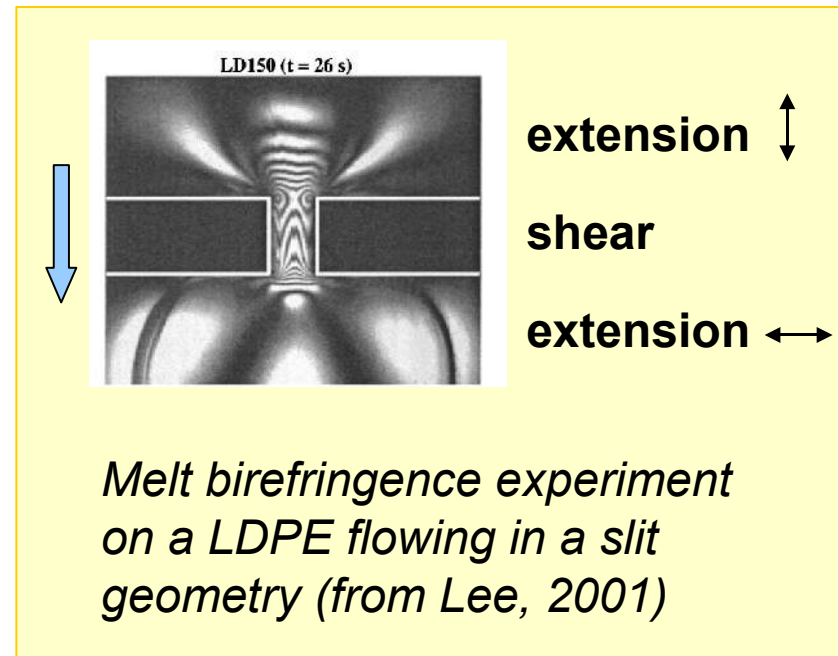
MPR



➤ Optical observations in slit geom.

- Birefringence → stress field
- Bright field → crystallisation

➤ X-rays observations in Be Capillary → microstructure



Why study flow-induced crystallisation (FIC) within an MPR?

The **final properties** of semi-crystalline polymers are determined by the **crystalline structure**.

In forming processes, such as **injection moulding** crystallisation occurs under flow.

Complex flow

Non-isothermal

Very high shear rates

The **MPR** provides a platform for performing controlled complex flow experiments that have the potential to be numerically simulated.

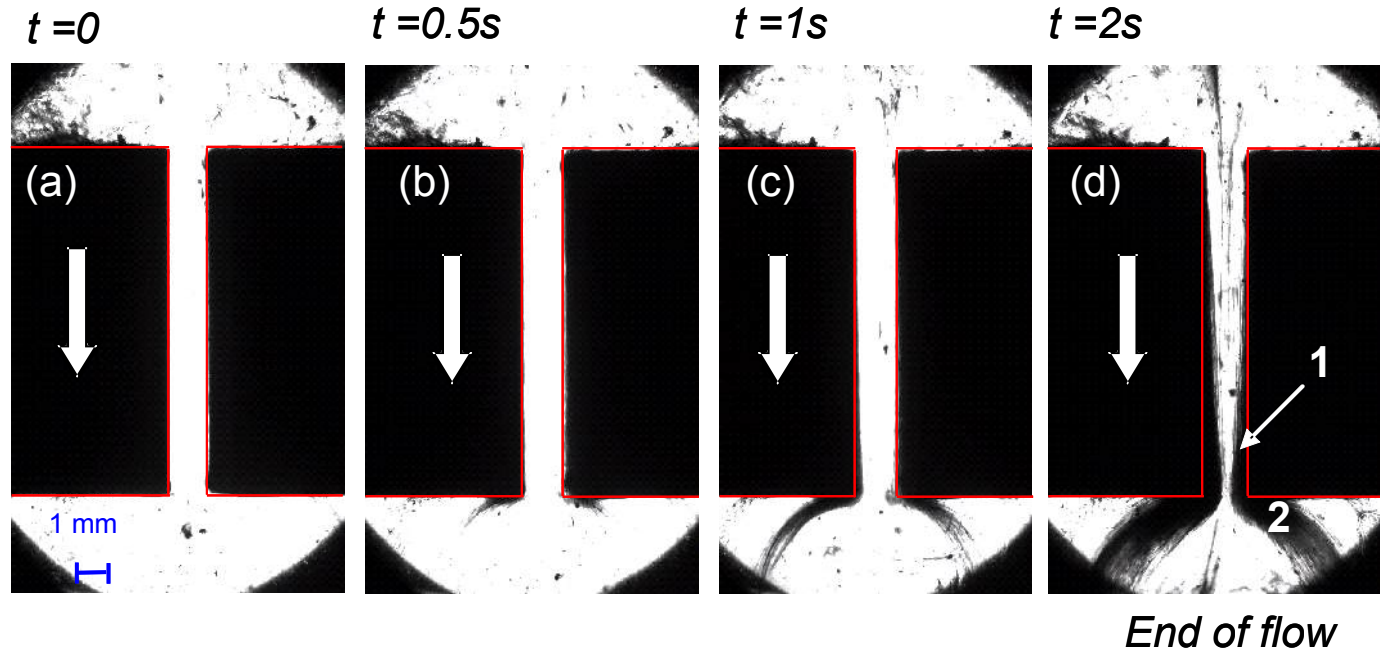
High shear rates, typical of industrial processes, are accessible.

FIC of HDPE

2D-slit ; T=130°C

$$\dot{\gamma}_{w,app} \approx 200 \text{ s}^{-1}$$

(observation
in bright field)



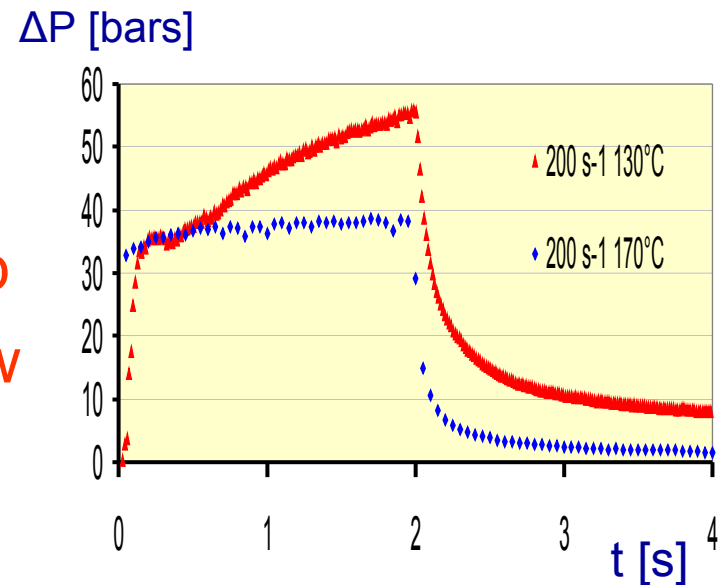
Fibrous crystals are generated at the walls:

1. The crystals accumulate
at the end of the duct



P build-up
during flow

2. Downstream: crystal “fangs”



Conclusions

- HDPE :**
- Fibrous crystallisation **during** the pistons movement.
 - The fibrous crystals form locally in the **high stress zones** of the flow
 - Crystallisation **coupled with the flow-field** and the stress distribution.

- PP :**
- The line-nuclei are generated during flow.
 - Most of the crystallisation process can occur **after** the flow (*see poster*).
 - **Qualitative diagrams** showing the crystal morphology in isothermal FIC MPR experiments as a function of T and wall stress were created.
 - **Numerical simulations** will enable to propose a model.

Acknowledgements

- Polymer Fluids Group



- EPSRC



Engineering and Physical Sciences
Research Council

- Marie Curie Actions



Thank you for your attention

➤ *Objectives:*

Understand and model the physical processes governing FIC

➤ *Materials:*

From “Commercial” to “Tailored-architecture” polymers

➤ *“Strategy”:*

- Characterise the materials.

- Identify the stress (τ), orientation (Φ) and stretch (λ) levels of the molecules at local positions in the flowing polymer by matching melt birefringence expts at T slightly $> T_c$ with numerical simulations.

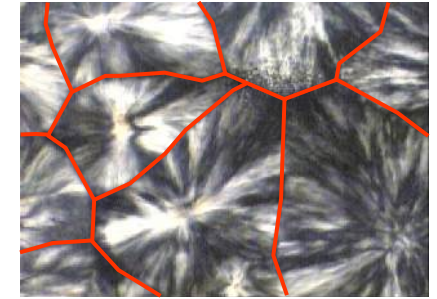
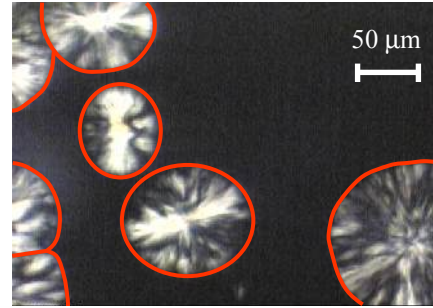
- Relate τ , Φ and λ to the nucleation and growth parameters of an Avrami-type crystallisation model.

Effect of flow on polymer crystallisation

Quiescent conditions:

Nucleation & Growth

nuclei → lamellae → spherulites



Optical observation of PP spherulites: growth & impingement

Applied Flow Field: *[Deviation from spherical coil configuration]*

Enhances Crystallisation:

→ Thermodynamics

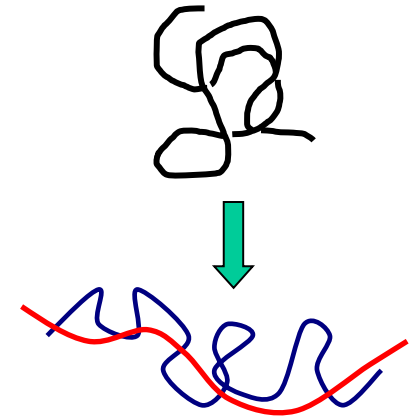
Increase in the T_c

→ Kinetics

Reduction of crystallisation time

→ Morphology

Elongated structures (fibrous crystals)



Why study FIC within an MPR ?

The **final properties** of semi-crystalline polymers are determined by the **crystalline structure**.

In forming processes, such as **injection moulding** crystallisation occurs under flow.

The **MPR** provides a platform for performing controlled complex flow experiments that have the potential to be numerically simulated.

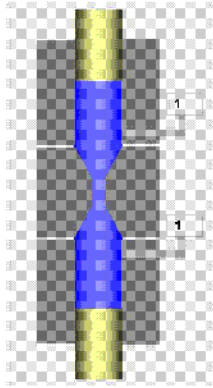
High shear rates, typical of industrial processes, are accessible.

➤ **Optical** observations in **slit geometries**

- **Birefringence** → **stress field**
- **Bright field** → **crystallisation**

➤ **X-rays** observations in **Be Capillary** → **microstructure**

- Complex flow
- Non-isothermal
- Very high shear rates



LD150 (t = 26 s)

extension \updownarrow

shear \longleftrightarrow

extension \longleftrightarrow

Melt birefringence experiment on a LDPE flowing in a slit geometry (from Lee, 2001)