

# Modeling of Anisotropic Polymers during Extrusion

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## Modeling of Anisotropic Polymers during Extrusion

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### Abstract

It is known that a liquid crystal polymer (LCP) melt aligns in the direction of the shear flow when it passes through an extrusion die. This alignment causes thin films of the anisotropic LCP material to display different properties in different directions. To overcome this problem, many complex die design technologies have been developed that involve moving surfaces. However, there is a clear need to develop a method of predicting crystal orientation (alignment) to aid in die design. This paper investigates different modeling methods, and develops a numerical modeling technique using FLUENT, to predict molecular alignment by correlating it to streamlines of flow. This model also incorporates the complex rheology of the LCP in predicting the resulting alignment. It is shown that for a new cross-flow design of the extrusion die that at around  $Re=500$  the maximum angle between the vectors is 80 degrees and at lower  $Re$  it does not satisfy the desired angle between the two flows.

### Introduction

Any rod like molecule or crystal tends to orient itself in the direction of shear as it moves with in a flow. Since polymers consist of long chains of molecules, they experience the same phenomena when flowing in the melt condition. For most polymers, the molecules change direction and distribute uniformly in space during the solidification. However due to the higher inertia of the crystals in the liquid crystalline polymers, the oriented crystals will keep their order during the solidification and as a result the final product will show anisotropy in the material properties.

This investigation focuses on film casting of LCPs and develops ways to prevent the anisotropy of properties in the final product. The challenge here is to alter the rheology of the polymer melt inside the die in order to have isotropic material properties in the plane of the film. The use of biaxial shear flow during extrusion, elongational strain after extrusion, electromagnetic field effects, and thermal treatment to develop isotropic films has been discussed by Lusignea et al. [1].

Farrell and Lawrence use co-rotating extrusion dies to produce biaxially oriented films of LCPs [2]. Due to the complexity of this type of dies and built-in instabilities during this extrusion process, a simpler stationary extrusion die is desirable.

### Geometry

An important parameter to be considered when designing the geometry of polymer dies is that the flow pattern inside the die highly influences the polymer extrusion process and any sharp edges inside the die which result in the flow to become turbulent should be avoided [3]. Turbulence inside the die increases the residual stresses which will be released after the extrudate leaves the die and causes instabilities in the extrusion process. The conventional die for extrusion of polymer films or the coat-hanger design is shown in Figure 1. These dies are designed such that the velocity of the extrudate coming out of the die is constant across the lip of the die.

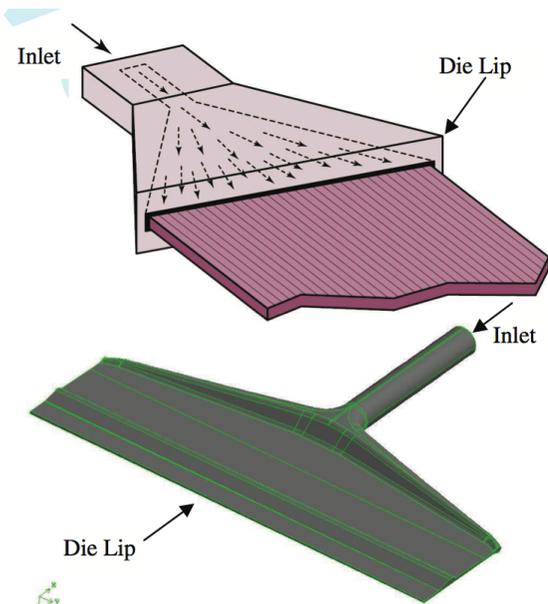


Figure 1. Schematic of a coat-hanger die (up) inside the actual die (down) [4]

In this study, simulation of the polymer flow inside this die geometry was performed with different material properties to investigate the difference between the rheology and the dependence of pressure drop on the material properties. Another type of dies which is designed to produce plane isotropic properties is corotating extrusion die. The co-rotating extrusion dies try to use the viscosity of the polymer to produce a laminar flow with a lateral shear flow that has a three dimensional profile at the lip of the die. This geometry, as shown in Figure 2, has two co-rotating cylinder which operate at high temperature and the polymer melt flows between them. The change in the direction of the crystallines is introduced through the shear effect of the cylinders.

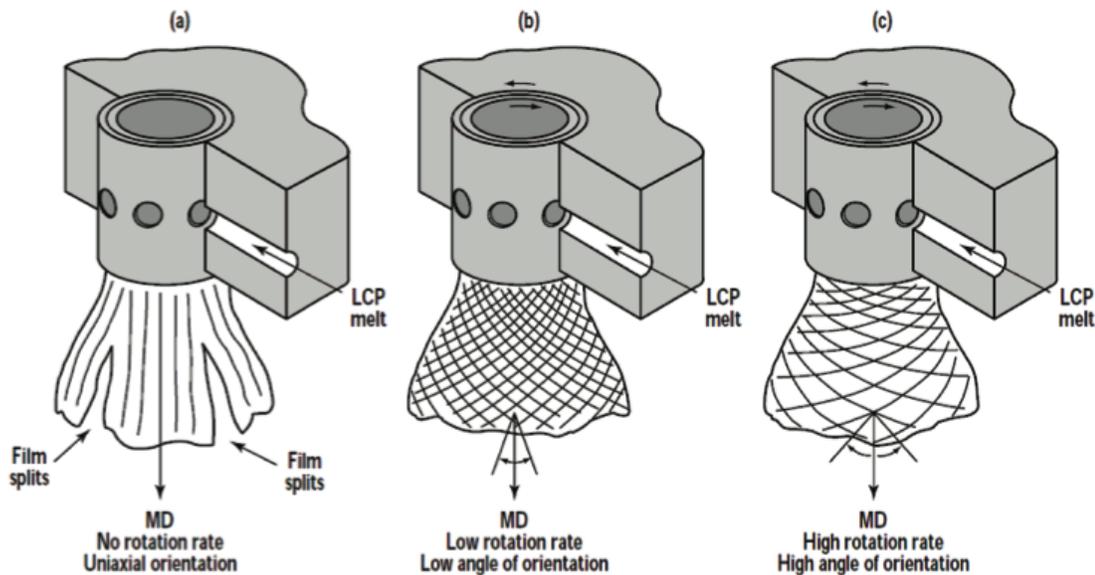


Figure 2. Geometry of the co-rotating biaxial die [4]

A new design for the die geometry introduced here uses cross flows to produce the required lateral shear across the thickness of the film. As a result, no moving part is used in the die. A simple geometry of cross-flow die consists of two cross flows which interact with each other along the mid-plane. The interaction of these two symmetrically positioned flows causes the mid-plane to exit the lip normal to the exit plane while from the midplane to the upper and lower planes velocity vectors change direction gradually. To construct this geometry, preprocessing software GAMBIT was used since it can export the geometry, mesh and boundary conditions to FLUENT. The idea of interacting cross flows was built with 20 channels in which the flow in half of them is perpendicular to the other half. These channels are open along the interface between them and provide the interaction of flows inside the die (Figure 3).

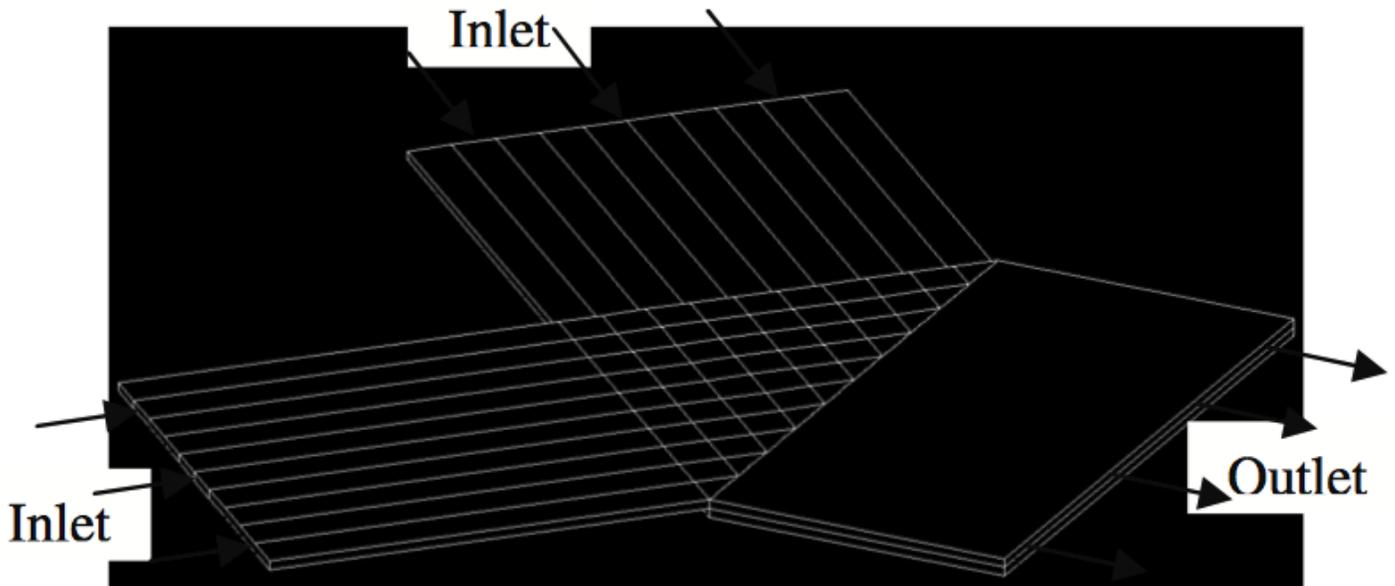


Figure 3. Geometry of the cross flow

This geometry can be constructed by machining the channels inside the upper and lower part of the die since the interface between them is open. It should also be noted that since the velocity of the polymer is inherently 3D in this flow, it is not possible to make a 2D geometry representing cross flows.

## Grid Generation

The required mesh to decompose the geometry to finite volumes was generated in GAMBIT. Structured cubic elements were generated for the geometry of the crossed flow with the consideration of the flow pattern. Using hexahedral elements, it is possible to achieve higher accuracy of the results with much fewer grids. In addition these types of grids are more desirable because modeling the free surface is only possible with structured grids normal to the approximate direction of flow. The maximum skewness of the generated grids is calculated to be 0.25 and the maximum aspect ratio of elements is 7.1, which are in the range of acceptable values. There are a total of 410,000 elements inside and outside the die. Figure 4 shows the constructed grid in different locations of the die.

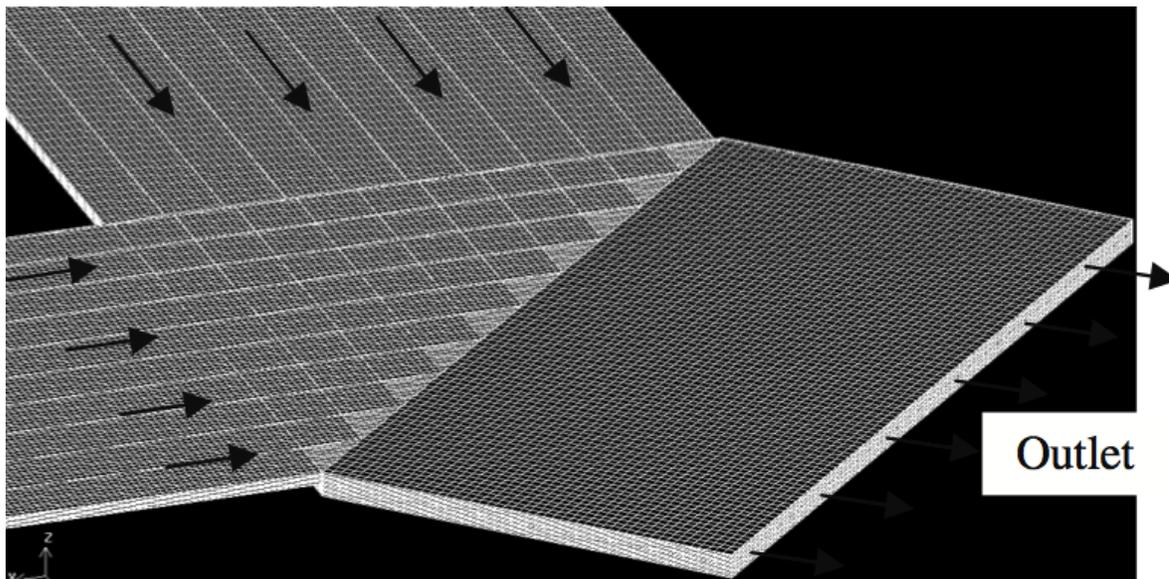


Figure 4. Structured grid on cross flow geometry

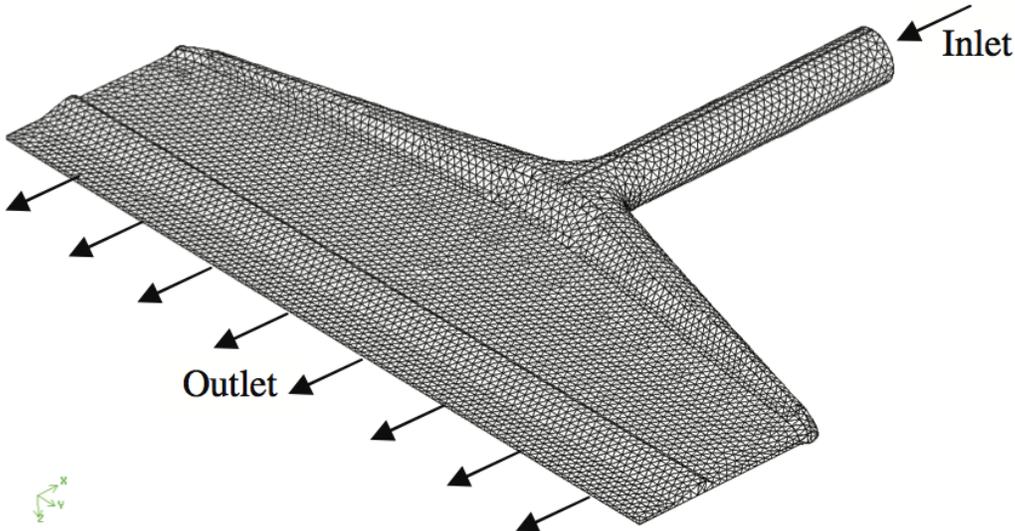


Figure 5. Unstructured grid on the geometry of the coat-hanger die

Due to the complexity of the coat-hanger die, unstructured hexahedral elements were chosen for decomposing the geometry into finite volumes. Figure 5 shows the generated mesh for the coat-hanger geometry (mesh density is reduced here for demonstration purpose). The mesh consists of 3,000,000 elements with the worst skewness of 0.777 and worst aspect ratio of 3.5 which are both in the range of acceptable limits.

## Material Properties and Boundary Conditions

Two different models for material properties were used to simulate the flow inside the die. The first model considers the fluid to be Newtonian and defines a constant viscosity. In this model stress and strain inside the flow are linearly proportional. For this case the viscosity of the fluid was taken to be 0.1 Pa.s for low viscosity and 220 Pa.s for high viscosity.

The second model was chosen based on the available experimental results for one type of LCP. An experiment based on capillary rheometer with capillary length of 20 mm and diameter of 1 mm has been done. This experiment was done under isothermal condition where the temperature was kept constant at 350°C. (Figure 6).

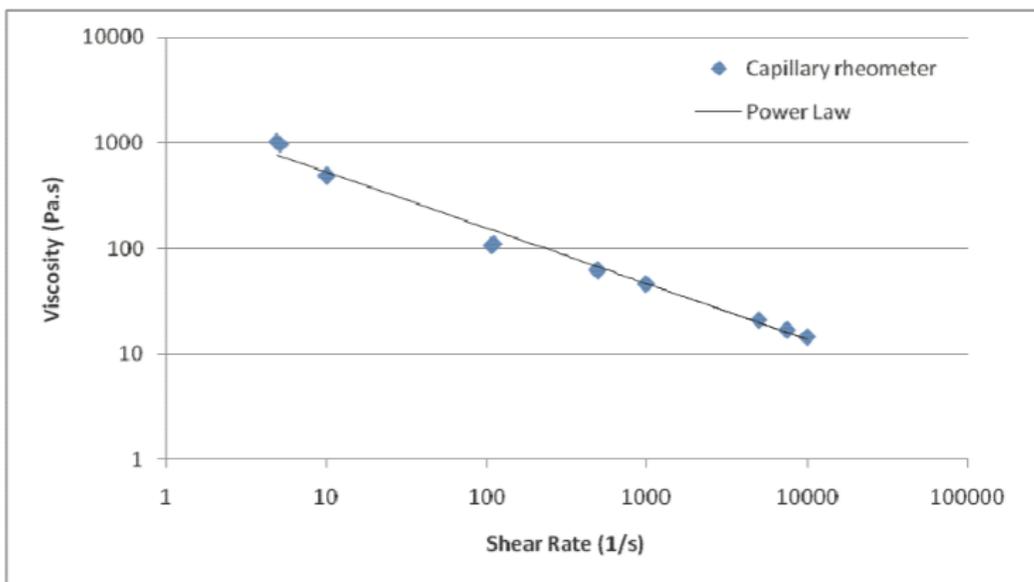


Figure 6. Viscosity of LCP as a function of shear rate used in the simulation

By looking at the curve of viscosity vs. strain rate on a log-log scale, it appears that it can be modeled by the power law model. Based on this information the nonNewtonian power law model was used as a second model to simulate the flow inside the die. In the power law model, equation (1) is used in the constitutive equation to model viscosity [5]:

$$\eta_{\min} < \eta = k\dot{\gamma}^{n-1} e^{T_0/T} < \eta_{\max} \quad (1)$$

To find the parameters to substitute in this equation, the automatic option of Polymat software was used. Moreover, for defining this model completely, the minimum and maximum values of viscosity should be given which was chosen to be the maximum and minimum values in the capillary experiment. (See Table 1).

Three different boundary conditions were used to define the boundaries of the geometry which include velocity inlet, walls and outlet. The channels inside the die were modeled as zero thickness walls with zero tangent and normal velocities. Also, since there is no free wall boundary condition, the flow outside the die is also considered to be through a channel. It should be noted that the interfaces between the upper and lower channels are defined as an interface which mean two flows have interaction.

| k    | n      | Max Viscosity | Min Viscosity | Temperature |
|------|--------|---------------|---------------|-------------|
| 1864 | 0.4651 | 1029          | 14            | 350°C       |

Table 1. Constants Used for Non-Newtonian Power Law Model

## Solution Considerations and Results

FLUENT uses finite volume method to solve the constitutive equations on the meshes [5]. One important consideration before beginning the simulation is the importance of inertia terms in the equations. Since most polymers have high viscosity, flow is mostly driven by pressure and the non-linear inertia terms can be neglected. The importance of inertia terms can be found by calculating the Reynolds number of the flow. In all the following simulations, the inertia terms were included. Ignoring the inertia terms helps with the convergence of the solution since these terms introduce non-linearity to the equations but it also reduces the accuracy of the results especially when Reynolds number is not small. Steady state first order pressure based implicit solver was used in all cases.

Figure 7 shows the pathlines colored by the pressure for the two cases of Newtonian constant viscosity and non-Newtonian power law viscosity models. For Newtonian flow, the value for the viscosity is chosen as the mean value of the viscosities obtained from the capillary rheometry experiment which is 220.1 Pa.s.

Figure 8 shows the velocity vectors at the lip of the cross-flow die geometry. As can be seen, velocity vectors change from -45 to 45 degrees across the thickness of the film. Velocity vectors are always tangent to the streamlines. In this study, the director of crystallines are tangent to the streamlines which means that almost the same biaxial pattern can be achieved using two cross flows instead of complicated rotating dies.

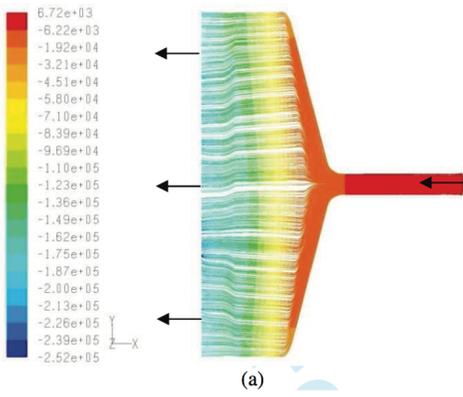


Figure 7. Pathlines colored by static pressure inside the coat-hanger geometry for the case of (a) constant viscosity and (b) power law

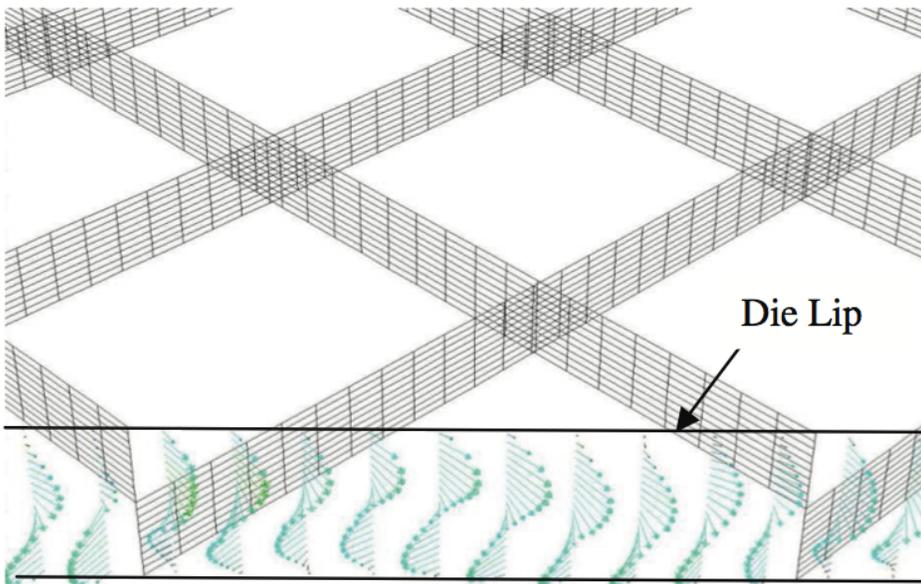


Figure 8. Velocity vectors across the thickness at the lip of the die (Newtonian model)

Figure 9 shows the result found by using the power law model based on capillary rheometer. As can be seen the change in direction of velocity vectors across the die is very small and hard to recognize except for the vectors in the vicinity of the fins.

The simulation of rheology inside the cross-flow die has been done using two different Newtonian and one non-Newtonian viscosity models. The first Newtonian model simulation is done using a constant viscosity of 0.1 Pa.s (See Table II).



Figure 9. Velocity vectors across the thickness at the lip of the die (non-Newtonian model)

It can be seen from the velocity vectors that in this case the relative importance of inertia terms forces the fluid to keep its direction until it exits the die lip. The second Newtonian model used the average value of the viscosities obtained during the capillary rheometry experiment which is 220.1 Pa.s. For this high value of viscosity, inertia forces are negligible compared to the viscous forces and flow reaches its fully developed condition in a very short distance and the only change in the velocity vectors are observed to be in vicinity of fins.

Since the maximum angle between two cross flows depends both on inertia and viscous forces, this angle is derived for several different Reynolds numbers based on the thickness of the each crossed flow channel with Newtonian model.

These results are shown in Figure 10. By increasing the Re of the flow, the relative importance of the inertia forces increases and we approach 90 degrees (from -45 to +45 degrees). On the other hand, at very low Re, flow approaches its fully developed condition and there will be no difference in the angle.

Figure 10 shows that there is a turning point at around  $Re=500$  when the angle is 80 degrees and at lower Re the maximum angle between the vector does not satisfy the desired angle between the two flows.

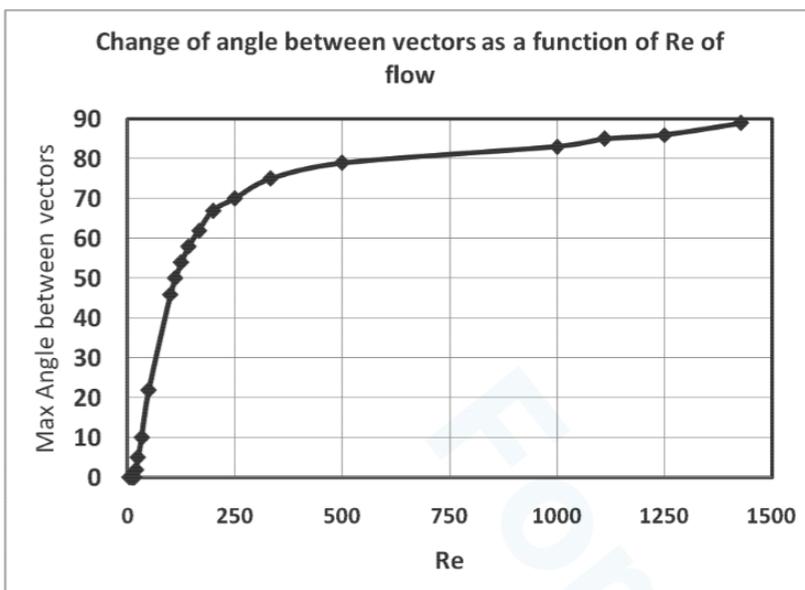


Figure 10. Change of angle between vectors as a function of Re of flow

The last simulation on the cross flow geometry is done using the actual experimental results of the capillary rheometer. In this case due to the shear thinning effect of the viscosity, the pressure drop is less than the pressure drop obtained for constant viscosity and also the velocity profile at the die lip is more flat.

## Discussion and Summary

The results of the simulations of the Newtonian and non-Newtonian material models for the coat-hanger and cross-flow die geometries are compared in Table 2. From these results it can be observed that it is possible to obtain the correct molecular orientation using this new crossflow die geometry but for the orientation of the crystalline. One solution for increasing the change in the direction of velocity vectors can be increasing the number of fins inside the die or increasing the angle between the two cross flows. It is also clear that the design of the die geometry is directly related to the material and processing characteristics such as viscosity and inlet velocities.

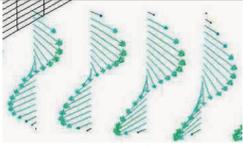
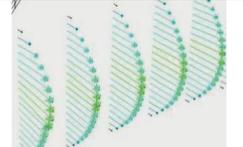
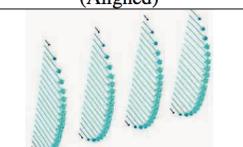
|                                       | $\Delta P$ (Pa) | Velocity profile   |
|---------------------------------------|-----------------|--|
| Newtonian coat-hanger                 | 258,720         | Aligned  |
| Power-law coat-hanger                 | 709,400         | Aligned  |
| Newtonian cross-flow (low viscosity)  | 2476            | <br>(Desired) |
| Newtonian cross-flow (high viscosity) | 4,207,000       | <br>(Aligned) |
| Power-law cross-flow                  | 3,224,000       | <br>(Aligned) |

Table 2. Summary of the simulation results

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