The Use of Computer Modelling Techniques for the Analysis of Solid and Melt Phase Polymer Processing

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Abstract: Application of computer modelling techniques to the melt and solid phase processing of polymeric materials is a challenging area of research for both academics and industrialist. In the Computer Modelling Research Centre at the IRC in Polymer Engineering, University of Bradford, we are actively researching all areas of processing from melt extrusion and injection moulding through to biaxial orientation of nanocomposite materials. The challenges such analyses pose have led to the construction of novel constitutive theories, employment of adaptive meshing for large strain applications, contact analysis with velocity dependent friction and the application of RVEs in the multiscale modelling of polymer nanocomposites. In this paper we describe some typical analyses from the Centre and an overview of the direction in which this demanding area of research is moving.

Keywords: Composites, Constitutive Model, Polymer.

1. Introduction

The Computer Modelling Research Centre at the IRC in Polymer Engineering, University of Bradford has for many years been involved in the analysis of polymer processing in both the melt and solid phases. Such analyses are of significant academic and industrial interests ranging from material description through novel constitutive theories, the definition of polymer – polymer and polymer – metal interfaces, composite materials containing soft and hard particulates with length scales down to the nanometer to the more practical analyses such as injection moulding of polyethylene, extrusion profile studies and mechanical performance of manufactured components. Within this paper we give some examples of current analyses.

2. Melt Processing and Final Product Performance of Composite Materials

For many years thermoplastics have been extensively used in a wide variety of applications because they are relatively cheap, inert and are easy to manufacture into component shapes, particularly by injection moulding. However, commodity polymers, such as polypropylene and Nylon, have relatively poor mechanical properties, which limits their use to non-structural applications at modest temperatures. Short glass fibre reinforcement is well established as a means of significantly improving on mechanical performance without compromising processability, and many glass filled polymer grades and products are commercially available. Apart from the fibre and matrix properties, the mechanical properties of the final component are crucially dependent on the fibre orientation distribution (FOD) developed during the process, as well as other aspects such as the resulting fibre aspect ratio and the fibre/matrix interface. As a fibre filled component's mechanical properties are significantly affected by the FOD there is a commercial desire to predict accurately this property during the design phase. The accuracy of this prediction is however constrained by limitations within mould filling analysis software, particularly in regions of three-dimensional flow such as ribbed or thick sections.

2.1 Melt Processing Analysis

At ANTEC 2004 (Caton-Rose, 2004) we presented work on fibre orientation predictions of an injection moulded ribbed plate using a midplane geometry definition within Moldflow MPI 4.1. By tailoring the thickness moment of interaction (D_z) and fibre interaction coefficient (c_i) we demonstrated that analysis software was capable of predicting FOD within this part to a reasonable degree of accuracy. It was however noted that the fibre orientation theory based on the work of Jeffery (Jeffrey, 1922) and modified by Folgar and Tucker (Folgar, 1987) had the tendency to over-predict the shell layer orientation or produce a thinner core region than seen experimentally. Tucker et al (Tucker, 2004) have further investigated this in terms of short flow paths and thicker section parts and suggested a further modification to the current theory.

The ribbed plate specimen, Figure 1, has been thoroughly investigated using an image analysis system based at Leeds University to evaluate fibre orientation at given locations across and along the specimen.

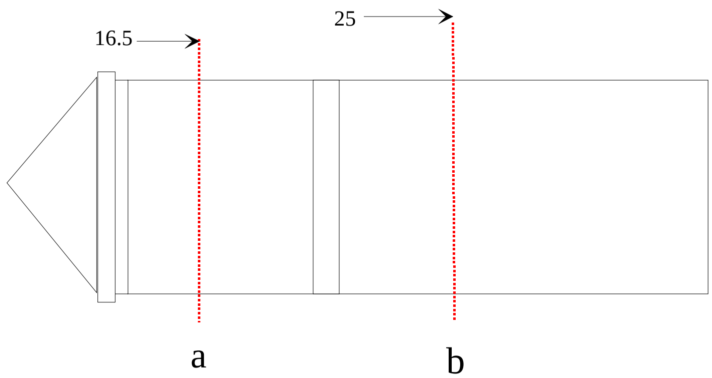

Figure 1. Ribbed plate specimen

For the purposes of model validation two locations either side of a transverse rib section were selected, 16 mm from the gate and 25 mm behind the rib. Fibre orientation distributions (FOD) from Moldflow MPI 5.0 predictions and measured values at location A are shown in Figure 2.

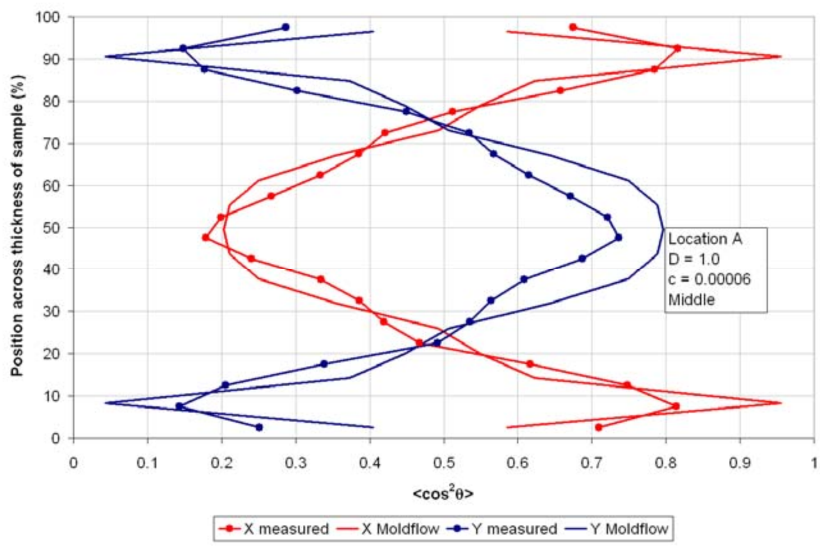

Figure 2. Fibre orientation measurement and predictions for location A

The orientations shown above were used together with micromechanical models to define the bending modulus of the composite material. The modelling approach has previously been validated using finite element techniques in collaboration with the group of Andrei Gusev at ETH Zurich, and combines the work of Qui and Weng to determine properties of the composite unit together with an aggregate model to predict the effects of disorientation. For simplicity the measured orientations at each location were averaged across the sample width and divided into 11 layers through the sample thickness. In contrast the Moldflow predicted orientations used the direct ABAQUS material model translator to convert elemental orientation into composite elastic material properties.

2.2 Solid Phase Deformation Validation

The three point bend tests used 10 mm wide sections taken from locations A and B and subjected to loading conditions shown in Figure 3.

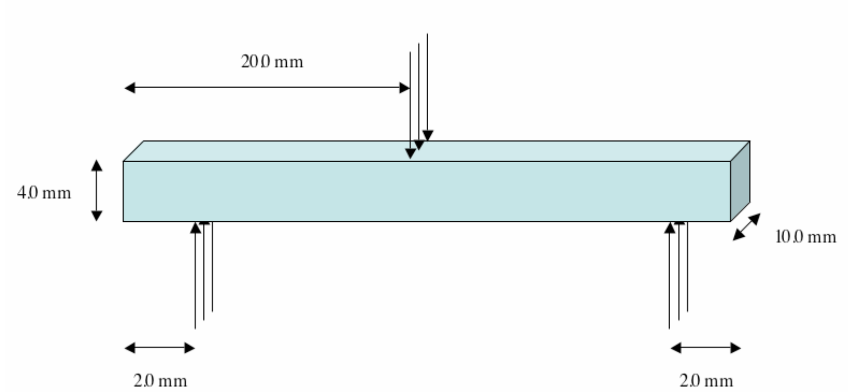

Figure 3. Three-point bending loading conditions

The resultant reaction force along the loading axis for both experimental testing and finite element analysis is shown in Table 1 for an imposed displacement of 1 mm.

Table 1. Three-point bending reaction forces along the loading axis

	Experimental	**FE model using Moldflow FOD**	**FE model using measured FOD**
Location A	162 N	437 N	193 N
Location B	127 N	361 N	155 N

Within Table 1 it can be seen that the measured FOD used within the micromechanical model was capable of predicting the response of the ribbed specimen to bending loads. The significantly increased loads shown by the Moldflow FOD model is however a cause for concern. On further investigation it was found that a large portion of this error could be attributed to the matrix material properties applied during model translation between Moldflow and ABAQUS. Material data from the manufacturer presented two values for the tensile modulus at different environmental conditions. Firstly, at room temperature and zero humidity, i.e. tested under dry ‘as moulded’ conditions, the tensile modulus of the matrix material was found to be 2800 MPa. The second value was also at room temperature but with a humidity of 50% and resulted in a tensile modulus of 1100 MPa, nearly three times that of the ‘as moulded’ test. The ‘as moulded’ value was used during the material model construction within the Moldflow – ABAQUS translator. Table 2 shows the effect of using the lower tensile modulus on the three-point bend analysis.

Table 2. Three-point bending reaction forces along the loading axis with adjusted matrix material properties

	Experimental	FE model using Moldflow FOD	FE model using adjusted material data	FE model using measured FOD
Location A	162 N	437 N	330 N	193 N
Location B	127 N	361 N	266 N	155 N

The adjusted matrix material property application within the Moldflow – ABAQUS translator showed a reaction force reduction of ~ 25%. The remaining over prediction of specimen response is probably due to the inaccuracy of the FOD predictions, due to shortcomings within the Folgar-Tucker model discussed previously.

3. Novel Constitutive Theories

Eyring devised his theory of plastic flow in the 1930s. In this physically-based model of thermally activated flow, plastic strain rate is driven by the stress in a highly nonlinear manner, such that the strain rate essentially increases exponentially with stress. An Eyring process may be viewed as a nonlinear dashpot. When it is combined in series with an elastic element, the resulting system exhibits phenomena that are associated with polymers. One such phenomenon is creep, where it has been shown that stress and temperature dependence of creep rate can be accurately modeled by Eyring processes. Stress relaxation in polymers may also be modeled by this system. When the Eyring/elastic system is subject to a constant rate of strain, the stress reaches a maximum value that depends on the applied strain rate; thus, the rate-dependent yield behavior of polymers is captured.

The Eyring analysis predicts an Arrhenius-type behavior of yield stress as a function of logarithm of strain rate. This was explored in detail for glassy polymer (Bauwens-Crowet 1972), who concluded that a material model featuring a single Eyring process gave an excellent representation of uniaxial tensile and compressive experimental data. For semi-crystalline polymer, more complex models have proved necessary. Working on the tensile creep behavior of polyethylene fibres, (Wilding, 1978) introduced a material model featuring a pair of Eyring processes, in which one of the processes became active at high stresses.

Tensile yield in polymers is often accompanied by an unstable deformation in the form of a neck or shear band. The existence or otherwise of an instability depends on the strain-rate sensitivity of the material, as is well understood (Sweeney, 1999). Since the Eyring model inherently includes strain rate sensitivity, and also provides a model of yield, it promises a means of predicting unstable behavior. In practice, this can be achieved by including the Eyring process in a constitutive equation that forms the basis of a numerical model of the tensile specimen. A general three-dimensional form of the Eyring process, including a flow rule, is then necessary.

Aside from the criterion that governs the existence or otherwise of an instability, there is the question of what geometric form it will take. In uniaxial stretching of plane tensile specimens, there are two possibilities: a neck, symmetric about the specimen axis; and a shear band, an area of localisation at an oblique angle to the axis. In this paper, we shall create finite element models of both kinds of instability, based on the same form of constitutive equation. The models will be discussed in relation to experimental results on shearbanding polycarbonate and necking polyethylene.

3.1 Experimental Validation

We have performed tensile tests on dumb-bell specimens of both polycarbonate and polyethylene, at 130 and 100°C respectively, using the same constant testing speed and specimen geometry. The polycarbonate deforms with a shear band, whereas the polyethylene deforms with an approximately symmetric neck. Table 3 shows the data used in the finite element model for the polycarbonate material. Some realistic levels of asymmetry are introduced into the finite element analysis, so that the shear band forms at the observed location at one end of the specimen gauge length, shown in Figure 4.

Table 3. Model parameters for polycarbonate

C / MPa	A / s^{-1}	V_s / MPa^{-1}	V_p / MPa^{-1}
385	2.0×10^{-18}	2.343	0.141

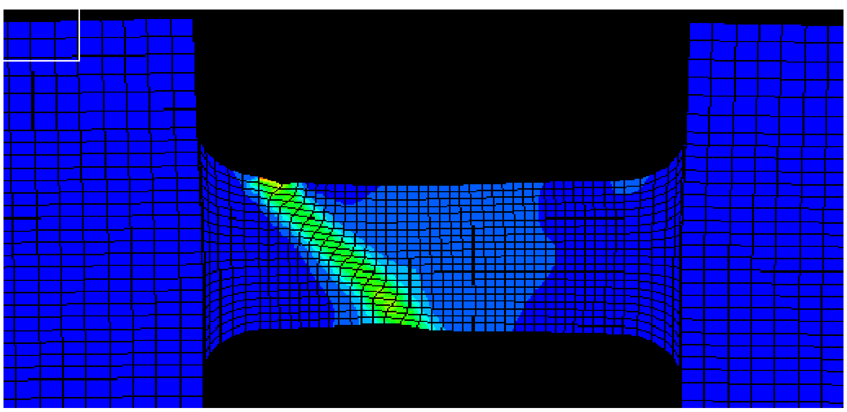

Figure 4. Contours of maximum principal strain – Table 1 properties

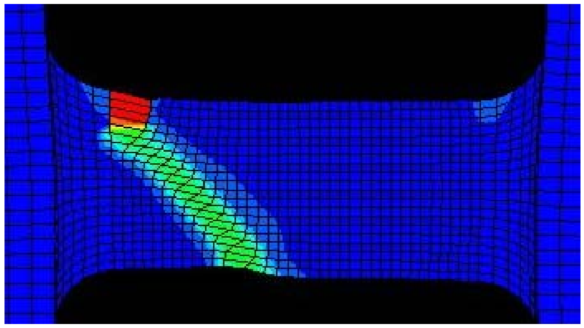

Figure 5. C = 170 MPa, otherwise as Table 1.

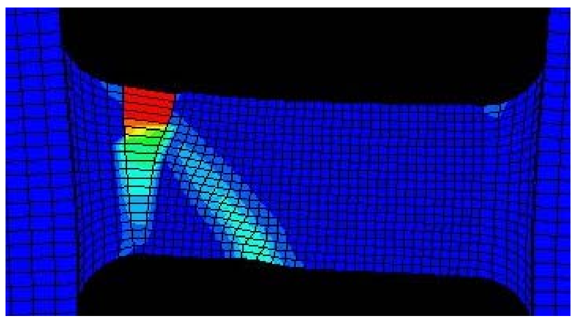

Figure 6. C = 165 MPa, otherwise as Table 1.

Polyethylene is much less stiff than polycarbonate, so in our model the value of the network stiffness C would be lower. Figures 4 to 6 show the transition from shear banding to necking, as C is reduced and as a greater proportion of the strain is taken up by the network. The plastic component of strain is controlled by the Levy-Mises flow rule, which is compatible with shearbanding, whereas the Gaussian network includes no such mechanism. Therefore, the geometry of the instability, being governed by the division of the strain into elastic and plastic components, tends towards shearbanding when the plastic strain is dominant and to a symmetric neck otherwise. The value of C for polyethylene is in fact much lower than in Figure 6, but strain fields follow a consistent trend, and realistic parameters do give predictions of necking.

4. Multiscale Analysis Using Statistical RVEs

Dramatic improvements in mechanical performance have been observed in polymer/clay nanocomposites, in which silicate platelets of nanoscale thickness are dispersed within a polymer matrix. The benefits over conventional composites derive primarily from the exceptionally large particle surface area achievable with the addition of only a small amount of particles (less than 5% by weight). In order to deal with the multiscale nature of the problem, a parameter-passing approach has been adopted, in which appropriate parameters determined from smaller-scale simulations act as the basis for larger-scale models. On the atomistic level, the properties of the clay platelets and the interface region are determined from molecular statics calculations using environmentally-dependant interatomic potentials. On the micron lengthscale, the nanocomposite is modelled using a statistical Representative Volume Element approach, in which a statistical ensemble of cells are generated according to given distributions in platelet shape, position and orientation (extracted from a large number of TEM micrographs). The effective mechanical properties of the composite system are determined using Finite Element Analysis for large strains, and passed up the lengthscale to be incorporated in models to predict processing behavior and final part properties. In relating the details at the molecular level with the material at the macroscopic level, it becomes possible to design materials to suit our needs.

4.1 RVE Description

Each RVE gives a statistical sample of the composite material (platelet and polymer) that simulates an infinite medium. This is achieved by the generation of a number of ensembles which contain random distributions of platelets within a polymer matrix, Figure 7 shows a typical arrangement. In addition to random platelet orientation the length, shape and stiffness of the filler particles can be adjusted as well as volume fraction.

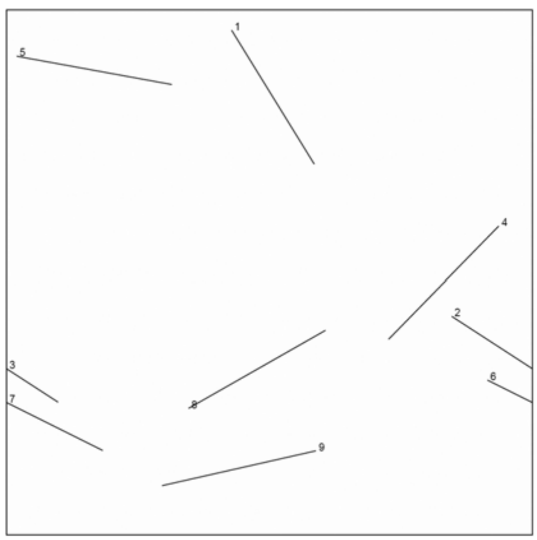

Figure 7. Typical RVE platelet geometry

Platelets of Smectic Silicate are defined by linear elastic properties whilst the polymer (polypropylene) uses a large strain, non-linear strain-rate dependent constitutive theory based on the work described in section 4. Any given ensemble can be meshed using 6-noded (quadratic triangular) elements to define the polymer and lamina (3-noded quadratic) elements for the platelets, or 6-noded quadratic triangular elements for the complete RVE, Figure 8.

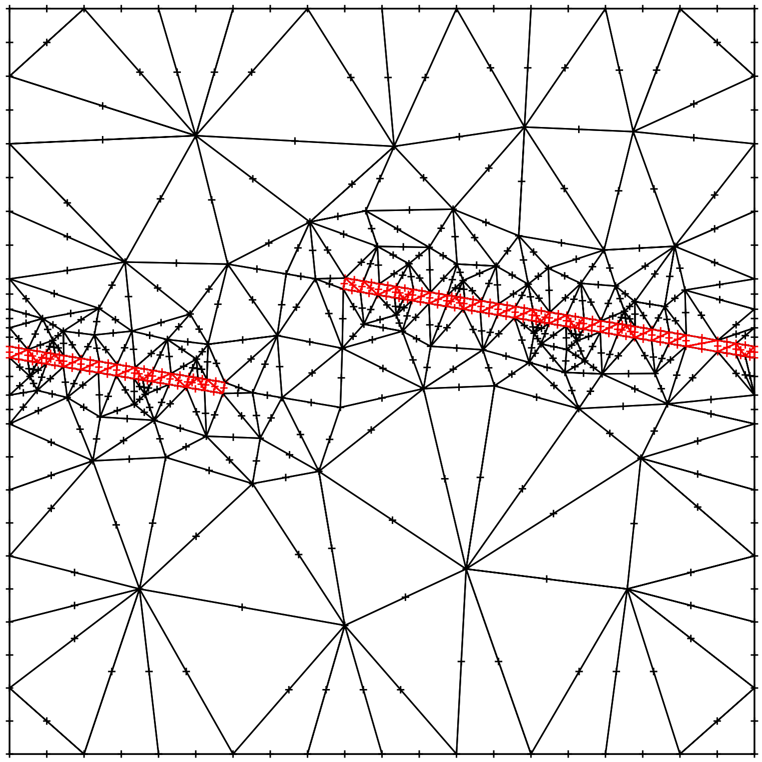

Figure 8. Meshed RVE

Application of various strain states such as uniaxial (x or y direction) and pure shear to the RVEs allows the determination of effective material properties of the composite. It should be noted that during these strain applications periodic boundary conditions are defined to ensure profiles are identical on opposing edges.

4.2 Application of Adaptive Meshing

Due to the large strain application adaptive meshing is required to improve stability and the extend solution window. Using ALE (Arbitary Lagrangian Eulerian) adaptive meshing built into ABAQUS explicit and converting the polymer material description into an elastic plastic formulation the overall solution of the RVE deformation has been improved. Figure 9 shows an example without adaptive meshing with the adaptive meshed case shown in Figure 10.

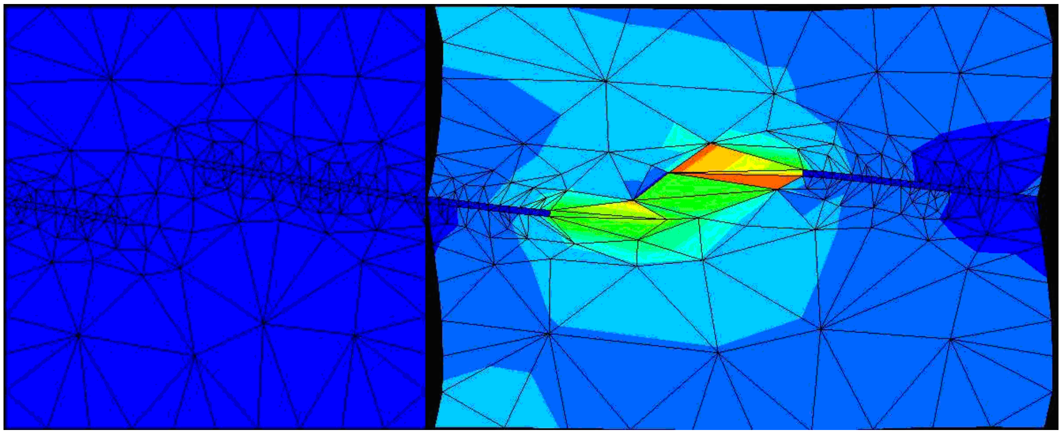

Figure 9. RVE deformation without adaptive meshing

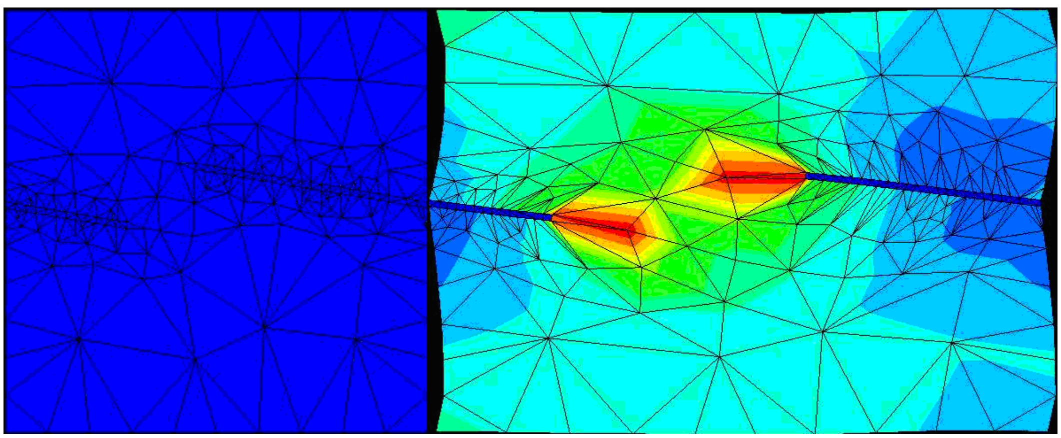

Figure 10. RVE deformation with adaptive meshing

5. Conclusions and Future Studies

Computer modelling techniques are an invaluable tool in the development of new processing schemes for polymeric materials. However dependent on the phase in which the process occurs, the specific material type and or the presence of additive materials each analysis should be considered carefully. Work within the Computer Modelling Centre in the IRC in Polymer Engineering, University of Bradford will continue to address issue such as melt to solid phase transitions in polymer processing, polymer surface interactions, micro and nano scale filler particles and their effect on macro-scale mechanical performance and constitutive theories to describe polymeric materials.

6. References

1. Bauwens-Crowet, C, and Bauwens, J-C, “The temperature dependence of yield of polycarbonate in uniaxial compression and tensile tests” J. Mat. Sci. **7**, 176 - 183., 1972
2. Caton-Rose, P., Coates, P.D., Whiteside, B., Hine, P., Duckett, A., Jittham, P., Chapman, C. and Smith, G., “Measurement and Prediction of Fibre Orientation Within a Three-Dimensional Flow”, SPE ANTEC Proceedings, Chicago, 2004
3. Folgar, F. and Tucker, C. L., J. Rheology, 31:751-784, 1987
4. Jeffery, G. B., Proc. Roy. Soc., A102:161-179, 1922
5. Sweeney, J. , Shirataki, H., Unwin, A.P. and Ward, I.M., “Application of a necking criterion to PET fibres in tension”, Journal of Applied Polymer Science **74** 3331-3341, 1999
6. Tucker, C.L. and Wang, J., Improved Fibre Orientation Predictions for Injection Molded Composites, PPS-20 Proceedings, Akron, 2004
7. Wilding, M.A. and Ward, I.M., “Tensile creep and recovery in ultra-high modulus linear polyethylenes” Polymer, **19,** 969 – 976., 1978