SAND2008-2039C

FINITE ELEMENT ANALYSIS OF MULTILAYER COEXTRUSION

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Multilayer coextrusion has become a popular commercial process for producing complex polymeric products from soda bottles to reflective coatings, and even ceramic structures where a sintering step is added to burn out the polymer from the ceramic paste. A numerical model of a multilayer coextrusion process is developed based on a finite element discretization and an arbitrary-Lagrangian-Eulerian (ALE) moving mesh implementation to understand the moving boundary problem associated with the polymer-polymer interface. The goal of this work is to have a numerical capability suitable for optimizing and troubleshooting the coextrusion process and circumventing flow instabilities such as ribbing, barring and variable layer thickness. Though these instabilities can be both viscous and elastic in nature, for this work a Newtonian description of the fluid is used. Models of varying degrees of complexity are investigated including stability analysis and direct finite element free surface approaches. The focus of this work will be a full 3D finite element free surface model that was built to examine a die design, which would allow for offset between the coextruded layers. In addition, particle tracking in the multiplication region is used to determine approximate layer thickness for a single fluid in steady flow.

Introduction

Multilayered coextrusion combines different polymers into a layered structure, which produces a composite material that has the union of properties from each of the material subcomponents. In this way, materials can be made that have much improved properties over single polymer extrudate. Current applications of multilayered coextrusion include packaging, protective coatings, and barriers [1]. For instance, these can be used for products such as soft drink bottles, which use an oxygen impermeable polymer adhered to a carbon dioxide impermeable polymer. This technique is also being explored for new technologies such as energy storage devices, sensors, displays, and membranes [2]. In our work, we are investigating multilayer coextrusion as a means to produce many-layered, nanoparticle-filled polymer structures for use as sensors or energy storage devices. These devices must have an offset to their layered structure such that one polymer essentially encapsulates the other polymer. Creating such an offset is not currently done commercially, and is thus a research issue.

To aid the manufacturing process, we are developing computational models of coextrusion to look at potential die design to create an offset structure, and issues such as layer non-uniformities and instabilities. Instabilities due to property differences between layers such as viscosity and elasticity and flow geometries can plague coextrusion processes [3, 4]. Models of varying degrees of complexity are investigated, though we limit ourselves to Newtonian rheology. We have undertaken linear stability analysis to understand possible ribbing and barring instabilities, though due to space limitations, these will not be discussed here. In this paper, we focus on the full 3D model that was built to examine a die design that would allow for offset between the coextruded layers and produce the necessary encapsulated phase for our project. In addition, particle tracking is investigated as an approximate method to determine layer thickness from a steady, single-fluid simulation.

Experimental

A schematic of our coextruder is given below in figure 1. Our inflow feed-block creates an eight-layer structure by having four inflow streams of each polymer. Gear pumps are added between the extruders and the feed-block to produce a uniform flow rate unlike the pulsed flow that results from the extrusion screw. Subsequent layers are created by using multiplication dies, which split the flow into two streams and restack to create more layers. The increase in the number of layers within the same cross-section decreases the thickness of each layer. After the splitter region, quieting regions are inserted to damp out any instabilities caused by the multiplication. Heaters with temperature controllers are used throughout the processing equipment and are critical to keeping the material flowing and relatively isothermal. Following these sections the layered structure will pass through a sheeting die onto a chilled roller, resulting in an approximately 1 mm tape.

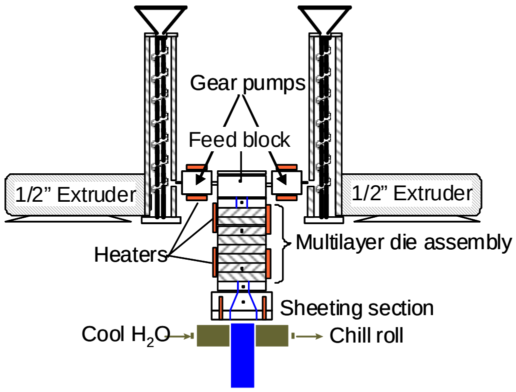

Figure 1: Schematic of Multilayer Coextruder

Thus far we have extruded 8, 16 and 32 layer structures and initially focused on a polyethylene/polystyrene system, though for future work we will investigate filled-systems of other more appropriate polymers. An example film is given in figure 2.

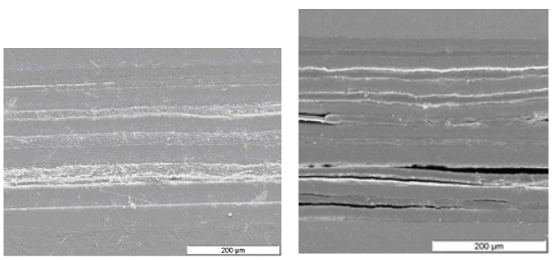

Figure 2: SEM images of an eight (left) and sixteen (right) layered structure produced by coextrusion of polyethylene and polystyrene. Delamination on the sixteen layer structure is from potting in epoxy and polishing for SEM, initial extruded layers are not delaminated.

Numerical Method

Equation of Motion

Here we solve the Navier Stokes equations for 3D fluid flow: conservation of momentum and mass for a Newtonian fluid of constant density.

$$\rho \frac{Du}{Dt} = \mu \nabla^2 u - \nabla p + \rho g$$
$$\nabla \cdot v = 0$$

The free surfaces associated with polymer-polymer and polymer-air interfaces are handled with a pseudo-solid mesh motion algorithm, which moves the mesh according to the material interfaces at these boundaries and elsewhere moves as a nonlinear elastic solid. For details of our mesh motion algorithm and implementation please see Cairncross et al., 2000 and Baer et al., 2000 [5,6].

Finite Element Implementation

The equations of motion and the mesh equations are solved with the finite element method, using GOMA, a computer code designed to solve free and moving boundary problems [7]. The equations are solved in a fully-coupled, Newton-Raphson manner. The Navier-Stokes equations were stabilized with the Dohrman-Bochev pressure stabilized pressure-projection [8] to allow for equal order, bilinear, interpolation of all variables; velocity, pressure and mesh. As these are large, 3D problems, direct solution methods for matrix inversion are precluded. Instead, we use Krylov-based iterative solver and preconditioning. For this work, we use an ILUT preconditioner with up to three levels of fill and a GMRES solve. The problem is solved in a steady manner, ignoring time derivative, as a stable coextrusion process must run at steady-state. Damping of 0.05 is necessary in order to achieve a convergent solution. The problem must be either rescaled or nondimensionalized in order to get good convergence of the iterative solvers.

For boundary conditions on the inlet and outlet planes, constant pressure conditions are applied. The inlet pressure on outer fluid is set at both inlet ports to be 2000 dyn/cm^2 while the inlet pressure on the inner fluid entry plane is set to 400,000 dyn/cm^2. At the outlet, the pressure is set on both fluid layers to be -440,000 dyn/cm^2. No slip is applied on all solid surfaces and the kinematic condition is applied on interfacial surfaces along with surface tension [5].

Results and Discussion

3D Free Surface Flow of Offset Die

The initial undeformed mesh to investigate the offset die to produce an encapsulated polymer is given in figure 3. Here we have simplified the coextruder down to a three-layer process. The blue inlet regions flow into the yellow regions, and are all the same material. The turquoise inlet flows into the red material. For this scoping simulation, the viscosities are set arbitrarily to be 100cp in the encapsulating polymer and 200cp for the inner fluid. The densities are assumed to be 1g/cm^3 for both polymers.

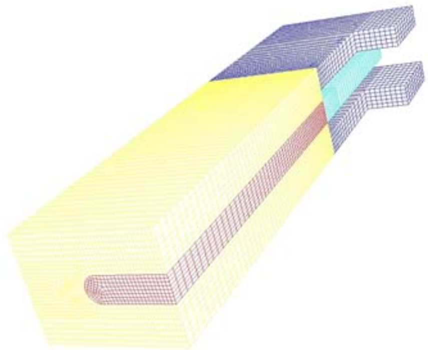

Figure 3: Undeformed mesh for offset die

Results for this problem are given in figure 4. Here we look at parameter continuation in the inflow pressure of the top blue material. From figure 4, we see an initially well-behaved free surface at high inlet pressure. However, as we decrease the pressure the yellow materials begins to swell and eventually wet the die lip of the blue material (figure 5). This can lead to extreme processing difficulties from inhomogeneous layer thickness to actual choking off of the blue fluid. This makes it clear how important it is to carefully meter and balance the flow rates in order to avoid processing issues.

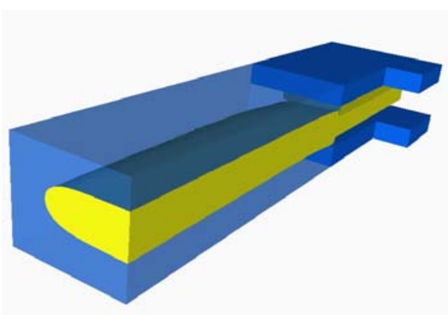

Figure 4: Free surface shape for a low inlet pressure for the yellow polymer.

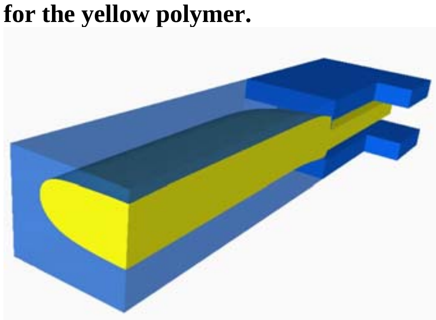

Figure 5: Free surface shape for a higher pressure where the yellow fluid is starting to wet the blue die.

Particle Tracking on Fixed Mesh Solution to Determine Layer Thickness

3D free surface flows can give us a great deal of information about the shape of the extrudate and how materials properties and processing parameters effect final film thickness and distribution. However, these calculations are very difficult to run requiring expert users. They also require a large amount of CPU time due to inclusion of mesh equation in the solution matrix. Thus, based on work by Kim et al, 2008 on recent coextrusion [9], we were intrigued by the idea of using particle tracking to estimate the layer thickness from a steady single-phase simulation. Here, we have undertaken such a simulation in a simplified geometry similar to our current coextruder, but containing a multiplication region to take the inflow two-layer structure to a four-layered one. The mesh for this geometry is given in figure 6.

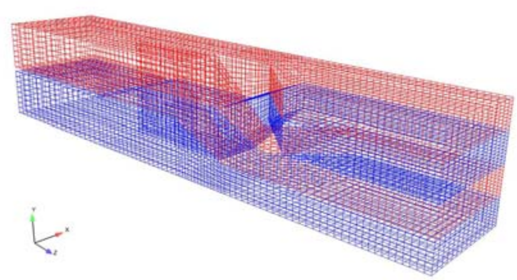

Figure 6: Coextruder mesh for initial multiplication creating a four-layered structure by splitting and restacking a two-layered one.

The drawbacks to this type of simulation are that all materials must have the same properties and there can be no interfacial tension or phase interface, which are all obvious gross simplifications. In contrast, these steady, single phase solutions are very easy to run and agile, but approximate, results for die design could be useful.

Results from this analysis where a closed-flow simulation is run and particles are tracked to indicate layer thickness are given in figure 7 (side view) and figure 8 (outflow plane).

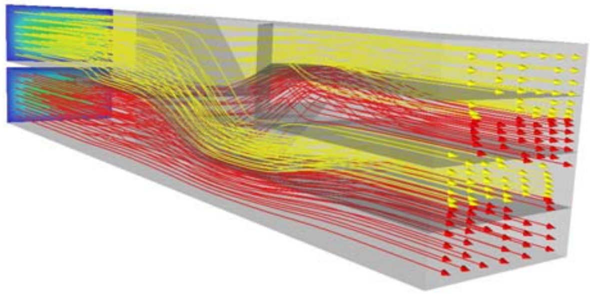

Figure 7: Particle tracking from inflow to exit through the multiplication die showing approximate layer thickness at outflow (side view).

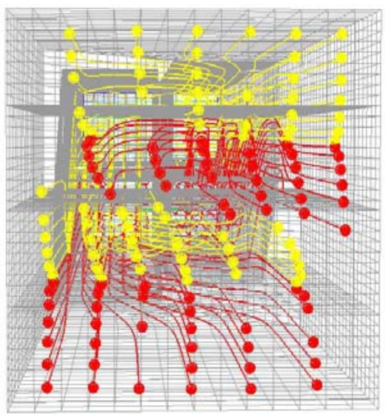

Figure 8: Particle tracking from inflow to exit through the multiplication die showing approximate layer thickness at outflow (outflow view).

From these figures, we can see that layer nonuniformity and potential encapsulation of the red phase by the yellow phase can be a problem even for polymers with identical properties, just due to the geometry of the flow.

Conclusions and Future Work

In this paper, we have presented a numerical study of multilayer coextrusion. We have developed an offset die design that can be used to create coextruded materials where one phase encapsulates the other for applications such as energy storage and sensors. We have investigated this design using a full 3D free-surface capability to determine the shape of the extrudate at outflow. This analysis has shown that careful metering of the polymers is critical to maintaining layer homogeneity. We have also examined an approximate approach where the material is assumed to be one phase and a steady problem is then analyzed with particle tracking to determine layer thickness.

In the near term, we plan on comparing the full 3D free-surface analysis to the particle tracking method to determine its range of applicability. We will also complete and document our 2D stability analysis. In the more distant future, we hope to look at non-Newtonian effects such as particle migration and viscoelasticity.

Acknowledgements

Thanks go to Mary White and Joel Lash for their complex meshing work, to Scott Spangler for his SEM images, and to Pat Notz for help with the visualization. Phil Cole and his graduate work were the inspiration for the project.

Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under Contract DE-AC04-94AL85000.

References

1. L. Lamnawar; A. Maazouz in *10th ESAFORM Conference on Material Forming*, edited E. Cueto and F. Chinesta, American Institute of Physics, 2007.
2. A.T. Crumm; J.W. Halloran *J. Mater. Sci*, 2007, *42*, 1336.
3. J. Dooley; L. Rudolf *PFFC online.com*, Jan. 2003, 73.
4. A.J. Rincon; A.N. Hrymak; J. Vlachopoulos *Int. J. Numer. Meth. Fluids*, 1998, *28*, 1159.
5. R.A. Cairncross; P.R. Schunk; T.A. Baer; R.R. Rao; P.A. Sackinger *Int. J. Numer. Meth. Fluids*, 2000, *33*, 375.
6. T.A. Baer; R.A. Cairncross; P.R. Schunk; R.R. Rao; P.A. Sackinger *Int. J. Numer. Meth. Fluids*, 2000, *33*, 405.
7. P.R. Schunk; P.A. Sackinger; R.R. Rao; K.S. Chen; T.A. Baer; D.A. Labreche; A.C. Sun; M.M. Hopkins; S.R. Subia; H.K. Moffat; R.B. Secor; R.A. Roach; E.D. Wilkes; D.R. Noble; P.L. Hopkins; P.K. Notz; "GOMA 5.0 – A Full-Newton Finite Element Program for Free and Moving Boundary Problems with Coupled Fluid/Solid Momentum, Energy, Mass, and Chemical Species Transport: User's Guide," Sandia National Laboratories, SAND2006-5375/1, October, 2006.
8. C.R Dohrmann; P.B. Bochev *Int. J. Numer. Meth. Fluids*, 2004, *46*, 183.
9. 9. S.J. Kim; K.H. Lim; P.C. Lee; and C. Macosko, presented at "The Society of Rheology 79th Annual Meeting – Salt lake City," 2007.