

Letter to the Editor: Steep wave fronts on extrudates of polymer melts and solutions

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It seems to us that the shape of the extrudate of polymers and polymer melts is very much like the wavy shapes one sees in core-annular flows of heavy oils in water. These flows are lubricated by the water and can be said to give rise to slip. A wave on the oil must steepen at its front and flatten at its rear because the pressure is larger in the front and lower in the back. Photographs of the ubiquitous asymmetric waves which are shown as a cartoon in Fig. 1 can be found in many places; for example, Oliemans and Ooms [1986], Feng, Huang and Joseph [1995].

The high and low pressures which are generated in the water as a wave on the oil pushes forward are much more intense when the gap is small; in a slipper bearing the pressure maximum is proportional to the reciprocal of the square of gap size. In journal bearings the low pressure on the back side is severe enough to produce cavitation even under rather mild conditions.

It is widely agreed that many polymer melts and solutions slip along the walls of capillary tubes (Benbow and Lamb [1963], Schreiber *et al.* [1966], Schowalter [1988], Hatzikiriakos and Dealy [1991], Chen and Joseph [1992], Migler *et al.* [1993], and many others). Despite the agreements, there remains substantial disagreement about why and how it slips. Moreover, the relation between wall slip and extrudate defects is even more controversial.

The word "slip" is rather general and is subject to different interpretations, all of which would appear to imply lubrication by different mechanisms. If a polymer melt or solution slips at a wall, it must slip on something. Even solids in dry friction are lubricated nearly everywhere by a thin fluid or air layer. Slip may arise as a failure of adhesion in which the bond between two materials, the polymer and the wall, is broken and the substance between which slipping takes place must give rise to lubrication, as in core-annular flow. The substance between sticks to the extrudate and to the wall. A substance between is also required by cohesive fracture in which bits of the fractured polymers remain on the wall. Another idea is that a thin layer of polymer softens as a consequence of instability associated with a nonmonotone stress-strain relationship in the constitutive equation. This also gives rise to a lubricating layer of less viscous material at the wall on which the polymer may slip. It is of interest to inquire what the substance is between and what are the dynamic effects of the lubricating layer.

Assuming, for the moment, the presence of a lubricating layer and a dynamic effect

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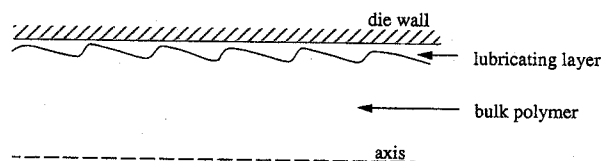


FIG. 1. A cartoon of the proposed wave shape (enlarged) in the interface between the bulk polymer and the thin lubricating layer in a die.

similar to the one which steepens wave fronts in core-annular flows, exacerbated by the surpassingly small gaps for slipping polymers, we expect that every wavy extrudate will have a steep wave front and a flatter rear. This idea seems to work well in all of the cases in which the direction of flow could be ascertained from photographs or the associated text. In fact, researchers in this subject do not say much about the direction of flow, and they do not look at the asymmetry of the wave as a window for understanding. We were able to know the direction of the extrudate only in the cases in which the die exit is shown. In all these cases the steep part of the wave advances as can be seen in Figs. 6(d), 6(e), 7(b), 7(c), 7(d), 8(f), 8(g), and 9(e) of Piau *et al.* [1990], Figs. 4(g), 5(d), 5(e), and 11(a) through 11(e) of Kissi and Piau [1990], and Fig. 4(b) of Piau *et al.* [1995]. Unpublished photographs of S. Kurtz and W. Schowalter (personal communications) are also consistent with the advances of the steep part. In other cases [Figs. 1.3, 1.4, 10.1, 10.2, and 10.3 of Benbow and Lamb (1963), Fig. 1(b) of DeSmedt and Nam (1987), Figs. 3(a), 3(b), and 8 of Kalika and Denn (1987), and Figs. 1(a) and 1(b) of Denn (1990)], we do not know which way the extrudate is going and we guess that it is moving so that the steep front of the wave advances. If we have guessed right we would judge that our idea is creditable and deserves further study.

The relation between wall slip and extrudate distortion is not clear. Loss of adhesion may not occur uniformly but at different spots; this could lead to the messy extrudates seen in photographs. A more organized loss of adhesion occurs as "stick-slip;" the slipping occurs on a lubricating layer and when the lubricated part comes out of the die it spurts. Even in the messy cases, the steep part of the wave appears to advance.

The hypothesis of this letter is that the dynamics of lubricating layers in slipping polymer melts and solutions is such as to produce an asymmetric wavy extrudate in which the steep side of the wave advances. A search of the literature reveals that this hypothesis is verified in all cases in which the direction of flow could be ascertained from data given in the paper and the direction of flow is here predicted from photographs of the wave shape in the cases in which this direction is not specified. The dynamics which give rise to the steep wave fronts in the extrudates is not well understood but may share features like those that produce the same type of shocklike waves on heavy oil in water in which the steep part of the wave advances. Perhaps the asymmetric form of waves on extrudates in slipping flow are as universal as they are in core-annular flow.

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Editor's Note: The letter to the editor by Joseph and Liu presents a hypothesis about the mechanism of extrusion instabilities. I invited authors whose work is referenced to respond if they wished. Stuart Kurtz's response, printed here, is the most extensive and incorporates thoughts expressed more briefly by others.

Comment on: "Letter to the Editor: Steep wave fronts on extrudates of polymer melts and solutions," by D. D. Joseph and Y. Joe Liu

One would think that after 40 or so years of studying sharkskin melt fracture and gross melt fracture in polymer melts and solutions that there would be at least some consensus among rheologists, polymer engineers, theoreticians and others on what is happening. That such consensus on mechanism does not exist does not preclude a general agreement of observations. Thus, old or new theories need to address what is known experimentally, both in terms of what fits and what does not fit.

Joseph and Liu propose a mechanism that is intended to explain the shape of melt fractured surfaces. This is something that has not been adequately addressed in the literature. Because it is another test of how good some theories are and may even differentiate theories, it is an important question. There still is a debate on whether the steep front occurs in all cases of melt fracture or whether some fractures may be symmetrical or reversed. Even if the surfaces are of the shape envisioned in the Joseph/Liu model, showing a steep wave front, does their proposed model fit other observations, models, and understandings?

In the case of Cogswell's model of fracture, whereby the shape of the surface is determined in the stretching and tearing process at the die exit, the Joseph/Liu model

seems to ignore this type of fracture. High speed video of the deformation process near the die exit might answer this question. Some of our studies and that of others suggest that there is a tearing process at the die exit associated with sharkskin melt fracture.

Observations that depth of melt fracture increases with die diameter at constant shear stress seem also to contradict a model of lubricated thin fluid or air layer. The Joseph/Liu analogy with the slipper bearing of a pressure maximum proportional to the reciprocal square of the gap size does not seem to fit the die diameter result.

Various studies on the interaction of polymer melts and wall materials by Dealy, Hatzikiriakos, Giacomini, Ramamurthy, Denn, and others show no evidence of a nearly inviscid layer adjacent to the wall, a necessary component of the Joseph/Liu model.

There are other observations and experiments that do not fit with the Joseph/Liu model. That is not to say that interactions of local pressure waves and slip processes going on inside the die should be ignored. On the contrary, perhaps the best way to obtain a consensus is to begin connecting the observed effects such as frequency, depth and shape of fractured surfaces with models of slip, bulk rheology and molecular rheology in a convincing manner. I think we have enough information to begin the process of building an integrated understanding.

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