Journal of Non-Newtonian Fluid Mechanics 195 (2013) 67–76

Contents lists available at SciVerse ScienceDirect

Journal of Non-Newtonian Fluid Mechanics

journal homepage: http://www.elsevier.com/locate/jnnfm

Numerical investigations of polyester coextrusion instabilities

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ARTICLE INFO

Article history:
Received 4 July 2012
Received in revised form 7 December 2012
Accepted 11 December 2012
Available online 30 January 2013

Keywords:
Coextrusion
Interface instability
Rheology
Spatio-temporal analysis

ABSTRACT

Interfacial instabilities occurring in the coextrusion process of molten polymers have been widely studied. The theoretical work based on the stability of 2D Poiseuille multilayer flows (invariant along the flow motion) have pointed out the convective nature of this instability (it is either amplified or damped in the flow direction). This behavior has been confirmed by experimental observations. As this instability is purely elastic (the Reynold number in coextrusion process is very small), it is necessary to introduce a viscoelastic constitutive equation for the polymers. Comparisons between experimental observations and theoretical approaches based on the Giesekus or White–Metzner models give good agreements for laboratory simple devices. However, the applicability of this approach for industrial coextrusion process remains an important challenge, because one has to deal with the influence of complex geometries, varying temperature and more realistic constitutive equations. As it is still very costly in computational time to perform 3D instationary computations for viscoelastic fluids, a methodology to analyze the appearance of interfacial instabilities in a complex geometry is proposed within this paper. First, a stationary 3D computation is performed in the real geometry for a single polymer. This computation allows to find zones in which the flow motion is essentially 2D, isotherm and with maximum values of the shear rate and, thus, the Weissenberg number. Instationary 2D finite element computations for two layer flows and a multimode differential viscoelastic model are then performed in these zones. Spatio-temporal analyses give information on the spatial behavior of interfacial disturbances. This methodology is applied to the coextrusion of two polyesters for which experimental observations are available. The simulations predict well the experimental results and show that the outlet zone can amplify low frequencies perturbations. Finally, the influence of processing parameters (temperatures, flow rates, etc.) and die geometries has been checked.

1. Introduction

The coextrusion process consists in extruding different polymers in the same die in order to get a multi-layer product which combines the properties (mechanical, optical, adhesion, barrier) of the different layers. It allows for example to substitute lacquer for steel protection in food packaging industry. Here, the coextruded film is composed of two layers, the bottom one providing good adhesion properties on the steel sheet substrate, and the upper one good printing property. The steel coated sheet will then be deep-drawn to shape for example a beverage can. The two polymer layers are constituted of various polyester patented formulations which present very different rheological properties. For some processing conditions, coextrusion instabilities are observed between the two layers. They induce unacceptable optical properties for the final steel coated product (Fig. 1).

Coextrusion instabilities have been experimentally investigated for a long time. Three kinds of experimental studies have been performed.

- In the first approach, the transition between stable and unstable processing conditions is investigated by examining the aspect of the extrudate obtained with different couples of commercial polymers [1–5]. The authors related stable and unstable processing conditions to thickness ratio as well as viscosity and first normal stress difference ratios at the interface.
- In the second approach, the coextrusion flow and the development of instabilities have been examined in a very long die between parallel plates: after stopping the rotation velocity of the two extruders, the die is quickly cooled down, then dismounted and the interface between the two incompatible polymers is carefully analyzed [6]. The instability is not visible at die entrance, but then develops and amplifies along the die length, pointing out its convective character.

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http://dx.doi.org/10.1016/j.jnnfm.2012.12.014

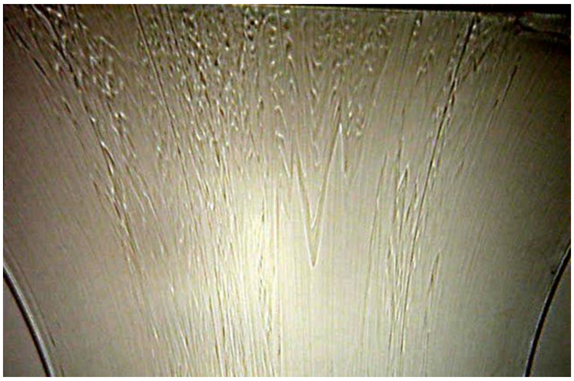

Fig. 1. Characteristic defect on an industrial coextrusion line.

– In the third approach, the instability is studied by looking at its development inside a partially transparent die for slightly unstable processing conditions, that is to say near the transition between stable and unstable coextrusion conditions. In Khomami and coworkers papers [7–11] evolution of the interface deviation along the die is observed through four optical windows for various wave numbers of the interface defect and various layer thickness ratios (or flow rate ratios). In Valette et al. paper [12] the same phenomenon has been observed, but the lateral walls of the channel die are now totally transparent, which allows a precise analysis of the progressive development of the instability. Interface perturbations are generated by adding small amplitude periodic oscillations of the flow rate at die entrance. These perturbations grow or decay as they travel downstream, pointing clearly out the convective nature of the interfacial instability [13].

From a theoretical point of view, the occurrence of such instabilities has been largely studied by means of a linear stability analysis of the two-layer Couette or Poiseuille flow. In such analysis, one studies the growth or decay rate of periodic and arbitrary small perturbations using the Gasters transformation [14]. In the twenty past years, stability analysis have been carried out for different viscoelastic constitutive equations (see for example [15–18]). All these studies point out that instability occurs even at very low Reynolds number and is mainly governed by the viscosity ratio, the elasticity ratio and the thickness ratio of the two layers of polymers.

More recently, authors use more realistic constitutive equations to compare linear stability analysis results with experiments. Ganpule and Khomami [19,20] use a 4-mode Giesekus constitutive equation, as Valette et al. [6] use a White–Metzner constitutive equation. There is a quantitative agreement with experimental observations both in terms of critical flow rate ratio and spatial amplification of interfacial disturbances.

For the first time Valette et al. [13] computed numerically the behavior in time and space of an interfacial perturbation generated at the inlet of a Poiseuille flow of two coextruded Maxwell fluids of high viscosity. The perturbation moves as a wave packet in the flow direction with a velocity different from that of the interface and is spatially damped or amplified according to parameter values (viscosity, elasticity and flow rate ratios). Due to the high viscosity of both polymers, the Reynolds number is close to zero and the driving parameter of the instability is the Weissenberg number which balances the first normal stress difference and the shear stress. As usual, theses previous non-dimensionless numbers are defined thanks to characteristic length and velocity of apparatus by taking

$$Re = \frac{\rho U_{\mathrm{ref}} L_{\mathrm{ref}}}{\eta}; \quad We = \lambda \frac{U_{\mathrm{ref}}}{L_{\mathrm{ref}}}$$

where η, ρ and λ are respectively the viscosity, the density and the relaxation time of fluid. For more complex fluids for which the relaxation time and viscosity are shear dependant, new expressions of theses non-dimensionless numbers are given at the end of the next section. This numerical analysis of the effect of a small perturbation was then compared to the linear stability analysis of the two-layer Poiseuille flow and a good agreement was found.

Time-dependent viscoelastic computations involve two major difficulties [21,22]. First, the non-linearity and the hybrid type of constitutive equations (elliptic and hyperbolic) make the convergence of the iterative algorithm difficult especially with geometrical or boundary conditions singularities. The second difficulty is the time-dependent computation of the interface. For these reasons, time dependent viscoelastic computations of multi-layer flow have been performed with rather simple rheological models, at moderate Weissenberg numbers and in simple geometries [13,23,24]. The competition between surface tension force and elastic stress for mono-layer flows is also explored in [25,26].

The aim of this paper is to perform a numerical study of the propagation of an interface perturbation in real die geometries and with realistic constitutive equations for the molten polymers. First we use a 3D finite element computation of the whole coat-hanger die geometry (with only one polymer). The velocity is maximal at the die inlet and at the die exit and it is supposed that instabilities will originate in these flow regions where the Weissenberg number is a maximum. In addition the flow pattern is 2D in these two flow regions. So we develop a 2D multilayer viscoelastic model with a precise capturing method of the interface. Then we apply this method to the inlet zone and the exit zone geometry of the coat-hanger die for various initial instability frequencies and for various processing conditions where defects are (or are not) experimentally observed. Finally we test some modifications of the geometry and of the processing conditions.

2. Presentation of coextrusion process and polymers

We consider a geometry composed of three zones (Fig. 2): the inlet zone is a feeding channel with a narrow width and an important thickness; then there is a distribution channel with a complex geometry (called *coat hanger* geometry) which allows to distribute regularly the polymer flow through the die width (350 mm). The third broad zone is of uniform thin thickness (1 mm).

The two coextruded polymers (PET1 and PET2) are polyester based patented formulations. They present very different rheological properties which have been measured using a cone and plate RMS 800 rheometer. Dynamic rheology measurements give the complex modulus (usually called G' and G'') for various temperatures. By assuming the Cox-Merz rule, viscosity η (the norm $\sqrt{(G'/\omega)^2 + (G''/\omega)^2}$ of complex viscosity) and relaxation time ($\lambda = G'/\omega G''$) are deduced. The bottom layer (PET1) in contact with the steel sheet has good adhesive properties and marked viscoelastic behavior. The upper layer (PET2) is classical polyester with a quasi-Newtonian behavior. Fig. 3 shows the master curve for the viscosity of both polymers at 260 °C and Fig. 4 the master curve for the relaxation time at the same temperature. The activation energy of both polymers is very different: E = 38 kJ/mole for PET1 and E = 61 kJ/mole for PET2.

This makes this coextrusion situation particularly interesting. At the reference temperature (260 °C) PET1 is much more viscous than PET2 within the extrusion processing range (typically 10 – 100 s^{-1}) but both viscosities become equivalent at higher rates. When decreasing the temperature the viscosity of PET2 increases