

Relationships between molecular structure and sharkskin defect for linear polymers

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Abstract

This work is focused on the sharkskin defect encountered during the extrusion of linear polymers. This defect is characterized by the development of surface cracks, perpendicular to the flow direction. Our purpose is to establish relationships between sharkskin onset and rheological and molecular properties of the extruded polymer. Starting from the elasticity theory of Griffith, we show that the period of the defect is proportional to its amplitude, and that the critical stress for the onset of sharkskin is a function of the plateau modulus, the weight average molecular weight and the molecular mass between entanglements. On the other hand, we show that the cracks propagation velocity is controlled by the extrudate velocity and the Rouse time. We explain that the critical shear rate for the onset of sharkskin depends on the longest time of the distribution of relaxation times, i.e. the tube renewal. Finally, the present approach allows to clarify the ambiguity of simultaneous apparition of sharkskin defect and wall slip, as reported in the literature.

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1. Introduction

In extrusion processes, the rates of industrial production are often limited by the onset of flow instabilities that perturb the shape of the extrudate. These instabilities can affect the surface or the whole volume of the extruded material, and they have been the subject of a large number of studies for more than 50 years. For detailed information, we refer the reader to relevant review papers published on this topic [1,2]. In the present study, we focus our attention on one of these flow instabilities, usually called sharkskin defect. It occurs generally at low shear rate, under the form of regular cracks on the surface of the extrudate, perpendicular to the flow direction. At the early beginning, a loss of gloss is observed, which means that the order of magnitude of the defect is that of the wavelength of the visible light, i.e. 0.4–0.8 μm . Then, by increasing the flow rate, the defect develops and becomes organized [3–6]. The surface appears as affected by regular cracks, whose period and amplitude increase progressively with flow rate (Fig. 1).

Many interpretations have been proposed to explain the origin of the sharkskin defect. Presently, it is now obvious that the defect takes its origin around the die exit, when the molten polymer is leaving the die. However, two main assumptions remain open. The first one, proposed by Cogswell [7] in 1977, considers the reorganization of the velocity profile around the exit singularity (transition from a steady state pressure flow in the die to a “plug flow” with flat profile in the extrudate). These changes in the velocity profile induce high elongational stresses in the skin region of the extrudate. If these stresses overcome the melt strength of the polymer, they lead to the fracture of the extrudate surface. This interpretation is supported by experimental observations [5,6,8–10] and numerical simulations [11–13]. The second interpretation is based on an experimental observation initially made by Kurtz [14]. The sharkskin defect seems to appear for a shear rate at which a change in slope may be observed in the flow curve (shear stress/apparent shear rate relationship). This idea, made then popular by Ramamurthy [15] and Kalika and Denn [16], leads to consider that the defect is due to an interfacial mechanism, corresponding to a strong slip at the die wall. In fact, Wang et al. [17] attributed a few years later the governing mechanism to the transition between weak slip and strong slip of the Brochard–de Gennes model [18]. In this

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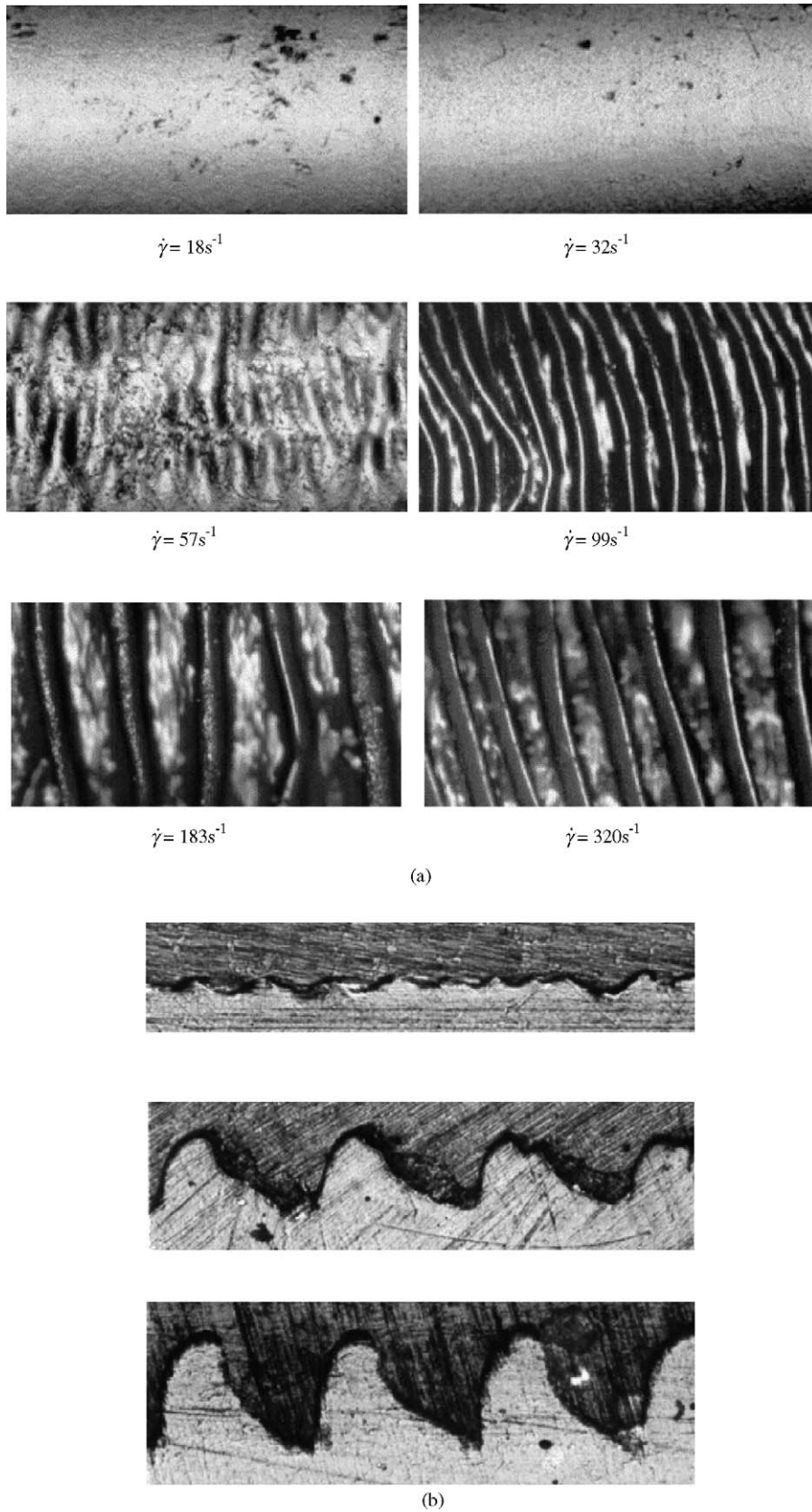


Fig. 1. Example of sharkskin defect. (a) Surface aspect for different shear rates; (b) corresponding cross sections parallel to the flow direction (from [4]).

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