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Test Method

A new double-slit rheometrical die for in-process characterization and extrusion of thermo-mechanically sensitive polymer systems

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The twin-slit rheological die can be operated in such a way that the total pressure drop is maintained constant while the shear rate in the measuring slit is changed. The device is particularly useful when coupled to an extruder and used to characterize materials that are sensitive to thermo-mechanical conditions. In the present work, the twin-slit die design was modified to a double-slit die where the measurement and extrusion channels have different geometries, and viscoelastic and morphological characterization can be performed. The flow analysis developed for twin channels was generalized to accommodate for different channel lengths and cross sections. The new set of equations was used to design the double-slit flow channel. Comparison of off-line with in-process data validated both the double-slit die and the experimental methodology. The practical utility of the new die was then demonstrated with the in-process rheological characterization of various polymer systems.

1. Introduction

As more sophisticated polymer materials and products need to be developed within shorter time-to-market, fast response characterization tools using small amounts of sample and capable of conveying data relating rheological response, process-induced morphology and engineering properties become increasingly necessary. A common approach is to use small-scale processing equipment coupled to on-line or in-line measuring sensors/devices [1,2].

Coupling instrumented capillary or slit dies to extruders for in-line measurements seems particularly promising. Not only shear viscosity [3,4] and normal-stress differences become accessible [5–8], but measurements are done at relatively higher shear rates (in excess of 10 s^{-1}), i.e., in the region of practical processing. Optical methods are non-invasive and recognized as powerful tools for morphology analysis. Thus, in-line light scattering studies of polymer blend morphology using a slit die with an optical window coupled to an extruder started over a decade ago [9–11]. For example, the authors coupled an extruder to a modular rheo-optical slit die with rheometrical (viscosity and normal stress differences) and optical capabilities (small angle light scattering (SALS) or polarized optical microscopy (POM)); together with the relevant downstream equipment, they also produce extrudates for subsequent appraisal of the engineering properties [5,12].

Coupling an extruder to a rheological die and measuring both the resulting throughput and pressure drop will yield a single point of the flow curve (viscosity vs. shear rate). To generate a range of shear rates, either the screw speed (single screw extruder) or the feed rate (twin screw extruder) must be changed. This action will modify the thermo-mechanical history of the material in the extruder (residence time, temperature, hydrodynamic stresses) and so may lead to changes in homogeneity, thermal stability and/or morphology. At each shear rate, a possibly distinct material could be characterized [13]. Rauwendaal and Fernandez [4] observed that their in-process measurements consistently yielded lower viscosity values compared to the results from off-line capillary and cone-and-plate rheometry. The differences were more pronounced at higher shear rates (i.e., higher screw speeds) and attributed to the shorter residence times between extruder and slit die allowing for less relaxation of the screw-induced effects. According to Vergnes et al. [13], changes in upstream flow conditions could explain the anomalous results (negative values of the power law index) obtained by some authors for in-line food rheology [14]. Other authors confirmed that the methodology had a small effect on the flow curves of low-density polyethylene melts, but yielded a considerable error in the case of foods [15,16] and other complex and structured fluids [17,18].

To circumvent this problem, various authors proposed slit die designs enabling the acquisition of various points of the flow curve in a single experiment. Pabedinskas et al. [19] used a slit die with constant width and a linear vertical taper, so that the strain rate increased gradually downstream (within a tenfold range). Assuming the flow of a power-law fluid, from the measurement of pressures at three axial

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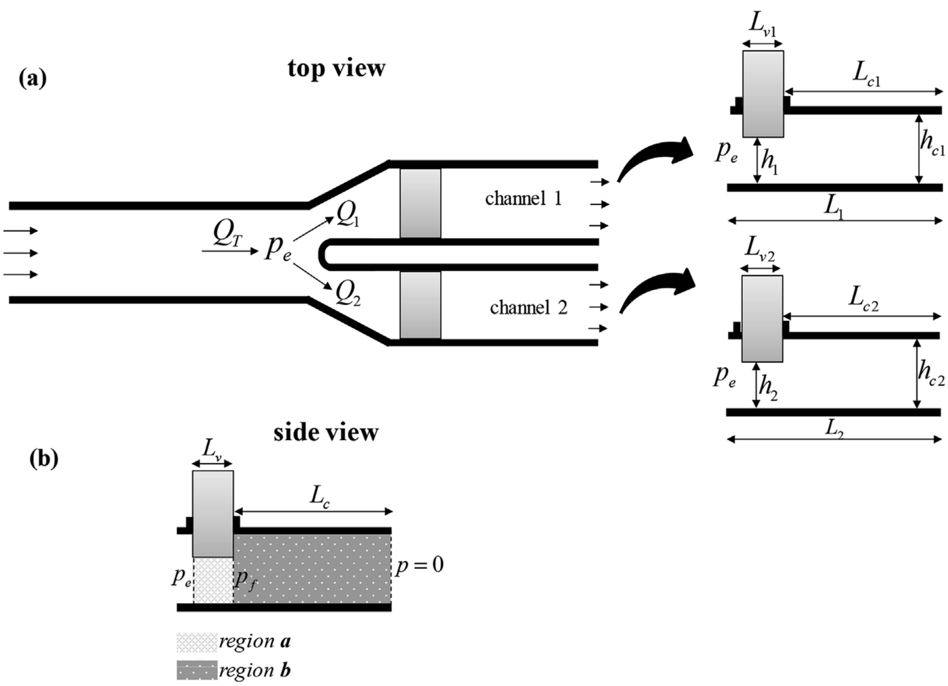

Fig. 1. Schematic representation of the twin channel slit following the approach and nomenclature of Vergnes et al. [13] (a). Left: top view, flow goes from left to right; valves are shaded in grey; Q_T is the total flow rate, Q_1 and Q_2 are the flow rates in channels 1 and 2, respectively; p_e is the entrance pressure. Right: side view of the two channels; valves move vertically; (b) flow regions considered to derive the relationship between Q_T and p_e.

locations of different heights, n and k could be extracted. Agreement with data obtained from off-line capillary rheometry was very good for low viscosity materials, but for higher viscosity samples the in-line values were consistently lower. Because of the convergence of the channel, the flow is not truly viscometric, and pressure differences will reflect both viscous and elastic responses [20]. Thus, the usefulness of this approach may be limited to low viscosity or little elastic fluids. Horvat et al. [21] developed a multi-step slit die in which the flow channel is divided into zones of different constant heights and uniform width. Each zone contains two flush mounted pressure transducers at distinct downstream locations. Experiments were reported for a three-step configuration, generating three shear rates spanning one order of magnitude. Due to the need to mount 6 pressure transducers, the total length of the slit was quite considerable (370 mm). However, with only two measurements being done per channel segment, the eventual effect of pressure or viscous dissipation on viscosity is not readily identifiable. Moreover, the width/height ratio of the channels was smaller than 10, which is often considered as the threshold to guarantee the development of simple 1D viscometric flow. Coates et al. [22] attached a flow regulating valve to the exit of a slit. Although various configurations of this accessory were tried, it was not evident that a viscometric flow was attained. In practice, only low shear rates were tested (approximately 1 s^{-1}), and agreement with off-line data was only moderate. Kalyon et al. [23,24] developed an adjustable gap in-line rheometer consisting of a slit die with a movable plate for continuous height adjustment. In order to keep the flow rate constant, the device must be coupled to extruders operated in starve-fed mode. Hochstein et al. [25] modified a multiple-step slit die by mounting the lower half of the slit on a wedge that can slide horizontally, and thus vary the gap. Also, a flow valve positioned near to the die exit was used to control the total pressure drop. Apart from the limitations already mentioned above, it is worth noting that variations in the total pressure drop created by these solutions to generate different shear rates will modify the flow conditions upstream of the die.

The design proposed by Springer et al. [26] aimed at inducing variations of shear rate while maintaining constant the total pressure drop. The entry flow channel is divided into two parallel identical slits, one being used for the measurements (i.e., with at least two pressure transducers). Flow distribution in the two slits is controlled by vertically moving valves located at the entrance of each channel. To change the shear rate at the measuring slit, the corresponding valve is moved simultaneously with the other one, but in opposite directions, in such a way that the pressure at the entry channel remains constant. Shear rates were varied over two orders of magnitude, but no direct comparison with off-line data was apparently performed. Later, Vergnes et al. [13] re-visited the concept and developed an analytical model for a power law fluid relating the aperture of the two valves that forces constant flow rate and constant entrance pressure. Della Valle et al. [27,28] and Lach [29] successfully used this type of rheological die, whose major drawback seems to be the waste of material during measurements. Li et al. [15] used a modified design with perpendicular, rather than parallel channels. Recently, Robin et al. [30] fitted a parallel slit die with adjustable slit heights (by means of the slide solution developed by Hochstein et al. [25]). The valves were located close to die exit, allegedly to avoid generating a high amount of shear and energy dissipation upstream of the measurement zone, as well as to avoid premature gas bubble nucleation in the starchy melt. However, locating valves near to discharging end of the die may jeopardize the viscometric nature of the flow, as seen above.

Due to its considerable advantages, namely maintaining the total pressure drop while the shear rate in the measuring slit is changed, the concept of the twin-slit rheological die is adopted here. However, both the geometry and the construction were modified in order to improve the practical utility and the range of measurements of the device. The measuring slit was made suitable for rheo-optical characterization [5,12], enabling the determination of both viscoelastic and morphological characteristics. The second slit was used for conventional extrusion, but its width was made larger, so that standard tensile specimens could be cut from the extrudate. Thus, the length and width of the two slits will be distinct. In order to support the new design, the analysis of Vergnes et al. [13] will be extended to non-identical double-slits.

The outline of the paper is as follows. First, Vergnes's analysis is extended to non-identical double-slits. Then, the geometry of the flow channels, construction of the die and experimental set-up are presented. Next, the theoretical predictions are compared with the experimental data. Finally, the new die is used to measure the viscoelastic behavior of several polymer systems.