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Stability of cast-film extrusion of various metallocene polyethylenes

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1- Introduction

The Cast Film process is one of the most widely used polymer processing technology to produce polymer films. A polymer melt is extruded through a flat die and then stretched in air by a chill roll to generate a thin film as illustrated in Figure 1. The molten film is quenched on the surface of the chill roll (typically water-cooled and chrome-plated) by an air knife or a vacuum box.

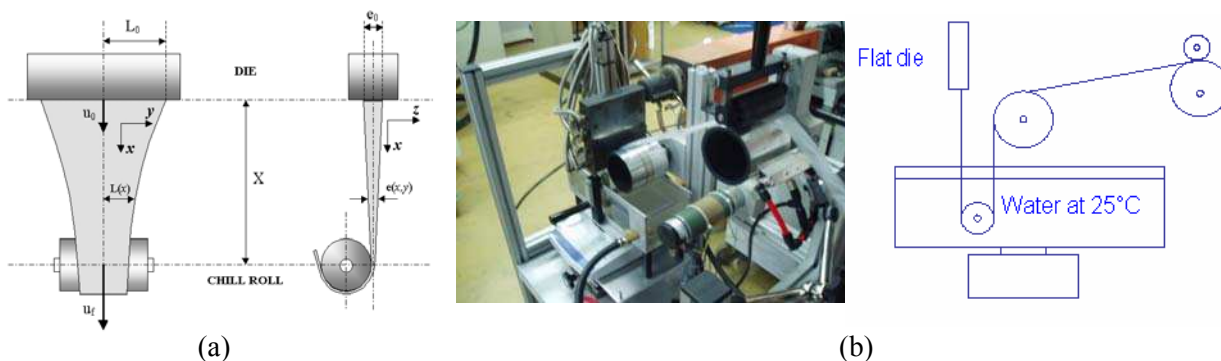


Figure 1: General view of (a) the Cast Film process, (b) the Cemef experimental device

The cast film process may induce defects depending on the drawing conditions. The Draw Resonance is the most critical one and consists of periodic fluctuations of the thickness and the width of the film. Several experimental and numerical studies were undertaken in order to model the process stability as a function of adimensional numbers (1). In this work, we use an experimental cast film line to study the stability of several mPE and a Ziegler Natta polyethylene.

The experimental extrusion line is composed of a single-screw Haake extruder and a vertical fish tail die of dimensions (100mm x 1mm). The film is pulled by a first roll in water and drawn with a second roll in the air. The traditional chill roll is replaced by a water bath disposed on a balance (Figure 1 b). Therefore it is possible to estimate the fluctuations of the drawing force by the measuring of the weight variations. The molten polymer crystallizes as soon as it enters the water bath.

2 – Experimental study

The stability analysis was done for 4 different resins. Two polyethylene resins, mPE1 and mPE2, obtained by metallocene catalysis, were selected as well as a third metallocene polyethylene with a different macromolecular architecture, mPE3. A Ziegler-Natta catalysis polyethylene, PE ZN, which is a reference polyethylene in the cast industry, was also studied. The main characteristics of the polymers and their relaxation times are summarised in Table 1.

| Resins | Density ρ (g/cm ³) | M_n (g/mol) | M_w (g/mol) | λ Carreau (s) |
|--------|-------------------------------------|---------------|---------------|-----------------------|
| mPE1 | 0.934 | 26600 | 67200 | 1.94 (200°C) |
| mPE2 | 0.95 | 25700 | 69000 | 2.32 (200°C) |
| PE ZN | 0.935 | 22600 | 70400 | 0.05 (200°C) |
| mPE3 | 0.927 | 23000 | 68000 | 0.02 (200°C) |

Table 1 : Description of the resins

All extrusions were carried out according to the same experimental protocol: an extrusion temperature of 260°C at the die exit and a drawing distance of 25 mm. Only the extrusion velocity varied from 16 rpm to 256 rpm. Extrusions of the mPE1 and mPE2 resins did not present any particular problem and a complete study of the processability behaviour has been done. On the contrary, mPE3 and PE ZN were very difficult to process under the same conditions.

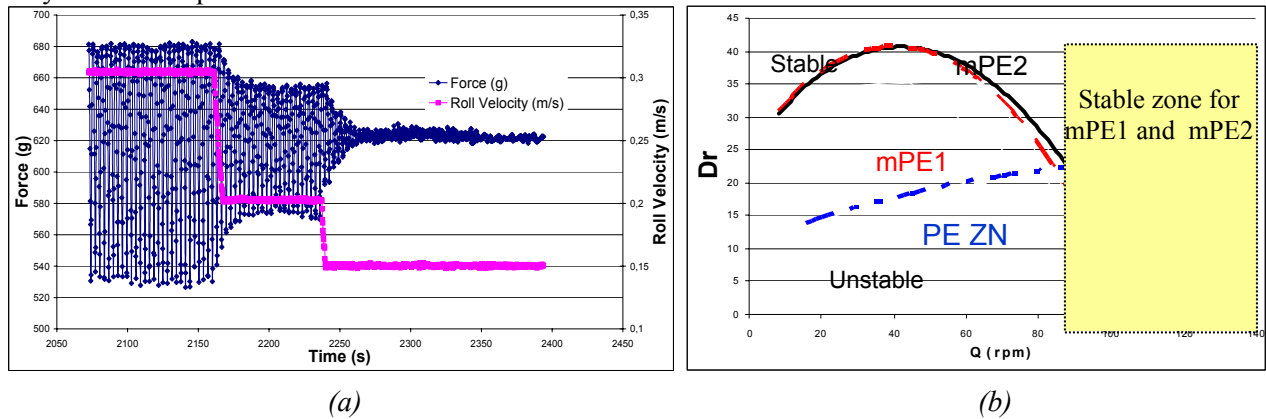


Figure 2: (a) Unstable/stable transition for mPE2 : $Q=90.5$ rpm, $Dr=20, 13$ and 10 ,
(b) Stability curves for 3 resins

In this experiment, we can identify with a high degree of accuracy the stability limit curve. The instabilities are reduced when the draw ratio Dr , defined as the ratio between the chill roll velocity (u_f) and the extrusion velocity at die exit (u_0), is reduced and disappear completely with low values of Dr (Figure 2a). This type of behavior is a perfect example of a transition from a stable domain towards an unstable one. The stability analysis was supplemented by measurements of film width variations. They follow the same evolution according to the drawing rate

Periodic instabilities appear at short drawing distances and high drawing rates. They are also influenced by the polymer rheology and the thermal process conditions. In order to compare the different polymers, the different stability curves were plotted (Figure 2b). A master curve appears whose shape is quite different from the one deduced from a linear stability analysis in 1D (1) or from a direct unstationnary 2D numerical simulation by (2).

3. Discussion

Periodic instability (Draw Resonance) is encountered at higher drawing rates for mPE1 and mPE2 than for PE ZN. This may be linked to the marked viscoelasticity of mPE1 and mPE2, which contributes to the stabilization of the process as shown by Silagy et al (1, 2). The mPE2 resin has a slightly more stable behaviour than mPE1, which presents atypical periodic instabilities. The disappearance of instability, for mPE1 and mPE2, at high flow rates could correspond to the transition from the unstable zone to the so-called unatteignable zone.

In conclusion, this kind of experimental device allows a very good description of metallocene polyethylene behaviour in the cast film process and makes it possible to define their stability limit.

4 References

1. D. Silagy, Y. Demay, J-F Agassant, *Analysis of the film casting process*, J. Non-Newtonian Fluid Mech. 79 (1998) 563-583.
2. D. Silagy, *Etude expérimentale et modélisation du procédé d'extrusion de film à plat de polymère*. Thèse de Doctorat en Sciences et Génie des Matériaux (1996), ENSMP.