

## Rheological Parameters to Predict the Extrusion Coating Performance of LDPE

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### ABSTRACT

Changes in the molecular structure have a great impact on the rheological and processing properties of a polymer. Therefore these parameters are extremely significant in understanding a wide range of polymer processing operations.

In extrusion coating a thin molten polymer film is coated on to some kind of substrate. The extrusion coating performance is related to the neck-in and draw-down properties of the polymer film.

This paper describes how oscillatory measurements can be used to characterize LDPE. The zero shear viscosity, the steady state compliance and the activation energy have been found to be useful parameters to predict extrusion coating performance of LDPE.

### INTRODUCTION

At high extrusion coating speed, even a minor disturbance on the melt web will cause major quality problems, which very rapidly lead to large quantities of waste. Therefore, higher coating speeds require high and even quality to avoid waste due to polymer edge instability and web break.

The rheology-related phenomena that can cause problems in extrusion coating is neck-in and draw-down.

The neck-in is the reduction of the web width and can cause uncoated parts of the substrate. Neck-in is less if melt elasticity is high.

The draw-down is the ability of the melt to be drawn at high line speeds to thin films without breaking. Draw-down is favoured by a melt that is more viscous than elastic.

In optimizing the extrusion coating process it is of most importance to balance the elasticity of the polymer.

Melt viscosity and elasticity of LDPE are dependant on the molecular structure, such as molecular weight (Mw), molecular weight distribution (MWD) and long-chain branching (LCB).

### MATERIALS AND METHODS

In this study 42 extrusion coating grades of LDPE from different suppliers have been characterized. All LDPE's have been high pressure autoclave produced, with a Melt Flow Index (MFI) between 6-8 g/10 min. (ASTM D1238) and density in the range of 917-920 kg/m<sup>3</sup>.

The rheological measurements were performed on a Bohlin Controlled Stress Melt Rheometer. Polymer granules were melted, compressed and cut into discs with a diameter of 25 mm and a thickness of 1 mm. Parallel plates were used and measurements were done under nitrogen atmosphere.

Dynamic measurements were performed at 150, 170 and 190°C respectively in the linear viscoelastic region (determined by stress sweep measurements) with frequency sweep between 6-0.004 Hz.

The  $\tan\delta$ ,  $G'$  and  $G''$  were used to shift curves to a reference temperature of 170°C by the use of the IRIS<sup>1</sup> software. From the

shift factors the horizontal and vertical activation energies were determined by the use of the Arrhenius equation.

For some of the LDPE's dynamic measurements were also performed at a frequency sweep between 0.004-0.001 Hz at 170°C.

Creep and recovery measurements at 5 Pa and 170°C were also done on some of the grades.

## RESULTS AND DISCUSSION

In linear viscoelasticity a relationship for the elasticity can be written as:

$$\Psi_0 = 2 \eta_0^2 J_e^0 \quad (1)$$

One objective in this study was to calculate the elasticity using this relationship by determining the zero shear viscosity and the steady state compliance from dynamic measurements.

From master curves at 170°C the complex viscosity has been extrapolated to  $\omega \rightarrow 0$  where  $G''/\omega = \eta_0$ .

For the steady state compliance the following approach has been made.

It can be shown that in the limit of low frequencies, where  $\omega \rightarrow 0$  the following relationship is valid:

$$G'(\omega) = J_e^0 [G''(\omega)]^2 \quad (2)$$

For linear polymers the level of  $G'$  at low  $G''$  can therefore be taken as a measure of MWD because it is proportional to the steady state compliance.

A method proposed by Shroff et al.<sup>2</sup> uses this relationship to characterize MWD. A polydispersity index, MWDER, is defined as:

$$\text{MWDER} = C_1 G'(\text{at } G''_{\text{ref}}) \quad (3)$$

In this study, good results have been obtained with  $G''_{\text{ref}} = 500$  Pa and  $C_1 = 1.78 \cdot 10^{-3}$ .

For LDPE where the modulus shows temperature dependence this method needs to be redefined. This is done, also described by Shroff et al.<sup>2</sup>, by multiplying the modulus with a factor  $\omega^{s-1}$  where  $s$  is defined as:

$$s = 1 - E_b/E_a \quad (4)$$

The  $E_a$  and  $E_b$  are the horizontal respectively vertical activation energy.

This method were then used to establish a relationship between MWDER and steady state compliance for the LDPE's. Steady state compliances were determined from creep and recovery measurements and from dynamic measurements at low frequencies, down to 0.001 Hz, where  $J'$  approach  $J_e^0$ . The IRIS<sup>1</sup> software was also used to calculate the steady state compliance from the relaxation time spectrum. The results are given in Fig. 1.

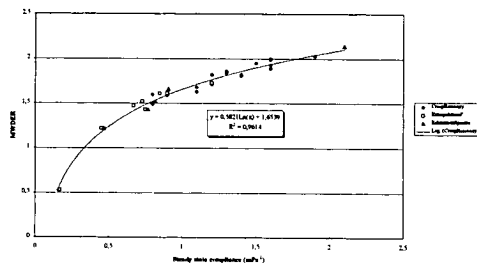


Figure 1. Relationship between MWDER and steady state compliance.

From dynamic measurements it is now possible with this procedure, to determine the steady state compliance,  $J_e^0$ , at 170°C.

Finally, the activation energy ( $E_a + E_b$ ), from shifting the curves to the reference temperature of 170°C, is plotted versus linear elasticity ( $2\eta_0^2 J_e^0$ ). Results from measurements of all 42 LDPE samples are given in Fig. 2.

From extrusion coating performances it has been shown that LDPE's having high linear elasticity and low activation energy, exhibit low draw-down properties. On the

contrary, LDPE's with low linear elasticity give high neck-in.

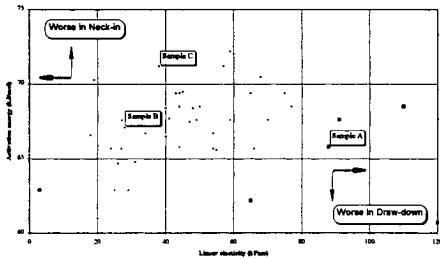


Figure 2. Relationship between rheological parameters and extrusion coating performance of LDPE.

Three different samples of LDPE produced in the same reactor, were tested in a pilot coating line with respect to draw-down and neck-in. The results are presented in Table 1.

Table 1. Draw-down (DD) and neck-in (NI) results from three LDPE samples.

Sample	DD (m/min.)	NI (mm)
A	390	40
B	500	80
C	535	70

The samples were also tested by the rheological method described, and the results are shown in Fig. 2.

It is difficult by rheological methods to characterize the molecular structure of LDPE because it contains long-chain branches. These long-chain branches have impact on both the relationship between the zero shear viscosity and Mw and the relationship between the steady state compliance and MWD.

One thing that has been confirmed for these LDPE grades by gel permeation chromatography (GPC), is that grades at the right side of the plot (high linear elasticity, >80 kPa) exhibit broader MWD. The

steady state compliance for these grades are in the range of 1.9 - 2.6 mPa<sup>-1</sup>.

Grades at the left side of the plot (low linear elasticity, <10 kPa) have shown to have more narrow MWD. Steady state compliance being in the range of 0.1 - 0.5 mPa<sup>-1</sup> for these grades.

## CONCLUSION

LDPE from high pressure autoclave reactor for extrusion coating with a MFI from 6 to 8 g/10 min., and with a density of 917-920 kg/m<sup>3</sup> has for a long time been considered as a uniform commodity.

However, as the evolution goes towards faster coating lines, it has been possible to detect differences in processing performance between different LDPE grades.

The parameters MFI and density gives very limited information about the processing performance of the product. Very often a resource demanding test run of the LDPE material in pilot or production coating line is necessary in order to determine the processability.

Extrusion coating performance can be conveniently, and considerably less resource demanding, determined by the rheological method-described-in-this paper.

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