Editor's Note: Per-Åke Clevenhag and Claes Oveby of Tetra Pak Carton Ambient AB were awarded both the 2004 TAPPI PLACE Division Best Paper Award and the Dragan Djordjevic Award for their paper "Rheological indicators to predict the extrusion coating performance of LDPE." TAPPI's PLACE (Polymers, Laminations, Adhesives, Coatings, and Extrusions) Division is made up of hundreds of technical professionals working in flexible packaging, laminations, adhesives, coatings, and extrusions.

This paper, which has been reprinted below, was selected by conference attendees as the best out of more than 100 papers presented at the 2004 TAPPI PLACE Conference. TAPPI's Extrusion Coating Committee also selected this paper to receive the Dragan Djordjevic award, which honors the best paper about extrusion coating presented at the conference. Originally published in the January, 2005 edition of "The PLACE," the division's online newsletter, the full presentation, including additional figures, can be accessed at http://www.tappi.org/index.asp?pid=30717&ch=4.

Both awards were presented at the 2005 TAPPI PLACE Conference held September 25-29, 2005, in Las Vegas, Nevada, USA. For information about the 2006 TAPPI PLACE Conference, which is scheduled for September 17-21, 2006 in Cincinnati, Ohio, USA, or to learn more about TAPPI's PLACE division, visit http://www.tappi.org/tappi/place/index.asp.

Rheological indicators to predict the extrusion coating performance of LDPE

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LDPE (low density polyethylene) from high-pressure autoclave reactors for extrusion coating with a melt flow rate (MFR) range from 7 to 9 g/10 min with a density of 917 to 920 kg/m³ has for many years been a uniform commodity. As evolution goes toward faster coating lines, differences in processing performance between various LDPE grades have occurred. Various LDPE grades behave differently in the coating process due to different rheological properties.

Thirteen grades of autoclave LDPE from different suppliers were tested in two pilot extrusion coating lines on three different occasions. All grades had MFR between 7 and 9 g/10 min and densities of 918 to 920 kg/m³. The melt temperature was monitored to 295–305°C by means of an infrared camera. In principle, the methodology was as follows. Extruder screw rpm was set to give 10 g/m² at 150 m/min to obtain stable processing conditions. The line speed was then increased in steps of 50 m/min until the web broke. The line speed at which the web broke (draw-down speed) and the neck-in were reported. This was made in duplicate for each grade of LDPE.

The parameters MFR and density give very limited information on the processing performance of a product. A resource demanding test run of LDPE material in a pilot or production coating line is often necessary to determine processability. Extrusion coating performance can be conveniently and considerably less resource demanding when determined and predicted by the rheological method described in this paper.

Application: *A new rheological test method allows prediction of processing performance of LDPE for extrusion coating.*

Changes in the molecular structure have 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 some kind of substrate. At high extrusion coating speed, even a minor disturbance on the melt web causes major quality problems, which very rapidly lead to large quantities of waste. Therefore, higher coating speeds require polymers with high and even quality in order to avoid waste due to polymer edge instability and web break.

The rheology-related phenomena that may cause problems in extrusion coating are neck-in (NI) and draw-down (DD). The neck-in is the reduction of the film width and may cause uncoated areas on the substrate. The neck-in is less if the melt elasticity is high. The draw-down is the ability of the melt to be drawn to thin films without breaking. Draw-down is favored by a melt that is more viscous than elastic.

In optimizing the extrusion coating process it is of utmost importance to balance the elasticity of the polymer. This paper describes how oscillatory measurements can be used to characterize LDPE. The G' and η_0 have been found to be useful parameters to predict the extrusion coating performance of LDPE.

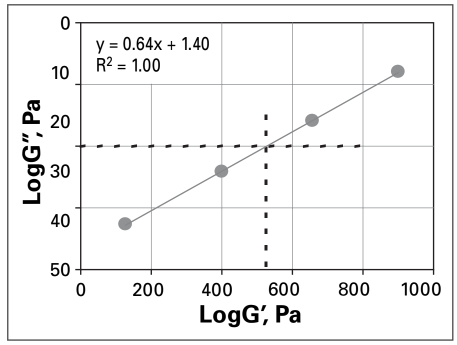
MATERIALS AND METHODS

Rheological Measurements

The rheological measurements were performed on a StressTech controlled stress melt rheometer. Polymer pellets were melted,

LDPE	MFR, g/10 min.	G', Pa	η_0, Pas	Predicted DD, m/min.	OBSERVED DD AND NI, m/min.	OBSERVED DD AND NI, mm
1	7.1	105.1	4220	414	400	99
2	7.0	132.7	4640	194	200	71
3	8.0	108.2	3580	405	365	91
4	6.8	123.4	5000	257	300	87
5	7.2	120.9	4280	292	300	84
6	7.1	117.2	4420	317	325	85
7	7.5	118.5	4280	311	290	80
8	7.2	108.5	4230	388	350	88
9	8.9	105.2	3220	435	550	—
10	7.0	132.7	4640	194	170	—
11	8.0	108.2	3580	405	430	—
12	6.8	116.3	4900	314	390	—
13	7.2	120.9	4280	292	322	—
14	7.1	117.2	4420	317	352	—
15	7.0	106.5	4575	396	400	—
16	7.5	118.3	5565	284	290	—
17	8.3	105.4	3635	425	375	—

I. Summary of the data measured and generated at the pilot extrusion coating trials.



1. The log loss modulus (G") versus log storage modulus (G') for the determination of the G' at G" equal to 500 Pa. Modulus measured at 170°C in a frequency sweep between 20 and 0.01 Hz.

	LDPE1 Mean	LDPE1 Std	LDPE2 Mean	LDPE2 Std
MFR 2.16 kg 190°C, g/10 min.	7.41	0.21	7.62	0.24
G' 170°C, Pa	110.3	1.59	121.5	3.42
η_0 170°C, Pas	4217	170	4757	262
Neck-in, mm	93	1.5	80	3.9
Draw-down, m/min.	374	11	277	30

II. Data from monitoring of 2 LDPE grades with MFR of 7.4 and 7.5 and density of 920 kg/m³. Neck-in and draw-down are calculated from G' and η_0 by the relationship (2) and (3).

compressed and stamped 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. Oscillatory measurements were performed at 170°C in the linear viscoelastic region with a frequency sweep between 20 and 0.01 Hz. When a material undergoes oscillatory stress with frequency, the response can be expressed in terms of a storage modulus, G' (G prime) a loss modulus, G" (G double prime) and a complex viscosity, η^* (Eta star).

Extrusion Coating Trials

Thirteen grades of autoclave LDPE from different suppliers were tested in two pilot extrusion coating lines at three different occasions. All grades had MFR between 7 and 9 g/10 min. and densities of 918-920 kg/m³. The melt temperature was monitored to 295-305°C, by means of an infrared camera. The methodology was in principle the following. Extruder screw rpm was set to give 10 g/m² at 150 m/min. in order to obtain stable processing conditions. The line speed was then increased in steps of 50 m/min. until the web broke. The line speed at which the web broke (draw-down speed) and the neck-in were reported. This was made in duplicate for each grade of LDPE.

G' and η_0 Monitoring of Two LDPE Grades

In order to determine the product consistency and possible correlation between G' or η_0 and MFR, two LDPE suppliers were selected to send samples from 2 lots to Tetra Pak each week during a period of 4 months for G' and η_0 measurement. The suppliers provided Tetra Pak with the MFR data (2.16 kg; 190°C).

RESULTS AND DISCUSSION

G' is determined at G" 500 Pa by the use of a Cole-Cole plot (**Fig. 1**). The log G" is plotted versus the log G' in the range of 200-900 Pa of the G". A linear relation is obtained and the G' can be determined at G" equal to 500 Pa (log500 = 2.7).

Zero shear viscosity, η_0 (Eta zero) is determined by extrapolation by the use of a 3 parameter Cross equation:

$$\eta^* = \eta_0/(1 + (\tau\omega)n) \qquad (1)$$

The curve fit calculation is preferably done with the Microsoft Excel solver.

The results from the extrusion coating trials were analyzed by the use of a multivariate data analysis software. G' and η_0 were set as variables and observed draw-down as the response (17 observations).

The model diagnostics evaluation gave the following results:

R^2 = 0.76 (estimates goodness of fit)
Q^2 = 0.72 (estimates goodness of prediction)

From the multivariate data analysis the following relationship was found:

$$DD = 1311 - 7.55^*G' - 0.022^*\eta_0 \quad \text{(m/min.)} \qquad (2)$$

A relationship between draw-down and neck-in, with an R^2 of 0.93, was also found:

$$NI = 0.13^*DD + 44 \text{ (mm)} \qquad (3)$$

MFR showed no correlation to observed draw-down. An R^2 of 0.37 was achieved when observed draw-down was plotted versus measured MFR.

The results from the oscillatory measurements, the pilot extrusion coating trials and the results fro m the predictions are given in **Table I**.

The results from the monitoring of the two LDPE grades are given in **Table IIC** .

Both grades showed MFR 190°C within specification. There was a significant difference in G' between the two grades, both in level and variation. LDPE1 shows better consistency and can be used for higher line speeds. LDPE2 exhibits lower neck-in but shows more fluctuations. There were no correlations found between G' or η_0 and MFR ($R^2 < 0.1$).

CONCLUSIONS

LDPE from high-pressure autoclave reactors for extrusion coating with an MFR from 7 to 9 g/10 min., and with a density of 917-920 kg/m^3 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 various LDPE grades.

The parameters MFR and density gives very limited information on the processing performance of the product. Very often a resource demanding test run of the LDPE material in a 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 and predicted by the rheological method described in this paper. **TJ**

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