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PROPERTIES PREDICTOR FOR HDPE/LDPE/LLDPE BLENDS FOR SHRINK FILM APPLICATIONS*

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ABSTRACT: A technique that involves design of experiments was developed to generate a set of equations that predicts processing, mechanical and shrink properties of HDPE/LDPE/LLDPE blends. The results are presented in an easy-to-use spreadsheet that can be used even in pocket computers.

KEY WORDS: polymer blends, shrink film, selector, LDPE, LLDPE, octene, butene, HDPE, BUR, FLH, MFI, modulus, tear strength, puncture resistance, rupture strength, toughness, gloss, haze, effect of processing variables.

INTRODUCTION

IT IS WELL known that a good shrink film depends on selecting the proper material and operating conditions [1,2,6]. Different levels of MD and TD shrink can be obtained for the same material by simply modifying film orientation by changing blow-up ratio (BUR), frost line height (FLH), and drawdown ratio (DDR). On the other hand, although fractional-MFI low density polyethylene (LDPE) is commonly used for this application, other materials such as high density polyethylene (HDPE) and linear low density polyethylene (LLDPE) are added to gain specific properties, such as rigidity, tear strength, and puncture resistance.

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Figure 2 appears in color online: http://jpf.sagepub.com

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The above means there are many variables to evaluate. In an industrial operation, it may be very costly in terms of production loss and human effort to use all these variables (including resin selection, blend proportion, operating conditions) to achieve the desired final properties. The objective of this research is to provide a tool that processors can use to minimize the time and resources needed to develop a film for specific applications.

EXPERIMENTAL PROCEDURE

In this work, a design of experiments was conducted where shrink-film LDPE, octene- and butene-LLDPE, and HDPE from Polinter, Venezuela were blended in various proportions. These blends were extruded at several BUR, FLH and thicknesses to study both the effects of material and operating conditions. The results were used to develop relations between these variables and final properties and incorporated in an easy-to-use spreadsheet.

The experimental design was conducted with the following constraints:

(a) HDPE content was limited to 10% by weight.
(b) LLDPE proportion was limited to 30% by weight.
(c) BUR was varied between 2.0 and 4.5.
(d) FLH was varied between 3 and 6 die diameters.
(e) Thickness was varied between 50 and 90 μm.

The above constraints were chosen to represent the most common conditions used in the processing industry, where LDPE is used as the major component and butene- and octene-LLDPE (C4-LLDPE and C8-LLDPE, respectively), and HDPE are used to gain specific properties. DOE software Echip™ v. 7.0 was used to design and analyze experiments.

In order to reduce the number of experiments, two variables were introduced: Cooling time ($T_e$) and Stretch ratio ($R_a$), which can be defined for a single lip air ring as follows:

$$T_e = \left[ \left( \frac{FLH}{vf - vo} \right) \ln \left( \frac{vf}{vo} \right) \right]$$

$$R_a = \frac{2h}{e(BUR)^2}$$
respectively, where \( v_f \) is the film take-up speed, \( v_o \) is the film speed at die exit, \( h \) is the die lip gap, and \( e \) is film thickness. This relation is deduced from basic principles from mass and momentum conservation and from assuming a linear velocity profile from die exit to FLH, Newtonian materials, and uniform film cooling [3].

To validate the above simplification, several extrusion conditions with different BUR, FLH and thickness – but with similar \( T_e \) and \( R_a \) – were simulated using software for modeling bubble formation [4]. Results from this simulation are shown in Figure 1 for one of the earlier cases evaluated with a larger diameter (200 mm) die. It shows MD and TD deformation as film exits the die and reaches FLH. It can be seen that the deformation pattern (and therefore orientation) are similar despite the fact that different BUR, FLH, and thickness are used in both cases.

**Figure 1.** Numerical simulation of the deformation patterns for two different extrusion conditions that had similar \( T_e \) and \( R_a \). \( (T_e = 16.9 \) and \( R_a = 3.3) \).
Differences between results can be attributed to non-uniform cooling and non-Newtonian material behavior of polyethylene which were not considered in deductions for $T_e$ and $R_a$.

The experimental design allowed developing regression equations for the following variables: extruder specific power and related operating conditions, tensile properties (modulus, strength, and elongation in MD and TD), MD and TD tear resistance, puncture resistance, optical (haze, gloss, and transparency), and MD and TD shrinkage. The properties were fitted to the following model:

\[
\text{Property} = a_0 + a_1 \times \text{HDPE} + a_2 \times \text{LDPE} + a_3 \times \text{CCM} + a_4 \times T_e \\
+ a_5 \times R_a + a_6 \times \text{HDPE} \times \text{LDPE} + a_7 \times \text{HDPE} \times \text{CCM} \\
+ a_8 \times \text{HDPE} \times T_e + a_9 \times \text{HDPE} \times R_a + a_{10} \times \text{LDPE} \times \text{CCM} \\
+ a_{10} \times \text{LDPE} \times T_e + a_{11} \times \text{LDPE} \times R_a + a_{12} \times \text{CCM} \times T_e \\
+ a_{13} \times \text{CCM} \times R_a + a_{14} \times T_e \times R_a
\]

(3)

where $a_i$ are the fitting coefficients, HDPE and LDPE are the HDPE content and LDPE content in the blend, respectively, CCM is an integer variable (0 for no LLDPE, 1 for C4-LLDPE, and 2 for C8-LLDPE), and $T_e$ and $R_a$ are the cooling time and stretch ratio, respectively. All these results were consolidated in a single, easy-to-use Microsoft Excel™ worksheet; however, the same sheet can easily be developed in other spreadsheet programs.

Table 1 lists the resins employed and its basic properties. Blends were prepared in a low intensity mixer (Lodige, model TK-150). Extrusions were done in a tubular film single screw extruder (Dolci, with a 40 mm barrel diameter, 24\,$l$/d, a 3:1 compression ratio screw with a mixing zone, and equipped with a 80 mm diameter die with a die gap of 1.2 mm). Mass flow rate of 30 kg/h was held constant in all cases. Temperature profile in extruder was set at 160/170/180/190/190°C, and 200/210/210°C in the die.

<table>
<thead>
<tr>
<th>Material</th>
<th>MFI (dg/min)</th>
<th>Density (kg/m³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LDPE</td>
<td>0.27</td>
<td>922</td>
</tr>
<tr>
<td>LDPE</td>
<td>0.80</td>
<td>922</td>
</tr>
<tr>
<td>HDPE</td>
<td>0.65</td>
<td>966</td>
</tr>
<tr>
<td>C4-LLDPE</td>
<td>1.00</td>
<td>919</td>
</tr>
<tr>
<td>C8-LLDPE</td>
<td>0.80</td>
<td>918</td>
</tr>
</tbody>
</table>
The design of the experiment comprised of 38 extrusions, which were carried out in a random manner. Extruder was purged in all cases for at least 1 h. When a less viscous blend was added, the screw was removed and cleaned. Samples were taken 30 min after steady conditions were reached.

Film tensile properties were measured in a universal tensile tester (Instron, model 5500) according to ASTM D882. Tear (ASTM D1922) and puncture (ASTM D2582) properties were measured using a Thwing-Albert Elmendorf tear tester. Impact properties were measured under ASTM D1709 in an instrumented falling weight impact tester (Rosand, Type 4). Haze and transparency were measured according to ASTM D1003 in a hazemeter (Haze Gardner, model XL211). Gloss was measured under ASTM D2457 (HunterLab, model Gloss III). Shrink properties were measured in a test bath (Aminco, model 10-276) under ASTM D2732.

RESULTS AND DISCUSSION

For all properties, a good regression coefficient was obtained. In all cases, $R^2 > 0.7$ and in most cases, it exceeded 0.85, considered good given the natural variation in lot quality, machine fluctuations, and measurement uncertainty. Equation (3) was simplified as much as possible for each property, to avoid very complex models that were difficult to use, by setting some $a_i$ to zero when the particular coefficient has little effect on response.

Although the models developed were complex, some general trends can be established. For example, Table 2 shows the expected changes from a base condition ($BUR = 3$, $FLH = 36 \text{ cm}$, die gap $= 1.2 \text{ mm}$, $70 \mu\text{m}$ thickness, $80 \text{ mm}$ die diameter, and $30 \text{ kg/h}$ mass flow rate). This base condition, located near the center of the experimental domain evaluated in this project, was subjected to incremental changes (around 10%) in key variables. It is important to note that this trend may change if another base point is selected.

Figure 2 shows the selector screen employed to analyze all the measured properties. The user inputs the required fields (blend composition, die characteristics, and operating conditions) and the program returns the main variables, including a cost estimate per kg of blend. This selector has been extensively tested at Indesca labs with pilot blends as well as in selected processors using industrial machines. Owing to the good results, the selector is currently used to design blends for specific applications and to choose appropriate operating conditions.
Table 2. Expected changes from a base condition. Horizontal arrows indicate less than $\pm 5\%$ change in property. A single arrow means a 5–10\% change in property, double arrow means a 10–15\% change, triple arrow means greater than 15\% change.

<table>
<thead>
<tr>
<th>Property</th>
<th>Adding 10% HDPE</th>
<th>Adding 10% C4- LLDPE</th>
<th>Adding 10% C8- LLDPE</th>
<th>Increasing BUR by 0.5</th>
<th>Increasing FLH by 10%</th>
<th>Increasing die lip gap by 10%</th>
<th>Increasing thickness by 10%</th>
<th>Increasing $R_a$ by 10%</th>
<th>Increasing $T_e$ by 10%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tensile modulus MD</td>
<td>↑↑↑↑</td>
<td>↓</td>
<td>↓</td>
<td>↑</td>
<td>↑</td>
<td>↓</td>
<td>↓</td>
<td>↑</td>
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</tr>
<tr>
<td>Tensile strength MD</td>
<td>⇐</td>
<td>↑↑↑↑</td>
<td>↑↑↑↑</td>
<td>↑↑↑↑</td>
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<td>↓</td>
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<tr>
<td>Ultimate elongation MD</td>
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<td>↑↑↑↑</td>
<td>↑↑↑↑</td>
<td>↑↑↑↑</td>
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<tr>
<td>Toughness MD</td>
<td>⇐</td>
<td>↑↑↑↑</td>
<td>↑↑↑↑</td>
<td>↑↑↑↑</td>
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<tr>
<td>Tensile modulus TD</td>
<td>↑↑↑↑</td>
<td>⇐</td>
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<tr>
<td>Tensile strength TD</td>
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<tr>
<td>Ultimate elongation TD</td>
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<tr>
<td>Toughness TD</td>
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<tr>
<td>Tear resistance MD</td>
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<tr>
<td>Tear resistance TD</td>
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<tr>
<td>Puncture resistance</td>
<td>⇐</td>
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<tr>
<td>MD shrink</td>
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<td>TD shrink</td>
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<td>Gloss</td>
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<td>Transparency</td>
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<tr>
<td>Haze</td>
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as a starting point when processors look for property improvement. This tool allows establishing conditions which can comprise all the requirements for a specific application (rigidity, toughness, shrink, tear, optical, etc.).

As expected, MD and TD modulus were increased as HDPE was added. On the contrary, LLDPE addition was beneficial for tensile strength properties; however, shrink properties were negatively affected. In turn, operating conditions (increasing BUR, for example) may reduce the impact of LLDPE and/or HDPE addition on shrink properties. This means that compromises might be found for a given application.

Tear properties behaved in a more unexpected manner. Addition of 10% of C4-LLDPE or C8-LLDPE resulted in decrease of puncture resistance and showed no effect in tear properties. However, if the addition level reached 30%, an important increase of tear and puncture properties was observed, which may be related to a synergistic effect (not shown in Table 2). This effect has been found before for similar blends [5].
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REFERENCES


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