Effect of blown-film fabrication variables on mechanical properties and heat sealing performance of polyethylene blends

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Basic mathematical models and response surface graphs have been derived to illustrate the interrelationships between blown-film fabrication variables with heat seal and mechanical properties of 3-mil films made from blends of LLDPE and LDPE.

One of the most challenging activities in the technical service and development of polyolefin films is to relate resin and film fabrication conditions to final film performance in specific end-use applications. Although some work has been done on correlating resin parameters such as melt index, molecular weight distribution, and density to film properties (*1-4*), very little and very old information is available on the effect of fabrication variables on mechanical properties (*5-7*) and heat-seal performance in form/fill/seal (F/F/S) applications.

Heat sealing performance has been related in the past to film surface contamination with grease (oil), powders, surface additives, and corona treatment levels (*5*). Several studies have looked at the heat sealing window of low-density polyethylene (LDPE) films and its relation to sealing parameters (pressure, temperature, and time) (*6, 7*). Others have studied the effect of sealing bar geometry (*8*).

Some more fundamental studies have found that film morphology is not only a result of the polymer characteristics; it also can be strongly affected by shear rate and orientation of the molecules inside the die and during the drawdown process (*9-11*). Also, the quenching rate significantly affects the nucleation and growth of crystals occurring in the blown-film process. For this reason, we believe that the variations in film morphology produced by different blown-film fabrication conditions make an important contribution to film properties.

Most film processors around the world are faced with the challenge of optimizing their plant extrusion process to attain maximum productivity. This means maximizing the blow-up ratio and output (revolutions per minute) while minimizing extrusion temperatures. This combination usually does not lead to optimum film quality. For this reason, many processors unknowingly sacrifice film performance at the expense of productivity.

With all of this in mind, and focusing on Latin American F/F/S markets, this study has a two-fold objective. The first, which is presented in this paper, is to understand the effect of fabrication variables on mechanical and heat sealing properties of film made from linear low-density polyethylene (LLDPE)/LDPE blends currently used throughout Latin America. The second part, not included in this paper, will correlate its effect on end-use performance, utilizing high- and low-quality F/F/S equipment encountered in Latin America.

To minimize the number of trials, we have chosen a Box-Behnken-design experimental methodology to derive mathematical models describing the desired correlations. Response surface plotting was used to obtain graphical representation of the main variables studied.

Experimental

A statistically designed study was devised to determine the interrelationships between LLDPE/LDPE blends, melt temperature, blow-up ratio, and screw speed (revolutions per minute). These independent variables were selected in a low to high range to provide processors with realistic fabrication conditions and resin blend ratios for predicting final film properties. The characteristics of the resins used are shown in **Table I**.

The blown-film extrusion equipment used was a standard LDPE/LLDPE line with microprocessor control. The independent variable settings listed in **Table II** were selected after verifying that all the setting combinations were technically attainable. The range of blends chosen represented common blend ratios used for automatic F/F/S applications.

During the trial, the frostline was allowed to equilibrate at various heights to obtain a stable bubble and realistic run condition. As the fabrication variables and resin blend varied, bubble stability was limited by the design of the fixed dual-air lip system and deep-dish forming cone.

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I. Resin characteristics

	Melt index, g/10 min	Density, g/cc
Octene based LLDPE	1.0	0.920
High-pressure LDPE	0.22	0.919

Slip and anitblock master batch at standard level for F/F/S applications.

II. Experimental design

	Variables settings Low	Medium	High
LDPE/LLDPE blend, %	25	50	75
Melt temp., °F	400	425	450
Blow-up ratio	1.9	2.3	2.7
Screw speed, rpm	40	60	80

III. Full regression analysis results for MD Elmendorf tear

R^2 = 0.90798

Stepwise selection

Variables in model	Coeff.	F-Remove	Variables not in model	P. Corr.	F-Enter
SGTEST.DLX	136.849	138.7164	SGTEST.RPM	0.2213	1.0304
SGTEST.BUR	32.5129	7.8299	SGTEST.MTEMP	0.0977	0.1927
SGTEST.DLXBUR	47.4723	5.5642	SGTEST.DLXRPM	0.2080	0.9042
SGTEST.DLX2	108.063	46.1315	SGTEST.DLXMTEM	0.1292	0.3396
SGTEST.BUR2	-25.0914	2.4871	SGTEST.RPMBUR	0.2192	1.0091
			SGTEST.RPMTEM	0.1772	0.6480
			SGTEST.BURMTEM	0.0835	0.1403
			SGTEST.RPM2	0.0580	0.0674
			SGTEST.MTEM2	0.2328	1.1462

Model fitting results

Independent variable	Coefficient	Std. error	T-value	SL
CONSTANT	309.176794	13.41678	23.0440	0.0000
SGTEST.DLX	136.849361	11.619272	11.7778	0.0000
SGTEST.BUR	32.512948	11.619272	2.7982	0.0108
SGTEST.DLXBUR	47.472323	20.125169	2.3589	0.0281
SGTEST.DLX2	108.063275	15.910343	6.7920	0.0000
SGTEST.BUR2	-25.091403	15.910343	-1.5770	0.1297

R^2 (adj.) = 0.8861

Further analysis of variance for variables in the order fitted

Source	Sum of squares	DF	Mean sq.	F-Ratio	P-Value
SGTEST.DLX	224732.972	1	224732.97	138.72	0.0000
SGTEST.BUR	12685.101	1	12685.10	7.83	0.0108
SGTEST.DLXBUR	9014.486	1	9014.49	5.56	0.0281
SGTEST.DLX2	85249.588	1	85249.59	52.62	0.0000
SGTEST.BUR2	4029.303	1	4029.30	2.49	0.1297
Model	33571.449	5			

Consequently, at the high revolutions per minute, high melt temperatures, and low blow-up ratios, a more stable bubble was obtained by running at a high frostline.

Even though the microprocessor control allowed a precise and efficient control of the extrusion process, conditions were allowed to stabilize for at least 30 minutes before a sample was collected.

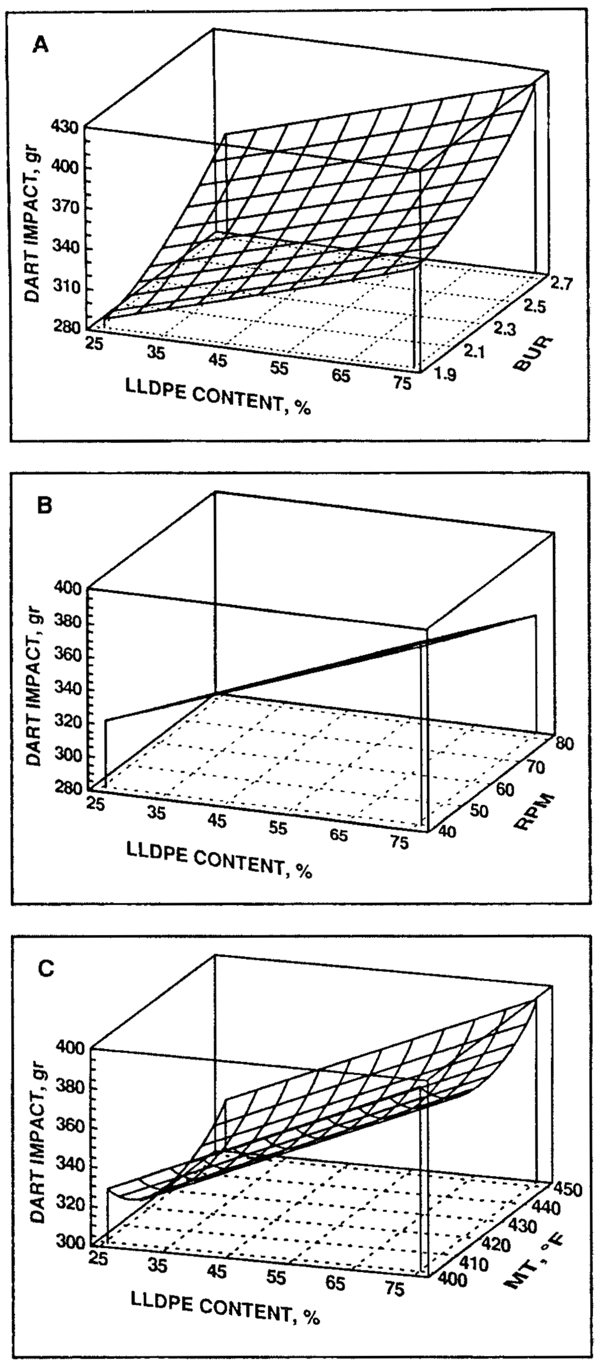
Most of the film properties were measured using American Society for Testing and Materials (ASTM) methods.

Statistical methodology

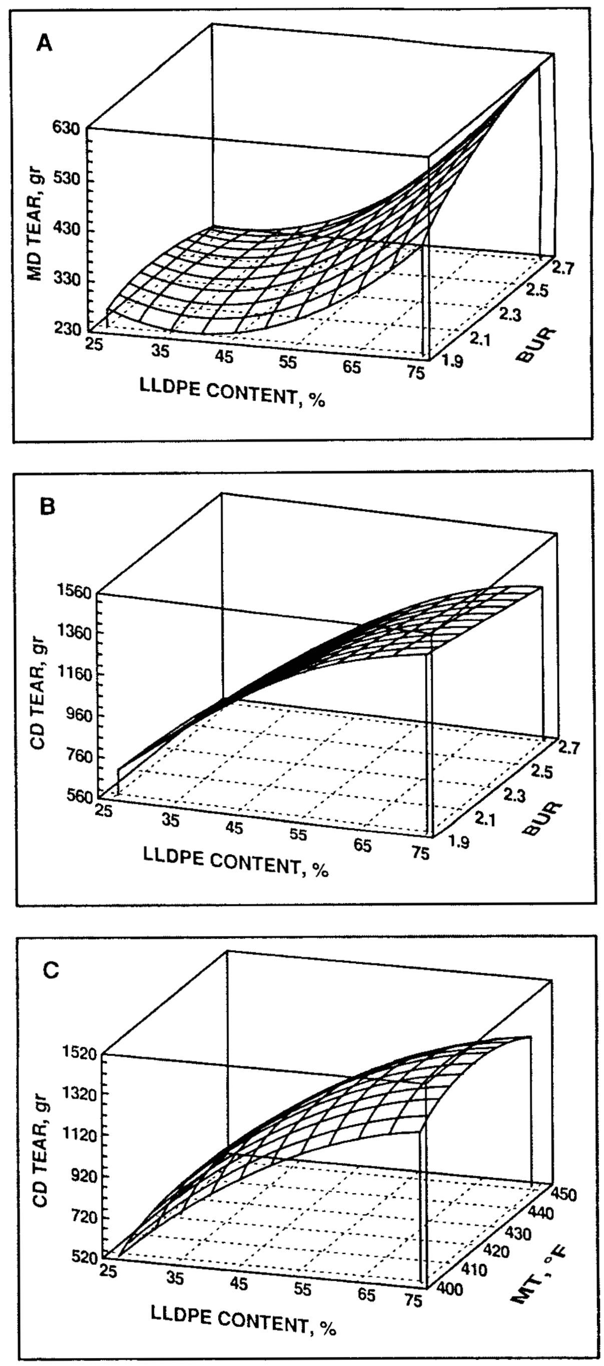
A Box-Behnken designed experiment with four variables at three levels was selected for this study. This design is a portion of a 3^k full-factorial design that allows for the development of empirical mathematical models correlating dependent with independent variables. This design has several advantages:

- Estimates main effects, second order terms and quadratic terms

1. Dart impact (MT = 425°F, RPM = 60 in Fig. A., BUR = 2.3 in Figs. B and C)



2. MD and CD tear (MT = 425°F in Fig. B, RPM = 60 in Figs. B and C, BUR = 2.3 in Fig. C)



- Is a rotatable design (precision response is independent of direction)
- Contains replicates to calculate experimental error.

A total of 24 runs and three center points (replicates) were necessary for this three-level, four-variable design.

This study seeks to understand the effect of four resin and fabrication condition variables (independent variables) on the mechanical and heat sealing properties of the films produced (dependant variables):

1. Blow-up ratio (BUR)
2. Melt temperature (MT)
3. Screw revolutions per minute (RPM)
4. LLDPE/LDPE blend ratio (BR).

Among the key properties studied are: tensile, Elmendorf tear, 2% Secant modulus, dart impact, puncture resistance, optical properties, heat seal strength, and hot tack.

A semirandom experimentation order was devised to avoid bias in the characteristics of the film samples collected. Three of the independent variables used—RPM, BUR, and MT—were randomly ordered because they could be easily changed. The fourth, BR, was not; instead, all samples of the same blend ratio were run together. This was purposely done to avoid the possibility of contamination by continuously and randomly switching from one blend to another. We think that this run order contributed to the quality (blend uniformity) of the samples obtained and significantly shortened the stabilization time of the extruder between samples.

The physical property and heat seal data obtained for the 27 samples collected were analyzed using the Statgraphics software package to obtain full regression fitted models for each dependant variable as a function of the film fabrication variables mentioned. An analysis of variance was made for each model and for each variable in the models to check their validity.

A lack-of-fit test was manually performed for each model, using both the three center points (replicates) data and more complete historical reproducibility data available at our laboratory for all the different physical properties and for the extrusion line used. In this test, we compare the inherent experimental error of the extrusion process and the film testing to the error term estimated by the analysis of variance for the full regression. If the test concluded a significant lack-of-fit, then variables were added or eliminated from the model to improve the fit.

The models (equations) obtained were then used to generate multiple-response surface graphs (three-dimensional plots), which show the simultaneous effect of two dependant variables on a particular property studied while fixing the other two dependant variables at specific values. This type of graphic representation allows a rapid visual indication of the property trends as the film fabrication conditions are varied. For more precise responses, "contour plots" were also made.

Results and discussion

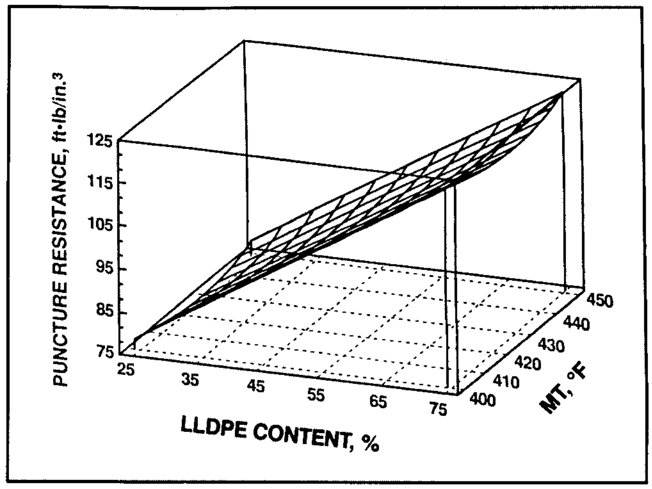
In this section, we will analyze the regression analysis results as they relate to polyethylene extrusion and morphology and to statistical validity.

Regression analysis and analysis of variance results

The R^2 values for most of the variables studied in the regression analysis are between 0.7 and 0.9, indicating the percent of the variation in a specific dependant variable that is explained by the model. Although the closer R^2 gets to 1 (100%) the better, values of 0.7–0.9 can be considered a very good fit of the model to the experimental data.

However, to measure how well a specific model fits in the real world, we need to test some hypothesis on the statistical data. **Table III** contains an example of the full regression analysis for MD Elmendorf tear. The criteria of acceptance of each variable into the model was set at values for the F-test greater than 2.3. This F-value is the ratio of the mean square value of the full regression to that of the residuals (the difference between observed and predicted values). The number obtained is then compared to a probability F-table value, which lets us define the confidence level with which the variation observed in the experimental data is described by a specific variable in the model.

3. Puncture resistance (RPM = 60)



The significance level (SL) or P-value give an idea of the relative importance (impact) of the different terms in the model. A SL value of 0.13 for BUR^2 means that we have 87% confidence that this term is significant to the model. So, the closer the SL value is to zero, the higher the significance level.

Whenever a particular response surface graph is analyzed, it is important to note that the different terms in the models are responsible for the shape of the surfaces. The main effects reflect the linear portion (i.e., Coeff. 1 × RPM, Coeff. 2 × BR) and are represented as flat surfaces. The curvature seen in some response surfaces is given by the quadratic terms (i.e., RPM^2, BR^2) in one or both variables being plotted, and the trends reversal (twisted surfaces) is given by first order interactions of the two variables (RPM × BR). So, depending on the relative SL of each of these contributions, we can assess the relative confidence we should have in the individual trends observed.

Physical property results

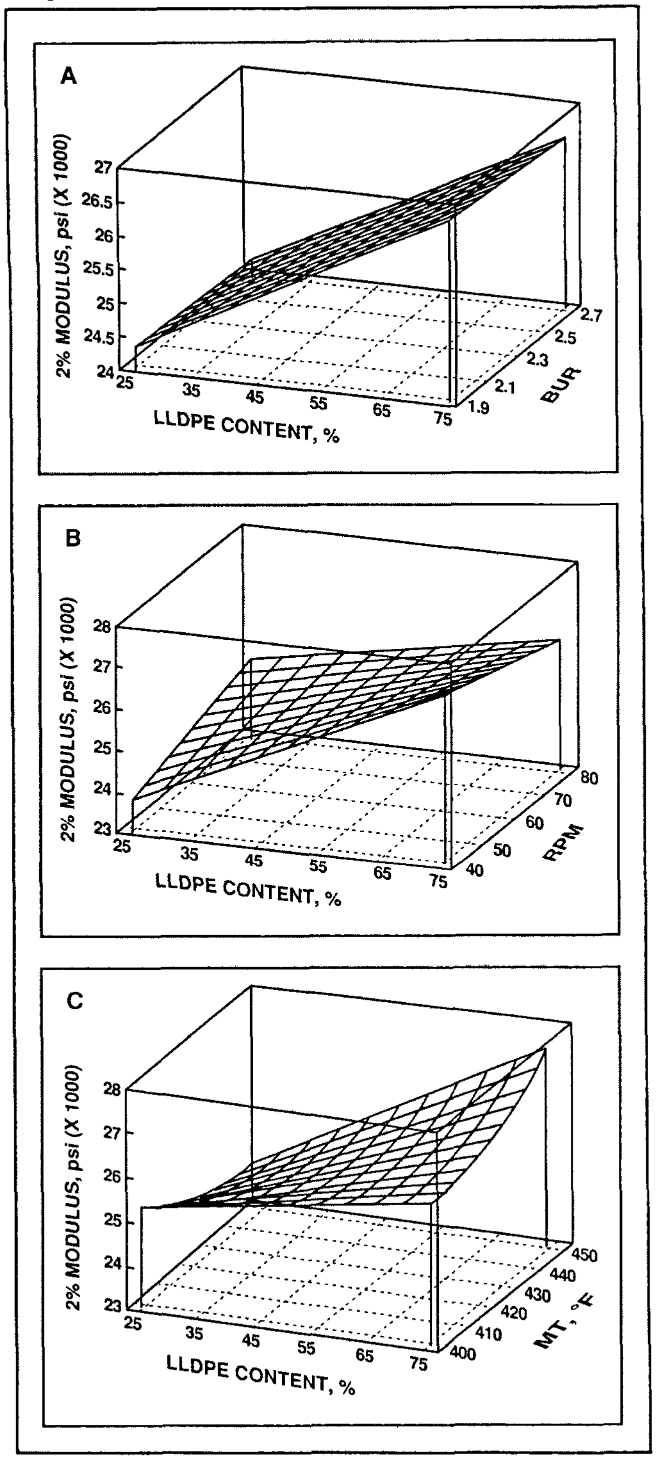
The figures in this paper are a selection of response surface graphs built from the key model equations. These models and graphs represent trends and are only statistically valid in the ranges selected for each variable and for similar types of equipment and resin blends. Any extrapolation to other resins, types of equipment, or extrusion ranges must be handled with care.

By describing and explaining some of the trends observed in the key properties analyzed, we hope to highlight the tremendous amount of information that can be derived from this 27-run experimental design. Each surface graph can only plot two variables at a time. For each model containing a term on each of the four dependant variables studied, we would need up to six surface graphs to fully cover the different variable combinations.

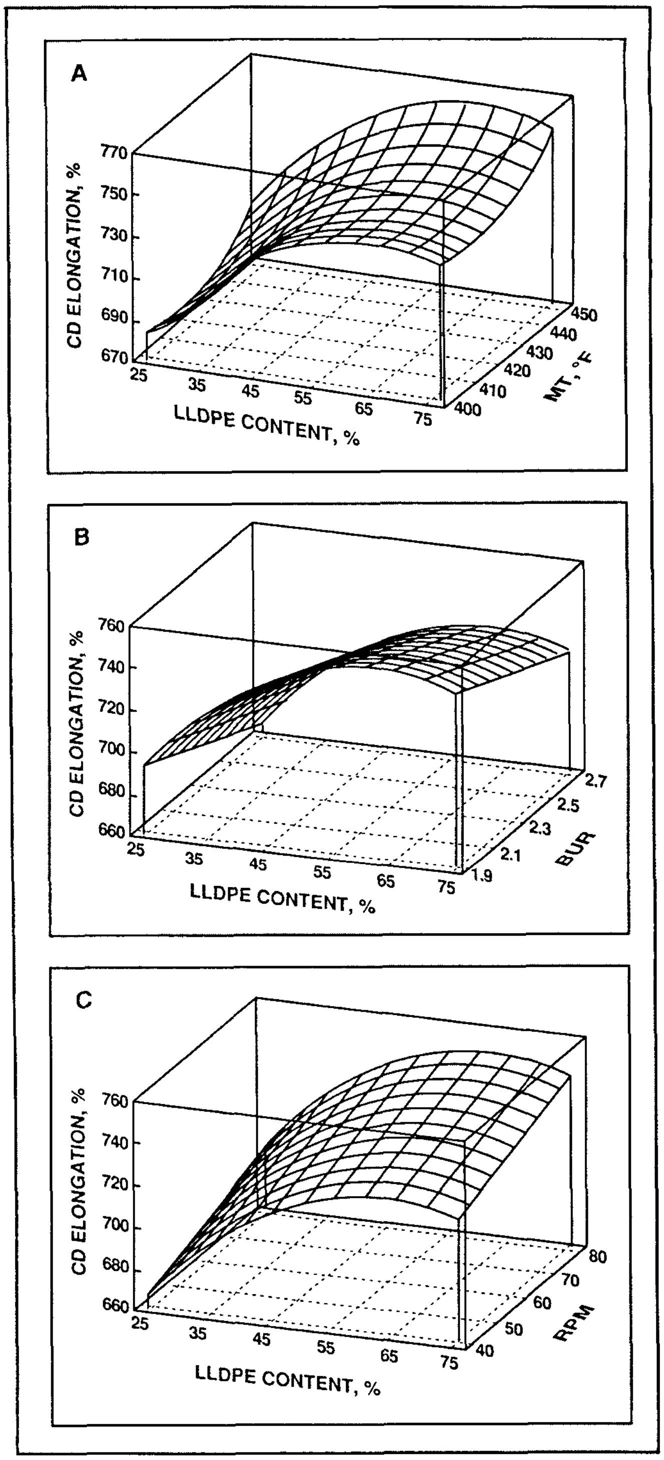
Dart impact (ASTM D-1709)

This is a critical property in many polyethylene film applications. As shown in **Fig. 1A**, dart impact increases approximately 16–18% by increasing the amount of

4. 2% Secant modulus (MT = 425°F in Figs. A and B, RPM = 60 in Figs. A and C, BUR = 2.3 in Figs. B and C)



5. CD elongation (RPM = 60 in Figs. A and B, BUR = 2.3 in Figs. A and C, MT = 425°F in Figs. B and C)



LLDPE in the blend from 25% to 75% or by increasing the BUR from 1.9 to 2.7. Simultaneously increasing both variables has an additive effect, increasing dart impact by about 32%. There does not seem to be any synergism between these two variables.

Figures 1B and 1C show the effect of RPM (output rate) and melt temperature, respectively. We can see that the higher dart impact values are obtained at the low RPM setting for the whole LLDPE/LDPE blends range.

MD Elmendorf tear (ASTM D-1922)

Figure 2A shows the combined effect of LLDPE/LDPE blend ratio and BUR on the MD tear strength of a 3-mil film. We can see that the strongest effect is, as expected, the LLDPE blend ratio. BUR shows to have a different trend depending on the blend ratio. At 25% LLDPE, it drops slightly when increasing the BUR. For 75% LLDPE, the trend reverses, and MD tear increases by 20% by increasing the BUR in the same range.

This trend reversal can be explained by the fact that cooling limitations for the low-BUR sample at 25% LLDPE did not allow us to maintain our frost line height (FLH) in the targeted range of 18 in. to 24 in. Therefore, this particular sample had a higher frost line height, allowing time for the induced orientation to relax and for crystals to grow. In our opinion, if the FLH is maintained the increasing BUR should favor MD tear at any blend ratio.

CD Elmendorf tear

As shown in **Figs. 2B and 2C**, the CD tear is strongly related to the blend ratio and has very little dependance on either BUR or MT.

Puncture resistance (Dow internal method)

This property, as shown in **Fig. 3**, has a linear response with the LLDPE blend ratio. RPM and MT had no significant effect on puncture.

MD 2% Secant modulus (ASTM D-790)

This property, related to the density and crystallinity of the films, is a critical property in F/F/S equipment. As shown in **Fig. 4**, MD modulus increases in a linear way, less than 10%, with increasing LLDPE content. This is due to the slightly higher density of this material compared to the LDPE resin used. MT and RPM seem to have a very small effect on film crystallinity and, therefore, on modulus.

CD elongation at break

In the blown-film process, CD tensile yield strength is usually lower than that in the MD direction because of the lower orientation experienced in this direction. So, whenever a F/F/S pouch filled with a liquid is squeezed, it experiences yield, elongation, and breakage mainly in the CD (weakest) direction. For this reason, CD tensile properties, especially elongation, are considered to give a practical indication of film toughness. This elongation criteria is commonly used by many end users (milk packagers, for instance) in Latin America who do not have film testing facilities. A pouch burst test is performed to evaluate this property.

Figure 5 indicates that CD elongation is maximized by using a 65–75% LLDPE blend at high MT, low BUR, and high RPM settings.

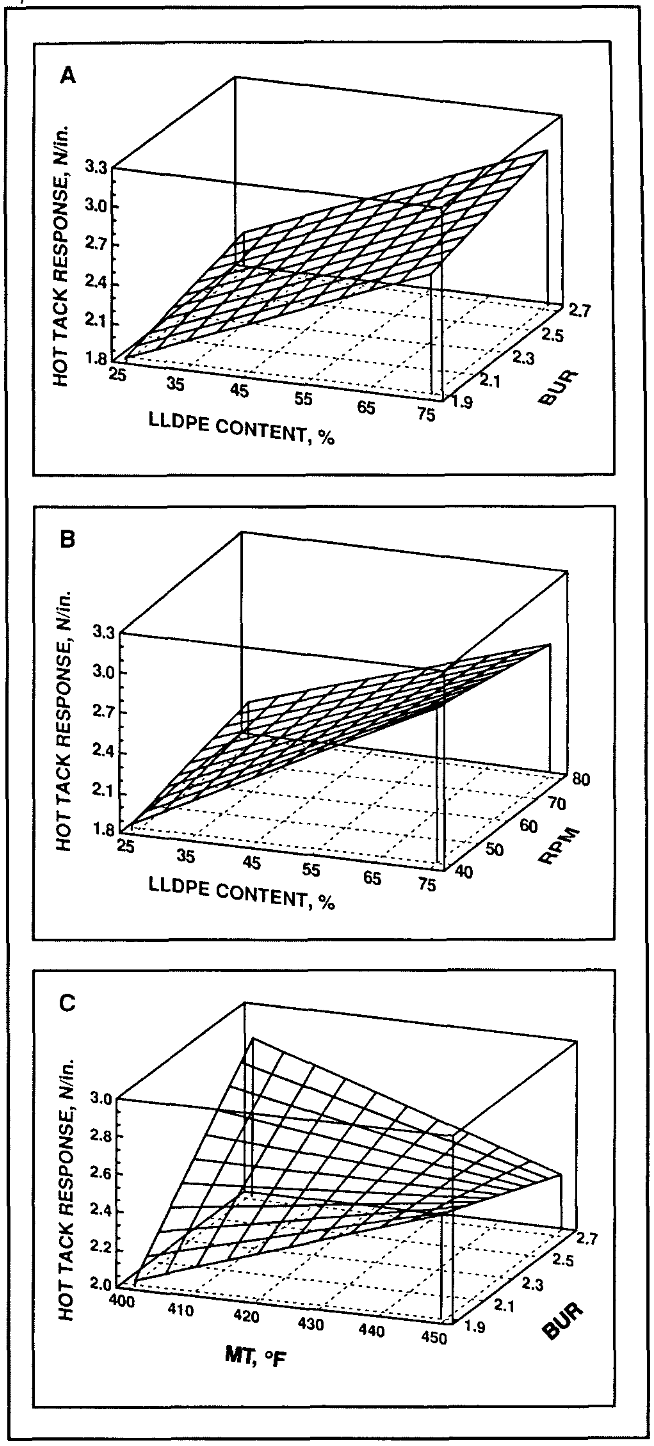
Optical properties

Optical properties are very important in many F/F/S packaging applications for obvious reasons. From our studies of these properties, we concluded that maximum gloss and minimum haze is obtained by the following combination of parameters:

- High LLDPE content
- High BUR
- High MT
- Low RPM.

In this case, optical properties are better for high LLDPE levels since the type of LDPE used in the blends is a fractional melt index, with higher haze and lower gloss than the 1.0-melt index octene LLDPE. Apart from this aspect, the other variables confirm previous trends known for 100% LDPE films.

6. Hot tack response (RPM = 60 in Figs. A and C, MT = 425°F in Figs. A and B, BUR = 2.3 in Fig. B, LLDPE content = 50% in Fig. C)



Heat seal properties

The most critical variable in F/F/S applications is represented by the film hot tack, which measures the ability of a film to offer heat seal resistance immediately after the seal is made. Hot tack is temperature dependent and reaches a maximum at a specific temperature.

To generate a hot tack surface response curve, we need to define a criterion to choose the value that better describes its performance. In this study, we have chosen the hot tack values at 230°F, extracted from the hot tack vs. temperature curves for the 27 samples. Although most polyethylene films do not exhibit the highest hot tack at 230°F, most F/F/S machines are operated at the lowest possible sealing temperatures to maximize line speeds. We selected 230°F because it is 10°F below the maximum hot tack value for most films in this study.

Figure 6 shows the different surface curves for hot tack at 230°F. As we can see, a combination of high LLDPE content, high BUR, and low MT are the ideal settings to obtain maximum hot tack at the lowest sealing temperatures. This same combination of fabrication conditions is favorable to optimize other heat seal parameters such as heat seal strength and maximum hot tack.□

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