Effects of Cast Film Fabrication Variables on Structure Development and Key Stretch Film Properties

J. A. DEGROOT, A. T. DOUGHTY, K. B. STEWART, and R. M. PATEL*

Polyolefins Research and Development, Dow Chemical, USA, Freeport, Texas 77541

SYNOPSIS

The properties of films produced by the cast or blown film processes can be altered by varying the fabrication parameters. An experimental design is used to determine the effect of cast film fabrication variables on the performance of LLDPE stretch films. A three-variable Box–Behnken designed experiment was conducted to study the effects of air gap, melt temperature, and line speed on the key cast stretch film properties. In addition, the differences in molecular orientation in the films were studied using optical birefringence and shrinkage methods. The key film properties are correlated with the fabrication conditions using a statistical analysis program. The results of this study are explained in terms of web tensile stresses before solidification and the degree of molecular orientation developed in the film due to the stresses. © 1994 John Wiley & Sons, Inc.

INTRODUCTION

The two most common methods of film production are the blown film and cast film processes. In the cast film process, a polymer melt is extruded through a flat or slot die. The molten web is "pinned" against a chrome-plated, water-cooled roll by an air knife or vacuum box. The roll "chills" the film instantly and the film is slit and wound. As a result of the very high cooling rates, film of excellent optical quality is obtained. One of the end uses of cast films is pallet wrapping. Films used in this application are called stretch cling films. The pallet wrapping application utilizes a machine, called a stretch wrapper, which prestretches the stretch cling film to as high as 250–300% elongation and wraps it around a pallet of goods. Today, most films in the pallet wrapping application are composed primarily of LLDPE.

Stretch film manufacturers sell a variety of films that offer different benefits to the end-user. Some films focus on high yield, stretching as high as 250–300% on a pallet, whereas other films offer high load retention properties. High load retention films are generally more difficult to stretch but are better able to maintain the integrity of a load. Others tout properties such as better optics, differential or one-sided cling, and high puncture or tear resistance. These different properties can be achieved in several ways: One method is to vary the intrinsic properties of the LLDPE resins used in the films, such as molecular weight and its distribution, and the comonomer, its type, and its distribution, to achieve the desired film properties. Typically, higher melt index or lower average molecular weight resins give higher film extensibility and lower load retention. In the case of coextruded films, the layers of the films can be varied in thickness and composition to achieve different properties.[1-3] Stretch film properties can also be altered by varying the film fabrication parameters.

Several papers have dealt with the effects of fabrication conditions on blown stretch film properties.[4,5] This article focuses on the effects of varying cast film fabrication parameters. A three-variable Box–Behnken-designed experiment was conducted to study the effects of fabrication conditions on the key cast stretch films properties. In addition, the differences in molecular orientation in the films were studied using optical birefringence and shrinkage methods. The key film properties are correlated with the fabrication conditions using a statistical analysis program.

* To whom correspondence should be addressed.

Journal of Applied Polymer Science, Vol. 52, 365–376 (1994)
 CCC 0021-8995/94/030365-12

EQUIPMENT AND MATERIALS

This study was conducted using a solution process ethylene/octene-1 copolymer. The molecular weight and molecular weight distribution of the linear low-density polyethylene (LLDPE) resin were determined using the melt-flow properties, melt index (I_2) and melt flow ratio (I_{10}/I_2), respectively. The value of I_2 was determined by ASTM D-1238 (Condition E, 2.16 kg) and the value for I_{10} was obtained by ASTM D-1238 (Condition N, 10 kg). The resin density was determined using ASTM D-1248. The resin chosen for the study had an I_2 = 2.3 g/10 min, I_{10}/I_2 = 7.6, and a density of 0.917 g/cc. A single blended lot of material was used throughout the entire study.

All films in this study were fabricated on an Egan coextrusion cast film line. The line consisted of a 64 mm (2.5 in.) 24 : 1 length/diameter (L/D) extruder, a 89 mm (3.5 in.) 32 : 1 L/D extruder, and a 51 mm (2.0 in.) 24 : 1 L/D extruder. The polymer streams from the extruders fed through a Dow design A/B/C feedblock into a 762 mm (30 in.) Johnson coat hanger, flex lip die. Although the same resin was run in each extruder to produce the single-component films, the pumping rates of the extruders were adjusted to maintain a 15%/70%/15% film layer ratio throughout the experiment. The die gap was approximately 0.50 mm (0.020 in.). The film contacted two chrome-plated chill rolls that had a finish of 2–4 RMS. An air knife, operating at a constant pressure of 102 mm H_2O (4.0 in. of H_2O), and air jet edge pinners were used to keep the film in contact with the primary chill roll. A CMR 2000 microprocessor was an integral part of the system for controlling and monitoring the equipment conditions. A Fife model OSP-2-40 beta thickness gauge was used to monitor film thickness.

All the film samples were produced at a nominal thickness of 20 microns (0.8 mil) with an overall gauge variation of ±5%. The majority of the film rolls were taken at 457 mm (18 in.) trimmed width; however, at the 191 mm (7.5 in.) air gap, 356 mm (14 in.) rolls were taken due to the large amount of web neck-in. Given the different sample roll widths, a correlation was developed for roll width vs. ultimate stretch and roll width vs. load retention (Figs. 1 and 2). As evidenced by the data, there is essentially no difference in ultimate stretch as the roll width varies. Load retention, on the other hand, increases linearly with roll width. These comparisons were made at the midpoint run conditions of the designed experiment. It was assumed that the same correlations would apply to all run conditions in this study.

EXPERIMENTAL

A Box–Behnken experimental design was conducted to investigate the effect of air gap, melt temperature, and line speed on the stretch performance of the LLDPE films. Each independent variable was evaluated at three response levels. The actual ranges of these response levels are provided in Table I.

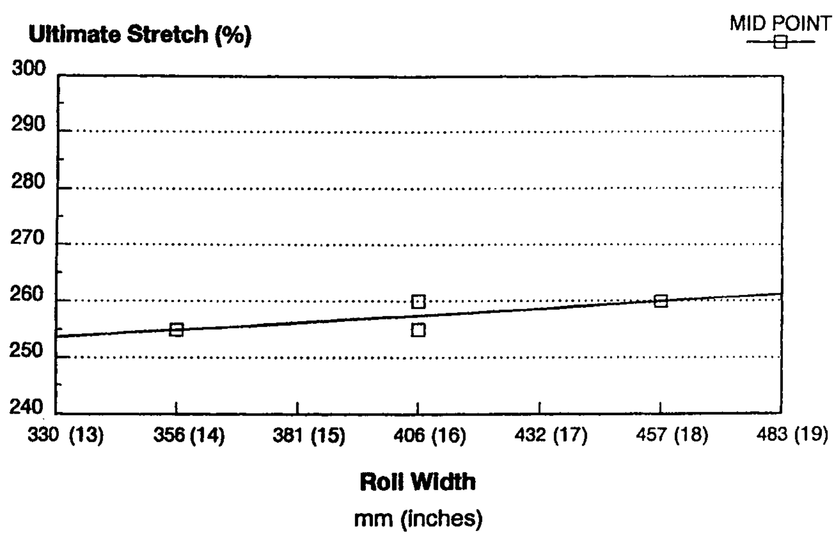

Figure 1 Plot of ultimate stretch vs. roll width.

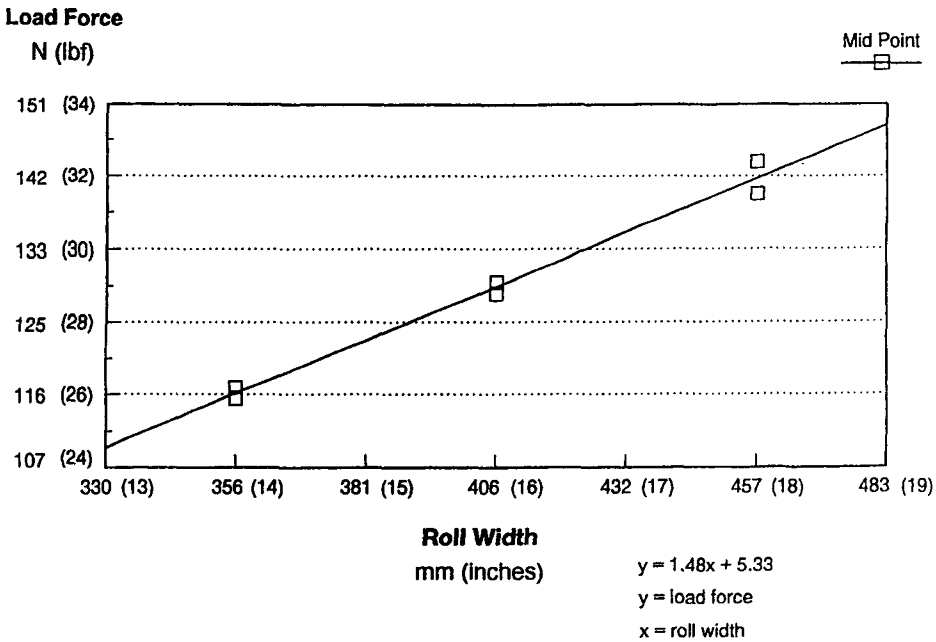

Figure 2 Plot of load retention vs. roll width.

Air gap is defined as the distance the film traverses from its exit at the die to the point it contacts the primary chill roll. During this experiment, the polymer extrudate exited the die vertically and contacted the chill roll at a location described as 9 o'clock, regardless of the air gap (Fig. 3). The film web was held in contact at this position using an air knife and air jet edge pinning. The melt temperatures of the polymer streams exiting the three extruders were held equal to each other to ensure a homogeneous melt temperature entering and, therefore, exiting the die. The die zones were set equal to the desired melt temperature. The extruder barrel zone temperatures were adjusted at different pumping rates and line speeds to maintain the desired melt temperature and a minimum temperature delta across each melt stream. A homogeneous melt temperature per polymer stream is considered to be important to achieving consistent film quality and performance.[6] The melt temperatures of the polymer streams were measured in the three adapter pipes leading into the combining adapter using variable depth melt thermocouples. Melt temperature is defined as the average of five measurements across the melt stream. Line speed was set and controlled by the CMR 2000 microprocessor. A constant 20 micron (0.8 mil) film thickness was achieved by varying the extruder rpm while maintaining the specified layer ratios. All other moveable or variable line parame-

Table I Independent Variable Ranges

	SI Units	English Units
Air gap	63.5–190.5 mm	2.5–7.5 in.
Melt temperature	260–288°C	500–550°F
Line speed	183–305 mpm	600–1000 fpm

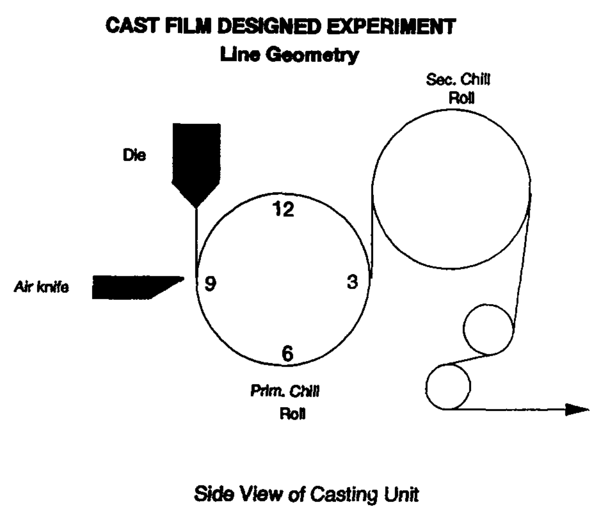

Figure 3 Schematic of the side view of the film casting unit.

Table II Processing, Structure, and Properties Data for Cast Film Designed Experiment

Exp.	Air Gap		Melt Temperature		Line Speed		Ult. Stretch	Load Force		Unst. Cling	Puncture	Elmendorf Tear MD	% MD	
No.	mm	In.	C	F	mpm	fpm	(%)	N	lbf	(g)	(in *lbf)	(g)	Shrink	$\Delta_{12} \times 10^3$
1	127	5	274	525	244	800	260	149	33.6	83	37.8	338	—	—
2	127	5	261	502	305	1000	235	152	34.1	67	36.7	407	—	—
3	127	5	259	498	183	600	257	143	32.1	86	36.1	422	—	—
4	127	5	286	547	183	600	297	137	30.9	99	40.1	317	—	—
5	127	5	288	551	305	1000	277	141	31.8	103	38.4	418	—	—
6	127	5	273	523	244	800	270	145	32.7	87	40.8	322	—	—
7	63.5	2.5	273	523	183	600	220	173	38.8	75	35.1	365	87.2	2.22
8	63.5	2.5	287	548	244	800	220	169	38.1	78	41.6	291	87.4	2.22
9	63.5	2.5	274	526	305	1000	205	191	42.9	55	36.4	359	87.8	2.27
10	63.5	2.5	261	501	244	800	195	196	44.0	52	34.3	358	88.1	2.48
11	127	5	272	522	244	800	265	150	33.7	77	41.2	394	—	—
12	190.5	7.5	287	548	244	800	305	131	29.4	114	39.2	291	84.2	1.41
13	190.5	7.5	273	524	183	600	305	132	29.6	86	40	335	—	—
14	190.5	7.5	274	525	305	1000	275	125	28.1	95	38.3	383	—	—
15	190.5	7.5	257	495	244	800	260	136	30.5	94	40.7	414	84.7	1.68
16	127	5	273	523	244	800	260	144	32.4	80	40	437	—	—

ters were measured and held constant throughout the 16 designed runs.

The key stretch performance properties that were studied included ultimate stretch, load retention, unstretched cling, puncture resistance, and Elmendorf tear. Ultimate stretch or ultimate elongation was determined using a Lantech H-Series power prestretch wrapper. Ultimate stretch is the point at which the film fails between the prestretch rollers as the percentage of prestretch is increased at a constant dancer bar tension. This value is considered to be indicative of the degree of extensibility achievable in pallet-wrapping. Note that the ultimate stretch was measured at very high strain rates, approximately 1600 1/min, as achieved in pallet wrapping. The film elongation at break at low strain rates, as measured by an Instron tensile tester, did not correlate well with the ultimate stretch and was not used.

Load retention or load holding force determines the ability of a film to maintain the integrity of a load. Load force was determined by prestretching the films to 200% and then further stretching them to 225% total on-pallet elongation onto an angle iron frame to which a Revere HPS load cell was attached. Three layers of film were wrapped around the frame and the force exerted on the load cell was recorded at 0, 1, 5, and 10 min. The decay in load force reached an approximate equilibrium between 5 and 10 min; therefore, 10 min was designated as the final load force value. The cling of the film samples was determined according to ASTM D-4649. Puncture resistance was measured using the ASTM test (probe size = 0.5 in.), and Elmendorf tear was determined by ASTM D-1922.

Among a variety of experimental techniques, optical birefringence and shrinkage measurements are relatively quick and easy methods for characterizing orientation in cast and blown films. Birefringence gives a measure of total, both crystalline and amorphous, orientation in the sample,[7] whereas shrinkage can be taken primarily as a measure of amorphous segment orientation and amorphous chain extension. Film shrinkage is due to the orientation frozen into the film as a result of the stresses the polymer experiences during fabrication. Samuels[7] showed that the shrinkage of poly(ethylene terephthalate) fiber was proportional to the orientation in the amorphous phase. Geleji et al.[8] showed that in the overall context tensile strength, elongation, and shrinkage depend on amorphous orientation. Hence, shrinkage can be taken as amorphous segment orientation. Shrinkage measurements involve the unrestrained melting of cast film samples to induce relaxation of the oriented (and extended) amorphous segments, which ultimately results in reduced film dimensions. During shrinkage, some slippage of the molecules may also occur; therefore, the shrinkage method is not an absolute indication of the original amount of amorphous orientation. Nevertheless, shrinkage measurements are still considered useful for comparing amorphous orientation in films. Film shrinkage was measured by applying a thin layer of Dow Corning 200 silicon oil on a tray and placing 4 × 4 in. square samples on it. The tray was kept in a forced, hot air oven for 10 min, and from the final film dimensions, percent machine direction (MD) and cross direction CD shrinkage were calculated. Since cast films are primarily uniaxially oriented, only MD shrinkage is of relevance and only in-plane birefringence (Δ_{12}) needs to be measured. The in-plane birefringence was measured using a polarizing microscope, the Senarmont compensator, and a green filter.

Table III Analysis of Variance (ANOVA) Regression

	Sum of Squares	DF	*F*-Ratio	*P*-Value
Ultimate stretch				
A: Melt temp	2957.9	1	119.8	0.0000
B: Air gap	11974.3	1	484.9	0.0000
C: Line speed	1154.9	1	46.8	0.0000
BB	1142.9	1	46.3	0.0000
Total error	271.6	11	—	—
R-squared = .98; adjusted *R*-squared = .98				
Load force				
A: Melt temp	15.1	1	23.2	0.0010
B: Air gap	263.5	1	404.5	0.0000
C: Line speed	4.9	1	7.5	0.0229
AB	6	1	9.2	0.0141
BC	8.7	1	13.4	0.0053
BB	25.8	1	39.7	0.0001
Total error	5.9	9	—	—
R-squared = .98; adjusted *R*-squared = .97				
Unstretched cling				
A: Melt temp	1247.6	1	40.5	0.0001
B: Air gap	2032.5	1	66	0.0000
C: Line speed	133.6	1	4.3	0.0639
BC	217.4	1	7.1	0.024
AA	267	1	8.67	0.0147
Total error	308.1	10	—	—
R-squared = .92; adjusted *R*-squared = .89				

RESULTS AND DISCUSSION

The data from the 16 experimental runs were analyzed using a statistical analysis program. Table II provides the ultimate stretch, load force, cling, and clarity values for each of the runs. The four center points of the experiment illustrate the excellent reproducibility achieved during this experiment. The reproducibility of these points and the other samples in the experiment resulted in very good correlations ($R^2 > .90$) between the independent and dependent variables.

The regression analysis or analysis of variance (ANOVA) data for ultimate stretch in Table III shows that the most influential variables are air gap and melt temperature, followed by line speed and a quadratic effect of air gap. The effect of air gap, melt temperature, and line speed on the ultimate stretch values are shown graphically in Figures 4 and 5. The range of ultimate stretch values in this experiment varied from 195% at a 63.5 mm (2.5 in.) air gap, 266°C (500°F) melt temperature, and 244 mpm (800 fpm) line speed to 305% at a 191 mm (7.5 in.) air gap, 288°C (550°F) melt temperature, and 244 mpm (800 fpm) line speed. This is a significant variation in extensibility and can greatly affect the performance of a film on-pallet. Films with higher ultimate stretch can be stretched on-pallet to greater percentages without breaking. This is highly desirable for applications in which minimal film usage and minimal operator interaction are very important. It can be seen from Figures 4 and 5 that as air gap and melt temperature increased the film extensibility increased. An increase in line speed, however, resulted in a decrease in film extensibility.

As one might expect, there are trade-offs to a highly extensible film. The main trade-off is the amount of load holding force that a film is able to exert on the pallet of wrapped goods. The ANOVA data for load retention (Table III) again show that air gap and melt temperature were the most significant variables affecting load force, followed by line speed. The R^2 value of .96 suggests that the model is a very good fit for the data. Air gap, melt temperature, and line speed had the opposite effect on load retention to which they had on ultimate stretch (Figs. 6 and 7).

The in-plane birefringence and machine direction (MD) shrinkage data of the samples produced at the extreme fabrication conditions and having extremes of properties are tabulated in Table II. The birefringence values are the average of three measurements and shrinkage values are the average of two measurements. It can be seen from Table II that MD shrinkage varied from about 84 to 88% for the

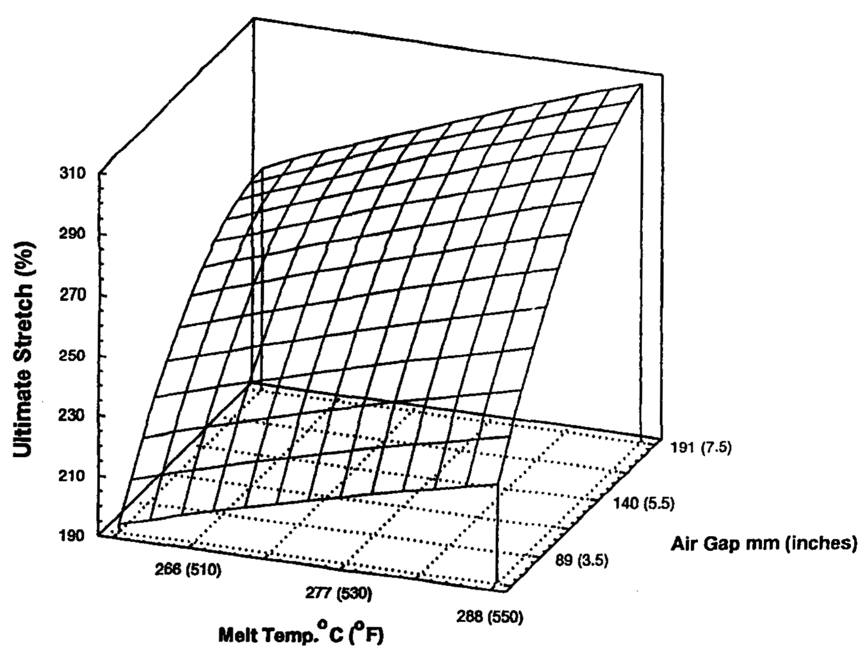

Figure 4 The 3-D surface plot of ultimate stretch as a function of melt temperature and air gap.

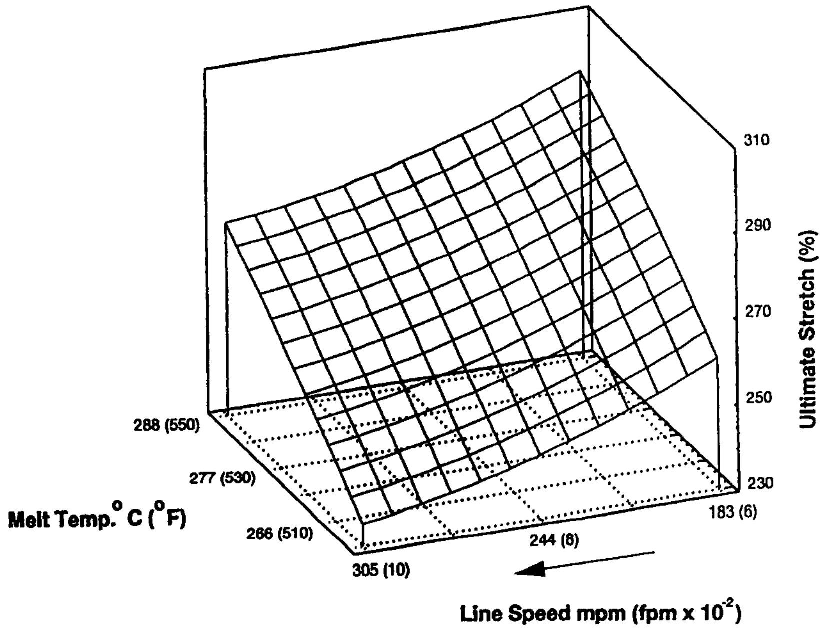

Figure 5 The 3-D surface plot of ultimate stretch as a function of melt temperature and line speed.

samples. It is interesting to note that only about a 4% change in MD shrinkage was observed for the samples with extremes of properties. Nevertheless, the shrinkage data were reproducible and correlated very well with the key film properties as explained below. The plot of percent MD shrinkage as a function of melt temperature at two air gaps is shown in Figure 8. It can be seen that at a given melt tem-

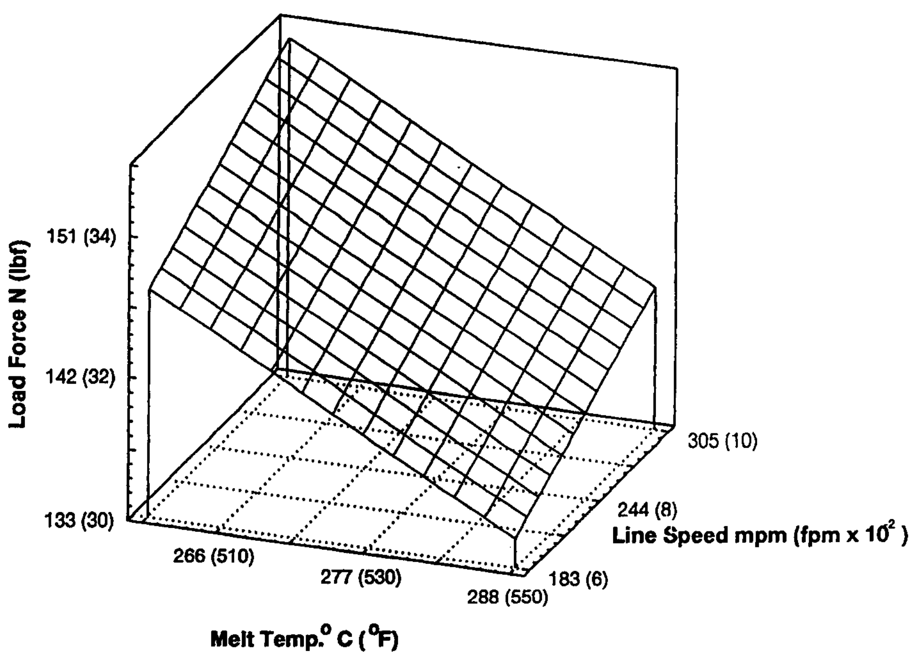

Figure 6 The 3-D surface plot of load retention as a function of melt temperature and line speed.

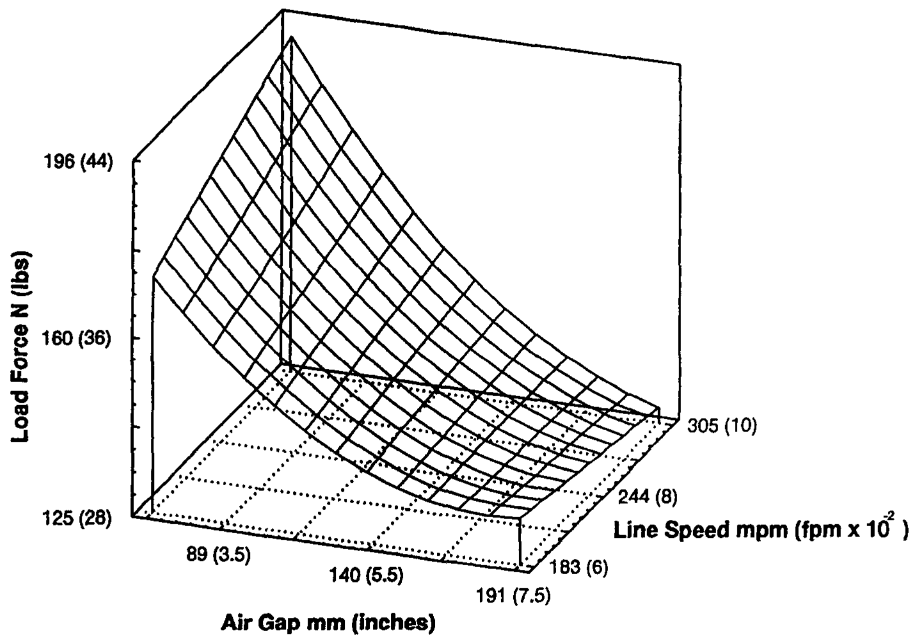

Figure 7 The 3-D surface plot of load retention as a function of air gap and line speed.

perature a larger air gap resulted in significantly lower MD shrinkage. For a given air gap, an increase in melt temperature resulted in slightly lower MD shrinkage. Table II shows that the films with higher MD shrinkage tended to give higher in-plane birefringence. This is because a higher uniaxial web stress results in higher orientation of both the amorphous and the crystalline segments in the MD of the cast film.[9]

The effect of fabrication variables on the degree of molecular orientation developed in the films and on ultimate stretch and load retention can be ex-

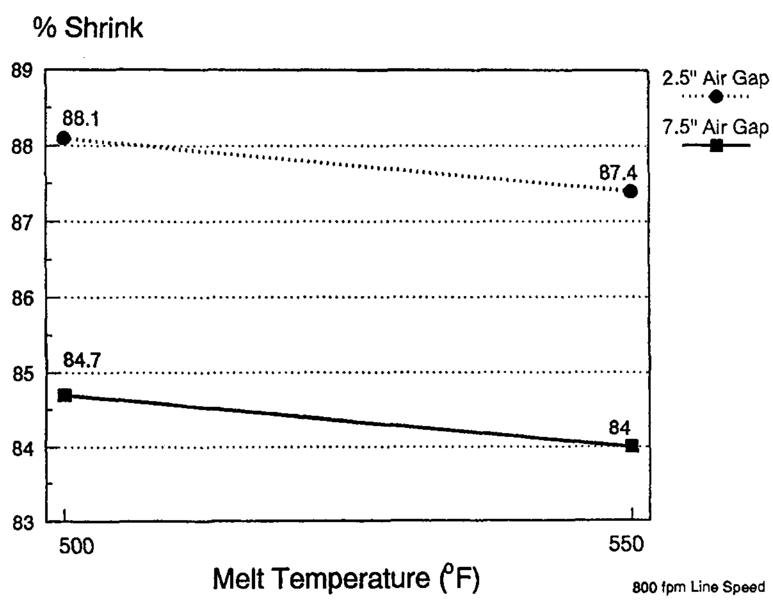

Figure 8 Percent MD shrinkage as a function of melt temperature at two air gaps.

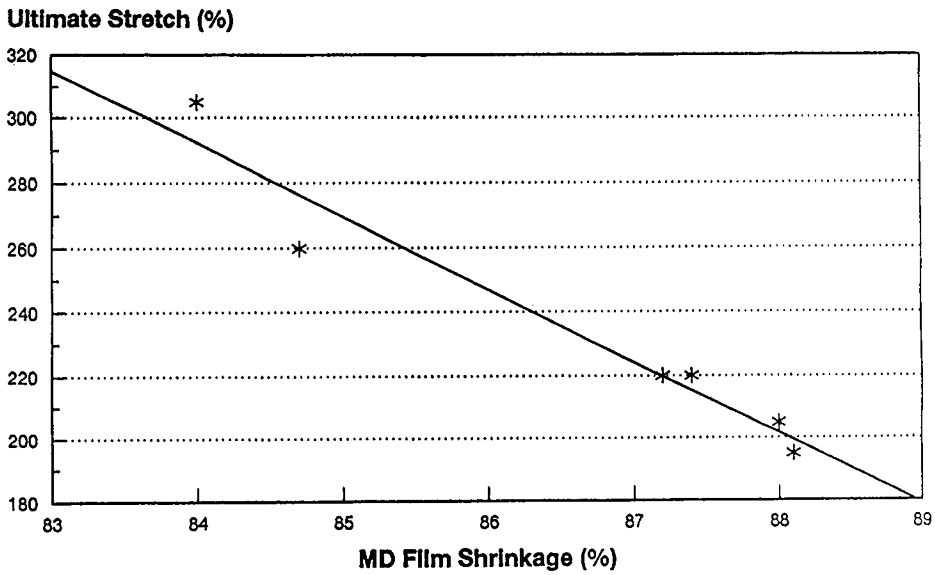

Figure 9 Plot of ultimate stretch vs. MD shrinkage.

plained in terms of the amount of stresses in the molten web just before solidification. A higher melt temperature results in a lower polymer melt viscosity and lower tensile, or extensional, stresses in the molten web. This would result in lower amorphous and crystalline phase orientation in the film, thereby, giving higher extensibility. Likewise, a larger air gap would give lower elongational rates and, hence, lower extensional stresses in the molten web due to the viscoelasticity of the molten polymer.[10] This would also result in lower orientation in the film, thereby giving higher extensibility but lower load retention. A smaller air gap and a lower melt temperature, as well as a faster line speed, induce higher web tensile stresses and higher orientation in the film as explained above. This translates into

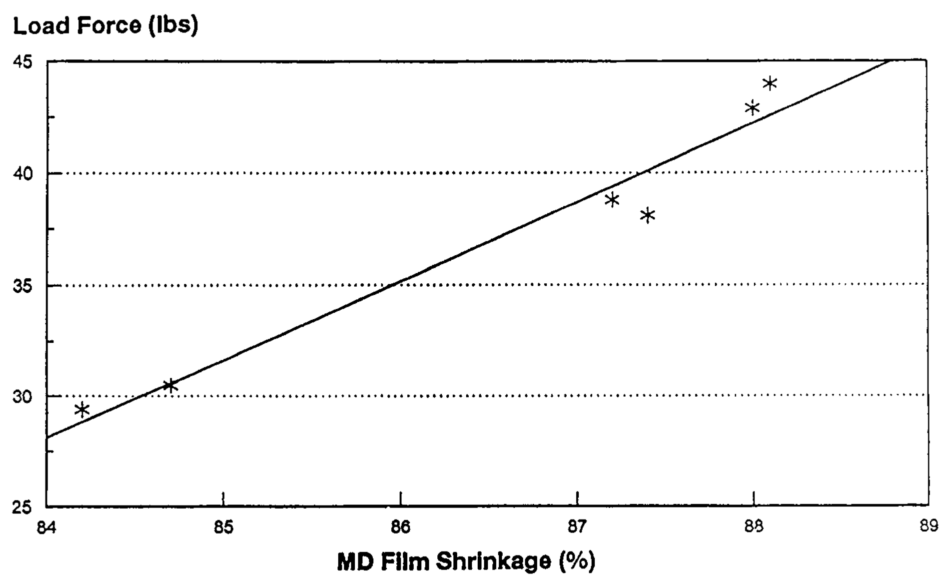

Figure 10 Plot of load retention vs. MD shrinkage.

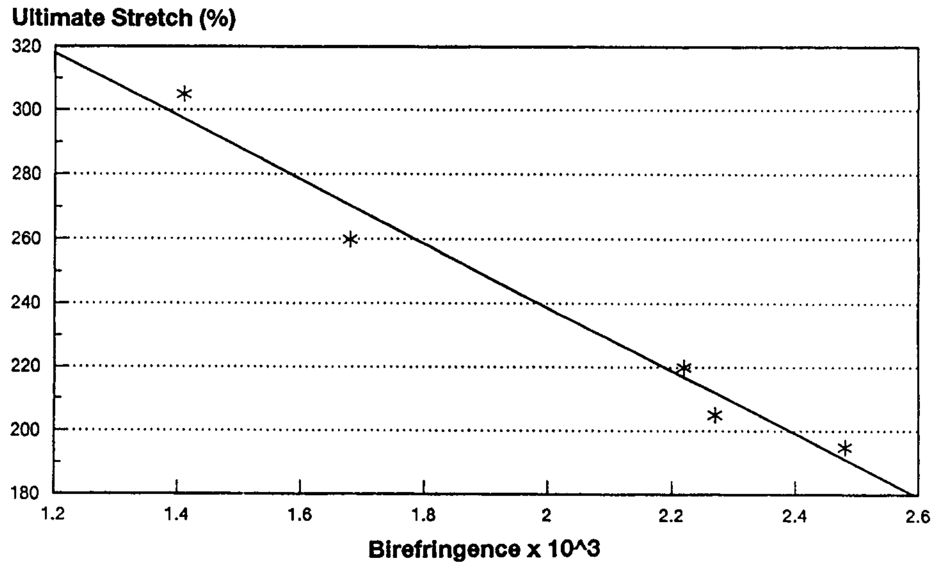

Figure 11 Plot of ultimate stretch vs. in-plane birefringence.

a film that is more resistant to stretching but provides greater integrity to the load being wrapped due to higher load retention.

Note that the orientation developed in the die is minor compared to the orientation induced by melt drawing. The shear flow that occurs in the die is referred to as "weak flow"[10,11] and it induces minimal orientation. Deformations that can generate a high degree of stretching and orientation are referred to as "strong flows."[10,11] The extensional flow occurring between the die and chilled roll is a "strong flow"[11] and it develops most of the orientation in the web. Any orientation developed in the die would be overridden by the extensional flow. Thus, the orientation developed in the die does not influence the orientation finally frozen into the film.[12,13]

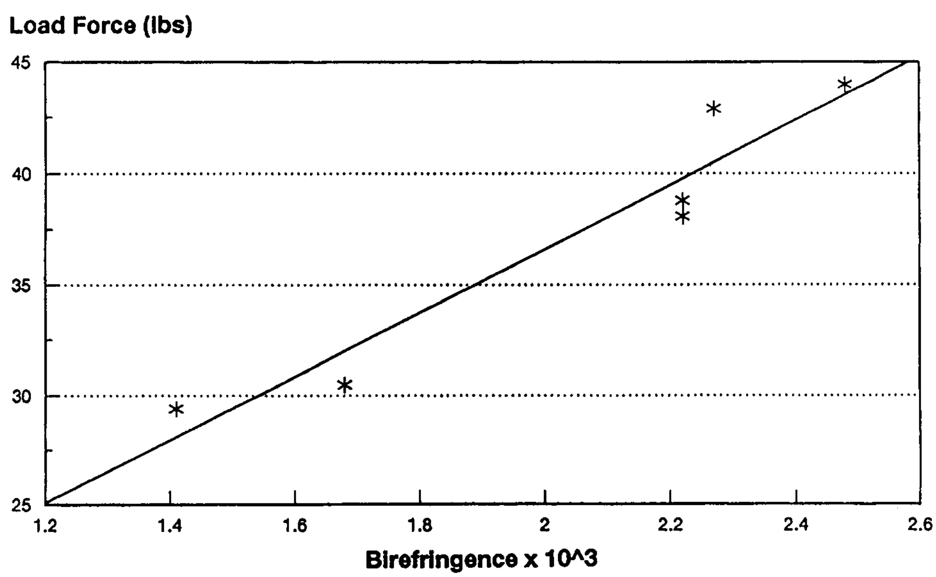

Figure 12 Plot of load retention vs. in-plane birefringence.

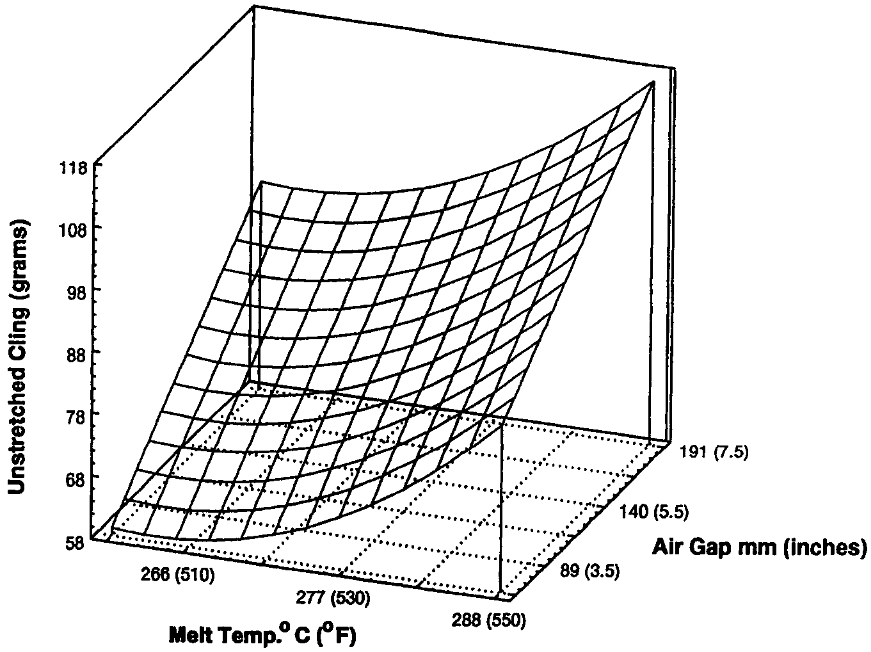

Figure 13 The 3-D surface plot of unstretched cling as a function of melt temperature and air gap.

It is the stresses in the molten web just before solidification that determine both amorphous and crystalline phase orientation. For semicrystalline films, orientation in crystalline and amorphous phases can be, and usually is, different. A higher stress just before web solidification would lead to higher chain orientation in both amorphous and crystalline phases.[9] Note that for amorphous chain segments it is the chain extension that leads to orientation. A higher amorphous phase orientation or chain extension, as measured by MD shrinkage, would lead to lower film extensibility and higher load retention. Plots of ultimate stretch and load retention as a function of MD shrinkage are shown as Figures 9 and 10, respectively. It can be seen that lower ultimate stretch and higher load retention values were obtained at higher MD shrinkage. Similar correlations were also observed using in-plane birefringence data as seen from Figures 11 and 12.

Another property that is very critical to the on-pallet performance of cast stretch films is the amount of film surface cling. Cling can be achieved by adding a tackifying agent, such as polybutene (PIB), or it can be inherent in the resins used to make the film. The inherent cling of stretch resins can vary significantly depending upon the intrinsic properties of the resin.[14] The resin chosen for this experiment typically has low inherent cling; however, during the designed experiment, the cling was altered drastically (50–110 g) by merely changing the fabrication parameters. As shown by the regression analysis data in Table III, the cling was most affected by air gap and melt temperature. When the air gap and melt temperature are increased, the unstretched cling also increases (Fig. 13). Raising the melt temperature and increasing the air gap may facilitate easier migration of the inherent cling material to the surface of the film.

The puncture resistance and MD Elmendorf tear values for the experimental runs are provided in Table III. The puncture values ranged from 3.9 J (34.3 in.-lb) to 4.7 J (41.6 in.-lb), whereas Elmendorf tear varied from 291 to 437 g. Statistical analysis of the data resulted in no correlation between the independent variables and the puncture and tear properties. However, high puncture and tear resistance are characteristics typical of films made from ethylene/octene-1 LLDPE resins.[15]

CONCLUSIONS

Based on the results of the cast film designed experiment, the following can be concluded:

(a) Ultimate stretch and load retention are primarily affected by melt temperature and air gap, with line speed as a secondary factor. A higher melt temperature, larger air gap, and slower line speed result in greater film extensibility and lower load retention.

(b) The effects of fabrication parameters on key stretch film properties can be explained in terms of web tensile stresses at solidification and the amorphous orientation developed in the stretch film due to the stresses. MD shrinkage and optical birefringence measurements correlated very well with the ultimate stretch and the load retention of the film samples.

(c) Model correlation coefficients (R^2) ranged from .92 to .99, indicating that the data fit the models with minimal error.

(d) Unstretched cling is directly proportional to both melt temperature and air gap. Careful control of these variables are essential to achieving consistent cling.

This study has also demonstrated that one can achieve a wide variety of stretch film properties with a single resin by altering the cast film fabrication conditions. Determining the optimum fabrication conditions, however, depends upon the desired stretch film properties as well as the limitations of the film manufacturer's fabrication equipment.

REFERENCES

1. G. W. Wilson, U.S. Pat. 5019315 (May 28, 1991).
2. W. Briggs et al., U.S. Pat. 4399180 (Aug., 1983).
3. W. Briggs et al., U.S. Pat. 4418114 (Nov., 1983).
4. G. Panagopoulos et al., in *The Effect of Blown Film Fabrication Variables on Cling and Stretch Film Performance, ANTEC,* 1991, 1345–1350.
5. P. M. German, in *Polymers, Laminations, and Coatings Conference, TAPPI,* 1988, pp. 445–495.
6. T. I. Butler and S. E. Pirtle, in *Polymers, Laminations, and Coatings Conference, TAPPI,* 1991, pp. 87–112.
7. R. J. Samuels, *Structured Polymer Properties,* Wiley, New York, 1974.
8. F. Geleji, L. Koezy, L. Fulop, and G. Bodor, *J. Polym. Sci. Symp.,* **58,** 253–273 (1977).
9. A. Keller and M. J. Machin, *J. Macromol. Sci.,* **B1,** 41 (1967).
10. J. M. Dealy and K. F. Wissbrun, *Melt Rheology and Its Role in Plastics Processing,* Van Nostrand Reinhold, New York, 1990.
11. R. I. Tanner and R. R. Huilgol, *Rheo. Acta,* **14,** 959 (1975).
12. N. D. Huck and P. L. Clegg, *SPE Trans.,* **1,** 121 (1961).
13. J. J. C. Picot, *Polym. Eng. Sci.,* **24,** 415 (1984).
14. K. A. Simmons, Eur. Pat. Appl. 0287272 (Oct. 19, 1988).
15. L. D. Cady, *Plast. Eng.,* **January,** 25 (1987).

Received June 30, 1993
Accepted October 5, 1993