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Research on Polyethylene Liner Films

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ABSTRACT

This paper describes the evaluation of several polyethylene films that are suitable as liners for baby diapers. The chemical and physical characteristics of the films are discussed. Also discussed are polyblends as a route to films having unusual and superior mechanical properties.

INTRODUCTION

In looking at prospects for polymers in the future, Usmani [1, 2] forecast that polymers made from the simplest hydrocarbon precursors (such as ethylene, propylene, other olefins) will grow in total usage at a higher rate than the more complex and higher costing polymers (such as ABS and nylon). Growth of ethylene and propylene polymers will be supported by intensive research to improve the physical properties, performance characteristics, and processing of these lowest cost polymers via filling, polyblending, grafting, cross-linking, etc.

Polyethylenes [3] prepared by mass polymerization at high temperature and high pressure with peroxide catalysts (low density polyethylenes) are highly branched and contain about 30 branches per 1000 carbon atoms. They have a density of ~ 0.92 g/cm³, a melting point

of $\sim 115^{\circ}\text{C}$, a crystallinity of $\sim 40\%$, and a heat fusion of ~ 12 cal/g. On the other hand, polyethylenes polymerized at low temperature and low pressure, using the Ziegler or Phillips catalyst (high density polyethylenes), have much greater linearity and contain on an average < 1 branch per 1000 carbon atoms. They have a density of ~ 0.96 g/cm³, a melting point of $\sim 135^{\circ}\text{C}$, a crystallinity of $> 95\%$, and a heat of fusion of ~ 48 cal/g. Recently, Union Carbide's new low pressure process has made it possible to produce LDPE at low pressures and temperatures by copolymerizing ethylene with various α -olefins. Very few or no long-chain branchings are produced in the resulting resins.

A lightly white pigmented low density polyethylene film is generally used as a liner for baby diapers. This paper describes the evaluation of several polyethylene films to determine their suitability as liners for baby diapers. Special emphasis has been placed on high speed tensile data and film orientation. Finally, we discuss polyblends as a route to films having unusual and superior mechanical properties.

EXPERIMENTAL WORK ON POLYETHYLENE RESINS

Besides providing a moisture barrier and improved appearance, the liner film should possess excellent impact strength, puncture resistance, tensile strength, and tensile elongation.

Seven low density polyethylene resins were chosen for developmental work. The following analysis were made: Melt index (MI), density, differential scanning calorimetry (DSC) measurement, vinyl acetate content (VA) determination, infrared spectroscopic analysis, and gel permeation chromatographic (GPC) analysis.

Melt Index

The melt indexes of the resins were measured, in triplicate, by ASTM-1238 using Monsanto's capillary rheometer #7288 at 190°C under a 2160 g static loading (see Table 1).

Density

The resins were compression molded and the densities were obtained by the displacement method in a silicone fluid of 0.7579 density at 25°C (see Table 1).

Vinyl Acetate Content Determination

Infrared examination of the seven resins revealed that two of them contained vinyl acetate. Therefore, the VA content was determined on two resins by wet chemistry as follows.

TABLE 1. Melt Index, Density, and VA Content of Polyethylene Resins

Resin	MI (g/10 min)	Density (g/cm ³)	VA (%)
A	2.200	0.9185	0.00
B	2.170	0.9179	0.00
C	1.250	0.9203	3.25
D	0.795	0.9172	0.00
E	0.904	0.9201	0.00
F	1.150	0.9169	4.90
G	0.942	0.9212	0.00

In a 500-mL Erlenmeyer flask, an accurately weighed 2-g sample was placed. Then 100 mL of toluene was added and the mixture was heated to 80-90°C for the resin to go into solution. The mixture was cooled to 50°C and then 50 mL of a standardized 0.5 N KOH (alcoholic, 1:1 methanol/ethanol) was added. The mixture was refluxed with constant stirring for 2 h. It was then cooled and titrated while still warm with a standardized 0.5 N methanolic HCl solution using phenolphthalein as the indicator. A blank was also run. The VA content was calculated as follows (see results in Table 1):

$$\begin{aligned} \% \text{ VA} &= \frac{(\text{mEq KOH} - \text{mEq HCl}) \times 8.61}{\text{sample weight}} \\ &= \frac{[(\text{mL KOH} \times \underline{N} \text{ KOH}) - (\text{mL HCl} \times \underline{N} \text{ HCl})] \times 8.61}{\text{sample weight}} \end{aligned}$$

DSC Measurements

DSC was used to determine the melting point, T_m , and the heat of fusion, ΔH_f , on all polyethylene resins. The results of DSC measurements are shown in Table 2. Note the lower T_m of Resins C and F due to their VA content.

GPC Measurements

GPC chromatograms of polyethylenes were run in trichlorobenzene at 147°C and 0.5% concentration. Calculated values (Table 3) are based on polystyrene's standard angstrom extended chain length.

TABLE 2. DSC Results on Polyethylene Resins

	A	B	C	D	E	F	G
Cycle 1:							
T_m ($^{\circ}\text{C}$)	106	106	98	106	110	100	109
$T_{\text{crystallization}}$	90	90	83	89	94	83	93
ΔH_f (cal/g)	18.5	16.2	13.2	18.4	18.8	13.6	18.0
Cycle 2:							
T_m ($^{\circ}\text{C}$)	107	107	101	106	110	101	109
$T_{\text{crystallization}}$	90	90	83	89	94	83	93
ΔH_f (cal/g)	17.8	16.7	12.7	17.2	18.6	13.0	17.1

TABLE 3. GPC Data on Polyethylenes

Resin	Average \AA extended chain length		
	Weight average, \bar{A}_w	Number average, \bar{A}_n	Dispersity (\bar{A}_w/\bar{A}_n)
A	5240	726	7.21
B	5230	687	7.61
C	4720	756	6.24
D	6220	544	11.40
E	5330	673	7.92
F	4220	750	5.62
G	5210	716	7.28

COMPOSITION AND CHARACTERIZATION OF FILMS

Composition

Polyethylene resins were compounded with SiO_2 and TiO_2 , and 2.5 mil thick films were made therefrom (see Table 4). It was found that

TABLE 4. Composition of Liner Films

Film	Filler composition (%)	
	TiO ₂	SiO ₂
A	3.68	1.06
B	3.60	0.71
C	3.55	0.72
D	3.53	0.34
E	4.30	0.65
F	4.12	0.88
G	3.28	1.35

TiO₂ filler decreases the strength properties of polyethylene and therefore a TiO₂ concentration > 4% is not desirable.

High-Speed Tensile Data

Eight-ply test samples were stretched at 1090%/s using a 1-in. gauge length with ultimate elongation at 6 in. to determine yield stress (σ_Y), fail stress (σ_F), and elongation at fail (see Table 5). Films A and G appear to be deficient in elongation at these rates. The other films are very similar and consistent as evidenced by the very small standard deviations.

Film Orientation

The mechanical properties of polyethylene films, and particularly toughness, depend upon a balanced orientation. We therefore determined the orientation of the films by the following method.

Strips 30.48 × 2.54 cm were carefully cut out along the machine direction (=), the cross machine direction (⊥) and the 45° angle to the machine direction (∠45°). Strips were made in duplicate from all films. A 3-g binder clip was fastened at both ends of every strip. These strips (one set) were then placed in a hanging position in an oven with good temperature control. The effective length of the hanging films was 28.3 cm. Slow heating of the circulatory oven was then started. The temperature of the oven was stabilized at 80°C. The temperature of the oven was maintained at 80°C until no visible change

TABLE 5. High Speed Tensile Data on Liner Films

Film	σ_y (psi)	σ_F (psi)	% Elongation at failure	Remarks
A	493 \pm 2	\sim 490	218-272	6 of 8 plies failed in 3 of 4 samples
B	491 \pm 3	No fail	> 600	
C	442 \pm 0	No fail	> 600	
D	469 \pm 10	\sim 470	500-> 600	1 ply in 1 of 4 samples failed
E	482 \pm 3	\sim 480	500-> 600	1 ply in 1 of 4 samples failed
F	456 \pm 28	No fail	> 600	
G	500 \pm 20	560	200-> 600	6 of 8 plies failed in one sample, 2 of 8 plies in one sample

in the lengths of the strips was noticed. The oven was then cooled to room temperature and the lengths of the strips were measured.

The other set of the strips was arranged in the oven in a similar way. They were heated to 85°C, maintained there, and then cooled for measurement. The data and its treatment are given in Table 6.

The change in length (ΔL) was obtained by subtracting the original length at RT from the final length in \parallel , $\angle 45^\circ$, and \perp at 80 and 85°C. In Fig. 1, ΔL for \parallel , $\angle 45^\circ$, and \perp directions for 85°C test are indicated on the ordinate axis. It is assumed that along the machine direction (\parallel) the strip will shrink due to recovery from the original orientation. Strength will be indicated for the \perp direction.

The ratings of orientation/strength at 80 and 85°C are indicated in Fig. 1. It will be noted from Table 6 and Fig. 1 that ΔL for $\angle 45^\circ$ direction is very low (nearly zero in all films), indicating the validity of our testing.

From these tests we conclude that Films C and B are highest and Film F lowest in orientation.

CONCLUSIONS

Our conclusions are as follows:

No impact improving additives, such as ethylene/propylene copolymers or butyl rubber, are needed to formulate baby diaper liner films.

TABLE 6. Orientation (zero tensile strength) Data

Film	Temperature (°C)	Original length (cm) at RT	Final length, L (cm), after heat treatment in		Change in length, ΔL (cm), after heat treatment in	
			=	$\angle 45$	=	$\angle 45$
A	80	28.3	27.6	27.6	-0.7	-0.7
	85	28.3	27.6	27.8	-0.7	-0.5
B	80	28.3	25.8	28.3	-2.5	0.0
	85	28.3	25.7	28.3	-2.6	0.0
C	80	28.3	23.6	28.4	-4.7	+0.1
	85	28.3	19.5	28.4	-8.8	+0.1
D	80	28.3	27.9	28.0	-0.4	+0.5
	85	28.3	27.3	28.4	-1.0	+0.7
E	80	28.3	27.7	28.3	-0.6	0.0
	85	28.3	28.2	28.3	-0.1	0.0
F	80	28.3	27.9	28.4	-0.4	+0.1
	85	28.3	27.9	28.7	-0.4	+0.4
G	80	28.3	27.5	28.0	-0.8	-0.3
	85	28.3	27.3	28.0	-1.0	-0.3

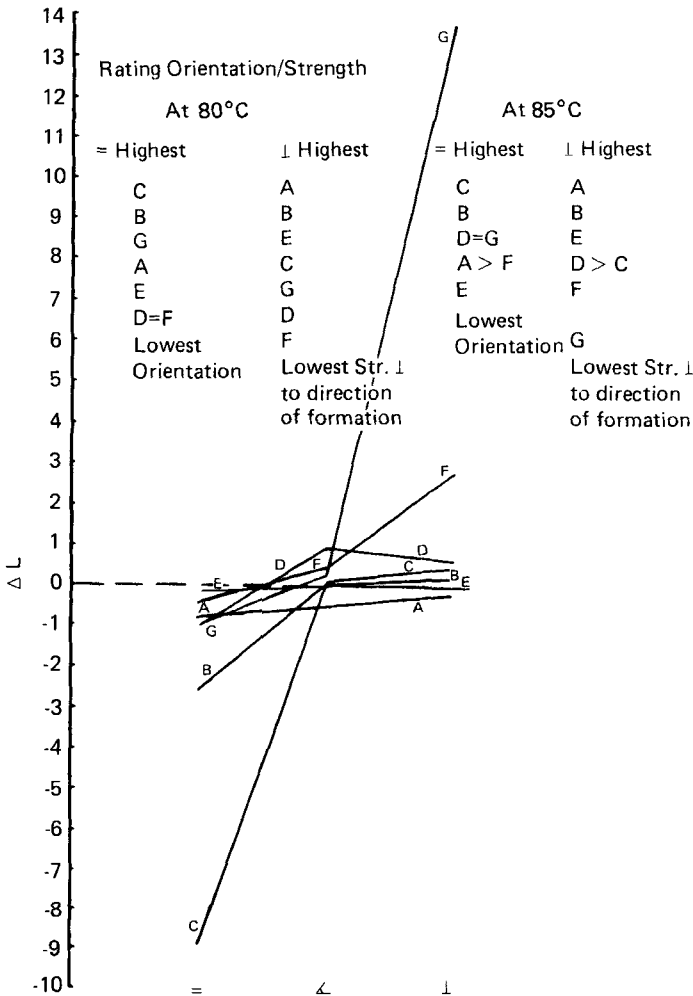


FIG. 1. Data reduction in orientation study by a graphical method.

Films containing vinyl acetate are tough with superior high speed tensile stress and % elongation. Orientation also leads to superior films (Films B and C), as would be expected.

Although vinyl acetate-containing polyethylene films are suitable as baby diaper liners, it is possible to develop polyblends as a route to films having unusual and superior mechanical properties. Opacity produced by polyblending can be also utilized to reduce or eliminate pigmentation. Two composition systems are worth investigating.

1) Propylene-ethylene/vinyl acetate polyblends: Blends of crystalline polypropylene and E/VA copolymers (15-20% VA) can be melt blended to produce compatible products. 2) Rubber modification of polyethylene: Tough films from low density polyethylene can be obtained by poly-blending polyethylene with 10-20% rubber [4].

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