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Development of Balloon Films Strong Enough to Prevent Bursting at High Altitude

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Constant volume polyethylene balloons are very useful tools for scientific observations. However, they have a tendency to burst during their ascent across the jet stream. In our experiment in 1959¹⁻³ aimed at determining the cause of this phenomena, it became clear that balloon films undergo violent tensile stresses due to impulsive high loads occurring in the jet stream. The present paper is concerned with obtaining balloon films good enough to prevent the balloon from bursting, with the relations between the mechanical properties of films and forming conditions of tubular film process studied. For controlling their mechanical orthotropies, a new idea is introduced in the low-density polyethylene tubular film process termed the "forming ratio." Also, for the crystal orientation in films possible to extend biaxially at low temperature, the b-axis law is induced from our uniaxial tensile test results of balloon films at room and low temperatures and many other references for deformations of high-density polyethylene (HDPE) plane spherulites. The balloon bursting ratio at high altitude successfully reached zero. Then a series of trial shapings to obtain larger extensible films at low temperature was made. This purpose was accomplished as expected.

I. Introduction

THE mechanical properties of light and reliable films that can prevent a balloon from bursting in high altitude at low temperature are analyzed with the following five main factors: 1) selection of polymers; 2) mechanical orthotropy of films; 3) crystal orientation in films; 4) crystal or lamella size; and 5) molecular weight and its distribution in polymers.

In the first factor, the lowest glass transition temperature of polymers is most important. Low-density polyethylene (LDPE) is one of the most suitable polymers from this point of view, and is generally used for present balloons. The second factor is regarded as the ratio of the molecular chain numbers oriented in the machine direction to the corresponding chain numbers oriented in the transverse direction. Concerning this relation, the forming ratio is introduced by the author^{4,5} in the tubular LDPE film process. The third factor is closely related to the extensibility of films at low temperature. The *b*-axis law is introduced by the author.⁶ The fourth and fifth factors are concerned with the increase in the stiffness of LDPE films at low temperature.

Using the forming ratio as a design parameter, yearly improvements in scientific balloon films have been made. These improvements have been evident in high-altitude balloon survival statistics.^{7,8}

It was anticipated that in the previous year the balloon bursting ratio at high altitude would reach zero, and that the forming ratio would still be about 1.5. Therefore, a series of trial formings by the tubular film process was attempted to shape more equibiaxially orthrotropic LDPE films with extensibility at low temperature under conditions subject to the five main factors just mentioned. As expected, the film in these had excellent properties at low temperature. Some of these properties are described.

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II. Introducing the Forming Ratio into LDPE Tubular Film Processes

Until recently, inflation or blowup ratio has been commonly used as a shaping parameter for film properties, but these are an unsatisfactory indication of the mechanical orthotropy of films. Therefore, the author has attempted to develop a more suitable and convenient shaping parameter for estimating their mechanical orthotropy.

This new parameter, β , is shown on the vertical section of the inflating bubble of this process in Fig. 1. Melted polymer is extruded from P to a cylindrical tube through the die gap e, and the melt tube is expanded by enclosed air in the bubble. Also, this bubble is cooled by blowing air from the outside, J. The melt tube changes to a solid-state film on the frost line by instantaneous crystalization that is almost amorphous at this stage. This deformation is going on at the same time as biaxial expanding continuous decreasing temperature. Hence, it may be possible to apply the homogeneous strain hypothesis at this stage and look upon it as an affine transformation. Also, there is no mass defect because the melt does not contain vaporizing components.

Therefore, an expression of constant mass can be developed in Fig. 1

$$c^2 e \rho_0 = abt \rho \tag{1}$$

where

c = square element area of melt on the die lips

e = gap of die lips

a = transverse direction (TD) length deformed to rectangular film from melt element square

 b = machine direction (MD) length deformed to rectangular film from melt element square

t = film thickness

 ρ_0 = density of melt

 ρ = density of film

As $a \neq 0$, $t \neq 0$, and $\rho \neq 0$,

$$b/a = (c/a)^2 \cdot (e/t) \cdot (\rho_0/\rho) \tag{2}$$

we put Eq. (2) in the form

 $b/a = \beta$, $a/c = \alpha$ (inflation ratio), and $e/t = \gamma$

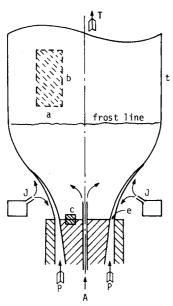


Fig. 1 Introducing the forming ratio on the inflation bubble in tubular film process.

Also, the value of ρ_0/ρ , is approximately equal to 0.8 under the actual conditions of LDPE turbular film processes.

$$\beta \approx 0.8(\gamma/\alpha^2) \tag{3}$$

This is the ratio of the MD deformation of the melt in the process to the TD one, and is nearly equal to the ratio of oriented molecular chain numbers in the MD to those in the TD. Therefore, β may be regarded as the parameter to explain the mechanical orthrotropy of LDPE films. This β is termed the forming ratio, which has not been used previously, and is one of the important points in this study.

III. Proving Significance of β by Experiments

In order to confirm the relation of β to mechanical orthotropies of films, we performed experiments on films—nine kinds of which had been shaped under various forming conditions. They were shaped in the Plastic Research Laboratory of Mitsubishi Petro-Chemical Co. Ltd., and

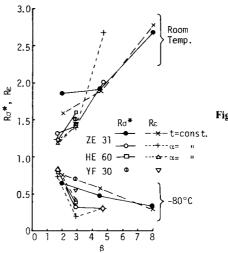


Fig. 2 β vs $R\sigma^*$ or $R\epsilon$.

tested under uniaxial tension on each TD and MD at room temperature, and at -80° C by Kawata Laboratory, Institute of Space and Aeronautical Science of the University of Tokyo. The shape of specimens were Dambel type No. 4 in Japanese Industrial Standard (JIS). These strain speeds were all 500 mm/min.

These conditions and results are shown in Table 1 where ϵ_B and σ_B are breaking strain and stress, respectively, $R\epsilon$ is the ratio of TD ϵ_B to MD ϵ_B , $R\sigma^*$ that of TD σ_B ' to MD σ_B ', and σ_B ' is true breaking stress at room temperature. However, specimens do not uniformly elongate in the case of tension at -80° C; their ϵ_B ' and $R\epsilon$ are calculated in the same way as the tension results at room temperature. Values of uniaxial tension are means for each of the five specimens.

The correlation β vs $R\epsilon$ and $R\sigma^*$ are shown in Fig. 2. In this figure, two groups of broken lines representing both room temperature and -80° C converge, respectively, at the (1,1) point. Thus, β is useful as a parameter for estimating mechanical orthotropies of LDPE films shaped in tubular film processes.

IV. Decrease in Balloon Bursting at High Altitude by Improvement in β

Since 1966, about twenty to thirty scientific balloons were launched every year by the Institute of Space and

Table 1 β of trial shaped LDPE films and their $R\epsilon$. $R\epsilon$

	β	α	tm, μ		−80°C					Room temperature				
LDPE				TD MD	€ _B ,	$R\epsilon^a$	$\frac{\sigma_B}{\text{kg/mm}^2}$	σ _B ,' kg/mm ²	Rσ*b	€ _B , 0%0	$R\epsilon$	σ_B , kg/mm ²	σ _B ,' kg/mm ²	Rσ*
ZE 31, high polymer HE 60, copolymer	8.08	1.5	26	T M	42 141	0.30	3.44 5.92	4.88 14.22	0.34	953 344	2.77	3.00 2.66	31.59 11.81	2.67
	4.55	2.0	26	T M	82 144	0.57	3.32 5.26	6.04 12.83	0.47	890 474	1.88	3.22 2.90	31.88 16.65	1.91
	2.02	3.0	26	T M	58 78	0.74	3.48 4.80	5.50 8.54	0.64	867 546	1.59	3.12 2.52	30.17 16.28	1.85
	4.73	2.5	16	T M	46 153	0.30	3.64 7.10	6.31 17.96	0.30	750 281	2.67	2.66 2.96	22.61 11.28	2.00
	2.91	2.5	26	T M	38 194	0.20	3.68 5.34	5.08 15.70	0.32	851 613	1.39	3.02 2.88	28.72 20.53	1.40
	1.73	2.5	44	T M	62 85	0.73	3.28 3.50	5.31 6.48	0.82	940 756	1.24	3.22 2.98	33.49 25.51	1.31
	2.91	2.5	26	T M	108 255	0.42	3.48 5.30	7.24 18.82	0.38	816 570	1.43	3.54 3.04	32.43 20.37	1.59
	1.73	2.5	44	T M	143 175	0.82	3.26 3.64	7.92 10.01	0.79	941 807	1.17	3.46 3.27	36.22 29.57	1.22
YF 30, low polymer	2.91	2.5	26	T M	29 54	0.54	3.22 3.92	4.15 6.04	0.69	917 643	1.43	2.78 2.53	28.29 18.80	1.50

^a $R\epsilon$ = breaking strain ratio, TD/MD. ^b $R\sigma^*$ = breaking true stress ratio, TD/MD. True stress $\sigma_B' = \sigma_B (100 + \epsilon_B) / 100$.

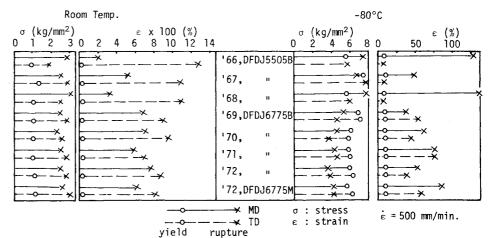
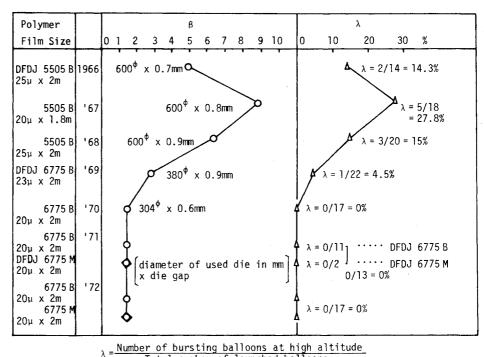


Fig. 3 Uniaxial tension test results ofballoon films used for 1966-1972 at room temperature and -80° C.



Total number of launched balloons

Fig. 4 Progress of β for used film from 1966 to 1972 and corresponding λ in every year.

Aeronautical Science of the University of Tokyo. We set about to improve β for balloon films from the preceding year, and results of uniaxial tensile test for the films which were used are shown in Fig. 3 for room temperature and -80° C. The progress of that improvement from 1966 to 1972 is shown in the left half of Fig. 4, including polymer kinds, film size, and diameter and gap of die used every year. Unfortunately in 1967 β increased beyond that of 1966 because of trouble in the extruder used previously; this value of β is identical to that of balloon films used in 1960 which was the first time the author engaged in the development of the Japanese Rockoon system. After 1968, β decreased year after year.

In the right-half of Fig. 3, the progress of λ is shown, where λ is the ratio in percentage of the number of balloons bursting at high altitude to the total number of launched balloons made by our films every year. Yearly changes in λ correspond very closely to changes in β , as seen in the figure. Although there are many factors involved in balloon bursting, namely, balloon size and structure, kinds of polymer films, launching conditions, payloads, etc., this close correspondence is spectacular. In 1970, λ finally reached zero, and β reached 1.4

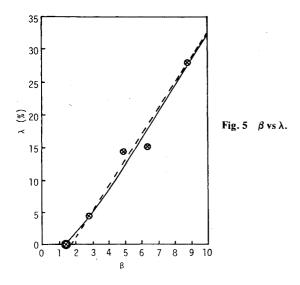
A plot of β vs λ is shown in Fig. 5. The total number of launched balloons every year is not enough from the statistical population point of view, but it is clear that the

correlation between β and λ is very close. Thus, the result of this study not only enables us to prevent bursting of balloons at high altitude, but also can lead to an increase in the performance of scientific balloons in the future.

V. Inducing the b-Axis Law at Low Temperature

As mentioned previously, the balloon bursting ratio λ successfully reached zero in 1970, but the forming ratio β was still 1.4. Figures 3 and 4 indicate this may be due to an increase in biaxial extensibility at low temperature, which has not yet been sufficiently studied.

Accordingly, the results of uniaxial tensile test at -80° C are investigated. Transverse direction speciments are able to elongate enough at room temperature, but they break at the vicinity of upper yield points at -80° C. When β decreases to 2.4 in 1969, they are able to barely elongate at -80° C. This is thought to be a problem of the crystal orientation in films. It is predicted by theoretical studies 10,11 that polyethylene single lamella is apt to tilt toward the b-axis direction, but this contradicts the above-mentioned phenomena as seen in Fig. 3, because the b-axis orientation in first-stage films is preferential to the TD.



The author has investigated many other references concerned with the deformation process of HDPE spherulites $^{12-17}$ under a tensile force, and with tensile tests of HDPE films at low temperature. As a result, it became clear that deformations of lamella are easiest by a tensile force with a 45 deg inclination angle to the *b*-axis direction; most difficult with a *b*-axis directional one, but possible with a perpendicular one. The tension tests of LDPE balloon films used in 1966 and 1968 at -80° C indicate that the relationship between HDPE spherulite deformations and tensile force angles are applicable. Therefore, it may be regarded as a law concerning deformations of LDPE films at low temperature. This induced law is termed the *b*-axis and is also one of the important points in this study.

VI. Trial Shaping of an Approximately Equibiaxial Orthotropic and Biaxial Extensible LDPE Film at Low Temperature

As described in Sec. I, the crystal orientation in films is the third main factor to be considered in obtaining desirable LDPE film properties. In order to obtain biaxially extensible LDPE films at -80° C, the *b*-axis law indicates that crystals should be preferentially oriented perpendicular with the film surface. This crystal orientation must be obtained at the blow ratio of 10.3:1 (inflation ratio 6.6) from Lindenmeyer's studies. ¹⁸ But is will be desirable for the inflation ratio to be less than 6 to maintain the stability of the inflation bubble.

The fourth main factor regarding crystal size has been studied on HDPE films by Kobayashi. 19 The fifth factor regarding molecular weight and its distribution may be parameterized almost exclusibely by the melt index (MI).

A. Trial Inflation Under Conditions Subject to the Five Main Factors as a Whole

We have made a series of trial-shaped films utilizing optimum forming conditions with a nominal film thickness of 20 μ . The LDPE material is DFDJ 6775 M (MI = 0.1₅, ρ = 0.927) of Nippon Unicar Company Limited. These five films and forming conditions are shown in Table 2. Each of the tensile test results at room temperature and -80° C are shown graphically in Fig. 6. Also, heating contraction specimens are shown in Fig. 7.

B. Some Interesting Results

1. Gradual Variation of the Upper Yield Point in Load-Elongation Diagrams at $-80^{\circ}\mathrm{C}$

At room temperature, every load-elongation diagram for the five trial shaping films is not very different, but there are very interesting changes in the vicinity of the upper yield point at -80°C.

The upper yield point peak decreases gradually from the 45-6-1 to 45-6-4 sample films as seen in Fig. 8. The 45-6-5 sample film is shaped under the usual conditions and a point of frost line is reheated by a jet of about 90°C hot air. This part of the inflation bubble suddenly is extended in the TD. Transverse direction tensile specimens of this part are 16 μ thickness different from 22 μ thickness of the other nonreheated part film. The TD load-elongation diagram of this part is shown by the dot-dash line, 45-6-5', in Fig. 8. It is similar to TD or MD diagrams of the 45-6-3 sample film. The most optimum

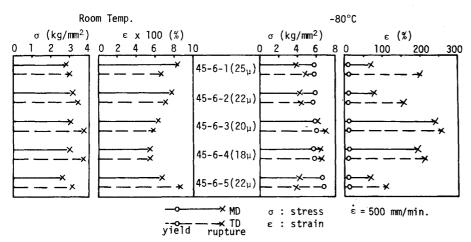


Fig. 6 Uniaxial tension test results for trial shaped film in 1970.

Table 2 Trial shaping conditions and β' (heating contraction ratio)

	Sample film no.	Film size, $\mu \times \text{cm}$	Diameter of die, mm	e, mm	α	β	β΄	Die temp., °C	Take-up speed, m/min
	45-6-1	25 × 42	75	0.5	3.57	1.26	1.06	200~230	12.5
Polymer	-2	22×49	75	0.5	4.16	1.0_{5}°	1.05	$200 \sim 230$	12.5
DFDJ 6775 B	-3	20×59	75	0.5	5.0°_{1}	0.8_{0}^{5}	1.0°_{9}	$200 \sim 230$	18.0
$MI = 0.1_5$	-4	18×62	75	0.5	5.26	0.80	0.9°_{7}	$200 \sim 230$	16.0
$\rho = 0.917$	-5	22 × 52	75	0.5	4.4	0.9_{3}°	1.33	$200 \sim 230$	8.0

a)

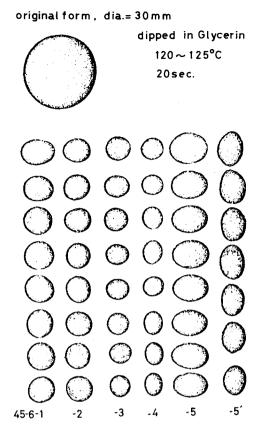


Fig. 7 Heating contraction specimens of trial shaped films.

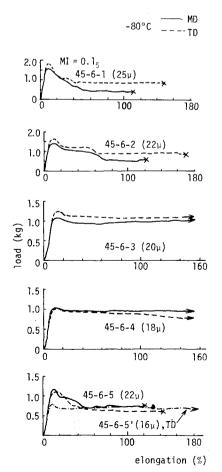
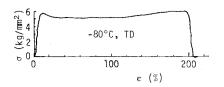
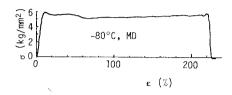
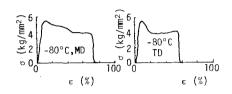


Fig. 8 Gradual variation of the upper yield point in load-elongation diagrams at -80° C for each trial shaped film (shaped in 1970).







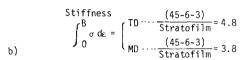


Fig. 9 Typical stress-strain diagrams²⁰: a) Trial shaped film (45-6-3); b) Stratofilm (19 μ thickness, imported in 1970).

sample film is 45-6-3 and does not have a large peak at the yield point. Then, it may be concluded that the peak is caused by crystal size and orientation.

The stiffness of this 45-6-3 trial-shaped film is largest, and is 4.8 times as large in the TD as that of the famous balloon Stratofilm, and 3.8 times in the MD, as seen in Fig. 9. The circular diaphram test by 100 mm diam specimens at -80° C compare 45-6-3 film with Stratofilm in Fig. 11. These quantitative comparisons are already reported in Ref. 6 and 21, together with true breaking stress and strain. Values of this 45-6-3 trial-shaped film are larger than any other tested films.

2. Crystallic Oreintation

Pole figures for two crystallographic directions, (200) *a*-axis, and (020) *b*-axis for two trial-shaped films 45-6-3 and 45-6-5 and one early stage balloon films are measured. These are shown in Fig. 10.

The b-axis for the early stage balloon film DFDJ 5505 B (used in 1968) preferentially orient to the TD. This film broke on the yield point at -80° C, as seen in Fig. 3. This phenomena supports the b-axis law. Pole figure (200) of 45-63 trial-shaped best film preferentially orients to the perpendicular with the film surface, as expected. Biaxial extensibility of this film at -80° C is largest as mentioned previously, which also supports the b-axis law.

Pole figure for crystallographic directions (110), (200), and (020) of a nondeformed sheet as extruded from the die after the 45-6 series trial shaping showed that no particular orientation for a nondeformed sheet is noted in any direction. However, the orientation of high-polymer molecule chains in the flowing melt that has just been extruded from the die lips is not yet sufficiently known; the author assumed that there are no orientations in this melt at the time inducing the forming ratio. Pole figures of this nondeformed sheet confirm this assumption.

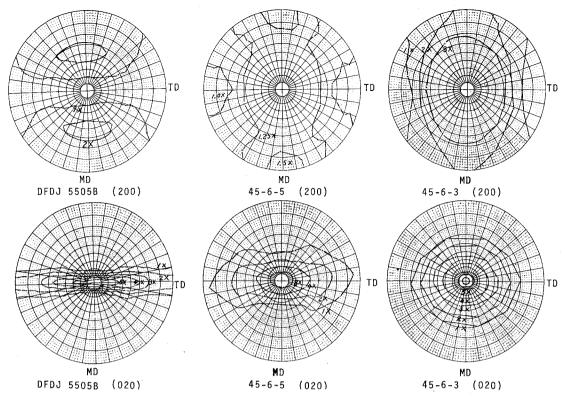
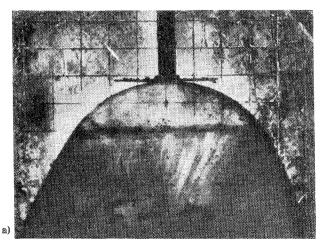


Fig. 10 Pole figures of early stage balloon film and two kinds of trial shaped films as 45-6 series.



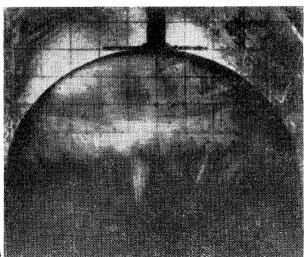


Fig. 11 Shapes just before bursting of inflated diaphragm specimens at -80° C: a) Stratofilm (19 μ thickness, imported in 1970); b) Trial shaped film 45-6-3 (20 μ thickness).

VII. Conclusions

Several conjectures have been proposed for the balloon bursting at high altitude. The true cause of this phenomena has been made clear by our test flight of the special prepared balloon for that purpose in 1959 and uniaxial tensile test results of LDPE balloon films at the ground.

Therefore, the author was interested in making large, biaxially extensible plastic films with large stiffness at low temperature as light and reliable reinforcement method for balloons, and analyzed the properties of those films into five main factors. The most serious problem in the then used films was the intense mechanical orthotropy. In order to correct this problem, the author introduced the forming ratio into the tubular films process as a convenient parameter for the mechanical orthotropy of shaped films.

The progress of this forming ratio for balloon film and corresponding balloon bursting ratio at high altitude was exactly as shown in Figs. 5 and 6; however, there were many factors involved in balloon bursting. This extreme correspondence was very spectacular. Thus, the goal of preventing scientific balloons from bursting at high altitude had been met, but the forming ratio was still 1.4 and not yet equibiaxial. This anticipation was made in the previous year, and it was considered that the approximate equibiaxial films large extensible at low temperature would be more effective to improve the performance of balloons.

Therefore, the author again introduced the b-axis law, as previously mentioned. If this law is correct, the b-axis orientation of lamellae in biaxially extensible LDPE films at low temperature must be preferentially perpendicular with film surfaces. This orientation had been made by Lindenmeyer. His forming ratio was not clear, of course, but the blow ratio was described for 10.3. When it was attempted that more advisable films at low temperature with LDPE of low MI were experimentally shaped under conditions subject to five main factors as a whole, his blow ratio was taken into consideration. This experimental shaping was made symmetrically centering nominal film thickness of $20~\mu$, namely 45-6-3, which was shaped at the inflation ratio about 5 and the forming ratio about 0.8. The stiffness of this film shaped

under most optimum conditions exceeds four times as large as that of Stratofilm (19 μ thickness, imported in 1970 from Winzen Inc.), which had been famous as a film to prevent balloon bursting at high altitude in that time. And circular diaphragm test specimens of 100 mm in diameter of the former extend far smoother than that of the latter at -80° C, as shown in Fig. 11. Also, it is considered that gradual variations of the upper-yield points as shown in Figs. 8 and 9 suggest the effect of crystal size and orientation to the extensibility at -80° C. From these results, we may conclude that the effect of not only preventing balloon bursting but also improving balloon performances will become even stronger.

Thus, expected results have been successfully accomplished. Therefore, that idea may be quite all right that the properties of advisable films to prevent balloon bursting at high altitude consist of five main factors. Also, these experimental results support the validity of the present study concerning the relations between the mechanical properties and conditions of the tubular film process based on the forming ratio and b-axis law.

In the future, it will be looked for that the results of this paper will contribute to reform not only balloon films but generally used films.

Acknowledgment

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