The Forming Properties of High Molecular Weight Polyethylene

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SYNOPSIS

The formabilities of cold-rolled high molecular weight polyethylene (HMWPE) sheets have been studied by measuring their plastic anisotropy ratio (R value), strain-hardening exponent (n value), strain distribution, and the forming limit diagram (FLD). The deep drawability of the polymer is improved by rolling. After 40% or more reduction in thickness by cold rolling, the HMWPE sheet could be deep-drawn into a cylindrical cup. The results of R value measurement indicate that the R value is responsible for improved drawability. Cold rolling also increases the n value but decreases the strain gradient. Stretch forming tests have also been carried out, and the results show that cold working could also improve the stretchability of this polymer. The results of the FLD are in agreement with the other properties studied. The mechanical properties, environmental stress cracking resistance, and shape, size, and property stability of the deep-drawn HMWPE cups have also been investigated.

INTRODUCTION

The cold rolling of polymeric materials is a useful method to modify the mechanical and other properties of polymers. The effect of rolling on the mechanical properties of polyethylene was studied by Rothschild and Maxwell.¹ The results showed that cold rolling increased the tensile strength and slightly decreased the ultimate elongation in the rolling direction. The effects of cold rolling on the mechanical properties of some other polymers have also been reported.²⁻⁵

Deep drawing and stretch forming are two important forming processes that are widely used in the metal industry. It has been reported that for metals, the plastic anisotropy ratio (so-called R value) controls the ability of a metal to be deep-drawn, and the strain-hardening exponent (n value) determines the ability of a metal to be stretched.⁶⁻⁸ Besides, strain distribution measurement and the forming limit diagram (FLD) are also widely used in the investigation of metal formability.^{9,10} Rela-

tively limited published data, however, are available concerning the deep drawing and stretch forming of cold-rolled polymeric materials. In addition, it is known that different modes of stress can be produced in the blank during the two forming processes, in particular, the deep drawing process.

A typical deep drawing equipment which consists of a punch, a die, and a blank holder is shown in Figure 1. During the deep drawing process, the material at the center of the blank under the nose of the punch is wrapped around the profile of the punch and is subjected to biaxial tensile stress due to the action of the punch. The material in the outer portion (i.e., the flange) is subjected to a compressive stress in the circumferential direction and a tensile stress in the radial direction. As the blank passes over the die radius, it is subjected to bending stress and unbending when it becomes part of the cup wall, and biaxial tension stresses are believed to exist in the cup wall between the punch nose and the die.

In the present work the formability of cold-rolled high molecular weight polyethylene (HMWPE) sheets was studied by measuring the R and n values as well as strain distribution and the forming limit diagram. The effect of different modes of stress dur-

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Figure 1 The general set up of the deep drawing process.

ing deep drawing on the deep-drawn HMWPE cups was also investigated. Mechanical properties such as dart falling impact strength and hardness were measured. Environmental stress cracking and shape, size, and property stability of the cups were also studied.

EXPERIMENTAL

Materials

Commercially available 2.0-mm-thick extruded HMWPE sheets were used in this study.

Rolling

Initial blanks 130 mm square were biaxially rolled at room temperature, i.e., they were first rolled at 0° to one edge of the blank and followed by rolling in the mutually perpendicular direction (90° direction) to the same edge of the blank. This process was repeated until the required thickness was obtained. The machine used was a Hille laboratory two-roll mill with 114-mm-diameter and 147-mmlong rolls revolving at 30 rev/min. The reduction in thickness per pass was 0.05 mm.

Deep Drawing and Stretch Forming

Deep-draw cupping tests and biaxial stretch forming tests of unrolled and rolled circular specimens were performed on a hydraulically operated Hille universal sheet metal testing machine. For deep drawing, a flat-nose punch was used whereas, for stretch forming, a hemispherical nose punch was employed. The diameter of both punches was 50 mm. The punch speed employed was 10 mm/s.

Tensile Test and Determination of R and n Values

Tensile tests and the determination of R and n values were performed on an Instron tensile testing machine. Rectangular specimens with dimensions of 130 mm length and 20 mm width were prepared at 0°, 45°, and 90° to one of the rolling directions. The straining speed was 20 mm/min.

Plastic Anisotropy Ratio

The plastic anisotropy ratio (R) is defined as

$$R = \frac{\ln(w_f/w_0)}{\ln(t_f/t_0)}$$
(1)

where w_0 and t_0 are the initial width and thickness of the specimen and w_f and t_f are the final width and thickness, respectively. Since the volume of the material remains constant during plastic deformation, the formula can be written as

$$R = \frac{\ln(w_f/w_0)}{\ln(l_0 w_0/l_f w_f)}$$
(2)

where l_0 and l_f are initial and final lengths of the specimen, respectively.^{8,11}

The component of normal anisotropy is defined as

$$\bar{R} = \frac{1}{4} \left(R_0 + 2R_{45} + R_{90} \right) \tag{3}$$

If R is greater than unity, the material could resist thinning; this would lead to better drawability. For many materials, there is a variation of the R value for different directions in the sheet. This variation of R value within the plane of the sheet is called planar anisotropy (ΔR) and is responsible for earing in deep-drawn cups:

$$\Delta R = \frac{1}{2} \left(R_0 + R_{90} - 2R_{45} \right) \tag{4}$$

Strain-Hardening Exponent (n Value)

The true stress-strain curves for many materials can be approximated by the Ludwick-Holloman equation:

$$\sigma = K\epsilon^n \tag{5}$$

where K is a constant and n is termed the strainhardening exponent.

The n value is a measure of the strain-hardening capacity of the material and indicates the ability of

the material to spread the strain away from the most heavily strained zones to regions of lower strain, thus avoiding the formation of local regions of high strain which lead to necking and ultimate fracture. Obviously, a high n value is desirable in stretch forming.

In the measurement of R and n values, 50 mm gauge length was marked on the specimens. The width and thickness of the specimens were measured to within ± 0.01 mm as a function of elongation, at four points within the gauge length, and average values were obtained.

From the measured values of width and thickness, the cross-sectional area of the specimen can be calculated at any elongation and thus true stress can be determined. The true strain is defined as $\epsilon = \ln (l_f/l_0)$, where l_0 and l_f are the initial and final lengths, respectively.

By changing eq. (5) into a log-log plot, a straight line is obtained, from the slope of which n can be determined. A mean value of n was obtained from the equation

$$\bar{n} = \frac{1}{4}(n_0 + 2n_{45} + n_{90}) \tag{6}$$

Strain distribution can be used to assess the formability of metals⁹ and polymers.^{12,13} Silk screen printing was used to imprint a series of concentric circles with 3 mm increment in radius successively on the HMWPE blanks. After forming, the deformation of the concentric circles differs with different portions of the blank. The dimensions between adjacent concentric circles before and after forming were measured. A Mitutoyo traveling microscope was used to measure the distance to within ± 0.01 mm. The percentage engineering strain can be calculated by the following equation:

$$d_f - d_0$$

where d_f = the final distance between adjacent concentric circles and d_0 = the initial distance between adjacent concentric circles.

The strain distributions of the specimens are shown by the relationship between the measured percentage strains and the radial locations of the specimens.

Forming Limit Diagram (FLD)

It is known that the limit or failure strains in material forming can be represented by an FLD which shows the onset of localized necking over all possible combinations of strains in the plane of the sheet. The magnitude and shape of the forming limit curve therefore provide some guide to formability, and so are commonly used as a diagnostic aid in failure analysis in metal forming,¹⁰ and recently in polymer forming.¹² Circular gridded HMWPE sheets of different sizes were stretch formed to the extent that stress whitening on the specimens was observed with a 50 mm diameter hemispherical punch. An unrolled sample with 1.10 mm thickness and a 50% rolling reduction sample with approximately the same thickness (1.15 mm) was used to study the FLD. Ten specimens with 120 mm length and different widths starting from 30 to 120 mm with increments of 10 mm were used for each sample as illustrated in Figure 2. Deformed circles lying both in the stress whitened region and just outside of it were measured to obtain the major strain and the minor strain. The FLD was then drawn to fall below the strains in the stress whitened zones, and above the strains found just outside these zones.

The falling dart impact test method was used to investigate the impact strength of the drawn cups. The test machine consisted of a support stand and electromagnetic chuck to support the dart above a table at a known height. The value of impact energy was determined by the weight and height that can break, crack, or split the plastic sheet under test. The normalized mean failure energy (NMFE) was calculated in accordance with BS 2782 method 352 D. The falling dart impact strength of the bottom of the drawn HMWPE cups was measured. For comparison, the impact strength of the unrolled and rolled specimens were also determined.

As the HMWPE specimens neither broke nor cracked during testing, the appearance of a stress



whitening spot on the surface of the specimen was used as a criterion of failure.

Hardness Test

A Durometer (A scale) was used to determine the Shore hardness of the rolled and unrolled HMWPE sheet samples as well as the bottom and wall of the deep-drawn cups. A specially machined flat-nose cylindrical fixture matching with the internal profile of the deep-drawn cup was used to support the bottom and the wall of the deep-drawn cups during hardness measurement.

Environmental Stress Cracking (ESC)

The rolled and unrolled HMWPE sheets were cut to strips of 20×40 mm. A special fixture was used to apply external tensile stresses to the strips. The rolled and unrolled strips with and without external stresses as well as the drawn cups were immersed in several solvents such as acetone, trichloroethylene, carbon tetrachloride, alcohol, and xylene. The specimens were observed closely for cracking and the time required for cracking was recorded.

Shape, Size, and Property Stability Study

High molecular weight polyethylene cups were heated in an oven at 40 and 50° C, respectively. The dimensions of the cups were checked hourly outside the oven. After heating for 12 h the cups were withdrawn from the oven and conditioned for 24 h at room temperature before testing the hardness as well as the dart falling impact strength.

RESULTS AND DISCUSSION

Tensile Properties

The tensile stress-elongation curves for specimens prepared at 0° , 45° , and 90° to one of the rolling directions for the HMWPE sheets which had been cold-rolled biaxially to different reductions in thickness were recorded. No markedly difference in orientations for different directions (i.e., at 0° , 45° , and 90° to the chosen rolling direction) was observed, and the stress-elongation curves for the unrolled specimens at different directions were almost identical. This implies that the directionality of the unrolled specimens due to extrusion was not significant. Hence only the tensile stress-elongation curves for specimens at the 0° direction are shown (Fig. 3). It can be seen that the curve for the unrolled specimen shows a conspicuous yield point. At 40% on higher rolling reductions, the yield point disappears. In addition, the tensile strength of the rolled specimens also increases.

Another effect of rolling on HMWPE sheet was that stress whitening was eliminated when the material was reduced in thickness by 40% or more. This implies that, after rolling, HMWPE sheets could resist higher stress. The elimination of stress whitening by cold rolling will be discussed in another paper.¹⁴

Deep Drawability

It is known that deep drawing and stretch forming are two important forming processes. In this section the deep drawability of the HMWPE sheets after rolling will be discussed using the results of deepdraw cupping tests and the determination of plastic anisotropy ratio (R value). The stretch formability will be discussed later.

The Effect of Rolling on Deep Drawability

Circular specimens of different blank diameters were deep-drawn using a constant-diameter punch. Rolled specimens of different percentage reductions (from 0 to 60% with increments of 10%) were deep-drawn under constant test conditions of blank clamping pressure and punch travel speed, with tool clearance varying between 1.05 and 1.25 t_0 depending on the blank thickness t_0 .

The ratio D_B/d_p was used as a measure of the deep drawability of the material, where D_B is the maximum blank diameter which can be drawn into a cup and d_p is the punch diameter. The relationship between the deep drawability and percentage rolling reduction of HMWPE is shown in Figure 4. It is



Figure 3 Nominal stress versus percentage elongation.



Figure 4 Deep drawability versus percentage rolling reduction.

evident that there is an increase in deep drawability with rolling reduction. The use of lubricant improves significantly the deep drawability of the material. The blank holding pressure also affects the deep drawability. It can be observed from Figure 4 that, for the lubricated specimens with 60% rolling reduction and different holding pressures, the D_B/d_p values concentrate in one point. This was due to the largest blank diameter which could be cut from the 60% rolled specimens was limited to 127 mm. In fact, according to our observation that the D_B/d_p ratio could be greater than this value and should be different for different holding pressures.

The Effect of Rolling on the Plastic Anisotropy Ratio

As shown in Figure 3, the stress-elongation curves of the unrolled samples and the samples with lower rolling reductions for HMWPE exhibit yield point coupled with necking. Thus the change in width and thickness of the specimen is not uniform. In view of this, the R values of only samples with 40, 50, and 60% rolling reductions which showed uniform changes in width and thickness were measured. The results are shown in Figure 5. Rolling increases significantly the \overline{R} value, which changes with elonga-



Figure 5 Normal anistropy versus percentage elongation.

tion. It is observed that the \overline{R} values of HMWPE with the indicated rolling reductions are all greater than unity. This is in agreement with the results of the deep-drawing tests, i.e., HMWPE sheets with 40% or higher rolling reductions could be deepdrawn into a cup successfully.

It is interesting to find that the normal anisotropy of HMWPE is fairly high. It has been reported¹⁵ that zinc and lead have an \overline{R} value of 0.2, steels range from 1 to 2, copper and brass range from 0.8 to 1, and titanium has a value of 6. Thus, the drawability of the rolled HMWPE sheets can compete with most metals. The planar anisotropy (ΔR) values determined for the rolled specimens are shown in Table I. The planar anisotropy property affects earing in deep-drawn cups. An ideal material for deep drawing is that which possesses high \overline{R} and low ΔR values, preferably with ΔR equal to zero. According to those criteria, HMWPE sheets with 40% or higher rolling reductions could be considered as superior materials for the deep-drawing process.

Stretch Formability

The stretch formability of the HMWPE sheets will be discussed in this section using the measured nvalues, the changes of strain distribution, and the results of the stretch forming tests.

The Strain Hardening Exponent **n** and Cold Rolling

For the same reason mentioned in the previous section, only the n values of the specimens with higher rolling reductions were measured. The results are listed in Table II. It can be seen that the n value of HMWPE increased when the rolling reduction changed from 40 to 50%; a further 10% increment in rolling reduction showed no effect on the n value. This indicates that rolling can increase the n value, but it seems that the increment is not in direct proportion to rolling reduction. In general, an n value

Table I Values of Planar Anisotropy (ΔR) for Biaxially Rolled HMWPE Sheets

Elongation (%)	ΔR , Rolling Reduction (%)				
	40	50	60		
10	-0.30	0.24	0.06		
20	-0.29	0.28	0.15		
30	-0.29	0.29	0.17		
40	-0.31	0.37	0.17		

Table IIThe n Values of HMWPE

Rolling Reduction (%)	n
40	0.25
50	0.28
60	0.28

of more than 0.20 is considered to be desirable for stretch forming. Thus, the HMWPE sheets could be regarded as suitable for stretch forming, provided that they were first subjected to rolling reductions of 40, 50, or 60%.

Strain Distribution and Cold Rolling

Strain distribution investigations were performed on the rolled and unrolled HMWPE blanks. Specimens with concentric circle marking were stretchformed under dry condition to approximately the same depth for comparison. It is realized that the depth of forming affects the maximum strain value and strain distribution of a specimen. Due to the limitation of the sheet metal testing machine, it was impossible to obtain accurately the same forming depth for every test.

A previous study¹³ indicates that when the strain is adjusted by dividing by the depth of forming, the depth effect was found to be eliminated. The results of adjusted percentage strain versus radial location are shown in Figure 6. It can be seen that the maximum strain value is not at the center of the blanks. On increasing the rolling reduction, the maximum strain value decreases, and the uniformity of stain distribution increases. These results imply that after rolling the HMWPE sheet has the ability to spread the strain away from the most heavily strained zones to regions of lower strain; thus the strain distribution is more uniform. It thus avoids, as far as possible, the formation of local regions of high strain, which leads to necking and ultimate fracture. In short, the stretch formability of the material is improved by rolling.

Stretch Forming Tests

In the stretch forming test, the depth of penetration when fracture occurs is generally taken as a measurement of the formability of the material. It may be regarded as the maximum depth of the material that can undergo plastic deformation by stretching.

All the unrolled and rolled HMWPE specimens of different thickness reductions were stretched formed on the Hille machine under dry conditions



Figure 6 Adjusted strain distribution of HMWPE sheets with different rolling reductions.

and with Shell Tellus oil as lubricant. It is known that in stretch forming the edges of the blanks are clamped tightly and the part is formed entirely by multidirectional stretching over the contour of the hemispherical punch. Hence the thickness of the blank will affect the extent of stretch forming. Considering the effect of thickness, the depth of penetration (DP) is adjusted by dividing by the blank thickness t. The DP/t value increases with percentage rolling reduction as shown in Figure 7. However, the use of lubricant in the stretch forming of HMWPE did not produce a significant effect.

From the results of the measured n values and strain distribution, it can be seen that rolling increases the n value and the uniformity of strain distribution, whereas, in the stretch forming tests, cold rolling increases the Dp/t value. All these results indicate that cold rolling improved the stretch formability of the HMWPE sheets.

Forming Limit Diagram (FLD)

Studies on the FLDs of the HMWPE sheets have also been carried out. The FLDs of two HMWPE



Figure 7 Depth of penetration over blank thickness as a function of percentage rolling reduction.



Figure 8 Forming-limit curve for two HMWPE samples

samples, an unrolled sample and a sample with 50% reduction, with approximately the same thickness are shown in Figure 8. It can be seen that the major strain values of the 50% rolling reduction sample are larger than those of the unrolled sample; hence the 50% rolling reduction sample has a larger safety forming zone. This indicates that the formability (i.e., including deep drawability and stretch formability) of the HMWPE sheets is greatly improved after being rolled to 50% thickness reduction. These results are in agreement with the deep-drawing cupping tests and stretch forming tests as well as the various properties such as the R and n values and strain distribution measurements mentioned in the previous sections.

Properties of the Deep-Drawn Cups

As mentioned in the Introduction, it is known that different modes of stress can be produced in the blank during the deep-drawing process. The effect of these different modes of stress was studied by measuring the mechanical properties such as dart falling impact strength and hardness of the deepdrawn cups. In addition, environmental stress cracking and shape, size, and property stability of the cups were also investigated.

The Effect of Cold Work on Falling Dart Impact Strength

The results of falling dart impact strength tests for HMWPE specimens with different rolling reductions and the drawn cups are shown in Table III. It can be seen that the dart falling impact strength of the HMWPE sheets increases with rolling reduction. For the sheets with 50% rolling reduction and the cups drawn from these sheets, no definite data could be obtained. The weight used for testing the 40%rolling reduction sheets was 0.64 kg where a stress whitening spot was observed. However, for the 50%rolling reduction sheets and the cups drawn from these sheets, even a 1 kg weight (the highest weight available) was used but no whitening could be seen.

The above results imply that the dart falling impact strength of HMWPE was improved by cold rolling and also that the drawn cups still retained high strength.

Results of Hardness Measurement

The Shore hardness of HMWPE is shown in Table IV. From the results obtained, it can be seen there is not much change in hardness for HMWPE samples.

The Effect of Cold Working on Environmental Stress Cracking

As mentioned in the subsection Hardness Test, the rolled and unrolled HMWPE strips with and without external stresses as well as the drawn cups were immersed in several solvents such as acetone, trichloroethylene, carbon tetrachloride, alcohol, and xylene to study the stress cracking. The specimens had been immersed in these solvents for 1 month and no stress cracking could be observed. It is known that high density polyethylene has good resistance to acetone, alcohol, medium resistance to carbon tetrachloride and xylene, and relatively poor resistance to trichloroethylene.¹⁶ Hence, from the results obtained it seems that the ESC of HMWPE is somewhat different from high density polyethylene. and that cold working has neither positive nor negative effect on the worked HMWPE products with respect to environmental stress cracking.

Environmental stress cracking was considered to be one of the pressing problems facing the plastics industry.³ Williams and Ford³ reported that when cross-rolled linear low density polyethylene was immersed in a bath of ethyl alcohol, a marked im-

1 able III Dart Failing Impact Strength of nim wPE Sam	Table III	Dart Falling	Impact Strength	of HMWPE	Samples
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		Sheets			Cups	
Rolling reduction (%) NMFE (J/mm)	0 0.81	30 1.08	40 1.57	50 > 1.57	(Drawn from 50% rolling reduction sheets) > 1.57	

	Shore Hardness, Rolling Reduction (%)			
	0	30	40	50
Sheet	67	66	64	64
Cup bottom			66	64
Cup wall			65	64

Table IVThe Shore Hardness ofHMWPE Specimens

provement on ESC resistance could be seen when compared with the unrolled specimens. They concluded that large cold reductions nullified the effect of the environment. Bigg et al.¹⁷ reported that the dimensional and weight stability of high density polyethylene in 100°C polybutane oil improve dramatically when the polymer is uniaxially rolled.

However, the present study does not show positive or negative effect on ESC from cold working. This indicates that ESC is a complex phenomenon and needs further investigation.

Results of Shape, Size, and Property Stability Studies

High molecular weight polyethylene cups were first heated in an oven at 50° C for 12 h, significant changes in shape were observed. A lower temperature (i.e., 40° C) was then used to heat the cups and no distortion of the cups could be seen when heated at this temperature for 12 h.

The falling dart impact strength and hardness of the HMWPE cups after heat treatment were also measured. The results indicated that there were no changes in these properties for HMWPE cups. These studies suggest that deep drawn HMWPE cups could be used at ambient temperature without losing shape and size, as well as mechanical properties.

CONCLUSIONS

- 1. From the results obtained, it can be concluded that the 40% or higher thickness reduction HMWPE sheets have higher tensile strength. The deep drawability of these sheets were greatly improved as indicated by the higher Rvalue and D_B/d_p value as well as the successfully drawn cups. The application of lubricant during deep drawing improved the drawability of cold rolled HMWPE sheets.
- 2. Cold rolling also improved the stretch formability of the HMWPE material as shown by

the increase in n value, the uniformity of strain distribution, and also the results in the stretch forming tests.

- 3. The results in 1 and 2 are in agreement with the FLD which shows that the formability (including deep drawing and stretch forming) of the HMWPE sheets is greatly improved after being rolled to 50% thickness reduction.
- 4. Although different types of stress were produced in the blank during the deep drawing process, no harmful effect on the properties of the drawn cups could be observed. The advantages resulting from cold rolling were retained, and the HMWPE deep-drawn cups possessed high dart falling impact strength and sufficient hardness.
- 5. Cold working has no significant effect on the environmental stress cracking for HMWPE material.
- 6. From the results of shape, size, and properties study, it was evident that HMWPE drawn cups could not change in shape or size at ambient temperature.

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