Ultradrawing Properties of Gel Films of Ultrahigh-Molecular-Weight Polyethylene and Low-Molecular-Weight Polyethylene Blends Prepared at Various Formation Temperatures

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ABSTRACT: The ultradrawing behavior of ultrahigh-molecular-weight polyethylene/low-molecular-weight polyethylene film specimens prepared at various concentrations and formation temperatures was studied. The critical draw ratio (D_{rc}) of $UL_{-0.7}$ film specimens was found to depend significantly on the formation temperature used to prepare the film specimens. At any fixed drawing temperature, the D_{rc} values of $UL_{-0.7}$ specimens prepared at various formation temperatures increased significantly as the formation temperatures were reduced. In fact, with an optimum drawing temperature of 95°C, the D_{rc} values of $UL_{-0.7}$ specimens prepared at a formation temperature of 0°C reached 488, about 50% higher than that of $UL_{-0.7}$ specimens prepared at a formation temperature of 95°C. These interesting phenomena were investigated in terms of the thermal, birefringence, and tensile properties of these undrawn and drawn $UL_{-0.7}$ specimens. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 89: 3728–3738, 2003

Key words: drawing; gels

INTRODUCTION

High-modulus and high-tenacity fibers have recently been obtained from flexible polymers by various methods such as solid-state extrusion,[1,2] ultradrawing,[3] surface growth,[4,5] and gel spinning.[6–8] Among these processing methods, gel spinning[6,9] has attracted much attention since its invention in the 1970s because of its availability in the production of commercial high-performance fibers of ultrahigh-molecular-weight polyethylene (UHMWPE). The key element in obtaining high-strength UHMWPE fibers is to find a way to ultradraw the gel fibers to an ultrahigh draw ratio after the gel spinning process. In fact, it has been found that tensile strengths and moduli of ultradrawn UHMWPE gel specimens improve consistently with increasing achievable draw ratios.[4,6,9–24] Many other ultradrawing methods have also been developed since then, and a surprising draw ratio of 350, a tensile strength of 6.5 GPa, and Young's modulus of 220 GPa have been achieved recently.[24] However, these tensile properties are still well below the theoretical tensile strength and modulus reported for a perfect polyethylene crystal, 32 GPa[11] and 324 GPa,[25–27] respectively. Investigations[8,10,18–22,27–29] have been performed to improve the achievable draw ratios and the corresponding tensile properties of UHMWPE gel specimens. The drawability of gel specimens depends significantly on the compositions of the solutions from which the gels are made.[11–14,28] The achievable drawability is reduced significantly when gel films are prepared from solutions with concentrations deviating from their critical values. In these, the numbers of entanglements in the coherent network structure of the gel films are too many or too few to yield the maximum extension of UHMWPE during the gel-deformation processes. However, several authors[22,30–32] have reported that the drawing temperature and rate can markedly affect the maximum achievable draw ratio of solution-grown UHMWPE crystal mats. At a fixed drawing rate, the achievable draw ratios reach a maximum value when each film specimen is drawn at a temperature near its optimum temperature (T_{op}).[32] In fact, the T_{op} values of each film sample increase consistently with the drawing rate. The achievable draw ratio of each film sample drawn at a constant rate and a temperature near T_{op} is called the optimum achievable draw ratio (D_{raop}), which reaches another maxi-

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TABLE I
Compositions of the Gel Solutions and Gel-Formation Temperatures (T_f's) Used to Prepare the Film Samples

Film sample	Weight ratio of UHMWPE/LMWPE	Concentration (g/dL)	T_f (°C)
$UL_{-0.6-0°C}$	98/2	0.6	0
$UL_{-0.7-0°C}$		0.7	
$UL_{-0.8-0°C}$		0.8	
$UL_{-0.9-0°C}$		0.9	
$UL_{-1.0-0°C}$		1.0	
$UL_{-0.6-35°C}$	98/2	0.6	35
$UL_{-0.7-35°C}$		0.7	
$UL_{-0.8-35°C}$		0.8	
$UL_{-0.9-35°C}$		0.9	
$UL_{-1.0-35°C}$		1.0	
$UL_{-0.6-65°C}$	98/2	0.6	65
$UL_{-0.7-65°C}$		0.7	
$UL_{-0.8-65°C}$		0.8	
$UL_{-0.9-65°C}$		0.9	
$UL_{-1.0-65°C}$		1.0	
$UL_{-0.6-95°C}$	98/2	0.6	95
$UL_{-0.7-95°C}$		0.7	
$UL_{-0.8-95°C}$		0.8	
$UL_{-0.9-95°C}$		0.9	
$UL_{-1.0-95°C}$		1.0	

TABLE II
C_c and D_{ra} of U/L Gel Films Drawn at Various Temperatures

Sample	C_c	D_{ra} of Gel Films Drawn at 85°C	95°C	105°C	115°C
$UL_{-0.6-0°C}$	0.74	385	421	398	332
$UL_{-0.7-0°C}$		400	488	419	361
$UL_{-0.8-0°C}$		389	463	404	348
$UL_{-0.9-0°C}$		334	393	361	305
$UL_{-1.0-0°C}$		310	377	324	289
$UL_{-0.6-35°C}$	0.74	372	403	380	311
$UL_{-0.7-35°C}$		385	429	393	334
$UL_{-0.8-35°C}$		379	410	382	322
$UL_{-0.9-35°C}$		321	365	325	301
$UL_{-1.0-35°C}$		300	354	305	267
$UL_{-0.6-65°C}$	0.74	323	350	331	289
$UL_{-0.7-65°C}$		352	376	364	317
$UL_{-0.8-65°C}$		339	359	347	300
$UL_{-0.9-65°C}$		301	324	318	277
$UL_{-1.0-65°C}$		273	312	286	254
$UL_{-0.6-95°C}$	0.74	306	316	302	272
$UL_{-0.7-95°C}$		313	326	312	301
$UL_{-0.8-95°C}$		308	321	309	284
$UL_{-0.9-95°C}$		283	308	277	254
$UL_{-1.0-95°C}$		272	291	263	237

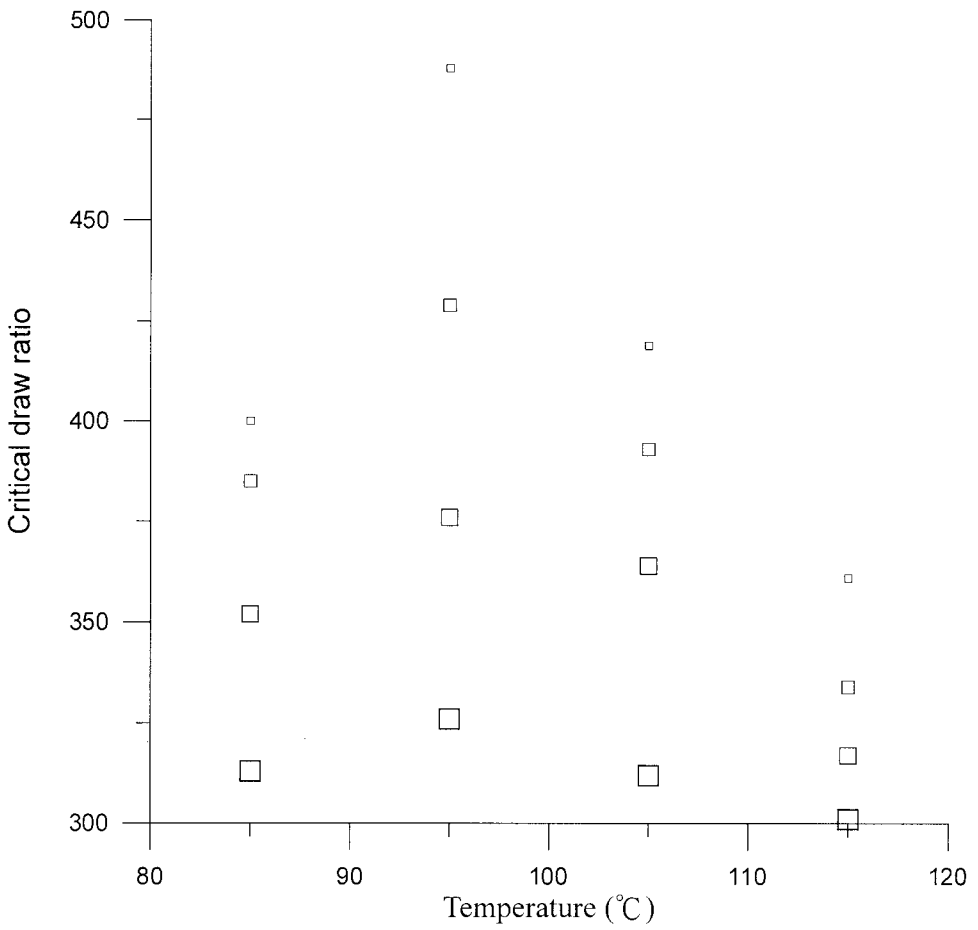

Figure 1 D_{rc} values of (▫) $UL_{-0.7-0°C}$, (□) $UL_{-0.7-35°C}$, (□) $UL_{-0.7-65°C}$, and (☐) $UL_{-0.7-95°C}$ specimens drawn at various temperatures.

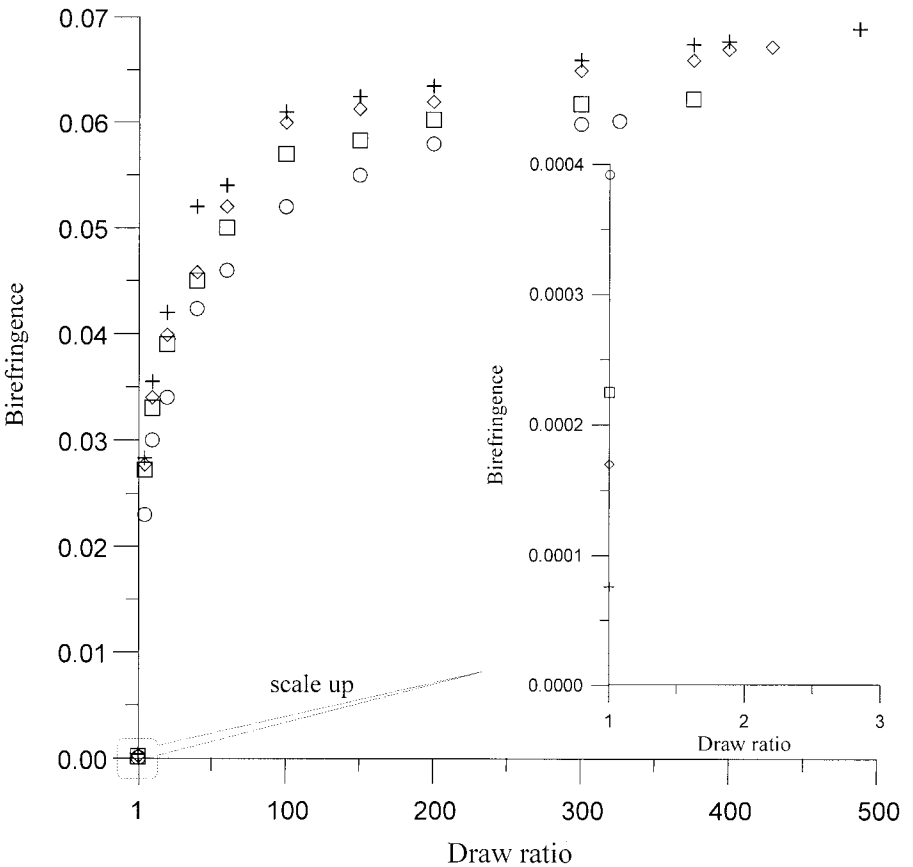

Figure 2 Birefringence values of various draw ratios of (+) $UL_{-0.7\text{-}0°C}$, (◇) $UL_{-0.7\text{-}35°C}$, (□) $UL_{-0.7\text{-}65°C}$, and (○) $UL_{-0.7\text{-}95°C}$ specimens drawn at 95°C.

mum value as the drawing rates approach an optimum value. Our recent investigations[22] have further found that the D_{raop} values of gel films can further be improved with a two-stage drawing process, in which the gel specimens are drawn at another T_{op} value after being drawn at 95°C, up to the transition draw ratio of 40.

In addition to the gel solution compositions and drawing conditions, it is generally recognized that the conditions used in the formation process after the spinning or solution casting of gel solutions can also have a significant influence on the morphology, microstructure, and drawing properties of the specimens formed during the aforementioned processes.[6,33–39] In this study, a systematic study of the influence of the formation temperature on the ultradrawing properties of UHMWPE/low-molecular-weight polyethylene (LMWPE) film specimens prepared from gel solutions of UHMWPE and LMWPE blends was carried out. The formation temperatures were found to have a significant effect on the drawability of the UHMWPE/LMWPE film specimens. Further investigations, including birefringence, thermal, and tensile experiments, were performed on the film specimens to further clarify the possible deformation mechanisms accounting for the interesting drawing properties found in this study.

EXPERIMENTAL

Materials and sample preparation

The UHMWPE resin used in this study is associated with a weight-average molecular (M_w) of 4.5×10^6, and it is called resin U in the following discussion. The linear LMWPE used in this study, called resin L, is a linear high-density polyethylene and is associated with an M_w of 5.0×10^4. Bruce Lu of Yung Chia Chemical Industrial Corp. (Kaohsiung, Taiwan) kindly supplied both UHMWPE and LMWPE resins. UHMWPE and LMWPE were mixed at a weight ratio of 98:2 and then were dissolved in decalin at 135°C for 90 min, and 0.1 wt % di-*t*-butyl-*p*-cresol was added as an antioxidant. The compositions of the gel solutions prepared in this study are summarized in Table I. The hot homogenized solutions were poured into an aluminum tray and cooled in a temperature-controlled oven so that gel films would form at a constant temperature. The gel-formation temperatures used in this study were 0, 35, 65, and 95°C, respectively. The deca-

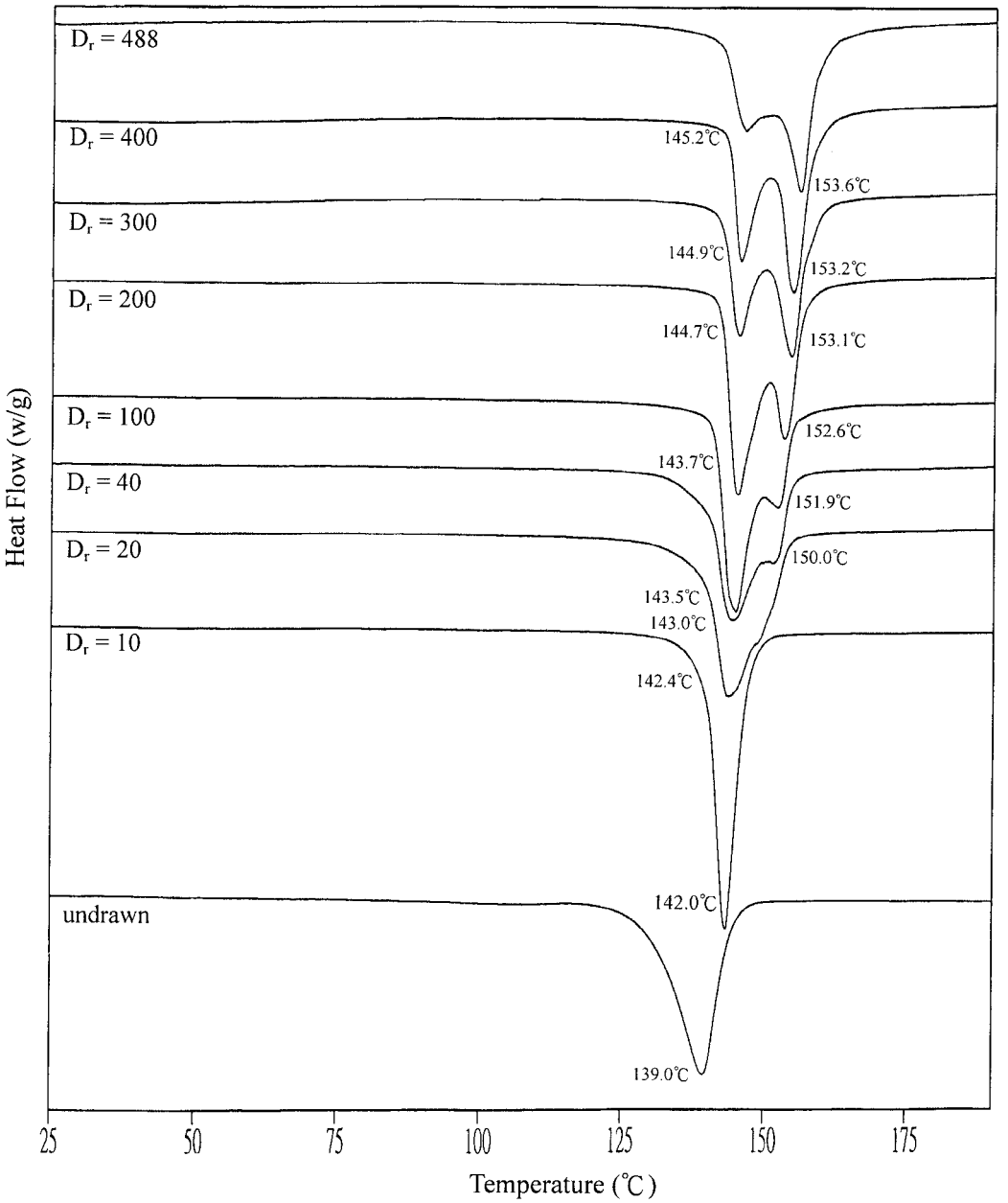

Figure 3 DSC thermograms of various draw ratios of $UL_{-0.7\text{-}0°C}$ gel films drawn at 95°C.

lin was then evaporated from the gels prepared at various formation temperatures in the temperature-controlled oven for more than 48 h. The dried gel films were immersed in ethanol for the removal of the antioxidant and residual traces of decalin; about 0.015 g of the residual solvent was present in 1 g of each completely dried film specimen. The prepared gel films had a thickness of around 250 μm.

Determination of the viscosity and critical concentration (C_c) of the solution

The viscosities of the polymer solutions were determined at 135°C with a Brookfield model LVDV-II$^+$ viscometer. As reported in our previous publication,[19] two distinct regions were found in plots of the reduced viscosities against the concentrations of the polymer solutions. The reduced viscosities increased slightly with the concentration in region 1, which was associated with low concentrations. However, the reduced viscosities increased dramatically as the concentrations of the solutions reached their critical values. The region associated with concentrations higher than C_c was called region 2. The value of C_c was determined by the intersection of two straight lines drawn parallel to the two distinct regions shown in these plots. The C_c values of solutions prepared in this study were determined in our previous studies[18] and are described later in the Results and Discussion section.

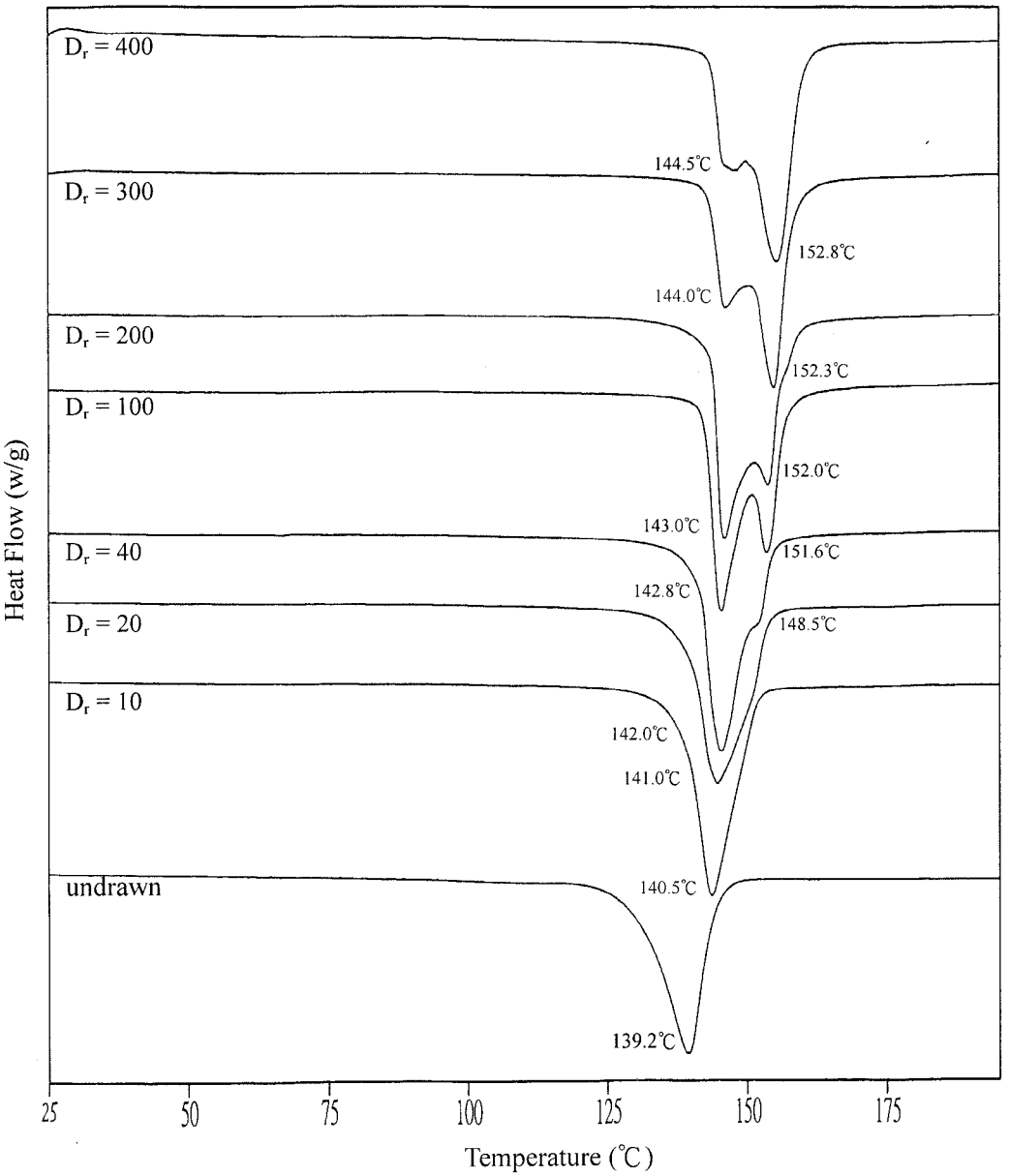

Figure 4 DSC thermograms of various draw ratios of $UL_{-0.7\text{-}35°C}$ gel films drawn at 95°C.

Drawing and tensile properties of the gel films

The strip specimens used in the drawing experiments were cut from the dry gel films and then stretched on a Tensilon RTA-1T testing machine equipped with a temperature-controlled oven at a crosshead speed of 20 mm/min. The specimens were 30 mm long and 10 mm wide. The specimens, prepared at different formation temperatures, were drawn at various temperatures (i.e., 85, 95, 105, and 115°C) to determine the temperature dependence of the drawability of the gel films. The draw ratio of each specimen was determined as the ratio of the marked displacement after and before drawing. The marked displacement before drawing was 5 mm. The tensile properties of the undrawn and drawn gel films were also determined with a Tensilon RTA-1T testing machine at 28°C and at a crosshead speed of 20 mm/min.

Birefringence and thermal analysis

The birefringence of the undrawn and drawn gel films was measured with a TFM 120 AFT polarizing microspectrometer. The thermal behaviors of all the samples were determined on a DuPont 2000differential scanning calorimeter. All scans were carried out at a heating rate of 10°C/min under flowing nitrogen at a flow rate of 25 mL/min. Samples weighing 0.5 or 10 mg were placed in standard aluminum sample pans for the determination of their melting temperatures and crystallinity percentages. The crystallinity per-

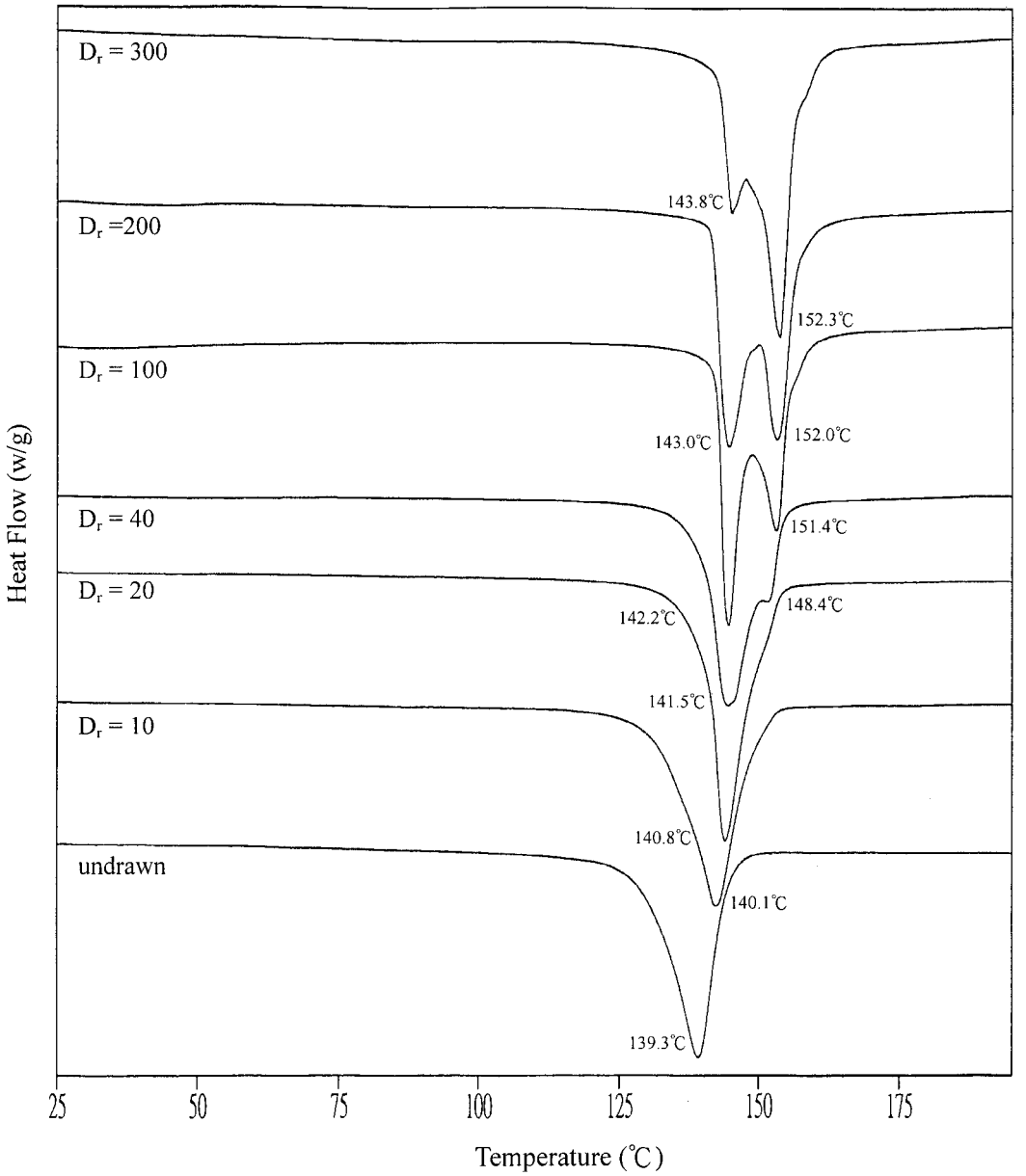

Figure 5 DSC thermograms of various draw ratios of $UL_{-0.7\text{-}65°C}$ gel films drawn at 95°C.

centages of the specimens were estimated with baselines drawn from 40 to 170°C and a perfect heat of fusion of polyethylene of 293 J/g.[23]

RESULTS AND DISCUSSION

Drawing properties of UHMWPE/LMWPE film specimens prepared at various formation temperatures

Table II summarizes the achievable draw ratios (D_{ra}) of the UHMWPE/LMWPE film specimens prepared at various concentrations and formation temperatures. Similar to those found in our previous investigations,[18–22] at any fixed drawing temperature, the achievable draw ratios of each of the gel film series prepared at a fixed formation temperature approached a maximum value when they were prepared at concentrations close to their C_c values. These achievable draw ratios obtained for specimens prepared near the C_c values are called the critical draw ratio (D_{rc}) in the following discussion. However, at any fixed drawing temperature, the D_{rc} values of the specimens prepared at various formation temperatures increased significantly as the formation temperatures were reduced (see Fig. 1 and Table II). For instance, at a drawing temperature of 85°C, the D_{rc} values of $UL_{-0.7}$ specimens increased from 313 to 400 as their formation temperatures were reduced from 95 to 0°C.

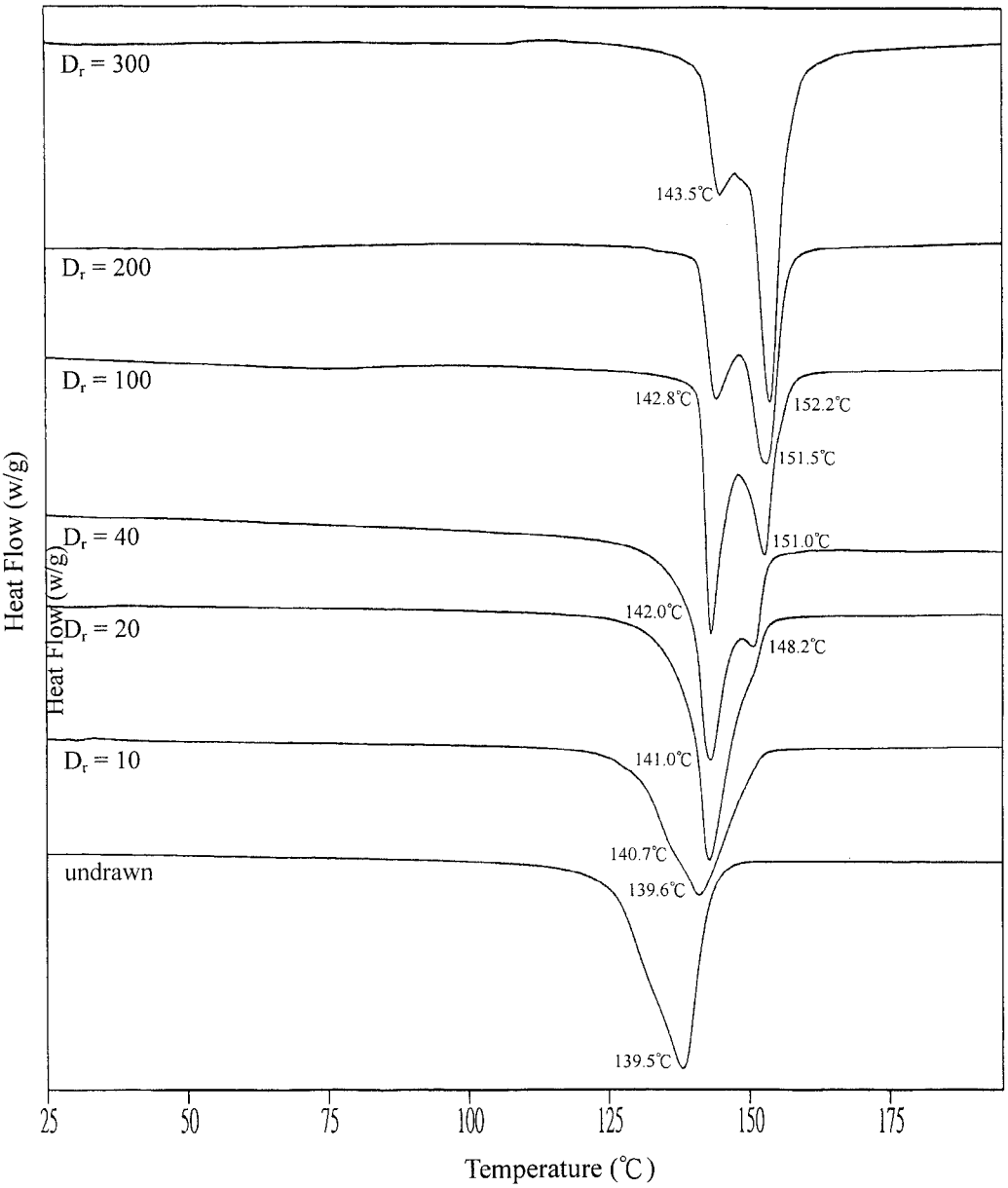

Figure 6 DSC thermograms of various draw ratios of $UL_{-0.7\text{-}95°C}$ gel films drawn at 95°C.

These specimens were drawn at various temperatures so that we could determine the temperature dependence of the values of D_{rc}, and we found that 95°C was the optimum drawing temperature for yielding the highest D_{rc} of $UL_{-0.7}$ specimens prepared at various formation temperatures. For instance, the D_{rc} values of the film specimens prepared at a formation temperature of 0°C (i.e., $UL_{-0.7\text{-}0°C}$ specimens) increased from 400 to 488 as the drawing temperatures increased from 85 to 95°C. However, the D_{rc} values of $UL_{-0.7\text{-}0°C}$ specimens significantly decreased from 488 to 361 when the drawing temperatures increased from 95 to 115°C. These D_{rc} values obtained for $UL_{-0.7}$ specimens drawn at T_{op} = 95°C are called the optimum critical draw ratio (D_{rcop}) in the following discussion. The D_{rcop} values of $UL_{-0.7}$ specimens prepared at a formation temperature if 0°C were about 14, 30, and 50% higher than those of $UL_{-0.7}$ specimens prepared at 35, 65, and 95°C, respectively (see Fig. 1).

Birefringence and thermal analysis of UHMWPE/LMWPE film specimens prepared at various formation temperatures

Typical birefringence values of various draw ratios of $UL_{-0.7}$ specimens drawn at 95°C are shown in Figure 2. Similar to those found in our previous investigations,[18–22] the values of the birefringence of $UL_{-0.7}$ specimens initially increased dramatically with the increasing draw ratio. The increasing rate of birefrin-

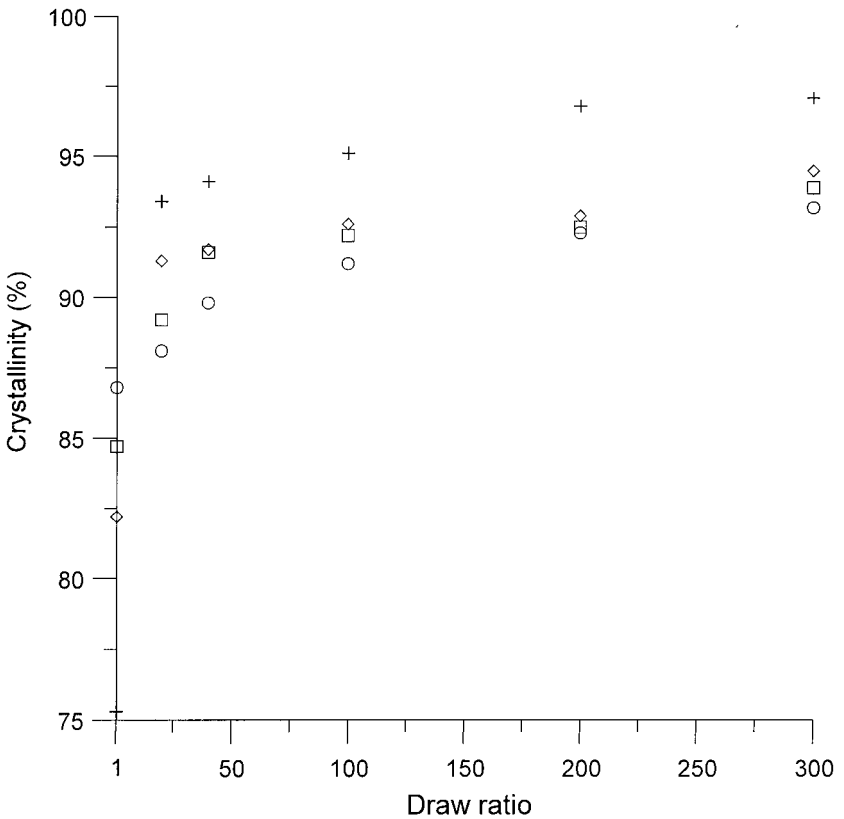

Figure 7 Crystallinity values of various draw ratios of (+) $UL_{-0.7\text{-}0°C}$, (◇) $UL_{-0.7\text{-}35°C}$, (□) $UL_{-0.7\text{-}65°C}$, and (○) $UL_{-0.7\text{-}95°C}$ specimens drawn at 95°C.

gence (IRB) became slower when the draw ratios of the drawn gel films were greater than about 20. In fact, IRB decreased consistently with the increasing draw ratio until its value reached about 200. After this value, IRB remained approximately constant with the increasing draw ratio. However, the birefringence values of the undrawn $UL_{-0.7}$ specimens remained almost constant and decreased slightly as their formation temperatures decreased from 95 to 0°C (see Fig. 2). However, after the drawing experiments, the drawn $UL_{-0.7}$ specimens prepared at lower formation temperatures always exhibited higher birefringence values than those of drawn specimens with the same draw ratio but prepared at higher formation temperatures. For instance, at a draw ratio of 100, the birefringence value of $UL_{-0.7\text{-}0°C}$ specimens was about 3, 8, and 20% higher than those of $UL_{0.7\text{-}35°C}$, $UL_{0.7\text{-}65°C}$, and $UL_{0.7\text{-}95°C}$ specimens, respectively. A similar tendency was observed for these drawn $UL_{-0.7}$ specimens with other fixed draw ratios (see Fig. 2).

Typical differential scanning calorimetry (DSC) thermograms of various draw ratios of $UL_{-0.7\text{-}0°C}$ specimens drawn at 95°C are summarized in Figure 3. The main melting endotherm, with a peak temperature at about 139°C, was found for $UL_{-0.7\text{-}0°C}$ specimens drawn at 95°C. The peak temperature associated with the main melting endotherm increased significantly with the draw ratio, and a small shoulder at a temperature of around 148°C was found on the right of the main melting endotherm when the $UL_{-0.7\text{-}0°C}$ specimens were stretched to a draw ratio of about 20. This small shoulder continued to grow into another melting endotherm, and the temperature associated with this new melting peak increased up to about 153.6°C as the draw ratio reached about 488. As shown in Figures 4–6, similar thermal properties were found for $UL_{-0.7\text{-}35°C}$, $UL_{-0.7\text{-}65°C}$, and $UL_{-0.7\text{-}95°C}$ specimens drawn at 95°C, respectively. However, the peak temperatures associated with the main melting endotherms of the undrawn $UL_{-0.7}$ specimens decreased slightly from 139.5 to 139.0°C as their formation temperatures decreased from 95 to 0°C. Similarly, the crystallinity percentages of the undrawn $UL_{-0.7}$ specimens decreased significantly from 86.8 to 75.3% as their formation temperatures decreased from 95 to 0°C (see Fig. 7). However, after the drawing experiments, the peak temperatures of the main and newly developed melting endotherms and the crystallinity percentages of the drawn $UL_{-0.7}$ specimens prepared at lower formation temperatures were always higher than those of drawn $UL_{-0.7}$ specimens having the same draw ratio but prepared at higher formation temperatures. For instance, at a draw ratio of 100, the peak temperatures of the main and new melting en-

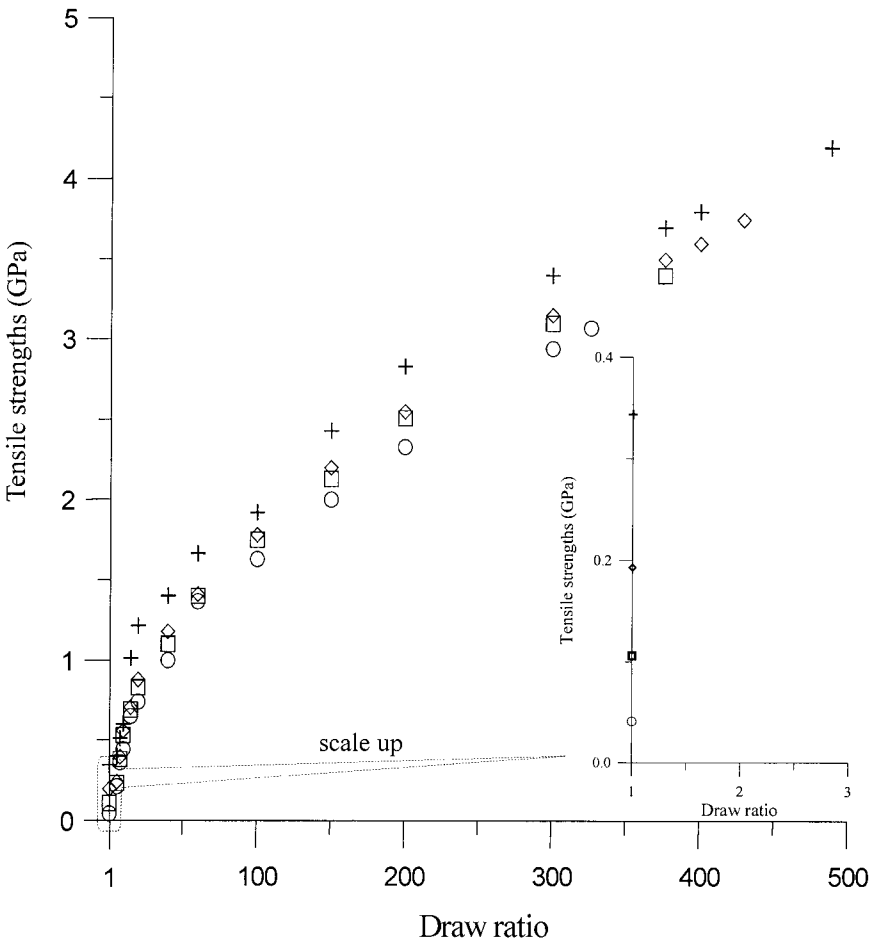

Figure 8 Tensile strengths of various draw ratios of (+$UL_{-0.7\text{-}0°C}$, (◇) $UL_{-0.7\text{-}35°C}$, (□) $UL_{-0.7\text{-}65°C}$, and (○) $UL_{-0.7\text{-}95°C}$ specimens drawn at 95°C.

dotherms increased from 142.0 and 151.0°C to 143.5 and 151.9°C, respectively, and the crystallinity percentages increased from 91.2 to 95.1% as the formation temperatures of drawn $UL_{-0.7}$ specimens were reduced from 95 to 0°C (see Fig. 7).

It is not completely clear what accounts for the interesting drawing, birefringence, and thermal properties previously described. It is generally recognized that the crystallization temperature can have a significant influence on the crystallization kinetics and crystalline morphology of polymers. Several investigations[40–46] have indicated that crystals obtained at low crystallization temperatures have a low degree of perfection and that these crystals can partially melt and recrystallize during the course of thermal analysis scans to yield thicker and/or more perfect crystals. On the basis of these premises, it is reasonable to believe that the low values of the crystallinity percentage, birefringence, and melting temperature found for $UL_{-0.7}$ specimens prepared at low formation temperatures were due to their low formation and/or crystallization temperatures because the molecular mobility of the UHMWPE molecules was reduced with decreasing temperatures that could inhibit the crystallization of the polymers at a low crystallization temperature. As a result, less perfect crystals with lower crystallinity percentage, birefringence, and melting temperature values were obtained when $UL_{-0.7}$ specimens were prepared at lower formation temperatures. Presumably, during the drawing process, these less perfect crystals and oriented structures of $UL_{-0.7}$ specimens could more easily be melted, disentangled, orientated, and effectively pulled out of folded lamellar crystals than those more perfect crystals and oriented structures of $UL_{-0.7}$ specimens prepared at higher formation temperatures, and so their drawability could be significantly improved over those of $UL_{-0.7}$ specimens prepared at higher formation temperatures.

Tensile properties of various draw ratios of UHMWPE/LMWPE film specimens prepared at various formation temperatures

As shown in Figures 8 and 9, the tensile strengths and moduli of each $UL_{-0.7}$ specimen were found to improve consistently as the draw ratios increased. Like

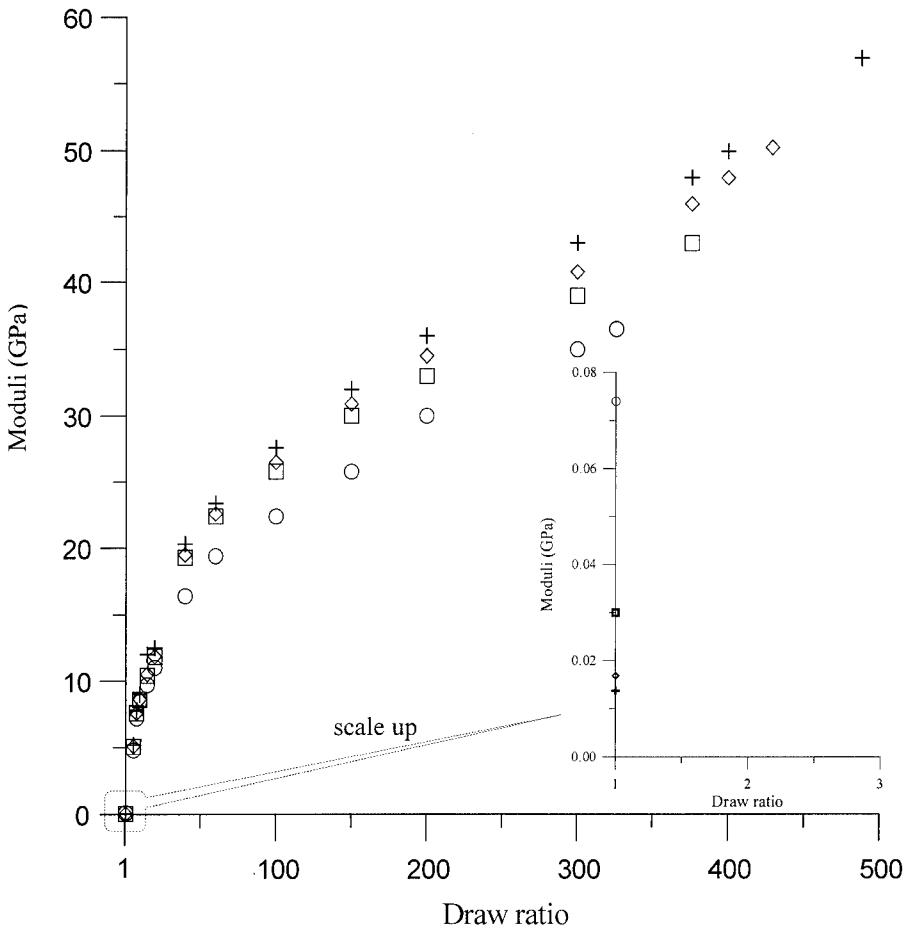

Figure 9 Moduli of various draw ratios of (+) $UL_{-0.7\text{-}0°C}$, (◇) $UL_{-0.7\text{-}35°C}$, (□) $UL_{-0.7\text{-}65°C}$, and (○) $UL_{-0.7\text{-}95°C}$ specimens drawn at 95°C.

the birefringence and thermal properties, the tensile strengths and moduli of the undrawn $UL_{-0.7}$ specimens remained almost constant and only slightly changed as their formation temperatures were reduced from 95 to 0°C. However, after the drawing experiments, the tensile strengths and moduli of the drawn $UL_{-0.7}$ specimens prepared at lower formation temperatures were significantly higher than those of specimens prepared at higher formation temperature but stretched to a same draw ratio. For instance, at a draw ratio of 100, the tensile strengths and moduli of $UL_{-0.7\text{-}0°C}$ specimens were about 7.8–17.8% and 4.2–23.2% higher, respectively, than those of drawn $UL_{-0.7}$ specimens prepared at other formation temperatures (see Figs. 8 and 9).

It is generally believed that the mechanical properties of drawn specimens depend mainly on the degree of orientation of the drawn specimens, as their molecular weights are constant.[29] As mentioned previously, the drawn $UL_{-0.7}$ specimens prepared at lower formation temperatures always exhibited higher values of the birefringence, crystallinity percentage, and melting temperature than drawn $UL_{-0.7}$ specimens having the same draw ratios but prepared at higher formation temperatures. These results suggest that a good orientation of UHMWPE molecules and more perfect crystals along the drawing direction had a beneficial influence on the tensile properties of the drawn $UL_{-0.7}$ specimens, which were obtained by the preparation of the $UL_{-0.7}$ specimens at a lower formation temperatures and drawing at an optimum drawing temperature.

CONCLUSIONS

At any fixed drawing temperature, the D_{rc} values of the $UL_{-0.7}$ specimens prepared at various formation temperatures increased significantly as the formation temperatures were reduced. In fact, with an optimum drawing temperature of 95°C, the D_{rc} values of $UL_{-0.7}$ specimens prepared at a formation temperature of 0°C were about 50% higher than those of $UL_{-0.7}$ specimens prepared at a formation temperature of 95°C. Further investigations found that the undrawn $UL_{-0.7}$ specimens prepared at low formation temperatures always exhibited lower values of the crystallinity percentage,

birefringence, and melting temperature than those prepared at higher formation temperatures. In contrast, the values of the crystallinity percentage, melting temperature, and birefringence of the drawn $UL_{-0.7}$ specimens prepared at lower formation temperatures were always significantly higher than those of drawn $UL_{-0.7}$ specimens having the same draw ratio but prepared at higher formation temperatures. Like the birefringence and thermal properties, the tensile strengths and moduli of the undrawn $UL_{-0.7}$ specimens only slightly decreased as their formation temperatures were reduced from 95 to 0°C. However, after the drawing experiments, the tensile strengths and moduli of the drawn $UL_{-0.7}$ specimens prepared at lower formation temperatures were significantly higher than those of specimens prepared at higher formation temperatures but stretched to the same draw ratio. Presumably, during the drawing process, these less perfect crystals and oriented structures of $UL_{-0.7}$ specimens could more easily be melted, disentangled, oriented, and effectively pulled out of folded lamellar crystals than those more perfect crystals of $UL_{-0.7}$ specimens prepared at higher formation temperatures; therefore, the drawability of $UL_{-0.7}$ specimens prepared at lower formation temperatures could significantly be improved.

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