Ultradrawing Behavior of One- and Two-Stage Drawn Gel Films of Ultrahigh Molecular Weight Polyethylene and Low Molecular Weight Polyethylene Blends

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ABSTRACT: The ultradrawing behavior of gel films of plain ultrahigh molecular weight polyethylene (UHMWPE) and UHMWPE/low molecular weight polyethylene (LMWPE) blends was investigated using one- and two-stage drawing processes. The drawability of these gel films were found to depend significantly on the temperatures used in the one- and two-stage drawing processes. The critical draw ratio (λ_c) of each gel film prepared near its critical concentration was found to approach a maximum value, when the gel film was drawn at an "optimum" temperature ranging from 95 to 105°C. At each drawing temperature, the one-stage drawn gel films exhibited an abrupt change in their birefringence and thermal properties as their draw ratios reached about 40. In contrast, the critical draw ratios of the two-stage drawn gel films can be further improved to be higher than those of the corresponding single-stage drawn gel films, in which the two-stage drawn gel films were drawn at another "optimum" temperature in the second drawing stage after they had been drawn at 95°C to a draw ratio of 40 in the first drawing stage. These interesting phenomena were investigated in terms of the reduced viscosities of the solutions, thermal analysis, birefringence, and tensile properties of the drawn and undrawn gel films. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci 70: 149-159, 1998

Key words: ultradrawing behavior; ultrahigh molecular weight polyethylene (UHM-WPE); low molecular weight polyethylene (LMWPE); one- and two-stage drawing process; drawability; critical draw ratio (λ_c) ; critical concentration

INTRODUCTION

Great progress has been made in the processing of high-performance fibers of ultrahigh molecular weight polyethylene (UHMWPE) since the 1970s.¹⁻¹¹ Among these processing methods, the gel spinning/casting method^{2,3} has attracted much attention because of its availability in production of high-strength/modulus fibers commercially. The method, by ultradrawing the gel specimens prepared by quenching solutions of UHM-WPE, is referred to as the gel-deformation method.

The drawability of the gel specimens was found to depend principally on the compositions of solutions from which the gels were made.^{12–15} The achievable drawability was reduced significantly as the gel films were prepared from 6 solutions whose concentrations deviated from their critical values, at which the numbers of entanglements in the coherent network structure of the gel films are too many or too few to yield the maximum

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extension of the UHMWPE during the gel-deformation process.^{12,13} In our recent study,^{14,15} the achievable draw ratios of the gel films of each weight ratio of low molecular weight polyethylene (LMWPE) to UHMWPE approached a maximum value when they were prepared at concentrations close to their critical concentrations. These critical draw ratios (λ_c) of the gel films prepared near their critical concentration were found to decrease significantly with an increasing amount of LMWPE added to the gel films. However, the addition of a small but optimum amount of LM-WPE in the films of UHMWPE/LMWPE blends can significantly improve their λ_c over that of gel film prepared from a pure UHMWPE gel solution, and this improvement in λ_c is further enhanced with decreasing lengths of short-chain branches of LMWPEs.¹⁵ Such fibers and ultradrawn gel films of UHMWPE/LMWPE blends are very important commercially, because the production rate of high-modulus fibers prepared from UHM-WPE gels is far below that commercially required and the drawability of gel films prepared from a pure UHMWPE solution can be lower than those of gel films prepared from UHMWPE/LMWPE blends because the enormous entanglements of UHMWPE gel films may prohibit the drawability of the gel films. However, very few investigations have been reported with regard to the preparation and drawing of the gel films of UHMWPE/ LMWPE blends. 14-19

In this study, the drawing behavior of gel films of plain UHMWPE and UHMWPE/LMWPE blends was investigated using one- and two-stage drawing processes. During the two-stage drawing process, varying temperatures were used in the second drawing stage after drawing gel films up to a draw ratio of 40 at 95°C. The effects of the temperature dependence of drawability, birefringence, and thermal behavior of gel films of UHM-WPE/LMWPE blends drawn by one- and twostage drawing processes were investigated.

EXPERIMENTAL

Materials and Sample Preparation

The UHMWPE resin used in this study is associated with a weight-average molecular weight (\overline{M}_w) of 4.5×10^6 , which will be referred to as resin U in the following discussion. On the other hand, the linear LMWPE used in this study will be referred to as resin L, which is a linear highdensity PE and is associated with an \overline{M}_w of 8.9 $\times 10^4$. Resins U and L were kindly supplied by Mr. Bruce Lu of the Yung Chia Chemical Industrial Corp. Mixtures of varying weight ratios of UHMWPE to LMWPE were dissolved in decalin at 135°C for 90 min, in which 0.1 wt % of 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 an oven to form the gel films at 35°C. The decalin was then evaporated from the gel in the oven. The dried gel was immersed in ethanol to remove the antioxidant and any residual trace of decalin. The prepared gel film had a thickness of about 250 μ m. No gel film can be obtained from pure LMWPE solutions, which is possibly due to their low molecular weight and lack of entanglements in the LMWPE solutions. The percentage of crystallinities of the fragments of the dry gels of pure L prepared at concentrations ranging from 5 to 10 g/dL are about 80%.

Determination of Viscosity and Critical Concentration of the Solution

The viscosities of the polymer solutions were determined at 135°C with a Brookfield viscometer Model LVDV-II⁺. As shown in our previous publication,^{14,15} two distinct regions were found on the plots of reduced viscosities against concentrations of the polymer solutions. The reduced viscosities increase slightly with concentration in region 1, which are associated with low concentrations. However, the reduced viscosities increase dramatically as the concentrations of the solutions reach their critical values. The region associated with concentrations higher than the critical concentration (C_c) is referred to as region 2. The value of C_c was determined by the intersection of the two straight lines drawn parallel to the two distinct regions shown in these plots. The values of the C_c of the solutions prepared in this study were determined in our previous studies,^{14,15} and will be described later in the Results and Discussion section.

Birefringence and Thermal Analysis

Birefringence of the drawn and undrawn gel films was measured by using a polarizing microspectrometer Model TFM-120 AFT. The thermal behavior studies of all samples were performed on a

Sample	Weight Ratio UHMWPE/LMWPE	Concentration (g/dL)		
$\begin{array}{c} U_{0.6} \\ U_{0.7} \\ U_{0.8} \\ U_{0.9} \\ U_{1.0} \end{array}$	100/0	$0.6 \\ 0.7 \\ 0.8 \\ 0.9 \\ 1.0$		
$\begin{array}{c} UL_{A\text{-}0.6} \\ UL_{A\text{-}0.7} \\ UL_{A\text{-}0.8} \\ UL_{A\text{-}0.9} \\ UL_{A\text{-}1.0} \end{array}$	99.5/0.5	0.6 0.7 0.8 0.9 1.0		
$\begin{array}{c} UL_{B-0.6} \\ UL_{B-0.7} \\ UL_{B-0.8} \\ UL_{B-0.9} \\ UL_{B-1.0} \end{array}$	99/1	0.6 0.7 0.8 0.9 1.0		
$\begin{array}{c} \rm{UL}_{\rm{C}\text{-}0.6} \\ \rm{UL}_{\rm{C}\text{-}0.7} \\ \rm{UL}_{\rm{C}\text{-}0.8} \\ \rm{UL}_{\rm{C}\text{-}0.9} \\ \rm{UL}_{\rm{C}\text{-}1.0} \end{array}$	98/2	$0.6 \\ 0.7 \\ 0.8 \\ 0.9 \\ 1.0$		
$\begin{array}{c} {\rm UL}_{\rm D\text{-}0.6} \\ {\rm UL}_{\rm D\text{-}0.7} \\ {\rm UL}_{\rm D\text{-}0.8} \\ {\rm UL}_{\rm D\text{-}0.9} \\ {\rm UL}_{\rm D\text{-}1.0} \end{array}$	95/5	$0.6 \\ 0.7 \\ 0.8 \\ 0.9 \\ 1.0$		
$\begin{array}{c} {\rm UL}_{{\rm E}\text{-}0.7} \\ {\rm UL}_{{\rm E}\text{-}0.8} \\ {\rm UL}_{{\rm E}\text{-}0.9} \\ {\rm UL}_{{\rm E}\text{-}1.0} \\ {\rm UL}_{{\rm E}\text{-}1.1} \end{array}$	90/10	$0.7 \\ 0.8 \\ 0.9 \\ 1.0 \\ 1.1$		
$\begin{array}{c} \mathrm{UL}_{\mathrm{F}\text{-}0.9} \\ \mathrm{UL}_{\mathrm{F}\text{-}1.0} \\ \mathrm{UL}_{\mathrm{F}\text{-}1.1} \\ \mathrm{UL}_{\mathrm{F}\text{-}1.2} \\ \mathrm{UL}_{\mathrm{F}\text{-}1.3} \end{array}$	80/20	$0.9 \\ 1.0 \\ 1.1 \\ 1.2 \\ 1.3$		
$\begin{array}{c} UL_{G\text{-}1.1} \\ UL_{G\text{-}1.2} \\ UL_{G\text{-}1.3} \\ UL_{G\text{-}1.4} \\ UL_{G\text{-}1.5} \end{array}$	70/30	$1.1 \\ 1.2 \\ 1.3 \\ 1.4 \\ 1.5$		

 Table I
 Compositions of Solutions of UHMWPE

 and UHMWPE/LMWPE Blends

DuPont differential scanning calorimeter (DSC) Model 2000. 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 10 and 0.5 mg were placed in standard aluminum sample pans for determination of the degree of crystallinity and melting temperature, respectively. The degrees of crystallinity of all the samples were estimated using baselines drawn from 40 to 170° C and a perfect heat fusion of 293 J/g.²⁰

Drawing Experiments

The strip specimens used in the drawing experiments were cut from the dried gel films and then stretched on a Tensilon testing machine Model RTA-1T equipped with a temperature-controlled oven at a crosshead speed of 20 mm/min. The dimensions of the specimens were 30 mm in length and 10 mm in width. The specimens were first drawn at varying temperatures to investigate the temperature dependence of the drawability of the UHMWPE/LMWPE gel films at a fixed drawing rate of 20 mm/min. The method of drawing specimens at a fixed temperature throughout the drawing process will be referred to as the one-stage drawing process in the following discussion. The temperatures used in the one-stage drawing process were 35, 65, 85, 95, 105, and 115°C. As will be shown later, the one-stage drawn gel films exhibited an abrupt change in birefringence and thermal properties as their draw ratio reached about 40. Presumably, the birefringence, thermal, and drawing behaviors of the gel films can be correlated to the molecular mobility and drawing temperatures used in the drawing processes. To further investigate the effect of the drawing process and temperature on the drawability of the gel films, these gel films were drawn at other fixed temperatures, after they had been drawn at 95°C, up to the transition draw ratio of 40. This type of drawing experiment will be referred to as the two-stage drawing process in the following discussion. The drawing temperatures used in the second drawing stage drawing process were 65, 115, and 125°C. 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 using a Tensilon testing machine Model RTA-1T at 28°C and a crosshead speed of 20 mm/min.

RESULTS AND DISCUSSION

Drawing Properties of One-Stage Drawn Gel Films of UHMWPE and UHMWPE/LMWPE Blends

The concentration and temperature dependence of achievable draw ratios (D_r) of one-stage drawn



Figure 1 The λ_c of (\diamondsuit) U, (\bigtriangleup) UL_A, (\Box) UL_B, (\bigcirc) UL_C, (\bigstar) UL_D, (\bigtriangledown) UL_E, (\bullet) UL_F, and (+) UL_G sample series drawn at varying temperatures.

gel films of plain UHMWPE and UHMWPE/LM-WPE blends are summarized in Figure 1 and Table II. The D_r of the gel films drawn at any fixed temperature approached the maximum values, when they were prepared at concentrations close to their C_c (see Table II). It is generally recognized that gel samples prepared near their C_c are associated with an optimum number of tie molecules and with UHMWPE molecules appropriately buried in the lamellar crystals, which thus can be extended to the highest D_r during the drawing process, because the UHMWPE molecules can be effectively pulled out of the lamellar crystals without significant breakage of the stress-concentrated tie molecules. These achievable D_r , obtained for samples prepared near their C_c , will be referred to as the critical draw ratios (λ_c) in the following discussion (see Table II). At each drawing temperature, the λ_c of many of the gel films containing less than 5 wt % of LMWPE in UHM-WPE are significantly higher than that of the gel film of pure UHMWPE (see Tables I and II). These results suggest that the presence of optimum amounts of LMWPE in the gel films of UH-MWPE/LMWPE blends can significantly improve their λ_c . This improvement in the λ_c of these gel films has been attributed^{14,15} to a suitable reduction of the number of entanglements within gel films caused by the addition of an optimum

amount of LMWPE in the UHMWPE/LMWPE blends.

On the other hand, it is worth noting that the maximum critical draw ratio (λ_{cmax}) of each gel film drawn at 20 mm/min was obtained when it was drawn at an "optimum" temperature ranging from 95 to 105°C (see Fig. 1). The λ_c of each gel film decreased significantly as the drawing temperature deviated from the "optimum" range. For instance, the λ_c of sample UL_{C-0.7} increased from 10 to 360 as the drawing temperatures increased from 35 to 95°C, but decreased to 295 as the drawing temperatures further increased to 115°C. Presumably, these interesting phenomena can be correlated to the temperature dependence of the molecular motions of PE molecules in the amorphous and crystalline phases during the drawing processes. It is well known that the molecular mobility of PE molecules originating from the crystalline and amorphous regions can be activated with increasing temperature. Three relaxation processes of PE have been extensively studied.^{21–23} It is generally considered that the β $(\sim -20^{\circ}\mathrm{C})$ and $\gamma\text{-transitions}$ $(\sim -120^{\circ}\mathrm{C})$ are due to the motion of branches^{24-27} and to the crankshaft motion of short polymer segments requiring a minimum of three methylene units $^{23,27-29}$ in the amorphous matrix, respectively. In contrast, the α -transition (~ 45°C) is attributed to motions or

	C_c	D_r of Gel Films Drawn at					
Sample		35°C	65°C	85°C	95°C	105°C	115°C
${f U_{0.6}} {f U_{0.7}}$	0.70	4 7	50 68	$\begin{array}{c} 145\\ 162 \end{array}$	$216\\240$	209 234	201 228
U _{0.8}		5	60	152	224	215	201
$U_{0.9}$ $U_{1.0}$		5 4	55 53	$\frac{125}{110}$	$\frac{221}{200}$	$\frac{209}{204}$	195 190
ULARG		4	75	165	234	226	215
UL _{A-0.7}	0.71	8	87	185	295	253	232
UL _{A-0.8}		6	80	174	244	234	210
$UL_{A-0.9}$		6	74	168	215	210	198
$UL_{A-1.0}$		5	71	138	206	203	194
$\mathrm{UL}_{\mathrm{B-0.6}}$		5	89	172	234	225	214
$UL_{B-0.7}$	0.73	8	106	209	330	276	243
UL _{B-0.8}		7	93	184	245	235	225
UL _{B-0.9}		6	75	176	229	219	213
$UL_{B-1.0}$		5	68	171	216	203	199
$\mathrm{UL}_{\mathrm{C}\text{-}0.6}$		5	105	189	277	242	222
$\mathrm{UL}_{\mathrm{C}\text{-}0.7}$	0.76	10	141	220	360	301	295
$UL_{C-0.8}$		8	132	200	288	286	268
UL _{C-0.9}		7	121	196	268	254	250
$UL_{C-1.0}$		6	104	176	238	229	221
$\mathrm{UL}_{\mathrm{D-0.6}}$		6	95	177	210	193	186
$\mathrm{UL}_{\mathrm{D-0.7}}$		8	110	208	236	225	205
$\mathrm{UL}_\mathrm{D-0.8}$	0.84	6	130	214	295	283	276
$UL_{D-0.9}$		6	110	164	285	268	259
$UL_{D-1.0}$		5	92	150	240	234	221
$\mathrm{UL}_{\mathrm{E}\text{-}0.7}$		5	93	150	195	189	168
$UL_{E-0.8}$		7	101	152	204	201	197
$UL_{E-0.9}$	0.85	6	107	167	229	221	216
$UL_{E-1.0}$		5	99	140	213	211	201
$UL_{E-1.1}$		5	89	135	209	195	186
UL_{F} -0.9		4	54	101	185	157	134
$\mathrm{UL}_\mathrm{F-1.0}$		5	78	134	194	176	158
$UL_{F-1.1}$	1.13	6	85	145	200	194	172
$UL_{F-1.2}$		5	68	124	175	166	111
$\mathrm{UL}_{\mathrm{F}\text{-}1.3}$		5	51	110	160	143	108
$UL_{G-1.1}$		3	48	78	115	109	101
$UL_{G-1.2}$	1 00	5	55	98	121	138	124
$UL_{G-1.3}$	1.28	5	67	110	144	156	134
$UL_{G-1.4}$		4	50	88	136	122	102
$UL_{G-1.5}$		3	48	75	130	108	98

Table II C_c and Achievable D_r of Gel Films Drawn at Various Temperatures

deformations in the interfacial region (tie molecules loops, folds, etc.), which originate from chain mobility in the crystal.²³ In addition to these molecular motions, some of the less perfect crystals start to melt at drawing temperatures well above the α -transition temperature. These relaxation processes and molecular motions, due to the partial melting of less perfect crystals, can help the disentanglement of the UHMWPE molecules and effectively pull the UHMWPE molecules out of the lamellar crystals without significant breakage of the stress-concentrated tie mol-



Figure 2 Birefringence of various D_r of samples UL_C-0.7 drawn at (\Box) 35°C, (\triangle) 65°C, (\blacktriangle) 95°C, and (\bigcirc) 115°C.

ecules. Therefore, the λ_c of each gel film improves as the drawing temperatures increase from 35 to 95°C. At further higher temperatures, the crystals are melted to an extent that many of the UHMWPE molecules can be easily disentangled in the melt regions during the drawing process and the stress can no longer distribute relatively evenly along the tie molecules and crystal boundaries. Under such circumstances, the UHMWPE molecules cannot be effectively pulled straight out of the lamellar crystals. As a consequence, the λ_c of each gel film decreases as the drawing temperatures increase from 95 to 115°C.

Birefringence and Thermal Analysis of One-Stage Drawn Gel Films

Typical birefringence of the undrawn and onestage drawn gel films are shown in Figure 2. At each drawing temperature, the values of the birefringence of sample $UL_{C-0.7}$ initially increase dramatically with the D_r and then slightly increase to their achievable D_r . For instance, at a drawing temperature of 95°C, the increasing rate of birefringence becomes slower when the D_r of the gel film is greater than about 40. To define the "transition" value of the D_r , two lines were drawn parallel to the initial and final regions of the curves shown in Figure 2. The intersection of the two lines is defined as the "transition" D_r . As shown in Figure 2, the value of the transition D_r is about 40 for gel films drawn at various temperatures. On the other hand, it is interesting to note that sample $UL_{C-0.7}$ drawn at higher temperatures always exhibited higher values of birefringence than those of samples drawn at the same D_r but at lower temperatures (see Fig. 2). Presumably, this can be due to the higher mobility of these UHMWPE molecules at higher temperatures such that the UHMWPE molecules can be more easily oriented along the drawing direction during the drawing processes. However, it is not clear why these drawn gel films exhibited an abrupt change in birefringence properties as their D_r reached about 40.

Typical DSC thermograms of these gel films drawn at varying temperatures and D_r are shown in Figure 3. At each drawing temperature, a main melting endotherm with a peak temperature of about 138°C was found for the undrawn gel films. The main melting temperature of the gel films then increases significantly as the D_r increases. Most interestingly, a small shoulder at a temperature of around 148°C was found on the right of the main melting endotherm when these gel films were stretched to a D_r of about 40 at each drawing temperature. In fact, this small shoulder continues to grow into another melting endotherm, and the temperature associated with this new melting peak increases up to about 152°C as the gel film was drawn up to a D_r of 360 at 95°C [see Fig. 3(c)]. Similar behavior was found for gel films drawn at other temperatures [see Figs. 3(b)–(e)]. The interesting double-melting behavior found above apparently is correlated to the birefringence transition as a function of the D_r observed previously. It is generally recognized that the orientation of the polymer chain and reduction of crystal defects during drawing can enhance the perfection and melting temperature of the crystal. However, it is not completely clear what accounts for the above interesting melting behavior and the abrupt change in birefringence and thermal properties as the D_r of the gel film reaches around 40.

Drawing Properties of Two-Stage Drawn Gel Films of UHMWPE and UHMWPE/LMWPE Blends

The achievable D_r of two-stage drawn gel films are summarized in Table III and Figure 4. Similar to those found for single-stage drawn gel films, the achievable D_r of two-stage drawn gel films approached the maximum values, when they were prepared at concentrations close to their C_c



Figure 3 DSC thermograms of varying D_r of sample UL_{A-0.7} drawn at (a) 65, (b) 85, (c) 95, (d) 105, and (e) 115°C.

(see Table III). At any temperature used in the second drawing stage, the λ_c of two-stage drawn gel films containing less than 5 wt % of LMWPE in UHMWPE are also significantly higher than those of the two-stage drawn gel film of pure UHMWPE (see Fig. 4). On the other hand, it is worth noting that the λ_c of two-stage drawn gel films can be further improved as the temperatures used in the second drawing stage ($T_{\rm sec}$) increase to about 115°C. The λ_c of two-stage drawn gel films then decrease significantly as $T_{\rm sec}$ continues to increase to 125°C. In fact, some of the λ_c of the two-stage drawn gel films are even higher than the maximum λ_c obtained using a one-stage

drawing temperature of 95°C (see Fig. 4 and Table III). For instance, the value of the λ_c of sample UL_{C-0.7} drawn at a $T_{\rm sec}$ of 115°C reaches 425, which is about 20% higher than the maximum λ_c (~ 360) obtained for sample UL_{C-0.7} drawn at 95°C. In contrast, the values of λ_c of the two-stage drawn gel films could not even reach 100, when they were drawn at a $T_{\rm sec}$ 65°C. These values are significantly lower than those of corresponding samples drawn using a one-stage drawing temperature of 95°C. These results suggest that the λ_c of the two-stage drawn gel films can be further improved to be higher than those of the corresponding one-stage drawn gel films as $T_{\rm sec}$ is in-



Figure 3 (Continued)

creased to the optimum temperature, which must be higher than the temperature used in the onestage drawing process.

Birefringence of Two-Stage Drawn Gel Films of UHMWPE and UHMWPE/LMWPE Blends

Typical plots of the birefringence of two-stage drawn gel films as a function of D_r are shown in Figure 5. At a fixed D_r of more than 40, the birefringence of the two-stage drawn sample UL_{C-0.7} was found to improve consistently with increas-

ing $T_{\rm sec}$ after being drawn to a D_r of 40 at 95°C. For example, at a fixed D_r above 40, the value of the birefringence of sample UL_{C-0.7} drawn at a $T_{\rm sec}$ of 125°C is about 5–10% higher than that of the corresponding one-stage drawn sample drawn at 95°C. In contrast, at a D_r of 60, the value of the birefringence of a sample drawn at a $T_{\rm sec}$ of 65°C decreases to about 5% lower than that of corresponding one-stage drawn sample drawn at 95°C. It is not completely clear what accounts for these interesting phenomena of the two-stage drawn gel films. Nevertheless, these results suggest that



Figure 3 (Continued)

the two-stage drawing process can provide the gel films with better drawability and birefringence than can the one-stage drawing process, as long as $T_{\rm sec}$ is optimum and high enough to provide a better and suitable molecular mobility from the crystalline and amorphous regions during the drawing process of the gel films.

Tensile Properties of One- and Two-Stage Drawn Gel Films of UHMWPE and UHMWPE/LMWPE Blends

The tensile strengths and moduli of one- and twostage drawn gel films were found to improve consistently as the D_r increased (see Figs. 6 and 7). At a fixed D_r , the tensile strengths and moduli of the one- or two-stage drawn gel films were also found to improve substantially as the drawing

Table III C_c and Achievable D_r of Two-Stage Drawn Gel Films

		D_r of Gel Films Drawn at a $T_{ m sec}$ of			
Sample	C_c	65°C	115°C	125°C	
$\begin{array}{c} U_{0.6} \\ U_{0.7} \\ U_{0.8} \\ U_{0.9} \\ U_{1.0} \end{array}$	0.70	56 76 70 68 64	290 356 331 321 295	281 292 273 251 244	
$\begin{array}{c} {\rm UL}_{\rm A-0.6} \\ {\rm UL}_{\rm A-0.7} \\ {\rm UL}_{\rm A-0.8} \\ {\rm UL}_{\rm A-0.9} \\ {\rm UL}_{\rm A-1.0} \end{array}$	0.71	61 77 72 66 65	330 399 385 375 363	303 359 312 286 251	
$\begin{array}{c} {\rm UL}_{\rm B\text{-}0.6} \\ {\rm UL}_{\rm B\text{-}0.7} \\ {\rm UL}_{\rm B\text{-}0.8} \\ {\rm UL}_{\rm B\text{-}0.9} \\ {\rm UL}_{\rm B\text{-}1.0} \end{array}$	0.73	65 78 75 72 68	320 405 382 374 365	339 365 359 313 295	
$\begin{array}{c} {\rm UL_{C-0.6}} \\ {\rm UL_{C-0.7}} \\ {\rm UL_{C-0.8}} \\ {\rm UL_{C-0.9}} \\ {\rm UL_{C-1.0}} \end{array}$	0.76	68 86 75 69 67	$363 \\ 425 \\ 407 \\ 385 \\ 374$	342 384 358 321 304	
$\begin{array}{c} {\rm UL}_{\rm D\text{-}0.6} \\ {\rm UL}_{\rm D\text{-}0.7} \\ {\rm UL}_{\rm D\text{-}0.8} \\ {\rm UL}_{\rm D\text{-}0.9} \\ {\rm UL}_{\rm D\text{-}1.0} \end{array}$	0.84	65 62 82 68 65	375 389 400 356 286	321 301 360 281 270	
$\begin{array}{c} \rm{UL}_{E\text{-}0.7} \\ \rm{UL}_{E\text{-}0.8} \\ \rm{UL}_{E\text{-}0.9} \\ \rm{UL}_{E\text{-}1.0} \\ \rm{UL}_{E\text{-}1.1} \end{array}$	0.85	62 65 70 60 52	286 310 349 332 256	287 299 319 278 240	
$\begin{array}{c} {\rm UL}_{\rm F\text{-}0.9} \\ {\rm UL}_{\rm F\text{-}1.0} \\ {\rm UL}_{\rm F\text{-}1.1} \\ {\rm UL}_{\rm F\text{-}1.2} \\ {\rm UL}_{\rm F\text{-}1.3} \end{array}$	1.13	50 59 62 53 48	279 278 291 268 253	243 263 281 253 241	
$\begin{array}{c} UL_{G\text{-}1.1} \\ UL_{G\text{-}1.2} \\ UL_{G\text{-}1.3} \\ UL_{G\text{-}1.4} \\ UL_{G\text{-}1.5} \end{array}$	1.28	$47 \\ 48 \\ 58 \\ 51 \\ 46$	253 265 274 240 219	186 215 226 203 186	



Figure 4 λ_c of two-stage drawn gel films draw at a T_{sec} of (\triangle) 65°C, (\bigcirc) 95°C, (\blacktriangle) 115°C, and (\diamondsuit) 125°C.

temperatures used in the one- and two-stage drawing processes increased. As mentioned previously, the degree of orientation and/or birefringence of the UHMWPE molecules increased significantly with the D_r and improved substantially with the drawing temperatures used in the first and second drawing stages, when the gel films were stretched to a fixed D_r . These results suggest



Figure 5 Birefringence of varying D_r of sample UL_C. 0.7 drawn using a T_{sec} of (\bigcirc) 65°C, (\triangle) 95°C, (+) 115°C, and (\diamondsuit) 125°C.



Figure 6 Tensile strengths of two-stage drawn sample UL_{A-0.7} drawn of a T_{sec} of (\diamond) 65°C, (\bigcirc) 115°C, and (\triangle) 125°C, and of one-stage drawn sample UL_{A-0.7} with a drawing temperature of (\blacklozenge) 65°C, (+) 85°C, (\Box) 95°C, (\Leftrightarrow) 105°C, and (\bigtriangledown) 115°C.

that the orientation of the UHMWPE molecules along the drawing direction has a beneficial influence on the tensile strengths and moduli of these gel films.



Figure 7 Moduli of two-stage drawn sample UL_{A-0.7} drawn at a T_{sec} of (\diamond) 65°C, (\bigcirc) 115°C, and (\triangle) 125°C, and of one-stage drawn sample UL_{A-0.7} with a drawing temperature of (\blacklozenge) 65°C, (+) 85°C, (\Box) 95°C, (\overleftrightarrow) 105°C, and (\bigtriangledown) 115°C.

CONCLUSIONS

The achievable D_r of the one-stage drawn gel films of the UHMWPE and the UHMWPE/LM-WPE blends approached the maximum values when they were prepared at a concentration close to their C_c . At a fixed drawing rate, the λ_c obtained for each gel film prepared near its C_c was found to reach a maximum value as it was drawn at an "optimum" temperature ranging from 95 to 105°C. The λ_c of each gel film was reduced significantly as the drawing temperature deviated from the "optimum" range. The birefringence and thermal properties of these one-stage drawn gel films drawn at each temperature exhibited an abrupt change as their D_r reached about 40. Based on this transition D_r , the λ_c of the two-stage drawn gel films can be further improved as the temperatures used in the second drawing stage increased to an optimum temperature of 115°C. In fact, the λ_c of two-stage drawn sample can be improved to about 20% higher than the maximum λ_c obtained for the one-stage drawn sample. Presumably, the temperature dependence of the drawability of the one- and two-stage drawn gel films is correlated to the temperature dependence of molecular mobility of PE molecules originating from the crystalline and amorphous regions during the drawing process. On the other hand, the tensile strengths and moduli of each one- and two-stage drawn gel films were found to improve consistently with increasing D_r or to increase with drawing temperatures used in the first and second drawing stages, when they were stretched to a fixed D_r . These improved tensile properties are attributed to the increased orientation of the UHMWPE molecules along the drawing direction.

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