

Modeling of Creep and Shrinkage Behavior of Polymeric Films Used in Magnetic Tapes

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ABSTRACT: For ever increasing high recording densities of magnetic tape drives, improved dimensional stability of the polymeric films used as magnetic tape substrates is required. During storage and use, creep and shrinkage occur simultaneously and it needs to be minimized. To obtain constitutive relationships for creep and shrinkage, these contributions need to be separated and modeled. A mathematical model based on Kelvin–Voigt models has been developed to characterize simultaneous creep and shrinkage behavior to obtain the constitutive relationships for creep and

shrinkage. Experiments have been performed to separate out creep and shrinkage effects and this model has been used to compensate the effect of shrinkage on creep data and to get true creep data. The experimental creep and shrinkage data of various films have been modeled to obtain viscoelastic parameters. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 91: 78–88, 2004

Key words: creep; shrinkage; Kelvin–Voigt model; viscoelastic properties; films

INTRODUCTION

Poly(ethylene terephthalate) (PET) and poly(ethylene naphthalate) (PEN) polymeric films are widely used in magnetic storage tapes as substrate materials.¹ The production of biaxially oriented PET and PEN polymeric films involve melt extrusion of the polymers, rapid quench from the extrusion temperatures, a subsequent heating and stretching of the films, and finally, a heat-setting or stabilization process.¹ A schematic of the manufacturing process and the molecular changes with various operations are shown in Figure 1(a). In essence the PET and PEN resins are extruded between T_m (melting temperature) and $T_m + 70^\circ\text{C}$, through a high-precision filter (with filter diameter of 3–10 μm ^{1,2}) onto a cold quenching drum to give an amorphous glassy film. The amorphous film is then heated between the glass-transition temperature (T_g) and $T_g + 70^\circ\text{C}$ and drawn in two or three steps of drawing to achieve the desired modulus and strength. The draw ratio in the machine direction (MD) is 4 to 6, and the draw ratio in the transverse direction (TD) is about 3 to 6. The heat-setting temperature and time are selected preferably between 190 to 250°C and 1 to 60 s, respectively, to achieve desired crystallinity and relaxation of residual stresses (to be discussed later). A

schematic of the temperature, strain, and stress variation during various stages of manufacturing is shown in Figure 1(b). Finally the film is cooled to ambient temperature and wound onto compliant hubs.

Internal stresses are developed in the film during various stages in manufacturing. After extrusion, internal stresses are almost zero. However, during drawing the polymer molecules are stretched away from their balanced state and experience internal stresses. The internal compressive stresses in the film that develop during biaxial drawing are not fully recovered after heat setting. These internal stresses remain frozen at ambient temperature because of high intermolecular friction and are referred to as “residual stresses.” The relaxation of residual stresses results in shrinkage during storage and use of these films [Fig. 2(a)]. The shrinkage is accelerated at elevated temperature because of the high mobility of the polymer molecules and their segments.

Crystallization occurs during the drawing and heat-setting processes at a temperature above the T_g of the film. The typical crystallinities in PET and PEN films are 40–50% and 30–40%, respectively.³ The crystalline and amorphous structure of the film is highly oriented along the direction of higher draw ratio [as shown in Fig. 1(a)]. If the draw ratio in MD is greater than that in TD, crystallites are more likely to orient toward the MD direction. Correspondingly, the relaxation of residual stresses results in rotation of crystallites away from the MD direction, as shown in Figure 2(b).

Magnetic tapes require thin substrates and high areal densities (track density \times linear density) to meet the demand for the advanced magnetic storage de-

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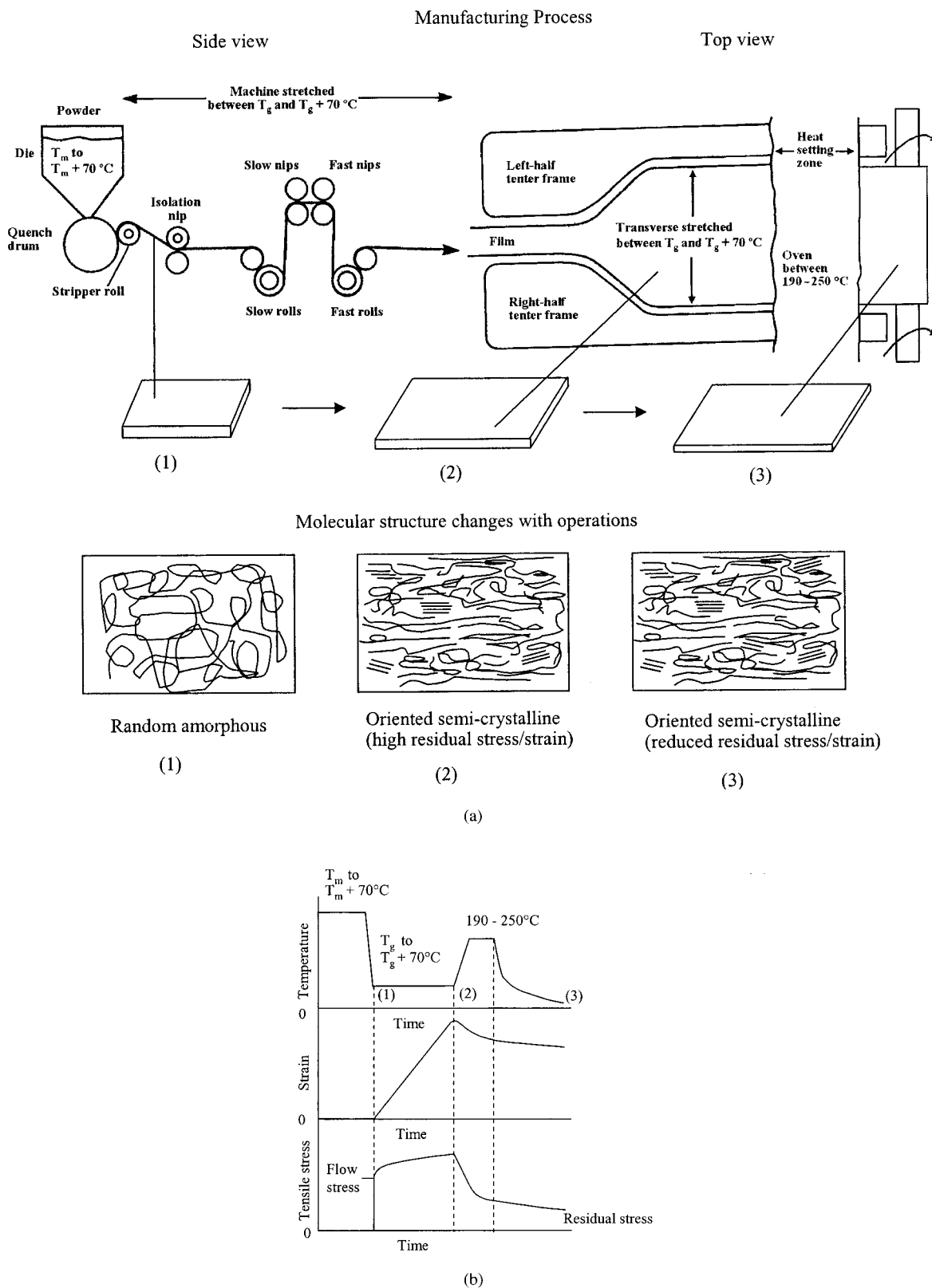


Figure 1 (a) Manufacturing process of polymeric film and the molecular structure changes with various operations,¹ and (b) temperature, strain, and stress variation during different stages of manufacturing.

vices with higher volumetric densities, especially for computer data-storage tapes. For high areal density, a substrate with high mechanical properties and high

dimensional stability under various environmental conditions is required. For high recording densities, reversible (thermal and hygroscopic expansions) and

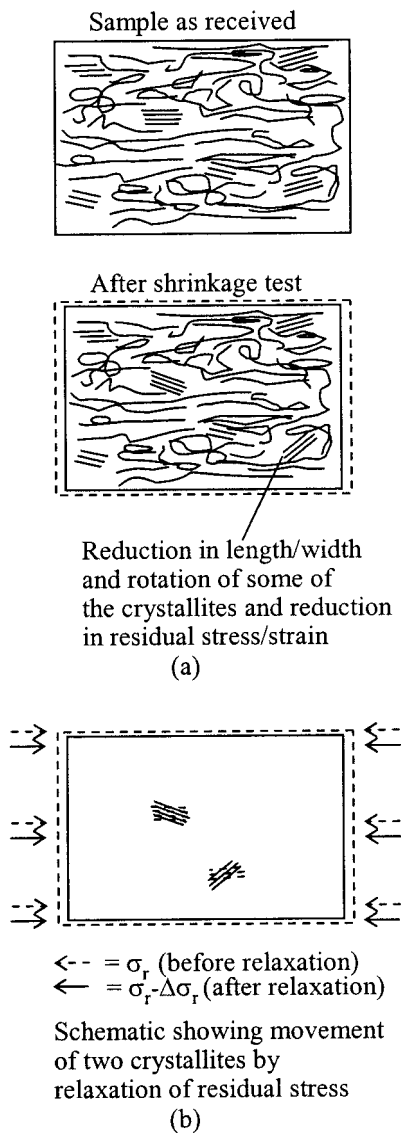


Figure 2 (a) Schematic showing changes in the structure during shrinkage test and (b) showing movement of two crystallites by relaxation of residual stress.

irreversible (creep and shrinkage) deformations of the substrates must be minimal during the storage on a reel and use in a drive.¹ The study of creep and shrinkage behavior is important for estimating the dimensional stability. To minimize stretching and damage during use, the substrates should be a high-modulus and high-strength material with low creep and shrinkage characteristics. If the substrates creep or shrink a lot, then long-term reliability problems are encountered, and thus it is required to minimize these deformations.^{1,3,4}

Bhushan and coworkers^{1,3,4} carried out long-term creep and shrinkage studies of various polymeric substrates. Bhushan¹ and Weick and Bhushan⁵ demonstrated a technique to obtain constitutive relationships by mathematical modeling of the experimental creep

data. The creep data was modeled using a maximum of three Kelvin-Voigt elements, and the creep compliance values and the time constants for the three elements were obtained. The assumption in the modeling was that the shrinkage of the film during creep was small and could be ignored. However, shrinkage occurs independently from the time a film is manufactured and it should have some contribution to the measured creep.

The objective of the research was to develop a mathematical model to characterize simultaneous creep and shrinkage behavior. Experiments were carefully designed to separate out creep and shrinkage effects and the model has been used to compensate for the effect of shrinkage on creep data and to get true creep data. Several PET, PEN, and ARAMID films were tested and viscoelastic parameters were obtained.

EXPERIMENTAL

Sample selection

A list of polymeric substrates studied during the experiment with their thicknesses and symbols are given in Table I.⁴ PET films include: Standard PET, Tensitized PET(1) and (2), Supertensitized PET, and Advanced PET. A 14- μm -thick Standard PET film is a typical substrate used for videotapes. Various 6.1- μm -thick tensitized type of PET films include: Tensitized PET(1) and (2) and Supertensitized PET, which are drawn to different degrees of tensilization in MD. PEN films have 6.2 μm thickness and include three kinds of films: Standard PEN, Tensitized PEN, and Supertensitized PEN, with different degrees of tensilization in MD. Standard PET and PEN films are used for long-play videotapes and tensitized films are used for computer-storage tapes. ARAMID film has 4.8 μm thickness and is used for magnetic tapes with thinner substrates. The glass-transition temperatures, based on differential scanning calorimetry measurements for PET, PEN, and ARAMID films are typically reported as 80, 120, and 280°C, respectively.³ Advanced PET film, an experimental film, has a higher glass-transi-

TABLE I
List of Magnetic Tape Substrates Used in This Study

Sample	Symbol	Thickness (μm)
Standard PET	Standard PET	14.0
Tensitized PET (1)	T-PET(1)	6.1
Tensitized PET (2)	T-PET(2)	6.1
Supertensitized PET	ST-PET	6.1
Advanced PET	Adv PET	6.0
Standard PEN	Standard PEN	6.2
Tensitized PEN	T-PEN	6.2
Supertensitized PEN	ST-PEN	6.2
ARAMID	ARAMID	4.8

tion temperature of about 115°C for higher thermal stability. The PET and PEN films are manufactured by biaxially drawing with a ratio of about four times in both machine direction (MD) and transverse direction (TD) during manufacturing.^{3,4} On the other hand, AR-AMID film is manufactured using a solution-casting process and then drawn slightly using a drawing process, which makes it more expensive than PET film ($\sim 3\times$); PEN film is also slightly more expensive than PET film ($\sim 1.3\times$).⁴ Further details on the substrates can be found in Ma and Bhushan.⁴

Experimental apparatus and procedure

The experimental apparatus and the test technique used to perform creep and shrinkage tests were previously described by Higashioji and Bhushan³ and Ma and Bhushan.⁴ The samples were tested for creep for 100 h, shrinkage for 100 h, and shrinkage for 100 h followed by creep for another 100 h. The environmental conditions used were 55°C and uncontrolled humidity (5–10% RH) and 55°C and controlled humidity (80% RH). An elevated temperature of 55°C was selected in this study because it is the upper limit of the operating envelope for tape drives.⁴

The creep tests were performed at a constant stress of 7 MPa, which is a typical stress applied to tapes in tape drives during use. It has been shown that creep deformations of the films used in this study, at this stress, remain in the linear viscoelastic regime.¹ Before applying this 7-MPa stress, the samples were conditioned at the test temperature without application of load for 1 h. Next a preload of 0.5 MPa was applied to the sample and conditioned for stabilization in the same test condition. During this stabilization period, typically of 2 h, the output signals from the apparatus were monitored until they were steady. Such a conditioning procedure has an effect on the creep behavior; for instance, the samples lose their long-term memory.^{1,3,5} The creep compliance $D(t)$ of the sample was obtained by dividing the time-dependent strain $\epsilon(t)$ obtained from the experiment with the constant applied stress σ .

$$D(t) = \frac{\epsilon(t)}{\sigma} \quad (1)$$

The shrinkage tests were performed at a minimal applied stress of 0.5 MPa, to hold the sample in place without causing any substantial creep in the sample.^{1,3,4} The samples were conditioned for 1 h in an environmental condition, without applying any load. Next a constant stress of 0.5 MPa was applied to the samples and the samples were allowed to shrink in the environmental condition. The shrinkage was obtained by measuring the decrease in the strain in the samples.

RESULTS AND DISCUSSION

Mathematical model

Creep compliance data for the polymeric films are generally modeled using a generalized Kelvin–Voigt model that has the following mathematical form:

$$D(t) = D_0 + \sum_{i=1}^n D_i [1 - \exp(-t/\tau_i)] \quad (2)$$

where $D(t)$ is the tensile creep compliance as a function of time, D_0 is the instantaneous compliance at $t = 0$, D_i represents the discrete compliance terms for each Kelvin–Voigt element, and τ_i represents the discrete compliance times for each Kelvin–Voigt element.

Based on this model, for a constant stress of magnitude σ applied at $t = 0$, the instantaneous response of a viscoelastic solid will be a sudden strain of magnitude $\epsilon_0 = \sigma D_0$. This is followed by a retarded (or delayed) response that can be attributed to the additional exponential terms in the equation. More specifically, each i th element of the model contributes to a delayed compliance of magnitude $D_i [1 - \exp(-t/\tau_i)]$, and the amount of this delay is directly related to the magnitude of the retardation time τ_i .^{1,5}

However, modeling the creep data using this technique does not take into account the effect of shrinkage that occurs simultaneously with the creep during a creep test. To accommodate the shrinkage effect, a new model has been proposed, which is shown in Figure 3. The model consists of creep and shrinkage units, both of which are Kelvin–Voigt elements. In the creep and shrinkage units, the spring is assumed to be a Hookean spring and the dashpot to be a Newtonian dashpot, which implies in the spring the stress is proportional to the strain and in the dashpot the stress is proportional to the rate of strain.⁶ It is also assumed that the contribution of each Kelvin–Voigt element is additive and independent of each other. This also implies that contributions of creep and shrinkage units are independent of each other and are additive. Thus in Figure 3, “+” is shown between the creep and shrinkage units.

The creep unit functions in the same way as discussed earlier in this section. During a shrinkage process the shrinkage unit is driven by a large change in compressive residual stress ($\Delta\sigma_r$), which was developed during manufacturing of the film. Because of shrinkage, the compressive residual stress relaxes and its magnitude decreases with time. The speed with which the system relaxes is also a function of temperature and humidity. An increase in temperature and humidity results in decrease of viscosity of the fluid in the dashpot unit and consequently the relaxation is more rapid. Continued shrinkage explains why some

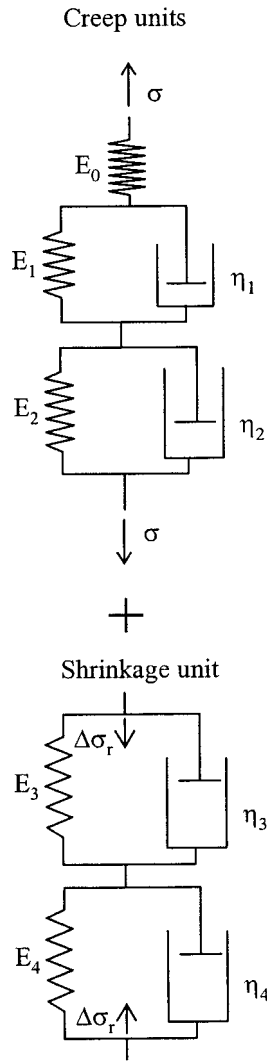


Figure 3 Creep and shrinkage models based on the Kelvin-Voigt model, combined for creep and shrinkage test.

substrates develop a negative slope during the creep test at elevated temperatures.^{3,4}

When a constant external stress (σ) is applied to this model, the creep unit will elongate, whereas the shrinkage unit will contract as a result of the change in residual stress. The net creep will therefore be given by addition of contribution of the two units. A general trend of creep and shrinkage curves and the effect of the addition of the two is shown schematically in Figure 4.

Curve fitting and data analysis

Based on the proposed model, the net strain of polymeric film is

$$\epsilon(t) = \epsilon_0 + \sum_{i=1}^n \epsilon_i [1 - \exp(-t/\tau_i)] - \sum_{i=n+1}^m \epsilon_i [1 - \exp(-t/\tau_i)] \quad (3)$$

where subscript $i \leq n$ corresponds to the creep unit and subscript $i > n$ corresponds to the shrinkage unit; and where $\epsilon(t)$ is the strain of the film as a function of time, ϵ_0 is the instantaneous strain at $t = 0$, ϵ_i represents the discrete strain terms for each Kelvin-Voigt element, τ_i represents the discrete times for each Kelvin-Voigt element, and $\epsilon_i = D_i \sigma$.

Experimental results were fitted to eq. (3) using a nonlinear least-squares technique known as the Levenberg-Marquardt method. This method finds the best-fit parameters for a given function by minimization of χ^2 by an iterative process.⁷ The initial strain immediately after loading (ϵ_0) was subtracted from all the data points of the experimental result curve. The resulting data were then used to find the viscoelastic parameters of the film. The trial values for the viscoelastic parameters (ϵ_i and τ_i) were estimated by observing the nature of slope of the curve to be fitted. The number of trial values is two for each Kelvin-Voigt element. The points at which there is a distinct change in the slope of creep/shrinkage curve and the point at which the slope nearly stabilizes were selected as an input to the program, which is based on the

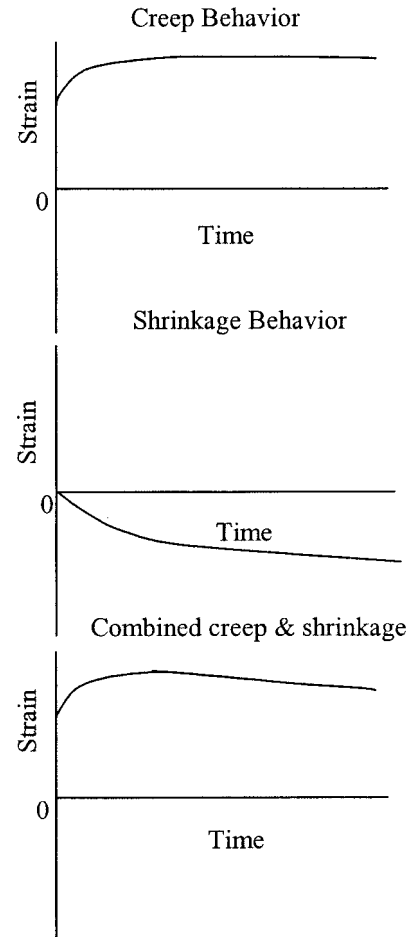


Figure 4 Schematics showing effect of simultaneous occurrence of creep and shrinkage.

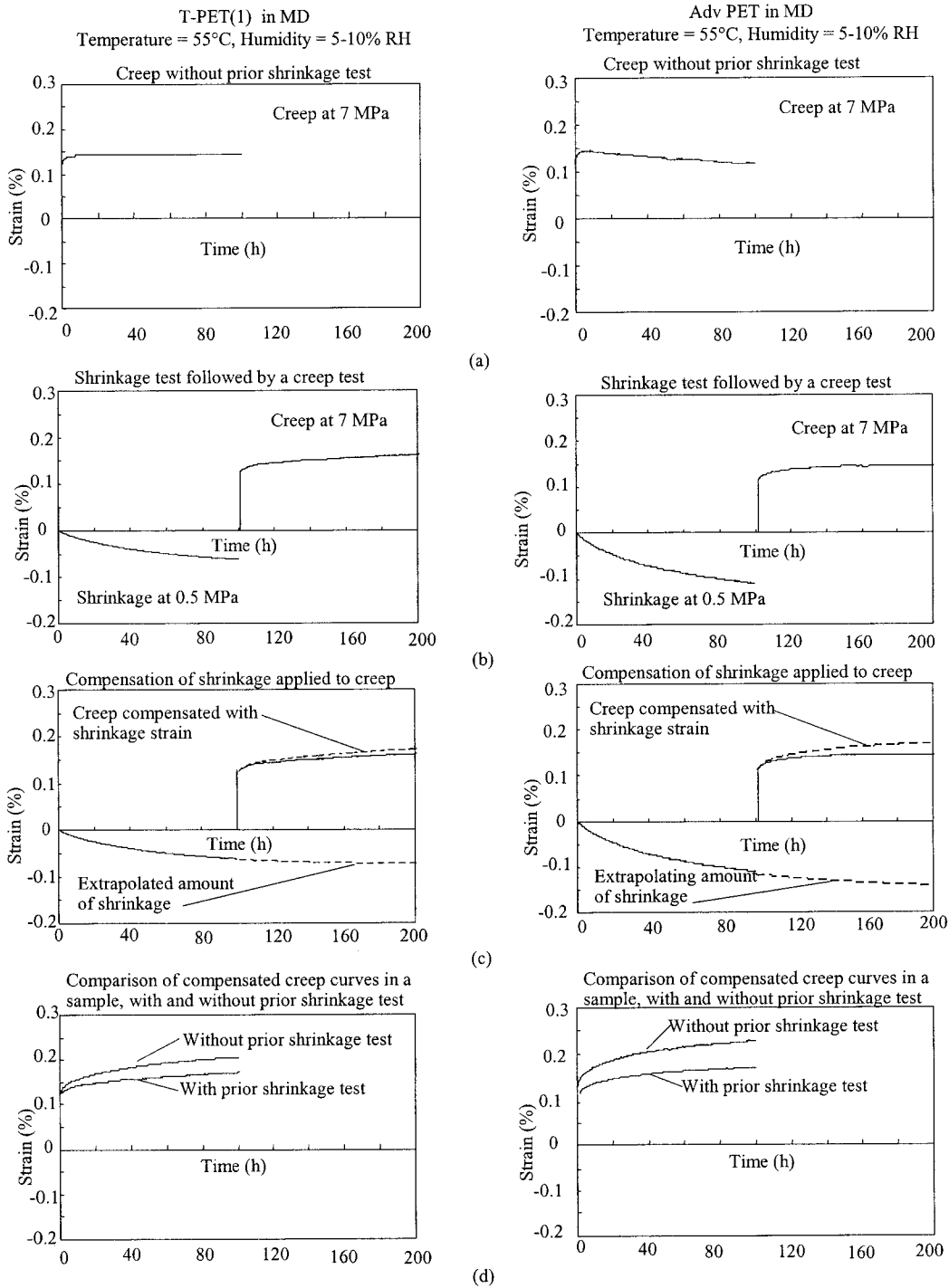


Figure 5 Long-term creep and shrinkage curves on PET samples to show simultaneous occurrence of creep and shrinkage and derivation of true creep data at 55°C and 5–10% RH (uncontrolled humidity).

Levenberg–Marquardt method. (For example, in the top left graph in Fig. 5, to be presented later, the points selected as the initial trial values are strains at 10 and 80 h.) An iterative process was carried out to obtain the converged values. The converged parameters obtained by this process were used to check the goodness of fit of the curve obtained from the model with the experimental curve before they were finally se-

lected. The values of parameters ϵ_i and τ_i were expected to be on the order of the range in the experimental data. If the fitted values were far off, new initial estimates were made and the iterative process was carried out again. The number of Kelvin–Voigt elements selected for creep and shrinkage units is two each, in the model presented in this study. If the fit was not believed to be good, the number of Kelvin–

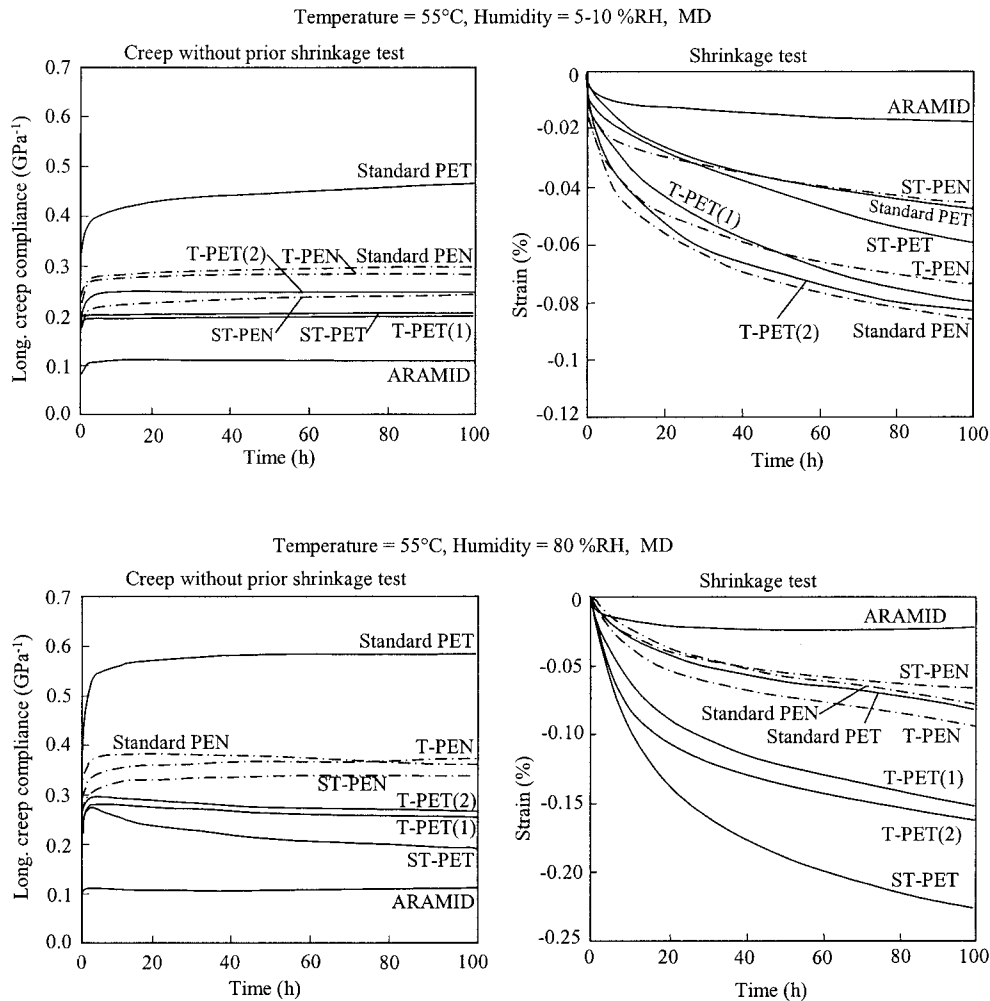


Figure 6 Long-term creep and shrinkage results for various polymeric films at 55°C and 5–10% RH (uncontrolled humidity) and 55°C and 80% RH controlled humidity.⁴

Voigt elements in the model would have been increased.

Experiments to separate creep and shrinkage effects

Higashioji and Bhushan³ and Ma and Bhushan⁴ reported that for some polymeric films, the slope of the creep curve became negative after some time during the creep test (as will be shown later in Fig. 6). Figure 5(a) shows representative creep data for two samples: Tensitized PET(1) and Advanced PET, which exhibit negative slopes to different extents at 55°C and 5–10% RH in MD. Because the slope of a true creep curve should always be positive, a negative slope implies that the sample was undergoing a stress-relaxation process or a shrinkage process. To verify this, a shrinkage test followed by a creep test, each for 100 h, were performed on both samples. The purpose was, if the substrates were allowed to shrink for 100 h, the substrates might become completely relieved of residual

stresses, and then if creep test was performed on such samples, it would give a measure of true creep. The test results in Figure 5(b) show a positive slope of creep curve after the “shrinkage followed by a creep test.” Because of the shrinkage test, a large decrease in the residual stress caused a large amount of shrinkage in the first 100 h and during the next 100 h, because of the presence of only a small magnitude of leftover residual stress, the films shrank only slightly. Thus the test result showed an increase in creep and positive slope in both films. It can be inferred that a completely stress-relieved film or a film that has undergone a shrinkage test for a sufficiently long time, if used for a creep test, will give a measure of true creep.

Because the slope of the shrinkage curve did not stabilize even toward the end of 100 h of the experiment; the film was not completely relieved of the residual stress. To compensate for the shrinkage that the film will undergo during the creep test, the shrinkage data were mathematically modeled. The shrink-

TABLE II
Parameters Obtained from Creep Without Prior Shrinkage Test Data^a

Substrate	D_0 (GPa ⁻¹)	D_1 (GPa ⁻¹)	τ_1 (h)	D_2 (GPa ⁻¹)	τ_2 (h)	ε_3 (%)	τ_3 (h)	ε_4 (%)	τ_4 (h)
A. Temperature = 55°C, humidity = 5–10%RH, MD									
Standard PET	0.300	0.0941	0.9	0.102	31.4	-0.00327	28.7	-0.0185	30.8
T-PET(1)	0.171	0.0216	0.3	0.0269	99.7	-0.00375	64.5	-0.00683	114.1
T-PET(2)	0.186	0.0206	0.3	0.0494	2.1	-0.00124	1.8	-0.00301	3.8
ST-PET	0.186	0.0153	0.3	0.0163	57.5	-0.00370	32.3	-0.00449	118.7
Standard PEN	0.229	0.0480	1.1	0.0283	47.1	-0.00011	1.3	-0.00302	57.4
T-PEN	0.214	0.0564	1.1	0.0527	59.7	-0.0228	208.8	-0.0296	187.5
ST-PEN	0.171	0.0544	1.2	0.0351	41.2	-0.00686	1.9	-0.00482	44.0
ARAMID	0.0714	0.0105	8.5	0.0337	1.2	-0.00351	20.2	-0.00160	23.2
B. Temperature = 55°C, humidity = 80%RH, MD									
Standard PET	0.429	0.125	1.4	0.101	31.1	-0.0144	44.0	-0.0395	47.3
T-PET(1)	0.243	0.0393	1.1	0.0360	18.9	-0.0151	29.2	-0.0310	29.4
T-PET(2)	0.257	0.0226	0.4	0.0230	2.1	-0.00801	34.8	-0.0180	34.8
ST-PET	0.229	0.107	1.5	0.0264	45.0	-0.0510	3.2	-0.0740	44.5
Standard PEN	0.329	0.0493	1.5	0.0826	36.2	-0.0438	75.0	-0.0481	75.1
T-PEN	0.286	0.0590	1.5	0.102	23.2	-0.0260	23.3	-0.0274	22.7
ST-PEN	0.257	0.0461	0.8	0.0591	12.0	-0.00663	12.1	-0.0106	11.9
ARAMID	0.100	0.0156	0.8	0.0453	386.6	-0.00450	6.7	-0.00528	20.1

^a Data modeled using 2 creep and 2 shrinkage units:1 unit = 1 Kelvin-Voigt element.

age data from the “shrinkage test followed by a creep test” were modeled using a two Kelvin-Voigt element shrinkage model and ε_i and τ_i values were obtained for the shrinkage unit. Using ε_i and τ_i values of the shrinkage unit, the amount of shrinkage the substrate would undergo in the next 100 h was extrapolated using the shrinkage model and this amount was added to the creep data in the next 100 h, to get a “creep compensated with shrinkage strain,” as shown in Figure 5(c). This approach can be used to compensate the creep obtained without a prior shrinkage test

because the shrinkage initially occurs at a fast rate and after some time (close to 100 h) it nearly stabilizes. Thus the difference between the actual shrinkage in the substrate (after 100 h) and the extrapolated shrinkage will be considerably less and the compensation factor will not have a significant amount of error in it. The creep compensated with shrinkage strain gives a measure of true creep that a polymeric film will undergo.

During the creep test, shrinkage also occurs but occurs at the load used in the creep test. If shrinkage

TABLE III
Parameters Obtained from Shrinkage Test Data^a

Substrate	ε_3 (%)	τ_3 (h)	ε_4 (%)	τ_4 (h)
A. Temperature = 55°C, humidity = 5–10%RH, MD				
Standard PET	-0.0134	4.0	-0.0395	52.0
T-PET(1)	-0.0165	0.7	-0.0704	44.2
T-PET(2)	-0.0232	1.5	-0.0615	32.3
ST-PET	-0.0133	0.7	-0.0688	89.9
Standard PEN	-0.0308	1.8	-0.0575	36.0
T-PEN	-0.0259	0.9	-0.0487	33.0
ST-PEN	-0.0193	0.7	-0.0308	54.2
ARAMID	-0.00899	1.0	-0.00942	47.8
B. Temperature = 55°C, humidity = 80%RH, MD				
Standard PET	-0.0301	8.2	-0.0673	83.1
T-PET(1)	-0.0682	8.3	-0.113	76.4
T-PET(2)	-0.0889	7.3	-0.102	79.0
ST-PET	-0.104	8.8	-0.154	61.9
Standard PEN	-0.0359	13.7	-0.107	217.9
T-PEN	-0.0480	9.7	-0.0917	158.7
ST-PEN	-0.0298	8.4	-0.0475	65.4
ARAMID	-0.0072	0.6	-0.0170	10.2

^a Data modeled using 2 shrinkage units.

data taken at 0.5 MPa is to be subtracted, true compensation of shrinkage does not occur if shrinkage is load dependent. To study the effect of external load on shrinkage, a comparative study was carried out. Shrinkage data from the shrinkage test at 0.5 MPa were added to the "creep without prior shrinkage test" data obtained at 7 MPa. Because conditioning times for creep (3 h) and shrinkage (1 h) were different, shrinkage data were discarded for the first 2 h. The compensated creep curve obtained by this method was compared with the compensated creep curve obtained by the method discussed in the previous paragraph in which shrinkage occurs in a sample, which is assumed to be relaxed. Given that a relaxed sample does not shrink much, the effect of external load during the creep test on shrinkage will be substantially less. The results are shown in Figure 5(d). The two comparison curves are represented by "without prior shrinkage test" and "with prior shrinkage test" curves. The two curves show similar trends but do not overlap, so it is believed that the external load in the case of without prior shrinkage test has an attenuating effect on shrinkage. The attenuation is believed to be attributable to the presence of entanglement in chains of macromolecules of the polymeric substrate. Because of the entanglements in the macromolecule chains, under external loading some of the macromolecule chains tend to uncoil and straighten; and as a result some of the neighboring coils will also be uncoiled and straightened. Because of the large chain length of macromolecules, the shrinkage still continues independently in the film. However, because of the stretching of chains the magnitude of shrinkage is now reduced. Thus if shrinkage (at 0.5 MPa) is added to the creep (at 7 MPa) without a prior shrinkage test, without taking into account this attenuation effect, the amount of creep obtained is higher.

True creep in a polymeric sample can be obtained by first performing a shrinkage test at the lowest possible load (~ 0.5 MPa) for long enough hours so that the residual stresses are essentially relieved and then performing a creep test on this completely relaxed sample. The process of addition of shrinkage at 0.5 MPa to creep at 7 MPa does not give true creep that the film will undergo at 7 MPa. However, this does provide a reasonable comparison of the creep behavior of various samples. The results of this study follow.

Analysis of creep and shrinkage data

The creep and shrinkage at different environmental conditions are shown in Figure 6.⁴ During the test, creep in the film increases with temperature and humidity. The humidity has a plasticizing effect on the films, which enables the macromolecule chains of the polymer to slip past one another, thus resulting in the increased creep. A similar plasticizing behavior was

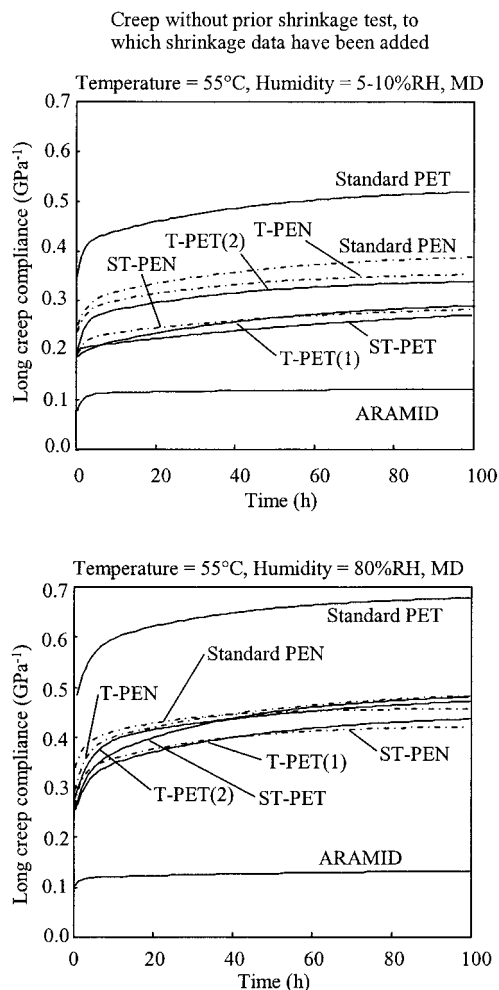


Figure 7 Creep curves without prior shrinkage test curves, with shrinkage data added.

observed in the shrinkage tests under the same environmental conditions. The tendency of the films to shrink increased with the increase in temperature and humidity. A discussion on comparison of creep and shrinkage behavior of each film shown in Figure 6 is given in Ma and Bhushan.⁴

The "creep without prior shrinkage test" data were modeled using the model shown in Figure 3. The shrinkage data were modeled using a two Kelvin-Voigt element shrinkage model. The viscoelastic parameters obtained by modeling the creep without prior shrinkage test data and shrinkage data are shown in Tables II and III, respectively. As mentioned in the previous section, that compensating the creep data at 7 MPa with shrinkage data at 0.5 MPa gives a reasonable comparison of the creep behavior of various samples, the creep data compensated with shrinkage at 0.5 MPa have also been mathematically modeled. The compensated creep curves are shown in Figure 7. Given that conditioning times for creep (3 h) and shrinkage (1 h) were different, shrinkage data

TABLE IV
Parameters Obtained from Creep Without Prior Shrinkage Test Data, to Which Shrinkage Data Had Been Added^a

Substrate	D_0 (GPa ⁻¹)	D_1 (GPa ⁻¹)	τ_1 (h)	D_2 (GPa ⁻¹)	τ_2 (h)
A. Temperature = 55°C, humidity = 5–10%RH, MD					
Standard PET	0.24	0.0510	1.5	0.0800	42.3
T-PET(1)	0.13	0.0104	2.4	0.0732	50.2
T-PET(2)	0.13	0.0537	1.6	0.0580	37.8
ST-PET	0.13	0.0129	0.4	0.0729	94.5
Standard PEN	0.17	0.0398	2.4	0.0696	45.4
T-PEN	0.16	0.0381	1.9	0.0531	36.9
ST-PEN	0.13	0.0276	1.7	0.0495	57.2
ARAMID	0.05	0.0289	1.5	0.00708	41.9
B. Temperature = 55°C, humidity = 80%RH, MD					
Standard PET	0.32	0.0829	3.1	0.0772	37.0
T-PET(1)	0.17	0.0564	2.9	0.0872	42.5
T-PET(2)	0.19	0.0812	3.9	0.0759	66.4
ST-PET	0.17	0.0634	2.7	0.1100	37.1
Standard PEN	0.23	0.0397	1.8	0.0500	22.1
T-PEN	0.20	0.0660	2.6	0.0823	48.1
ST-PEN	0.18	0.0513	1.7	0.0652	29.6
ARAMID	0.07	0.0130	1.1	0.0121	60.8

^a Data modeled using 2 creep units.

were discarded for the first 2 h. The viscoelastic parameters for the compensated creep curves are shown in Table IV.

Tables II, III, and IV contain parameters D , ε , and τ , which were obtained after modeling. Creep parameters in Tables II and IV and shrinkage parameters in Tables II and III are somewhat comparable, as expected. The creep compliance (D) values of Kelvin-Voigt elements correspond to the creep unit; strain (ε) values of Kelvin-Voigt elements correspond to the shrinkage unit. The time constants (τ) in a similar manner represent the time constant for the respective units. The D_0 value is the creep compliance at time $t = 0$ and is obtained because of the elastic property of the film and D_1 , D_2 , $\varepsilon_{3'}$ and ε_4 correspond to the viscoelastic properties. The time constant values indicate when the elements would reach 63.2% of their final value. The higher time constant value implies that the element will reach a steady state after a lengthy time. In addition to this it can also be deduced that contribution of the element with higher time constant will be negligible during the initial phase of experiment.

CONCLUSIONS

The viscoelastic property of various films was studied through the results of creep and shrinkage experiments. Creep and shrinkage were observed to occur simultaneously during the creep experiments and some of the Tensilized PETs and an Advanced PET at 55°C showed a tendency to shrink more than they creep in a creep test. An experiment—"shrinkage test

followed by a creep test"—was designed to prove that shrinkage and creep occur simultaneously and independently of each other and the overall behavior of the creep is a result of the creep and shrinkage that occur in the film. The test results on Tensilized PET(1) and Advanced PET at 55°C and uncontrolled humidity (5–10% RH) showed that after the experiment, the creep curves showed a positive slope against the negative slope, which was obtained when the sample was tested for creep without a prior shrinkage test on the sample.

A mathematical model based on the Kelvin-Voigt model has been proposed to explain the phenomenon of simultaneous creep and shrinkage. The proposed model incorporates a shrinkage unit subjected to a large change in residual stress to take into account the contribution of shrinkage. Based on the model the experimental data were curve fitted using the Levenberg-Marquardt method of nonlinear least-squares fitting to obtain viscoelastic parameters for creep and shrinkage data. A technique has been shown to calculate the true creep in a film, with the use of experiment and the proposed model.

Comparison of compensated creep curves obtained after compensation of shrinkage to "creep without prior shrinkage test" data and "shrinkage test followed by a creep test" data showed attenuation of shrinkage with the external load. The films shrank more at 0.5 MPa compared to that during any creep test. An explanation to this behavior is given in terms of ease relaxation in entangled macromolecular chains in the films under external load.

Creep and shrinkage curves at various environmental conditions were presented. Both creep and shrinkage experienced a plasticizing effect at high humidity and temperature. A compensated creep curve was obtained by addition of shrinkage at 0.5 MPa to the creep. Viscoelastic parameters to study the constitutive relationship of creep and shrinkage obtained by modeling different creep and shrinkage data have been tabulated.

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