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The Role of Stress Concentrations in Accelerated Creep and Sorption-Induced Physical Aging

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### The Role of Stress Concentrations in Accelerated Creep and Sorption-Induced Physical Aging

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Accelerated creep in cyclic humidity environments is a curious phenomenon that has been established for nearly 40 years but has no widely accepted explanation. The purpose of this paper is to promote a sorption-induced stress concentration mechanism. We show that materials creep more under a cyclic load than at a constant average load and that cyclic sorption creates localized load cycling. We argue that this is sufficient to account for the observed degrees of accelerated creep. We interpret sorption-induced physical aging as a similar phenomenon that is the result of creep sensitivity to load and of residual stresses that are built up during sorption. Finally, we demonstrate that there is a type of work hardening resulting from the rapid relaxation of residual stresses when a material is loaded.

### THE ACCELERATED CREEP PHENOMENON

Creep is the time dependent deformation of a sample held under a constant load. It is well established that paper exhibits much higher rates of creep at higher moisture levels [1,2]. Nevertheless, paper creeps faster when loaded in a cyclic humidity environment than it would at the high moisture content extreme of the cycling. Figure 1 documents a typical experiment conducted in our laboratory. A tensile load equal to 25% of 50% RH breaking strength was applied to a paper sample conditioned at 80% RH. After three hours, the humidity was cycled between 80 and 30% RH at one hour intervals. The creep strain curve shown is the average of three runs. Along with the variable humidity creep history, we present the average of two creep responses of similar samples subjected to the same load, but maintained at 80% RH.

In Figure 1, we have plotted the creep strain versus the logarithm of the time lapse since application of the load. We did this because paper, like many other materials, exhibits a creep regime over which creep strain is roughly linear with the logarithm of time for many orders of magnitude of time [1,2]. Thus, a large portion of the creep response can be characterized by a single creep rate parameter (the log time strain rate). Notice that, after cycling begins, a straight line can be linked between points at the same phase in the cycling. In this way, the cycling creep

can be expressed as another constant creep rate in logarithm of time. We adopt from Wang et al. [3] the following definition: accelerated creep ratio is the log time creep rate under cyclic humidity divided by the log. time creep rate at high humidity if the sample were left in the high humidity state. Drawing a line tangent to the top of the hydrated portion of the cyclic humidity curves and comparing its slope to 80% RH curve just before cycling starts yields an accelerated of about 5.4 for this paper sample. If, as in this case, the accelerated creep ratio is greater than one, we say that the material undergoes accelerated creep. Clearly, the magnitude of accelerated creep, so defined, will depend on the cycling parameters. However, in our work, it appears to be relatively insensitive to creep load; for this sample, the accelerated creep ratio was 6.6 at 15% of breaking load and 7.5 at 35% of breaking load.

Accelerated creep is a curious and interesting phenomenon. Most people are surprised when confronted with materials that strain much more during cycling than in their most compliant state. Accelerated creep also has unfortunate practical ramifications; paper structural elements creep much more and fail much sooner in an inconstant environment than when damp but stable [4]. Many paperboard products are overbuilt as a precaution. Obviously, we need to understand the causes of accelerated creep so that we can intelligently devise strategies to blunt its consequences.

### **EXPERIMENTAL PROCEDURES**

The measurements reported in Figure 1, as well as all the subsequent sheet measurements, were conducted in a tensile creep tester that we had constructed for this work. It is an environmental chamber with tensile mounting stations for five specimens. The chamber has input ports for moist and dry air. A sensor monitors the humidity inside the chamber and reports to a computer, which controls valves on the air ports in order to reproduce a prescribed relative humidity regime. All of the results reported were conducted on 60  $g/m^2$  handsheets formed from a 86 CSF, thermomechanical pulp (TMP). The 50% RH tensile strength was 7,400 N/m. Specimens are preconditioned at 90% RH for 72 hours, then subjected to 20% RH for 24 hours, and stored at 50% RH. A sample is cut and placed in an alignment jig. The jig also supports clamps that sandwich the specimen at both ends. Epoxy is applied between the sample ends and the clamp faces. After the epoxy cures, the sample-clamp assembly is removed from the jig. The sample is 2.5 cm wide and has a 14.0 cm free run between clamps. The glass front face of the environmental chamber is removed, and one clamp fixture is inserted into a slot at the top on the chamber. The sample and clamp assembly are now hanging from a mounting bar. There is a hole in the bottom clamp. We span the bottom clamp with a yoke. A peg is inserted through holes in the yoke arms and the clamp hole suspending the yoke from the clamp. A rod projects from the bottom of the clamp. The rod extends out of the chamber through a small hole in the bottom. The rod passes through an LVDT (linear variable differential transformer) coil that is mounted below the environmental chamber. An LVDT core forms a section of the rod positioned inside the LVDT coil. A hook hangs from the end of the rod below the LVDT. Weights can be attached to the hook to attain the desired tensile load. A nut, which is threaded onto the rod just above the hook, rests on a holding bar that supports the total dead weight load until the experiment begins.

We use this tensile tester for three types of experiments. For constant load, constant humidity experiments the samples are sealed in the chamber by attaching the glass front face. Humidity control begins, and the sample is preconditioned in an unloaded state at the test value for at least 16 hours. The test commences when the holding bar is pneumatically lowered. The computer monitors the LVDT output and calculates the creep strain history over the period of the experiment. For cyclic humidity tests, the computer is programmed to change the chamber RH at fixed intervals. The unloaded sample is preconditioned, as before, at the initial RH setting. We perform constant humidity, variable load experiments by manually changing the weights hanging from the hooks at regular time intervals.

We also report single-fiber accelerated creep measurements on Kevlar fibers. We used a single fiber apparatus developed by Wang and Dillard that is well documented in the literature [5]. Briefly, a vertical glass tube with wet and dry air ports acts as an environmental chamber. A test fiber is suspended down the center of the tube. Its bottom end is epoxied to the core of an LVDT. The core passes through a small hole in the bottom of the chamber into an externally mounted LVDT coil. Small extra weights can be suspended from the core. A computer controls the air ports and monitors the LVDT, the chamber temperature, and relative humidity.

### ACCELERATED CREEP HISTORY

Accelerated creep work on wood products began in the early 1960's when Armstrong, Kingston, and Christensen [6-8] reported that wooden beams experience excess bending creep under cyclic humidity conditions. This was the first recognition that cellulosic materials display additional compliance as a result of sorption. Experimenters, such as Pickett [9], previously made similar observations on concrete structures. About ten years later, accelerated creep was demonstrated by Byrd [10-12] to be important in paper. Subsequently, many others [4,13-19] have contributed to the verification and extension of paper experimental observations. The surprising experimental results precipitated a flurry of theoretical conjecture. However, today, after nearly forty years of squabbling, there is no generally accepted mechanism.

In the 1960's, accelerated creep in wood was widely attributed to a previously unknown coupling of moisture transport and mechanical compliance [20-22]. Accelerated creep attained the status of a totally new phenomenon requiring its own special explanation. In various ways, it was argued that moisture diffusion directly causes a decrease in compliance. It was sometimes explained that transport of moisture through the structure induces extra bond breakage which leads to extra creep. This explanation took a major hit in 1972 when Armstrong [23] published creep experiments on hollow wood cylinders. He demonstrated that there was no accelerated creep when a constant moisture gradient was established from the inside to the outside of a hollow cylinder. Much later, Bazant [24] made a clever distinction between "micropore" and "macropore" diffusion that saved the transport concept from total destruction. However, at the time, the theorists moved on.

Led by Ranta Maunus [25], the next generation of investigators argued that changing moisture content drives accelerated creep in wood. Instead of postulating a link between moisture transport and compliance, they claim that the increase in creep rate is a function of the

time rate of change of moisture content. The consequences of Armstrong's experiments are dodged by invoking a different, new physical phenomenon. This perspective continues to be in the mainstream of wood research: accelerated creep is handled as a consequence of a decrease in compliance triggered directly by a change in moisture content [26-28]. Specialized, cell wall mechanisms are sometimes proposed to account for the putative sorption rate dependence of creep [29,30].

Padanyi [31,32] has done some very provocative thinking concerning the influence of moisture history on the mechanical properties of paper. He made a connection to "physical aging", an established phenomenon in the study of polymer rheology. In its standard form, physical aging of polymers (Struik [33]) refers to the progression of an amorphous polymer to equilibrium after it goes below its glass transition temperature. Above the glass transition temperature, an amorphous polymer is in a relatively high entropy state with considerable "free volume" and with appreciable polymer chain backbone mobility. Its compliance is high. It is argued that there is not an immediate passage to a new equilibrium configuration when the polymer temperature falls below its glass transition. Thermodynamic state change requires polymer backbone motion, and this requires free volume. At first, thanks to the residual free volume, transition progresses rapidly. However this is a "self-retarding" process. The rate of the process greatly depends on the free volume, and the free volume is steadily decreasing. The low-temperature equilibrium state is approached gradually, and the polymer continually loses compliance. This slow progression to glassy equilibrium is called physical aging. It differs from other polymer processes (such as chemical aging) in that the original state is recoverable by cycling back above the glass transition temperature.

Padanyi's insight was that aging could be triggered by moisture changes as well as temperature changes. At 50% RH, the glass transition temperature of amorphous cellulose is above room temperature. However, it falls as moisture content increases. Padanyi points out that, according to Salmen and Back [34], glass transition reaches room temperature at about 11-13% moisture content. Thus, quickly passing from high to low moisture content should produce a non-equilibrium state with more free volume than that of the slowly approached equilibrium state. The material after desorption would be more compliant than at equilibrium. As it aged, its compliance would decrease. Padanyi did the standard aging creep tests after dropping moisture content rather than temperature. The experimental results were remarkably similar to aging curves: creep compliance decreased with age, master creep curves could be built in the normal way, and the slope of the shift in log time versus the log age was nearly one. This is very interesting, but not terribly startling. The real surprise is the desorption treatment. Padanyi made creep tests at 50% RH at variable times after exposing the samples to low humidity. Although the shift in creep curves was slightly less when "deaged" dry, the creep compliance curves were again very similar to thermal aging curves. This cannot be rationalized by appeal to a glass transition explanation. Nevertheless, although standing on less solid ground, one can still make the same general argument. The amorphous polymer will have different thermodynamic states in dry and 50% RH conditions. It will not immediately reach equilibrium after transition from dry to its 50% RH moisture content, and the nonequilibrium state may be more compliant. Buttressed by his aging experiments, Padanyi proposes that accelerated creep is a manifestation of sorption-induced physical aging. During moisture cycling amorphous portions of the amorphous polymer structure are continually being "deaged" and are thus more compliant at every juncture than at equilibrium. This is a persuasive argument, and it has given us considerable pause.

In the study of paper accelerated creep, there are adherents to the Ranta Maunus [25] and to the Padanyi "aging" explanations [35]; however, many more workers make appeal to paper-specific, fiber-level mechanisms. As examples, Soremark and Fellers [17,18] present a "physico-mechanical" model that employs moisture induced stress redistribution to generate extra dislocations, whereas Haslach [15] offers a long explanation that concentrates on anisotropic swelling and the motion at the interfiber bonds. Caulfield [36], coming from a physical chemistry point of view, contends that a fundamental connection between stress-strain hysteresis and sorption hysteresis allows the two processes to interfere and produce accelerated creep.

Accelerated creep is observed in a variety of materials and structures. It has been reported in wood [6], concrete [9], paper [10], polyurethane foams [37], Kevlar composites [3] and Kevlar fibers [3]. Although concluded otherwise by Haslach [15], we would like to add regenerated cellulose films to the list. Figure 2 documents accelerated creep experiments on cellophane strips in tension. The sample was a 24 µm thick unplasticized P00 cellophane film from UCB Films. It was moisture preconditioned in the manner of the paper samples of Figure 1. We applied a load in the machine direction equal to 7.5% of its 50% RH tensile strength (436 N/m). After 3 hours of creep at 80% RH, the samples were cycled in hour intervals from 80 to 30% RH. The wet and cyclic curves of Figure 2 are the averages of two runs. Even though cellophane experiences more hygroexpansion than paper, its accelerated creep ratio (2.6) was less than half that of the TMP paper samples. Unlike the paper sample, it displays creep recovery in the dry state, and it lacks the cusp at the bottom of the first dry step. Cellophane creep appears to be less susceptible to creep amplification by changing environment, but it clearly does undergo accelerated creep. The contrary report of the earlier work [15] is likely due to an experimental detail. Our environmental chamber takes only 1 minute to transverse wet to dry and 5 minutes to go dry to wet, whereas Haslach changed humidity at a much slower rate. We will discuss the importance of cycle transition time and duration later.

Accelerated creep seems to be a general response of hydrophilic materials of macroscopic size. However, with the exception of aramid fibers (Kevlar in particular) [3,38] and perhaps thick cellulose acetate butyrate fibers [38], single fiber work yields negative results. Accelerated creep is not observed in wood pulp fibers [39,40], Nylon 6,6 fibers [3,41], poly(methylmethacrylate) fibers [38], or Rayon fibers [41].

### ACCELERATED CREEP MECHANISM

Our selection of an accelerated creep model is restricted by basic considerations. First, we do not want to postulate new physical law. We contend that everyday explanations should be exhausted before we elevate accelerated creep to the status of an independent phenomenon. Also, we prefer a single picture that encompasses all occurrences. Therefore, we avoid explanations that rely on material-specific mechanisms. One of these might address some

occurrences, but a group of special stories would be needed for a full explanation. Below is our proposal for a general theory resting on established physics.

We assert that accelerated creep is a direct result of sorption-induced swelling combined with a common property of the constitutive creep behavior. In short, cyclic humidity changes can cause localized cyclic loading, and many materials creep more under a cycling load than at a constant, average load. One of the ways that cyclic sorption induces cyclic stress can be appreciated with the help of Figure 3. There, hypothetical moisture and stress profiles through a single sheet under tensile load are represented at a series of times. At time A, the sample is uniformly damp and the stress is evenly distributed across the sheet. At time B, the surrounding humidity has been reduced, and the sample is drying from the outside to the inside. The material is less swollen at the surface. Therefore, to keep more or less uniform strain through the sample, the tensile stress will be above average on the outside and below average on the inside, somewhat as depicted in the time B sequence. After the sample has been dry for a sufficient period (time C), the stresses equilibrate. However, during rewetting (time D) the moisture and stress profiles are reestablished, but their trends are the reverse of those experienced at time B. Thus, moisture cycling causes stresses in the two regions to cycle, out of phase, above and below the average value. Of course, in actuality the situation will be more complex. Nevertheless, extra creep will occur at the high stress locations causing the stress to relax in those regions and tending to even out the stress profile. Properly timed moisture cycling will continually interrupt the equilibrating processes and perpetuate load cycling throughout the structure. We call accelerated creep resulting from this kind of stress cycling "moisture-gradient-driven" accelerated creep. So far, the moisture gradient has been specified to be through the sheet. In the case of paper, moisture content during sorption may also differ between the central and outer portions of each fiber. This would produce fiber-level moisture gradients and load cycling. Whatever the details of the process, moisture gradients must exist during sorption, and cyclic moisture gradients will lead to local cyclic stress histories.

There is a second mechanism for sorption-induced load cycling. If the material response to moisture is heterogeneous, load cycling can follow from moisture cycling, even if the material sorbs instantly and there are no moisture gradients. Suppose that some fibers swell more than others, or that fibers swell more in the radial direction than in the axial direction, or that the sheet swells differently in different layers. When the environment cycles dry, the tensile load will be concentrated in the more moisture-sensitive load-bearing elements. These stress concentrations will relax out, until the environment goes damp. Then, the less hygroexpansive elements get the high stresses. Again, but in a different way, moisture cycling leads to localized load cycling. We call this "heterogeneity-driven" accelerated creep.

We claim that it is a common feature of the creep constitutive relation of many materials that total creep is greater under cycling load than at the constant average load. We will discuss this in more detail later. For now, we merely point out that this material property combined with the cyclic loading that accompanies humidity cycling will produce extra creep. Accelerated creep (as defined above) will arise if this effect overwhelms the loss in creep due to the reduction in creep rate at lower average moisture content.

We are not the first to single out creep compliance and swelling. To underscore the antiquity of the idea, we quote a 1942 passage from Pickett [9]: "an increase in creep accompanying non-uniform shrinkage or swelling is a natural consequence of the fact that the sustained-stress-vs.-strain curve of <u>concrete</u> is not linear". In the wood and paper literature, this is a neglected explanation, but we did find one modern paper (Selway and Kirkpatrick [42]) conveying like sentiments: "Changes in moisture can cause rapid transient increases in stress and as the creep rate is a highly non-linear function of stress there can be significant increases in creep rate". To these authors, the term, non-linear creep, implies that the constant load creep response increases more than proportionately with the creep load. There is a distinction between this and our requirement for the creep constitutive behavior. This will become apparent later, when we introduce a material that does not creep disproportionately at higher loads, but does creep more when the load is cycled, and does experience accelerated creep. Nevertheless, the two explanations are very similar. Thus, our contribution is to expand and refine the approach. to address known and new experimental results, and to promote the explanation. Also, there are parallels between the mechanism of Soremark and Fellers [17, 18] and what we call heterogeneity-driven accelerated creep. Both appeal to heterogeneous hygroexpansion and the resulting stress concentrations, but their third leg is extra dislocations, whereas ours is the natural stress dependence of creep compliance.

Be aware that there are predictive as well as theoretical distinctions between this picture and the moisture transport and moisture rate change mechanisms of the wood scientists. For us, increased creep is not an inevitable response to moisture changes or to moisture transport. These will not lead to accelerated creep unless they cause cyclic stress concentrations, and there is the right kind of creep behavior. A swelling material with extra compliance in cyclic loading is susceptible to accelerated creep. However, these are necessary but not sufficient conditions. For accelerated creep to actually happen, we contend the moisture cycling must result in stress gradients. Also, the cyclic sorption processes must result in stress gradients in the material for a significant portion of the cycle time. In the case of moisture-gradient-driven accelerated creep, this puts constraints on the ratios of sorption time to cycle time and sorption time to relative humidity ramp time. If the ramp time is not short compared to sorption time, no large stress gradients will arise. Also, stress concentrations will endure for a significant portion of the creep time only if the cycle time is not too long or too short compared to the sorption time. Optimum conditions for accelerated creep occur when the environment is switched rapidly and sorption time is the order of cycle time. Moisture gradients are not necessary to produce stress gradients under the heterogeneity-driven accelerated creep scenario, therefore, the dependence on sorption time is not so great. However, cycle time cannot be much shorter than sorption time, and cycle time cannot be long, compared to the time for stress relaxation.

### VARIABLE LOAD CREEP CONSTITUTIVE EQUATION

Since we hold that accelerated creep is a manifestation of the extra creep experienced under varying load, we must demonstrate the phenomenon. Figure 4 documents cyclic creep load experiments at constant 80% humidity on the 60 g/m<sup>2</sup> TMP handsheets like those of Figure 1. The cyclic creep test began with a load of 25% of the 50% RH breaking load. It was sustained for 2 hours, then the load was cycled between 35 and 15% of 50% RH breaking load.

We performed three cycles, and each load was maintained for 1 hour. After cycling, we returned the load to 25% of breaking load and recorded additional data. The displayed curve is the average of three runs. For comparison, we superimposed average creep curves at constant 15, 25, and 35% of tensile strength. Notice that the load cycled samples suffered considerably greater total creep strain at the end of the load cycling period than they would have under their average load of 25% of breaking strength. To support the generality of the observation, we include Figure 5 which documents the same kind of testing for the unplasticized cellophane of Figure 2. The effect is at least as strong as in paper.

We wanted to assess whether moisture generated stress concentrations explanations can reproduce accelerated creep behavior in form and magnitude. To do this, we first needed a rough model of creep under varying load. Paper is a nonlinear visco-plastic material with fading memory. The mechanical action at any time is a complex function of its state of being at all previous times. There are formal ways of expressing this general mechanical action, but they are far too complex for simple calculations, and we are without specific, practical parameters. Instead, we will employ an approximation that allows us to construct creep curves under inconstant load from standard creep curves at different loads and to obtain an estimate of the role of stress concentrations in accelerated creep.

When out of the tertiary creep regime, creep rate under constant load is a monotonically decreasing function of time. One way to express this behavior for a constant load creep experiment would be to say that creep rate is a function of stress and strain  $(d\varepsilon_c/dt = f_t(\sigma,\varepsilon_c))$ , where  $f_t(\sigma,\varepsilon_c)$  decreases as  $\varepsilon_c$  increases. Viewed in this manner, it appears that a fixed stress is less effective in generating creep as strain increases. At a given stress,  $f_t(\sigma,\varepsilon_c)$  could be constructed from an experimental creep curve, and  $f_t(\sigma,\varepsilon_c)$ , could be determined over the range of  $\varepsilon$  and  $\sigma$  from a series of creep curves at different loads. If we take a leap of faith and assume that  $f_t(\sigma,\varepsilon_c)$  is (within limits) valid regardless of the stress and strain history, we could then construct creep behaviors under varying loads from  $f_t(\sigma,\varepsilon_c)$  as determined by constant-load creep curves.

To confirm that this works fairly well, take a closer look at the creep curves of Figures 4 and 5. These are creep strain plots; the initial elastic strains were removed. The cyclic load curve contains elastic components when the load differs from its initial value. We claim that we can construct the cyclic curves from the constant load curves. As an example, consider the creep in the first high load application of the cyclic load. This should be reproduced by the constant high curve beginning at the same creep strain. We drew arrows between points on the two curves to show the corresponding creep responses beginning at the same load and creep strain and proceeding for the same time period. Notice that there is a good match and that subsequent high load sections of the curve are reproduced similarly. To get the creep during a low load excursion, we would have to transfer to the constant low load curve at equal creep strain. This would be far to the right of the extent of the graph, at which point the low load curve would be nearly flat. Thus, the construction generates essentially no creep (or creep recovery) at the lowest load. To find the creep behavior after cycling, we should switch (again off the graph) to the middle curve at the final cycling strain giving, as observed, a lesser creep rate than on the middle

load curve at the same time. The construction makes a good rendition of the major features of the cyclic load curve. Creep during the high and middle load dosages is well reproduced; however, the creep recovery transpiring at the lowest load is missed. Please appreciate that switching between creep curves at the same creep time would grossly underpredict cyclic load creep.

To proceed, we need a constitutive equation for constant load creep rate:  $f_t(\sigma, \varepsilon_c)$ . Brezinski [1, 2] demonstrated, a long time ago, that one can construct "master creep" curves for papers. That is, creep compliance curves at different loads coincide if they are shifted a distance in the logarithm of time that is proportional to the applied load. The load shift coefficient,  $\alpha$ , is a material property. So, knowing  $\alpha$ , we need just one creep curve in order to reproduce creep under varying loads. Again leaning on Brezinski, we accept that there are two regimes in which paper creep can be empirically approximated with simple equations. At low loads and short times, creep curves mimic a power law behavior:  $\varepsilon_c/\sigma_o$  is proportional to the power, a, of time, where a is about 1/4. When the load is higher and/or the time is greater, creep depends on the logarithm of time as

$$\varepsilon_c / \sigma_o = A \ln(Bt + 1)$$
 (1)

When the creep action is safely in one of these regions, we can use the proper creep constitutive equation, the master creep construction, and the assumption that creep rate only depends on present load and creep strain to write a creep constitutive equation that is valid for changing load conditions. For the log-linear regime, this unfolds as below. The master creep curve condition is that the change in creep compliance with log time is proportional to the change with stress, or

$$\alpha \, \mathrm{d}(\varepsilon_c/\sigma_o)/\mathrm{dlnt} = \mathrm{d}(\varepsilon_c/\sigma_o)/\mathrm{d}\sigma_o \qquad (2)$$

Inserting Eqn. (1) into Eqn. (2) under the assumption that A is independent of stress gives

$$\alpha Bt = t \, dB/d\sigma_{o} \tag{3}$$

A solution of Eqn. (3) is  $B = B_o e^{\alpha \sigma_o}$ . Therefore, the general creep curve valid over the range of initial loads is

$$\underbrace{\left|\frac{\varepsilon_{c}}{\sigma_{o}} = \operatorname{Aln}(B_{o}e^{\alpha\sigma_{o}}t+1)\right|}_{,}$$
(4)

This is not yet in a form that we can use to propose a general  $f_t(\sigma, \varepsilon_c)$ . It explicitly contains the time from load initiation, which has no unique meaning in a variable load experiment. We are looking for a differential equation that depends only on the current values of stress and creep strain and that can be integrated to trace behavior over time. We find a time to strain conversion by taking the derivative of Eqn. (4) with respect to time:

$$\frac{d(\varepsilon_c / \sigma_o)}{dt} = AB_o e^{\alpha \sigma_o} / (B_o e^{\alpha \sigma_o} t + 1)$$
(5)

Using Eqn. (4) to eliminate time from Eqn. (5) gives the creep strain rate in terms of the creep stress and the strain at the time of interest:

$$\frac{d\varepsilon_c}{dt} = \sigma_o AB_o e^{\alpha \sigma_o} e^{-\varepsilon_c / A\sigma_o}$$
(6)

Generalizing this to arbitrary stress histories, we have an expression for  $f_1(\sigma, \varepsilon_2)$ , viz.

$$\mathbf{f}_{t}(\sigma,\varepsilon_{c}) = \sigma \mathbf{A} \mathbf{B}_{o} e^{\alpha \sigma} e^{-\varepsilon_{c}/A\sigma}$$
(7)

In the power law creep regime ( $\varepsilon_c/\sigma_o = Ct^a$ ), an almost identical derivation gives the creep rate constitutive behavior of Eqn. (8):

$$f_t(\sigma, \varepsilon_c) = a(C_o \sigma)^{1/a} e^{\alpha \sigma} (\varepsilon_c)^{(a-1)/a}$$
(8)

where  $C = C_0 e^{\alpha \alpha \sigma}$ . These are our proposed creep constitutive equations in differential form. The parameters, A, B<sub>0</sub>,  $\alpha$ , C<sub>0</sub>, and a, are material properties affected by the environment. A major shortfall of Eqns. (7) and (8) is that they do not predict creep recovery. This is not the only problem. Notice that stress is in the denominator of the exponential term in Eqn (7). Generally, this leads to tiny creep rates at low stresses, but if during cycling we reach a state in which stress and strain are of opposite signs, the strain rate can blow up. This is impossible to justify physically. We rightly have reservations about both equations in situations allowing stress and creep of opposite polarity. We cannot extrapolate between creep strains in tension and compression. Nonetheless, we don't need a perfect constitutive equation; we just want one that is good enough to test our accelerated creep mechanism. We will strictly avoid using Eqn. (7) or (8) when a material is cycling between tension and compression.

### ACCELERATED CREEP MODELING

We just argued that moisture-induced stress concentration mechanisms are operative, but we have not demonstrated their efficacy. Can they account for the total amount of accelerated creep observed? When is moisture-gradient-driven accelerated creep important and when is heterogeneity-driven accelerated creep important? To address these questions, we devised a mechanical model that, although far too simple to replicate detailed mechanical action, incorporates all the ingredients necessary for this kind of accelerated creep. We reasoned as follows. The history of hygroexpansion depends on the location in the sample. A minimal degree of heterogeneity can be modeled with two separate elements each standing for a different section of a sample. The sections of a real material that are sharing load are bound together so that they creep as a whole, as shown in Figure 3. Therefore, we decided to model the two sections as elements in parallel (See Figure 6). The elements are constrained to deform together. To maintain equal deformation, the individual loads are allowed to vary under the constraint that their sum equals the overall load. The differential constitutive equation for each section will be expressed as  $d\varepsilon = f_s(F)dF + f_t(F,\varepsilon_c)dt + \beta dm$ . The first term accounts for the immediate elastic response to a change in load; the second term takes care of creep; and the third handles hygroexpansion. The creep term will have the form of Eqn. (7) or (8) depending on which best fits the constant load creep curves, and linear elasticity ( $f_s(F) = 1/E$ ) will be accepted. In representing moisture-gradient-driven accelerated creep, the moisture content as a function of time will differ between sections. The constitutive coefficients will be given different values and different moisture sensitivities to demonstrate heterogeneity-driven accelerated creep.

To assess moisture-gradient-driven accelerated creep in our humidity chamber, we mentally subjected the two-section construction of Figure 6 to ramped-wave moisture cycling experiments. The moisture content in the leading section progresses linearly between two moisture states over a time equal to one half the sample sorption time plus the time for the chamber to change humidity. The trailing section is treated to the same moisture ramp, but delayed one half the sorption time. We chose the humidity sequence of the simulation to correspond to our experimental program: the load is applied with both strings at the high humidity extreme; after a period of time, the relative humidity is alternated repeatedly from wet to dry at a regular rate. We wrote a Mathcad program to do the appropriate numerical integrations. Model coefficients (hygroexpansion, sorption time, wet and dry moduli, creep regime, and wet and dry creep and master creep coefficients) were taken from experiments. Intermediate moisture content values were linearly extrapolated from the wet and dry numbers.

First, we tried to simulate the paper accelerated creep experiment documented by Figure 1. The results are presented in Figure 7. The values of the model parameters for this and subsequent accelerated creep models are listed in Table I. The power law creep model was used because it made a better fit to the wet and dry, constant-humidity creep curves. Notice that the slope of the wet creep curve with log time (Figure 1) does not level off as time increaes. In Figure 7, the upper graph contains the creep strain curves for a wet, a dry, and a humidity-cycled sample. A significant amount of accelerated creep was generated, but it was less than the experiment of Figure 1. The shape of the numeric cyclic creep curve bears similarities to the experiment on the desorption excursions. Both exhibited a cusp at the bottom of the first moisture drop only and displayed relatively flat creep curves during subsequent dry periods. However, model-made wet creep sections were not as rounded as those in the experiment. Insights in the accelerated creep process are gained by viewing the lower graph. Here, the first three cycles of the load (relative to even distribution) on Element 1 are plotted versus linear time. Element 1 represents the portion of the sample that responds first to environmental change. When the humidity cycles low, Element 1 dries first. Its sorption-induced shrinkage is restrained by Element 2, therefore, the dry cycle begins with Element 1 carrying the majority of the tensile load. Due to the sensitivity of creep to load, there is increased creep rate in Element 1 during this time period. When Element 2 dries, it also shrinks and starts to carry more of the load. Because of the extra early creep in Element 1, Element 2 ends up with most of the load after both elements are dry. During the all-dry period, the load relaxes toward an even distribution as a result of the higher creep rate in the element with higher load. As Element 1 goes wet again, it

tries to expand and more of the load is transferred to Element 2. Now, Element 2 experiences its creep burst. When all is wet again, Element 1 is a bit overloaded. As always, in constant moisture conditions the load distribution relaxes to an equilibrium, in this case, equal distribution of load.

Notice that the amplitude of the load excursions from even distribution increase with cycle number. This is a general observation that carries through to all moisture-gradient-driven and heterogeneity-driven simulations. When the load is concentrated in one element, the creep section of that element deforms rapidly ameliorating the imbalance. The creep constitutive behavior dictates decreased equal-load creep rates at higher strains. Therefore, as accelerated creep continues, the material becomes less responsive to sorption-created load imbalances, and the load imbalances get progressively larger. This phenomenon could be important in the failure of paper in an inconstant environment. Load-dependent failure mechanisms could be triggered in an even more virulent manner as humidity cycles.

In tensile moisture-gradient-driven accelerated creep, the high loads are experienced by an element when it is in the dry state. Thus, the dry creep constitutive equation at high load and large strain dominates the model prognosis for accelerated creep. The dry creep rate at higher strains must be extrapolated from constant-load creep experiments conducted over reasonable time periods. To demonstrate the sensitivity to creep rate extrapolations, we present, in Figure 8, a repeat of the simulation with the creep constitutive equations taken from fits to the log linear form rather than the power law. (In this case, the power law curve gave a better match to the data, but its extrapolation is still in question.) There is still extra elongation in cycling humidity, but the prediction of accelerated creep is greatly muted. As modeled, moisture-gradient-driven accelerated creep is highly dependent on the extrapolation of dry creep behavior to high strain.

In a compression creep moisture-gradient-driven simulation, the high load periods would occur in wet elements. An element creeps more when wet, thus, you might expect greater accelerated creep in compression. However, there is another phenomenon that compensates. The wet element also has a lower elastic modulus. This allows for more elastic strain reducing the buildup of load. The moisture dependence creep accentuates accelerated creep in compression, whereas the moisture dependence of modulus adds to accelerated creep in tension. A theoretical comparison of accelerated creep in tension and compression depends on the details of the moisture dependence of all parts of the constitutive equations. We do not have experimental compression results; therefore, we cannot compare.

Now, we turn to heterogeneity-driven simulations. Again, there are two model elements. This time we change the moisture simultaneously in the two elements, but they can have different hygroexpansivities and elastic moduli and different sensitivities of parameters to moisture. Since we do not have a method for experimental determination, we have to estimate the degree of heterogeneity. When we visualize heterogeneity-driven accelerated creep in paper, we picture the sorption-induced load cycling caused by the anisotropy in fiber hygroexpansion. Single fiber hygroexpansion is much greater in the lateral directions than along the axis. The axes of fibers bonded together in a sheet will not, in general, be aligned. Thus, along the direction of tensile load, they will have different coefficients of hygroexpansion. Moisture cycling will induce

fiber-level load cycling due to the incompatibilities of hygroexpansion at the bonds. If we assume that the component of hygroexpansion at an angle  $\theta$  to the fiber axis is proportional to  $\sin^2\theta$ , we can show by a straightforward geometrical calculation that in a sheet of random fiber orientation the average ratio for bonded fibers of the component of hygroexpansion along the tensile axis is 2.47. For our heterogeneity-driven simulation documented as Figure 9 (along with the already-measured, overall sheet parameters), we made the second element 2.47 times more hygroexpansive than Element 1, 2.0 times as elastically compliant, and 1.67 times more sensitive in modulus to moisture. Again, we get accelerated creep of the same magnitude as in the experiments. The wet-creep sections match experiment better in that they are more rounded; however, unlike experiment and moisture-gradient-driven modeling, there is now significant dry-time creep. Referring to the second cycle of the bottom graph of Figure 9, we observe a different pattern in load cycling. As the sheet dries, Element 2 tries to shrink more and take on extra load. On the other hand, during wetting it expands more and yields load to Element 1. During the wet periods and dry periods the heavily loaded element creeps more, and the load distribution relaxes toward an equilibrium. As before, the load excursions increase with time, but only on the more hygroexpansive element. The magnitude of heterogeneity-driven accelerated creep is not as dependent on the creep extrapolations; we do see less accelerated creep with log-linear fits, but the decrease is much less than in the moisture-gradient-driven example.

We present combined moisture-gradient-driven and heterogeneity-driven accelerated creep modeling in Figure 10. The sorption lag parameters are the same as in Figure 7, and the heterogeneity parameters are as in Figure 9. Now, we generate accelerated creep that is about equal to experiment. Each mechanism gives us roughly the same level of extra creep; therefore, we must conclude that both processes are important. In the modeling, we have artificially associated the element that responds first to the environment with the stiffest, least hygroexpansive element. Of course, we made many other compromises because of ignorance and for the sake of simplicity. Any near numeric match is fortuitous. One can easily point to assumptions that cause the calculated accelerated creep to be more or less powerful than in real life. We are not claiming to have an accurate model. We do believe, however, that the modeling supports our contention that both moisture-gradient-driven and heterogeneity-driven accelerated creep function in paper.

Next, we modeled the PD00 cellophane in the machine direction. It creeps more and is more hygroexpansive than paper, but shows less accelerated creep. It has the opportunity for moisture-gradient-driven accelerated creep from cycling moisture profiles through the thickness direction of the sheet. However, without fiber-fiber bonding, a comparable heterogeneity-driven mechanism is difficult to rationalize. Figure 11 is the moisture-gradient-driven creep modeling which compares favorably with the experiment of Figure 2. We used a power law creep equation because it fit the data better. You might expect less tensile moisture-gradient-driven accelerated creep in cellophane because creep decreases more with moisture content. However, it also has greater hygroexpansivity, a larger master creep stress coefficient, and a larger dependence of modulus on moisture content. All these enhance accelerated creep and overcompensate for the loss of creep rate upon drying. Without an additional kick from heterogeneity, paper should

experience less accelerated creep than cellophane. This is more evidence that cycling stress gradients in paper have two sources.

### ARMSTRONG TEST

As discussed in the history section, Armstrong [23] showed that accelerated creep does not occur when samples are subjected to steady-state moisture gradients. This observation is clearly compatible with the proposed mechanism which requires a continually changing moisture content to maintain local load cycling. Nonetheless, to remove any doubts, we made a model of creep in which one element was taken dry and the other was maintained wet. As in Figure 10, TMP sample properties and combined mechanisms were used for this simulation. After the sample creeps in the wet state for 1.5 hours, Element 1 dries. It remains dry, whereas Element 2 never leaves the wet state. In this way, creep under a steady moisture gradient is assessed. The results are documented in Figure 12. Just after Element 1 dries, there is a creep spurt. However, as time proceeds, the load distribution relaxes to an equilibrium in which both elements creep at the same rate. The final steady-state moisture-gradient creep rate is intermediate between the wet and dry values. Clearly, repeated moisture cycling is needed to make a sample elongate more than it would in its wet state.

### FIBER ACCELERATED CREEP

Conforming to tradition, we arbitrarily maintain that accelerated creep occurs only when creep is greater in a cyclic humidity environment than in the wet state. This is a high threshold for onset of the phenomenon. Sorption-induced stress concentrations often lead to significant additional creep. However, if this extra creep is not sufficient to overcome the loss in creep rate from dry state rigidity, the experiment is still classified as a nonoccurrence of accelerated creep. Stress concentrations always accompany sorption, and they always contribute, somewhat, to creep, even though they may not result in "accelerated creep."

Accelerated creep, as defined, is usually not observed in single-fiber tensile creep tests [3,38,40]. We think this is because there is no opportunity for paperlike heterogeneity-driven accelerated creep and most fibers sorb quickly in comparison to the chamber RH transition times and the cycle times. When a material sorbs more rapidly than the environmental chamber changes state, moisture gradients in the material are greatly attenuated. When material sorption times are small relative to cycle time, stress concentrations exist for only a small portion of the creep time, reducing the extra creep from stress concentrations. We demonstrate these phenomena by running the cellophane moisture-gradient driven model (Figure 11) again, but with a sorption time of 30 seconds rather than 10 minutes. As shown in Figure 13, we no longer observe accelerated creep. The deviations from the equal load state are smaller and persist for a lesser portion of the cycle time.

Aramid fibers [38] are exceptions to the no fiber accelerated creep rule. Kevlar (and presumably the other aramid fibers) sorb very slowly compared to the fibers that do not show accelerated creep. Kevlar fiber sorption times are on the order of tens of minutes [43] giving

them the potential for moisture-gradient-driven accelerated creep in chambers that change RH in seconds or minutes and cycle humidity in hours. However, from our point of view, there is still a problem. It is the normally large sensitivity of creep to load that powers the extra creep from load cycling that we rely on to explain accelerated creep. But, constant load creep in Kevlar is not very sensitive to load. As published by Ericksen [44] and confirmed in our laboratory, the constant load creep rate at equal times after load application increases very slowly with load for Kevlar 29. In fact, Kevlar 29 creep compliance decreases substantially with load [44]. On the other hand, we found that Kevlar 29 fiber creep depended little on moisture content. Therefore, cyclic loading would have less dry-state creep loss to overcome in creating accelerated creep. Early on, we realized that Kevlar was a challenge to the mechanism, and we committed to do experiments to resolve the issue.

The previous accelerated creep work on Kevlar fibers by Wang et al. [3,38,45] was done above room temperature in an oven. We discovered that Kevlar 29 fibers also exhibit accelerated creep at room temperature. Thus, working solely with Kevlar 29, we were able to proceed without the annoyance of conducting experiments in an oven. Figure 14 documents a typical accelerated creep run on a 1.5 denier (about 15  $\mu$ m in diameter) fiber that was equilibrated to 90% RH for 8 hours, loaded with a 0.056 N weight, held at 90% RH for 2 hours, then cycled between 90 and 10% RH at one-hour intervals. The cyclic humidity creep performance is as reported by Wang et al.; however, it is much different from that of paper and cellophane. Initially, the extra sorption-induced creep obliterates the normal hygroexpansive length changes. After many cycles, a slight negative hygroexpansion appears.

According to conventional wisdom [46], Kevlar tensile deformation is dominated by rotation of its crystallites toward the fiber axis. Tensile load application induces an immediate elastic rotation and a time-dependent creep rotation. Crystallite rotation stiffens the fiber, reducing elastic and creep compliance. Moisture apparently decreases the crystallite angle thereby reducing the length of the fiber. This is clearly a very different deformation process than we experience with paper. The constitutive equation approximations used above would be inappropriate. However, the influence of moisture on crystallite angle and the relatively long-time sorption process gives the opportunity for consequential sorption-induced load cycling to accompany RH cycling. For this to produce accelerated creep, Kevlar must creep more under a cycling load than at a constant average load. The sensitivity of creep rate to load in a standard creep test is not the crucial observation. In Figure 15, we present the results of a cyclic load test on a Kevlar 29 fiber. At 90% RH a 0.0559 N load is applied for 2 hours. Then, we cycle the load between 0.0304 and 0.0813 N at one-hour intervals. After three cycles, we return the load to 0.0559 N and continue the creep test. In this case, the Kevlar behavior resembles that of the cellulosics (Figures 4 and 5). Cyclic loading causes additional creep. After load cycling, the sample creep rate is less than it would have been under a constant average load. Kevlar is indeed a much different material, and we have not contributed a coherent description of its load-dependent deformation. However, it has all the requisite properties for moisture-gradient-driven accelerated creep, and from a mechanistic perspective we are comfortable with its accelerated creep behavior.

### **MOISTURE-INDUCED PHYSICAL AGING AND WORK HARDENING**

We assert that moisture-induced stress concentrations cause accelerated creep. Padanyi [31, 32] and others [35] maintain that moisture-induced physical "deaging" causes accelerated creep. To us, "deaging" by moisture cycling is not an explanation; it is a problem that begs its own solution. We believe that accelerated creep and moisture deaging are closely related phenomena and that both can be understood in terms of moisture-induced stress concentrations. We will directly address the desorption "deaging" experiments since these are the more perplexing; however, the same arguments would apply to absorption "deaging".

We begin by presenting experiments that confirm and slightly extend Padanyi's desorption "deaging" observations. Figure 16 communicates our "deaging" results. First we did a creep test at 42% of the 50% RH breaking load on one of our TMP samples that had been at 50% RH for a long time. Its creep curve is designated by the "old" label. We dried this sample (and others like it) at 10% RH for an hour. After returning to 50% RH and waiting 45 minutes, we repeated the creep test. These samples generated the high creep compliance curves of Figure 16. We "deaged" some of these again and redid the creep after 45 minutes at 50% RH. As reported by Padanyi, the creep curves repeated. For other samples in the group we waited 18 hours before applying load and recorded creep curves intermediate between "old" and "0.75 hours." To demonstrate the extent of creep mechanical conditioning achieved in these tests, we then repeated creep tests on the "18-hour" samples without "deaging." This endeavor produced the "work-hardened" curves. The creep load exposures are clearly sufficient to do mechanical conditioning. However, a simple dry cycle transforms a sample that would have exhibited work hardening in a subsequent creep test to a "young" sample of high creep compliance. All this transpires, and there is no possible appeal to a glass transition mechanism. Amazing!

Papers [47], along with metals and plastics [48], sustain residual stresses. That is, due to the uneven nature of cooling and/or drying and/or inhomogeneity, a significant stress distribution exists through the material even under no-load conditions. Paper seems to display contradictory behaviors: it creeps and it maintains residual stresses. A material that creeps is yielding to load over time. When residual stresses arise, such a material should respond locally to relieve those stresses. However, this may not happen quickly. Paper creep rate is very sensitive to load. When load is low, paper does not creep rapidly and relatively low levels of residual stresses persist. Nonetheless, as discussed above, the creep rate increases greatly as load increases, allowing paper to both have residual stresses and exhibit significant creep under external load. Our explanation of accelerated creep hinged on the notion that the distribution of load through the paper plays a large role in the determination of the overall creep rate. By the same token, residual stresses should assist creep. Under an overall applied load, the extra creep in regions of high stress will overcompensate for the loss of creep in regions of low load. When a piece of paper encounters a sorption event, residual stresses are established making the paper more creep compliant. As time progresses, these residual stresses slowly relax away, producing a more creep resistant sample. If, at any time, the sorption event recurs, the sample returns to the more compliant (higher residual stress) state.

Our accelerated creep modeling apparatus can mimic the physical aging scenario and thereby elucidate the mechanics. First, we employed our standard two-element model to demonstrate that moisture-gradient-driven stress concentrations can establish the residual stresses that will increase creep compliance in a physical aging experiment. (There can also be heterogeneity-driven residual stresses, but we present just the one example.) Since model elements must pass between tension and compression in a "deaging" simulation, we could no longer use our paper-based constitutive equations. Instead, we adopted the Eyring creep element [49] (creep rate proportional to the hyperbolic sine of a constant times the load) which is the outcome of an elementary application of statistical mechanical to strain in a polymer under load. It is sufficiently load sensitive to drive physical aging and work hardening. The following exercises are intended only to clarify mechanisms that we believe are active in paper.

We begin with a sample unloaded and without residual stresses. We dry it out. Assuming that one portion dries before the other, residual stresses arise in the sample. After awhile, we rewet the sample, reworking the residual stress distributions, and ending the "deaging" treatment. The sample "ages" in the damp state until we begin a creep test. The creep strain generated is compared to that of an "old" sample without residual stress, i.e., one that has been in the damp state forever. The top graph of Figure 17 shows the overall elongation of a sample that is "deaged" for one time period, "aged" for 10 time periods, then subjected to creep for another 10 time periods. For comparison, an elongation curve of an old, not-deaged sample is included (dotted line). The bottom portion of Figure 17 depicts the load in Element 1 relative to the elastic load needed to give a strain equal to the hygroexpansive strain. At the start of the "deaging" process, only Element 1 is dry. This reduces its equilibrium length, forcing it to go into tension and Element 2 to go into compression. This is a fairly high load, and, in the beginning, the residual stresses relax out rapidly. By the time Element 2 also flips dry, Element 1 is elongated and Element 2 is shortened. Thus Element 1 is in compression in the all-dry state. Now the residual stress is smaller and relaxation is much slower. When Element 1 returns to the wet state, it tries to expand and goes into deep compression. Residual stresses are large, and the creep segment of Element 1 is quickly shortened, whereas creep segment of Element 2 lengthens. This causes Element 1 to be in tension in the following all-dry, "aging" state. The residual stresses are relatively small and persist throughout the "aging" period. After 10 time periods, a creep load sufficient to give an elastic elongation equal to 25% of the hygroexpansion is applied. Both samples immediately respond elastically and creep over time. The top graph of Figure 17 reveals that residual stresses in the "deaged" sample lead to a faster creep rate, but Figure 18 better demonstrates the "aging" effect. There, the creep portion of the "deaging-aging" experiment is plotted versus log time for three samples. The most creep compliance curve was "aged" only 1.0 "deaging" time. The middle curve sample was 100 "deaging" times old when load was applied; whereas the "old" sample had no residual stresses which is the state it would reach if stresses relaxed forever.

At high applied loads, small load differences make great differences in creep rate; therefore, stress variations relax out rapidly. This means that residual stresses can be removed by holding a sample under load then releasing it. In this way, a creep exposure can make a sample less creep compliant. In other words, a type of creep work hardening transpires by the removal of residual stresses. Figure 19 is a mathematical demonstration of this phenomenon. In order to set up residual stresses, two samples are subjected to a "deaging" experience. Shortly after "deaging", a load sufficient to give an elastic strain equal to 35% of hygroexpansion is applied to one of the samples. It creeps for 10 "deaging" times at which time the load is removed. In the final free state, there is less load on Element 1 for the sample that endured creep than for the sample that was untreated. Thus the "work-hardened" sample will be less compliant in a later creep test.

### CONCLUSIONS

The moisture-induced stress concentration mechanism can explain accelerated creep, sorption-induced physical aging, and one type of creep work hardening.

Our main point is that accelerated creep is just creep, pure and simple. If one properly handles material creep response, spatial and temporal sorption changes, hygroexpansion, heterogeneity, and moisture dependence of properties, accelerated creep will naturally emerge from the analysis. In the future, a better understanding of accelerated creep will come not from the postulation of special mechanisms but rather from a better understanding of the creep and sorption processes.

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### TABLE I

Figure #	7	8	9	10	11
Hygroexpansion. $(\beta_1 E_1 + \beta_2 E_2)\Delta m/(E_1 + E_2)$ : (%)	0.2	0.2	0.13	0.13	0.6
sorption time (min)	12	12	0	12	12
$\sigma/[(\beta_1 E_1 + \beta_2 E_2)\Delta m]$	1	1	1	1	1
Time shift: $\alpha(\beta_1 E_1 + \beta_2 E_2) \Delta m$ wet and dry	9.2	9.2	9.2	9.2	11.5
dry creep exponent (ad)	0.22		0.22	0.22	0.20
wet creep exponent (aw)	0.31		0.31	0.31	0.31
dry creep coef. $(Co(\beta_1E_1+\beta_2E_2)\Delta m)^{1/a}/\beta\Delta m (1/s)$	2.2*10 <sup>-5</sup>		2.2*10 <sup>-5</sup>	2.2*10 <sup>-5</sup>	3.0*10 <sup>-8</sup>
wet creep coef. $(Co(\beta_1E_1+\beta_2E_2)\Delta m)^{1/a}/\beta\Delta m$ (1/s)	4.7*10 <sup>-3</sup>		4.7*10 <sup>-3</sup>	4.7*10 <sup>-3</sup>	7.3*10 <sup>-4</sup>
dry creep coef. (A( $\beta_1 E_1 + \beta_2 E_2$ ) $\Delta m$ )		0.07			
wet creep coef. $(A(\beta_1E_1+\beta_2E_2)\Delta m)$		0.37			
dry creep coef. Bo (1/s)		0.057			
wet creep coef. Bo (1/s)		0.028			
E <sub>dry</sub> /E <sub>wet</sub> for Element 1	2	2	1.5	1.5	3
E <sub>dry</sub> /E <sub>wet</sub> for Element 2	2	2	2.5	2.5	3
$\beta_2/\beta_1$	1	1	2.5	2.5	1
E <sub>2</sub> /E <sub>1</sub>	1	1	0.6	0.6	1
E is elastic modulus, $\beta$ is coefficient of hygroexpansion, $\sigma$ is total stress, $\Delta m$ is % change in moisture content, $\beta\Delta m$ is hygroexpansion					
content, pain is hygroexpansion					

### Model Parameters Used in Simulations for Results Shown in Figures 7-11.

80-30% RH Accelerated Creep and 80% RH Creep for TMP Samples at 25% of 50% RH Breaking Load

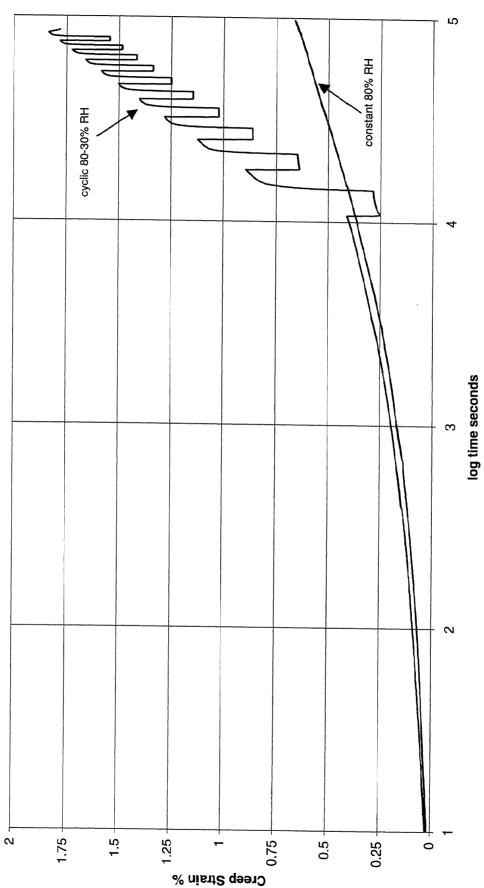
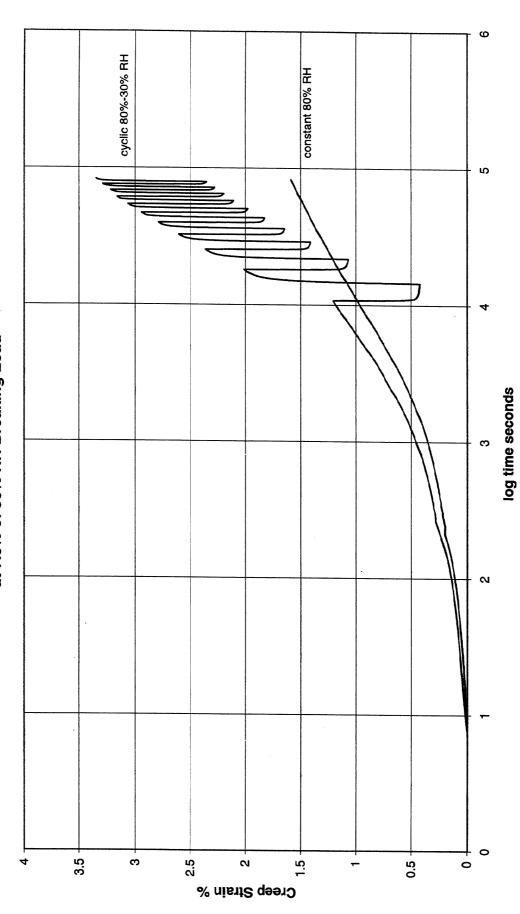
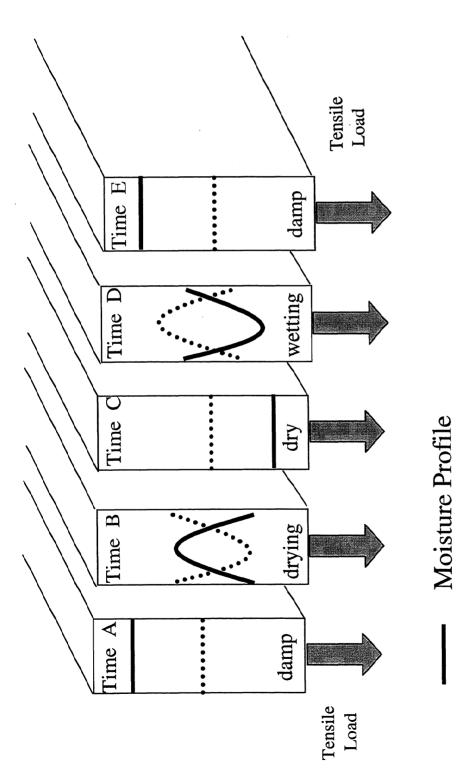


Figure 1

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80%-30% RH Accelerated Creep and 80% RH Creep for MD PD00 Cellophane at 7.5% of 50% RH Breaking Load



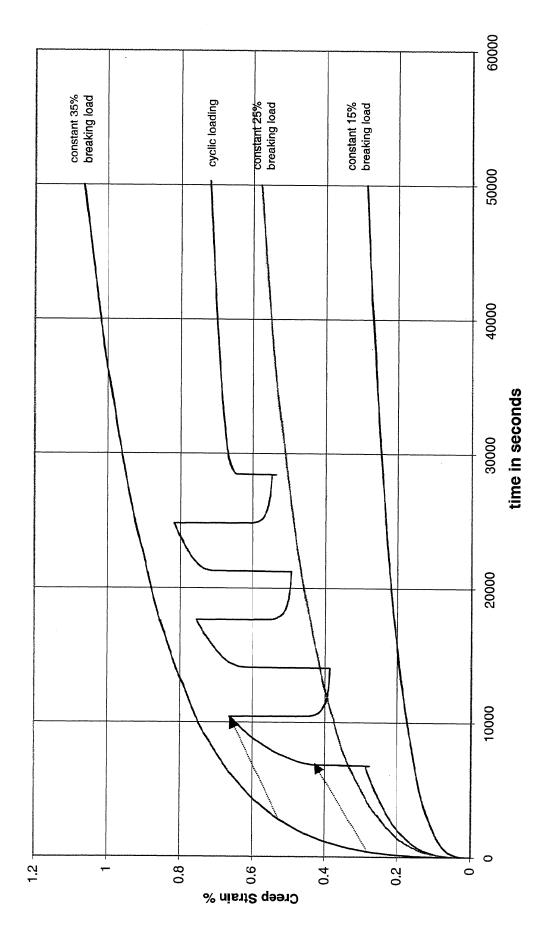


Hypothetical Moisture and Stress Profiles through a Sample Under Tensile Load over a Sorption Cycle

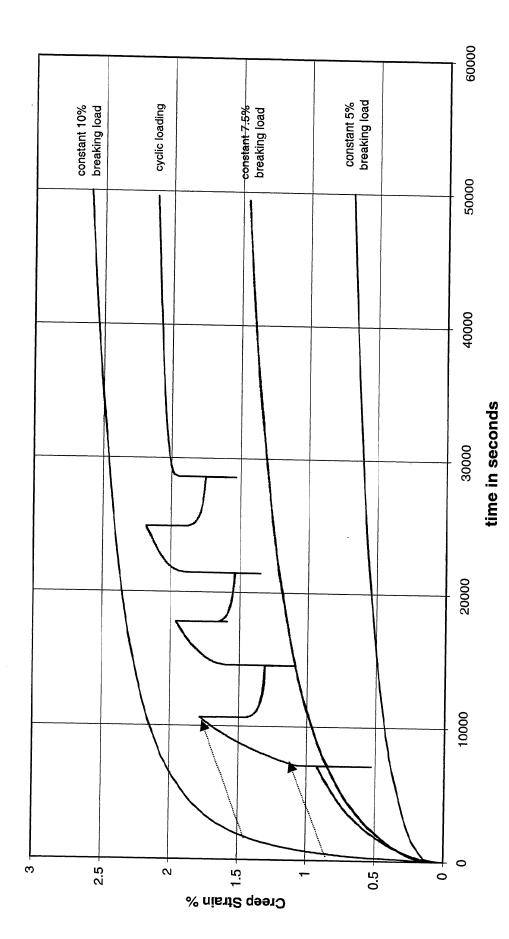
**Stress Profile** 

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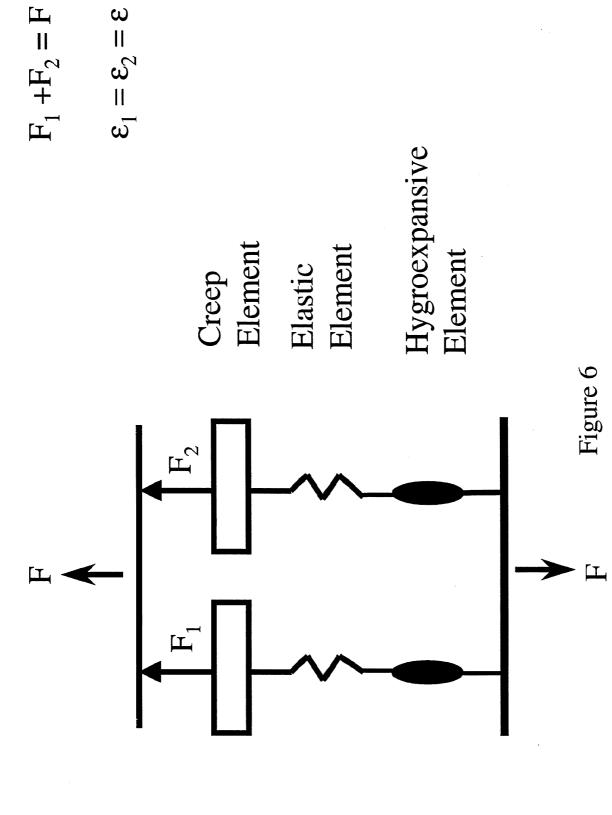
80% RH Load Cycling for TMP Samples between 15% and 35% of 50% RH Breaking Load



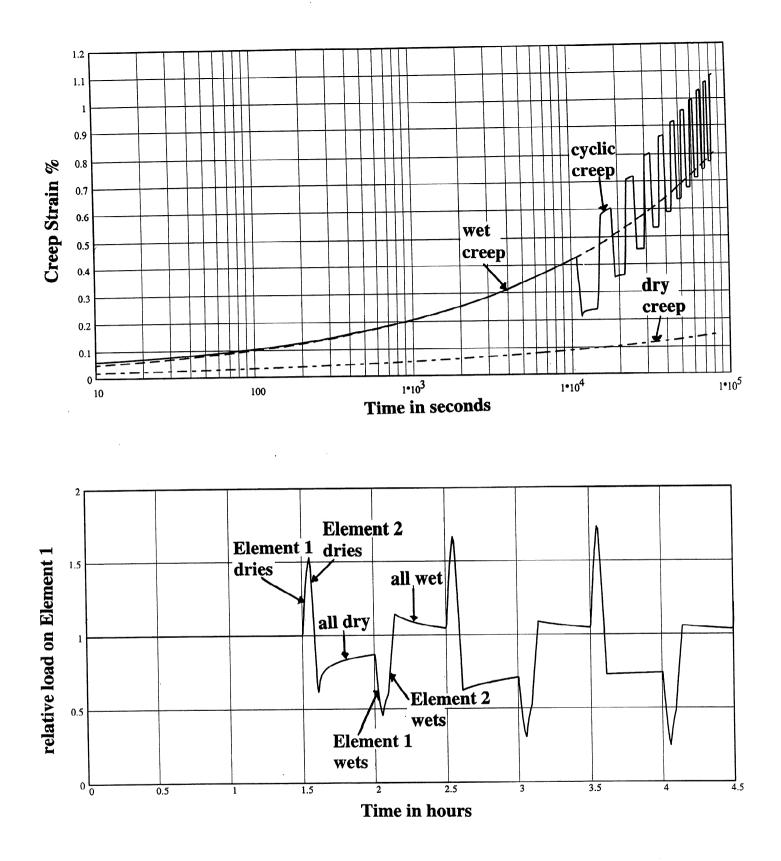
80% RH Load Cycling of PD00 Cellophane in the MD between 10% and 5% of its 50% RH **Breaking Load** 



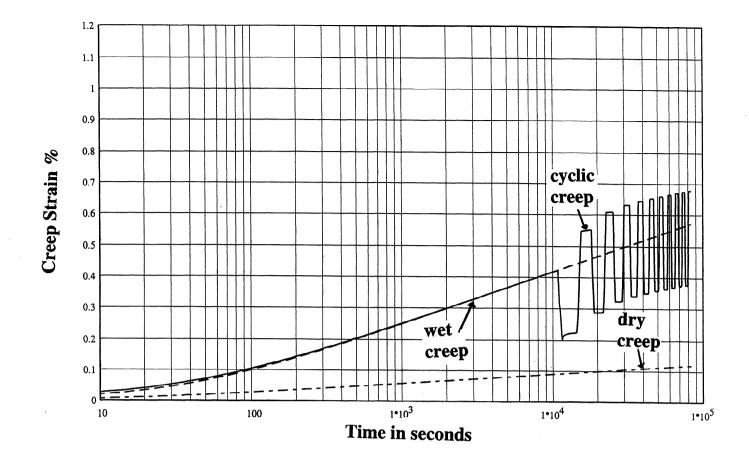




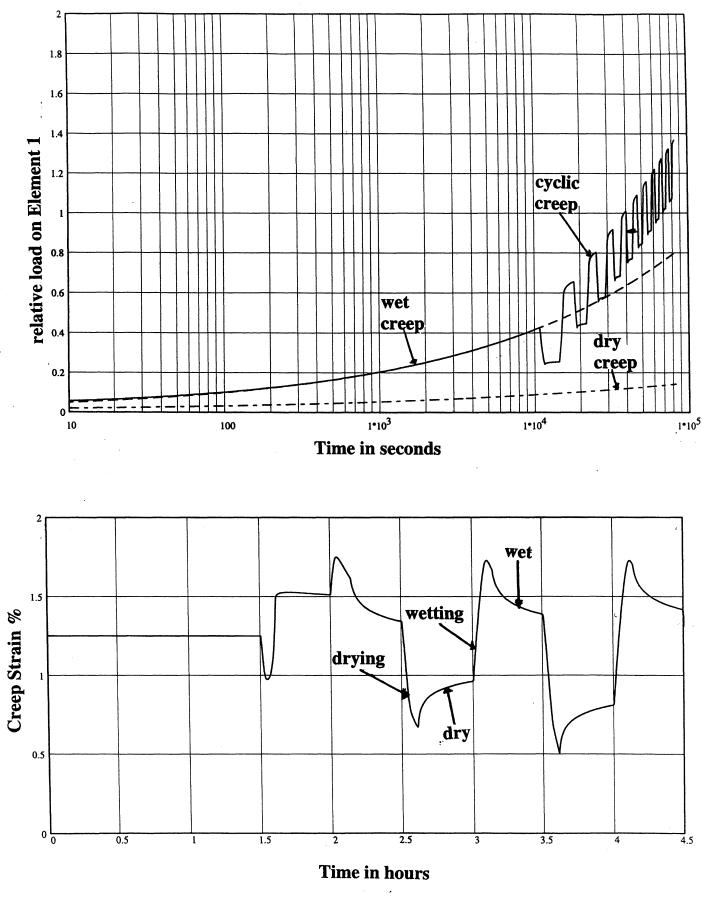
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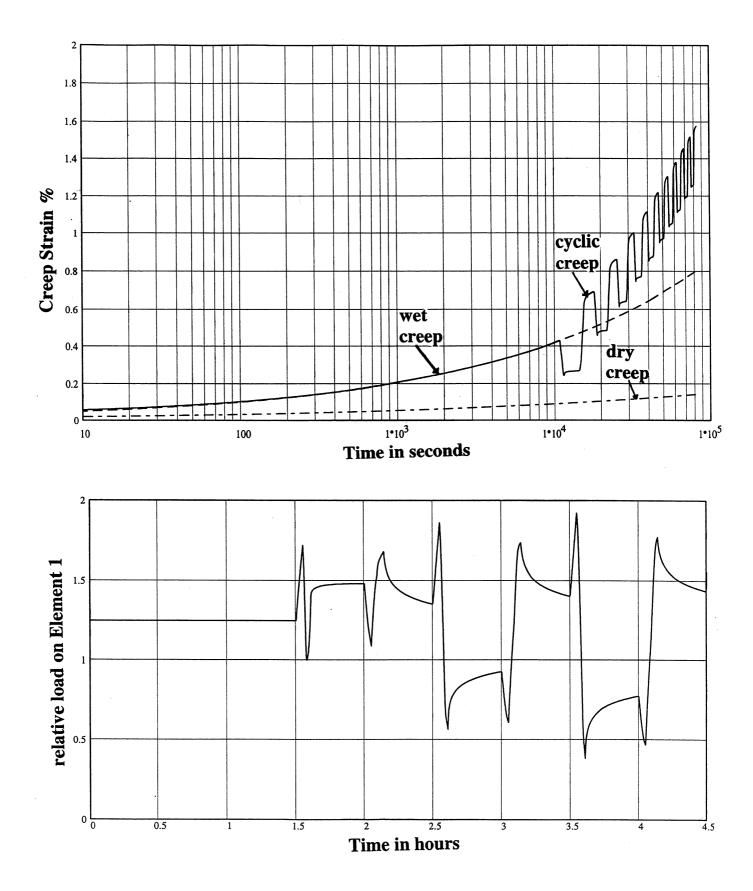
Moisture-Gradient-Driven, Power Law Accelerated Creep Modeling Using Material Properties of TMP Paper of Figure 1



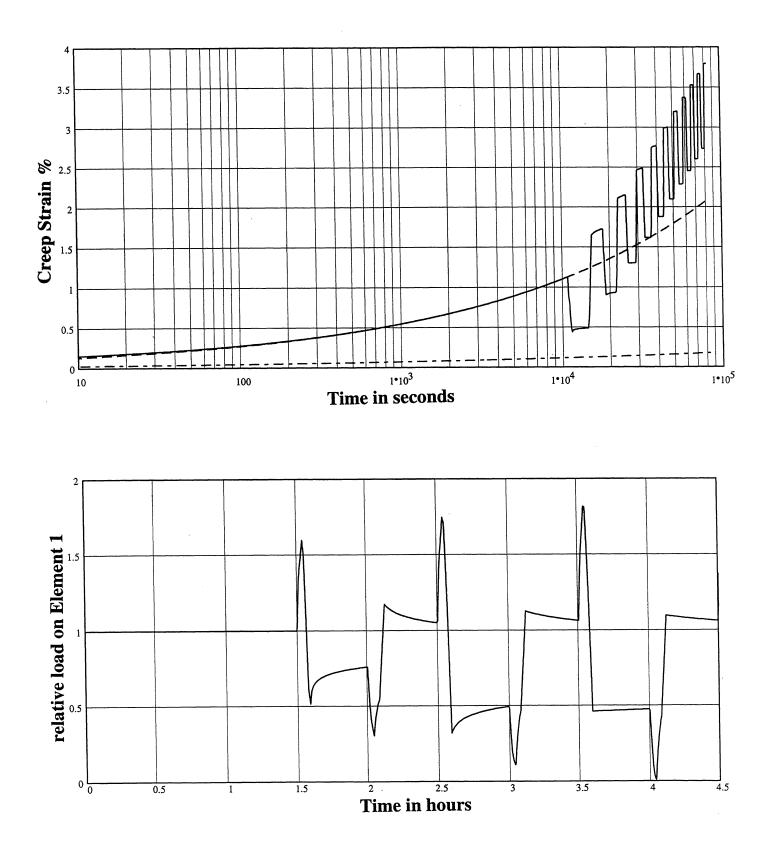
Moisture-Gradient-Driven, Log. Linear Accelerated Creep Modeling Using Material Properties of the TMP Paper of Figure 1



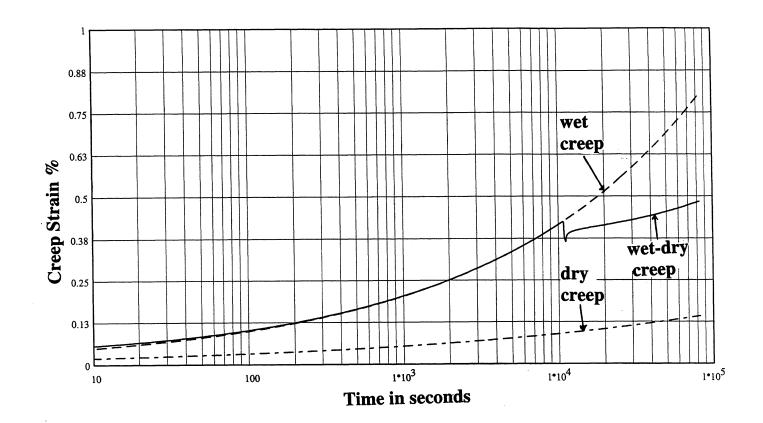
Heterogeneity-Driven, Power Law Accelerated Creep Modeling Using Material Properties of the TMP Paper of Figure 1

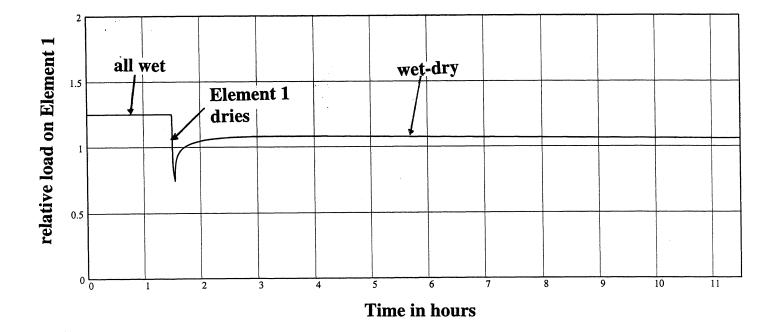




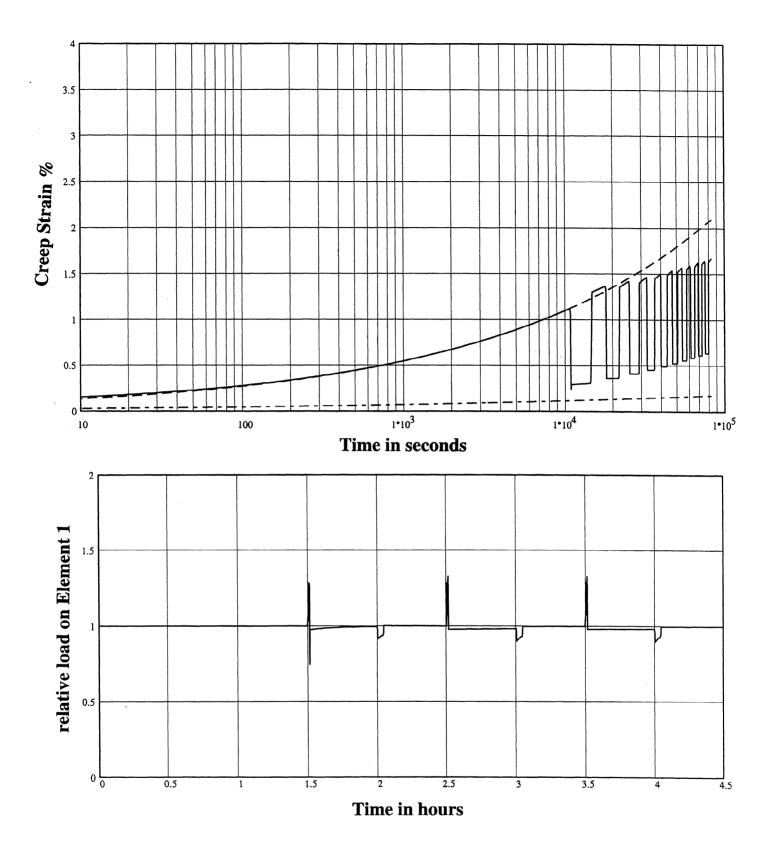


Moisture-Gradient-Driven, Power Law Accelerated Creep Modeling Using Material Properties of the MD PD00 Cellophane

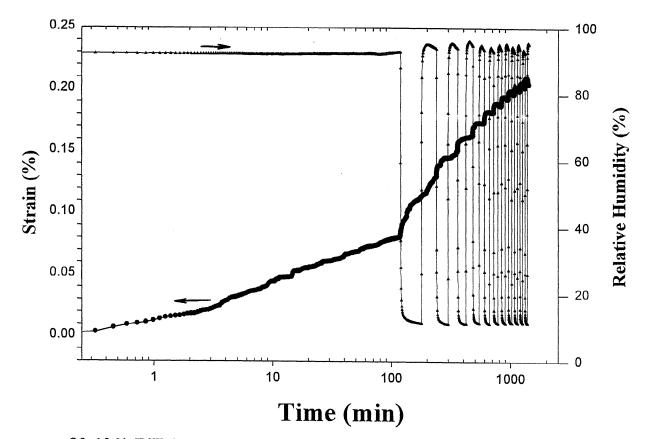




Moisture-Gradient-Driven Plus Heterogeneity-Driven, Power Law Armstrong Modeling Using Material Properties of the TMP of Figure 1



Moisture-Gradient-Driven, Power Law Accelerated Creep Modeling Using Material Properties of the MD PD00 Cellophane with an Artificially Small Sorption Time



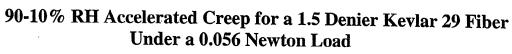
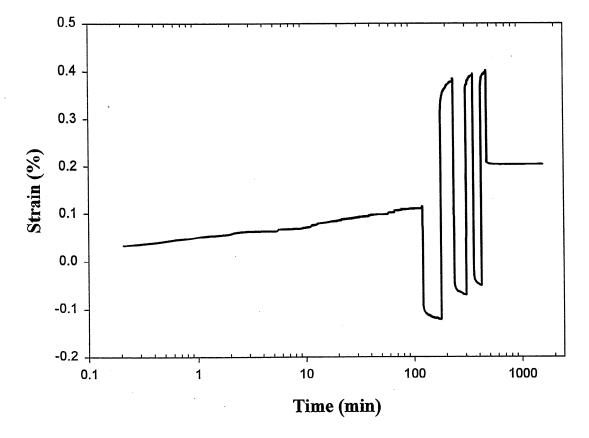
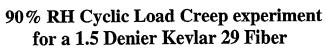
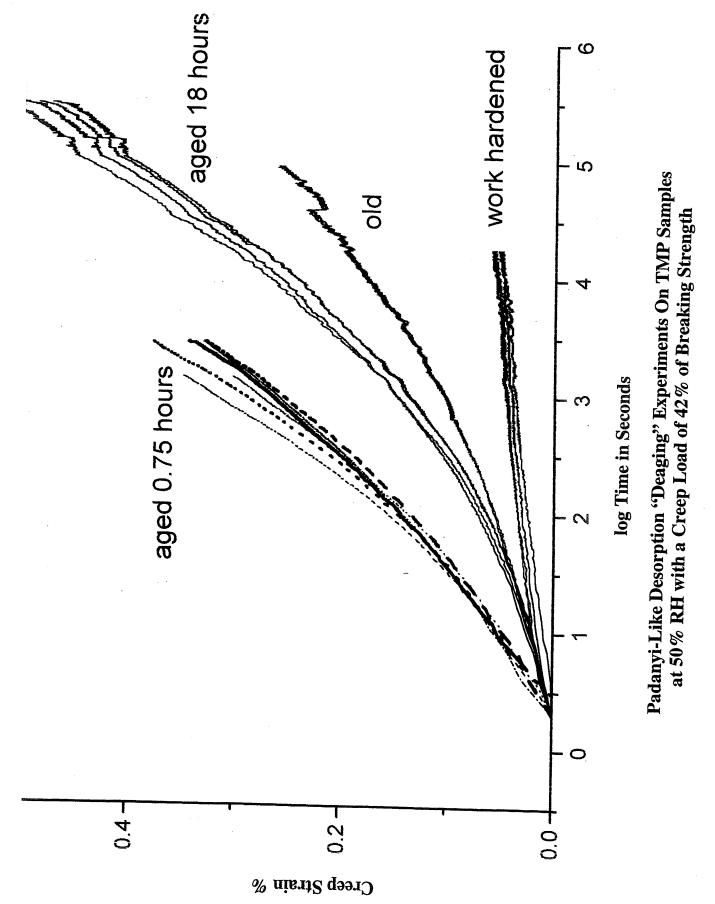
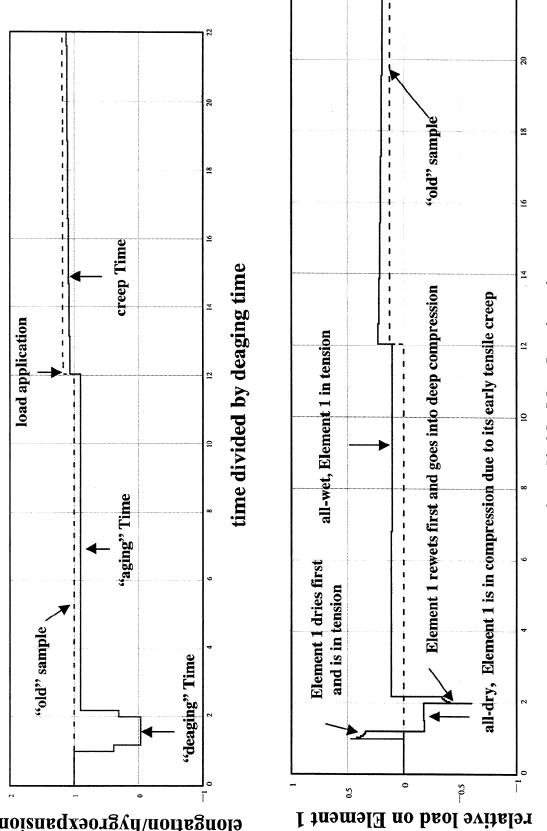


Figure 14









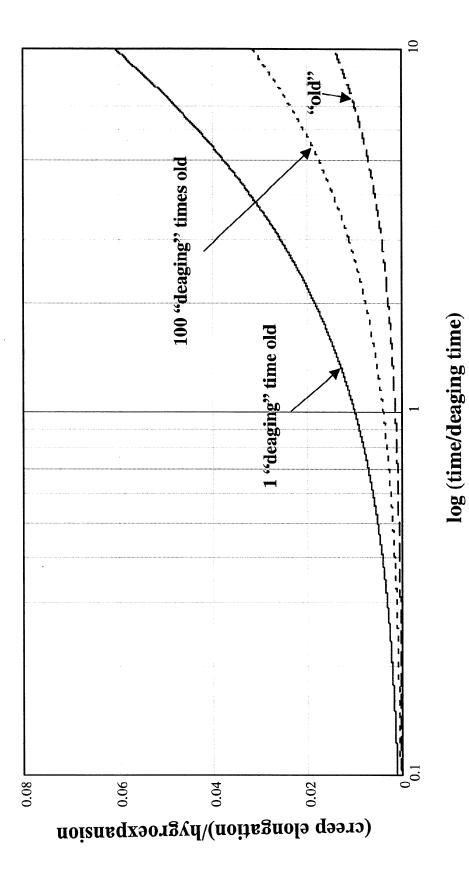


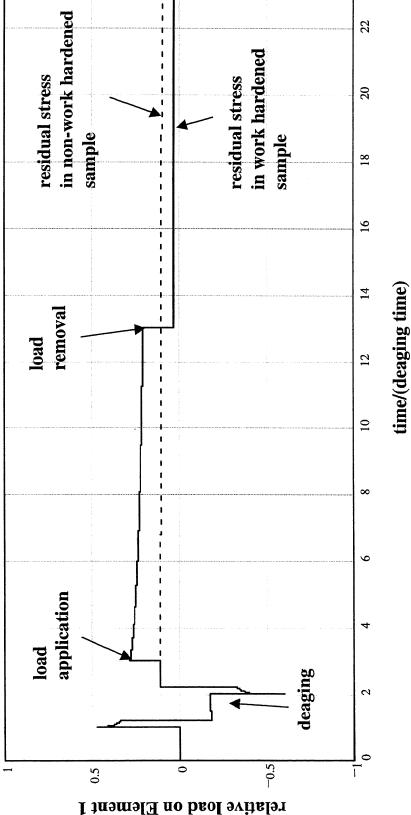


elongation/hygroexpansion



# Mathematical Model of the Creep Portion of a "Deaging" Experiment with Different "Aging" Times





## Mathematical Model of the Reduction in Residual Stresses by the Application of Load over Time