The mechanics of moisture accelerated tensile creep in paper

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ABSTRACT: Tensile creep of paper is accelerated in a variable relative humidity environment, but the causes of this nonequilibrium response to force and moisture are not well understood. A hypothesis is proposed for the physical mechanisms underlying moisture accelerated tensile creep of paper. This hypothesis, involving all levels of paper structure, is based on experimental results on paper and other polymers from the literature, and additional variable humidity, tensile, creep tests on handsheets and on unfibered cellophane.

KEYWORDS: Accelerated tests, cellophane, creep, creep tests, handsheets, humidity, moisture content, paper, tensile properties, variables.

Paper is most often used in a variable relative humidity environment. The interaction of load and moisture change in paper can cause many practical problems even under constant loads. These include compressive buckling collapse of the walls of corrugated cartons stacked in warehouses lacking humidity control, and the tensile creep of newly manufactured paper as the web is pulled through the press and dryer sections toward the reel. The mechanical response of paper to forces in a variable relative humidity climate is quite different from that in a constant relative humidity environment. The response of paper in variable relative humidity has been explored experimentally by several researchers, but no generally accepted explanation of the physical mechanisms underlying such behavior exists.

In particular it has been observed that, if during a creep test the relative humidity is cycled, the creep strain is larger than it would be in any constant relative humidity test after a given period of time. A constant relative humidity creep test at the maximum relative humidity in the cycle might have been expected to produce the largest creep strain at any given time. In this sense, the variation in the relative humidity accelerates the creep. Ever since Byrd *(1)* reported this behavior for paper in 1972, various explanations have been offered, none of which has been completely satisfactory. Here a new hypothesis is proposed for the underlying mechanisms in the case of moisture accelerated creep in which a constant in-plane tensile load is applied to a paper sheet but the ambient relative humidity is allowed to cycle over a large range. This hypothesis was developed from results in the literature and qualitative experiments performed by the author.

In the following sections, the creep experiments performed by the author are described, the hypothesis describing the underlying physical mechanisms which cause moisture accelerated creep is summarized, and the evidence for this hypothesis is presented.

The existence of moisture accelerated creep implies that one should not use, say, constant 90% RH creep as a worst case test in paper design. Better understanding of the underlying physics of this problem is required before useful design controls and descriptive mathematical models can be created.

Creep experiments

Moisture accelerated creep is shown by the three tensile creep tests in **Fig. 1**, one at a constant 30% RH, one at a constant 90% RH, and one as the RH cycled from 30% to 90% to 30% at a rate of 1/3% RH/min

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through four such cycles. In the latter, the load was applied at the initiation of cycling. The variation in relative humidity accelerates the creep, causing the tensile specimen to creep more than at a constant relative humidity equal to the highest relative humidity reached in the cycle. Large creep strains are obtained in these tests, often up to 4% without failure.

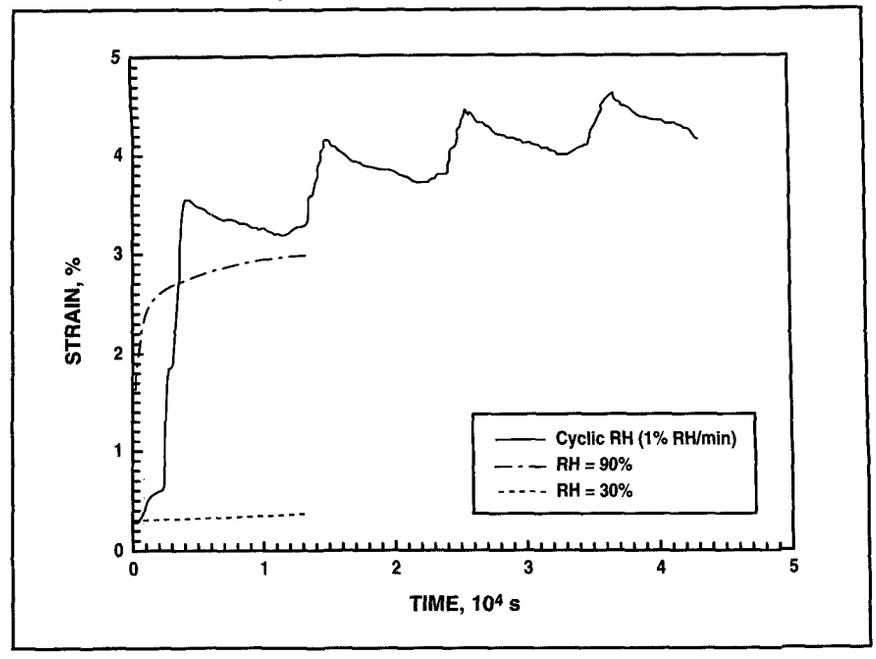
In all of the author's tests reported here, the paper tested was a southern hardwood handsheet for which the pulp was beaten to 208 mL Canadian Standard Freeness. The specimens measured 25.4-mm (1 in.) wide by 228.6-mm (9 in.) long. The initial length between the clamps was 177.8 mm (7 in.). The specimen thickness was about 0.1 mm (0.004 in.). This thickness is small enough that the moisture diffusion can be assumed to be nearly instantaneous.

Stress–strain tests determined failure loads of 117.4 N (26.4 lb), 99.6 N (22.4 lb), and 74.4 N (16.72 lb) at 30%, 50%, and 90% constant relative humidity respectively and at 23°C. Creep tests were performed at 25% of the failure load for a constant 50% RH 24.9 N (5.60 lb), a stress of approximately 9.65 MPa (1400 psi). This load was chosen to ensure that the specimen would pass through at least four relative humidity cycles before fracture. The specimens were conditioned for one day at 30% RH. The relative humidity cycles ranged between the initial 30% and a maximum of 90% at various constant rates between 1/4% RH/min and 1% RH/min at 23°C.

Experiments were conducted in a MicroTenny temperature–humidity chamber, measuring about 480 mm on a side. The humidity is sensed by a wet bulb–dry bulb system. The desired chamber humidity and temperature as a function of time are electronically controlled through an operator determined program.

The specimen is fastened horizontally between a fixed clamp and a clamp attached to a Microslide cross-roller bearing slide, which has a rated 0.003 coefficient of friction. This system is mounted on an aluminum frame which is placed inside the humidity chamber. The load is applied through a horizontal cable attached to the slide which is connected over a pulley system to a dead weight outside the sealed chamber. Flat pressure clamps with rubber liners on top and bottom are used to secure the specimen. A lip is machined on each clamp to help line up the specimen. A bubble level attached to the stand and leveling screws on the legs of the stand are used in conjunction with an adjustable dual pulley system through which the load is applied to ensure a horizontal load on the specimen. Deformation is read by an optical digital encoder which is accurate to 0.01 mm (0.0004 in.). The encoder has four emitter–detector pins and a glass diffraction grating to output a digital signal to a counter and then to a computer. Simultaneously, an output voltage from the chamber representing current humidity and one representing time is stored in the computerized data acquisition system.

1. Moisture accelerated creep under a variable relative humidity



Hypothesis for the physical mechanisms of moisture accelerated creep

A hypothesis is required which can explain the differing behavior in constant and variable relative humidity creep. Moisture accelerated creep appears in fibrous materials in which the fiber exhibits anisotropic swelling. The hypothesis must also account for the influence of microcompressions in the interfiber bonds and the observed changes in fibril angle. The success of empirical models for the creep of paper based on logarithmic functions must be explained.

A wood pulp fiber can be viewed as a composite of an amorphous hemicellulose and lignin matrix surrounding mostly crystalline cellulose microfibrils. The microfibril crystals are of finite length since they are periodically broken by amorphous cellulose regions along their length. The amorphous regions in the fibrils and the matrix are the sites of moisture sorption. In variable relative humidity, the unbalanced ingress and exit of moisture molecules and the increase in temperature due to sorption excite the matrix molecules into

nonequilibrium states. The molecules change their spatial configuration, opening intermolecular spaces. This process generates a large amount of free volume in the amorphous matrix within the pulp fiber. Such free volume generation within the matrix swells the fiber anisotropically in the direction transverse to the fiber axis due to the fiber's lamellar structure. The swelling may be further increased by lamellae separation.

The excessive transverse swelling in the fiber during a variation in relative humidity produces a large axially transverse force in the fiber. The strength of the transverse swelling force is increased by the fact that the matrix will not allow the fibrils to shear past one another to reduce swelling and lengthen the fiber. Lignin is highly crosslinked and therefore both stiff and shear resistent. The transverse force tends to increase the fibril angle, and a single fiber unconstrained by the paper fiber network shortens. This force is large enough to overcome the tensile force, induced in the fiber by the creep load, which tries to reduce the fibril angle.

In the paper sheet, the tensile force in each fiber increases since the interfiber bonds with crossing fibers in the sheet resist individual fiber longitudinal contraction.

This, in combination with forces induced by the mutual transverse fiber reswelling of crossed fibers, disrupts the interfiber bonds. These forces in each interfiber bond relieve the compressive stress maintaining the microcompressions, that the creep load alone could not release in a constant relative humidity environment. The prestresses causing microcompressions are created by transverse fiber contraction in the interfiber bonds as the sheet dries during manufacture.

This disruption of the interfiber bonds reduces the energy which must be supplied by the creep load to induce creep strain in the sheet. If the swelling and tensile load are large enough, the specimen creeps, a delayed elastic creep behavior involving release of the prestress causing the microcompression. This unstable prestress can be recovered by reducing the load and drying the sheet.

Each interfiber bond with its microcompressions requires a different energy to activate creep, in large part due to the different angles at which the two fibers forming the bond cross. The activation energy for each bond depends on its bonded area and the strength of the microcompressive stress in it. This large range of creep activation energies and the delayed elastic behavior accounts for the success of empirical logarithmic models for the mechanical behavior of paper.

The large creep strain increase in a variable humidity environment is due to motion in the interfiber bonds. Once the interfiber bonds are loosened by the free volume swelling induced release of microcompressions, there are three possibilities. At a low energy level the microcompressions merely unwrinkle, and slack produced by partial release of the microcompressions is taken up in the strain, but the integrity of the interfiber bond is maintained. There can be partial bond breaking of interfiber bonds at somewhat higher energy levels. At even higher energy levels, the fibers can fully separate at an interfiber bond and the bonded area can subsequently slip along the fiber. If the creep load is then reduced and the moisture is held constant, the bond reforms at a new location along the fiber as the specimen goes to equilibrium. This accounts for unrecoverable creep strains in moisture accelerated creep. Some of the strain is a delayed elastic strain recoverable by fibril angle reduction and reformation of microcompressions.

In contrast, during constant relative humidity creep, fiber elongation accounts for much of the strain. The tensile load reduces the fibril angle and dominates any transverse force in the fiber.

Evidence for the hypothesis

The hypothesis proposes that several supramolecular motions are involved in moisture accelerated tensile creep. The evidence will be presented beginning with the smallest structures.

Free volume generation

The fundamental difference between constant and variable moisture content tensile creep lies in the generation by variable relative humidity of excessive free volume within the amorphous regions of the fiber. In any polymer at thermodynamic equilibrium, the chain molecules tend to be closely packed. Free volume is generated between molecules when the material is in a nonequilibrium state. This free volume swells the material.

A wood fiber is a composite of polymers including cellulose, hemicellulose, and lignin. The fiber is built of layers composed of fibrils wound at an angle to the fiber's longitudinal axis. The fibrils are in turn made of microfibrils oriented along the longitudinal axis of the fibrils and bound in an amorphous matrix of hemicellulose and lignin. The microfibrils are composed of a crystalline state of long chain cellulose molecules in which the cellulose chains are believed to lie parallel to the longitudinal axis of the microfibrils. Since moisture sorption occurs only in the amorphous regions, and free volume cannot easily be generated in the highly ordered crystalline regions, the free volume is generated in the amorphous matrix.

When the relative humidity is constant, the moisture in a specimen is in kinetic equilibrium with the surrounding atmosphere. The specimen is subjected to a dynamic process of moisture molecules continually entering and leaving the specimen. This process is in balance so that there is no net change of moisture content for a fixed atmospheric moisture vapor pressure, i.e., constant relative humidity.

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The kinetic moisture balance is broken in variable relative humidity so that the specimen is in a moisture nonequilibrium state. Free volume may be generated by long chain molecules rotating away from their equilibrium configuration, by spaces left by exiting moisture molecules, or by spaces created by the entrance of moisture molecules.

Furthermore, moisture adsorption in cellulose is exothermic *(2)*. The change in specimen temperature as the moisture content either increases or decreases excites the molecular structure away from thermal equilibrium.

The point at which most free volume is generated is visible in creep tests of paper sheets. The largest change in total strain during a 30-90-30% cyclic relative humidity test occurs in the increasing relative humidity portion of the first cycle (Fig. 1). A similar pattern [*(3)*, Fig. 2], was observed when a tensile specimen was allowed to creep at a constant 30% RH, the conditioning relative humidity, for 2 h before beginning humidity cycling. The largest strain rate again coincided with the beginning of humidity cycling.

The rapid increase in strain in the first relative humidity cycle results from the immediate creation of most of the total free volume. During the first portion of the initial variable humidity cycle, the paper is in a dynamic nonequilibrium state because the humidity rate of change abruptly changes from zero during conditioning to a constant rate greater than zero. The humidity suddenly accelerates when the humidity cycle is begun. Subsequent constant rate change in humidity merely preserves the free volume already generated and prevents the paper specimen from relaxing to equilibrium.

Transverse fiber swelling is aided by free volume increase in amorphous regions between the circumferential lamellae in the S_2 layer, which results in their separation. The wood fiber cell wall is built of concentric lamellae composed of fibrils, as shown by the splitting of the S_2 layer into from two to ten layers by beating *(4)*. The possibility of lamellae separation is supported by Caulfield's *(5)* proposal that crosslinking between adjacent lamellae to prevent the entry of moisture and separation of existing hydrogen bonds would explain both the lack of swelling and the increased wet strength of formaldehyde crosslinked papers. Such binder crosslinks reduce the transverse swelling of the fibers. This fits with Back, Salmen, and Richardson's *(6)* independent suggestion, based on experiments with textile fibers, that a binder may reduce the moisture-accelerated creep effect in paper.

Anisotropic swelling and change in fibril angle

Moisture-accelerated creep is found only in fibrous materials whose fibers exhibit anisotropic swelling. Such a fiber swells more in the transverse than in the longitudinal direction of the fiber. The creation of free volume in the amorphous matrix induces transverse swelling which, in turn, causes a transverse force on the lamellae in the fiber. Increases or decreases in the fibril angle are a consequence of the competition of this transverse force and forces in the fiber produced by the creep load. Transverse swelling increases the fibril angle.

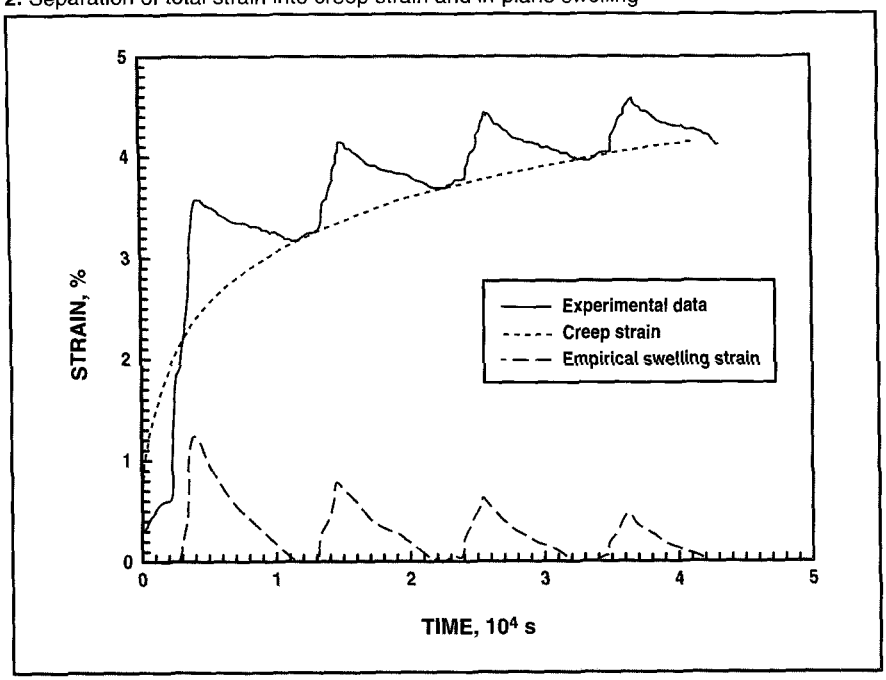
Byrd *(1)*, who provided the first experiments on the acceleration of tensile creep due to variable ambient relative humidity in paper, found that the fibril angle decreases in constant moisture but increases in cyclic relative humidity, tensile creep tests. The fibril angle, the angle between the fibril direction in the S_2 layer and the longitudinal axis of the wood fiber, typically ranges between 10° and 30°. Byrd found that in constant moisture content creep of his southern pine, bleached, kraft handsheets at 90% RH, the fibril angle decreased from 19.2° to 16.8°. However, for creep of sheets in an environment of relative humidity cycling between 35% and 90% RH, the fibril angle increased from 19.2° to 21.1°. These results are supported by Jentzen's *(7)* study of single fibers dried under a constant tensile load in which he concluded that the transverse shrinkage of the fiber causes a decrease in fibril angle. Hill's *(8)* X-ray diffraction measurements also showed that a constant tensile load on a dry pulp fiber decreases the fibril angle.

In Byrd's creep tests of a single fiber, the single fiber elongated under constant moisture content creep, but it shortened under variable moisture content creep. This result is consistent with the observed fibril angle changes. Also in variable relative humidity, an individual fiber shortened more than a fiber in a sheet, indicating that interfiber bonds prevent some contraction.

The transverse swelling force in variable moisture creep dominates the tensile force and increases the fibril angle. In constant relative humidity creep, there is little transverse force and the tensile force in the fiber decreases the fibril angle.

The role of the lignin and hemicellulose matrix in the fibril angle increase is suggested by comparing the behavior of Kevlar®. Wang *et al. (9)* have shown that Kevlar fibers also swell anisotropically and that sheets made of Kevlar exhibit moisture accelerated creep. In contrast to paper, Kevlar is not a composite. Kevlar is nearly 100% crystalline. According to Ericksen *(10)*, Kevlar 29 and 49 fibers consist of a system of sheets composed of a nearly axially oriented fibrillar crystallite structure, in which the fibrils are tied together by other molecules. The crystallites, which are from 6 nm to 18 nm long, are bundled in a mutually parallel orientation into the fibrils, much as in cellulosic fibers. The sheets in Kevlar lie in a plane containing the fiber axis, while the lamellae in wood pulp lie in circumferential sheets around the fiber axis.

2. Separation of total strain into creep strain and in-plane swelling



Ericksen proposed that creep in Kevlar fibers is due to crystallite rigid body rotation with respect to the fiber axis, permitted by shear along the crystallite boundaries. The amount of shear depends on the motion of hydrogen and van der Waals bonds along the crystallite boundaries. There may be voids or amorphous material in the boundary regions. The variable relative humidity causes free volume generation on these boundaries and facilitates creep. The tensile creep force aligns the crystallite fibrils closer to the fiber axis since no matrix, as in paper, restricts this motion, and therefore the Kevlar fiber lengthens during moisture accelerated creep. In a wood fiber, such shear is prevented by the matrix.

Microcompressions and delayed elasticity

Variable relative humidity creep strain is accelerated by the interaction of transverse fiber swelling with microcompressions in the interfiber bonds, which partially reverses the drying process. This behavior is reflected in the successful logarithmic creep models which, in turn, indicate that delayed elasticity is involved.

Page and Tydeman *(11)* showed that interfiber bonds form before the web shrinks in drying. Page's *(12, 13)* microscopic observations of the interfiber bonds show that as the paper dries, each swollen, wet fiber contracts anisotropically with up to 30% transverse shrinkage, but only 1–2% longitudinal shrinkage. This transverse contraction of a crossing fiber induces compressive stresses in the contact area of the already formed bond between two fibers and wrinkles the fiber with the least compression resistance in the bond area. The wrinkles are approximately transverse to the fiber longitudinal axis. The formation of such microcompressions can cause from 2% to 15% in-plane shrinkage in an unrestrained sheet. The microcompressions form sites of unrelaxed stress in the fibers in a dried sheet.

Delayed elastic strain is due to stress induced relaxation from an unrelaxed state. The relaxed state can be returned to the original unrelaxed state by a "back stress" from the surrounding material when the load is removed, and some of the creep strain is recovered. This concept was originally used to describe the creep behavior of amorphous glasses. The unrelaxed state might be represented by a relative minimum of an energy function from which the material can be removed by a stress which draws it over an energy barrier into a lower minimum. The process, therefore, depends on an activation energy. Argon *(14)* showed that, in delayed elasticity, if the activation energies involved vary over a large range and if a nearly equal number of sites have each activation energy, then the strain of the bulk material is a logarithmic function of time. Several researchers have independently fit creep data for paper by logarithmic models.

To apply these ideas to variable moisture-accelerated creep, the creep and hygroexpansive components of the total experimental in-plane axial strain must be separated. Byrd *(1)* measured the relation between in-plane swelling and moisture content. During his creep tests, he measured the current moisture content and then subtracted the corresponding swelling to determine the creep strain component at any given time. Byrd fit the creep portion of the strain in his tests by a logarithmic equation of the following form:

$$\varepsilon = a + b \ln (t)$$

Haslach, Pecht, and Wu *(15)* used a different method of determining the creep portion of the total strain since the moisture content during their tests was not measured. On every cycle, when the relative humidity returned to the conditioning relative humidity of 30%, the strain at that time was assumed to be only creep strain. A logarithmic curve was fit through the strain values at 30% RH. At any time, this logarithmic curve was subtracted from the total strain to determine the hygroexpansion in the load direction (**Fig. 2**). Even though this approach is in effect the opposite to that of

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Byrd's, both methods produce a logarithmic mathematical model for the creep component of strain.

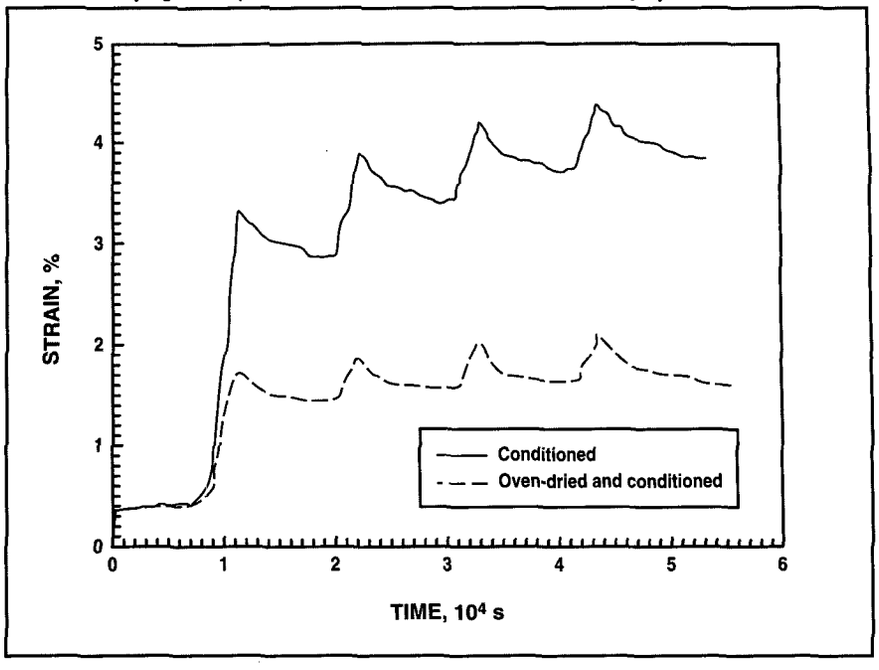
Also, Pecht and Haslach *(16)* fit the stress-strain data of Gunderson, Considine, and Scott *(17)* at various constant relative humidities with a viscoelastic model involving a logarithmic function. Pecht *(18)* had previously used a logarithmic model for Brezinsky's *(19)* constant moisture content creep data. The success of a logarithmic model fit to moisture–load interactions in paper is evidence that delayed elasticity is involved in paper behavior.

The microcompressions in the interfiber bonds are the most likely physical basis for this fit to moisture-accelerated creep strain. The stress which relaxes during delayed elastic creep is the compressive stress in the microcompressions. The creep activation energy is that required to deform the interfiber bonds in the paper sheet which deformation, in turn, produces specimen elongation. Since the interfiber bonded area varies over a large range due to the arbitrary crossing angles and since the network restraints on each interfiber bond also vary greatly from fiber to fiber, there must be a large range of creep activation energies within the paper sheet.

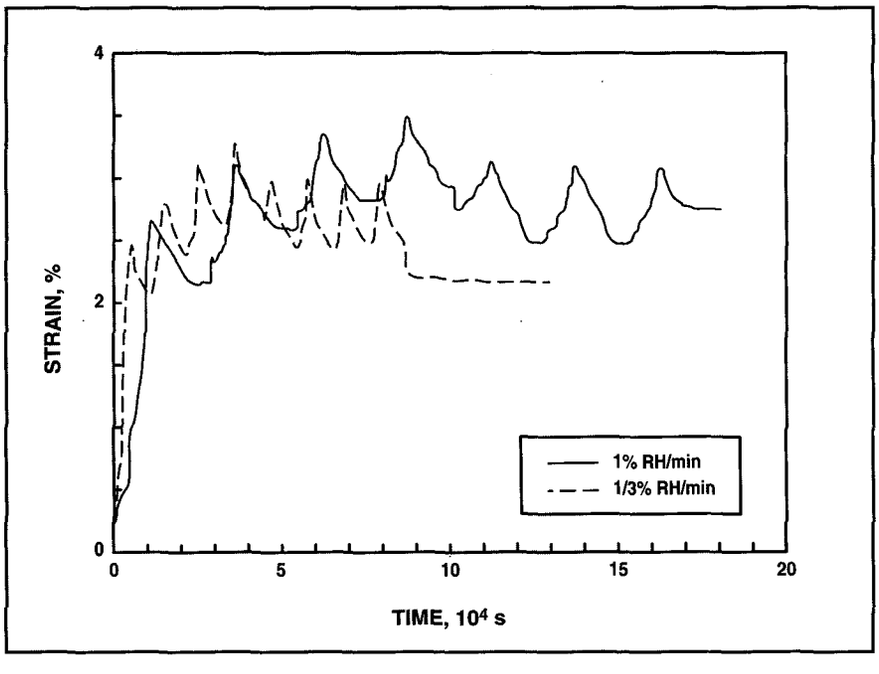
Some creep strain can be recovered by oven drying the specimen. After a specimen was exposed to four variable relative humidity cycles, it was oven dried at 105°C, and then again subjected to four variable relative humidity cycles (**Fig. 3**). The oven drying caused some of the microcompressions to reform. That the specimen shows the same qualitative behavior in the second set of cycles, with a large strain increase in the first cycle, supports the idea that the microcompressions play a key role in moisture accelerated creep.

Creep strain can also be recovered by reducing the load on the specimen in the absence of any other operation on the specimen (**Fig. 4**). When the load was cut in half after the first four cycles, the strain was of constant maximum magnitude in each cycle and therefore due only to hygroexpansion. There was no longer any creep strain since the load did not apply enough energy to the bonds to activate creep. The load was then completely removed and the relative humidity held constant. Over time a large portion of the total strain was recovered, a delayed elastic strain.

3. Effect of drying a sample at 105°C between two sets of humidity cycles



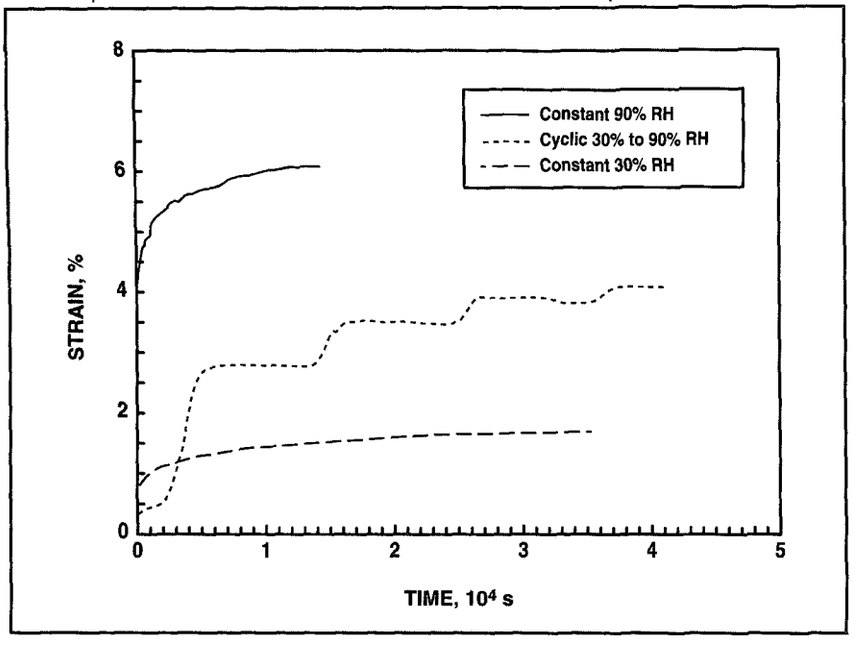
4. Effect of load reduction during a cyclic relative humidity creep test



Interfiber bond behavior

The large magnitude of the moisture accelerated creep strain is due

5. A cellophane sheet does not exhibit moisture-accelerated creep



to interfiber bond disruption. That the interfiber bonds deform can be deduced from Byrd's measurements of the bonded area. Using the ideas of Nordman *(20, 21)*, Byrd *(1)* experimentally related light scattering from the paper surface to the behavior of the interfiber bonds under moisture influenced creep. The light scattered from a paper sheet is assumed proportional to the unbonded area in the sheet. Constant relative humidity creep had a small effect on the light-scattering coefficient from the sheet. The scattering coefficient decreased about 2% at 0.7% strain for the lowest load, and increased back to its original value at higher loads. This suggests that for low creep loads under constant relative humidity, the reduction in the proportion of bonded area might be due to an increase in the total area by fiber lengthening. At the higher loads, since the net change in unbonded area is nearly zero, such fiber lengthening might be balanced by some unwrinkling of microcompressions and partial bond fracture.

Byrd *(1)* reported that cycling the relative humidity during creep increases the scattering coefficient at low strains. Higher loads resulted in a slight reduction in scattering coefficient. For the 40% of maximum load, the scattering coefficient increased by about 7% at 1.1% strain. At the higher loads there is less of a scattering coefficient increase than at the lower loads for cyclic relative humidity. In fact, at 70% load, Byrd reports that the change in scattering coefficient was nearly zero but the final strain was 2.4%.

A mechanism to explain this pattern is that the release of microcompressions due to transverse swelling increases the bonded area. The new bond area is unaffected by low creep loads. At higher creep loads, the bonds completely broke, the bonding area slid to a new position and the bond reformed. This is supported by the creep behavior of individual fibers. Under constant moisture content, the creep strain of a single fiber increases logarithmically with respect to time. But under cyclic relative humidity, the single fiber length at the maximum relative humidity in the cycle is smaller after the first cycle. Then after several cycles it remains constant *(1)*. In a sheet, the interfiber bonds resist any individual fiber contraction. But, in combination with the higher creep load, the attempt of the fiber to contract induces tension in the fiber and shear in the bond which, after the bonded area breaks free, causes that contact area to slide along the fiber. In later cycles, the fiber no longer tries to contract, allowing an interfiber bond to reform at the new position. The measured change in unbonded area, and thus scattering coefficient, would then be very small.

Hygroexpansion and moisture accelerated creep

Further evidence for the role of swelling in the interfiber bonds lies in the behavior of the hygroexpansive strain accompanying moisture accelerated creep. Soremark and Fellers *(22)* found accelerated creep in corrugated board with kraft liners in small strain bending under a cyclic relative humidity environment (40–80% RH). They found that tensile stresses reduce hygroexpansion over multiple cycles, while compressive stresses increase it.

The separation of strain into creep and hygroexpansive components by Haslach, Pecht and Wu *(15)* also shows that the hygroexpansive strain decreases in subsequent relative humidity cycles in a tensile specimen (Fig. 2). The tensile load "uses up" in creep some of the strain that would be available for swelling at any particular time. On later cycles, hygroexpansion appears damped by the load. Gunderson *(23)* reported that the presence of a load on a paper specimen does not significantly affect the amount of moisture adsorbed; therefore, a reduction in moisture sorbed is not the explanation. Both hygroexpansion and moisture accelerated creep must involve the same physical mechanism, most likely the deformation of the microcompressions in the interfiber bonds.

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Nonfibrous cellulose sheets

The importance of the fibered network structure of paper is shown by large strain creep tests performed on cellophane by Haslach and Wu *(24)*. These tests are in contrast to the small strain sonic tests performed by Berger and Habeger *(2)* on cellophane to investigate the polymeric properties. Samples of 0.254-mm thick cellophane were subjected to a constant dead load at a constant 30%, a constant 90%, and four cycles between 30% and 90% RH at a rate of change of 1% RH/min. During the cycles, the relative humidity was held constant for 60 min each time it reached 30%. The temperature was 23°C. The specimen was conditioned first at 30% RH for 24 h to allow it to come to moisture equilibrium. The dead load (500 g) used in these tests was one sixth of the failure load in a uniaxial stress-strain test at 50% RH. The moisture content was not measured during the test.

The cyclic relative humidity tests of cellophane did not exhibit significant accelerated creep, even though cellophane is made of cellulose. **Figure 5** shows superimposed constant 30%, constant 90%, and the 30–90% cyclic relative humidity creep curves in which the cyclic curve lies between the two constant relative humidity curves. The strain measured experimentally is due to both hygroexpansion and creep. This result verifies that the network structure of paper is a significant factor in moisture accelerated creep. Free volume is generated in the cellophane since there is a rapid increase in creep during the increasing portion of the initial 30–90% cycle on the cyclic creep curve. Strain is recovered in cellophane when the load is reduced due to molecular relaxation and consequent loss of free volume.

Comparison to other hydrophilic materials

All cellulosic materials, including wood, cotton, and wool, exhibit moisture accelerated creep. However, drawn nylon fibers are hydrophilic, but nylon sheets do not exhibit moisture accelerated creep. For the cyclic relative humidity tests on nylon 6,6 fibers reported by Wang *et al.* [*(9)*, Fig. 8], the creep curve oscillated between and touched the boundaries formed by the two creep curves at the constant relative humidities defining each extreme of the cycle. Creep in drawn nylon fibers may be due to the breaking of hydrogen bonds in the amorphous region by moisture and stress and subsequent molecular motion. A drawn nylon fiber does not have a fibril or lamellar structure which allows for anisotropic swelling.

Wood exhibits moisture accelerated creep. Some have proposed that this is due to the creation of slip planes within the wood structure. Creep would be accelerated by the creation of free volume along these planes.

In each of the materials showing moisture accelerated creep, free volume causes swelling which, in turn, facilitates slip of substructures. The slip mechanism differs in each material depending on how the fibers are bonded to each other. ■

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