THE PRACTICAL INFLUENCE OF HETEROGENEITY ON TENSILE ACCELERATED CREEP IN PAPER

Douglas Coffin and Chuck Habeger

ABSTRACT

According to theoretical work done by the authors (5,6), sheet heterogeneity can affect accelerated creep in paper. Experiments were conducted to see whether these influences have any impact on everyday papermaking decisions. We report that single-ply sheets made from furnishes of mixed fiber type do not experience significant additional accelerated creep. However, we conclude that a sheet formed from plies of different furnishes can exhibit extra accelerated creep because of a mismatch in hygroexpansivity. An attempt to create fiber microcompressions by Hobart mixer treatment did not significantly change the accelerated creep performance.

In addition to the analysis of heterogeneity, we argue with appeal to experiment that accelerated creep depends on the ratio of sorption time to moisture cycle time.

Application:
Papermakers should be aware that layering different furnishes in multi-ply board can contribute to the premature failure of the product under the influence of load and cyclic humidity. A similar concern is not raised for paper made from blends of different furnishes.

INTRODUCTION

Accelerated creep is an established phenomenon in wood (1), paper (2), and other materials (3,4). Hydrophilic materials generally creep faster at a high relative humidity (RH) than at a low RH. However, under the proper circumstances, creep can be greater in an environment that is continually cycled between low and high RH levels than when it is held at the high humidity extreme. When this is observed, the material is said to experience accelerated creep. In **Figure 1**, the results of a typical accelerated creep experiment are plotted in terms of creep strain versus log time. The sample was maintained under tensile load for three hours at 80% RH. Then, keeping the load constant, the humidity was cycled at one-hour intervals between 80 and 30% RH. Notice that the creep under cyclic conditions is far above the time-projected constant 80% RH creep curve.

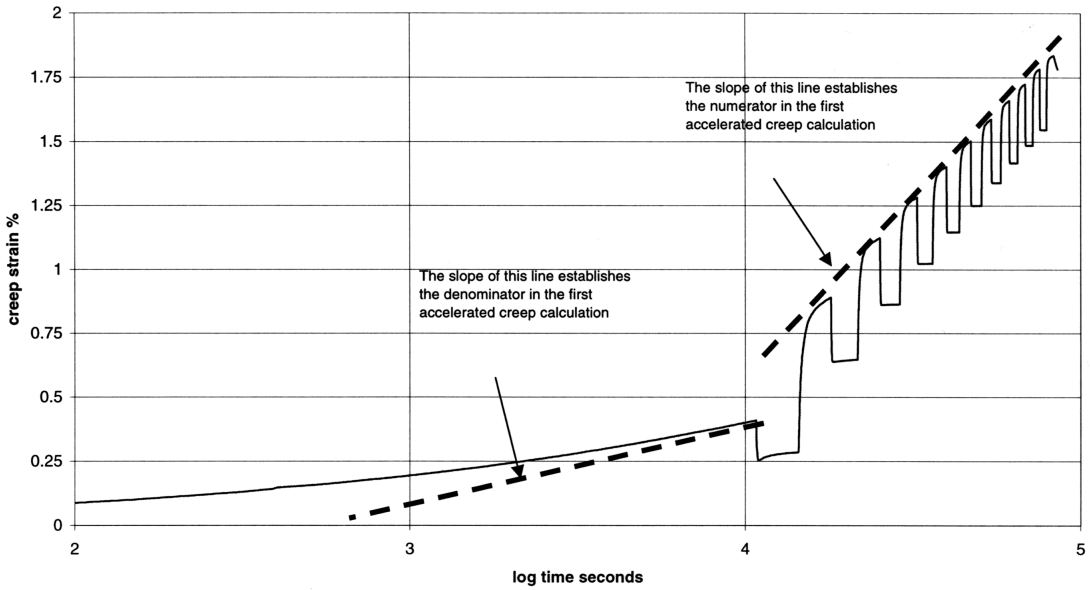


Figure 1. 80-30% RH accelerated creep of sample A at 25% of 50% RH breaking load.

The driving mechanism for accelerated creep is a contentious subject (5). We advocate a cyclic moisture-induced cyclic stress explanation (3,5,6,7). For us, the driving force for accelerated creep is the localized cyclic loading caused by moisture cycling. During cyclic sorption, a portion of the sheet experiences elevated stresses levels while the stress in other portions is depressed. We demonstrate that materials, which undergo accelerated creep, also experience more creep in cyclic load testing than they would if held at a constant, average load. We argue that accelerated creep is the result of this nonlinear creep constitutive behavior and the localized load cycling produced in cyclic humidity environments. The differential swelling induced by moisture gradients is one clear source of load cycling. In our view, sheet heterogeneity in response to moisture could be another cause for non-uniform loading. We undertook the experimental work reported below to assess whether "heterogeneity-driven accelerated creep" is of practical interest to papermakers.

To envision the heterogeneity type of accelerated creep, imagine adjacent structural elements in the paper that are parallel to the direction of the applied load. If these elements have different moisture sensitivities, stresses will redistribute during sorption, and moisture cycling will lead to out-of-phase load cycling of each element and in turn to extra creep. If this extra creep overcompensates for the low creep rates at low moisture content, a sample in a cycling moisture environment can creep more than another sample held at the highest moisture content (5,6). As an example, consider a sheet made of two separate plies under a constant tensile load. Let the hygroexpansion of one of the plies be greater than the other ply. As the sheet absorbs moisture, one side attempts to expand more than the other. Part of the tensile load is shifted to the less hygroexpansive ply. Upon desorption, the reverse occurs: the more moisture sensitive ply carries most of the tensile load. During moisture cycling, the loads on each of the two plies cycle out-of-phase. This same kind of action might also take place, but on a fiber level, if fibers of differing hygroexpansivity are bonded together. Wood fibers expand at least an order of magnitude more laterally than axially with the uptake of water (8); thus identical fibers aligned crosswise will also cycle their load sharing in a cyclic humidity chamber. In a like manner,

heterogeneity in the moisture dependence of stiffness and creep compliance can also generate load cycling and accelerated creep.

Three types of heterogeneity (through-sheet heterogeneity, fiber-to-fiber heterogeneity, and fiber-level heterogeneity due to fiber anisotropy) are therefore possible candidates for initiating load cycling and accelerated creep. We decided to conduct experiments that would shed light on the importance of these mechanisms and on the degree that papermakers can influence accelerated creep by altering heterogeneity parameters. To do this, we procured furnishes that produced papers which responded differently to changing relative humidity. We formed blended sheets and multi-ply sheets of furnishes composed of fibers of different hygroexpansivity and compared their accelerated creep performance to that of sheets made of a single fiber type.

We wanted to work with common fibers having significant differences in hygroexpansion. Although drying restraint has a major influence on hygroexpansion (9), we cannot expect different sections of our samples to have different drying histories; therefore, we could not reasonably select drying restraint as a variable for our accelerated creep experiments. Hygroexpansion, defined as change in percent length divided by change in percent moisture (10), depends on furnish (11). Mechanical pulps shrink less than low yield chemical pulps during drying, and the dimensional stability of mechanical pulps does not benefit as much from restrained drying (9). Fully restraint-dried TMP sheets are about 10% more hygroexpansive than fully restrained bleach kraft sheets (9). In our accelerated creep testing, we are changing relative humidity; therefore, we are interested in the change in length induced by changes in relative humidity. For a given change in relative humidity, sheets made from mechanical pulps undergo a 10% greater change in moisture content than sheets made of chemical pulps. Therefore, we concluded that fully restrained sheets formed from mechanical and chemical pulps would be different in their RH-based hygroexpansions, and that these differences would be representative of those encountered in papermaking. We investigated the influence of through-sheet heterogeneity by testing samples made by wet pressing plies of different furnish combinations and then drying them under restraint. Fiber-to-fiber heterogeneity was assessed by comparing the performances of single-ply handsheets produced from blended furnishes with those formed from pure furnishes.

We have argued (5,6) that the anisotropy in a wood fiber's response to moisture leads to a fiber-level heterogeneity in the sheet that contributes to paper accelerated creep. We experimentally address the influence of fiber anisotropy by looking at the accelerated creep of sheets made of fibers treated to be less anisotropic. Wood fibers are filament-wound structures of crystalline cellulose mircofibrils held together by a lignin-hemicellulose matrix. They are stiff and moisture insensitive in the axial direction and compliant and hygroexpansive radially. High-consistency mixing curls and microcompresses wood fibers (12). The curl, but not the microcompressions, can be removed by subsequent low consistency treatment. The microfibrillar structure is buckled and folded at the microcompressions. This preferentially reduces axial stiffness and increases axial moisture sensitivity. As suggested by Derek Page, we used a Hobart kitchen mixer (13) as a means to induce microcompressions and reduce fiber anisotropy.

SAMPLES

We chose a newsprint TMP and a bleached softwood kraft pulp as the test furnishes for our blend-ply experiments. These furnishes were received as never-dried pulps, and we did no refining. After removing the fines, the freenesses were measured as 721 CSF for the BKS and as 767 CSF for the TMP. Sheets of 60 g/m^2 were formed on a Noble and Wood 8-inch square handsheet mold. The sheets were wet pressed (individually and in stacks of four) at a common pressure of 50 psi for 5 minutes. All sheets were restraint-dried on a static drum drier. Single-ply handsheets were made from the TMP furnish (sheets A), the BKS furnish (sheets B), and a 50-50 retained-on-the-wire blend of the TMP and BKS fibers (sheets AB). The D sheets were formed from the BKS pulp after it was processed in the Hobart mixer at 20% consistency for two hours followed by mixing in a British Disintegrator at 1.2% consistency to remove curl. Four-ply handsheets were formed with TMP plies (sheets AAAA), BKS plies (sheets BBBB), two TMP plies in the middle and a BKS ply on each side (sheets BAAB), and two BKS plies in the middle and a TMP ply on each side (sheets ABBA).

We used the variable-humidity creep tester (5,6) to measure hygroexpansion of the samples between 30 and 80% RH. We applied a tensile load of 100 grams (400 grams for four-ply samples). This was large enough to straighten the samples, but it was insufficient to cause appreciable elastic or creep strain. The 100-gram load supplies only about 5% of the ultimate tensile load of the weakest sample. After conditioning with several humidity cycles, we recorded the strain difference between 80 and 30% RH as the hygroexpansion. With this technique, the standard deviations in the hygroexpansions were about .01%. Pertinent average physical properties are provided in **Table I.**

Sheet Type	Density	Basis Weight	Tensile Strength	Stretch	Specific Longitudinal Sonic Modulus	Hygroexpansion 30-80% RH
	g/cm^3	g/m^2	kN/m	%	km^2/s^2	%
A	0.314	61.9	1.84	1.6	4.98	0.39
B	0.317	61.9	0.68	1.1	3.95	0.32
AB	0.305	59.6	1.41	2.0	4.81	0.36
D	0.491	60.3	1.69	2.4	6.79	0.35
AAAA	0.332	254	8.41	2.7	5.23	0.34
BBBB	0.356	250	3.36	1.9	4.84	0.30
ABBA	0.333	256	5.58	2.2	4.94	0.32
BAAB	0.347	258	5.81	2.5	4.73	0.33

Table I. Physical properties of samples.

ACCELERATED CREEP TESTING

Notice that there is a considerable difference in tensile strength between samples. In previous testing, we observed that accelerated creep depends on tensile load. Accelerated creep measurements could be compared at the same nominal load or at the same portion of tensile strength. Each approach has its advantages, so we decided, in all cases, to do it both ways.

In conformation to convention (5,6), we normally quantify accelerated creep as the ratio of the creep versus log time slope taken after and before humidity cycling begins (see Figure 1). In truth, for these paper samples, this decision is arbitrary and warrants reevaluation. First consider more closely the present method. Figure 1 is a representative accelerated creep plot. Accelerated creep is taken as the ratio of the slope of the line tangent to the local maximums of the cyclic creep curve to the slope of the constant humidity creep curve in the early part of the experiment. Notice that the construction in the cyclic humidity portion of the experiment forms an almost straight line, whereas the slope of the creep in the initial high-humidity regime is not constant. That is, at these levels of loads and humidity cycling, creep is log linear under cyclic humidity, but it is not in the initial wet state. Therefore, the process of slope determination is not obvious for the wet state. We have arbitrarily defined it as the tangent to the creep line just prior to the beginning of humidity cycling. Other materials, such as Kevlar fibers and Nylon 6,6 (4,5,6), show log time linear creep in both regimes, and this determination of accelerated creep is more consistent.

At relatively small loads and short times, creep in paper follows the power law; it only becomes log linear at long times or high loads (14) or, as we have demonstrated, under cyclic humidity. Power law creep forms a straight line when log creep is plotted versus log time. The same data as in Figure 1 is plotted in **Figure 2**; however, the logarithm of creep strain is now on the y-axis. This time, the wet-state creep is linear, and the cyclic humidity portion is nearly linear. We could have just as logically used the log-log plot to define accelerated creep. Notice, from Figure 1 or Figure 2, that a significant portion of the extra elongation induced by moisture cycling comes during the first rewetting cycle. Both of these accelerated creep calculations ignore this part of the creep. They take the cyclic humidity slope between peaks, after the first rewetting. This is arguably inappropriate. For paper, creep rates fall as creep increases. A sample that has a large response upon first rewetting will have a smaller subsequent creep rate. A calculation that doesn't take this into account could misrepresent relative accelerated creep rates. A third alternative is to use the slope of the line between the beginning and the end of the cyclic humidity strain of the log-log plot as the numerator in the accelerated creep calculation (see Figure 2). This slope clearly depends on the length of the test. However, it does account for all the first rewetting creep, and we used a common humidity cycling history for all tests. We assessed accelerated creep using all three methods, but although it is somewhat arbitrary, we have decided to report the results using the third definition. The final conclusions drawn are obtained on values averaged from the results of all three methods, but are also evident in the reported results from the third method.

We did the accelerated creep testing in a controlled-environment, tensile creep tester described previously (5,6). The samples were mounted in the chamber and equilibrated to 80% RH. The tensile load was applied and held for three hours, at which time the RH was cycled between 30 and 80% RH at one-hour intervals. The sample lengths were monitored, and creep strain was determined as a function of time. We made from two to four replications (generally three) of each test and averaged the results. The standard deviations in the accelerated creep numbers were about 0.1. The accelerated creep numbers for A, B, and AB at 25% of the sample's 50% RH tensile strength and at 25% of the 50% RH strength of sample B are presented in **Table II**. Also listed are the creep rates during cyclic moisture in terms of percent creep per decade of time after initiation of load.

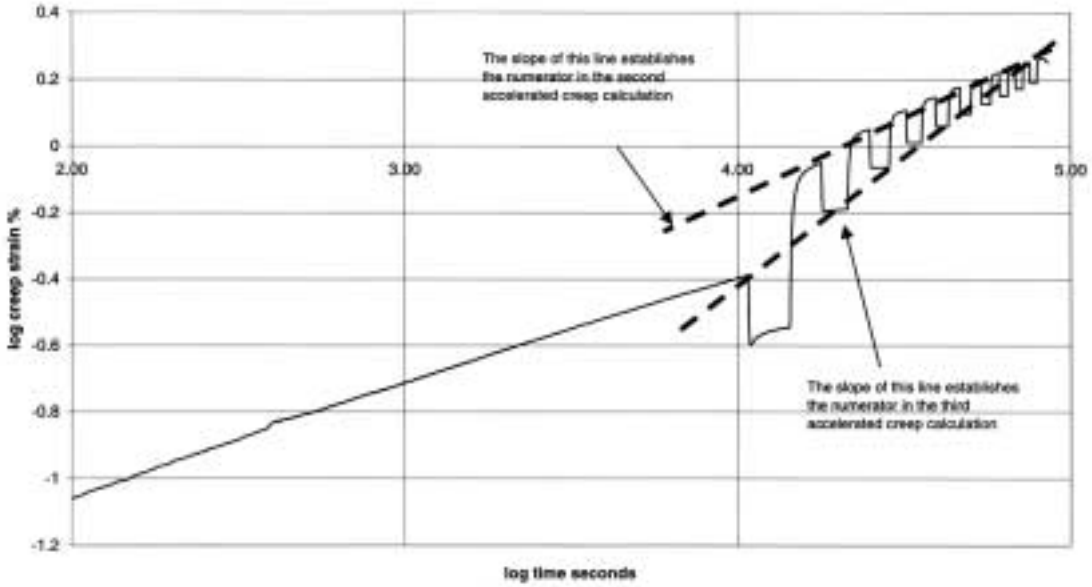


Figure 1. 80-30% RH accelerated creep of sample A at 25% of 50% RH breaking load.

Sheet Type	Accelerated Creep at 25% of Sample's Tensile Strength	Accelerated Creep at 25% of Sample B's Tensile Strength	Cyclic Creep Rate at 25% of Sample's Tensile Strength %/decade	Cyclic Creep Rate at 25% of Sample B's Tensile Strength %/decade
A	2.37	2.86	1.77	0.51
B	2.26	2.26	0.50	0.50
AB	2.34	2.52	1.31	0.54

Table II. Blend accelerated creep results.

As in the blend testing, we made multi-ply accelerated creep comparisons at the same load and at the same portion of tensile strength. In addition to the 25% load work, we did test at 35% of the tensile strength of BBBB at 50% RH. The results are listed in **Table III**. The cyclic moisture creep rates in percent per decade are stated in parenthesis after the accelerated creep numbers.

Sheet Type	Accelerated Creep and cyclic creep rate at 25% of sample's tensile strength	Accelerated Creep and cyclic creep rate at 25% of sample BBBB's tensile strength	Accelerated Creep and cyclic creep rate at 35% of sample BBBB's tensile strength
AAAA	1.85 (1.42)	1.96 (0.53)	2.03 (0.83)
BBBB	2.13 (0.56)	2.13 (0.56)	2.08 (1.06)
ABBA	2.07 (1.22)	2.34 (0.62)	2.27 (0.85)
BAAB	2.38 (1.11)	2.55 (0.60)	2.38 (1.05)

Table III. Multi-ply accelerated creep results.

The comparisons between accelerated creep coefficients and cyclic creep rates with and without treatment in the Hobart mixer follow as **Table IV**.

Sheet Type	Accelerated Creep at 25% of Sample's Tensile Strength	Accelerated Creep at 25% of Sample B's Tensile Strength	Cyclic Creep Rate at 25% of Sample's Tensile Strength %/decade	Cyclic Creep Rate at 25% of Sample B's Tensile Strength %/decade
B	2.26	2.26	0.50	0.50
D	2.21	2.31	1.05	0.31

Table IV. Treated accelerated creep results.

DISCUSSION

We did achieve (see Table I.) a moderately higher hygroexpansion in the TMP (A) sheets than in the bleached kraft softwood (B) sheets. This gave the hoped-for opportunity to assess the role of moisture sensitivity heterogeneity in accelerated creep. We think that accelerated creep is caused by load cycling that is driven by humidity cycling. All other things being equal, we expect the more hygroexpansive sheet to exhibit more accelerated creep. Notice from Table II that there was more accelerated creep recorded for the A sheets than the B sheets. In all, we made six (three ways to determine coefficients at two loads) different comparisons between accelerated creep of the A samples and the B samples. In all cases, the A samples exhibited more accelerated creep. The average ratio of A to B accelerated creep is 1.23 with a standard deviation of 0.28.

In Table II and in three of the four other comparisons, the accelerated creep numbers for the TMP-BKS blend sheets were intermediate between the pure TMP and pure BKS. The average of AB accelerated creep divided by the mean of the A and the B accelerated creep is 0.99 with a standard deviation of 0.04. Clearly, heterogeneity due to fiber type is not producing a noticeable influence on accelerated creep. This observation supports the premise that the major part of the influence (if any) of fiber level heterogeneity on cyclic moisture induced load cycling comes from fiber anisotropy rather than from fiber variety. The axial to radial difference in fiber properties dwarfs the difference between fiber types. Therefore, we should not expect the overall level of load cycling to be significantly increased by the introduction of relatively small fiber-to-fiber differences.

Comparing Tables II and III you will find that we observed different accelerated creep rates in the four-ply than in the single-ply sheets. This is because of the selected cycle time. Accelerated creep depends on the ratio of sorption time to cycle times (5,6). If moisture cycles too rapidly in relation to sorption time, moisture gradient cycling will not penetrate the sheet and through-sheet load cycling will not occur in the interior of the sheet. On the other hand, if the moisture content is maintained at the extremes for too long, moisture gradients and stress concentrations will be established for only small portions of the total creep time, and accelerated creep will be attenuated. When cycle time and sorption time are of the same order of magnitude accelerated creep is maximum. This is demonstrated in **Figure 3**. Here an accelerated creep test was conducted on A and AAAA sheets. To begin with, the cycle times were only 20 minutes long. After four cycles the cycle time was increased to 80 minutes, and four more cycles later it was increased to 320 minutes. In the beginning, during rapid cycling, the faster sorbing A sheets experienced greater accelerated creep. However, by the time of 320 minute cycles, the A sheets are sorbing too rapidly relative to the cycle time, and the creep in the AAAA sheets overtook that in the A sheets. Because of the effect shown in Figure 3, accelerated creep comparisons between the single-ply and four-ply sheets based on the results shown here are meaningless.

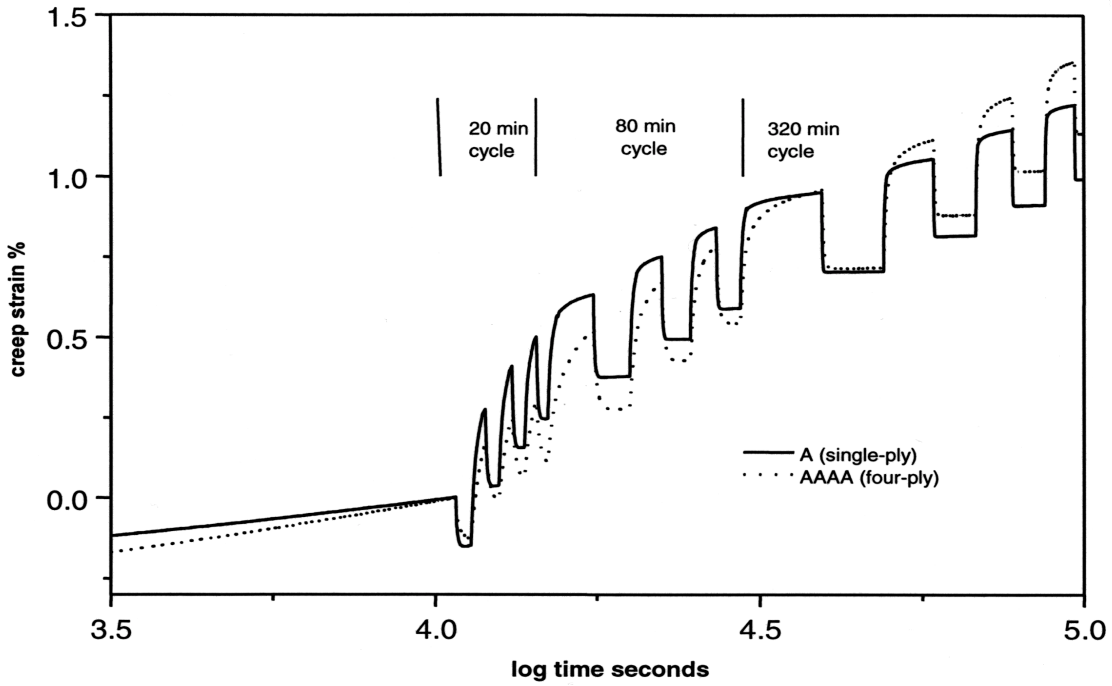


Figure 3. Accelerated creep for sample A and AAAA for three cycle times.

The construction of through-sheet heterogeneity by wet pressing TMP and BKS sheets together did influence accelerated creep. In all, but one (35% of BBB strength using the standard, liner-creep determination), of the nine (three methods at three loads) cases, the mixed-ply sheets had greater accelerated creep than either of the four-ply sheets made of sheets of the same furnish. The average of the sum of the mixed-ply sheets to the sum of the pure-ply sheets is 1.12 with a standard deviation of 0.07. From our perspective (5,6), the added multi-ply heterogeneity caused enough extra load cycling to perceptibly augment accelerated creep. A good example of this effect is given in Table III. At a loading level of 25% of the BBBB tensile strength, the creep rate per decade and the accelerated creep ratio were fairly well matched for the pure-furnish, multi-layer sheets, but the mixed ply sheets had faster creep rates. This is a clear case where producing a mixed furnish sheet lead to worse performance than sheets made of either pure furnish.

Anisotropy in fiber hygroexpansion is a possible contributor to accelerated creep in paper (5,6). We conjectured that, if this is major factor, microcompression of unrefined pulp by Hobart mixer treatment might reduce this anisotropy and thereby reduce accelerated creep. The comparison of B and D sheets is an attempt to assess this hypothesis. The three coefficient average ratio of D to B accelerated creep is 0.97 with a standard deviation of 0.09. Thus, we detected no large effect. The comparison is complicated by the fact that the Hobart mixer apparently did significant refining causing the D sheets to be denser and stronger than the B sheets. Also, based on our modeling (5,6), there is a maximum in accelerated creep when it is plotted as a function of fiber anisotropy. If the stiffness anisotropy is too great, most of the load will always be carried by the stiffer element, and no load cycling occurs. If the heterogeneity is too small, then elements move together, and again there is no anisotropy-driven load cycling. It is hard to guess where on the accelerated creep vs. fiber anisotropy curve the untreated and Hobart-mixer-treated samples lie, and it is hard to predict which should be larger.

SUMMARY

We were able to produce handsheets of different furnishes having different hygroexpansivities and different accelerated creeps. A blend of furnishes produced samples of intermediate hygroexpansion and intermediate accelerated creep. We therefore believe that blending of fiber types will have neither a corrupting nor a salutary influence on accelerated creep. However, we predict that multi-ply sheets of furnishes with different hygroexpansivities will experience additional accelerated creep when compared to the average of multi-ply sheets of a single furnish. Papermakers should consider degradation of product performance in cyclic humidity environments when they produce multi-ply products with layers of different hygroexpansion. In addition, it appears that accelerated creep is most potent when moisture cycle time is of the order of sorption time.

Acknowledgements: We thank the member companies of the Institute of Paper Science and Technology for their financial support. John Waterhouse and Derek Page made contributions to the direction of this research. Miranda Bliss made our handsheets, and Kennisha Collins did the creep testing.

LITERATURE CITED

1. Armstrong, L. D. and Kingston, R. S. T., *Nature* 185 (4716): 862 (1960).
2. Byrd, V. L., Tappi 55(2): 247(1972)
3. Pickett, G. J., J. Amer. Concrete Inst. 13(4): 257(1942)
4. Wang, J. Z., Vipul, D., Glasser, W., and Dillard, D. A., in *High Temperature and Environmental Effects on Polymeric Composites* ASTM STP 1174, C. E. Harris and T. S. Gates Eds. Amer. Soc. for Testing and Mater., Philadelphia: 186(1993).
5. Habeger, C., and Coffin, D. *J. Pulp Paper Sci.* 26(4): 145(2000)
6. Coffin, D. and Habeger, C., in Proceedings: Moisture and Creep Effects on Paper, Board, and Containers, Grenoble, France, March 18-19, 1999
7. Selway, J. W., and Kirkpatrick J., in, Proceedings: Cyclic Humidity Effects on Paperboard Packaging, FPL Madison, WI: 31(1992).
8. Nanko, H., and Tada, Y., Proceedings of the 1995 International Paper Physics Conference: 159(1995).
9. Nanri, Y., and Uesaka, T., Tappi 76(6): 62(1993).
10. Uesaka, T., Moss, C., and Nanri, Y., *J. Pulp Paper Sci.*18(1): J11(1992).
11. Salmen L., Proceedings of the Eleventh Fundamental Research Symposium Held in Cambridge: September 1997: 374(1997).
12. Page, D., Seth, R., Jordan, B., and Barbe, M., Papermaking Raw Materials (Punton, ed.), Vol. 1: (Mechanical Eng. Publns. Ltd.). 183(1985).
13. Page, D., Pulp Paper Mag. Can. 67(1): T2(1966).
14. Brezinski, J. P., Tappi 39(2): 166(1956).

Received: January 20, 2000
Accepted: September 26, 2000

This paper was accepted for abstracting and publication in the March 2001 issue of *TAPPI JOURNAL*.

TAPPI Website: www.tappi.org