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MECHANO–SORPTIVE CREEP OF PAPER

– Influence of drying restraint and fibre orientation

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ABSTRACT

A four–point bending stiffness tester was used to study the creep behaviour of corrugated board panels under constant and cyclic relative humidity. By substituting one linerboard in a single–wall corrugated board with steel ribbons, creep tests on linerboard under compression and under tension were also carried out. The results were evaluated using isochronous stress–strain curves relating to both constant and cyclic relative humidities. The isochronous curves showed a straight line relationship at low stress levels, for both constant and cyclic relative humidities. This means that the linerboard could be described as a linear viscoelastic material in both tension and compression at constant relative humidity.

The initial slope of the isochronous curves, the creep stiffness index, was lower under the cyclic relative humidity than at the constant relative humidity equal to the highest used in the cyclic creep experiments, a phenomenon called "mechano–sorptive creep". It is worth noting that, as opposed to many other investigations, mechano–sorptive creep was demonstrated for paper subjected to comparatively low strain levels. No difference was observed between creep stiffness indices in tension and compression under a constant climate, but

under a cyclic relative humidity the compression creep stiffness index was found to be lower i.e. the creep rate was higher in compression. A freely dried paper was found to have a lower compression creep stiffness index both at constant and under cyclic relative humidity than a paper dried under restraint. In machine–made sheets, the compression creep stiffness index was higher in the MD than in the CD both at constant and under cyclic relative humidities. For panels of corrugated board, isochronous bending moment–curvature curves were used to describe creep behaviour under pure bending. The results are discussed in relation to a model previously proposed (1) to describe the cyclic creep behaviour of paper.

INTRODUCTION

Several hygroscopic materials under stress exhibit, when subjected to a change in moisture content, a creep rate that is higher than the creep rate of the same material at a humidity equal to the highest moisture content used in the cyclic experiments. This phenomenon is referred to as "mechano–sorptive creep". The effect was first observed in wooden beams (2–5) and later in many other organic polymers (6, 7).

Mechano–sorptive phenomena in corrugated boards and linerboards and their influence on the stacking strength of corrugated boxes have been demonstrated in many experimental studies (8–14).

In a recent study on the creep of corrugated board under pure bending and of linerboard under tension and compression, the concept of "stress–induced hygroexpansion", SIH, was introduced (1). SIH has the effect that the hygroexpansion is higher under compression stresses and lower under tension stresses than in an unloaded sample. SIH explains why a beam of corrugated board exhibited its largest deflection in a dry climate. A physico–mechanical model, based on the fact that paper consists of hygroscopic cellulose fibres with anisotropic swelling, that describes SIH and mechano–sorptive phenomenon was also proposed. When the sample is under stress, the model further assumes a redistribution of stresses and the existence of and a change in the number of dislocations in the paper structure at different structural levels, and it deals with both compression and tension modes of applied stress.

Creep tests on corrugated board under pure bending and on linerboard under tension and compression under constant and cyclic relative humidity are reported in the present paper. By substituting one of the linerboards in the corrugated board with steel ribbons it has also been possible to evaluate creep properties of the linerboards under tension and compression in bending experiments. The results are evaluated using stress–strain curves at constant times, so called isochronous curves. The isochronous curve concept has been used earlier for creep studies of paper at constant relative humidity (15, 16).

Both in compression and in tension, the isochronous stress–strain curves show a linear relationship at low stress levels and constant relative humidity. This indicates that paper under low strain may be described as a linear viscoelastic material. The same straight line relationship was also found for low stresses and cyclic relative humidity. At higher strain levels, the stress–strain isochronous curves levelled off.

The initial slope of the isochronous curves, the creep stiffness index, was lower under a cyclic relative humidity than at a constant relative humidity equal to the highest humidity in the cyclic creep experiments. This is another way of describing the well known "mechano–sorptive creep" phenomenon. In the present study, the effects of drying conditions and fibre orientation of linerboard on the mechano–sorptive creep are demonstrated.

The tensile and compression creep stiffness indices at constant relative humidity were found to be the same. Under a cyclic relative humidity, however, the compression creep stiffness index was found to be lower than the tensile creep stiffness index.

A freely dried paper was found to have a lower compression creep stiffness index both at constant and under a cyclic relative humidity than a sheet dried under restraint. Creep experiments on machine–made sheets showed that the tensile and compression creep stiffness indices were higher in the MD than in the CD both at constant and under cyclic relative humidities. Mechano–sorptive effects on beams of corrugated board were also demonstrated.

The results are discussed in relation to an earlier proposed physico–mechanical model (1).

MATERIALS AND METHODS

Materials

In the creep experiments, three different types of beam constructions were used (fig. 1):

* Steel/linerboard combination with linerboard under compression.
* Steel/linerboard combination with linerboard under tension.
* Single–wall corrugated board with linerboard under compression and tension.

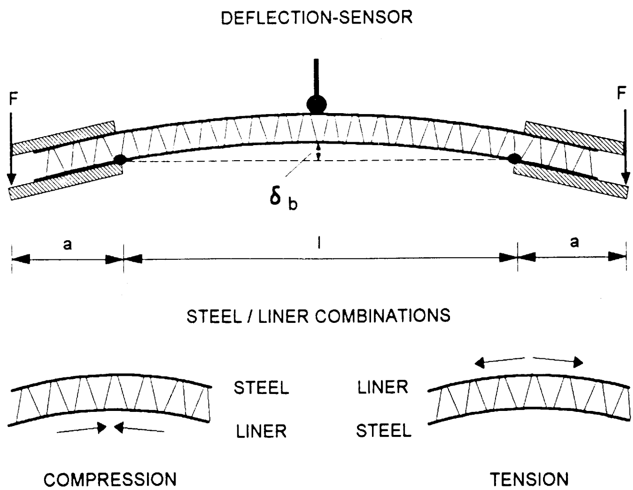

Figure 1 Measuring principle and the three different types of creep experiments.

Steel is insensitive to changes in relative humidity and comparatively insensitive to creep, from which it follows that the deflection of the steel/linerboard beam was governed by the hygroexpansion, change in elastic properties and creep of the linerboard. Depending on the way in which the test piece was mounted in the bending tester, the linerboard was subjected either to compression or to tension.

Both laboratory sheets and oriented machine-made kraft linerboards were used together with a commercial corrugated board.

The laboratory sheets were made in a dynamic sheetformer (17) using unbleached kraftpulp (kappa=80) beaten to 25 SR. The sheets were pressed and then dried both freely and under restraint (Table I). The freely dried sheets were dried on a heated cylinder against which they were held with a felt under low stress. To prevent any shrinkage during the drying under restraint, an STFI plate drier was used. The principle is described in reference (18). The oriented machine-made sheets were taken from the production on PM 1, ASSI Kraftliner, Sweden.

When hand-made or machine-made boards were tested, the sheets were glued to the flute tops on one side of a machine-made corrugated fluting using starch adhesive. The corrugated fluting was obtained by passing a release paper through the corrugator between the linerboard and the fluting when a single-face corrugated board was run. In this way a part of the web (width 200 mm) was obtained with no bonding between the linerboard and the fluting. The currugated fluting was then cut into pieces (200 x 100 mm) and the flute tops dipped into starch that was spread on a glass plate (approximately 15 g/m^2 wet). The board to be tested was then placed on the same flute tops and a pressure was applied (4 kN/m^2) until the glue was completely dry (approximately 6 hours). The steel ribbons were then glued to the other side using epoxy adhesive. Commercial corrugated board (single-wall, C-flute) using kraftliner in both the SF and DB positions was also used. Data for the materials used are summarized in Table I.

		Laboratory-made sheets		Oriented Machine-made sheets	Steel	Commercial corrugated board	
		Freely dried	Restrained dried			SF-liner	DB-liner
	Grammage [g/m2]	200	200	186	780	200	200
Tensile Stiffness index [MNm/kg]	MD	10.64	14.22	10.09	25	10.34	10.23
	CD	4.59	6.16	3.56	25	3.55	3.36
Strain at break [%]	MD	3.62	2.52	2.03	-	1.90	1.99
	CD	4.41	2.46	3.77	-	3.62	4.01

Table I Properties of materials.

Methods

Creep tests were conducted on a commercial four–point bending stiffness tester as described earlier (1). The tensile and compressive creep were monitored by recording the change in deflection with time of the test beam which was subjected to a pure bending force. From the change in deflection, the strain was then calculated.

Since all measurements were made in the machine direction of the corrugated board, i.e. the flute pipes were perpendicular to the direction of applied bending load, the contribution of the corrugated medium to the stiffness of the corrugated board was negligible (1). The stiffness of the corrugated board was thus governed by the tensile stiffness of and the distance between the linerboards.

The stiffness tester was placed in a cabinet in which the temperature was maintained constant to ± 0.25°C and the relative humidity to ± 0.5 %. The cabinet was programmed to change the relative humidity from 40 to 80 %, or vice versa, every fourth hour at a temperature of 27°C. This temperature was chosen slightly higher than the ambient room temperature to ensure adequate stability of the humidity control. Before the measurements were started, the specimens were subjected to two days of cycling climate before the load was applied in the middle of a 40 % relative humidity cycle. This preliminary procedure was adopted in order to release possible stresses after the specimen had been glued together and mounted in the measuring device.The procedure was adopted for the samples subsequently tested at a constant humidity as well as for those subjected to a cyclic climate.

The tensile stiffness index was determined for the linerboards before preparation of test panels and for the constituent linerboard in the corrugated board. In the latter case, the corrugated medium was cut in the middle of the corrugations using a razor blade. Tensile properties were evaluated in accordance with SCAN-test procedures on a 100 mm long test piece at a strain rate of 1.7% per second (19). The tensile stiffness index was evaluated from the maximum slope of the curve of force per unit width and unit grammage versus strain.

Principles of evaluation

All creep data were evaluated by plotting them in the form of isochronous stress-strain curves, i.e. mutually connected values of stress and strain at a constant time (20, 21).

As an example, points A_1, A_2 and A_3 were taken from three different strain-time curves at a creep time of t in Fig 2a and replotted to one stress-strain curve in Fig 2b. If the initial part of the isochronous stress-strain curve was a straight line the slope was denoted the creep stiffness index. The following conventions are used for the different variables used:

Stress:	$\sigma^{w} = F/b \cdot w$	Nm/kg	[1]
Creep stiffness index:	$E^{w,cr}(t) = \sigma^{w}/\varepsilon(t)$	Nm/kg	[2]

Where:

- F = force, N
- b = test piece width, m
- w = test piece grammage, kg/m^2
- ε = strain, m/m
- t = time, s

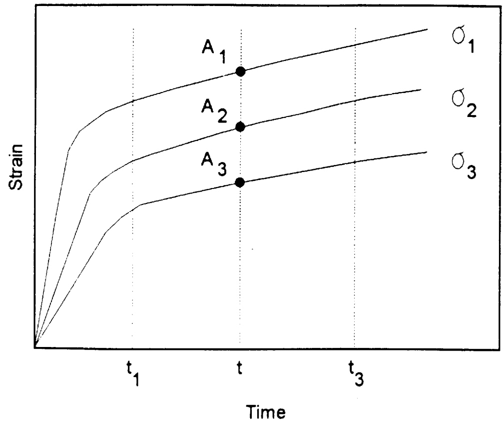

Figure 2a Strain vs time for different stress levels, A_1-A_3 are points on the different curves at time t.

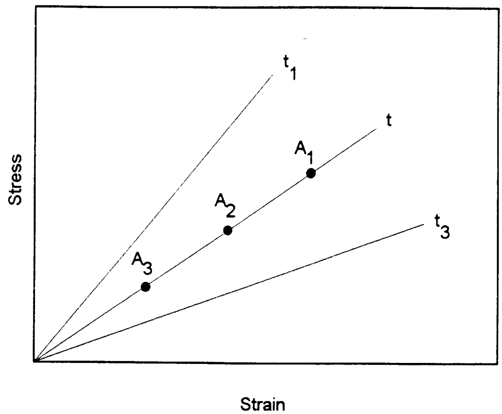

Figure 2b Stress vs strain at different times, so called isochronous curves. A_1-A_3 from Fig 2a gives the isochronous curve at time t.

RESULTS

The aim of the present study has been to compare the creep behaviour of paper and corrugated board under constant and cyclic relative humidity using isochronous stress–strain curves. Comparisons have been made between tension and compression creep and the effects of both drying conditions and fibre orientation have been demonstrated. The creep behaviour of commercial corrugated boards has also been studied.

Mechano-sorptive creep

A typical recording of a creep test, for a steel-linerboard combination with the linerboard under compression, is shown in Fig 3. The test piece was first subjected to a cyclic climate for two days. The specimen showed a large change in deflection each time the climate was changed due to the hygroexpansivity of the linerboard. After the specimen had been held at a relative humidity of 40 % for two hours the load was applied at point A where the creep starts. The relative humidity was then kept constant for a further two hours to point B where the relative humidity was increased to 80% and the strain started to increase. The total change in strain was then the combined effect of the relatively rapid expansion of the paper through moisture adsorption and a decrease in elastic properties together with the minor creep response (1). The latter contribution could be isolated in the last part of the humidity cycle as a slight decrease in strain. The humidity then continued to cycle between 40 and 80 % relative humidity and the same principle pattern regarding strain was observed.

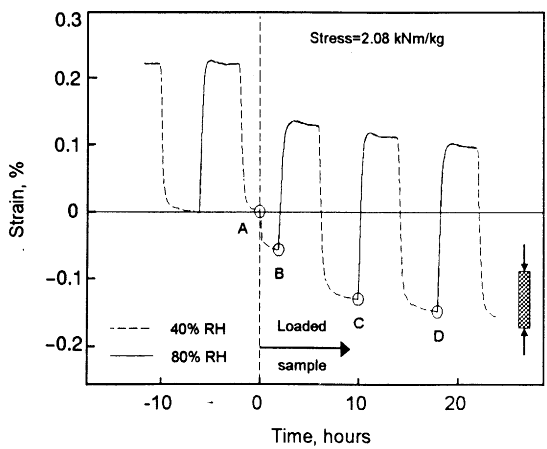

Figure 3 Strain vs time for a steel/linerboard combination subjected to a cycled climate with the linerboard under compression.

In Fig 4, the creep behaviour under compression of freely dried sheets subjected to a cyclic relative humidity, curve 1 (40/80 %), is compared with the creep behaviour of samples subjected to a constant relative humidity, curve 2 (80 %) and curve 3 (40 %). The strain in curve 1 was determined at the end of each low humidity level indicated by points B, C, D and so on in Fig 3. The reference for the strain determination was in each case the strain at the moment when the load was applied. It is evident that in the specimen subjected to the highest relative humidity, curve 2, the creep rate was lower than in the specimen subjected to a cyclic relative humidity, curve 1. The different times of creep can be related to the number of cycles, as shown on the abscissa, since the cycle time was constant. Note that cycle number 0 is equivalent to two hours of loading at constant relative humidity (40 %).

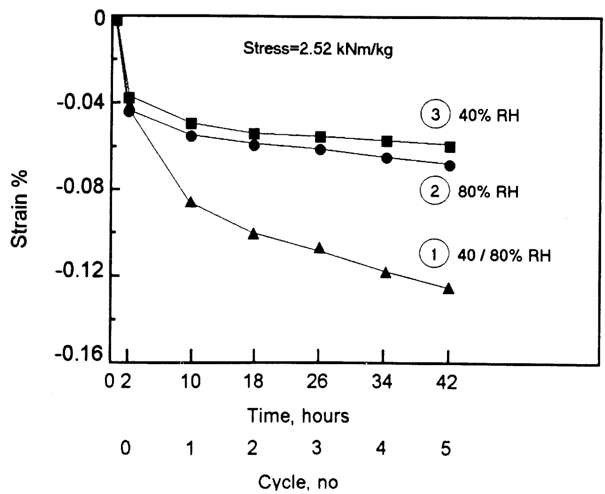

Figure 4 Strain vs time for specimens subjected to constant climate (2 and 3) and to a cycled climate (1).

Figure 5 shows isochronous curves for freely dried laboratory sheets under a compressive load. At these low levels of strain, there is a linear relationship between stress and strain for the three different number of cycles, including number 0 representing constant relative humidity. Cycle number 0 represents the strain increase A to B and cycle number 1 the strain increase A to C in Fig. 3, and so on. The slope of the line is the creep stiffness index as defined by Eq. [2]. A decrease in compression creep stiffness index with increasing number of cycles is observed, i.e. at a given stress the strain increases with increasing number of humidity cycles.

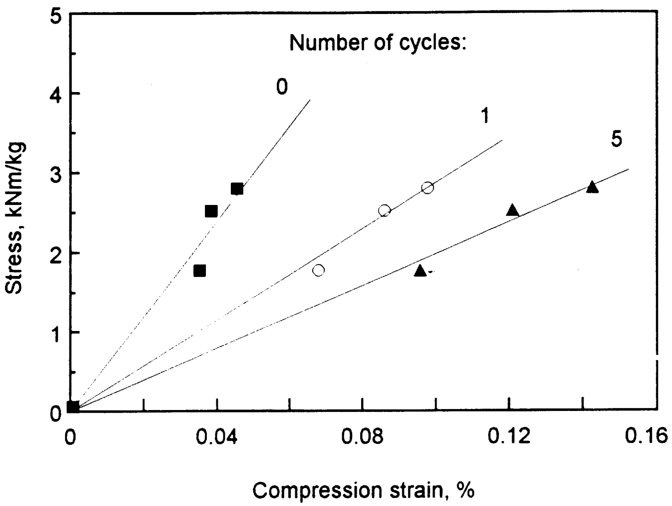

Figure 5 Isochronous curves for a linerboard under compression subjected to different numbers of climatic cycles.

The isochronous curves can also be used to illustrate the mechano-sorptive effect, as in Fig. 6. Curve 1 represents the isochronous curve at a constant relative humidity of 80 % and a creep time of 42 hours and curve 2 represents the same time of creep but under a cyclic climate. The lower compression creep stiffness index of curve 2 illustrates that the creep is more rapid under a cyclic climate than at the highest constant climate used in the cyclic experiments, curve 1.

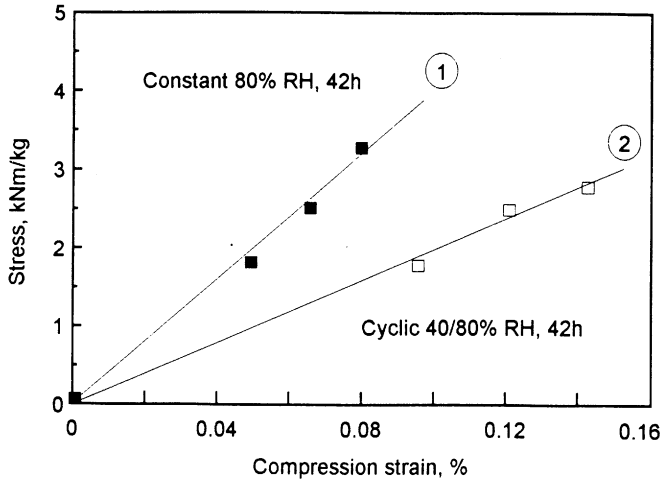

Figure 6 Isochronous cruves for a freely dried linerboard under compression subjected to a constant climate (1) and to a cycled climate (2). Total creep time is 42 hours.

Tension and compression

The difference in creep between tension and compression is illustrated in Fig.7. These isochronous curves refer to laboratory sheets dried under restraint. No difference can be seen between the tensile and compression creep stiffness indices at constant relative humidity. Under a cyclic relative humidity however the compression creep stiffness index was less than the tensile creep stiffness index, i.e. the creep rate was higher in compression than in tension.

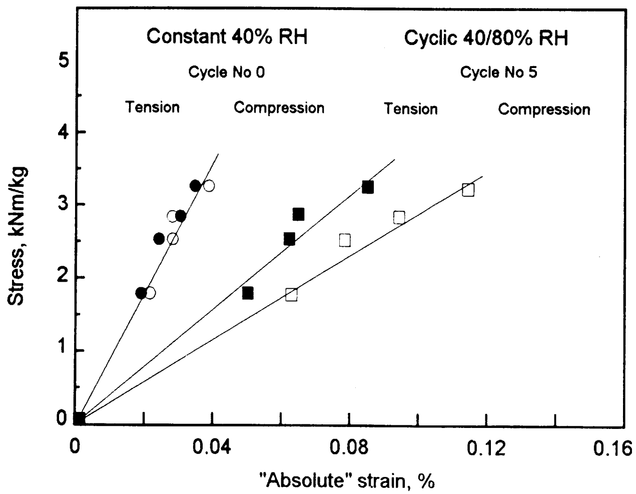

Figure 7 Isochrones for tension and compression modes, both at constant climate (cycle No 0) and under cycled climate (cycle No 5). The sheets were dried under restraint.

Drying conditions

Freely dried paper and paper dried under restraint were compared in the compression mode (Fig. 8). In this case, a difference was noted in compression creep stiffness index both at constant and under cyclic relative humidity. The compression creep stiffness indices were lower for the freely dried sheets than for the sheets dried under restraint. This was expected, at least at constant relative humidity (cycle 0), since the tensile stiffness index of a freely dried paper was lower than that of paper dried under restraint (Table I).

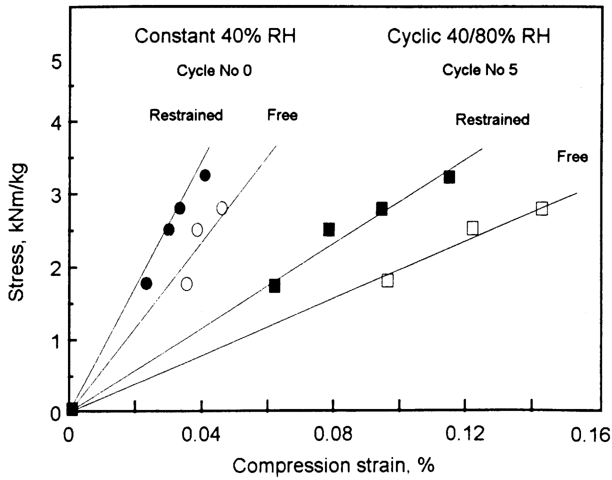

Figure 8 Isochronous curves for freely dried sheets and restrained dried sheets loaded in compression, both at constant climate (cycle No 0) and under cycled climate (cycle No 5).

Fibre orientation

The influence of fibre orientation on the compression creep stiffness index at constant and under cyclic relative humidity was studied on machine-made sheets by comparing MD and CD behaviour. Fig. 9 shows that the compression creep stiffness index in the MD was higher both at constant and under cyclic relative humidity than that in the CD. This was also expected, since the tensile stiffness index was lower in the CD than in the MD (Table I).

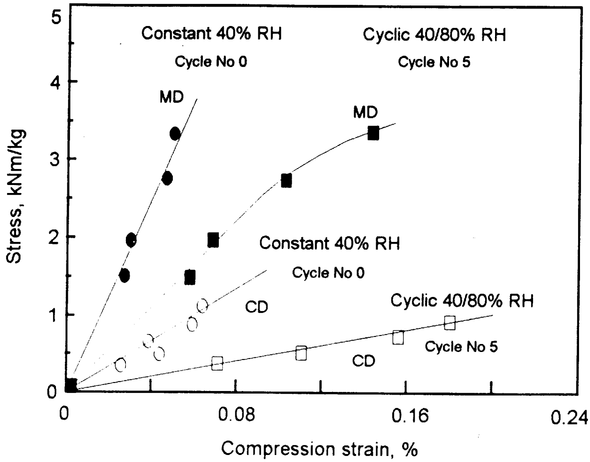

Figure 9 Isochronous curves for oriented machine-made sheets, loaded in compression. The isochronous curves are shown for both constant climate (cycle No 0) and cycled climate (cycle No 5).

Corrugated board

Mechano–sorptive creep has also been demonstrated for corrugated board in the machine direction, i.e. with the flute pipes perpendicular to the direction of the applied bending load. The results are illustrated in Fig. 10 for two different applied loads, where the curvature of the beam is plotted versus time. Increasing curvature corresponds to an increase in tension and in compression strain for the two linerboards in the panel. The load was applied at point A (=zero time) at 40 % relative humidity. After two hours creep at constant relative humidity, the curvature had increased up to B, at which point the relative humidity was increased to 80 % relative humidity. At this relative humidity, the curvature increased further up to point C. In the subsequent humidity cycles the curvature increased during each 40 % period and decreased during each 80 % period, as reported earlier (1). Fig. 10 illustrates that the overall creep was lower when the bending moment was decreased, although the same general pattern was recognized. Note that, after the first humidity cycle, the curvature of the beam under stress is greater in the dry than in the humid climate.

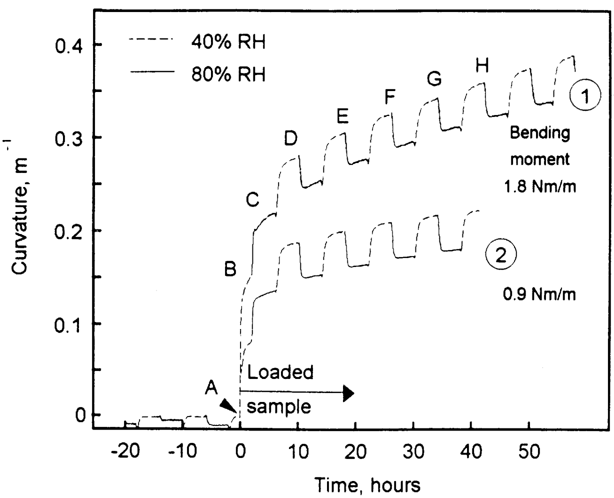

Figure 10 Curvature as a function of time for corrugated board samples subjected to a cycled climate. A load was applied at point A.

The envelopes of the creep curves, points A, B, D, E and so on, were replotted as bending moment versus curvature isochronous curves (Fig. 11) instead of as stress versus calculated strain. This was necessary because the strains in the two linerboards were not the same since one was under compression and the other under tension. The isochronous curves are shown in Fig. 11 for three different numbers of cycles, where cycle 0 represents constant relative humidity. Initially all the curves are linear, but at higher curvatures they tend to level off. It is further seen in Fig. 11 that the greater the bending moment the greater is the difference in curvature between constant and cyclic climates. At higher bending moments, the beams collapsed through buckling of the linerboard on the compression side between two flute tops. The curvature at collapse was always approximately 0.9 m^{-1} for this type of corrugated board.

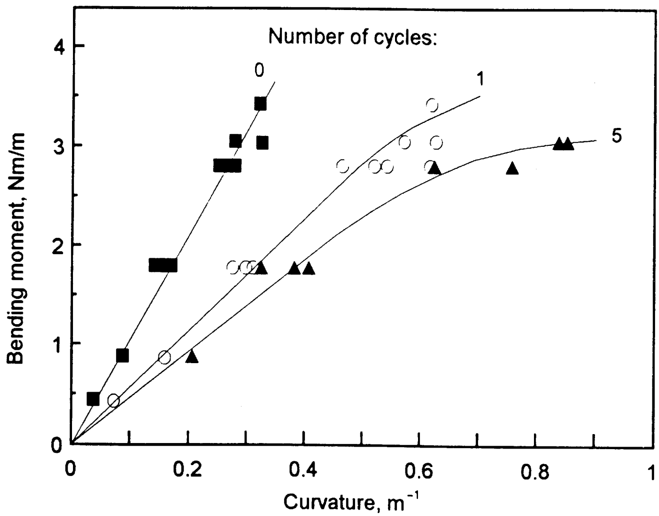

Figure 11 Isochronous curves for a corrugated board subjected to different numbers of climatic cycles.

In Fig. 12, both curves represent a creep time of 42 hours. The steeper slope was obtained for samples subjected to a constant relative humidity of 80 %. The lower slope was obtained when the board was subjected to cyclic relative humidity.

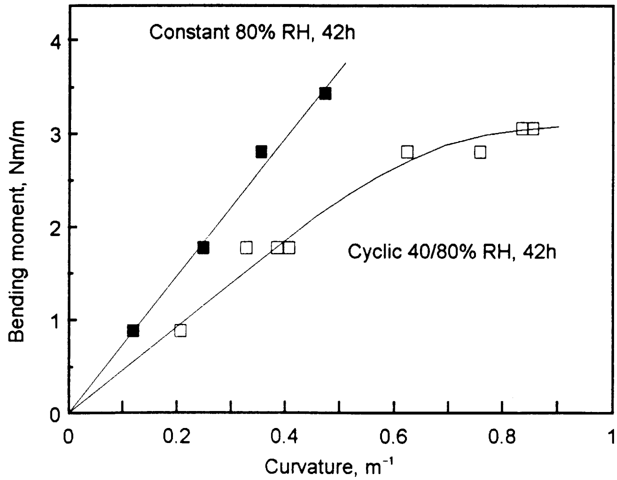

Figure 12 Isochronous curves for a corrugated board subjected to a constant climate and to a cycled climate. Total creep 42 hours.

DISCUSSION

The result of the creep studies on linerboard under compression and tension and of the studies on corrugated board under pure bending were evaluated using isochronous stress–strain and bending moment–curvature curves, respectively. This way of describing creep in a constant climate is often used for engineering purposes in the plastic industry (20, 21) and has also been used for paper at a constant relative humidity (15, 16). The initial linear slope of the isochrone at a low strain level is called the creep stiffness index ($E^{w,cr}(t)$), i.e. the material can be treated as a linear viscoelastic material. In this study, the tensile and compression creep stiffness indices were found to be the same at a constant ambient climate.

The isochronous curve concept was also tested with respect to its ability to describe the creep behaviour under a cyclic relative humidity. A striking result was that the envelope of the strain–time curve could also be expressed as a isochronous stress–strain curve. Furthermore, at low strain levels, the stress–strain relationship showed a linear relationship just as it did at constant relative humidity. Accordingly the term "creep stiffness index" is here also used in the cyclic relative humidity experiments. Isochronous creep curves may be used to compare the responses of different materials to a cyclic relative humidity. The isochronous curve concept makes it possible to describe mechano–sorptive creep quantitatively and to create a foundation for setting up design criteria for corrugated boxes.

In an earlier paper (1), a physico–mechanical model was proposed for paper that describes mechano–sorptive creep and shows that compression stresses increase and tension stresses decrease hygroexpansion. The model was based on the fact that paper consists of hygroscopic cellulose fibres with anisotropic swelling. It further assumed that there is a redistribution of stresses during swelling and deswelling and that there are dislocations in the paper structure at different structural levels. The model was applied to phenomena occurring at low strain levels.

Dislocations are created in the fibres in many stages of the papermaking process e.g. during chipping and beating. During the drying of paper, the fibres shrink in the lateral direction which leads to compression failure in the fibres bonded at right angles and thus new areas of dislocation are formed (22, 23). During restrained drying, the free fibre segments are straightened out and become load-bearing in their longitudinal direction (24–26). During this process, the number of dislocations in the fibre segments decreases. Free drying of the sheets gives less straightend fibre segments and leaves more dislocations in these areas. As a consequence, the sheet exhibits a greater hygroexpansion and lower stiffness (27–30).

When a sheet of paper is subjected to a change in atmospheric relative humidity, anisotropic swelling or deswelling occurs in the structure. These changes in swelling are likely to lead to a redistribution of stresses in the paper structure on the molecular or fibre level. If the paper is subjected to an external static load, the number of dislocations will be affected, predominantly in the direction of load, to a different degree each time a moisture change takes place, due to a redestribution of stresses. Compression stresses increase and tension stresses decrease the number of dislocations and thus the strain is affected. In both cases the creep rate increases.

Mechano-sorptive creep

In the present work this mechano-sorptive creep phenomenon has been demonstrated for linerboard under compression and corrugated board under pure bending, at low strain levels. The results have been expressed as strain-time curves (Fig. 4) and in a more comprehensive form as isochronous stress-strain curves e.g. Fig. 6 or isochronous bending moment-curvature curves e.g. Fig. 12. If the isochronous curve at 42 hours and the highest constant relative humidity used is compared with the corresponding isochronous curve for a cyclic climate (Fig. 6) it is found that the creep stiffness index in a cyclic climate is lower, i.e. the strain is greater at a given stress under a cyclic relative humidity. This mechano-sorptive creep effect has also been demonstrated for corrugated board under bending (Fig.12). The proposed physico-mechanical model suggests that the increase in creep in a cyclic relative humidity is due to a change in the number of dislocations during swelling and deswelling of the fibres. It is worth noting that, as opposed to other investigations, mechano–sorptive creep is here demonstrated and interpreted for paper subjected to comparatively low strain levels.

Tension and compression

The study has also revealed that under a cyclic climate the creep stiffness index is lower in compression than in tension, although no difference was observed at constant relative humidity (Fig.7). That the creep stiffness indices for paper under compression and under tension are the same in a constant climate has been reported earlier (16). Compressive mechano–sorptive creep is the result of an increase in the number of dislocations under stress, and tensile mechano–sorptive creep is the result of a decrease in the number of dislocations. When the data for cycles 0 and 5 (Fig. 7) are compared, it is concluded that new dislocations are created faster in compression than existing dislocations disappear in tension when the relative humidity is cycled. This means that there are fewer dislocations to eliminate in tension than there are potential new dislocations to create in compression.

The assumption that the increase in number of dislocations in compression is faster than the decrease in tension explains in accordance with the proposed model, the earlier observation that the stiffness decreased more (13%) in compression than it increased (9%) in tension, (1).

Drying conditions and fibre orientation

Two exploratory studies were performed in order to illustrate the influence of drying conditions and fibre orientation on creep behaviour.

At a given compression stress, the strain recorded in cycle 0 at constant relative humidity was less for the sheet dried under restraint than for the freely dried sheet (Fig.8). The same applied in cycle 5, i.e. the compression creep stiffness index for sheets dried under restraint is higher both at constant and under cyclic relative humidity. This was expected for cycle 0 since the tensile stiffness was higher in the sheet dried under restraint (Table I). The compression creep stiffness index in cycle 5 decreased to approximately 1/3 of its value in cycle 0 for both freely dried sheets and sheets dried under restraint.

The results of fibre orientation studies (Fig. 9) showed that the compression creep stiffness index was higher in the MD than in the CD both at constant and under cyclic relative humidity. The decrease in compression creep stiffness index was approximately 1/3 from cycle 0 to 5, i.e. the change in creep stiffness index due to a cyclic climate was not influenced dramatically by the drying conditions or by the fibre orientation.

Corrugated board

In the experiments on creep in corrugated board, it was found that the deflection of the beam was greater in the dry climate than in the humid climate after the first humidity cycle. This behaviour is explained by the existence of a stress-induced hygroexpansion, SIH (1). When a compressive force is applied, the hygroexpansion increases in the direction of the applied stress and, when a tensional force is applied, the hygroexpansion decreases. This observation and the fact that the degree to which the hygroexpansion is affected under stress is greater than the sum of the elastic and creep responses explains the greater deflection at the lower relative humidity. The exception in the first high humidity period when the deflection increased (Fig.10, B–C) is explained by the large creep response during this first humidification, and is supported by the results in Fig.11. It can be seen in this figure that the greatest increase in deflection occurs during the first cycle (cycle 0 to 1), after which the increases in the following humidity cycles are comparatively small (cycle 1 to 5). Thus the contribution from creep in the first cycle was so large that the stress–induced hygroexpansion was less than the sum of the elastic and creep responses.

Figure 12 shows that mechano–sorptive creep can be illustrated in isochronous bending moment–curvature curves. Thus the isochronous curve concept is not limited to stress–strain relations but can also be applied to more direct curvature measurements on panels of corrugated board under bending stress. This technique may be useful in studies of the performance of corrugated containers.

Conclusions

* Mechano–sorptive creep effects are present at all strain levels.

* A creep stiffness index in tension and compression can be defined for both constant and cyclic relative humidities.

* The creep stiffness index is lower for samples subjected to cyclic relative humidity than for those subjected to a constant relative humidity.

* The creep stiffness index is the same in tension and compression at constant relative humidity, but under a cyclic relative humidity the compression creep stiffness index is lower than tensile creep stiffness index, i.e. the creep–rate is higher in compression.

* A board dried under restraint has a higher compression creep stiffness index than a board dried without restraint both at a constant and under a cyclic relative humidity.

* A commercial linerboard has a higher compression creep stiffness index in the MD than in the CD both at a constant and under a cyclic relative humidity.

* Corrugated board under bending shows a linear relationship in isochronous bending moment–curvature curves at low stress levels.

* The isochrone concept provides a good technique for studying the long–term loading behaviour of paper and paper products.

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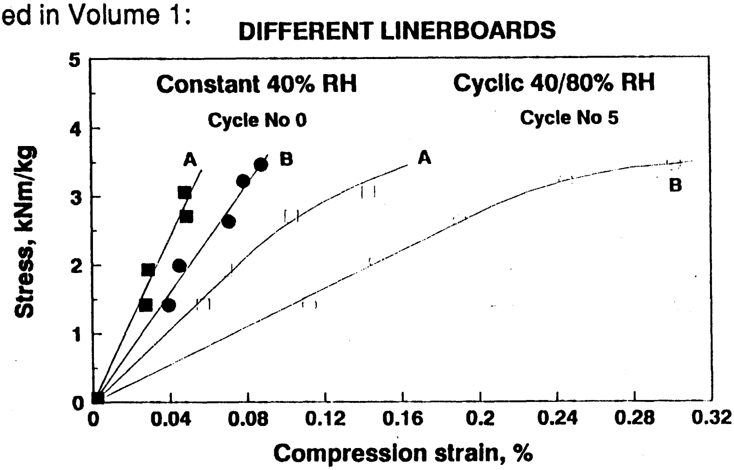
Transcription of Discussion

MECHANO-SORPTIVE CREEP OF PAPER - INFLUENCE OF DRYING RESTRAINT AND FIBRE ORIENTATION

C Soremark, C Fellers & L Henriksson

Supplementary information given at the presentation that is not included in Volume 1:



Isochronous curves for two different paper grades subjected to a constant and a cyclic climate. (Paper A mainly virgin fibers, paper B mainly recycled fibers.)

Comparison of stiffness ratios evaluated by different testing procedures

Ratio (Property)	Tensile [1] Stiffness	Creep Stiffness [2] Constant RH [3]	Creep Stiffness [2] Cyclic RH [4]
Tension/ Compression	1.0	1.0	1.4
Restrained/ [5] Freely	1.3	1.4	1.5
MD/CD [5]	2.8	4.0	5.1
Sample A/ [5] Sample B	1.5	1.6	2.0

1. Standard tensile testing
2. Creep experiments
3. RH = 40%, creep time 2 hours
4. RH - 40/80%, creep time 42 hours
5. Compressive loading

Prof B Lyne, Royal Institute of Technology, Sweden

I don't think I fully understood your model. When the humidity in paper is increased and the fibres have dislocations such as microcompressions and kinks, then the fibres swell transversally the kinks and dislocations tend to straighten out. This is one of the mechanisms for hygroexpansion in paper. Don't you have fewer dislocations in paper with an increase in moisture?

C Soremark

No. If you increase the moisture content, you must not forget that the sample is under compression stress all the time. When you change the moisture content, increasing it for instance, in those disordered areas or kinks - there would be a swelling but those are also the weak points. There exists a stress distribution such that there will be viscoelastic flow of the material in certain areas - a real deformation under the external pressure so the number of dislocations will increase. What I have not shown here is that the reverse takes place when you have it under tensional force. So under tension and you are changing the moisture content, some of these dislocations or disordered areas will be eliminated. This has been shown before and will also affect the tensile stiffness of the paper in the dry state. As a result of what I said which is in accordance with your question - under compression the hygroexpansion will increase due to an increased number of dislocations so there is still a coupling between the number of dislocations and the hygroexpansion. The hygroexpansion is higher in a paper under a compressive load.

B Lyne

When you moisturise a fibre it is like blowing up a tyre or inner tube that is kinked. When you blow the inner tube up it becomes smooth and straight.

C Soremark

The swelling will then cause that the yield points in different areas are reached so the material will flow and that is why you increase the number of dislocations.

I Kartovaara, Enso-Gutzeit Oy, Finland

If I remember correctly, some research that is being done in ESPRI you get exactly the same mechano-sorptive phenomenon also in the case when you do not have actual cyclic change in moisture content in the sample. In this situation both sides of the samples are exposed to different relative humidities where you have a flow of water through the sample but each layer of the sample is all the time at the same moisture content. I think that because a mere flow of water molecules can cause this phenomenon, you must also have a mechanism which operates without swelling and drying.

C Soremark

Yes. This is absolutely true. When you have a moisture gradient over the paper there would be a net flow of water through the paper but each layer of the paper there will be at the same moisture content. So there is no change in swelling with this procedure. You should differentiate between two things here - some are referring to a rapid or fast responses just when the moisture is changing transient effects. We are discussing the equilibrium values.

According to your example there was no accelerated creep until you changed the moisture content on one of the two sides. So there was no accelerated creep in the experiment you referred to.
It is only when you get a moisture change within the sample that there is an accelerated creep.

I K Kartovaara
I hope there is someone in the audience who remembers the results but my impression was that there is a mechano-sorptive phenomenon also under those conditions.

C Soremark
No, there was not.

Dr K Ebeling, Kymmene Corporation, Finland
Is there a thermodynamic principle that guides the water molecules to those inter- and intra-fibre hydrogen bonds, where the stress relaxation due to addition of water molecule(s) will be at a maximum?

C Soremark
What we have suggested is a physico-mechanical model. Of course, you can discuss this in terms of free volume but we are not measuring what is happening during the change of moisture in the sample. We are not discussing the transient effects in creep. We are discussing what happens when you go from one level to another level in moisture content, and in this case back again.

Dr D Page, Pulp & Paper Research Institute of Canada
I am delighted that you have invoked microcompressions as a possible explanation of this mechanism. There are some things that concern me though - firstly I believe, that this mechano-sorptive effect does occur in other materials like bulk nylon which do not actually have any microcompressions in them to the best of my knowledge, and I feel that the key to the understanding of this is going to be in a little sentence which I spotted in your Paper. This says that there is a redistribution of stresses in the paper structure at the molecular or fibre level. I think it is the molecular level that is important. I suggest what is happening is very simply that when you create bonds by drying down to 40% relative humidity and then you humidify again, the bonds which break when you re-humidify are not the same as the bonds which form when you dry. I am talking now at a molecular level. They are not the same ones because they are being stressed and so the ones which are more favourable to forming in the stressed state will form and this will repeat itself in exactly the same way that a caterpillar moves along by a little hump and it can shear across a surface by breaking and reforming different contacts with a twig. It is more that kind of molecular mechanism which is probably going to be the explanation.

Prof E Back, Feedback Consulting E&E Back KB, Sweden
Professor J Kubat** pointed out some 30 years ago that the so called mechano-sorptive effect also called transient effects during the absorption or de-sorption of moisture underload did occur also in polymers on absorption or desorption of softeners from the gas phase. There were a couple of such polymers tested and that indicates some of the general nature***. Is it realistic to describe the

phenomenon as you have done here with microcompressions and stress concentrations in fibres without having made the corresponding experiments, with eg a cellulose acetate film or a similar material?

** Kubat J., and Lindberg B; Appl Polymer Sci 9, (1965) p 2651
*** Back E L., Salmen L and Richardson G; Svensk Papperstidn 86 (1983):6 R 61-71

C Soremark
Yes. I agree with you. As Derek has said it should be put down to the molecular level and the explanation should be found there. We have not had the objectives primarily to describe the process in detail.

Dr A Nissan, Westvaco Corporation, USA
I think Derek Page highlighted a very important point. It is not only applicable to structures like paper. I believe polystyrene shows this type of behaviour in an atmosphere of benzene that changes in concentration. So it need not be hydrogen bonding because it does seem to apply to other systems.

I do think an experiment with cellophane might be very useful to see what a homogeneous nonfibrous structure shows. I believe it is shown by cement and water and of course it was originally discovered with wood. I believe in Australia in 1960? In other words it seems to be a more universal phenomenon and perhaps not totally explainable by paper structure and water. Other systems which lack the heterogeneous structure of paper and specifically the hydrogen bonding system also show similar phenomena. Therefore an experiment should cover them as well.

C Soremark

That is true. I do not have the intention to go so deep into the theory. There have been review articles in the literature eg, Wang (Ref 6&7) and this phenomena has been seen as well in concrete as in different organic materials. All that are common when you find the mechano-sorptive creep phenomena. It is that it has to be hydrophilic or hydroscopic and show anisotropic swelling so you will find it for instance in Kevlar fibres. You will not find it in Nylon 6.6 because it does not have any anisotropic swelling but it is still hydrophilic. It seems to be hydrophilicity in combination with anisotropic swelling that are needed for mechano-sorptive creep under the influence of moisture changes.

C Fellers, STFI, Sweden

I would like to make a few comments. We realise that this phenomenon might be explained on different structure levels and we could discuss that at length. Our contribution to explain mechano-sorptive creep is that we are saying that the effect is due to non-uniformity of the swelling in the structure and that you are forming some sort of dislocations on a molecular level which then spread to other structural levels. D Gunderson should have part of the credit for this view because he once gave me a piece of paper that had been exposed to cyclic creep. I noticed a lot of undulations on the surface and a lot of structural changes. The paper exposed to cyclic creep has a much higher strain to failure compared to normal papers. There are structural effects happening.

Dr K Ebeling, Kymmene Corp, Finland

The Fundamental Research Committee re-published a year ago the collected works of Barkas. I think he was one of the first to show that when you stretch wood material or paper it tends to pick up moisture from the surrounding air and when placed under compression it releases some moisture. Thus, there is this stress induced moisture absorption/desorption phenomena together with what you have been explaining. I do not think that you need to introduce a new concept of non-uniformity to account for what you have observed. It is just redistribution of stresses on very small structural level and redistribution of moisture in those stressed areas. This is why I have asked: "Do you know if there is a thermodynamic principle that will tell the water molecule that this specific molecule is now needed here because here it can give the greatest relief (relaxation)"?

C Soremark

I know about the work of Barkas, and he states that moisture content increases during tension and it has also been demonstrated experimentally - in a very elegant experiment by D. Gunderson - that there is a slight moisture increase in a sample under tension. In this case remember that the stress level was the same all the time. The global stress level was the same. There may be differences on a structural level. It could be that there is a migration of water from one area to another. Is that what you suggested?

K Ebeling

Yes. As soon as the stress is redistributed there is a migration of water due to the Barkas mechanism. The question is -Is there some thermodynamic address to that water?

C Soremark

I do not know the answer for the moment. There could be.