Micro-mechanically based modeling of mechano-sorptive creep in paper

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Preface

The work presented in this thesis has been carried out between February 1999 and July 2004 at KTH Solid Mechanics, Royal Institute of Technology, Stockholm, Sweden.

I would like to thank my supervisors Prof. Peter Gudmundson and Prof. Sören Östlund who initiated the project and provided excellent guidance and support during the work. I would also like to thank Mr. Hans Öberg and Mr. Bertil Dolk for the design and manufacturing of experimental equipment, and the Paper physics group at STFI-Packforsk, especially Prof. Christer Fellers and Dr. Joel Panek[1], for many valuable discussions.

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Finally I want to thank my colleagues for creating a stimulating atmosphere at the department, and my family who have always supported me.

Stockholm, September 2004

Johan Alfthan

[1]Now at Iggesund Paperboard, Workington, UK

List of appended papers:

Paper A:

J. Alfthan, P. Gudmundson and S. Östlund, “A micro-mechanical model for mechano-sorptive creep in paper”, *J. Pulp Paper Sci.*, 28(3):98-104 (2002)

Paper B:

J. Alfthan, “A simplified network model for mechano-sorptive creep in paper”, *J. Pulp Paper Sci.*, 29(7):228-234 (2003)

Paper C:

J. Alfthan, “The effect of humidity cycle amplitude on accelerated tensile creep of paper”, Report 366, KTH Solid Mechanics, Stockholm (2004), submitted for international publication

Paper D:

J. Alfthan and P. Gudmundson, “Linear constitutive model for mechano-sorptive creep in paper”, Report 375, KTH Solid Mechanics, Stockholm (2004)

Abstract

The creep of paper is accelerated by moisture content changes. This acceleration is known as mechano-sorptive creep, which is also found in wood and some other materials. Mechano-sorptive creep has been known for several decades but it is still not well understood, and there is no generally accepted model explaining the effect.

In this thesis, it is assumed that mechano-sorptive creep is the result of transient redistributions of stresses during moisture content changes in combination with non-linear creep behaviour of the material. The stress redistributions are caused by the anisotropic hygroexpansion of the fibres, which will give a mismatch of hygroexpansive strains at the bonds and hence large stresses each time the moisture content changes. This redistribution will lead to an uneven stress state. If the creep of the material depends non-linearly on stresses this will give an increase in creep rate where the stresses are high, that is larger than the decrease of creep rate where stresses are low, so in average there will be an increase in creep rate. The stress distribution evens out as the stresses relax during creep, and the moisture content has to change again to create a new uneven stress state and maintain the accelerated creep.

Two different network models based on this mechanism are developed in this thesis. Numerical simulations show that the models produce results similar to the mechano-sorptive creep found in paper. In the first model it is assumed that creep takes place in the fibre-fibre interfaces at the bonds, in the second the creep of the fibres themselves is accelerated. The second model is further developed. Experiments verify model predictions of the dependence of the amplitude of moisture changes.

The second model shows a linear relationship between mechanical load and deformation, although creep of the fibres depends non-linearly on stresses. This linear behaviour is also found in applications. Further analysis shows that the mechanical load can be treated as a small perturbation of the internal stress state caused by moisture content changes. This can be used to develop a linearized model, from which a continuum model can be derived. This leads to a reduction of the necessary number of variables, and a significant increase in speed of calculations. Hence, this linearized continuum model can be used as a constitutive law of paper in problems with complicated geometries, for example a corrugated board box in varying humidity.

Contents

1 Introduction

Packages of paper often have to withstand loads for a long time. Consequently, the time dependent deformation, creep, must be considered in design of packages. The creep of paper is affected by humidity. Higher humidity, and hence higher moisture content, means more creep. In addition, creep is accelerated by varying humidity, Fig. 1. This phenomenon is known as *mechano-sorptive creep*, or *accelerated creep*. The total deformation after a few humidity cycles exceeds the deformation at any constant humidity.

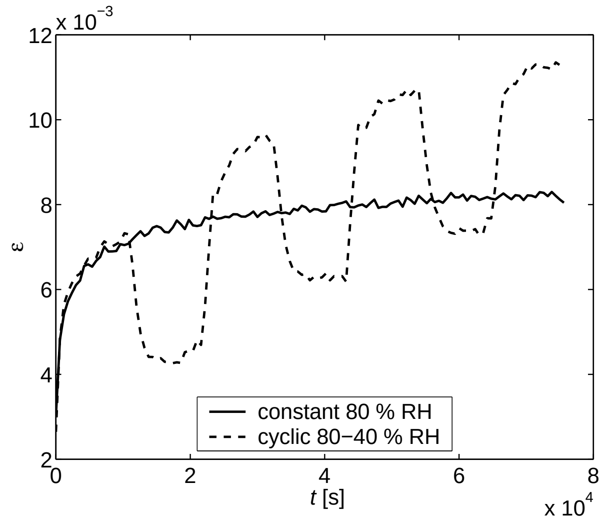


Fig. 1. *Example of mechano-sorptive creep. Results from tensile creep tests (specific stress 4.9 kNm/kg) in cyclic and constant relative humidity are shown.*

Mechano-sorptive creep was independently reported in the literature for wool [1, 2] and wood [3, 4] in the late 1950s and early 1960s. Ten years later it was also found in paper [5, 6]. In the 1990s mechano-sorptive creep was also observed in some synthetic fibres, for example Kevlar [7–9]. A similar phenomenon is found in concrete [10].

The most important load cases in packages are compression and bending, and many of the experimental results reflect this. The mechanical properties (except the initial stiffness) of paper are different in tension and compression, and this is true for mechano-sorptive creep as well [11, 12]. This probably arises from the structural properties of the fibre network. The compressive load case involves more structural mechanisms. The models in this thesis are hence first and foremost aimed at explaining mechano-sorptive creep in tension. It is assumed that mechano-sorptive creep is a result of heterogeneous hygroexpansion producing a transient redistribution of stresses during moisture content changes, which in combination with non-linear stress dependence will lead to an acceleration of creep. Mechano-sorptive creep is then a natural consequence of the regular creep properties of fibres and papers.

2 Previous work

2.1 Observations

The first reported observation of mechano-sorptive creep in wood was made in 1960 by Armstrong and Kingston [3]. It had previously been observed that moisture content changes in wood increased the creep rate and the total creep. Armstrong and Kingston was able to confirm this observation for small wooden beams. They studied creep in bending at three different moisture conditions: Never dried wood at constant moisture content, never dried wood allowed to dry and dried wood at constant moisture content. They found that creep of the specimens allowed to dry was at least twice that of specimens held at constant moisture content. This discovery resulted in a more extensive investigation carried out by Armstrong and Christensen [4]. In this investigation bending of wooden beams of different sizes was studied during moisture cycling between a completely dry state and a wet state. The result is shown in Fig. 2.

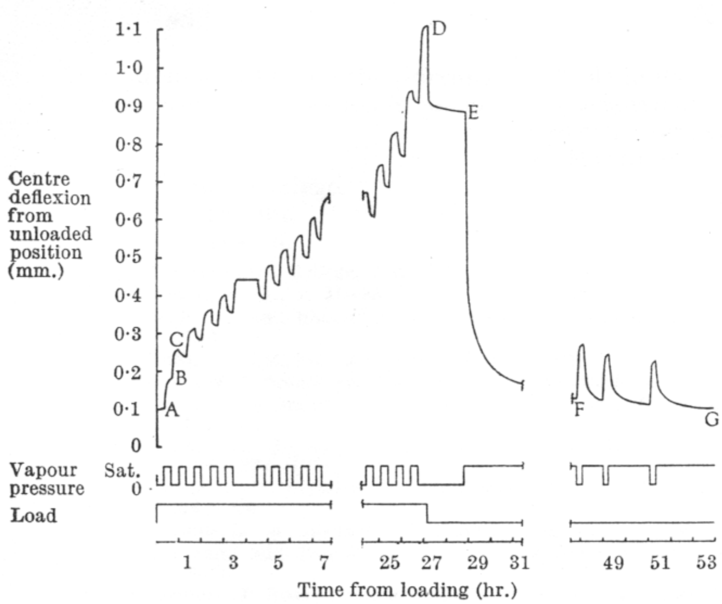


Fig. 2. *Effect of changes in moisture content on bending of wooden beams under load according to Armstrong and Christensen [4].*

When the specimens were loaded in the dry state, very slow creep was observed after the initial, elastic deflection. During the first adsorption the deflection increased rapidly. The deflection increased further during the following desorption, and during subsequent moisture content cycles, the deflection of the specimens decreased during each adsorption and increased during desorption. The final result was a total deflection, that was several times larger than the initial deflection. According to the authors, the sorption rate did not affect the increase in deflection. Drying of never dried wood specimens confirmed the independence of sorption rate. If the range of the moisture content cycles was made smaller, the total deflection decreased, but the principal behaviour remained. If the deformed specimens were unloaded in the dry state, little recovery other than the initial deflection occured. However, if the moisture content was changed to the wet state after the unloading, a large recovery occurred. With further moisture cycling after unloading, the recovery was slightly enhanced.

Since the initial investigation, many experimental studies on mechano-sorptive creep in wood have been carried out and described in the literature [13–32]. Mechano-sorptive creep has also been found in wood based materials, like particle-board [33, 34] and hardboard [34].

The first observations of mechano-sorptive creep in paper were reported in 1972 by Byrd [5, 6], who studied creep in tension and compression, and showed that paper exhibits mechano-sorptive creep, Fig. 3. These studies were followed by many others [11, 12, 35–50].

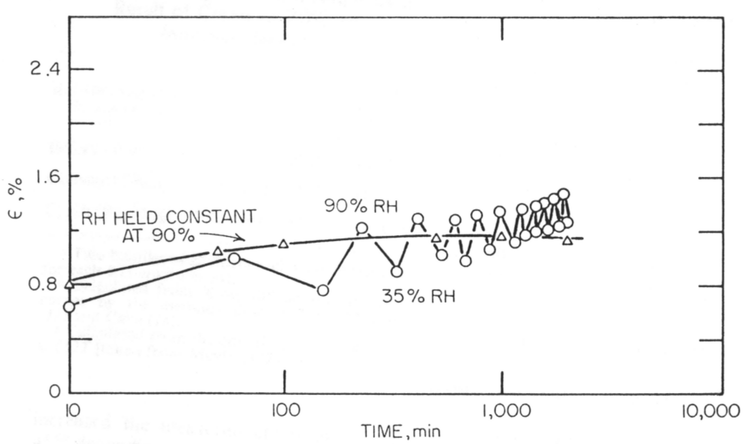


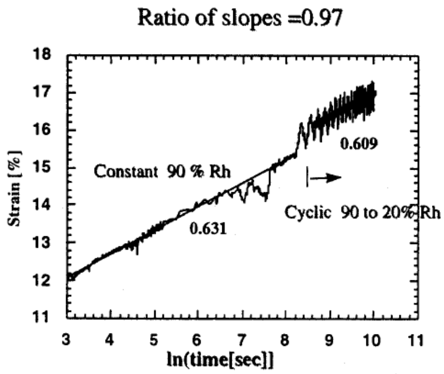
Fig. 3. *Creep response of paper specimens under tensile load in cyclic and constant humidity according to Byrd [5].*

Neither pure cellulose [43, 51] nor pulp fibres [46, 52] exhibit mechano-sorptive creep under the same conditions as wood and paper. Coffin and Boese [46] carried out creep tests in tension of single fibres and hand-sheets made of fibres from the same source, and found that the single fibres did not exhibit mechano-sorptive creep, while the hand-sheets did, Fig. 4. However, if the moisture cycles are adjusted so moisture gradients will arise in the test specimen at each absorption and desorption, both pure cellulose (cellophane) [47, 48, 53] and ramie fibres [53] show accelerated creep.

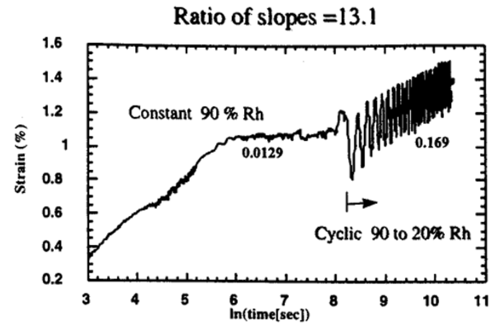
Mechano-sorptive creep was found in wool in the late 1950s [1, 2], and in some synthetic fibres in the 1990s: Aramid fibres, for example Kevlar [7–9, 48, 53–55], Kevlar/epoxy composites [7] and cellulose acetate butyrate [9] exhibit mechano-sorptive creep. A common property of these fibers is that they contain hydrogen bonds. Most researchers claim that nylon do not exhibit mechano-sorptive creep [7, 9, 41], but a (very) small effect is obtained if the moisture cycles are adjusted to the sorption time [53]. Concrete exhibits mechano-sorptive creep during drying [10].

Armstrong and Kingston [13] studied mechano-sorptive creep during bending of wooden beams and in tensile and compressive tests. Hearmon and Paton [14] investigated the creep of wood in bending and shear. Mechano-sorptive creep was found for all load cases. Armstrong and Kingston [13] also found good correlations between the range of moisture content changes and the additional creep taking place, and that creep would increase for moisture content changes in both directions.

Söremark *et al.* [11, 44] found that the creep rate was higher in compression than in tension for corrugated board under cyclic humidities. The corrugated board was loaded in pure bending. In order to make creep tests in pure tension or compression, the liner-board on



(a) Fibres



(b) Handsheets

Fig. 4. *Typical creep responses of (a) fibres and (b) hand-sheets according to Coffin and Boese [46]. The ratio of slopes during constant and cyclic humidity condition are used to quantify the acceleration of creep.*

one side of the corrugated board specimen was replaced with a steel ribbon.

Fellers and Panek [12] have found that creep in compression is accelerated by humidity cycles with very small amplitudes. They could not find a lower limit for the amplitude where the mechano-sorptive creep vanish. The increase in creep rate levels off as the humidity amplitude increases.

Experiments by Armstrong [19] showed that a constant moisture gradient, with a steady state flow of water, does not produce mechano-sorptive creep. A hollow specimen of wood was designed. An air pump, connected to the hollow core of the specimen, was used to get different humidities on the inside and on the outside, producing a constant moisture gradient through the specimen. Back *et al.* [36, 37] confirmed that moisture gradients by themselves do not produce mechano-sorptive creep in stress relaxation tests on hardboard and sack paper.

When the humidity is cycled, the increase in deformation during each cycle decreases as long as no failure takes place [14, 21, 24–26], see Fig. 5, and according to Hunt and Shelton the deformation eventually will reach a limiting value, which they called the *creep limit* [24–26], see Fig. 6.

Armstrong and Kingston [13] observed that microscopic failures developed in bending and compression at lower loads during moisture cycling compared to constant moisture, and Hearmon and Paton [14] found that their specimens broke at lower load if the humidity was cycled. Hoffmeyer and Davidson [27, 28] made an extensive microscopic study of wood specimens, and found that the number of microscopical compression failures in the fibre walls increases during mechano-sorptive creep.

According to Armstrong *et al.* [4, 13] mechano-sorptive creep is almost time independent – the sorption rate and length of cycles have little effect on the creep rate. The range of moisture content changes seems to be more important.

Gunderson and Tobey [38] carried out creep tests on paper in tension. They compared different cyclic humidity conditions, and found that the deformation after a given number of cycles was almost the same independent of the time spent to change humidity from one level to another and the time spent at each level. From this they drew the conclusions that mechano-sorptive creep is almost time-independent and that a stress gradient caused by a

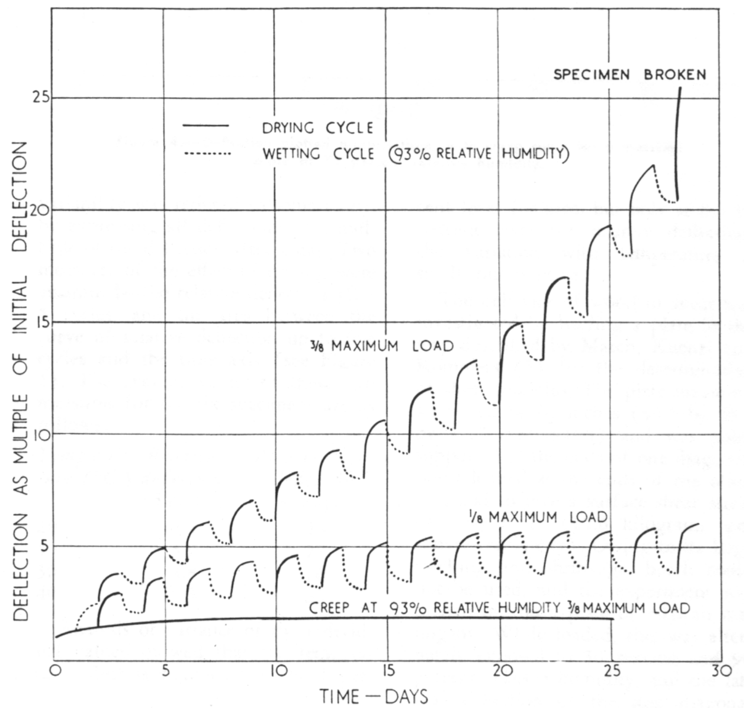


Fig. 5. *Deflection of wooden beams in the experiments of Hearmon and Paton [14].*

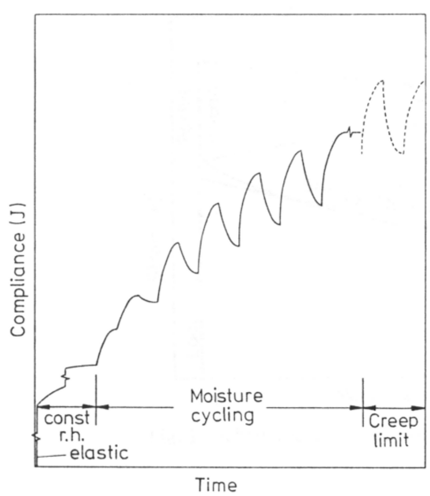


Fig. 6. *Schematic graph of creep experiment with moisture cycling according to Hunt [26]. The creep limit is shown to the right.*

moisture gradient within the web can not explain mechano-sorptive creep, as different rates of change should have created different gradients.

On the other hand, the sorption rate and cycle time have been found to be important by other researchers [1, 2, 36, 37, 48, 49]. Back *et al.* [36, 37] claim that the creep increases with increasing sorption rate. This claim is supported by results obtained by the authors [37], but also by experiments on wool [1, 2]. Coffin and Habeger [49] have reported that the sorption time and cycle time have to be of the same magnitude to get the maximum creep rate.

Padanyi [39, 40] carried out dynamic mechanical analysis (DMA) on liner-board, i.e. the paper used as faces in corrugated board. In DMA a cyclic strain is applied to the test specimens. In these tests, the strain lags behind the stress by an phase angle δ for a viscoelastic material, and a loss factor is defined as $\tan(\delta)$. Padanyi found a transient increase of the loss factor every time the moisture was changed. This is equivalent to an increase in creep compliance. Other researchers have got similar results [32, 41]. However, Salmén and Fellers [41] found the transient increases of the loss factor not only in paper but also in nylon, which does not exhibit mechano-sorptive creep. Hence they drew the conclusion that this kind of testing does not really reflect the material properties. Padanyi [39, 40] also found that by exposing paper samples to high humidity, the compliance of the material could be increased.

Olsson and Salmén [50] have used IR-spectra to measure molecular orientation in low grammage paper during creep. They found that different parts of the molecules were activated during creep at constant and cyclic humidity conditions.

Haslach *et al.* [42] tried to separate creep and hygroexpansion in mechano-sorptive creep tests, but they also state that this separation can be made in different ways. Söremark *et al.* [11, 44] found that the hygroexpansion changes during loading, giving more hygroexpansion in compression and less in tension. These studies show that there is no straight forward way to separate hygroexpansive strains from creep strains. Clearly the superposition principle does not hold when the moisture changes during loading. The applied stress seems to change the hygroexpansion. Söremark *et al.* [11, 44] introduced the term stress-induced hygroexpansion (SIH) to describe this.

The coupling between moisture content changes and stresses, and the difficulties to find a way to separate their influence suggest that mechano-sorptive creep is a result of some kind of non-linearity in the material. Habeger *et al.* [47, 48, 53, 55] have found that the creep of all the materials that exhibit mechano-sorptive creep have non-linear stress dependence. However, both regular and mechano-sorptive creep seems to be linear in stress at load levels encountered in applications [44, 56].

To sum up, mechano-sorptive creep is found in several materials: Wood, paper, wool, synthetic fibres and concrete. Creep is accelerated and failure is observed at lower loads if humidity is cycled. Changing moisture content is necessary for mechano-sorptive creep – a constant moisture gradient does not produce mechano-sortive creep. It is debated whether mechano-sorptive creep is time-dependent or not. Some of the observations suggest a non-linear mechanism.

2.2 Modeling

The experimental investigations considering mechano-sorptive creep are often combined with empirical models based on experimental data. These models are usually intended for a specific application, and hence they often have a limited applicability. Mechanisms explaining mechano-sorptive creep have also been proposed, but the effects of these mechanisms are however rarely quantified, and none of them has been generally accepted.

The reasons for modeling mechano-sorptive creep of paper are to be able design packages and predict their life-time and to be able to optimize the material.

2.2.1 Empirical models

Simple empirical models describing mechano-sorptive creep may be obtained by fitting simple equations, for example linear functions of logarithmic time, to experimental data [7–9, 21, 54]. Models of this kind can be used only to quantify the mechano-sorptive creep and compare different materials.

Leicester [16] proposed a rheological model for mechano-sorptive creep in wooden beams during drying. This model was a modified Maxwell model for viscoelasticity, where the dashpot element had been substituted by a special mechano-sorptive creep element.

In the model, the total deflection Δ of a wooden beam was assumed to be the sum of an elastic component Δ_e and a mechano-sorptive component Δ_m according to

$$\Delta = \Delta_\mathrm{e} + \Delta_\mathrm{m} \tag{1}$$

$$\Delta_\mathrm{e} = KP \tag{2}$$

$$-\frac{\mathrm{d}\Delta_\mathrm{m}}{\mathrm{d}m} = Pf(m) \tag{3}$$

where P is a load parameter, K is a constant, m is the moisture content and $f(m)$ is a function of moisture content. Validation showed that the behaviour of drying wood could be described by this model [16–18]. The model is only valid during drying.

More sophisticated models based on experimental data are achieved if constitutive relations between strain and stress are developed. Ranta-Maunus [20] developed a generalized viscoelastic model for wood that includes moisture changes. In the one-dimensional case the constitutive equation is given by

$$\begin{aligned} \varepsilon(t) = & \int_0^t J(t-\tau)\,\mathrm{d}\sigma(\tau) \\ & + \int_0^t \left(K(t-\tau)\sigma(\tau) + L(t-\tau)\left(\sigma(t)-\sigma(\tau)\right)\left(u(t)-u(\tau)\right)\right)\mathrm{d}u(\tau) \end{aligned} \tag{4}$$

where ε is the strain, σ is the stress, u is the moisture content, J is the viscoelastic compliance, K and L are kernel functions describing the mechano-sorptive behaviour and t and τ are time variables. In Eq. (4), the material is assumed to be unloaded up to the time $t = 0$. Different values of K during absorption and desorption are necessary to describe mechano-sorptive creep.

Following Ranta-Maunus' ideas, Mårtensson [29, 57] developed an incremental strain-stress relationship for wood. In the one dimensional case the total strain, ε, is given by the strain rate equation

$$\dot{\varepsilon} = \dot{\varepsilon}^\mathrm{e} + \dot{\varepsilon}^\mathrm{s} + \dot{\varepsilon}^\mathrm{c} + \dot{\varepsilon}^\mathrm{ms} + \dot{\varepsilon}^\mathrm{msr} \tag{5}$$

where ε^e is the elastic strain, ε^s is the swelling strain, ε^c is the creep strain, ε^ms is the mechano-sorptive creep strain and ε^msr is the mechano-sorptive recovery strain. For the mechano-sorptive creep strain, Mårtensson proposed a relation between strain, stress and moisture content given by

$$\dot{\varepsilon}^\mathrm{ms} = m\sigma\left|\dot{u}\right| \tag{6}$$

where σ is the stress, $\dot{u}$ is the sorption rate and m is a mechano-sorptive parameter. The parameter m is assumed to depend on the current moisture content u, but also on its history.

The mechano-sorptive recovery strain in Eq. (5) is given by

$$\dot{\varepsilon}^{\mathrm{msr}} = -L\left|\frac{\sigma^{*}-\sigma}{\sigma^{*}}\right|\varepsilon^{\mathrm{mst}}\dot{u} \tag{7}$$

where $\varepsilon^{\mathrm{mst}} = \varepsilon^{\mathrm{ms}} + \varepsilon^{\mathrm{msr}}$, σ^{*} is the stress averaged from the time when $\varepsilon^{\mathrm{mst}}$ was zero to current time, and L is a mechano-sorptive recovery parameter, which is assumed to be a positive constant if the moisture content increases and the current stress is less in magnitude than or opposite to the average stress. Otherwise L is zero.

The strength of the constitutive models developed by Ranta-Maunus and Mårtensson is that they are relatively easy to apply to real problems as they treat the material as a continuum. Similar models have later been developed for different applications [30, 31, 45, 58].

Urbanik [45] suggested a model to describe deformation of corrugated board subjected to a constant compression load during humidity cycling. In this model the total deformation is the sum of the hygroexpansion $X_{\mathrm{h}}(t)$ and the mechano-sorptive creep $X_{\mathrm{c}}(t)$. The mechano-sorptive creep function was derived from the assumptions that the mechano-sorptive creep rate is proportional to the magnitude of the rate of change in moisture content and that the moisture content is proportional to the hygroexpansion, leading to the equation

$$\frac{\mathrm{d}X_{\mathrm{c}}}{\mathrm{d}t} = \mu\left|\frac{\mathrm{d}X_{\mathrm{h}}}{\mathrm{d}t}\right| \tag{8}$$

where μ is a mechano-sorptive creep constant. In this model the load is included in the constant μ. The model was found to fit experimental data well.

2.2.2 Physically based models

The empirical models presented may be useful in some applications, but they can not explain the mechano-sorptive effect. To do this, the mechanisms behind the behaviour have to be found. The mechanisms suggested in the literature can roughly be divided into mechanisms that increase the creep compliance of the material and mechanisms that increase the loads on the material.

Nordon [2] suggested that hydrogen bonds in the material are broken during moisture sorption. This will lead to increased molecular mobility. Nordon further assumed that there is an equilibrium molecular state at each moisture content level. It will take the molecules a certain time to reach this state. The increased mobility will lead to accelerated creep, but when the equilibrium state is reached the creep slows down again. Similar ideas have been adopted by many researchers and many models based on bond breaking and molecular mobility have been suggested. Gibson [15] proposed that a non-equilibrium state will occur during absorption and desorption, in which hydrogen bonds will be broken and recreated by the interaction with migrating water molecules. This breaking and recreation will cause the material, in this case wood, to be more compliant during moisture changes, as the molecular mobility increases.

The hydrogen bond model was questioned by Armstrong [19] and Grossman [59]. They claimed that if the hydrogen bond mechanism causes mechano-sorptive creep, then moisture transport itself would be enough to give the effect. However, the experiments by Armstrong

[19] showed that steady state flow does not produce accelerated creep, consequently the hydrogen bond mechanism could not be responsible for mechano-sorptive creep.

The hydrogen bond theory was then developed to take the result of Armstrong's experiments [19] into account. According to Bažant [60] a common property of materials that exhibits mechano-sorptive creep is that they are porous with a broad range of pore sizes. Bažant suggests that water molecules in very small pores, which he called micro-pores, are strongly bounded with hydrogen bonds to the pore walls, thus taking up a large portion of an applied load. However, if moisture is transported through these micro-pores the bonds are disrupted, leading to accelerated creep. This will happen both in absorption and desorption, as water flows to or from larger pores. To explain the lack of mechano-sorptive creep during steady state flow, it was suggested that a steady state flow gives moisture transport only in larger pores of the material, and hence no disruption of hydrogen bonds.

A simpler explanation is that the hydrogen bonds are formed and disrupted at the same rate during moisture transport without changes in moisture content, leading to equilibrium, and hence no mechano-sorptive creep takes place [61]. A change in moisture content disturbs the equilibrium, which will lead to accelerated creep.

Schaffer [62] and van der Put [63] also claim that moving water will change the compliance of the material by bond breaking. However, in their models they include the moisture content change in a way similar to Ranta-Maunus [20]. This is done without much physical motivation, but the problem with steady state flow is avoided.

Materials like polymers have a glass transition temperature, where the equilibrium molecular configuration changes. If the material is cooled rapidly through this temperature it is possible that the equilibrium state is not reached immediately. Instead an excess volume, called the *free volume*, is locked in the material. The free volume increases the molecular mobility, and hence the creep compliance of the material. Gradually the molecule configuration will reach the equilibrium and the compliance will decrease. This process is called *aging*. The material can be *de-aged* by raising the temperature above the transition temperature and then lowering it again. It has been suggested that rapid moisture content changes also create free volume, and that this is the reason for mechano-sorptive creep [39, 40, 54].

Padanyi [39, 40] found that exposure of paper to high humidity had an effect similar to the de-aging procedure described above. The glass transition temperature is lowered by high moisture content, so it is actually possible to de-age paper by raising moisture instead of temperature. Based on this, Padanyi argues that mechano-sorptive creep is an effect of de-aging. It is however possible to get the same effect when the humidity is lowered. Padanyi assumed that de-aging takes place in this case also, but this conclusion is not supported by the original free volume theory, as pointed out by Habeger and Coffin [48].

During mechano-sorptive creep in bending and compression, local compression failures take place [13, 27, 28]. Microscopic compression failures in the fibre wall form distinct planes, called *slip planes*, which increase the compliance of the material. Hoffmeyer and Davidson [27] proposed a slip plane mechanism for mechano-creep in wood. The number of slip planes is a function of stress level, moisture content and duration of load. Hoffmeyer and Davidson suggested that moisture content changes should be added to this list. The theory was verified by experiments [27, 28], but it is not possible to explain mechano-sorptive creep in tension with this theory. It is however possible that different mechanisms explain mechano-sorptive creep in compression and tension [61].

In the early works by Pickett [10], Mackey and Downes [1] and Armstrong and Christensen [4] it was suggested that transient stresses are introduced in the material during changes in moisture content, adding to the stresses caused by external load, thereby giving an increase creep. Pickett [10] and Mackay and Downes [1] assumed that a swelling gradient causes these stresses as moisture penetrates the material. Pickett shows that this gradient in combination with non-linear material behaviour leads to additional deformation. Armstrong and Christensen [4] claim that their smallest specimens were too small for any moisture gradients to occur, but they assumed that there might be other ways to introduce transient stresses in the material. These ideas were later developed by other researchers [11, 36, 37, 44, 47–49, 55].

Back *et al.* [36, 37] assumed that moisture gradients always are present during moisture changes. In this case the differences in swelling between different parts of the material would give an uneven stress distribution, that may produce accelerated creep. The differences in stress relaxation rates between areas with different stresses will eventually even out the stresses. Thus the moisture gradient have to change to maintain the accelerated creep.

Söremark *et al.* [11, 44] suggested a model combining the slip plane model [27] and transient stresses during changes in moisture content. In paper dislocations, or kinks, are created during the papermaking process and during drying. The dislocations affect the mechanical properties of the paper. For example the creep rate is increased. Söremark *et al.* proposed that these dislocations are responsible for the accelerated creep during moisture cycling. If paper is subjected to static load during moisture cycling, swelling and deswelling occurs, leading to redistributions of stresses within the fibres and the fibre network. In compression every moisture change will increase the number of dislocations according to Söremark *et al.* The increase in dislocations causes length decrease in the direction of loading and increases creep rate, which together lead to very fast creep in compression. In tension the dislocations decrease, which causes length increase but decreases the creep rate. These two effects counteract each other, but according to the authors the result is a slightly increased creep rate. Thus the model explains mechano-sorptive creep, and explains the differences between compressive and tensile loading.

A mechanism combining transient stresses with free volume increases was proposed by Haslach [43, 64]. Haslach discusses how the anisotropic hygroexpansion of the fibres in paper will cause additional stresses at the interfibre bonds. According to Haslach the hygroexpansion is increased by free volume created by moisture changes. This will further increase the stresses at the bonds, which lead to disruption of the bonds, which in turn will lead to a weaker material and higher creep compliance.

The mechanism proposed by Söremark *et al.* [11, 44] was also suggested by Coffin and Boese [46] as a possible explanation for the difference between single fibres, which do not exhibit mechano-sorptive creep and have few dislocations, and hand-sheets, which do exhibit mechano-sorptive creep and have many dislocations due to the drying process. Another possible explanation to this difference in creep behaviour is that accelerated creep is caused by the non-linear creep behaviour of the fibres in combination with large transient stresses, which do not appear in single fibre creep [46–48, 55]. This is basically a further development of Pickett's ideas [10]. At each change of moisture content, stresses are redistributed because of variations of hygroexpansion in the material, caused by moisture gradients or material heterogeneities. This will lead to higher stresses at some parts of the material and lower at other. Because of the non-linear creep behaviour the increase in creep rate in areas with high stresses will be larger than the decrease in the areas with low stresses, leading to higher average creep rate. The process is repeated at each change of moisture content.

Habeger and Coffin [47, 48, 55] have showed that a very simple rheological model can exhibit accelerated creep if the model includes variations of hygroexpansion. Their model is shown in Fig. 7.

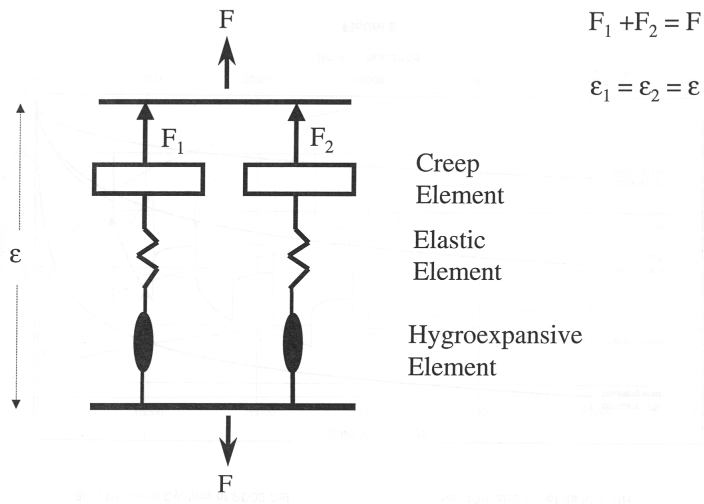


Fig. 7. *The mechanical model for moisture cycling problems suggested by Habeger and Coffin [48].*

The model in Fig. 7 consists of two sections, which represent different parts of the material. Each section is composed of a creep element, an elastic element and a hygroexpansive element. The two sections are restrained to deform together. The strain rate of each section is given by

$$\frac{\mathrm{d}\varepsilon}{\mathrm{d}t} = \frac{1}{E}\frac{\mathrm{d}\sigma}{\mathrm{d}t} + f_{\mathrm{t}}(\sigma, \varepsilon_{\mathrm{c}}) + \beta\frac{\mathrm{d}m}{\mathrm{d}t}, \tag{9}$$

where indices have been left out for simpler notation and the stress σ is used instead of the forces F_1 and F_2 in Fig. 7. In Eq. (9), E is the elastic modulus, $f_{\mathrm{t}}(\sigma, \varepsilon_{\mathrm{c}})$ is the creep rate, a non-linear function of stress and possible the creep strain ε_{c}, β is the hygroexpansion coefficient and m the moisture content.

The model by Habeger and Coffin will exhibit accelerated creep if the creep law,

$$\frac{\mathrm{d}\varepsilon_{\mathrm{c}}}{\mathrm{d}t} = f_{\mathrm{t}}(\sigma, \varepsilon_{\mathrm{c}}), \tag{10}$$

is non-linear in stress σ, and the hygroexpansive strains βm in the two sections are different in magnitudes or out of phase. Material heterogeneities would give different magnitudes while a moisture gradient would give a phase difference. There are many possible forms of the function $f_{\mathrm{t}}(\sigma, \varepsilon_{\mathrm{c}})$, and Habeger and Coffin used

$$f_{\mathrm{t}}(\sigma, \varepsilon_{\mathrm{c}}) = k\sigma^n \tag{11}$$

$$f_{\mathrm{t}}(\sigma, \varepsilon_{\mathrm{c}}) = \sigma A B_0 e^{\alpha\sigma} e^{-\varepsilon_{\mathrm{c}}/A\sigma} \tag{12}$$

$$f_{\mathrm{t}}(\sigma, \varepsilon_{\mathrm{c}}) = a(C_0\sigma)^{1/a} e^{\alpha\sigma} (\varepsilon_{\mathrm{c}})^{(a-1)/a} \tag{13}$$

where k, n, A, B_0, C_0, α and a are material parameters. Eq. (11) is simple and useful for discussion on the general behaviour of the model, but this equation will not give a creep behaviour that resembles paper very much. Hence Habeger and Coffin continued to Eqs (12–13), which they derived from creep curve fitting equations by Brezinsky [65]. These creep laws do however behave very strange if the sign of the stress changes [48]. None of Eqs (11–13) include creep recovery.

With their model, Habeger and Coffin were able to simulate the accelerated creep of paper during moisture cycling. The measured accelerated creep of TMP samples is shown in Fig. 8, and Fig. 9 shows the simulated accelerated creep. In the simulation the creep rate was assumed to be given by Eq. (13) and a moisture gradient, i.e. a phase difference in the cyclic moisture content of the sections, creates the necessary stress redistribution.

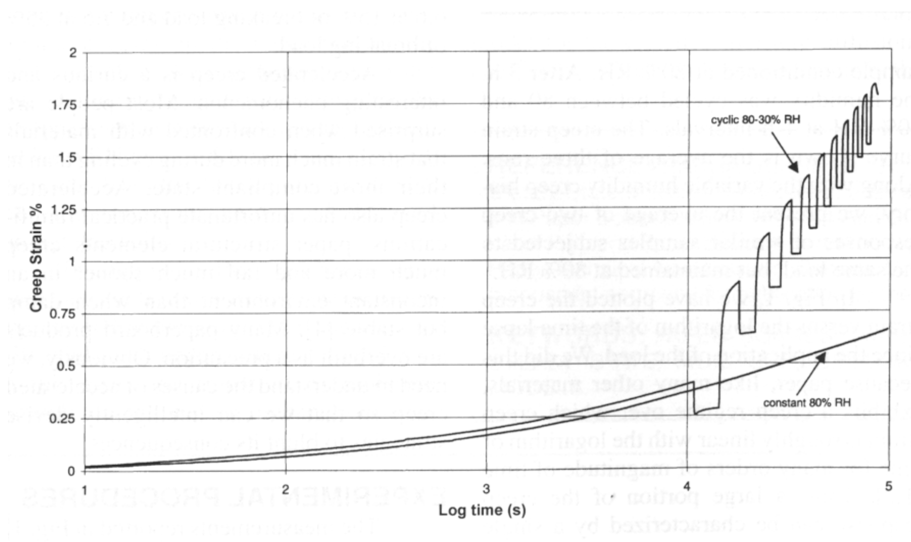


Fig. 8. *Measured accelerated creep of TMP samples [48].*

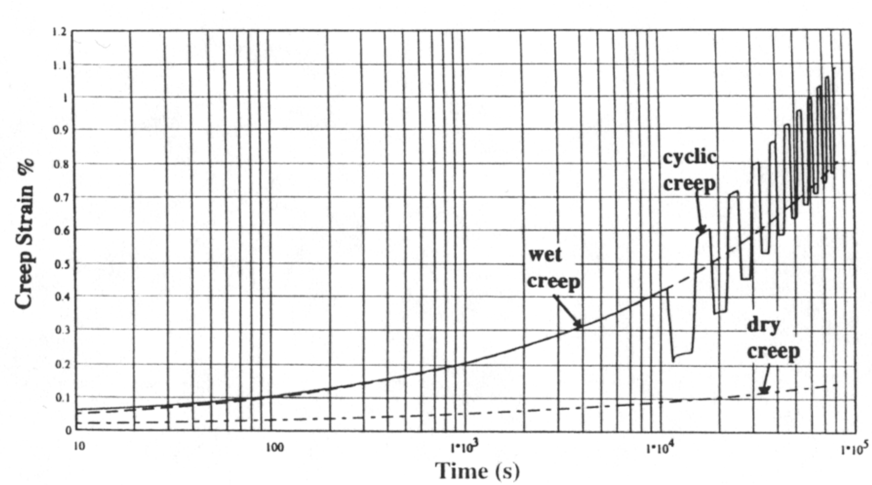


Fig. 9. *Simulation of accelerated creep of TMP [48]. The accelerated creep was assumed to be driven by a moisture gradient and a creep law according to Eq. (13) was used.*

3 Micro-mechanical modeling of mechano-sorptive creep

In this thesis it is assumed that mechano-sorptive creep is a result of inhomogeneous hygroexpansion and non-linear creep as suggested by Pickett [10] and Habeger and Coffin [48]. Similar models have also been suggested for metal-matrix composites during temperature cycling [66, 67]. The major advantage in comparison to other models is that accelerated creep turns out to be a natural consequence of regular creep and not a completely new phenomenon.

In all constitutive modeling, an appropriate length scale must be chosen, see Fig. 10. At the largest scale, a box is considered. Few, if any, models for mechano-sorptive creep uses this length scale. Continuum models [20, 45, 57, 58] can be used to model the material, in this case the corrugated board or liner. The model of Habeger and Coffin constitutes an intermediate step between continuum and the fibre network, as some kind of heterogeneity is needed. Slip plane models [11, 27, 28] are based on a smaller scale, fibre network or fibres, while the hydrogen-bond model [15] and free volume model [39, 40] fall on the molecular level.

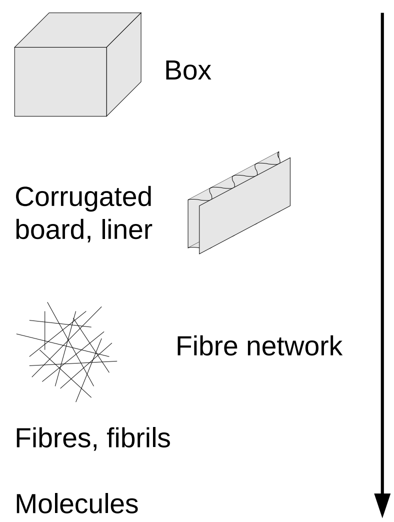


Fig. 10. *Different length scales, going from the box at the top down to molecules. An appropriate level must be chosen for modeling.*

The fibre network seems like a suitable level for modeling of mechano-sorptive creep. The continuum models for corrugated board or liner do not explain the physics behind mechano-sorptive creep. Models on fibre level or below are probably not necessary as neither the fibres themselves nor pure cellulose exhibit significant accelerated creep [46, 51, 52]. In this work, the hygroexpansion heterogeneity at the bonds is assumed to drive the accelerated creep.

In Papers A and B, two different micro-mechanical models for mechano-sorptive creep are developed. In Paper A, shear stresses in the bond interfaces between the fibres are assumed to cause the accelerated creep. A model of a fibre-fibre bond is developed, and included as a bonding element in a random fibre network model. In Paper B, normal stresses are redistributed between the fibres at the bonds. The analysis is simplified by assuming that the average strain of any fibre is equal to the macroscopic strain of the paper in the direction of the fibre. Both models show accelerated creep similar to that of paper. The model in Paper A is very sensitive to the rate of moisture content changes, while the model in Paper B is rather insensitive.

The model in Paper B predicts that the acceleration of creep is very small for small moisture cycle amplitudes, then increase rapidly for intermediate amplitudes and finally increase slowly for large amplitudes. In Paper C the effect of moisture cycle amplitudes was investigated experimentally. The prediction from Paper B is verified, and a quite good fit between the theoretical model and the experimental creep curves is obtained.

In Paper B it is also found that the deformation of the network model is proportional to the applied mechanical load for small loads typically found in applications, i.e. the accelerated creep is linear in stress although non-linear creep on fibre level cause the acceleration. This is in accordance with experimental results, see for example Panek *et al.* [56]. It is found that the applied mechanical load produces stresses that are an order of magnitude smaller than the internal stresses produced by the moisture content changes. In Paper D, the applied loads are hence treated as perturbations to the stress state caused by moisture changes, and a linearized model for mechano-sorptive creep of paper is derived. Furthermore, the linearization makes it possible to derive a continuum model from the network model. The number of variables and equations is reduced, and consequently the speed of calculations is increased. The linearized model is valid for loads found in many applications. The continuum model can be used as a constitutive law in problems with complex geometries, for example corrugated board boxes exposed to varying humidity.

4 Conclusions and discussion

The work presented in this thesis shows that mechano-sorptive creep may be a result of transient stresses produced by heterogeneous hygroexpansion at the fibre-fibre bonds in paper in combination with creep behaviour that depends non-linearly on stresses. The major advantage of this mechanism is that mechano-sorptive creep turns out to be a natural consequence of regular creep. It has been demonstrated that the accelerated creep from this model is linear in the stress applied to the paper, as is also found in experiments. This feature has been used to derive a linear constitutive model.

However, there are some problems that have to be addressed to get an accurate description of mechano-sorptive creep. There is a need for accurate non-linear creep laws for fibres and paper, and reliable methods to find parameters for these creep laws. The residual stresses at the bonds from drying processes have been neglected in this work. These stresses may be of the same magnitude as the internal stresses produced by the moisture content changes, and could then have a large influence on the creep behaviour.

It has not been shown that the mechanism presented here is the only possible mechanism for mechano-sorptive creep, although it is very likely to give a major contribution to the acceleration of creep during moisture changes. The difference between compressive and tensile creep has not been investigated. It might be a structural effect of the network. The difference is also to a lesser extent present in regular creep, so it might be the same effect that is enlarged by the creep acceleration.

5 Summary of appended papers

Paper A: A micro-mechanical model for mechano-sorptive creep in paper

In this paper, stresses created at bonds due to anisotropic swelling during absorption and desorption of moisture, in combination with non-linear creep, are proposed to be the cause for mechano-sorptive creep. Two simplified models are first discussed in order to demonstrate the suggested mechanism. A three-dimensional fibre network model composed of elastic fibres and inelastic bonds is then studied by finite element calculations. The relative sliding in the bonds is described by a non-linear creep model which in combination with anisotropic fibre hygroexpansion results in accelerated creep of the network.

Paper B: A simplified network model for mechano-sorptive creep in paper

A simplified network model for mechano-sorptive creep is presented. The model resembles Cox's model for fibrous materials, but creep and influence of bonds are included in addition to the elastic behaviour of the fibres. Three different creep laws describing the creep of individual fibres are applied in the simulations of creep of the network.

Results from simulations using the model are presented. The influence of the amplitude of moisture content changes is discussed. It is shown that the model may produce macroscopic strains that are linear in stress, even though the creep of the fibres is non-linear. This may explain why both regular creep and mechano-sorptive creep at small loads appear to be linear in stress.

Paper C: The effect of humidity cycle amplitude on accelerated tensile creep of paper

In this paper, the effect of different amplitudes of the moisture content on mechano-sorptive creep is investigated experimentally and numerically.

Tensile creep tests are made in a climate chamber. Low basis weight isotropic sheets are used in the tests. The moisture content history is measured during each creep test using a balance placed in the climate chamber.

The experimental results are compared to predictions using a theoretical network model. A brief description of the model is given. In the model it is assumed that the anisotropic hygroexpansion of the fibres produces large stresses at the fiber-fiber bonds when moisture changes. The resulting stress state will accelerate creep if the material obeys creep laws that are non-linear in stress. A good agreement between the theoretical model and the experimental creep curves is obtained.

Paper D: Linear constitutive model for mechano-sorptive creep in paper

It is assumed that mechano-sorptive creep is an effect of transient stresses produced during moisture content changes in combination with non-linear creep behaviour of the fibres. The stresses produced by the moisture content changes are often much larger than the applied mechanical loads. If this is the case, the mechanical loads are only perturbations to the internal stress state, and it will appear as if the mechano-sorptive creep is linear in stress. It is possible to take advantage of this feature. In the present report the pure moisture problem is first solved.

The mechanical load is then treated as a perturbation of the solution to the moisture problem. Using this strategy, it is possible to linearize a non-linear network model for mechano-sorptive creep and to formulate a continuum model. As a result, the number of variables in the model is reduced. This is a significant improvement as it will be possible to use the linearized model to describe the material in a finite element program and solve problems with complicated geometries.

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