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DEVELOPING A DEEPER UNDERSTANDING OF THE CONSTITUTIVE BEHAVIOR OF PAPER

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ABSTRACT

A constitutive equation for the mechanical behavior of paper is presented. Since the initial work in the late 1940's rheology studies of paper seem to come in waves and the results have not always been viewed favorably. With the benefit of fifty years of literature and the relative speed of modern computing, a more robust constitutive model is certainly within reach. The presented model captures the essence of the mechanical response and links the stress-strain curve to relaxation and creep in a reasonable manner. Experimental results are provided to illustrate the important components of the behavior and the model can easily be generalized to include more subtle features.

INTRODUCTION

How far has the mechanics of paper progressed in the last fifty years? The effects of process variables such as pressing, refining, and drying shrinkage on the elastic modulus and tensile strength are largely heuristic, and one questions general applicability. Computers have made it possible to simulate papermaking, converting, and printing but are limited by the employed constitutive equations. New experimental methods give detailed insights into paper structure that were previously only imagined, but applying these tools to gain insights into the mechanical behavior has only just begun. We sit on the verge of a renaissance in the science of paper; yet we have not fully unraveled the fundamentals of its mechanical response: even something as simple as the stress-strain curve is largely a mystery or in the least cause for debate. It is time that we develop this deeper understanding so that the advanced tools of the 21st century can be fully utilized. This paper focuses on developing a more robust constitutive equation for paper that could be utilized for characterization and modeling.

The essential features required to model paper were clearly defined fifty years ago. A gathering of scientists at the first FRC symposium was bent on understanding the mechanical response of paper and attributing observations to our beloved fibers and bonds and the actions of beating and shrinkage. In his critical review, Steenberg [1] stated

> *Most people will associate the concept of mechanical properties of paper with properties like bursting or tensile strength, tear resistance or folding endurance. The mechanical properties of paper is of course a much wider subject.*

He continued with

> *. . . At the present advanced state of paper technology, current technological ultimate strength testing procedures no longer give information sufficiently detailed and highly correlated with paper properties as exhibited in actual use. Very few, if any, of these tests measure properties having sufficiently well defined physical interpretations to enhance theoretical analysis of the influence of beating on the mechanical properties of paper. This is much more the case with tests of the nondestructive type, determining properties like stiffness, modulus of elasticity, etc. This latter field, holding good promises for rapid theoretical and practical results, has not been studied to the extent its significance seems to justify.*

Those statements stand equally well today. Fifty years later, most papermakers still view mechanical properties in terms of measures of strength. Elastic modulus may be the main exception having found its way into the hearts of some papermakers. When dealing with papermaking processes, converting operations, and end-use performances, one is in general pleased to keep the paper from failing. Therefore, strength measurements serve only as limits but cannot be used for evaluations of real processes. As pointed out by Steenburg [1], pre-rupture response of paper is of most interest and relevance.

Furthermore, the theorist should be concerned in modeling pre-rupture behavior from which prediction of failure naturally follows. The sophisticated modeling that is available today will be most useful once the proper constitutive equations are incorporated into them. Whereas micromechanical modeling yields insights but lacks applicability, brute force computational modeling of networks is somehow unsatisfying and yields little insight.

By reading the proceedings of the first FRC symposium, one learns many things about the mechanical response of paper. For example, considering the time-dependent nature of paper is necessary [1], and the study of the shape of the stress-strain curve can be insightful [2]. Experimental results demonstrated that reflectance increases as paper is strained and especially during yielding indicating increase in surface area and implying loss of bonding [3]. Furthermore, drying stresses and shrinkage play a large role in property development [4]. Of more interest in this first symposium are the comments in the discussion section of these papers [1–4], where one gains an appreciation for the thorough understanding of the stress-strain behavior that these scientists possessed while at the same time realizing there was much uncertainty and disagreement amongst them.

The following passage from a discussion between Steenburg, Van den Akker, and Andersson illustrates the disposition at the time. The discussion was centered on the idea of using critical velocity as some test independent quantification of mechanical response.

> DR. VAN DEN AKKER: *There remains the important point are we interested in the particular combination of basic factors underlying the critical velocity? We should be more interested in some other combination, depending on the requirements.*
> MR. O. ANDERSSON: *I think that no specific combination has any preference. If so, why should not this one have it? We have just been told why it was chosen. On the other hand, I have a feeling that the argument is irrelevant.*
> PROF. STEENBERG: *As a matter of fact, I hope that someday we will not be interested in any combination at all; we will be interested in the elementary basic concepts.*
> DR. VAN DEN AKKER: *I agree with that.*

Although they held different views of how to approach modeling the mechanical behavior of paper, they both expressed a hope that someday in the future the focus would be on fundamentals. Full fundamental understanding of the tensile behavior of paper is not yet in our grasp. There are many complicating factors and uncoupling each one in a systematic manner will be a long and tedious process. I do not possess this deeper understanding, and I consider myself a typical Paper Physicist of the 21st century. Therefore, I believe it necessary to go back to the beginning, further investigate the constitutive behavior of paper and develop a more fundamental understanding.

The contribution made in this article, is what I believe the first necessary step to gaining a deeper understanding of the mechanical response of paper. Structural theories, micromechanics, and network models will likely provide the final steps to getting the "elementary basic concepts", but we are still missing a continuum model that captures the basic essence of the tensile behavior. The following is not focused on fitting empirical data or trying to understand the effect of process or structural parameters. Neither is it geared towards assigning responsibility of behaviors to fibers, bonds, materials, or structure. The focus is on developing a constitutive equation of paper that captures the essence; as a continuum. One could argue that this is not fundamental and what science could possibly develop from this. My response is that the mechanical behavior of paper is a product of its constituent materials as affected by fiber and sheet structure, and before the individual influences can be properly accounted for, one should be able to adequately express the effective response with some physics and mathematics.

BACKGROUND

Take any paper and conduct a constant rate of deformation tensile test, the results are surprisingly similar with exceptions being for papers that have undergone a process to create severe structural changes as in creping. The initial response between deformation and load is fairly linear. At some point, the tangent to the load versus deformation curve decreases substantially giving the impression that the paper has yielded. After yielding, the paper typically shows strain hardening up to the point of failure. If one were to plot slope versus deformation it would typically decrease with increasing strain. The tensile stress-strain curves are similar in nature to many materials, and one cannot really glean the necessary information needed to develop an appropriate model. Figure 1 provides an example showing typical results for constant rate of deformation tensile tests for a printing paper. Both MD and CD curves are shown as well as the curves for three different rates of strain. These curves are typical stress-strain curves for paper. A cursory glance shows that MD and CD can be drastically different and the faster the rate of deformation the stiffer the response. Also one notes, that a single stress-strain curve does not suffice, and is just a single representation of the more complex mechanical response of the paper. It is not until a change in the loading history is instituted that the paper reveals its true behavior.

From a historical perspective, the decade leading up to the first FRC

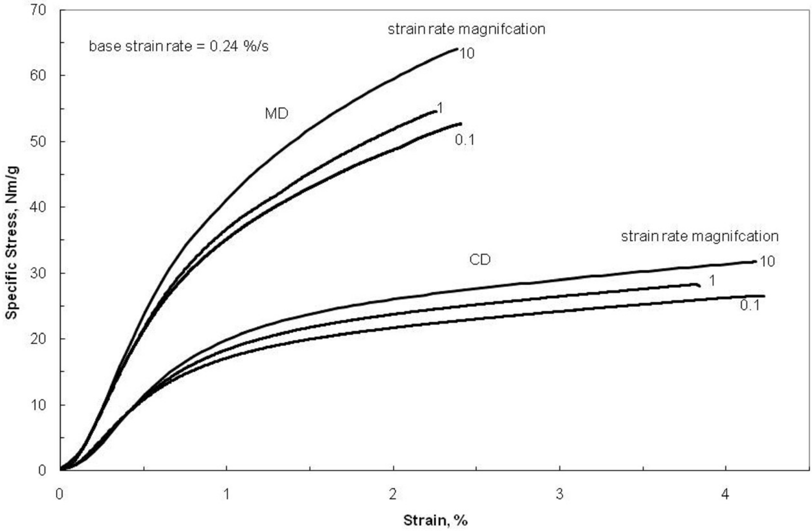

Figure 1. Example of MD and CD stress-strain curves at three magnifications of strain-rate. (A 107 g/m^2 printing paper.)

conference was the era dedicated to the rheological treatment of paper. There was a flourish of activity by researchers conducting tests, reporting and interpreting results, and applying models [5–14]. It is from this work that much of the insights can be determined. For example, Steenberg [5] showed that paper exhibits many of the classic behaviors observed for materials classified as viscoelastic including: stress-strain hysteresis in loading and unloading, stress relaxation, creep, and stress and strain recovery. In addition, paper exhibits both recoverable and unrecoverable deformation. Figure 2, illustrates the behaviors reported in [5] that would be observed when one begins to vary the loading history of paper.

The graph in Figure 2, illustrates a tensile response commonly observed for paper. The solid loading curve A to B would be the first-loading curve for the case of a constant rate of deformation and if not interrupted would continue along its path until failure. If at point B, the rate of deformation is reversed the paper would unload to a load of zero (C). The path from BC shows that there was potential energy stored in the sheet at B it is released upon unloading. Part of the deformation is recoverable. If the load is allowed to remain zero, strain will continue to recover from C to D, indicating a delayed-elasticity. The strain represented by the length AD is the permanent set and indicative of inelastic or plastic deformation. If the paper is then re-loaded at

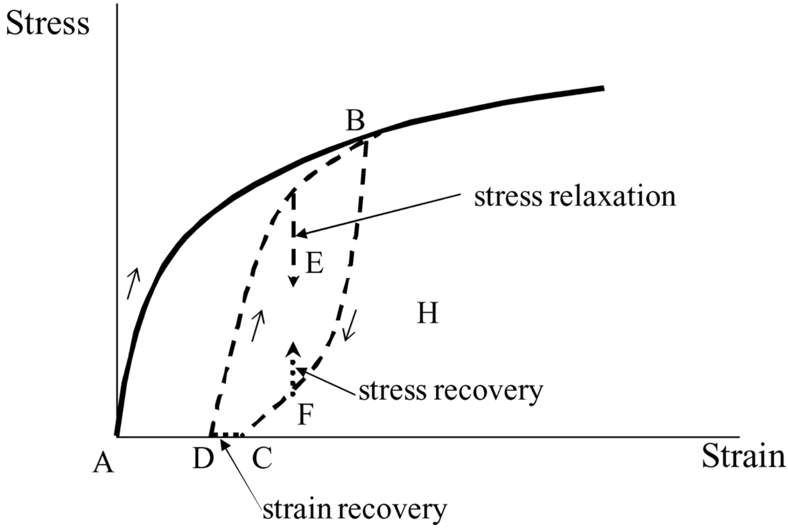

Figure 2. Observed mechanical responses of paper in tensile testing.

the same constant rate of strain, the tensile curve will resemble the initial loading but have a higher apparent yielding. If the strain is held constant at point E stress relaxation occurs. If during unloading the strain is held constant at point F, stress will recover. Upon reloading, the dashed curve will meet with the solid curve the two curves will coincide as strain increases. The fact that stress-relaxation and stress-recovery occur at the same strain level could be construed as indicative that there is a permanent delayed-elastic response in the material. This is not necessary and will be addressed later. Although not shown in Figure 2, paper also exhibits creep or elongation over time at fixed load level [13].

From the previous work [5–12] in the rheology era, requirements of a continuum model can be stated as follows:

- Paper exhibits both recoverable (elastic) and nonrecoverable (inelastic) deformation
- The elastic response has components with both long and short characteristic time scales.
- The mechanical response is time, moisture, and temperature dependent
- The mechanical response is not linear with load level.
- The apparent internal relaxation of stress is one that decays with the logarithm of time.

- In general, paper retains most of its properties even as it approaches failure.
- Existing rheological models are inadequate for general application.

Towards the end of the rheology era, Rance [12] implied that disappointment and skepticism had stymied the development of making gains in understanding the mechanical response of paper. He believed that this was because (1) methods were borrowed from other fields without allowances for differences in material, (2) rheology deals mainly with sub-fracture response and in paper failure is more commonplace, and (3) too much focus is placed on "arbitrary analysis of phenomenological curves portraying macrobehavior, with too little analysis in terms of observable behavior." He further states that it is impossible to attribute any fundamental significance to the shape of the stress-strain curve. He agrees that a nonlinear Eyring viscosity approach can be used to fit at least the recoverable response of paper, but suggests that one cannot ignore the inelastic or plastic deformation. Although Rance [14] says no model can capture the creep response, he does emphasize that the behavior is nonlinear, not fully recoverable, and likely tied to the nonlinearity in the stress-strain curve. In terms of stress relaxation, Rance points out the there is no reason to expect paper to asymptote to a constant delayed-elastic response. It is interesting that that he emphasizes that the relaxation processes in paper tend to decay with the logarithm of time, but on the surface seems against developing a model based on this evidence. Rance [14] supported a model based on progressive accumulation of damage in the sheet attributable to inter-fiber bond breaking.

Van den Akker [15] completed a review of the tensile behavior of paper in 1970. He stated that the viscoelastic models that were being developed in the rheology era of activity [1940's–1950s] had been abandoned because of "their failure to be heuristic and account for all the important phenomena." Upon reflection, it seems that the discontent voiced by Rance [14] and Van den Akker [15] is misplaced. It is not that the continuum approach is invalid, but the assumptions of the models were inadequate. Clearly, assuming the deformation is simply the addition of an instantaneous elastic response and a delayed-elastic response, even if is it nonlinear, will not capture the observed plasticity in paper. Furthermore, the delayed-elastic response will likely require a spectrum of relaxation times to be adequate. Part of the issue was that early researchers adopted existing three element models but did not generalize them. It is not that a phenomenological approach to model paper is unworkable. My current stance is similar to that voiced by Steenburg in the 1950's. The behavior of paper is generally well-behaved enough to be treated as a continuum, and if the appropriate behaviors are accounted for a reasonable model that captures the full mechanical response of paper should emerge.

During the latter half of the 1950's and the 1960's perhaps because of the seeming failure of modeling, work intensified on determining the role of fibers and bonding on the tensile response of paper. Many of the test results from the rheology era were suspect and inherently flawed due to the equipment in use at the time. For example, the practical work of Wink *et al.* [16] restored common sense to tensile testing when they refuted earlier claims [11, 14] that span length drastically affected the mechanical response of paper. As equipment and tests methods improve, the mechanical response must be re-evaluated. In the 1960's work did continue [17–20] but little progress on modeling was made.

At the third FRC symposium, Algar [21] provided a comprehensive review of work completed to that date on understanding and modeling the tensile response of paper. The focus was on how the models incorporate behavior in terms of fibers and bonding rather on the adequacy of the model to provide a prediction. There is a pertinent statement in the discussion that followed the paper [21].

> Dr O. L. Forgacs: *On listening to the discussion of the stress/strain theories of paper, I wonder whether we are not losing sight of the purpose of this work. There are surely two objectives, one short-term and one long-term. The short-term one is to arrive at a simplified and approximated theory for the stress/strain curve, which is immediately useful to applied problems. Therefore, any theoretical treatment, whether based on springs and dashpots or any other device, is quite acceptable, if and only if it fulfills a pragmatic function. The long-term objective is to obtain a true insight into the behaviour of the paper network under applied stresses. It seems to me that, to accomplish this, the arrangement and behaviour of the individual fibre elements and the bonds under stress must be thoroughly understood before a realistic model for theoretical treatment can be constructed. Is it not possible that the controversy today over the choice of models arises through our occasional neglect to define objectives?*

I believe that because the first objective was never really satisfied, the second has also suffered.

At the fifth FRC symposium, Dotson [22] presented an overview of paper mechanics geared towards the role of fundamental parameters. Ebeling [23] presented work on energy consumption during the straining of paper. He provides a good summary of proposed mechanisms for the inelastic deformation observed in the stress-strain curve as viscous flow, pre-rupture breakage of inter-fiber bonds, or shear deformation in the inter-fiber transfer of load.

His results indicate that the inelastic deformation is a thermodynamically irreversible process and linked to permanent structural changes in the material.

Two FRC symposiums later, Seth and Page [24] presented an insightful view of the role of bonding on the stress-strain curve of paper. In my view, this is one of the most significant contributions to the field. The work they presented clearly shows that the shape of the stress-strain curve is not significantly affected by changes in degree of bonding. The concept of a network efficiency factor that completely accounts for differences in the stress-strain curve due to bonding simplifies the whole concept of how to study paper. By analyzing the results of Brezinski [13], Coffin [25] demonstrated that the efficiency factor applies equally well to creep. DeMiao and Patterson [26] found the same result applied even in a cyclic humidity environment. In terms of modeling, it means that simply adding a load magnification factor to the equation accounts for the influence of bonding on the sub-fracture behavior. It also implies that scaling the stress by modulus has merit.

In the 1980's there was a renaissance of rheological studies of paper [27–34]. The work ranged from combining springs, dashpots, and sliders in different combinations [28, 29] to incorporating fitted creep compliance curves. These models were applied to either monotonic loading cases or creep and were not adequate at capturing the full effects of time and load. What emerged from this work once again was dissatisfaction with mechanical modeling of paper as a continuum.

The work of Skowronski and Roberston [28, 33, 34] was fairly detailed. In addition to separating elastic, viscoelastic, and plastic deformation, they focused on how the slope of the stress-strain curve changes in various loading and unloading scenarios. They conclude that the plastic deformation is a strong function of initial strain and less dependent on the duration of imposed load. Although they note that the plastic deformation is rate or time dependent. They note that the recoverable deformation is a "relatively pure viscoelastic response" [33]. They suggest that a quantitative model is unattainable requiring spectra of relaxation times and inclusion of activation and failure processes. They also suggest that it is a challenge to attempt to devise a hypothetical mechanical model producing results similar to that observed for paper [33].

In the last ten years, the need to have constitutive models for computer simulations has once again brought on new developments. For example, Lif and Fellers [35] developed a linear viscoelastic model and Xia *et al.* [36] developed an elastic-plastic model having nonquadratic yield surfaces. Mäkelä and Östlund [37] developed an isotropic plasticity equivalent material model. A somewhat different approach using damage mechanics was taken by Isaksson *et al.* [38]. Even with these sophisticated models, different sets of parameters must be chosen if the conditions are altered too much. The models are meant for modeling multi-axial states of stress and can be used in simulations if correctly tuned, but they have been adopted from other materials and general applicability is not known.

From the perspective of fifty years since Steenberg's review [1] it seems likely that an adequate continuum model for paper can be developed. The behavior is indeed regular and predictable. It is just a matter of capturing the right behaviors with mathematics. To be adequate the paper must exhibit the behaviors shown in Figure 2. It should display the correct creep, stress-relaxation, and loading-unloading behavior with one set of parameters. Some of these essential behaviors are presented in the following section.

EXPERIMENTAL OBSERVATIONS

Scaling of stress

To develop an adequate model it is the functional shape that needs to be developed. A normalization of the stress-strain curve may be useful. Arlov [2] proposed normalizing stress-strain curves to match at the failure point, but for a model of subfracture behavior this does not seem appropriate. Seth and Page [24] found that normalizing sheet by the modulus accounted entirely for differences in shape due to processing thought to affect only bonding. Kajanto [39] also suggested normalizing the stress by elastic modulus and referred to the result as the linear elastic stain. Here we will normalize stress by some measure of modulus, but refrain from calling it linear elastic strain.

Scaling the stress by the initial modulus makes sense when comparing different sheets and different directions within a sheet. Magnitude differences will be eliminated and differences in yielding and hardening will be evident. Figure 3 provides MD and CD curves for Newsprint showing high degree of anisotropy. The curves were scaled by the elastic modulus, taken as the maximum slope of the stress-strain curve, and the resulting curves are shown in Figure 4. Figure 4 indicates that for this sheet, the basic shape of the MD and CD tensile stress-strain curves are not that different. The largest difference is that in the MD stretch is significantly lower in magnitude. It appears that MD retains stiffness and if failure had not occurred would likely have slightly higher yielding stress, and perhaps a higher slope in strain hardening. For this paper, one could approximate that the MD and CD functions have the same shape.

An example of normalized MD and CD curves of another paper containing

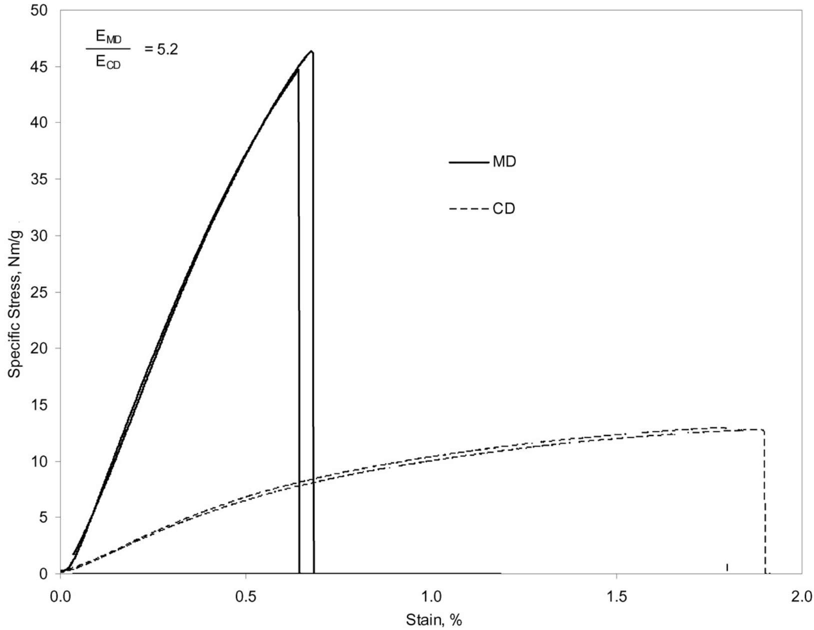

Figure 3. MD and CD stress strain curves for Newsprint, grammage=44 g/m^2.

75% groundwood and having less orientation is shown in Figure 5. Again, the main difference in MD and CD is the stretch. The MD appears to have higher strain hardening, but it is not dramatically different than CD. Figure 5 also shows a cycle of unloading, recovery, and re-loading. The CD accumulated slightly more permanent strain. Comparing the results of Figures 4 and 5 reveals that the newsprint yields at a lower normalized stress than the groundwood paper. Therefore, different papers can be expected to yield different sets of model parameters.

Figures 3–5 show results are for papers that are fairly brittle in both MD and CD. In the MD, these papers do not have large inelastic strains and so scale to CD fairly well. For papers containing a large fraction of kraft fibers, there is more ductility and larger differences in MD and CD will likely result. Figure 6 provides normalized curves for MD and CD for the same paper used for the curves shown in Figure 1. This more ductile paper shows significant difference in MD and CD. The MD has less softening and can reach proportionately the same strength as CD. An extreme case is shown in Figure 7, for a multi-ply paperboard, with a grammage of 205 g/m^2. In this case, the

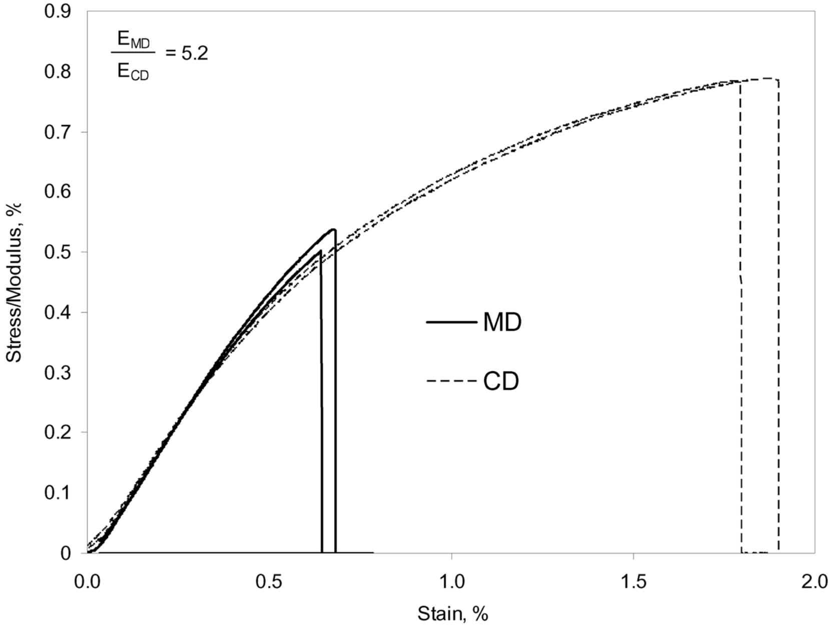

Figure 4. Newsprint stress-strain curves from Figure 3 scaled by modulus.

MD experiences much less yielding than CD and proportionately can carry more load.

The results shown in Figure 7 are similar to those shown in Figure 6. Note that for the paper represented in Figure 7, a loss of CD stiffness occurs as strain approaches failure. Many papers do not have such an extreme change in loading behavior and first loading curves and last loading curves are much more similar. Presumably the sheet represented by the results shown in Figure 7 experiences structural changes or damage in the CD under the action of straining.

If one were to scale the curves shown in Figure 1 for the different strain-rates, the result would not clarify any behavior. Even with magnitude order differences in rate of loading, the maximum slope does not vary that much. The variability from test to test yields results that are sporadic. Sometime, the curves scale and remain fairly unchanged in appearance or order. Sometimes, a test conducted at a slower rate has a modulus reduced enough such that when scaled, the curve appears to have less yielding than a higher rate test. One would expect that slower rates of deformation would cause more relaxation, as shown in Figure 1, even though the properties of the material are the

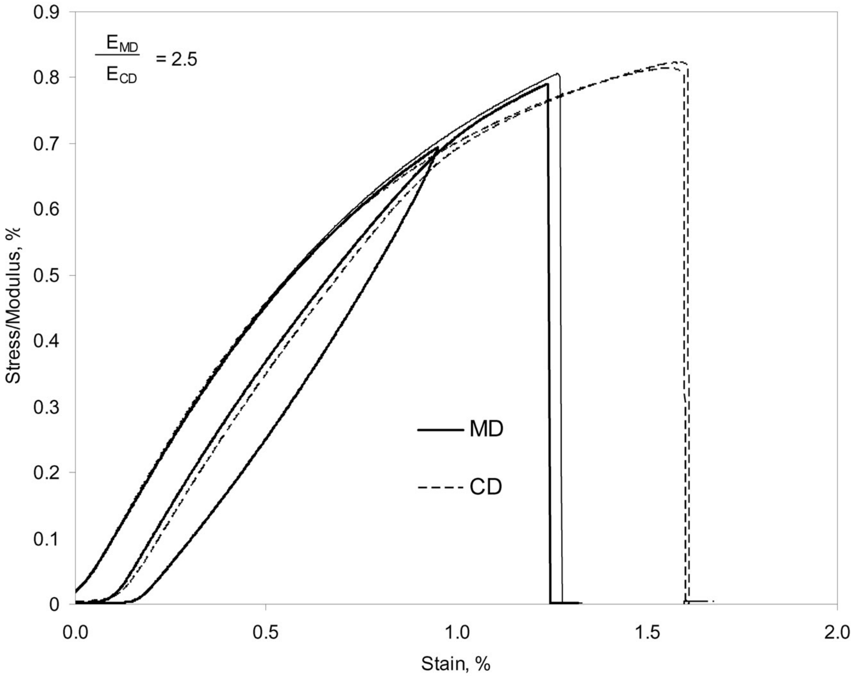

Figure 5. MD and CD stress strain curves for a commercial paper with 75% gw, grammage =76 g/m^2.

same. The scaling of stress should be to some property of the material, not the measured slope. In Figures 3 to 7, the maximum slope was taken as a measure of elastic modulus and the difference between MD and CD for the various papers are indicative of structural changes, thus the scaling makes sense. Scaling of individual curves should not be done for comparisons of results for the same paper with different test conditions. Each paper needs one scaling factor.

Initial modulus

One can determine an elastic modulus from a tensile test as the maximum slope in the initial part of the curve. Others could argue that there is not a material property that is the elastic modulus, but that the shorter the time involved in the test the stiffer the response of the material. Results from typical tensile tests do not show this result, because it takes a finite amount

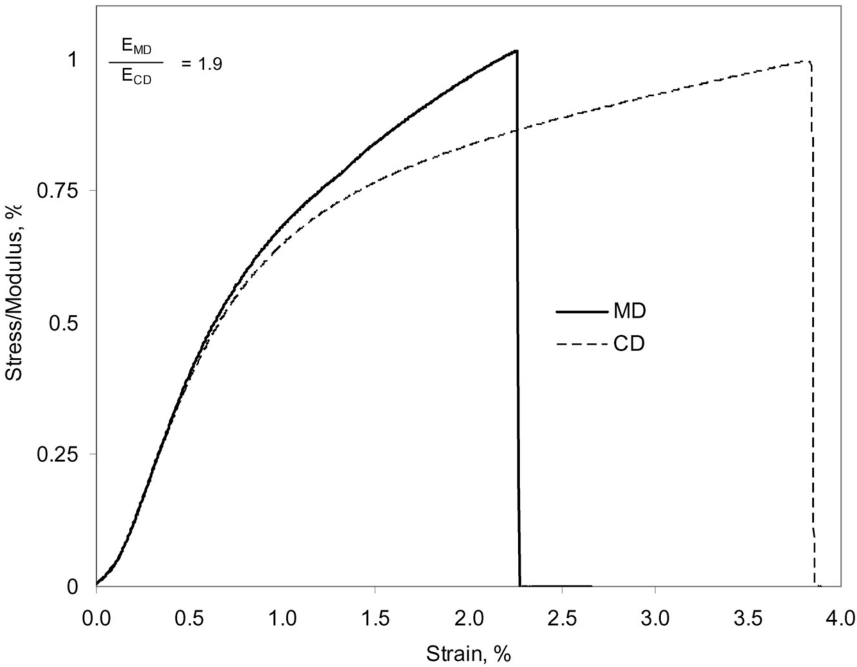

Figure 6. MD and CD normalized curves for a writing paper, grammage = 107 g/m^2.

of deformation to fully load the sample. In other words, one must pull out the slack in order to fully engage the sheet. Therefore, for a typical stress-strain curve of paper the initial slope starts low, it increases with strain until it is fully engaged, and then decreases as relaxations dominate. Figure 8 shows how the slope of the stress-strain curve varies with strain for two-different rates of elongation. The insert figure shows the corresponding stress-strain curves. Note the initial increase in slope as the sample is engaged.

The ideal model presented the current paper ignores slack, and thus one must make a decision on how to model the initial response of the paper. One choice would be to allow the slope to be constant for small strain. A second choice would be to continue the curvature of the elastic modulus at lower times. These options are shown in Figure 9. The first choice, corresponds with the idea of an instantaneous elastic modulus, but would present a problem for the relaxations at small times. The second choice would be consistent with allowing for viscous relaxations even at small time scales. In a numerical

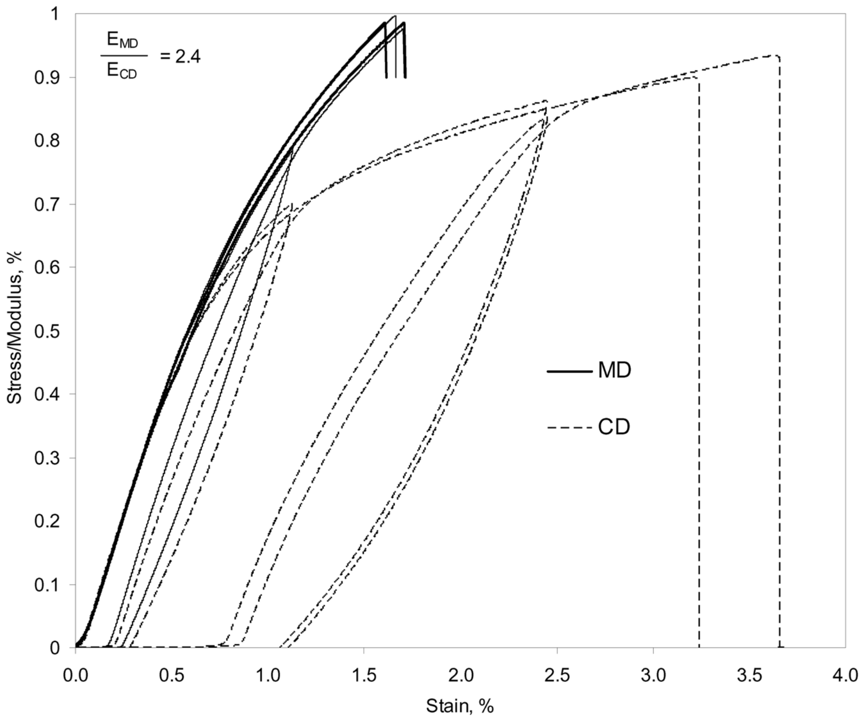

Figure 7. MD and CD scaled stress-strain curves for a multi-ply paperboard, grammage=205 g/m^2.

model, behavior at small time scales is folded into the instantaneous elastic modulus and a cut-off upper frequency.

Slack in the initial response of the tensile results is due to several factors and could be inherent in the paper or an artifact of testing. Artifacts of testing include compliance in the apparatus or nonuniform clamping of the sample. Inherent features of the paper include structural factors such as fiber micro-compressions or out-of-plane displacements like creping or cockle. Depending on the source and degree of slack, it could be accounted for in the model or ignored. For the papers shown here, it is ignored. At the other extreme, say for creped paper, it could not be ignored.

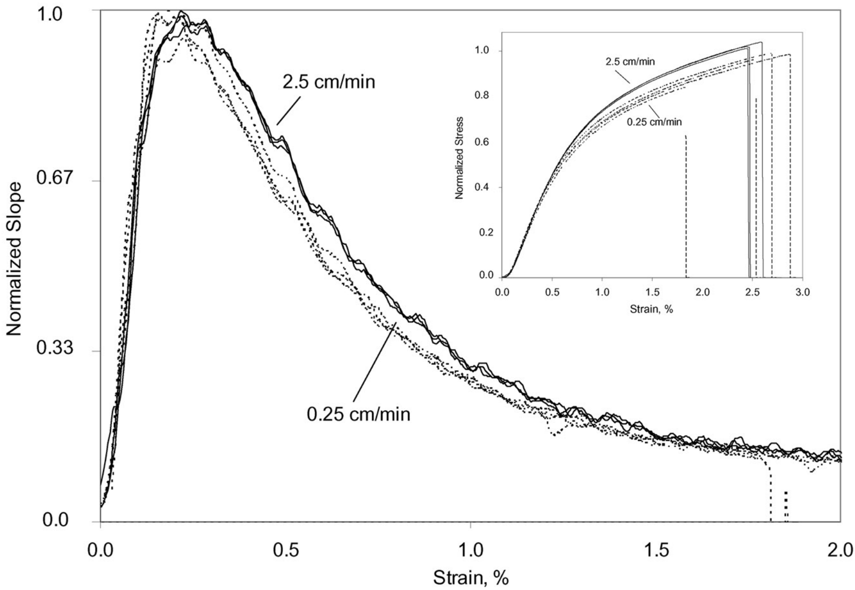

Figure 8. Typical plot of the slope of stress-strain curve versus strain for paper at two different rates or deformation.

Separation of elastic and inelastic deformation

For a model, it will be convenient to separate the recoverable (elastic) and non-recoverable (inelastic) deformation. Experimentally, this can be done by subjecting the paper to a series of loading, unloading, recovery, and reloading experiments [33]. Figure 10 provides the results of this type of loading for a CD specimens of copy paper.

The reloading curves shown in Figure 10 reveal generally parallel curves indicating both that permanent deformation has occurred, and the initial stiffness of the material has been maintained. Compare this to the CD reloading curves shown in Figure 7 for the paperboard. In Figure 7, the initial slope decreases and more relaxation occurs along with inelastic deformation. Figure 7 is a somewhat severe case and likely due to damage in the structure as a result of the straining. Many papers behave in a manner more similar to that shown in Figure 10. It seems reasonable that an initial material model would correspond to that shown in Figure 10 and inclusion of structural features to account for damage would capture the behavior shown in Figure 7.

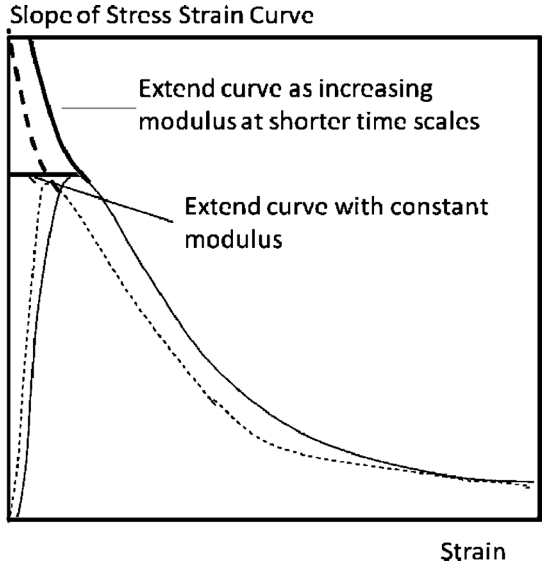

Figure 9. Possible extensions of slope when ignoring slack in stress-strain curve.

The data represented in Figure 10 can be utilized to estimate the amount of permanent deformation as a function of total strain. After the recovery period, the point of load increase on the next reloading provides an offset for determining the inelastic strain. It is assumed that the majority of the inelastic strain occurred during the previous loading and any additional inelastic strain occurring during the unloading is negligible. Therefore, the difference between the total maximum strain just before the previous unloading and the permanent set provides an estimate of the total elastic strain composed of both fast and delayed responses. Figure 11 shows the estimate of the components of the total strain.

There are three estimates shown in Figure 11. The permanent strain is determined by shifting the re-loading curve to superimpose with the initial loading curve. The squares represent a shift to match the initial slack of both curves. The diamonds represent a shift to match the point of maximum slope for each curve. The lines shown in Figure 11 represent a shift and then subtraction of the last loading curve from the initial loading curve. The agreement between the methods is adequate. The last method would be convenient to use because it would only require initial loading and a final loading near failure to determine the permanent strain.

Figure 11 indicates that the paper is approximately elastic up to a strain of

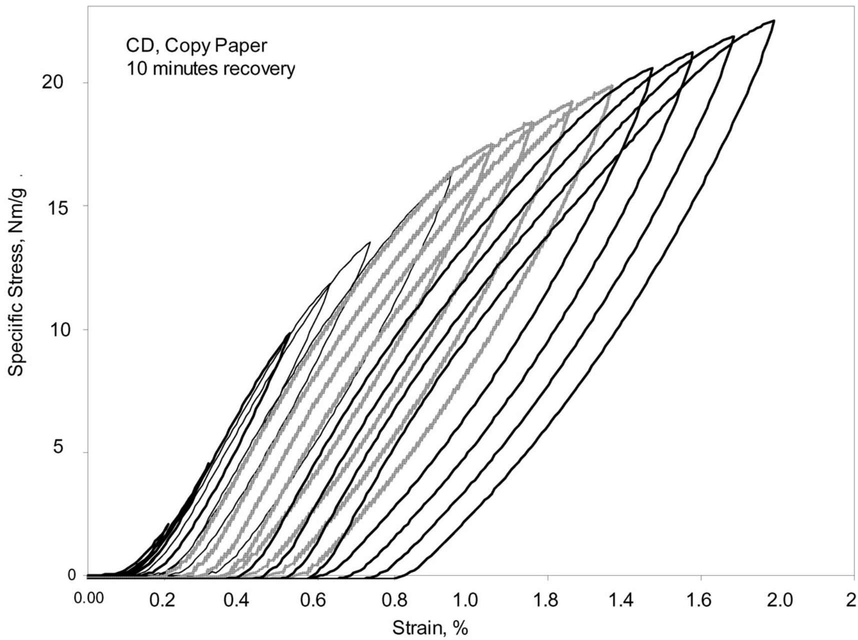

Figure 10. Stress-strain curves for periods of loading, unloading and recovery.

0.5%, then the rate of accumulation of inelastic strain increases. This is in agreement with the general observation found for many papers. It should be noted though that some permanent deformation occurs even at small initial straining.

Another way to separate out the elastic and inelastic behavior of the material is to first load the sample to a strain level such that permanent deformation occurs. If upon reloading the sample is kept at strains well below the previously strained state, the majority of the deformation will be recoverable. By allowing for recovery for each subsequent, reloading one can verify that the previous deformation was recoverable. Figure 12 provides a graph illustrating this concept for load cycling at three different rates of deformation.

By studying the elastic response separately from the inelastic response one can determine, the requirements for a model. As expected the elastic component of the deformation is found to be time dependent and nonlinear with load. The nonlinearity can easily be observed with relaxation data that shows proportionally higher rates of relaxation at higher load levels as shown in

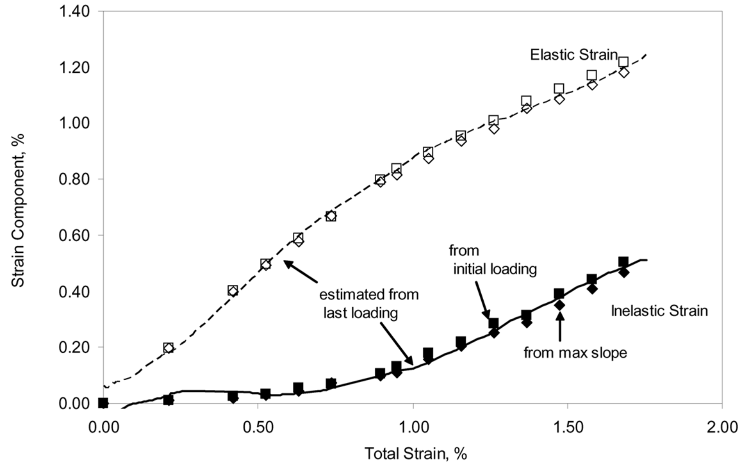

Figure 11. Estimation of elastic and inelastic component of strain corresponding to Figure 10. Curves are taken directly by subtracting initial loading curve from last loading curve.

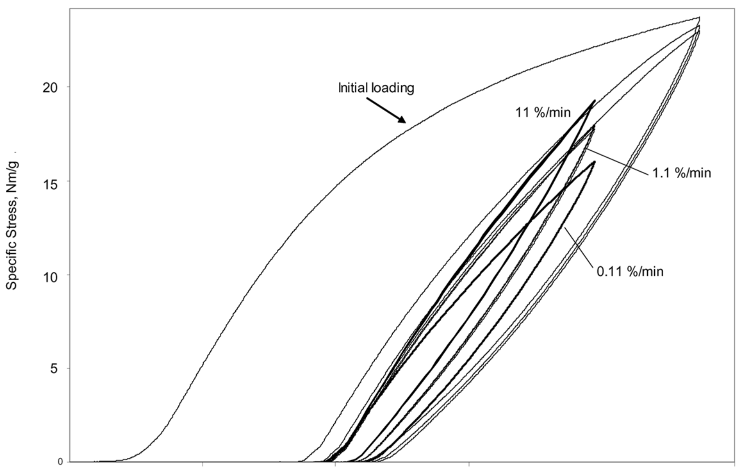

Figure 12. Observing elastic response by first removing inelastic deformation.

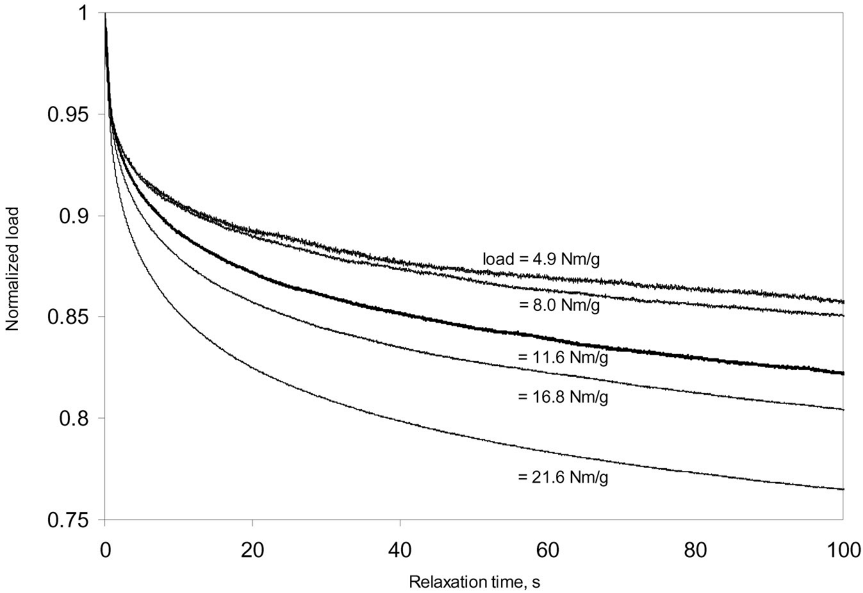

Figure 13. Influence of load level on recoverable stress relaxation.

Figure 13. For these tests, the initial loading was followed by a period of stress relaxation and then unloading. The subsequent relaxations were kept to a time of 100 seconds followed by unloading and recovery. The absence of additional inelastic strains was verified with the next reloading.

One may wonder if the inelastic strains are time dependent, but clearly inelastic creep [13] shows this to be the case. In addition, stress relaxation can be utilized to demonstrate that the inelastic behavior is time dependent [33]. A series of tests were completed where a sample was loaded to an initial strain, unloaded and allowed to recover. Then the sample was sent through a series of reloading, stress relaxation, unloading, and recovery. With each additional step, the time for stress relaxation was increased. Figure 14 shows these results along with the relaxation curve for a similar sample.

The results in Figure 14 were normalized to the initial maximum load reached at the target strain. The subsequent relaxations have lower normalized loads because more inelastic deformation accrued during the previous relaxation step. The curves in Figure 14 appear to converge to a steady logarithmic decay of stress. After the initial loading cycle with no relaxation there was an inelastic strain of 1.0%. Figure 15 displays the increase in the inelastic

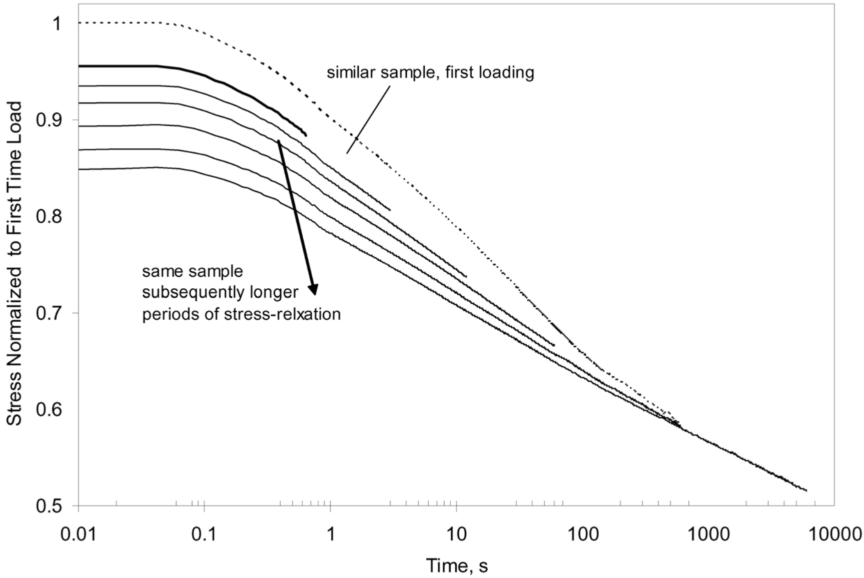

Figure 14. Stress relaxation curves for sequentially longer periods of relaxation.

strain with additional relaxation times. Clearly, a significant portion of the inelastic strain is time-dependent.

The final point to be addressed for the purpose of developing a model is the question of whether paper exhibits a sustained delayed-elastic response in stress-relaxation or whether the load will continue to decay to zero. The rate of decay of load shown in Figure 14 is logarithmic with time, but that rate does not appear to be slowing down. If the long term stress relaxation is due to inelastic behavior one could easily expect the process to continue. To date, I have not found results that would indicate the delayed-elastic state is stable for long times, nor has a test been carried out for sufficient time to establish if the relaxation will continue to zero. My own relaxation testing carried out for more than sixty hours was not conclusive. At intermediate time scales, the delayed-elastic response does ehibit stress recovery. These stress recoveries also appear to be converging to a point where it would converge with the long-time scale inelastic stress relaxation. Since both the inelastic relaxation and the delayed-elastic relaxation decay logarithmically with time and the inelastic decay appears to be at a lower rate than the delayed-elastic response, it may not be that important for some modeling situations. If the timescale of interest is intermediate, a model with only delayed-elastic deformation could

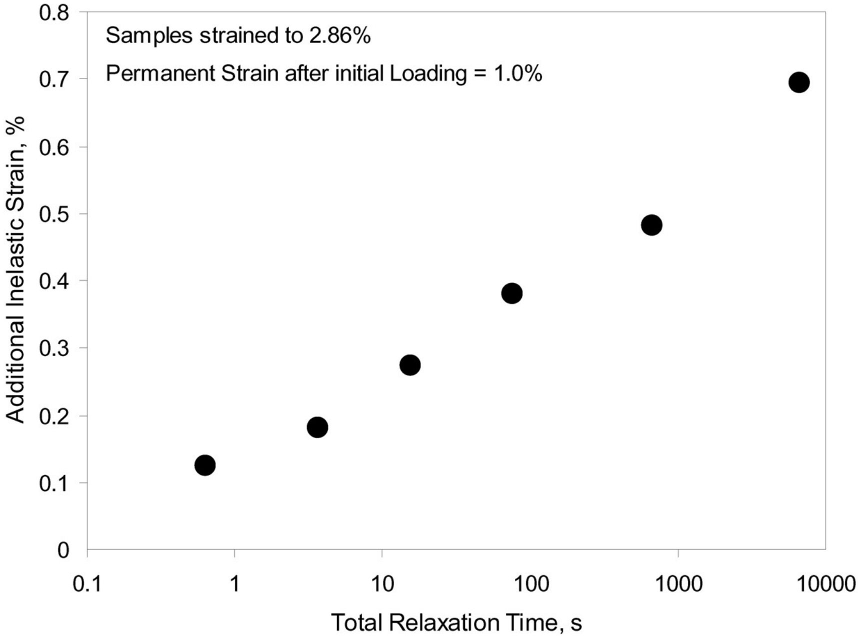

Figure 15. Additional inelastic strain due to stress relaxation as a function of total relaxation time.

be adequate. On the other hand, to be consistent with the secondary creep it would seem more prudent to include time-dependent inelastic behavior.

From the observations reported here, the essential features required for a model can be summarized as follows:

- Both elastic and inelastic deformations are time dependent and nonlinear.
- Inelastic strain accrues even for small loadings, but the rate of accrual increases at a certain strain level.
- Because of slack in the sample and/or limitations of test equipment, the low-load and essentially the short time response cannot adequately be determined in a standard tensile test.
- Scaling to some measure of initial elastic modulus appears reasonable to remove much of the variability between various papers and load directions.
- The rate of relaxations in the material decays logarithmically with time.
- Some papers experience subfracture damage that would alter effective properties, but a model that does not account for this would capture the behavior of many papers.

UNIAXIAL LOADING MODEL FOR PAPER

Constitutive equation

The starting point for a constitutive equation for paper should rely on polymeric behavior regardless of the entire structural hierarchy of the fibers and network. The impetus for the constitutive equation presented here is the Eyring model based on the premise that an applied force causes a linear shift in the potential energy barrier disturbing dynamic equilibrium and causing flow in the direction of the applied force of a polymer [40]. The early rheology work dealing with paper also utilized an Eyring type response [9]. To generalize this behavior, The Eyring mechanism is interpreted such that the characteristic time constants of the material are load dependent. This dependence on load was observed by Brezinski [13] for creep in paper. Based on the load dependence of creep compliance, Habeger and Coffin [41] developed empirical constitutive equations for creep that explicitly showed the exponential dependence of load on creep compliance. In essence, load acts to shift the response to smaller logarithms of time. Therefore, it seems justified both from a fundamental stance and experimental evidence to introduce a load activated time constant as

$$\tau = \tau_0 e^{-a\sigma}, \tag{1}$$

where τ_0 and a are material properties [25].

For the model, the total strain, ε, is taken as the sum of an instantaneous elastic component, ε_e, a delayed-elastic component, ε_{de}, and the inelastic component, ε_{ie}:

$$\varepsilon = \varepsilon_e + \varepsilon_{de} + \varepsilon_{ie}. \tag{2}$$

The instantaneous elastic response is the approximation for the effective response of events occurring at time-scales smaller than those accounted for in the model. Because of the load dependence of the time constant, an incremental or differential constitutive equation is well-suited for describing the response. The elastic component is described with a finite spectrum of relaxation times (τ_i) and moduli (E_i) as

$$\frac{d\varepsilon_e}{dt} + \frac{d\varepsilon_{de}}{dt} = \frac{d}{dt}\left(\frac{\sigma}{E_0}\right) + \sum_{i=1}^{N}\left(\left(\frac{E_0}{E_i}\right)\left(\frac{\sigma}{E_0}\right) - \varepsilon_{de_i}\right)\frac{e^{a_i\left|\frac{\sigma}{E_0} - \frac{E_i}{E_0}\varepsilon_{de_i}\right|}}{\tau_i} \tag{3}$$

Equation (3) is analogous to a series of nonlinear Kelvin units, where Equation (1) is utilized to describe the relaxation except with the change that only the viscous component of the stress is activating the relaxation. It is important that the nonlinearity be utilized using only the viscous stress component. It is this component that can go into compression in unloading and would account for differences in loading and unloading.

The inelastic response, taken from Habeger and Coffin [41] and Coffin [25] is written as

$$\frac{d\varepsilon_{ie}}{dt} = \frac{\sigma A}{E_0 \tau_{N+1}} e^{a_{N+1}|\sigma|/E_0} e^{-\left(E_0\varepsilon_{ie}/A\sigma\right)} \tag{4}$$

Equation (4) gives the expected log-linear secondary creep response for paper. Just as Equation (3) is taken as a summation of different time constants, Equation (4) could just as easily be written as a summation of inelastic contributions at different characteristic time constants. For the present model, only one element, as reflected in Equation (4) was utilized.

Equations (2) to (4) represent a model to describe the uniaxial response of paper at all pre-failure loading and time scales. Instead of the exponential function in Equation (1) a hyperbolic cosine function is just as effective and would provide for a smooth transition between tension and compression. The exponential function is utilized here because the coefficient a_1 is chosen to be different in tension and compression and thus a step-function is required and the benefit of a hyperbolic cosine is lost.

To use Equations (3) and (4) to simulate the tensile response, the following scheme is used. Initial conditions are specified for the stress and strain. Then at each increment of time either the total stress or strain is specified. Each component of the incremental strain is calculated from equation (3) and (4) using the strains from the previous time interval. The updated unknown stress or strain is determined from the relationship

$$\sigma = E_0\left(\varepsilon - \varepsilon_{de} + \varepsilon_{ie}\right). \tag{5}$$

With this incremental solving scheme any loading history can be evaluated. The model is not elegant, but it is rather simple, has few parameters, and is easily evaluated using a computer. It will yield results for both tensile and compressive loading. For the results presented here, only an Excel spreadsheet was utilized.

Note, that the natural scaling factor for stress in the model is the instantaneous elastic modulus, E_0. This is a magnification factor for the stress and would render similar scaling as shown in Figures 4–6. The inelastic strain is governed by three parameters, A, a_{N+1}, and τ_{N+1}. For creep, A gives the slope of the logarithmic secondary creep response, a_{N+1} determines the degree of nonlinearity, and τ_{N+1} is the characteristic time constant at no load. All the inelastic parameters could be obtained from forming and fitting the master creep curve [25]. Each element of the delayed-elastic response has three parameters, E_i, a_i, and τ_i. In creep, the E_i control the magnitude of the delayed-elastic creep strain, the a_i control the degree of load nonlinearity, and the τ_i are the characteristic time constants at no load.

In order to simplify the delayed-elastic response, so that only three parameters need to be chosen for the entire array of elements represented by the summation in Equation (3), a delayed-elastic response as a function of time scale is introduced. Assume that between two cut-off time constants the steady-state delayed–elastic response is a linear function of the time constant. Then one element is chosen for each decade of time constant. The modulii, E_i, is determined such that the effective elastic modulus varies linear with the logarithm of time. Thus, one only need specify an upper and lower time constant and the corresponding effective elastic modulii at these levels. The modulus for the lower time constant cut-off can be chosen as a large number so that the overall modulus is just slightly less than E_0, thus one only need specify the total delayed-elastic modulus at the upper cut-off time constant, E_f. The elastic modulus of each element is then obtained by using the fact that the inverses of the moduli add to give the inverse of the effective moduli. The value of a_i is chosen to be the same for all delayed-elastic elements, but it can take on different values for tension and compression.

The model easily handles tension and compression. In unloading, compressive stresses are created in the viscous element of the delayed-elastic response. Since paper is expected to be more compliant in compression the value of a_1 is chosen to be higher in compression than tension. Although not reflected in the results here, the value of a_{N+1} can be assigned to have different values in tension and compression.

Fitting of model

This model was utilized to see how well it would fit the tensile response in the CD for a commercial copy paper. The values utilized in the model are given in Table 1. Creep tests were not carried out for the present study, the value for the inelastic time constant was taken from previous work [25]. The time constants for the elastic deformation were then taken to span up to one order of time less than the creep response. The lower limit of time scale is limited to be sufficiently larger than the time-increment used for the numerical evaluation of Equation (3).

Table 1. Model fitting parameters for a CD specimen of copy Paper.

Parameter	*Values*
[τ_1 to τ_{12}]	[10^{-3} to 10^8] seconds
E_f/E_0	0.36
a_i i = 1 to 12	3600 Tension 12600 Compression
a_{13}	5100
A	.085
τ_{13}	10^9 seconds

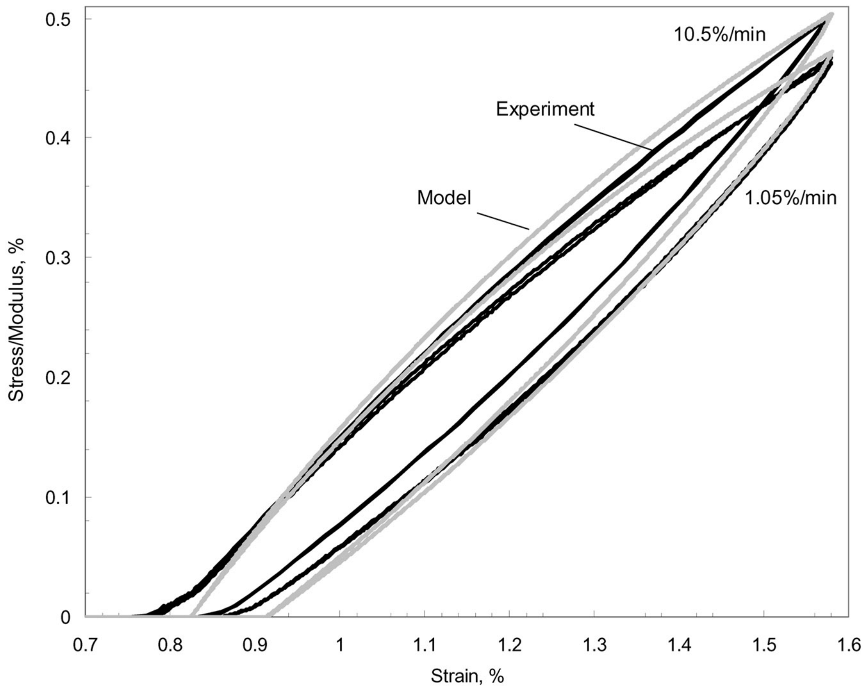

Figure 16. Comparison of model and experiment for elastic-deformation.

The recoverable response was determined by comparing the model to the stress-strain curves shown in Figure 16. In this case, the sample was first pre-loaded to remove the inelastic strain. A load cycle at two-different rates of loading was conducted. All the a_i for the delayed-elastic elements were chosen

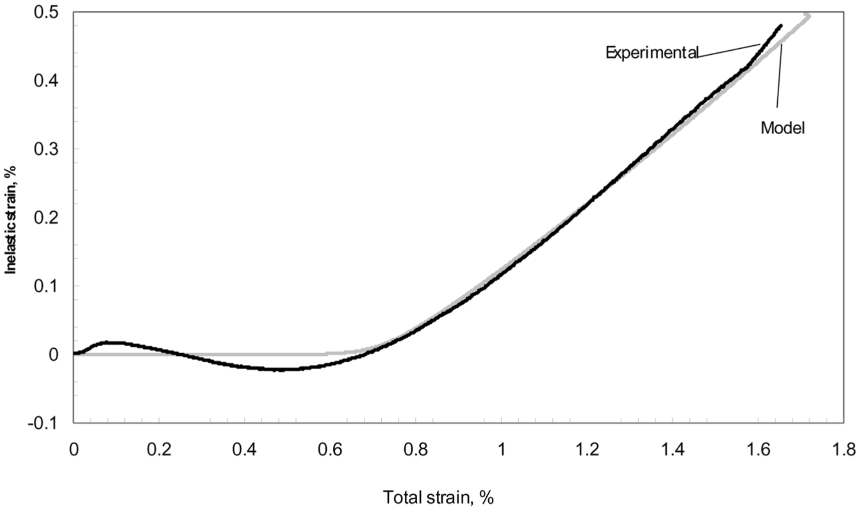

Figure 17. Comparison of model and experimental for inelastic deformation.

to be the same. The value of E_{12}/E_0 and a_1 was then varied to give a reasonable approximation for both the elastic and total stress-strain curves. The comparison for delayed-elastic response is shown in Figure 16. For the inelastic strains the value of A and a_{13} were determined from the graph of inelastic versus-total strain curves shown in Figure 17. The value of a_{13} determines the strain when yielding begins and the slope is governed by the value of A.

A comparison of the model to the total stress-strain curve is given in Figure 18 illustrates the model does provide a reasonable simulation of the stress-strain curve. The elastic response is a bit too stiff, and the inelastic response is a bit under predicted, but overall it is satisfactory.

Figure 19 shows that without changing the model parameters, stress-relaxation is also adequately predicted. The model provides a reasonable representation of paper treated as a continuum.

Parametric Study

Using a model such as presented here, provides a foundation to determine the effect of parameters on the mechanical response. Figure 20 to 24 show how the various model parameters affect the stress-strain curve. From Figure 20, it is clear that a_{N+1}, the nonlinear inelastic term, controls yielding, and from the

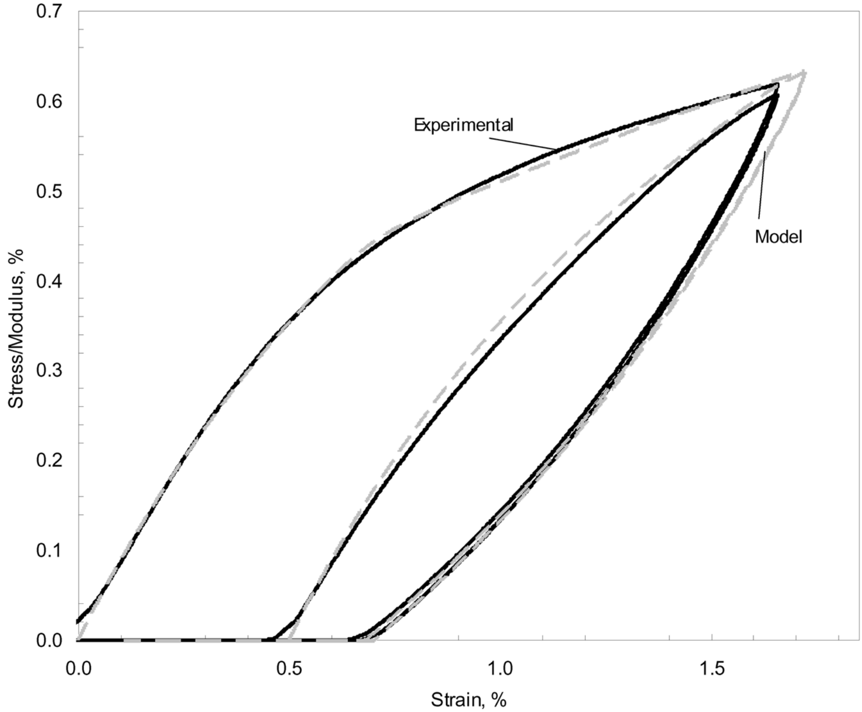

Figure 18. Comparison of stress-strain curve for model and experiment.

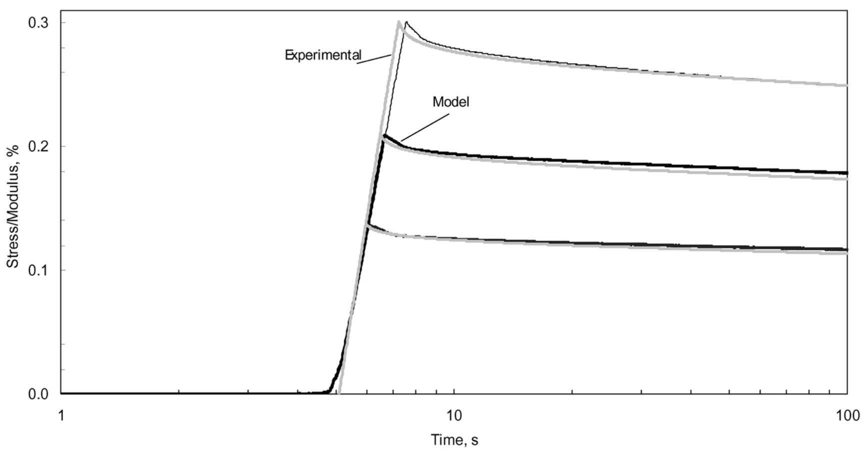

Figure 19. Comparison of model to experiment for CD stress relaxation.

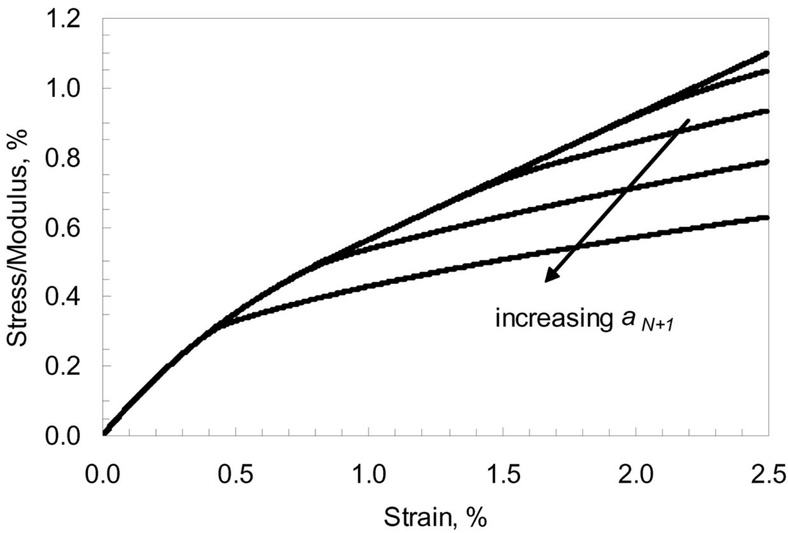

Figure 20. Influence of a_{N+1} on Stress-strain curve; yield point.

results given in Figure 21, the slope of post yielding is controlled by A. Changes in the delayed-elastic response due to changes in the parameters are shown in Figure 22 to 24 for a sinusoidal load application. The parameters affect both the dynamic modulus and the amount of energy dissipated in a cycle.

GENERALIZATION OF MODEL

The model as presented here is adaptable. A spectrum of characteristic times could be added to provide for very different elastic and inelastic behaviors. Different ranges of time constants could be utilized. The difference in tension and compression is easily handled. An efficiency factor is introduced by simply scaling the initial modulus. A failure criterion can be imposed.

Slack or severe structural changes as imposed by creping would best be incorporated by modeling the structure. Nonuniform loading would give slack and severe misalignment of the loading direction and principal material axes would give the hardening observed in stretching creped materials. Otherwise, additional phenomenological elements could be added to mimic these features.

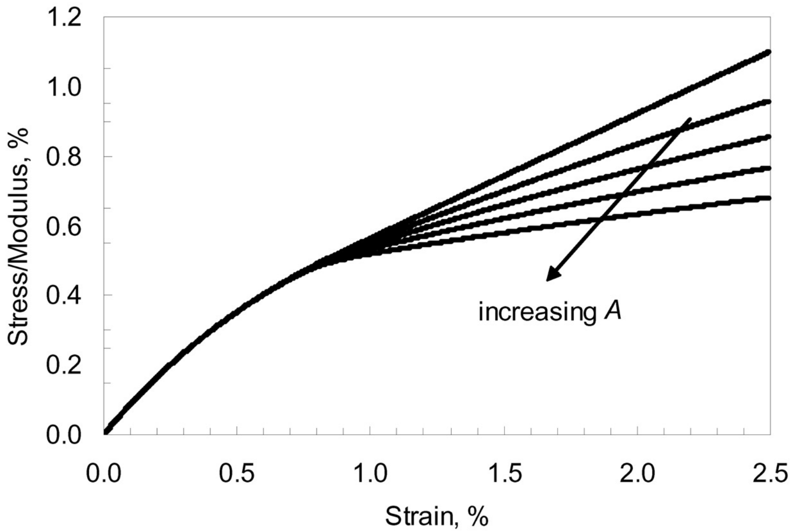

Figure 21. Influence of A on stress-strain curve; slope of strain-hardening region.

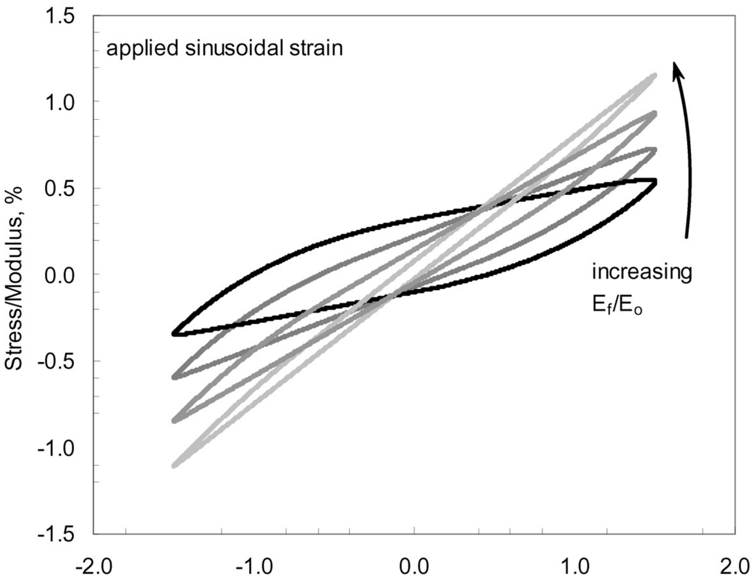

Figure 22. Influence of E_f/E_0 on the delayed-elastic response.

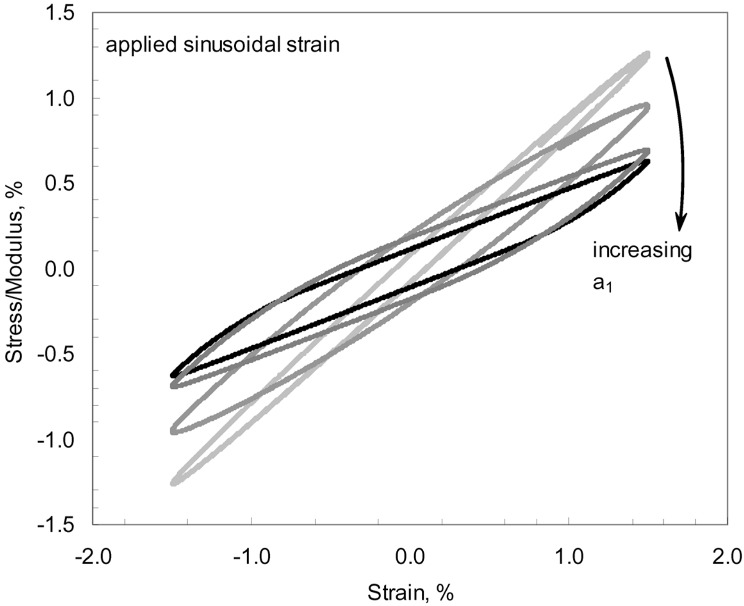

Figure 23. Influence of a_1 on the delayed-elastic response.

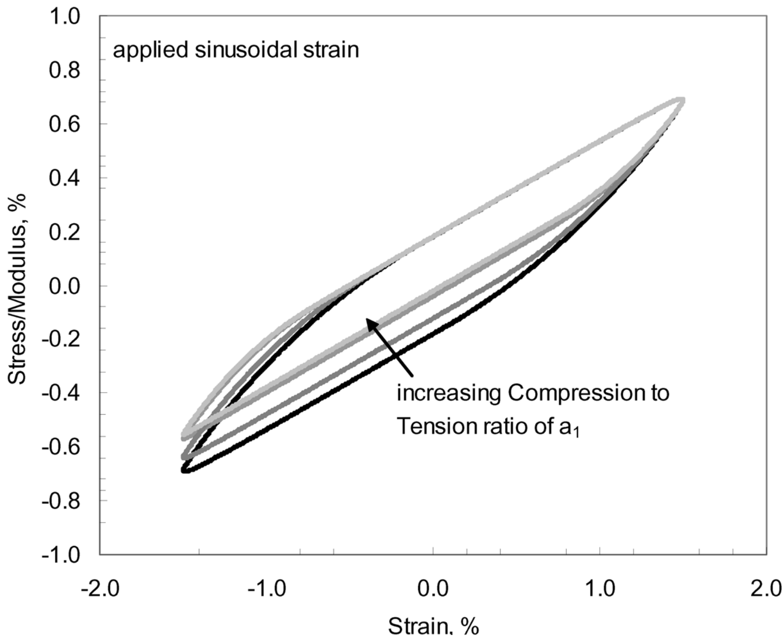

Figure 24. Influence of compression being more nonlinear than tension on delayed-elastic response.

The extension to multi-states of stress requires thought. Clearly the elastic components could be treated with an orthotropic material model. Poisson compliance terms could be added to Equation (3). Similar relationships could be introduced for shear. The problems that remain to be addressed are how to couple the inelastic strain tensor to stress and how to consider the load non-linearity with multi-axial states of stress. One must determine the correct effective stress term to include in the nonlinearity. Experimental guidance is lacking and this is work for the future.

CONCLUSIONS

By accounting for load nonlinearity, recoverable and nonrecoverable deformation, appropriate time-scale responses, and differences in tension and compression, a continuum model will provide a reasonable representation for the mechanical behavior of paper. The model parameters provide a connection between the stress-strain curve of paper and stress-relaxation, recovery, and creep. The Eyring-like load dependent time constant is an appropriate way to account for nonlinearity.

The model captures the influence of load and time in a manner that is consistent with observations. The secondary creep parameters are directly related to the yielding and strain-hardening exhibited in the stress-strain curve. An advantage of the model is that no yielding criterion needs to be specified. The yielding and creep is a natural consequence of irrecoverable relaxations that occur in the paper. The chosen form of the nonlinearity allows for plastic deformation to accrue with load and time. It is the load nonlinearity in creep that dominates the strain at which yielding occurs. The slope of strain-hardening is directly related to the slope of the log-linear secondary creep response. When the load is sufficiently high, large amounts of inelastic deformation can occur at short time scales.

In order to model the stress-strain curve, a spectrum of recoverable relaxation times is required. These load dependent relaxation times provide for a delayed-elastic response that has the curvature observed in testing.

By evaluating the model one can develop new hypotheses about the mechanical response of paper. The model predicts that the reloading curves should converge with the original loading curve at the point of unloading. If full recovery has not occurred, the reloading path will be steeper but still meet at the same point. Experiments of reloading after partial recovery support this. On the other hand at large deformations, the re-loading converges with the stress-strain curve at larger strains as shown by the dark curves of Figure 10. This implies that additional inelastic behavior has occurred or that due to changes in the material the response has slightly changed. To capture this behavior would require some change in the model. Thus, the model can be just as useful by determining what it can not explain.

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Transcription of Discussion

DEVELOPING A DEEPER UNDERSTANDING OF THE CONSTITUTIVE BEHAVIOR OF PAPER

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Department of Paper and Chemical Engineering,
Miami University, Oxford, OH, USA, 45056

Mark Kortschot University of Toronto.

Doug, you said that you had 6 parameters altogether, although I saw a_i, with i varying from 1 to 12, is that so?

Doug Coffin

They are all fixed at the same constant.

Mark Kortschot

So all of the a terms are fixed?

Doug Coffin

Yes, as you delve into more detail, you might find that at shorter times, the a_is might decrease. Right now, I do not see where that is needed, but I can imagine at really small time scales there maybe less non-linearity. Right now, it is fixed. Every time you add an element then you are going to have to add two parameters, but if you choose a function to fit those, then that's less parameters to go into the model. So the function I chose for a_i is constant, so it is one parameter, the function I chose for the modulus is a straight line – two parameters.

Discussion

Mark Kortschot

So, 6 is the total number, in other words.

Doug Coffin

6 is the total number.

Mark Kortschot

And so you don't need very many experiments to find your 6 parameters.

Doug Coffin

Exactly!

Mark Kortschot

Thanks.

Mikael Nygårds KTH

Nice presentation. I was wondering though, if you are going to have this model of everything, how does it cope with the out-of-plane deformation? Now you are presenting a lot of the in-plane properties; would it be the same behavior, and how can it be compared to have it as a multi-axial model?

Doug Coffin

This is one-dimensional and, just like any other constitutive equation, once you go from one dimension you have got to decide how you are going to do that. So you are either going to need to come up with some kind of effective stress maybe, or you are going to have to come up with the right coupling terms and the right geometric term, so that it all matches. That, as I said in the paper, is work that needs to be done, but it is not impossible. It could work just as well as other models work.

Mikael Nygårds

But do you think the behavior would be the same?

Doug Coffin

Do you think when we go out-of-plane, it is all of a sudden going to be plastic deformation? I think that we are dealing with polymers here and so I think they are going to behave like polymers.

Myat Htun Mid-Sweden University

Thank you very much for your paper. You took up Börje Steenberg's work; I think we have forgotten that paper is a viscoelastic or even plastic material. We have to regard paper as a polymer; it is a natural polymer. You have crystalline and also amorphous components in it, besides the paper structure, of course. So I think this is a very good re-introduction of old knowledge and I think the younger generation might pick this up and do better.

Doug Coffin

With computers today, you can implement those models and change them and generalize them so that you can easily back out. If we try to do this analytically – 50 years ago you kept the models down to 2 or 3 elements so that you could solve it, and then they are not adequate to describe paper. Now, it takes 10 seconds with an Excel program to generate 10,000 data points to plot out a stress/strain curve – the computation is easy. What's important is how we put it together in a right way to make it work.

Kit Dodson University of Manchester

Thanks, Doug. I well remember the arguments at the time of Steinberg proposing the springs and dashpots. They were really quite fierce, certainly here in Oxford in '61. The point is that he was misrepresented by those who wished to misrepresent him, and there is a lot of good in his approach. The whole idea of a model, and it really does not matter how you do the modelling, is that it enables you to go somewhere you cannot easily go with an experiment and to make a statement that is valuable. Now the trick, it seems to me, that would demonstrate the value of your new approach, would be to dream up some simple enough coupling to a 3D structure, superimpose it on the mass map and look at strain and moisture changing. You should be able to come up with a statement about a transformation of the network that would be useful.

Doug Coffin

Okay! Thank you. And I think the other thing is it links creep to the stress-strain curve in an adequate way. There have been other attempts before to say from the stress-strain curve how do we predict the creep response? They haven't worked very well. I think this will connect it well. The trouble you are always going to run into though is the variability. That the sample-to-sample variability is going to have such an impact that the longer time you spend looking at it, the more it is magnified because of the log scale. So, a little bit of variability at short time is going to create huge differences in the end. But we know the connection is there, it is now a matter of handling the variability to show exactly why we think we do not have good correlations, when we actually do.