Stress Relaxation of a Paper Sheet under Cyclic Load: An Experimental and Theoretical Model

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Received October 21st, 2010; revised November 12th 2010; accepted November 19th, 2010.

ABSTRACT

Mechanical experiments have been performed to study the dynamic stress relaxation of a paper sheet material mainly used in food packaging industry. The material was cyclically tensile-loaded with a strain range between 2.4% and 4%. The time period for each cycle was 400 seconds. It was found that stress will decrease when the number of cycles increases in the case of upper load and vice versa in the case of lower load. At the same time, the stress to strain curves followed the same pattern as the one from the previous cycle. The stress relaxation behavior of each cycle has been analyzed and the dynamic relaxation modulus was derived. An improved model is proposed to describe the dynamic relaxation behavior of the paper sheet. This model shows a very good fit to the experimental results and trends of prediction are observed. Furthermore, the physical description of this model and the variation by the cycles is discussed.

Keywords: Relaxation, Cyclical Load, Paper Sheet, Improved Relaxation Model

1. Introduction

Paper as a material is subjected to a complicated loading pattern from its birth in the drying operation to the converting operation and a possible collapse in an end-use situation. In recent years, several works have been done to study the mechanical properties of the paper sheet [1-5]. Corresponding material models have been created for paper or carton materials in [2-4] and [5]. In liquid food packaging companies, it is also important to avoid the swelling of paper-based liquid food packages within their given storage time. Therefore it is important to study the relaxation behavior of the paper sheet. Experiments and characterization of the nonlinear viscosity of some composites and paper fiber composites have been performed in [6-9] where creep tests and Schapery’s constitutive law have been used to determine the viscoelastic nonlinearity parameters. Inherently, a more sophisticated testing with regard to the time-dependent behavior of the paper with constant deformation (stress relaxation) and at cyclic deformation (dynamic behavior), might give a better understanding of the difference in the performance of this type of material during a given time scale. Therefore, our work addresses the stress relaxation phenomenon under cyclic loading on the paper sheet used in the food packaging industry.

Thin sheets having no bending stiffness are in general complex types of material which do not always satisfy the classical plane stress theory [1-9]. Their unusual elastic properties behavior is the expression of this complexity. Like in geo-materials [10-12], gels [13], granular material [11], and even sometimes liquids [13], long-time recovery relaxation phenomena and anomalous softening of the resonance frequency with strain are some examples of the elastic properties of this class of material. This was demonstrated on thin sheets having extremely small bending stiffness for the first time by Mfoumou et al. [14], and is the motivation of a further investigation of the stress relaxation process on a paper sheet in this work.

Earlier works have been done to analyze relaxation and reverse relaxation in synthetic fibers under cyclic loading [15,16], as well as their potential analytical models for prediction of relaxation and reverse relaxation and deformation-recovery process [17]. These works pointed out that during the dynamic loading of fibers initially having no bending stiffness there was a viscoelastic effect, which was clear from the increased elongation following the number of load cycles and from the hysteresis curve of load versus strain. Vangheluwe [15] argued that the existence of viscoelastic effects during
dynamic loading influences the relaxation behavior afterwards, and that the existence of reverse relaxation seems to be due to viscoelastic effects from the previous dynamic loading of the specimen.

Traditionally, the relaxation behavior of different materials can be described by several different models as listed in [18]. The viscoelastic properties of the material under investigation have recently been illustrated by Mfoumou [19]. The relaxation curves were fitted to a series of exponential terms derived from a Maxwellian model [20,21] which has a similar response to the Prony series as discussed in [7]:

$$Y(t) = \sum_{i=1}^{n} a_i \exp(-b_i t)$$  \hspace{1cm} (1)

where \(Y(t)\) is the decaying parameter (force, stress or apparent modulus), \(t\) the time and \(a_i\) and \(b_i\) constants characteristic of the material. Based on this model, the paper sheet investigated clearly exhibits a nonlinear viscoelastic behavior since both the number of terms and the constants depended on the deformation history and level. The number of terms reported was 2 involving 4 constants; this makes comparison of curves difficult because all the constants are independent. In this paper, an improved model will be introduced, analyzed and discussed. Here, only the relaxation stress at the upper strain of load cycles is considered in the analysis. We believe that once the relaxation model and its relation to the cycle are clear, the reversed relaxation can possibly be modeled in a similar way.

2. Experimental Method and Results

2.1. Sample Geometry and Test Setup

The test specimens are rectangular strips, 250 mm long, 15 mm wide, with a thickness of 100 µm. The samples were placed in a conditioned environment at 23°C and an atmospheric humidity of 40% during at least three days prior to the tests. The MTS Tensile Test Machine used is shown in Figure 1. The load on the sample is recorded by a piezo-electric load cell mounted between the sample and the crosshead of the machine. This load cell (with maximum 2.5 kN) is used together with a pair of pneumatic clamps. The grip separation is set to the specimen length. Each specimen is clamped at the upper and lower ends to create a fixed-fixed boundary condition on a tensile test machine.

2.2. Tensile Test Measurement

A (low-frequency) cyclic loading was applied to the samples. This conditioning was made of repeated step perturbations at two different strain levels. Each step perturbation consisted of first moving the crosshead to 1 mm elongation at a speed of 1 mm/s, holding the crosshead for 200 seconds, unloading the sample by moving back the crosshead to 0.6 mm elongation at the same speed as in the loading process, and again, holding for 200 seconds. The subsequent step perturbations, shown in Figure 2, were performed with the extension between 0.6 mm and 1 mm. As the loading and unloading processes last for less than one second, we assume the phenomenon can be considered as a cyclic deformation.
relaxation of the sample is therefore monitored and analyzed at constant strains corresponding to the higher strain level.

2.3. Stress Relaxation Monitoring

The measurement of stress relaxation is usually made by placing the specimen in series with a spring of sufficiently great stiffness so that it undergoes negligible deformation compared with the specimen. In our experimental setup, the spring is the element of the load cell transducer, which enables a direct measurement of stress. Figure 2 shows the time-dependent variation of the strain and measured stress for cycles between 2 and 6. Here, the first cycle has been removed since there is often an intermittent behavior which is not always repeatable.

3. Analysis and Discussion

3.1. Relaxation Modulus

Figure 3 shows the time-dependent relaxation modulus of the above mentioned 5 cycles. Relaxations moduli here are calculated by simply dividing the upper stress value by the strain. Note that the horizontal axis is a logarithm of time. Curves are moved so that each upper strain is supposed to start at time zero. As can been seen, the relaxation modulus is reduced as the loading time increases during each load. The same pattern is repeated for all the following load cycles.

More completely, the experiment time was extended to one hour in Figure 4 for the purpose of getting a complete picture of the shape of the relaxation curve for the paper sheet studied. Irregularities are observed, indicating the end of the relaxation process. Such fluctuating as shown in Figure 4 has also been found in other materials and Kolseth et al. [22] described it as the presence of stress in inhomogeneities arising from insufficient annealing.

3.2. Analytical Modeling of the Observed Phenomenon

Figure 5(a) redraws the stress relaxation of 5 cycles (marked with data) at the constant strain of the upper level as shown in Figure 2. Again each curve is moved so that the load cycle starts at time zero. Based on these results, a mathematical description is discussed below.

An ideal mathematical representation of a physical phenomenon is based on the following criteria:

- The number of constants is kept at a minimum;
- The constants and the equation components bear meaningful physical information;
- The equation is sensitive to physical changes in the system but insensitive to arbitrary parameters;
- The mathematical form of the equation is as simple as possible.

To apply these conditions to our relaxation curves, an improved model has been suggested as below:

\[ \sigma^{(i)}(t) = a + e^{b(i)}e^{d(i)t} \]  

The super index \( i \) is only used to indicate the cycle. We have \( \lim_{t \to \infty} \sigma^{(i)}(t) = a \). So there is no need to estimate \( a \) by statistical methods.

Thus, the number of parameters that need to be estimated is \( 1 + 2i \), where \( i \) is the number of cycles. The model is thus slightly more complex than the simplified model, but it will be shown that the fit is sufficiently better that it is worth the extra complexity. The method of least squares is used for finding the parameters in Table 1 and \( d = 0.05823 \).

As can be seen, \( d \) is small, and this means that the
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Figure 5. (a) Stress relaxation curves of paper sheets at successive loading histories together with the data from the improved model; (b) The residuals (differences between measurements and the model) of different cycles. The residuals are very small, and appear rather independent.

The empirical formula in Equation (2) can be suggested to describe an experimentally measured relaxation law.

In order to make it more comparable to the standard relaxation laws that are known, Equation (2) takes the form:

$$\sigma = a + Ge^{-pt\alpha}$$

or

$$\frac{\sigma - a}{G} = e^{-pt\alpha}$$

(4)

with, $G = e^b$, $p = -c$ and $\alpha = d$.

Table 1. Coefficients used for the least squares method.

| Cycle | $i=2$ | $i=3$ | $i=4$ | $i=5$ | $i=6$ |
|-------|-------|-------|-------|-------|-------|
| $b^{(i)}$ | 2.819 | 2.659 | 2.612 | 2.593 | 2.556 |
| $c^{(i)}$ | 0.6741 | 0.5692 | 0.5399 | 0.5324 | 0.5081 |
| $b^{(i)}+c^{(i)}$ | 2.145 | 2.090 | 2.072 | 2.061 | 2.048 |

where $\tau$ is a parameter to be optimized, yielding a total of $2 + 2i$ parameters. If one prefers to see the model in this way, then we simply set $\tau = 1.6$ rather than optimizing over $\tau$.

Since the model is unstable around $t < d$, to study the internal irreversible changes, it may be more interesting to use $\sigma^{(i)}(d) = a + e^{b^{(i)}+c^{(i)}}$, than to use $\sigma^{(i)}(0) = a + e^{b^{(i)}}$. Thus $b + c$, which steadily decrease, may be an appropriate statistic for studying the internal irreversible changes.

As can be seen in Figure 5(a) where the solid line shows the values calculated by Equation (3), the improved model follows the measured values satisfactorily.

In Figure 5(b), we have gathered residuals (i.e. the differences between values from measurements and the improved model) from all cycles, and observe that the residuals are, along the entire timeline, fairly evenly scattered around zero. Cycle 2 clearly has the largest residuals, in particular around $t = 60$ and $t = 95$. Considering the data in cycle 2, one can see that the measurements between $t = 40$ and $t = 70$ appear to lie on a different curve than the other data points of cycle 2. Most likely these measurements are flawed, causing the residuals of cycle 2 to be larger than the residuals of the other cycles. Also, the removal of some of these “flawed” data points from cycle 2 would lower $b^{(i)}$ and increase $c^{(i)}$, so that parameter values from cycle 2 would not differ much from the parameter values of the other cycles.

We also observe that for each cycle, the residuals soon after $t = 0$ are among the largest residuals for that cycle, which is no surprise as measurements soon after $t = 0$ are less accurate than other measured results.

4. Further Discussion of the Improved Relaxation Model

The model is not very stable for $t < d$. For example, $td$ increases relatively sharply in the interval $0 < d$, as $0^d = 0$ while $d^d \approx 1$. The measurements in each cycle span over 198.4 seconds, and rather than assuming that the first measurement is made at $t = 0$, we have assumed that it is made at $t = 1.6$, so that the last measurement is made at $t = 200$. If one assumes that our measurements actually start at time 0, this would result in a large discrepancy between the model and our measurements for small values of $t$, and in particular for the first measurement. Thus one could argue that the improved model for relaxation is actually

$$\sigma^{(i)}(t) = a + e^{(b^{(i)}+c^{(i)})(t-\tau)^d}$$

(3)
It means that the stress $\sigma$ approaches its equilibrium value $\sigma = a$ according to the temporal law that will reach one $t \to \infty$ at $e^{-\mu a}$. On the side, at $t = 0$, $\sigma = a + G$.

Notice that this relaxation law in (4) differs significantly from the standard relaxation law (see [23-25]):

\[
\sigma = a + Ge^{-\frac{t}{T}} \quad \text{or} \quad \frac{\sigma - a}{G} = e^{-\frac{t}{T}} \tag{5}
\]

which satisfies the ordinary differential equation:

\[
\frac{d\sigma}{dt} = \frac{\sigma - a}{T} \tag{6}
\]

As it is known, $T$ is the “relaxation time”, $a$ is the “equilibrium stress” and $a + G$ is the initial stress.

To reconstruct the differential equation of type Equation (6) by empirical relaxation law, Equation (4) is differentiated to get:

\[
\frac{d\sigma}{dt} = -\frac{(\sigma - a)}{\left(\frac{t}{\mu a}\right)} \tag{7}
\]

Comparing this equation with Equation (6), we can conclude that formally the governing equation is the same. But the relaxation time is now time-dependent. Now define:

\[
T = T(i) = \left(\frac{t}{\mu a}\right)^{1-a} \tag{8}
\]

Equation (7) can be rewritten by a similar way as Equation (6).

As discussed before, $\alpha \ll 1$ is the characteristic time that increases with an increase in “physical time” $t$. If formally $\alpha = 1$ in Equation (8), the equation will reduce to $T = \frac{1}{\mu} = const.$

Consequently, at $\alpha = 1$, our empirical law transforms to the usual one as in Equation (5) in which.

\[
T = T_i = \frac{1}{\mu} \tag{9}
\]

Correspondingly, the Equation (7) transforms to Equation (6).

We can come to two important conclusions as follows from this consideration:

1). $T(i) \gg T_i = 1/\mu$, because we know $\alpha \ll 1$. This means that the relaxation of the paper sheet is much slower than the usual relaxation process as soon as $i \gg \alpha$.

2). $T(i)$ increases with time as can be seen by Equation (8). This means that the more time that has passed since the beginning of the process, the slower will the stress/tension of the paper decrease/relax.

The second form of governing equation can now be derived which does not contain time $t$ in the coefficients. Rewriting Equation (4) as:

\[
\ln\left(\frac{\sigma - a}{G}\right) = \alpha \ln t \tag{10}
\]

After differentiation on time $t$, we get:

\[
\frac{d\ln}{\ln G} = \alpha \ln\left(\frac{G}{\sigma - a}\right) \tag{10}
\]

or

\[
\frac{d\ln Y}{\ln \xi} = \alpha Y \tag{11}
\]

Where

\[
Y = \ln \left(\frac{G}{\sigma - a}\right), \xi = \ln t
\]

This logarithmic dependence on time indicates a slowness of the relaxation process.

5. Conclusion

From the experimental tests and theoretical modeling it can be concluded that:

- The paper sheet clearly shows the relaxation behavior. This behavior cannot be described by the well known Maxwell model since its relaxation is much slower than the usual relaxation process in materials like metals.
- Paper relaxation, moreover, demonstrates the presence of a wide spectrum of relaxation time.
- The relaxation modulus is reduced by the loading time and loading cycles with the same pattern. This can be explained by the nonlinearity of the stress and strain relation of the paper sheet.
- An improved relaxation model: $\frac{\sigma - a}{G} = e^{-\mu a}$ shows very close correspondence with the experimental results and its governing equation is similar to that of the Maxwell model but with variable relaxation time.
- This model can also describe the behavior of the material under a wide range of material conditioning (loading levels) without becoming inconsistent.
- The main characteristic of the improved model is that the parameters play a major role in the overall response of the material. One of the important ad-
vantages is the possibility to anticipate the asymptotic limiting value at which the stress will tend at infinite time.

6. Acknowledgements

The authors would like to thank Professor Oleg Rudenko and Professor Claes Hedberg for valuable discussions during the work. The Swedish Foundation for Knowledge and Competence Development and Blekinge Institute of Technology in Sweden are acknowledged for their financial support.

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