

# Branching of strain histories for nonlinear viscoelastic solids with a strain clock

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**Summary.** An important class of constitutive equations for nonlinear viscoelastic response utilizes the concept of a strain clock. The clock takes the form of a material time variable which is defined in terms of the strain history and which increases faster than physical time. Important consequences of the strain clock are that stress relaxation and creep occur faster as strain increases, and the stress may not increase monotonically with time. In this work, we discuss whether this non-monotonic response implies that strain histories may branch into multiple histories.

## 1 Introduction

There has been a great deal of interest in a particular class of constitutive equations for the nonlinear viscoelastic response of amorphous polymers, such as polycarbonate. The dominant feature of this class of constitutive equations is a reduced time variable by means of which stress relaxation occurs faster with increasing strain. This variable defines a relation between a material time scale and the laboratory time scale, and is often referred to as a “strain clock”.

The present work is concerned with an important consequence of the “strain clock”. With this constitutive equation, it is possible to produce plots of stress vs. strain which are not monotonic. The stress can reach a local maximum, decrease to a local minimum and then increase. In other terms, a given stress can correspond to several strains. This suggests that for some stress histories there may occur a time at which the corresponding strain history may branch or bifurcate. The purpose of the present work is to provide a proper interpretation of the non-monotonic stress-strain plots within the context of the class of constitutive equations considered here and then to show that branching of strain histories does not occur.

For the sake of brevity of presentation, results are developed using the form of the constitutive equation for one-dimensional response. This constitutive equation is introduced in Sect. 2. In Sect. 3, examples of non-monotonic stress-strain plots are developed for the constitutive equation. Section 4 contains a discussion of the proper interpretation of these non-monotonic stress-strain plots, and how they do not imply the occurrence of branching of strain histories. The issue of branching of strain histories is addressed in Sect. 5. It is shown, for one-dimensional response, that branching is not possible. Concluding comments are presented in Sect. 6.

## 2 Nonlinear viscoelastic constitutive equation

The constitutive equation for nonlinear viscoelastic response considered here has the following form in the case of one-dimensional response:

$$\sigma(t) = \int_{0^-}^t G[\xi(t) - \xi(s)] \frac{d\varepsilon(s)}{ds} ds = \varepsilon(0) G[\xi(t)] + \int_0^t G[\xi(t) - \xi(s)] \frac{d\varepsilon(s)}{ds} ds, \quad (1)$$

in which

$$\xi(t) = \int_0^t \frac{dx}{a(\varepsilon(x))}. \quad (2)$$

$\xi(t)$  represents a reduced time (also referred to as pseudo, intrinsic, or material time) and introduces nonlinear dependence on strain into the constitutive equation. The three-dimensional version can be found in [1].

The properties of  $a(\varepsilon)$  are similar to those of the time-temperature shift function; namely  $a(0) = 1$  and  $a(\varepsilon)$  is a monotonically decreasing function of  $\varepsilon$ . For deformation histories such that the strain increases from zero with time,  $\varepsilon(t)$  is small in an initial time interval. Then  $a(\theta) \approx 1$  and Eq. (2) implies that  $\xi(t) \approx t$ . It follows from Eqs. (1) and (2) that the response approximates that given by linear viscoelasticity. As strain continues to increase with time,  $a(\varepsilon)$  decreases and  $\xi(t)$  increases faster than the physical time  $t$ . This results in an acceleration of stress relaxation.

The form for  $a(\varepsilon)$  used in previous work is the Dolittle shift function

$$\log a = \frac{b}{2.303} \left( \frac{1}{f} - \frac{1}{f_0} \right), \quad (3)$$

where  $b$  is a material property,  $f = f(\varepsilon)$  and  $f_0 = f(0)$ . It is assumed here that  $f(\varepsilon)$  monotonically increases with  $\varepsilon$ , in which case  $a(\varepsilon)$  monotonically decreases with  $\varepsilon$ . For example, Knauss and Emri [2], [3] assumed that  $f(\varepsilon) = f_0 + c\varepsilon$ . Typical values for the constants are  $b = 0.16$ ,  $c = 1.0$  and  $f_0 = 0.01$ , for which it is found that  $a(0.001) = 0.2335$  and  $a(0.002) = 0.0695$ . The value of the shift function can thus become very small and  $d\xi/dt = a$  can become very large as  $\varepsilon$  increases.

Use will be made of the alternate form for Eq. (1) obtained by integration by parts,

$$\sigma(t) = G[0] \varepsilon(t) + \int_0^t \frac{\dot{G}[\xi(t) - \xi(s)]}{a(\varepsilon(s))} \varepsilon(s) ds, \quad (4)$$

where  $\dot{G}[s] = dG[s]/ds$ .

## 3 Non-monotonic stress-strain plots associated with the constitutive equation

Non-monotonic stress-strain plots can be constructed for a class of strain histories, discussed below, which are initially zero and then monotonically increase, i.e.  $\varepsilon(0) = 0$  and  $\dot{\varepsilon}(t) > 0$ ,  $0 \leq t$ . The stress history is given by Eq. (1). The stress-strain plot is then determined in terms of the parameter  $t$  through the expressions for  $\sigma(t)$  and  $\varepsilon(t)$ . In other terms, the stress-strain plot is constructed by plotting the stress at each time versus the strain at the same time.

The slope of the stress-strain plot is given by

$$\frac{d\sigma}{d\varepsilon}(t) = \frac{\dot{\sigma}(t)}{\dot{\varepsilon}(t)}. \quad (5)$$

An expression for  $\dot{\sigma}(t)$  can be found from Eq. (1) with  $\varepsilon(0) = 0$  and Eq. (2),

$$\begin{aligned} \frac{d\sigma}{dt}(t) &= \dot{\sigma}(t) = G[0]\dot{\varepsilon}(t) + \int_0^t \dot{G}[\xi(t) - \xi(s)]\dot{\varepsilon}(t)\dot{\varepsilon}(s) ds \\ &= G[0]\dot{\varepsilon}(t) + \frac{1}{a(\varepsilon(t))} \int_0^t \dot{G}[\xi(t) - \xi(s)]\dot{\varepsilon}(s) ds. \end{aligned} \quad (6)$$

Since  $\dot{\varepsilon}(t) > 0$ ,  $G > 0$ ,  $\dot{G} < 0$  and  $a(\varepsilon) > 0$ , the first term in Eq. (6) is positive and the integral term is negative.

Consider first, for the purpose of comparison, linear viscoelastic response, which is obtained by setting  $a(\varepsilon) = 1$  and  $\xi = t$  in Eq. (6). For constant strain rate histories, where  $\dot{\varepsilon}(t) = \dot{\varepsilon}_0$ , Eq. (6) reduces to  $\dot{\sigma}(t) = \dot{\varepsilon}_0 G(t)$ . Since  $G(t) > 0$ , it follows from Eq. (5) that  $d\sigma/d\varepsilon > 0$  for all strains, and the stress-strain plot is monotonic. In contrast, for nonlinear viscoelastic response with a strain clock,  $a(\varepsilon)$  becomes very small as  $\varepsilon(t)$  increases, and the integral term can become dominant. Thus, because of the reduced time variable  $\xi(t)$ ,  $\dot{\sigma}(t)$  and  $d\sigma/d\varepsilon$  can become negative and the stress-strain plot can become non-monotonic.

Next, consider a strain history of the form

$$\varepsilon(t) = \dot{\varepsilon}_0 t + \sum_{i=1}^N \dot{\varepsilon}_i (t - t_i) h(t - t_i), \quad (7)$$

in which  $h(t - t_i)$  is the Heaviside step function and  $\dot{\varepsilon}_i$  is a positive constant. This strain history has a constant strain rate in each time interval, for  $(t_{i+1} - t_i)$ , with the strain rate increasing in subsequent time intervals,

$$\dot{\varepsilon}(t) = \dot{\varepsilon}_0 + \sum_{i=1}^N \dot{\varepsilon}_i h(t - t_i). \quad (8)$$

For linear viscoelastic response,

$$\dot{\sigma}(t) = \dot{\varepsilon}_0 G(t) + \sum_{i=1}^N \dot{\varepsilon}_i G(t - t_i) h(t - t_i). \quad (9)$$

According to Eq. (5),  $d\sigma/d\varepsilon > 0$  and the stress-strain plot is monotonic. For the nonlinear viscoelastic response with a strain clock, the preceding discussion again shows that the stress-strain plot can become non-monotonic.

Non-monotonic stress-strain plots are frequently presented in the literature for experiments in which specimens are subjected to constant strain rate histories,  $\varepsilon(t) = \dot{\varepsilon}_0 t$ ,  $0 \leq t$ , with  $\dot{\varepsilon}_0$  constant (see, for example, [2] or [3]). Most discussions of yield in polymers are based on stress-strain plots corresponding to the strain history. Another example was presented by Yee and DeTorres [4] who compared the results of two strain histories with positive piecewise constant rates. The first history had a low rate followed by a high rate, and the second had a high followed by a low rate. Force-elongation plots were constructed for both histories by the procedure discussed above. A non-monotonic plot was obtained for each case, and the plots were different.

#### 4 Non-monotonic stress-strain plots for elasticity and viscoelasticity

Non-monotonic stress-strain plots, such as in Fig. 1, have been considered in the context of nonlinear elasticity by Ericksen [5], who discussed stability for different strain states corresponding to the same stress. Such plots can be used to construct strain histories which branch, in the following manner. Consider the stress history shown in Fig. 2a, in which the stress increases to the maximum value  $\sigma_B$  at  $B$  and then decreases. The corresponding strain history, constructed using Fig. 1, is shown in Fig. 2b. As the stress reduces from  $\sigma_B$ , the strain branches at  $\varepsilon_B$  and decreases along the branch  $BA$ , increases along the branch  $BC$ , or jumps to the branch indicated by  $D$ .

This suggests that non-monotonic  $\sigma - \varepsilon$  plots for nonlinear viscoelastic response, such as developed in Sect. 3, also indicate the branching of strain histories. The purpose of the discussion is to point out that they do not indicate branching of strain histories. The reason is that non-monotonic  $\sigma - \varepsilon$  plots for nonlinear viscoelasticity are fundamentally different from non-monotonic  $\sigma - \varepsilon$  plots for nonlinear elasticity.

In order to see this difference, let Fig. 1 represent a non-monotonic  $\sigma - \varepsilon$  plot for nonlinear elasticity, and let  $\sigma = f(\varepsilon)$  denote the stress-strain relation. The  $\varepsilon$ -axis denotes the set of possible strain values and the  $\sigma$ -axis denotes the set of possible stress values. The relation  $\sigma = f(\varepsilon)$  represents an equation of state that defines the mapping from the set of possible strains to the set of corresponding stresses. The  $\sigma - \varepsilon$  graph is the visualization of the set of all possible combinations  $(\sigma, \varepsilon)$  determined by the equation of state. These are known in advance when considering the response of an elastic material.

Next consider viscoelasticity, and let  $\sigma(t) = \mathfrak{F}(\varepsilon(t); \varepsilon(s)|_{0-}^{t-})$  denote the constitutive relation. This notation is introduced so as to distinguish between the strain history up to time  $t$ ,  $\varepsilon(s)$ ,  $0- < s < t$ , and the strain  $\varepsilon(t)$  at time  $t$ . This constitutive relation defines a mapping from the space of strain histories to the space of stress histories. Recall the results for monotonic strain histories discussed in Sect. 3, and let Fig. 1 now represent a  $\sigma - \varepsilon$  plot constructed as discussed there. The  $\sigma - \varepsilon$  graph for, say, a constant strain rate history relates one point in the space of strain histories to the corresponding point in the space of stress histories. It is **not** a visualization of the mapping from the set of possible strains to the set of corresponding stress values. It is a cross plot of one strain and one stress history in which time is eliminated.

One can visualize any number of strain histories, and for each it is possible to construct a  $\sigma - \varepsilon$  plot. The plots will all differ, and some will be non-monotonic. Each plot is valid only for the strain history for which it is constructed. Moreover, each such plot still relates one point in strain space to one point in stress space. The stress history is not specified, but its evolution as time increases is determined through the use of the constitutive expression  $\sigma(t) = \mathfrak{F}(\varepsilon(t); \varepsilon(s)|_{0-}^{t-})$ . Thus, the  $\sigma - \varepsilon$  plot is not known in advance, but is constructed as the stress evolves.

The  $\sigma - \varepsilon$  plots discussed so far are valid only for specified strain histories. They are not valid when a stress history is specified. When this is the case, the  $\sigma - \varepsilon$  plot is constructed as time increases by determining the evolution of the strain from the constitutive equation. The resulting  $\sigma - \varepsilon$  plot is valid only for the specified stress history. As before, the  $\sigma - \varepsilon$  plot is not known in advance, but is constructed as the strain evolves.

In summary,  $\sigma - \varepsilon$  plots for nonlinear elasticity are known a priori because they represent an equation of state.  $\sigma - \varepsilon$  plots for viscoelasticity are not known a priori, and are developed along with the response to a specified strain or stress history.

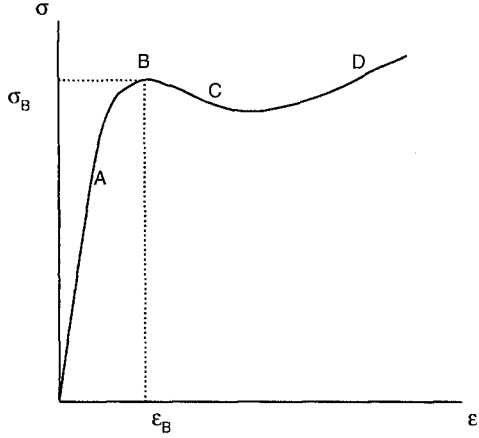


Fig. 1. A non-monotonic stress-strain plot

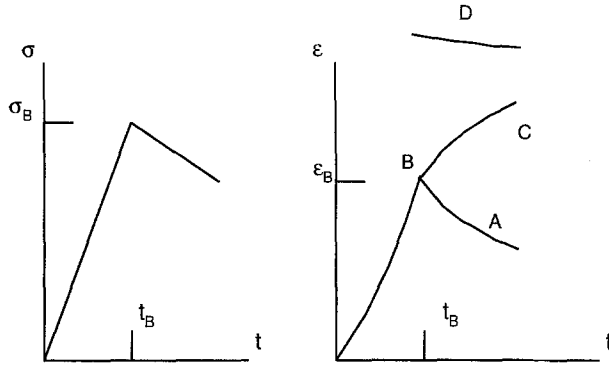


Fig. 2. A strain history with branches constructed from a non-monotonic stress-strain plot

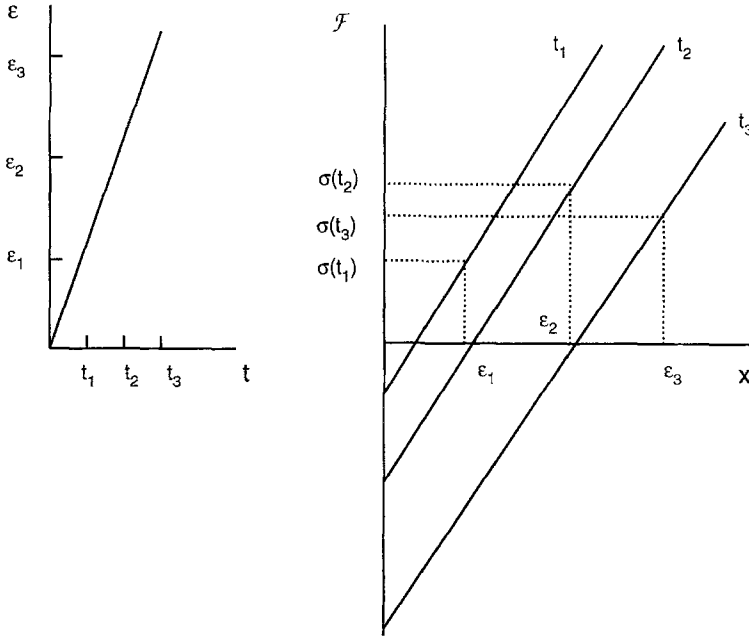
This discussion of non-monotonic  $\sigma - \varepsilon$  plots with branching of strain histories raises the questions: Is branching of strain histories possible for nonlinear viscoelastic response? If so, how are the branches determined using  $\sigma(t) = \mathfrak{F}(\varepsilon(t); \varepsilon(s)|_{0-}^{t-})$ ? This will be discussed next.

## 5 Analysis

The possibility is now considered that there can be branching of strain histories for the “strain clock” constitutive equation presented in Sect. 2. The method of analysis is similar to that used by Bernstein and Zapas [6], and Wineman [7].

The constitutive relation in Eqs. (1) or (4) has the form  $\sigma(t) = \mathfrak{F}(\varepsilon(t); \varepsilon(s)|_{0-}^{t-})$ , in which  $\mathfrak{F}(\varepsilon(t); \varepsilon(s)|_{0-}^{t-})$  is regarded as having two independent variables,  $\varepsilon(t)$  and the preceding strain history  $\varepsilon(s)$ ,  $0 \leq s < t$ , denoted by  $\varepsilon(s)|_{0-}^{t-}$ . The strain  $\varepsilon(t)$  is considered to be independent of the preceding strain history  $\varepsilon(s)$ ,  $0 \leq s < t$ , so that there may be a jump in strain at time  $t$ , i.e.,  $\varepsilon(t) \neq \varepsilon(t-)$ . Define the function  $\mathfrak{F}(x; \varepsilon(s)|_{0-}^{t-})$ , the **jump response function**, which gives the stress at time  $t$  for a strain of  $x$  at time  $t$ . The plot of  $\mathfrak{F}$  vs.  $x$  at time  $t$  is called the **jump curve** corresponding to time  $t$ . The preceding strain history  $\varepsilon(s)$ ,  $0 \leq s \leq t$ , acts as a parameter in determining the jump curve at time  $t$ . At time  $t$ , when the stress  $\sigma(t)$  is specified, the strain  $\varepsilon(t)$  is the solution of

$$\sigma(t) = \mathfrak{F}(x; \varepsilon(s)|_{0-}^{t-}). \quad (10)$$



**Fig. 3.** Use of the jump response function to construct the non-monotonic stress response to a constant strain rate history

Let  $[0, T^*]$  be an initial time interval and let  $\mathfrak{F}(x; \varepsilon(s)|_{0-}^{t-})$  be monotonic in  $x$  for each time  $t \in [0, T^*]$ . Then Eq. (10) has a unique solution for  $\varepsilon(t)$  at each time  $t$ , and the strain history does not branch during this time interval.

Suppose there is a time interval  $[T_1, T_2]$  in which  $\mathfrak{F}(x; \varepsilon(s)|_{0-}^{t-})$  is not monotonic in  $x$ . For example, for each time  $t \in [T_1, T_2]$  the jump curve has a form as shown in Fig. 1. Consider a situation similar to that discussed at the beginning of Sect. 4. For some time  $t' \in [T_1, T_2]$ , let

$$\sigma(t') = \max_x \mathfrak{F}(x; \varepsilon(s)|_{0-}^{t'-}). \quad (11)$$

At a later time  $t'' > t'$ ,  $t'' \in [T_1, T_2]$ , let

$$\sigma(t'') < \max_x \mathfrak{F}(x; \varepsilon(s)|_{0-}^{t''-}). \quad (12)$$

Then the equation

$$\sigma(t'') = \mathfrak{F}(x; \varepsilon(s)|_{0-}^{t''-}) \quad (13)$$

has several solutions, and the strain history has branched at time  $t'$ .

Now consider the constitutive equation of Sect. 2. The most convenient form of this equation for the present purposes is that in Eq. (4). The jump response function has the form

$$\mathfrak{F}(x; \varepsilon(s)|_{0-}^{t-}) = G[0]x + \int_0^t \frac{\dot{G}[\xi(t) - \xi(s)]}{a(\varepsilon(s))} \varepsilon(s) ds. \quad (14)$$

The  $\mathfrak{F} - x$  plot is a straight line with positive slope  $G[0]$  and has an intercept along the  $\mathfrak{F}$ -axis which is given by

$$\int_0^t \frac{\dot{G}[\xi(t) - \xi(s)]}{a(\varepsilon(s))} \varepsilon(s) ds. \quad (15)$$

Let Eq. (14) be substituted into Eq. (10). At each time  $t$ , Eq. (10) has a unique solution for the strain  $\varepsilon(t)$ . Thus, the strain history does not branch at any time.

This discussion explains how a non-monotonic stress history is generated by a constant strain rate extensional history. Let attention be restricted to strain histories such that  $\varepsilon(s) > 0$ ,  $s > 0$ . Then since  $\dot{G} < 0$ , the straight line intersects the negative  $\mathfrak{F}$ -axis. For a monotonically increasing strain history,  $a(\varepsilon)$  becomes very small and the distance of the intercept from the origin increases. Figure 3 shows such a strain history and the  $\mathfrak{F} - x$  plots for several times. As the strain increases, the intercept given by Eq. (15) moves faster along the negative  $\mathfrak{F}$ -axis. The stress  $\sigma(t) = \mathfrak{F}(\varepsilon(t); \varepsilon(s)|_{0-}^t)$  is seen to increase at first and then decrease.

## 6 Summary

The constitutive equation considered here is linear in the strain, with nonlinearity arising by means of a “strain clock”, i.e. a strain dependent time like variable. For such a constitutive equation it is possible to develop non-monotonic  $\sigma - \varepsilon$  plots in one dimensional response. A discussion has been presented as to why such plots have an interpretation which is entirely different from  $\sigma - \varepsilon$  plots in elasticity and do not give the response of the material under general conditions. In particular, the discussion leads to the conclusion that non-monotonicity of a  $\sigma - \varepsilon$  plot does not necessarily indicate that a strain history will branch at some time.

A method of analysis was used to determine the possibility of branching which considers the mathematical structure relating strain histories and stress histories. It was shown that branching of strain histories is not possible for a constitutive equation of the type studied here, which is linear in the strain. The results are readily extended to the three dimensional form of the constitutive equation. However, branching may be possible in constitutive equations with a “strain clock” which are nonlinear in the strain tensor, as would occur for finite strains. A study of these constitutive equations is beyond the scope of the present work. The method of analysis presented here may be of use for these more complicated equations.

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