A Brief Review of Some Approaches to Hysteresis in Viscoelastic Polymers

H.T. Banks
Center for Research in Scientific Computation
North Carolina State University
Raleigh, NC 27695-8205

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Abstract

We give a brief review of hysteresis in viscoelastic polymers. The efforts surveyed range from phenomenological to molecular modelling with applications involving recent efforts on elastomers to biotissue.

1 Introduction

In control and systems theory, delay or hereditary systems or systems with memory (hysteresis) have played an important role for many years because of the early realizations by Minorsky and others [40, 41, 56, 57, 58, 60] that feedback design based on dynamics wherein one ignores any delays may fail catastrophically to stabilize or control a system in which delays or hysteresis are present in the dynamics. This is true whether the hysteresis is a fundamental part of the underlying dynamics or a part of the input or control operator. For the latter there is a growing body of literature [11, 12, 13, 45, 54, 68] on the Preisach and related theories for hysteretic control input such as arises in smart material systems [27, 39, 64]. Applications in which delays and hysteresis play a basic role in the underlying dynamics include sustained efforts in biology with early efforts employing delay systems [1, 2, 16, 17, 28, 29, 30, 31, 32, 42, 51, 53, 55, 62] and more recent investigations involving hysteretic probabilistic structures [5, 7] as well as classical materials and electromagnetics research (see [8] and the references therein). These applications drove a substantial amount of mathematical and computational research on hysteretic systems in the last half of the 20th century, e.g., see [26, 40, 41, 46, 47] among the many books, research monographs, and research articles written. Here we shall focus on the delays or hysteresis arising in the fundamental dynamics of the systems to be stabilized or controlled. In particular we consider viscoelastic materials that are polymeric in nature. This includes a wide range of materials

of current importance such as (rubber or silicone based) filled elastomers and all types of biotissue (soft tissue, ligaments, cartilage, etc.).

2 Viscoelasticity

The mathematical modelling of *viscoelasticity* (sometimes also loosely referred to as *hysteresis*) in materials using ideas from elasticity has attracted the attention of a large number of investigators over the past century. Among significant contributors (see the many references in [33, 34, 35, 37, 38, 52, 59, 61, 65, 67, 69, 70]) have been some of the true giants from the fields of engineering and material sciences. One of the most widely used empirical models for viscoelasticity in materials is the Boltzmann convolution law [22, 35, 37, 38, 69], one form of which is given in equation (1)

$$\sigma(t) = g_e(\epsilon(t)) + C_D\dot{\epsilon}(t) + \int_{-\infty}^{t} Y(t-s)\frac{d}{ds}g_v\left(\epsilon(s), \dot{\epsilon}(s)\right) ds, \tag{1}$$

where ϵ is the infinitesimal strain, Y is the convolution memory kernel, and g_e and g_v are nonlinear functions accounting for the elastic and viscoelastic responses of the elastomers, respectively; for summaries and further references, see Chapter 2 of [38] as well as [22]. This form of model, when incorporated into force balance laws, results in *integro-partial differential equations* which are most often phenomenological in nature as well as being computationally challenging both in simulation and control design. This stress-strain law implies that the stress depends not only on the current strain and strain rate but also on the history of the strain and the strain-rate. It is very important to note that the stress-strain law (1) contains various standard *internal strain* or *internal variable* formulations as special cases. The anelastic displacement field (ADF) models of Lesieutre [48, 49] for composite materials exhibiting both elastic and anelastic displacement fields are formulated on the assumption that the host elastic material contains anelastic materials with internal strains ε_1 which are elastic strain driven. That is, the constitutive laws have the form

$$\sigma(t) = E\varepsilon(t) - E_1\varepsilon_1(t), \tag{2}$$

where the internal strain is given by

$$\dot{\varepsilon}_1(t) + \frac{1}{\tau}\varepsilon_1(t) = c_2\varepsilon(t), \quad \varepsilon_1(0) = 0, \tag{3}$$

or equivalently,

$$\varepsilon_1(t) = \int_0^t c_2 e^{-\frac{t-s}{\tau}}\varepsilon(s)ds.$$

Several generalizations of this formulation exist, e.g., Johnson, et al., [43, 44], suggest that the internal strain is strain *rate* driven, i.e.,

$$\dot{\varepsilon}_1(t) + \frac{1}{\tau}\varepsilon_1(t) = c_2\dot{\varepsilon}(t). \tag{4}$$

The Boltzmann-type law (1) (under appropriate assumptions on the past memory from $-\infty$ to 0) corresponds to an internal strain model of the form

$$\dot{\varepsilon}_1(t) + \frac{1}{\tau}\varepsilon_1(t) = \frac{d}{dt}g_v(\varepsilon(t), \dot{\varepsilon}(t)), \quad \varepsilon_1(0) = 0. \tag{5}$$

This form is often chosen since one finds that neither (3) nor (4) provide laws that readily describe experimental data, especially in the cases of filled elastomers, biotissues and other molecular polymers.

3 Fung's Quasi-linear Models

Fung, in his extensive efforts [38] with biomechanics and biotissue, develops and presents the quasi-linear viscoelastic constitutive equation

$$S_{ij}(t) = \int_{-\infty}^{t} G_{ijkl}(t-\tau)\frac{\partial S_{kl}^{(e)}[\bar{E}(\tau)]}{\partial \tau}d\tau, \tag{6}$$

where S_{ij} is the Kirchoff stress tensor, $\bar{E}$ is the Green's strain tensor, G_{ijkl} is a reduced relaxation function, and $S_{kl}^{(e)}$ is the "elastic" stress tensor. For the scalar components G_{ijkl}, Fung proposes the reduced relaxation function $G(t)$ given in the form

$$G(t) = \left\{1 + C[E_1(\frac{t}{\tau_2}) - E_1(\frac{t}{\tau_1})]\right\}[1 + c\ln(\frac{\tau_2}{\tau_1})]^{-1}. \tag{7}$$

Here $E_1(z) = \int_z^\infty \frac{e^{-t}}{t}dt$, C represents the degree to which viscous effects are present, and τ_1 and τ_2 represent fast and slow viscous time phenomena. We note that the internal strain variable formulation (2), (5) is equivalent to the constitutive relationship proposed by Fung if one considers an approximation of the relaxation function G by a sum of exponential terms. Various internal strain variable models are investigated in [3] and a good agreement is demonstrated between a two internal strain variable model (e.g., of the form $\sigma = E\varepsilon - E_1\varepsilon_1 - E_2\varepsilon_2$) and undamped simulated data based on the Fung kernel G.

Since its introduction, this quasi-linear viscoelastic (QLV) theory of Fung has been applied successfully in stress-strain experiments to several types of biological tissue. A benefit to using (6) as a constitutive equation is that, unlike simpler models for viscoelasticity, it allows for the consideration of a continuous spectrum (e.g., see the discussions in [38]) of relaxation times and frequencies (this is also true of the probabilistic-based internal variable approach developed in [23] and described below). (The need for a continuum of relaxation times in certain materials was observed many years ago [36, 63, 66, 70].) While Fung's theory has been successfully employed for fitting hysteretic stress-strain curves, for control applications one is interested in using it in a full dynamical model. Unfortunately, the QLV, as presented by Fung, leads to exceedingly difficult computations within full dynamical partial differential equations, especially in estimation and control problems. This motivated the development of the internal variable approach described in [3, 23, 48] (which permits discrete approximation to a continuum) in attempts to approximate well the corresponding *dynamic responses* even in cases where the *stress-strain curves alone* do not produce adequate approximations – see [38].

The probabilistic based internal variable alternative [23] to Fung's kernel involves a parameter dependent kernel with a continuous distribution of parameters and internal variables. In the case of a finite combination of Dirac δ distributions, one obtains a finite summation of exponential functions as the approximation kernel (see the discussions below). This method can be extended to allow for consideration of a continuous spectrum of relaxation times and frequencies by utilizing absolutely continuous parameter distributions in place of the δ distributions.

4 Internal Variables

The internal variable approach to overcome both conceptual and computational challenges is consistent with the belief that hysteresis is actually a manifestation of the presence of multiple scales in a physical or biological material system that is frequently modelled (and masked) with a phenomenological representation such as an hysteresis integral for the macroscopic stress-strain constitutive law. The *internal variable* modelling leads to an efficient computational alternative for the corresponding integro-partial differential equation models. In addition, it provides a "molecular" basis for the models (for a comparison of models of viscoelastic damping via hysteretic integrals versus internal variable representations, see [22] and the references therein).

Our own interest in viscoelasticity in polymeric materials has been motivated by projects in our Industrial Applied Mathematics Program with at least two of our industrial partners: The Lord Corporation and Medacoustics, Inc. The collaborations with polymer scientists and engineers at Lord involved the dynamic modelling of filled rubbers which experimentally exhibit both significant hysteresis and nonlinearity in tensile and shear deformations as depicted in the sample stress-strain curves in Figure 1. The efforts with engineers at Medacoustics used some of the viscoelastic models we have investigated in attempts to understand the propagation of arterial stenosis induced shear waves in composite biotissue in a sensor development and characterization project.

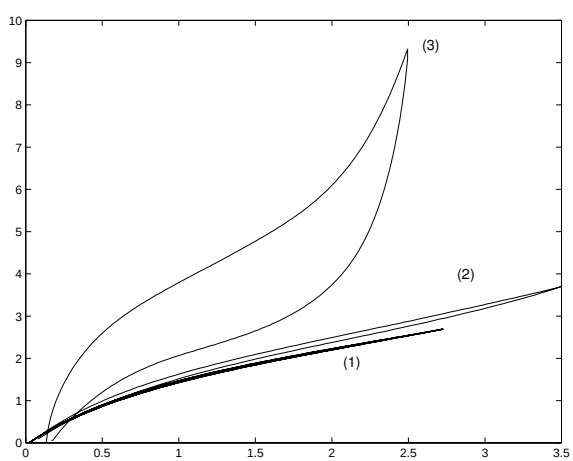

Figure 1: Experimental stress-strain curves for (1) unfilled, (2) lightly filled and (3) highly filled rubber in tensile deformations.

In some of our earlier efforts [24, 25], the models for hysteretic damping in elastomers employed a phenomenological Boltzmann-type constitutive law of the form (1). As explained in [20, 24], our nonlinear materials undergoing large deformations required the use of *finite* (as opposed to *infinitesimal*) strain theories [61]. However, since the nonlinearity between the stress and finite strain is an unknown to be estimated (using inverse problem algorithms) and since the finite strain can be expressed in terms of known nonlinearities as a function of the infinitesimal strain (at least in the problems of interest here), one can effectively formulate the problem as one of estimating the unknown nonlinearity between stress and infinitesimal strain (see [24]). Hence one can develop models for stress in terms of infinitesimal strain. Our previous efforts as summarized in [20, 21] have shown, through comparison with experimental data, that the best fit to filled elastomer data occurs when g_e and g_v are cubic, along with Y as a *distribution* of decaying exponentials. We subsequently [18, 19, 20, 21] developed nonlinear models based on stick-slip "molecular" ideas of Johnson and Stacer [43] and Doi and Edwards [34] which resulted in a form for g_e, g_v and Y in (1) that matched the empirical findings reported in [20, 24, 25]. These models allow for multiple relaxation times present in polymer strands of composite materials within a virtual compartmental model of entangled chemically cross-linked/physically constrained system of long chain "molecules". While accounting for multiple relaxation parameters, these models do not include physically or chemically based parameters in representations of the polymer strands.

We mention briefly two recent advances: (i) a new constitutive model [10] that has been developed which combines the virtual stick-slip continuum "molecular-based" ideas of Johnson and Stacer [43] with the Rouse bead chain (see Figure 2 below) ideas as described in Doi and Edwards [34]; (ii) a two dimensional version [14, 15, 50] of a model that accounts for stenosis driven shear wave propagation in biotissue.

5 A Stick-Slip/Rouse Hybrid Model

The early models and the nonlinear extensions of the Boltzmann law did not provide insight into the underlying mechanisms for tensile and/or shear deformations in filled rubber or biotissue. This is not unexpected since the approaches described above are based on pseudo-phenomenological formulations. We then [19, 20, 21] turned to a different approach based on molecular arguments which, as we shall see, lead precisely to the class of models based on a Boltzmann hysteresis formulation. As usual, one begins with force and moment balance and seeks constitutive laws for the viscoelastic stress term σ_{visco} in

$$\sigma(t;\tau) = \sigma_{elast}(\varepsilon, \dot{\varepsilon}) + \sigma_{visco}(\varepsilon_1(\cdot)),$$

where $\varepsilon = \frac{\partial u}{\partial x}$ is the infinitesimal strain and ε_1 is an "internal strain" variable on which σ_{visco} depends in an hysteretic manner. As described above, we found that a reasonable description of the data of interest could be given with the typical stress-strain relationship

$$\sigma(t) = g_e(\varepsilon(t), \dot{\varepsilon}(t)) + \int_0^t \gamma e^{-\frac{t-s}{\tau}} \frac{d}{ds} g_v(\varepsilon(s), \dot{\varepsilon}(s)) ds, \tag{8}$$

where τ is a relaxation parameter, g_v is defined with cubic polynomials and $g_e = g_e(\frac{\partial u}{\partial x}) + C_D \frac{\partial^2 u}{\partial t \partial x}$. We have already observed that this expression is equivalent to

$$\sigma(t) = \tilde{g}_e(\varepsilon(t), \dot{\varepsilon}(t)) + \gamma \varepsilon_1(t; \tau),$$

where, for a given "relaxation parameter" τ, the internal strain $\varepsilon_1(t; \tau)$ satisfies (5). In fact, we found that highly filled rubbers required multiple relaxation times τ_1, τ_2 in an approximation to (7) to obtain good model fits to the data. As one might expect, molecular based formulations, where microscopic relaxation parameters vary across the population of molecules in the material, lead to internal dynamics of the form (5),(8) that involve multiple values of τ. When combined with a Prohorov metric framework (see [4, 6]) for uncertainty in internal dynamics, these ideas lead to the computational models we have used. Indeed, the molecular based approach leads to a general class of models with uncertainty or randomness in the stress

$$\sigma(t, x; P) = \tilde{g}_e(\varepsilon(t, x), \dot{\varepsilon}(t, x)) + \gamma \int_{\mathcal{T}} \varepsilon_1(t, x; \tau) dP(\tau), \tag{9}$$

where P is a probability distribution over the set $\mathcal{T}$ of possible relaxation parameters, and $\varepsilon_1(t; \tau)$ satisfies, for each $\tau \in \mathcal{T}$,

$$\dot{\varepsilon}_1(t, x; \tau) + \frac{1}{\tau} \varepsilon_1(t, x; \tau) = \dot{\varepsilon}(t, x) h(\varepsilon(t, x)).$$

For the reptation model derivation in [19], one begins with the Doi/Edwards [34] stick-slip molecular models as embodied in the continuous tube reptation models of Johnson/Stacer [43] wherein polymer materials such as rubber are postulated to be composed of two types of molecules. In tensile deformations, one denotes by $L(t)$ the length of *chemically cross-linked* or CC molecules, while $\ell(t)$ denotes the length of *physically constrained* or PC molecules. To use stick-slip models in continuum simulations of reptation in rubbers, one considers networks of "cells" or boxes of parallel-sided CC boxes and PC boxes with sides of length (principal stretches)

$$\lambda_c = 1 + \varepsilon = 1 + \frac{\partial u_c}{\partial x}, \quad \lambda_p = 1 + \varepsilon_1 = 1 + \frac{\partial u_p}{\partial x},$$

respectively. Here u_c denotes the deformations of the CC box and u_p denotes the deformations of the PC box. Using a linear stick-slip assumption as in [43], and strain energy densities based on experiments of Young and Danik (see [18, 20] for details), one obtains as a limit of PC response to step tensile deformations of the CC molecules, the $\varepsilon, \varepsilon_1$ coupled dynamics

$$\dot{\varepsilon}_1 + \frac{1}{\tau} \varepsilon_1 = \dot{\varepsilon} \frac{1 + \varepsilon_1}{1 + \varepsilon}.$$

However, if one replaces the linear assumption of [18] by a nonlinear stick-slip hypothesis (which is the basis of the work in [19]), one obtains a more general nonlinear, dynamical relationship between ε and ε_1 given by

$$\dot{\varepsilon}_1 + \frac{1}{\tau} \varepsilon_1 = \dot{\varepsilon} f((1 + \varepsilon_1)/(1 + \varepsilon)).$$

Expansion and truncation of higher order terms lead to equations of the form

$$\dot{\varepsilon}_1 + \frac{1}{\tau}\varepsilon_1 = \dot{\varepsilon}(\alpha_0 + \alpha_1\varepsilon + \alpha_2\varepsilon^2 + \alpha_3\varepsilon^3), \tag{10}$$

which are of the same form as the internal variable model (5),(8) with g_v a cubic polynomial. For the corresponding contributions to σ from the strain energy densities of Young-Danik/ Johnson-Stacer with the nonlinear stick-slip hypothesis, one obtains a contribution to the rate independent strain g_v^s (after expanding f in a Taylor series and dropping higher order terms) of the form

$$g_v^s(\varepsilon, \varepsilon_1) = g_{cubic}(\varepsilon) + \gamma_1\varepsilon_1,$$

where ε_1 is as before (i.e., the internal strain satisfying (10)). Thus, the total stress-strain relationship can be written in the form (9). If the measure P of (9) has atoms at τ_1 and τ_2, (i.e., the measure is composed of Dirac measures concentrated at τ_1 and τ_2), then the constitutive law leads precisely to the model

$$\sigma(t, x; P) = \tilde{g}_e(\varepsilon(t, x), \dot{\varepsilon}(t, x)) + \gamma_1\varepsilon_1(t, x; \tau_1) + \gamma_2\varepsilon_2(t, x; \tau_2),$$

which was used in the data fits in [3, 20]. Further details on this modeling approach can be found in [9, 21]

We next developed a new constitutive model which combines the "molecular-based" ideas of Johnson and Stacer with the Rouse bead chain ideas and explained its relation to the Boltzmann phenomenological models.

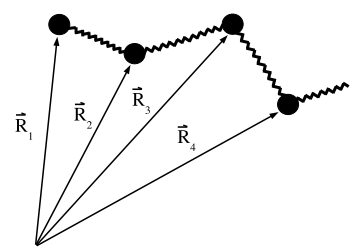

Figure 2: Representation of vectors for a bead-spring polymer molecule.

The new molecular-based constitutive model, in which polymer chains are treated as Rouse type strings of interconnected beads (a reasonable approximation for many materials), permits the incorporation of many important physical parameters (such as temperature, segment bond length, internal friction, and segment density) in the overall hysteretic constitutive relationship. Its form is similar to that developed in [19, 20] and does have the general form (1) of Boltzmann type, even though the kernel is *not* of convolution type. The resulting model does, however, provide a molecular basis for the earlier pseudo-phenomenological stick-slip models.

We give only a brief outline of the new constitutive model here; more details of the derivation can be found in the report [10]. We model a polymer material undergoing directional deformation by assuming it is composed of two *virtual* compartments as depicted in Figure 3.

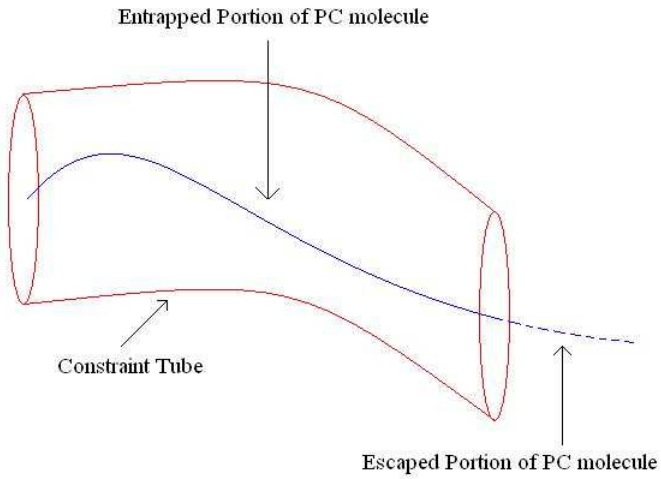

Figure 3: PC molecule entrapped by the surrounding constraining tube.

One compartment consists of a *constraining tube* which is a *macroscopic* compartment containing both CC (chemically cross-linked) and PC (physically constrained) molecules. The other compartment is *microscopic* in nature and consist of those PC molecules aligned with the direction of the deformation. These molecules will at first "stick" to the constraining tube and be carried along with its motion, but will very quickly "slip" and begin to "relax" back to a configuration of lower strain energy. In the model derivation one computes the contributions of both "compartments" to the overall stress of this polymer material undergoing deformations to obtain the constitutive law.

6 Stenosis-Driven Shear Wave Propagation in Biotissue

Finally, we turn to recent results on the viscoelastic models for propagation of stenosis-driven biotissue waves mentioned above. In the biotissue efforts [14, 15, 50], we employ an internal variable formulation of Boltzmann type hysteresis laws to investigate the propagation of stenosis generated waves in biotissue where it has been demonstrated that a viscoelastic (as opposed to an elastic) formulation is important and that waves generated in a two-dimensional cylindrical geometry with inner radius partial occlusions can be readily modelled and simulated.

Specifically we have carried out efforts on two and three dimensional models that employ an internal variable approach to model wave propagation. To motivate this, we recall [3] that coronary artery disease (CAD) is caused by atherosclerosis, the gradual accumulation of plaque along the walls of an artery. This buildup, known as a stenosis, restricts the flow of blood, leading to a decrease in the oxygen supply to the heart muscle. It is well known that

arterial stenoses produce sounds due to turbulent blood flow in partially occluded arteries. In principle, turbulent normal wall forces exist at and downstream from an arterial stenosis, exerting pressure on the wall of the artery which then causes a small displacement in the surrounding body tissue. The goal is to model the propagation of the wave generated from the stenosis to the chest wall, and ultimately, to create an inverse problem methodology which can be utilized to determine the location of an arterial stenosis. In [14, 15, 50] we also discuss comparison of the viscoelastic model to an elastic one as well as present typical simulations for a biologically motivated example.

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