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Nonlinear viscoelastic solids

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Abstract

An overview of constitutive equations and models for fracture and strength of nonlinear viscoelastic solids is given. Recent work is emphasized. Research needs are listed in the concluding section.

1. Introduction

Many materials of engineering importance exhibit significant nonlinear viscoelastic behavior in physical environments and under mechanical loading of practical interest. Plastics and rubber, with and without reinforcement (Lai and Bakker, 1995; Tuttle et al., 1995; Ha and Schapery, 1998), asphalt concrete (Uzan, 1996), polycrystalline ice (Schapery, 1997b) and biological materials (Fung, 1993) are examples. Viscoelastic behavior manifests itself in various ways, including creep under constant load, stress relaxation under constant deformation, time-dependent recovery of deformation following load removal, time-dependent creep rupture and frequency-dependence of fatigue strength. If, following load removal, a measurable portion of the deformation does not vanish after a long period of time, and this residual deformation is affected by the time under prior loading, then this deformation is commonly called viscoplastic. Viscoelastic solids, as discussed here, may or may not exhibit viscoplasticity.

Our attention here is mostly on mechanical behavior of monolithic and composite materials, and not on behavior of engineering structures made of these materials. There are two main sections in this paper; the first is on constitutive equations and second is on fracture and strength models. The emphasis is on nonlinearities, rather than linear effects. These nonlinearities may be due to intrinsic, locally nonlinear stress-strain behavior that exists practically down to the atomic or molecular scale, or may be due to the combined effect on macro-stress-strain equations of many defects that are large enough to be modeled using local continuum mechanics such as distributed microcracks or dislocations. Procedures

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for handling geometric nonlinearities (large strains and/or rotations) through use of appropriate stress and strain measures are well-established (Haddad, 1995; Arruda and Boyce, 1993; O'Dowd and Knauss, 1995), and thus will not be addressed.

It is quite common now to account for linear viscoelastic behavior of some materials, such as polymers, when analyzing their short and long-term performance. However, the assumption of linearity is often used even when nonlinear behavior is significant because the nonlinearities are not well understood and/or useful experimental and theoretical means are not available for nonlinear viscoelastic characterization and analysis. The rapid growth of computing power and availability of advanced finite element software that allows for the use of customized material models appear to have removed the computational aspect as a roadblock to progress in accounting for nonlinear viscoelasticity in many applications (Shaw et al., 1997). Instead, it is the experimental and realistic theoretical characterization of mechanical behavior of the materials where the greatest needs are at this time. Primarily theoretical aspects are covered here, while another paper in this volume is on developments in experimental solid mechanics (Knauss).

There is no attempt to provide a historical perspective or an exhaustive review of the literature. Instead, in most cases we list in a bibliography only relatively recent publications that can give the interested reader a more detailed picture of the current state-of-the-art and a more complete listing of publications.

2. Constitutive equations

Nonequilibrium thermodynamics has proved to be very helpful in constructing general and specific mathematical models that interrelate *histories* of the stress tensor, strain tensor, temperature and other parameters, such as moisture in polymers (Schapery, 1997a; Lustig et al., 1996; Weitsman, 1990). The approaches are essentially of two types: functional thermodynamics and state-variable thermodynamics. In the first one, scalar response quantities, like the free energy, are expressed as functionals of the strain (stress) tensor, temperature, etc. and then rules of functional differentiation are given for expressing the stress (strain) tensor in terms of the free energy. In the second type a more traditional approach is used in which the free energy is expressed as a function of current values of strain (stress), temperature and other variables, including so-called internal state variables (ISVs). The ISVs may be identified with specific, local micro-structural rearrangements or suitably defined averages. Rate effects are introduced through evolution equations, which usually relate time rates-of-change of ISVs to thermodynamic forces; the latter are derivatives of the free energy with respect to each ISV. In some special cases the rate equations may be explicitly solved to obtain the ISVs in terms of the history of 'external' quantities like the stress or strain tensor and temperature. From this result one may explicitly eliminate the ISVs from the constitutive equations, thus obtaining a result similar to that from the functional thermodynamic theory in which stress (strain) is expressed as a functional of strain (stress) and other relevant variables like temperature.

An advantage of the state variable approach is that physical theories, such as rate process and dislocation models, may be introduced directly in the formulation of the evolution equations. Also, many if not all ISVs may produce only small changes in energy; then, one may use a power series expansion for free energy in which third and higher order terms in the ISVs are neglected (Schapery, 1997a). Although considerable progress has been made in the thermodynamic formulations, there are many assumptions that have to be made to arrive at constitutive equations which are simple enough to carry out a practical program of experimental evaluation of the various material constants and functions. What is needed are more realistic, physically-based theories for establishing the free energy and evolution equations in terms of fundamental material parameters as well as the stress or strain state.

Explicit representations of the stress-strain behavior have been developed from thermodynamics or purely mathematical assumptions and expressed as either single or multiple-integrals of stress or strain history (Haddad, 1995). The latter form comes from a functional expansion much like a Taylor series expansion of a function. Based on studies of actual mechanical behavior to-date the author does not believe one needs to use the more complex characterization involving multiple integral expansions.

It is generally accepted that transient creep or stress-relaxation is significant over many decades of time for most nonlinear viscoelastic solids. This implies many ISVs, and associated time constants, are needed in the characterization, with at least one needed for each decade of time. Although the physical bases for such broad-spectrum behavior is reasonably well understood, there do not appear to be any widely accepted physical models which show explicitly how nonlinear effects enter this behavior. There are, of course, various models that have been proposed and applied with various degrees of success achieved under limited conditions of loading (e.g. Gates and Sun, 1991; Hasan and Boyce, 1995; Kody and Lesser, 1997; Lai and Bakker, 1996; Lesser and Kody, 1997; Popelar et al., 1990). Molecular dynamic models help to provide such information, but much remains to be done before they can provide the type of information needed for modeling nonlinear viscoelastic behavior.

Consider, for purposes of illustration, the present state of affairs of physically-based models for an important class of materials: amorphous, glassy polymers. Suppose the temperature of a polymer is initially above the so-called glass transition temperature, Tg, and then the temperature is taken below Tg; the polymer goes from a rubbery state to a glassy state with a much higher modulus. The viscoelastic behavior when $T < Tg$ is dependent on how rapidly the material is cooled and on the time that has elapsed since cooling to a fixed temperature; this dependence on time after cooling is called *physical aging* (Bradshaw and Brinson, 1999; McKenna et al., 1995). With further decreases in temperature both the aging and viscoelastic effects under mechanical loading become weaker. Also, the material becomes more brittle.

Physical aging is generally attributed to a slowly decreasing *free volume*. Free volume is identified with molecular packing irregularities which are distributed nonhomogeneously throughout the material. The larger the free volume, the greater is the mobility of molecular response to external loading. Below Tg this implies that rate or time effects diminish with diminishing free volume. The rate of collapse of overall volume is itself a viscoelastic process, but one that is nonlinear in that it depends strongly on the free volume. Limited models exist for predicting the overall volume in terms of free volume; the overall volume is assumed to be the sum of free volume and occupied volume, the latter being dependent on temperature, but not on time. Existing physically-based mathematical models appear to provide good quantitative predictions of overall volume over only a relatively narrow temperature range close to Tg (McKenna and Simon, 1999). A controversial issue is concerned with how applied stress affects the physical aging. Some investigators have interpreted their data to mean that application of a sufficiently high stress rejuvenates the material; thus, following stress removal, the material ages like one with a larger free volume than if the high stress had not been applied. However, Waldron and McKenna (1995) and Lee and McKenna (1990), in a series of experiments on changes in tensile and shear compliance and volume change following a quench, provide data that strongly support the view that the material is not rejuvenated. After the stress is removed, the instantaneous volume follows the same curve that would have existed without prior stressing. During the time stress is applied the aging rate, as implied by the change in shear compliance, is affected.

With or without physical aging, the only generally accepted physical model for nonlinear material behavior of glassy polymers at small strains is based on free volume (Losi and Knauss, 1992). In this model all nonlinear behavior comes from only the material's time scale through a free volume-dependent reduced time. Both dilatational and distortional stress (or strain) invariants appear to be needed to obtain observed experimental behavior; thus, the free volume is not related in a one-to-one fashion to

overall volume (Popelar and Liechti, 1997). There is no consensus on whether free volume depends directly on current stress or strain or is a function of the history of one or the other.

The conventional free volume model implies all material relaxation times are affected equally by the nonlinearity, just as they are affected by temperature for thermorheologically simple materials. While there is considerable experimental support for this uniform effect on the material time parameters, there does not appear to be any satisfying theoretical explanation of this simplicity (and conditions for which it is not true) for the glassy state. There are many experimental results that show sources of material nonlinearity exist beside that affecting the time-scale (Schapery, 1997a). Implications of these results, in terms of state-variable thermodynamics, is that the free energy contributes to the nonlinearity through third and high order terms in strain or stress. If all nonlinearity is in the time scale, it is only the entropy production rate that is a thermodynamic source of material nonlinearity. There does not appear to be any molecular model for explaining nonlinearity in the free energy at small strains. Under certain conditions some glassy polymers may undergo large strains without breaking (Boyce et al., 1988, 1994). Molecular-based theory of material stiffening has been successful in predicting overall stiffening under uniaxial and multiaxial stresses (Tomita and Tanaka, 1995; Liang and Liechti, 1996); this stiffening is similar to that for rubber, and is attributed to the stretching of polymer molecules that approach their limit of extensibility, while being restrained by entanglements or crosslinks.

Viscoelastic materials with a reinforcement phase, like particle-reinforced rubber, fiber-reinforced plastic and asphalt concrete, may undergo considerable stable, distributed microcracking (including separation between matrix and reinforcements) prior to overall fracture. Growth of this damage and resulting overall nonlinear behavior must be taken into account to make realistic strength and durability predictions in many applications. An assumption that all nonlinearity is due to microcracking has enabled realistic, practical constitutive models to be developed. Viscoelastic microcrack growth equations provide, in-effect, nonlinear evolution equations for use in a state variable thermodynamic formulation of constitutive equations for uniaxial and multiaxial loading. Such equations have been successfully used to characterize mechanical behavior of solid propellant (highly-filled rubber) (Park and Schapery, 1997; Ha and Schapery, 1998), asphalt concrete Park et al. (1996), swirl-mat glass reinforced epoxy (Abdel-Tawab et al., 1997) and polycrystalline ice (Schapery, 1991). Except for solid propellant, most experimental characterization and evaluation of the nonlinear theory has been for uniaxial loading and simple stress histories. Damage-induced anisotropy may be quite large, especially in initially isotropic materials. Although there are some theoretical models available for predicting this anisotropy (Schapery, 1999), again there is very little related experimental work.

Micromechanical modeling has helped to provide useful measures of continuum damage and how it affects homogenized mechanical response. However, quantitative predictions of various types, including how volume fraction and particle size distribution affect damage and other sources of nonlinearity in soft matrices, as well as the effect of high stress gradients, are practically non-existent. Only now has computational mechanics reached the point where such things can be calculated for linear elastic composite systems (Fu, 1998; Babuska et al., 1998).

3. Fracture and strength models

Although much progress has been made in modeling failure of materials exhibiting elasto-plastic behavior, this is not true for nonlinear viscoelastic solids except in special cases when viscoplastic deformation dominates the behavior or damage growth is of a very simple type involving only one mode (e.g. dominant opening mode crack). However, as in the case of constitutive equations, theory has been useful for constructing those measures of damage that are responsible for the failure and for predicting failure in *linear* viscoelastic materials under complex loading by combining effects of special types of

loading (such as constant stress and cyclic loading under constant amplitude and frequency). For limited materials, loading conditions and temperature ranges existing approaches have been successful in predicting failure, including failure at long times of loading from short-time experimental characterization using temperature-reduced time (Miyano et al., 1997; Reeder et al., 1999). For composites, such as laminates of fiber-reinforced plastic, there may be multiple damage modes (e.g. matrix cracking, fiber breakage, delamination) that grow at different relative rates under different load histories (e.g. fatigue compared to constant load) (Dutta and Hui, 1996; Moore and Dillard, 1990; Raghaven and Meshii, 1996; Reifsnider et al., 1998). Unless the different modes and their growth are properly modeled one can hardly have confidence in predictions far from the conditions used in the experimental characterization program. Chemical and physical aging, if significant, obviously adds significant further complexity.

Damage and other sources of softening (Bazant, 1995; Schapery and Sicking, 1995; Wu and van der Giessen, 1994) may produce strain localization, in which failure is initiated because of a resulting deformation instability. An important example of this behavior for unidirectional fiber composites is that of a local shear instability (often called fiber microbuckling) which is a result of initial fiber waviness and reduction of the principal composite shear modulus as waviness grows locally due to axial compressive loading. Typically, this failure process leads to initation and growth of a kink band, followed by global failure if the composite is not constrained, such as by other layers in a multidirectional laminate. Significant progress in theory and experiment for predicting the compressive strength (point of shear instability) has been made for elastic-plastic and viscoplastic behavior (Vogler and Kyriakides, 1999; and Hsu et al., 1999). Progress has also been made for failure prediction of nonlinear viscoelastic composites (Schapery, 1993), but at this time experimental work seems to be limited to only monotonic, uniaxial loading.

Prediction of the growth of individual cracks in viscoelastic (and other) materials requires knowledge of both crack growth resistance (which is usually defined by behavior of the highly nonlinear and damaged material at the crack tip) and the crack driving force (originating from the continuum around the crack tip). For some materials, especially composites, the surrounding material may be in a damaged state. This view of the problem implies that one can identify a crack tip zone which may be characterized by itself, assuming of course the appropriate local constraints are recognized. Whether or not this is always or usually the case is not a settled issue. Nevertheless, it is a standard assumption in available models (Knauss, 1993a, 1993b; Bazant et al., 1997; Li et al., 1997). Generalizations of the J-integral have been proposed as crack driving forces (Schapery, 1990), but so far this approach considers only opening mode crack growth and is restricted to limited types of nonlinear viscoelastic behavior.

4. Research needs

Research activities on nonlinear viscoelastic solids have been quite intense during the past decade. During the 1960s there was also intense activity; but it tapered off, leaving many issues unresolved until the most recent period. Some developments that are believed to have stimulated the current level of activities are: rapidly growing computing power, enabling the use of nonlinear viscoelastic constitutive equations in processing, manufacturing and structural analyses; increasing availability and applications of polymers and polymer composites in structures ranging from the micro- to the macro-scale; growing demand for better structural performance, reliability and durability; and experimental findings that have highlighted the shortcomings of existing models of mechanical behavior. With these points in mind, the primary needs may be summarized as follows, in which there are needs for both experimental and theoretical studies:

- Better understanding of molecular and microscale rearrangements that influence nonlinear deformation and fracture behavior, especially for amorphous and semi-crystalline polymers and composites, in the range between linear viscoelastic behavior and high-strain viscoplastic behavior.
- A closer collaboration of the mechanics and materials science communities.
- Mechanical behavior and aging studies for development of nonlinear constitutive equations and failure models considering effects of multiaxial stresses and their histories, including studies on the effect of rotation of principal stress or strain axes; transient and cyclic temperature effects (and, for polymers, effects of plasticizers such as moisture).
- Identify input histories for use in experimental studies which enable critical evaluation of theoretical models.
- Effects of distributed damage and other softening mechanisms, especially localization and related stress gradient and size effects.
- Interaction of damage modes and its effect on strength and durability.
- Improved models of crack tip material behavior for use in crack growth models.
- Improved crack driving forces that account for crack tip loading in opening and mixed-mode crack growth in general nonlinear viscoelastic solids.
- Accelerated characterization of deformation, strength and lifetime.

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