Research Summary:
Object Oriented Finite element analysis for materials science*: A tool for viscoelastic polymer composite deformation analysis

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Abstract

A public domain code "Object Oriented Finite element analysis for materials science" (OOF) has been extended to include tools for analysis of viscoelastic materials. Utility of these tools has been discussed along with possible applications in this publication. Added features in OOF include means to quantitatively analyze the spatiotemporal response of a composite polymeric material in dynamic as well as in static deformation conditions. These coupled with the existing features of OOF, in particular, the complete analysis of mechanical characteristics of materials provide a comprehensive tool for the studies of timedependent behavior of variety of materials including polymeric solid composites, polymer nanocomposites, polymer blends, block copolymers, and so on. The viscoelastic module draws its strength from the underlying OOF architecture to provide a macroscopic evaluation of mechanical properties using microstructural details. An application of this module for deformation analysis is the characterization of mechanical behavior a polymer nanocomposites. The deformation behaviour of polymer composite depends on the combined characteristic relaxation times of its consituents as well as its microstructural details. Results of analysis are expected to provide better insight into the role of microstructure as well as the role of interphase on the average mechanical properties and their variations.

1 Introduction

Polymeric composites show a very wide range of mechanical responses, determined by the microstructure and inherent mechanical response of the constituent materials. The mechanical response of constituent polymer materials itself is determined by the orientation and the ease of motion of component macromolecules [1]. This concerted motion of the macromolecules manifests usually as a macroscopic viscoelastic deformation response of the polymeric material. In general, the viscoelastic response could encompass of a range of relaxation time scales reflecting the multiple time and length scales of dynamics of the macromolecular structure on deformation. When multiple phases are present as in the case of a composite along with the constituent properties, the inteface properties also play a crucial role in dictating the mechanical behaviour of the entire composite. The mechanical behaviour often differs on the surface, interface or within small domains of a composite and any one these behaviour could drastic alter macroscopic deformation behaviour of the sample.

From the point of view of applications, it is imperative that the interactions between the different deformation mechanisms are well understood and charac-

*Originally developed by Steve Langer, Ed Fuller, Craig Carter and Edwin Garcia. Center for Theoretical and Computational Materials Science, National Institute of Standards and Technolgy. For more details, http://www.ctcms.nist.gov/oof/

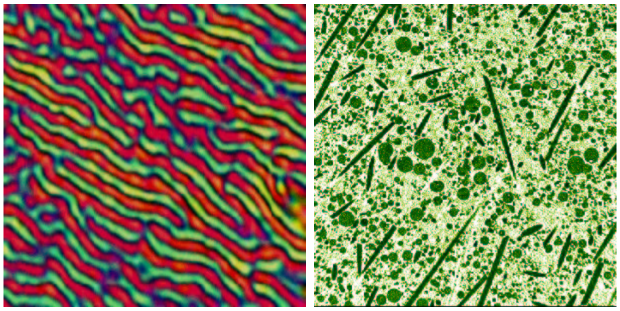

Figure 1: Microstructures that can analysed with viscoelastic module for OOF. The microstructure on the left and right correspond to a PS-PMMA thin film and glass filled epoxy composite material respectively.

terized accordingly. One area of importance for viscoelastic composite material is the structural materials with desirable dissipation characteristics. Reduction of noise or damping vibration is a crucial aspect for components in many device applications. Alternatively, a small disipation composite may be useful in dimensional stability of the composite material structure. These design goals are usually acheived by tuning the properties of the constituents of the composite or microstructural topology [2] therby altering the damping charcteristics of the polymer composite.

Realization of the design goal of a device for its deformation properties often depends on the model chosen for characterizing the composite material. There are several analytical models in the literature for the estimation the composite properties from a given set of constituent properties with a simple microstructure[3]. These models are restricted to simple morphologies and microstructures and usually in the limit of small volume fractions of inclusions. There also have been attempts to extend to higher volume fraction of inclusions inthe composite but the success is still limited to either uniformly random structures or uniformly ordered structures. Most of the real composite microstructure fall in between these two extremes.

Finite element methods provide an excellent alternative for evaluation of the material response, particularly when the material has a complex microstructure which cannot be easily handled by analytical means. Owing to its capability to provide a information on the average properties of the material along with the spatial distribution of micromechanical quantities, FEM is widely used to model and test the material qualities, particularly in the area of polymer rheology. The analytical methods provide a cost effective way to infer the bounds on the properties of the composite material for a given set of constituent materials and the FEM methodology extends it to pinpoint the composite microstructure that shows the desired charactristic of the composite material.

2 Viscoelastic behaviour

A distinctive feature of the mechanical behavior of polymers is their time dependent response to the applied stress or strain which depends on the rate of loading. The behavior can be thought of as being in between the ideal solid and liquid behaviour. At low temperatures and high strain rates, these materials display solid like Hookean behaviour and in high temperatures and low strain rates, polymers exhibit liquid like, viscous behaviour.

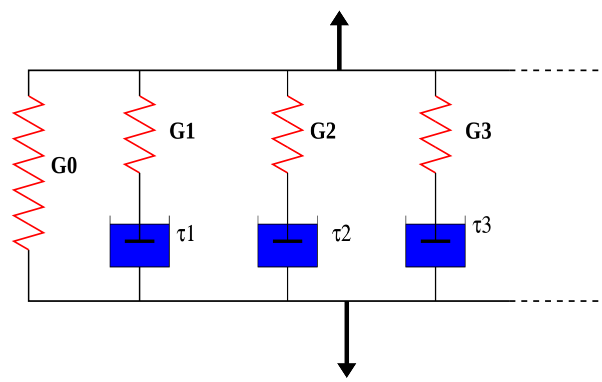

Figure 2: Standard linear solid - spring and dashpot model for viscoelastic behavior.

In the intermediate ranges of temperature and strain rates, the deformation is usually more complicated. This deformation behaviour is usually modelled with the assumption that the deformations are small and can be decomposed to purely elastic and purely viscous components. This is the assumption of linear viscoelastic behaviour and this combined with the Boltzmanns superposition principle leads to a simpler picture of deformation behaviour of polymeric material. There are a variety of spring-dashpot models which recover the deformation behaviour of viscoelastic material at appropriate experimental conditions. We chose standard linear solid model, which is basically a several Maxwell element constructed in parellel, for the description of linear viscoelasticity (see Figure 2). This model recovers both solid and liquid like behavior in appropriate experimental conditions.

An intergral representation of the constitutive equation for the viscoelastic response is given as

$$\sigma(t) = \int_{\infty}^{t} \mathbf{G}(t')\dot{\epsilon}(t-t')dt' \quad (1)$$

where dot refers to time derivate.

The relaxation function or the kernel is given as

$$G(t) = G_{\infty} + \sum_{i=1}^{N} G_i exp(-\frac{t}{\tau_i}) \quad (2)$$

where G_{∞} is the long term modulus (solid like), G_i's are the partial modulii with corresponding relaxation time scales τ_i. This expression forms the Prony series for the relaxation function.

For the simpler case of deviatoric stress,

$$\mathbf{S}(t) = \mathbf{G}_{\infty}\epsilon(t) + \sum_{i=1}^{N} \mathbf{G}_i \int_{\infty}^{t} exp(-\frac{(t-t')}{\tau_i})\frac{\partial \mathbf{e}(t')}{\partial t}dt'. \quad (3)$$

3 Finite element method for viscoelastic behavior

The integro-differential constitutive equation as given in previous equation is difficult to handle numerically, as it requires entire history of process to be stored for the evaluation of current state of the deformation. Even with a cutoff of few hundreds of timesteps in memory would enormously slow down the computational speed, as for every step in memory entire state of the system needs to be stored. We improved upon an algorithm originally proposed by the Zienkiewics and Taylor [4], wherein the total time is decomposed into several intervals. To evaluate the current deformation state, the integrals need to be evaluated only for the current time interval, and a recursion provides the information about state of the system in previous time step. The details of this routine are given elsewhere. This eliminates the necessity of storage of entire history and helps increasing the speed of simulation process.

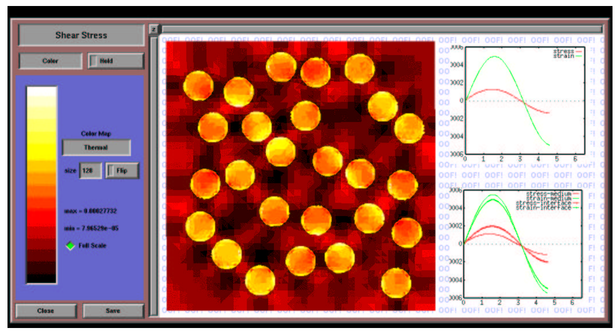

Figure 3: A typical OOF output : inhomogenous spatial distribution of stress component σ_{xy} along with the temporal evolution of average micromechanical quantities. This snapshot is from a DMA analysis of glass beads in a polymeric matrix.

Object oriented finite element analysis for material science, originally, could handle both thermal and deformation behaviour analysis of homogenous as well as composite materials. In this work, we have introduced a set of dynamic and static test methods for the analysis of time dependent viscoelastic deformations.

The description of the local fields subjected to the constraints provided by the microstructural details and the boundary conditions results on a set of equations for the nodes of the finite element mesh, given by

$$\mathbf{KU} = \mathbf{F} \quad (4)$$

where $\mathbf{K}$ is the stiffness matrix of the material, $\mathbf{U}$

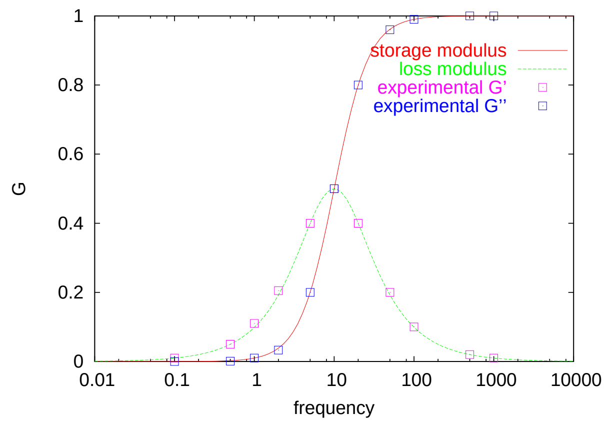

Figure 4: Comparison between the experimental results (square markers) and exact analytical results (lines) for a viscoelastic solid with single relaxation time.

is the vector of nodal positions and **F** is the vector of body forces and forces arising from loading conditions. This forms the simplest description of the finite element process. The details of the finite element implementation can be found elsewhere.

The deformation state of the system or the local fields evolve with time and these spatiotemporal solutions for the integrodifferential equation (eqn. 3) are obtained using using a Newton Ralphson solver. Since, the tangent matrices necessary for the Newton Ralphson solver are independent of state variables, the solver converges rapidly, hence simplifying the computation. The boundary conditions for the system are determined by the kind of experiment chosen and by independent specification. The viscoelastic response for a time varying loading is obtained by succesively solving for the resulatant finite element mesh and updating the stress fields recursively.

The model is validated using several different conditions. To verify the model and the working of the finite element method, a dynamical mechanical analysis simulation for a homogenous solid is studied. The Prony series was restricted to a single term to keep the analysis simple. The results are compared with exact analytical solutions as shown in Figure. 4.

4 Static and dynamic experiments

4.1 Static experiments

The viscoelastic behaviour as defined previously is a dynamic phenomena, that is, the response of the sytem varies with time. The 'static' experiments in this case refers to the condition on loading or deformation applied: constant loading in creep expermeents or the constant applied deformation in the case of stress relaxation experiments. Both of the above experiments can be performed on a microstructure. The object oriented technique with which the OOF package has been built, allows also to superimpose two different loads in different directions.

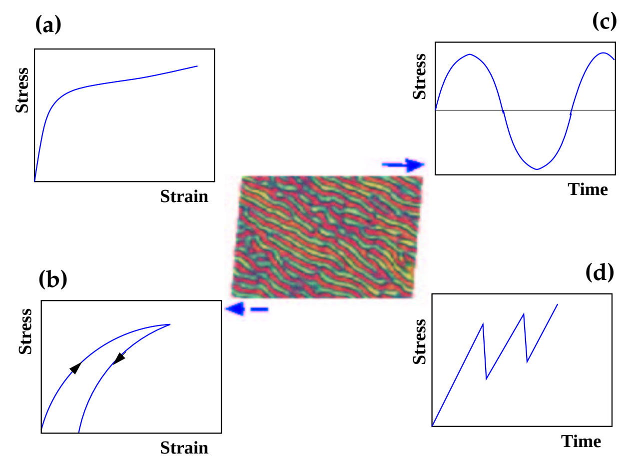

Figure 5: Kinds of virtual experiments that can be performed (a)stress-strain relationships, (b)loading - unloading experiments, (c) dynamical mechanical analysis (d) constant strain rate experiments or combinations of different loading conditions.

4.2 Dynamical Mechanical Analysis

Often applications involving composite materials require response to variable loading at high or low frequencies depending on the application. (For example as a dampner, the composite would be expected to absorb the high frequency component of loading without any sign of degradation.) So it is pertinent to analyze composite behaviour in these kinds of deformation conditions. Modified OOF (this work) incorporates a suite of tests including dynamical mechanical tests wherein the frquency, initial phases of the applied oscillatory deformation field can be specified.

The variations of stress and strain are given by (in complex notation)

$$e(t) = e_0 exp(i\omega t) \tag{5}$$

$$\sigma(t) = \sigma_0 exp(i(\omega t + \delta)) \tag{6}$$

where δ is the phase lag or phase angle.

The overall complex modulus is then given by,

$$G^* = \frac{\sigma_0}{e_0} exp(i\delta) \tag{7}$$

The real part of the complex modulus (G_1) is referred to as the storage modulus as it is relates to the amount elastic energy stored in the system. The imaginary part of the complex modulux (G_2) is referred to loss modulus and is measure of energy dissipation due to viscous flow in each cycle of deformation.

In a homogenous viscoelastic material with a single time scale involved, the complex modulii can be derived easily. Figure 4 shows a comparison between the analytically derived spectra and the results from finite element analysis for a homogenous sample.

In addition to the static experiments and DMA tests, constant strain rate experiments can also be performed on a polymeric composite. These also would provide information regarding the dissipation of energy and limiting behaviour of the composite structures.

Results of the virtual experiments in OOF are normally presented as spatial distribution of the micromechanical quantities. Complete imformation about the local deformations are embedded in these distributions. For the polymeric composites, along with the all the field component distriutions, the dissipated energy distribution, and the stored energy distributions can be quantitatively studied. Several

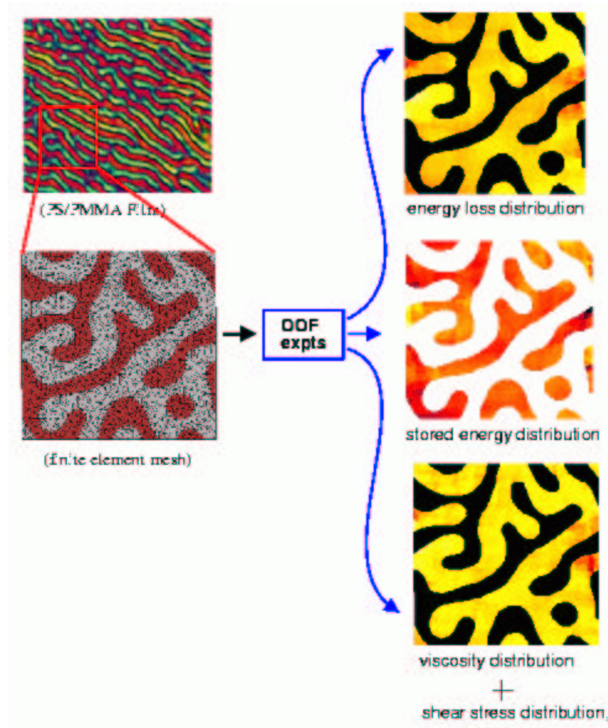

Figure 6: A typical OOF virtual experiment

other quantifiers for local deformation like the local viscosity have also been incorporated.

Several applications could be envisaged which canmake use of this viscoelastic deformation analysis module. For example, effect of microstructural details for inclusions in polymeric matrix : soft inclusions on a hard matrix, as in rubber toughened polymer acrylic material and hard fillers on a soft matrix like a carbon black on polymer composite. In addition, we also investigate the effect of interfacial properties, specifically the relaxation time scales, on these composite materials. As discussed in preceeding sections, the interphase plays a crucial role in determining the deformation properties of the composite system. For example, in core-sheel PMMA/rubber system, good adhesion between the acrylic matrix - a coploymer based upon poly(methylmethacrylate) - and the rubber particle is particularly important in acheiving high performance. The toughening particles are composed of a core of acrylic particles with a outer shell bonded with rubber. The study of the dynamical response of these composite are expected to provide insights into the roles of the components in the toughening behvior. Morphology and microstructure also play important roles in designing the desired mechanical property for a composite. The stress distribution for a deformation process depends on these factors and can ultimately decide the strength the material.

5 Conclusions

The public domain code OOF has been extended to include tools for analysis of viscoelastic materials while retaining all its powerful features. Added features in OOF include means to quantitatively analyze the spatiotemporal response of a composite polymeric material in dynamic as well as in static deformation conditions. These coupled with the existing features of OOF, in particular, the complete analysis of mechanical characteristics of materials provide a comprehensive tool for the studies of timedependent behaviour of variety of materials including polymeric solid composites, polymer nano composites, polymer blends, block copolymers, and so on. The viscoelastic module draws its strength from the underlying OOF architecture to provide a macroscopic evaluation of mechanical properties using microstructural details.

The correlation between the macroscopic mechanical property on the processing conditions can also be analysed thorugh the dependence of microstructural details on the processing conditions. This is also being envisaged as a tool for the analysis of evolution of phase separating of polymer mixtures in thin films and for the study of interfacial interactions. The strength of this module lies in its apparent simplicity and easy to use approach with menu driven functional capabilities. However, a limitation of this module, in its present form, is its use of linear viscoelastic formalism to calculate the mechanical response. Work is underway to eliminate this limitation by utilizing available experimental data to the fullest extent as inputs to individual component material of a composite.

6 References

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