Implementation of a Viscoelastic Material Model to Simulate Relaxation in Glass Transition

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Abstract: Glass relaxation occurs in a range of temperature during transition from equilibrium to super-cooled liquid. Viscoelastic material model can be applied to simulate glass behavior in the glass transition regime. COMSOL Multiphysics software 4.3b can simulate the stress relaxation with the WLF (Williams-Landel-Ferry) shift function, but no structural relaxation. In this study we developed a framework of material constitutive model to implement the full viscoelastic model with both structural and stress relaxation in COMSOL. The fictive temperature, thermal strain, viscoelastic stress can be calculated simultaneously by solving a set of coupled equations. The results are also compared to the ANSYS simulation.

Keywords: glass viscoelastic deformation simulation, stress relaxation, structural relaxation, finite element method (FEM)

1. Introduction

Glass relaxation occurs in a range of temperature during transition from equilibrium to super-cooled liquid. Viscoelastic material model can be applied to simulate glass behavior during the glass transition regime and to predict the glass deformation and stress evolution. Viscoelasticity is the property of materials that exhibit both viscous and elastic characteristics when undergoing deformation. The generalized viscoelasticity theory involves two kinds of phenomena: stress relaxation and structural relaxation. Structural relaxation describes the time-dependent change in volume due to the temperature change as well as the thermal history. Stress relaxation involves both viscous and elastic deformation in the dynamic stress-strain response. The dependence of the viscoelastic properties on the temperature can be accounted for by the thermorheologically simple (TRS) assumption, using the shift function concept. COMSOL Multiphysics software 4.3b can simulate the stress relaxation with the WLF (Williams-Landel-Ferry) shift function, but no structural relaxation. In this study we developed a framework of material constitutive model to implement the full viscoelastic model with both structural and stress relaxation in COMSOL. The structural relaxation is captured by the concept of fictive temperature, first proposed by Tool[1]. The fictive temperature is solved by a set of differential equations. Besides the WLF shift function we can use any kind of shift functions. A shift function widely used in glass simulation is the Tool-Narayanaswamy shift function[2], which considers the influence of fictive temperature.

In this paper we will show the theory of stress relaxation, structural relaxation during glass transition and their implementation in COMSOL Multiphysics 4.3b. The framework of implementing full viscoelastic model with both stress relaxation and structural relaxation in COMSOL Multiphysics 4.3b is demonstrated through a numerical example of glass tube residual stress evolution during a non-uniform cooling process. The simulation results using the framework are also compared to the ANSYS simulation results. Discussion and conclusions are presented in the end.

2. Stress Relaxation

Stress relaxation describes the time-dependent change in dimensions due to applied loadings. In general the stress relaxation can be put in this form

$$\boldsymbol{\sigma}(t) = \int_0^t \mathbf{D}(t-t')\frac{d\boldsymbol{\varepsilon}(t')}{dt'}dt', \quad (1)$$

where $\boldsymbol{\sigma}$ is the stress, $\mathbf{D}$ is the modulus matrix and $\boldsymbol{\varepsilon}$ is the strain. We see that the stress is an integration of the previous history, or history dependent. In other words, the stress has "memory effects". For a shear stress component σ_{ij} we have

$$\sigma_{ij}(t) = \int_0^t 2G(t-t')\frac{d\varepsilon_{ij}}{dt'}dt'. \quad (2)$$

The relaxation shear modulus function, $G(t)$, can be approximated by a Prony series as shown below[4,5]

$$G(t) = G(\infty) + (G(0) - G(\infty)) \sum_{n=1}^{N} \mu_n e^{-\frac{t}{\tau_n}}, \qquad (3)$$

where N is the number of terms, τ_n are the stress relaxation times and μ_n are the weight coefficients, satisfying

$$\sum_{n=1}^{N} \mu_n = 1 \cdot \qquad (4)$$

In a special case when N=1 and assuming $G(\infty)$=0, Eq. (2) reduces to

$$\sigma_{ij}(t) = 2G(0) \int_0^t e^{-\frac{t-t'}{\tau}} \frac{d\varepsilon_{ij}}{dt'} dt', \qquad (5)$$

which is the solution of the following differential equation

$$\frac{d\sigma_{ij}}{dt} + \frac{\sigma_{ij}}{\tau} = 2G(0) \frac{d\varepsilon_{ij}}{dt'} \cdot \qquad (6)$$

Eq. (6) is actually a description of a single element Maxwell model consisting of a spring with modulus G and a dashpot with viscosity η, and $\tau = \eta / G$[3]. For a step strain stress relaxation test where ε_{ij} jumps from 0 to a constant value ε_0 at time t=0 and keeps constant at time t>0, the analytical solution for Eq. (5) is

$$\sigma_{ij}(t) = 2G(0) \varepsilon_0 e^{-\frac{t}{\tau}}. \qquad (7)$$

A quick check of the stress relaxation simulation result using COMSOL Multiphysics 4.3b in comparison with the analytical solution (7) is shown in Figure 1. In this test the step strain is applied at time t=0.001. The COMSOL simulation result agrees very well with the analytical solution.

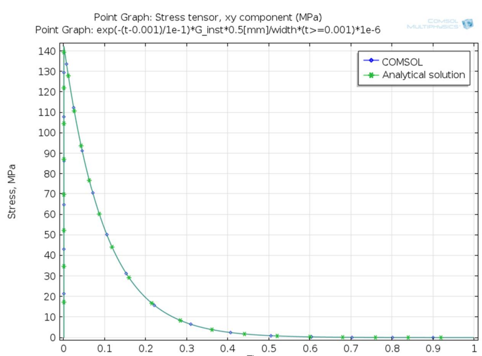

Figure 1. Step-strain stress relaxation result comparison.

2. Structural Relaxation

In reality the stress relaxation time τ is not constant. It has strong dependence on the temperature. A common assumption is that the material is thermorheologically simple (TRS). It means that the change of temperature can be transformed to the change in the time scale, by shifting the relaxation time with a shift function Φ[3]. One of the widely used shift function is the WLF (Williams-Landel-Ferry) shift function

$$\log 10(\Phi) = \frac{-C_1 (T - T_{ref})}{C_2 + (T - T_{ref})}, \qquad (8)$$

where C_1 and C_2 are material constants, T_{ref} is usually the glass transition temperature. WLF shift function is the only shift function implemented in COMSOL Multiphysics 4.3b. Another shift function is

$$\Phi = \exp\left[\frac{H}{R}\left(\frac{1}{T_{ref}} - \frac{1}{T}\right)\right], \qquad (9)$$

where H is the activation energy, R is the universal gas constant, T_{ref} is the reference temperature. The stress relaxation time will be scaled as

$$\tau = \tau_0 / \Phi, \qquad (10)$$

where τ_0 is the stress relaxation time at T=T_{ref}.

However, the shift function Φ itself can be influenced by the structural rearrangement during glass transition. Tool[1] proposed the concept of fictive temperature T_f to characterize the structural rearrangement, described as

$$\frac{dT_f}{dt} = \frac{T - T_f}{\lambda}, \qquad (11)$$

where λ is the structural relaxation time, scaled as

$$\lambda = \lambda_0 / \Phi, \qquad (12)$$

similar to the stress relaxation. Here λ_0 is the structural relaxation time at T=T_{ref}.

Narayanaswamy[2] then proposed a shift function that considers the influence of fictive temperature, as shown in Eq. (13).

$$\Phi = \exp\left[\frac{H}{R}\left(\frac{1}{T_{ref}} - \frac{x}{T} - \frac{1-x}{T_f}\right)\right], \qquad (13)$$

where x is another material constant. Eq. (13) is the Tool-Narayaswamy shift function, widely used in glass viscoelastic simulations. It is observed that if x=1 or T=T_f, Eq. (13) simplifies to Eq. (9).

Taking into account of structural relaxation, the thermal strain is calculated as the following[5]

$$\frac{d\varepsilon^{th}}{dt} = \alpha_g \frac{dT}{dt} + (\alpha_l - \alpha_g) \frac{dT_f}{dt}, \qquad (14)$$

where α_g and α_l are CTEs (coefficients of thermal expansion) at glass state and liquid state respectively. The thermal stress changes will affect the mechanical stress in the structure. Stress relaxation and structural relaxation are therefore closely coupled through shift function, fictive temperature and thermal strain.

In a general application, there will also be multi-term Prony series for the structural relaxation, Eqs. (11) and (12), to resolve different modes of structural relaxation. They need to be rewritten as

$$\frac{dT_{f,n}}{dt} = \frac{T - T_{f,n}}{\lambda_n}, \tag{15}$$

where

$$\lambda_n = \lambda_{0,n} / \Phi, \tag{16}$$

are the structural relaxation times. The fictive temperature is calculated as

$$T_f = \sum_{n=1}^{M} w_n T_{f,n}. \tag{17}$$

Here M is the number of terms for the structural relaxation and w_n are the weight coefficients, satisfying

$$\sum_{n=1}^{M} w_n = 1. \tag{18}$$

3. Numerical Example

Since stress relaxation and structural relaxation are closely coupled, both of them need to be considered in simulating the glass viscoelastic deformation during the glass transition stage.

Only stress relaxation is implemented in COMSOL Multiphysics 4.3b with a generalized Maxwell model[4]. In order to simulate the structural relaxation, two domain ODEs are added. One is for the fictive temperature components, Eq. (15) and the other is for thermal strain, Eq. (14).

This simulation example solves the stress evolution when a glass tube with 3mm thick wall cools from above glass transition temperature to below glass transition temperature, shown in Figure 2. The outer surface cools faster than the inner surface, inducing a temperature gradient through the thickness. During the non-uniform cooling process, the temperature gradient will generate thermal expansion mismatch and thus thermal stress which will relax with time. But the stress relaxation rate also varies with the temperature changing. Residual stress will be left in the glass tube when it eventually reaches the room temperature.

The temperature profiles at the inner surface and outer surface are shown in Figure 2. The temperature distribution through the thickness can be determined as[6]

$$T(r) = T_i + (T_o - T_i)\frac{\ln\left(r/r_i\right)}{\ln\left(r_o/r_i\right)}, \tag{19}$$

where T_i, T_o are the temperature at the inner surface and outer surface respectively, r_i, r_o are the radiuses of the inner surface and outer surface respectively and r is the radial coordinate, shown in Figure 4.

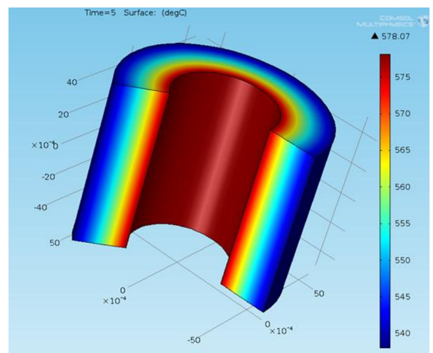

Figure 2. Geometry of a glass tube cooling with different rates at inner and outer surfaces.

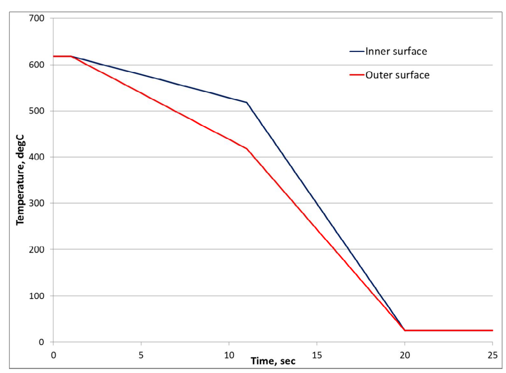

Figure 3. Temperature history at inner and outer surfaces.

A 2D axisymmetric solid mechanics module is set up for simulating the problem, shown in Figure 4. A symmetry boundary condition is applied at the bottom edge. The tube is free of mechanical loadings and is only subject to the thermal loadings defined above.

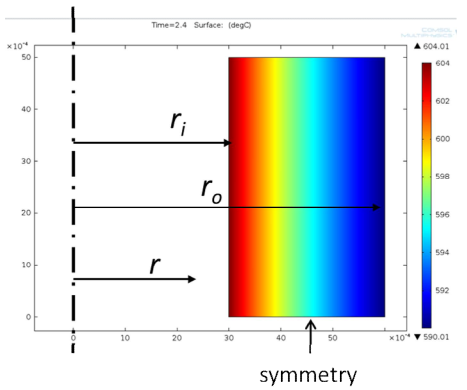

Figure 4. 2D axisymmetric solid mechanics model.

The glass viscoelastic material property is taken from the ANSYS verification manual VM200 G-11 glass material property[5]. Young's modulus E=72.5GPa, Poisson ratio ν=0.3. 3 terms of Prony series for stress relaxation are used: relaxation time $\tau_{0,n}$=0.0689, 0.0065, 0.0001 s and weights μ_n=0.422, 0.423, 0.155. 6 terms for structural relaxation are used: relaxation times $\lambda_{0,n}$=3.0, 0.671, 0.247, 0.091, 0.033, 0.008 s and weights w_n=0.108, 0.443, 0.166, 0.161, 0.046, 0.076. Tool-Narayaswamy shift function, Eq. (13), is used, with parameters H/R=6.45e4 K, x=0.53, T_{ref}=618 degC. CTEs at liquid state and glass state are 34.3e-6, 6.47e-6 respectively.

The stress relaxation parameters are added to the Generalized Maxwell Model branches under the linear viscoelastic material component of the solid mechanics module in COMSOL Multiphysics 4.3b. The thermal effects are enabled for thermal strain influence. The thermal strain to be used for stress calculation need to be replaced by the thermal strain variable calculated from the domain ODE implementing Eq. (14) using the Equation View, as shown in Figure 5.

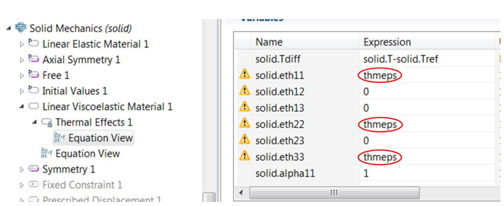

Figure 5. Replacement of thermal strain with the variable (thmeps is the name) from the domain ODE, implementing Eq. (14).

The shift function can also be replaced by any user defined function as shown in Figure 6. In this example, we used the Tool-Narayaswamy shift function, i.e., Eq. (13), defined in Figure 7.

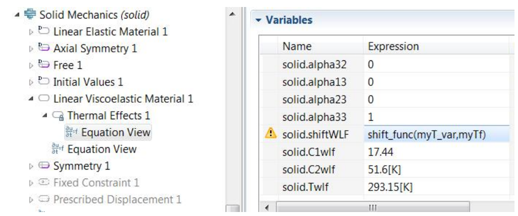

Figure 6. Replacement of shift function with the user defined function.

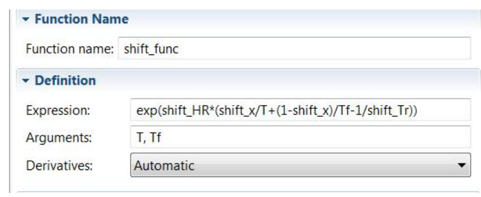

Figure 7. Function definition of Tool-Narayanaswamy shift function.

The whole coupled system is then solved using the time-dependent solver to obtain the solutions, including fictive temperature, thermal strain, structure displacement and stress.

The solutions of fictive temperature and thermal strain at the center of the inner surface are reported in Figures 8 and 9, in comparison with ANSYS solution using the same conditions. It is seen that these two results agree very well, indicating that we captured the structural relaxation behavior well with the fictive temperature and thermal strain solutions.

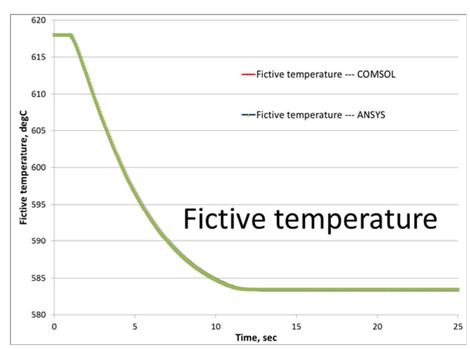

Figure 8. Comparison of fictive temperature at center inner surface between COMSOL and ANSYS solutions.

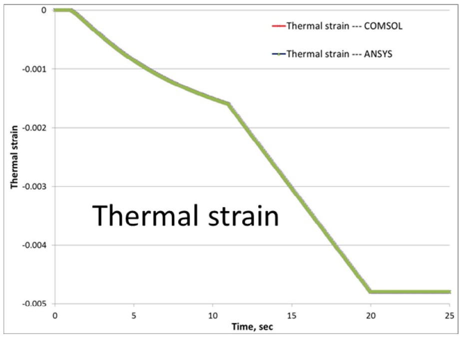

Figure 9. Comparison of thermal strain at center inner surface between COMSOL and ANSYS solutions.

The fictive temperatures at the inner surface and the outer surface can be found in Figure 10. Since the outer surface is cooled at a faster rate, the fictive temperature at the outer surface is frozen at a higher value than that in the inner surface.

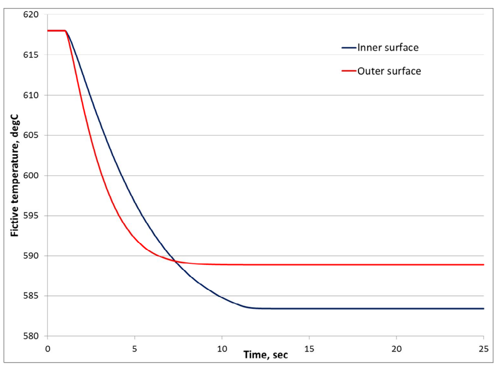

Figure 10. Comparison of fictive temperature at inner surface and outer surface.

The stress evolution at the center inner surface is plotted in Figure 11. Good agreement between COMSOL simulation results and ANSYS simulation results is again observed. The hoop stress and axial stress show the similar behavior of changing, although the hoop stress is greater than the axial stress in magnitude. Looking at the temperature profile in Figure 3, there are two stages: before time=11sec, the inner-outer temperature gradient is increasing; after time=11sec, the inner-outer temperature gradient is decreasing. In these two stages, the stresses behave differently. As shown in Figure 12, the inner surface shows compressive stress and the outer surface shows tensile stress in the first stage. And it reversed in the second stage. The inner surface shows tensile stress and the outer surface shows compressive stress in the end. It is showing the effect of tempering: strengthening the outer surface by putting in compressive stress. It may increase the risk of breaking at the inner surface, since tensile stress is generated there.

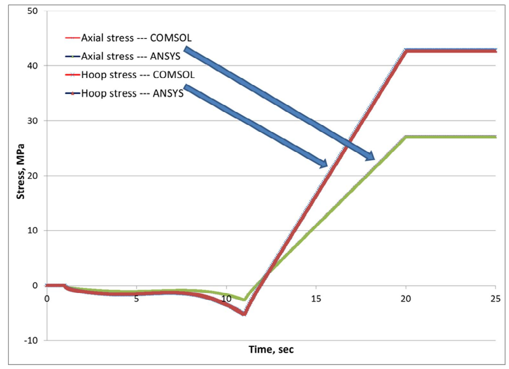

Figure 11. Axial and hoop stress at the center inner surface as a function of time.

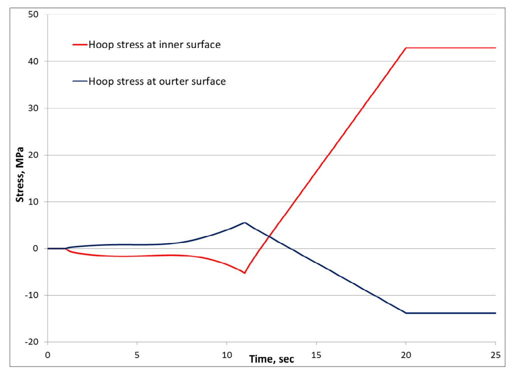

Figure 12. Comparison of hoop stress at the inner surface and outer surface.

4. Discussions and Conclusions

In this paper we demonstrate a framework of material constitutive model to implement the full viscoelastic model with both structural and stress relaxation in COMSOL Multiphysics software 4.3b. For the structural relaxation simulation, the fictive temperature and thermal strain solutions are obtained by solving the added domain ODEs. Any type of shift function can be defined using the method presented in this work. Particularly the Tool-Narayanaswamy shift function is implemented in this work, to include the fictive temperature dependence in the shift function. Viscoelastic stress is solved efficiently with the

COMSOL model and the results compare well with the ANSYS results.

This framework of viscoelastic material model simulating both stress relaxation and structural can then be extended to simulate more sophisticated glass forming process. As shown in the numerical example presented, the viscoelastic simulation of glass forming gives the information of how fictive temperature, thermal strain, and stress evolve with the thermal history. This information can be a good guidance for understanding and improving the glass manufacturing process.

5. References

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