Elastic-viscoplasticity modeling of the thermo-mechanical behavior of chalcogenide glass for aspheric lens molding

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ABSTRACT: Chalcogenide Glass (ChG), as an alternative material in place of single-crystal germanium, is increasingly used in thermal imaging, night vision and infrared guidance systems, etc., owing to their excellent formability through Precision Glass Molding (PGM). The deformation mechanisms of these glasses at the molding temperature involve elasticity, plasticity and viscous flow, which call for a new theoretical model to assist the design of PGM process. This paper investigates the thermo-mechanical properties of $Ge_{22}Se_{58}As_{20}$ at the temperature above its softening point and establishes a new elastic-viscoplasticity model to describe its thermo-mechanical behaviors. After determining the model parameters through cylindrical compression tests, the new constitutive model is implemented in finite element method (FEM) of PGM to form an aspheric ChG lens. And the agreement of displacement-time curves between experimental and simulation results exhibit the validity of the proposed elastic-viscoplastic constitutive model.

KEYWORDS: Chalcogenide Glass (ChG); Precision Glass Molding (PGM); Thermo-mechanical; Elastic-viscoplasticity; Finite element method (FEM)

1. Introduction

Infrared materials are gaining growing attention for their wide applications in martial and civil fields,[1] which include thermal imaging, night vision and infrared guidance systems, etc. In the past decades, though single-crystal germanium is rare and expensive, it has been used exclusively in infrared optical system, which is machined by single point diamond cutting.[2] Chalcogenide Glass (ChG), which is made of mainly chalcogens (S, Se and Te) and some other elements, shows a lot wider transmission wave band from near to far infrared wavelength. In comparison, oxide glasses are limited to near and intermediate infrared wave band because of the strong infrared absorption of metal-oxygen bond vibrations. Compared with single-crystal germanium, ChG is much more economic and has excellent properties of athermalization and achromatism.[3] Due to the amorphous nature, the viscosity of ChG decreases to an optimum range for molding during heating.[4] Therefore for ChG lens, Precision Glass Molding (PGM) can be employed and is much more efficient and less costly than single point diamond cutting,[5] the only choice for single-crystal germanium.[2] ChG is now deemed as an alternative for single-crystal germanium in various infrared systems.[6]

Though PGM for oxide glasses has been relatively mature, it is still in its infancy and needs particular treatment for ChG. Directly using the constitutive model of oxide glass to describe the thermo-mechanical behavior of ChG could be problematic since the molding temperature for oxide glass is between the yielding point and the softening point, at which the viscosity is about $10^{10.76}$ dPa·s[7] and $10^{7.65}$ dPa·s[8], respectively. However the molding temperature is above the softening point for ChG since the viscosity is still too high below softening point and can cause breakage in molding.[9] This leads to the most distinct thermo-mechanical behavior between these two materials and the necessity of developing a new constitutive model for ChG. Most previous works have used the viscoelastic constitutive model in studying glass molding process. Jain and Yi have used the generalized Maxwell model to represent the stress relaxation behavior during the forming stage of oxide glass molding, and applied this numerical model to Finite Element Method (FEM) to predict molding results.[10] Ruan and Zhang have developed a Metropolis stochastic process for the glass transition and thermo-forming simulations of selenium, and investigated the microscopic origin of viscosity by simplifying an amorphous system to a mixture of many independent atomic subsystems to be applied on the amorphous selenium.[11,12] Liu *et al.* have studied the imprint technology of ChG with the generalized Maxwell model to represent the stress relaxation behavior.[13] In our earlier studies, Maxwell model, Kelvin model and Burgers model have been introduced to explain the viscoelasticity of creep and stress relaxation behavior of oxide glass,[14,15] which has already been used in aspheric glass lens[16,17] and glass microstructure array molding.[18,19] In this work, our studies are extended to ChG molding. ChG presents non-flowing state above softening temperature and incomplete recovery deformation in pressing stage, which indicates plastic deformation. So far, there has been no elastic-viscoplastic constitutive model used in either oxide glass or ChG. But such model has been applied in describing thermo-mechanical behaviors of asphalt,[20,21] amorphous polymers,[22] and metallic glasses.[23-25]

In this study, an elastic-viscoplastic constitutive model of ChG is established. We use the high-temperature cylindrical compression test to determine model parameters and then implement the constitutive model in FEM simulation of PGM for molding aspheric ChG lenses. The numerical simulation allows us to accurately predict the optimal molding conditions in virtue of adjusting molding parameters.

2. Methodology

2.1. Cylindrical compression test of ChG

A PGM process has four stages: heating, pressing, annealing and cooling,[26] as schematically shown in Fig. 1 This paper focuses on the pressing stage to investigate the elastic-viscoplastic deformation at high temperature.

The cylindrical compression tests are carried out by using an ultraprecision glass molding machine PFLF7 (SYS Corp., Japan). As shown in Fig. 2, the cylindrical glass preforms are compressed between a pair of tungsten carbide molds with flat surface. A complete set of mold contains a lower mold, an upper mold, an inner sleeve to guide the motion of mold and an outer sleeve to locate the lower mold. The molds and a ChG preform are assembled before pressing. Nitrogen gas is purged into the molding chamber to prevent the molds and ChG from oxidation at high temperature. Three bundles of resistance rods are inserted in each of die holders to heat the complete set of mold as well as the glass to the molding temperature, which is several tens of centigrade degrees above its softening temperature. The temperature of each mold holder is monitored and controlled with a accuracy of ±1°C. The lower mold remains stationary, while the upper mold is moved by a pneumatic system, of which the pressure of the pneumatic actuator is regulated by an air compresser with an accuracy of ±0.01 MPa. The pressing force can be calculated knowing that the piston diameter is 76 mm. During experiments, the position of the upper mold is recorded by a position sensor with a measurement accuracy of ±1 μm. The displacement range of the upper mold is adjusted by the height difference between the inner and outer sleeves.

2.2. Theory of elastic-viscoplasticity

At the molding temperature, ChG exhibits elastic-viscoplastic behavior which is a combination of viscoelastic and viscoplastic mechanisms. As the ChG is an isotropic material, the elastic-viscoplastic behavior can be simplified as a combination of spring, dashpot and slider, representing elasticity, viscosity and plasticity, respectively. The constitutive relations of these three elements are respectively:

$$\sigma_e = E\varepsilon_e \tag{1}$$

$$\sigma_v = \eta\dot{\varepsilon}_v \tag{2}$$

$$\sigma_p = \sigma_f \operatorname{sign}\left(\dot{\varepsilon}_p\right) \tag{3}$$

where σ_e, σ_v, σ_p are the elastic stress, viscous stress and plastic stess, respectively, σ_f is the yield stress of the friction element, ε_e, ε_v, ε_p are corresponding strains, E, η, are elastic modulus and viscosity,[27] and $\operatorname{sign}\left(\dot{\varepsilon}_p\right)$ is sign function, being 1, 0, and -1 when the plastic strain rate $\dot{\varepsilon}_p$ is positive, zero and negative.

For different materials, these three elements can be connected in series or parallel. When connected in series, the strain rate of the whole model equals to the sum of all the basic components' strain rate, and the stress of every basic components is the same as the whole model. By contrast, when connected in parallel, the stress of the entire model is the sum of all the elements' stress, and the strain rate of every element is the same as the whole model.[28] To indicate the elastic-viscoplasticity, all of the three bodies must be used, such as Bingham model,[29] which is shown in Fig. 3(a). However, there is neither viscoelastic deformation nor viscous flow in the model before the stress of slider exceeds the yield point, which is not the case for an actual molding process. Kelvin model, which consists of a spring element and a dashpot element in parallel, usually describes viscoelastic motion of oxide glass,[15] as shown in Fig. 3(b). In order to describe all deformation mechanisms of ChG, the above two models are combined as a Schofield-Scott Blair model,[30] as shown in Fig. 3(c). The instantaneous elastic and plastic deformation, delayed elastic and plastic deformation and viscous flow coexist in this model. The constitutive equation based on the Schofield-Scott Blair model is:

$$\begin{cases} \dfrac{\eta_1}{E_2}\dot{\varepsilon} + \varepsilon = \dfrac{\eta_1}{E_1 E_2}\dot{\sigma} + \dfrac{E_1 + E_2}{E_1 E_2}\sigma, \quad \sigma < \sigma_f \\ \dfrac{\eta_1}{E_2}\ddot{\varepsilon} + \dot{\varepsilon} = \dfrac{\eta_1}{E_1 E_2}\ddot{\sigma} + \dfrac{1}{E_2}\left(1 + \dfrac{E_2}{E_1} + \dfrac{\eta_1}{\eta_2}\right)\dot{\sigma} + \dfrac{1}{\eta_2}\sigma - \dfrac{\sigma_f}{\eta_2}, \quad \sigma \geq \sigma_f \end{cases} \tag{4}$$

where E_1, E_2, are elastic moduli and η_1, η_2 represent the viscosities of the elastic-viscoplastic material. As the ChG preform is compressed at an uniform temperature, the pressing stage can be treated as isothermal process without heat transfer.[14]

3. Experimental results and discussion

3.1. Experimental results of cylindrical compression test

In this work, ChG $Ge_{22}Se_{58}As_{20}$ specimen of height 2.8 mm and diameter 7.8 mm is subjected to compression. Its thermal and mechanical properties are listed in Table 1. Fig. 4 shows the displacement *w* of the upper mold under different pressing forces where the *w* is defined as $w(t) = y(t) - y(0)$, where $y(t)$ is the position of the upper mold. The cylindrical compression test is conducted at the temperatures of 382°C and 392°C. And the maximum displacement of the upper mold is set to 1.4 mm. The pressing time increases when the temperature is lower, owing to the increase of elastic modulus and viscosity at the lower temperature.[4,31]

3.2. Elastic-viscoplastic deformation

The true strain ε_t, also called logarithmic strain or Hencky strain,[32] considering an incremental strain, can be derived by Eq. (5):

$$\varepsilon(t) = \int_{z_0}^{z_t} \frac{dz}{z} = \ln(1 - \frac{w(t)}{z_0}) \tag{5}$$

where z_0, z_t are the initial and instantaneous heights of the cylindrical specimen. Both stress and strain should be negative in compression but are sign-revered in the following figures for convenience. The changes of strain with time during deformation under different pressing forces are shown in Fig. 5. The increase of strain rate with time, as shown in Fig. 5, associated with reduction of stress owing to the increase of contact area between mold and glass, is a clear indication of delayed elastoplastic deformation, which becomes more significant at the lower molding temperature or under the smaller pressing force.

The true stress $\sigma(t)$ can be derived by Eq. (6):

$$\sigma(t) = \frac{F}{\pi\rho_0^2} e^{\varepsilon(t)} \tag{6}$$

where F is the molding force, and ρ_0 is the radius of glass preform. The changes of stress with time under different pressing forces are shown in Fig. 6. It is noted that the stress decreases more rapidly under higher pressing force.

Consider the small strain scenario, the first order Taylor expansion of Eq. (6) leads to:

$$\varepsilon(t) = C\sigma(t) - 1, \tag{7}$$

where $C = \pi\rho_0^2 / F$ is a constant. Substituting Eq. (7) into Eq. (4) and considering the cases of $\sigma < \sigma_f$ and $\sigma \geq \sigma_f$ result in:

$$\sigma(\mathrm{t}) = \{[(E_1 + E_2 - CE_1E_2)\sigma_0 + E_1E_2 - CE_1\eta_1]e^{-\frac{(E_1+E_2-CE_1E_2)t}{\eta_1}} - E_1E_2 + CE_1\eta_1\} / (E_1 + E_2 - CE_1E_2), \quad \sigma < \sigma_f \tag{8a}$$

$$\sigma(\mathrm{t}) = \sigma_f + C\eta_2 + \{[(E_1\eta_1 + E_1\eta_2 + E_2\eta_2 + C_1)(\sigma_0 - \sigma_f) - CC_1\eta_2 - CE_1\eta_1\eta_2 + 2\eta_1\eta_2\dot{\sigma}_0 - C_3 - C_2]e^{-\frac{(E_1\eta_1+E_1\eta_2+E_2\eta_2-C_1)t}{2\eta_1\eta_2}}\} / 2C_1 - \{[(E_1\eta_1 + E_1\eta_2 + E_2\eta_2 - C_1)(\sigma_0 - \sigma_f) + CC_1\eta_2 - CE_1\eta_1\eta_2 + 2\eta_1\eta_2\dot{\sigma}_0 - C_3 - C_2]e^{-\frac{(E_1\eta_1+E_1\eta_2+E_2\eta_2+C_1)t}{2\eta_1\eta_2}}\} / 2C_1, \quad \sigma \geq \sigma_f \tag{8b}$$

Where

$C_1 = \sqrt{E_1^2\eta_1^2 + 2E_1^2\eta_1\eta_2 + E_1^2\eta_2^2 - 2E_1E_2\eta_1\eta_2 + 2E_1E_2\eta_2^2 + E_2^2\eta_2^2}$,

$C_2 = CE_2\eta_2^2$, $C_3 = CE_1\eta_2^2$,

and σ_0 is the initial stress when the upper mold just contacts the glass surface and $\dot{\sigma}_0$ is the stress rate at t = 0. From Fig. 6, the values of σ_0 and $\dot{\sigma}_0$ can be obtained. σ_0 is the same at different molding temperature and $\dot{\sigma}_0$ reduces with the increase of molding temperature and pressing force, as shown in Table 2. With them, Eq. (8) can be utilized to determine model parameters from these compression tests.

The least square nonlinear curve fitting method is used to fit the experimental data of $\sigma(t)$ and both Eqs. (8a) and (8b), pertaining to the cases of $\sigma < \sigma_f$ and $\sigma \geq \sigma_f$ respectively, are used and compared. As shown in Fig. 7, the curves rendered by Eq. (8a) are obviously inconsistent with the experimental data. Therefore it can be concluded that the stress in compression test is equal to or larger than the yield stress. Then the initial portion of stresses $\sigma(t)$ is fitted based on the situation of $\sigma \geq \sigma_f$ and Eq. (8b), which leads to the model parameters as listed in Table 3. It is noted that all parameters, including yield stress, moduli and viscosities reduce with the increase of temperature.

For large strain, Eq. (6) is substituted into Eq. (4), leading to:

$$\frac{\eta_1}{E_1E_2}\ddot{\sigma} + \frac{1}{E_2}\left(1 + \frac{E_2}{E_1} + \frac{\eta_1}{\eta_2}\right)\dot{\sigma} + \frac{1}{\eta_2}\sigma - \frac{1}{\sigma} + \frac{\eta_1}{E_2\sigma^2} - \frac{\sigma_f}{\eta_2} = 0, \quad \sigma \geq \sigma_f \tag{9}$$

which can be solved using the Fourth-order-Runge-Kutta method with the obtained parameters (Table 2 and Table 3). Introducing a new variable s for converting Eq. (9) to two first-order equations:

$$\begin{cases} \dot{\sigma} = s, & \sigma(0) = \sigma_0 \\ \dot{s} = -\frac{E_1}{\eta_1}\left(1 + \frac{E_2}{E_1} + \frac{\eta_1}{\eta_2}\right)s + \frac{E_1 E_2}{\eta_1 \eta_2}\sigma - \frac{E_1 E_2}{\eta_1 \sigma} + \frac{E_1}{\sigma^2} - \frac{E_1 E_2 \sigma_f}{\eta_1 \eta_2}, & s(0) = \dot{\sigma}_0 \end{cases} \tag{10}$$

And their finite difference form with the discretized time step h is:

$$\begin{cases} \sigma_{n+1} = \sigma_n + (h/6)(K_{11} + 2K_{12} + 2K_{13} + K_{14}) \\ s_{n+1} = s_n + (h/6)(K_{21} + 2K_{22} + 2K_{23} + K_{24}) \end{cases} \tag{11}$$

$$\begin{cases} K_{11} = s_n \\ K_{21} = -\frac{E_1}{\eta_1}\left(1 + \frac{E_2}{E_1} + \frac{\eta_1}{\eta_2}\right)s_n + \frac{E_1 E_2}{\eta_1 \eta_2}\sigma_n - \frac{E_1 E_2}{\eta_1 \sigma_n} + \frac{E_1}{\sigma_n{}^2} - \frac{E_1 E_2 \sigma_f}{\eta_1 \eta_2} \\ K_{12} = s_n + hK_{21}/2 \\ K_{22} = -\frac{E_1}{\eta_1}\left(1 + \frac{E_2}{E_1} + \frac{\eta_1}{\eta_2}\right)\left(s_n + \frac{h}{2}K_{21}\right) + \frac{E_1 E_2}{\eta_1 \eta_2}\left(\sigma_n + \frac{h}{2}K_{11}\right) - \frac{E_1 E_2}{\eta_1(\sigma_n + hK_{11}/2)} \\ + \frac{E_1}{(\sigma_n + hK_{11}/2)^2} - \frac{E_1 E_2 \sigma_f}{\eta_1 \eta_2} \\ K_{13} = s_n + hK_{22}/2 \\ K_{23} = -\frac{E_1}{\eta_1}\left(1 + \frac{E_2}{E_1} + \frac{\eta_1}{\eta_2}\right)\left(s_n + \frac{h}{2}K_{22}\right) + \frac{E_1 E_2}{\eta_1 \eta_2}\left(\sigma_n + \frac{h}{2}K_{12}\right) - \frac{E_1 E_2}{\eta_1(\sigma_n + hK_{12}/2)} \\ + \frac{E_1}{(\sigma_n + hK_{12}/2)^2} - \frac{E_1 E_2 \sigma_f}{\eta_1 \eta_2} \\ K_{14} = s_n + hK_{23} \\ K_{24} = -\frac{E_1}{\eta_1}\left(1 + \frac{E_2}{E_1} + \frac{\eta_1}{\eta_2}\right)(s_n + hK_{23}) + \frac{E_1 E_2}{\eta_1 \eta_2}(\sigma_n + hK_{13}) - \frac{E_1 E_2}{\eta_1(\sigma_n + hK_{13})} \\ + \frac{E_1}{(\sigma_n + hK_{13})^2} - \frac{E_1 E_2 \sigma_f}{\eta_1 \eta_2} \end{cases} \tag{12}$$

The comparisons between experimental and theoretical results are shown in Fig. 8. It's noted that when the pressing force is 156 N, the agreement between experimental and theoretical results is relatively poor since the applied stress is closer to the yield stress σ_f. This may imply that the yield stress is not a constant or the yield stress determined from the scenario of small strain is not accurate. In the cases that the applied stress is much larger than the yield stress, as shown Fig. 8(b) and (c), the agreement between experimental and theoretical results are excellent. These results indicate that the Schofield-Scott Blair model is able to fit the stress curves under different pressing forces.

4. Aspheric lens molding experiments and simulation verification

The aspheric lens molding experiments are carried out at 382°C and 392°C under the pressing forces of 156 N, 468 N and 779 N, respectively. The aspheric precision molds, a ChG preform and a molded aspheric glass lens which are used in the molding experiment are shown in Fig. 9 and Fig. 10. Since the pressing force has little effect on the shape accuracy, so only one (156 N) of these three pressing forces is selected for descriptions. The formed lenses have an excellent surface finish of Ra = 8 nm, and the surface profiles of formed lenses are consistent with the designed value. To demonstrate the conformity and replication fidelity of the formed lenses, the profile error between molds and formed lenses are compared and shown in Fig. 11. It shows that both the upper and lower surface finish of formed lenses are better than the molds because the formed lens does not fill in between surface asperities of molds. However, the higher molding temperature leads to the better conformation between mold and glass surface and the larger roughness of the molded lens, indicating that the surface finish of the molded lens can be improved at the lower molding temperature. On the other hand, the radius of curvature of the formed lens is less than the molds due to the shrinkage of glass in the annealing and cooling stages, and there is a shrinkage inducing depression in the lower surface center of formed lens, where the cooling rate is larger than other places. And this shrinkage becomes greater with the increase of molding temperature.

The validity and accuracy of the proposed Schofield-Scott Blair model is checked with aspheric lens molding experiments and simulations. The FEM simulations are performed using a commercial nonlinear FEM software ABAQUS/Explicit, which is suitable for solving thermal-mechanical coupled problems. It can be used to visualize the glass molding process and monitor some difficult-to-measure variables including strain/stress distribution and internal temperature variation. To reduce calculation time, the simulation model is simplified to be a two-dimensional (2D) axisymmetric model. In the model, three blocks assemble from top to bottom representing upper mold, glass and lower mold respectively, as shown in Fig. 12. The Schofield-Scott Blair model has been established in Sections 2 and 3 is used to describe the thermo-mechanical property of the ChG material and the molds are described using the elastic constants of tungsten carbide. In order to take thermal effect into consideration, the steps are set to dynamic, temperature-displacement field coupling and explicit. The contact between the upper mold and glass preform is set to surface-to-surface contact with friction coefficient 0.1. The lower mold is fixed and a constant pressing force is applied on the upper mold to press the glass with initial field temperature of the whole model is 25°C, which was the same condition with the aspheric lens molding experiment. To reduce computational cost, elements' size of molds (0.1 mm) is twice as large as elements' size of glass (0.05 mm). The 4-node thermally coupled quadrilateral and bilinear element, with reduced integration point and hourglass control, is selected.

Shown in Fig. 13 are the snapshots of distribution of equivalent stress (von Mises stress) in course of molding. The comparison of experimental and simulated displacement-time curves is shown in Fig. 14. The discrepancies between simulation and experimental results at the beginning of molding process as shown in Fig. 14 are due to the initial contact condition. In the simulation, the contact area gradually increases from the point contact and the distribution of stress is initially localized near the contact region, as shown in Fig. 13(a). At this early stage, the stresses are large and stress relaxation predicted based on our theoretical is slower than the experimental result, as shown in Fig. 8 (cf. the cases of 468 and 779 N). This is the reason that the deformation at early stage is slower than the experimental result. And this discrepancy increases with the pressing force. On the other hand, at the later stage of aspheric lens molding, the contact area becomes large and the stress becomes small, as shown in Fig. 13(b). It is noted from Fig. 8 (cf. the case of 156 N) the theoretical stress relaxation could become faster than the experimental result when stress becomes small. This explains why the simulated deformation in aspheric lens molding can catch up (or even overrun) the experimental deformation rate.

5. Conclusions

To describe the thermo-mechanical behavior of ChG in PGM, the Elastic-viscoplastic has been established and characterized using cylindrical tests. The constitutive model is then implemented in FEM simualtion of PGM process, which is verified by molding experiments. The main findings of the present work are summarized as follows:

(1) The Schofield-Scott Blair model is suitable for describing the variations of stress and strain against time. The elastic, plastic and viscous parameters of the model can be obtained by cylindrical compression tests.

(2) The surface finish and profile accuracy of the formed aspheric lens by PGM are of very high quality. The small deviation of surface profile from the designed one is due to the shrinkage at annealing and cooling stages, which is greater at the higher molding temperature.

(3) The agreement of displacement-time curves between experiment and simulation of aspheric lens molding verifies the elastic-viscoplastic constitutive model. Using the FEM simulation, material flow and stress distribution in ChG during molding process can be visualized, which will help optimizing the PGM process parameters for an optimum forming accuracy.

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