

ORIGINAL ARTICLE

Modeling of thermo-viscoelastic material behavior of glass over a wide temperature range in glass compression molding

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Abstract

In glass compression molding, most current modeling approaches of temperature-dependent viscoelastic behavior of glass materials are restricted to thermo-rheologically simple assumption. This research conducts a detailed study and demonstrates that this assumption, however, is not adequate for glass molding simulations over a wide range of molding temperatures. In this paper, we introduce a new method that eliminates the prerequisite of relaxation functions and shift factors for modeling of the thermo-viscoelastic material behavior. More specifically, the temperature effect is directly incorporated into each parameter of the mechanical model. The mechanical model parameters are derived from creep displacements using uniaxial compression experiments. Validations of the proposed method are conducted for three different glass categories, including borosilicate, aluminosilicate, and chalcogenide glasses. Excellent agreement between the creep experiments and simulation results is found in all glasses over long pressing time up to 900 seconds and a large temperature range that corresponds to the glass viscosity of $\log(\eta) = 9.5 - 6.8$ Pas. The method eventually promises an enhancement of the glass molding simulation.

KEYWORDS

FEM simulation, glass molding, thermo-rheologically simple, thermo-viscoelastic modeling

1 | INTRODUCTION

Glass, recently redefined as a nonequilibrium, noncrystalline condensed state of matter,¹ contains a glass transition, the region at which it strongly exhibits viscoelastic behaviors. Owing to enormously precious properties including high hardness, thermal resistance, refractive index, light permeability, and stability to extreme environment changes, such as humidity and temperature, glass has been replacing polymers in many optical applications. Today, optical components are challenging for glass optic manufacturers toward

increasing geometrical complexity, higher precision, and production volume. Accordingly, conventional fabrication techniques via grinding and polishing become no longer feasible. Instead, replicative manufacturing or glass molding promises a distinct satisfaction of such ever-increasing demands. This technology enables the fabrication of precision glass optics, where the desirable shape is readily achieved after a very short molding cycle and requires no subsequent machining steps, such as grinding and polishing.

In the glass compression molding, a hot glass blank is pressed in between a precision mold pair and then is cooled

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down to a solid state. Depending on the temperature difference between the glass and mold during pressing, the glass molding process is distinguished by isothermal glass molding or precision glass molding (PGM) and nonisothermal glass molding (NGM).² Regardless, the glass undergoes the heating stage to a liquid-like state at nearly softening temperature (S_p) in PGM or even above S_p in NGM and the following pressing and cooling stages to a solid-like state, typically below transition temperature (T_g), where its desirable shape is mostly achieved.

In this temperature regime, viscoelastic properties dominate the deformation of the glass under thermo-mechanical loads. Previous studies demonstrate essential contributions of viscoelastic properties to the final shape of molded lenses.^{3–5} Numerical predictions of the final lens shape requires a coupling thermo-mechanical constitutive model to comprehend thermo-viscoelastic responses of glass deformation over the entire molding process. For mechanical modeling of the viscoelastic responses, a large number of rheological models have been investigated in the field of glass molding simulation over years. In the early days, behavior of glass at the molding temperature was commonly considered the pure viscous flow, where flow stress is proportional to the strain rate.^{6–8} Other studies use either simple Maxwell or Kelvin model to approximate the stress relaxation and creep responses, respectively.^{9,10} Those simple models are chosen to qualitatively study the form deviation of molded glass shapes and stress characteristics. A later study by Zhou et al reports that neither Maxwell nor Kelvin model satisfactorily describes the experiment data in the temperature regime of glass molding, but Burgers model—a combination of both Maxwell and Kelvin model—does.¹¹ The Burgers model, in fact, can be represented by a two-term generalized Maxwell model with an identical mathematical description.¹² The generalized Maxwell model, a combination of several Maxwell elements in parallel, has been widely utilized in many recent studies to characterize the viscoelastic property of glass above the transition temperature.^{13–15} The sufficient number of Maxwell elements is determined by fitting the relaxation data, either gained by stress relaxation experiments or derived from creep experiments. It is emphasized that the number of Maxwell elements strongly depends on the temperature chosen for the experimental characterization. For example, a six-term generalized Maxwell model is commonly found for investigating temperatures in the vicinity of T_g ,¹⁶ while three terms are used at higher characterizing temperatures.¹⁷

In addition, the thermal-mechanical modeling of the viscoelastic responses necessitates an incorporation of the temperature effect into the mechanical models. Most of the present studies relied on thermo-rheologically simple (TRS) modeling approach in the field of glass molding simulation.^{18–20} The TRS assumption is widely employed due to the ease of numerical

implementation where the coupling temperature-mechanical model can be simplified as a solely mechanical model by incorporating the so-called reduced time, defined by a shift function. It is emphasized that this implementation is valid if and only if the shape of stress relaxation functions at every temperature, when plotted against a logarithmic time, is fully identical. However, Scherer stresses that, albeit a good approximation near the glass transition, it fails at high temperature where the glass exhibits liquid-like viscoelasticity.²¹ In particular, sensitivity studies of the TRS assumption highlight that non-negligible deviation occurs in the numerical predictions of the molded lens shape²² or creep displacements.^{5,23}

Finally, to characterize the thermo-viscoelastic material behavior of the glass, the most common approach, saying *conventional approach*, is to perform experiments, either creep or stress relaxation, at several temperatures. The stress relaxation experiment is likely more favorable because the model's parameters, for example, the generalized Maxwell model, can be directly obtained by fitting the stress relaxation data measured at one specific temperature, called the reference temperature.²⁴ The disadvantage of this approach is that the stress relaxation experiments are mostly conducted at temperatures in the vicinity of T_g , which are quite far from the actual molding temperature, normally near S_p . In this context, the viscoelastic properties at the actual molding temperature are determined by using the shift function. In contrast, the characterization of viscoelastic properties at the real molding temperature is restricted to the creep experiment. At such high temperature, in fact, the glass becomes liquid-like where its relaxation time is extremely fast ($\tau \ll$ seconds) so that the data obtained from the stress relaxation experiment is not sufficient and reliable for postprocessing. Based on the creep approach, the mold displacements are acquired and used to compute creep compliances.^{25,26} The drawback of this approach is that the temperature-dependent viscoelasticity is not accomplished. Instead, the modulus relaxations need to be derived from the creep compliance functions using inter-conversion theory. Relying on the creep setup, in addition, previous studies realize a non-negligible discrepancy between the experiment and numerical prediction of the creep displacements.^{26,27} According to their thoughts, the discrepancy is attributed to friction at the glass–mold contact interface. Nevertheless, satisfactory verifications of the creep displacement cannot be obtained by varying the friction coefficients. A recent study by Yu et al shows that if the frictional disturbance can be diminished from the creep data so that mere purity of the viscoelastic property is secured by creep experiments, good agreement can be found.²⁸

Three major contributions are aimed in this research. First, a comprehensive study on the conventional characterization of the thermo-viscoelastic properties derived from the creep experiments is presented. The study aims to demonstrate the rationale for the mismatches between the simulation and creep

experiments reported in earlier studies; and the following is to provide a solution to such existing problem. Furthermore, the thermo-mechanical modeling using TRS over a wide range of molding temperature is examined. The inadequacy of TRS over the wide temperature range is then elaborated. Finally, this research introduces a new thermo-viscoelastic modeling approach to overcome this deficit. According to the proposed method, the prerequisite of the relaxation functions and shift factors by using the TRS assumption to characterize the thermo-viscoelastic properties can be completely eliminated. Experimental validations are carried out for three different glass types commonly used in compression molding for glass optic production. Excellent agreement between experiment and simulation results found in all three glasses strengthens the validity of our method.

2 | THEORY OF 3D LINEAR THERMO-VISCOELASTICITY

The constitutive thermo-rheological model of a viscoelastic material determines the relationship among stress, strain, strain rate, and temperature. In view of mechanical modeling, the stress–strain relation can be either described by differential forms or hereditary integrals.²⁹ The integral representation is commonly chosen as its convenience for incorporating temperature into the mechanical model and flexibility to manipulate the measured viscoelastic properties, ie, creep and stress relaxation. For linear viscoelastic theory, the relation between stress and strain is linear and time dependent, represented by the following three-dimensional (3D) integral forms, using Boltzmann superposition principle:

$$\sigma(t) = \int_{0^-}^t \mathbb{E}(t-t') \frac{\partial \epsilon(t')}{\partial t'} dt', \quad (1)$$

$$\epsilon(t) = \int_{0^-}^t \mathbb{J}(t-t') \frac{\partial \sigma(t')}{\partial t'} dt', \quad (2)$$

where σ and ϵ are stress and strain tensor, respectively; $\mathbb{E}$ and $\mathbb{J}$ are relaxation modulus—and creep compliance tensors, respectively; t is the actual observation time, and t' is an arbitrary time prior to t .

The rate of relaxation rapidly varies with the change of temperature. For incorporating the temperature into the mechanical formulation, the conventional approach relies on the concept of time-temperature superposition principle, mainly based on the TRS property. Based on this concept, a single parameter ξ is introduced to combine the effects due to both time and temperature. Then, Equations (1) and (2) in varying temperature becomes:

$$\sigma(t) = \int_{0^-}^t \mathbb{E}(\xi - \xi') \frac{\partial \epsilon(t')}{\partial t'} dt', \quad (3)$$

$$\epsilon(t) = \int_{0^-}^t \mathbb{J}(\xi - \xi') \frac{\partial \sigma(t')}{\partial t'} dt', \quad (4)$$

where ξ is the so-called “reduced time”, which is related to the real time t by a temperature shift factor $a(T)$ and a reference temperature T_r . In case of transient temperature conditions, ie, temperature varying with time, the reduced time is defined as follows:

$$\xi = \int_0^t a(T(t')) dt', \quad (5)$$

where the shift factor is given by:

$$a(T) = \frac{\tau(T_r)}{\tau(T)}. \quad (6)$$

In Equation (6), τ is the relaxation time. Here $a(T)$ defines the horizontal shift from the relaxation curve of the computed temperature T to that of the reference temperature T_r , called the *master curve*.

As mentioned earlier, characterization of the viscoelastic responses can be performed by either the creep or stress relaxation experiments. In fact, both these phenomena are the viscoelastic properties of the same material; therefore, they can be interconverted. According to the interconversion theory, the relation between creep compliance and relaxation modulus can be defined as a compact form in s -domain by applying Laplace transform:

$$\hat{\mathbb{E}}(s) \hat{\mathbb{J}}(s) = \frac{1}{s^2}. \quad (7)$$

Such a relation can be written in the time domain by performing the inverse Laplace transform:

$$\int_{0^-}^t \mathbb{E}(t-t') \mathbb{J}(t') dt' = \int_{0^-}^t \mathbb{J}(t-t') \mathbb{E}(t') dt' = t. \quad (8)$$

Using Equation (7), the relaxation modulus is derived from the creep compliance on the s -domain, and following the relaxation modulus on the time domain $\mathbb{E}(t)$ is defined by the inverse Laplace transform, using Equation (8). Then, considering the TRS assumption, the shift factor is determined from the modulus relaxations similar as the stress relaxation approach.

Finally, in 3D formulation of the viscoelastic responses, the stress–strain relation, eg, Equation (1), is commonly

expressed in a form where the deviatoric and volumetric parts are separated. In this regard, the shear and bulk (volumetric) relaxation moduli are employed instead of Young modulus. Instantaneous shear and bulk moduli can be computed from Young modulus and Poisson ratio. In the viscoelastic regime, shear deformation is typically more dominant than bulk deformation³⁰; hence, time-independent bulk modulus, ie, no relaxation, is commonly agreed. Accordingly, the shear relaxation modulus and its temperature-dependent characteristic are the most essential requirements for the characterization of the thermo-viscoelastic properties.

3 | CONVENTIONAL CHARACTERIZATION OF THERMO-VISCOELASTIC MATERIAL BEHAVIOR OF GLASS

3.1 | Experimental procedure

The characterization of the glass viscoelastic properties is based on the creep approach using the uniaxial compression setup. The creep experiments are conducted in a precision glass-molding machine Toshiba GMP 207HV. Borosilicate SUPRAX[®] 8488, a “long” glass type, is chosen for the investigation of the thermo-viscoelastic responses over a wide temperature range. Thermal and mechanical properties of this glass are provided in Table 1. The glass specimen has a cylinder shape. In addition, a high-temperature resistance glassy-carbon SIGRADUR[®] is used as the mold material for two essential purposes. First, this mold material allows the molding experiments at high temperatures, even above S_p , without any glass sticking problem. Second, by fine polishing, the surfaces of the mold pair is mirror-like with the roughness of $R_a = 2 \text{ nm}$.³¹ Such ultraprecision mold surface enables a small friction coefficient at the glass-mold interface, below 0.1.³² As indicated in the previous works,^{5,26,28}

the low friction coefficient is essential to minimize the error that leads to the impurity of the viscoelastic properties derived from the creep experiments due to the frictional deformation of the glass. Figure 1 exhibits the polished glassy-carbon molds and the glass specimen used for the creep experiments. Besides, demonstrations of pressed glasses at high molding temperatures in the vicinity of S_p are presented.

Creep experiments are performed at four specific temperatures ranging from 650°C to 790°C, which corresponds to the viscosity range $\eta = 10^{9.5} - 10^{6.8} \text{ Pas}$. Such viscosity range, from slightly above T_g to nearly S_p , is of interest for this study because this is a typical molding temperature range for PGM. Besides, the glass deformation from a preform to nearly final shape is mostly occurred in this temperature range by NGM,^{33,34} before it is sent to a separate annealing oven. Please note that the experimental characterization of the viscoelastic properties is conducted for equilibrium glasses. The glass specimens are slowly heated up with a rate of 5 K/min and then undergone a sufficient long soaking, approximately 20 minutes, in order to erase the thermal history of the specimens before the creep experiments. Within the chosen temperature range, this soaking time is expected to satisfactorily equilibrate the glass specimens. For the creep compression, a pressing time of 900 seconds is held for all experiments. The mold displacement is recorded and used in the following steps to derive the thermo-viscoelastic properties of the glass. Figure 2A introduces the mold displacements of four testing temperatures.

3.2 | Computational procedure of creep compliance

The computational procedure to obtain the viscoelastic properties from the creep displacements has been presented in the earlier works.^{25,26} For the completeness of the paper, most

TABLE 1 Material properties of glass

Properties*	Suprax 8488	Xensation 3D	IRG 27
E (GPa)	67	83	16.77
ν (—)	0.20	0.22	0.24
ρ (kg/m ³)	2310	2490	3200
λ (W/mK)	1.2	1.22	0.33
α (10 ⁻⁶ /K)	4.3	8.5	22.5
C_p (J/kgK)	750	890	482
T_g (°C)	540	505	197
S_p (°C)	808	720	N/A

*Nomenclature: E , modulus of elasticity; ν , Poisson's ratio; ρ , density; λ , thermal conductivity; α , thermal expansion coefficient; C_p , specific heat; T_g , glass transition temperature; S_p , softening temperature.



FIGURE 1 Demonstrations of the polished glassy carbon molds, glass specimen, and pressed glass samples at the molding temperature in the vicinity of S_p [Color figure can be viewed at wileyonlinelibrary.com]

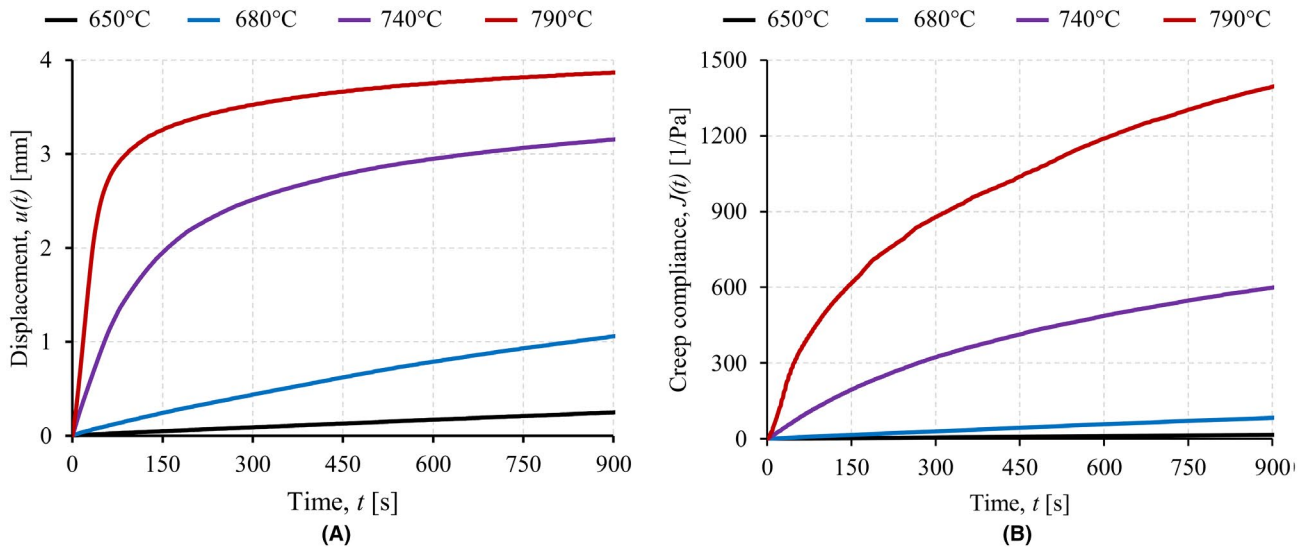


FIGURE 2 A, Creep displacements of uniaxial compression experiments; and (B) creep compliances computed from the displacements [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com/doi/10.1111/jace.16963)]

important aspects are highlighted. First, true stress and true strain are taken to account for the large deformation of the glass specimens. By using the recording displacement history $u(t)$ and the initial length of the specimen L_0 , the true strain $\varepsilon(t)$ is given as follows:

$$\varepsilon(t) = \ln \frac{L(t)}{L_0} = \ln \frac{L_0 - u(t)}{L_0}. \quad (9)$$

Following the computation of the true strain, the true stress $\sigma(t)$ is calculated by using the initial cross-section A_0 of the specimen and applying force F :

$$\sigma(t) = \frac{F}{A(t)} = \frac{F \cdot L(t)}{A_0 \cdot L_0} = \sigma_0 \cdot \frac{L(t)}{L_0} = \sigma_0 \cdot \exp(-\varepsilon(t)), \quad (10)$$

Here, please note that the applying force $F = 250\text{N}$ is remained for all experiments of the entire temperature range. This deployment is aimed to avoid the effect due to the pressure-dependent viscosity,³⁵ which is not considered in this study.

The true stress is calculated by considering a constant volume of the deformed glass specimen. Theoretically, the creep test requires an instantaneous applying stress σ_0 that holds constantly during the entire experiment. Under experimental conditions, however, such requirement cannot be fulfilled. Since the machine solely controls the applying force, the significant increase of the cross-section during the deformation of the glass specimen results in a decrease of the compression stress. To account for the change of the true stress as a variable with time, $\sigma(t)$, the stress input is approximated by the sum of serial constant stress inputs σ_i .²⁹ According to the Boltzmann superposition principle, the strain output under the variable stress input equals to the sum of all discretized

strain outputs ε_i resulting from each corresponding discretized stress inputs σ_i .

Now, it is important to point out that only vertical displacements of the mold, ie, one-dimensional (1D) data are acquired by the uniaxial compression setup. Thus, Equation (2) needs to be rewritten in the 1D form. In addition, by separating the initial applied stress ($\sigma(0) = \sigma_0$) and the variable stress, for the sake of creep compliance computation, an alternative form of Equation (2) becomes:

$$\varepsilon(t) = \int_0^t J(t-t') \frac{\partial \sigma(t')}{\partial t'} dt' + J(t) \sigma(0). \quad (11)$$

This convolution integral can be solved by a finite difference scheme using discretized representations of the true stress σ_n , true strain ε_i , and creep compliance J_i , defined as follows:

$$\varepsilon_i = \sum_{n=1}^i J_{i-n} \left(\frac{\sigma_n - \sigma_{n-1}}{\Delta t} \right) \Delta t + J_i \sigma_0. \quad (12)$$

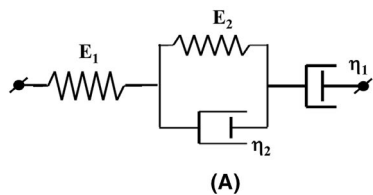
Rearranging Equation (12), the discretized creep compliance J_i is given as follows:

$$J_i = \frac{\varepsilon_i}{\sigma_0} - \frac{1}{\sigma_0} \sum_{n=1}^i J_{i-n} \left(\frac{\sigma_n - \sigma_{n-1}}{\Delta t} \right) \Delta t. \quad (13)$$

Thus, from the recording displacement inputs and the initial geometry of the glass specimens, using Equations (9-10), the creep compliances $J(t)$ are achieved by solving Equation (13) iteratively. Figure 2B provides the creep compliances computed from the displacement inputs $u(t)$.

3.3 | Determination of relaxation modulus

The relaxation modulus can be achieved if the creep compliance function is known, according to Equation (7). Since the creep compliance contains discrete data, the creep function is realized by curve fitting. The fitting of the creep compliance data requires a material model. For example, generalized Kelvin with three terms is used in previous studies.^{17,26} For the liquid-like creep behavior characterized at high temperature range investigated in this study, instead, the 4 element-Burgers model is employed. The phenomenological representation of the Burgers model is shown in Figure 3A. The creep function $J(t)$, accordingly, is defined as follows:



— Fitting data 790°C × Exp. data 790°C — Fitting data 740°C
 □ Exp. data 740°C — Fitting data 680°C △ Exp. data 680°C
 — Fitting data 650°C ○ Exp. data 650°C

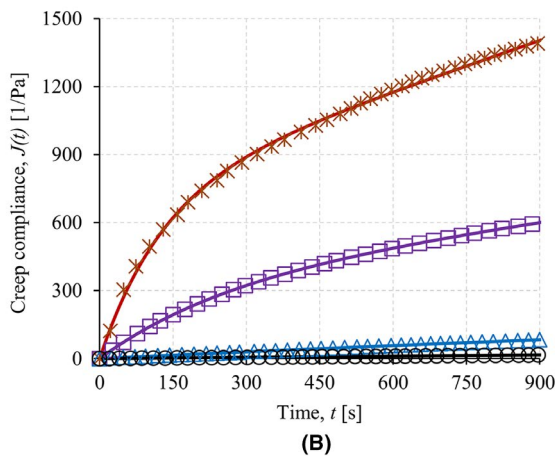


FIGURE 3 A, Phenomenological Burgers model; and (B) fitting of creep compliance using the Burgers model [Color figure can be viewed at wileyonlinelibrary.com]

TABLE 2 Relaxation parameters of Suprax 8488

Temperature (°C)	A, Coefficients computed by Equation (15A)				B, Revised coefficients using Equation (15B)			
	Weight factors, g_i (—)		Relaxation time, τ_i (s)		Weight factors, g_i (—)		Relaxation time, τ_i (s)	
	g_1	g_2	τ_1	τ_2	g_1	g_2	τ_1	τ_2
650	0.999611	3.89e-4	0.7556	286.79	0.999689	3.1e-4	0.6045	286.77
680	0.999884	1.16e-4	0.1354	355.10	0.999908	9.2e-5	0.108	335.09
740	0.999971	2.90e-5	9.35e-3	1432.85	0.999908	2.8e-5	7.48e-3	1432.84
790	0.999984	1.60e-5	2.28e-3	1122.511	0.999987	1.3e-5	1.83e-3	1122.507

$$J(t) = \frac{1}{E_1} + \frac{1}{E_2} \left(1 - e^{-E_2 \cdot t / \eta_2} \right) + \frac{t}{\eta_1}, \quad (14)$$

where E_1 , E_2 , η_1 and η_2 are the fitting parameters of the Burgers model.

There are several advantages of utilizing the Burgers model. First, as demonstrated in Figure 3B, the Burgers model illustrates an outstanding fitting accuracy over the entire temperature range of investigation. An earlier study of the author has indicated that the Burgers model precisely predicts the creep displacements of B270i glass in a wide molding temperature range, which correspond to viscosity $\eta = 10^{11.7} - 10^7$ Pas.³⁶ Second, it is emphasized that because the Burgers model consists of a Maxwell and a Kelvin element, it is capable of representing both creep and stress relaxation phenomena. As a result, the relaxation function can be directly computed from the fitting parameters obtained from the creep compliance. This property helps to avoid the second fitting of the relaxation modulus to attain the parameters of the relaxation modulus function as pursued in most of the previous works.^{25,26} Moreover, the Burgers model is mathematically equivalent to a two-term Prony series; therefore, the parameters obtained from the creep compliance function (Equation 14) can be used to compute the Prony series parameters, ie, weight factor g_i and relaxation time τ_i ($i = 1, 2$), in a straightforward manner. Details of the mathematical equivalence between the Burgers model and two-term Prony series are described in Appendix A. When the Prony series parameters are known, the material model can be simply implemented into most of the commercial FEM simulation software, eg, ABAQUS, using the hereditary integral formulation. Finally, compared to generalized Maxwell model with 3-6 terms commonly employed in other works, the Burgers model contains the fewest fitting parameters (4 parameters) that bring essential advantages for the proposed modeling method of the thermo-viscoelastic material behavior presented in Chapter 4.

Since four parameters of the model are already realized by fitting the creep compliance using Equation (14), the shear parameters of the Burgers model, G_1 , G_2 , μ_1 and μ_2 , can be calculated as follows:

$$G_1 = \frac{E_1}{2(1+\nu)}, G_2 = \frac{E_2}{2(1+\nu)}, \mu_1 = \frac{\eta_1}{2(1+\nu)}, \mu_2 = \frac{\eta_2}{2(1+\nu)}. \quad (15A)$$

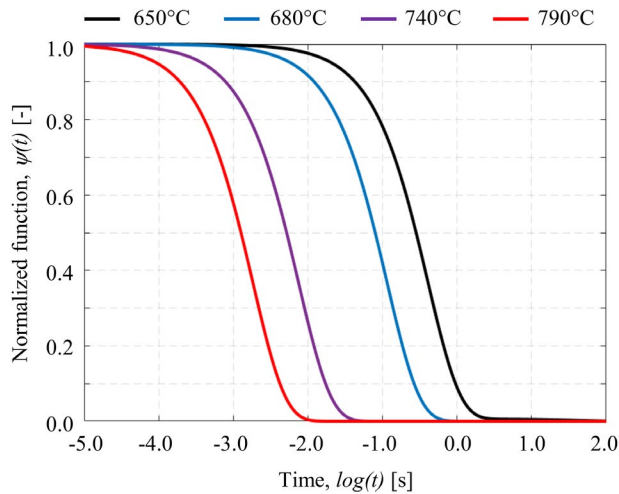


FIGURE 4 Normalized modulus relaxation functions of four testing temperatures [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

Using the shear parameters in Equation (15A), Prony series coefficients, including weight factors g_i and τ_i ($i = 1, 2$), are achieved as demonstrated in Appendix A (Equations A6 and A9). Table 2A provides the computed coefficients of four investigating temperatures. Please note that as these coefficients are calculated directly from the creep compliance parameters, the second fitting of the relaxation modulus curves is no longer necessary. Using these coefficients, Figure 4 plots the normalized shear relaxation moduli ($\psi(t) = G(t)/G_0$) of four testing temperatures, accordingly.

3.4 | Numerical verification of shear relaxation parameters

A Finite Element (FE) model is established to verify the shear relaxation parameters derived from the uniaxial creep experiments. Process inputs and boundary conditions of the FE model are described in Figure 5. For the numerical verification of the creep displacements, ie, the glass thickness changes, only the glass specimen is considered. The glass has a cylinder shape and is modeled as an axisymmetric deformable body. Vertical displacements of bottom surface are constrained, while the top surface is coupled with the applying force F taken from the experiments. Since mold parts are not necessarily included in the FE model, friction between the glass and molds is neglected. In addition, avoidance of the dimensionality effect on the numerical verifications is particularly taken. Before each creep experiment, the initial length and diameter of the glass specimens are measured with an accuracy of 3 digits. Then, these geometrical inputs are set for the respective simulation of the same experiment temperature. The numerical verification is performed using ABAQUS/CAE

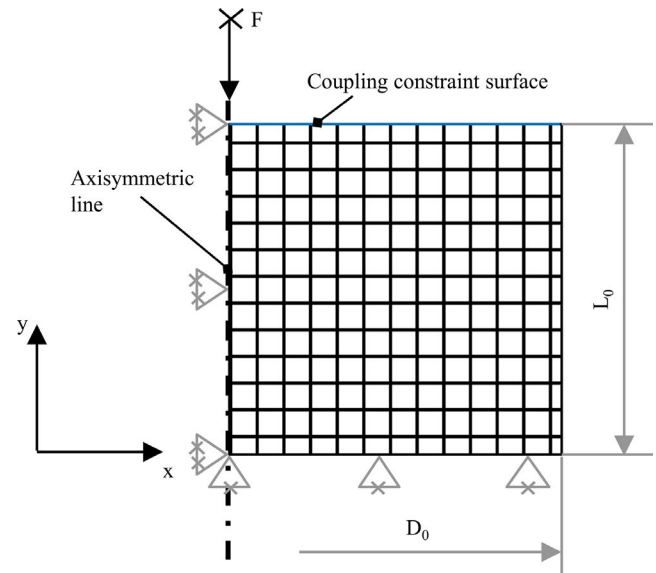


FIGURE 5 Representation of FE model for numerical verifications [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

2017. The glass properties and viscoelastic parameters are given in Tables 1 and 2A.

Figure 6A presents the simulation results of the creep displacements. When compared with the experiment data, however, the simulation results show obvious disagreements found in all testing temperatures. In fact, the numerical predictions of the creep displacement are always underestimated. As mentioned earlier, observations of such discrepancy were reported in the previous works.^{26,27} In their studies, the mismatch is believed due to friction at the glass and mold contact surfaces. They argued that the displacement acquired by the compression setup is not purely viscoelastic creep deformation of the glass but contains a frictional resistance at the interface. The presence of friction violates the nature of the creep data used for the derivation of the viscoelastic parameters, and it causes the discrepancy. Several studies investigated the influence of the friction on the viscoelastic constants using the creep compression setup. It is demonstrated that the accuracy of the obtained viscoelastic parameters can be enhanced if the friction between the glass and mold is sufficiently small,²³ as approached in this research, or the error due to the friction can be dismissed from the displacement data.²⁸

The authors agree that the friction at the glass-mold interface, which cannot be avoided by the compression setup, may violate the pure creep deformation due to viscoelastic property of an investigating material. In this study, however, another reason that crucially results in the disagreement between the experiment and simulation results is elucidated. The argumentation is that if the creep displacement is used as the input data for the derivation of viscoelastic parameters, and the numerical verification—indeed a reverse computation—must bring an identical result. Furthermore, because such input displacement comprises the deformation due to friction, the

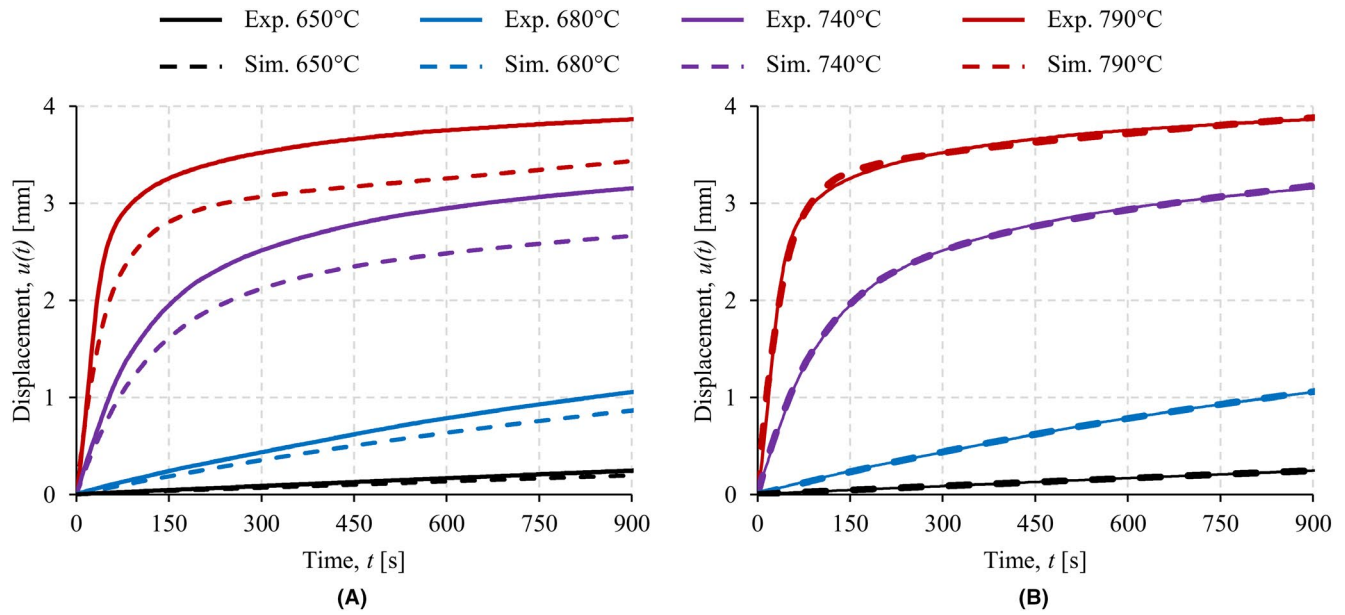


FIGURE 6 Verification of creep displacements using: (A) parameters computed by Equation (15A); and (B) revised parameters using Equation (15B) [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com/doi/10.1111/jace.16963)]

friction at the glass-mold interface should not be reconsidered in the FE simulation for the verification.²⁵ According to the thoughts, the simulation and experiment displacements have to be identical. So why do such large discrepancies observed in Figure 6A come from? Described in Sections 3.2 and 3.3, the shear relaxation modulus is derived from the uniaxial displacement, ie, the 1D experiment data, and is computed using the 1D viscoelastic formula (Equation 15A). Viscoelastic theory performed in ABAQUS simulation platform is developed for generalization of the integral representation to three dimensions.³⁷ In 3D formulation, the stress tensor is split into volumetric and deviatoric parts for the ease of modeling the nature of volume-preserving materials such as viscoelasticity. Here, it is emphasized that while the volumetric (dilatational) tensor carries out the bulk or volumetric deformation of the materials, the deviatoric tensor solely accounts for the shear deformation, meaning that no volumetric changes are taken. Meanwhile, as discussed earlier, the bulk deformation is time-independent, ie, no viscoelastic property. For this reason, the Poisson's ratio given in Equation (15A) must be set to 0.5 instead. This implementation is essential to correct the computation of the shear parameters of the constitutive model given in the 3D viscoelastic formulation while taking no account of the deformation due to viscoelastic bulk.³⁸ The time-independent bulk modulus, in fact, results in an elastic response,⁵ and therefore it can be accounted by the elastic component of the Burgers model (E_1). Eventually, the shear relaxation parameters are defined as shown in the following revised equation:

$$G_1 = \frac{E_1}{2(1+\nu)}, G_2 = \frac{E_2}{2(1+0.5)}, \mu_1 = \frac{\eta_1}{2(1+0.5)}, \mu_2 = \frac{\eta_2}{2(1+0.5)}. \quad (15B)$$

Table 2B provides the revised Prony series coefficients calculated from shear relaxation parameters using Equation (15B). Based on these coefficients, simulations are rerun using the same setting as described in Figure 5. Figure 6B plots the simulation results and the experiment creep displacements. It is clear that great agreements of all experiments are found. Please remind the underestimation of the simulated creep displacements observed in Figure 6A. This underestimation results from the contribution of the bulk deformation implicit in the shear parameters by Equation (15A), which is certainly not when the deformation of the viscoelastic bulk is excluded from the computation of the shear parameters using Equation (15B). The validated results emphasize the essential implementation of the shear relaxation coefficients in the 3D formulation for the simulation of the glass molding processes.

Up to now, the viscoelastic parameters derived from the creep displacement at individual testing temperature are fully validated. For the glass molding simulation, however, temperature-dependent viscoelastic parameters are required. For modeling the thermo-viscoelastic responses, most of the previous studies relied on the TRS behavior of the viscoelastic materials. The next step focuses on an examination of this thermo-viscoelastic modeling approach over the temperature range chosen in this study.

3.5 | Numerical verification of thermo-viscoelastic parameters using TRS

Thermo-viscoelastic modeling using TRS necessitates a shift function. The shift function is determined from the shear relaxation modulus functions varying with temperatures (Figure 4). First, the master curve at an arbitrary experiment temperature—the

reference temperature T_r —is selected. In this study, the shear relaxation function at 680°C is considered. Following, this master curve is shifted to the relaxation curves of other temperatures along the abscissa. Relying on the Williams-Landel-Ferry (WLF) equation, the shift function is defined as follows:³⁹

$$\log(a(T)) = \frac{-C_1(T - T_r)}{C_2 + (T - T_r)}, \quad (16)$$

where C_1 and C_2 are constants obtained by fitting the shifts computed from the master curve to each individual temperature. Figure 7 exhibits the curve fitted by WLF equation and the constants.

Figure 8 provides simulation results of the creep displacements using temperature-dependent viscoelasticity by using the WLF shift function. At the reference temperature ($T = 680^\circ\text{C}$), the experiment and simulation results are certainly validated, exactly the same as observed in Figure 6B. However, discrepancies between the simulation and experiment data occur at other temperatures. The discrepancy becomes larger and non-negligible at temperatures further from the reference temperature. This evidence indicates the inadequate implementation of the TRS behavior for a large temperature range. Consequently, numerical predictions of the molded glass characteristics, eg, form deviation or stress, are not reliable.

The underlying cause of the observed discrepancies stem from an improper assumption using the TRS over the range of studying temperature. In principle, the application of the TRS is valid if and only if the master relaxation curve and the shifted curves are fully identical, because only a horizontal shift is acted by the TRS theory. In order to examine this requisite, Figure 9A plots the relaxation functions at four testing experiments (solid lines) and the relaxation curves shifted from the master curve (dashed lines). The plots reveal that the modulus relaxation function and the shifted curve are not fully identical. While two data have a good match within the first relaxation response, ie, fast

relaxation time ($\tau_1 \approx 10^{-5} - 10^{-2}$ seconds), an obvious distinction can be recognized when one looks a bit closer on the long-term relaxation response ($\tau_2 \approx 10^{-2} - 10^3$ seconds) as indicated in the zoomed plot (Figure 9B). Similar findings can be seen in the work of Zhou et al²⁶ and Yu et al²⁸; however, impacts of such nonidentical relaxation response in the long term on the creep displacements have not been studied. It is obvious that the first relaxation period occurs extremely short (<0.01 second) compared to the total pressing time. In contrast, the second relaxation period, though it weighs only a small proportion of the entire relaxation spectrum, holds the entire creep experiment, starting at very early stage (0.01 second) till the end of the pressing time (900 seconds). Therefore, the second relaxation stage definitely contributes a substantial dominance, while the first relaxation barely plays a role in the overall deformation of the glass in the creep experiments. According to the definition of the TRS (Equation 6), only one relaxation time—the first relaxation time τ_1 —is mainly involved in the determination of the shift factor because of its dominant overlapping ratio. The second relaxation times, however, have not the same temperature dependence, leading to the dissimilarities between the master and shifted curves when using the shift factors computed from the first relaxation times. Such inadequate characteristic of the TRS makes it insufficient to represent the temperature-dependent viscoelastic property over a wide temperature range. Clearly seen in Figure 8, even a small difference of relaxation response in the long-term relaxation time can bring substantial deviations in the numerical computation of creep displacements.

Several studies attempt to enhance the model predictions by not only shifting the master curve horizontally but also vertically, called *Thermo-Rheologically Complex*, so that the vertical differences can be reduced.⁴⁰ However, it is stressed

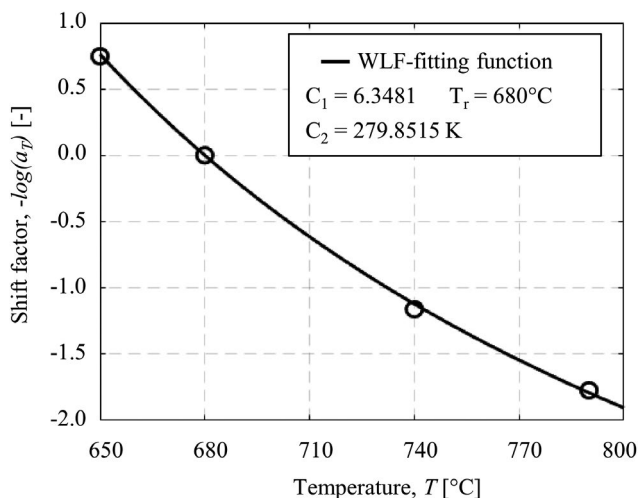


FIGURE 7 WLF-fitting function and constants

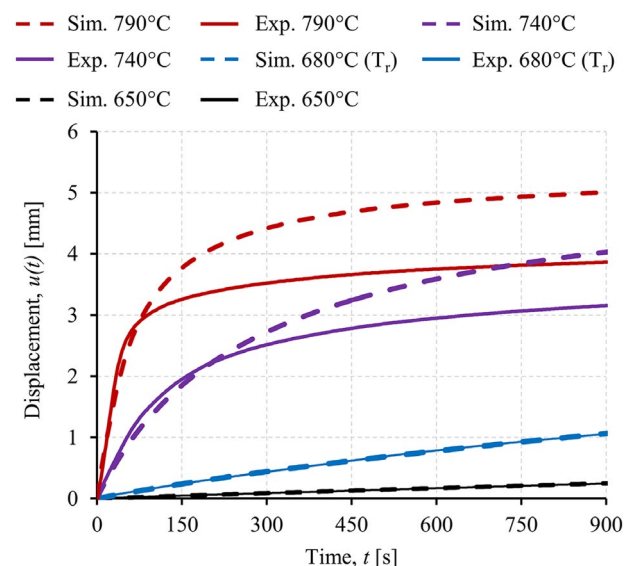


FIGURE 8 Simulation results of creep displacements using thermo-rheologically simple [Color figure can be viewed at wileyonlinelibrary.com]

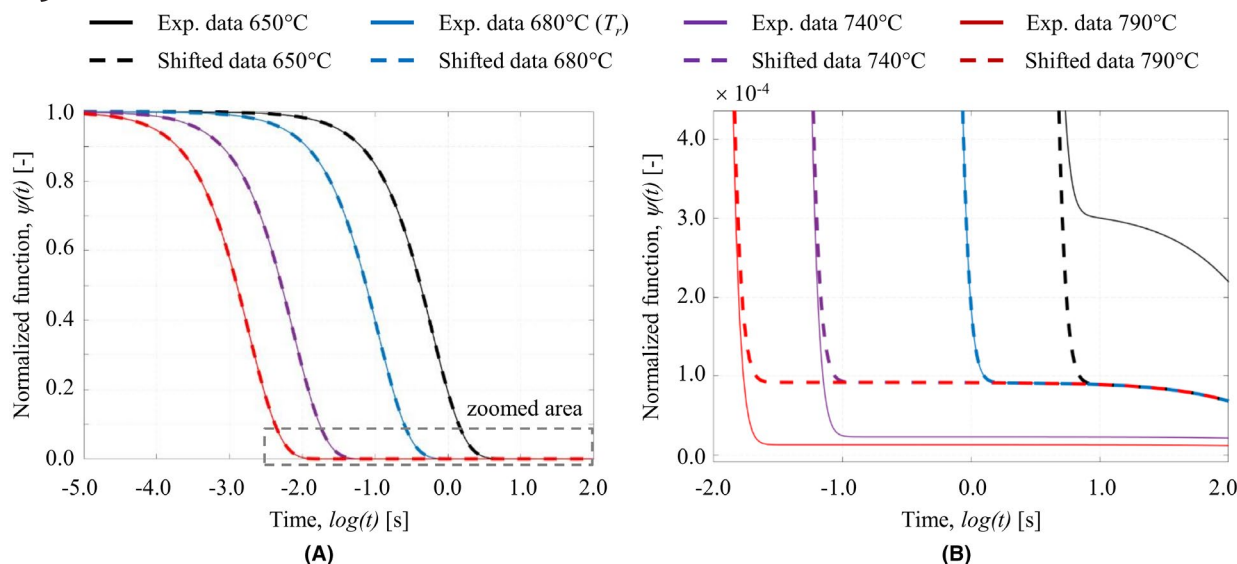


FIGURE 9 A, Relaxation functions and shifted data using WLF function; and (B) zoomed plot of relaxation functions at large time scales [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com/doi/10.1111/jace.16963)]

that the vertical shift, again, is another assumption, and it is only valid if the master and shifted curves are fully vertically identical. In the following chapter, an alternative approach is introduced to improve the modeling of temperature-dependent viscoelasticity without assuming that the thermorheological material behavior is simple or complex.

4 | PROPOSED THERMO-VISCOELASTIC MODELING APPROACH

This chapter introduces a new modeling approach of the thermo-viscoelastic behavior of glass materials. The proposed method aims at the characterization of the temperature-dependent viscoelastic parameters without using the relaxation moduli and shift function. To characterize the viscoelastic properties at a specific temperature, ie, *isothermal rheology*, the earlier verifications demonstrate that the rheological Burgers model is fully adequate to represent the creep response (Figure 6B). For incorporating temperature into the rheological model, ie, *thermorheology*, the central point is to pursue a unique set of Burgers model parameters, called Burgers parameters, which are dependent on temperature, and this unique parameter set is able to represent the glass viscoelastic responses over the entire temperature range.

As previously presented, the relaxation parameters, ie, relaxation time τ_i and weight factors g_i ($i = 1, 2$), can be directly calculated from four Burgers parameters (Equations A6 and A9; Appendix A). It means that if there exists a temperature-dependent function for each individual Burgers parameter, the relaxation parameters at any temperature can be determined while the prerequisite of the relaxation functions and shift factors is no longer necessary.

4.1 | Determination of Burgers parameters

Our method focuses on finding a parameter set of the Burgers model, $\varphi = \varphi(E_1(T), E_2(T), \eta_1(T), \eta_2(T))$, which is able to represent both creep and stress relaxation over the entire temperature range. Using the creep compliances at four experiment temperatures (Figure 3B), the Burgers parameters of each temperature are obtained by a nonlinear curve fitting. Figure 10 presents the fitting values of these parameters in natural logarithmic scales varying with temperatures. Here, it is noted that the first spring of the Burgers model, E_1 , represents the instantaneous viscoelastic response, which is accounted for the Young modulus of glass materials. This parameter can be temperature

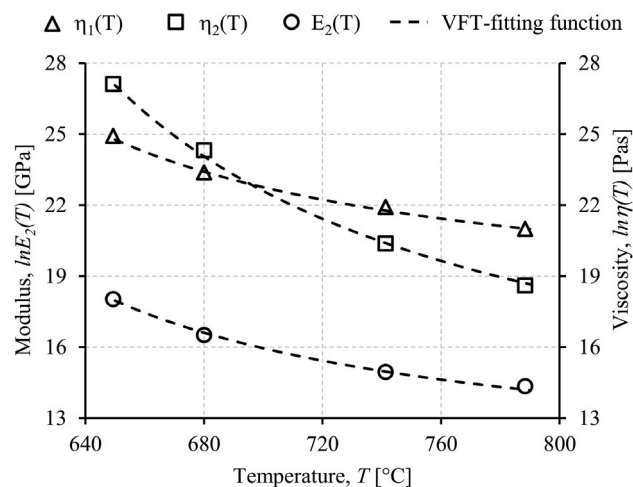


FIGURE 10 Temperature-dependent parameters of the Burgers model

dependent, measured by several available techniques, such as Brillouin light scattering³ or impulse excitation.⁴¹ However, since most of glass molding simulations consider a constant Young modulus, usually available in glass datasheet, the parameters E_1 is assumed to be temperature independent. Thus, this value is constant given at room temperature (Table 1). The relations of other parameters, ie, E_2 , η_1 and η_2 , with temperature, plotted in Figure 10, reveal a unique trend where the properties of these parameters are decreasing with increasing temperatures.

In the next step, such relations between each Burgers parameter and temperature demand a best mathematical description, ie, a relevant temperature-dependent model. In this regard, it is necessary to figure out a realistic model using the fewest possible number of fitting parameters. Since the observing trends shown in Figure 10 are similar to that of the glass viscosity, existing models for the equilibrium viscosity of glass can be a good reference. In the transition region, most of the glass exhibits non-Arrhenius behavior, and a sufficient model to represent such behavior requires at least three parameters. In this study, the major focus puts on three-parameter models only, able to describe the universal physics of the viscoelastic responses. A model in the form of Vogel-Fulcher-Tammann (VFT) equation is chosen given as follows:

$$\ln(\varphi) = A(\varphi) + \frac{B(\varphi)}{(T - T_0(\varphi))}, \quad (17)$$

where T is the absolute temperature, and φ is each of the three temperature-dependent parameters of the Burgers model, ie, E_2 , η_1 and η_2 . In other words, each Burgers parameter can be determined at any temperature by the VFT function. Advantages of the proposed method are to increase the freedom of each model parameter when they are coupled with temperature, whereas all thermo-mechanical coupling parameters using the TRS property are restricted to a single temperature-dependent variable—the shift factor. Please note that the similar approach can be applied to any other rheological models instead of the Burgers model.

The VFT-fitting function in Equation (17) is chosen because each coefficient contains universal physics; though it is commonly known as an empirical estimation of viscosity with temperature dependence in the glass transition. In fact, the three coefficients, A , B , and T_0 , in the VFT form correspond, respectively, to (a) the viscosity at infinite temperature; (b) a function curve defining the fragility of a glass-forming material; and (c) a temperature of a glass-forming liquid at which its entropy, if the liquid is cooled sufficiently slow, coincides with the crystal entropy, defined as the Kauzmann temperature.⁴² Understanding the physical meaning of each coefficient helps to minimize the number of fitting coefficients.

4.2 | Minimization of number of fitting coefficients

Using the VFT-fitting equation, the three temperature-dependent Burgers parameters are obtained, each of which contains three unknown coefficients. Thus, the total number of fitting coefficients is nine, including $A(E_2)$, $B(E_2)$, $T_0(E_2)$; $A(\eta_1)$, $B(\eta_1)$, $T_0(\eta_1)$ and $A(\eta_2)$, $B(\eta_2)$, $T_0(\eta_2)$. It is stressed that one can use all nine coefficients to determine each Burgers parameter as the temperature dependence.

For the benefit of computational effort, however, the next step is to find a less possible number of the fitting coefficients. First, all three Burgers parameters would have the same Kauzmann temperature since it is the unique property of a glass material; hence, $T_0(E_2) = T_0(\eta_1) = T_0(\eta_2) = T_0$. Besides, as observed in Figure 10, the variation of temperature-dependent viscosity $\eta_1(T)$ is mostly identical to that of $E_2(T)$. From the mathematical view using the VFT equation, both coefficients would have a similar coefficient B —the fragility, meaning that $B(\eta_1) = B(E_2)$. Finally, we assume that the Kelvin component of the Burgers model has the same infinite viscosity, permitting $A(\eta_2) = A(E_2)$. According to this thought, the number of fitting coefficient reduces to five, which are $A(\eta_1)$, $B(\eta_1)$, $A(\eta_2)$, $B(\eta_2)$, and T_0 .

As illustrated in Figure 10, the VFT model using a set of five fitting coefficients $\mathfrak{R} = \mathfrak{R}(A(\eta_1), B(\eta_1), A(\eta_2), B(\eta_2), T_0)$ yields excellent fits for all three Burgers parameters over the temperature range of investigation. It means that each Burgers parameter can be determined at any specific temperature. By using these parameters, eventually, either creep or relaxation responses can be directly identified without awareness of the relaxation functions and shift factors.

4.3 | Thermo-viscoelastic modeling procedure

Based on the discussions in Sections 4.1 and 4.2, a thermo-viscoelastic modeling approach is established as in the following:

The procedure starts with creep experiments conducted at a certain number of chosen temperatures n (eg, $n=4$, Figure 3B) in the range of glass molding. Corresponding mold displacements u_k^{exp} over entire recording time t are acquired, while k is the index of the displacement ($k=1-n$). Goal of this study is the development of a computational method to estimate the creep displacements called u_k^{cal} , using the single set of Burgers parameters φ . Objective of the parameter estimation procedure is to minimize the difference between the estimated and experiment displacements. More specifically, the estimation procedure is converted into an optimization problem so as to determine the unique parameter set φ by matching the

estimated and experiment displacements simultaneously at all temperatures.

In fact, providing that the parameter set ϕ is known, the creep displacements are achieved according to the computational procedure presented in Section 3.2. Furthermore, the single set of Burgers parameters is able to compute all n displacements at different temperatures by using the temperature-dependent relation defined by Equation (17). Therefore, the computation of creep displacements over the experiment temperature range eventually requires determining the set $\mathfrak{R}$ of 5 unknown coefficients as discussed in Section 4.2.

A computational solver is developed for this purpose. Inputs fed into the solver consist of five unknown coefficients, the glass property (E, ν) and the experiment data, ie, recording forces and displacements. Outputs are the computed displacements u_k^{cal} . At first, u_k^{cal} are computed using an arbitrary set of coefficients. Following, they are compared with the experiment data u_k^{exp} . Providing the difference is bigger than an acceptable tolerance, a new coefficient set $\mathfrak{R}$ is updated to correct the computed displacements iteratively until the following relation is satisfied:

$$u_k^{\text{exp}}(\zeta) = u_k^{\text{cal}}(\zeta). \quad (18)$$

In this formulation, $u_k^{\text{exp}}(\zeta)$ and $u_k^{\text{cal}}(\zeta)$ are vectors containing the experiment and computed displacements, respectively, taken at a given time ζ ($0 < \zeta < t$) and at the k -th temperature; t is the recording time of the displacement. The time-displacement data obtained from creep experiments are highly nonlinear as shown in Figure 2. The overdetermined system of linear equations cannot be solved for Equation (18). Therefore, the determination of the coefficient set $\mathfrak{R}$ is necessarily required a powerful nonlinear optimization. In this study, the Levenberg-Marquardt algorithm is utilized for the parameter estimation. Accordingly, the optimization procedure serves to estimate the computational displacements iteratively until the difference can be sufficiently negligible to satisfy Equation (18).

To calculate the displacements from the coefficient set, the solver consists of 3 modules. The first module is to determine the Burgers parameters from the coefficient set. At this point, note that the Burgers parameters can be converted into relaxation parameters in the form of Prony series, for the ease of model implementation into an FEM commercial software, such as ABAQUS. In the following step, as the Burgers parameters are known, the creep compliances are computed in the second module. Lastly, the third module is employed to calculate the displacements from the computed creep compliances. Detail of the computational procedure is described in the flowchart provided in Appendix B.

TABLE 3 Thermo-viscoelastic coefficients

Coefficients	Suprax 8488	Xensation 3D	IRG 27
$A(\eta_1)$	17.347 68	18.0983	12.5106
$B(\eta_1)$	8316.032	8852.178	10 053.35
$A(\eta_2)$	10.543 54	16.357	12.689
$B(\eta_2)$	18 531.4523	3385.455	3865.25
T_0	515	823	476

4.4 | Numerical validations

Numerical validations of the proposed method are conducted in ABAQUS. The FE model shown in Figure 5 is used to simulate the creep displacements at four testing temperatures. Viscoelastic parameters are provided in Table 3. The viscoelastic parameters are implemented for the simulation in ABAQUS via a UMAT user-subroutine. Figure 11A presents the simulation results, which greatly agree with the experiment data. Compared to the large discrepancies observed in Figure 8, the proposed method obviously enhances the simulation accuracy.

In order to demonstrate the efficiency of this method, further simulations are performed to predict the creep displacements at other temperatures that are not involved in the VFT-fitting function. For this reason, creep experiments from 650°C to 790°C with an increment of 10°C are validated. Using the same viscoelastic parameters in Table 3, FEM simulations are run at every experiment temperature. Figure 11B presents the simulation results and experimental data. Exceptionally good agreement is found within the entire temperature range chosen for the validation.

Furthermore, to intensify the appropriateness of the method, other glass types, commonly used in PGM, are investigated. Beside the borosilicate glass SUPRAX[®] 8488, two other glass categories of SCHOTT, aluminosilicate Xensation[®] 3D and infrared chalcogenide IRG 27 (As_2S_3), are chosen. Thermo-mechanical properties of these glass materials are provided in Table 1. Figures 12 and 13 introduce the creep experiments and numerical validations of the Xensation[®] 3D and IRG 27, respectively. For those plots, the dashed lines in Figures 12A and 13A present the numerical verifications at the testing temperatures chosen for obtaining the Burgers parameters by the proposed method. The VFT-fitting functions of those parameters are shown in Figures 12B and 13B, and the corresponding values are provided in Table 3. Besides, the solid lines exhibit the validation of the creep displacements at temperatures that are not involved in the fitting functions. These results demonstrate that accuracy of the numerical predictions at other temperatures within the glass molding range is satisfied. The validation results of three different glass categories strengthen the validity of the proposed method in characterizing and modeling the

thermo-viscoelastic nature of glass for FEM simulation in the glass molding process.

In addition to considering the VFT model, it is emphasized that other existing equilibrium viscosity models can be valid for the proposed method. To demonstrate, the MYEGA viscosity model, recently introduced by Mauro

et al.⁴³ is investigated. Similar as the VFT form, this model contains three fitting parameters that are used to obtain the temperature-dependent Burgers parameters. Figure 14 compares the simulation of creep displacements of the MYEGA and the VFT models where exceptionally identical results are found. In future work, the validity of the equilibrium

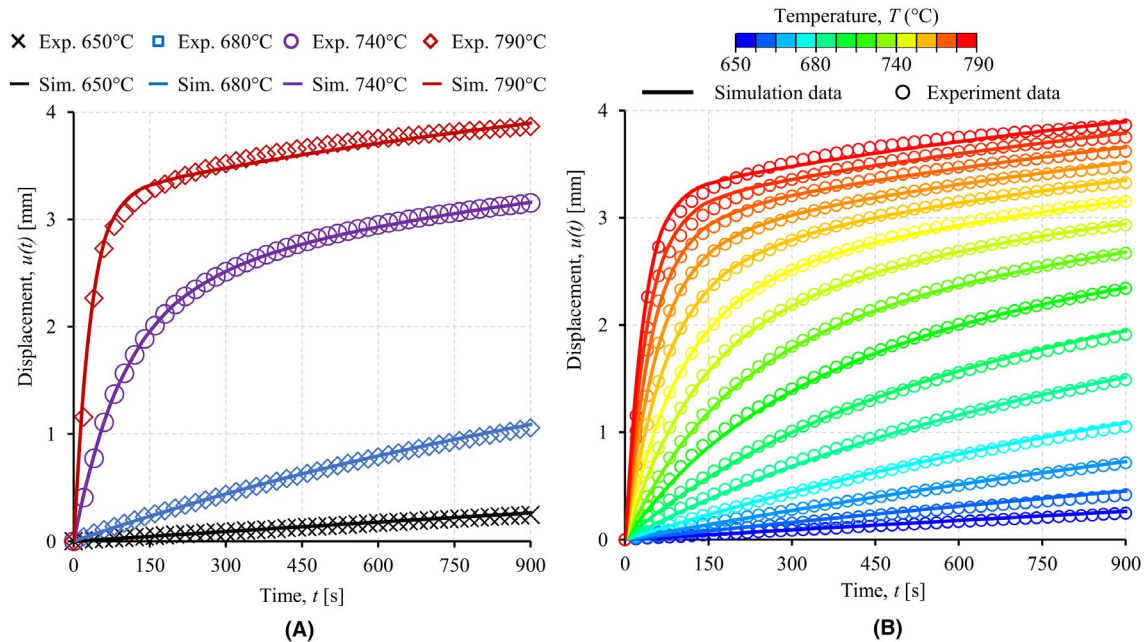


FIGURE 11 A, Validations of creep displacements for Suprax 8488 glass at four testing temperatures; and (B) validations over temperature range from 650°C to 790°C [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com/doi/10.1111/jace.16963)]

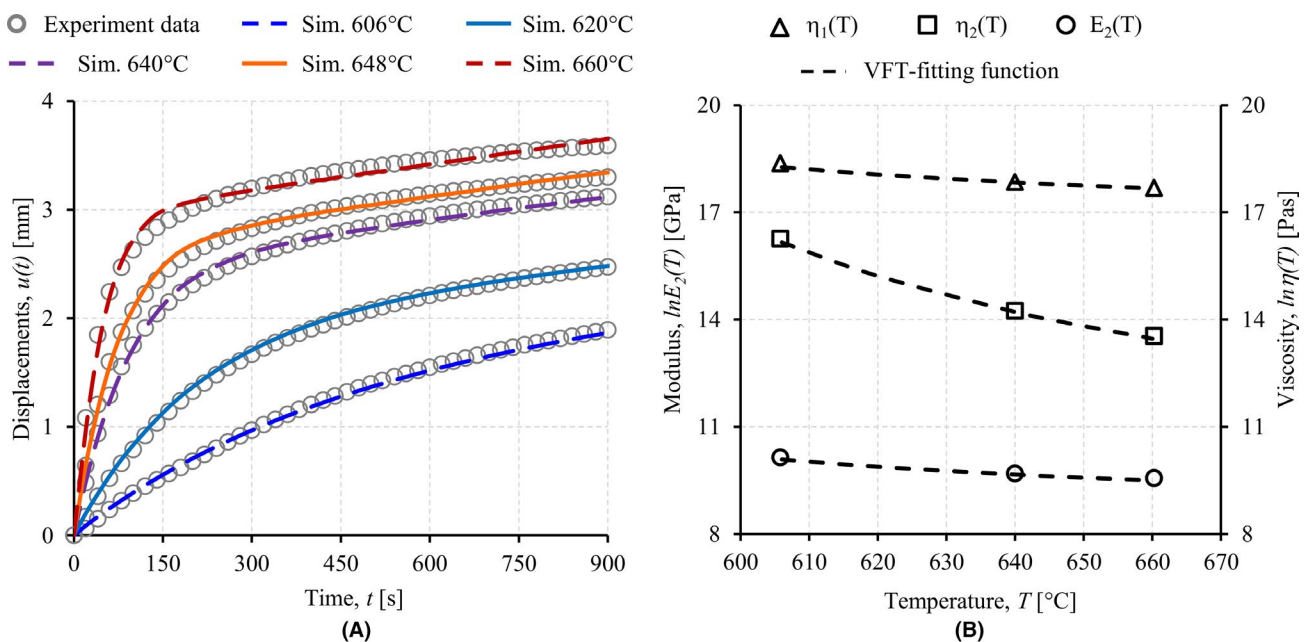


FIGURE 12 A, Validations of Xensation 3D glass; and (B) temperature-dependent Burgers parameters [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com/doi/10.1111/jace.16963)]

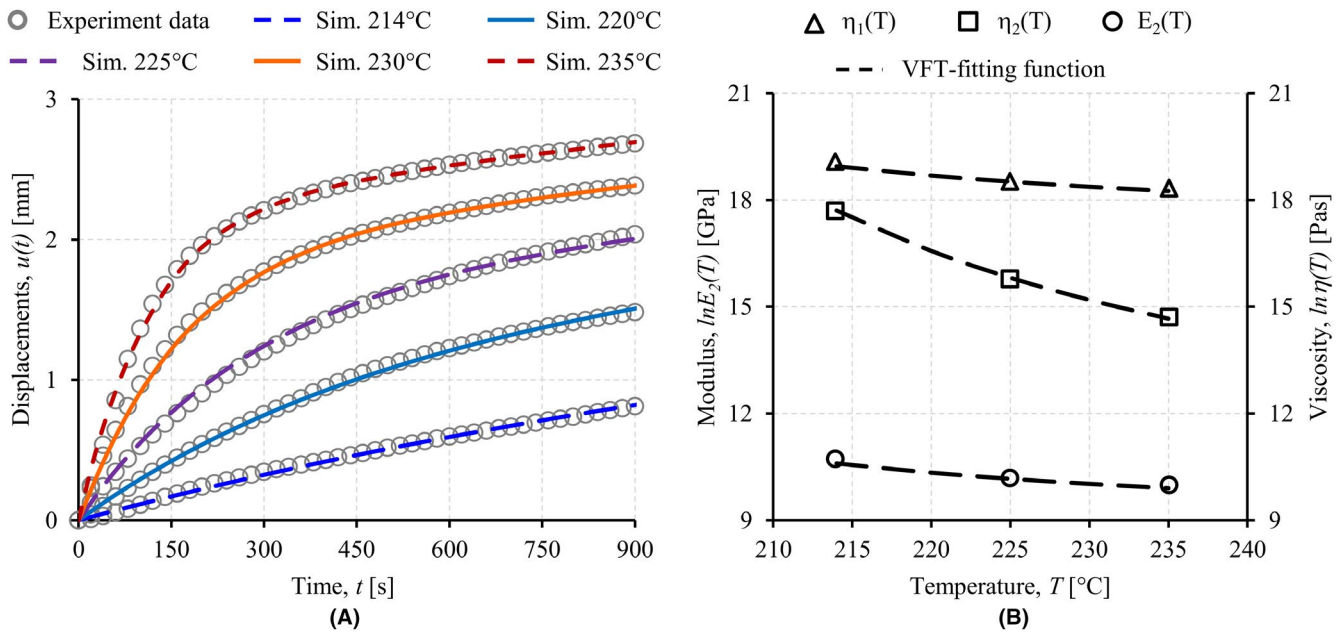


FIGURE 13 A, Validations of IRG 27 glass; and (B) temperature-dependent Burgers parameters [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

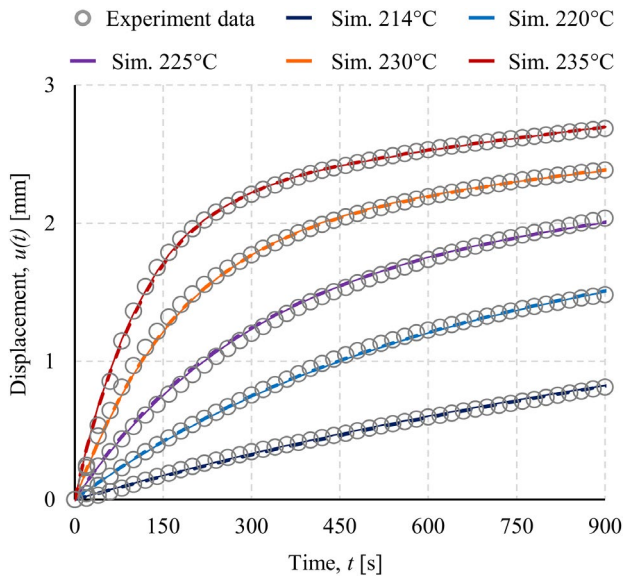


FIGURE 14 Comparison of MYEGA (solid line) and VFT model (dashed line) for IRG 27 glass [Color figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

viscosity models needs to further examine a broader temperature range, particularly for temperatures near T_g and below.

Finally, the methodology is independent from the choice of material models defined by simulation users. For example, the generalized Maxwell model with multiple Maxwell terms $n \geq 3$ are commonly used in many previous studies. We emphasize that the same modeling procedure can be truly adopted for other user-defined material models.

5 | CONCLUSIONS AND FUTURE RESEARCH

An accurate model to characterize the thermo-viscoelastic properties is among the most successful keys for the simulation of the PGM process. In this paper, present restrictions on the modeling of the viscoelastic material behavior have been resolved. For the simulation of the viscoelastic responses, a correct implementation of the relaxation parameters derived from the creep experiment data is elucidated. The finding gets rid of the mismatch between the simulation and experiment data of the creep displacements. For modeling the temperature-dependent viscoelasticity, a detailed examination reveals the deficiency of the TRS assumption over a wide temperature range of glass molding. To overcome this deficit, the paper presents a novel method for modeling the thermo-viscoelastic material behavior of glass. The proposed method permits a determination of the thermo-viscoelastic parameters without using the relaxation functions and shift factors. The constitutive Burgers model demonstrates its sufficient representations of the viscoelastic responses over the range of molding temperature chosen in this study. Temperature is incorporated into each Burgers parameters in the form of VFT function to account for the temperature-dependent viscoelasticity. The method illustrates that a single set of five coefficients satisfactorily characterizes the thermo-viscoelastic properties of the glass. By multiple curve fitting of the creep displacement data, the parameter set is achieved. Using this single set, the viscoelastic responses, either creep or stress

relaxation, at any temperature within the glass molding range can be directly determined. Thanks to the mathematically equivalent advantage of the Burger model and the two-term Prony series, the five temperature-dependent viscoelastic parameters are implemented in the FEM simulation tools via UMAT, using the hereditary integral approach in a straightforward procedure. The validity of the proposed method is steadily solidified by excellent agreements between the experiments and simulation results of three different glass types in a broad temperature range.

To widen the applications of our modeling method, future work will examine the validity of fitting models in a broader temperature range of glass transition and below. In this context, we will focus on the implementation of nonequilibrium viscosity models, such as the Mauro-Allan-Potuzak model,⁴⁴ in order to incorporate thermal history and thermo-viscoelastic properties into a single constitutive model of glass materials.

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APPENDIX A

MATHEMATICAL EQUIVALENCE BETWEEN THE BURGERS MODEL AND THE TWO-TERM PRONY SERIES

Constitutive equation of the Burgers model in the differential form is given as (Findley et al²⁹):

$$\sigma + \left(\frac{\eta_1}{E_1} + \frac{\eta_1}{E_2} + \frac{\eta_2}{E_2} \right) \dot{\sigma} + \frac{\eta_1 \eta_2}{E_1 E_2} \ddot{\sigma} = \eta_1 \dot{\epsilon} + \frac{\eta_1 \eta_2}{E_2} \ddot{\epsilon}. \quad (\text{A1})$$

Using Laplace transformation, the solution to creep and stress relaxation responses can be obtained:

$$\epsilon(t) = \frac{\sigma_0}{E_1} + \frac{\sigma_0}{\eta_1} t + \frac{\sigma_0}{\eta_2} \left(1 - \exp \left(-E_2 \cdot t / \eta_2 \right) \right), \quad (\text{A2})$$

and

$$\sigma(t) = \frac{\epsilon_0}{A} \left[\left(q_1 - \frac{q_2}{\tau_1} \right) e^{-t/\tau_1} - \left(q_1 - \frac{q_2}{\tau_2} \right) e^{-t/\tau_2} \right], \quad (\text{A3})$$

where q_1, q_2 , and $\bar{A}$ are the parameters defined as follows:

$$q_1 = 2\eta_1; q_2 = 2 \frac{\eta_1 \eta_2}{E_2}; \bar{A} = \sqrt{p_1^2 - 4p_2}, \quad (\text{A4})$$

with

$$p_1 = \left(\frac{\eta_1}{E_1} + \frac{\eta_1}{E_2} + \frac{\eta_2}{E_2} \right); p_2 = \frac{\eta_1 \eta_2}{E_1 E_2}, \quad (\text{A5})$$

and τ_1, τ_2 are the relaxation times, given as follows:

$$\tau_1 = \frac{2p_2}{(p_1 - \bar{A})}; \tau_2 = \frac{2p_2}{(p_1 + \bar{A})}. \quad (\text{A6})$$

Following, shear parameters of the Burgers model are calculated by using Lamé constants:

$$G_1 = \frac{E_1}{2(1+\nu)}, G_2 = \frac{E_2}{2(1+\nu)}, \mu_1 = \frac{\eta_1}{2(1+\nu)}, \mu_2 = \frac{\eta_2}{2(1+\nu)}. \quad (\text{A7})$$

By inserting Equation (A7) into Equation (A3), the shear relaxation modulus is gained:

$$G(t) = \frac{G_1 \tau_1 \tau_2}{(\tau_1 - \tau_2)} \left[\left(\frac{G_2}{\mu_2} - \frac{1}{\tau_1} \right) e^{-t/\tau_1} + \left(\frac{1}{\tau_2} - \frac{G_2}{\mu_2} \right) e^{-t/\tau_2} \right]. \quad (\text{A8})$$

Let us define two new parameters g_1, g_2 , called weight factors:

$$g_1 = \frac{\tau_1 \tau_2}{(\tau_1 - \tau_2)} \left(\frac{G_2}{\mu_2} - \frac{1}{\tau_1} \right), g_2 = \frac{\tau_1 \tau_2}{(\tau_1 - \tau_2)} \left(\frac{1}{\tau_2} - \frac{G_2}{\mu_2} \right). \quad (\text{A9})$$

Then, Equation (A8) is expressed in a compact form:

$$G(t) = G_1 \left[g_1 \cdot e^{-t/\tau_1} + g_2 \cdot e^{-t/\tau_2} \right]. \quad (\text{A10})$$

Easily to realize that $g_1 + g_2 = 1$. Therefore, the shear relaxation modulus of the Burgers model (Equation A10) is mathematically equivalent to the two-term Prony series.

APPENDIX B

COMPUTATIONAL PROCEDURE FOR THE DETERMINATION OF TEMPERATURE-DEPENDENT VISCOELASTIC PARAMETERS

FIGURE B1 Flowchart of the computational procedure for the determination of temperature-dependent viscoelastic parameters

