Model Description of the Viscosity of Glass-Forming Chalcogenides

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➢Introduction

Understand the viscosity behavior is of primordial importance in the processing of glass-forming materials. Concerning the temperature dependence of the viscosity, many theories and models have been proposed till now [1]. Some years ago, we proposed the Bond Strength-Coordination Number Fluctuation (BSCNF) model [2, 3]. This model describes the temperature dependence of the viscosity or relaxation time in terms of the mean bond strength, mean coordination number and their fluctuations of the structural units that form the melt. In the past, this model was applied to analyze the viscosity of different type of materials. An example is shown in Fig. 1.

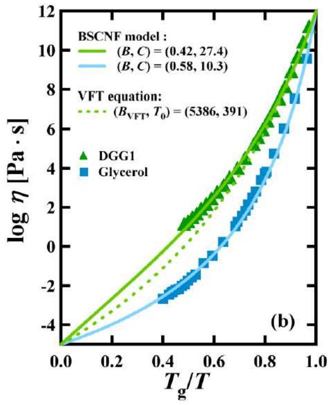

Fig. 1 Example of application of the BSCNF model [3].

Recently, a modified version of the BSCNF model was applied to analyze the unusual temperature dependence of the viscosity observed in some metallic glass-forming systems [4,5]. The result is shown in Fig. 2 (a). In the modified version, the description is done by controlling the average coordination number between the structural units.

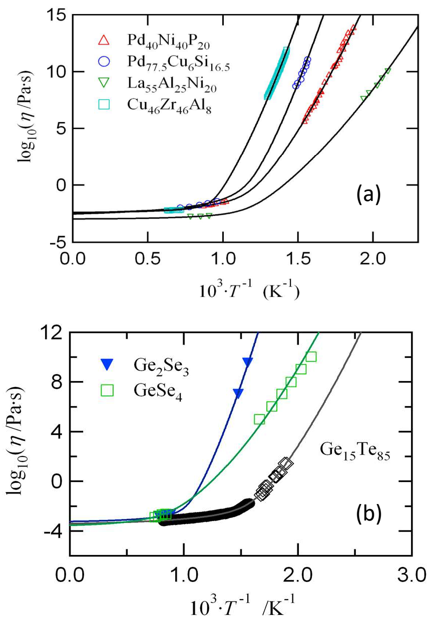

Fig. 2 Viscosity of metallic and chalcogenide glass forming systems described by the modified BSCNF model [4,5].

Interestingly, analogous viscosity pattern have been also reported in chalcogenide glass forming systems [6]. The description based on the modified BSCNF is shown in Fig. 2 (b) [4]. The pattern shown in Fig. 2, can not be described by the well known equations, for instance the VFT expression.

➢Model and Result

In the present study, a different approach to describe the temperature dependence of the viscosity of chalcogenide systems is presented. Recent studies on fragile-to-strong transition indicate that the phenomenon is related with some kind of structural transformation [6,7]. We incorporate this information into our BSCNF model by assuming that the interaction between the structural units changes at a temperature T^*. According to the BSCNF model, the temperature dependence of the viscosity is written as [2,3]

$$\ln\left(\frac{\eta}{\eta_0}\right)=\frac{C\left(\frac{T_g}{T}\right)+C\left(\frac{T_g}{T}\right)^2\left\{\left[\ln\left(\frac{\eta_{T_g}}{\eta_0}\right)+\frac{1}{2}\ln(1-B)\right]\frac{(1-B)}{C}-1\right\}}{1-B\left(\frac{T_g}{T}\right)^2}-\frac{1}{2}\ln\left[1-B\left(\frac{T_g}{T}\right)^2\right]$$

$$C=\frac{2\gamma(1-B)}{2\gamma+\sqrt{B}(1+\gamma^2)}\left[\ln\left(\frac{\eta_{T_g}}{\eta_0}\right)+\frac{1}{2}\ln(1-B)\right]\qquad \gamma=\frac{|\Delta E|/E_0}{|\Delta Z|/Z_0}\qquad B=\frac{(\Delta Z)^2(\Delta E)^2}{R^2T_g^2}$$

$E_0, \Delta E$: mean value and fluctuation of energy of the structural units

$Z_0, \Delta Z$: mean value and fluctuation of coordination number between the structural units

η_{T_g}, η_0 : viscosity at T_g and at the high temperature limit

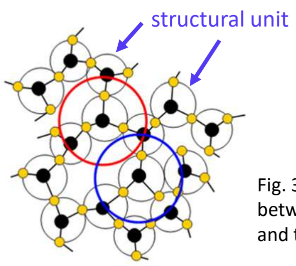

Fig. 3 Schematic representation of the model. The interconnection between the structural units are described by the energy $E = E_0 + \Delta E$ and the coordination number $Z = Z_0 + \Delta Z$.

In the description we adopt the values $T_g/T^* = 0.6$ and $\gamma = 1$ irrespective of the materials. The result of the model is illustrated in Fig. 4 for the case of $Ge_{15}Te_{85}$ and As_2Se_3. The result indicates that if we know the value of $\ln\left(\eta_{T_g}/\eta_0\right)$, only 1 free parameter (B at T_g) suffices to describe the data of As_2Se_3. On the other hand, for the case of $Ge_{15}Te_{85}$, we need 2 free parameters (B at T_g and at the high temperature limit). It should be noted that B has a clear physical meaning, that is, it reflects the fluctuation of the connections between the structural units.

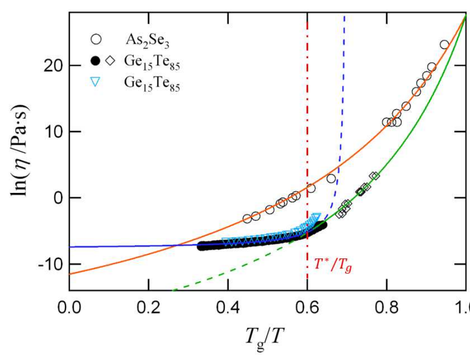

Fig. 4 Temperature dependence of the viscosity in As_2Se_3 and $Ge_{15}Te_{85}$. Experimental data are taken from [6,7] and the references therein.

➢Conclusion

- A model that is able to describe the unusual temperature dependence of the viscosity has been proposed.
- The model indicates that the temperature dependence of the viscosity of some materials such as $Ge_{15}Te_{85}$ can be described by considering that the interconnection between the structural units changes at a certain temperature T^*. Probably, such change is related with the fragile-to-strong transition.

➢References

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