

Understanding the Vogel-Fulcher-Tammann law in terms of the bond strength-coordination number fluctuation model

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It is shown that an expression similar to the Vogel-Fulcher-Tammann (VFT) equation of viscosity can be derived from the bond strength-coordination number fluctuation (BSCNF) model. The parameters B_{VFT} and T_0 of the VFT equation, $\eta = \eta_0 \exp[B_{\text{VFT}}/(T - T_0)]$, are related directly to the parameters of the BSCNF model; $B_{\text{VFT}} \cong E_0 Z_0 / R$ and $T_0 \cong |\Delta E| |\Delta Z| / R$, where E_0 and Z_0 are the mean values of the bond strength and the coordination number of the structural units, and ΔE and ΔZ are their fluctuations and R is the gas constant. According to the BSCNF model, the viscous flow occurs by breaking or twisting the bonds that connect the structural units of the melt. From the analytical expression obtained in the present study, an expression for the strength parameter D can be also derived. Namely, D is written as $D = B_{\text{VFT}} / T_0 \cong (E_0 Z_0) / (|\Delta E| |\Delta Z|)$. This relation indicates that D includes information of the fluctuations that characterizes the disorder of glass-forming materials.

Keywords: VFT Equation; Supercooled Liquids; Viscosity; Fragility

1. Introduction

The clarification of the mechanism of glass transition and structural relaxation in supercooled liquids is one of the most challenging and intriguing subjects in condensed matter physics [1,2]. Usually, when a liquid is cooled fast enough to avoid its crystallization, the viscosity η or the relaxation time τ of the liquid increases drastically until the glass transition occurs. During the cooling process, it is believed that the size of cooperativity or heterogeneity grows as well [3,4]. The relaxation occurring during this process is usually called α -relaxation. One of the most frequently used expressions to describe the non-Arrhenius temperature dependence in the α -relaxation is the Vogel-Fulcher-Tammann (VFT) equation [5-7]. The VFT expression of the viscosity is given by

$$\eta = \eta_0 \exp\left(\frac{B_{\text{VFT}}}{T - T_0}\right) \quad (1)$$

where T is the absolute temperature, η_0 is the value of the viscosity at the high temperature limit, and B_{VFT} is a quantity related with the activation barrier. T_0 is the so called "Vogel temperature" or "ideal glass transition temperature" at which the viscosity or the relaxation time diverges, or the configurational entropy of the supercooled liquid vanishes [1]. The relation between T_0 and the Kauzmann temperature T_K [8] has been controversial [1,9,10], since they are related with the kinetics and the thermodynamics of supercooled liquids.

The VFT expression has been used widely as a convenient tool to analyze the experimental data of the temperature dependence of the transport coefficients such as viscosity, diffusion coefficient, electric conductivity [11-13]. It has three adjustable parameters. For instance, η_0 , B_{VFT} and T_0 in the case of Eq. (1). However, the physical origin of the VFT-behavior has not been fully understood even nowadays. For instance, it has been

discussed [14] that the diverging behavior of the VFT equation, particularly for fragile liquids, is not reflected in the actual measurements of the structural relaxation and the glass transition.

In our previous studies, a model for the viscous flow of the melt has been proposed. The model which is called bond strength-coordination number fluctuation (BSCNF) model [15-17] describes the temperature dependence of the viscosity that extends from strong to fragile glass-forming liquids in terms of the mean bond strength E_0 , the mean coordination number Z_0 , and the fluctuations, ΔE , ΔZ , of the structural units that form the melt. According to this model, the viscosity is written as [15]

$$\eta = \frac{\eta_0}{\sqrt{1-Bx^2}} \exp \left[\frac{Cx + Cx^2 \left[\left\{ \ln \left(\frac{\eta_{T_g}}{\eta_0} \right) + \frac{1}{2} \ln(1-B) \right\} \frac{(1-B)-1}{C} \right]}{1-Bx^2} \right], \quad (2)$$

where

$$B = \frac{(\Delta E)^2 (\Delta Z)^2}{R^2 T_g^2} \quad \text{and} \quad C = \frac{E_0 Z_0}{RT_g}. \quad (3)$$

Here, x is given by $x=T_g/T$, where T_g is the glass transition temperature. η_{T_g} is the value of the viscosity at T_g and $R=8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ is the gas constant. Recently, it has been shown [17] that the BSCNF model reproduces the experimental data better than the VFT equation. One reason for such a success is that the BSCNF model given in Eq. (2) has four adjustable parameters, namely, η_0 , η_{T_g} , B and C . However, once the value of viscosity at T_g is fixed as $\eta_{T_g}=10^{12} \text{ Pa} \cdot \text{s}$ [2,18] (or the relaxation time τ at T_g can be also fixed as $\tau_{T_g}=10^2 \text{ s}$ [11]), the adjustable parameters of the BSCNF model reduces to η_0 , B and C , which is the same to the number of fitting parameters used in the VFT equation. Furthermore, if the usual value $\eta_0 =10^{-5} \text{ Pa} \cdot \text{s}$ [2,19] (or $\tau_0=10^{-14} \text{ s}$ [11,14]) is adopted, the number of fitting parameters reduces

further to two. The BSCNF model based on two adjustable parameters shows also a reasonable agreement with the experimental values. The application of the BSCNF model to various kinds of glass-forming liquids has provided new viewpoints to the structural relaxation in addition to the notions provided by the traditional and other models such as the Adam-Gibbs (AG) model [20], the free volume theory [13,21], the Avramov-Milchev (AM) model [22,23], and the Mauro-Yue-Ellison-Gupta-Allan (MYEGA) equation [24], etc. For instance, it has been shown [25] that the quantity N_B which is defined in the framework of the BSCNF model is closely related with the notion of cooperatively rearranging region (CRR) in the theory of Adam and Gibbs. The details of the BSCNF model are discussed in our previous works [15-17].

The main purpose of the present paper is to show that an expression similar to the VFT equation can be derived analytically from the BSCNF model. In a previous study, it was found empirically [17] that under certain condition, the viscosity behavior described by the BSCNF model becomes almost the same to that described by the VFT equation. However, the theoretical foundation that relates the BSCNF model to the VFT equation was not clear. In this paper, the background of such a relation is clarified. As will be shown later, the obtained expression enables one to discuss the physical meanings of the parameters B_{VFT} and T_0 from a point of view that differs from the models proposed by others. The interrelation obtained here gives also another approximated expression for the strength parameter $D=B_{VFT}/T_0$ by using the parameters of the BSCNF model, E_0 , Z_0 , ΔE and ΔZ . In this paper, the implications of these parameters to D are also discussed.

2. VFT-like expression derived from the BSCNF model

The BSCNF model was introduced based on the physical picture that the viscous flow of the melt occurs by breaking or twisting the bonds between the structural units [15]. Within the framework of the BSCNF model, the following relationship between the parameters of the BSCNF model and the shift factor, $\ln(\eta_{T_g}/\eta_0)$, holds at T_g [15,25],

$$\frac{1}{2RT_g} \left\{ Z_0 \frac{(\Delta E)^2}{E_0} + E_0 \frac{(\Delta Z)^2}{Z_0} \right\} = \left\{ \ln \left(\frac{\eta_{T_g}}{\eta_0} \right) + \frac{1}{2} \ln(1-B) \right\} \frac{(1-B)}{C} - 1. \quad (4)$$

Using Eq. (4), the BSCNF model expressed by Eq. (2) can be rewritten as

$$\eta = \frac{\eta_0}{\sqrt{1-Bx^2}} \exp \left[\frac{Cx + Cx^2 \left(\frac{1+\gamma^2}{2\gamma} \sqrt{B} \right)}{1-Bx^2} \right], \quad (5)$$

where γ is the ratio between the fluctuations normalized by their mean values,

$$\gamma = \frac{|\Delta E|/E_0}{|\Delta Z|/Z_0}. \quad (6)$$

In a previous study we have noted that in the case where $\gamma=1$, the viscosity behavior described by the BSCNF model becomes indistinguishable from that of the VFT equation [17]. From a physical point of view, $\gamma=1$ gives the equality between the normalized fluctuations of the bond strength and the coordination number of the structural units. For the case where this condition is violated (i.e., $\gamma \neq 1$), the BSCNF model reproduces the experimental data of fragile liquids better than the VFT equation (See, ref. [17]).

When $\gamma=1$, Eq. (5) becomes

$$\eta = \frac{\eta_0 T}{\sqrt{T^2 - T_F^2}} \exp \left(\frac{T_D}{T - T_F} \right) \quad (7)$$

where

$$T_D = \frac{E_0 Z_0}{R} \quad \text{and} \quad T_F = \frac{|\Delta E| |\Delta Z|}{R}. \quad (8)$$

We note that Eq. (7) is an expression similar to the VFT equation, except for the pre-exponential part. In Table 1, the values of B_{VFT} , T_0 , T_D and T_F for the glass-forming materials investigated in the present study are given. These values have been obtained by fitting the experimental data by both, the VFT equation and the BSCNF model. The materials cover various types of glass-formers such as inorganic [18,19,26-28], organic [29], metallic [30], ionic [29,31] and polymers [32]. Other material parameters such as T_g , the fragility index m and the value of $\ln(\eta_{T_g}/\eta_0)$, are also given in Table 1.

Figure 1 shows a direct comparison between the values of B_{VFT} and T_D for the materials listed in Table 1. Meanwhile, Fig. 2 shows a comparison between T_0 and T_F . From both figures, we can see that B_{VFT} and T_0 correspond to T_D and T_F , respectively:

$$B_{\text{VFT}} \cong T_D \quad \text{and} \quad T_0 \cong T_F. \quad (9)$$

From Fig. 1, we recognize a slight deviation from the straight line given by $B_{\text{VFT}} = T_D$ in fragile systems. The relative deviation between B_{VFT} and T_D evaluated as $\varepsilon_D = |T_D - B_{\text{VFT}}| / B_{\text{VFT}} \times 100$ is within approximately 3% for all the glass-forming materials investigated. On the other hand, we can see from Fig. 2 that the value of T_F is almost equal to the value of T_0 for the glass-forming materials ranging from strong to fragile liquids. The relative deviation $\varepsilon_F = |T_F - T_0| / T_0 \times 100$ is within 1%, which is smaller compared with the values of ε_D .

Concerning the physical meanings to these parameters, the relations given in Eq. (9) provide a clear interpretation. B_{VFT} corresponds to the mean bond strength per structural unit, $B_{\text{VFT}} \cong E_0 Z_0 / R$, and the Vogel temperature T_0 is related directly to the fluctuations of the bond strength and the coordination number of the structural units, $T_0 \cong |\Delta E| |\Delta Z| / R$. Recently, Hecksher *et al.* [14] have insisted that there is no any indication of divergence in a finite

temperature range as suggested by the VFT equation. On the other hand, the relation given by $T_0 \cong |\Delta E||\Delta Z|/R$ suggests that the non-Arrhenius temperature dependence described by the VFT equation arises from the fluctuations of the binding energy and coordination number of the structural units. From the above discussion, Eq. (9) provides a clue to understand the physical background concerning the origin of the deviation from the Arrhenius temperature dependence, which has been the subject of debate for many years. Concerning the relation $B_{\text{VFT}} \cong E_0 Z_0/R$, it also gives a physically sound picture to the effective activation energy or the pseudoactivation energy RB_{VFT} [33].

As mentioned above, a distinct difference between the VFT equation and the VFT-like expression given by Eq. (7) lies in their pre-exponential parts. To compare directly the VFT-like expression derived here with the VFT equation, Eq. (7) is rewritten as

$$\eta_{\text{BSCNF}} = \eta_0 \exp\left(\frac{\beta}{T - T_F}\right) \quad (10)$$

where

$$\beta = T_D - \frac{T - T_F}{2} \ln\left\{1 - \left(\frac{T_F}{T}\right)^2\right\}. \quad (11)$$

Besides, we express Eq. (10) as follows,

$$\eta_{\text{BSCNF}} = \eta_0 \exp\left(\frac{B_{\text{VFT}}}{T - T_0} \delta\right) \quad (12)$$

where

$$\delta = \frac{\beta(T - T_0)}{B_{\text{VFT}}(T - T_F)} \approx \frac{\beta}{B_{\text{VFT}}}. \quad (13)$$

In the approximation of Eq. (13), we have used the relation given in Eq. (9), $T_0 \cong T_F$. Fig. 3 shows the behavior of δ plotted as a function of T_g/T for the materials, GeO₂ (No.2), As₂Se₃ (No.6), Sorbitol (No.15), TPG (No.17) and DPG (No.18). It is noted in Fig. 3 that $\delta \approx 1$ holds

over the whole temperature range. That is, $\beta \approx B_{\text{VFT}}$. To be more precise, Fig. 3 indicates that for the fragile systems a slight deviation from the line $\delta = 1$ is observed in the high temperature part. However, this does not affect the main result obtained above.

As shown in Fig. 4, the VFT-like expression of viscosity given by Eq. (7) (or Eq. (12)) is indistinguishable from the behavior of the VFT equation. In Fig. 4, it is shown that the experimental data of the viscosity and the relaxation time are fitted well by both, Eq. (1) and Eq. (7). As we can see, the behavior of the VFT-like expression derived here from the BSCNF model behaves almost similarly to that described by the VFT equation. Hence, Eq. (12) can be approximated as $\eta_{\text{BSCNF}} \approx \eta_0 \exp[B_{\text{VFT}}/(T - T_0)]$, that is, the behavior of Eq. (7) behaves quite similarly to that of the VFT equation given in Eq. (1). This gives a theoretical background that the VFT law can be derived from the BSCNF model.

In the literature, different expressions for the VFT-like equations have been employed. For instance, the expression for the viscosity given as $\eta = A_\eta T^{1/2} \exp[B_\eta/(T - C_\eta)]$ [32,34], or for the ionic conductivity given as $\sigma = A_\sigma T^{-1/2} \exp[-B_\sigma/(T - C_\sigma)]$ [35,36], among others, have been used. Here, $A_{\eta(\sigma)}$, $B_{\eta(\sigma)}$, and $C_{\eta(\sigma)}$ are constants depending on materials and are used as fitting parameters to the experimental data. These expressions and the VFT-like expression given by Eq. (7) have a common property, that is, the temperature-dependent term is contained in their pre-exponential parts. However, the present study suggests that irrespective of the difference in the pre-exponential parts, the VFT-behavior or the diverging temperature dependence is mainly determined by the exponential term. Now, we have seen above that the diverging temperature T_0 is almost equal to T_F , which carries information on the fluctuations. The situation does not change if other expressions for the transport coefficients are used. That is, C_η and C_σ are also related with fluctuations.

3. Implications to the strength parameter D

The VFT equation expressed as $\eta = \eta_0 \exp[DT_0/(T - T_0)]$ has been also often used in the analysis of experimental data [2,9,37-41], where $D=B_{\text{VFT}}/T_0$ is the so called “strength parameter” or “fragility parameter” [38-41]. According to Eq. (8) and Eq. (9), D can be rewritten as

$$D \cong D_{\text{F}}, \quad (14)$$

where

$$D_{\text{F}} = \frac{E_0 Z_0}{|\Delta E| |\Delta Z|}. \quad (15)$$

Equation (15) gives a new interpretation to D , that is, it gives the ratio of the mean binding energy per structural unit to their fluctuations. It is known that the value of D classifies conveniently the glass-forming materials into strong and fragile systems [38-41]. For the strong liquids, it takes a large value (typically $D \geq 100$). While for the fragile liquids it takes a small value ($D < 10$). In a paper by Hodge [42], it has been pointed out that in the case of Arrhenius limit (i.e., $T_0 \rightarrow 0$), the value of D must be infinite, since B_{VFT} takes a finite value. However, a small finite value of T_0 compared with T_g has been observed even for the strong systems such as SiO_2 [19,23,40]. On the other hand, Eq. (15) suggests that the value of D should have a finite value, and indicates also that D reflects the fluctuations of the bond strength and the coordination number of the structural units that exist in glass-forming materials. In other words, the finite value of the fluctuations incorporated in D characterizes the structural disorder of glassy materials.

Note that Eq. (15) is similar to the expression obtained by Vilgis [43], $D=(z_0/\Delta z)^2/4$. Here, z_0 and Δz are the average value and the fluctuation of the coordination number of the

atoms [43,44]. In the BSCNF model, the coordination number Z is a quantity related with the coordination between the structural units.

Here, we discuss briefly a potential usability of Eq. (15). It was shown above that in the case of $\gamma=1$ or $|\Delta E|/E_0=|\Delta Z|/Z_0$, the temperature dependence of viscosity or relaxation time described by the BSCNF model and the VFT equation behave similarly. Thus, for this case we obtain from Eq. (15)

$$\frac{|\Delta E|}{E_0} = \frac{|\Delta Z|}{Z_0} = \frac{1}{\sqrt{D_F}}. \quad (16)$$

It is a theoretical relation which provides a tool to obtain additional information to the conventional strength parameter. The value of D is usually determined by fitting the measured data with the VFT equation. However, not all the data can be reproduced well with the VFT equation. In such a case, it is probable that the equality given in Eq. (16) is violated, i.e., $|\Delta E|/E_0 \neq |\Delta Z|/Z_0$. In one of our studies [45], we have discussed the bonding nature of chalcogenide semiconducting glasses such as Ge-S and Ge-Sb-S(Se) by comparing with other studies by Tanaka [46,47] and Tichý and Tichá [48]. The studies have provided clues to understand the degree of network connectivity and the dimensionality reflected in the medium range structure of glassy materials [46,49]. It has been discussed in ref. [45] that the information of the structure in the chalcogenide systems Ge-S and Ge-Sb-S(Se) can be obtained from the analysis of the viscosity based on the BSCNF model. The analysis has indicated that for these systems, we have $\gamma > 1$ by considering their composition dependence. This result suggested that the chalcogenide glassy semiconductors are possibly characterized by $|\Delta E|/E_0 > |\Delta Z|/Z_0$.

In the present formulation of the model, the quantities E_0 , Z_0 , ΔE and ΔZ are considered to be temperature independent. This assumption has been adopted to avoid the increase of free

parameters and to investigate the extent that the present model can reproduce the observed behavior of the viscosity ranging from strong to fragile liquids. Other theoretical models such as the AM model [22,23] and the MYEGA equation [24,29] use also temperature independent parameters. It should be pointed out, however, that in principle, the quantities used in the BSCNF model could depend on temperature. The effect of the temperature on the parameters such as fluctuations, ΔE and ΔZ , is expected to be weak in strong systems. In fragile systems, in contrast, its role is expected to increase. In the present paper, however, the effect of temperature on the parameters is not considered.

4. Conclusions

In the present study, it was shown that an expression similar to the VFT equation can be derived analytically from the BSCNF model. The derived analytical expression was slightly different in the pre-exponential part compared with the original VFT expression, while the parameters of the VFT equation, B_{VFT} and T_0 , were almost equal to $E_0 Z_0 / R$ and $|\Delta E| |\Delta Z| / R$, respectively. It was confirmed that the behaviors of the BSCNF model and the VFT equation behave almost similarly under the condition of $\gamma=1$, i.e., $|\Delta E| / E_0 = |\Delta Z| / Z_0$. In this regard, the analytical relationships given in the present study provide a theoretical background to the relation that connects the VFT equation and the BSCNF model. In addition, the relation derived in the present study enables one to represent also the strength parameter, $D = B_{\text{VFT}} / T_0$, in terms of the parameters of the BSCNF model, $D \approx D_F = (E_0 Z_0) / (|\Delta E| |\Delta Z|)$. This relation predicts that for the glass-forming materials whose viscosity data are not reproduced well with the VFT equation, the equality between the normalized bond strength and coordination number of the structural units is violated, $|\Delta E| / E_0 \neq |\Delta Z| / Z_0$. This is a theoretical relation that

suggests the link between the microscopic structural connectivity and the macroscopic viscosity parameters of glass-forming materials.

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Figure Captions

Fig. 1: Comparison between the values of B_{VFT} and T_{D} given in Eq. (8). The straight line is given by $B_{\text{VFT}} = T_{\text{D}}$. The relative deviation calculated by $\varepsilon_{\text{D}} = |T_{\text{D}} - B_{\text{VFT}}| / B_{\text{VFT}} \times 100$ is within approximately 3% for all the glass-forming materials investigated.

Fig. 2: Comparison between the values of T_0 and T_{F} given in Eq. (8). The straight line is given by $T_0 = T_{\text{F}}$. The relative deviation calculated by $\varepsilon_{\text{F}} = |T_{\text{F}} - T_0| / T_0 \times 100$ is within 1% for all the materials.

Fig. 3: Temperature dependence of δ defined in Eq. (13). The inset shows an enlarged view.

Fig. 4: Temperature dependence of the viscosity for GeO_2 [26], As_2Se_3 [27], and of the relaxation time [29] for Sorbitol, Tripropylene glycol (TPG), and Dipropylene glycol (DPG). The experimental data are taken from the references indicated. The materials investigated here are the same to those shown in Fig. 3. Note that the curves calculated by the VFT equation and the BSCNF model behave similarly and are indistinguishable.

Table Caption

Table 1: Parameters of the glass-forming materials investigated in the present study. The values of B_{VFT} and T_0 are determined in this study, or some of them are taken from the literature. The values of T_{D} and T_{F} are determined by using the relations given in Eq. (8). The values of $\ln(\eta_{T_{\text{g}}}/\eta_0)$ with * are replaced by those of the relaxation time, $\ln(\tau_{T_{\text{g}}}/\tau_0)$. The fragility index m_{cal} is calculated from the BSCNF model [15,17].

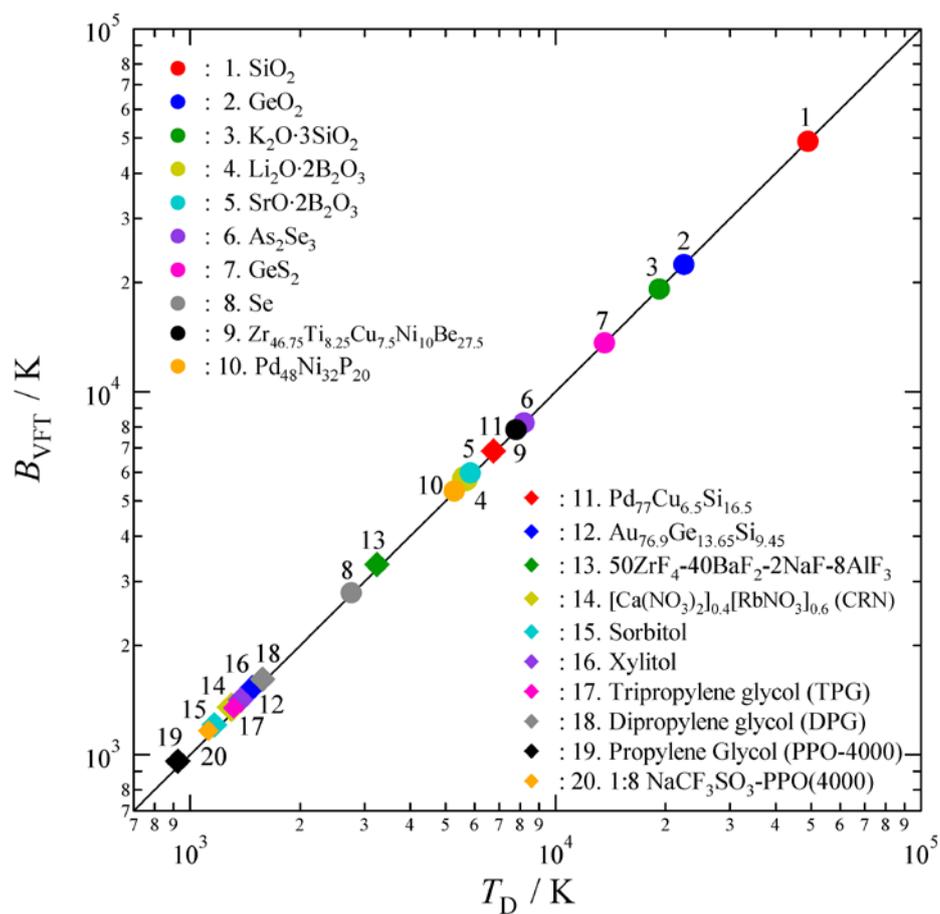


Fig. 1

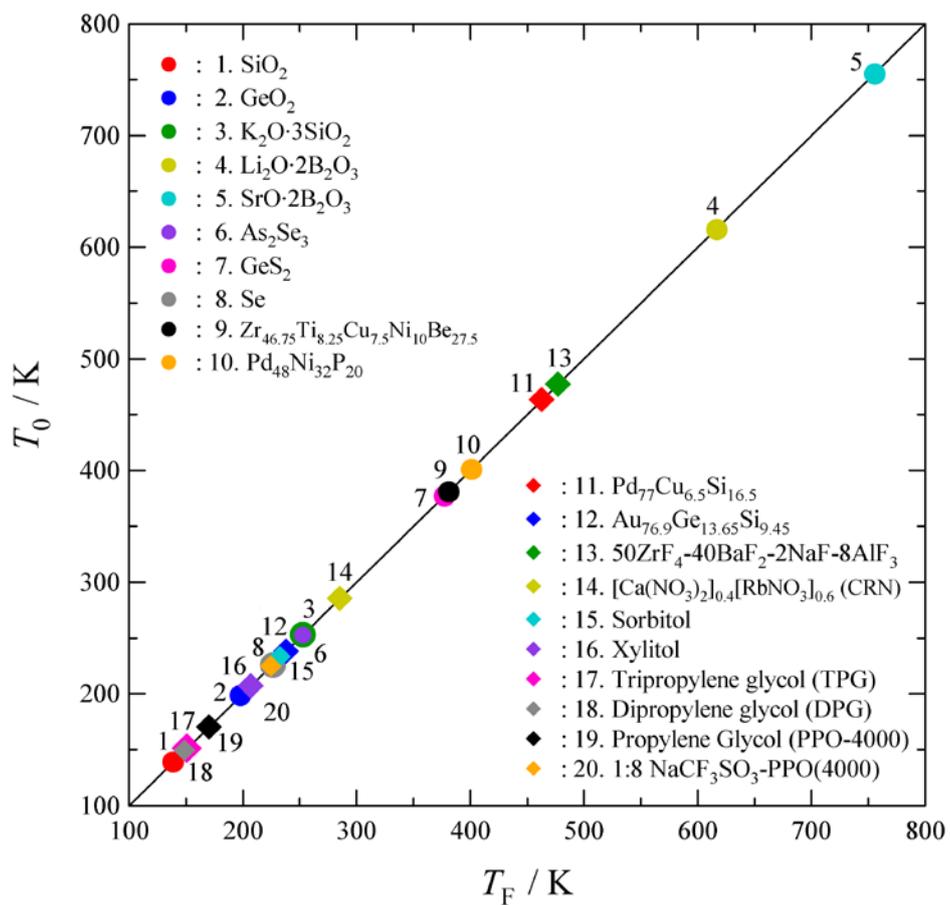


Fig. 2

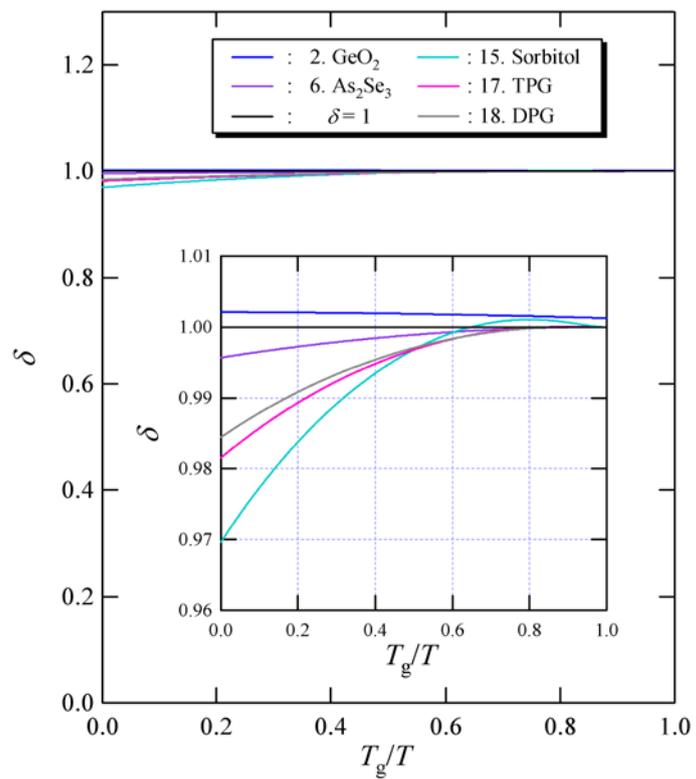


Fig. 3

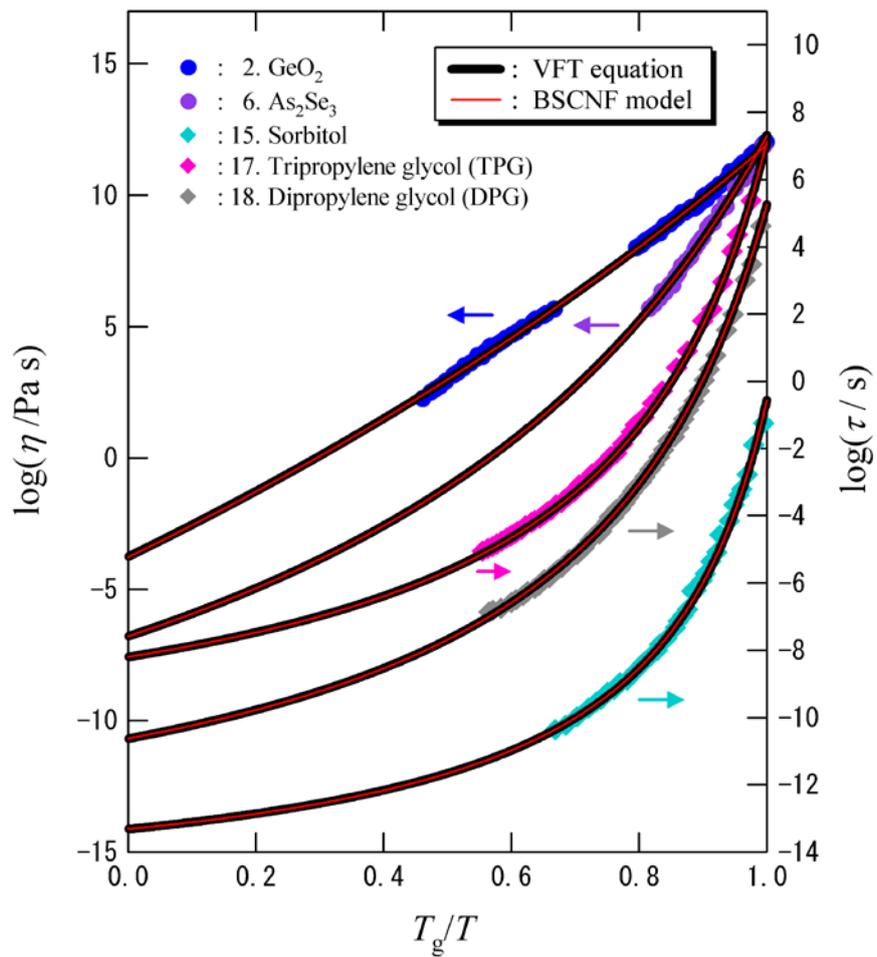


Fig. 4

No.	Glass-forming material	T_g (K)	B_{VFT} (K)	T_0 (K)	$m_{cal.}$	$\ln(\eta_{T_g}/\eta_0)$	T_D (K)	T_F (K)	Ref.
1.	SiO ₂	1450	48939	139	17.9	37.3	48938	139	[19]
2.	GeO ₂	820	22410	199	20.7	36.1	22457	198	[26]
3.	K ₂ O·3SiO ₂	760	19190	253	24.6	37.8	19172	253	[19]
4.	Li ₂ O·2B ₂ O ₃	763	5750	616	88.2	39.1	5647	617	[19]
5.	SrO·2B ₂ O ₃	911	5968	755	97.0	38.3	5846	756	[19]
6.	As ₂ Se ₃	440	8218	253	44.7	43.9	8183	253	[27]
7.	GeS ₂	726	13650	377	35.4	39.1	13605	377	[18]
8.	Se	295	2795	226	74.7	40.4	2757	226	[28]
9.	Zr _{46.75} Ti _{8.25} Cu _{7.5} Ni ₁₀ Be _{27.5}	596	7848	381	43.9	36.5	7792	381	[30]
10.	Pd ₄₈ Ni ₃₂ P ₂₀	566	5333	401	48.2	32.3	5266	401	[30]
11.	Pd ₇₇ Cu _{6.5} Si _{16.5}	620	6852	463	74.9	43.6	6773	463	[30]
12.	Au _{76.9} Ge _{13.65} Si _{9.45}	285	1523	238	85.3	32.4	1485	238	[30]
13.	50ZrF ₄ -40BaF ₂ -2NaF-8AlF ₃	579	3325	477	80.4	32.6	3247	478	[31]
14.	[Ca(NO ₃) ₂] _{0.4} [RbNO ₃] _{0.6} (CRN)	333	1345	285	84.4	28.0 *	1301	285	[29]
15.	Sorbitol	274	1205	233	85.3	29.4 *	1168	233	[29]
16.	Xylitol	248	1410	207	90.3	34.4 *	1376	207	[29]
17.	Tripropylene glycol (TPG)	189	1341	151	76.2	35.3 *	1316	151	[29]
18.	Dipropylene glycol (DPG)	193	1609	149	69.7	36.6 *	1584	149	[29]
19.	Propylene Glycol (PPO-4000)	201	955	170	90.1	31.4	928	171	[32]
20.	1:8 NaCF ₃ SO ₃ -PPO(4000)	262	1158	225	96.0	31.3	1124	225	[32]

Table 1