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Article

On the Use of Generalised Coordinates to Describe the Temperature Dependence of Viscosity and Relaxation Time in the Glass Transition Region

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Abstract

It is shown that the empirical Williams-Landel-Ferry (WLF) and Vogel-Fulcher-Tammann (VFT) formulas, as well as the semiempirical formulas of other researchers, for the viscosity η in the glass transition region are, in fact, hyperbolic functions of temperature with corresponding relationships between their parameters. The hyperbolic dependence can be derived from the universal expansion of $\ln(\eta)$ into a Taylor series in a small temperature parameter near the glass transition temperature. The applicability of the principle of corresponding states follows from this expansion. A new two-parameter formula in the form of a second-degree polynomial is proposed for $\ln(\eta)$ in the glass transition region. This formula contains physically significant parameters and adequately describes the available experimental data for individual glass-forming substances.

Keywords: viscosity; relaxation time; glass transition; Williams–Landel–Ferry (WLF) equation; Vogel–Fulcher–Tammann (VFT) equation; principle of corresponding states; glass transition activation energy

1. Introduction

Viscosity is a property of fundamental importance both for the molten and solid states of materials quantifying the resistance of material to flow and indicates the ability to dissipate momentum [1–3]. For example, in glass technology the viscosity determines melting, working and annealing temperatures, rate of refining, maximum use temperature, and crystallization rate, while in nature it governs the behaviour of Earth mantle and volcanic lava flow rates [4–10]. The viscosity of glass-forming liquids and amorphous solids exhibits a characteristic and often dramatic temperature dependence that has motivated decades of theoretical and experimental investigation. Understanding the microscopic origins of viscous flow is essential not only for fundamental condensed-matter physics but also for the processing, stability, and long-term performance of technological glasses. Early physically motivated descriptions of viscosity focused on the structural rearrangements required for flow in network-forming materials. Mott proposed that viscous flow in vitreous silica is governed by the thermally activated rupture of Si–O bonds, with the number of broken bonds increasing with temperature and enabling local structural rearrangements [11]. This model established the principle that flow in network glasses is controlled by the energetics of bond rupture rather than by collective configurational entropy. A complementary structural interpretation

was introduced earlier by Douglas, who proposed that the bridging oxygen atom between two silicon atoms can occupy two metastable positions separated by an energy barrier [12]. Flow occurs when the Si–O–Si bond breaks, allowing the oxygen to transition between these positions. Douglas’s bistable-oxygen model was one of the first to treat viscous flow as a sequence of discrete, thermally activated configurational transitions, anticipating later developments in energy-landscape theory. Importantly, it framed viscosity as a property emerging from the local topology of the network, rather than from long-range structural order. The role of defects in mediating viscous flow was further rectified by Doremus, who demonstrated that amorphous silica contains SiO molecular defects and other structural irregularities that act as flow units [13]. In this defect-mediated picture, viscosity is governed by the concentration, mobility, and activation energies of such defects, whose formation is itself temperature dependent. Doremus’s work provided a microscopic basis for understanding deviations from simple Arrhenius behaviour and helped explain why even “strong” glasses exhibit subtle non-Arrhenius features near the glass transition. These ideas were unified in the Douglas–Doremus–Ojovan (DDO) model, developed in detail in [14] and expanded to account for the effects of radiation on viscosity in [15,16]. The DDO model integrates Mott’s bond-breaking concept, Douglas’s bistable oxygen positions, and Doremus’s defect-mediated transport into a single theoretical framework. It treats viscous flow as a process controlled by the temperature-dependent concentration of broken bonds and network defects, with activation energies determined by the local topology of the silicate network. The DDO model has been successfully applied to complex various nature multicomponent systems including industrial and nuclear-waste glasses as demonstrated in [17–21]. Parallel to these structural models, thermodynamic theories have sought to relate viscosity to configurational entropy or free volume. The free-volume theory of Cohen and Turnbull [22] and the Adam–Gibbs model, in which viscosity is inversely related to configurational entropy [23], have been widely applied to fragile glass formers. These models emphasize collective rearrangements and the growth of cooperatively rearranging regions as the glass transition is approached. Although conceptually distinct from bond-breaking models, they share the idea that viscosity reflects the availability of structural degrees of freedom and the energetic cost of accessing them. Energy-landscape approaches, pioneered by Goldstein [24] and later developed by Stillinger and Weber [25], further emphasized the role of the multidimensional potential-energy surface in determining relaxation and flow. These models provided a conceptual bridge between microscopic structural rearrangements and macroscopic transport properties. A major conceptual advance came with the development of topological constraint theory (TCT), which treats the glass network as a mechanical truss whose rigidity is determined by the number and type of atomic constraints [26,27]. In this framework, viscosity and relaxation are governed by the balance between constraints and degrees of freedom, and the glass transition can be interpreted as a rigidity transition in a topologically disordered network. This perspective was followed by the description of the glass transition as a topological phase transition in disordered systems [28–38] formally being based on Kantor-Webman theorem which established that “the rigidity threshold of an elastic percolating network is identical to the percolation threshold” and hence providing explanation of ductile to brittle transition on vitrification which is a feature of the calorimetric glass transition [39], and also linking topological defect formation to viscosity and relaxation [40,41]. Most recently, the topological description has been refined adding a generic criterion of melting based on the mathematical set theory which assigns to bonding system of solids a

Hausdorff-Besicovitch dimensionality $D = 3$ and to bonding system of melts a fractal one $D = 2.5$ [42,43]. These studies reinforce the idea that viscosity is fundamentally controlled by the topological state of the network, with flow occurring through the creation, annihilation, and migration of topological defects. A significant modern contribution to the topological understanding of amorphous materials comes from Zaccone and co-workers, who provided the first computational discovery of well-defined topological defects as mediators of plasticity in amorphous solids [44]. These defects—non-affine rearrangement zones associated with local coordination anomalies—serve as the elementary carriers of irreversible deformation. Their identification provided a microscopic

foundation for understanding how local topological irregularities govern flow, yielding, and relaxation in disordered networks. Against this historical and theoretical background, the widespread use of empirical and semiempirical viscosity equations—most notably the Williams–Landel–Ferry (WLF) [45] and Vogel–Fulcher–Tammann (VFT) [46–48] formulas—can be understood in a new light. Although often treated as purely empirical, these relations encode the same underlying physics: the temperature dependence of structural degrees of freedom, defect populations, and network rigidity. As shown in the present work, both WLF and VFT can be expressed as hyperbolic functions of temperature, emerging naturally from a universal Taylor expansion of $\ln(\eta)$ near the glass transition temperature. This expansion clarifies the relationships among the parameters of WLF, VFT, and related formulas, and it provides a theoretical basis for the principle of corresponding states in glass viscosity. Building on this insight, we introduce a new two-parameter quadratic expression for $\ln(\eta)$ in the glass transition region. This formula contains physically meaningful parameters, is consistent with the structural, thermodynamic, and topological models discussed above, and accurately describes available experimental data for a wide range of glass-forming substances. The goal of this paper is hence to demonstrate that the known empirical relationships are special cases of a more general approach based on the Taylor series expansion of the logarithm of viscosity near the glass transition temperature, and to propose a new two-parameter formula whose coefficients have a direct physical meaning.

2. Modelling the Temperature Dependence in the Glass Transition Region

To describe the temperature dependence of viscosity η or relaxation time τ in the glass transition region, two empirical equations are most often used. The Williams-Landel-Ferry (WLF) equation:

$$\ln a_T = -C_1 \frac{T-T_g}{T-T_g+C_2} ; \quad a_T = \frac{\tau(T)}{\tau(T_g)} \cong \frac{\eta(T)}{\eta(T_g)}, \quad (1)$$

where C_1 and C_2 are empirical constants, T_g is the glass transition temperature, τ is the relaxation time at temperature T [45]; or the Vogel-Fulcher-Tammann (VFT) equation:

$$\ln a_T = \left(\frac{B_0}{T-T_0} - \frac{B_0}{T_g-T_0} \right), \quad (2)$$

where B_0 and T_0 are empirical constants [46–48]. Recently, a number of empirical formulas have been proposed (e.g. [49–51]). In particular, the principle of corresponding states has been demonstrated for metallic glasses if the value of (T_A/T) is plotted on the abscissa axis as a temperature parameter [51]. The temperature T_A is a significant physical parameter; when cooling liquid metal at T_A , the cooperative nature of thermal motion begins to form. The authors [51] propose to define it in a macroscopic experiment as the temperature at which a deviation from the Arrhenius law is observed for viscosity. There is a clear correlation between T_A and T_g : $T_A \approx 2T_g$. As the second parameter of the empirical formula describing $\lg(\eta)$ in the glass transition region, the value of $\lg(\eta_0)$ was used in the work, where the average value of $\eta_0 = nh$, n is the particle density, and h is Planck's constant. The formula proposed for viscosity (in decimal logarithms) has the following form:

$$\lg \eta = \lg \eta_0 + T_A/T. \quad (3)$$

For $\ln a_T$ this gives

$$\ln a_T = \alpha \left(\frac{T_A}{T} - \frac{T_A}{T_g} \right), \quad (4)$$

where $\alpha \approx 2.3026$ is the conversion factor from decimal to natural logarithms. The relationships between the empirical constants of the WLF, VFT equations and equation (4) follow from their graphical interpretation (Figure 1).

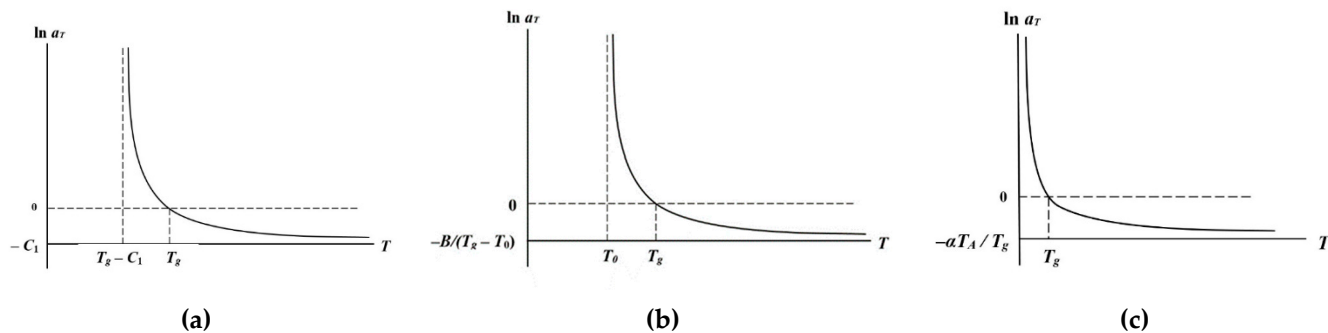


Figure 1. Graphs of the $\ln a_T$ function: (a) – according to the WLF equation, which can be represented as: $\ln a_T = -C_1 \frac{T-T_g}{T-T_g+C_2} = -C_1 \left(1 - \frac{C_2}{T-T_g+C_2}\right)$; (b) – according to the VFT equation (2); (c) – according to equation (4) in accordance with formula (3) [51].

It becomes obvious that all these empirical formulas correspond to the hyperbolic dependence of $\ln a_T$ on temperature. Each formula contains, in addition to T_g , two “adjustable” parameters. Comparison of the WLF and VFT formulas yields $C_1 = B_0/(T_g - T_0)$; $T_g - C_2 = T_0$. Comparison of the formula of the authors [51] and the WLF leads to the equality of the coefficients $C_1 = \alpha T_A/T_g$ at $C_2 = T_g$. Comparison of the VFT equation and (4) formula yields $T_0 = 0$ and $B_0 = \alpha T_A$.

Thus, the function $\ln a_T$ has been successfully described by a number of authors as a hyperbolic function of T by varying two hyperbola parameters and using the value of T_g as the third parameter. Moreover, unlike the well-known purely empirical relationships of the WLF and VFT, the authors [51] attribute a specific physical meaning to the parameters of their formula. Obviously, a hyperbolic dependence on T can be transformed into a linear dependence on $1/T$ in certain coordinates.

3. Theoretical Background: Taylor Series Expansion

Note that the WLF and VFT equations do not explicitly satisfy the principle of corresponding states unless we assume that the coefficients C_2 in the WLF formula and T_0 in the VFT formula are proportional to T_g . The former was proven from general considerations in the work of one of the authors [52] and discussed in detail in [53]. It was shown in [52] that the universality of the WLF equation, similar to the universality of Hooke's law, follows from the expansion of the function of interest to us in terms of a small parameter in a Taylor series near the glass transition temperature. In the case of Hooke's law, as is known, this is the expansion of the energy of an atom (molecule) in a solid near the equilibrium position in terms of the deformation of an interatomic bond (for a uniform deformation equal to the deformation of the sample as a whole). In the case of $\ln \eta$, the series expansion can be carried out near T_g in terms of a small parameter $\delta = (T - T_g)/T_g$:

$$\ln \eta(T) = \ln \eta(T_g) + A\delta + B\delta^2 + \dots \quad (5)$$

Restricting ourselves to the quadratic term in δ in the expansion, we obtain

$$\ln a_T = \ln \frac{\eta(T)}{\eta(T_g)} \approx A\delta + B\delta^2, \quad (6)$$

where $A = T_g \left. \frac{d \ln \eta}{dT} \right|_{T_g}$, $B = \frac{1}{2} T_g^2 \left. \frac{d^2 \ln \eta}{dT^2} \right|_{T_g}$, at the same time $A < 0$.

The condition

$$|B\delta/A| \ll 1, \quad (7)$$

allows one to use the approximation $(1 + B\delta/A) \approx (1 - B\delta/A)^{-1}$ and obtain [42] the WLF formula, in which the coefficients are equal

$$C_1 = A^2/B, \quad C_2 = -AT_g/B. \quad (8)$$

The relationship between the coefficients A and B and the coefficients of the VFT formula can be easily obtained from the above-mentioned relationship of the latter with the coefficients C_1 and C_2 .

The coefficient $C_1 \approx 36 - 40$ and depends weakly on the nature of the glass [54]. Comparison of formulas (1) and (4) yielded $C_1 = \alpha T_A/T_g$; considering that in [51] $T_A \approx 2T_g$, we find that for the metallic

glasses studied by the authors, the constant C_1 in the VFT formula also depends practically weakly on the type of metal and $C_1 = \text{const} \approx 4.6$ – an order of magnitude smaller than for inorganic glasses. The principle of corresponding states according to formula (3) is ensured by the rigid relationship between T_A and T_g . Figure 2 shows experimental data for a number of glasses, confirming the dependence $C_2 = \beta T_g$, where the coefficient β is the same for glasses of the same class.

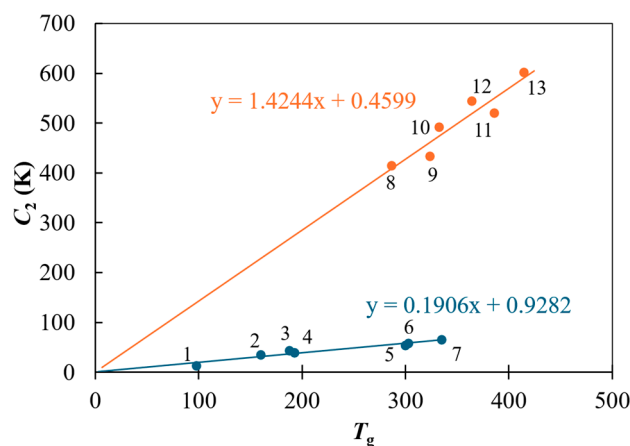


Figure 2. Dependence of the parameter C_2 of the WLF equation on the glass transition temperature T_g for different classes of glass-forming systems. Group I [55]: propanol – (1), cellulose tributyrat in dimethyl phthalate 43 wt. % – (2), propylene glycol – (3), cellulose tributyrat in dimethyl phthalate 21 wt. % – (4), natural rubber – (5), selenium – (6), ethyl methacrylate – (7). Group II [46]: B_2O_3 – 98 %, Li_2O – 2 % – (8); B_2O_3 – 94.9 %, Li_2O – 5.1 % – (9); B_2O_3 – 93 %, Li_2O – 7 % – (10); B_2O_3 – 87.8 %, Li_2O – 12.2 % – (11); B_2O_3 – 89.6 %, Li_2O – 10.4 % – (12); B_2O_3 – 84.8 %, Li_2O – 15.2 % – (13).

It should be noted that the requirement that the approximating straight line passes through the origin is a strict consequence of the principle of corresponding states. However, actual experimental data demonstrate a noticeable scatter from the ideal linear dependence. The clearest correlation is observed for polymers (propanol, propylene glycol, cellulose mixtures), as well as for selenium, ethyl methacrylate and borate glass. For many other glass-forming systems (e.g., for the glasses from Table 1), the scatter of points can reach 20–30%, which is due to both errors in determining C_2 and the influence of specific chemical interactions not taken into account in the simple model.

Table 1. Parameters of the WLF equation and coefficients A and B for some chalcogenide and silicate vitreous systems based on experimental data from [57–61].

Glass		$T_{g,}$	$-A$	B	$\Delta T,$	C_1	$C_2,$	$U_g, \text{kJ/mol}$
mol. %		K			K		K	
As_2S_3	AsI_3							
					[57]			
100	0	453	78.8	75.7	94.3	54.7	305.8	206,0
95	5	443	74.0	70.5	93.0	50.2	285.8	184,9
90	10	431	76.6	67.1	98.4	53.0	274.4	190,0
85	15	418	97.0	117.9	68.8	58.8	248.7	204,3
75	25	394	90.0	107.4	66.0	55.1	229.6	180,5
65	35	372	101.5	179.5	42.1	58.6	234.6	181,0
55	45	353	86.9	326.3	18.8	-	-	-
50	50	343	96.7	486.9	13.6	-	-	-
45	55	337	91.5	78.4	78.7	-	-	-

Averaging:			-	94.0	129.8	-	-	-	-
Sb	Ge	Se	[58]						
10	5	85	351	50.1	63.0	55.8	23.9	204.4	69.8
5	15	80	408	72.1	82.8	71.1	36.9	191.6	125.0
20	10	70	424	93.8	150.4	52.9	43.2	200.3	152.3
20	15	65	489	85.9	125.6	66.9	47.5	271.9	193.1
22	15	63	520	94.4	80.7	121.7	36.8	270.8	158.8
Averaging:			-	81.8	108.2	-	-	-	-
SiO ₂	B ₂ O ₃	Na ₂ O	[59]						
74	13	13	868	91.3	120.7	131.3	44.0	395.4	317.1
SiO ₂	Li ₂ O	Na ₂ O	[60]						
66.7	13.3	20	656	68.7	24.7	364.9	63.1	431.2	344.0
SiO ₂	Al ₂ O ₃	Na ₂ O	[61]						
70	10	20	810	68.0	66.4	165.9	32.0	398.2	215.6
Averaging:			-	84.1	127.7	-	-	-	-

Nevertheless, even in these cases, the general trend remains, and the deviations are random and do not violate the main conclusion about the scalability of the WLF parameters with the glass transition temperature. The absence of a systematic dependence on composition in individual samples is probably associated with the narrow range of T_g changes and cumulative error, but does not contradict the principle of corresponding states.

Since $(T_g - C_2) = T_0$ (Figure 1), the validity of the principle of corresponding states for the VFT formula is thus proven. Indeed, experimental data for glasses of the same class can be quite successfully represented by a single curve in generalized T/T_g coordinates (Figures 3 and 4).

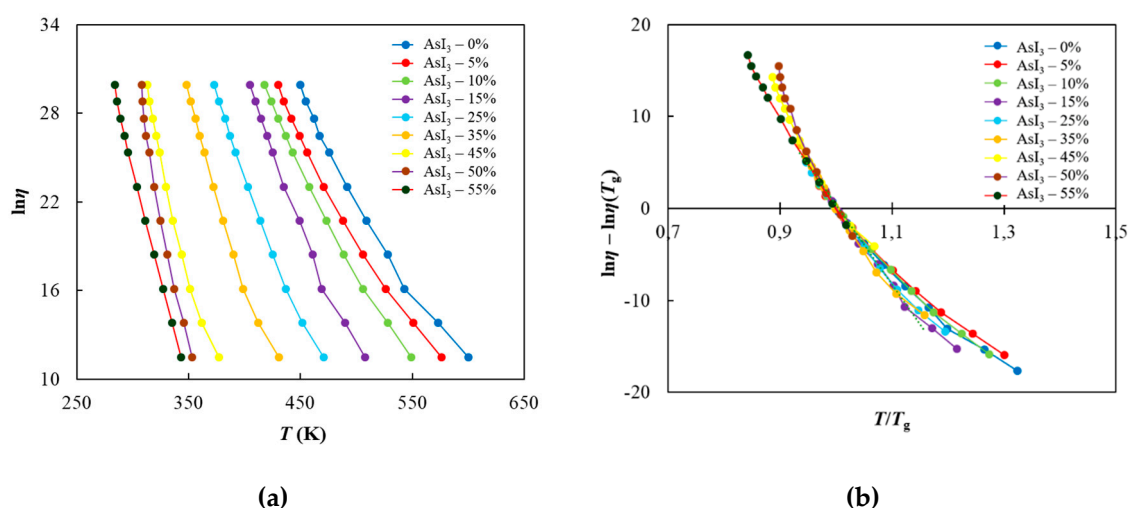


Figure 3. The temperature dependence of viscosity in the glass transition region (a) –for chalcogenide glasses with different additive contents [57]; (b) – the same data in generalized T/T_g coordinates.

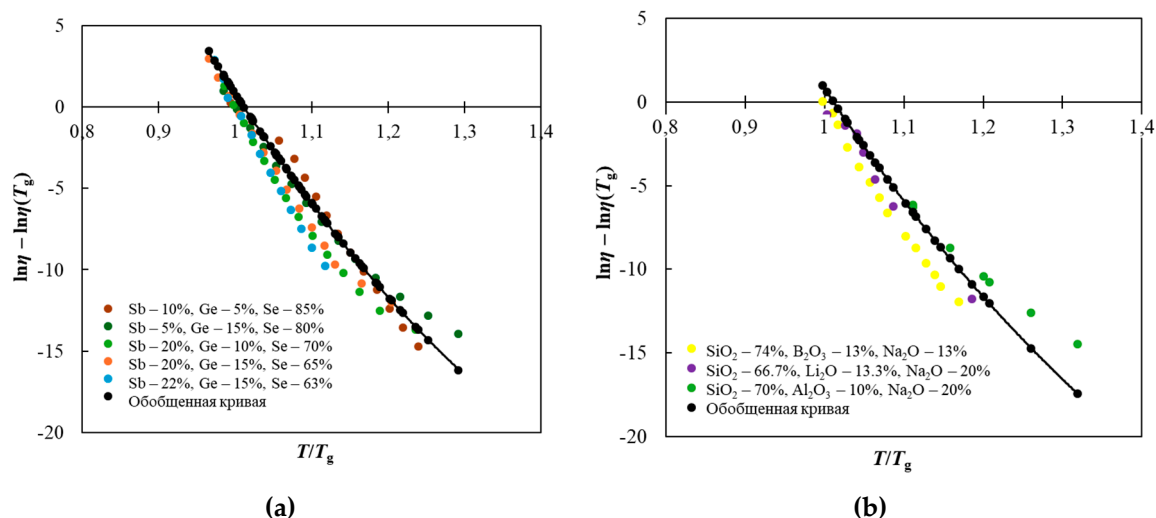


Figure 4. The temperature dependence of viscosity in generalized coordinates: (a) –for chalcogenide glasses of a different composition, data from [48] were used; (b) – for silicate glasses of three compositions, data from [59] were used. Solid curves are the result of averaging.

The $\ln(\eta)$ function can be represented as a second-degree polynomial (6) if the term cubic in the Taylor series δ can be neglected. In fact, the possibility of using such an expansion of this function into a rapidly converging series is ensured by the sharp temperature dependence of the viscosity in the glass transition region, which is close to a linear line.

The experimental values of the coefficients A and B in Table 1 allow us to estimate the temperature range ΔT above and below T_g , in which the term quadratic in δ is an order of magnitude smaller than the linear term, and the second-degree polynomial coincides with the hyperbolic dependence of the WLF or VFT. From the condition $|B\delta/A| = 0.1$, where $\delta = (T - T_g)/T_g$, and at the interval boundaries $T = T_g \pm \Delta T/2$, we obtain

$$\Delta T = 0.2T_g \left(\frac{A}{B} \right). \quad (9)$$

Formula (6) can be directly applied to describe experimental data on viscosity or relaxation time in the glass transition region. Instead of hyperbolic dependences, a second-order polynomial with respect to the generalized coordinate $\delta = (T - T_g)/T_g$ can be used, also with two empirical constants (A and B):

$$\ln a_T = A\delta + B\delta^2 = A(T/T_g - 1) + B(T/T_g - 1)^2. \quad (10)$$

The advantage of this approach is that the coefficients A and B have a clear physical meaning: they are related to the first and second derivatives of the logarithm of viscosity with respect to temperature at the glass transition point, and therefore to the activation energy and its temperature dependence.

Furthermore, obtaining the coefficients A and B is a relatively simple process. Standard programs easily characterize a specific curve with a polynomial of not only the second degree, but also the third degree (if a more precise description of the experiment is required).

In Figures 3b, 4a, and 4b, it is sufficient to shift the coordinates on the x-axis by one to the left, i.e., move the intersection of the curve with the axis to the origin, and we obtain the $\ln(a_T(\delta))$ dependence.

Below (Figures 5 and 6) are some experimental curves from Figures 3 and 4 in $\ln a_T(\delta) - \delta$ coordinates and the corresponding second-degree polynomials describing these curves. Table 1 shows the curves. 1 shows the values of the coefficients A and B for some chalcogenide and silicate glasses, as well as the temperature range ΔT near T_g , for which our approximation turns into the WLF formula with an accuracy of 10%.

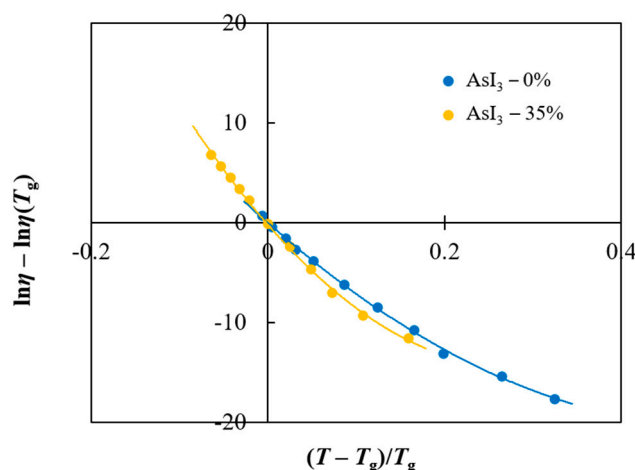


Figure 5. The experimental data in Figure 3a for glasses of two compositions and the corresponding second-degree polynomials with coefficients: for AsI3 0% $A = -78.79$; $B = 75.74$; for AsI3 35% $A = -101.52$; $B = 179.53$.

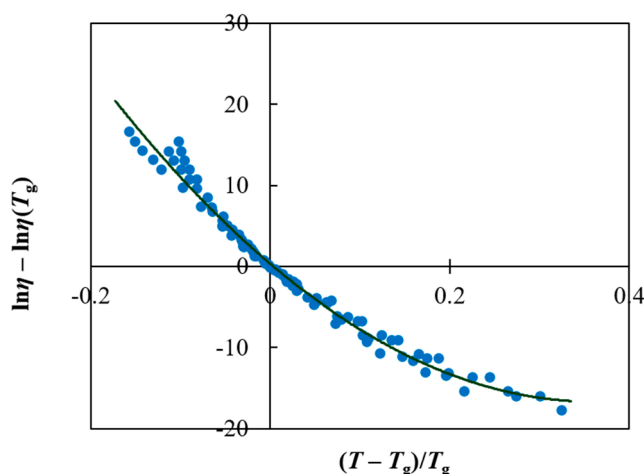


Figure 6. A second-degree polynomial (solid curve) describing all the experimental data in Figure 3a in generalized coordinates $(T - T_g)/T_g$ at $A = -94.0$; $B = 129.8$.

It is important that the coefficients A and B allow us to directly calculate the temperature dependence of the effective activation energy of the glass transition process $U_{ef} = RT \ln(\eta)$ [62].

4. Calculation of Effective Activation Energy

A sharp increase in viscosity in the glass transition region is usually considered as a consequence of an increase in the effective activation energy of the process $U_{ef} = RT \ln(\eta)$. Using the VFT equation (2), we obtain for the effective activation energy [62]:

$$U_{ef} = RT \frac{B_0}{T - T_0}. \quad (11)$$

If we express the effective activation energy through the parameters of the VLF equation, then, taking into account the relations $B_0 = C_1 C_2$ and $T_0 = T_g - C_2$, we obtain

$$U_{ef} = RT \frac{C_1 C_2}{T - T_g + C_2}. \quad (12)$$

From equality (12) follows the relationship for calculating the activation energy of the glass transition process $U_g = U(T_g)$ at the glass transition temperature $T = T_g$

$$U_g = \frac{C_1 C_2 R T_g}{T_g - T_g + C_2} = C_1 R T_g. \quad (13)$$

Finally, the calculation of U_{ef} using our proposed formula (10) gives:

$$U_{ef} = \frac{U_g T}{T_g} + RT \left[A(T/T_g - 1) + B(T/T_g - 1)^2 \right], \quad (14)$$

or, in the form of a third-degree polynomial in T ,

$$U_{ef} = T \left[\frac{U_g}{T_g} + R(B - A) \right] + T^2 \frac{R(A - 2B)}{T_g} + T^3 \frac{RB}{T_g^2}. \quad (15)$$

As follows from Table 1, $A - 2B \approx -A$. Taking this into account, the derivative of U_{ef} with respect to T can be written as follows:

$$\frac{dU_{ef}}{dT} \approx \frac{U_g}{T_g} + R(B - A) + 2R(A - 2B) \left(\frac{T}{T_g} \right) + 3RB \left(\frac{T}{T_g} \right)^2. \quad (16)$$

Some numerical refinements are naturally possible with proposed formulae for example aiming to account for the correlation between T_A and melting temperatures T_m : $T_A = kT_m$ where $k = 1.1 \pm 0.15$ instead of that used above $T_A \approx 2T_g$ [63–65].

5. Conclusions

The applicability of the law of corresponding states to the viscosity of glasses of a given composition in the glass transition region has been demonstrated using a number of glass-forming systems. This applicability is substantiated by the possibility of expanding $\ln(\eta)$ into a Taylor series in terms of relative temperature near the glass transition temperature, regardless of the glass transition mechanism. Known empirical and semiempirical dependences of $\ln(a\tau)$ are, in fact, hyperbolic functions of temperature and can be derived from this expansion. Certain relationships exist between their coefficients. It is proposed to use a second-degree polynomial, along with hyperbolic dependences, to describe experimental data on viscosity and relaxation time in the glass transition region. The coefficients of the polynomial, unlike empirical constants, have a physical meaning and are directly related to the effective activation energy of the glass transition process.

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Data Availability Statement: The dataset used for calculations were obtained from other publications, all of which are cited appropriately in the text.

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