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Article

Relaxation Character of Glass Transition in Binary Oxide Glasses Containing Lead and Boron

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Abstract: We have examined the temperature dependence of the viscosity of lead and boron containing binary oxide glasses in the context of transition from the liquid to the vitreous state, including analysis of kinetic criterion of glass transition. Both the temperature interval determining the glass transition and the structural relaxation time were found near the glass transition temperature of PbO-SiO₂, PbO-GeO₂, B₂O₃-Bi₂O₃, B₂O₃-Li₂O, B₂O₃-BaO, B₂O₃-Na₂O₃ glasses based on Williams-Landel-Ferry equation.

Keywords: viscosity; glass transition criterion; liquid-glass transition; Williams-Landel-Ferry equation; glass transition

1. Introduction

The glass transition being a universal feature of amorphous state of matter is analyzed with novel insides aiming to reveal its nature. Transformation of a liquid on cooling into a glass (i.e. vitrification) can take place at melt cooling rates rapid enough that crystallization is kinetically avoided. Glass transition phenomena are observed universally, and any liquid can be in practice vitrified provided that the rate of cooling is suitable selected so that crystallization is avoided kinetically. Experimentally the glass transition is usually observed as a second order-like phase transformation when the material volume and entropy are continuous functions of temperature exhibiting kinks at glass transition, and have discontinuities observed for their derivatives. Namely these characteristics are used in practice to detect where transformation occurs with “calorimetric glass transition” typically used as a standard term, see Chapter 3.2 of Ref. [1]. Understanding the nature of glass transition is, however, difficult due to almost undetectable changes in the structure of amorphous materials using conventional techniques despite the qualitative changes in characteristics such as embrittlement and extremely large change in the time scale of relaxation processes [2]. As for the second-order thermodynamic transition which occurs at the calorimetric glass transition temperature its nature is related to the structural changes in the bonding system of materials and effectively unveiled by the mathematical set theory which assigns a Hausdorff-Besicovitch dimensionality D of bonding system equal to 3 alike for crystals and a fractal one equal to 2.5 in the molten state, see e.g. Table 3 of Ref. [3], and Refs. [4,5] for details. The Kantor-Webman theorem which states that “the rigidity threshold of an elastic percolating network is identical to the percolation threshold” is then providing explanation of ductile to brittle transition which is a feature of the calorimetric glass transition [6]. The concept of dimensionality change at glass transition is receiving further support via statistical analysis of random processes activated by temperature in various materials including metals and polymers [7,8].

Often the glass transition is however considered as just a gradual although considerable change of material viscosity with an arbitrarily defined glass transition temperature at which the equilibrium viscosity of the melt reaches 10^{12} Pa·s [3]. The definition of a glass as an amorphous material at high

viscosities is inconsistent for at least the reasons that (i) the viscosity is a continuous function of temperature in contrast with derivative thermodynamic parameters such as heat capacity, and (ii) the viscosity is not necessarily equal to 10^{12} Pa·s varying 4 orders of magnitude (10,000 times) from $10^{8.8}$ Pa·s to 10^{13} Pa·s at the calorimetric glass transition temperature, see e.g. Table 4 of Ref. [9]. Also, in many marginal glass-formers equilibrium viscosity of 10^{12} Pa·s (or any other similarly high value) can never be reached owing to competing crystallization process.

Viscosity quantifies the resistance of material to flow and indicates the ability to dissipate momentum where microscopically it arises because of a transfer of momentum between fluid layers moving at different velocities [10–13]. Therefore, the tighter the layers are bound to each other the more difficult their motion occurs and hence the resulting viscosity is higher. In principle the viscous flow is mediated by flow defects so that the viscosity is inversely proportional to their concentration. There are at least three crossover temperatures in the viscosity-temperature dependence which are directly related to structural changes which occur in glass-forming systems [14,15]. Viscosity of glass-forming systems changes by many orders of magnitude, attaining values typically exceeding 10^{12} in the vitreous state and decreasing to less than 10 Pa·s at the melting temperature [9–15]. The typical viscosity temperature dependencies of glass-forming systems show two distinct Arrhenius-type behaviors at high and low temperatures (at high-viscosity and low-viscosity ends) and a temperature-dependent activation energy of flow caused by structural rearrangements (Figure 1).

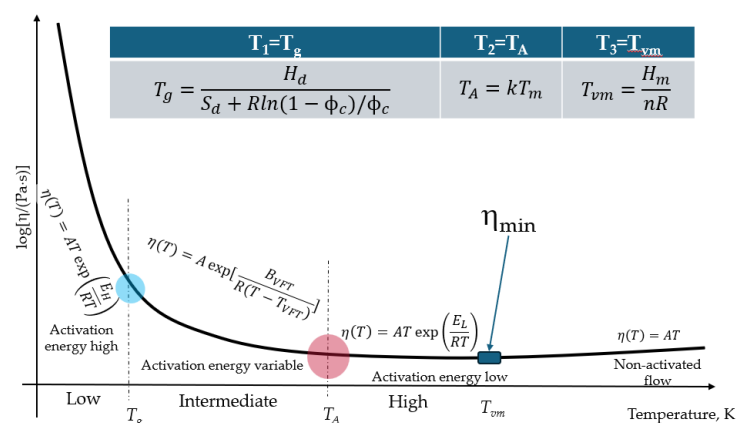


Figure 1. Generic temperature dependence of the viscosity of glass-forming systems (modified after [15]).

The first crossover temperature T_1 occurs at the high-viscosity end and is equal to glass transition temperature T_g above which the thermodynamically unstable yet kinetically stable glass transforms into a metastable supercooled liquid. The second crossover temperature T_2 occurs at the low-viscosity end and is the temperature above which the liquid becomes fully depolymerized, it is found to be about 10 – 15% higher than the melting temperature T_m . The third crossover temperature T_3 occurs when the liquid reaches its minimum possible viscosity, and the viscous flow becomes non-activated. Estimations show that T_3 is very high and can barely be reached in practical uses, although at temperatures close to T_3 the contribution of the non-activated regime of the flow must be accounted in calculations [14,15].

In various classes of amorphous substances in the glass transition region, freezing of the mobility of various kinetic units occurs differently, which is associated with the different nature of particle interactions. However, it is surprising that the basic laws of the transition from the liquid state to glass are the same for different glass-forming systems, regardless of their nature. This is confirmed by the presence of universal rules and equations in the glass transition region [16–22]. As glass-forming melts cool in the glass transition range, their flow resistance i.e. the viscosity increases sharply. As a first approximation is typically assumed that they vitrify upon reaching the same ultimate viscosity $\eta_g = \eta(T_g) = 10^{12}$ Pa·s [3]. Mazurin has showed [23] that for most of the studied glasses the temperature T_g corresponds to viscosity values η_g in the range from 10^{11} to 10^{12} Pa·s. Although

these values fluctuate with noticeable spread observed the approximate within logarithmic scale constancy $\eta_g = \text{const}$ is better fulfilled in glassy systems of one class [23]. Within the crossover temperatures $T_g \leq T \leq T_A$ the viscosity of amorphous materials is a complex function of temperature (see Figure 1) where most often the Vogel–Fulcher–Tammann (VFT) model is used to approximate its temperature dependence although the Douglas–Doremus–Ojovan (DDO) double-exponential model (see Chapter 8 of Ref. [14]) model is that one providing the most exact data with correct both high and low temperature asymptotes [14,15]. Another classical model is the Williams–Landel–Ferry (WLF) equation [9,17,22,24,25]:

$$\eta = \eta_0 \exp\left(\frac{B_0}{T-T_0}\right), \quad (1)$$

$$\log a_T = -C_1 \frac{T - T_r}{T - T_r + C_2}, \quad (2)$$

where η_0 and B_0 are temperature-independent constants and material-dependent parameters, a_T is the relative viscosity $a_T = \eta(T)/\eta(T_g)$, C_1 and C_2 are empirical temperature-independent constants, T_r is the reference temperature typically taken as the glass transition temperature $T_r = T_g$. The validity of WLF equation is confirmed for various glass-forming systems including polymers, organic, and inorganic substances, and metallic amorphous alloys [26–33].

This work is devoted to the study of the glass transition of binary oxide systems PbO–SiO₂, PbO–GeO₂, B₂O₃–Bi₂O₃, B₂O₃–Li₂O, B₂O₃–BaO, B₂O₃–Na₂O₃ based on WLF approach and the discussion of some aspects of the liquid-glass transition, in particular, the kinetic criterion of glass transition, using them as an example.

2. Methodology Based on WLF Equation

Let us check the linearity of the dependence $y = -(T - T_g)/\ln(a_T)$ on temperature departure from glass transition $x = (T - T_g)$. We use experimental data [34] on the temperature dependence of the viscosity of PbO–SiO₂ glasses with results shown in Figure 2.

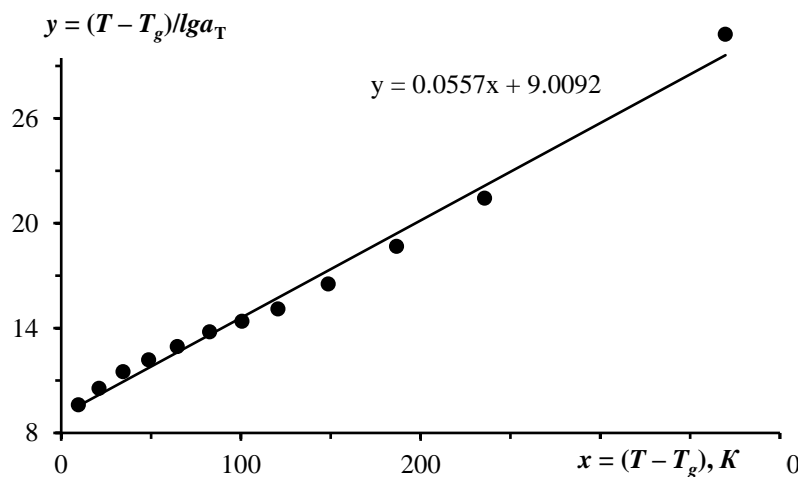


Figure 2. Temperature dependence of viscosity of lead-silicate glasses in coordinates corresponding to the Williams-Landel-Ferry equation. PbO content is 62.29 mol.%.

Analysis of Figure 2 shows that for one of the compositions of the studied lead-silicate glasses, the dependence $y(x)$ in the glass transition region is linear. This indicates the applicability of the WLF equation. Similar linear dependences were obtained for other glass compositions. From these dependences, the parameters C_1 and C_2 of the WLF equation were determined, and on their basis, the characteristics of the glass transition process were calculated (Tables 1 and 2).

Table 1. Parameters of the WLF equation C_1 and C_2 for lead-silicate and lead-borate glasses, as well as characteristics of the glass transition process based on experimental data from [34,35].

№	Glass		C_1	C_2, K	T_g, K	$\delta T_g, K$	f_g	$C_g \cdot 10^3$	τ_g, s
	mol. %								
	PbO	SiO ₂							
1	40.27	59.73	34.3	211.8	720	14.2	0.029	8.3	284.1
2	42.07	57.93	35.7	233.3	711	15.0	0.028	7.8	300.3
3	44.64	55.36	36.8	226.6	704	14.2	0.027	7.5	283.5
4	47.25	52.75	36.6	207.7	693	13.0	0.027	7.6	260.8
5	48.73	51.27	36.5	189.3	685	11.9	0.027	7.6	238.8
6	49.54	50.46	36.5	189.2	681	11.9	0.027	7.6	238.6
7	50.24	49.76	37.8	208.0	675	12.7	0.026	7.3	253.4
8	51.19	48.81	37.2	190.4	673	11.8	0.027	7.4	235.3
9	52.25	47.75	37.2	182.8	669	11.3	0.027	7.4	225.8
10	55.02	44.98	38.5	179.3	658	10.7	0.026	7.1	214.4
11	57.53	42.47	38.4	168.1	643	10.1	0.026	7.1	201.2
12	60.16	39.84	39.1	157.0	637	9.2	0.026	7.0	184.7
13	62.29	37.71	41.2	161.2	630	9.0	0.024	6.5	179.9
	PbO	GeO ₂							
1	15	85	32.2	520.8	712	16.2	0.013	3.1	323.2
2	18	82	38.2	633.2	711	16.6	0.011	2.5	331.5
3	20	80	34.6	561.9	711	16.3	0.013	2.9	325.2
4	25	75	33.0	418.8	714	12.7	0.013	3.0	254.1
5	27	73	35.9	455.9	712	12.7	0.012	2.7	254.0
6	33	67	34.9	413.6	695	11.9	0.012	2.8	237.2
7	35	65	54.9	751.0	686	13.7	0.008	1.6	273.5
8	40	60	28.9	321.9	671	11.1	0.015	3.6	222.5
9	42	58	41.2	483.0	660	11.7	0.011	2.3	234.6
10	45	55	46.1	549.0	646	11.9	0.009	2.0	238.0
11	48	52	56.6	627.0	634	11.1	0.008	1.6	221.4
12	50	50	73.7	873.6	635	11.8	0.006	1.1	237.0

Table 2. Parameters of the WLF equation C_1 and C_2 for various borate glasses and characteristics of the glass transition process based on experimental data from [21–24,36–39].

№	Glass		C_1	C_2, K	T_g, K	$\delta T_g, K$	f_g	$C_g \cdot 10^3$	τ_g, s
	mol. %								
	B ₂ O ₃	Bi ₂ O ₃							
1	79	21	46.4	517.9	717	11.2	0.009	2.0	223.3
2	78.2	21.8	33.9	385.7	717	11.4	0.013	2.9	227.4
3	77.21	22.79	44.2	437.4	716	9.9	0.010	2.1	197.9
4	76.95	23.05	64.9	713.8	716	11.0	0.007	1.3	220.1
5	72	28	40.1	439.2	715	11.0	0.011	2.4	219.0
6	69	31	63.4	620.6	709	9.8	0.007	1.4	195.9
7	64.03	35.97	63.4	620.6	697	9.8	0.007	1.4	195.9
8	62.2	37.8	99.7	982.5	694	9.9	0.004	0.8	197.1
9	57.98	42.02	27.5	266.6	670	9.7	0.016	3.8	194.2
10	56.91	43.09	59.0	580.9	661	9.8	0.007	1.5	196.9
11	43.19	56.81	85.4	765.5	616	9.0	0.005	1.0	179.3
	B ₂ O ₃	Li ₂ O							
1	98	2	25.3	400.5	286	15.9	0.017	4.2	317.2

2	94.9	5.1	27.6	433.0	323	15.7	0.016	3.8	313.6
3	93	7	29.4	492.3	332	16.7	0.015	3.5	334.5
4	89.6	10.4	31.7	544.6	364	17.2	0.014	3.2	343.2
5	87.8	12.2	29.8	520.0	386	17.4	0.015	3.4	348.7
6	84.8	15.2	38.2	601.6	414	15.8	0.011	2.5	315.0
	B₂O₃	BaO							
1	98.44	1.56	25.5	394.3	524	15.5	0.017	4.2	309.1
2	98.13	1.87	15.3	231.2	530	15.1	0.028	8.0	302.8
3	95.52	4.48	17.3	259.9	659	15.0	0.025	6.8	300.2
4	91.55	8.45	21.6	355.4	720	16.5	0.020	5.2	329.7
5	90.44	9.56	20.2	317.4	732	15.7	0.022	5.6	314.9
6	87	13	45.9	855.2	735	18.6	0.009	2.0	372.7
7	85.8	14.2	24.6	403.6	739	16.4	0.018	4.4	327.5
	B₂O₃	Na₂O							
1	94.6	5.4	21.9	367.4	293	16.8	0.020	5.1	336.2
2	92	8	23.1	377.1	319	16.3	0.019	4.7	325.9
3	89.3	10.7	23.1	377.1	343	16.3	0.019	4.7	325.9
4	86.5	13.5	19.9	307.7	373	15.4	0.022	5.7	308.9
5	84.7	15.3	29.2	457.0	391	15.7	0.015	3.5	313.2
6	78.7	21.3	28.3	375.7	455	13.3	0.015	3.7	265.3
7	76.8	23.2	21.0	255.4	460	12.2	0.021	5.3	243.4
8	75.7	24.3	20.8	253.5	460	12.2	0.021	5.4	243.3
9	75.4	24.6	21.6	253.6	463	11.8	0.020	5.2	235.0
10	72.6	27.4	22.6	261.1	466	11.5	0.019	4.9	230.5
11	67.4	32.6	20.4	215.8	467	10.6	0.021	5.5	211.5
12	61.5	38.5	23.4	241.5	461	10.3	0.019	4.7	206.6

* Note that: $\delta T_g = C_2/C_1$, $f_g = 1/C_1$, $C_g = f_g / \ln(1/f_g)$, $\tau_g = (C_2/C_1)q$.

3. Relaxation Character of Glass Transition

Analysis of Tables 1 and 2 shows that the parameter of the WLF equation C_1 in the studied glasses demonstrates a low dependence on their nature. For example, in lead-silicate glasses with different component contents, the value of C_1 in the first approximation turns out to have an insignificant spread $C_1 \approx \text{Const} \approx 34 - 40$. Similar value can be obtained from the assessment of $C_1 \approx \ln(\eta_g/\eta_0)$ accounting that typically $\eta_0 \approx 10^{-3} - 10^{-5}$ Pa·s which gives $C_1 \approx 35 - 39$. The weak dependence of C_1 on the nature of the glass is due to the fact that the viscosity at the glass transition temperature η_g and its so-called high-temperature limit η_0 remain practically constant for different glasses. We note however that there is not any high-temperature limit of viscosity (see Figure 1), instead there is always a minimum of it [40–42].

It follows from the interpretation of the WLF equation within the framework of the delocalized atom model [22,43,44] that the parameter C_1 is the reciprocal of the fraction of the fluctuation volume f_g frozen at the glass transition temperature: $C_1 = 1/f_g$. Thus, the volume fraction of the fluctuation volume f_g , obtained from the data on the parameter C_1 , weakly depends on the nature of the amorphous substances (Tables 1, 2): $f_g \approx 0.01 - 0.03$.

In the glass transition range the molecular rearrangements occur so slowly that structural changes in the melt during cooling cannot keep up with the change in temperature which is an indication of non-ergodic regime. As a result, alike the viscosity, the relaxation time of the structure $\tau(T)$ increases sharply, which is directly related to the cooling rate of the liquid $q = (dT/dt)$. In 1951, Bartenev [45], based on general reasoning, put forward the following relationship between these quantities, known as the kinetic criterion of glass transition:

$$q\tau_g = C, \quad (3)$$

where τ_g is the structural relaxation time at the glass transition temperature T_g , C is an empirical parameter with the dimension of temperature (see also Refs. [22] and [46]). Here and below, q should be understood as the absolute value of the rate of change of temperature $|q|$ during cooling ($q < 0$) of the melt or heating ($q > 0$) of the glass.

In the relaxation theory of Volkenstein and Ptitsyn [22,47–49], it is assumed that particles in a glass-forming system can exist in two states: ground and excited ones. The states are separated by an energy barrier which is typical for utilizing the two-state modelling of glass transition [4,5,50]. The solution of the compiled kinetic equation shows that at a certain temperature, called the glass transition temperature moving particles freeze in the ground state. In this approach the glass transition of the system occurs when the transitions of particles between the ground and excited states cease and the rate of transition of particles through the energy barrier becomes comparable to the rate of cooling of the system, i.e. the ergodic to non-ergodic transition occurs. This leads to the fact that the particles "get stuck" in the ground state and the system transit into a glassy state. The glass transition temperature is then found from the condition:

$$\left(\frac{d\tau}{dT}\right)_{T=T_g} = -\frac{1}{q} \quad (4)$$

In this theory, the kinetic criterion for the liquid-glass transition is the glass transition equation [26]:

$$q\tau_g = \delta T_g \quad (5)$$

The designation of the right-hand side of this equality as δT_g was proposed by Nemilov as Schmelzer noted in [51]. The value δT_g characterizes the temperature range of vitrification i.e. the transition from liquid to glass during cooling. The parameter of the Bartenev equation C in the Volkenstein-Ptitsyn theory (see equation (3)) acquires the meaning of the temperature interval δT_g .

Let us consider the estimate of the parameter of the glass transition equation δT_g for the studied glasses. Substitution of the dependence $\tau(T)$ from the WLF equation into the relation (4) leads to equality

$$q\tau_g = \frac{C_2}{C_1} \quad (6)$$

comparison of which with the glass transition equation (5) allows us to obtain a formula for calculating the temperature interval δT_g based on the data on the parameters of the WLF equation:

$$\delta T_g = \frac{C_2}{C_1} \quad (7)$$

For the studied glasses (Tables 1, 2) the value of δT_g is as high as 9 – 17 K.

The second parameter of WLF equation C_2 is equal to the ratio of f_g to the coefficient of thermal expansion of the fluctuation volume at the glass transition temperature β_f [26]:

$$C_2 = \frac{f_g}{\beta_f} \quad (8)$$

where the product $\beta_f \cdot T_g$ is a single-valued function of f_g :

$$\beta_f T_g = f_g \ln(1/f_g) \quad (9)$$

Accounting that $C_1 = 1/f_g$, (7) and for (7), (8) and (9) we see that the parameter of the glass transition equation δT_g is determined by the glass transition temperature and the fraction of the fluctuation volume f_g :

$$\delta T_g = \frac{f_g}{\ln(1/f_g)} T_g \quad (10)$$

Since for glasses of the same class it is assumed that $f_g \approx \text{const}$, a linear correlation between the values of δT_g and T_g is hence expected. As we can see from Figure 2, for lead-silicate glass the temperature interval δT_g , within which the transition from liquid to glass occurs, linearly depends on the glass

transition temperature T_g . An approximate linear correlation is observed for lead-germanate (PbO-GeO_2) and borate glasses ($\text{B}_2\text{O}_3\text{-Bi}_2\text{O}_3$, $\text{B}_2\text{O}_3\text{-Li}_2\text{O}$, $\text{B}_2\text{O}_3\text{-BaO}$, $\text{B}_2\text{O}_3\text{-Na}_2\text{O}_3$) as shown in Figures 2 and 3.

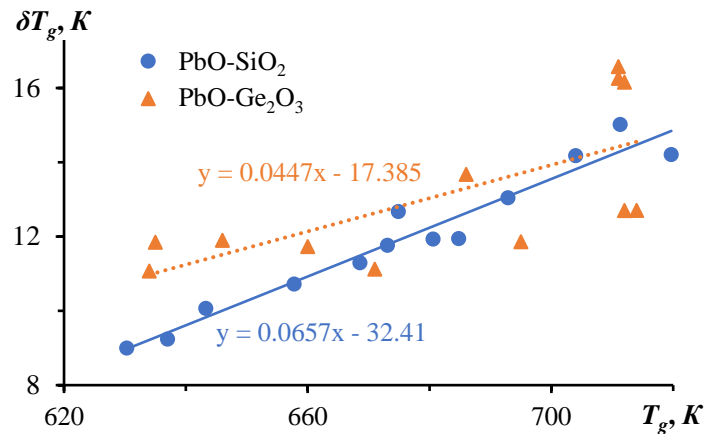


Figure 3. Correlation between the temperature interval δT_g , characterizing the glass transition range, and the glass transition temperature T_g for lead-silicate and lead-germanate glasses.

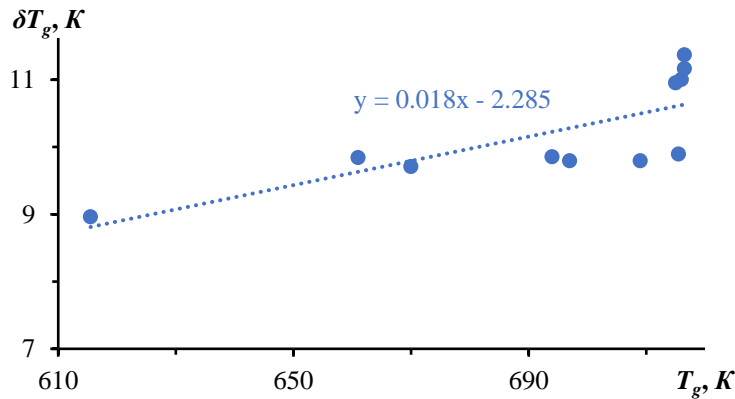


Figure 4. Example of correlation between δT_g and T_g for $\text{B}_2\text{O}_3\text{-Bi}_2\text{O}_3$ glasses

4. Generalization of Schmelzer Vitrification Criterion

Schmelzer has proposed a characteristic time change in temperature: $\tau_T = T/q$. At $\tau \ll \tau_T$ the liquid is in a state of thermodynamic equilibrium, and at $\tau \gg \tau_T$ – in a frozen glassy state [51,52]. It is then assumed that the liquid-glass transition (vitrification) corresponds to the condition: $\tau \approx \tau_T$. Based on these considerations Schmelzer formulated the following kinetic criterion for glass transition:

$$\left[\frac{1}{T} \left(\frac{dT}{dt} \right) \tau \right]_{T=T_g} = C_3, \quad C_3 \approx 1 \quad (11)$$

The glass transition equation (5) in the account of expression (10) for the temperature interval δT_g , receives the following interpretation:

$$q\tau_g = \frac{f_g}{\ln(1/f_g)} T_g \quad (12)$$

Dividing both parts of this equality by the glass transition temperature T_g with an average value of $f_g \approx 0.02$ we arrive at a generalized version of the kinetic glass transition criterion [30]:

$$\frac{q\tau_g}{T_g} = C_g = \frac{f_g}{\ln(1/f_g)} \quad (13)$$

which, taking into account that $q = (dT/dt)$, can be written as an expression for determining the transition temperature T_g :

$$\left[\frac{1}{T} \left(\frac{dT}{dt} \right) \tau \right]_{T=T_g} = C_g, \quad C_g \approx 5 \cdot 10^{-3} \quad (14)$$

The equation (14) is a generalization of the Schmelzer glass transition criterion (11). Based on it we conclude that the constant C_3 acquires a certain physical meaning and a different numerical value:

$$C_3 = C_g = \frac{f_g}{\ln(1/f_g)} \approx \text{const} \approx 5 \cdot 10^{-3} \quad (15)$$

Constancy in the first approximation of C_g is observed in glass of the same family (Table 1). Hence it follows that the value of f_g (as well as of C_g) is determined by the viscosity $\eta_g \approx \text{const}$ at T_g and the conventional high-temperature viscosity limit $\eta_0 \approx \text{const} \approx \eta_{\min}$ [12,13,15,42,53]:

$$\frac{1}{f_g} = \ln \left(\frac{\eta_g}{\eta_0} \right) \quad (16)$$

The interpretation of the generalized criterion (13), (14) is as follows: the liquid passes into a frozen glassy state (vitrifies) when the cooling rate q reaches a certain constant fraction C_g of the characteristic cooling rate $q = (T_g/\tau_g)$, closely related to the relaxation time of the structure τ_g at the glass transition temperature T_g , e.g. see for details [30].

5. Structural Relaxation Time

The glass transition temperature, although weakly i.e. logarithmically [16,22], depends on the cooling rate of the melt q . The temperature at which the viscosity $\eta_g \approx 10^{12}$ Pa·s is achieved is conventionally considered as the glass transition temperature T_g , and the corresponding cooling rate

$$q = 3 \text{ K/min} = 0.05 \text{ K/s}, \quad (17)$$

which is considered a standard cooling rate [17,23,40,54]. Approximately the same cooling rate is almost universally used as well in the dilatometric investigations of inorganic glasses and polymers. Due to the weak dependence of T_g on q , small fluctuations of q around the standard value (17) practically do not affect the value of T_g . Therefore, it is usually assumed that the vast majority of available data on T_g actually relate to the standard cooling rate (17) with few data for other cooling rates q . Using formula (6) at the standard cooling rate $q = 0.05$ K/s and data on the parameters of the WLF equation C_1 and C_2 , one can calculate the structural relaxation time τ_g at the glass transition temperature as follows:

$$\tau_g = \frac{C_2}{qC_1} \quad (18)$$

Tables 1 and 2 show the results of calculating the structural relaxation time for lead and boron containing glasses. It is seen that τ_g depends on the nature of the glass-forming substance.

Let us now estimate the calculated values of the structural relaxation time. For this we calculate τ using the Maxwell formula: $\tau = \eta/G$, where G is the shear modulus. For most inorganic glasses, the value of the shear modulus G is approximately $(20 - 25) \cdot 10^9$ Pa and remains virtually unchanged with temperature as at $T = T_g$ it is only 3 – 5% less than at room temperature. For a significant number of oxide inorganic glasses, at a standard cooling rate of $q = 0.05$ K/s, the logarithm of viscosity at the glass transition temperature is $\lg(\eta_g, \text{Pa}\cdot\text{s}) = 12.76 \pm 0.26$. From these data on the values of G_∞ and η_g at $T = T_g$, at the level of qualitative estimates, we have [3,14,19]:

$$\tau_g = \frac{\eta_g}{G_\infty} \approx 10^2 \text{ s}, \quad (19)$$

which by the order of magnitude coincides with the results of calculation using formula (18) based on the data on the parameters C_1 and C_2 of WLF equation.

6. Conclusions

The regularities of liquid transition to the glassy state were considered using lead and boron containing glasses as an example. It is established that the fraction of the fluctuation volume f_g frozen

at the glass transition temperature T_g varies within small limits for the studied systems in the first approximation, since the value of f_g weakly depends on the nature of glasses. Based on the data on the parameters of the Williams-Landel-Ferry equation at a standard cooling rate, the temperature interval δT_g , within which the liquid passes into glass (vitrifies), is calculated. For the studied glasses, the value of δT_g is as high as 9 – 17 K. The obtained results contribute to a deeper understanding of the glass transition mechanism and allow us to establish quantitative relationships between the parameters of the empirical equations and the fundamental characteristics of glass-forming systems. This is also important for predicting the properties of new vitreous materials and optimizing the technological processes for their production.

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