

INVITED SCIENTIFIC REPORT 2

RELAXATION ASPECTS OF THE GLASS TRANSITION

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On relaxation nature of glass transition in amorphous materials

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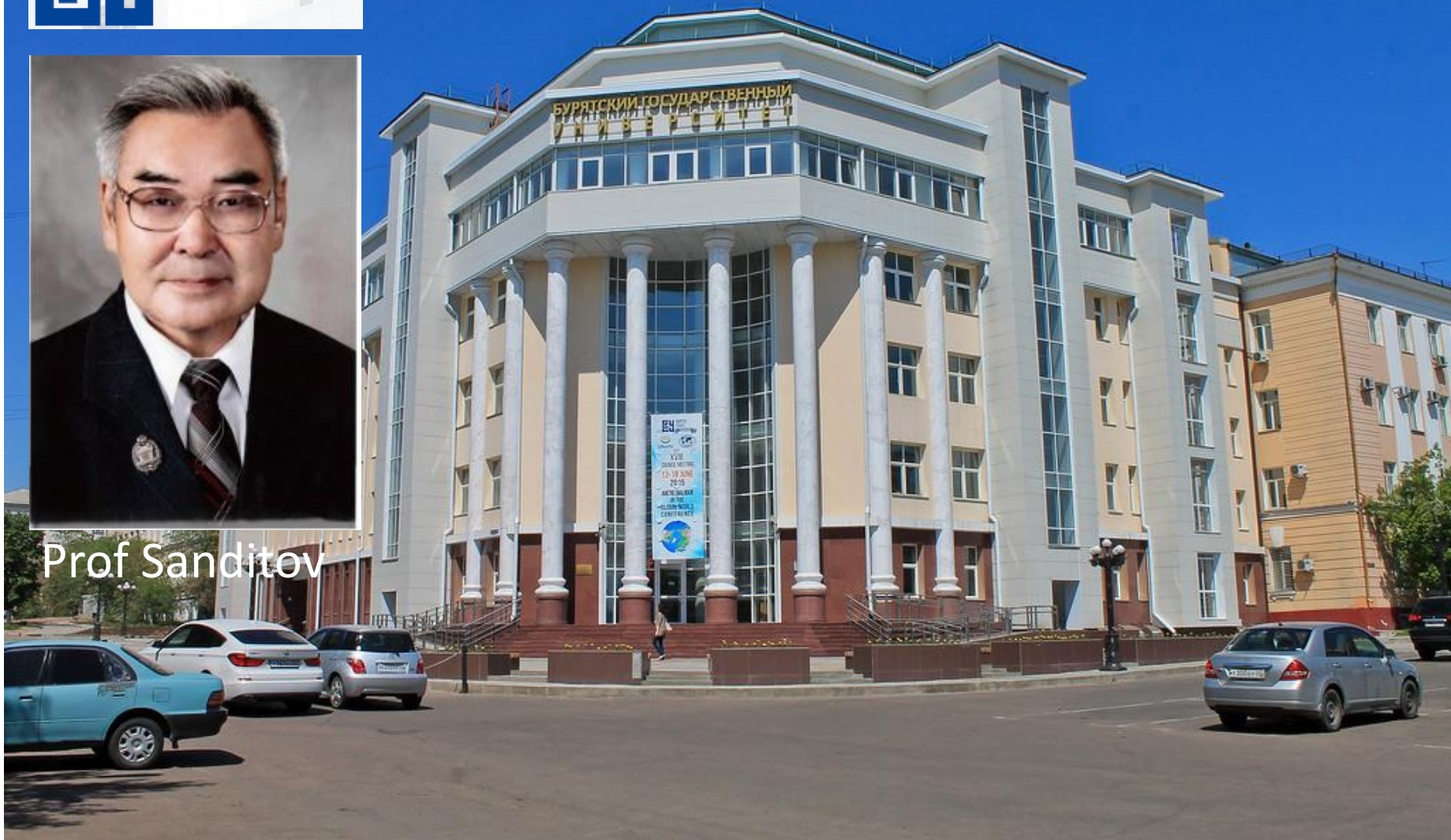




BURYAT
STATE
UNIVERSITY



Prof Sanditov



Buryat State University named after Dorjee Banzarov



Lake Baikal



Olkhon Island



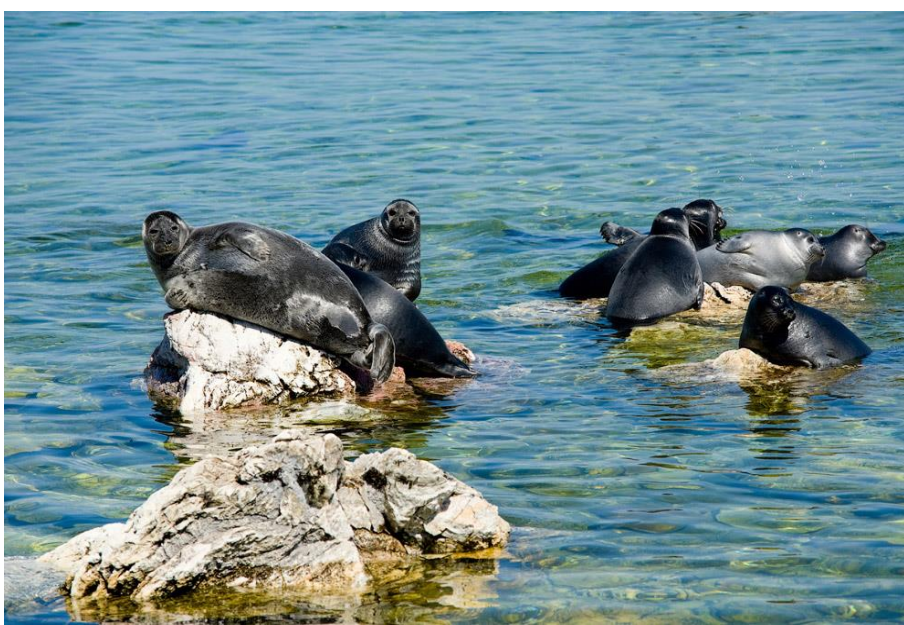
Lake Baikal is the largest freshwater lake by volume in the world, containing 22–23% of the world's fresh surface water. With 23,615.39 km³ of fresh water, it contains more water than the North American Great Lakes combined. With a maximum depth of 1,642 m, Baikal is the world's deepest lake. It is considered among the world's clearest lakes and is considered the world's oldest lake — at 25 million years, making it the most ancient lake in geological history.



Republic of Buryatia, Ulan-Ude, Buddhist temple



Ulan-Ude, Monument of Lenin



- Lake Baikal is the largest freshwater lake by volume in the world, containing 22–23% of the world's fresh surface water.
- With a maximum depth of 1,642 m, Baikal is the world's deepest lake.
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Russia, Republic of Buryatia, Lake «Baikal»

Background

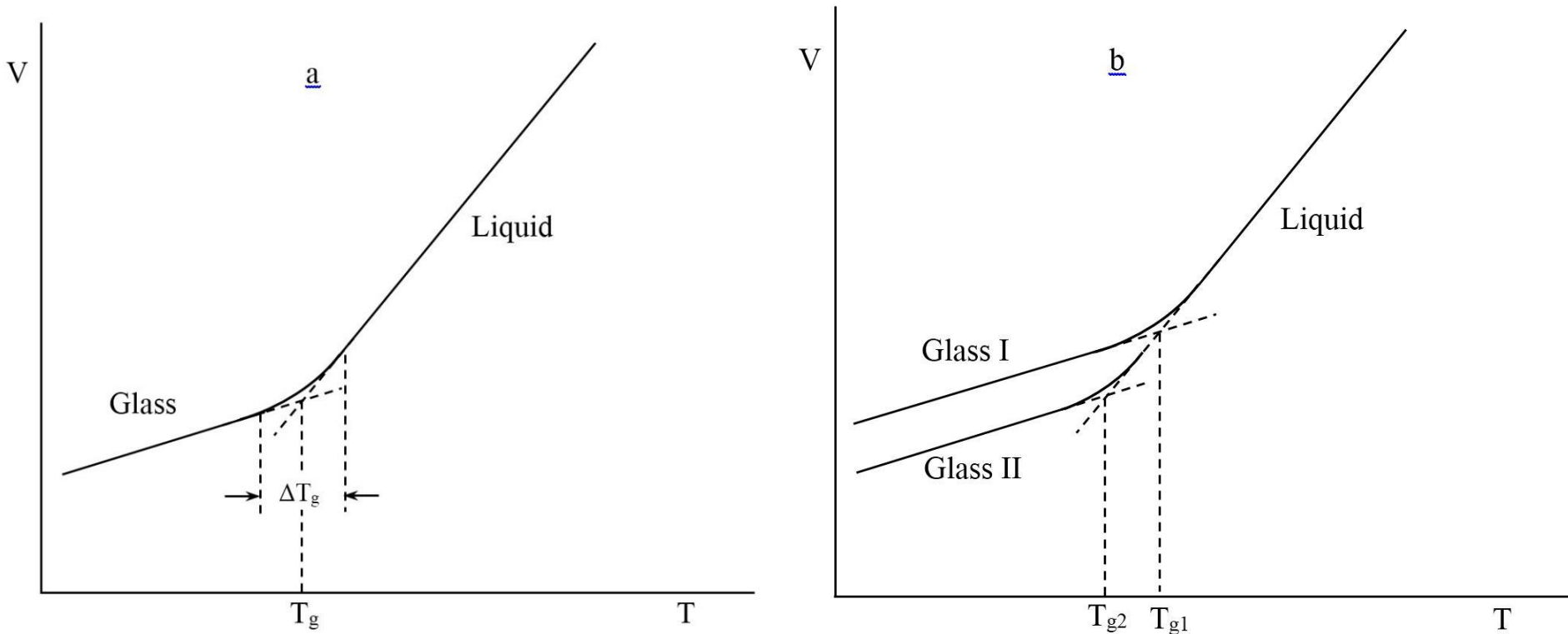


Fig. 1.1. Volume change during the liquid–glass transition in the process of cooling. ΔT_g – glass transition region, T_g – glass transition temperature. T_{g1} – corresponds to cooling rate q_1 , and T_{g2} – to cooling rate q_2 , $q_2 < q_1$.

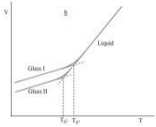
Bartenev proposed in 1951 the equation that is often called the main equation of glass transition:

$$q\tau_g = C, \quad \tau_g \approx (1 \div 2) \cdot 10^2 \text{ s},$$

G.M. Bartenev, Dokl. Akad. Nauk SSSR 76 (1951) 227–230.

This equation is successfully applied in relaxation spectrometry of polymers and glasses as a condition of structural relaxation transition at $T = T_g$.

The glass transition temperature T_g depends on the cooling rate of the glass-forming melt $q = dT/dt$. The lower the cooling rate, the lower the glass transition temperature.



$$\frac{1}{T_g} = a_1 - a_2 \ln q$$

Bartenev-Rittland equation

G.M. Bartenev, Dokl. Akad. Nauk SSSR 76 (1951) 227–230.

H.N. Ritland, J. Am. Ceram. Soc. 37 (1954) 370–378.

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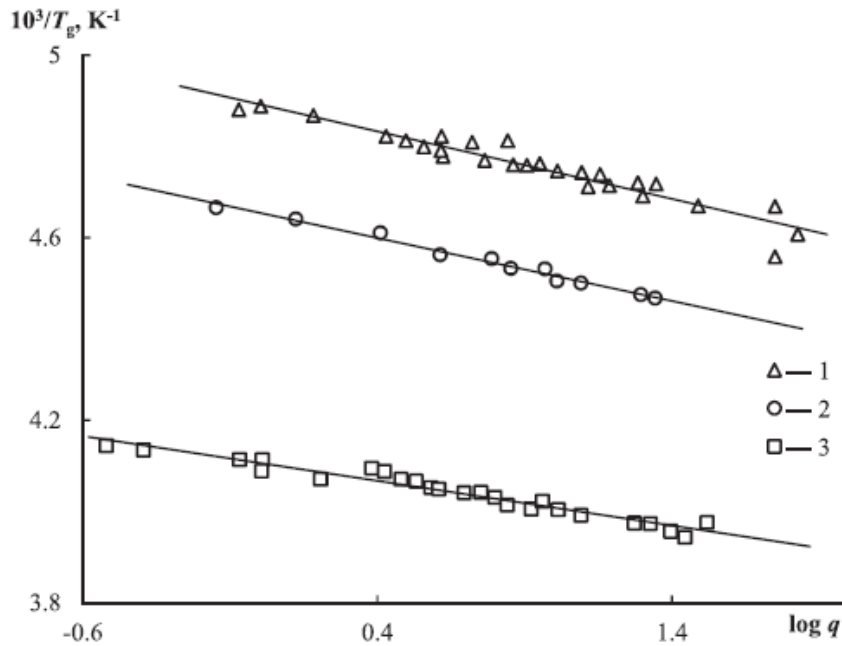


Fig. 1.2. Dependence of the reciprocal value of T_g in the logarithm of cooling rate of rubbers SKS-30 (1), SKN-18 (2) and SKN-40 (3) [44].

$$\frac{a_2}{a_1} \approx \text{const} \approx 0.027 \div 0.035.$$

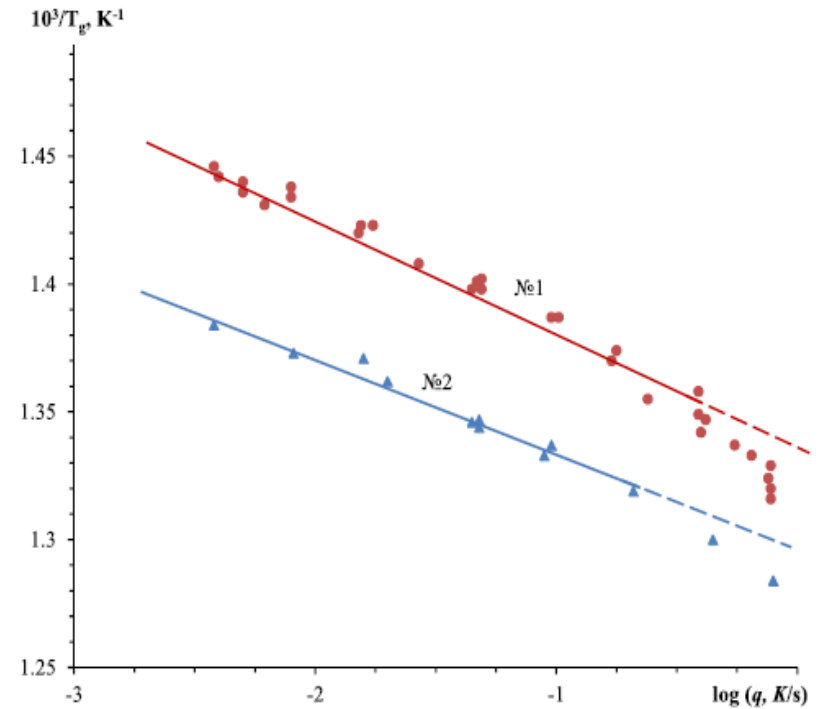


Fig. 1.3. Dependence of T_g on cooling rate of melt for lead-silicate glasses № 1 and № 2 in coordinates $1/T_g - \log q$ according to data from Bartenev and Lukyanov [45].

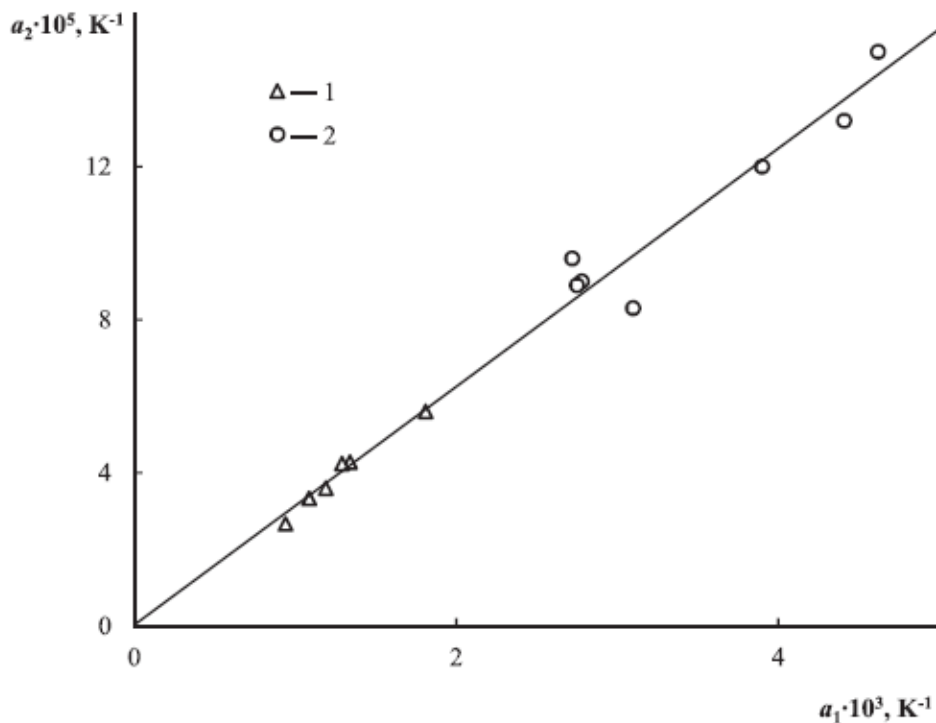


Fig. 1.4. Linear correlation between Bartenev equation parameters a_1 and a_2 . 1 – inorganic silicate glasses; 2 – amorphous organic polymers.

$$\frac{1}{T_g} = a_1 - a_2 \ln q$$

$$\frac{a_2}{a_1} \approx \text{const} \approx 0.027 \div 0.035.$$

Weak dependence: e.g. a changes of q by a order of magnitude (10 times) typically result in about 3% change of T_g ($\Delta T_g/T_g \sim 0.03$)

Bartenev equation parameters (1.8) and fraction of fluctuation volume f_g for anoxic and a number of oxide glasses [49].

Glass	T_g, K	$a_1 \cdot 10^3, K^{-1}$	$a_2 \cdot 10^5, K^{-1}$	a_2/a_1	f_g
Se-Bi	309	3.12	10.45	0.034	0.031
Se-Ga	315	3.09	9.29	0.030	0.027
In-Se	316.7	3.09	5.54	0.018	0.017
As-Sb-Se	444.8	2.15	6.74	0.031	0.028
GeO ₂	762	1.23	3.45	0.028	0.026
P ₂ O ₅ -TeO ₂	578	1.67	4.99	0.030	0.027
SiO ₂ -Al ₂ O ₃ -B ₂ O ₃ -P ₂ O ₅ -MgO-Na ₂ O-K ₂ O	1064	0.89	3.20	0.036	0.032

Theories

Mandelstamm–Leontovich theory

$$V = V(P, T, \xi)$$

$$\frac{d\xi}{dt} = -\frac{1}{\tau}(\xi - \xi^e)$$

$$|q| \cdot \tau_g = \Delta T_0$$

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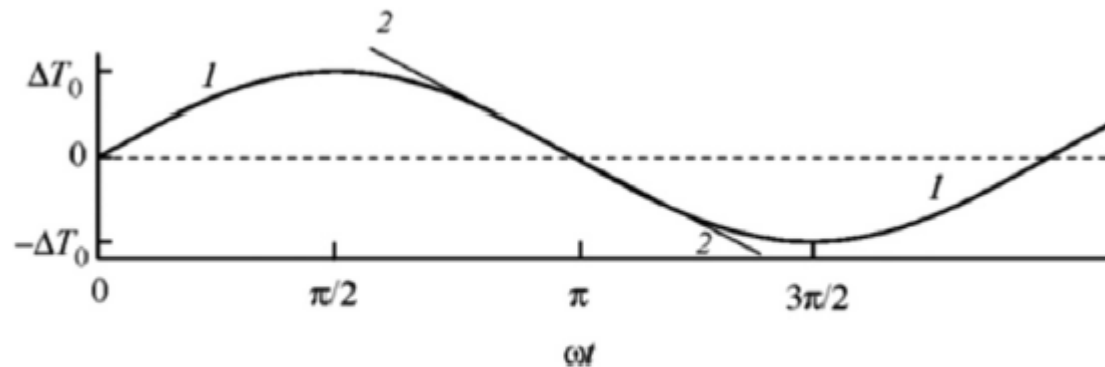


Fig. 2.1. To the explanation of the interrelation between periodic temperature change and its approximation in the form of linear function on certain intervals. 1 – sinusoid, Eq. (2.5); 2 – linear parts of the curve as corresponding to approximation $dT/dt = \Delta T_0 \cdot \omega \cdot \cos(\omega t)$ (see text).

Relaxation theory of Volkenshtein–Ptitsyn

$$\frac{dn_2}{dT} = -\frac{1}{q\tau}(n_2 - n_{20}).$$

Condition for glass transition

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

$$|q|\tau_g = \frac{1}{\Psi(T_g)}$$

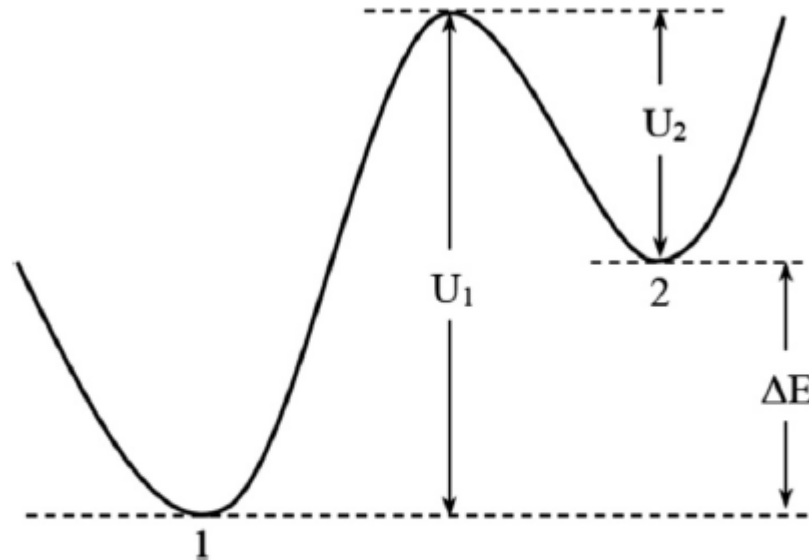


Fig. 3.1. The theoretical scheme of Volkenshtein-Ptitsyn [6]. 1 – ground state of the particle, 2 – its excited state. U_1 and U_2 – kinetic barriers for the $1 \rightarrow 2$ and $2 \rightarrow 1$, respectively, $\Delta E = (U_1 - U_2)$, the energy of the excited state is greater than the energy of the ground state by an amount ΔE .

Configuron percolation theory

A configuron is an elementary configurational excitation in an amorphous material, formed by breaking of a chemical bond and the associated strain-releasing local adjustment of centres of atomic vibration. Formation of a broken bond can be represented as a reaction when a lattice phonon is absorbed by a bond resulting in a un-relaxed broken bond which after relaxation of the surrounding environment results in formation of a quasiparticle termed a configuron (o):

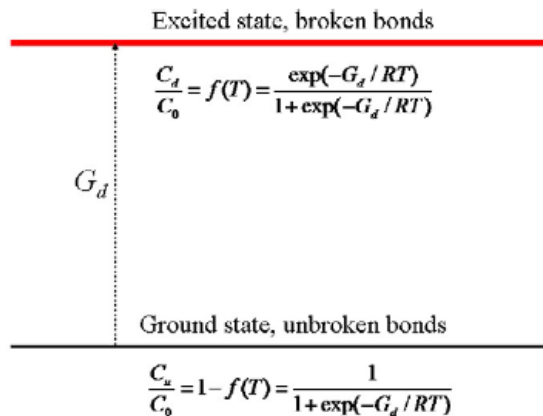
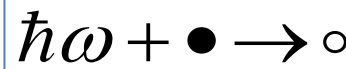


Figure 2. Two-level state equivalent to disordered system of bonds of an amorphous material.

J. Phys.: Condens. Matter 18 (2006) 11507–11520

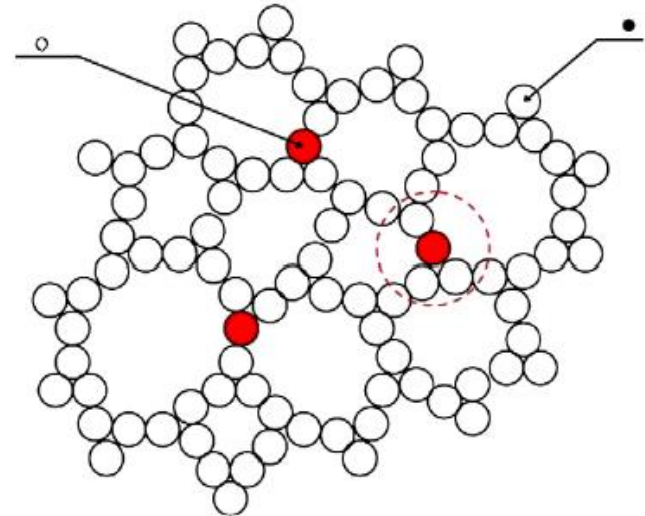


Fig. 2. CBL of A_2B_3 with several broken bonds (non-relaxed configurons). The system is shown before structure relaxation which will involve rearrangements near the broken bond (dashed circle).

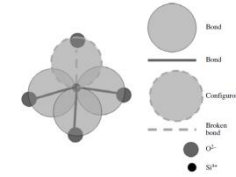


Fig. 3. Schematic diagram of the replacement of the configurational tetrahedron in silica by the bond tetrahedron. A configuran corresponds to the breaking of one bond in silica.

$$T_g = \frac{H_d}{S_d + R \ln[(1 - f_c)/f_c]}$$

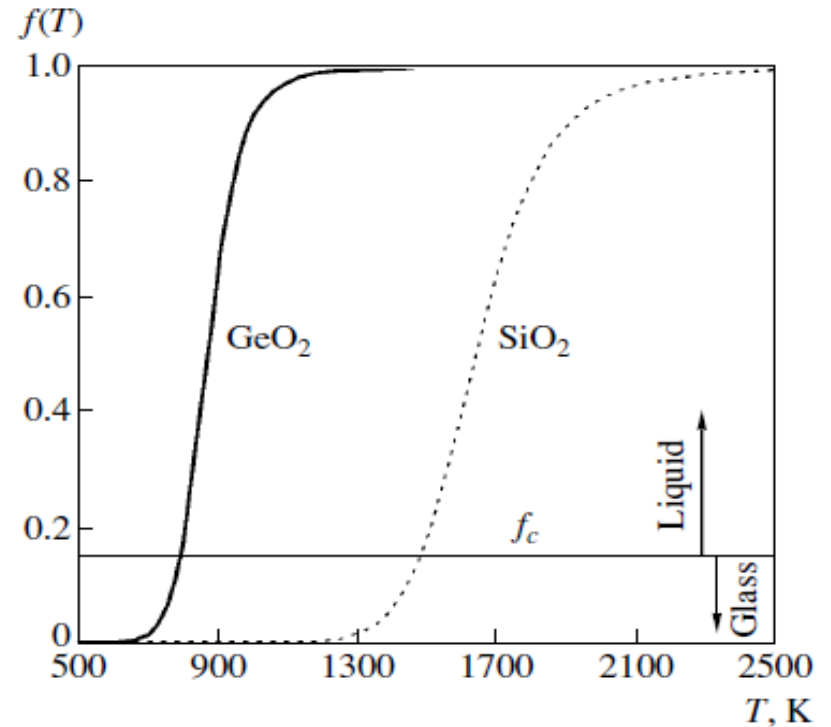
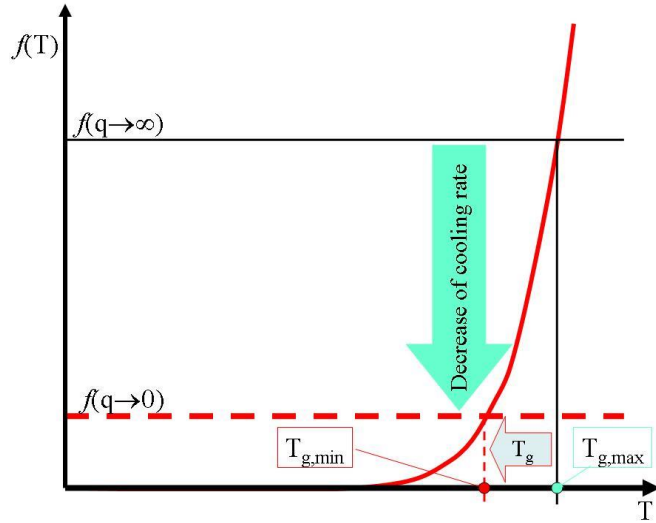


Fig. 4. Degree of brokenness of bond networks in GeO₂ and SiO₂ as a function of temperature.

$$\frac{1}{T_g} = \frac{S_d}{H_d} + \frac{[\ln(1/\vartheta_c) + \ln((V_c - V_d)/V_d)]R}{H_d} - \frac{mR}{H_d} \ln\left(\frac{q}{q_0}\right)$$

$$\frac{1}{T_g} = a_1 - a_2 \ln q$$

Estimation of δT_g

Williams-Landel-Ferry equation for relaxation and viscosity

$$\ln a_T = -C_1 \frac{T - T_g}{T - T_g + C_2}, \quad a_T = \frac{\tau(T)}{\tau(T_g)} \cong \frac{\eta(T)}{\eta(T_g)}, \quad \left(\frac{d\tau}{dT} \right)_{T=T_g} = -\frac{1}{|q|}$$

$$q\tau_g = \frac{C_2}{C_1},$$

from which follows the formula for calculating δT_g using parameters of the WLF equation C_1 and C_2

$$\delta T_g = \frac{C_2}{C_1}.$$

$$\delta T_g = (8 \div 12) \text{ K}$$

Fragility and δT_g

$$\delta T_g = \left(\frac{1}{m} \right) T_g$$

$$m = \left. \frac{d \ln \eta(T)}{d(T_g/T)} \right|_{T=T_g}$$

Sanditov equation

$$\frac{\delta T_g}{T_g} \cong \frac{f_g}{\ln(1/f_g)}$$

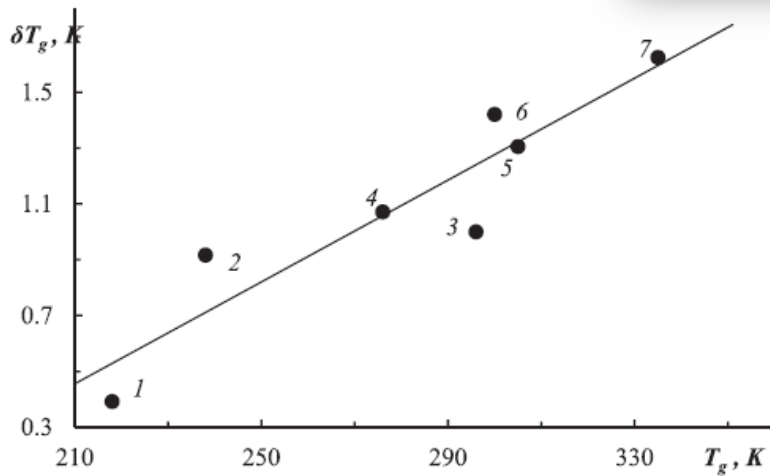


Fig. 7.2. Linear correlation between δT_g and T_g for amorphous organic polymers. 1 – polyhexene-1, 2 – polyurethane, 3 – polyvinylchloroacetate, 4 – polymethylacrylate, 5 – polyvinyl acetate, 6 – natural rubber, 7 – methacrylate ethyl. Data from [36].

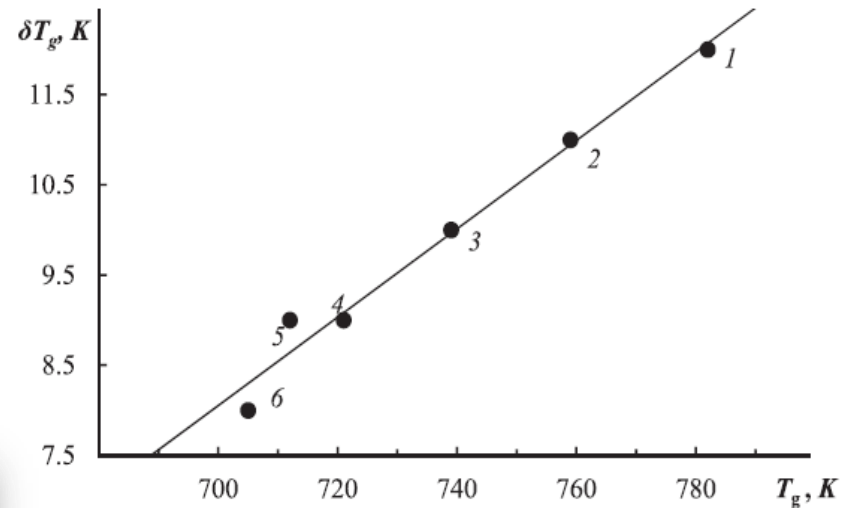


Fig. 7.1. Linear correlation between δT_g and T_g for sodium silicate glasses. Content Na₂O, mol%: 1 – 15, 2 – 20, 3 – 25, 4 – 30, 5 – 33, 6 – 35. Data from [49].

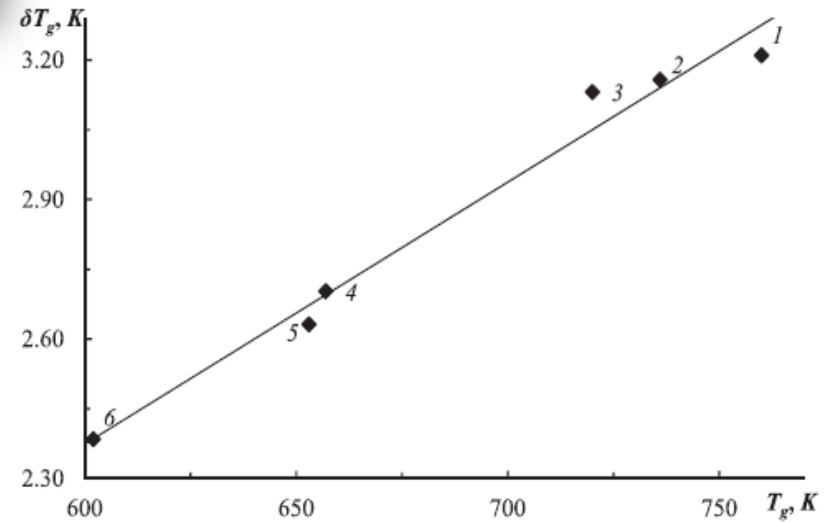


Fig. 7.3. Correlation between δT_g and T_g for metallic glasses. Data from [77,78]. 1 – Fe₈₃B₁₇, 2 – Fe₈₀P₁₃C₇, 3 – Fe_{41.5}Ni_{41.5}B₁₇, 4 – Pd₈₂Si₁₈, 5 – Pd_{77.5}Cu₆Si_{16.5}, 6 – Pd₄₀Ni₄₀P₂₀.

Nemilov estimations

$$\delta T_g = - \left(\frac{\partial T}{\partial \lg \eta} \right)_{T=T_g} = - \left(\frac{\Delta T}{\Delta \lg \eta} \right) = \frac{T_{12} - T_{13}}{\lg \eta_{12} - \lg \eta_{13}}$$

Overestimated data $\delta T_g = (T_{12} - T_{13}) \cong 20 \text{ K}$

Volkestein-Ptitsyn estimations

$$\delta T_g = \frac{1}{2.3} (T_{12} - T_{13}) = (6 - 13) \text{ K}$$

Calculation of temperature range δT_g for silicate glasses and amorphous polymers.

Glass (content, mol%)	$T_{12},$ K	$T_{13},$ K	$\delta T_g, \mathbf{K}$ $\delta T_g = T_{12} - T_{13}$	$\delta T_a, \mathbf{K}$ $\delta T_g = \frac{1}{2.3}(T_{12} - T_{13})$	$\delta T_g = \frac{C_2}{C_1} \mathbf{K}$ (6.8)	C₁	C₂, K	$\tau_g,$ c
Sodium-silicate glasses Na₂O – SiO₂ [49]								
15 Na ₂ O–85 SiO ₂	819	790	29	13	12	36	430	239
20 Na ₂ O–80 SiO ₂	792	766	26	11	11	36	390	217
25 Na ₂ O–75 SiO ₂	769	745	24	10	10	35	355	202
30 Na ₂ O–70 SiO ₂	749	727	22	10	9	35	322	184
33 Na ₂ O–67 SiO ₂	738	717	21	9	9	35	304	174
35 Na ₂ O–65 SiO ₂	726	705	21	9	8	35	291	166
Window glass [19]	846	825	21	9	8	36	305	160
Poly-alkali silicate glass [76]								
69.04SiO ₂ ·30.96Na ₂ O	736	718	18	8	7	46	340	147
79.29SiO ₂ ·12.97Na ₂ O·7.75Li ₂ O	700	683	17	7	7	45	315	140
43.22SiO ₂ ·9.55Na ₂ O·47.23CsO	721	704	17	7	6	31	200	129
71.59SiO ₂ ·24.4Na ₂ O·4.01Li ₂ O	695	681	14	6	6	36	231	128
Amorphous polymers [58]								
Polyisobutylene	–	202	6	–	2.7	38	104	54
Polyvinyl acetate	–	305	9	–	1.3	36	47	26
Polyvinylchloroacetate	–	296	9	–	1.0	40	40	20
Polymethyl acrylate	–	276	8	–	1.1	42	45	22
Polyurethane	–	238	7	–	0.9	36	33	18
Natural rubber	–	300	9	–	1.4	38	54	57
Methacrylate polymers								
ethyl	–	335	10	–	1.6	40	65	32
n-butyl	–	300	9	–	2.5	39	97	50
n-octyl	–	253	8	–	2.9	37	107	58

Note: Composition of window glass (mass%) [40]: SiO₂ – 72.7; CaO – 8.6; MgO – 3.4; Al₂O₃ – 1.3; Na₂O – 13.6; K₂O – 0.4; $\tau_g = C_2 / qC_1$, C_1 и C_2 – parameters of the Williams-Landel-Ferry equation, $q = 0.05$ K/s.

inorganics

$$q\tau_g \approx (5 - 10) \text{ K}$$

organics

$$\delta T_g \approx (1-3) \text{ K}$$

Model of delocalized atoms

D.S. Sanditov, J. Exp. Theor. Phys. 115 (2012) 112–124.

Fluctuation volume

$$\Delta V_e = N_e \Delta v_e.$$

Fraction of fluctuation volume

$$f = \frac{\Delta V_e}{V}$$

$$\frac{\delta T_g}{T_g} \approx \frac{f_g}{\ln(1/f_g)}$$

WLF constants and delocalized atoms model:

$$C_1 = \frac{1}{f_g}, \quad C_2 = \frac{f_g}{\beta_f}, \quad f_g = (\Delta V_e/V)_{T=T_g} \text{ is the fraction of fluctuation volume frozen}$$

The fraction of fluctuation volume frozen at glass transition is practically a constant

$$f_g = \left(\frac{\Delta V_e}{V} \right)_{T=T_g} = \frac{1}{C_1} \approx \text{const} \approx 0.024 - 0.028$$

A double-exponent equation for viscosity

$$\eta = \eta_o \exp \left\{ \frac{\Delta F_\infty}{kT} + \left[\exp \left(\frac{\Delta \varepsilon_e}{kT} \right) - 1 \right] \right\}$$

WLF equation parameters C_1 , C_2 and the glass transition characteristics of amorphous substances.

Amorphous substance	T_g , K	C_1	C_2 , K	$\delta T_g = \frac{C_2}{C_1}$, K	f_g	$\frac{\delta T_g}{T_g} \cdot 10^3$	$\delta T_g \cdot K$ $\frac{\delta T_g}{T_g} \cong \frac{f_g}{\ln(1/f_g)}$	τ_g , c	C_0	$\left(\frac{N_g}{N}\right)_{T=T_g}$, %
Sodium silicate glass Na₂O–SiO₂ [49]										
Na ₂ O, mol%										
15	782	36	430	12	0.028	7.8	6.1	240	0.5	3.5
20	759	36	390	11	0.028	7.8	5.9	220	0.5	3.4
25	739	35	355	10	0.028	7.8	5.8	200	0.5	3.5
30	721	35	322	9	0.028	7.8	5.6	180	0.5	3.5
33	712	35	304	9	0.028	7.8	5.6	180	0.6	3.5
35	705	35	291	8	0.028	7.8	5.5	160	0.6	3.5
Metallic glass (amorphous alloys) [77]										
Pd ₄₀ Ni ₄₀ P ₂₀	602	39	93	2.4	0.026	7.1	4.3	48	0.8	3.6
Pt ₆₀ Ni ₁₅ P ₂₅	500	37	95	2.6	0.027	7.5	3.7	52	0.8	3.6
Pd _{77.5} Cu ₆ Si _{16.5}	653	38	100	2.6	0.026	7.1	4.6	52	0.8	3.7
Fe ₈₀ P ₁₃ C ₇	736	38	120	3.2	0.026	7.1	5.2	64	0.8	3.6
Amorphous organic polymers [58] and selenium [5]										
Polyvinyl acetate	305	36	47	1.3	0.028	7.8	2.4	61	0.8	3.5
Natural rubber	300	38	54	1.4	0.026	7.1	2.1	57	0.8	3.6
Methacrylate ethyl	335	40	65	1.6	0.025	6.8	2.3	50	0.8	3.6
Selenium	303	32	58	1.8	0.031	8.9	2.7	44	0.8	3.6
Low molecular weight organic glass [61]										
Propanol	98	41	25	0.6	0.024	6.4	0.6	12	0.7	3.6
Prothylene Glycol	160	44	40	0.9	0.023	6.1	1.0	18	0.7	3.7
Glycerol	185	42	53	1.3	0.024	6.4	1.2	26	0.7	3.9

Note: $f_g = 1/C_1$, $\tau_g = C_2/C_1q$.

Sanditov equation of viscosity

D.S. Sanditov, JETP 110 (2010) 675–688.

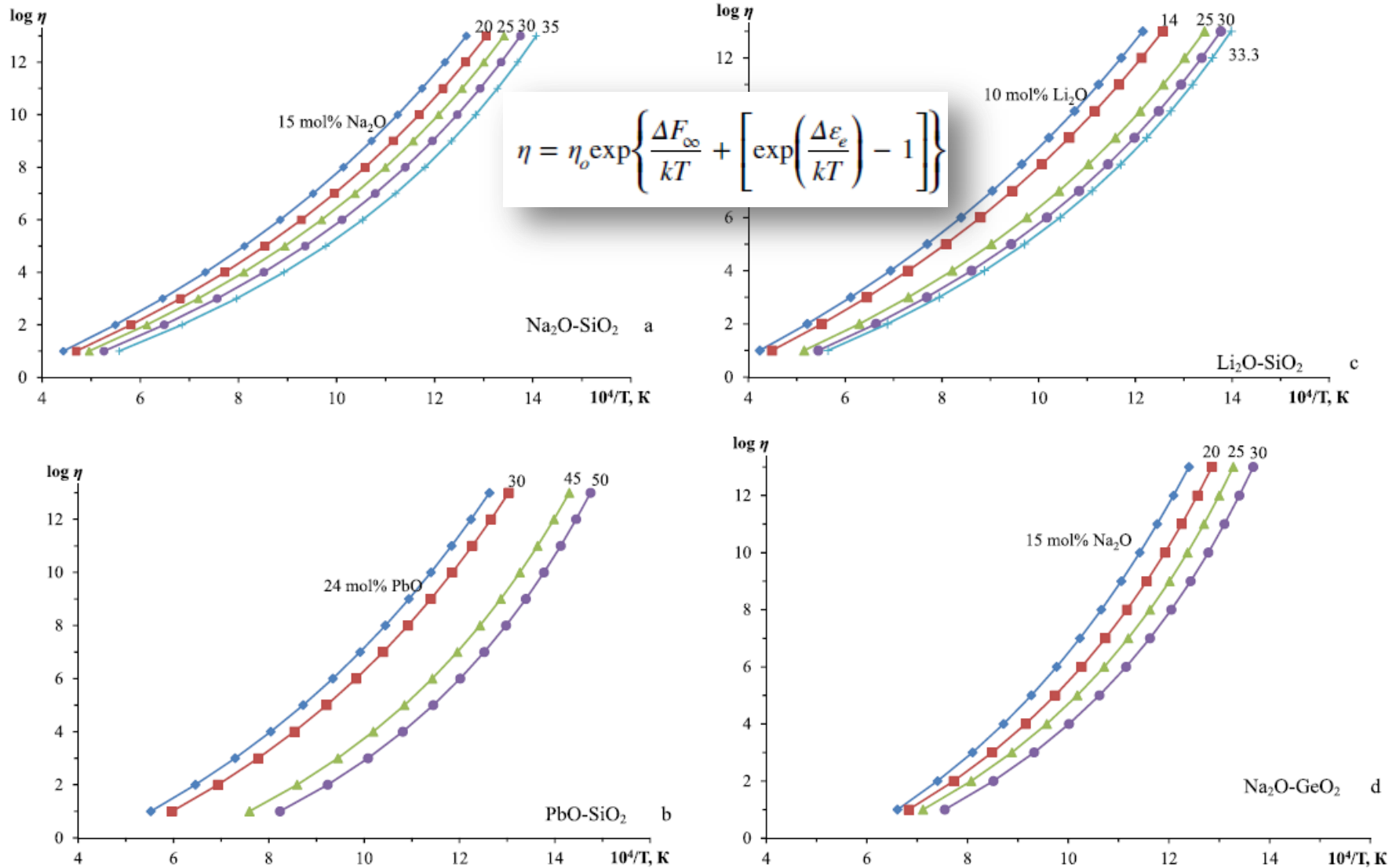


Fig. 12.2. Temperature dependence of the viscosity of sodium silicate (a), lead silicate (b), lithium silicate (c) and sodium germanate (d) glasses. Points – experimental data, curves – calculation by the Eq. (12.11).

Characteristics of viscous flow of inorganic glasses R_2O-SiO_2 ($R = Li, Na, K$), $PbO-SiO_2$, Na_2O-GeO_2 and $Na_2O-B_2O_3$.

R_2O ($R =$ Li, Na, K), PbO, mol%	$-\log$ η_0 [Π]	$\Delta\epsilon_e$, kJ/mole	ΔF_∞ , kJ/mole	T_g , K	ΔF_η (T_g), kJ/ mole	f_g	$\Delta\epsilon_e$, kJ/mole (13.8)
Li ₂ O	Li ₂ O-SiO ₂						
10	2.55	20	127	814	245	0.028	24
14	2.57	20	120	788	237	0.028	23
25	2.41	19	91	738	219	0.028	22
30	2.25	19	78	721	212	0.028	21
33,3	2.23	19	71	708	208	0.028	21
Na ₂ O	Na ₂ O-SiO ₂						
15	2.53	19	118	783	235	0.028	23
20	2.35	19	101	759	225	0.028	23
25	2.36	19	94	739	219	0.028	22
30	2.36	19	87	721	214	0.028	21
33	2.26	19	78	712	209	0.028	21
K ₂ O	K ₂ O-SiO ₂						
13	2.33	19	121	795	235	0.028	24
15	2.31	19	117	793	232	0.028	24
20	2.14	19	100	759	222	0.025	23
25	2.22	19	90	739	217	0.025	23
PbO	PbO-SiO ₂						
25	3.15	21	100	785	245	0.027	24
30	2.95	21	79	761	234	0.027	23
45	3.34	20	57	696	218	0.027	21
50	3.33	19	42	674	212	0.027	20
Na ₂ O	Na ₂ O-Ge ₂ O						
15	3.81	22	79	801	259	0.026	24
20	3.21	22	58	773	241	0.027	23
25	3.10	21	49	749	232	0.027	22
30	3.12	21	40	727	225	0.027	22
Na ₂ O	Na ₂ O-B ₂ O ₃						
10	3.45	18	47	618	194	0.026	19
15	3.93	20	44	680	221	0.026	21
20	3.41	21	26	727	229	0.026	22
25	3.77	22	19	735	237	0.026	22
30	4.45	23	3	748	250	0.025	23

$$\eta = \eta_0 \exp \left\{ \frac{\Delta F_\infty}{kT} + \left[\exp \left(\frac{\Delta \epsilon_e}{kT} \right) - 1 \right] \right\}$$

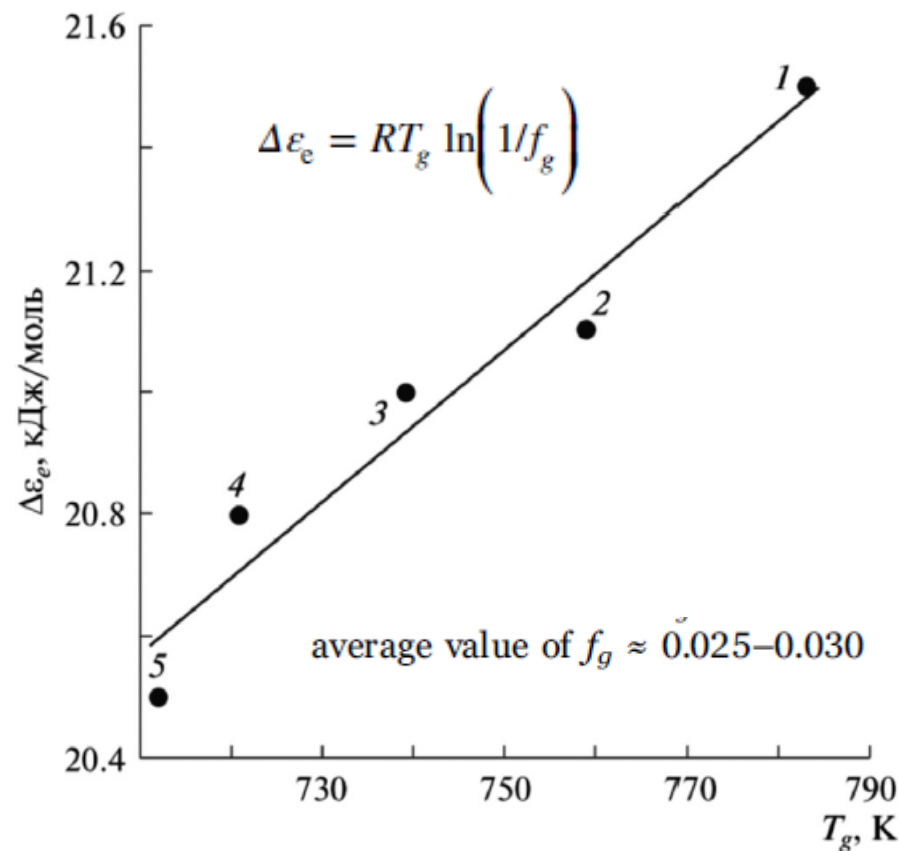


Fig. 13.4. Linear correlation between atom delocalization energy ($\Delta\epsilon_e = RD$) and glass transition temperature (T_g) of sodium-silicate glasses Na_2O-SiO_2 . Content of Na_2O , mol %: 15(1); 20(2); 25(3); 30(4).

Configuron percolation theory

$$\eta(T) = A_1 T \left[1 + A_2 \exp\left(\frac{H_m}{RT}\right) \right] \left[1 + C \exp\left(\frac{H_d}{RT}\right) \right]$$

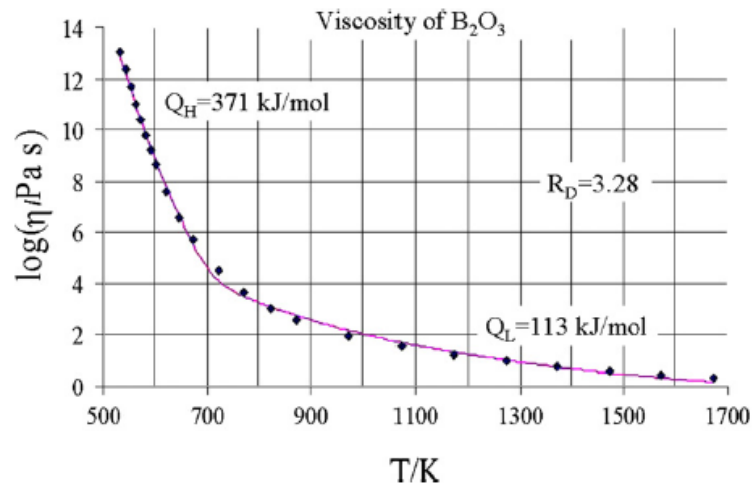
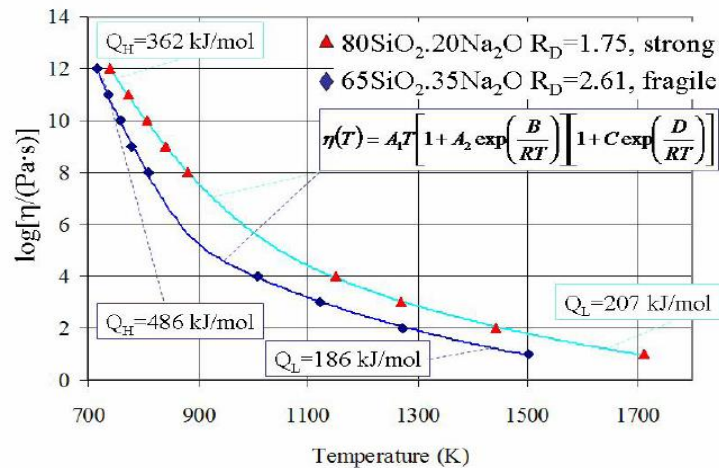
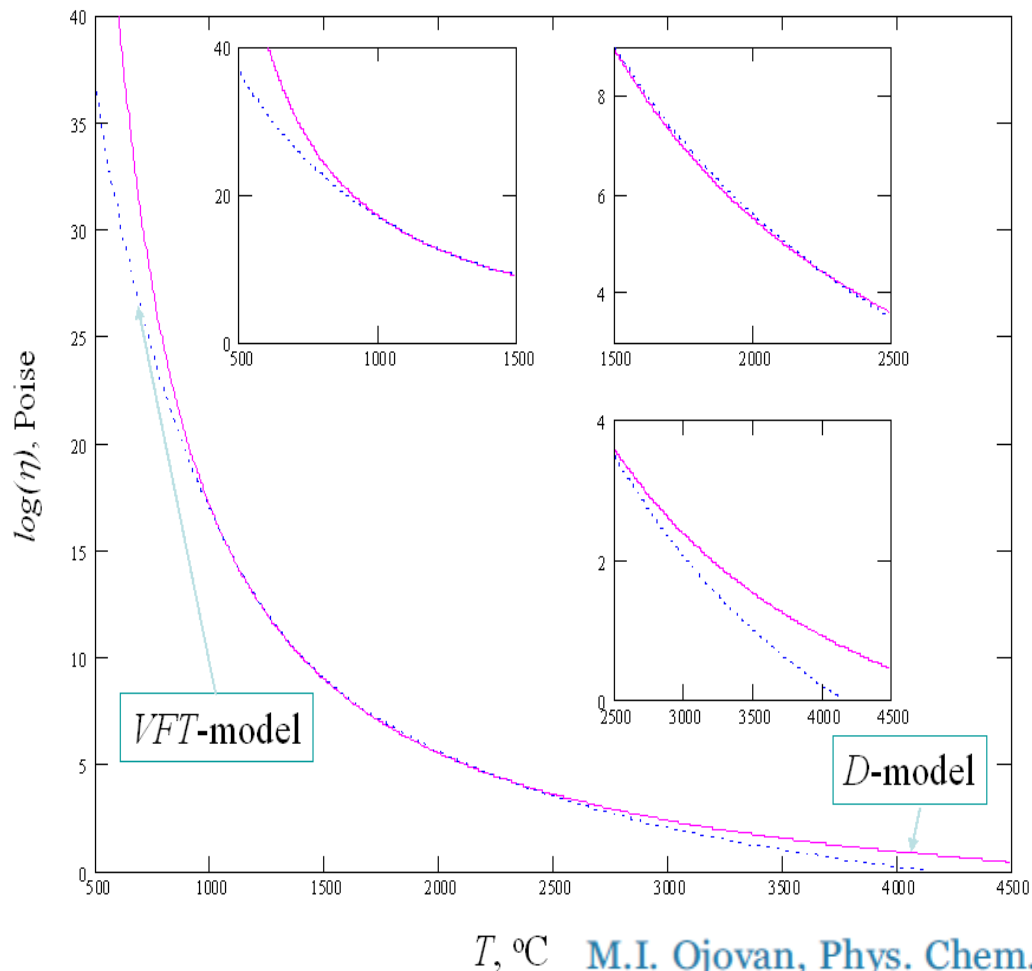


Fig. 4. Viscosity of amorphous B₂O₃.

Kinetic criterion of glass transition

Gutzov suggested the criterion: $\left(\frac{1}{T} \left| \frac{dT}{dt} \right| \tau \right) \Big|_{T=T_g} = C_3$, with $C_3 \approx 1$.

It gives unrealistic $q\tau_g \approx 800 \text{ K}$ (Nemilov).

It is suggested: $\frac{q\tau_g}{T_g} = C_g$,

where value of C_g is actually a universal constant

$$C_g = \frac{\delta T_g}{T_g} = \frac{f_g}{\ln(1/f_g)} \approx \text{const} \approx 0.007.$$

In terms of Gutzov equation e.g.:

$$\left(\frac{1}{T} \left| \frac{dT}{dt} \right| \right) \tau \Big|_{T=T_g} = C_g, \quad C_g \approx 7 \cdot 10^{-3}$$

Generalized Bartenev equation

$$\frac{1}{T_g} = a_1 + b_1 \ln\left(1 - \frac{\ln q}{b_2}\right)$$

$b_1 = R/\Delta\varepsilon_e$; $b_2 = \ln(C/\tau_0)$; $a_1 = b_1 \ln b_2$ $\tau_0 \approx 10^{-12}$ s and $C \approx 10$ K $b_2 = \ln(C/\tau_0) \approx 30$

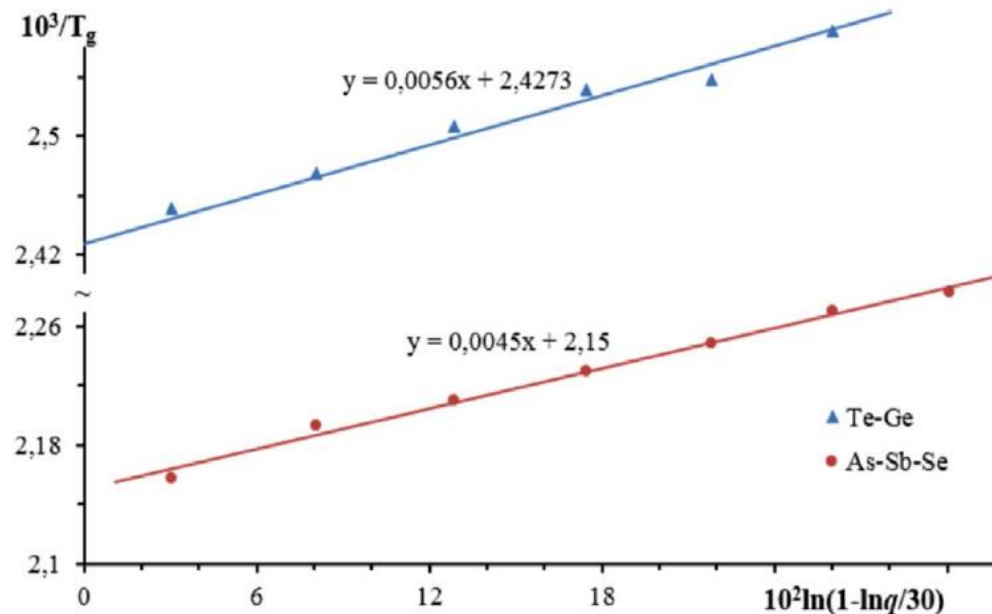


Fig. 11.1. Dependence of T_g on cooling rate for glasses As-Sb-Se and Te-Ge in the coordinates of Eq. (11.3): $1/T_g - \ln[1 - (\ln q/30)]$. Data from [49]: (Mahadevan S., 1986 and Lasocka M., 1976). Parameters of Eq. (11.3) for As-Sb-Se: $a_1 = 2,1 \cdot 10^{-3} \text{ K}^{-1}$, $b_1 = 4,5 \cdot 10^{-3} \text{ K}^{-1}$; and for Te-Ge: $a_1 = 2,4 \cdot 10^{-3} \text{ K}^{-1}$, $b_1 = 5,6 \cdot 10^{-3} \text{ K}^{-1}$.

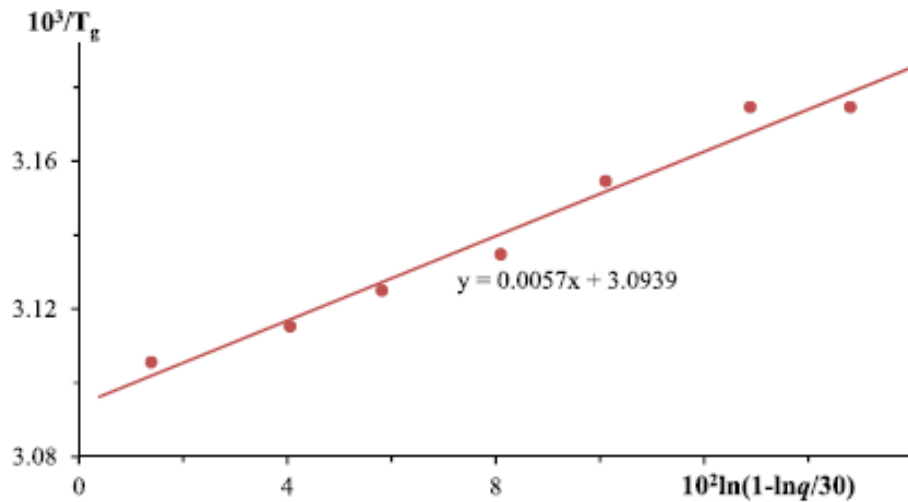


Fig. 11.2. Dependence $1/T_g$ on $\ln(1 - \ln q/30)$ for Se-Ga glass. Data from [49]: (El-Oyoun M.A., 2003). Parameters of Eq. (11.3): $a_1 = 3 \cdot 10^{-3} \text{ K}^{-1}$, $b_1 = 5,7 \cdot 10^{-3} \text{ K}^{-1}$.

Sanditov equation

$$\frac{1}{T_g} = a_1 + b_1 \ln\left(1 - \frac{\ln q}{b_2}\right)$$

When $\ln q \ll 30$ this reduces to original Bartenev equation

$$\frac{1}{T_g} = a_1 - a_2 \ln q$$

$$a_2 = b_1/b_2.$$

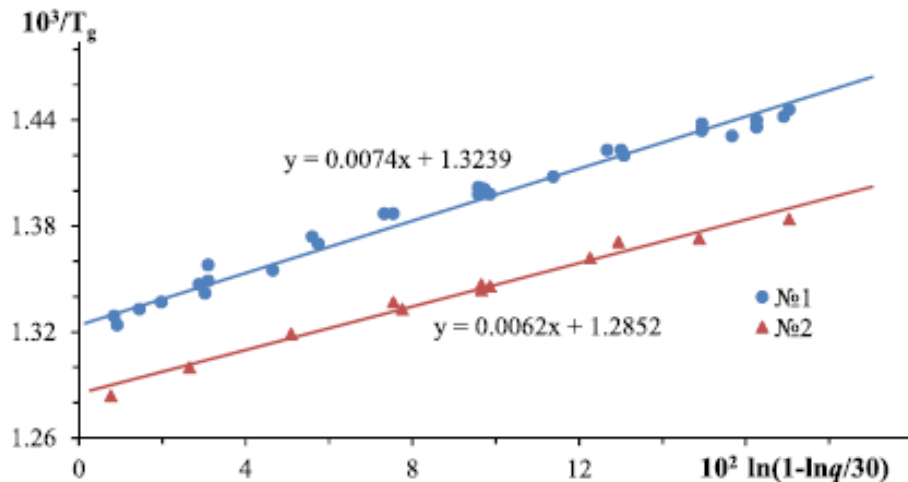


Fig. 11.3. Dependence of the glass transition temperature on the heating rate for lead silicate glasses №1 and №2 in coordinates $1/T_g - \ln(1 - \ln q/30)$. Data from [45]. Parameters of Eq. (11.3) for glass № 1: $a_1 = 1,3 \cdot 10^{-3} \text{ K}^{-1}$, $b_1 = 7,4 \cdot 10^{-3} \text{ K}^{-1}$; and glass № 2: $a_1 = 1,2 \cdot 10^{-3} \text{ K}^{-1}$, $b_1 = 6,2 \cdot 10^{-3} \text{ K}^{-1}$.

Conclusions

- Glass transition of liquid has a pronounced relaxation character and obeys kinetic laws.
- The relation between structural relaxation time τ and cooling rate $q = dT/dt$ is expressed by glass transition equation: $q\tau_g = \delta T_g$.
- Calculation of temperature interval δT_g using the data on parameters of Williams-Landel-Ferry equation and the model of delocalized atoms for inorganic oxide glasses is in an agreement with the product $q\tau_g \approx (5 - 10) K$.
- An analogous statement is true for organic glasses, whose δT_g have lower values $\delta T_g \approx (1 - 3) K$.
- Bartenev equation is true at relatively low cooling rates $\ln q \ll 30$.

- Fraction of the fluctuation volume f_g , calculated from the data on the dependence of T_g on cooling rate of the melt coincides with calculation of f_g from the data on viscosity near the glass transition region.
- A refined version of kinetic criterion of glass transition is proposed.
- Excited delocalized atoms are responsible for viscous flow of glass-forming materials, their concentration during cooling decreases and in the glass transition region reaches negligibly small values (around 3%), which is equivalent to freezing.

