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Experimental investigations of varying the temperature parameters in the glass-transition range for glass-metal composites when heated

O.N. Lyubimova ✉, **M.A. Barbotko**, **A.A. Streltcov**

Far Eastern Federal University, Vladivostok, Russia

✉ berms@mail.ru

Abstract. The article is concerned with the preliminary experimental investigation of varying the linear extension of glass in composition of the glass-metal composite on basis of steel 20 (st20) and glass S52-1 when heated to transition temperature from the solid state to the high viscous one. The characteristic temperatures of glass-transition (T_g) and softening (T_f) and the glass transition from the solid state to the high viscous one (ΔT_g) were determined. In order to determine the basic parameters in the approximated model of the coefficient of linear thermal expansion, the linear rate of variation was fixed in the vicinity of the glass-transition temperature and softening temperature. The simulation was based on the Williams-Landel-Ferry relation and Sanditov's model of delocalized atoms. The statistical processing of results was carried out using small samples, and the experimental investigation of changing the temperature parameters under conditions of varying the rate of heating was planned.

Keywords: glass-transition temperature, thermal expansion coefficient, glass-metal composite

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Introduction

The application of the glass-metal connections in the instrument making industry, production of new composite and construction materials on basis of glass and metal and problems of determining the thermophysical and mechanical properties in the temperature interval of glass transition in the similar connections from the liquid state to the solid one under cooling (heating) determine the actuality of their experimental investigations [1–5]. When producing the glass-metal connections with the use of methods of the heat treatment, the contact is created by way of drawing together of jointing surfaces as a result of plastic deformation when heated one or both connectable elements [6–8]. The description of the regularities of contact production, methods of regulating the mechanical properties of connections and behavior under the subsequent temperature loads is associated among other things with the study of varying the properties within a wide temperature range. For the glass component of connection, the temperature interval (ΔT_g) in which the processes of softening (liquidity) and glass-transition take place is the interval in which many properties of glass vary most hardly. On the part of high temperatures, it is limited to the temperature of liquidity (T_f) while; on the part of low temperatures to the glass-transition temperature (T_g) (Fig. 1). The glass-transition temperature and zone ($\delta T_g = T_{12} - T_{13}$, T_{12} and T_{13} are temperatures corresponding to the viscosity

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$\eta(T) = 10^{12}$ and 10^{13} $(Pa \cdot s)$) are the most frequent and convenient criterion for analysis of properties of glasses [9–14].

The searches of correct computing methods of T_g and δT_g for different glasses are the urgent problems and the articles [14-18] are concerned with the review of main methods. The peculiarities of the glass deformation in the $\Delta T_g = T_f - T_g$ interval (from experiments in elongation of the glass thread at the constant load [4]) are composed of three main deformation components: totally reversible instantaneously elastic; partially reversible slow elastic and totally irreversible of viscous flow. It was experimentally proved that the basic external factors determining the glass formation and, accordingly, such important mechanical characteristics as the temperature coefficient of linear extension, elastic modulus and stressed-deformed state are the rates of sample cooling and heating (q) and its sizes [8,19,20]. The experimental and theoretical investigations of the basic parameters of glass transition are carried out for glasses of different compositions. The number of the works on studying the parameters of glass transition in the glass in composition of the glass-metal connections under conditions of the formed special stressed state is precious little [21,23] due to the theoretical difficulties and absence of the unified theory for analytic description of the glass-transition phenomenon and construction of the correlation dependences between its parameters. The strong majority of the experimental data belongs to the standard rate of cooling $(q = 3\ Kmin^{-1} = 0.05\ K\ s^{-1})$, however, the real technology processes demand to study the wider interval of the rate variation together with the influence of the dependence of the heating (cooling) rate on temperature.

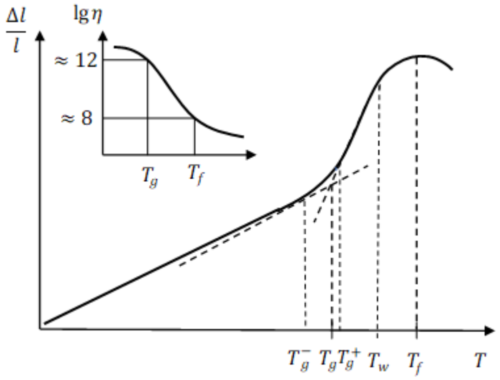
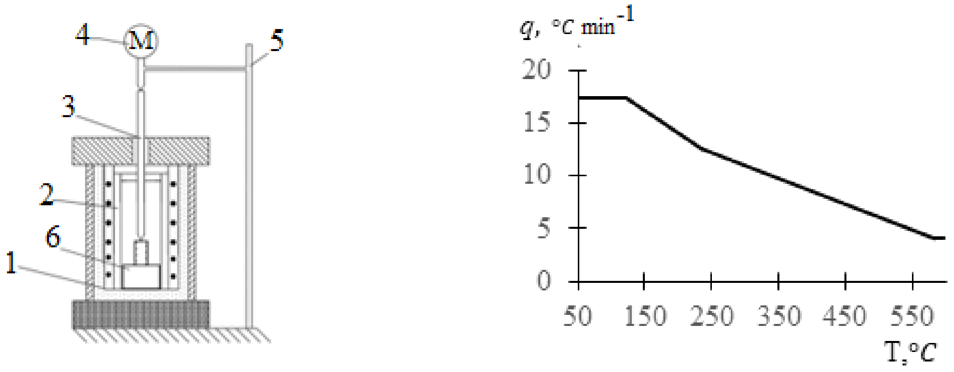


Fig. 1. Action of temperature on the linear sizes of glass and temperature dependence of viscosity with the mean value in the characteristic points

Experimental method of investigation

The experimental investigations of parameters in the process of forming the physical contact between the glass and steel in the course of soldering in the technology of producing the cylindrical soldered joints and glass-metal composite stem [8] in the plant by type of the cargo viscometer with prescribed loading have shown that, when measuring the shrinkage under heating and postcooling, the change in the rate of the linear size of sample and temperature $T_f$are fixed in the interval corresponding to $\Delta T_g = T_f - T_g$. When determining the parameters of the mechanical relaxation in the connections of glass with other materials, the characteristics of varying the linear sizes are the critical parameters, for which reason the problem of developing the method of the experimental determination of the following values $(T_g, \delta T_g, \Delta T_g$ and $T_f)$ was set.

The determination of the listed parameters for the glass-metal composite was carried out by the method of the thermal linear expansion. The samples in the form of round bars with the diameter of 10 mm (diameter of the core out of glass S52-1 and is 8 mm and depth of the metal enclosing envelope made of steel 20 is 1 mm) and with the length of 50 mm were located into the lower part of the piped muffle furnace with temperature deviation of not more than 1 °C (Fig. 2) along the length of the allocated sample, quartz stem transmitting the readings of elongation was arranged exactly to diameter of the glass core. The error in measurement of temperature reached 0.5 °C while the accuracy of the sample elongation registering was 1 μm. The rate of the sample heating was aligned with the diagram in Fig. 2(c).



(a) (b)

Fig. 2. (a) Scheme of plant. 1– muffle furnace, 2– sample, 3– quartz stem, 4– micrometric indicator Calibron 132002– (M), 5– support, 6- quartz rest; (b) rate of temperature variation when heated (q, °C min^{-1})

Discussion of results

For comparison with data obtained for the glass-metal composite, the analogous experiments were carried out for the glass (S52-1) samples with the diameter of 8 mm and length of 50 mm. For exclusion of the thermal prehistory, the samples were previously annealed within 30 minutes at a temperature exceeding the softening temperature T_ω ($550\ °C$) by 20-30 °C, afterward cooled with constant rate not exceeding 3 °C · min^{-1} to the temperature lower by 100 °C than the above temperature, thereafter the cooling of samples was carried out with the rate of 15 °C· min^{-1}. For computing the average coefficient of the linear temperature expansion (α) in any interval of temperatures $T_1 - T_2$, it is necessary to divide the difference of the values of relative elongation $(\Delta l/l)_{T_2} - (\Delta l/l)_{T_1}$ by the consistent interval of temperatures $T_2 - T_1$ [24,25]. In Figure 3, the results of determining the coefficient of linear expansion when heated two types of samples: 1-3 is the glass S52-1 and 4-5 – the same glass in composition of the glass-metal composite for several samples.

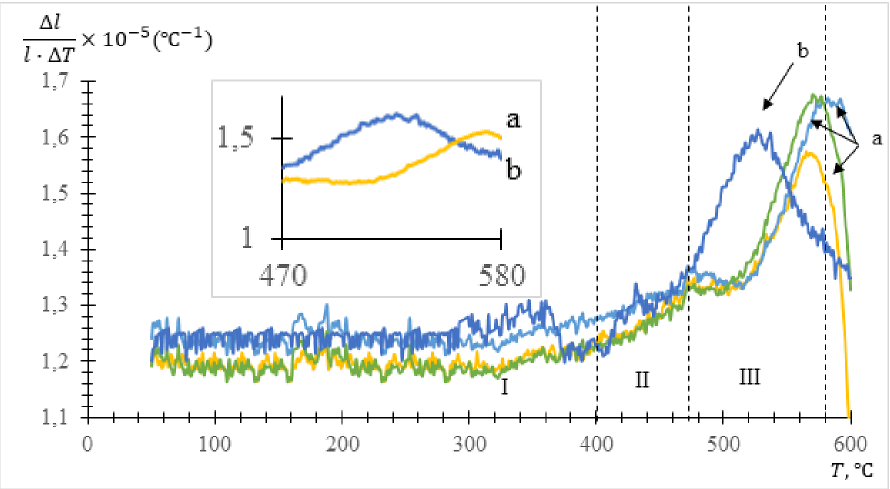
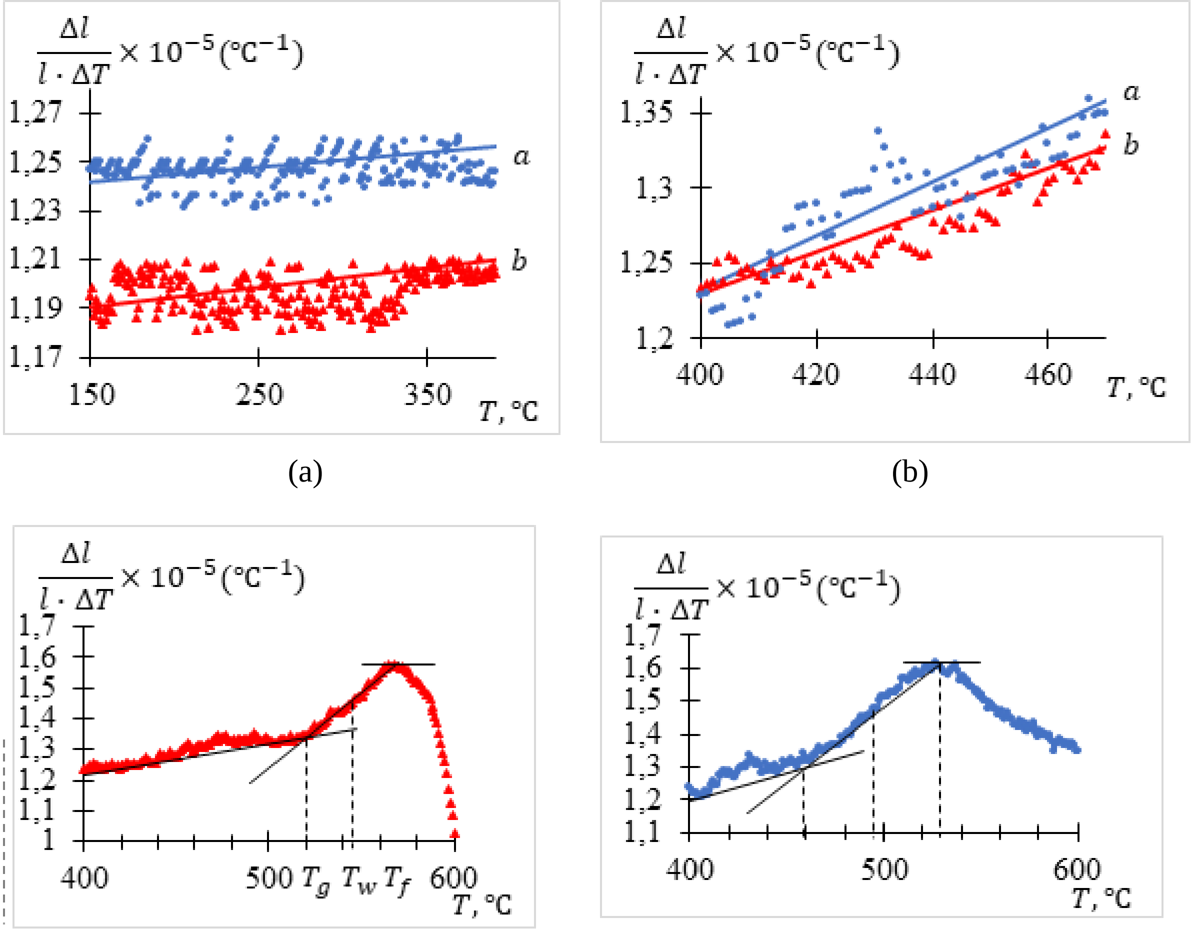


Fig. 3. Dependence of the coefficient of linear temperature expansion on temperature: (a) glass S52-1; (b) glass-metal composite (GMC)



(a) (b)

(c) (d)

Fig.4. Correlation dependences of the coefficient of linear temperature expansion in different intervals of temperatures: (a) 150–400 °C, a - r=0.8901, b - r=0.9354; (b) in the interval 400–480 °C, a - r=0.9207, b - r=0.9567; (c) *Tg*=519 °C, *Tw*=533 °C, *Tf*=564 °C; (c) *Tg*=464 °C, *Tw*=498 °C, *Tf*=529 °C

It is noticeable that the character of changing α of glass differs from that of glass-metal composite by the occurrence of the interval II in which its increase is noticeable. In the graph, 3 zones can be distinguished: I – interval to $\approx 400\,°C$, in this interval, α remains virtually unchanged; the character of changing α of glass differs from that of the glass-metal composite by the occurrence of interval II in which its increase is noticeable; III – interval ≈ 470–580 °C corresponds to S-curve, in this interval, the high increase of the rate of changing the linear dimension is noticeable. In order to determine the temperature parameters, namely III interval was studied (Fig. 3). For each experimental sample, data in each interval were approximated using different dependences (Fig. 4): linear ones on the intervals I and II (Fig.4(a,b)), curves 2-nd and 4-th orders in cases of different characters of data change at the beginning of interval (Fig. 4(c,d)). Apart from the above dependences, the important characteristics for approximation of the coefficient of linear temperature expansion in the transition interval include the angle coefficients of the correlation right lines. In the III interval, the glass-transition temperature was determined subject to approximation type. The softening temperature T_ω was determined as the average value in the interval $[T_g, T_f]$, the interval $\delta T_g = T_g^+ - T_g^-$ is found as the difference of the extreme values of temperature when using the approximation by the linear regression in the intervals II (determination of T_g^-) and III (determination of T_g^+) and the intersection point of these correlation dependences determines T_g. The flow temperature was determined as the maximum point after approximation of α curve of 2nd order in the III interval (Table 1).

Table 1. Basic results of statistical processing

glass	T_g, ℃		T_f, ℃		T_ω, ℃		δT_g, ℃		ΔT_g, ℃	
	M_x	D_x	M_x	D_x	M_x	D_x	M_x	D_x	M_x	D_x
S52-1	518.5	512.4–532.6	566.6	561.2–572.0	532.4	529.2–536.5	8.3	7.4-9.8	45.2	35.9–53.1
GMC	468.5	462.4–472.6	526.6	525.2–533.4	501.2	496.2–516.5	10.6	8.4-12.7	52.1	49.9–56.0

The continuation of experimental investigations of the temperature parameters was associated with increasing the sample for improvement of quality of the statistical processing and subsequent evaluation of the relaxation time τ_g at the glass-transition temperature using the Bartenev formula [26] generalized by Nemilov [12] as a result of analysis of the relaxation theories of Volkenstein-Ptitsyn and Mandelstam-Leontovich [15] $q\tau_g = \delta T_g$, and study of new kinetic criteria of glass transition [14] $C_g = \frac{\delta T_g}{T_g} = \frac{f_g}{\ln\left(\frac{1}{f_g}\right)}$, where $f_g = \left.\frac{\Delta V_e}{V}\right|_{T=T_g}$ is a fraction of the fluctuation volume frozen at the glass-transition temperature, in this event, the temperature expansion coefficient of the fluctuation volume β_f at temperature $T = T_g$, in the model of delocalized atoms in the articles of D.S. Sanditov [25] $\beta_f T_g = f_g \ln\left(\frac{1}{f_g}\right)$, parameters of this relation are determined by the constants $C_1 = \frac{1}{f_g}$, $C_2 = \frac{f_g}{\beta_f}$, permitting to estimate in the Williams-Landel-Ferry (WLF) equation the relative time of relaxation (relative viscosity) a_T $ln a_T = -C_1 \frac{T-T_g}{T-T_g+C_2}, a_T = \frac{\tau(T)}{\tau(T_g)} = \frac{\eta(T)}{\eta(T_g)}, q\tau_g = C_2/C_1$.

According to the data of Table 1, the parameters C_1 and C_2 and characteristics of glass transition for S52-1 and glass-metal composite can be determined (Table 2). The derived estimates for glass S52-1 are aligned with experimental data obtained in the articles [13,25].

Table 2. Parameters of the VLF equation ($q = 6\ Kmin^{-1} = 0{,}1\ Ks^{-1}$)

Glass	T_g, K	δT_g, K	$\delta T_g/T_g$	f_g	β_f, K^{-1}	C_1	C_2, ℃	τ_g, c
S52-1	792	8.3	$10.4 \cdot 10^{-3}$	0.0349	$14.7 \cdot 10^{-5}$	28.7	236	80
GMC	741	10.6	$14.3 \cdot 10^{-3}$	0.0445	$8.2 \cdot 10^{-5}$	22.5	542.9	240

Conclusions

In the present paper, the method for determination of temperature (T_g) and interval of glass-transition (δT_g) necessary for determining the relaxation time (τ_g) and analytic approximation of the coefficient of linear temperature expansion in the glass-transition interval was worked out using the measuring technique of shrinkable displacements for the glass-metal composite (GMC). The investigations of the listed parameters are necessary for analytic computations of the tensely-deformed state of the glass-metal connections and glass-metal composites in the state change interval using the method proposed in the work [27] which determines the plan and perspective of continuation of the further experimental investigations of the glass-metal composite.

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THE AUTHORS

Lyubimova O.N.
e-mail: berms@mail.ru

Barbotko M.A.
e-mail: barbotko.ma@dvfu.ru

Streltcov A.A.
e-mail: streltsov.aa@students.dvfu.ru