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E.V. Karasik, Yu.S. Hordieiev

CALCULATION OF THERMAL EXPANSION, GLASS TRANSITION TEMPERATURE AND GLASS DENSITY IN THE SYSTEM RO–Al_2O_3–B_2O_3–SiO_2 (WHERE RO=BaO, SrO, CaO, MgO, ZnO)

Ukrainian State University of Chemical Technology, Dnipro, Ukraine

Glasses of the system RO–Al_2O_3–B_2O_3–SiO_2 are used as a base for the fabrication of heat-resistant nonmetallic materials and general-purpose products. The purpose of this work is to develop mathematical models for calculating the temperature coefficient of linear expansion, glass transition temperature and density as a function of the composition of glass in the oxide system RO–Al_2O_3–B_2O_3–SiO_2 where RO=BaO, SrO, CaO, MgO, ZnO. The disadvantage of the known models is that the range of their application is limited by the quantitative content of components in the glass. At the same time, an increase in the sample size of experimental compositions made it possible to obtain more accurate mathematical models for calculating these properties. The glasses included in the experimental sample are distinguished by a wide range of temperature coefficient of linear expansion (from 30 to $105 \cdot 10^{-7}$ K^{-1}). The glass transition temperature of these glasses is within the range of 580–710^{0}C, which allows a reasonable approach to the choice of temperature regime for the formation of the structure vitreous and glass-ceramic materials for different functional purposes. The mathematical models were developed with the use of the experimental and statistical method. The obtained mathematical models are adequate to the experimental data and allow calculating the thermal expansion, glass transition temperature and density of glasses; the mean-square deviations of temperature coefficient of linear expansion, glass transition temperature and density being $\pm 1.9 \cdot 10^{-7}$ K^{-1}, $\pm 16.0^0$C and ± 0.06 g cm^{-3}, respectively. Their accuracy is sufficient for the development of basic glass compositions for various functional purposes.

Keywords: borosilicate glass, glass transition temperature, density, thermal expansion, mathematical modeling, additive coefficients.

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Introduction

The glasses of the system RO–Al_2O_3–B_2O_3–SiO_2 (RO=BaO, SrO, CaO, MgO, ZnO) are widely used as a basic glass for the manufacturing of refractory nonmetallic materials and multi-purpose products. In particular, such glasses are used to obtain heat-resistant glass-ceramic materials, including radio-transparent ones [1–4]. They also serve as the base for the fabrication of heat-resistant protective coatings for special alloys in the manufacturing of rocket engines [5–7].

As commonly known [8], the quality indicators and processibility of the obtained refractory nonmetallic materials designed for various functional purposes directly depend on the values of their temperature coefficient of linear expansion (TCLE), glass transition temperature (t_g) and density (d). When choosing the most preferred components and their content in the compositions of basic glasses, it should be remembered that the glasses with low TCLE values (~$30 \cdot 10^{-7}$ K^{-1}) can be used in the manufacturing of heat-resistant materials, whereas the glasses with the thermal expansion close to those of the alloys ($110–130) \cdot 10^{-7}$ K^{-1} are recommended to prepare protective coatings [6,7]. Glass transition temperature characterizes the transition temperature of the material into a glassy state. It should be 200–300^{0}C lower than the aging temperature of the metal alloy under operating conditions at high temperatures in order to prevent a decrease in its mechanical characteristics during firing. The density of a glass is to be taken into account in the process of preparation

of materials and products, which are subjected to weight limitation requirements. In addition, an important condition for fabrication of such glasses is their processibility and the possibility of melting at relatively low temperatures (1300–1400°C), which is also taken into account when choosing the basic components and their content in glass compositions with a given set of performance characteristics.

The models developed earlier [9,10] allow determining the values of density and glass transition temperature only indirectly. At the same time, an increase in the sample size of glass compositions will provide more accurate mathematical model for calculation of the thermal expansion [11,12]. Furthermore, the use of wider limits of the content of basic components will significantly expand the scope of their application.

In this regard, the purpose of this paper is to obtain the mathematical models for calculating the TCLE, glass transition temperature and density of glasses in the oxide systems $RO–Al_2O_3–B_2O_3–SiO_2$ (where RO=BaO, SrO, CaO, MgO, ZnO) as a function of their composition.

Calculations

The development of the mathematical models was carried out by the method of multiple correlations [12–14]. The regression equation in the following form was used to describe the dependence between the properties and compositions of the multicomponent glasses:

$$\hat{y}=\sum_{i=1}^{n} b_i x_i, \qquad (1)$$

where $\hat{y}$ is the calculated value of glass properties; b_i are the regression coefficients; and x_i are the content of the components in the glass (mol.%).

The values of regression coefficients were estimated by the least square method [12] based on three experimental samples of the glass compositions with the known values of temperature coefficient of linear expansion, glass transition temperature and density, which were drawn with the use of the SciGlass electronic database [15].

The approximation accuracy by a regression equation (1) of the experimental values of properties was estimated by comparison of the residual dispersions S^2_{res} with the dispersions relative to the mean values of S^2_y. The specified dispersions were calculated according to the following formulae:

$$F=\frac{S^2_y}{S^2_{res}},\quad S^2_y=\frac{\sum_{i=1}^{n}(y_i-\bar{y})^2}{n-1},\quad \bar{y}=\frac{\sum_{i=1}^{n} y_i}{n},$$

$$S^2_{res}=\frac{\sum_{i=1}^{n}(y_i-\hat{y}_i)^2}{n-l}, \qquad (2)$$

where F is the calculation value of Fisher's criterion; S^2_y is the sampling variance of the values of properties; S^2_{res} is the residual variance; y is the sample mean of the property; y_i is the experimental value of the i-th glass property; $\hat{y}_i$ is the calculated value of the i-th glass property; n is the size of the experimental sample; l is the number of coefficients in the regression equation (it is equal to 8, which is the number of components).

The more the value of S^2_y exceeds the value of S^2_{res}, the more accurately the regression equation approximates the experimental data.

Results and discussion

The numerical characteristics of the initial experimental samples as well as the limits of the content of components in the glasses are given in Tables 1 and 2.

Results of statistical analysis are presented in Table 3.

It follows from the data of Table 3 that developed equations adequately describe the experimental data and allow calculating the TCLE, glass transition temperature and glass density within specified limits of the component content with sufficiently high accuracy.

It was found that barium oxide made the most important contribution to the increase in the temperature coefficient of linear expansion (Table 2, Fig. 1,a). This, in turn, suggests its positive effect on

Table 1

Numerical characteristics of experimental samples

Properties	Amount of sample, n	Range of property values	Sample mean, $\bar{y}$	Mean-square deviation, S_y
TCLE, $\alpha\cdot10^7$, K^{-1}	612	25–118	58.9	23.57
Glass transition temperature, t_g, ^0C	343	530–730	639.1	43.73
Density, d, g cm^{-3}	756	2.17–4.47	3.11	0.597

Table 2

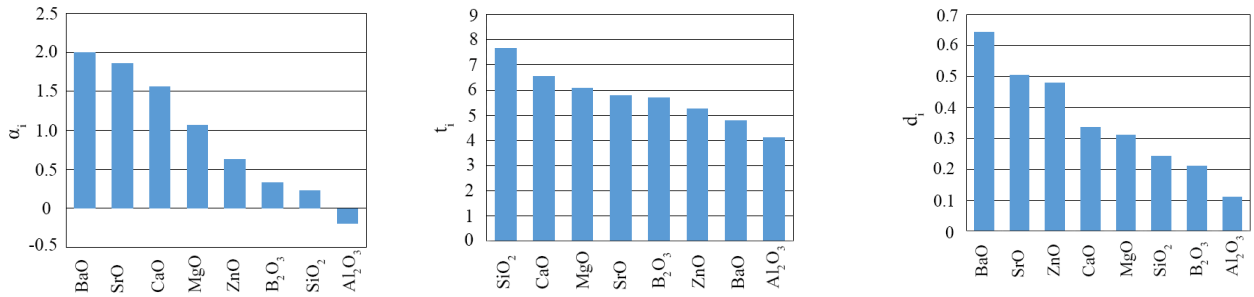
Regression coefficients and their mean-square deviations for calculation of properties of glasses under study according to Eq. 1

Oxide	TCLE, α_i	Glass transition temperature, t_i	Density, d_i	Limits of component content, mol.%
SiO_2	0.230±0.008	7.674±0.071	0.0244±0.0002	20–70
B_2O_3	0.333±0.013	5.705±0.108	0.0212±0.0003	5–44
BaO	2.006±0.010	4.801±0.073	0.0644±0.0003	0–55
SrO	1.865±0.016	5.787±0.106	0.0507±0.0004	0–45
MgO	1.071±0.018	6.092±0.217	0.0312±0.0004	0–35
CaO	1.561±0.012	6.553±0.087	0.0336±0.0003	0–50
ZnO	0.633±0.013	5.260±0.100	0.0481±0.0005	0–50
Al_2O_3	–0.200±0.050	4.122±0.417	0.0111±0.0015	0–10

Table 3

Results of statistical analysis of the regression equations

Properties	Sample variance, S_y^2	Residual variance, S_{res}^2	Fisher's criterion, F	Mean deviation, $\hat{y}_i - y_i$
TCLE, $\alpha \cdot 10^7$, K^{-1}	555.6	6.13	90.64	±1.9
Glass transition temperature, t_g, ^0C	1912.3	395.8	4.83	±16.0
Density, d, g cm^{-3}	0.356	0.007	50.86	±0.06



a **b** **c**

Fig. 1. Partial contribution of oxides to the values of TCLE (a), glass transition temperature (b) and density (c) of glasses

the formation of a continuous and flawless protective coating on special chromium-nickel alloys for rocket engines, the TCLE of which is within the range of $(110–130)\cdot10^{-7}$ K^{-1}.

At the same time, aluminum oxide is most preferable for the use of basic glasses of the system $RO–Al_2O_3–B_2O_3–SiO_2$ to fabricate radio-transparent heat-resistant glass ceramics.

The highest partial contribution to the values of glass transition temperature is made by SiO_2 (Fig. 1,b). This fact should be taken into account, first of all, when choosing the compositions of glass bonds for protective coatings, since an increase in the content of silicon dioxide will lead to undesirable rise in the temperature of formation of the protective coatings.

Barium and strontium oxides make the maximum contribution to the increase in density of products (Fig. 1,c). The partial contribution of aluminum oxide to the density value is minimal, as is the case with the TCLE and glass transition temperature.

The proposed mathematical models make it possible to substantiate theoretically the need to introduce a particular component into the composition of glass-ceramic materials, and receive accurately the calculated weight of the designed products made of them.

Table 4 shows some practical compositions of borosilicate glasses, which properties vary in wide ranges. Analysis of the calculated and experimental data (Table 5, Fig. 2) testifies to a strong correlation

Table 4

Chemical compositions of glass

No.	Content of components, mol.%							
	SiO_2	B_2O_3	BaO	SrO	MgO	CaO	ZnO	Al_2O_3
1	62.0	8.0	0.0	7.0	7.0	7.0	0.0	9.0
2	35.0	10.0	35.0	0.0	0.0	15.0	0.0	5.0
3	40.0	12.0	0.0	39.0	0.0	0.0	9.0	0.0
4	49.7	22.2	20.0	0.0	0.0	2.5	1.5	4.1
5	69.3	9.8	1.3	0.0	4.7	5.6	0.0	9.3
6	30.0	10.0	20.0	0.0	0.0	0.0	40.0	0.0
7	42.5	21.2	27.2	0.0	0.0	0.0	0.0	9.1

Table 5

Calculated and experimental values of glasses properties

No.	Calculated values							Experimental values		
	$\alpha \cdot 10^7$, K^{-1}			t_g, ^0C		d, g cm^{-3}		$\alpha \cdot 10^7$, K^{-1}	t_g, ^0C	d, g cm^{-3}
	1	2	3	1	2	1	2			
1	47	49	53	687	684	2.59	2.61	45	684	2.60
2	104	110	100	613	573	3.88	3.92	105	619	3.88
3	92	89	91	648	593	3.64	3.60	95	643	3.71
4	63	64	64	645	657	3.18	3.17	62	640	3.24
5	34	38	41	697	692	2.42	2.46	32	701	2.41
6	76	77	85	594	541	4.15	4.00	79	585	4.15
7	70	74	68	615	602	3.34	3.38	71	610	3.32

Note: The numbering of columns stands for the calculated values of the properties obtained with the use of the developed partial coefficients (1), with the use of the partial coefficients given in works [11,12] (2) and with the use of the partial coefficients given in work [13] (3).

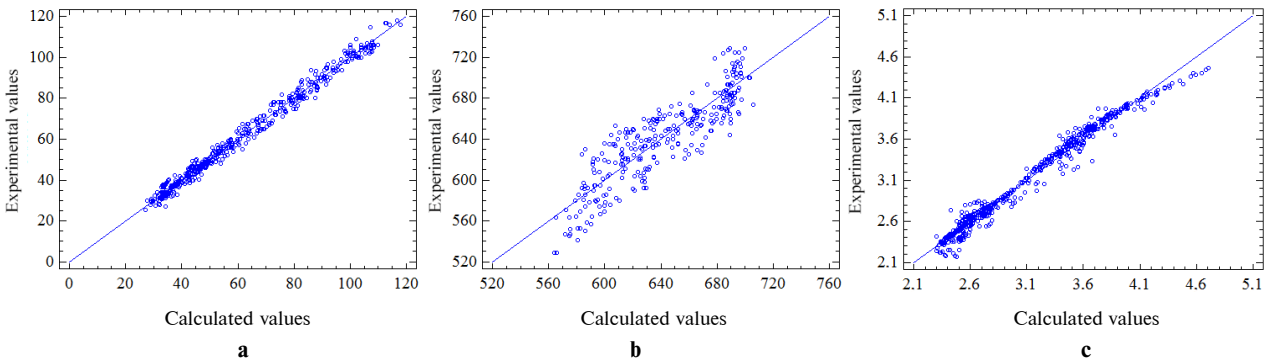


Fig. 2. Relationship between experimental and calculated values of the TCLE (a), glass transition temperature (b) and density (c) of glasses

between them. It is also confirmed by the correlation coefficients, which are 99.85%, 99.90% and 99.93% for the TCLE, glass transition temperature and glass density, respectively.

Therefore, analysis of the values of regression coefficients in the equations, which represent the estimates of the partial contributions of oxides, can be a basis for the selection of the most preferable components and their contents in the compositions of the basic glass types in the system $RO-Al_2O_3-B_2O_3-SiO_2$ (RO=BaO, SrO, CaO, MgO, ZnO) with a given set of performance characteristics.

Analysis of the obtained coefficients and their comparison with the calculations made earlier [11–13] allows drawing a conclusion about higher accuracy of the developed mathematical model, which is confirmed by the results given in Table 5. Thus, mean-square deviation for the values of thermal

expansion is $\pm 1.9 \cdot 10^{-7}$ K^{-1} (Table 3), which is significantly lower than the value given in ref. [13].

Chosen limits of the content of components of the system $RO-Al_2O_3-B_2O_3-SiO_2$ (RO=BaO, SrO, CaO, MgO, ZnO) give an opportunity to calculate the basic composition of glasses with a wide range of TCLE values from 30 to $105 \cdot 10^{-7}$ K^{-1}. In this case, their glass transition temperature varies in the range of 580–710^{0}C, which allows using the substantiated approach to the choice of temperature regime for the formation of the structure glassy and glass-ceramic materials for various functional purposes.

Conclusions

By means of the multiple correlation method, the mathematical models were developed for multicomponent glassws in the oxide system $RO-Al_2O_3-B_2O_3-SiO_2$ (RO=BaO, SrO, CaO, MgO, ZnO) with the following mean-square deviations: TCLE=$\pm 1.9 \cdot 10^{-7}$ K^{-1}, t_g=$\pm$16.0^{0}C, d=$\pm$0.06 g cm^{-3}. These models adequately describe the experimental data with the accuracy sufficient for the development of basic compositions of glasses for various functional purposes. It will allow solving a set of practical problems with appreciable time and cost saving during experimental research.

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РОЗРАХУНОК ТЕПЛОВОГО РОЗШИРЕННЯ, ТЕМПЕРАТУРИ СКЛУВАННЯ ТА ЩІЛЬНОСТІ СТЕКОЛ У СИСТЕМІ $RO-Al_2O_3-B_2O_3-SiO_2$ (ДЕ RO=BaO, SrO, CaO, MgO, ZnO)

О.В. Карасик, Ю.С. Гордєєв

Стекла системи $RO-Al_2O_3-B_2O_3-SiO_2$ використовуються як базові для одержання тугоплавких неметалічних матеріалів і виробів широкого призначення. Метою даної роботи було одержання математичних моделей для розрахунку температурного коефіцієнта лінійного розширення, температури склування та щільності в залежності від складу скла в оксидній системі $RO-Al_2O_3-B_2O_3-SiO_2$, де RO=BaO, SrO, CaO, MgO, ZnO. Недолік відомих моделей полягає в тому, що діапазон їх застосування обмежений кількісним вмістом компонентів у склі. У той же час, збільшення обсягу вибірки експериментальних складів дозволило отримати більш точні математичні моделі для розрахунку зазначених властивостей. Стекла, що увійшли в експеріменальную вибірку,

відзначаються широким діапазоном значень температурного коефіцієнта лінійного розширення (від 30 до $105 \cdot 10^{-7}$ K^{-1}). Їх температура склування при цьому знаходиться в межах 580–710^{0}C, що дає можливість обгрунтованого підходу до вибору температурного режиму формування структури склоподібних і склокерамічних матеріалів різного функціонального призначення. Розробку математичних моделей виконували експериментально-статистичним методом. Отримані математичні моделі адекватні експериментальним даним і дозволяють розраховувати термічне розширення, температуру склування та щільність стекол із середньоквадратичними відхиленнями для температурного коефіцієнта лінійного розширення $\pm 1{,}9 \cdot 10^{-7}$ K^{-1}, температури склування $\pm 16{,}0^0$C, щільності $\pm 0{,}06$ г/см^3. Їх точність достатня для розробки базових складів стекол різного функціонального призначення.

Ключові слова: боросилікатне скло, температура склування, щільність, термічне розширення, математичне моделювання, адитивні коефіцієнти.

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E.V. Karasik, Yu.S. Hordieiev *

Ukrainian State University of Chemical Technology, Dnipro, Ukraine

* **e-mail: yuriihordieiev@gmail.com**

Glasses of the system RO–Al_2O_3–B_2O_3–SiO_2 are used as a base for the fabrication of heat-resistant nonmetallic materials and general-purpose products. The purpose of this work is to develop mathematical models for calculating the temperature coefficient of linear expansion, glass transition temperature and density as a function of the composition of glass in the oxide system RO–Al_2O_3–B_2O_3–SiO_2 where RO=BaO, SrO, CaO, MgO, ZnO. The disadvantage of the known models is that the range of their application is limited by the quantitative content of components in the glass. At the same time, an increase in the sample size of experimental compositions made it possible to obtain more accurate mathematical models for calculating these properties. The glasses included in the experimental sample are distinguished by a wide range of temperature coefficient of linear expansion (from 30 to $105 \cdot 10^{-7}$ K^{-1}). The glass transition temperature of these glasses is within the range of 580–710^{0}C, which allows a reasonable approach to the choice of temperature regime for the formation of the structure vitreous and glass-ceramic materials for different functional purposes. The mathematical models were developed with the use of the experimental and statistical method. The obtained mathematical models are adequate to the experimental data and allow calculating the thermal expansion, glass transition temperature and density of glasses; the mean-square deviations of temperature coefficient of linear expansion, glass transition temperature and density being $\pm 1.9 \cdot 10^{-7}$ K^{-1}, $\pm 16.0^0$C and ± 0.06 g cm^{-3}, respectively. Their accuracy is sufficient for the development of basic glass compositions for various functional purposes.

Keywords: borosilicate glass; glass transition temperature; density; thermal expansion; mathematical modeling; additive coefficients.

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