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The use of regression models to describe the temperature coefficient values of linear expansion and technological temperatures of $CaSO_4$-KPO_3-$Na_2B_4O_7$ low-melting glass system

A Stolbovsky[1, 2, 3]

[1] Ural Federal University named after the first President of Russia B.N. Yeltsin, 19, Mira str., Yekaterinburg, Russia
[2] M.N. Miheev Institute of Metal Physics, UB RAS, 18, S. Kovalevskoy str., Yekaterinburg, 620108, Russian

E-mail: [3] stolbovsky@imp.uran.ru

Abstract. Low-melting noncrystallizing glasses are used as solders for connections of various metallic and nonmetallic parts. However, theoretical data on glass formation and physical parameters of low-melting glasses are still limited. The use of modelling allows to simplify the selection of a glass system as a solder by the mathematical description of the dependencies of both that the temperature coefficient values of linear expansion and other specific temperatures, which are necessary for a practical application. Therefore, the fourth-order regression polynomial models are offered for describing dependences of the temperature coefficient values of linear expansion, the softening temperature and the glass transition temperature from the component concentrations of the $CaSO_4$-KPO_3-$Na_2B_4O_7$ low-melting glass system in the glass formation region. The model involver both pair and ternary interactions of the glass system's components. The analysis of the obtained results showed that the concentration of the CaSO4 component has the highest effect on the temperature coefficient values of linear expansion.

1. Introduction

At present, the application of a leak-free and reliable bonding of parts, which are made of various kind of materials with each other is required in modern instrument engineering. Low-melting noncrystallizing glasses are used as solders for connections of various metallic and nonmetallic parts [1]. The lower softening temperature of such solders allows to perform a junction at quite low temperatures in vacuum that will prevent the oxidation and deformation at soldering [1-2]. In addition, the creation of strong junctions between parts requires from solders that the temperature coefficient values of linear expansion (TCLE) must be comparable, the complete spreading temperature during heat treatment, as a rule, must not exceed above 700 °C and the softening temperature must be below 450–600°C as well [2].

Considerable experimental material concerning low-melting glasses in numerous glass-forming systems has already obtained [2-6]. The data about the properties such as TCLE, the softening temperature (T_s) and the glass transition temperature (T_g) for various glass systems is not generalized.

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That leads to difficulties in choosing low-melting glasses for practical application in industry as solder.

At the same time, theoretical data on glass formation and physical parameters of low-melting glasses are still limited [3,7]. Therefore, the availability of data on TCLE and the technological temperatures for various compositions would allow to make an accurate selection of the composition with the most preferred properties for a specific application.

As represented in the work [8], the use of modelling allows to simplify the selection of a glass system as a solder by the mathematical description of the dependencies of both TCLE and other specific temperatures, which are necessary for a practical application.

In the previous study [9], the possibility of using the ternary $CaSO_4$-KPO_3-$Na_2B_4O_7$ low-melting glass system as a glass-solder material was shown. The aim of the present paper is to study the possibilities of using a regression model for predicting TCLE and the technological temperatures' values depending on the glass composition.

2. Materials and methods

The results of the study [9] devoted to the investigation of the $CaSO_4$-KPO_3-$Na_2B_4O_7$ system have been taken. According to the regression analysis recommendations [10-12], polynomial regression was chosen as a model for fitting the experimental data. The chi-squared (χ^2) test as the quality factor to estimate the fitting accuracy was chosen because χ^2 is a more sensitive characteristic in comparison with the coefficient of determination R^2 [12].

The general view of the N-order polynomial with pair and ternary interactions, which was used for fitting experimental data is represented by the following expression:

$$\begin{gathered}\alpha = C_0 + C_x x + C_y y + C_z z + C_{xy} xy + C_{xz} xz + C_{yz} yz + C_{xyz} xyz + \\ + C_{x2} x^2 + C_{y2} y^2 + C_{z2} z^2 + C_{xy2} x^2 y^2 + C_{xz2} x^2 z^2 + C_{yz2} y^2 z^2 + C_{xyz2} x^2 y^2 z^2 + \\ \ldots \\ + C_{xN} x^N + C_{yN} y^N + C_{zN} z^N + C_{xyN} x^N y^N + C_{xzN} x^N z^N + C_{yzN} y^N z^N + C_{xyzN} x^N y^N z^N\end{gathered}, \quad (1)$$

where α is TCLE value ($\alpha \cdot 10^{-7}$ K^{-1}), various indexes – the calculated model coefficients, x is the concentration in mole fractions of $CaSO_4$, y – KPO_3 and z – $Na_2B_4O_7$, accordingly.

The same expression (1) was used for fitting T_s (°C) and T_g (°C) dependences as well.

The calculation was carried out in two stages. The first stage was the calculation of the full model with expression (1). At the second stage, coefficients, which were a few orders of magnitude less than 1 and other model coefficients as well, were excluded from the model to simplify one. Then the simplified model was recalculated to estimate accuracy. If it was necessary the process of simplifying was repeated.

3. Results and discussion

The regression analysis of experimental data allows to make the TCLE and technological temperatures' values interpolation between the experimental points and estimate the relationships between a glass composition and these dependent variables. Such approach will allow to consider the surface features in multidimensional space without deep theoretical studies.

As shown by the regression analysis of the experimental dataset, the TCLE, T_s and T_g dependences on components composition of the glass system are well described by means of the regression model in the form of a fourth-order polynomial in contrast to the second-order one for the $MgSO_4$-KPO_3-$Na_2B_4O_7$ system [8]. The results of approximation for the TCLE, T_s and T_g as calculated coefficients of the model are shown in Table 1. The numerical values are represented in exponential form. The sign in front of the model's coefficient indicates a direction of an influence.

It should be noted that suggested regression models do not have a physical basis. That is statistical description only. However, such statistical representation allows to estimate an influence of components on the resulting value. That can be important for understanding when knowledge of physical foundations does not exist, or it is significant limited.

Moreover, the use of the models for practical application should be restricted in glass formation region [9]. It is important since the represented models does not take into account the limitation of the composition. In order to avoid an incorrect estimation of values of the TCLE and the technological temperatures, should use the following set of expressions for the components of the glass system:

$$\begin{cases} x \in (0.0; 0.3) \\ y \in (0.0; 0.8) \\ z \in (0.0; 0.9) \\ x + y + z = 1 \end{cases}, \tag{2}$$

where x is the concentration in mole fractions of $CaSO_4$, y – KPO_3 and z – $Na_2B_4O_7$, accordingly.

Table 1. Results of approximation.

Model's coefficients	$\alpha \times 10^{-7}$, K^{-1}	T_s, °C	T_g, °C
C_0	-	3.39E+02	-
C_x	-	-	3.81E+02
C_z	-	-	2.45E+02
C_{xy}	-	6.04E+02	7.62E+02
C_{yz}	3.76E+02	7.99E+01	4.99E+02
C_{xyz}	2.05E+03	-	-
C_{z2}	2.92E+02	1.31E+01	6.74E+01
C_{xy2}	-	1.65E+01	9.14E+02
C_{xz2}	2.13E+03	-	-
C_{y3}	2.09E+02	5.12E+01	2.27E+02
C_{xy3}	5.83E+03	-	-
C_{yz3}	-	-	7.77E+02
C_{xyz3}	1.03E+00	1.01E+00	-
C_{x4}	9.59E+03	-	-
C_{y4}	-	-	2.22E+02

To visually estimate α, T_s and T_g, colored ternary diagrams of these properties can be used. Figure 1 is represented the calculated values of the TCLE, T_s and T_g for the glass formation region of the $CaSO_4$-KPO_3-$Na_2B_4O_7$ system, which are indicated by colored areas on the ternary diagrams. It can be seen than the TCLE is variated in very wide range of $140\text{-}270 \cdot 10^{-7}$ K^{-1}. The values of the glass transition temperature are in the range of 270-390°C and the values of the softening temperature in the range from 350 to 490°C. These are lower values of T_s and T_g, which are usually required for glass solders [2].

In comparison with the $MgSO_4$-KPO_3-$Na_2B_4O_7$ system [8], the replacement of the $MgSO_4$ component expanded the range of the TCLE values. At the same time, the second-order regression polynomial model, which was used for the $MgSO_4$-KPO_3-$Na_2B_4O_7$ system, did not allow to describe the dependencies adequately. That can be caused by a different effect of Mg^{2+} and Ca^{2+} cations on the glass structure.

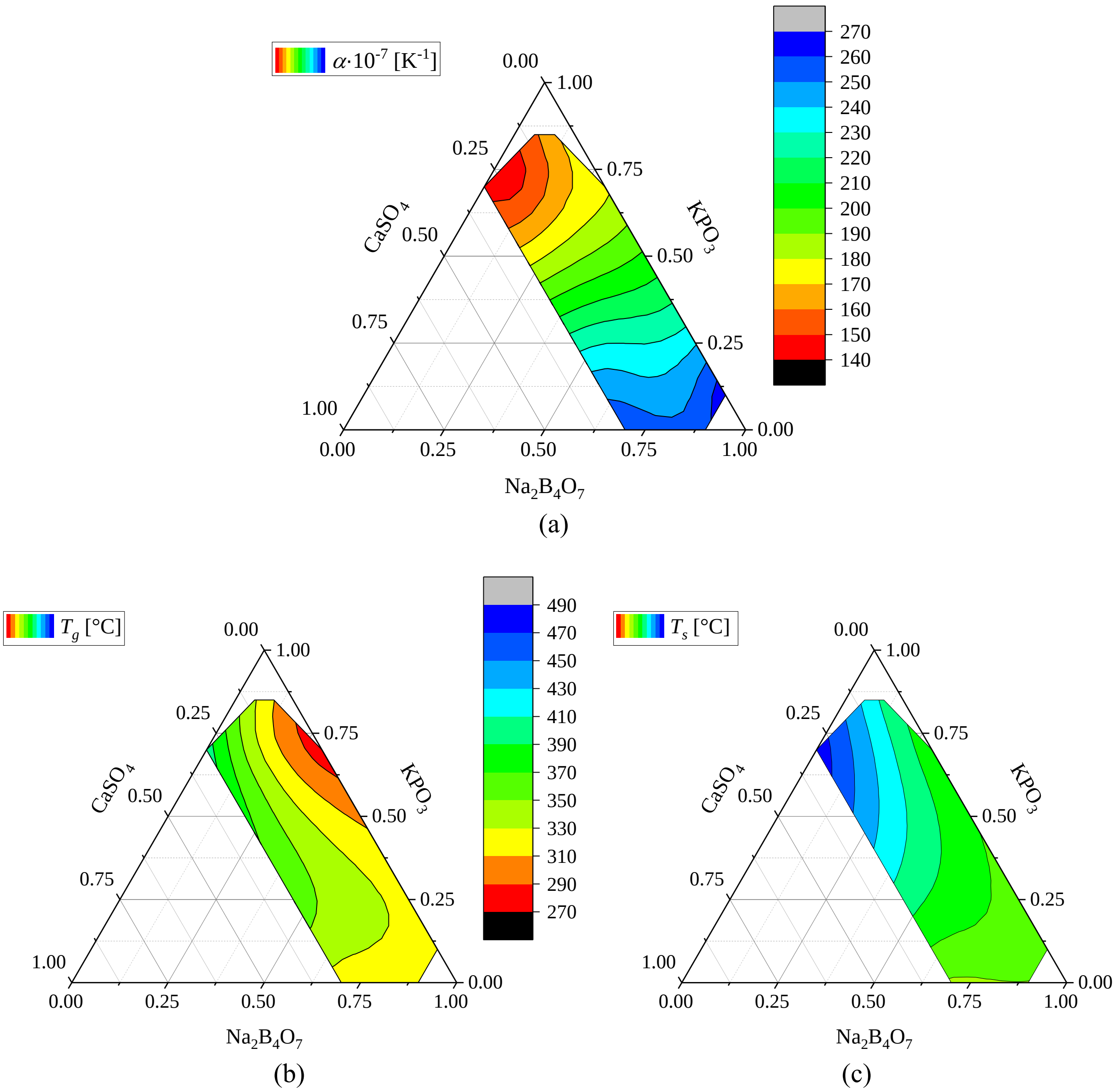

Figure 1. The glass formation region of the $CaSO_4$-KPO_3-$Na_2B_4O_7$ system with calculated values of (a) – TCLE; (b) – the glass transition temperature (T_g) and (c) - the softening temperature (T_s), which are represented by colored areas

Besides, it can be seen (Table 1) that, in the TCLE model, among the coefficients at the fourth-order magnitude of components, the coefficient corresponding to the $CaSO_4$ component is presented only. Moreover, this coefficient is the highest that may also indicate differences in the influence of Mg^{2+} and Ca^{2+} cations on the glass structure additionally.

4. Summary

It was established that the second-order regression polynomial model, which was used for the MgSO4-KPO3-Na2B4O7 system, did not allow adequately to describe the dependencies of TCLE and the technological temperatures' values of the $CaSO_4$-KPO_3-$Na_2B_4O_7$ system. The fourth-order regression polynomial model is offered for describing dependences of the TCLE, the softening temperature and the glass transition temperature from the concentration in mole fractions of $CaSO_4$, KPO_3 and $Na_2B_4O_7$ components of the glass system. The proposed model takes into account pair and ternary interactions of the glass system's components.

As shown by the analysis of the obtained results, in comparison with the $MgSO_4$-KPO_3-$Na_2B_4O_7$ system, the replacement of the $MgSO_4$ component with $CaSO_4$ allowed to expand the range of the TCLE values. In addition, it was found, the $CaSO_4$ component has the highest effect on the TCLE that may points out at different influence of Mg^{2+} and Ca^{2+} cations on the glass structure.

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