

# **On the development of a viscosity model for molten multicomponent slag systems with several glass-forming, amphoteric and modifier oxides**

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Keywords: viscosity modelling, molten slags

## ABSTRACT

Viscosity of multicomponent slags is one of the crucial physicochemical properties for many industrial processes and technologies. While the viscosity-temperature relationship of completely molten slags can well be described by an Arrhenian-type law, the compositional dependence of viscosity is still not fully understood, especially in compositional regions with the lack of experimental data (e.g. at the glass-former concentration of 90 molar pct or higher). In the present work major compositional relationships of the activation energy of viscous flow have been revised for numerous binary and ternary slag systems on the basis of experimental data collected and stored in the viscosity database OxiVis. For example, it has been found that the activation energy relevant to the polymerisation effect in binary systems of the MO-GFO type, where MO is the modifier oxide (i.e. monovalent  $K_2O$ ,  $Na_2O$ ,  $Li_2O$  and bivalent  $CaO$ ,  $MgO$ ,  $BaO$ ,  $MnO$ ,  $PbO$  etc) and GFO is the glass-forming oxide ( $SiO_2$  or  $B_2O_3$ ) is not always proportional to the third power of  $X_{GFO}$  (contrary to the Urbain formalism) and can vary significantly dependent on the slag system. Also, the activation energy of the charge compensation effect has been revised for ternary systems of the AO-MO-GFO and AO-MO type, where AO is the amphoteric oxide ( $Al_2O_3$ ,  $Fe_2O_3$  or other). A simple polynomial model has been proposed on the basis of the carried-out analysis for estimation of viscosities of multicomponent slag systems. The model has been optimised against the experimental viscosities in the selected binary, ternary, and higher-order oxide systems, and has been shown reasonable agreement with the experimental data.

## INTRODUCTION

Viscosity of molten oxide mixtures commonly known in industry as slags plays the critical role in many industrial and natural processes. The main problem with experimental determination of viscosity is a high melting point of various oxides ( $>1500^\circ C$ ) and extreme reactivity of oxides in the liquid state. Therefore, modelling of viscosity can be applied to avoid experimental difficulties.

A number of practical viscosity models have been developed over last 30-40 years to calculate viscosity of multicomponent oxide melts (e.g. Kondratiev 2002, Jung 2014). Among others, structural viscosity models are based on the melt structure usually described by concentrations of various structural units (e.g. bridging and non-bridging oxygens, silica tetrahedra, the second nearest neighbour bonds etc). However, these models are complicated, often cannot be used without additional data, and demonstrate good agreement mostly for multicomponent melts.

It is well known that, based on their effect on viscosity, all oxides can roughly be divided into three groups: glass-forming oxides (GFO), modifier oxides (MO) and amphoteric oxides (AO). Glass-forming oxide melts have much higher viscosity than the rest of oxide melts due to formation of interconnected, continuous structural network in the molten state. This continuous structural network is sometimes referred to as the "fully polymerised melt". Modifiers usually break (or "depolymerise") that continuous network into fragments and thereby lower the melt viscosity. Amphoteric oxides can either break the network or form a quasi-network structure depending on the other melt components.

One of the less complicated, polynomial models based on the above idea is the so called Urbain formalism (Urbain 1981), in which the Weymann-Frenkel equation (1) is used for the viscosity-temperature dependence, and the compositional dependence of the parameter  $B$  in the ternary  $Al_2O_3$ - $CaO$ - $SiO_2$  system is expressed as the polynomial (2) of the molar fraction of  $SiO_2$  and the ratio of  $Al_2O_3$  and  $CaO$ :

$$\log \eta = \log A + \log T + \frac{B}{T}, \quad (1)$$

$$B = \sum_{i=0}^3 \sum_{j=0}^2 b_{ij} \left( \frac{X_{Al_2O_3}}{X_{Al_2O_3} + X_{CaO}} \right)^j X_{SiO_2}^i. \quad (2)$$

The Urbain-type models (e.g. Kondratiev 2001, Forsbacka 2007) demonstrate good agreement for calculating viscosity of the systems with one GFO ( $\text{SiO}_2$ ), one AO ( $\text{Al}_2\text{O}_3$ ) and a number of divalent MOs (CaO, MgO, MnO, etc). In the binary and ternary systems with monovalent modifiers ( $\text{K}_2\text{O}$ ,  $\text{Na}_2\text{O}$ ) this formalism is not generally capable to describe experimental data with a reasonable accuracy, and other, more complex models have to be developed (e.g. Jak 2009). To the authors' best knowledge the Urbain formalism has not been applied to the systems with two or more GFO. The major problem of development of phenomenological viscosity models is the experimental data, its existence, amount, and quality.

In the present work the OxiVis viscosity database (Kondratiev 2021) was used to extract and analyse experimental data for unary, binary and ternary systems. The OxiVis database is a big collection of the experimental viscosity data developed and maintained by one of the authors for 20 years. It contains over 50,000 experimental points for over 9,000 chemical compositions of 54 different oxides.

The main task considered in the present work is to construct a simplest possible polynomial model that is still capable to describe experimental data on viscosity of unaries, binaries, and ternaries with a reasonable accuracy. Thus, some non-linear viscosity effects other than polymerisation encountered in binary oxide systems, though be discussed, will not be taken into account during modelling. First, the available experimental data for binary systems will be analysed, and a polynomial model will be proposed for binary silicate, borate and other systems that takes into account different viscosity growth at the GFO concentrations approaching 1. Then additional terms will be proposed and discussed for the parameter  $B$  (equations 1-2) in the ternary systems of the AO-MO-GFO type. Finally the model will be tested on high-order systems.

## EXPERIMENTAL EVIDENCE ON THE PRESENCE OF LINEAR REGIONS IN BINARY SYSTEMS

Viscosity of binary systems, in which one component is a GFO, are generally characterised by the so-called polymerisation effect, which results in a rapid growth of viscosity by several orders of magnitude as the GFO concentration tends to 1. In terms of the activation energy (proportional to the parameter  $B$  in Eq. 1) it results in an increase of the activation energy to the value that corresponds to the GFO parameter. In the Urbain formalism the polymerisation effect is expressed as the third power polynomial of the  $\text{SiO}_2$  molar fraction (see Eq. 2). However, the exact form of the effect is not well investigated for most of binaries, especially at higher GFO concentrations (see, for example, Kondratiev 2021).

Analysis of the collected experimental data for binary silicates revealed the presence of a nearly linear region at the  $\log \eta$  vs  $X_{\text{GFO}}$  dependence in each binary silicate system. The width of this region can expand from 0-0.3 to about 0.7-0.8  $X(\text{SiO}_2)$  and depends on a system. Figure 1 provides the experimental data for five silicate binary systems at constant temperatures against the molar fraction  $X(\text{SiO}_2)$  (between 0 and 0.8) along with the linear fit for logarithm of viscosity. At  $X(\text{SiO}_2) > 0.7$  a rapid growth of viscosity (the so-called polymerisation effect) usually takes place. At  $X(\text{SiO}_2) < 0.3$  a similar, but less pronounced effect can be observed in systems like  $\text{Al}_2\text{O}_3\text{-SiO}_2$  or  $\text{PbO-SiO}_2$ .

Interpolation of the linear fit of the viscosity logarithm against  $X(\text{SiO}_2)$  was carried out for each system in selected silicate binaries ( $\text{Al}_2\text{O}_3\text{-}$ ,  $\text{BaO-}$ ,  $\text{CaO-}$ ,  $\text{K}_2\text{O-}$ ,  $\text{MgO-}$ ,  $\text{MnO-}$ ,  $\text{Li}_2\text{O-}$ ,  $\text{Na}_2\text{O-}$ , and  $\text{PbO-SiO}_2$ ). The interpolation procedure was applied to all the experimental viscosity values at  $X(\text{SiO}_2)$  from 0.3 to 0.7, approximately, and to all the temperatures above the melting point. Table 1 shows the linear fit coefficients ( $\log A$  &  $B$ ) obtained for each system and the root mean squared errors (RMSE) of the interpolation. Also the coefficient of determination ( $R^2$ ) is included. It can be seen that the fit "quality" ( $R^2 > 0.8$ , RMSE is low) is good for most of the systems, which implies that the quasi-linear regions exist at certain intervals of  $X(\text{SiO}_2)$  in silicate binaries. Also, it can be seen that the parameters of the linear fit are similar to each other: the parameter  $\log A$  is negative for all systems, and the parameter  $B$  is positive for all systems except for  $\text{K}_2\text{O-SiO}_2$ .

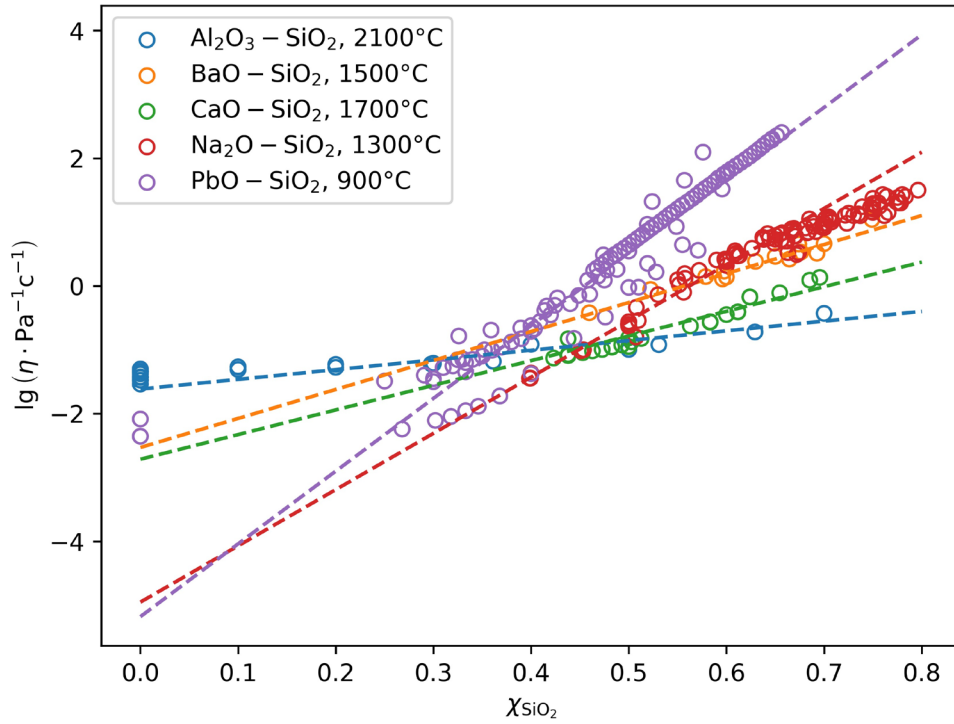


FIG 1 – The quasi-linear regions in the (Al<sub>2</sub>O<sub>3</sub>, BaO, CaO, Na<sub>2</sub>O, PbO)-SiO<sub>2</sub>.

Table 1. Linear fit parameters and errors for selected silicate systems (equation 1).

System	End-member	log(A/Pa*s)	10 <sup>3</sup> ·B, kK	RMSE	R <sup>2</sup>
Al <sub>2</sub> O <sub>3</sub> -SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	-2.538	2.189	0.104	0.833
	SiO <sub>2</sub>	-4.792	11.146		
BaO-SiO <sub>2</sub>	BaO	-5.029	4.430	0.062	0.987
	SiO <sub>2</sub>	-5.157	12.708		
CaO-SiO <sub>2</sub>	CaO	-6.839	8.137	0.172	0.834
	SiO <sub>2</sub>	-3.292	8.756		
K <sub>2</sub> O-SiO <sub>2</sub>	K <sub>2</sub> O	-6.302	-0.896	0.172	0.973
	SiO <sub>2</sub>	-5.540	17.028		
Li <sub>2</sub> O-SiO <sub>2</sub>	Li <sub>2</sub> O	-5.775	3.065	0.103	0.969
	SiO <sub>2</sub>	-3.227	9.411		
MgO-SiO <sub>2</sub>	MgO	-2.801	0.524	0.101	0.987
	SiO <sub>2</sub>	-6.454	15.189		
MnO-SiO <sub>2</sub>	MnO	-6.601	6.812	0.118	0.811
	SiO <sub>2</sub>	-0.110	2.925		
Na <sub>2</sub> O-SiO <sub>2</sub>	Na <sub>2</sub> O	-6.923	3.100	0.153	0.983
	SiO <sub>2</sub>	-3.319	11.289		
PbO-SiO <sub>2</sub>	PbO	-8.468	3.857	0.293	0.921
	SiO <sub>2</sub>	-1.606	9.175		

Figure 2 shows the “log(Viscosity) vs 1/T” dependences of the pure SiO<sub>2</sub> obtained during interpolation for each silicate binary system. It can be seen that only two systems do not follow the mean trend: K<sub>2</sub>O-SiO<sub>2</sub> and MnO-SiO<sub>2</sub>. And PbO-SiO<sub>2</sub> is also slightly out at higher temperatures. A possible reason for deviation of the K<sub>2</sub>O-SiO<sub>2</sub> and MnO-SiO<sub>2</sub> systems is the lack of experimental data at X(SiO<sub>2</sub>) < 0.5 for K<sub>2</sub>O-SiO<sub>2</sub> and at X(SiO<sub>2</sub>) > 0.5 for MnO-SiO<sub>2</sub>. The PbO-SiO<sub>2</sub> system can be characterised by a large scatter in the experimental data at X(SiO<sub>2</sub>) from 0 to 0.5. On the basis of the above analysis one set of parameters can be selected for the pure SiO<sub>2</sub> end (shown in Fig.2 as the mean line).

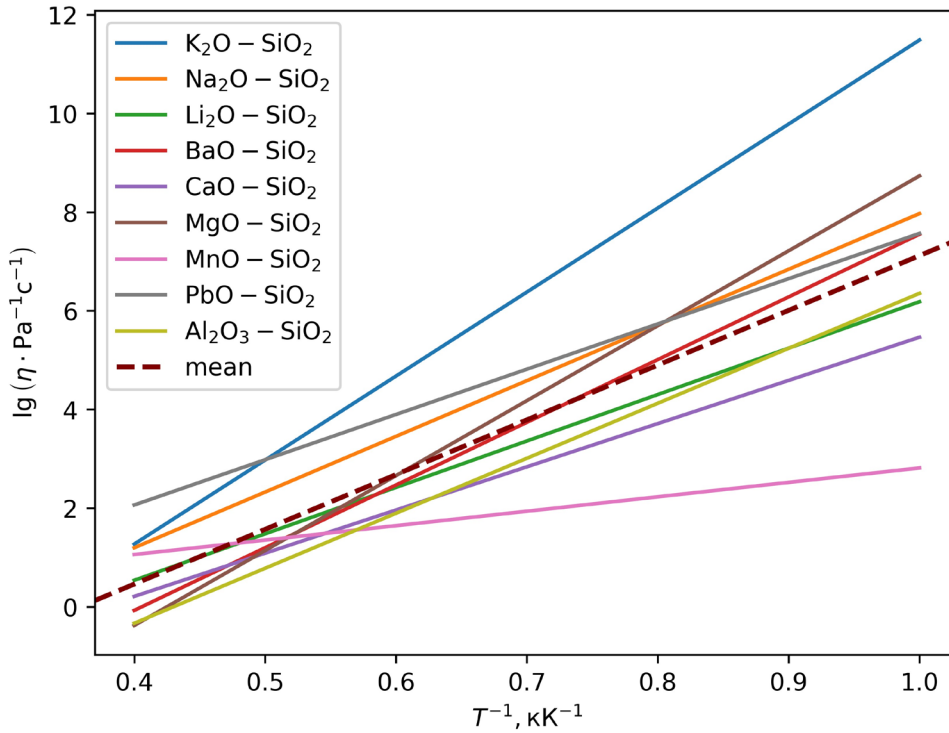


FIG 2 – Viscosity of pure SiO<sub>2</sub> obtained from each system optimisation.

Among all binaries molten silicates are by far the most investigated systems due to their significance for industry and geology. Binary borates, germanates and phosphates are less investigated, although these systems may also be important for glass-making industry or nuclear waste vitrification. Nevertheless, a lot of silicate systems are investigated only in a narrow compositional range (e.g. the CaO-SiO<sub>2</sub> viscosities are measured only in the range  $0.4 \leq X(\text{SiO}_2) \leq 0.7$ ), at relatively low melting temperatures (Kondratiev 2021). In the present study a number of binary silicate and borate systems will be analysed as well as few other GFO-containing systems.

## MODEL CONSIDERATIONS

In the present study the classical Arrhenius equation (in a logarithmic format) is used to describe the temperature dependence of viscosity of molten oxide systems:

$$\log \eta = \log A + \frac{B}{T}, \quad (3)$$

where the parameters  $A$  and  $B$  depend on chemical composition. The  $B$  parameter is linked to the activation energy of viscous flow.

Parameters of Eq. (3) can easily be calculated for those pure oxides, for which the experimental data is available. For the rest of pure oxides it is possible to use estimations from various viscosity models or molecular dynamics simulations (Kondratiev 2024).

The compositional dependence of viscosity in a binary system consists of two parts (called “ideal” and “excess” by the analogy to thermodynamics):

$$\log \eta = \log \eta^{id} + \log \eta^{ex} \quad (4)$$

$$\log \eta^{id} = X_1 \log \eta_1^{id} + X_2 \log \eta_2^{id} = X_1 \log A_1^{id} + X_2 \log A_2^{id} + \frac{X_1 B_1^{id} + X_2 B_2^{id}}{T} \quad (5)$$

$$\log \eta^{ex} = X_1^{\alpha_1} \log \frac{A_1^0}{A_1^{id}} + X_2^{\alpha_2} \log \frac{A_2^0}{A_2^{id}} + \frac{X_1^{\alpha_1} (B_1^0 - B_1^{id}) + X_2^{\alpha_2} (B_2^0 - B_2^{id})}{T} \quad (6)$$

Consequently, the following dependence can be written for the parameter  $B$  in a binary system MO-GFO:

$$B = B_1^{id} X_1 + B_2^{id} X_2 + (B_1^0 - B_1^{id}) X_1^{\alpha_1} + (B_2^0 - B_2^{id}) X_2^{\alpha_2}, \quad (7)$$

where indices 1 and 2 refer to the first and second components of a binary system,  $B^{id}$  describe the linear part of the experimental data and  $B^0$  are the pure oxide values.

One of relatively well investigated binary systems without GFO is  $\text{Al}_2\text{O}_3\text{-CaO}$ , in which viscosity as a function of composition has a maximum approximately at  $\text{Al}_2\text{O}_3/\text{CaO} = 1$ . This non-monotonous behaviour is due to formation of quasi-tetrahedra  $(\text{AlCa}_{0.5})\text{O}_4^{4-}$  that are linked together and generate a network similar to the silica tetrahedral network. This effect is usually referred to as the charge compensation effect in ternary systems, while it is not investigated in similar binary systems such as  $\text{Al}_2\text{O}_3\text{-K}_2\text{O}$  or  $\text{Al}_2\text{O}_3\text{-Na}_2\text{O}$ .

As mentioned above, viscosity in the ternary systems of the AO-MO-GFO type is also characterised by the charge-compensation effect, when an amphoteric cation (e.g.  $\text{Al}^{3+}$ ) coupled with a mono- or divalent modifier cation (e.g.  $\text{Na}^+$ ) form additional quasi-network interlinked to the silica tetrahedral network, which results in an increase of viscosity. This effect is much more pronounced for monovalent cations ( $\text{K}^+$ ,  $\text{Na}^+$ ,  $\text{Li}^+$ ), but also found in systems with divalent cations (e.g.  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ).

In a binary system AO-MO and a ternary system AO-MO-GFO the charge compensation term is proposed in the following form:

$$B \sim B^{ChC} \cdot \left( \frac{X_{AO}}{X_{AO} + X_{MO}} \right)^{\gamma_1} \cdot \left( \frac{X_{MO}}{X_{AO} + X_{MO}} \right)^{\gamma_2}, \quad (8)$$

where  $\gamma_1$  and  $\gamma_2$  are the adjustable parameters.

Thus, in the ternary system AO-MO-GFO, the parameter  $B$  (the Arrhenius activation energy reduced by the universal gas constant  $R$ ) has the following general form:

$$B = B_{AO}^{id} X_{AO} + B_{MO}^{id} X_{MO} + B_{GFO}^{id} X_{GFO} + (B_{AO}^0 - B_{AO}^{id}) X_{AO}^{\alpha_{AO}} + (B_{MO}^0 - B_{MO}^{id}) X_{MO}^{\alpha_{MO}} + (B_{GFO}^0 - B_{GFO}^{id}) \left[ \frac{X_{AO}}{X_{AO} + X_{MO}} X_{GFO}^{\beta_{AO}} + \frac{X_{MO}}{X_{AO} + X_{MO}} X_{GFO}^{\beta_{MO}} \right] + B_{AO \cdot MO}^{ChC} \left( \frac{X_{AO}}{X_{AO} + X_{MO}} \right)^{\gamma_{AO}} \left( \frac{X_{MO}}{X_{AO} + X_{MO}} \right)^{\gamma_{MO}}. \quad (9)$$

## RESULTS AND DISCUSSION

Only a part of the results obtained for binaries and ternaries is presented and discussed here. Other results are still to be obtained and might be included into the conference presentation.

## Viscosities of binary silicates

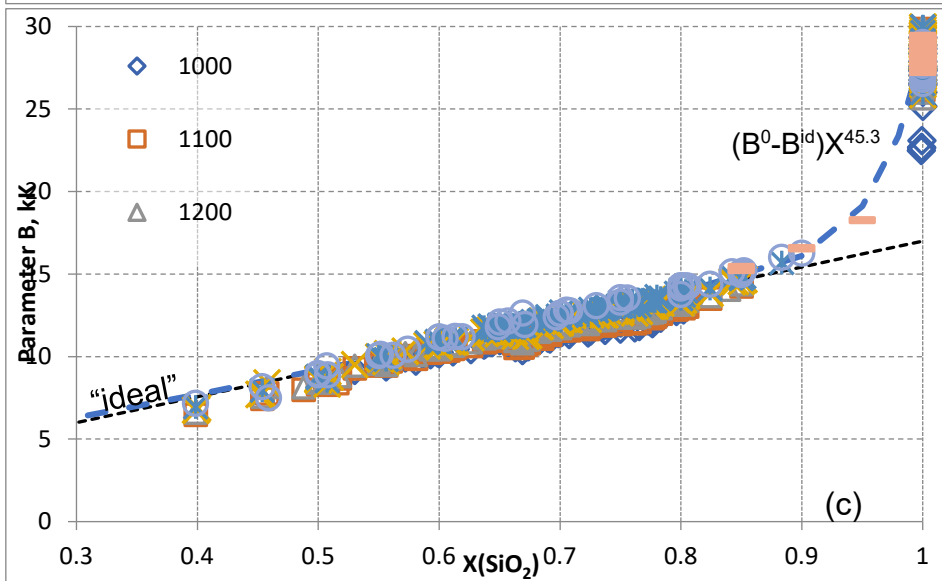
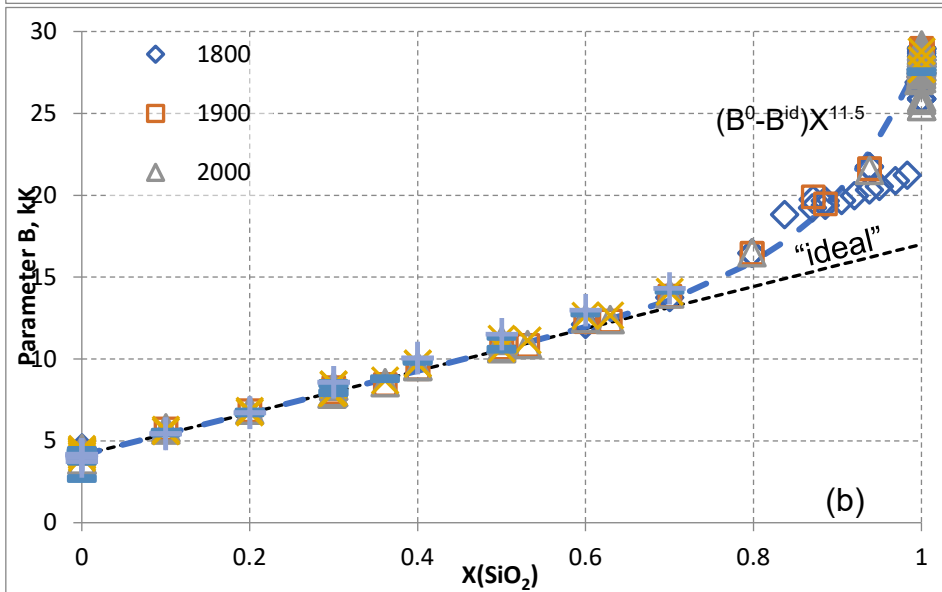
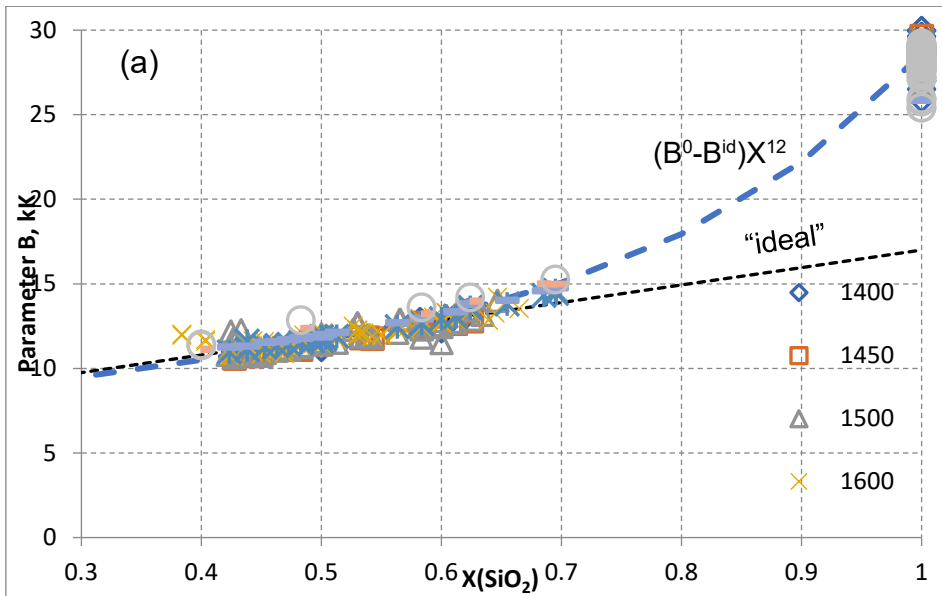
Tables 2-3 represent the model parameters for unary systems and binary silicate systems, respectively. Parameters for several pure oxides were taken from the review by Kondratiev (2024). In the present work the parameters of those pure oxides, for which no experimental data are available, were made equal to the corresponding linear parameters obtained as described above. Also, the  $\alpha$  parameters for these oxides were not calculated, since it is not possible without experimental data. The “ideal” SiO<sub>2</sub> parameters listed in Table 3 were calculated as the average for all systems from Table 1 except K<sub>2</sub>O-SiO<sub>2</sub> and MnO-SiO<sub>2</sub>.

TABLE 2 – Model parameters for pure oxide melts.

	Al <sub>2</sub> O <sub>3</sub>	PbO	SiO <sub>2</sub>	BaO	CaO	K <sub>2</sub> O	Li <sub>2</sub> O	MgO	MnO	Na <sub>2</sub> O
log(A <sup>0</sup> /Pa*s)	-3.20	-4.02	-7.73	-5.86	-6.18	-8.56	-4.31	-5.21	-3.73	-6.83
B <sup>0</sup> (kK)	4.13	2.30	28.54	5.24	5.85	7.49	0.06	4.53	0.74	4.50
Reference	Kondratiev 2024			This work						

TABLE 3 – Model parameters for binary silicate melts obtained in this work.

System	Coefficients			
	log(A <sup>id</sup> /Pa*s)	B <sup>id</sup> (kK)	$\alpha_1$	$\alpha_2$ (SiO <sub>2</sub> )
SiO <sub>2</sub>	-3.72	10.85	-	-
Al <sub>2</sub> O <sub>3</sub> -SiO <sub>2</sub>	-3.07	1.63	3.46	11.46
BaO-SiO <sub>2</sub>	-5.86	5.24	-	14.69
CaO-SiO <sub>2</sub>	-6.18	5.85	-	12.00
K <sub>2</sub> O-SiO <sub>2</sub>	-8.56	7.49	-	67.98
Li <sub>2</sub> O-SiO <sub>2</sub>	-4.31	0.06	-	23.51
MgO-SiO <sub>2</sub>	-5.21	4.53	-	12.00
MnO-SiO <sub>2</sub>	-3.73	0.74	-	10.00
Na <sub>2</sub> O-SiO <sub>2</sub>	-6.83	4.50	-	45.26
PbO-SiO <sub>2</sub>	-5.52	1.22	8.66	9.23





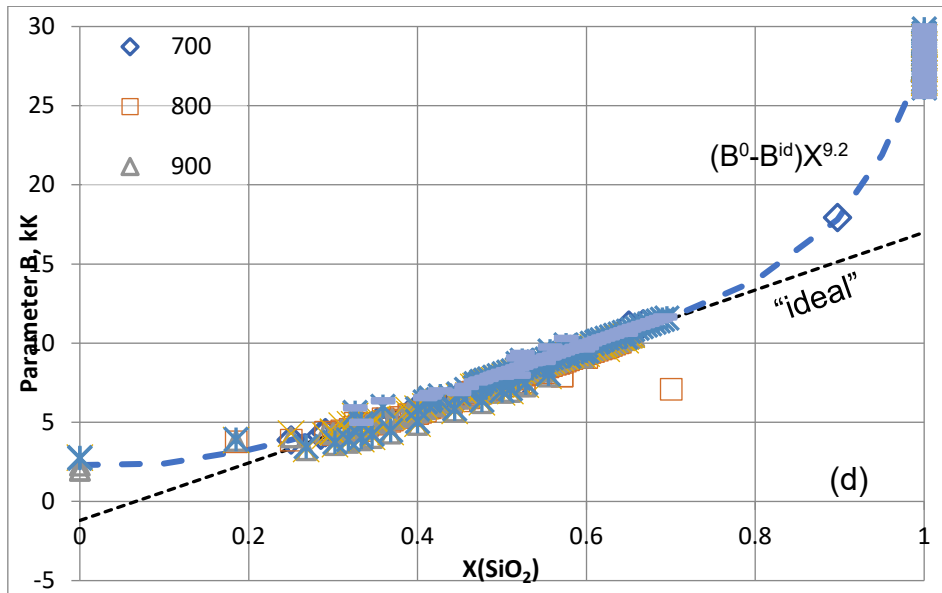


FIG 3 – The parameter  $B$  as a function of the  $\text{SiO}_2$  molar fraction for (a)  $\text{CaO-SiO}_2$ , (b)  $\text{Al}_2\text{O}_3\text{-SiO}_2$ , (c)  $\text{Na}_2\text{O-SiO}_2$ , (d)  $\text{PbO-SiO}_2$ .

The parameter  $B$  in binary silicate systems is expressed by Equation 7, and the values of the parameters of Eq. 7 can be found in Table 3. The polymerisation effect is introduced into the model simply as the difference between the pure oxide and linear (“ideal”)  $B$  parameters multiplied by the  $\text{SiO}_2$  concentration to a certain power (the  $\alpha$  parameter). Figures 3(a-d) show four examples of the compositional dependence of the parameter  $B$  (Eq. 7) for the  $\text{CaO-SiO}_2$ ,  $\text{Al}_2\text{O}_3\text{-SiO}_2$ ,  $\text{Na}_2\text{O-SiO}_2$  and  $\text{PbO-SiO}_2$  systems. Different symbols denote different temperatures (in  $^\circ\text{C}$ ). It can be seen that the polymerisation effect is more pronounced in the  $\text{Na}_2\text{O-SiO}_2$  than in the  $\text{Al}_2\text{O}_3\text{-SiO}_2$  or  $\text{PbO-SiO}_2$  systems. The region that corresponds to the polymerisation effect in the  $\text{CaO-SiO}_2$  is not investigated, so it is problematic to assume the exact form of the compositional dependence with increasing  $X(\text{SiO}_2)$ . Therefore the parameter  $B$  was chosen arbitrarily from 10 to 15. For the  $\text{PbO-SiO}_2$  system two structural effects can be observed: the polymerisation effect at higher  $\text{SiO}_2$  and similar but less pronounced at lower  $\text{SiO}_2$ , with the quasi-linear region at intermediate  $\text{SiO}_2$ .

This linear region at intermediate  $\text{SiO}_2$  concentrations in binary silicate systems may be considered as the compositional region, in which no structural rearrangement takes place. Increasing  $\text{SiO}_2$  leads to the polymerisation effect, while  $\text{SiO}_2$  decreasing results in other, similar to polymerisation, effect of structural rearrangement. Both effects result in viscosity increasing. Polymerisation is a type of clustering, structural network formation. At the other end also some clustering occurs.

Figures 4-12 represent the experimental and calculated viscosities in the  $\text{Al}_2\text{O}_3\text{-SiO}_2$ ,  $\text{CaO-SiO}_2$ ,  $\text{K}_2\text{O-SiO}_2$ ,  $\text{Li}_2\text{O-SiO}_2$ ,  $\text{MgO-SiO}_2$ ,  $\text{MnO-SiO}_2$ ,  $\text{Na}_2\text{O-SiO}_2$ , and  $\text{PbO-SiO}_2$  systems. It can be seen that the model describes the experimental data with a reasonable accuracy, although only 2-3 model parameters are used for each binary system.

Figures 4-12 demonstrate reasonably good agreement with the available experimental data. Perhaps, a poorest, but still reasonable agreement among all systems is shown in the  $\text{Al}_2\text{O}_3\text{-SiO}_2$  system (Figure 4). This is mainly due to slightly lower values of the  $\text{SiO}_2$  ideal parameters than for the  $\text{Al}_2\text{O}_3\text{-SiO}_2$  system (see Table 1), and also because a low-temperature experimental data is described with a better agreement than the viscosity data at higher temperatures.

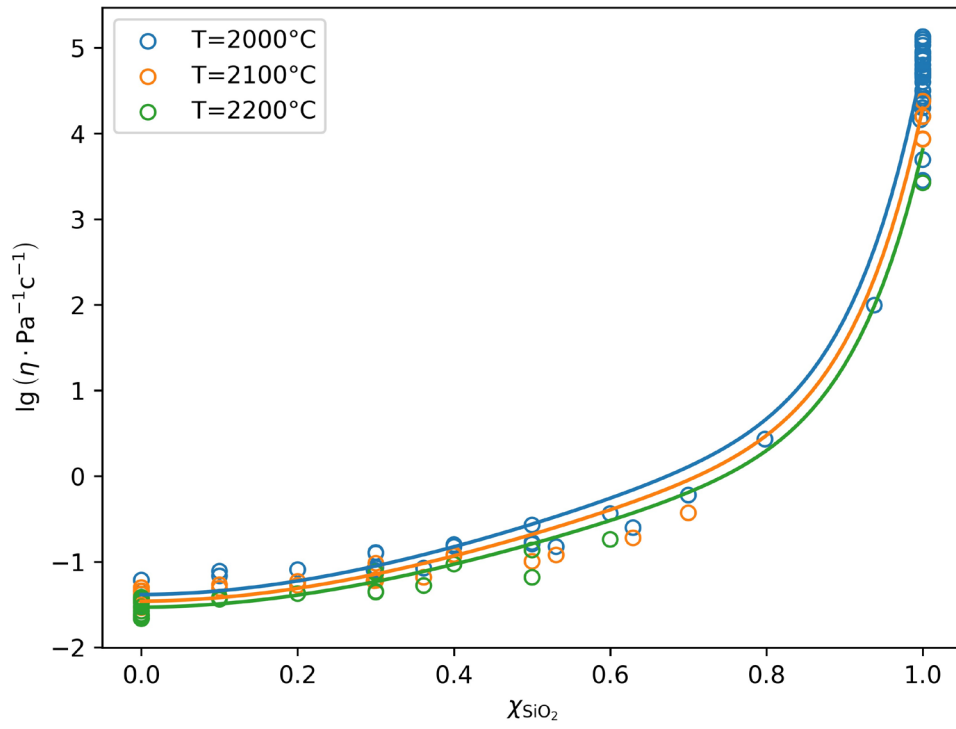


FIG 4 – Viscosities of the Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system.

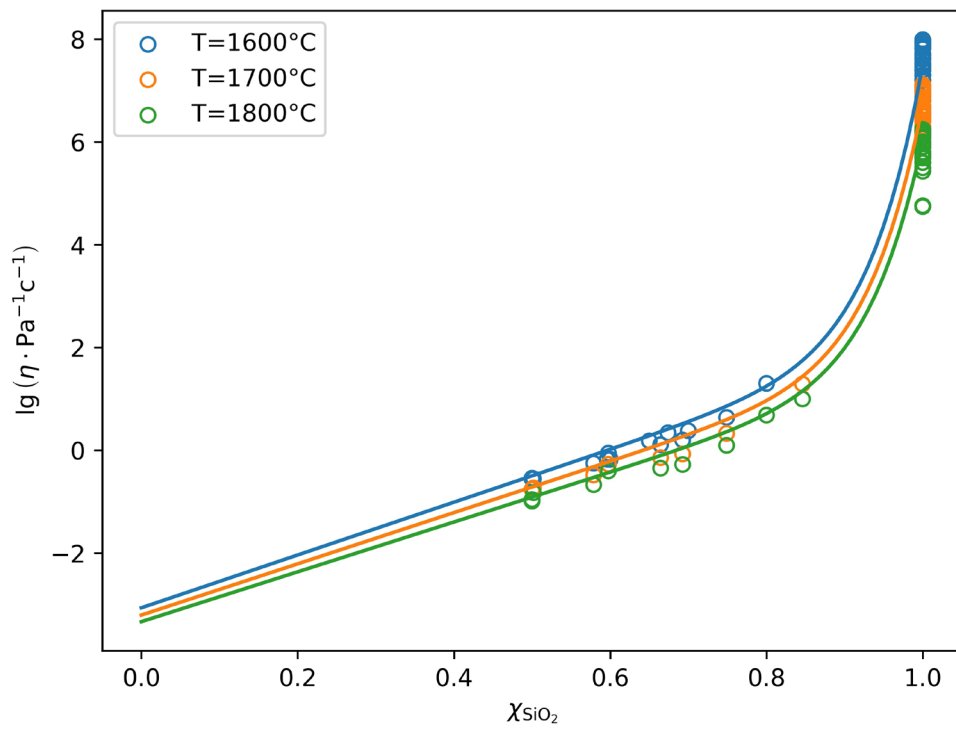


FIG 5 – Viscosities of the BaO-SiO<sub>2</sub> system.

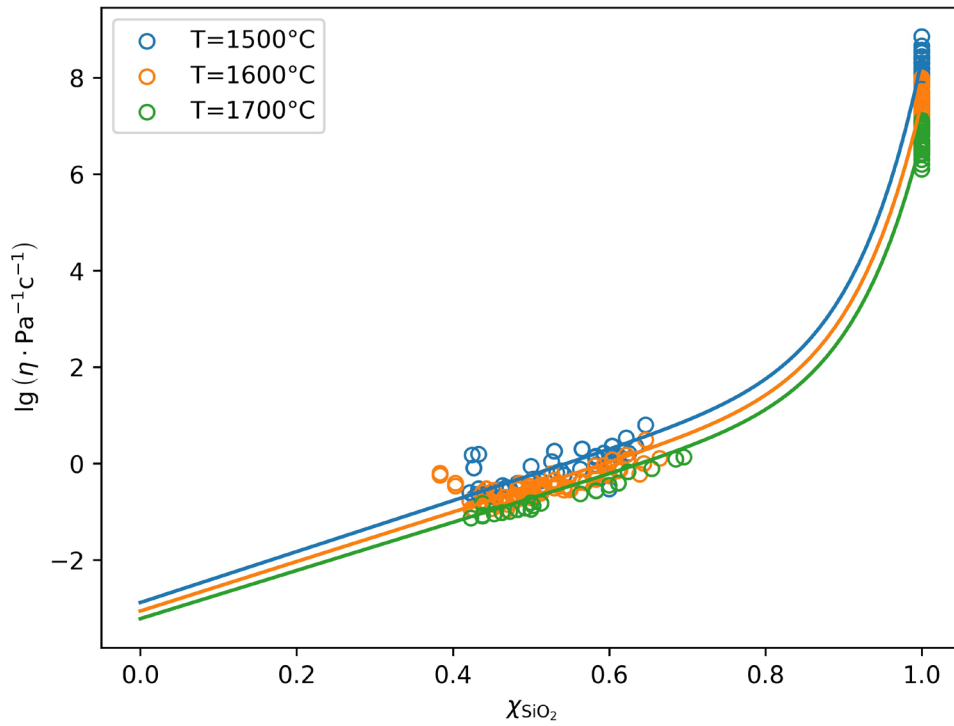


FIG 6 – Viscosities of the CaO-SiO<sub>2</sub> system.

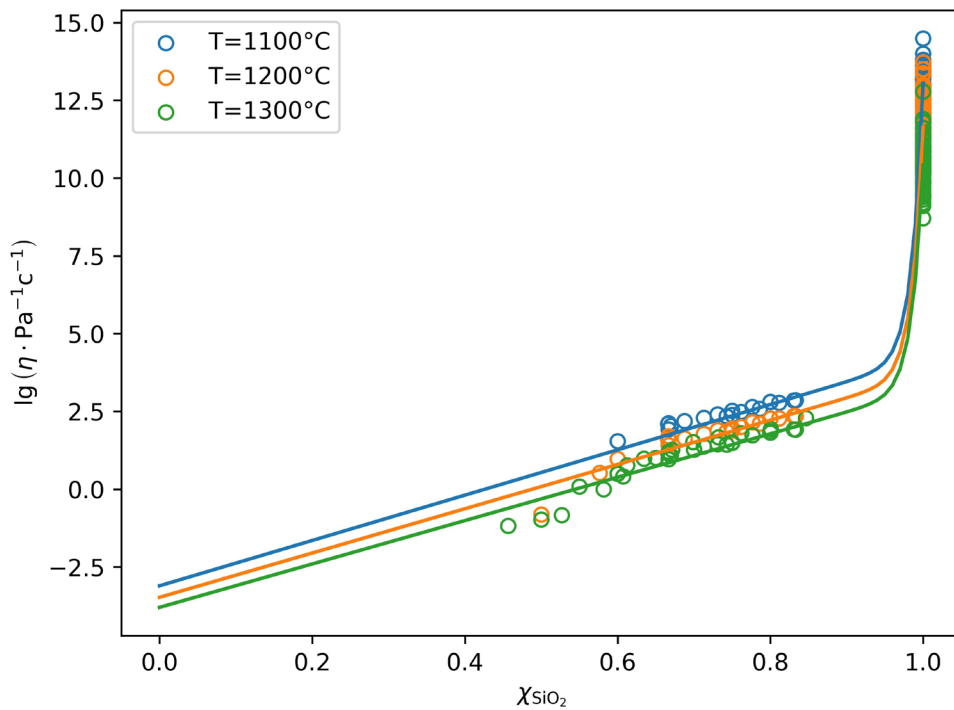


FIG 7 – Viscosities of the K<sub>2</sub>O-SiO<sub>2</sub> system.

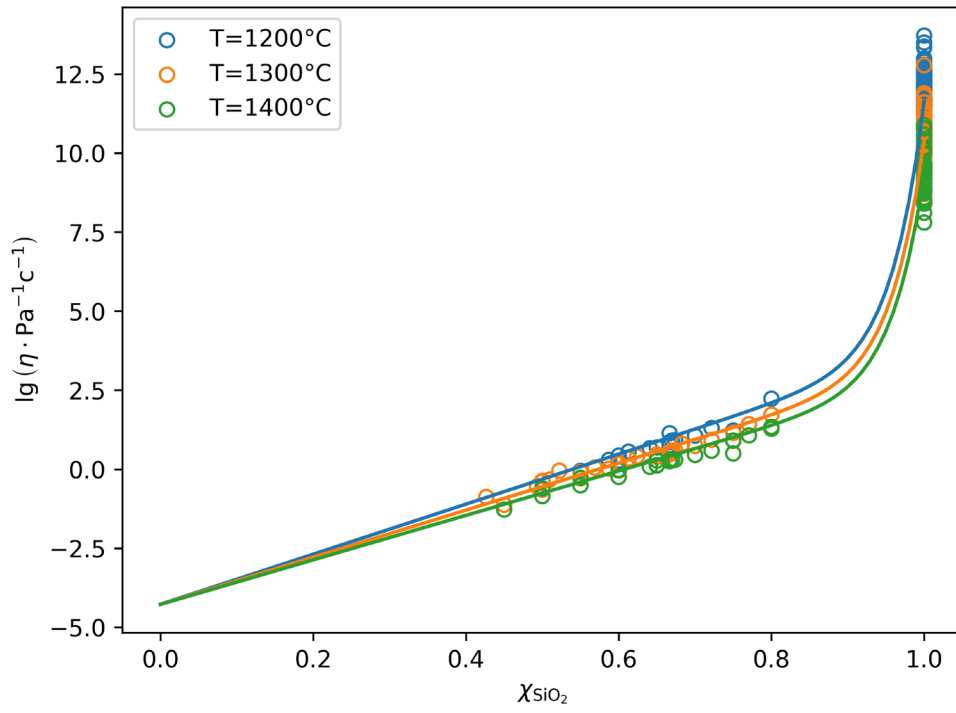


FIG 8 – Viscosities of the  $\text{Li}_2\text{O-SiO}_2$  system.

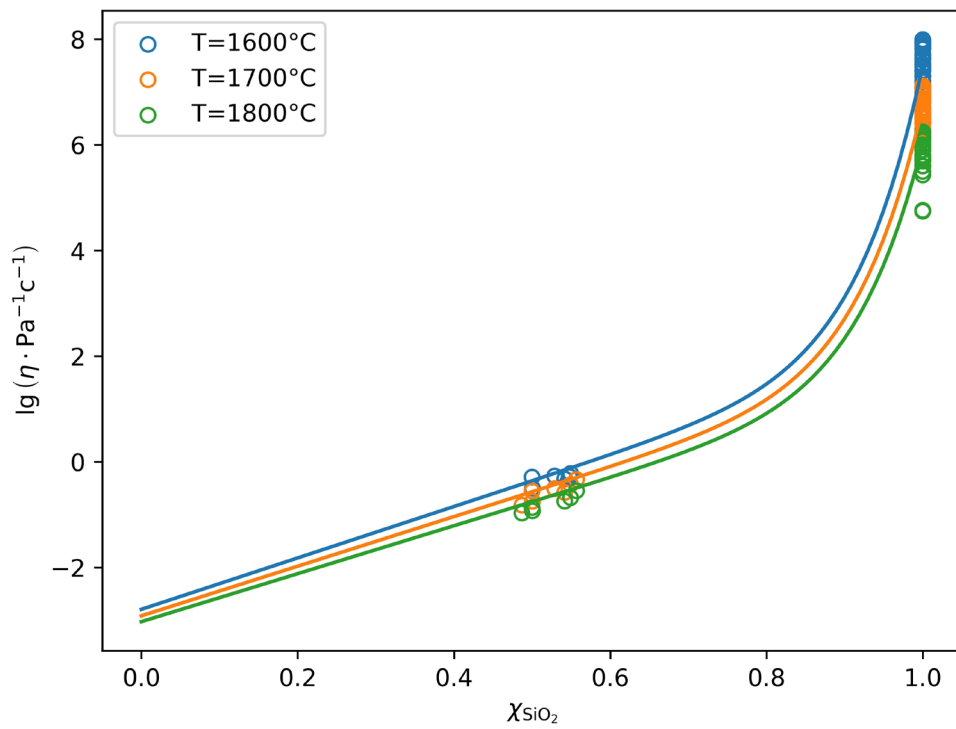


FIG 9 – Viscosities of the  $\text{MgO-SiO}_2$  system.

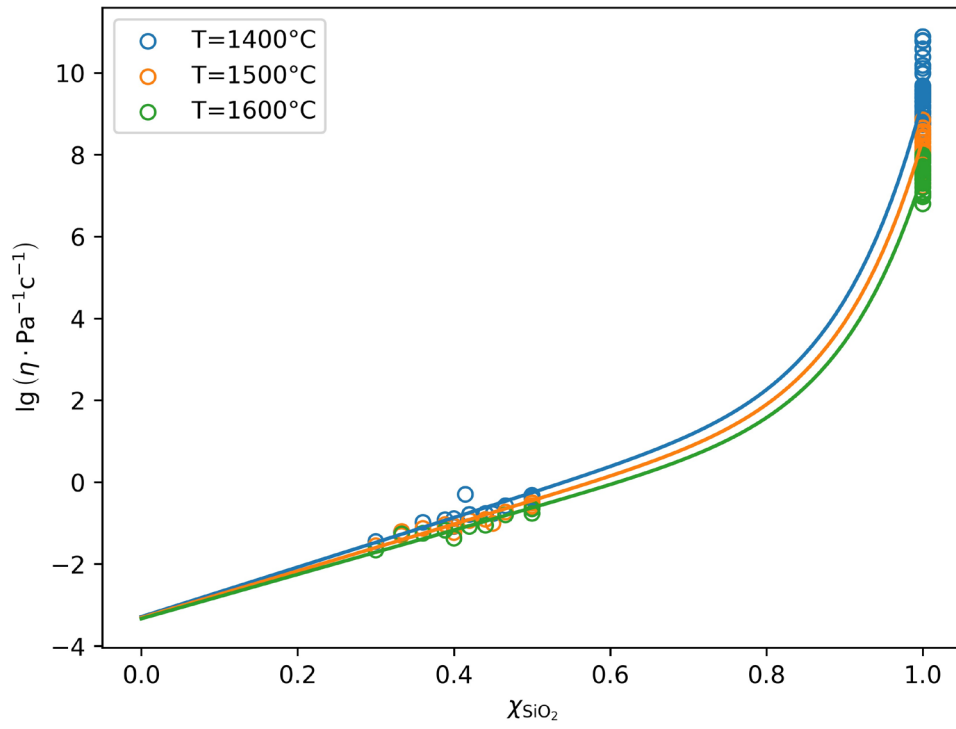


FIG 10 – Viscosities of the MnO-SiO<sub>2</sub> system.

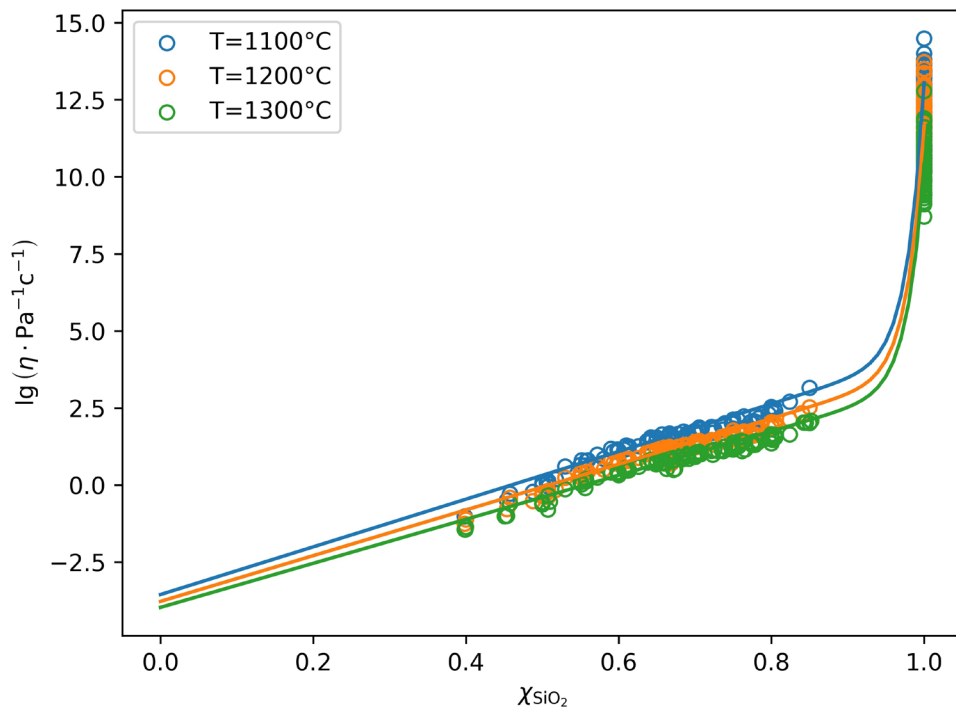


FIG 11 – Viscosities of the Na<sub>2</sub>O-SiO<sub>2</sub> system.

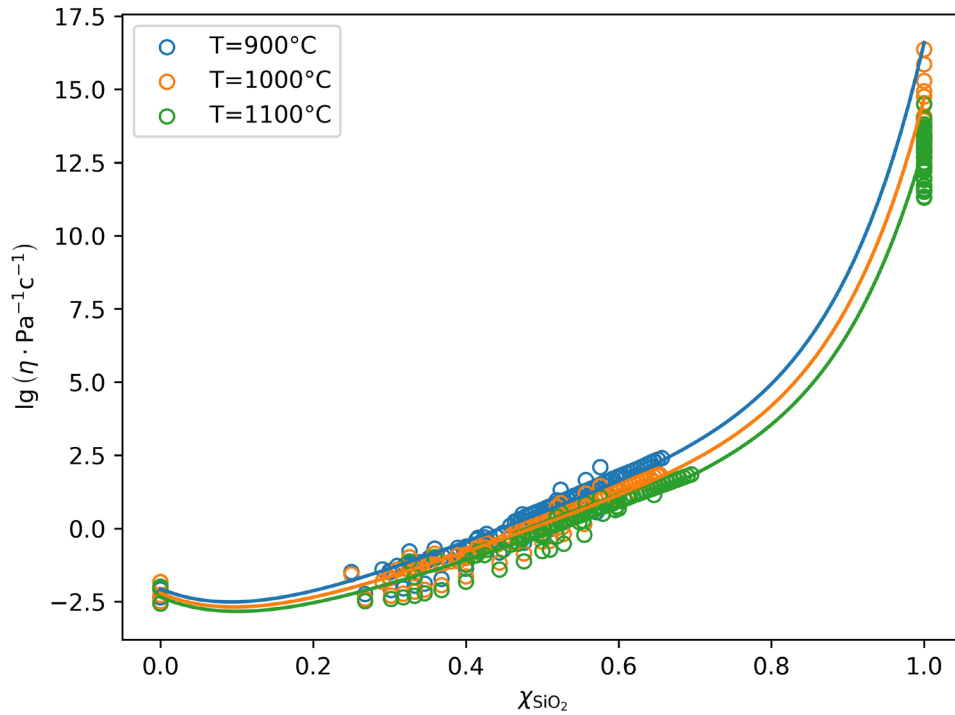


FIG 12 – Viscosities of the PbO-SiO<sub>2</sub> system.

### Viscosities of binary borates and other binaries

Table 4 represent the model parameters for B<sub>2</sub>O<sub>3</sub> and B<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>. The parameters for pure B<sub>2</sub>O<sub>3</sub> are calculated using the experimental data at temperatures above the melting point and the Arrhenius equation (1). The experimental data for borates are scarce and controversial, the only reliable data is found to be present in the B<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system. The B<sub>2</sub>O<sub>3</sub> “ideal” parameters were optimised mainly on the basis of B<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>. The other end-member parameters for molten B<sub>2</sub>O<sub>3</sub>-BaO, B<sub>2</sub>O<sub>3</sub>-Na<sub>2</sub>O, B<sub>2</sub>O<sub>3</sub>-PbO were taken from the corresponding silicates listed in Table 3.

TABLE 4 – Model parameters for binary borate melts.

System	Coefficient					
	log(A <sup>0</sup> /Pa*s)	B <sup>0</sup> (kK)	log(A <sup>id</sup> /Pa*s)	B <sup>id</sup> (kK)	α <sub>1</sub>	α <sub>2</sub>
B <sub>2</sub> O <sub>3</sub>	-2.14	3.96	-0.83	2.06	-	-
B <sub>2</sub> O <sub>3</sub> -SiO <sub>2</sub>	-	-			4.64	4.61

Figures 13-15 represent the results of model calculation for two borate systems (B<sub>2</sub>O<sub>3</sub>-PbO and B<sub>2</sub>O<sub>3</sub>-Na<sub>2</sub>O). Borate melts are less investigated and are characterised by larger experimental scatter than silicate melts. It can be seen that the model cannot describe complex compositional effects present in the borate binary systems. It should be mentioned that the model parameters for PbO and Na<sub>2</sub>O (the linear part) were taken from the corresponding silicate binaries, while the model parameters for B<sub>2</sub>O<sub>3</sub> were obtained on the basis of the B<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system.

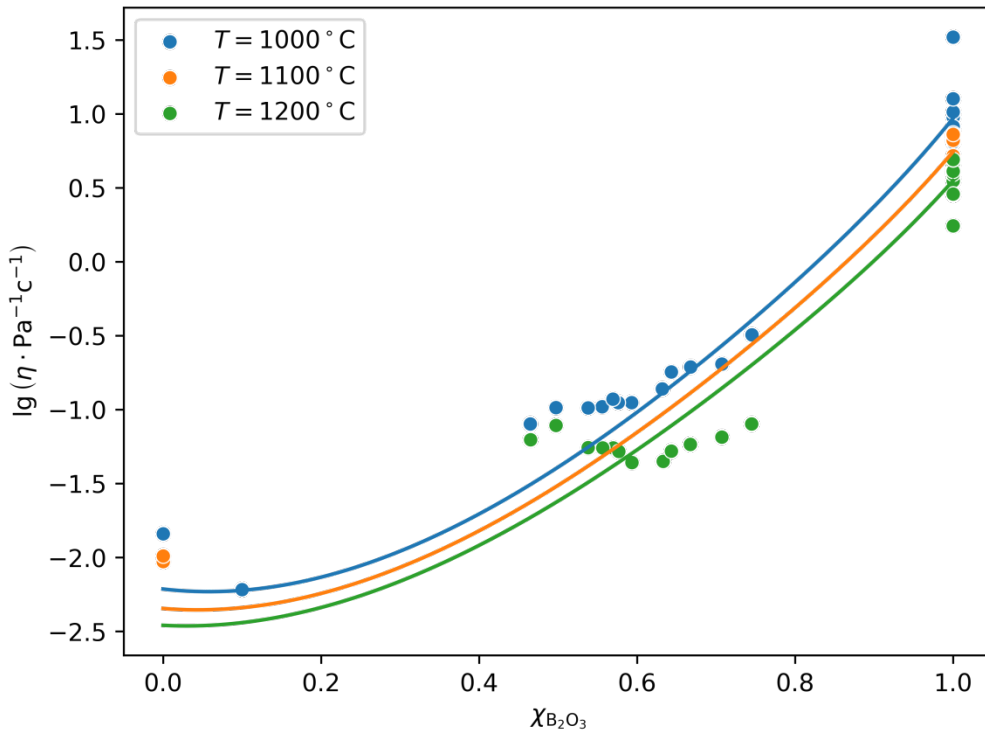


FIG 13 – Viscosities of the  $B_2O_3$ -PbO system.

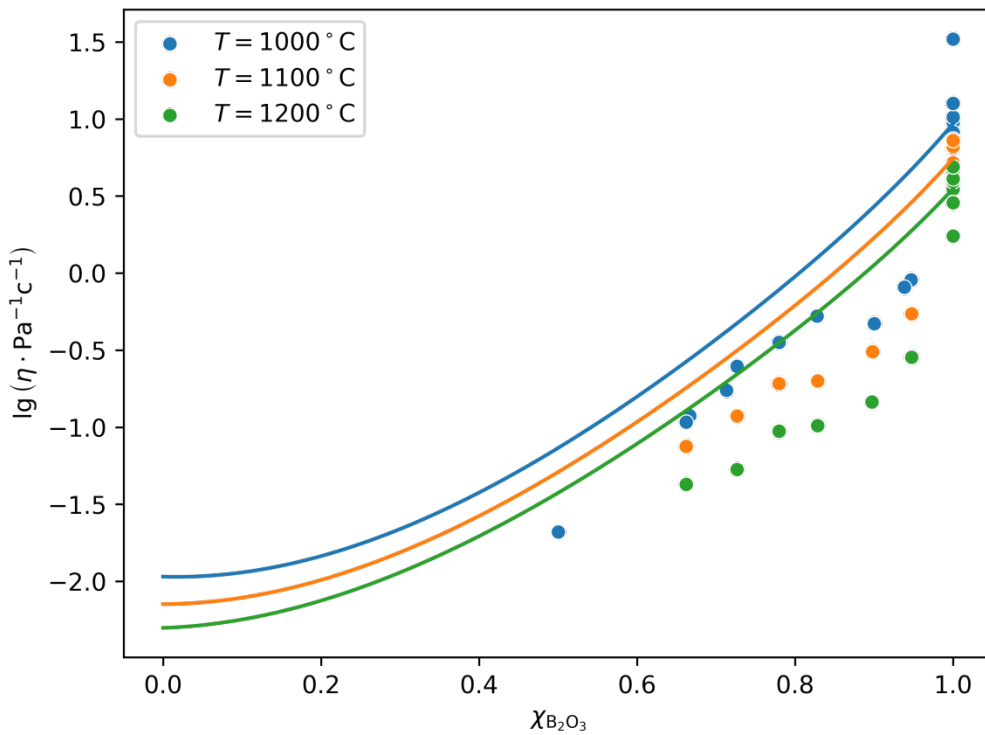


FIG 14 – Viscosities of the  $B_2O_3$ -Na<sub>2</sub>O system.

Viscosity of the  $B_2O_3$ -SiO<sub>2</sub> melt is shown in Figure 15. It can be seen that agreement between the experimental and calculated viscosities is reasonably well.

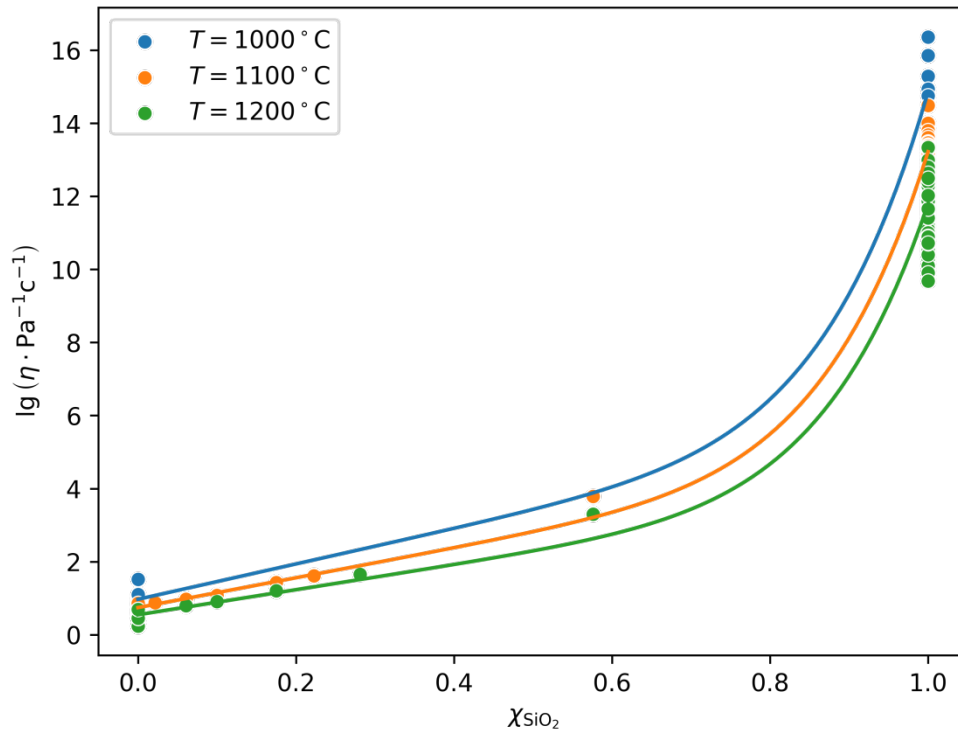


FIG 15 – Viscosities of the B<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> system.

Viscosity of the Al<sub>2</sub>O<sub>3</sub>-CaO system is shown in Figure 16 as a function of the Al<sub>2</sub>O<sub>3</sub>/(Al<sub>2</sub>O<sub>3</sub>+CaO) ratio. Parameters are listed in Table 5. The agreement between the experimental and calculated values is reasonable.

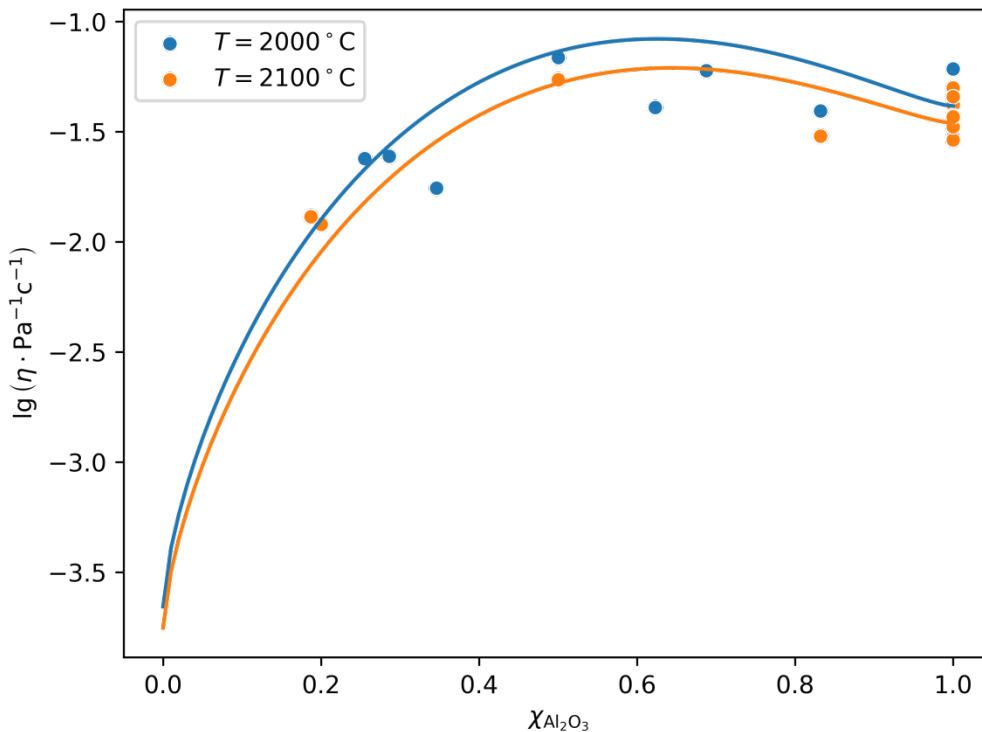


FIG 16 – Viscosities of the Al<sub>2</sub>O<sub>3</sub>-CaO system.

### Viscosities of ternary systems

Table 5 provides the model parameters for the charge compensation term in one binary system, Al<sub>2</sub>O<sub>3</sub>-CaO, and two ternary systems, Al<sub>2</sub>O<sub>3</sub>-Na<sub>2</sub>O-SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>-CaO-SiO<sub>2</sub>.



TABLE 5 – The charge compensation model parameters for Al<sub>2</sub>O<sub>3</sub>-CaO and ternary oxide melts.

System	Coefficient		
	B <sup>ChC</sup> (kK)	γ <sub>1</sub>	γ <sub>2</sub>
Al <sub>2</sub> O <sub>3</sub> -CaO	10.66	0.64	1.12
Al <sub>2</sub> O <sub>3</sub> -Na <sub>2</sub> O-SiO <sub>2</sub>	829.45	4.53	3.20
Al <sub>2</sub> O <sub>3</sub> -CaO-SiO <sub>2</sub>	3.57	1	1

Viscosity of the Al<sub>2</sub>O<sub>3</sub>-Na<sub>2</sub>O-SiO<sub>2</sub> is presented in Figure 17. The agreement between the calculated and experimental values is good.

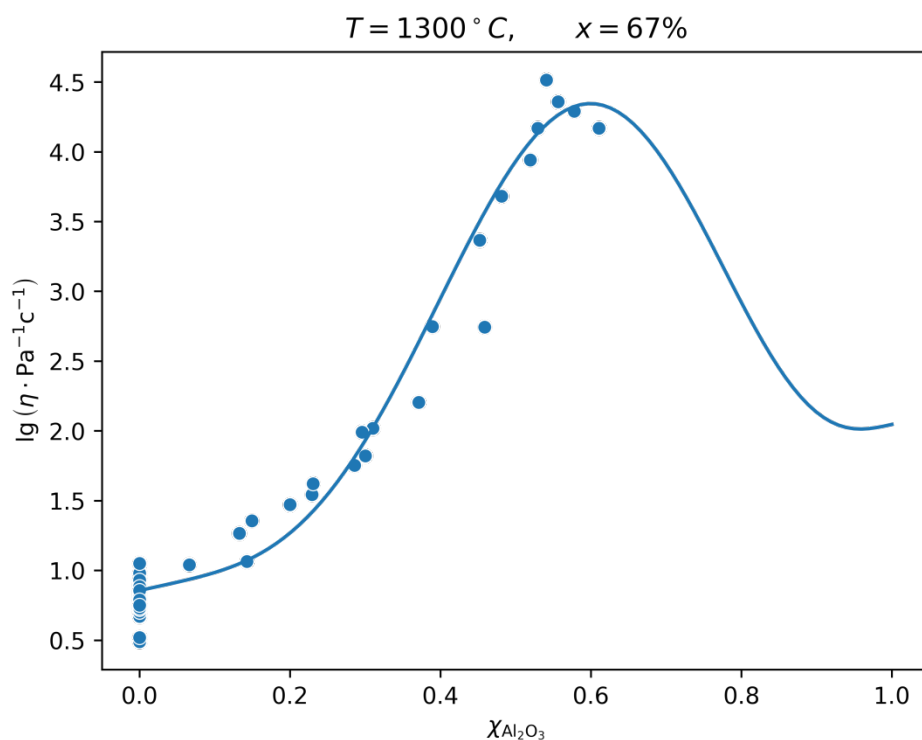


FIG 17 – Viscosities of the Al<sub>2</sub>O<sub>3</sub>-Na<sub>2</sub>O-SiO<sub>2</sub> system at X(SiO<sub>2</sub>)=0.67 and T=1300°C.

Experimental viscosities of Al<sub>2</sub>O<sub>3</sub>-CaO-SiO<sub>2</sub> are shown in Figure 11 with preliminary coefficients of the charge compensation term. It can be seen that agreement between calculated and experimental viscosities is reasonable.

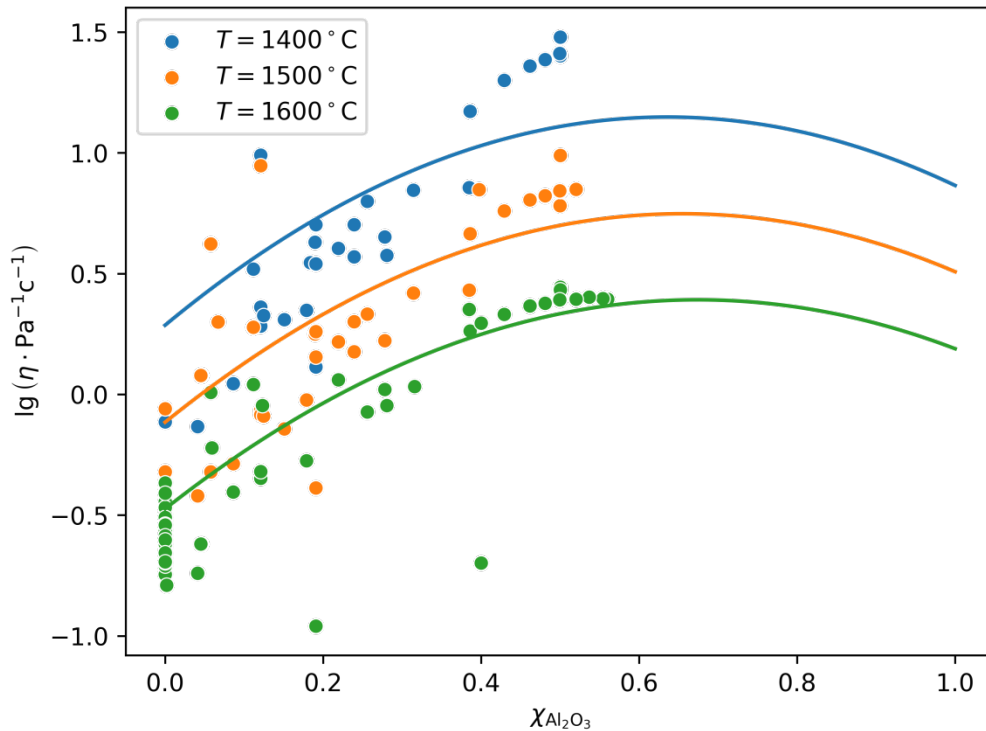


FIG 18 – Viscosities of the  $\text{Al}_2\text{O}_3\text{-CaO-SiO}_2$  system at  $X(\text{SiO}_2)=0.5$  and  $T=1400, 1500, 1600^\circ\text{C}$ .

The experimental data and calculated viscosities for the other AO-MO-GFO systems (e.g.  $\text{Al}_2\text{O}_3\text{-K}_2\text{O-SiO}_2$ ,  $\text{Al}_2\text{O}_3\text{-Li}_2\text{O-SiO}_2$ ,  $\text{Al}_2\text{O}_3\text{-MgO-SiO}_2$ ) might be added to the conference presentation.

## CONCLUSIONS

The simple polynomial model is constructed on the basis of analysis of experimental data in binary silicate systems. The model introduces a linear compositional dependence and polymerisation effect for silicate systems. In ternary systems a charge compensation term is added to describe the experimental data.

A significant rise of viscosity with approaching pure  $\text{SiO}_2$  (the so-called polymerisation effect) is described reasonably well in different binary silicate systems.

The presence of viscosity maximum in ternary systems of the AO-MO-GFO type (the charge compensation effect) is described reasonably well in two ternaries ( $\text{Al}_2\text{O}_3\text{-Na}_2\text{O-SiO}_2$  and  $\text{Al}_2\text{O}_3\text{-CaO-SiO}_2$ ) systems.

More complex viscosity effects (e.g. in binary borates) are not described. However, viscosity in the  $\text{B}_2\text{O}_3\text{-SiO}_2$  system is described reasonably well.

## ACKNOWLEDGEMENTS

This work was financially supported within the framework of the State Program 'Chemical Thermodynamics and Theoretical Materials Science' at the Lomonosov Moscow State University (N°121031300039-1).

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