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General method for calculating the properties of oxide glasses and glass forming melts from their composition and temperature

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A new method for calculating various properties of glasses and glass forming melts has been proposed. The method covers such properties as viscosity and standard points, density, heat capacity and enthalpy, refractive index and its dispersion, coefficient of thermal expansion, surface tension, elastic moduli, and ultrasonic velocities. Temperature dependences of the properties (except for elastic and acoustic properties) could also be calculated. The above mentioned properties could be calculated for glasses and glass forming melts belonging to various oxide systems: 64 oxides can be included in the calculation. The method is based on assuming chemical equilibria between structural groups, which are found in glass forming melts. The results of calculations have been compared with the experimental data for properties of about 100 000 glasses and glass forming melts using the MDL SciGlass information system. The results of the comparison show that the proposed method allows the prediction of properties with a precision as high, on the whole, as the best known methods.

The first system for the calculation of glass properties from their composition was proposed by Winkelmann & Schott^(1–4) at the end of the 19th century. Since then the calculation of glass properties has been the subject of hundreds of publications. The most recent attempt to summarise them is a monograph by Volf⁽⁵⁾ published in 1985. It describes the best known methods, such as those of Gehlhoff & Thomas,^(6–8) Gilard & Dubrul,⁽⁹⁾ Huggins & Sun,⁽¹⁰⁾ Appen,^(11,12) Demkina,^(13,14) Gan Fuxi,^(15,16) etc. Since 1985 some new methods have been proposed to extend the possibilities of such calculations (Refs 17–22, etc).

The importance of solving this problem is so evident that it does not need any substantiation. Nevertheless, the solution is far from being complete. ‘Prediction of the glass properties of a multicomponent system from its chemical composition is desirable’⁽²³⁾ – these or similar words begin many recent

papers on this subject.

Even calculation of viscosity, the most important glass property determining conditions for the majority of glass making processes, has until very recently only been possible in the narrow range of compositions. Lakatos and co-workers,^(24–29) Lyon,⁽³⁰⁾ Mazurin *et al.*,⁽³¹⁾ Hrma and co-workers,^(32,33) Okhotin,⁽³⁴⁾ Bottinga⁽³⁵⁾ and others have all proposed methods for the calculation of viscosities of glass forming melts. However, the area of applicability of all these methods is significantly restricted¹ (some details about these methods are given in Ref. 36).

One of the most important practical tasks is the development of general methods allowing calculation of many properties of glasses and glass forming melts over a wide range of compositions and temperatures with a relatively high precision. This might considerably simplify a search for compositions of glasses with desired sets of properties. The development of such a method is the subject of the work presented here. The result of this work is the proposal of a new general method for calculating the properties of oxide glasses and glass forming melts.

The details of the proposed method have been described in the author’s previous papers.^(37–46) However, a reader of these papers (except for the Ref. 37) cannot get a general impression on the overall method. Explanation of the backgrounds and main features of the method is the subject of this paper.

Characterisation of known approaches to calculate properties from composition

Additive methods

The majority of the known methods for calculating the properties of oxide glasses from composition are based on additive formulae that represent the calculated property as a linear function of oxide concentrations.

¹ It should be noted that if the authors of the method do not specify the range of applicability (see Ref. 25 as an example), this does not mean that this area is unlimited – actually it is restricted by the area of the experimental data used by the authors of the methods.

Additive methods allow quite a large number of oxides to be included in calculation (see, for instance Ref. 47), if their concentrations vary within relatively narrow ranges. However the composition dependence of the majority of glass and melt properties is not quite linear over wider composition ranges. Thus there is a need to correct the calculation algorithm. To take deviations from simple additivity into account, the authors of many methods (Refs 9–14 and many others) consider partial coefficients, which vary with composition.² Such an approach is effective in silicate systems. One can divide them into several sections, so that all coefficients of the additive formula may be considered as constants in any of them.

The known attempts to extend this approach to non-silicate glasses (for example, Refs 15, 16, 48) have been less successful (see Refs 49, 50). The failures are caused by the fact that the dependence on composition of glass properties in silica free systems is more complicated than in silicate systems. Hence it is difficult to separate out compositional areas, in which the additive formulae are approximately followed, and adjustment of the parameters in the formulae is sometimes impossible because of the lack of initial experimental data.

Methods based on polynomial regression

Quite a few authors (Refs 23, 51–53, etc.) attempted to use polynomial equations to describe the concentration dependences of glass properties. However, so far such polynomial equations have been for either for binary and ternary systems, or within narrow concentration ranges. The possibility of developing a general method for the calculation of properties of multicomponent glasses over wide compositional ranges using polynomial equations seems questionable in many respects; this is discussed in Ref. 49.

Structural–chemical models

The known structural–chemical models (Refs 54–56, etc) are based on the idea, first stated by Müller (to the best of the author's knowledge), that the structure of oxide glasses could be described in terms of structural groups (see the detailed consideration of his approach in Ref. 57). These groups are multiatomic complexes formed as a result of chemical interactions between glass components. It is suggested that all structural units in glasses keep a constant chemical composition independent of the surroundings of a complex.

Within the majority of the structural–chemical models all supposed chemical reactions leading to formation of structural groups are considered as irreversible. Their order and directions are defined by empirical rules. These rules are mostly based on the analysis of structural data obtained by NMR and other direct structural methods. Thus such models allow calculation of the presumed concentrations of all assumed structural groups contained in any particular glass. In Refs 58, 59 the formulae for calculating density and

some other properties of borate and borosilicate glasses from their composition are proposed using one of the structural–chemical models.⁽⁵⁶⁾ Some backgrounds of using the structural models for calculation of other glass properties are presented in Ref. 60.

From the standpoint of abstract science, structural–chemical models have some advantages, firstly, the fact that they quite precisely reflect the interrelation 'composition–structure–property.' However these models are not of practical importance, since calculations by them are available only when direct data on glass structure are present. So far, such data are available only for very small number of the simplest glass forming systems, to which the real possibilities of calculation are confined.

Thermodynamic model

There are many publications devoted to calculation of thermodynamic properties of glass forming substances (see Refs 61–63, etc). These works are outside the scope of the present article. However, recently Shakhmatkin and co-workers (see Refs 64–66 where references to previous papers are given) have proposed a thermodynamic model for calculation of physical (non-thermodynamic) properties of glasses, such as density, electrical conductivity, etc. Therefore, it seems reasonable to consider this model briefly.

The ideology and mathematical apparatus of this model are very close to the structural–chemical models described above. The main distinctive features of the thermodynamic model consist of the following. Firstly, it is postulated that the composition of all structural groups coincides with the composition of thermodynamically stable compounds. Secondly, all data required to determine the parameters expected for calculating the assumed concentrations of structural groups must be available from thermodynamic data, and the calculation is performed using the chemical equilibria equations.

The main obstacle to a wide practical use of the model of Shakhmatkin and co-workers for the calculation of glass properties is the lack of data on thermodynamics of glass forming substances. At present, such information is only available for glass forming substances with the simplest chemical compositions, and most likely the situation will probably change little within the near future. The prospects for practical use of this model are therefore not very encouraging.

Other models

Besides the above stated models, some other models describing the dependence of glass properties on composition, such as molecular dynamic models,^(67–69) the phase model⁽⁷⁰⁾ and others have been proposed. These models are, as a rule, exclusively of theoretical interest. These models have the same limitation as the calculations by structural–chemical and thermodynamic models in that they require data that are not known over a wide compositional area and thus are not of practical importance.

To summarise, it is clear that in spite of quite significant differences between the methods for calculat-

² Thus, the mentioned models are not quite linear. However, the authors of the models often call them 'additive', having in mind that the concentration dependences of the calculated properties are, in whole, close to linear.

ing glass properties from composition, they have a common defect that limits their applicability, namely that there is a lack of initial data required to determine the parameters used in the calculation. In other words, it is not the theoretical basis of the method but the presence or absence of the data required to determine the parameters that limits the possibility of extending the calculation to a wide range of compositions.

It is obvious that calculation of glass properties over a wide enough composition area can only be constructed on an empirical basis. In this case it is possible to use a very large amount of the known data on the properties of oxide glasses and melts to determine the parameters used in the calculations. At present, the most complete source of such data is the MDL SciGlass information system,⁽⁷¹⁾ which contains the data on numerous properties of 222 590 glasses and glass forming melts taken from 3699 patents and 11 521 original papers published by 13 339 authors in 828 journals and books from the end of XIX to the beginning of XXI century.³

Two significant remarks need to be made about this data and the proposed method of calculation.

Firstly, a large body of the initial data itself does not guarantee a correct determination of all parameters used within the calculation. That is possible only when the total number of parameters is not large, and parameters themselves are not related to narrow composition ranges. Otherwise we will face the same problems as the authors of the additive and polynomial methods.

Secondly, while developing even an empirical method of calculation it would be erroneous to ignore the known information on glass structure. This information has been repeatedly involved⁽¹⁰⁻¹⁷⁾ and, in the author's opinion, should be involved to correctly account for the interrelations between composition, structure and properties and thereby to improve the precision of calculation of glass properties from composition. However, the role of the data on glass structure should be an auxiliary one, and the absence of these data should not prevent the calculations being performed.

The proposed method

Range of applicability

The proposed method allows the calculation of properties of single phase (i.e. non-liquated and non-crystallised) oxide glasses and melts.

The following quantities may be calculated: glass transition temperature, viscosity (from 10^2 to $10^{13.5}$ dPas), density (from 0 to 2000 K), thermal expansion coefficient (from 200 K to the lower boundary of the glass transition range), surface tension of melts (from 1200 to 1900 K), as well as the following quantities at 20°C: refractive index, mean and partial dispersion, Abbé number, Young's modulus, shear modulus, and longitudinal and transverse ultrasonic velocities.

Calculations can cover almost all known types of oxide glasses: silicate, germanate, borate, aluminate, phosphate, vanadate, molybdate, niobate, arsenite,

tellurite, tungstate, etc, including mixed systems. In total 64 oxides are included in the calculations: Li_2O , Na_2O , K_2O , Rb_2O , Cs_2O , Cu_2O , Ag_2O , Tl_2O , BeO , MgO , CaO , SrO , BaO , MnO , CoO , NiO , SnO , ZnO , CdO , PbO , FeO , CuO , B_2O_3 , Al_2O_3 , Ga_2O_3 , Fe_2O_3 , Cr_2O_3 , Ti_2O_3 , In_2O_3 , La_2O_3 , Y_2O_3 , Sc_2O_3 , Gd_2O_3 , Sm_2O_3 , Nd_2O_3 , Pr_2O_3 , Tb_2O_3 , Dy_2O_3 , Er_2O_3 , Yb_2O_3 , As_2O_3 , Sb_2O_3 , Bi_2O_3 , SiO_2 , GeO_2 , ThO_2 , VO_2 , HfO_2 , SeO_2 , TeO_2 , VO_2 , PbO_2 , TiO_2 , SnO_2 , ZrO_2 , CeO_2 , MnO_2 , P_2O_5 , V_2O_5 , Nb_2O_5 , Ta_2O_5 , MoO_3 , WO_3 , and SO_3 .

In describing methods for the calculation of glass properties, it is customary to specify concentration limits for components, i.e. the concentration ranges, within which it is possible to guarantee acceptable precision. However, when describing the current method which covers almost all known oxide glass forming systems such an approach is hardly acceptable. Oxide systems are too diverse to specify unified oxide concentration limits for them. I believe that the problem of applicability of the proposed method should be solved separately for each composition area. This can be done using MDL SciGlass,⁽⁷¹⁾ it is necessary to select experimental data on the glasses belonging to the composition area of interest from the database, and to compare these data with the results of calculations. If the MDL SciGlass database does not contain the data on the glasses of interest, this probably means that nobody has studied them, and in this case the only reliable way to evaluate an applicability of any method of calculation, including the proposed one, is to carry out new experimental research.

The author is quite aware that at present not all specialists on glass have an access to the SciGlass information system. However, it is necessary to refer to SciGlass since only by using the computer program included within SciGlass one can use the proposed method of the calculation in practice.

To give a general idea of the possibilities of the method, some generalised information on the precision of calculations of the above mentioned properties is given at the end of the paper.

Description of the calculation algorithm

As mentioned above, the algorithm generalises the partial algorithms described in Refs 37–46 where detailed information for them (including examples of calculation) is given. A full description of the algorithm is given in Ref. 37. This algorithm has been included in MDL SciGlass,⁽⁷¹⁾ and thus users of this database can perform the calculations for arbitrary glass compositions and/or compare the results of calculations with the published experimental data for arbitrary concentration ranges.

The most important features of the algorithm are discussed in the following sections.

Classification of properties by the method of calculation

The proposed method is empirical. When developing the method both the direct dependences of properties on composition and temperature and the correlations between different properties of glasses and melts were

³ MDL SciGlass also includes the subroutines for calculation of properties of glasses and glass forming melts from their composition and temperature using more than 100 known methods; the proposed method is among them.

considered. Accordingly, all calculated quantities were classified into two groups, 'basic' and 'derived' ones. The 'basic' properties are calculated directly from composition and temperature. 'Derived' quantities are calculated indirectly, i.e. expressed in terms of 'basic' properties.

The group of 'basic' properties includes quantities that have been studied within the widest composition area. They include viscosity and its temperature coefficient at temperature T_{13} corresponding to the viscosity $\eta=10^{13}$ dPas, thermal expansion coefficient at 160°C, surface tension at 1400°C, as well as density, refractive index, mean dispersion, Young's modulus, and heat capacity (all at 20°C). All other properties are considered as 'derived' ones.

'Basic' properties are conventionally divided into 'structure sensitive' properties (heat capacity of glasses at 20°C and surface tension of melts at 1400°C) and 'structure insensitive' ones (the seven other 'basic' properties). The reason for this classification is that precise calculation of 'structure sensitive' properties requires consideration of structural factors, whereas they may be ignored when calculating 'structure insensitive' properties.

Calculation of 'structure insensitive' properties

Structure insensitive properties are calculated using atomic additivity formulae

$$P = \sum_{i=1}^N g_{i,p} m_i n_i \sum_{i=1}^N m_i n_i \quad (1)$$

where P is the property being calculated; i is the index of the oxide; N is the number of types of oxides forming the glass in question; m_i is a molar fraction of the i -th oxide; n_i is a number of atoms in the formula of the i -th oxide and $g_{i,p}$ is a partial coefficient for the i -th oxide.⁴ When calculating surface tension (σ) the quantity calculated by Equation (1) is σ^{-1} ; this allows a considerable improvement of the precision of calculations at high concentrations of surface active oxides.

Calculation of 'structural sensitive' properties Structure sensitive properties are calculated by considering the concentrations of structural groups in glass. Two types of structural groups are considered.

(1) *Coordination polyhedra* that are generated by network forming (usually polyvalent) cations and oxygen. A polyhedron can be chemically combined (but can also be free) with one or more cations of a low valency, acting as modifiers. The following complexes are examples of coordination polyhedra: $\text{SiO}_{4/2}$, $\text{SiO}_{3/2}\text{ONa}$, $\text{BO}_{3/2}$, $\text{BO}_{4/2}\text{Na}$.

(2) *Associates* formed as a result of joining several polyhedra of various types, e.g. $\text{AlO}_{4/2}\text{Na} \cdot 2\text{SiO}_{4/2}$, $\text{BO}_{4/2}\text{Na} \cdot \text{BO}_{3/2}$.

A simplified mathematical model is used to calculate the concentrations of these structural groups. This model involves a system of empirical equations simi-

lar to the equations used in the theory of chemical equilibria.

Since, to date, such equations have not been used in the empirical methods for calculating glass properties, it seems appropriate to consider a specific example. Let us assume that there are structural groups of three types in a glass, namely, polyhedra A and B, as well as an associate AB formed from them by the reaction



The equilibrium equation for reaction (2) is

$$K = \frac{a_{\text{AB}} m_{\text{AB}}}{a_{\text{A}} m_{\text{A}} a_{\text{B}} m_{\text{B}}} \quad (3)$$

where K is the equilibrium constant, a (with the subscripts corresponding to the participants in a reaction) is an activity coefficient. After adding the equations of the material balance⁵ to Equation (3) one can obtain a system of equations whose solution is a set of concentrations of all structural groups.

We rewrite Equation (3) as

$$\kappa \equiv \frac{K a_{\text{A}} a_{\text{B}}}{a_{\text{AB}}} = \frac{m_{\text{AB}}}{m_{\text{A}} m_{\text{B}}} \quad (4)$$

In the proposed approach Equation (4) is considered as an empirical equation, and the value of κ – as its adjustable parameter. Being empirical the equation, of course, loses its thermodynamical sense and makes no claim to have one. Similar equations are written for all the chemical reactions under consideration. The values of κ for the majority of the reactions⁶ are considered as constants, i.e. independent of composition and temperature.

Replacement of the thermodynamic equations of equilibria by their empirical analogues expressed in the form of Equation (4) at once gives several desired results. Firstly, the number of parameters used to calculate the concentrations of structural groups are reduced to one per reaction. Secondly, the values of parameters are not attached to any specific composition area. Thirdly, these values can be determined both from experimentally measured relationships between glass properties and composition and data on glass structure. Thus, the proposed approach completely meets the above stated requirements in respect of parameters.

The possibility of determining the κ values from the data on glass structure is especially important for describing the chemical reactions accompanied by the change of the coordination number of boron. A fraction of fourfold-coordinated boron is a paramount structural characteristic which has a pronounced effect on almost all properties of glasses containing boron oxide. Judging from the data presented in MDL SciGlass, boron oxide is a constituent of one third of

⁴ In the formula for calculation of glass heat capacity the partial coefficients obtained by Khalimovskaya-Churkina⁽⁴⁴⁾ are used.

⁵ The equations of material balance are not included here to simplify the argument.

⁶ For some reactions with R_2O_3 the value of k is considered as a function of temperature; details are given in Refs 41 and 42.

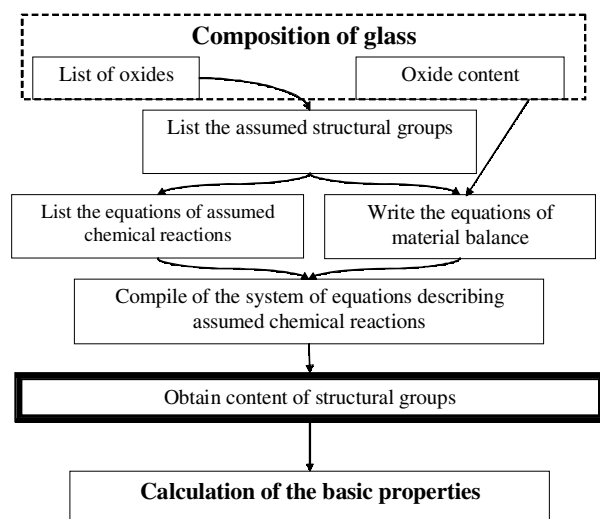


Figure 1. Schematic diagram of the proposed algorithm for calculating 'basic' properties of glasses and melts

all oxide glasses so far studied. That is why correctly accounting for this structural factor in many respects determines the practical value of the method as a whole. Therefore the values of κ for the reactions involving boron oxide were determined from direct data for the fraction of fourfold-coordinated boron, which were obtained by NMR.⁽⁴³⁾

The values of κ for other reactions were determined from the experimental data on the most comprehensively studied 'structural sensitive' glass properties, first of all viscosity and glass transition temperature.^(43,46)

A flow diagram of the calculation of 'structural-sensitive' properties from glass composition and temperature is given in Figure 1. According to this scheme, first, based on the list of oxides present, we compile the list of all structural groups that can be formed from these oxides.^(43,46) Then we compile the list of all the possible chemical reactions involving these given structural groups.⁷ Using these lists we compile a set of equations similar to the equations of chemical equilibria, as well as the equations of material balance. As a result, we obtain a system of equations. Solving the latter we determine the concentrations of all structural groups. Next, based on these results, we calculate the values of 'basic' properties by using the empirical formulae. These formulae are most complicated for viscosity.⁽³⁷⁻⁴⁶⁾ To calculate other properties Equation (1) is used, where structural groups are substituted for the oxides.

Calculation of 'derived' properties The interrelations between basic and derived properties are shown in Figure 2. Detailed specification of the formulas describing these interrelations is beyond the scope of this paper; they are given in detail in Refs 37-46. Three examples demonstrating the expediency of calculating one property via another one are given below.

⁷ For simplicity, in the proposed method only the reactions in which no more than three types of cations take part are considered.

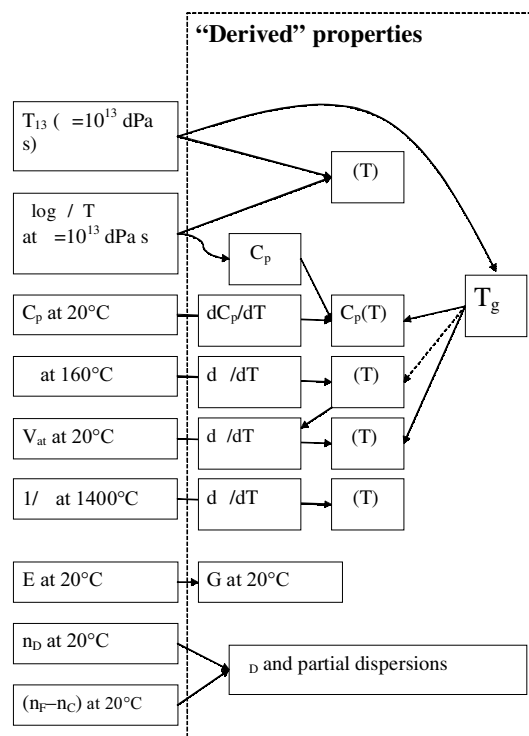


Figure 2. Interrelations between 'basic' and 'derived' properties. Designations: η -viscosity, T_g -glass transition temperature, V_{at} -atomic volume (i.e. volume per 1 g-atom of a given glass), ρ -density, α -thermal expansion coefficient, σ -surface tension, C_p -heat capacity, ΔC_p -jump of heat capacity in the glass transition range, E -Young's modulus, G -shear modulus, H -microhardness, n -refractive index (subscripts denote wavelengths), v -Abbé number.

(a) Refractive index n_D , mean dispersion $n_F - n_C$, and Abbé number v_D are related by the formula

$$v_D = \frac{n_D - 1}{n_F - n_C} \quad (5)$$

It is obvious that, when any two of these three properties have been determined, it is possible to calculate the third one. Nevertheless, in many known methods, including the methods by Appen⁽¹²⁾ and Demkina,⁽¹⁴⁾ the partial coefficients are obtained for all three quantities. Such an approach leads to a factor of 1.5 increase in the number of adjustable parameters. Such an increase could be justified, if it resulted in the improvement of precision; that is possible theoretically, but does not happen in practice. It therefore seems advisable to calculate only two quantities directly from composition and the third one from Equation (5).

(b) Shear modulus G and Young's modulus E are related by the relationship

$$G = E/2(1 + \mu) \quad (6)$$

in which Poisson's ratio μ for almost all known oxide glasses varies in the range from 0.16 to 0.32. Substituting the mean value 0.24 in Equation (6), we obtain

$$G \approx 0.4E \quad (7)$$

Hence, knowing one of the moduli, we can estimate the other one by Equation (7). The root mean square

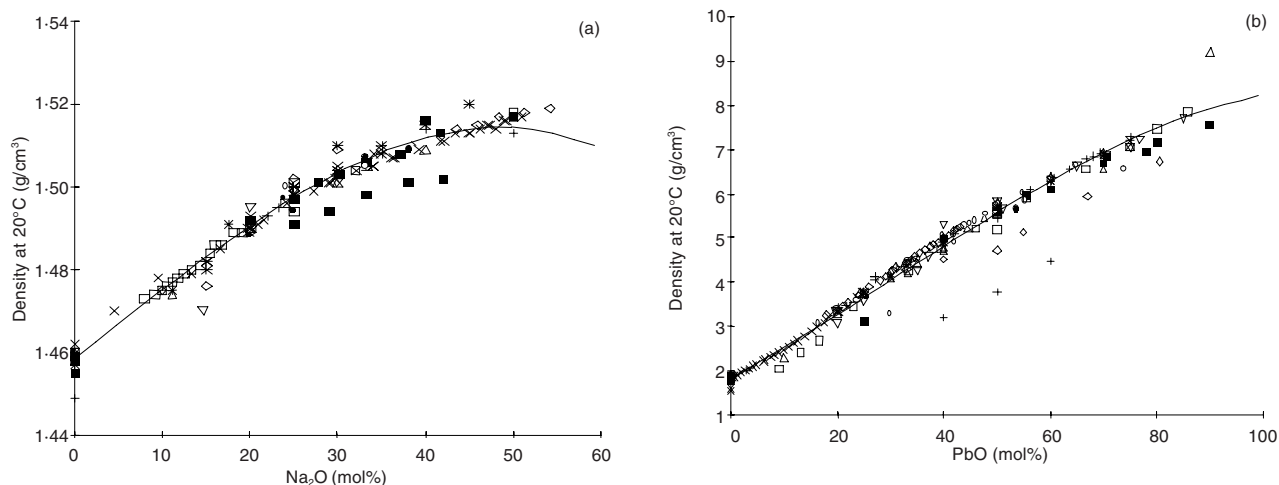


Figure 3. Concentration dependence of glass properties in the systems (a) $\text{Na}_2\text{O}-\text{SiO}_2$ and (b) $\text{PbO}-\text{B}_2\text{O}_3$. The points correspond to the experimental data given in MDL SciGlass (different signs correspond to data by different authors). The lines correspond to the results of calculations using the proposed method. The graphs are obtained directly from the MDL SciGlass program

error of such approximation is only 2.5%.⁸ This is many times less than the error of calculating any of these moduli from glass composition by any *known* method. Thus, the independent calculation of both moduli from composition, accompanied by duplicating the number of adjustable parameters (as in the methods of Appen⁽¹²⁾ and Demkina⁽¹⁴⁾), cannot result in greater precision. Accordingly, it seems advisable to calculate only one value directly from composition, that value being the Young's modulus which has been studied in more detail.

(c) The heat capacity of melts, $C_{p,\text{melt}}$, in contrast to heat capacity of glasses, $C_{p,\text{g}}$, depends strongly on structure. However it is impossible to determine partial coefficients of $C_{p,\text{melt}}$ for the majority of structural groups, because the required experimental data are absent. To surmount this obstacle within the proposed method the heat capacity of melts is calculated using

$$C_{p,\text{melt}} = C_{p,\text{g}}(T_g) + \Delta C_p \quad (8)$$

where $C_{p,\text{g}}(T_g)$ is the heat capacity of glass at glass transition temperature T_g , and ΔC_p is the jump in the heat capacity in the glass transition range, which in fact contains the structure dependent component. The value of ΔC_p is calculated via the temperature coefficient of the log viscosity in the glass transition range.

This algorithm (see details in Ref. 46) does not contain parameters whose determination requires experimental data on heat capacity of melts. Hence the small number of these data is no longer an obstacle to extending the calculation to a wide composition area.

Thus, using the known relationships between different properties of glasses and melts makes it possible to improve precision and generality of the calculations by the proposed method.

Checking the reliability of the initial data used to determine parameters While processing large arrays of data obtained from numerous sources, the problem of

⁸ The figure is obtained after treating the array of experimental data on oxide glasses, for which both moduli were measured (~1800 compositions) after excluding clearly erroneous results, that give values of Poisson's ratio of more than 0.5 which is physically impossible.

data reliability is highlighted. The results represented in Figure 3 show how serious this problem is. Only two examples of concentration dependence of the properties of glasses in binary systems are shown in the plots. It is clear from the plots that the data of different authors for the same property of glasses of the same composition sometimes differ significantly from each other.

There is a range of reasons for the discrepancies in the published data including a change of the chemical composition of glass during melting (volatilisation of some components, dissolution of a crucible material, redox and other reactions); phase separation processes (liquation, crystallisation); failure to correctly carry out property measurements and misprints in published data. However, whatever the cause, the outliers that 'falling out' of the general dependences are blunders, and their use in determining parameters leads to distorted results.

It is obviously impossible to eliminate all blunders from such a large (six figure) data array. Nevertheless it is possible to decrease the number of erroneous data in the array if one can compare them with other data. Such comparison, as a rule, is possible only within the simplest glass forming systems that contain no more than three components. In complex systems it is much more difficult to identify erroneous data, because there are often no data to compare them with.

Based on the above, to improve reliability of calculations I considered it expedient to determine the values of all parameters only from the data on the glasses and melts belonging to binary and ternary systems. Digital and visual comparison of these data with each other by using MDL SciGlass made it possible to select the most reliable data and eliminate the most doubtful ones.

The data relating to the glasses and melts with more complex compositions were used only to test the method. This enabled checking of the 'predictive power' of the approach, i.e. the possibility of using the approach outside the limits of the composition area within which parameter values had been determined. Taking into account the generality of the proposed method, such testing is of great importance.

Table 1. Root mean square absolute (ΔP_{avg}) and relative (δP_{avg}) deviations of calculated property values from the experimental data given in MDL SciGlass

Property	Multiplier	T (°C)	Number of compositions	ΔP_{avg}	δP_{avg} (%)
Viscosity characteristics K	-	T_1	2383	107	-
		T_{7-6}	3384	45	-
		T_{13}	1672	32	-
		T_g	16679	43	-
		Total**	51677	-	-
Density of glasses, g/cm ³	-	20	32178	0.138	4.0
Density of melts, g/cm ³	-	800	456	0.133	4.2
		1400	534	0.106	4.3
		Total**	2335	0.131	4.2
		55±5	1556	10.2	14.8*
Thermal expansion coefficient of glasses, K ⁻¹	10 ⁻⁷	160±10	6595	8.4	12.0*
		210±10	4579	8.6	11.7*
		Total**	27424	11.3	14.3*
		900	659	26	16.9
Surface tension of melts, mN/m	-	1300	1128	23	10.4
		Total**	3038	28.5	13.0
		20	310	103	16.2
		1000	151	168	13.3
Heat capacity of glasses and melts, J/kg K	-	1200	121	111	8.6
		Total**	1438	114	11.4
		T_g	136	2.3	22
		20	26122	0.017	1.0
ΔC_p , J/kg K	-	T_g	136	2.3	22
Refractive index (n_D)	-	20	9124	12.9	7.1
Mean dispersion (n_F-n_C)	10 ⁻⁴	20	9189	2.4	6.1
Abbé number (n_D)	-	20	3770	7.5	12.9
Young's modulus, GPa	-	20	1971	2.5	12.2
Shear modulus, GPa	-	20	1689	0.04	18
Poisson's ratio	-	20			

* For $\alpha \geq 30 \times 10^{-7} \text{ K}^{-1}$.

** Rows 'total' include other temperatures that are not presented in the table. In the column 'Number of compositions' glasses studied at several temperatures are accounted for only once.

Discussion

Priven & Mazurin⁽⁵⁰⁾ compared some glass property values (namely, density, thermal expansion coefficient, and refractive index) calculated by the proposed method and other known methods with experimental data presented in SciGlass. They showed that the proposed method is superior to the known methods both in precision and generality for these properties.

Tables 1–5 give statistical data for all properties being calculated. Absolute (ΔP) and relative (δP) root mean square deviations of the calculated values of property P from the experimental data presented in SciGlass are used as numeric characteristics of the calculation error. In fact, these deviations are the sum of calculation and experimental errors.

It was stated above that there are some blunders among the published experimental data on glass properties. To reduce their influence on root mean square deviations, the following procedure was used (its advantages and disadvantages are discussed in Ref. 50). The blunders were excluded by Student's criterion, t

$$|P_{exp} - P_{calc}| < \Delta P_{avg} t(p, n) \quad (9)$$

where p is the level of significance (taken as equal to 0.05), n is the number of the evaluated compositions, ΔP_{avg} is root mean square deviation of the calculated (P_{calc}) from the experimental (P_{exp}) values of the property P . Tables 1–5 list the data obtained after blunders were excluded.

General evaluation of the calculation error

In Table 1 the root mean square deviations of the calculated properties from the experimental data are given. Experimental data on the properties of glasses and

melts containing the 64 above stated oxides in all combinations and ratios were given as the initial data. In other words, the comparison was performed in almost all investigated composition area of oxide glasses. The total number of glass and melt compositions being compared exceeded 100000.

At first sight the root mean square deviations given in the table seem to be rather high. However, when evaluating the data presented in Table 1, one should remember that even after excluding the blunders the root mean square deviations remain a sum of experimental and calculation errors, i.e. they include an experimental error. The latter includes not only the error involved in measuring a property, but also the errors in the published concentrations of oxides, caused by changes in a chemical composition of glasses on melting. Thus, the real error of calculation makes up only a part of the values presented in the table. Some estimations (for example, processing of the array of data obtained with chemically analysed glasses⁽⁵⁰⁾) indicates that this accounts for 30–50% of the error.

Taking this into account, I consider the root mean square deviations presented in Table 1 as quite acceptable for solving practical problems over a wide area.

Comparison of the proposed method with the method by Appen

It is well known, that Appen's method⁽¹²⁾ is among the best for calculating properties of silicate glasses; Ref. 50 cogently confirms this thesis.

In his monograph Appen⁽¹²⁾ gives the values of calculation errors that are several times less than the values presented in Table 1. However a direct comparison of these values with our data is not quite correct, since the method of Appen covers a significantly narrower

Table 2. Comparison of the proposed method with Appen's method within the limits of application of Appen's method using the data presented in MDL SciGlass

Property	Number of compositions	ΔP_{avg} Proposed method	Method by Appen
Density (ρ , g/cm ³) at 20°C	6879	0.043	0.038
Thermal expansion coefficient ($\alpha \times 10^7$, K ⁻¹) at 210°C	1583	6.49	6.44
Refractive index (n_D) at 20°C	6063	0.0076	0.0068
Mean dispersion ($n_F - n_C$) $\times 10^4$	1571	4.3	5.2
Abbé number (n_D) at 20°C	1579	1.5	3.0
Young's modulus (E , GPa) at 20°C	1155	4.3	6.4
Shear modulus (G , GPa) at 20°C	452	1.8	3.0
Surface tension (σ , mN/m) at 1300°C	253	17	13

ΔP_{avg} and δP_{avg} mean absolute and relative root mean square deviations

compositional area than the method proposed. In this connection it was advisable to perform a direct comparison between the proposed method and Appen's method using the same experimental data. To do this all data on glasses with compositions lying within the concentration limits specified by Appen were selected from SciGlass. The property values of these glasses were then calculated by both methods.

The blunders in the initial data were revealed by the procedure described above. In the process, for the purity of the comparison, only the data that were qualified as blunders with respect to both methods were excluded from processing. Thus, the conditions for comparison were absolutely the same for both methods.

The results of the comparison are given in Table 2. It is clear from the table, that the proposed method as a whole is no less precise than Appen's method within the limits of applicability of the latter method.

Note that the proposed method proved to be about twice as precise as Appen's method when calculating

Table 3. Comparison of root mean square deviations of the calculated property values from the experimental data for glasses with and without SiO₂

Property or temperature	Glasses with SiO ₂			Glasses without SiO ₂		
	ΔP_{avg}	δP_{avg} (%)	N	ΔP_{avg}	δP_{avg} (%)	N
Viscosity characteristics, K						
T_1 ($\eta=10^1$ dPas)	111	-	2125	102	-	279
T_4 ($\eta=10^4$ dPas)	65	-	3068	31	-	232
T_7 ($\eta=10^7$ dPas)	44	-	1688	39	-	283
T_{10} ($\eta=10^{10}$ dPas)	32	-	1875	36	-	800
T_{13} ($\eta=10^{13}$ dPas)	31	-	1058	35	-	618
T_g	45	-	8784	42	-	7943
Density, g/cm ³						
293 K	0.091	3	18262	0.193	5.2	13689
1673 K	0.096	3.2	475	0.192	7.8	55
Thermal expansion coefficient, $\alpha \times 10^7$, K ⁻¹						
433±10 K	7	10.9	5041	13.1	16.5	1585
Optical properties						
n_D	0.013	0.8	17309	0.024	1.4	8784
$(n_F - n_C) \times 10^4$	8.8	5.3	5327	20.5	9.4	3789
v_D	1.8	4.4	5409	3.3	8.3	3833
Elastic moduli, GPa						
E	6.7	8.8	2711	9.6	23.1	1073
G	2	7.2	1212	3.6	19.9	774
Surface tension, mN/m						
1573 K	24	8.8	878	20	14.6	251
Heat capacity, J/kg K						
293 K	149	25.5	213	162	13.8	114

ΔP_{avg} and δP_{avg} mean absolute and relative root mean square deviations, N is the number of compositions

Table 4. Comparison of root mean square deviations of calculated property values from experimental data for glasses with and without B₂O₃

Property or temperature	Glasses without B ₂ O ₃			Glasses with B ₂ O ₃		
	ΔP_{avg}	δP_{avg} (%)	N	ΔP_{avg}	δP_{avg} (%)	N
Viscosity characteristics, K						
T_1 ($\eta=10^1$ dPas)	110	-	2103	120	-	305
T_4 ($\eta=10^4$ dPas)	63	-	2024	61	-	1265
T_7 ($\eta=10^7$ dPas)	46	-	1227	38	-	734
T_{10} ($\eta=10^{10}$ dPas)	37	-	1569	29	-	1113
T_{13} ($\eta=10^{13}$ dPas)	33	-	1032	28	-	632
T_g	44	-	10081	42	-	6650
Density, g/cm ³						
293 K	0.14	3.9	20138	0.13	4.1	11983
1273 K	0.17	4.7	380	0.1	4.1	348
Thermal expansion coefficient, $\alpha \times 10^7$, K ⁻¹						
328±5 K	11	14.9	786	9.2	14.4	769
Optical properties						
n_D	0.017	1	15196	0.017	1	10916
$(n_F - n_C) \times 10^4$	19	10.3	3401	10.4	6.6	5701
v_D	2.3	6.9	3396	2.4	5.6	5788
Elastic moduli, GPa						
E	7.6	13.4	2645	7.1	11.8	1125
G	2.2	10.2	1420	3	15	530
Surface tension, mN/m						
1473 K	22	10.4	367	20	12.8	409
Surface tension, mN/m						
293 K	120	20.2	218	129	22	99
1273 K	169	12.8	127	182	14.9	27

shear modulus and Abbé number. The peculiarity of these properties is that they are calculated directly from composition by Appen's method, and indirectly (via other properties) by the proposed method. We can see that the indirect method of calculation of shear modulus and Abbé number made it possible to calculate these properties with significantly greater precision than the direct method.

The precision of property calculations in different composition areas

Table 3 compares the precision of glass property calculations for silicate and non-silicate systems. One can see from the table that in silicate systems the root mean square deviations for the majority of properties are about half those in the systems which do not contain SiO₂ as a component.⁹ This can be easily explained. The properties of silicate glasses and melts have long been investigated, and a significantly greater amount of information about them has been accumulated than for non-silicate glasses and melts. In addition, the relationships between compositions and properties of glasses and melts in silicate systems are simpler than those in non-silicate systems. Therefore the influence of composition on the properties of glasses and melts in silicate systems has been more precisely evaluated than in non-silicate systems.

It also seemed advisable to estimate how correctly the proposed method accounts for the influence of the assumed changes of glass structure on properties. Such a procedure is most relevant with respect to glasses

⁹ Temperatures corresponding to fixed viscosity values are an exception for this rule. However, this is a formal exception and it is determined only by the fact that non-silicate glasses are, as a rule, 'shorter' than silicate glasses. Accordingly, smaller values of calculation errors for the stated temperatures for non-silicate glasses do not signify that their viscosity is calculated more precisely.

Table 5. Comparison of root mean square deviations of the calculated property values from the experimental data for glasses containing different number of components

Property or temperature	Number of components in glasses		More than 3 ΔP_{avg}	Number of compositions	Up to 3 ΔP_{avg}	Number of compositions
	Any ΔP_{avg}	Number of compositions				
Viscosity characteristics, K						
T_1 ($\eta=10^1$ dPas)	110	2405	104	1261	117	1133
T_4 ($\eta=10^4$ dPas)	62	3297	67	2354	50	947
T_7 ($\eta=10^7$ dPas)	44	1974	46	1151	41	820
T_{10} ($\eta=10^{10}$ dPas)	33	2678	35	1079	32	1599
T_{13} ($\eta=10^{13}$ dPas)	32	1675	34	510	31	1167
T_g	43	16733	52.4	5997	38.4	10764
Density, g/cm ³						
293	0.14	32178	0.12	11091	0.15	20996
1673	0.11	534	0.11	207	0.10	328
Thermal expansion coefficient, $\alpha \times 10^7$, K ⁻¹						
433±10	8.4	6595	7.5	4396	10.1	2190
Optical properties						
n_D	0.017	26122	0.017	13201	0.017	12907
$(n_F - n_D) \times 10^4$	12.9	9124	12.2	5950	15.0	3169
v_D	2.4	9189	2.4	5966	2.3	3222
Elastic moduli, GPa						
E	7.5	3770	8.0	1891	7.0	1884
G	2.5	1971	2.3	604	2.7	1369
Heat capacity, J/kg K						
293	112	314	106	85	123	231
1273	174	155	144	44	176	108
Surface tension, mN/m						
1573	23.2	1128	30.9	231	21.0	895

ΔP_{avg} means absolute root mean square deviation

containing boron oxide. In Table 4 the data on boron containing glasses are compared with the data on boron free glasses. It can be seen that the errors in the property calculations are close for both groups. In other words, the more complex calculations needed for glasses containing boron oxide do not result in a larger error. This indicates that the proposed method in general accounts correctly for the influence of boron oxide on the properties of glasses and melts over a wide composition area.

The precision of property calculations in systems containing different number of components

Table 5 compares the precision of property calculations for multicomponent and simple (one-component, binary, and ternary) glasses. As was stated above, only the data related to simple glasses were used to determine the parameter values used in the calculation. Thus, this test might to a certain extent answer the question about a possibility of the proposed method to predict properties of glasses and melts outside the glass forming systems that were involved in determining the parameters of the method.

The data presented in the table indicates that the precision of property calculations in both groups is almost the same. Consequently, the parameters used in the calculations are equally applicable to both simple and complex systems. This can be considered as a confirmation of generality of the proposed method, i.e. the possibility of its application in a wide composition area.

Conclusion

A new method to calculate the properties of oxide glasses and glass forming melts is proposed. The method enables calculation of viscosity, density, thermal expansion coefficient, surface tension, heat ca-

capacity, as well as some optical, elastic, and acoustical characteristics. The method can be applied in almost all composition and temperature ranges so far investigated.

Mathematical apparatus borrowed from the theory of chemical equilibria is used in the proposed method. The thermodynamic characteristics appearing in this theory (namely, constants of equilibria and activity coefficients) are replaced by empirical coefficients. The values of these coefficients are determined from the known data on glass structure and properties.

Substitution of empirical parameters for thermodynamic characteristics takes the calculation away from thermodynamical rigour but it allows extension of the calculations over an extremely wide composition area due to a many fold decrease in the number of the empirical parameters describing the chemical interactions between oxides.

Testing the method with the array of property data for more than 100000 glasses and melts available in the MDL SciGlass information system⁽⁷¹⁾ has shown that the proposed method is not only much more general than the other known methods but also compares well with their precision. The proposed method may be therefore considered as a single alternative to the other methods of calculating properties of oxide glasses and melts from their composition and temperature.

This does not mean that the known methods have completely lost their significance. As is shown in Ref. 50, in certain composition areas some of these methods allow prediction of glass properties more precisely than the proposed method. However, it seems more significant that the proposed method, being, as a whole, comparable in precision with the best of the other methods, covers a much wider composition area and enables calculation of many properties at a time. This

opens new possibilities of searching for promising composition areas of glasses with predefined, including unique, combinations of properties.

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