

INFLUENȚA GRADULUI DE COMPACTARE STRUCTURALĂ LA SCARĂ SUBNANOMETRICĂ ASUPRA UNOR PROPRIETĂȚI ALE STICLELOR DIN SISTEMUL OXIDIC $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{B}_2\text{O}_3$

THE INFLUENCE OF THE STRUCTURAL COMPACTNESS AT A SUB-NANOMETRIC SCALE ON SOME GLASS PROPERTIES OF THE $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{B}_2\text{O}_3$ SYSTEM

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In the $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{B}_2\text{O}_3$ system, six glasses were synthesized having the $(35-x)\text{Na}_2\text{O}\cdot x\text{Al}_2\text{O}_3\cdot 65\text{B}_2\text{O}_3$ molar composition, where $x = 0; 5; 10; 12.5; 15; 17.5$. For these synthesized glasses the following physical properties were measured: density; refractive index; linear thermal expansion coefficient; glass transition temperature. The structural compactness, the molar volume and the volume of oxygen ion, considered as structural characteristics for the first level of analysis (atomic), at a sub-nanometric scale, were also calculated.

The correlation analysis of the properties indicated that these structural characteristics, even at this atomic level, sub-nanometric, influence the physical properties considered at a macroscopic scale.

Moreover, the dependency graphs show a series of particular points (return points) that suggest structural transformation areas generated by the $[\text{BO}_3] \rightarrow [\text{BO}_4]$ and $[\text{AlO}_6] \rightarrow [\text{AlO}_4]$ transformations, according to the chemical composition of the glasses.

În sistemul oxidic $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{B}_2\text{O}_3$ s-au sintetizat 6 sticle, având formula chimică molară $(35-x)\text{Na}_2\text{O}\cdot x\text{Al}_2\text{O}_3\cdot 65\text{B}_2\text{O}_3$ ($x = 0; 5; 10; 12.5; 15; 17.5$). Pentru aceste sticle s-au măsurat următoarele proprietăți fizice: densitatea, indicele de refracție, coeficientul de dilatare termică liniară, temperatura tranziției vitroase. Totodată, s-au calculat mărimile: gradul de compactare structurală, volumul molar și volumul ionului de oxigen, considerate caracteristici structurale la un prim nivel de analiză (atomic), la scară subnanometrică.

Analiza de corelație a mărimilor a indicat că aceste caracteristici structurale, chiar la acest nivel atomic, subnanometric, influențează proprietățile fizice considerate, la scară macroscopică.

În plus, graficele de dependență prezintă o serie de puncte particulare (de întoarcere) care sugerează zone de transformări structurale generate de transformările $[\text{BO}_3] \rightarrow [\text{BO}_4]$ și $[\text{AlO}_6] \rightarrow [\text{AlO}_4]$, funcție de compoziția chimică a sticlelor.

Keywords: $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{B}_2\text{O}_3$ glasses, structural compactness, properties, correlations, sub-nanometric scale

1. Introduction

Regarding the vitreous systems, a problem of great interest is to establish theoretical computational relationships for the properties of glasses. Unfortunately, at the present level of our knowledge, such relationships can not be obtained by calling only fundamental laws, but, at most, in the case of very simple oxide systems.

The only successful approach to correlating properties with some characteristics of vitreous systems is provided by the mathematical statistics methods. A first such attempt is reported in the literature by Winkelmann and Schott (1890), cited in [1], and consists in various computational relationships for the properties according to the oxide composition of glasses. At the time, a multitude of computation formulas of properties have been obtained depending on the oxide composition or the basicity of the various vitreous systems of interest [1-4].

After 1960, the accumulation of

experimental data, as well as fundamental knowledge, highlighted the determinant role of the structure for evaluating the properties of glasses. Thus, the analysis of vitreous systems is increasingly done on the basis of implications:

oxide composition $\xrightarrow{\text{processing conditions}}$ vitreous structure \rightarrow properties of vitreous oxide system.

The structure is described as a set of structural entities with stronger interactions than those with the environment. Thus, from the property versus oxide composition approach, the property versus structural entity approach has been taken. For glasses of the $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{B}_2\text{O}_3$ system, the authors presented a series of applications such as the properties versus oxide composition [5] and properties versus structural entities such as NaO_6 , AlO_4 , AlO_6 , BO_3 and BO_4 [6,7].

At the same time, structural entities for any oxide system are defined in relation to "structural levels of approach" [8]. In the paper, for glasses

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with the molar chemical formula $(35-x)\text{Na}_2\text{O} \cdot x\text{Al}_2\text{O}_3 \cdot 65\text{B}_2\text{O}_3$, it is shown the dependency of some properties (such as the density, d , the refractive index, n , the linear thermal expansion coefficient, α , glass transition temperature, T_g) with structural entities, such as structural compactness, C , the molar volume, V_m and the molar volume of oxygen ion, $V_{\text{O}^{2-}}$.

2. Experimental

In the $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{B}_2\text{O}_3$ system, there were chosen six chemical compositions, noted from G0 to G5, shown in Figure 1.

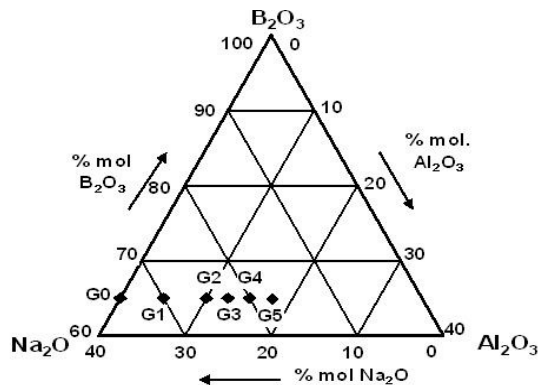


Fig. 1 - Placement in the $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{B}_2\text{O}_3$ ternary diagram of the molar oxide compositions of synthesized glasses / Plasarea în diagrama ternară $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{B}_2\text{O}_3$ a compozițiilor oxidice molare ale sticlelor sintetizate.

The preparation of the raw materials mixture, the melting conditions and the obtaining of the glasses are described by the authors in [5,6]. Measurements for the properties of interest are also presented. The values of the obtained experimental data were analyzed by comparison with other literature data, which are not numerous for the studied system [5,6]. Finally, a good consistency was obtained, which has validated the experimental data obtained.

3. Theoretical background

3.1. The hierarchy of the structure of the oxide systems

In a systemic approach, the vitreous systems can be structurally characterized at several levels. This approach envisages a certain degree of complexity, common to structural entities and which influences a number of properties of the vitreous system taken into consideration.

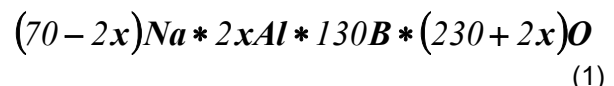
For vitreous oxide systems, the main structural levels - the types of structural entities and their typical structural characteristics are [8]:

- First structural level:
 - *Types of structural entities*: atoms (cations, anions) corresponding to the

chemical composition for the vitreous system analyzed;

- Associated structural characteristics (considered): electronic structure of constituent atoms, the ionization potential of atoms, the electronegativity, the atomic radius, (ionic, covalent), the polarizability, the structural compactness;
- Second structural level:
 - *Types of structural entities*: elementary coordination polyhedra;
 - *Structural characteristics*: hybridization type for the cation-oxygen chemical orbitals (X-O), coordination number CN (X/O), coordination polyhedron type, internuclear distances, X-O, angles between bonding orbitals (O-X-O; X-O-X), the effective charge of atoms that form chemical bonds, bond energies, ionicity of chemical bonds, oxide basicity, etc.;
- The third structural level:
 - *Types of structural entities*: "cluster" type of structural entities, with different dimensions;
 - *Structural characteristics*: the degree of polymerization associated with the macroanions formed by interconnecting the unitary coordinating polyhedra; the geometry, dimensions and frequency of the clusters; internuclear distances, angles between different bonding orbitals, type and intensity of interactions within and between macroanionic formations;
- The fourth structural level:
 - *Types of structural entities*: separate micro-phases (in certain vitreous oxide systems, depending also on the obtaining conditions);
 - *Structural characteristics*: chemical composition, geometry and dimensions of the separated micro-phases; the density at the surface of the glasses;

In the concrete situation of the studied glasses in this paper, their chemical composition can be described at atomic level in the form [6]:



On this basis, component structural entities are established at the first level of approach:

- sodium cations: $\frac{(70 - 2x) \cdot N_A}{430 + 2x}$
- aluminum cations:

$$\frac{2x \cdot N_A}{430 + 2x}$$

- boron cations: $\frac{(230 + 2x) \cdot N_A}{430 + 2x}$

- oxygen anions: $\frac{130 \cdot N_A}{430 + 2x}$

where N_A is Avogadro's number.

Note: In a first evaluation it was considered that all structural elements are cations (for Na, Al and B) and oxygen anions, respectively.

It should be noted that the analysis of the structure at this level is performed on a sub-nanometric scale. This is also characteristic of the second structural level. Mostly, third-level structural entities are placed on a nanometric scale. At the fourth level, structural entities may have varied dimensions ranging from a nanometric scale to a micrometric scale.

At the second structural level, the present elementary structural entities are: NaO₆, AlO₄, AlO₆, BO₃ and BO₄; Their frequency of occurrence is determined by the effective oxide composition of the glass [2,6,8].

At the third structural level, the presence of Na_uAl_vB_w entities, which so far could not be specifically defined from a chemical point of view (u, v, w - natural numbers)

A fourth level is not necessary for describing the structure of the studied glasses, because in the compositional studied field the phenomenon of separation of the vitreous microphases was not reported in the literature and was not manifested after the synthesis of the glasses in the laboratory.

3.2. Evaluation of the structural compactness

For any atomic (molecular) system, the compactness represents the fraction of the molar volume, V_m , occupied by the constituent atoms (molecules). For the compactness calculation, atoms are considered to be rigid, non-deformable

$$C = \frac{4\pi \cdot N_A \cdot \sum_{i=1}^N f_i \cdot r^3(X_i)}{3V_m} = \frac{4\pi \cdot N_A \cdot \sum_{i=1}^N f_i \cdot r^3(X_i) \cdot d}{3 \sum_{i=1}^N f_i \cdot M(X_i)} \quad (2)$$

$$C(x) = \frac{4\pi N_A}{3V_m} \cdot (f_{Na} \cdot r^3(Na) + f_{Al} \cdot r^3(Al) + f_B \cdot r^3(B) + f_O \cdot r^3(O)) \cdot 10^{-27} \quad (3)$$

$$C(x) = \frac{4\pi \cdot N_A}{3V_m} \cdot \frac{(70 - 2x) \cdot 0.098^3 + 2x \cdot 0.057^3 + 130 \cdot 0.025^3 + (230 + 2x) \cdot 0.14^3}{430 + 2x} \cdot 10^{-27} \quad (4)$$

$$C(x) = \frac{1756 + 10x}{(430 + 2x) \cdot V_m} = \frac{(1756 + 10x) \cdot d(x)}{(430 + 2x) \cdot (66.95 + 0.4x)} \quad (5)$$

spheres between which there are no interactions of any kind.

According to the definition [9], the compactness, C , of an atomic system with N components, X_i , $i = \overline{1, N}$, having the radius $r(X_i)$ and the atomic fractions f_i , is calculated with the relation 2, where $M(X_i)$ is the atomic (molecular) mass of the atom (molecule) i ; d - the density of the analyzed system; N_A - Avogadro's number.

However, in the case of real oxide systems, one can not ignore the fact that strong (chemical) interactions occur between atoms, which, in correlation with other factors, determine a certain structure of the system. Unfortunately, at the present stage of knowledge these values can only be established for very simple oxide systems.

4. Results and discussion

In a classical approach, when calculating the compactness of the vitreous oxide structure, the component atoms are considered as cations and anions (the "ionic" structural model).

The atomic chemical formula is used to calculate the structural compactness of the studied glasses, $C(x)$, when the relationship (2) is written as relationship 3.

Taking into account the values of the ionic radius presented in Table 1, $C(x)$ is calculated with the relation 4, where $r(x)$ is the ionic radius of the ion i , in nm; f_i - the atomic fraction of the atom i .

Finally, the expression is 5.

Table 1

Characteristics of interest for component atoms [9,10]
Caracteristicile de interes pentru atomii componenti

Element	Ion	Ionic radius, r_i , [nm]	Covalent radius, r_c , [nm]	Coordination number, CN (X/O)	Internuclear distance, $d(X-O)$, [nm]	Basicity, pB [%]	Type of chemical bonding, X-O
B	B ³⁺	0.025	0.081	3, [BO ₃] 4, [BO ₄]	0.136 0.144	44.8 54.3	covalent
Al	Al ³⁺	0.057	0.126	4, [AlO ₄] 6, [AlO ₆]	0.176 0.189	61.6 72.4	ionic
Na	Na ⁺	0.098	---	6, [NaO ₆]	0.230	89.0	ionic
O	O ²⁻	0.140	0.060	---	---	---	---

However, analyzing the data from Table 1 it results that the hypothesis of the ionicity of the chemical bonds in which the oxygen is present is largely verified for Na, partly for Al and to a less extent for B. In this case, if the values for the covalent radius of the boron and oxygen of the B-O bonds are considered, it results that the compactness calculated by the integral ionic model is greater than that obtained when the covalent bond B-O is considered to be fully covalent.

In fact, there are relatively few cases where X-O bonds are fully ionic or fully covalent. In a quantum chemistry approach, considering a chemical bond between the atom X and oxygen and their wave functions Ψ_X and Ψ_O involved in the formation of the chemical bond, characterized by hybridized orbitals sp^x and sp^y , respectively, (normalized and orthogonal orbitals), it can be written [11]:

$$\Psi_X = \frac{s + \sqrt{x} \cdot p}{\sqrt{1+x}} \quad (6)$$

$$\Psi_O = \frac{s + \sqrt{y} \cdot p}{\sqrt{1+y}} \quad (7)$$

where x and y are the hybridization parameters.

On this basis, it is possible to establish the relations for calculating the effective radius r_X and r_Y with which the two atoms participate in the bond, and which, together, give the internuclear distance [6].

$$r_X = \frac{r_X^s + x \cdot r_X^p}{1+x} \quad (8)$$

$$r_O = \frac{r_O^s + y \cdot r_O^p}{1+y} \quad (9)$$

in which r_i^s and r_i^p they represent the orbital radius s and p , respectively, for the atom i .

In relations (8) and (9) the radius of the ion without electron on the layer p was considered and r_i^p represents the covalent radius of the atom

i . The radius thus calculated are compared to other values presented in the paper [12], demonstrating a very good concordance.

This evaluation of the actual radius of the atoms involved in the chemical bond has the main advantage that no longer requires the evaluation of the ionic / covalent character and the degree of multiplication for the chemical bonds.

Although applicable to single and binary oxide glasses, the methodology can not generally be used for the complex oxide systems, due to difficulties in establishing the hybridization parameters. These can be determined experimentally using X-ray diffraction and taking into account the correlation of the hybridization parameters y with the X-O-X angles; at the same time, a theoretical evaluation method is the molecular dynamics.

Consequently, in the paper the compactness of the glasses was calculated with the relation (5), according to the ionic integral model.

For the synthesized glasses, the variation of the measured properties (d , n , α , T_g) is presented graphically according to the compactness, (Figures 2÷5) and according to the molar volume V_m (Figures 6÷9).

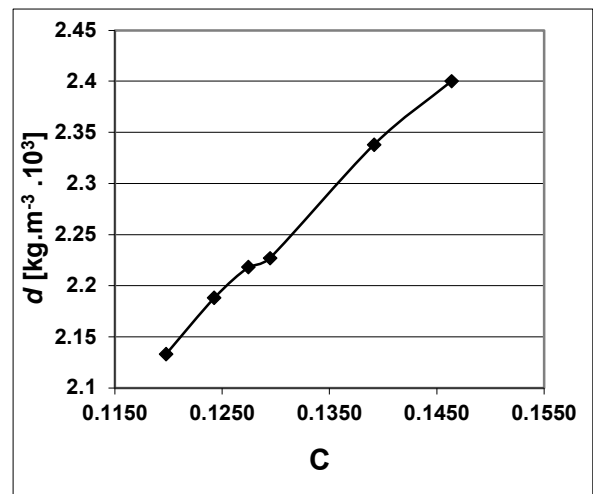


Fig. 2 - Density versus compactness / Densitatea versus compactitatea.

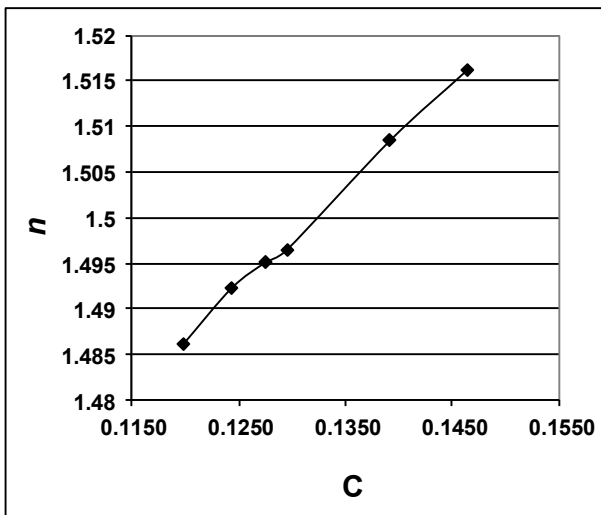


Fig. 3 - Refractive index versus compactness / *Indicele de refracție versus compactitatea.*

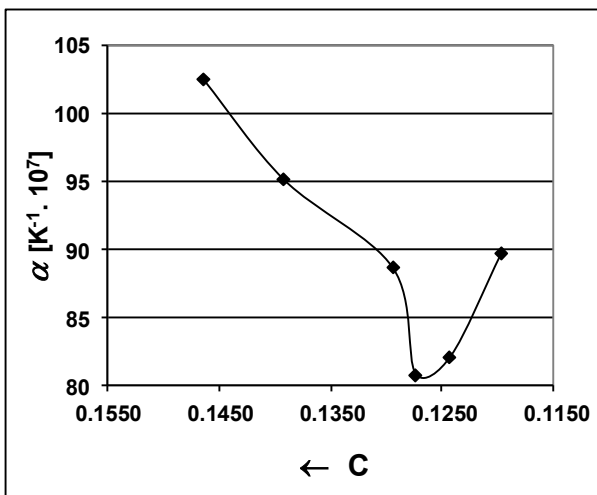


Fig. 4 - Linear thermal expansion coefficient versus compactness / *Coefficientul de dilatare termică liniară versus compactitatea.*

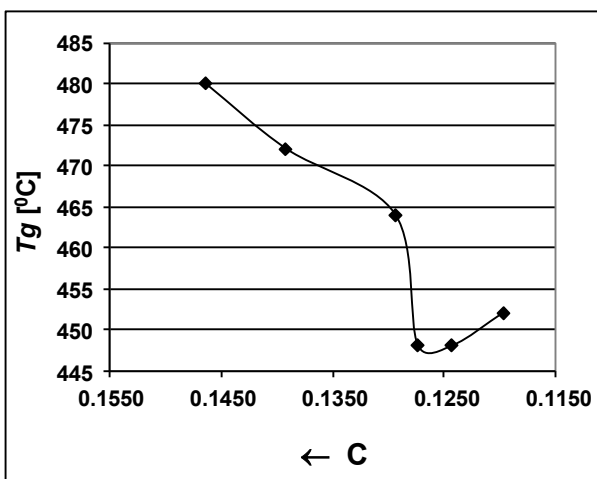


Fig. 5. - Glass transition temperature versus compactness / *Temperatura tranziției vitroase versus compactitatea.*

In an indirect way, the structural compactness (or the degree of structural packing)

can be evaluated with the molar volume, V_m , and the volume of oxygen ion, $V_{O^{2-}}$, computable with the relations:

$$V_m = \frac{\overline{M}}{d} = \frac{\sum_i x_i \cdot M_i}{d} \quad (10)$$

$$V_{O^{2-}} = \frac{V_m}{\sum_i x_i \cdot N_i} \quad (11)$$

in which \overline{M} is the average molar mass of the glass, in kg / mol; d – density of glass, in kg / m³; x_i – the molar fraction of oxide i ; M_i – the molecular weight of oxide i , in kg / mol; N_i – the number of oxygen atoms in oxide i .

Figures 6-9 show the graphical dependencies between d , n , α and, T_g , respectively, versus V_m .

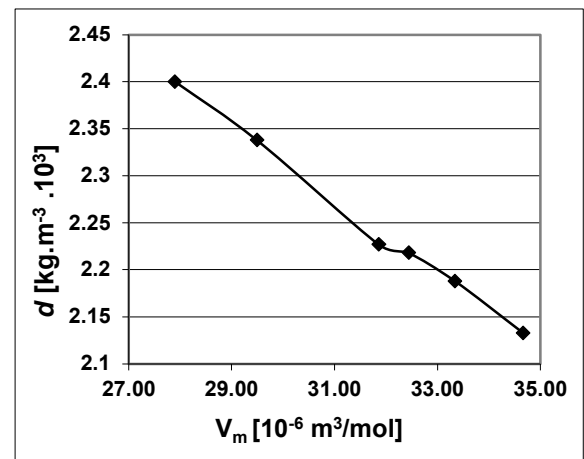


Fig. 6 - Density versus molar volume / *Densitatea versus volumul molar.*

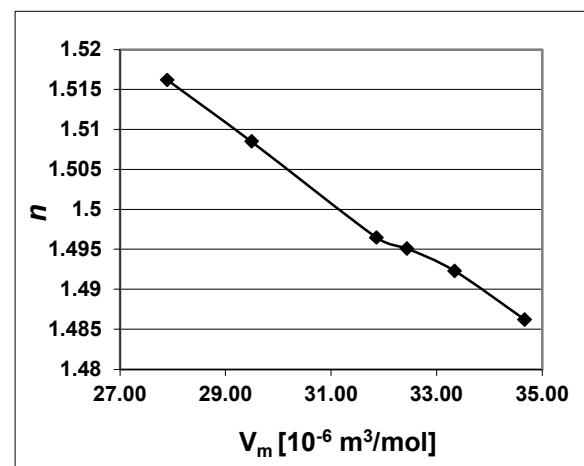


Fig. 7 - Refractive index versus molar volume / *Indicele de refracție versus volumul molar.*

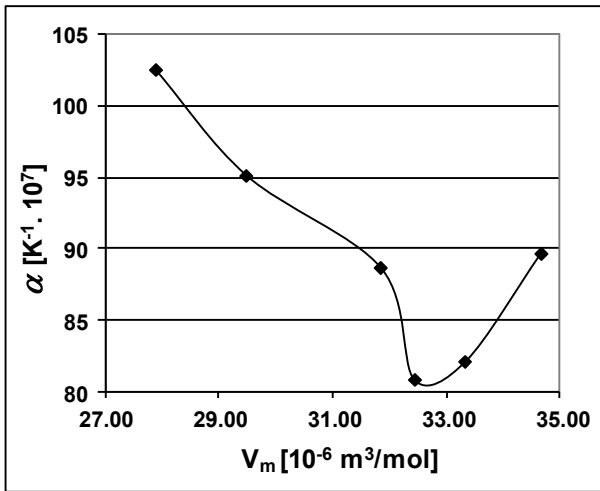


Fig. 8 - Linear thermal expansion coefficient versus molar volume / Coeficientul de dilatare termică versus volumul molar.

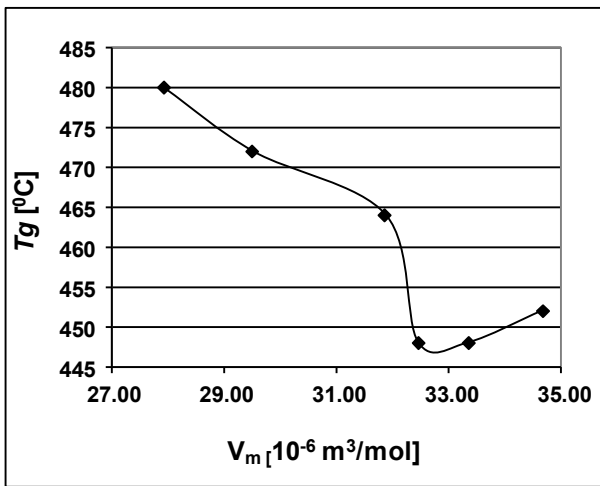


Fig. 9 - Glass transition temperature versus molar volume / Temperatura tranziției vitroase versus volumul molar.

To be remarked for several vitreous oxide systems, Makishima & Mackenzie [13] deduced relationships by which a series of mechanical properties (modulus of elasticity, E , compression modulus, K and Poisson's ratio, μ) are explicitly dependent of the structural compactness.

From the data presented in Figure 2 the density is calculated according to C –dependence which can be well approximated by a linear equation with $R^2 = 0.9965$ and $SD = 0.0067$:

$$d = 10.016 \cdot C + 0.9376 \quad (12)$$

The dependency of the density, d , with the compactness, C – see Figure 2 - was predictable if the relation (5) is taken into account, which for x in the range $0 \div 17.5$ can be written in a simplified form:

$$d = 17.8C \pm 4\% \quad (13)$$

Analogously, the variance of the refractive index, n , in relation to the compactness C can be represented by a linear function with $R^2 = 0.9976$ and $SD = 0.00061$:

$$n = 1.12 \cdot C + 1.3523 \quad (14)$$

The similar dependencies of d and n versus compactness were expected because they are part of the same class of properties and there is a linear dependence between them. Thus, Told quoted in the paper [1] verified this statement for 200 optical oxide glasses, achieving a deviation of no more than $\pm 2\%$.

At the same time, the linear thermal expansion coefficient α and T_g is placed in a common class of properties, if it is taken into account that T_g can be calculated from the dilatometry data. In fact, the graphs α versus C and T_g versus C show great similarity, including the existence of a turning point. This particular point is placed in the 10-12.5 mol% Al₂O₃ area when significant structural changes occur – see Figures 4 and 5.

A possible correlation α versus C can be deduced from Debye's state equation for solids [1]:

$$\alpha = 0.33 \cdot \gamma \cdot C_V \cdot \chi / V_m \quad (15)$$

where γ is Gruneisen's constant; C_V - Isochoric heat capacity; χ - compressibility.

The relationship (15) is also applicable to homogeneous vitreous oxide systems, which can be considered isotropic. It reflects a proportionality between α and C . This dependence is checked on a side of the graph in fig. 4, on the compositional range $0 \div 12.5$ mol. % Al₂O₃. For higher content of Al₂O₃ the dependence is inversed. In this area, where α increases as compactness decreases, the explanation could be the stronger influence of structural changes AlO₄ \rightarrow AlO₆ and BO₃ \rightarrow BO₄. These transformations are accompanied by a decrease in the bonding energy and, implicitly, by the increase of the expansion coefficient for the Al-O and B-O bonds.

In case of the dependence T_g versus C in the compositional area rich in Al₂O₃, the increasing of T_g as C is decreasing can also be explained by the fact that, in this area, the glasses (with a lower content of Na₂O and higher than Al₂O₃) are becoming more and more fusible. In conclusion, T_g , which for a large number of oxide glasses is a fraction of the melting temperature [1], has higher values.

By comparing, one by one, the figures 2-5 with the figures 6-9, it is noted that the graphs, d (n, α, T_g) versus V_m represent the mirror image for the homologous, d (n, α, T_g) versus C .

The explanation from the numerically point of view resides in the fact that, according to the relation (5), the properties V_m and C are inversely proportional.

From a phenomenological point of view, the shape of the graph in Figure 6 is explained on the basis of the definition relation (10) for V_m . The relationship d versus V_m can be approximated quite well by the linear equation:

$$d = -0.03955 \cdot V_m + 3.50125 \quad (16)$$

for which $R^2 = 0.9949$ and $SD = 0.0080$.

On the range $x = 0 \div 17.5$, the relationship (10) can be rewritten in the simplified form:

$$d = \frac{70.5}{V_m} \pm 3.5\% \quad (17)$$

Since n it is considered to vary linearly relative to d , it is also easy to understand the shape of the graph n versus V_m (Figure 7), which can be expressed by the function:

$$n = -0.0044 \cdot V_m + 1.63882 \quad (18)$$

for which $R^2 = 0.9936$ and $SD = 0.0010$.

For another property that can reflect the compactness of a structure, the volume of the oxygen ion, $V_{O^{2-}}$, the graphical dependencies d (n, α, Tg) versus $V_{O^{2-}}$ are similar to those illustrated in figures 6÷9. The explanation is that between $V_{O^{2-}}(x)$ and $V_m(x)$ there is a relationship of proportionality:

$$V_{O^{2-}} = V_m \cdot \frac{230 + 2x}{(66.95 + 0.4x)} \quad (19)$$

The relation (19) can be written more simply for $x = 0 \div 17.5$:

$$V_{O^{2-}} \cong 0.404 \cdot V_m \quad (20)$$

The calculation of relation (20) gives a maximum deviation 6.7%.

5. Conclusions

- The structure of vitreous oxide systems has a hierarchy. At the first structural (atomic) level among the associated characteristics, the structural compactness, C , the molar volume, V_m and the volume of oxygen ion $V_{O^{2-}}$ are calculable.
- For 6 glasses from the Na₂O-Al₂O₃-B₂O₃ system, the measured properties – density, refractive index, linear thermal expansion coefficient, glass transition temperature are dependent on the characteristics C , V_m and $V_{O^{2-}}$.

- From the obtained data it follows that:
 - the density and refractive index correlate strongly with the characteristics C , V_m and (by extension) $V_{O^{2-}}$, dependency functions being practically linear;
 - for the linear thermal expansion coefficient and for the glass transition temperature, it is revealed that they depend to a great extent on C și V_m ($V_{O^{2-}}$), but also put in evidence the influence of some other structural characteristics (type of cation/oxygen coordination and cation – oxygen bonding energy).
- The presented results show that a number of physical properties of glasses are strongly dependent on a series of structural characteristics on a sub-nanometric scale, such as those that measure the structural compactness.

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