DEPENDENŢA GRADULUI DE COMPACTARE STRUCTURALĂ DE CARACTERISTICILE COMPOZIŢIONALE ALE UNOR STICLE DIN SISTEMUL $Na_2O–Al_2O_3–B_2O_3$

DEPENDENCE OF STRUCTURAL COMPACTNESS UPON THE COMPOSITIONAL CHARACTERISTICS FOR THE GLASSES IN $Na_2O–Al_2O_3–B_2O_3$ SYSTEM

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Vitreous oxide systems can be described at several structural levels. Thus, the first considered is the atomic level, where typical structural entities are oxygen ions and various cations in fixed proportions. In this case, the determined properties of the glass are the molar volume, the molar volume of oxygen ion and compactness.

In our research work all these properties were calculated based on the experimental data obtained by analyzing glasses with (35–x) $Na_2O·xAl_2O_3·65B_2O_3$ molar formula, where x = 0; 5; 10; 12.5; 15; 17.5. Practically, there were established mathematical relations for quantities such as structural compactness - chemical composition, structural compactness – structural characteristics of the studied glasses, respectively.

Sistemele oxidice vitroase pot fi descrise la mai multe niveluri structurale. Astfel, un prim nivel considerat este cel atomic, când entităţile structurale caracteristice sunt ionii de oxigen şi diverşi cationi, în proporţii stabilite. În acest caz, proprietăţile sticlei determinate sunt volumul molar, volumul molar al ionului de oxigen şi compactitatea.

În lucrare, toate aceste proprietăţi au fost calculate pe baza datelor experimentale obţinute prin măsurarea unor proprietăţi pentru sticlele având formula molară (35–x)$Na_2O·xAl_2O_3·65B_2O_3$, unde x = 0; 5; 10; 12,5; 15; 17,5. Concret, s-au stabilit relaţii de calcul de tipul compactare structurală versus compoziţie chimică, respectiv, versus caracteristicile structurale ale sticlelor studiate.

Keywords: *Na_2O-Al_2O_3-B_2O_3 glass, molar volume, molar volume of oxygen ion, compactness, density, correlation*

1. Introduction

Ternary oxide glasses are the simplest systems containing 3 oxides types having different functions:

- network formative oxides, such as B_2O_3, SiO_2, P_2O_5, GeO_2 etc;
- network modifier oxides represented usually by the alkali or alkali earth oxides;
- intermediaries oxides can play the role of formative network or network modifier oxides, depending on the effective chemical composition of the glass, such as Al_2O_3, Ga_2O_3 etc.

Usually, the addition of an alkali oxide in a required composition glass determines the change of its properties. This is an extensive and cumulative effect. In some cases, properties modification is also generated by a qualitative effect, represented by structural changes. From this point of view, glasses can be divided into two categories.

In the first category are placed, for example, glasses of the type Na_2O-Al_2O_3-B_2O_3 where the change of the Na_2O content determines structural changes for both oxides, Al_2O_3 with intermediary role, which may be present in two forms (AlO_4 and AlO_6) and for the network formative oxide, B_2O_3 (as BO_3 and BO_4). A similar situation is found for the glasses based on GeO_2 (GeO_4 and GeO_6).

In the second category are placed the glasses in which the structural changes occur only in oxides with intermediary role. Such are the ternary glasses in $Na_2O–Al_2O_3–SiO_2$ system and $Na_2O–Al_2O_3–P_2O_5$ system.

Given the implications generally applicable for the vitreous systems concerning the triad oxide composition → structure → properties, it is useful to find quantitative relationship between the glass properties and the oxide composition, and between the glass properties and their structural characteristics, respectively.

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The molar formula for the analyzed glasses was $(35–x)Na_2O·xAl_2O_3·65B_2O_3$, where x = 0; 5; 10; 12.5; 15; 17.5. For these glasses, we aimed to find certain dependencies between the structural compactness versus their structural and chemical characteristics, respectively.

For the investigated glasses, we also established correlations between the molar volume, molar volume of oxygen ion and the above mentioned characteristics.

2. Experimental

Six glasses were synthesized, having the $(35–x)Na_2O·xAl_2O_3·65B_2O_3$ molar composition, where x = 0; 5; 10; 12.5; 15; 17.5. Figure 1 shows their location in the ternary phase diagram of the $Na_2O–Al_2O_3–B_2O_3$ system. One of the glasses, G0, is considered as reference (x = 0), having the oxide composition placed in the $Na_2O–B_2O_3$ binary system. In the ternary system, there were considered 5 oxide compositions, characterized by values of x between 5 and 17.5% mol. Na_2O.

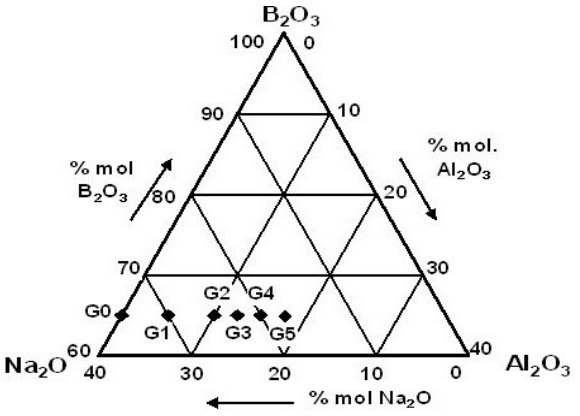

Fig. 1- The molar oxide compositions of the synthesized glasses in the $Na_2O–Al_2O_3–B_2O_3$ ternary diagram / *Compoziţiile oxidice molare ale sticlelor sintetizate în diagrama ternară $Na_2O–Al_2O_3–B_2O_3$*

The conditions and the effective way to obtain the glasses have been described in previous papers [1-3]. At the same time, for synthesized glasses there were measured: density, d; refractive index, n; glass transition temperature, Tg; linear thermal expansion coefficient, α; the wavenumber, $\overline{\nu}$, corresponding to the maximum absorption peak in the UV spectrum; on this basis, an experimental evaluation was made for glass basicity, pB_e, using the chemical and structural parameter "the experimental percentage basicity", pB_e, in %.

Comparing the values for experimental pB and for the calculated pB and using the fitting method we determined what fraction of Al_2O_3 is in coordination 4 (a) and in coordination 6 (1–a), respectively, and the fraction of B_2O_3 in coordination 3 (b) and, in coordination 4 (1–b), respectively. The values for density, a, 1–a, b, 1–b, useful in subsequent calculations, are presented in Table 1.

3. Theoretical background

3.1. Definition relations

In order to evaluate the structural compactness of the oxide systems one can use physical properties such as the molar volume, V_m, the molar volume of oxygen ion, $V_{O^{2-}}$, the compactness, C, the volume obtained by adding the volumes of oxide components, V^0, (in the limit case when the oxide components of glass would form an ideal solution).

The presented properties are calculated with the following relations [4]:

$$V_m = \frac{\overline{M}}{d} = \frac{\sum_i x_i \cdot M_i}{d} \quad (1)$$

$$V^0 = \sum \frac{x_i \cdot M_i}{d_i} = \sum_i x_i \cdot V_{mi} \quad (2)$$

$$\Delta V = V^0 - V_m \quad (3)$$

$$V_{O^{2-}} = \frac{V_m}{\sum_i x_i \cdot N_i} \quad (4)$$

Table 1

Evolution of the weight fraction, a, 1 – a, b, 1 – b and density, d, depending on the glass compositions in $(35–x)Na_2O·xAl_2O_3·65B_2O_3$ system / *Evoluţia fracţiilor gravimetrice a, 1 – a, b, 1 – b, precum şi a densităţii, d, în funcţie de compoziţia sticlelor în sistemul $(35–x)Na_2O·xAl_2O_3·65B_2O_3$.*

Glass no. / *Nr. sticlă*	x [% mol. Al_2O_3]	a	1 – a	b	1 – b	d [10^3 kg/m^3]
G0	0	0	0	0.53	0.47	2.400
G1	5	0.602	0.398	---	---	2.338
G2	10	0.655	0.345	0.516	0.484	2.227
G3	12.5	0.927	0.073	0.786	0.214	2.218
G4	15	1	0	1	0	2.188
G5	17.5	0.934	0.066	0.793	0.207	2.133

$$C = \sum_i \sum_j \frac{x_{ij} \cdot V_{ij} \cdot N_A}{V_m} \quad (5)$$

where $\overline{M}$ is the average molar mass of the glass, in kg / mol; d – the glass density, in kg / m^3; x_i – the molar fraction of the oxide i; M_i – molecular weight of the oxide i, in kg / mol; d_i – the density of the oxide component i; N_i – the number of oxygen atoms in the oxide i; V_{ij} – the volume of j ion from i oxide; x_{ij} – the atomic fraction of j ion from oxide i; N_A – Avogadro's number (6.023 x 10^{23} mol^{-1}).

3.2. The molar volume and derived quantities in the $Na_2O–Al_2O_3–B_2O_3$ system

Considering the chemical molar formula of the synthesized glasses and the definition relation (1) the molar volume $V_m(x)$ is then calculated with the formula (6), where $M_{(MO)}$ is the molecular weight of the oxide MO and $d(x)$ the density of glass with x % mol. Al_2O_3.

Considering a limit case that the glass is an ideal solution of the mixture of component oxides, the resulting additive volume V^0 is determined by the formula (7).

Table 2 shows the input data needed for calculating the property V^0 .

With these specifications, the calculation formula for V^0 is:

$$V^0 = 33.35 + 0.0182x \quad (8)$$

Consequently, the volume deviation ΔV for studied glasses is explained with the relationship:

$$\Delta V = 33.35 + 0.0182x - \frac{66.95 + 0.4x}{d(x)} \quad (9)$$

The calculation of the volume occupied by one mole of oxygen ions is made on the assumption that the oxygen ion radius is considerably higher than the cations existing in glass. Consequently, it is considered that they occupy the interstitial positions between oxygen ions, more voluminous.

Table 3 shows the ionic radii for the structure of studied glasses, the ionic ratio and the ionic ratio of volumes.

$$V_m(x) = \frac{(35-2x)\cdot M_{(Na2O)} + x \cdot M_{(Al2O3)} \cdot +65 \cdot M_{(B2O3)}}{100 \cdot d(x)} = \frac{66.95 + 0.4x}{d(x)} \quad (6)$$

$$V^0 = \frac{(35-2x)\cdot M_{(Na2O)}}{100 \cdot d_{(Na2O)}} + \frac{x \cdot M_{(Al2O3)}}{100 \cdot d_{(Al2O3)}} + \frac{65 \cdot M_{(B2O3)}}{100 \cdot d_{(B2O3)}} \quad (7)$$

$$V_{O^{2-}} = \frac{100 \cdot V_m}{1 \cdot (35-x) + 3x + 3 \cdot 65} = \frac{(66.95 + 0.4x)\cdot 100}{(230 + 2x)\cdot d(x)} \quad (10)$$

$$(70-2x)Na * 2xAl * 130B * (230+2x)O \quad (11)$$

$$C(x) = \frac{4\pi N_A}{3V_m} \cdot \left(f_{Na} \cdot r^3(Na) + f_{Al} \cdot r^3(Al) + f_B \cdot r^3(B) + f_O \cdot r^3(O)\right) \cdot 10^{-27} \quad (12)$$

$$C(x) = \frac{4\pi \cdot N_A}{3V_m} \cdot \frac{(70-2x)\cdot 0.098^3 + 2x \cdot 0.057^3 + 130 \cdot 0.025^3 + (230+2x)\cdot 0.14^3}{430+2x} \cdot 10^{-27} \quad (13)$$

$$C(x) = \frac{1756 + 10x}{(430+2x)\cdot V_m} = \frac{(1756+10x)\cdot d(x)}{(430+2x)\cdot(66.95+0.4x)} \quad (14)$$

Table 2

The input data needed to calculation the property V^0 / *Date primare de calcul necesare evaluării* V^0 .

Oxide/*Oxidul*	Mol. wt./ *Masă molară* $M_{(MO)}$	Density / Densitate $d(MO)$ [g/cm^3]	Molar volume / *Volum molar* $V_m(MO)$ [cm^3/mol]
Na_2O	62.0	2.60 [a]	23.85 [b]
α - Al_2O_3 (rhombohedral)	101.9	3.97	25.66
B_2O_3 (cubic)	69.62	1.81	38.46 [c]

[a] – the value used in calculations by Winkelmann and Schott, cited in [5]

[b] – in paper [6] the calculated molar volume for Na_2O is $26 \cdot 10^{-6}$ m^3/mol;

[c] – in paper [6] the calculated molar volume for $BO_{3/2}$ is $18.7 \cdot 10^{-6}$ m^3/mol, respectively $37.4 \cdot 10^{-6}$ m^3/mol for B_2O_3; in reference 10 from the paper [6], the molar volume for B_2O_3 is considered $38.6 \cdot 10^{-6}$ m^3/mol.

Table 3

The ionic radii of the anionic and cationic component [5]
Razele ionice ale anionilor şi cationilor componenţi [5]

Element	Ion	Ionic radius *Raza ionică [nm]*	Ionic ratio *Raport ionic, r*	Ionic ratio of volumes *Raport ionic al volumelor, r_V*
B	B^{3+}	0.025	0.179	0.0057
Al	Al^{3+}	0.057	0.407	0.067
Na	Na^{+}	0.098	0.700	0.343
O	O^{2-}	0.140	---	---

For glasses in the studied system $V_{O^{2-}}$ is calculated using the formula (10).

Based on the molar formula of the studied glasses, we can write their chemical formula related to the atoms (see relation (11)).

The "atomic" formula is used to calculate the structural compactness of glasses, $C(x)$ (see relations (12) and (13)), where $r(x)$ is the ionic radius of ion i, in nm; f_i – the atomic fraction of atom i. Considering $r(Na)$ = 0.098 nm; $r(Al)$ = 0.057 nm: $r(B)$ = 0.025 nm; $r(O)$ = 0.140 nm, we can write for $C(x)$ the final expression (14).

4. Results and discussions

The evolution of the properties according to the chemical composition of the glasses in Na_2O–Al_2O_3–B_2O_3 system may be understood in relation to the structural particularities, starting with the basic glass, B_2O_3.

In B_2O_3 glass the elementary structural entity is [BO_3]. At the next structural level, the glass is made of boroxol rings interconnected by some triangles [BO_3]. For a B_2O_3 glass with density $1.81.10^{-6}$ kg/m^3, the molar volume is $38.45.10^{-6}$m^3/mol.

The incorporation of network modifier oxides in B_2O_3 glass may have different effects, depending on the size and the intensity of the electrostatic field of the cations, but also according to the amount of oxide, as well.

At the same time, the newly introduced O^{2-} oxides, more basic, determine the conversion of some [BO_3] structural entity in the [BO_4] structural units. The maximum degree of conversion occurs in the glasses with a diborate chemical composition. At the same time, the maximum value is different, for example, in binary glasses M_2O - B_2O_3 depending to the alkali ion M type. Thus, for the borate glasses the type of elementary structural entities are [BO_3] and [BO_4].

Finally, the higher structural level complex structural entities are formed, such as pentaborate, triborate, thetraborate, methaborate, pyroborate and orthoborate groups [5,7].

However, due to certain sterical constraints (resulting from asymmetry) and to some electrostatic effects, some BO_3 – O – BO_4 bonds break, non-bridging oxygen O_{np} are appearing. They form BO_3 triangles, with one, two or three positions occupied by O_{np}. The alkali ions neutralize these negative charges, including those generated by the [BO_4] entities formation.

The conversion effect [BO_3] $\rightarrow$ [BO_4] is increasing the volume of covalent network for borate glasses.

At the same time, the cations of network modifier oxides are placed in the vacant spaces formed by the mainly covalent network B – O – B. The cations with smaller ionic radius and high electrostatic field (Li^+ case) determine, by interacting with oxygen ions from the network, a "constraint" applied to the network. The result is a decrease of the molar volume. At higher Li_2O percentages, the number of cations occupying the free interstitial positions is higher and they begin to run a "pressure" on the covalent structure. Consequently an ‘expansion’ of the network takes place, which determines an increase of the molar volume [8].

When the modifier oxide is Na_2O, the Na^+ cation has a radius similar to that of O^{2-}, and a relatively small electrostatic field. At low concentrations, these cations occupying vacant spaces exercise a low pressure on the network, which expands. At higher concentrations, the increase in the network volume is bigger. Consequently, there is an increase of the molar volume of glasses on a faster pace (for example, the $Na_2O – SiO_2$ system [8]).

If in the reference glass $Na_2O.2B_2O_3.Al_2O_3$ is added, using the molar chemical formula $(35–x)Na_2O{\cdot}xAl_2O_3{\cdot}65B_2O_3$ some structural changes take place. Thus, the replacement of Na^+ cations with Al^{3+} cations in the holes of covalent structure determines, at least at the lower concentration of Al_2O_3, a structural relaxation due to the decrease of Na^+ ions pressure on the network. As a result, it can reduce the oxide volume of glasses (ratio r_C/r_{O2-} is lower for Al^{3+} ions, but their electrostatic field is higher, relative to Na^+ ions).

At the same time, in the $Na_2O–Al_2O_3–B_2O_3$ system, for values $Na_2O/Al_2O_3 > 1$ the sodium ions are used mainly to form the [AlO_4] unitary structural entities, following the conversion [AlO_6] $\rightarrow$ [AlO_4]. (In $Na_2O–Al_2O_3–SiO_2$ system [5,9] the conversion is done for the ratio $Na_2O/Al_2O_3 < 1$).

As a result, the decrease in the availability of Na^+ ions for the reaction with B_2O_3 determines a reconversion [BO_4] $\rightarrow$ [BO_3], which implies a decrease in the network volume and, a decrease of the molar volume of glass as the Al_2O_3 percentage increases, respectively. But at the same time, following the formation of more and more [AlO_4] structural entities, involved in the formation of the vitreous structure, leads to an increase in its volume. Consequently, there is an increase of the molar volume of the glasses, with increase of Al_2O_3 percentage. Partial compensation of the antagonistic effects shown in

the oxide composition on the molar volume of the studied glasses generates a resultant leading to the increase of the molar volume.

A first consequence of this fact is that the structural compactness of glasses having the molar chemical formula $(35–x)Na_2O \cdot xAl_2O_3 \cdot 65B_2O_3$ decreases in inverse proportion to the Al_2O_3 content.

Secondly, although the molecular weight of the glasses increases with the increase of x, the molar volume grows more for the same x values, resulting inverse proportion dependence between the density of glasses and the Al_2O_3 percentage, according to the experimental data presented in Table 1.

Specifically, if x has the value from 0 to 17.5, the molecular weight of the glass increases by 10%, while the molar volume increases by about 27%.

On the basis of experimental data for the density of the synthetized glasses were calculated V_m and V^0. Their dependence on % mol. Al_2O_3 is shown in Figure 2. Figure 3 shows graphically the dependence of ε_r on x, where ε_r is defined by the relationship:

$$\varepsilon_r = (V^0 - V_m) \cdot 100 / V^0 \qquad (15)$$

Finally, from the equations for $V_{O^{2-}}$, expressed in 10^{-6} m^3/mol, it was plotted the graph of Figure 4, showing the $V_{O^{2-}}$ dependence on % mol. Al_2O_3.

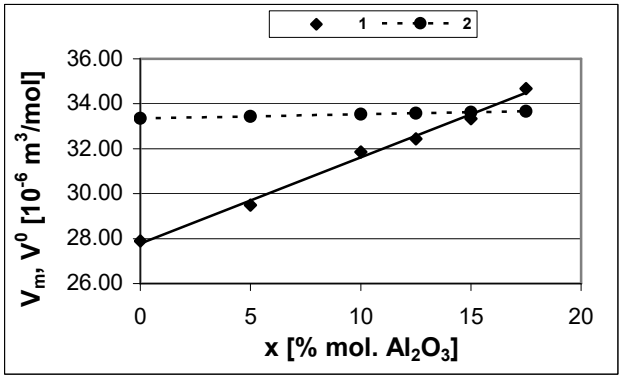

Fig 2 - The variation of V_m, (**1**) and of V^0, (**2**) with % mol. Al_2O_3 / *Variaţia V_m (1) şi a V^0 (2) cu % mol. Al_2O_3.*

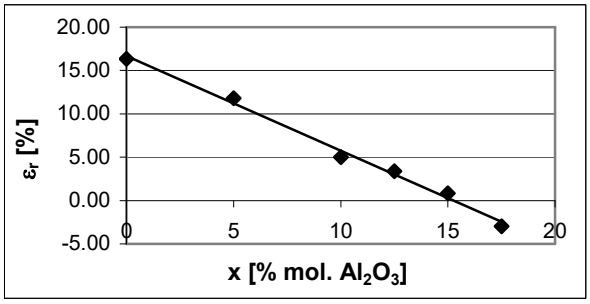

Fig. 3- The dependence of relative deviation, ε_r, on % mol. Al_2O_3 / *Abaterea relativă, ε_r, în funcţie de % mol. Al_2O_3.*

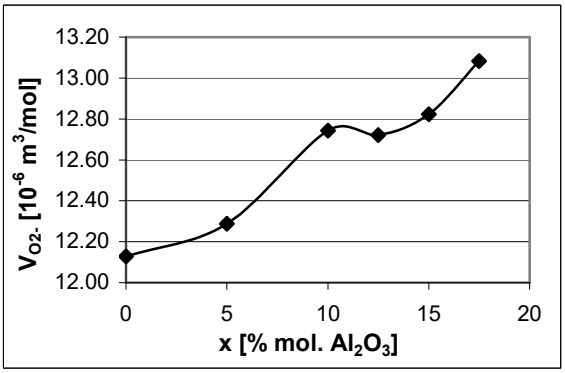

Fig. 4- The dependence of the molar volume of oxygen ion, $V_{O^{2-}}$, versus % mol. Al_2O_3 / *Variaţia volumului molar al ionului de oxigen, $V_{O^{2-}}$, în funcţie de % mol. Al_2O_3*

The graphs of Figures 2 ÷ 4 reveal three important aspects:

- the glasses of the studied system can be considered an ideal solution at 12.5 ÷ 17.5% mol Al_2O_3 concentrations, because V_m and V^0 have very close values and $\Delta V \to 0$;
- the molar volume of oxygen ion will change very little relative to x, showing a slight increase on the 0 ÷ 17.5% mol range;
- both V_m and V^0 show deviations from linearity for x = 15÷10% mol., indicating a number of structural changes also reported in [3].

Considering the relation (14), in Figure 5 we present the dependence of the compactness on % mol. Al_2O_3. It shows the decrease of the structural compactness with the increase of x. In this case, it mentions a number of structural changes, in particular in the range 10 ÷ 15 % mol. Al_2O_3.

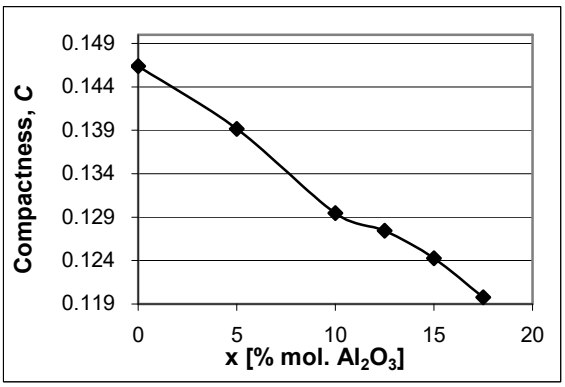

Fig. 5 - The dependence of structural compactness, C, on % mol. Al_2O_3 / *Compactitatea structurală, C, în funcţie de % mol. Al_2O_3.*

5. The dependence of the properties on composition, respectively on structural characteristics

Glass properties cannot be calculated on fundamental basis according to their oxide composition, to the structural characteristics, respectively, except some very simple oxide systems.

Since 1890, Winkelmann and Schott have elaborated a semiempirical methodology for calculation of some properties of oxide glasses P_j using the with relation [5]:

$$P_j = \sum_{i=1}^{n} f_{ij} \cdot x_i \qquad (16)$$

where x_i is the gravimetric or molar fraction of the oxide i; f_{ij} – specific additive factor of the oxide i and property j; n – number of oxide components of the glass.

For the studied oxide system we used the factors proposed by Winkelmann and Schott, and also by Appen [5] to calculate the density and the molar volume of the studied glasses, respectively.

The calculations have led to unsatisfactory results, probably because in determining of respective additive factors (chosen from literature) were not taken into account similar compositions to those studied in the paper.

Consequently, it was applied the mathematical regression method for the experimental density values (d). Taking in account the molar formula of the analyzed glasses was established a following functional dependency:

$$d = f_1 \cdot (35 - x) + f_2 \cdot x + f_3 \cdot 65 \qquad (17)$$

where f_1, f_2 and f_3 are unknown coefficients attached to the Na_2O, Al_2O_3 oxides, respectively B_2O_3 ones.

Relation (17) is written as:

$$d(x) = (35 \cdot f_1 + 65 \cdot f_3) - (35 \cdot f_1 - f_2) \cdot x \qquad (18)$$

or :

$$d(x) = A + B \cdot x \qquad (19)$$

where A and B are coefficients to be determined statistically.

Figure 6 shows the dependence of d on x.

The regression analysis leads to the numerical identification of the statistics coefficients: A = 2.4016; B = – 0.0151; R = – 0.99; SD = 0.015.

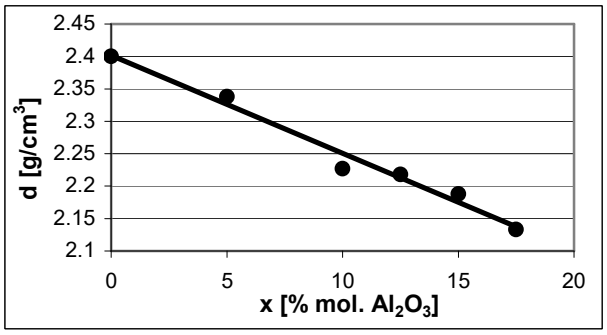

Fig. 6 -The density of the glasses function of the % mol. Al_2O_3 / *Densitatea sticlelor în funcţie de % mol. Al_2O_3.*

The main weakness of the relations type property - oxide composition resides in the fact that they cannot reveal the effect of certain structural changes (e.g. changes in the coordination number of some cations: B^{3+}, Al^{3+}, etc., in relation to oxygen. Therefore, the properties calculation requires to be conducted in relation to various structural entities of glass and not according to the oxide composition.

On the basis of relation (19) and taking into account the relation (6), (9), (10) and (14) it can be written the formula for the calculation of V_m, $V_{O^{2-}}$ and C, in relation with the oxide composition of the studied glasses. For example:

$$V_m = \frac{66.95 + 0.4x}{2.401 - 0.015x} \qquad (20)$$

$$V_{O^{2-}} = \frac{(66.95 + 0.4x) \cdot 100}{(230 + 2x) \cdot (2.401 - 0.015x)} \qquad (21)$$

$$C(x) = \frac{(1756 + 10x) \cdot (2.401 - 0.015x)}{(430 + 2x) \cdot (66.95 + 0.4x)} \qquad (22)$$

In another alternative, the explicit calculation of the molar volume can be done based on some structural characteristics of the vitreous system. In paper [3] for the analyzed system were determined explicit calculation relations for a number of properties. Thus, the density calculation formula is:

$$d = 7.276 \cdot f_{NaO6} + 4.184 \cdot f_{AlO4} + 4.059 \cdot f_{AlO6} + 0.13 \cdot f_{BO4} + 0.01 \cdot f_{BO3} \qquad (23)$$

where f_x is the fraction of structural entity x (respectively: NaO_6, AlO_4, AlO_6, BO_3 and BO_4);

Based on the relationship (1) for the molar volume V_m calculation the formula (24) can be written.

Analogously the relations for $V_{O^{2-}}$ and C are written according to the typical structural entities NaO_6, AlO_4, AlO_6, BO_3 and BO_4 (25, 26).

$$V_m = \frac{M(x)}{d(x,a,b)} = \frac{66.95 + 0.4x}{7.276 \cdot f_{NaO6} + 4.184 \cdot f_{AlO4} + 4.059 \cdot f_{AlO6} + 0.13 \cdot f_{BO4} + 0.01 \cdot f_{BO3}} \quad (24)$$

$$V_{O^{2-}} = \frac{(66.95+0.4x)\cdot 100}{(230+2x)\cdot(7.276\cdot f_{NaO6} + 4.184\cdot f_{AlO4} + 4.059\cdot f_{AlO6} + 0.13\cdot f_{BO4} + 0.01\cdot f_{BO3})} \quad (25)$$

$$C(x) = \frac{(1756+10x)\cdot(7.276\cdot f_{NaO6} + 4.184\cdot f_{AlO4} + 4.059\cdot f_{AlO6} + 0.13\cdot f_{BO4} + 0.01\cdot f_{BO3})}{(430+2x)\cdot(66.95+0.4x)} \quad (26)$$

Table 4

The values V_m obtained on different ways / *Valorile pentru* V_m *obţinute pe căi diferite.*

Glass no. / *Nr. sticlă*	x [% mol. Al_2O_3]	Molar volume / *Volumul molar*, V_m, [10^{-6} m^3/mol]				
		Experimental	Rel. (20)	ε_r [%]	Rel. (24)	ε_r [%]
G0	0	27.90	27.88	0.04	27.91	-0.04
G1	5	29.49	29.64	-0.52	29.97	-1.61
G2	10	31.86	31.52	1.07	31.92	-0.18
G3	12.5	32.44	32.51	-0.20	32.57	-0.41
G4	15	33.34	33.52	-0.55	33.34	0.00
G5	17.5	34.67	34.58	0.26	34.67	0.00

Table 5

The values $V_{O^{2-}}$ obtained in different ways / *Valorile pentru* $V_{O^{2-}}$ *obţinute pe căi diferite.*

Glass no. / *Nr. sticlă*	x [% mol. Al_2O_3]	Molar volume of oxygen ion / *Volumul molar al ionului de oxigen*, $V_{O^{2-}}$ [10^{-6} m^3/mol]				
		Experimental	Rel. (21)	ε_r [%]	Rel. (25)	ε_r [%]
G0	0	12.13	12.124	0.04	12.13	-0.04
G1	5	12.29	12.351	-0.52	12.49	-1.61
G2	10	12.74	12.608	1.07	12.77	-0.18
G3	12.5	12.72	12.747	-0.20	12.77	-0.41
G4	15	12.82	12.894	-0.55	12.82	0.00
G5	17.5	13.08	13.049	0.26	13.08	0.00

Table 6

The values C obtained in different ways / *Valorile pentru* C *obţinute pe căi diferite.*

Glass no. / *Nr. sticlă*	x [% mol. Al_2O_3]	Compactness / *Compactitatea*, C				
		Experimental	Rel. (22)	ε_r [%]	Rel. (26)	ε_r [%]
G0	0	0.1464	0.1465	-0.04	0.1463	0.04
G1	5	0.1392	0.1385	0.51	0.1370	1.58
G2	10	0.1295	0.1309	-1.08	0.1292	0.18
G3	12.5	0.1274	0.1272	0.20	0.1269	0.41
G4	15	0.1243	0.1236	0.55	0.1243	0.00
G5	17.5	0.1198	0.1201	-0.26	0.1198	0.00

Analyzing the date presented in Tables 4÷6 it results that both dependencies property versus oxide composition and property versus structural entities agree very well with the experimental values. However, a higher degree of correlation is recorded in case the structural entities of the glass are considered independent variables.

6. Conclusions

In the $Na_2O–Al_2O_3–B_2O_3$ oxide system, using the experimental data concerning the glass density having the molar formula $(35–x)Na_2O·xAl_2O_3·65B_2O_3$, where x = 0; 5; 10; 12.5; 15; 17.5, the following properties have been calculated: the molar volume, corresponding to hypothesis that the glasses are considered ideal solutions, the molar volume of oxygen ion and the structural compactness.

It shows that the first two properties increase with increasing of % mol. Al_2O_3, while for compactness is a decrease.

The dependencies property versus % mol. Al_2O_3 put in evidence a domain in the range of 10 ÷ 15% mol. where the most important structural changes occur, reflected by major deviations from linearity.

The main properties which reflect the degree of structural behavior were put into quantitative relations with the oxide composition, with the number of structural characteristics of the studied glasses, respectively. The results are compared with those obtained experimentally, resulting in a very good agreement.

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