

# DEPENDENȚA UNOR PROPRIETĂȚI TERMODINAMICE DE BAZICITATE PENTRU COMPUȘII OXIDICI DIN SISTEMUL $M_2O - SiO_2$ ( $M = Li, Na, K, Rb, Cs$ ) DEPENDENCE OF SOME THERMODYNAMIC PROPERTIES OF OXIDE COMPOUNDS ON THE BASICITY IN THE $M_2O - SiO_2$ ( $M = Li, Na, K, Rb, Cs$ ) SYSTEM

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The type, proportion and chemical-structural characteristics of compounds that are formed as a result of chemical reactions between oxides are determined by the chemical-structural characteristics of the starting oxides and the processing conditions (genesis route).

In present paper, we investigate the influence of basicity on the values of some thermodynamic functions. The considered thermodynamic functions correspond to the forming reactions of the oxide compounds in the alkali-silicate systems.

It was shown that there was no strong correlation between the considered thermodynamic functions (standard enthalpy of formation,  $\Delta H^0_{298}$ , standard Gibbs free energy of formation,  $\Delta G^0_{298}$ ) and the percentage of basicity, pB (%).

However, a strong correlation was determined between the "specific" thermodynamic properties and pB%. The specific thermodynamic properties were calculated by dividing the values of the standard (classic) functions to the number of moles of oxide in the compound which resulted from the reaction.

Tipul, proporția și caracteristicile chimico-structurale ale compușilor care apar în urma reacțiilor între diverși oxizi sunt determinate de două grupe principale de factori de influență: particularitățile chimico-structurale ale reactanților (oxizii de start) și condițiile de procesare (rută de geneză).

Deoarece reacțiile de formare a compușilor oxidici reprezintă reacții de neutralizare (de tip acid-bază) între oxizii reactanți, în lucrare se relevă în ce măsură bazicitatea sistemului determină valoric diversele funcții termodinamice atașate reacției de formare a compușilor din sistemele alcalino-silicatic.

Se evidențiază că, oarecum surprinzător, nu există o corelație puternică între funcțiile termodinamice considerate (entalpie standard de formare,  $\Delta H^0_{298}$ , și energia liberă standard de formare Gibbs,  $\Delta G^0_{298}$ ) și bazicitatea compușilor comensurată prin parametrul „ponderea bazicității”, pB, în %.

Totodată, o foarte strânsă corelație se manifestă între proprietățile termodinamice „specifice” și pB. Proprietățile termodinamice specifice s-au obținut raportând mărimile standard (clasice) la numărul de moli de oxid din compusul rezultat în urma reacției.

**Keywords:** Standard Gibbs free energy, Standard enthalpy, Basicity, Alkali-silicate systems

## 1. Introduction

It is generally accepted that the properties of solids are mainly determined by their stereochemical characteristics and by the nature and intensity of the interactions between the atoms in the chemical systems.

In oxide systems, the formation of some compounds occurs as a result of neutralizing reactions (between the reacting oxides considered 'basic' and the other partners, considered more 'acid'). The nature of the resulted products is determined by the acido-basic character of the reacting oxides and of the resulted oxide compounds, according to the corresponding phase diagrams [1].

A series of thermodynamic functions are used to describe the reactions between oxides: the

enthalpy of formation,  $\Delta H$ , the entropy of formation,  $\Delta S$ , the Gibbs free energy of formation,  $\Delta G$ , the enthalpy of reaction,  $\Delta^r H$ , the entropy of reaction,  $\Delta^r S$ , the Gibbs free energy of reaction,  $\Delta^r G$ . The values of these thermodynamic properties have been determined for most solid oxides, as well as for a series of oxide compounds [2, 3]. Additionally, the values were correlated with the chemical composition of the starting oxide system and with temperature [2, 4].

Thus, a strong interrelation is expected between the thermodynamic properties and the basicity of the oxide system.

Several approaches were made towards the qualitative and/or quantitative evaluation of the acido-basicity of chemical systems. Obviously, this interest was manifested for oxide materials, as well.

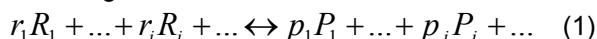
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For instance, papers [5-9] describe a series of synthetic approaches.

Baltă et al. propose to use 'the basicity percentage, pB %' as evaluating measure for the basicity of oxides and oxide combinations. This approach, having an integrative chemical-structural character, was used to correlate a series of structural characteristics of oxides with their basicity. In some oxide systems – either crystalline or vitreous - the dependence of some of the properties on the basicity was also emphasized [7-11].

## 2. Theoretical basis

A chemical reaction is generally carried on according to the scheme:



where  $R_1, \dots, R_i$  are the reactants;  $P_1, \dots, P_j$  are the products;  $r_1, \dots, r_i$  are the stoichiometric coefficients of the reactants;  $p_1, \dots, p_j$  are the stoichiometric coefficients of the products.

A series of thermodynamic functions can be associated to reaction (1), as follows [12]:

- The enthalpy of reaction,  $\Delta^r H_{T,P}$ :

$$\Delta^r H_{T,P} = \sum_j p_j \cdot H(P_j) - \sum_i r_i \cdot H(R_i) \quad (2)$$

where:  $H(P_j)$  is the molar enthalpy of the product,  $j$ , in kJ/mole;  $H(R_i)$  – molar enthalpy of the reactant,  $i$ , in kJ/mole; temperature  $T$  and pressure  $P$ , the state parameters. For standard conditions ( $T = 298$  K and  $P = 1$  atm.) the standard enthalpy of reaction,  $\Delta^r H_{298}^0$ , is defined. The standard enthalpy of reaction is calculated using the enthalpies of formation of the reactants,  $\Delta H_{298}^0(R_i)$ , and of the products,  $\Delta H_{298}^0(P_j)$ .

The exothermal reactions ( $\Delta^r H_{298}^0 < 0$ ) that occur with the increase of entropy ( $\Delta^r S_{298}^0 > 0$ ), are thermodynamically probable reactions (they evolve in the direction of product formation) at any temperature, meaning that  $\Delta^r G_{298}^0 < 0$  [2,12].

- The standard entropy of reaction,  $\Delta^r S_{298}^0$ , can be calculated based on the standard entropy of the reactants,  $S_{298}^0(R_i)$  and of the products,  $S_{298}^0(P_j)$ , in kJ/mole:

$$\Delta^r S_{298}^0 = \sum_j p_j \cdot S_{298}^0(P_j) - \sum_i r_i \cdot S_{298}^0(R_i) \quad (3)$$

The entropy is used to calculate the free energy of reaction. Also, it offers information regarding the direction of the transformations during the reaction.

- The standard free energy of reaction (Gibbs free energy),  $\Delta^r G_{298}^0$ :

$$\Delta^r G_{298}^0 = \sum_j p_j \cdot \Delta G_{298}^0(P_j) - \sum_i r_i \cdot \Delta G_{298}^0(R_i) \quad (4)$$

where  $\Delta G_{298}^0(P_j)$  is the standard free energy of

formation from of elements for products, in kJ/mole;  $\Delta G_{298}^0(R_i)$ , is the standard free energy of formation from elements for reactants, in kJ/mole.

If  $\Delta^r G_{298}^0 < 0$ , the reaction occurs spontaneously in the direct sense, determining the decrease of the free energy of the system.

The decrease of standard free energy correlates with a stronger tendency of converting the reactants into products.

The enthalpy, entropy and free energy, being thermodynamic functions, depend on the state parameters. Thereby, their variation with temperature  $T$  leads to defining other functions, -  $\Delta^r H_T^0$ , -  $\Delta^r S_T^0$ , and -  $\Delta^r G_T^0$  ( $P = \text{constant}$ ).

A particular importance is given to  $\Delta^r G_T^0$  because depending on it, the equilibrium constant expressed through activities,  $K_a$ , can be calculated:

$$\ln K_a = - \frac{\Delta^r G_T^0}{RT} \quad (5)$$

The evaluation of the acid-basic character of a chemical system has caught the attention of researchers since 18<sup>th</sup> century. Various theories that were trying to qualitatively and/or quantitatively evaluate the acid-basicity of substances are presented, for example, in papers [5-7, 9].

In oxide systems, chemical reactions are acid-base type; regarding these systems, a set of theories was developed in the last decades, allowing both the experimental measurement of basicity and its calculation based on the chemical composition.

Balta et al [8] suggest the basicity percentage, pB, for measurement of the basicity of oxide systems. Considering that  $O^{2-}$  has the highest electron donating power, it has been attributed the maximum basicity value, pB =100%, respectively.

The oxygen ions from various combinations, in which the effective charge is smaller than -2, thus determining, for the chemical bonds they participate to, a lower basic character. The basic character can be experimentally determined by spectroscopic means.

Therewith, pB for oxides can be calculated with the equation proposed by Balta and Radu [8]:

$$\lg pB = 1.9 \cdot (CN)^{0.02} - 0.023 \cdot \frac{P_i}{CN} \quad (6)$$

where CN is the coordination number of the  $M^{z+}$  cation relative to oxygen and  $P_i$  represents the ionization potential for the considered oxidation number of the cation,  $z^+$ , in eV. The  $P_i$  values are easily accessible from tables based on calculated or experimentally determined data.

For complex oxide systems (multicomponent) pB is calculated using the equation:

$$pB = \sum_i pB_i \cdot c_i \quad (7)$$

where  $pB_i$  is the basicity percentage of the  $i$ -th oxide and  $c_i$  is the mass fraction of the  $i$ -th oxide.

### 3. Results and discussion

#### 3.1. Enthalpy and Gibbs free energy versus basicity

A first attempt to quantitatively correlate the heat of formation  $Q$  with ionicity was performed by Pauling [13]. Thus, for molecular systems that contain simple chemical bonds, he proposed the equation ( $Q$  in kJ/mole):

$$Q = 96.3 \cdot \sum_{A,B} (X_A - X_B)^2 - 231.8 \cdot n_N - 108.8 \cdot n_O \quad (8)$$

in which  $X_A$  and  $X_B$  represent the thermodynamic electronegativities (Pauling) of atoms A and B that form the chemical bond A-B;  $n_N$  and  $n_O$  – the number of nitrogen and oxygen atoms in the molecule;  $\sum_{A,B}$  represents the sum of simple A-B type bonds.

Also, the ionicity  $I_{AB}$  of the A-B chemical bond is calculated using the equation:

$$I_{AB} = 100 \cdot \left( 1 - e^{-0.25 \cdot (X_A - X_B)^2} \right) \quad [\%] \quad (9)$$

The considered compounds of every oxide system are presented in Table 1, according to the information given by the phase diagrams [1, 2].

By correlating equations (8) and (9) it ensues:

$$Q = 385.2 \cdot \sum_{A,B} \ln \frac{100}{100 - I_{AB}} - 231.8 \cdot n_N - 108.8 \cdot n_O \quad [\text{kJ/mole}] \quad (10)$$

For the simplest chemical system, characterized by a single A-B type bond and not containing N and O, the variation of the heat of formation  $Q$  with bond ionicity is graphically presented in Figure 1.

By extension, emphasizing a similar correlation between the standard enthalpy of formation,  $\Delta H_{298}^0$  and the basicity percentage,  $pB$ , was pursued for a set of compounds in the  $M_2O - SiO_2$  ( $M = Li, Na, K, Rb, Cs$ ) oxide system.

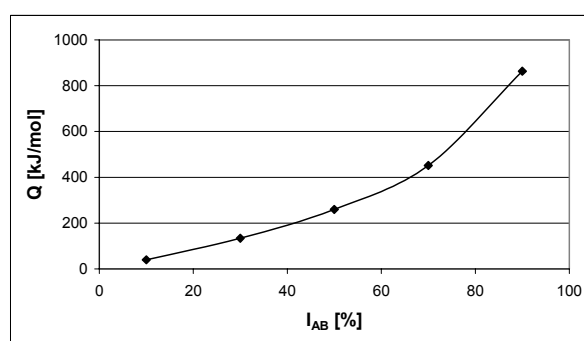


Fig. 1 - Variation of heat of formation  $Q$  with ionicity  $I_{AB}$  for a system with one A-B type bond / Variația căldurii de formare  $Q$  în funcție de ionicitatea  $I_{AB}$  pentru un sistem cu un singur tip de legătură A-B.

Table 1

Compounds  $xM_2O \cdot SiO_2$  that form in  $M_2O - SiO_2$  ( $M = Li, Na, K, Rb, Cs$ ) system / Compușii  $xM_2O \cdot SiO_2$  ce se formează în sistemele  $M_2O - SiO_2$

System / Sistem	Formed $xM_2O \cdot SiO_2$ oxide compounds / Compușii oxidici $xM_2O \cdot SiO_2$ formați						
	$SiO_2$	$x = 1/4$	$x = 1/3$	$x = 1/2$	$x = 1$	$x = 2$	$M_2O$
$Li_2O - SiO_2$	•	---	•	•	•	•	•
$Na_2O - SiO_2$	•	---	•	•	•	•	•
$K_2O - SiO_2$	•	•	---	•	•	•	•
$Rb_2O - SiO_2$	•	•	---	•	•	---	•
$Cs_2O - SiO_2$	•	•	---	•	•	---	•

In Table 2 are presented the primary data for the calculation of basicity using equation (6), respectively the oxidation number,  $z$ , ionization potential  $P_i$  in eV, and the coordination number of the cation related to oxygen, CN.

Table 2

Basicity percentage for alkaline oxides and  $SiO_2$  / Ponderea bazicității pentru oxizii alcalini și  $SiO_2$

Oxide / Oxidul	Oxidation number of the cation / Cifra de oxidare a cationului	$P_i$ [eV]	CN	$pB$ [%], eq. (6)
$Li_2O$	1	5.39	4	83.6
$Na_2O$	1	5.14	6	89.0
$K_2O$	1	4.34	9	94.2
$Rb_2O$	1	4.18	10	95.5
$Cs_2O$	1	3.90	12	97.5
$SiO_2$	4	45.13	4	49.4

The thermodynamic data used for  $\Delta H^0_{298}$  and  $\Delta G^0_{298}$  were taken from [2, 3].

Somehow surprisingly, the  $\Delta H^0_{298}$  versus pB plot show that the dependency of the functions of interest is non monotonic and relatively complex. It is difficult to explain the variation inside the same binary system, as well as to highlight possible influences of the nature of the alkaline oxide. Also, no general explanations that apply to all studied systems can be submitted, mainly because some plots are bimodal, and the plots corresponding to the most basic systems are unimodal and highly asymmetric. To exemplify, in Figures 2-6 are presented the dependency plots of  $\Delta H^0_{298}$  versus pB for the 5 studied systems.

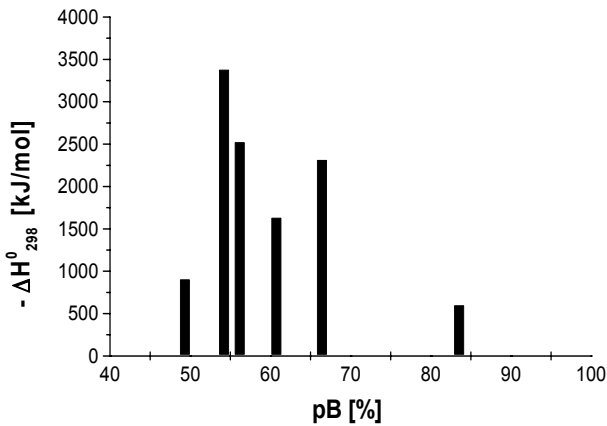


Fig. 2 - Variation of  $\Delta H^0_{298}$  with in the  $Li_2O - SiO_2$  system  
Variația -  $\Delta H^0_{298}$  cu pB în sistemul  $Li_2O - SiO_2$

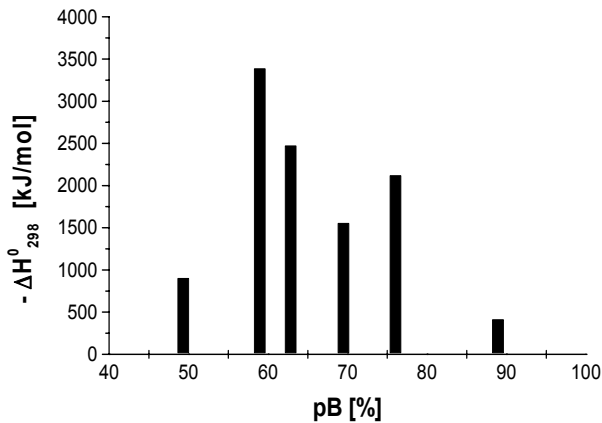


Fig. 3 - Variation of  $\Delta H^0_{298}$  with pB in  $Na_2O - SiO_2$  system  
Variația -  $\Delta H^0_{298}$  cu pB în sistemul  $Na_2O - SiO_2$ .

The conclusions presented here can be extrapolated for  $-\Delta G^0_{298}$  versus pB dependencies because between  $-\Delta G^0_{298}$  and  $-\Delta H^0_{298}$  there is a strong correlation, as illustrated as presented in Figure 7, with the regression equation ( $R^2 = 0.9998$ ):

$$-\Delta G^0_{298} = 0.9461 \cdot (-\Delta H^0_{298}) - 12.571 \quad (11)$$

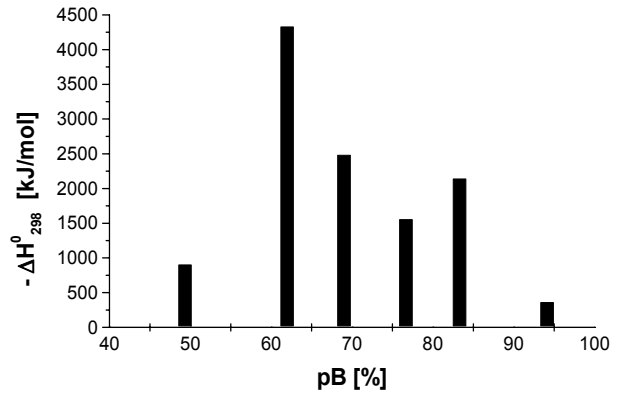


Fig. 4 - Variation of  $\Delta H^0_{298}$  with pB in  $K_2O - SiO_2$  system  
Variația -  $\Delta H^0_{298}$  cu pB în sistemul  $K_2O - SiO_2$ .

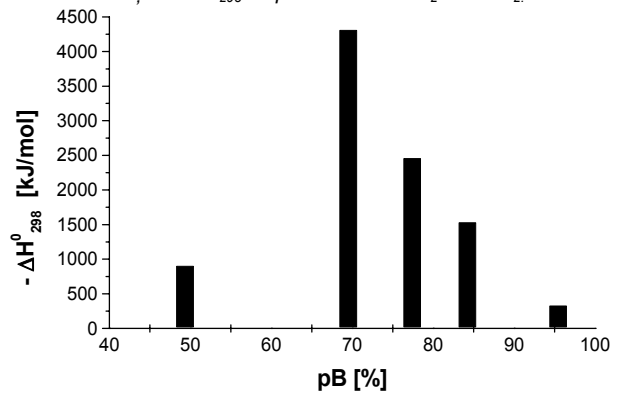


Fig. 5 - Variation of  $\Delta H^0_{298}$  with pB in  $Rb_2O - SiO_2$  system  
Variația -  $\Delta H^0_{298}$  cu pB în sistemul  $Rb_2O - SiO_2$ .

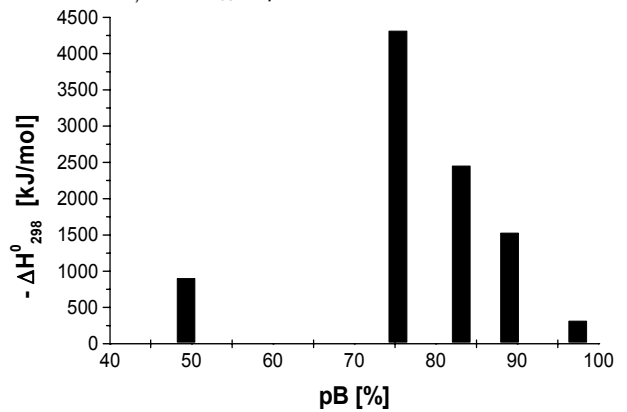


Fig. 6 - Variation of  $\Delta H^0_{298}$  with pB in  $Cs_2O - SiO_2$  system  
Variația -  $\Delta H^0_{298}$  cu pB în sistemul  $Cs_2O - SiO_2$ .

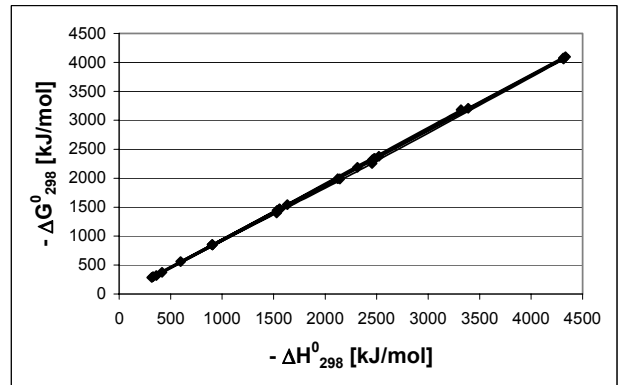


Fig. 7 - Correlation between  $-\Delta G^0_{298}$  and  $-\Delta H^0_{298}$  (in kJ/mole) in  $M_2O - SiO_2$  ( $M = Li, Na, K, Rb, Cs$ ) system / Corelația între  $-\Delta G^0_{298}$  și  $-\Delta H^0_{298}$  (în kJ/mol) în sistemele  $M_2O - SiO_2$  ( $M = Li, Na, K, Rb, Cs$ ).

### 3.2. „Specific’ thermodynamic functions

The explanation of the relatively weak correlation between  $-\Delta H_{298}^0$ ,  $-\Delta G_{298}^0$  and pB can be provided by considering equations (8) and (10). In these equations the heat of formation represents and extensive unit in relation to the number of bonds of the chemical system.

At the same time, for a given chemical system, the ionicity of the chemical bonds represents an intensive unit in relation to the number of same type A-B bonds. By extension, when  $X_xY_y$  oxide compound forms from two oxides X and Y, it can be stated that:

- the thermodynamic properties  $\Delta H_{298}^0$  and  $\Delta G_{298}^0$  are extensive unites in relation to the number of molecules x and y;

- the basicity, expressed through the basicity percentage, represents and intensive unit that does not depend on the quantity of substance.

For this reason, in order to obtain a uniformity of the units of interest, two „specific’ thermodynamic properties were introduced: **the specific enthalpy of formation**  $-\Delta H_{298}^0/N$  and **the specific free energy of formation**  $-\Delta G_{298}^0/N$ , both measured in kJ/mole, where  $N = x + y$ .

Somehow similarly, the paper [14] highlights the dependence of the standard enthalpy of formation on the optical basicity for the analyzed oxides [6, 14].

Figures 8 ÷ 12 graphically present the correlations  $-\Delta H_{298}^0/N$  versus pB and  $-\Delta G_{298}^0/N$  versus pB for the five considered oxide systems.

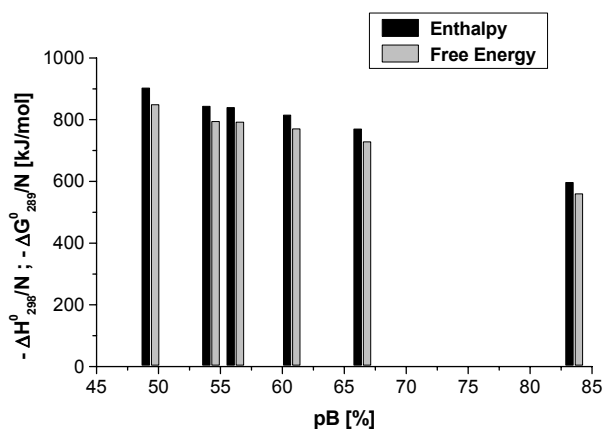


Fig. 8 - Variation of  $-\Delta H_{298}^0/N$  and  $-\Delta G_{298}^0/N$  with pB for  $Li_2O - SiO_2$  system / Variația  $-\Delta H_{298}^0/N$  și  $-\Delta G_{298}^0/N$  versus pB pentru sistemul  $Li_2O - SiO_2$ .

Regression equations that describe the correlations between the properties of interest are presented in the Table 3, for  $-\Delta H_{298}^0/N = f(pB)$ , and in the Table 4 for  $-\Delta G_{298}^0/N = f(pB)$ . The tested equations were of polynomial type:

$$y = a_0 + a_1x + a_2x^2 + a_3x^3 \quad (12)$$

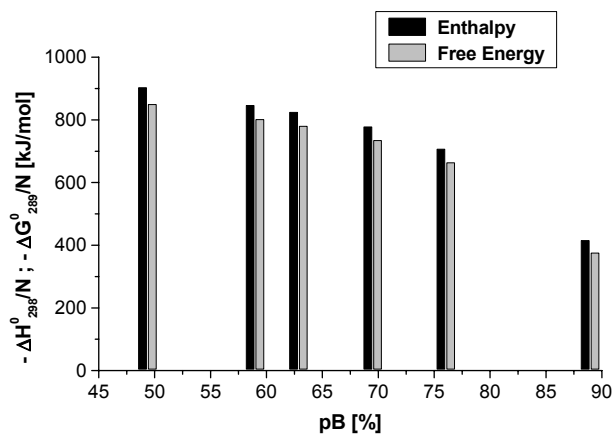


Fig. 9 - Variation of  $-\Delta H_{298}^0/N$  and  $-\Delta G_{298}^0/N$  with pB for  $Na_2O - SiO_2$  system / Variația  $-\Delta H_{298}^0/N$  și  $-\Delta G_{298}^0/N$  versus pB pentru sistemul  $Na_2O - SiO_2$ .

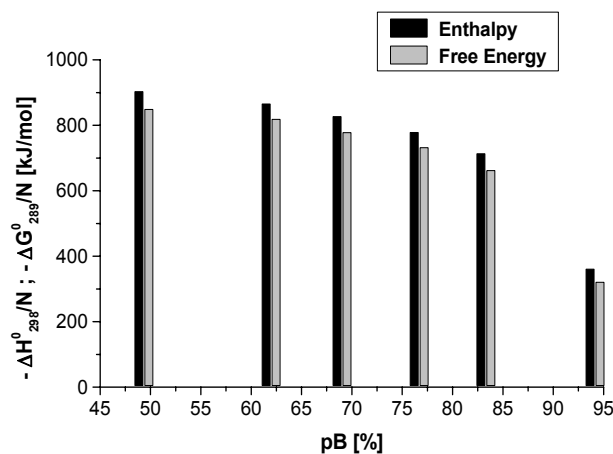


Fig. 10 - Variation of  $-\Delta H_{298}^0/N$  and  $-\Delta G_{298}^0/N$  with pB for  $K_2O - SiO_2$  system / Variația  $-\Delta H_{298}^0/N$  și  $-\Delta G_{298}^0/N$  versus pB pentru sistemul  $K_2O - SiO_2$ .

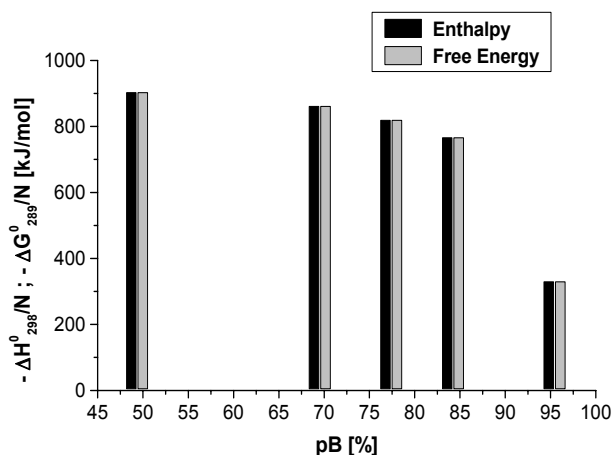


Fig. 11- Variation of  $-\Delta H_{298}^0/N$  and  $-\Delta G_{298}^0/N$  with pB for  $Rb_2O - SiO_2$  system / Variația  $-\Delta H_{298}^0/N$  și  $-\Delta G_{298}^0/N$  versus pB pentru sistemul  $Rb_2O - SiO_2$ .

Table 3

The statistic evaluation of the  $-\Delta H_{298}^0/N$  versus  $pB$  dependency in  $M_2O - SiO_2$  ( $M = Li, Na, K, Rb, Cs$ ) system  
 Evaluarea statistică a dependenței  $-\Delta H_{298}^0/N$  versus  $pB$  în sistemele  $M_2O - SiO_2$  ( $M = Li, Na, K, Rb, Cs$ ).

System / Sistemul	Polynomial coefficients / Coeficienții polinomiali				Correlation coefficient / Coeficient de corelație, $R^2$	Dispersion / Dispersia
	$a_0$	$a_1$	$a_2$	$a_3$		
$Li_2O - SiO_2$	1331.4	-8.669	0	0	0.9866	13.74
$Na_2O - SiO_2$	128.21	30.52	-0.305	0	0.9903	22.25
$K_2O - SiO_2$	-366.08	44.26	-0.384	0	0.9623	49.66
$Rb_2O - SiO_2$	7262.32	-297.20	4.545	-0.023	0.9967	26.90
$Cs_2O - SiO_2$	12324.8	-514.75	7.51	-0.036	0.9917	43.34

Table 4

Evaluarea statistică a dependenței  $-\Delta G_{298}^0/N$  versus  $pB$  în sistemele  $M_2O - SiO_2$  ( $M = Li, Na, K, Rb, Cs$ )  
 The statistic evaluation of the  $-\Delta G_{298}^0/N$  versus  $pB$  dependency in  $M_2O - SiO_2$  ( $M = Li, Na, K, Rb, Cs$ ) system.

System / Sistemul	Polynomial coefficients / Coeficienții polinomiali				Correlation coefficient / Coeficient de corelație, $R^2$	Dispersion / Dispersia
	$a_0$	$a_1$	$a_2$	$a_3$		
$Li_2O - SiO_2$	1256.38	-8.19	0	0	0.9822	14.98
$Na_2O - SiO_2$	18.33	32.14	-0.315	0	0.9915	20.45
$K_2O - SiO_2$	-417.23	44.11	-0.382	0	0.9665	45.86
$Rb_2O - SiO_2$	6930.74	-284.79	4.364	-0.022	0.9964	27.06
$Cs_2O - SiO_2$	10419.8	-433.29	6.36	-0.031	0.9904	44.57

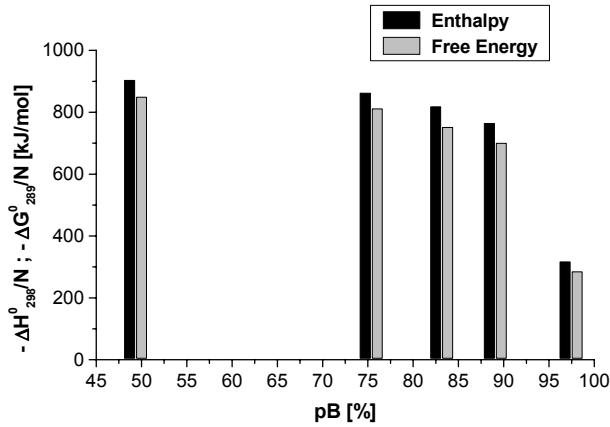
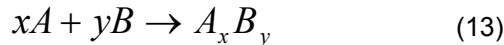


Fig. 12 - Variation of  $-\Delta H_{298}^0/N$  and  $-\Delta G_{298}^0/N$  with  $pB$  for  $Cs_2O - SiO_2$  system / Variația  $-\Delta H_{298}^0/N$  și  $-\Delta G_{298}^0/N$  versus  $pB$  pentru sistemul  $Cs_2O - SiO_2$ .

### 3.3. Calculation of the specific free energy of reaction as a function of basicity

Two oxides A and B are considered to react to form a compound  $A_xB_y$ , according to the reaction:



In this case, for the Gibbs free energy of reaction  $\Delta^r G_{298}^0$ , with respect to equation (13), we can write:

$$\Delta^r G_{298}^0 = \Delta G_{298}^0(A_xB_y) - x\Delta G_{298}^0(A) - y\Delta G_{298}^0(B) \quad (14)$$

in which  $\Delta G_{298}^0(X)$  is the Gibbs free energy of formation for the oxide compound or the oxide X, in kJ/mole.

A formula to calculate the specific Gibbs free energy of reaction  $\Delta G_{298}^0/N$  is obtained from its definition:

$$\frac{\Delta^r G_{298}^0}{N} = \frac{\Delta G_{298}^0(A_xB_y)}{N} - \frac{x\Delta G_{298}^0(A)}{N} - \frac{y\Delta G_{298}^0(B)}{N} \quad (15)$$

in which, with respect to equation (15), it can be considered that:

$$\frac{\Delta G_{298}^0(A)}{N} = F[pB(A)] \text{ and } \frac{\Delta G_{298}^0(B)}{N} = F[pB(B)] \quad (16)$$

Finally, results:

$$\frac{\Delta^r G_{298}^0}{N} = F(pB) - xF[pB(A)] - yF[pB(B)] \quad (17)$$

Equation (13) becomes operational considering that:  $A \equiv M_2O$ ;  $B \equiv SiO_2$ ; x and y gain values according to Table 1; the polynomial coefficients of  $F(pB)$  functions are presented in Table 4.

Also, the equation (13) can be useful in determining a dependency between the basicity of the compound that represents the product of reaction and the number of component moles, so that reaction (13) is more probable and the product more stable. For this it is needed that  $\Delta^r G_{298}^0 < 0$ , for which, out of equation (17) results:

$$F(pB) < xF[pB(A)] + yF[pB(B)] \quad (18)$$

In example, for the  $Li_2O - SiO_2$  system according to Figure 8 and data in Table 4, results:

$$F(pB) = 1257.38 - 8.19 \cdot pB \quad (19)$$

At the same time  $pB(Li_2O) = 83.6\%$  and  $pB(SiO_2) = 49.4\%$  resulting  $F[pB(Li_2O)] = 571.72$  kJ/mole and  $F[pB(SiO_2)] = 851.81$  kJ/mole respectively. With the values from equation (18) results the inequation:

$$pB > 153.4 - 104 \cdot x - 69.8 \cdot y \quad (20)$$

Equation (20) can be used for the prediction of some compounds in the  $Li_2O - SiO_2$  oxide system.

Practically, for a certain compound (known  $x$  and  $y$ ) to form and be thermodynamically stable, its basicity,  $pB$ , needs to respect the condition given by equation (20).

#### 4. Conclusions

In this paper alkali-silicate systems are studied, with direct reference to the correlation of some thermodynamic properties with the basicity of the oxide compounds that can form.

From the presented data results:

- The standard enthalpy of formation and the standard free energy of formation of oxide compounds- in accordance with literature data – do not exhibit an interpretable and generally applicable dependence on the basicity of the products of reaction (the basicity of the oxide compounds was evaluated through the basicity percentage,  $pB$  %).
- By relating the standard thermodynamic functions to the number of moles of oxide in the oxide compound, results a strong correlation with the basicity percentage, which is statistically described by polynomial functions.

- The specific free energy of reaction is strongly dependent on the basicity of the considered oxide system; moreover, predictions can be made about the direction in which a reaction between specific oxides evolves, the possibility that oxide compounds with a certain chemical composition can form, their degree of thermodynamic stability, all in function of basicity percentage  $pB$  of the reactants and products.

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