FORMAREA ŞI PROPRIETĂŢILE UNOR COMPUŞI OXIDICI ÎN RAPORT CU BAZICITATEA LOR
THE FORMATION AND THE PROPERTIES OF SOME OXIDE COMPOUNDS IN RELATION WITH THEIR BASIC CAPACITY

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The formation and the properties of the oxide compounds are determined by intrinsic factors (depending on the reactants) and extrinsic ones (the reaction conditions). The formation reactions are defined by a series of thermodynamic functions. In this paper we highlight the correlation between the Gibbs free energy of formation, the reaction energy and the basicity of the reacting oxides, respectively, evaluated through the basicity percentage, pB, in % in the $CaO - SiO_2$ system.

At the same time, for a series of properties (interaction with water, hydration degree, mechanical resistance to compression, shrinkage) of some mineralogical constituents of the Portland cement clinker it is highlighted their interdependence with basicity.

Formarea şi proprietăţile compuşilor oxidici sunt determinate de factori intrinseci (ai reactanţilor) şi extrinseci (condiţiile de reacţie). Reacţiile de formare se caracterizează cu o serie de funcţii termodinamice. În lucrare se pune în evidenţă corelaţia dintre energia liberă de formare Gibbs, respectiv, energia de reacţie şi bazicitatea oxizilor reactanţi, evaluată prin ponderea bazicităţii, pB, în %, în sistemul $CaO - SiO_2$.

Totodată, pentru o serie de proprietăţi (interacţia cu apa, gradul de hidratare, rezistenţa mecanică la compresiune, contracţia) ale unor constituenţi mineralogici ai clincherului de ciment portland se evidenţiază interdependenţa acestora cu bazicitatea.

1. Introduction

The main structural parameters of an oxide (oxide compound) refer to:

- the electronic structure of the constituent atoms and the hybridization type for the chemical bonds obtained; consequently, there are determined the forms of the coordination polyhedra, the interatomic spacing, the angles between the chemical bonds;

- the ionization degree (covalence) of the chemical bonds formed, and the electronic polarizability of the oxygen ion, respectively;

- the intensity of the electrostatic field, the binding energy O1s and the energy of the chemical bondings with the oxygen.

The structural particularities of the oxide compounds, but not only, finally lead to their properties. As a matter of fact, in time, correlations were shown between a series of properties of the oxide materials and some of the structural parameters presented [1 - 3].

At the same time, the oxides structural particularities determine their acid-basic character, which influences the reactivity and the formation of the oxide compounds. Other influential factors are represented by the reaction conditions (temperature, pressure, etc.). Hence, the necessity to evaluate in a quantitative way the basicity (acidity) of the oxides.

In this context, more theories and quantities were developed, in order to evaluate the acid-basic character of the oxides [4 - 6]. Among these, one seems to stand out, the ″basicity percentage pB parameter″, the importance of basicity, which determines, in %, the basic character of an oxide (oxide compound) or that of some complex oxide systems.

The importance of the basicity percentage, which is correlated to more structural parameters of the oxides [3, 7, 8] can be considered as an involving structural parameter which characterizes an oxide. It was successfully used to correlate some properties of the vitreous oxide systems to their basicity [3, 4, 6].
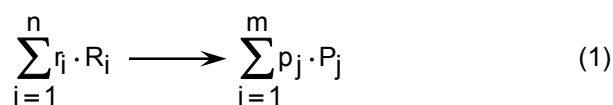
In this paper we try a first attempt to correlate a series of thermodynamic functions associated to the formation of the mineralogical constituents of some clinkers to their basicity. At the same time, we highlight the dependence existing between more properties of the mineralogical constituents and their basicity, evaluated through pB.

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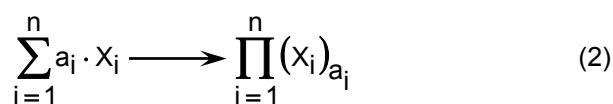
2. The theoretical basis

2.1. Thermodynamic aspects

A generalized chemical reaction can be written:

$$\sum_{i=1}^{n} r_i \cdot R_i \longrightarrow \sum_{j=1}^{m} p_j \cdot P_j \qquad (1)$$

where: R_i represents the reactant i; n - the number of reactants; P_j - the reaction product j; m - the number of the reaction products; r_i, p_j- the stoichiometric coefficients.

For some oxide compounds which represent the mineralogical constituents of various clinker types, reaction (1) is re-written under the particular form:

$$\sum_{i=1}^{n} a_i \cdot X_i \longrightarrow \prod_{i=1}^{n} (X_i)_{a_i} \qquad (2)$$

where: X_i is the oxide reactant i; n - the number of oxides which take part in the reaction; a_i - the stoichiometric coefficients; $(X_i)_{a_i}$ - an oxide compound of the type i.

It should be noticed that reactions of the (2) type relation can be considered "as a general rule". In fact, the oxide compounds can also result through the contribution of some secondary reactions followed by various intermediate oxide compounds, which eventually are consumed, when the thermodynamic equilibrium is reached.

The reactions described by the relation (2) develop according to:
- the chemical-structural particularities of the reacting oxides;
- the proportions between the reacting oxides;
- the obtaining conditions (temperature, pressure, time, cooling speed, etc).

All these influential factors representing the reaction parameters (of the genesis line) in conditions of thermodynamic equilibrium are included in a series of thermodynamic functions. These functions are:
- the enthalpy of reaction:

$$\Delta^r H_{T,P} = \sum_{j} p_j \cdot H(P_j) - \sum_{i} r_i \cdot H(R_i) \qquad (3)$$

where: $H(P_i)$ and $H(R_i)$ are the molar enthalpies of the reaction products, respectively, of the reactants, at constant temperature (T) and pressure (P). If T = 298K and P = 1 atm the standard enthalpy of reaction is defined, $\Delta^r H_{298}^0$,

which is calculated according to the enthalpies of formation of the reactants and reaction products;
- the standard enthalpy of reaction, $\Delta^r S_{298}^0$;
- the standard free energy of reaction (Gibbs free energy), $\Delta^r G_{298}^0$:

$$\Delta^r G_{298}^0 = \sum_{j} p_j \cdot \Delta G_{298}^0 (P_j) - \sum_{i} r_i \cdot \Delta G_{298}^0 (R_i)$$

$$(4)$$

where: ΔG_{298}^0 represents the standard free energy of formation from elements, in kJ/mol, for reactants and reaction products.

If $\Delta^r G_{298}^0$ < 0, the reaction takes place spontaneously and directly (leading to a diminution of the free energy). At the same time, the diminution of the standard free energy of reaction implies a tendency of more accentuated conversion of the reactants into reaction products.

2.2. The evaluation of the basicity of oxides

The atoms which, during a physical or chemical process receive electrons, have a more significant basic nature and become basic ions. An anion's basic nature is more significant as the number of the negative charges is bigger. In return, the atoms which give up one or more electrons become acid ions.

In the case of oxides, the O^{-2} ion is the carrier of the basic property. The chemical interactions between oxides are determined by the exchange of oxygens, which can be of three types: bridging (O^0), non bridging (O^-) and totally isolated (O^{-2}). Thus, in the case of oxides, acid-base reactions represent reactions of oxide reduction which lead to a new distribution of oxygen species compared to the initial one. The reaction is represented:

$$O^{-2} + O^0 \longrightarrow 2O^- \qquad (5)$$

Since in solids the effective charge of the oxygen is less than (-2), even fractional, reaction (5) can be written under the general form:

$$O_i^{-\delta_i} + O_j^{-\delta_j} \longrightarrow 2O_m^{-\delta_m} \qquad (6)$$

Relation (6) pictures the fact that reactions between oxides happen due to the interaction of i and j oxygen ions with different partial charges. At the end, in equilibrium conditions, oxygen ions result having a different average effective charge, δ_m. The equilibrium values are determined by the donor-acceptor power of the i and j ion electrons. Hence, it can be considered that the average

basicity depends on the difference of basicity between the oxygen ions types involved in the reaction.

The groundwork of the evaluation of the basicity of oxides is to consider the free oxygen O^{-2} [5] as having the maximum weight (virtual) of basicity pB = 100%. For oxides and complex oxide solids, the importance of basicity is calculated with the relations:

$$\lg pB = 1,9 \cdot (NC)^{0,02} - 0,023 \cdot \frac{P_i}{NC} \qquad (7)$$

$$pB = \sum_{i=1}^{n} pB_i \cdot c_i \qquad (8)$$

where: NC is the coordination number of the M^{+z} cation with regard to oxygen; P_i - ionization potential for the considered oxidation state z^+ of the corresponding cation; n – the number of component oxides; pB_i - the basicity percentage of the i-th oxide; c_i - the gravimetric fraction of i-th oxide.

As a matter of fact, this neutral condition of the scale for pB leads to choosing this indicator to evaluate the basicity of oxides, compared to others, like the optical basicity [9].

In Table 1 are shown the elements necessary to calculate the pB indicator for the main oxides which form the mineralogical constituents of the clinker in the systems CaO- SiO_2 and CaO- SiO_2 - Al_2O_3 - Fe_2O_3 (C - S - A - F).

In Table 1 the water was also mentioned, as various mineralogical constituents from the clinker interact to it. For the coordination number of the hydrogen related to the oxygen, value 1 is strictly correct in pure water. In fact, water contains a series of impurities, which implies an alteration of the coordination number of hydrogen to the value 2. The proportion of dicoordinate hydrogen can be determined by using spectrofotometric methods [10].

3. Correlations of free energy of formation - pB

In the oxide system CaO - SiO_2 (C - S) four

characteristic compounds are formed: C_3S, β-C_2S, C_3S_2, β-CS [2].

The thermodynamic properties of the compounds are determined by their chemical composition, the acid-basic condition of the formative oxides, the structural particularities of the reaction products, etc.

The attempts to correlate the enthalpy of formation ΔH_{298}^0 and the Gibbs free energy of formation ΔG_{298}^0 [11] with the chemical composition of the compounds - stated in % molars/gravimetrics CaO- have not been successful. The graphic representations indicated a low degree of correlation of the variables, difficult enough to be expressed in a quantitative relation.

A second level of correlation considered was the one referring to the thermodynamic property versus basicity. A first attempt in this direction was made by Pauling [12]. He established a quantitative relation between the enthalpy of formation of some simple substances and the electronegativity difference of the atoms forming simple chemical connections. Hence, given the quantitative relation between ionicity and the difference of electronegativity, it is practically established a correlation between the enthalpy of formation and basicity (for some molecular classes).

Surprisingly somehow, the extension of these interactions for the considered oxide compounds did not lead to cogent results. By calculating the basicity of the compounds with relations (7) and (8) it was shown in Figure 1 the dependence for ΔG_{298}^0 in proportion to pB, for the oxide compounds in system C-S.

The explanation appears from the fact that if X and Y represent two oxides featuring the reactants for the reaction product X_aY_b it can be shown that:

- the free energy of formation ΔG_{298}^0 (Gibbs), but also the enthalpy of formation ΔH_{298}^0, represents

Table 1

The calculus elements of the importance of basicity for oxides/ *Elemente de calcul a ponderii bazicității pentru oxizi*

Oxide *Oxidul*	Cation *Cationul*	The oxidation number of the cation *Cifra de oxidare a cationului*	P_i, eV	CN	pB, %
CaO	Ca	2	11.87	6	83.8
				8	88.4
SiO_2	Si	4	45.13	4	49.4
Al_2O_3	Al	3	28.44	4	61.6
				6	72.4
Fe_2O_3	Fe	3	31.69	4	59.1
				6	72.5
H_2O	H	1	13.6	1	38.7
				2	58.9
O^{-2}	-	-2	-6.5 - (7.37)	2	100 - (102.4)

Note. The values for P_i from Table 1 are taken from the paper [5], appendix 1 / *Valorile pentru P_i din tabelul 1 sunt preluate din lucrarea [5], anexa 1.*
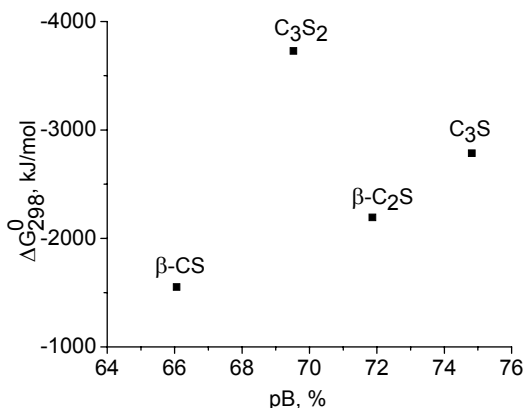
Fig. 1 - Variation ΔG_{298}^0 with pB for CaO - SiO$_2$ systems

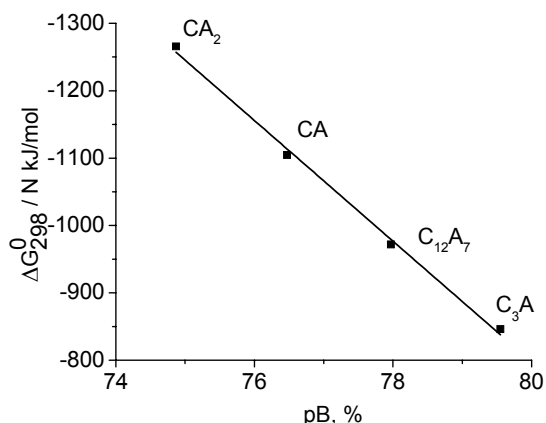*Variaţia energiei libere de formare cu ponderea bazicităţii în sistemul CaO - SiO$_2$.*

Fig. 3 - Variation ΔG_{298}^0/N with pB for CaO - Al$_2$O$_3$ systems

Variaţia energiei libere de formare specifică cu ponderea bazicităţii în sistemul CaO - SiO$_2$

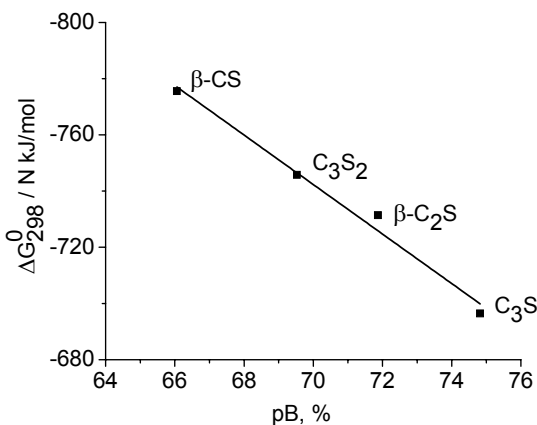$$\Delta G_{298}^0 /N = -7965.11 + 89.59 \, pB, R^2 = 0.999.$$

Fig. 2 - Variation ΔG_{298}^0/N with pB for CaO - SiO$_2$ systems

Variaţia energiei libere de formare specifică cu ponderea bazicităţii în sistemul CaO - SiO$_2$

$$\Delta G_{298}^0 /N = -1359.03 + 8.81, \, pB, R^2 = 0.978$$

Fig. 4 - Variation $\Delta^r G_{298}^0$/N with pB for CaO - SiO$_2$ systems

Variaţia energiei specifice de reacţie cu ponderea bazicităţii în sistemul CaO - SiO$_2$

extensive quantities in proportion to the number of X and Y oxides moles which form the compound X$_a$Y$_b$;

- the basicity percentage, pB, is an intensive quantity for the compound X$_a$Y$_b$, no matter the amount of the product involved in the reaction.

This is why the quantity ΔG_{298}^0 was transformed into an intensive capacity, called specific free energy of reaction. It is calculated by relating the quantity ΔG_{298}^0 to the number N = a + b of oxides moles composing the reaction product.

It should be noticed that, recently, in paper [13] it is highlighted a quantitative interdependence
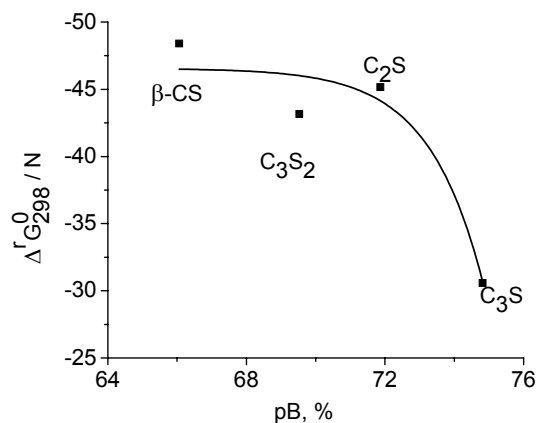
between the standard enthalpy of reaction related to the number of oxygens from an oxide compound and the optical basicity suggested by Duffy and Ingram [14].

The dependence specific free energy of formation, ΔG_{298}^0/N and pB is shown in Figure 2, by referring to the oxide system C-S. It can be noticed the existence of a high degree of correlation of the variables. This is also observed, for example, for the system CaO - Al$_2$O$_3$, according to the plotting from Figure 3.

In both cases, the specific free energy of formation declines linearly as the basicity of the compounds from the same system is bigger.

The same correlation was also analysed for

the specific energy of reaction $\Delta^r G^0_{298}/N$, according to the basicity pB (Figure 4).

The reaction energies, $\Delta^r G^0_{298}$, were calculated considering for the compounds from system C-S the following reactions [2]:

1. $CaO + SiO_2 \longrightarrow CaO \cdot SiO_2$
2. $3CaO + 2SiO_2 \longrightarrow 3CaO \cdot 2SiO_2$
3. $2CaO + SiO_2 \longrightarrow 2CaO \cdot SiO_2$
4. $3CaO + SiO_2 \longrightarrow 3CaO \cdot SiO_2$

Still, from Figure 4 it is obvious the fact that the variation of the specific energy of reaction is correlated to pB in a smaller proportion. The result seems amazing if taken into account the relation (4) and the data from Figure 2. Moreover, the same type of evaluations applied to systems M_2O - SiO_2, M = Li, Na, K, Rb, Cs [6] highlighted a high degree of correlation. In certain cases, the influence of other structural parameters not included in pB may interfere.

In the future the accumulation of new results will lead to a firm conclusion.

4. Correlations properties of mineralogical constituents - basicity

4.1. The reactivity of oxide compounds related to water. Correlations hydration degree - pB

Experimental data show that in the system CaO - SiO_2, the reactivity of the silicate compounds with water decreases within the series: C_3S > β-C_2S > C_3S_2 > β-CS.

In the literature, this result was explained in regard to the hybridization degree of the orbitals engaged in the chemical bonding from the compounds, the geometry of the coordination polyhedra, the electronegativity of the cation, the energy of the chemical bondings, etc. [2]

At the same time, interactions of the type water-oxide compound represent a chemical reaction between two compounds having a different basicity. In Table 2 it is displayed, for the series considered for the calcium silicates, the basicity evaluated by pB. In order to express the basicity, it was chosen the pB, which can be calculated with the relation (7), when knowing the electronegativity, the oxidation number, the coordination number and the ionization potential. For H_2O, in paper [10] it is shown that H atom can have coordination 1 in proportion to the oxygen. This action is strictly available for pure water which has pB = 38.7%. If the water contains impurities, hydrogen bridges - - - O — H— O- - - appear ,

when H has the coordination number 2; in this case, pB (H_2O) = 58.9% [10].

In Table 2 was mentioned the difference of basicity, ΔpB, between the silicate compound and water (taken with impurities). From the data presented, it is seen that the basicity of the compounds decreases continuously from C_3S to β-CS. In the same manner, the minimum value ΔpB also varies, the minimum value being proper to the compound CS. Thus, in a simple and uniform way, it is explained the reactivity of calcium silicates related to water.

These conclusions can be supported also from a structural point of view, with precise reference to the association degree of the structural entities of SiO_4 type. In this way, for more acid chains - C_3S_2 şi β-CS - a bigger association degree is appropriate. The structural chains are of Si_2O_7 type, Si_3O_9 rings, and $(SiO_3)_\infty$ chains respectively, for the forms α/β. Naturally, the extension of the structural polymerization degree of the compounds determines a deceleration of the reactions with water.

In this context, the correlation between the reactivity of the oxide compounds in proportion to water can be extrapolated also to the dependence of the hydration degree and basicity.

Calculating the pB for the mineralogical constituents of the Portland cement clinkers with relation (8) and knowing their hydration degree at various periods of time, [2] in Figures 7 - 10 it is indicated the interconnection degree of the two quantities. Durations of 3 - 7 - 28 - 90 days are taken into account. From Figures 7 - 10 it can be seen a very strong interdependence between the hydration degree and the basicity of the mineralogical constituents, indicated with pB. At relatively reduced periods of time, 3 - 7 days, the increase of pB implies a proportional augmentation of the hydration degree. This result is expected: the bigger the basicity of the compound, the higher the difference of basicities related to water. Hence, it results a stronger interaction, also more pronounced by the fact that impure water, more basic, is more reactive. At longer periods of time, (90 days, Figure 10), this interdependence is not available anymore. The explanation is given by the fact that, at longer periods of time, the hydration processes are almost finished. [2] With the exception of β-C_2S, for the rest of the mineralogical constituents, the hydration degree is situated in the interval 89 - 93% [2] and iB can no longer influence the process. This is a consequence of the fact that, from the four

Table 2

The difference of bacicities, ΔpB, between the silicate compound and water / *Diferenţa de bazicitate, ΔpB, între compusul silicatic şi apă*

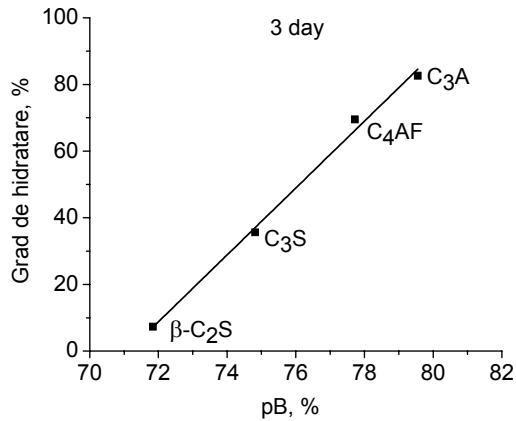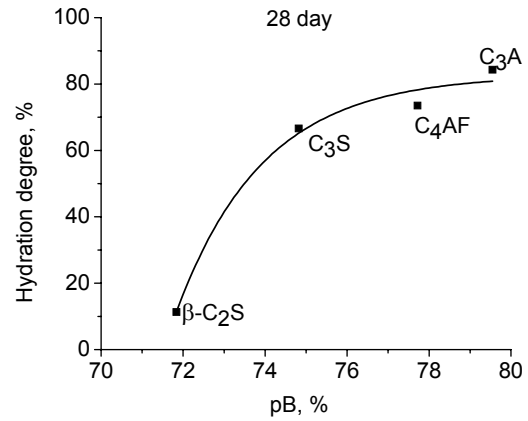| Silicate compound *Compusul silicatic*	C_3S	β-C_2S	C_3S_2	β-CS
pB, %	74.5	71.5	69.2	65.8
ΔpB, %	15.6	12.6	10.3	6.9

Fig. 5 - Variation of the 3 days hydration degree with the basicity percentage of the mineralogical constituents hydration degree / *Variaţia gradului de hidratare la 3 zile cu ponderea bazicităţii* = -713.8 + 10.03 pB, R^2 = 0.992

Fig. 7 - Variation of the 28 days hydration degree with the basicity percentage of the mineralogical constituents hydration degree / *Variaţia gradului de hidratare la 28 zile cu ponderea bazicităţii* =

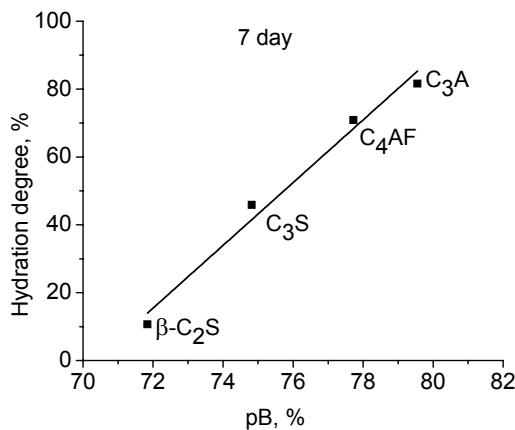$$82.73 - 3.32 \cdot 10^{16} \cdot e^{-0.47 \cdot pB}, R^2 = 0.965$$

Fig. 6 - Variation of the 7 days hydration degree with the basicity percentage of the mineralogical constituents hydration degree / *Variaţia gradului de hidratare la 7 zile cu ponderea bazicităţii* = -649.96 + 9.24 pB, R^2 = 0.974.
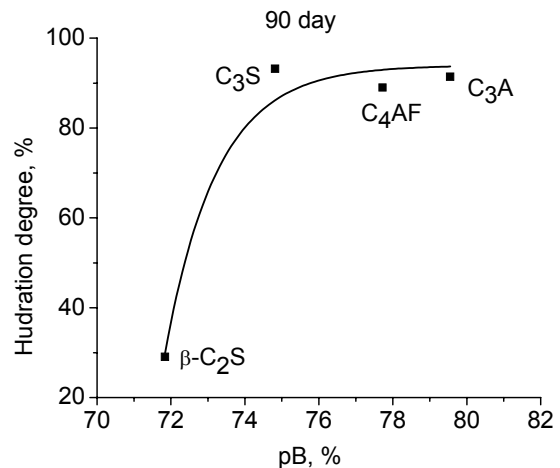

Fig. 8 - Variation of the 90 days hydration degree with the basicity percentage of the mineralogical constituents hydration degree / *Variaţia gradului de hidratare la 90 zile cu ponderea bazicităţii* =

$$93.94 - 8.29 \cdot 10^{23} \cdot e^{-0.70 \cdot pB}, R^2 = 0.927$$

mineralogical constituents, β-C_2S is the most acid. This way, its interaction with water, which has a more acid nature (no matter the shape), is smaller in proportion to the other oxide compounds (more basic).

Consequently, for β-C_2S, the most acid of the mineralogical constituents of the Portland cement clinkers, the reactivity in proportion to water is smaller. Hence, even after 90 days its hydration degree doesn't exceed 30% [2].

4.2. Correlations mechanical resistance - basicity to some oxide compounds

For some clinker types (cements) the mechanical compressive strengths are determined by the influence of some factors of intrinsic and extrinsic value, respectively. In the second category are included all the thermo technological

parameters associated to the genesis line (of processing), including environmental factors.

The intrinsic factors include a series of compositional and chemical-structural characteristics of the constituent oxide compounds. Thus, for Portland cement clinkers the main mineralogical constituents are C_3S, β-C_2S, C_3A and C_4AF. Their structural particularities determine a different behavior during the hydration - hydrolysis processes and those of consolidation. Finally, different levels of values are obtained for various properties of practical interest. For example, in Table 3 are presented the mechanical resistances to compression at various periods of time for the mineralogical constituents of the Portland cement clinker [2]. On this basis, in Figures 9 and 10 it is highlighted the dependence between the compressive strength (at 7, respec-

tively 28 days) and the basicity of compounds stated through pB. In the given charts it is underlined a dependence in the form of an inverted V, this being recaptured at the other periods as well.

The analysis of the results highlights the fact that C_3S represents the mineralogical compound which offers the clinkers bigger values of mechanical strength. It is an observation, confirmed by a multitude of experimental data, which reveals that C_3S, having an average value of basicity in proportion to the other constituents (in the series β-C_2S < C_3S < C_4AF < C_3A) determines big mechanical strength (both initial and final). According to table 2, C_3S also has the highest difference of basicity from all the mineralogical constituents in proportion to water.

At the same time, since β-C_2S is less basic than C_3S, it presents, as shown before, a weaker interaction with water, so a reduced hydration degree, implicitly a slower process of consolidation. Thus, for example, the energy of the Ca-O connection is bigger in β-C_2S than in C_3S; for the anhydrous silicates, this value declines as the proportion CaO/SiO_2, respectively, pB has bigger values.

Yet, although C_3A is described by a high rate of interaction with water and a huge release of heat during the exothermic hydration-hydrolysis processes, this has no important contribution to the values of mechanical strength. The explanation is given by the fact that C_3A, having the biggest value for pB (compared to the other mineralogical constituents of the Portland cement clinker), from a structural point of view presents a minimal polymerization degree, which doesn't favor the values of mechanical strength.

The explanation is also similar to the aluminate ferrite phase from the clinker (respectively C_4AF), which presents an activity in proportion to water, which declines in the series C_6A_2F - C_4AF - C_6AF_2 - CF_2, parallel to the diminution pf the pB values.

A V type variation is highlighted by the chart from Figure 11, where is shown the dependence of the contraction at 360 days for the mineralogical compounds of the Portland cement clinker, of their pB basicity. As expected, it is highlighted that C_3S presents a minimal contraction and C_3A a maximal one. As a matter of fact, the
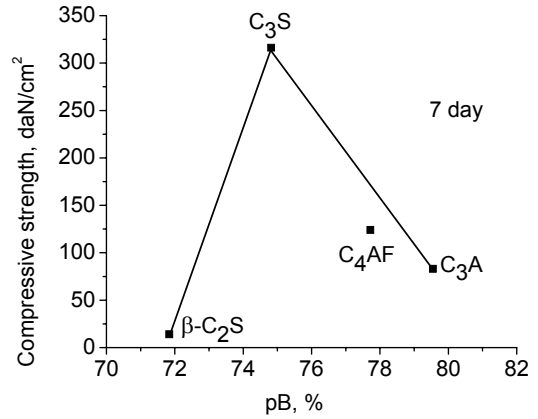

Fig. 9 - Variation of the 7 days compressive strength with the basicity percentage of the mineralogical constituents
Variația rezistenței la compresiune la 7 zile cu ponderea bazicității compușilor mineralogici
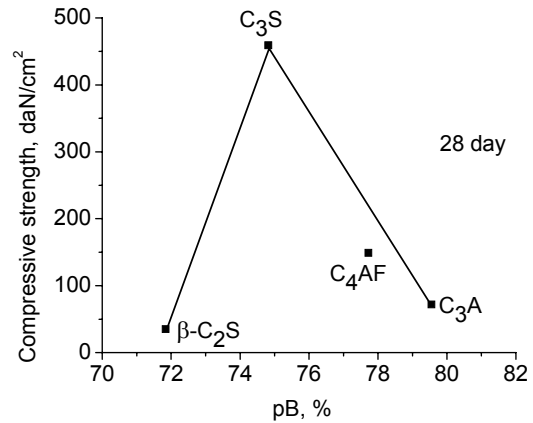

Fig. 10 - Variation of the 28 days compressive strength with the basicity percentage of the mineralogical constituents.
Variația rezistenței la compresiune la 28 zile cu ponderea bazicității compușilor mineralogici.

chart in Figure 11 is a mirror image of the chart of the mechanical strength type - pB (for example, see Figure 9).

Table 3

Compressive strength of some mineralogical compounds / *Rezistențele la compresiune a unor compuși mineralogici*

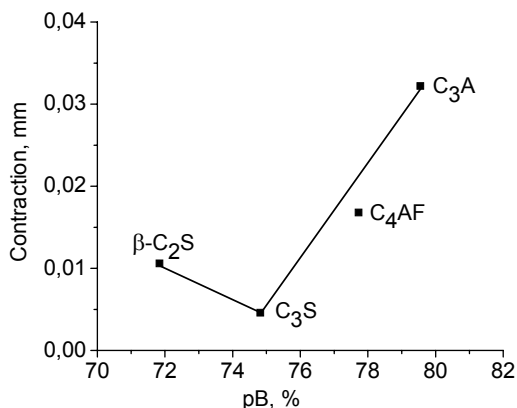| Mineralogical compounds *Compuși mineralogici*	Compressive strength, daN/cm², for / *Rezistența la compresiune, daN/cm², la*	
	7 day	28 day
C_3S	316	459
β-C_2S	14	35
C_3A	83	72
C_4AF	124	149

Fig. 11 - Variation of the 360 days contraction with the basicity percentage of the mineralogical constituents / *Variaţia contracţiei la 360 de zile cu bazicitatea compuşilor mineralogici.*

5. Conclusions

Present paper made possible the following conclusions:

- the existence of some relatively low correlations between the energy of formation of the compounds from the $CaO-SiO_2$ system and their basicity;

- the existence of a strong correlation between the Gibbs free energy of formation $\Delta G_{298}^0/N$ (which represents the proportion between $\Delta G_{298}^0/N$ and the number of moles from a compound) and the basicity of some oxide compounds from the system $CaO-SiO_2$;

- the dependence of some properties (the interaction degree with water, the hydration degree, compressive strength, the contraction) of some mineralogical constituents of the Portland cement clinker to their basicity (evaluated through the basicity percentage).

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