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THERMODYNAMIC MODELING OF THE FORMATION OF THE MAIN MINERALS OF CEMENT CLINKER AND ZINC FUMES IN THE PROCESSING OF TOXIC TECHNOGENIC WASTE OF THE METALLURGICAL INDUSTRY

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ABSTRACT

This paper presents the results of thermodynamic studies on the effect of temperature on the Gibbs energy of the formation of basic minerals of cement clinker (Ca_2SiO_4, Ca_3SiO_5, $Ca_3Al_2O_6$) with the simultaneous distillation of zinc (Zn_g) into the gas phase from mineral and man-made raw materials, in particular limestone and tailings from the enrichment of non-ferrous metals at the Balkhash processing plant. The research was carried out using the software package "HSC Chemistry 6", developed by the Outokumpu metallurgical company (Finland). During the conducted research it was established that the compounds of zinc oxide have an advantage in the process of formation under the influence of the temperature of the main clinker minerals on the zinc compounds in the sulphide form, for example, the forming temperature of the beginning of the reaction with the formation of Ca_2SiO_4 has virtually no difference, and when the formation mineral Ca_3SiO_5 forming temperature of the beginning of the reaction is 19,7% lower. And when The $Ca_3Al_2O_6$ mineral is formed, the formation temperature at the beginning of the reaction is lower by 17,5%, which is less energy-consuming.

Keywords: Thermodynamics, Gibbs Energy, Main Minerals of Cement Clinker, Zinc Fumes, Processing and Disposal of Toxic Technogenic Waste.

INTRODUCTION

At the present stage, calcium silicates are of great practical interest to the industrial industry. Main applications: production of cement and cement clinker, ceramics, refractory, and acid-resistant materials;

metallurgy; paint and varnish industry, dry building mixes. Every year they find their application in science, technology, and everyday life.[1] It is essential to reduce the consumption of fuel, heat, and electricity in the development of progressive science-based standards for their use, based on the introduction of the latest achievements of science and technology and modern technological solutions.[2-9] The solution to these problems in the production of silicate materials and products for various technical purposes makes the theoretical study of the processes underlying their production and operation more and more relevant. In this regard, the thermodynamic research method is of great importance, which allows not only to uniquely determine the energy parameters of the processes of silicate formation and synthesis of silicate materials but also, together with the study of the speed and mechanism of substance transfer, to obtain the necessary data on the rational management of technological processes for obtaining various technical silicates and similar substances.[1,10-13] The widespread use of computers and the introduction of mathematical and thermodynamic modeling methods in the practice of engineering and scientific research has led to the intensive development of many branches of science and technology, including the processes of clinker formation and sublimations of non-ferrous metals.[14-25] One of the most relevant areas that significantly expand the technological capabilities of clinker formation processes is the use of non-traditional sources of raw materials, which allows for high savings by reducing operations such as exploration of raw material deposits, development, and extraction of raw materials at the quarry, which helps to reduce the cost of clinker formation processes.[26-53]

Thus, research and scientific work aimed at reducing energy consumption and unit costs of raw materials, the involvement of man-made raw materials in the production cycle with a concomitant reduction of harmful effects on the flora, fauna and population of the region due to their disposal during the production of cement clinker and Zn sublimates are relevant. In this regard, we conducted thermodynamic studies of the effect of temperature on the Gibbs energy (ΔG_T^o) of the formation of the main minerals of cement clinker with simultaneous sublimation of zinc. In the simulation, zinc was used in sulfide and oxide forms.

EXPERIMENTAL

Thermodynamic studies were carried out using the HSC Chemistry 6 software package developed by the metallurgical company Outokumpu (Finland). The software package used in this work is based on the ideology of the European consortium SGTE (Scientific Group Thermodata Europe), which develops, maintains, and disseminates high-quality basic data. The SGTE structure is represented by research centers in Germany, Canada, France, Sweden, Great Britain, and the USA. The database of the software package contains information on 22000 individual substances.[19] To calculate the thermodynamic functions characterizing an individual substance, the standard values of enthalpy H_{298}, entropy S_{298}, and the coefficients of the polynomial A, B, C, and D, stored in the database, were used, from which the value of the molar heat capacity was calculated at an arbitrarily specified temperature T in accordance with the expression (1). The enthalpy of an individual substance at a temperature T, which differs from the standard one, equal to 298 K, was calculated by the formula:

$$H_T = H_{298} + \int_{298}^{T} C_p \, dT + \sum H_F \,, \quad (1)$$

Where H_{298} - is the value of the enthalpy of a given substance under standard conditions; C_p - molar heat capacity; $\sum H_F$ - enthalpy of phase transitions (polymorphic transformations, melting, evaporation). [1,2,19]

The entropy is defined as:

$$S_T = S_{298} + \int_{298}^{T} \frac{C_p}{T} dT + \frac{\sum H_F}{T} \,, \quad (2)$$

Where S_{298} - the value of the entropy of a given substance under standard conditions; C_p - molar heat capacity; $\frac{\sum H_F}{T}$ – entropy of phase transitions (polymorphic transformations, melting, evaporation). [1,2,19]

RESULTS AND DISCUSSION

As a result of the conducted thermodynamic studies, the Gibbs energy (ΔG_T^o) was calculated under the conditions of modeling the synthesis of the formation of the main minerals of cement clinker (Ca_2SiO_4, Ca_3SiO_5, $Ca_3Al_2O_6$) and zinc sublimates (Zn_g) in the temperature range of 500 - 1700°C from mineral and technogenic raw materials (limestone and tailings from non-ferrous metals), we considered the following chemical reactions:

$ZnS+2CaCO_3+SiO_2=Zn+Ca_2SiO_4+2CO+SO_2$; (3)

$ZnS+3CaCO_3+SiO_2=Zn+Ca_3SiO_5+3CO+SO_3$; (4)

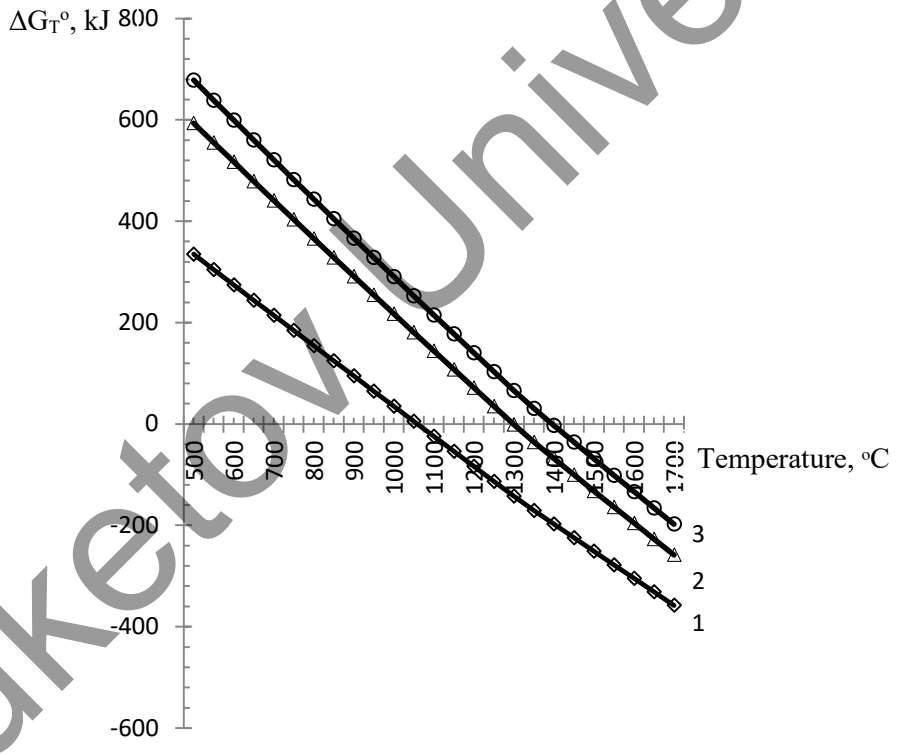
$ZnS+3CaCO_3+Al_2O_3=Zn+Ca_3Al_2O_6+3CO+SO_3$; (5)

$ZnO+2CaCO_3+SiO_2=Zn+Ca_2SiO_4+2CO_2+0,5O_2$; (6)

$ZnO+3CaCO_3+SiO_2=Zn+Ca_3SiO_5+3CO_2+0,5O_2$; (7)

$ZnO+3CaCO_3+Al_2O_3=Zn+Ca_3Al_2O_6+3CO_2+0,5O_2$; (8)

Based on calculations using the software complex «HSC Chemistry 6» of chemical reactions 3-8, the ΔG_T^o values of the formation of basic minerals and zinc sublimates were obtained, which theoretically can be seen from the above reactions. Based on the calculation results, we plotted the graphical dependences of the Gibbs energy on temperature. Figure-1 shows the dependence of Gibbs energy on temperature during the formation of the main minerals of cement clinker and zinc fumes in reactions 3-5. In these reactions, zinc is present as zinc sulfide (ZnS).

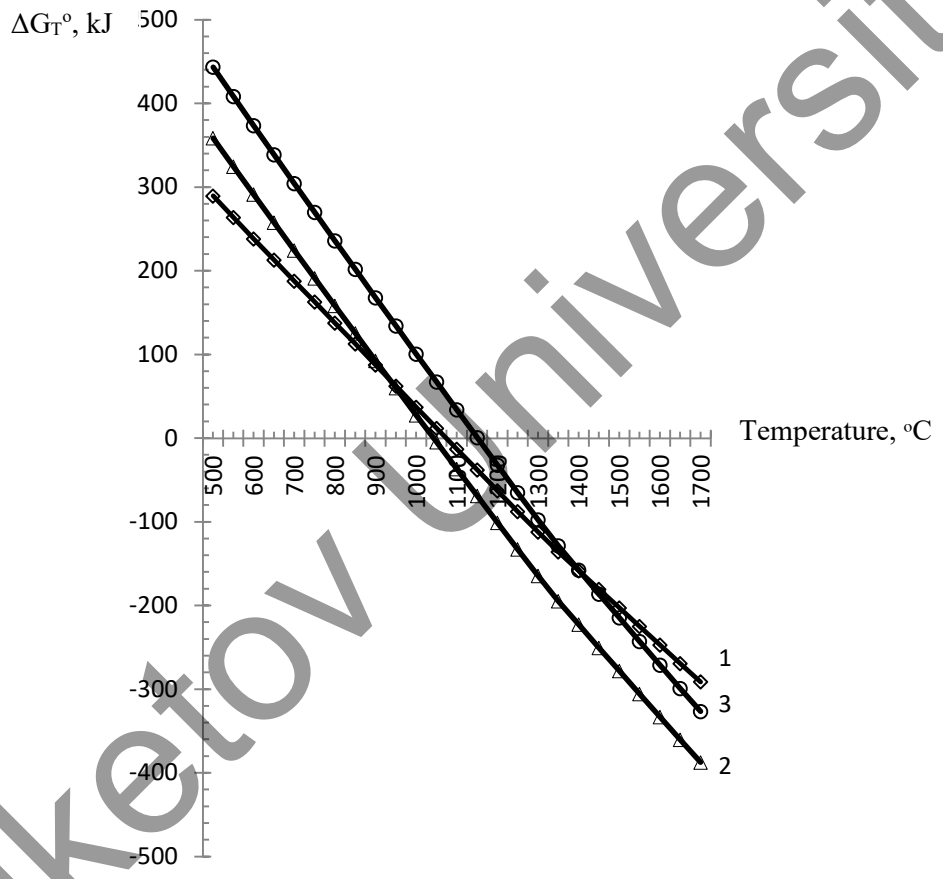


1- $ZnS+2CaCO_3+SiO_2=Zn+Ca_2SiO_4+2CO+SO_2$; 2- $ZnS+3CaCO_3+SiO_2=Zn+Ca_3SiO_5+3CO+SO_3$;
3- $ZnS+3CaCO_3+Al_2O_3=Zn+Ca_3Al_2O_6+3CO+SO_3$.

Fig.-1: Influence of Temperature on the Gibbs Energy of Formation of the Main Minerals of Cement Clinker in the Presence of Zinc Sulfide with Simultaneous Stripping of Zinc

As can be seen from Fig.-1, the distillation of zinc into the gas phase with the simultaneous formation of two calcium silicates is possible already at a temperature of 1059°C, at which the Gibbs energy reaches a negative value of -0,123kJ, which indicates the possibility of a reaction (3). The formation of three calcium silicates with the simultaneous distillation of zinc becomes possible at 1299°C when the Gibbs energy becomes negative -0,043 kJ, which favors the occurrence of reaction (4). The figure also shows the formation of three calcium aluminates with simultaneous sublimation of zinc, which becomes possible at a temperature of 1397°C with a negative Gibbs energy equal to -0,625 kJ, which indicates the

possibility of a reaction (5). Thus, it becomes obvious the possibility of obtaining the main minerals of cement clinker with simultaneous stripping of zinc into the gas phase for its further extraction and use; also from reactions 3-5 the possibility of obtaining SO_2-containing gases, which are an intermediate product for obtaining sulfuric acid, is visible. Moreover, the formation of the main minerals of cement clinker, depending on the onset of the reaction from temperature, is represented by the following series: Ca_2SiO_4>Ca_3SiO_5>$Ca_3Al_2O_6$ with a temperature range from 1059 to 1397°C. Figure-2 shows the dependence of the Gibbs energy on temperature during the formation of the main minerals of cement clinker and zinc fumes in reactions 6-8. In these reactions, zinc is present in the form of zinc oxide (ZnO). As can be seen from Fig.-2 the distillation of zinc into the gas phase with the simultaneous formation of two calcium silicates is possible already at a temperature of 1074°C, at which the Gibbs energy reaches a negative value of -0,022 kJ, which indicates the possibility of the reaction (6).



1- $ZnO+2CaCO_3+SiO_2=Zn+Ca_2SiO_4+2CO_2+0,5O_2$; 2- $ZnO+3CaCO_3+SiO_2=Zn+Ca_3SiO_5+3CO_2+0,5O_2$;
3- $ZnO+3CaCO_3+Al_2O_3=Zn+Ca_3Al_2O_6+3CO_2+0,5O_2$.

Fig.-2: Influence of Temperature on the Gibbs Energy of the Formation of the Main Minerals of Cement Clinker in the Presence of Zinc Oxide with Simultaneous Stripping of Zinc

The formation of three calcium silicates with the simultaneous distillation of zinc becomes possible at 1043°C when the value of the Gibbs energy becomes negative -0,067 kJ, which favors the occurrence of reaction (7). The figure also shows the formation of three calcium aluminates with the simultaneous sublimation of zinc, which becomes possible at a temperature of 1152°C with a negative Gibbs energy equal to -0,457 kJ, which indicates the possibility of the reaction (8). Thus, the possibility of obtaining the main minerals of cement clinker with simultaneous stripping of zinc into the gas phase for its further extraction and application is obvious. Moreover, the formation of the main minerals of cement clinker, depending on the start of the reaction on temperature, is represented by the following sequence:

Ca_3SiO_5>Ca_2SiO_4>$Ca_3Al_2O_6$ with a temperature range from 1043 to 1153°C. The obtained results have novelty and correspond to the scientific directions of the previously conducted research, complementing them.[54-61]

CONCLUSION

In the course of the studies carried out using thermodynamic modeling, it was found that:

- From mineral and man-made raw materials, in particular limestone and tailings of non-ferrous metals enrichment, it is possible to obtain the main minerals of cement clinker with simultaneous distillation of zinc into the gas phase, while reducing the anthropogenic impact of toxic waste on the population and the environment of the region;
- The effect of temperature on the Gibbs energy of the formation of the main minerals of cement clinker in the presence of zinc sulfide with simultaneous stripping of zinc and the production of SO_2 - containing gases, is represented by the following series of main minerals of cement clinker Ca_2SiO_4>Ca_3SiO_5>$Ca_3Al_2O_6$ with a temperature range from 1059 to 1397°C;
- The effect of temperature on the Gibbs energy of the formation of basic minerals of cement clinker in the presence of zinc oxide with simultaneous stripping of zinc is represented by the following series of basic minerals of cement clinker Ca_3SiO_5>Ca_2SiO_4>$Ca_3Al_2O_6$ with a temperature range from 1043 to 1153°C;
- In addition to the effect of temperature on the Gibbs energy of the formation of the main minerals of cement clinker and the simultaneous stripping of zinc, the process is also influenced by the form of the initial zinc in the form of a sulfide and oxide compound;
- Zinc compounds in the form of oxides have an advantage in the process of formation under the influence of the temperature of the main clinker minerals over zinc compounds in the sulfide form, for example, the temperature of the formation at the beginning of the reaction with the formation of Ca_2SiO_4 practically does not differ, and during the formation of the mineral Ca_3SiO_5 the temperature of formation of the beginning of the reaction 19,7% lower. And during the formation of the $Ca_3Al_2O_6$ mineral, the formation temperature of the onset of the reaction is 17,5% lower, which is less energy-consuming.

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