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Thermodynamic modeling of the synthesis of the main minerals of cement clinker from technogenic raw materials

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ABSTRACT

In this article, thermodynamic studies of modeling the formation of the main minerals of cement clinker as a function of the Gibbs energy (ΔG) on temperature were investigated. The temperature range of the studies was 873-1873 K. In the course of modeling studies, the reactions of formation of cement clinker minerals (C_2S - belite, C_3S - alite, C_3A – three calcium aluminates, C_4AF – four calcium aluminium ferrite) from standard raw materials and non – standard-technogenic raw materials containing non-ferrous metals were calculated. Equations describing the dependence of the Gibbs energy on temperature with approximation coefficients (R^2) equal to 0.99 are found. According to the results of the conducted studies, the beginning (Tb) temperatures of the course of the compared clinker formation reactions were determined, which were 1008 K for the standard reaction, and 1023 K for the non-standard reaction. It was found that the reactions begin to occur in the temperature intervals for the standard (1008 K – 1873 K), for the non - standard (1023 K - 1873 K). At the same time, the values at the initial stage of the course of the standard reaction have a more negative value of the Gibbs energy than the non-standard reaction. With a further increase in temperature, it becomes evident that the Gibbs energy of the non-standard reaction of mineral formation in the temperature range 1173-1873K becomes more negative in comparison with the values of the standard reaction and exceeds its values from 9 to 19.01%.

Keywords: Technogenic raw materials, processing waste, thermodynamics, Gibbs energy, cement clinker minerals, non-ferrous metals.

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Introduction

Over the long-term period of intensive development of all industries in Kazakhstan, including the mining and metallurgical complex (MMC), more than 26 billion tons of solid production waste have accumulated, which is annually replenished in dumps by another 1 billion tons. Most of them (58%, or 15,1 billion tons) are waste from the mining and metallurgical industries, considered an independent raw material base. In non-ferrous metallurgy (copper-aluminium, lead-zinc, gold-rare-metal industries), the total amount of waste reaches more than 5 billion tons, including rocks of associated mining and overburden - 72%, tailings of enrichment - 26 and metallurgical processing – 1,6 %. The area of land occupied by waste is more than 13 thousand hectares. The total amount of accumulated waste in the ferrous metallurgy of Kazakhstan (iron ore, chromium ore and manganese ore industries) is more than 6,2 billion tons, of which: associated mining and overburden — 92,8 %, enrichment-6,1 and metallurgical processing-1,1 %. The area of land occupied by waste is more than 15 thousand hectares [1].

The reasons for the formation of technogenic waste are the deterioration of the technological quality of ores, a significant lag in the technologies of extraction, processing and metallurgical processing from the changing characteristics of ore raw materials, an increase in the loss of mineral carriers of metals [2-10].

Society's consumption of natural resources is becoming an acute problem due to the quantitative growth of their use, which leads to environmental and other problems. The fact is that the country's MMC is special in terms of resources, since it is characterized by an increased level of resource consumption, which naturally increases as mining operations decrease, as well as due to the deterioration of the quality of minerals, mining-geological and mining-technical conditions for the development of deposits [1-15].

In Kazakhstan, the problem of increasing the resource productivity of MMC and its environmental safety has not yet been given due attention, because of which the traditional low level of productivity of the use of basic production resources and environmental safety provides reason to consider mining and metallurgical production as a whole as a factor hindering the overall economic and social development of the country [2, 15-27].

Solving the problem of increasing the resource productivity of MMC production and preserving the environment is becoming increasingly important. It will only increase over time, especially due to the increasing scarcity of natural resources as their available sources are exhausted. In accelerated scientific and technological progress, one of the main issues of economic policy is to ensure high production efficiency. With the ever-increasing shortage of natural resources, a special role is played by the rational, integrated and economical use of all types of raw materials. Therefore, in recent years, the recovery of raw materials from mining waste and their integrated use is a matter of national importance [3, 9-27].

The level of involvement of human-made industrial waste as secondary raw materials is frankly low, namely: in the context of ferrous metallurgy - 3,3%, in the context of the coal industry - 5,2% and in the context of non-ferrous metallurgy – 5,6%, which is a very low indicator concerning the developed economies of the world [3-28].

Most developed foreign countries have long practiced saving their mineral resources, intensively involving man-made deposits in processing, disposing of production waste, and developing technologies for processing this waste. For example, in the United States back in 1993, the share of secondary raw materials in the production of non-ferrous metals was: copper - 55 %, tungsten - 28 %, nickel - 25 %. A similar trend in secondary resources is observed in Canada, the United Kingdom, South Africa, Spain, and other countries [29].

In Canada, 40% of copper is recovered from copper mining waste containing 0,45% copper, thanks to new enrichment methods (heap acid leaching, heap pyrite and bacterial leaching). In the US, in the state of Montana, 2 tons of gold and 4 tons of silver are extracted annually from the Mandiski mine dumps containing 0,84 g/t of gold and 2,8 g/t of silver. In the state of Michigan, 60% of copper is extracted from the tailings of the enrichment (copper content – 0,3%). In Bulgaria, copper concentrate is obtained from waste containing 0,1 - 0,15% copper, the cost of which is 3 times lower than when it is obtained from natural raw materials. In the Republic of South Africa, 3,5 tons of gold and 696 tons of uranium per year are obtained from the dumps of gold recovery factories with a gold content of 0,53 g/t and 40 g/t uranium [29].

As the reserves of the developed fields are exhausted, human-made objects may become a priority. In some cases, the only source of mineral raw materials for numerous mining and metallurgical enterprises. At the same time, it should be borne in mind that the waste of mining and industrial production, representing a large reserve of raw materials for the extraction of metals and non-metals, at the same time are hotbeds of local or regional environmental pollution. The waste mass accumulated in dumps and tailings dumps with an average layer thickness of 20 m covers more than 1300 km^2. The annual increase in the area of alienated land is at least 85-90 km^2. The negative impact on the environment is manifested in an area that is 10 times or larger than the area occupied by waste. A significant part of the alienated land is located in industrially developed areas, often within the borders of settlements and large cities [29].

One of such multi-tonnage wastes of technogenic nature is the tailings from the enrichment of non-ferrous metal ores of the Balkhash processing plant located near the city of Balkhash (Kazakhstan), which during the economic activity of the processing plant for decades hurt the natural environment of the region. And from an ecological point of view, they are a hotbed of pollution of the surrounding natural environment, particularly the soil cover, the air basin, surface, and underground waters containing non-ferrous metals in their composition.

The Balkhash tailings storage facility is of particulary great interest, as it is one of the oldest in the industry and one of the largest tailings storage facilities in Kazakhstan. In addition, it is not located in a simple, arid zone, which significantly increases the impact of the tailing dump on the environment [1-3,15-29], it is in significant proximity to Lake Balkhash, which has a large, even enormous economic purpose [15-29].

The Balkhash processing plant, which owns the tailings storage facility, enriches copper-molybdenum ores from the Kounrad and Sayak deposits. During the day, the factory processes an average of 33,7 thousand tons of Kounrad and 9,5 thousand tons of Sayak ores. The enrichment waste is stored in the tailings storage facility.

In Kazakhstan, where 360-390 thousand tons of refined copper are produced annually [1-3, 30], the amount of copper ore flotation waste is growing by 1,8-2,5 million tons per year.

The result of many years of operation of the processing plant, which processes various ores, including copper-zinc ores, is the formation of production waste on the earth's surface on the territory of the enterprise or near it. Such formations are a relatively low content of non-ferrous metals and a decrease in the quality of processed ores. Natural leaching of stored formations [1-30].

In connection with the urgent problem of the need to dispose of waste from the Balkhash processing plant, based on the chemical composition of tailings from processing as secondary raw materials, they contain such valuable compounds and elements as SiO_2, Al_2O_3, CaO, Fe_2O_3, Cu, Zn; thus, they may be of interest to the silicate and metallurgical industries as aluminium - silicate - iron-containing raw materials and non-ferrous metals, respectively. Therefore, the tailings from the Balkhash processing plant are considered by us as secondary raw materials for their utilization by the method of complex processing as a raw material component in the high-temperature synthesis of cement clinker.

Study object, research methods, and calculation methods

Thermodynamic studies were carried out using the software package "HSC Chemistry 6.1", 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 is engaged in the development, support and distribution of high-quality databases. The SGTE structure is represented by specialized research centres in Germany, Canada, France, Sweden, the United Kingdom, and the United States. The database of the software package contains information about 22,000 individual substances [31].

To calculate the thermodynamic functions that characterize the individual, the substance used is stored in the database of the standard values of enthalpy H298, S298 entropy, and the polynomial coefficients A, B, C, D, by which the calculated value of the molar heat capacity at arbitrarily given temperature T according to equation (1).

Individual enthalpy of a substance at the temperature T, different from the standard, equal to 298 K, was calculated by the formula:

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

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

The entropy value is defined as:

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

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

Table 1 - Calculation of the Gibbs energy of the reaction $12CaCO_3+2SiO_2+2Al_2O_3+Fe_2O_3= =Ca_3SiO_5+Ca_2SiO_4+Ca_3Al_2O_6+Ca_4Al_2Fe_2O_{10}+12CO_2\uparrow$

T, K	*deltaH, kJ*	*deltaS, J/K*	*deltaG, kJ*	*K*	*Log(K)*
873.000	1447.213	1436.921	192.781	2.913E-012	-11.536
973.000	1379.154	1363.127	52.831	1.457E-003	-2.836
1073.000	1312.193	1297.626	-80.159	7.990E+003	3.903
1173.000	1253.513	1245.225	-207.136	1.678E+009	9.225
1273.000	1182.795	1187.380	-328.740	3.092E+013	13.490
1373.000	1110.083	1132.404	-444.708	8.317E+016	16.920
1473.000	1035.284	1079.827	-555.301	4.936E+019	19.693
1573.000	958.322	1029.283	-660.741	8.772E+021	21.943
1673.000	433.018	702.365	-742.039	1.479E+023	23.170
1773.000	347.830	652.963	-809.873	7.275E+023	23.862
1873.000	247.568	597.956	-872.403	2.147E+024	24,332

Results and its discussion

Having considered the chemical composition of the tailings from the enrichment, which according to various data [29, 30, 32,33] contains an average of SiO_2 - 53%, Al_2O_3 – 13,1%, CaO – 8,3%, Fe_2O_3 – 8,2%, MgO-0,55%, as well as residual non-ferrous metals CuS - 0,8%, ZnO - 0,5%. From which it follows that the tailings from the enrichment of non-ferrous metal ores may be considered as a secondary mineral raw material for the cement industry as an aluminum-silicate component of the raw material mixture during synthesis cement clinker. Thus, we have modeled the conditions for the formation of the main minerals of cement clinker (Ca_3SiO_5 - alite, Ca_2SiO_4 - belite, $Ca_3Al_2O_6$ - tricalcium aluminate, $Ca_4Al_2Fe_2O_{10}$ - four-calcium aluminoferrite) in the classic form. The standard chemical reaction was used as the basis of our research for comparison:

$$12CaCO_3+2SiO_2+2Al_2O_3+Fe_2O_3=Ca_3SiO_5+Ca_2SiO_4+Ca_3Al_2O_6+Ca_4Al_2Fe_2O_{10}+12CO_2\uparrow \quad (3)$$

The results of the thermodynamic calculation of the possibility of the standard reaction (3) are shown in Table 1.

Based on the results of the calculation of the Gibbs energy (ΔGTo) of the reaction (3), shown in Table 1, it can be seen that the reaction is limited and begins to flow at T=1073K, reaching a Gibbs energy value of -80,159 kJ and with increasing temperature becomes more negative reaching a value of -872,403 KJ at T=1873K. Based on the results of the obtained values of the Gibbs energy as a function of temperature, we we found an equation describing the dependence of the Gibbs energy on the temperature of the reaction (3) and were able to determine the temperature of the beginning (Tb) of the reaction (3). Thus, the dependence of the Gibbs energy on the reaction temperature (3) is described by the following equation (and has an approximation coefficient R^2=0,99):

$$\Delta G_T^o = -1437\ln(T) + 9933.6 \quad (4)$$

And the temperature of the beginning of the reaction (Tb) according to equation (4) was 1008K.

Based on the chemical composition of the tailings from the enrichment of non-ferrous metal ores, we considered the following non-standard reaction to obtain information on the formation of the main minerals of cement clinker with simultaneous distillation of zinc into the gas phase:

$$12CaCO_3+2SiO_2+2Al_2O_3+Fe_2O_3+MgO+ZnS+2CuO=Ca_3SiO_5+Ca_2SiO_4+Ca_3Al_2O_6+Ca_4Al_2Fe_2O_{10}+MgO+Zn\uparrow+2Cu+12CO_2\uparrow+SO_2\uparrow \quad (5)$$

The calculation of the Gibbs energy (ΔGT°) of the reaction (5) ais shown in Table 2. Table 2 shows that the non-standard reaction (5) is limited and begins to flow at T=1073K, reaching a Gibbs energy value of -71.262 kJ. With increasing temperature becomes more negative getting a value of -1077,296 kJ at T=1873K.

Based on the results of the obtained values of the Gibbs energy as a function of temperature, we

Table 2 - Calculation of the Gibbs energy of the reaction $12CaCO_3+2SiO_2+2Al_2O_3+Fe_2O_3+MgO+ZnS+2CuO=$ $=Ca_3SiO_5+Ca_2SiO_4+Ca_3Al_2O_6+Ca_4Al_2Fe_2O_{10}+ +MgO+Zn\uparrow+2Cu+12CO_2\uparrow+SO_2\uparrow$.

T. K	*deltaH. kJ*	*deltaS. J/K*	*deltaG. kJ*	*K*	*Log(K)*
873.000	1779.155	1738.591	261.364	2.293E-016	-15.640
973.000	1708.063	1661.508	91.415	1.236E-005	-4.908
1073.000	1638.050	1593.022	-71.262	2.947E+003	3.469
1173.000	1576.335	1537.916	-227.641	1.374E+010	10.138
1273.000	1502.639	1477.635	-378.390	3.370E+015	15.528
1373.000	1453.366	1439.856	-523.556	8.316E+019	19.920
1473.000	1375.424	1385.070	-664.784	3.768E+023	23.576
1573.000	1196.041	1266.296	-795.842	2.690E+026	26.430
1673.000	665.874	936.380	-900.690	1.330E+028	28.124
1773.000	575.810	884.147	-991.782	1.665E+029	29.222
1873.000	470.661	826.459	-1077.296	1.113E+030	30.046

found an equation describing the dependence of the Gibbs energy on the temperature of the reaction (5) and were able to determine the temperature of the beginning (Tb) of the reaction (5). Thus, the dependence of the Gibbs energy on the temperature of the reaction (5) is described by the following equation (and has an approximation coefficient R^2=0,99):

$$\Delta G_T^o = -1801\ln(T) + 12484 \qquad (6)$$

And the temperature of the beginning of the reaction (Tb) according to equation (5) was 1023K.

A comparison of reaction (3) with reaction (5) shows that new reaction participants have appeared in reaction (5), in particular magnesium oxide, zinc sulfide, and copper oxide. At the same time, the course of these reactions in the temperature range of 873-1873 K is limited. Based on the results of the calculations in Tables 1 and 2, their flow is possible in the temperature range of 1073-1873K when negative values of ΔG are reached and is almost identical at the beginning temperature (Tb) of the reaction (3 and 5), where Tb was 1008K and 1023K, respectively, which is not a significant difference in high-temperature synthesis. At a temperature of 1073 K, the reaction ΔG (3) has a value of -80,159 kJ, and the reaction ΔG (5) has a less negative value of -71,262 kJ, which indicates a slight advantage of reaction (3) overreaction (5). However, with a further increase in temperature from 1173K to 1873K, the ΔG of the reaction (5) begins to have more negative values (from 227,641 kJ to -1077,296 kJ, respectively) than the values of the ΔG of the reaction (3), from 207,136 kJ to -872,403 kJ, respectively. This indicates the intensification of the reaction (3) due to the presence of zinc compounds, which contributes as a mineralizer not only to the intensification of the clinker formation reaction [34-46], but also to a decrease in the formation temperature of clinker minerals by approximately 100-120 K, which in the future when synthesizing cement clinker minerals, will help to save energy of the process.

Conclusions

Thus, based on the conducted research, we can draw the following conclusions:

- reaction $12CaCO_3+2SiO_2+2Al_2O_3+Fe_2O_3=$ $=Ca_3SiO_5+Ca_2SiO_4+Ca_3Al_2O_6+Ca_4Al_2Fe_2O_{10}+12CO_2\uparrow$ is limited and begins to occur at T=1073K, at a Gibbs energy value of -80,159 kJ and reaches a value of ΔG -872,403KJ at T=1873K;
- Tb of the reaction $12CaCO_3+2SiO_2+2Al_2O_3+$ $+Fe_2O_3=Ca_3SiO_5+Ca_2SiO_4+Ca_3Al_2O_6+Ca_4Al_2Fe_2O_{10}+$ $+12CO_2\uparrow$ was 1008K and is described by the equation $\Delta G_T^o = -1437\ln(T) + 9933,6$;
- reaction $12CaCO_3+2SiO_2+2Al_2O_3+Fe_2O_3+MgO+$ $+ZnS+2CuO=Ca_3SiO_5+Ca_2SiO_4+Ca_3Al_2O_6+$ $+Ca_4Al_2Fe_2O_{10}+MgO+Zn\uparrow+2Cu+12CO_2\uparrow+SO_2\uparrow$ in the studied temperature range is limited and begins to flow at T=1073K, having a Gibbs energy of -71.262 kJ with an increase in the Gibbs energy to 1077,296 kJ at 1873K;
- Tb of the reaction $12CaCO_3+2SiO_2+2Al_2O_3+$ $+Fe_2O_3+MgO+ZnS+2CuO=Ca_3SiO_5+Ca_2SiO_4+Ca_3Al_2O_6$ $+Ca_4Al_2Fe_2O_{10}+MgO+Zn\uparrow+2Cu+12CO_2\uparrow+SO_2\uparrow$ was

1023K and is described by the following equation $\Delta G_T^o = -1801\ln(T) + 12484$;

- according to the reaction (5), it is theoretically possible to form the main minerals of cement clinker (similar to reaction 3), to distil zinc and sulfur-containing gases into the gas phase, for their further capture and processing.

- reaction (5) proceeds more intensively than reaction (3) in the temperature range 1173-1873K due to the participation of zinc in the reaction as an intensifier, which also contributes to a decrease in the process temperature by 100-120K compared to the standard reaction (3);

- at the initial stage of the flow at T=1073K, the reaction (3) has a more negative value of the Gibbs energy than the reaction (5), but with increasing temperature, it becomes obvious that the Gibbs energy of the reaction (5) in the temperature range 1173-1873K becomes more negative compared to the values of the reaction (3) and exceeds its values from 9 to 19,01%.

Conflicts of interest. On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Техногенді шикізаттардан цемент клинкерінің негізгі минералдарын синтездеуді термодинамикалық модельдеу

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ТҮЙІНДЕМЕ

Мақалада Гиббс энергиясының (ΔG) температураға байланысты цемент клинкерінің негізгі минералдарының түзілуін модельдеуге термодинамикалық зерттеулер жүргізілді. Зерттеуді модельдеу барысында стандартты шикізаттан және құрамында түсті металдар бар стандартты емес - техногенді шикізаттан цемент клинкерінің (C_2S - белит, C_3S – алит, C_3A – үш кальций алюминаты, C_4AF – төрт кальций алюмоферрит) негізгі минералдарының түзілу реакцияларына есептеу жүргізілді. Модельдеу барысында Гиббс энергиясының апроксимация коэффициенттері (R^2) 0,99-ға тең температураға тәуелділігін сипаттайтын теңдеулер табылды. Жүргізілген зерттеулердің нәтижелері бойынша клинкер түзілу реакцияларының басталу температурасы (Тб) анықталды. Негізгі стандартты реакция үшін (стандартты шикізатты қолдана отырып) 1008 К, ал стандартты емес реакция үшін (байыту қалдықтарын қолдана отырып) 1023 К құрады. Сондай-ақ, стандартты реакциялар (1008К – 1873К), стандартты емес (1023 К - 1873 К) реакциялар темпетатуралық интервалдарда басталатыны анықталды. Сонымен қатар, стандартты реакцияның бастапқы кезеңіндегі Гиббс энергиясының мәні стандартты емес реакцияға қарағанда теріс мәнге ие болады. Температура одан әрі жоғарылағанда 1173-1873 К температуралық интервалдағы стандартты емес минерал түзілу реакциясының Гиббс энергиясы, стандартты реакция мәндерімен салыстырғанда теріс болып, оның мәндері 9-дан - 19,01% - ға дейін асатыны айқын болады.

Түйін сөздер: техногенді шикізат, байыту қалдықтары, термодинамика, Гиббс энергиясы, цемент клинкерінің минералдары, түсті металдар.

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Термодинамическое моделирование синтеза основных минералов цементного клинкера из техногенного сырья

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АННОТАЦИЯ

В настоящей статье проведены термодиначические исследования моделирования образования основных минералов цементного клинкера в зависимости энергии Гиббса (ΔG) от температуры. Температурный интервал исследований составил 873-1873 К. В ходе моделирования исследований проведен расчет реакций образования основных минералов цементного клинкера (C_2S - белита, C_3S - алита, C_3A – трех кальциевого алюмината, C_4AF – четырех кальциевого алюмо феррита) из стандартного сырья и нестандартного – техногенного сырья, содержащего цветные металлы. В ходе моделирования найдены уравнения, описывающие зависимость энергии Гиббса от температуры с коэффициентами апроксимации (R^2) равными 0,99. По результатам проведенных исследований были определены температуры начала (Тн) протекания сравниваемых реакций клинкерообразования, которые составили для стандартной реакции (с использованием стандартного сырья) 1008 К, а для нестандартной (с использованием отходов обогащения) 1023 К. Установлено что реакции начинают протекать в темпетатурных интервалах для стандартной (1008 К – 1873 К), для нестандартной (1023 К - 1873 К). При этом значения энергии Гиббса на начальном этапе протекания стандартной реакции обладают более отрицательным значением чем нестандартная реакция. С дальнейшим увеличением температуры, становится очевидным, что энергия Гиббса нестандартной реакции менералообразования в температурном интервале 1173-1873К становится более отрицательной по сравнению со значениями стандартной реакции и превышает ее значения от 9 до 19,01%.

Ключевые слова: техногенное сырье, отходы обогащения, термодинамика, энергия Гиббса, минералы цементного клинкера, цветные металлы.

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