

Analysis of Non-Ferrous Metallurgy Waste Processing Capacity

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Abstract— Numerous non-ferrous metallurgy wastes are known (dumps, slags, sludge, dust, clinker, etc.), which, from an economic as well as an environmental point of view, are beneficial and must be disposed of. This book provides an analysis of known methods and proposes new solutions for the autonomous and joint processing of zinc production cake and molybdenum cake of the leaching plant of OJSC Almalyk MMC, which are technogenic deposits of unique polymetallic raw materials, which are practically not used to date. The relevance and novelty of the problem of processing such deposits in the steady growth of their volumes and the lack of effective processing technology. These deposits differ from natural mineral raw materials in that they are a product of technological processing and are composed of new mineralogical formations, more or less persistent and difficult to process. Operating enterprises and technologies often turn out to be unsuitable for waste processing and require reconstruction, modernization or re-equipment based on new progressive technological solutions. The solutions proposed by the authors make it possible to selectively extract copper, zinc, iron, gold and silver from the specified raw materials and define the technology as environmental protection and diversification.

Keywords— metallurgy, man-made waste, zinc, aluminum, molybdenum, acid, processing, thermodynamic changes.

1. INTRODUCTION

Scientific and technological progress in the modern world is accompanied by a sharp increase in the consumption of natural resources and a simultaneous increase in the amount of industrial waste, the problem of rational use of which is closely related to the efficiency of industrial production, environmental protection and new developments in the field of waste disposal [1-3].

The technologies used for waste disposal in developed countries are 90-98% focused on their disposal to landfills and tailing dumps, incineration in utilization power plants or inefficient use at existing metallurgical enterprises, the main disadvantage of which is dust and gas emissions and the associated losses of valuable elements and etc. In addition, landfills and tailing dumps require the allotment of significant land plots and violate the environmental situation in the adjacent areas [4-8].

Uzbekistan has confidently embarked on the path of searching, developing, improving and introducing technologies for processing mineral and technogenic secondary raw materials. The Almalyk Mining and Metallurgical Combine (AMMC) is the pearl of the country [1], convinced from his own experience that market relations call for initiative, reasonable risk, and the introduction of new developments, he chose for himself, incl. one of the activities is the involvement of industrial waste (dust from copper-smelting production, tailings of a copper processing plant, slags, clinker, etc.) into processing. Hundreds of thousands and tens of millions of tons of this and other wastes are generated and accumulated at the plant: in the dumps of only zinc production (stale clinker), the reserves of zinc are estimated at more than 10 thousand tons, copper over 9 thousand tons, iron over 150 thousand tons, as well as industrial quantities of precious metals (about 2 tons of gold and over 200 tons of silver) [9-14].

The dumps continue to grow: hundreds of thousands of tons of clinker are produced annually, and only a small part of it is processed according to the currently basic technology of reflective smelting of copper smelting production to deplete the slag melts with copper. At the same time, noble metals and copper are concentrated in the matte, a significant part of the zinc sublimes with the dust and gas flow, and the iron passes into slag. Formation of slag dumps, for which an acceptable utilization technology has not yet been found; dust and gas emissions requiring capture and cleaning, as well as the loss of zinc (partly with dust) and iron (completely with slags) require further research in the direction of searching and developing more advanced science-intensive technologies for processing clinker, which is considered as a polymetallic industrial raw material. World practice undoubtedly shows that the processing of such raw materials is economically more profitable if the principle of waste-free processing is observed [15-20].

Preliminary calculations based on experimental laboratory data confirm the economic and environmental efficiency of autonomous processing of clinker with the release of copper and zinc, as well as iron and precious metals. Given the extreme tenacity of clinker, this task seems to be not an easy one, requiring knowledge of mineralogy, chemistry and technology of behavior and processing of individual valuable components of clinker. Of particular scientific and applied interest in solving this problem is the development of a rational technology for processing clinker by hydrometallurgical methods: oxidative catalytic processes, autoclave methods, methods using plasma chemistry, ozone, ultrafine grinding of raw materials, etc. [21-23].

One of the problems in copper smelting is the capture, treatment and disposal of dust and gas emissions. Thus, during the processing of sulfide copper concentrates by the method of oxygen-flame smelting (OCP) at the AMMC, the process of oxidation of sulfide minerals into oxides is realized, which is carried out due to the exothermic reaction of the combustion of sulfide sulfur of raw materials in an oxygen atmosphere; moreover, the thermal regime of the furnace is characterized by an excess of heat against

the required amount according to the regulations of the melting process and is accompanied by the release of dust. To regulate the process temperature in practice, recycled dust is used (factories "Asko", "Toyo", "Tamanno" in Japan, "Koper-Cliff" in Canada, "Harjavatta" in Finland, Almalyk copper smelter in Uzbekistan), which is captured in boilers - utilizers and dry electrostatic precipitators from the dust and gas flows (DGF) leaving the furnace and through the pneumatic transport system are returned to the head of the process. Dusts captured in electrostatic precipitators mainly contain metal sulphates, which violate the autogenousness of melting and contribute to a decrease in the temperature of the process, that is, they eliminate the problem of excess heat in the melting furnace. Therefore, the dusts are often recycled by wrapping them in a melting furnace. Meanwhile, plants for autonomous processing of dust from copper-smelting industries in the Russian Federation (Kirovograd), in the USA (Wyoming), in Japan have been built and are successfully operating, where the idea of using only part of the dust as recycled dust has been implemented, and the rest is processed autonomously and shows high technical economic indicators. At the copper-smelting plant of OJSC Almalyk MMC, the topic of withdrawing a part of fine dust from circulation and their autonomous processing is still not considered. There are still no practical recommendations for the processing of fine dust from electrostatic precipitators at the AMMC copper smelter [24-44].

2. ANALYSIS OF EXISTING TECHNOLOGIES

When processing zinc sulfide concentrates using traditional technology, including oxidative roasting and sulfuric acid leaching, cakes are obtained, the main components of which are zinc, lead, copper, iron in the form of compounds and noble metals [45-47]. The main reason for the incomplete leaching of zinc during leaching is the formation of poorly soluble zinc ferrite $ZnFe_2O_4$.

The most widespread in practice is the method of processing zinc cakes by Waelz with the transfer of noble metals and copper into clinker. Clinker is processed at copper smelters together with copper concentrates / 1. A.P. Snurnikov "Hydrometallurgy of zinc", M.: metallurgy, 1981, p. 331. 2. N.V. Gudima, Ya.P. Shein "A short guide to the metallurgy of non-ferrous metals" M.: Metallurgy. 1975, p. 117-136 /. The Waelz process, despite significant improvements in recent years, has several disadvantages. The main ones are the high consumption of the carbonaceous reductant, the low content of copper and noble metals in the clinker, which complicates their processing [48-55].

For the extraction of precious metals from zinc cakes, flotation is used. A known method of extracting silver from zinc cakes by flotation, including preliminary conditioning of the pulp with tetrachlorethylene, which helps to remove elemental sulfur from the surface of minerals and eventually increase the content of silver in the concentrate / 3. RF patent 2496892 /.

Known hydrometallurgical methods for processing zinc cakes, based on the decomposition reactions of poorly soluble ferrites with sulfuric acid at atmospheric or elevated pressure. From solutions obtained by such leaching, iron and other impurities are precipitated by various methods, after which zinc is extracted in the general technological scheme / 1 /.

Known combined methods of processing zinc cakes, including flotation of cakes, roasting of flotation concentrate, leaching of roasted products and extraction of precious metals from middlings using various methods and reagents / 4-7. RF patents No. 2192488, 2172352, 2170773, 2153013 /. A common disadvantage of these methods is the multistage nature and high unit costs for the extraction of precious metals from relatively poor middlings [56-67].

A known method of processing zinc cakes / 8. RF patent No. 2175354 /, selected as a prototype and including the stages of flotation of cakes, roasting of flotation concentrate, high-temperature sulfuric acid leaching of roasting products and uncoiling of the solid residue in concentrated sulfuric acid at a ratio of the residue and acid of 1: 0.7-1: 1.5 and a temperature of 150- 170 ° C followed by leaching of the boil product in an aqueous solution with a chlorine concentration of 0.3-1 g / l. When using this method, the yield of the silver-containing product decreases and, accordingly, the silver content in it increases, and costs are reduced. The main disadvantages of the prototype are the low silver recovery in the final product and the multistage technology in general [68-74].

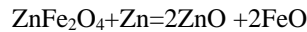
3. THE NATURE OF THE PROPOSED TECHNOLOGIES

3.1 STUDY OF THE POSSIBILITIES OF THE METALLOTHERMIC REDUCTION PROCESS OF ZINC FERRITE

Zinc cake (obtained at the acid leaching stage) contains about 20% zinc, indium, cadmium, lead, copper. The content of the main components varies within a fairly wide range, %: 17-20 Zn_{total} ; 4-6 Zn_{ZnS} ; $>1 Zn_{ZnSO_4}$; 2-5 Pb; 1,0-2,0 Cu; 0,2-0,4 Cd; 18-25 Fe; 4-6 S_{total} .

The reason why electric arc steel is difficult to process in furnace dust and zinc cakes is because it contains iron, zinc and silicates. Since the chemical and mineralogical composition of zinc production cakes and arc steel smelting furnaces are similar, finding ways to process them together is one of the current topics in metallurgy. For example, most of the zinc and iron in zinc cake and powder is in the form of zinc ferrite ($ZnO \cdot Fe_2O_3$), a compound that is insoluble in mineral acids and is not attracted to magnets. Therefore, it is necessary to find a way to process zinc cakes and electric arc furnace powder, so that zinc ferrite ($ZnO \cdot Fe_2O_3$) can be separated into zinc and iron. Currently, the most convenient and energy-efficient way to separate zinc and iron in zinc ferrite ($ZnO \cdot Fe_2O_3$) is to restore it with coke. The only thing to keep in mind is that the zinc that is formed after the recovery process evaporates quickly. This leads to the loss of precious non-ferrous metals [75-77].

Several studies have studied the thermodynamic possibilities of the recovery of zinc ferrite with zinc metal powder. Accordingly, the basic chemical reaction that takes place in a system can be described as follows:



The thermodynamic values of the starting materials are given in Table 1 [2].

Table 1: Appropriate thermodynamic quantities of substances (298 K)

Substance	ZnFe ₂ O ₄	ZnO	FeO	Zn
ΔH (kJ/mol)	-1182	-350.62	-272	-
ΔG (kJ/mol)	-1077	-320.66	-251.46	-
ΔS (J/(mol*K))	153.3	43.64	60.75	41.63

Using these values, we determine the probability that a chemical reaction will occur at several temperatures. To do this, we first consider the results of a chemical reaction under standard conditions:

$$\begin{aligned} \Delta H_{\text{reaction}}^{298} &= -2*350.62 - 2*272 + 1182 = -63.24 \text{ (kJ)} \\ \Delta G_{\text{reaction}}^{298} &= -2*320.66 - 2*251.46 + 1077 = -67.24 \text{ (kJ)} \\ \Delta S_{\text{reaction}}^{298} &= 2*43.64 + 2*60.75 - 41.63 - 153.3 = 13.85 \text{ (J)} : 1000 = 0.01385 \text{ (kJ)} \end{aligned}$$

From the results under standard conditions, it can be seen that the enthalpy value of the reaction is negative (exothermic), i.e. heat is released as a result of the reaction. The value of the free energy of the reaction is also negative, which means that the chemical reaction can flow independently under the given conditions. But in practice it is necessary to give activation energy to the starting materials for the beginning of each chemical reaction. How the increase in temperature after the onset of a chemical reaction affects the reaction rate has been determined:

$$\Delta G^T = -63.24 - T * 0.01385 \quad (1)$$

$$\begin{aligned} T=100 \text{ K}; \quad \Delta G^{100} &= -63.24 - 100 * 0.01385 = -64.385 \text{ (kJ)} \\ T=200 \text{ K}; \quad \Delta G^{200} &= -63.24 - 200 * 0.01385 = -66.01 \text{ (kJ)} \\ T=500 \text{ K}; \quad \Delta G^{500} &= -63.24 - 500 * 0.01385 = -70.165 \text{ (kJ)} \\ T=1000 \text{ K}; \quad \Delta G^{1000} &= -63.24 - 1000 * 0.01385 = -201.75 \text{ (kJ)} \end{aligned}$$

From the Gibbs energy values of the reaction, the values of the equilibrium constant were determined:

$$\begin{aligned} \text{Ln}(K_e) &= \frac{-\Delta G}{RT} \quad (2); \quad \text{or} \quad \text{Ln}(K_e) = \frac{(-1)*(-201.75)}{8.31*1000} = 0.0249; \\ \text{and} \quad K_e &= e^{0.0249} = 1.0225 \end{aligned}$$

When studying the thermodynamic possibilities of the reduction process of zinc ferrite with metallic zinc powder, it was found that at a temperature of 727 °C (1000 K) the equilibrium in the chemical reaction shifts to the right, that is, to the side of product formation. At the same time, thermal processing of raw materials with metallic zinc powder prevents contamination of the product with harmful (unnecessary) elements [78-80].

3.2 THERMODYNAMIC ANALYSIS OF ALUMINOTHERMIC REDUCTION OF METAL OXIDES

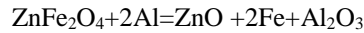
Aluminothermy is the process of extracting metals and non-metals (as well as alloys) from their oxides by reversing the metal with aluminum. Aluminothermia was first developed by Russian scientist opened by N.N. Beketov. Aluminothermy is used in the metallurgical industry to extract metals such as Cr, Mn, V, Zr, Ti from their oxides. A mixture of 8 moles of aluminum and 3 moles of magnetite (Fe₃O₄) is called termite, which is sometimes used to weld metals [81]. If a hot wire is mixed with a mixture of aluminum and iron oxides (FeO + Fe₂O₃), the course of a very intense exothermic reaction has been studied several times in practice. This process is widely used in the welding of train rails:



As a result of the reaction, the mixture is heated to 3500 °C. Therefore, recovery processes with aluminum are autogenous processes.

Metallic aluminum can also be used in the separation of non-ferrous metals. However, due to the high cost of aluminum, many metal oxides may not be economically viable. This situation requires metallurgists to look for cheap and secondary sources of aluminum. The results of chemical and mineralogical analysis of man-made wastes containing aluminum showed that the slags

formed in the secondary aluminum metallurgy contain powdered metallic aluminum. In the secondary metallurgical plants of aluminum production, the following compounds are found in the composition of salt slag from liquefaction furnaces: KCl – 22 %; NaCl – 13 %; Al₂O₃ – 34 %; SiO₂ – 3,5 %; CaO – 1,9 %; MgO – 4,8 %; Zn – 0,6 %, CuO – 0,4 %; metallic aluminum – 11 %; Fe₂O₃ – 2,6 %. From these slags, metallic aluminum is extracted by dry grinding or by dissolving its salts in water, which is then used as a reducing agent. For example, zinc ferrite can be reduced by aluminothermy to produce pure metallic iron and zinc oxide:



Since this chemical process is not available in the literature, its thermodynamic potential has been studied [82]. Accordingly, the thermodynamic magnitudes of the respective substances are shown in Table 2.

Table 2: Some thermodynamic values of the reactants

Substance	ZnFe ₂ O ₄	ZnO	Al ₂ O ₃	Fe	Al
ΔH (kJ/mol)	-1182	-350.62	-1675.7	-	-
ΔG (kJ/mol)	-1077	-320.66	-1582.26	-	-
ΔS (J/(mol*K))	153.3	43.64	50.92	27.15	28.34

The enthalpy, entropy, and Gibbs energy values of a chemical reaction for standard conditions were calculated according to Hess's law. The values of changes in this value at several temperatures are given in Table 3.

Table 3: Changes in thermodynamic values in the temperature range

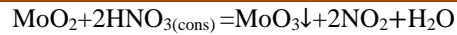
ΔH	R	ΔS	T	ΔG _T	LnK _e	K _e
-844,3	8,31	-0,061	100	-838,2	1,0087	2,742
-844,3	8,31	-0,061	200	-832,1	0,5007	1,6498
-844,3	8,31	-0,061	300	-826	0,3313	1,3928
-844,3	8,31	-0,061	400	-819,9	0,2467	1,2797
-844,3	8,31	-0,061	500	-813,8	0,1959	1,2163
-844,3	8,31	-0,061	600	-807,6	0,162	1,1758
-844,3	8,31	-0,061	700	-801,5	0,1378	1,1477

From the values in Table 3, it can be seen that energy is released in a chemical reaction (i.e., exothermic) and that the Gibbs energy value increases with increasing temperature. But the value of the equilibrium constant leads to a decrease. Because the reaction is exothermic, a decrease in temperature leads to an increase in the rate of the chemical reaction according to Le Chatelier's principle.

The process of magnetic separation is used to separate the components of the experiment. In this case, pure iron in powder form is extracted. The zinc oxide is used for hydrometallurgical selective smelting and alumina for wastewater treatment.

3.3 THERMODYNAMIC ANALYSIS OF MOLYBDENUM DIOXIDE ACIDICAL OXIDATION

The growing demand for molybdenum products requires the development of new technologies for the extraction of molybdenum from secondary raw materials and wastes of molybdenum production. Molybdenum extraction technology is available in the sector No. 5 of the "Scientific Production Association for the Production of Rare Metals and Hard Alloys" in Chikchik, owned by JSC Almalyk MMC, which is carried out by hydrometallurgical selective leaching. The process produces waste in the form of sludge, which also contains Mo, Re, Fe, Cu, Au, Ag - in the solid and soluble parts of the plant. These technologies produce hydrometallurgical wastes (cakes) that store molybdenum in the process. The results of spectrometric analysis showed that cakes containing non-ferrous and rare metals are a valuable man-made raw material. This cake contains the following components (%): 4.8 Mo, (including 2.1 oxidized and 2.7 sulfide); 1.2 Cu; 0.03 Re; 0.24 V as well as 9.5 Fe; 4.3 SiO₂; Residues of As, P and 6.0 ion exchange resins (used); industrial waste (stones, wood chips, etc.) and humidity of 42% and above [83]. In order to organize the efficient processing of this material, a method of selective melting of molybdenum-containing cakes in concentrated nitric acid solutions was carried out. The thermodynamic possibilities of this process were analyzed. Molybdenum in cakes is mainly in the form of slightly oxidized molybdenum dioxide (MoO₂). The reaction of its oxidation with nitric acid is as follows:



The thermodynamic values of the starting materials are given in Table 4.

Table 4: Appropriate thermodynamic quantities of substances (298 K)

Substances	MoO ₂	HNO ₃	MoO ₃	NO ₂	H ₂ O
ΔH (kJ)	-589.1	-133.91	-745.6	33.5	-228.597
ΔG (kJ)	-533.25	-73.78	-668.6	51.55	-242.827
ΔS (J)	46.28	266.78	77.74	240.2	188.724

The thermodynamic properties of the chemical reaction between nitric acid and molybdenum dioxide under standard conditions were studied:

$$\begin{aligned} \Delta H_{\text{Reaction}}^{298} &= -745.6 - 228.597 + 2 \cdot 33.5 + 589.1 + 133.91 \cdot 2 = -50.277 \text{ (kJ)} \\ \Delta G_{\text{Reaction}}^{298} &= -668.6 - 242.827 + 2 \cdot 51.55 + 533.25 + 2 \cdot 73.78 = -127.517 \text{ (kJ)} \\ \Delta S_{\text{Reaction}}^{298} &= 77.74 + 480.4 + 188.724 - 266.78 \cdot 2 - 46.28 = 0.167 \text{ (kJ)} \end{aligned}$$

The results of the calculations show that the chemical reaction releases heat (exothermic) during the process, and that the negative Gibbs energy output means that the process proceeds under standard conditions. We now determine the thermodynamic parameters of a chemical reaction over several temperature ranges:

$$\Delta G^T = -50.277 - T \cdot 0.167$$

Table 5: Thermodynamic values of the reaction in the temperature range 308 - 378 K

No	T	G ^T	LnK	Ke
1	308	-101,7	0,04	1,041
2	318	-103,4	0,039	1,04
3	328	-105,1	0,039	1,039
4	338	-106,7	0,038	1,039
5	348	-108,4	0,037	1,038
6	358	-110,1	0,037	1,038
7	368	-111,7	0,037	1,037
8	378	-113,4	0,036	1,037

From the thermodynamic values given in Table 5, it can be concluded that increasing the temperature by every 10 units in the chemical reaction between nitric acid and molybdenum dioxide reduced the rate of the chemical reaction. This is because the process of oxidizing molybdenum dioxide with nitric acid is exothermic and no temperature increase is required for this process. The optimum temperature for the process was set to 308 K (35 °C).

4. CONCLUSION

The main idea of the joint processing of cakes from a zinc and molybdenum plant and clinker from zinc production is based on the fact that these products are man-made raw materials of one enterprise and they are characterized by an identical composition in terms of the content of the main valuable components. projects based on practically the same hydrometallurgical methods with the difference that, due to the chemical inertness of clinker, the scheme of its processing involves the use of pyrometallurgical redistribution of "activation" of the material by sulfatizing low-temperature roasting. The joint processing of these products according to a unified scheme has shown the possibility of separating copper, zinc, iron and lead middlings enriched with noble metals using technology without pyrometallurgical processing in compliance with the principle of wastelessness and environmental safety of the project. This approach allows you to reduce capital costs for its implementation. The main operation of the proposed technological scheme for the combined processing of clinker and dust consists in oxidative sulfuric acid leaching of the mixture "clinker + dust" in a certain mass ratio in the presence of sodium nitrite and oxygen. Laboratory tests of this method have shown that the products of

leaching are a solution of copper and zinc sulfate, as well as a solid residue that concentrates iron (up to 28%) and precious metals. The processing of these products can be effectively implemented according to the above scheme. As a result of the practical implementation of the technology, the environmental orientation of the technology is ensured, which is achieved through the complex processing of a mixture of clinker and dust and the absence, as a result, of waste; a positive economic result is achieved, which is ensured by the resource-saving orientation of the technology due to the expansion of the raw material base of non-ferrous and ferrous metallurgy, the specialized infrastructure developed at the AMMC for the creation of an enterprise for processing the above raw materials and the availability of the necessary chemical reagents and qualified personnel for the operation of the enterprise.

5. ACKNOWLEDGMENT

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