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Application of Physical and Chemical Methods for Processing Slags of Copper Production

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ABSTRACT: The article deals with the problem of reducing copper losses from waste slags. A comprehensive solution to the problem with the use of physicochemical methods of influence on the melt is proposed. It is shown that the success of the depletion process depends on the sulfiding of oxidized copper compounds, the reduction of magnetite in the slag to wustite, and the creation of conditions for the coalescence of fine drops of matte. Creating these conditions makes it possible to reduce the residual concentration of copper to the level applied. This can be obtained poor matte, which is processed by pouring them into a melting furnace. Depleted slag is waste products and can be implemented in the construction industry. In fact, this makes it possible to abandon the creation of slag heaps.

KEYWORDS: slags, depletion process, sulphidation of oxidized compounds, magnetite reduction, bubbling of a bath, coalescence of small particles, poor matte, waste product, construction industry.

I. INTRODUCTION

The main problem of modern copper production in the world is the complex processing of slags, the utilization of valuable components and the increasing complexity of the use of raw materials. Over time, this problem will increase, as there is an all-round depletion of rich and easily opened ore deposits, an increased demand for environmental protection, an increase in the demand for non-ferrous metals, including copper and its accompanying elements.

The waste and intermediates of mining and metallurgical enterprises have accumulated a large number of tailings of the processing plants, copper slags, clinker from the processing of zinc cakes and other types of secondary man-made structures. These materials contain significant amounts of non-ferrous and noble metals, and in large part are outside the production cycle. Involving them in processing will allow enterprises to expand the resource base without increasing capital expenditures on mining and geological work and transportation of materials [1].

II. RELETED WORK

According to the traditional pyrometallurgical technology for producing copper, 4-5 tons of slags are produced per ton of metal. Although the copper content in slags is relatively small (0.5–3.5%), because of their large amount, metal losses are usually an impressive amount. In addition, it is necessary to take into account the conscious aspect of this problem. The fact is that slag heaps attack the natural territory, occupy thousands of hectares of farmland, pollute the air basin and disfigure the landscape. Assessment of damage from environmental pollution leads to a significant expansion of the boundaries of economic feasibility of the creation and use of low-waste technology. Taking into account this circumstance, the economic efficiency of combining production will undoubtedly increase, and new criteria arise for the formation of production structures in industry related to environmental protection [2].

In this regard, the problem of creating a rational and integrated technology for processing slags and industrial products of copper production is very important. The reasons for this technology should be based on the use of local secondary man-made structures and fit into the existing process line [3].



In world practice, a sufficiently large number of technologies for the processing of copper slags have been developed [4 – 6]. Almalyk Mining and Metallurgical Combine (AMMC) uses flotation slag processing technology.

The common disadvantage of all these methods is that they process the already solidified slags and do not use the heat of the molten state. During flotation enrichment, the extraction of copper from slags does not exceed 69 - 70%. At the same time, the specific productivity is lower when processing a mixture of copper ore and slag decreases by 22 - 25% in comparison with processing only ore. This also significantly increases the consumption of grinding balls and lining. In addition, the copper content in slag-tails slightly exceeds the permissible level (0.40 - 0.45%), which leads to irretrievable loss of copper and noble metals [7].

III. OBJECTS AND METHODS OF RESEARCH

We have conducted a study on the use of heat of the molten state and without significant changes in the technological regulations of copper smelting production.

From the literature it is known that in the slag during transportation to the slag heap a number of processes take place, leading to the partial precipitation of matte drops into the bottom phase. At the same time, the copper concentration in the lower part of the furnace is slightly higher than in the upper layers. This phenomenon we called thermogravity.

In many industrial processes, mechanical losses account for the bulk of the loss of non-ferrous metals with slag. Mechanically, the losses were represented by drops of metal (matte) of various size, which did not have time to stand out from the slag phase into the bottom (matte or metal) during the time the melts stay in the metal aggregate. The size of such drops in industrial slags ranges from 0.5 μm (micrometer) to 0.2 mm (millimeter). The bulk has a particle size of 10 - 100 microns.

The sedimentation rate of matte drops can be calculated based on the Stokes formula:

$$V = \frac{2}{9} \cdot \frac{(\rho_1 - \rho_2) r^2 g}{\eta_2} \cdot \frac{3 \cdot (\eta_1 + \eta_2)}{3\eta_1 + 2\eta_2}$$

where: v - the deposition rate of the drop, cm / s;

g — acceleration of gravity, $\text{g} \cdot \text{cm}/\text{s}^2$;

ρ_1 and ρ_2 - respectively, the density of a more or less dense liquid, g/cm^3 ;

η_1 and η_2 - viscosity of a more dense and less dense fluid, Poise ($\text{P} \cdot \text{s}$);

r - is the radius of the deposited drop, cm.

The calculation using this formula showed that during the transportation of slag into the bottom phase, only particles larger than 0.2 mm in size can settle [8].

IV. RESULTS OF THE RESEARCH AND THEIR DISCUSSION

We conducted studies that studied the rate of thermal termination of slag during its transportation to the slag heap. Laboratory responses on thermo gravitational depletion of slags in order to determine the main parameters of the technological process were performed using a laboratory unit located near the outlet chute of the smelting furnaces. The need for such an arrangement is due to the fact that it is impossible to study thermo gravitational depletion of slags after they are cooled and re-melted due to an irreversible change in the structure of the sulfide suspension and the ratio of mechanical and dissolved copper losses to slags [9]. Laboratory experiments on thermo gravitational depletion of slags were carried out at a cooling rate of 1, 2, 3, 4 and 5 $^{\circ}\text{C} / \text{min}$.

To compare the results, isothermal aging of such slag was carried out at 1250 $^{\circ}\text{C}$. The initial temperature was controlled by an optical thermocouple, and the temperature in the laboratory furnace was controlled by a thermocouple and secondary devices. The research results are summarized in table 1 - 3.

Table 1. The results of laboratory experiments on thermogravitational depletion at $dT / dt = 1 \text{ }^\circ\text{C} / \text{min}$

Slag type		Composition, %				T, $^\circ\text{C}$	Slag type		Composition, %				T, $^\circ\text{C}$
		Cu	S	SiO ₂	Fe				Cu	S	SiO ₂	Fe	
O T F	A	0,85	0,63	38,2	30,1	1305	R F	A	0,53	1,10	37,2	31,4	1305
	B	0,80	0,59	38,4	30,2	1250		B	0,51	1,05	37,4	31,6	1250
	C	0,62	0,51	38,9	30,5	1100		C	0,50	1,05	37,1	31,3	1100
	A	0,89	0,64	34,1	33,5	1300		A	0,52	1,15	35,8	32,1	1305
	B	0,85	0,60	34,4	33,6	1250		B	0,48	1,10	35,9	32,1	1250
	C	0,47	0,42	33,8	34,4	1100		C	0,57	1,10	35,1	30,0	1100
	A	0,98	0,78	32,5	36,8	1305		A	0,48	1,25	32,8	36,1	1305
	B	0,83	0,70	32,3	36,6	1250		B	0,47	1,15	32,6	36,4	1250
	C	0,60	0,32	32,8	36,6	1100		C	0,45	1,15	32,6	36,1	1100

Note: A - the original; B - isothermal exposure for 1.5 hours; C - slowly cooled; OTF – Oxygen-Torch Furnace; RF – Reverberatory Furnace.

Table 2. The results of experiments on thermogravitational depletion of slags at $dT / dt = 3 \text{ }^\circ\text{C} / \text{min}$

Slag type		Composition, %				T, $^\circ\text{C}$	Slag type		Composition, %				T, $^\circ\text{C}$
		Cu	S	SiO ₂	Fe				Cu	S	SiO ₂	Fe	
O T F	A	0,84	0,65	37,5	31,1	1300	R F	A	0,53	1,10	37,2	31,4	1305
	B	0,82	0,62	37,3	31,2	1250		B	0,51	1,05	37,4	31,6	1250
	C	0,65	0,58	37,2	30,9	1100		C	0,50	1,05	37,1	31,3	1100
	A	0,90	0,71	35,1	32,1	1300		A	0,52	1,15	35,8	32,1	1305
	B	0,81	0,70	35,2	32,2	1250		B	0,49	1,10	35,9	32,2	1250
	C	0,52	0,62	35,0	32,2	1100		C	0,48	1,10	35,1	30,0	1100
	A	0,88	0,75	32,2	35,2	1305		A	0,57	1,25	32,8	36,1	1305
	B	0,75	0,72	32,6	35,1	1250		B	0,48	1,15	32,6	36,1	1250
	C	0,54	0,62	32,1	35,4	1100		C	0,47	1,15	32,6	36,1	1100

Note: A – the original; B – isothermal exposure for 1.5 hours; B – slowly cooled; OTF – Oxygen-Torch Furnace; RF – Reverberatory Furnace.

Table 3. The results of laboratory experiments on thermogravitational depletion of slags at $dT / dt = 4 - 5 \text{ }^\circ\text{C} / \text{min}$

Slag type		Composition, %				T, $^\circ\text{C}$	Slag type		Composition, %				T, $^\circ\text{C}$
		Cu	S	SiO ₂	Fe				Cu	S	SiO ₂	Fe	
O T F	A	0,91	0,65	38,1	30,5	1305	O T F	A	0,93	0,70	37,2	31,1	1300
	B	0,85	0,60	38,1	30,4	1250		B*	0,83	0,65	37,1	31,3	1250
	C	0,68	0,52	38,1	30,4	1100		C	0,80	0,68	37,3	31,4	1100
	A	0,94	0,71	35,1	32,6	1300		A	0,88	0,69	34,9	33,1	1305
	B	0,86	0,70	35,4	32,8	1250		B*	0,82	0,64	34,9	33,1	1250
	C	0,61	0,65	35,2	32,6	1100		C	0,75	0,62	34,8	33,0	1100
	A	0,89	0,66	32,3	34,4	1305		A	0,84	0,72	33,1	34,6	1305
	B	0,82	0,61	32,3	34,5	1250		B*	0,84	0,63	33,1	34,7	1250
	C	0,60	0,47	32,2	34,5	1100		C	0,78	0,70	33,2	34,7	1100

Note: A - the original; B - isothermal holding at $dT / dt = 4 \text{ }^\circ\text{C} / \text{min}$; 1,5 hour; C - slowly cooled; B* - isothermal holding at $dT / dt = 5 \text{ }^\circ\text{C} / \text{min}$; OTF – Oxygen-Torch Furnace; RF – Reverberatory Furnace.

Analysis of the data presented in Tables 1–3 shows that ordinary thermogravity somewhat reduces the copper content in the upper layers of the slag bath with the production of a very poor matte in the bottom part (2.3–3.2% Cu). At the same time, the upper part requires additional processing, which means that it is necessary to create slag heaps

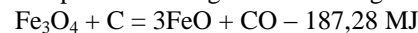
after all. When a slag is dumped, the bottom part must be collected separately and sent for remelting in a reflective furnace or for flotation. This creates additional difficulties and does not allow to solve the problem of processing in one technological cycle.

This situation can be explained by the fact that the reduction processes of slag magnetite, the sulfidization of oxidized copper compounds, and the coalescence of fine droplets of matte proceed extremely slowly.

During the real time of slag transportation to the dump (30–40 min), only a small amount of copper is deposited in the bottom phase. In addition, isothermal aging for 60–90 minutes is very expensive and has no technological perspective. And it is also necessary to take into account the fact that during transportation the temperature of slags decreases. This leads to the fact that the viscosity of the melt decreases and the deposition process becomes difficult [8].

As was shown earlier, the success of the depletion of copper slags can be achieved by reducing the slag magnetite, sulphiding the oxidized copper compounds and creating conditions for the coalescence of small droplets of matte into larger ones. Of particular importance is the process of coalition, since the radius of a drop of matte is included in the Stokes formula in the square. During the experiments, we tried to create conditions for the realization of the physicochemical processes listed above. In order to save time and money on analyzes in experiments, we analyzed only changes in the copper content in different parts of the slag bath. Moreover, earlier experiments showed that during normal thermogravity, the content of sulfur, silicon dioxide and iron varies slightly.

The reduction of slag magnetite can proceed through the following reactions:



The use of this technology is complicated by the fact that it requires coke, which is not produced in our republic.

From the theory of metallurgical production it is known that magnetite can be reduced by metallic iron:



Metallic chips, ferrous scrap, iron powder and many other types of secondary technogenic formations can be used as metallic iron [10].

We have conducted studies to determine the effect of steel chip additives on the degree of magnetite reduction and the copper content in the slag. For the research, a mixture of slag AGMK (Almalyk Mining and Metallurgical Combine), prepared for flotation concentration, was selected.

Copper content in the mixture was 0.85%, magnetite 12.45%. The research results are presented in Fig.1.

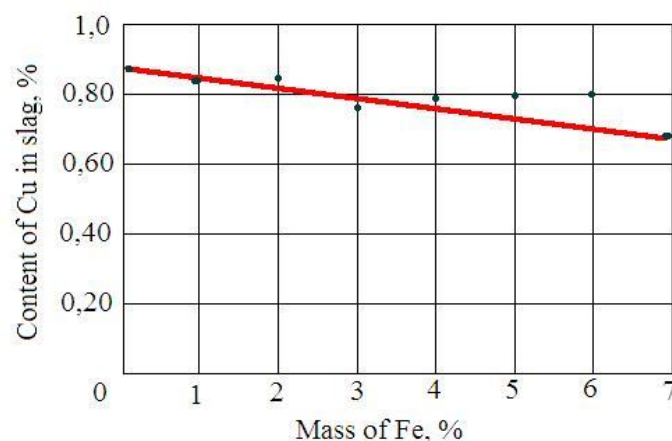


Figure 1. The effect of iron shavings on the copper content of the slag

As can be seen from Figure 1, the addition of iron shavings to the charge mixture only slightly reduces the copper content in the slag, and this process cannot be recommended for industrial introduction.

A significant amount of copper in the slag is in an oxidized state and in very small sizes. According to the Stokes formula, during the residence of the slag, only particles larger than 0.20 mm can settle in the bottom part of the ladle. Activating these two processes (sulphidation of oxidized copper compounds and coalescence of small droplets)

can occur when pyrite concentrate is added to the molten slag. To verify these positions, we carried out a series of experiments with the addition of various amounts of pyrite concentrate. The research results are presented in Fig.2.

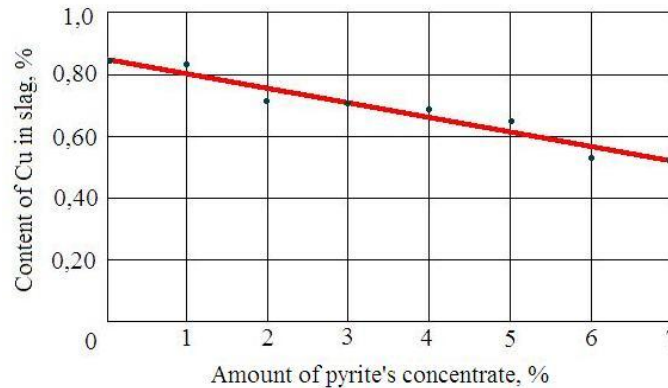
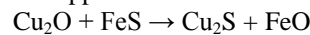


Figure 2. The effect of pyrite concentrate additives on the residual copper content in the slag

From the data in Figure 2 it can be seen that the residual copper content in the slag is reduced to 0.55 - 0.60%. This was due to the fact that pyrite dissociates in contact with molten slag by the reaction:

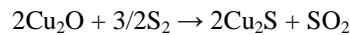


The resulting iron sulfide interacts with copper oxides:



The resulting copper sulfide can go into the bottom matte phase.

The reaction is also possible:



The SO₂ released by this reaction bubbles the slag bath and promotes the coalescence of small drops of matte into larger ones.

An excessive increase in the addition of pyrite concentrate increases the slag volume and dilutes the matte that is formed. For this reason, we conducted studies on the joint addition of metallic iron and pyrite concentrate to slag. The research results are presented in Fig.3.

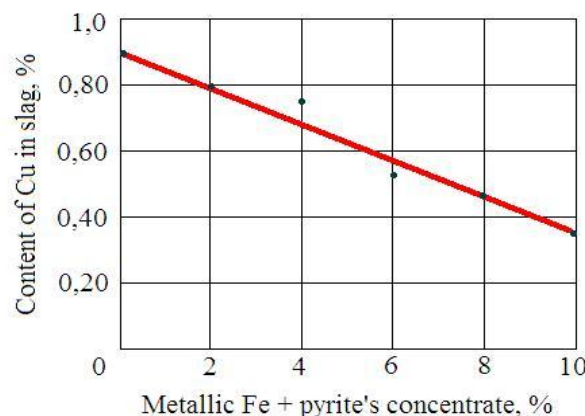


Figure 3. The effect of joint addition of metallic iron and pyrite concentrate in the ratio 1:4 on the residual copper content in the slag.

The research results presented in Fig. 3 showed that the joint addition of metallic iron and pyrite concentrate reduces the residual copper content in the slag 0.35 - 0.40%. These data practically coincide with the thermal possibilities of extracting copper from slags and their further processing is not effective. Such slags can be sold in the cement industry or in the construction industry.

In the absence of iron shavings at the enterprise, zinc production clinker can be used as a reducing agent, the composition contains more than 20% metallic iron.



V. CONCLUSION

It is the implementation of this method that is based on the fact that the composition of the zinc production clinkers contains a large content of the main carbon as a reducing agent and sulfur, the main copper sulphidizer. Due to the formed gases CO₂ and H₂O, when the hematite and magnetite interact with carbon and hydrogen, the slag mass does not increase. When magnetite is reduced, the viscosity of the slag decreases and this makes it possible to increase the sedimentation rate of the matte drops to the bottom phase. All of the taken results show that, implementing this research on an industrial scale it will increase the coefficient of productivity of copper producing plants.

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