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Study of the Factors Influencing the Decoppering Process of Non-Ferrous Metallurgy Slags: A Review

Shokhrukh Khojiev[1], Bakhriddin Berdiyarov[2], Dilfuza Yavkochiva[3], Jahongir Abduraimov[4]

[1]Senior Lecturer of the Department of Metallurgy, Tashkent State Technical University,
[2]Head of the Department of Metallurgy, PhD, Tashkent State Technical University,
[3]Assistant of the Department of Chemical and Food Technologies, Termiz branch of Tashkent State Technical University,
[4]Student of the Department of Chemical Technologies, Termiz branch of Tashkent State Technical University,
E-mail: hojiyevshohruh@yandex.ru

***Abstract**: The metallurgy of copper and other heavy non-ferrous metals is the leading link in the domestic non-ferrous metallurgy. The share of heavy non-ferrous metals in Uzbekistan accounts for a significant part of the gross output of the industry. The value of copper is increasing from year to year, especially in connection with the rapid development of energy, electronics, mechanical engineering, aviation, space and nuclear technology. Further development and the technical level of copper production largely determine the technical progress of many sectors of the national economy of our country, including microprocessor technology. To obtain copper, all kinds of smelting methods are used, for example, smelting copper concentrates in electric, reflective, shaft furnaces, using the process of converting copper mattes, thanks to autogenous suspended smelting, on matte, etc. Today there are several main processes of autogenous smelting: process "Noranda", "Warkra", "Mitsubishi", "Oxygen-flare" and Vanyukov. Unfortunately, the development of new designs of furnaces and various processes requires significant investments, and our enterprises do not have enough free funds. This paper will consider the technology for reducing the loss of copper with slags during smelting processes. Numerous parameters affect the copper content in slags, of which the following should be considered the most important: preparation of the charge, parameters of the technological mode of smelting, physicochemical properties of melts, design of units, organization of work, etc. It is very difficult to give an unambiguous assessment of the influence of all parameters acting separately or in aggregate, and is hardly possible at all. In this regard, we will try to assess individual process parameters from the point of view of their influence on the depletion indicators and the possibility of changing their values in real metallurgical smelting.*

Keywords— *metallurgy, slag, copper, recycling, decoppering, depletion process, factors, reducing-sulfiding complex.*

1. INTRODUCTION

Charge preparation plays a very important role in any metallurgical process. It would be ideal to use a bedding system for this purpose. However, this will require large capital investments for the construction of the charge house, there must be significant free areas, which is not feasible under the conditions of the operating Almalyk copper-smelting plant. In addition, in the concept of impoverishment we have adopted, it is also envisaged to work with stale materials and wastes from various industries, the special preparation of which will negatively affect the economic indicators of the process. For these reasons, no special expensive preparation of the charge is planned.

The manufacturability and feasibility of the developed technology will be closely related to the unit, which is taken as a basis. As shown earlier, the first liberated copper smelter is a reverberatory furnace. It is this furnace that is recommended for the depletion process and there are no other possibilities for varying equipment in Almalyk Mining and Metallurgical Complex (AMMC). It is also necessary to take into account the fact that at present in the production of copper there is a tendency towards the widespread decommissioning of reverberatory furnaces and their replacement with autogenous technology. But since most autogenous processes produce slags rich in copper, we believe that reverberatory furnaces can be used as depleting units without changing their design. They can process solid and liquid converter slags in conjunction with non-ferrous metal-containing reduction - sulfiding complexes.

The need to process converter slags in a separate cycle is due to the very nature of the pyrometallurgical method for producing copper, and it is unlikely that an economically viable technology for combining smelting and depletion in one autogenous unit will appear in the near future.

2. ORGANIZATION OF COPPER SLAG DEPLETION PROCESSES

The organization of work makes a certain contribution to the efficiency of the depletion process. However, there are practically no opportunities for a significant change in the currently used production organization system and, apparently, there is no need for this.

From the above it follows that the real lever for controlling the quality of the depletion process is the selection of the optimal technological mode of melting and, closely related to it, the physical and chemical properties of slag-matte melts.

The technological mode of melting primarily means: optimal slag and matte compositions, oxidation potential of the gas phase, temperature, as well as the physicochemical properties of melts, which are closely related to each other.

All of the above technological parameters have a significant effect on the copper content in the slags. However, not all of them can vary within wide limits, providing the possibility of influencing the final results of depletion. For example, the upper temperature limit is limited by the resistance of the refractories in an aggressive environment and the increase in the solubility of copper under these conditions. The lower limit is limited by the fluidity of the melts, the difficulty of separating slag and matte due to a sharp increase in viscosity with decreasing temperature, etc. Therefore, in real conditions of reflective redistribution, the temperature of the melts can vary within the range of 1150-1300 ° C, i.e. this factor cannot be used to effectively regulate the copper content in slags.

The composition of the gas phase is a very important factor affecting the copper content in slags. Working in neutral and reducing atmospheres is considered ideal. Although the amount of oxygen supplied to the fossil fuel combustion furnace can be easily adjusted, it is almost impossible to create the required atmosphere under real metallurgical conditions. The fact is that a reflective furnace is far from being a modern metallurgical unit and it is practically impossible to eliminate harmful leaks of secondary air in it. For this reason, the gas atmosphere inside the furnace above the melt will always be oxidizing and it is necessary to take special measures to reduce its harmful effect.

The composition of the matte directly affects the copper content in the slags. This position has been discussed in detail in the previous sections. However, there are no opportunities for its wide variation. The point is that when working for rich mattes, the copper content in the slags increases. When receiving a unit of production, work on a poor matte increases its volume and causes large losses of copper during the subsequent conversion operation. As a result, metallurgy is forced to work on matte containing 25-36% copper.

Thus, one of the real effective levers of quality control of the depletion process is the selection of an effective slag composition with optimal physicochemical properties.

3. Considerable aspects of the process of extracting copper from slag

The following The study of the literature on the processing of copper slag, the analysis of the practical work of copper smelters, as well as the results of his own research and development, allowed the author to establish that the success of the depletion process is largely determined by the solution of the following problems:

- reduction of slag magnetite to FeO;
- adjusting the slag composition and operating at the optimal content of such components as SiO_2, CaO, FeO, Fe_3O_4, etc.
- sulfidation of oxidized copper compounds and their transfer to the matte phase:
- bubbling the melt with gases in order to create conditions for the coalescence of fine matte droplets;
- transferring small drops of matte to the bottom phase by mixing with the matte;
- binding of sulfur into the condensed phase, which allows one to simultaneously reduce desulfurization to an acceptable limit and convert it to matte;

The complex of these measures will reduce the solubility of copper sulfide in the slag, extract dissolved copper into matte, convert oxidized copper into a sulfide form, and also create conditions for the deposition of the formed fine suspension of sulfides into the bottom phase.

Each of the above factors has its own specific features and possibilities for implementation in terms of influence on the final results of the depletion process.

The need to restore magnetite is due to the following circumstances. The oxidizing atmosphere of the furnace creates good conditions for the conversion of wustite to magnetite. At concentrations exceeding the solubility limit of Fe_3O_4 in slag, magnetite transforms into matte and creates great difficulties in its further processing. Within the limits of its solubility in slag (10-20%), magnetite significantly changes such properties of the melt as density and viscosity.

An increase in density makes it difficult to separate slag and matte, and this, in turn, leads to a significant increase in copper losses with slags.

An increase in the viscosity of the slag reduces its fluidity, creates great difficulties in removing the melt from the furnace, and significantly increases the amount of entangled metal particles. Hence the need for the maximum possible reduction of magnetite in the furnace.

Magnetite under conditions of metallurgical smelting can be reduced as a result of the following reactions:

$$Fe_3O_4 + Fe = 4FeO \quad (1)$$
$$Fe_3O_4 + C = 3FeO + CO \quad (2)$$
$$3Fe_3O_4 + FeS + 5SiO_2 = 5(2FeO \cdot SiO_2) + SO_2 \quad (3)$$

The presence of free silicon dioxide in the melt reduces the activity of wustite and promotes a more complete reduction of magnetite as a result of the fayalite formation reaction:

$$2FeO + SiO_2 = Fe_2SiO_4 \quad (4)$$

From the point of view of environmental protection, it is more expedient to restore magnetite according to reactions (1) and (2), since the development of reaction (3) leads to the release of free sulfur dioxide into the air atmosphere.

Under the AMMC conditions, for the implementation of reactions (1) and (2), it is possible to use clinker of zinc production, which contains free carbon and metallic iron in its composition. In addition to clinker, various carbon-containing materials (coke, coal), gaseous and liquid fuels, as well as non-ferrous metals with a high affinity for oxygen (for example, aluminum waste) can be used as a reducing agent. However, the use of clinker is more expedient, because in addition to reducing agents, it contains a large amount of non-ferrous and noble metals, which will also be extracted into the finished product and will significantly reduce the cost of processing. For these reasons, clinker was chosen as a reducing agent.

The successful implementation of the copper extraction process is possible only under operating conditions with an optimal slag composition.

Analysis of the factory practice data made it possible to establish that the content of FeO, SiO_2, CaO, Fe_3O_4 in them directly affects the optimality of the slag composition. Other oxides that make up the slag melt (ZnO, Al_2O_3, BaO) are not introduced into the charge as fluxes, but are included in the composition of the initial components in small amounts. Therefore, when assessing their effect on the loss of metals with slag, it is difficult to count on the possibility of active influence with the help of these components on the course of the metallurgical process.

Let us consider in more detail the influence of the main components of slag melts. V.Yu. Kupryakov gives the following data on the effect of slag composition on copper losses.

4. THE EFFECT OF TEMPERATURE AND OXYGEN ON THE COPPER LOSS WITH SLAG

Losses of copper with slags in the AMMC are as follows. In slags with a degree of acidity of 0.95 - 1.3 (29-35% - SiO_2) copper losses are greatest: total 0.47-0.54%; mechanical 0.22-0.28%, dissolved 0.21-0.29%. General (0.28-0.44%) and mechanical losses (0.23-0.36%) are also quite high in slags with an acidity of 1.9-2.5 (46-52% SiO_2). The smallest total (0.15-0.38%) and mechanical losses (0.05-0.15%) are observed in slags with a degree of acidity from 1.3 to 1.9 (36-45% SiO_2). The ratio of mechanical and dissolved copper losses for the first slags is about 1, for the second - about 3, and for the third - about 0.7.

From the graphs it follows that the total minimum losses are observed when the content of the slags is 42-45% SiO_2, the dissolved losses decrease as the content of this component increases. This effect of silica on losses is associated with a significant increase in the interfacial tension with an increase in the SiO_2 content. However, when the content of silica is over 45-46% (at a constant temperature of the slag), the viscosity increases sharply, the effect of which on the deposition of particles becomes greater than the effect of interfacial tension.

An increase in the content of calcium oxide to 12-13% reduces all forms of copper loss. This component has a greater effect on the reduction of dissolved losses, less on a decrease in mechanical losses. This is due to the fact that interfacial tension and viscosity decrease with increasing calcium oxide content.

Iron oxide, with an increase in its content from 26 to 52%, increases all forms of copper loss, which is explained by an increase in the solubility of copper in ferrous slags.

The data presented show that the minimum copper losses are achieved when slags of the following composition are obtained:

42-45 % SiO_2, 12-13 % CaO, 22-25 % FeO

It is to work with such a slag composition that the creation of compositions of reducing-sulfiding complexes will be directed.

To regulate the composition of the slag, limestone, tailings of the lead and copper concentrating plants of AMMC will be used. In these materials, along with the components regulating the composition of the slag, there is a noticeable amount of non-ferrous metals, the additional extraction of which will have a favorable effect on the technical and economic indications of the entire depletion process.

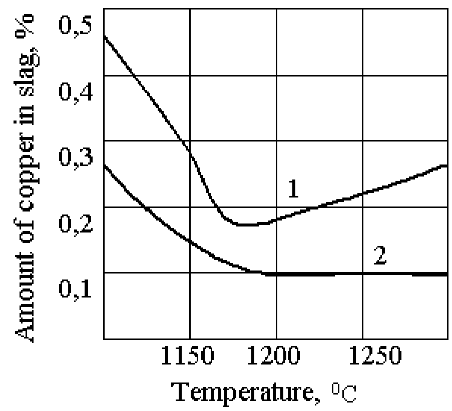

Fig.1. Influence of temperature on the loss of copper with slags

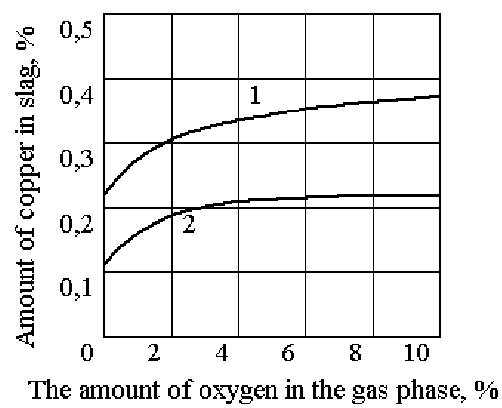

Fig.2. Influence of oxygen in the gas phase on the loss of copper with slags

The effect of temperature on copper loss depends on the composition of the slag (Figure 1). An increase in temperature determines a decrease in the content of copper in the acidic slag. As the temperature rises, the copper content in the main slag first decreases due to a decrease in mechanical losses, and then increases due to an increase in the solubility of copper. The effect of oxygen in the gas phase on the loss of copper with slags is shown in Fig.2. An increase in the oxygen concentration in the gas phase leads to an increase in the copper content in the slags. This can be

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explained by the formation of magnetite in the process of decopperization. Magnetite increases the oxidation potential of the slag, which leads to an increase in the solubility of copper in it.

Thus, for basic slags, there are optimal temperature conditions at which copper losses are minimal. For example, for the main slags of the AMMC, the optimal temperatures are 1180-1200 ° C. The effect of magnetite on the loss of copper with slags is shown in Figure 3.

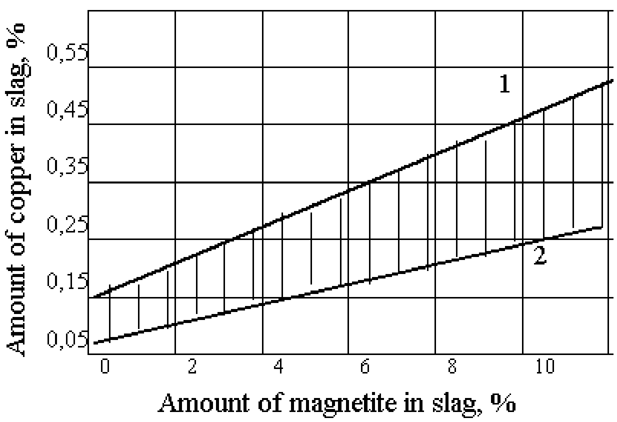

Fig.3. The effect of magnetite on the loss of copper with slags: 1-total losses; II-dissolved:
1 - 31 % SiO_2; 45,6 %, FeO; 2,6 % CaO; 7,7 % Al_2O_3
2 - 41,4 % SiO_2; 23,5 % FeO; 12,3 % CaO; 10,2 % Al_2O_3

An increase in magnetite content determines an increase in mechanical and dissolved copper losses with slag. So. with an increase in its concentration from 0 to 11%, mechanical losses increase from 0.05 to 0.23%, dissolved from 0.13 to 0.23%. For each percent of magnetite, the first losses increase by 0.016%, the second by 0.014%. The increase in mechanical losses of copper is associated with a deterioration in the conditions for coarsening the size of suspended particles, and an increase in dissolved losses is associated with an increase in the oxidizing potential of the slag with an increase in the concentration of magnetite.

The theoretical and practical provisions considered in the previous sections allow a reasonable approach to the choice of technological modes of smelting in order to minimize the loss of copper with slag.

The choice of the optimal slag composition is one of the most important reserves for increasing metal recovery. The slag must be sufficiently low-melting, liquid-flowing and have a low density. The solubility of non-ferrous metals in the slag should be minimal, and the interfacial tension at the matte-slag interface should be as high as possible. Moreover, the output of slag should be minimized.

Slags with a high concentration of iron oxides dissolve more non-ferrous metals. The interfacial tension on the matte border with such melts is low. All this leads to increased losses of copper, which are in such a melt, both in a dissolved state and in the form of a finely dispersed suspension.

The effect of iron oxides is much less harmful under reducing conditions, when the Fe^{3+} content is reduced to a minimum. In order to reduce the concentration of FeO, fluxes are introduced into the melt: quartz and limestone. An increase in the concentration of SiO_2 in the melt significantly reduces the solubility of non-ferrous metals in the slag and increases the interfacial tension of the system.

A prerequisite for depletion of the slag is its reduction in order to reduce Fe^{3+} in it and, at the same time, dissolved metals.

It is most rational to recover slags by blowing them with a reducing agent (gaseous, liquid or coal dust). Obviously, it is most convenient to use natural gas for recovery. However, the utilization rate of unconverted gas is very low. The reduction can be carried out in a fixed bed, for which a solid reductant is loaded onto the surface of the melt. Probably in this case, due to the low rate of mass transfer processes, the recovery will proceed slowly. Consequently, preference should be given to processes combining reduction with stirring of the melt.

Due to the oxygen dissolved in the melt, as well as the high oxidation potential of the gas phase, up to 10% of the copper in the slag is in the oxidized state. If you do not take special measures to sulfidize them, they will be almost completely lost with the slag. It is proposed to use zinc tailings from the AMMC lead concentration plant and phosphogypsum from the Almalyk chemical plant as sulfidizers.

The selection of the optimal composition of the slag, its reduction and sulfiding, although necessary, but insufficient operations for deep depletion. The fact is that the metal and sulfide particles formed during these operations are very small in size and for their sufficiently complete sedimentation, long exposures (10-15 hours) are required, which is not technologically and economically feasible.

The process of separating fine suspended particles from slag can be accelerated in various ways. In particular, bubbling of the bath with gaseous products of reduction reactions, mixing of slag with matte can be used. Both of these possibilities, to one degree or another, take place during the treatment of copper slags with reductive sulfiding complexes.

The technology for depletion of copper slag can be successfully implemented if it does not pollute the air basin with sulfur-containing gases outside the existing standards. To prevent the release of sulfur dioxide into the environment, limestone is introduced into the charge, which binds sulfur into a very strong compound CaS - calcium sulfide. Calcium sulfide, in turn, participates in the sulfidation of oxidized copper compounds according to the reaction:

$$Cu_2O + CaS = CaO + Cu_2S \qquad (5)$$

Thus, at the same time, two problems are solved: sulfiding oxidized copper compounds and preventing the emission of harmful gases into the air basin, which is technologically very advantageous.

When solving the complex problem of processing slags from copper production, it is very important to establish technological and economically viable boundaries for the depth of copper extraction. The ideal option is the complete disposal of copper and all associated valuable elements. However, in production this is an almost impossible task. The fact is that up to 10% of copper is embedded in the lattice of other compounds (for example, wustite) and it is impossible to extract it without completely destroying the structure of the solid. Part of the copper is in the form of very thin inclusions and, in a technologically acceptable time (1.5 - 2 hours), does not have time to settle into the bottom phase. This is about 20-40% of the total copper losses with slag. Part of the copper is in the slag in an oxidized state and does not have time to sulfidize and go into the matte phase. This part of the copper is irretrievably lost with slags. Thus, there are always objective reasons for the presence of a certain amount of copper in the slags and losses are inevitable here.

A study of the literature and analysis of the operation of operating copper smelters allows us to conclude that the economically and technologically acceptable residual copper content in slags is 0.30-0.35%. Deeper de-crimping causes great technological difficulties and is associated with significant economic costs. Experience shows that with such a residual copper content, it is possible to organize waste-free production by transferring decopperized slags to the construction industry.

5. CONCLUSION

Thus, on the basis of the analysis carried out, it can be concluded that in real conditions of the Almalyk Mining and Metallurgical Combine, the technology of extracting copper from slags under conditions of a minimum desulfurization can be carried out using reduction-sulfiding complexes (RSC) of two types:

- RSC containing a reducing agent and a sulfidizing agent in the form of hard-to-dissociate sulfates, during the melting of which matte formation occurs due to the reduction of sulfur and exchange reactions;

- RSC, the content of sulfur in the form of sulfide, but having the ability to maintain a low partial pressure of oxygen, which will lead to the binding of dissociated sulfur to sulfide compounds that form matte.

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