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Technology for processing balanced feed charge based on copper-, lead-containing products

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Introduction

One of the important tasks of sustainable development of non-ferrous metallurgy in Kazakhstan is the organization of new productions (technologies) aimed at the complex processing of substandard intermediate products and recycled materials of copper and lead production with maximum extraction of non-ferrous and associated valuable metals. This is since the increase in the content of toxic and carcinogenic arsenic [[1], [2], [3], [4], [5], [6], [7], [8], [9]] in sulfide concentrates [[10], [11], [12], [13], [14], [15], [16]] significantly affected its distribution between the gas, slag and matte phase in smelting processes [16]. As a result, large volumes of substandard intermediate products, recycled materials and man-made waste with increased arsenic content were formed. On the

one hand, this increased their negative impact on the environment and public health, which hinders their further processing. Serious pollution of the environment and significant harm to human health are caused by emissions of arsenic, especially $As₂O₃$. Arsenic control has become one of the important issues for all copper and lead smelters [17].

Based on the above, it can be stated that the dominant linear model of obtaining lead and copper from low-quality primary sulphide raw materials in the production of non-ferrous metals does not meet modern requirements of today. Industrial development of technologies for processing substandard copper-, lead-containing intermediate products and recycled materials of lead production is included in the number of the most important priorities for the innovative development of the mining and metallurgical industry of Kazakhstan.

 $=$ 96 $=$

In recent years, the circular economy model has been widely spread in the global economy within the framework of the concept of "zero waste". Systems for managing substandard multi-component intermediated products and man-made waste in many countries already meet the principles of a circular economy [18].

For the economy of Kazakhstan, the task of primary importance is to transfer the activities of the mining and metallurgical complex of the republic to a circular economy within the framework of the "zero waste" concept using resource-saving and energy-efficient technologies. The development and design of new technologies aimed at the complex processing of multi-component raw materials substandard intermediate products and man-made waste from lead production, seems to be very relevant.

The objective of the present research is to study the behavior of copper, lead, zinc and arsenic during the reductive-oxidative processing of balanced charge under conditions of technology scaling and to determine the optimal parameters that ensure high complex extraction of metals into targeted products: lead into the rough lead, copper into matte, zinc into slag.

The main objective of the research is to evaluate the selective extraction of Pb, Cu and Zn into targeted products.

Materials and research methods

The compositions of the initial products are given in Table 1.

The composition of the charge from various materials was carried out based on the sulfur content in the initial products, taking into account the production of matte with a high copper content. To obtain slag of optimal composition [19, 20], ensuring the minimum solubility of lead and copper in it, the required calculated amount of quartz flux (95% $SiO₂$) was added to the charge.

The following charge structure was adopted for calculating the average composition of the charge, %: lead cake -30 ; copper-lead matte -25 ; copper

slips -25 ; converter slag -10 ; zinc cake -5 ; quartz flux -5 .

Calculated composition of the average balanced charge, % by weight: 13 Cu; 43.5 Pb; 5.6 Zn; 7.8 Fe; 5.0 S; 1.6 As; 0.6 Sb; 6.0 SiO₂, others.

Experimental part: Installation diagram and procedure for conducting experiments

The experiments were carried out using a Nabertherm GmbH RHTV 120-150/16 hightemperature furnace, the general appearance of which is shown in Fig. 1.

1 – crucible with charge; 2 – alundum tube for blowing the melt; $3 -$ quartz reactor; $4 -$ plug; $5 -$ gas outlet hose

Fig. 1 - General view of the high-temperature furnace (A) and the design of the assembled installation (B)

The use of a large-capacity furnace made it possible to conduct experiments with the maximum amount of the initial charge, which was constant in all experiments – 500 g.

Technological experiments were carried out in two stages: the first was the reduction smelting of the charge by blowing with natural gas to obtain rough lead, slag and intermediate matte, and the second was the oxidizing blowing of the matte with atmospheric oxygen to obtain commercial copper matte and slag.

The experiments determined the influence of the consumption of natural gas, oxygen and the duration of melt blowing on the technological parameters of smelting.

Preliminary experiments have shown that at a charge melting temperature of 1473 K, the production of a fluid slag that would ensure the complete flow of physical and chemical processes of product formation and separation is not achieved. For complete homogenization of the melt, it was necessary to increase the holding time of the melting temperature regime to 20 minutes. From a practical point of view, an increase in the melting time leads to a decrease in the productivity of the technology as a whole, therefore, to optimize the process, all experiments were carried out at a higher temperature - 1523 K.

In the first stage, upon reaching the set temperature (1523 K), the melt was held for 10 minutes to obtain a homogeneous melt, after which the melt was blown with natural gas for a set time. The natural gas consumption varied within the range from 1 to 1.7, in fractions of the stoichiometric required amount (SRA) for the complete reduction of lead and zinc compounds (sulfates, sulfides, ferrites) to oxides with their subsequent conversion to slag. The melt blowing time was 5, 10, 15, 20 minutes. Upon completion of the melt blowing for a set time, the tube was raised (position above the melt), and the furnace was cooled in a stream of natural gas. After the furnace cooled, the crucible with the sample was removed from the quartz reactor. The resulting smelting products: rough lead, matte and slag were separated from each other and each product was subjected to elemental analysis for metal content.

The intermediate mattes contained increased levels of lead, zinc and iron sulphides. In order to

reduce their content in the mattes and obtain a highcopper matte, the intermediate matte was purged with oxygen in the second stage. The procedure for conducting the experiments was the same as in the case of blowing the melt with natural gas. The temperature of the experiments was 1523 K.

The dust yield in all experiments was calculated based on the difference between the amount of the initial sample and the sum of the amount of obtained smelting products.

Each experiment was repeated three times. After the experiments, the products were subjected to elemental analysis. The results of parallel experiments on the metal content showed good convergence (error +/- 0.5% abs.). Based on the averaged results of the product yield and the metal content in them, the material balances of the reducing and oxidizing smelting of the charge, as well as the consolidated material balance of the general technology for processing the balanced charge, were calculated.

Results and discussion

The results of the conducted technological experiments on the reduction smelting of the charge are presented in Fig. 2-5.

It should be noted that the consumption of natural gas and the duration of the melt blowing process are complementary. Research could focus solely on the influence of natural gas consumption, leading to the establishment of its optimal value. However, determining the time dependence of metal extraction allows for a qualitative assessment of the kinetic patterns of the interaction between the charge components and natural gas.

It has been established that the extraction of copper into the matte demonstrates a high recovery rate of over 98% at a natural gas consumption level of 1.0 times its stoichiometric requirement for the reduction of lead compounds (see Fig. 2).

Further increasing the consumption of natural gas for copper extraction into the matte has only a minor effect. This established pattern aligns well with the theory of the process. In the context of reductive smelting of the charge with natural gas, the reduction of lead sulfates and sulfides appears to be more advantageous.

 \equiv 98 \equiv

Figure 2 – Impact of natural gas consumption CH4 (ratio of SRA) and the melt blowing time (τ, min) on the extraction of copper into smelting products

Figure 3 – Impact of natural gas consumption CH⁴ (ratio of SRA) and the melt blowing time (τ, min) on the extraction of lead into the smelting products

 \equiv 99 \equiv

Figure 4 – Impact of natural gas consumption CH⁴ (share of SRA) and the melt blowing time (τ, min) on the extraction of zinc into the smelting products

Figure 5 – Impact of natural gas consumption CH4 (ratio of SRA) and the melt blowing time (τ, min) on the extraction of arsenic into the melt products

Considering that the main matrix of the charge is a lead cake consisting of lead, zinc and copper sulfates when developing the technology, we proceeded from the condition of ensuring their complete recovery by purging the melt with natural gas at high temperatures (1200 ° C). This approach is a fundamental difference from previous studies, where during the processing of a charge consisting of various lead production materials with a high arsenic content, the melt was first subjected to oxidative purging to maximize its removal, and only then to reductive purging to form liquid phases: rough lead, matte and slag.

During the reduction purge of the melt with natural gas, favorable conditions are created for the reduction of sulfates by reaction:

$$
MeSO_4+CH_4 = MeO+CO_2\uparrow + SO_2\uparrow + H_2O\uparrow,
$$

Me – Pb, Cu, Zn. (1)

The resulting metal oxides, interact with the components of sulfide materials (copper-lead matte, copper slips), by reactions (2), (3):

$$
2PbO + PbS = 3Pb + SO2 \uparrow, \qquad (2)
$$

Cu₂O + FeS = Cu₂S + FeO, (3)

The liquid phases of rough lead and matte are formed.

At the temperatures involved in the process, while lead sulfide is present in the melt, other sulfides are not reduced by natural gas.

Under conditions of intensive bubbling of the melt with natural gas, lead compounds are reduced to metal, resulting in the formation of a liquid phase of raw lead. The completeness of raw lead formation depends on the amount of natural gas used for the reduction of lead compounds.

In the smelting of a balanced charge, the main losses of lead are related to its solubility in the matte. Experiments have shown that the minimum lead content in the matte is achieved when the resulting final copper matte contains the least amount of lead and iron sulfide. In $Cu₂S-MeS$ alloys with a high content of copper sulfide, the solubility of lead is minimal and is primarily determined by its physical solubility. As the content of PbS and FeS in the sulfide alloy increases, the solubility of lead in them rises, with the proportion of chemical losses of lead dominating due to the reactions between lead and iron sulfide [21].

The final extraction of copper into matte is affected by the distribution of copper between matte and rough lead. Under conditions of reductive smelting of a balanced charge with natural gas, an intermediate matte with a copper content of 55.3% was obtained. With further oxidative smelting of matte with air, due to the creation of conditions for the complete oxidation of iron and zinc sulfides, the copper content in the matte increased to 76.3%.

Based on the results of large-scale laboratory melts, the following optimal technological parameters are recommended for practical application:

– melt blowing time: with natural gas – 20 min; with oxygen – 20 min;

- gas consumption: $CH_4 - 1.7$ times exceeding its consumption from the SRA for the reduction of lead compounds; oxygen -1.4 times exceeding its consumption from SRA for the oxidation of zinc and iron sulfide;

– melting temperature – 1250 °C.

With optimal technological parameters, the following technological indicators were achieved:

– yield of smelting products, % of the total charge:

- rough lead 38.3;
- copper matte -15 ;
- $slag 31.2;$
- \bullet dust, gases -15.5 .

– composition of melting products, % by weight:

 \blacksquare rough lead – 99.34 Pb; 0.18 Cu; 0.08 Sb; others.

■ copper matte – 76.3 Cu; 0.93 Pb; 0.52 Zn; 0.23 Fe; 20.4 S; 0.05 As; 0.03 Sb; other.

 \blacksquare slag – 22.03 Fe; 20.2 SiO₂; 4.0 CaO; 9.12 Zn; 0.19 Cu; 0.55 Pb; other.

– extraction of metals into targeted products:

- \bullet lead into rough lead -97.6% ;
- copper into matte 98.6%;

• zinc into slag -56.8% ; into matte -1.7 ; into dust, gases -41.5 .

• arsenic and antimony into dust - 97.4% and 90%, respectively.

Conclusions

The possibility of implementing reductiveoxidative smelting of a balanced charge is demonstrated. The optimal parameters of the technology are established: time of blowing the melt with natural gas – 20 min; with oxygen – 20 min; CH_4 consumption – 1.7 times exceeding its consumption from the SRA for reduction of lead compounds; oxygen consumption $-$ 1.4 times exceeding its consumption from the SRA for oxidizing zinc and iron sulfide; temperature – 1523 K.

With optimal process parameters, the following product yield was obtained, % of the total charge: rough lead $-$ 38.3; copper matte $-$ 15; slag $-$ 31.2; dust, gases – 15.5. Copper matte with a high copper content (more than 76%) and a minimum impurity content were obtained: 0.93 Pb; 0.52 Zn; 0.23 Fe; 0.05 As; 0.03 Sb.

High rates of complex selective extraction of metals into target products have been achieved: lead in the rough lead $-$ 97.6%; copper in matte $-$ 98.6%; zinc in slag – 56.8%, in matte – 1.7%, in dust, gases -41.5% ; arsenic and antimony in the dust $-$ up to 97.4% and 90%, respectively.

The developed technology for processing balanced charge can be used for processing substandard intermediates of copper and lead production.

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

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Investigation, Software, Validation. M**. Kurmanseitov:** Writing- Reviewing.

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Құрамында мыс, қорғасын бар өнімдер негізінде теңдестірілген шихтаны қайта өңдеу технологиясы

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Технология переработки сбалансированной шихты на основе медь-, свинецсодержащих продуктов

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