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Distribution of Cu, Pb, Zn and As between the products of the reduction melting of slag from the Vanyukov furnace in the conditions of the Balkhash copper smelter

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Abstract. The results on the distribution of Cu, Pb, Zn and As between the products of the reduction melting of slag from the Vanyukov Furnace (VF) in the conditions of the Balkhash Copper Smelter (BCS) are presented. Optimal technological parameters of the process of two-stage reducing depletion of slags by natural gas in the presence of coal at a temperature of 1300°C. have been established. It is shown that under conditions of two-stage reductive depletion of VF slags, the residual metal content in the reduced slag is: copper – 0.35%; lead – 0.08%; zinc – 0.18%; traces of arsenic. High recovery of metals into target products has been achieved: copper in matte – up to 98%; gold, silver in matte – 96%; arsenic in dust at the first stage of slag treatment with natural gas – up to 99% and lead, zinc in dust at the second stage – up to 98%. The results obtained were used in the development of an innovative technological platform for a comprehensive waste-free technology for processing current and accumulated BCS slags to produce marketable products.

Keywords: VF slag, recovery, coal, natural gas, extraction, valuable metals, gas and coal consumption.

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1. Introduction

In modern metallurgy, where autogenous melting processes are actively used [1-6], the issue of processing emerging slag waste is acute. According to the information given in [7], approximately 2.2 tons of slag are formed during the production of each ton of metal. The disposal or reuse of these significant volumes of slag creates a number of environmental and spatial difficulties. The urgency of recycling these wastes is determined not only by economic, but also by environmental factors. The accumulation of slag heaps occupies significant territories, posing a threat to the environment, including pollution of soils, water resources and atmospheric air.

Environmental issues related to the storage and disposal of slags are coming to the fore. The leaching of toxic elements from slag masses into groundwater, dusting of landfills, as well as the release of greenhouse gases under certain storage conditions are just some of the negative consequences. Due to stricter environmental regulations and increased public attention to issues of sustainable development, the search for effective methods of slag processing is becoming a priority.

In the context of the cyclical economy and the desire for waste-free production, the processing of metallurgical slags

opens up new opportunities for the extraction of valuable components and their reuse. Slags contain a wide range of valuable various elements that can be returned to the production cycle or used in other industries [8, 9, 10].

Today, there are various technologies for processing autogenous smelting slags, ranging from simple crushing methods and use in road construction to more complex processes involving the extraction of valuable components such as non-ferrous and rare metals [11, 12]. The choice of a specific technology depends on the chemical composition of the slag, its physical properties, and economic feasibility. However, despite the variety of solutions offered, the problem of efficient and cost-effective slag processing remains relevant, requiring further research and development.

Industrial slags formed as a result of melting a complex copper-containing charge in a furnace, as well as stored slags, are a significant source of secondary raw materials. Reducing enrichment is a promising method of slag processing aimed at extracting valuable components such as non-ferrous and precious metals, mineral phases, and preparing them for further use.

The research carried out in the work is aimed at the development and optimization of reduction depletion technologies for various types of industrial slags. The developed technology of two-stage reduction processing of slags will

increase the efficiency of extraction of valuable components and reduce energy consumption, and is an important area of resource conservation and sustainable production development. The introduction of technology for processing VF slags in the conditions of the Balkhash Copper Smelter (BCS) will not only reduce the negative impact on the environment, but also ensure the economically beneficial use of secondary resources.

Currently, six large Vanyukov furnaces are successfully operating in Russia and Kazakhstan, processing over 2.5 million tons per year of copper, copper-nickel sulfide ores and concentrates, with an annual capacity of about 500 thousand tons of copper. Technological and design solutions were developed and tested at Vanyukov's experimental furnaces located in Norilsk, the Urals (SUCS) and Balkhash (the Kazakhmys Corporation Copper Smelter).

The versatility of purge mode control allows the Vanyukov furnace to be used for processing both copper-rich and zinc-containing slags formed during lead production. The technology remains applicable to different types of slags, requiring only minor adjustments.

The Vanyukov process, based on emulsion pyrometallurgy, has a number of advantages: simple and economical preparation of raw materials, eliminating drying, grinding and mixing; high productivity, reaching 4 tons of charge per hour per 1 m² of furnace cross-section, due to mixing of matte with oxygen-air blast (O₂ content from 40 to 96%, consumption up to 1000 m³/(m²·h)); intensification of heat and mass transfer, ensuring thermodynamic phase equilibrium and reliable forecast of raw material processing indicators; the possibility of creating multi-chamber devices for solving complex tasks.

The process is effective for the two-stage processing of slags formed in the production of copper and lead, to produce matte, lead-zinc sublimates and demetallized slag. The resulting slags, which are difficult to process using other processing methods, comply with the requirements of the Romelt process, developed at the Moscow Institute of Steel and Alloys and similar to the Vanyukov process.

The Romelt process can be used for processing iron silicate slags obtained after two-stage processing of copper-produced slags. This will make it possible to combine the extraction of copper, lead and zinc in the Vanyukov furnace with the production of an iron-carbon alloy from depleted slag and ensure waste-free technology.

The choice of the Vanyukov process for complex slag processing is due to:

- its suitability for processing iron-containing materials with a low content of iron oxides containing copper and volatile metals;
- using coal and natural gas as a fuel and reducing agent;
- the possibility of integrating the Vanyukov furnace with copper and lead smelting furnaces, as well as with the Romelt furnace.

The integrated resource-saving technology proposed in the study is focused on the organization and expansion of production for processing the generated and stored industrial slags of the BCS.

The key goal is the complex extraction of valuable metals into target products and the expansion of the range of marketable products.

To achieve this goal, in this work, the patterns of distribution of Cu, Pb, Zn, and As between the products of VF slag smelting under deeply reducing conditions, in relation to the conditions of the BCS, are investigated.

The study of the distribution patterns of elements such as Cu, Pb, Zn, and As in the products of VF slag smelting is important for optimizing the technological process. Understanding how these elements behave under deeply reducing conditions allows us to develop effective methods for their extraction and separation, as well as minimize the loss of valuable components.

The results of the research conducted in relation to the conditions of the BCS will make it possible to adapt the Vanyukov process to the specific characteristics of the processed slags and achieve maximum efficiency in extracting valuable metals. The introduction of integrated resource-saving technology will allow BCS not only to solve environmental problems related to slag storage, but also to make additional profits through the production of new types of products.

2. Materials and methods

The experiments were carried out with VF slag from the Balkhash Copper Smelter (BCS) of the composition, % by weight: 2.1 Cu; 1.02 Pb; 4.22 Zn; 0.1 As; 38.4 Fe; 30.3 SiO₂; 4.1 CaO; 5.1 Al₂O₃; 0.5 MgO; others.

To obtain the optimal slag composition, % by weight: 30 SiO₂; 50 FeO; 10 CaO; others, it was adjusted by adding lime.

To purge the slag melt, natural gas was used with a content, % vol.: 92.6 CH₄; 4.07 C₂H₆; 1.07 C₃H₈; others. Shubarkul coal with a content of 74% carbon was used as a solid reducing agent.

X-ray phase analysis of powder slag samples was performed using a D8 Advance device (Bruker AXS GmbH), α-Cu, X-ray tube voltage 40/40. The processing of the obtained diffractogram data and the calculation of interplane distances were carried out using EVA software. The samples were decrypted and the phases were searched using the Search/match program using the ASTM Card Database.

The elemental analysis of slag samples was determined using an inductively coupled plasma mass spectrometer Agilent 7700 Series ICP-MS (USA).

A Neophot microscope (Carl Zeiss AG, Germany) is used for mineralogical studies of polymetallic samples.

A distinctive feature of the conducted research is the implementation of a two-stage depletion of high-sulfur slags under deeply reducing conditions of purging the slag melt with natural gas in the presence of coal.

The method of conducting the experiments was as follows. At the first stage, a beryllium crucible with an initial slag sample of 100 g was loaded into a quartz reactor and placed in a heating furnace. After installation, the furnace was heated to a set temperature of 1300°C. From the moment the furnace was heated, continuous purging of the reactor's free zone with natural gas began. The position of the alund purge tube is above the melt. After the slag was melted, the blast tube was immersed in the melt, and its purging began. The consumption of natural gas was set based on the calculation of exceeding its consumption by 20% of the theoretically necessary for the reduction of arsenic oxide As₂O₅ to the volatile compound As₂O₃. The gas flow rate was monitored using a rotameter.

The experiments were carried out on the installation shown in Figure 1.

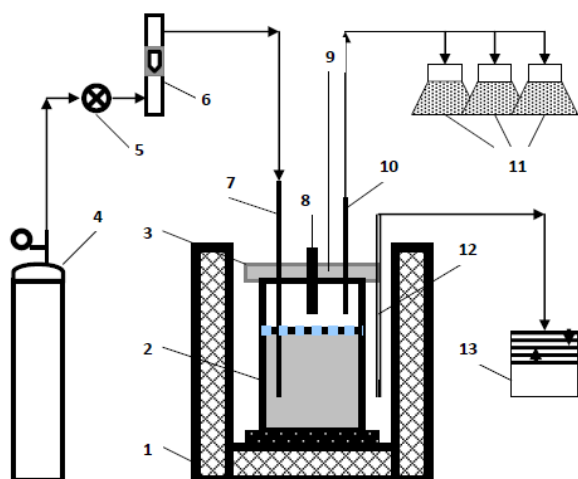


Figure 1. Schematic diagram of an installation for two-stage depletion of VF slags: 1 – Nabertherm GmbH RHTV 120-150/16 high temperature furnace; 2 – alund crucible; 3 – lid; 4 – natural gas cylinder; 5 – valve; 6 – rheometer; 7 – tube for supplying gas to the melt; 8 – coal loading window; 9 – slag melt; 10 – gas discharge; 11 – vessels with water; 12 – thermocouple PP-1; 13 – KSP-4

Preliminary experiments have shown that after purging the melt with natural gas for 5-10 minutes, a high extraction of arsenic into the gas phase is achieved. It was also found that during the prescribed time, the residual arsenic content in the slag reaches a minimum value and remains practically at the same level, regardless of the time of further purging.

The compositions of the slags did not undergo significant changes after their treatment with natural gas. The obtained results served as the basis for achieving the equilibrium of reducing depletion of slag by natural gas for arsenic. According to the results of preliminary experiments, the optimal time for purging slag with natural gas at the first stage for all subsequent experiments was assumed to be 10 minutes.

After a predetermined time of purging the slag with natural gas, the tube was removed from the melt and the second stage of reducing depletion of the slag began. The method of conducting the experiments was as follows. A pre-crushed coal powder was loaded onto the surface of the melt at a rate exceeding the set flow rate from the stoichiometrically required amount (SRA) for the reduction of lead and zinc oxide. Next, a purge tube was lowered into the melt and purging with natural gas began. The gas consumption was selected based on the calculation of exceeding its consumption by 10, 20, 30, 40, 50 and 60% of the SRA to reduce lead and zinc oxides to a metallic state. Each experiment was repeated three times in order to achieve reliable results. After purging, the crucible with the melt was settled for 20 minutes to separate the slag and matte, after which the crucible was removed from the furnace. The smelting products (slag and matte) were subjected to elemental analysis and mineralogical studies.

3. Results and discussion

The results of the elemental analysis of the slags obtained after purging the melt with natural gas at the first stage showed a slight decrease in the content of lead in the slags – from 1.02 to 0.9% and zinc – from 4.22 to 4.03%.

Figure 2 shows the dynamics of changes in the As content in the slag during purging of the melt with natural gas.

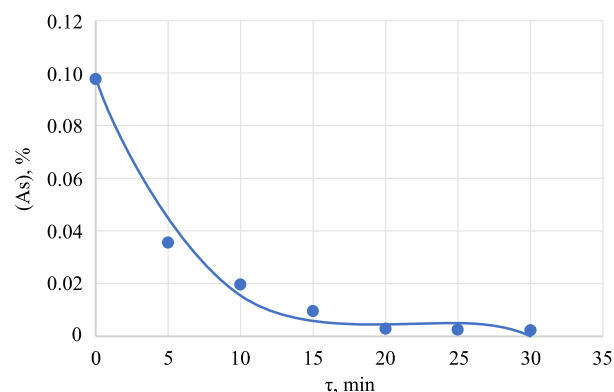


Figure 2. Dynamics of changes in the As content in the slag during purging of the melt with natural gas

It was found that even when purging the melt for 10 minutes, almost complete removal of arsenic from the slag is achieved.

The achievement of thermodynamic equilibrium between the slag melt and the matte at the second stage of slag depletion was determined by the stabilization of the gas phase. Preliminary experiments have shown that a state close to the thermodynamic equilibrium of the slag–matte system is achieved within 20 minutes. With an increase in natural gas consumption from SRA, the recovery of PbO and ZnO is enhanced by the influence of both coal and natural gas. At the same time, the extraction of lead and zinc from slag into dust increases dramatically (Figure 3).

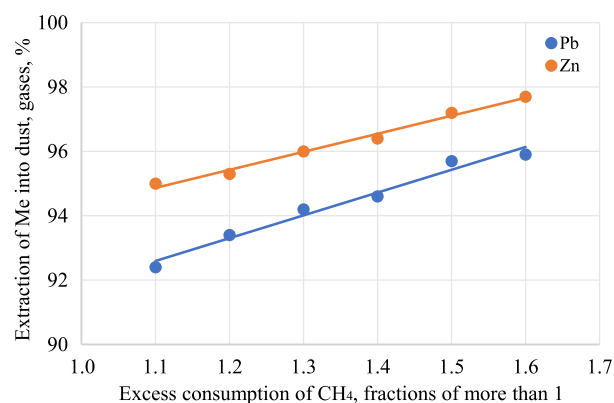


Figure 3. Dependence of the extraction of Pb, Zn into dust on the consumption of natural gas from the SRA, a fraction of more than 1

The resulting dusts are solid fine particles (sublimes) formed as a result of condensation of volatile metals and their compounds (oxides) from the vapor-gas phase with a decrease in temperature. With intensive continuous mixing of the melt with natural gas, favorable conditions are created for the coalescence of matte inclusions dispersed in the slag, which leads to an increase in copper extraction into the matter.

The results of mineralogical studies of the grinding of solid slag samples in each series of experiments showed the presence of ferrites in them, represented by compounds of iron oxides with calcium oxides, aluminum and, in a mini-

imum amount, copper oxide, in the form of delafossite (CuFeO_2). It was found that after depletion, the content of magnetite in the slags decreases from 15-18% to 5-7%. This indicates a deep reduction of magnetite by carbon from coal and natural gas.

The revealed dependencies are fully consistent with the results shown in Figures 2 and 3. With an increase in natural gas consumption exceeding the SRA for the reduction of lead, zinc and arsenic oxides, their maximum extraction into combustion is achieved, which, all other things being equal, increases dust yield. At the same time, the excess consumption of natural gas and carbon is spent on the recovery of slag magnetite. The destruction of magnetite is accompanied by the formation of iron oxide, which binds to silica in a stable phyllyte $\text{FeO} \cdot \text{SiO}_2$. The additional introduction of lime into the charge reduces the viscosity of the slag, which creates additional favorable conditions for reducing mechanical losses of copper with slag.

The yield of slag depletion products was, % of the weight of the initial charge: matte – 2.5; dust, gases – 10.4.

The dust yield in the experiments was determined by the difference between the amount of the corresponding metals in the initial slag and their amount in the resulting slag and matte (based on the calculation of the material balance of each experiment). The insignificant yield of matte is explained by the dominant extraction of copper from slag by reducing its mechanical losses.

The optimal consumption of natural gas, which ensures a minimum metal content in the slag, corresponds to a consumption exceeding its value from the SRA by 30% for the reduction of their oxides (Figure 4).

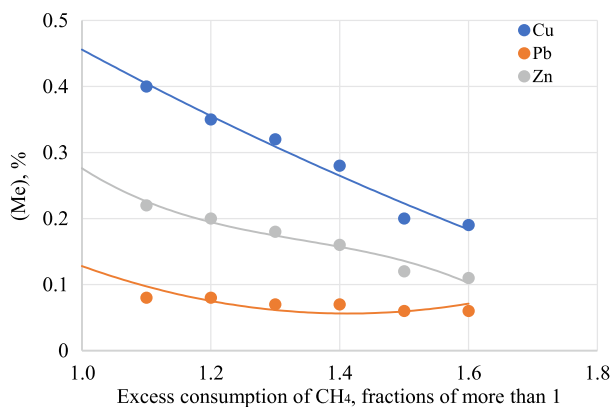


Figure 4. Dynamics of changes in the content of Cu, Pb, Zn in slag depending on the consumption of natural gas

At the same time, the minimum content of copper in the slag (0.35%) and lead, zinc (0.08% and 0.18%, respectively) is achieved.

Assay analysis of depleted slag for the content of Au and Ag in it showed their minimum values: 0.13 and 7.4 g/t, which indicates their maximum extraction into matte as a result of coalescence of mechanical suspensions of matte.

Based on the conducted experiments and the results of the material balance of each experiment, the distribution of non-ferrous metals and arsenic between the products of reducing depletion of slag by natural gas in the presence of coal is calculated, depending on the consumption of natural gas. The results are shown in Figure 5.

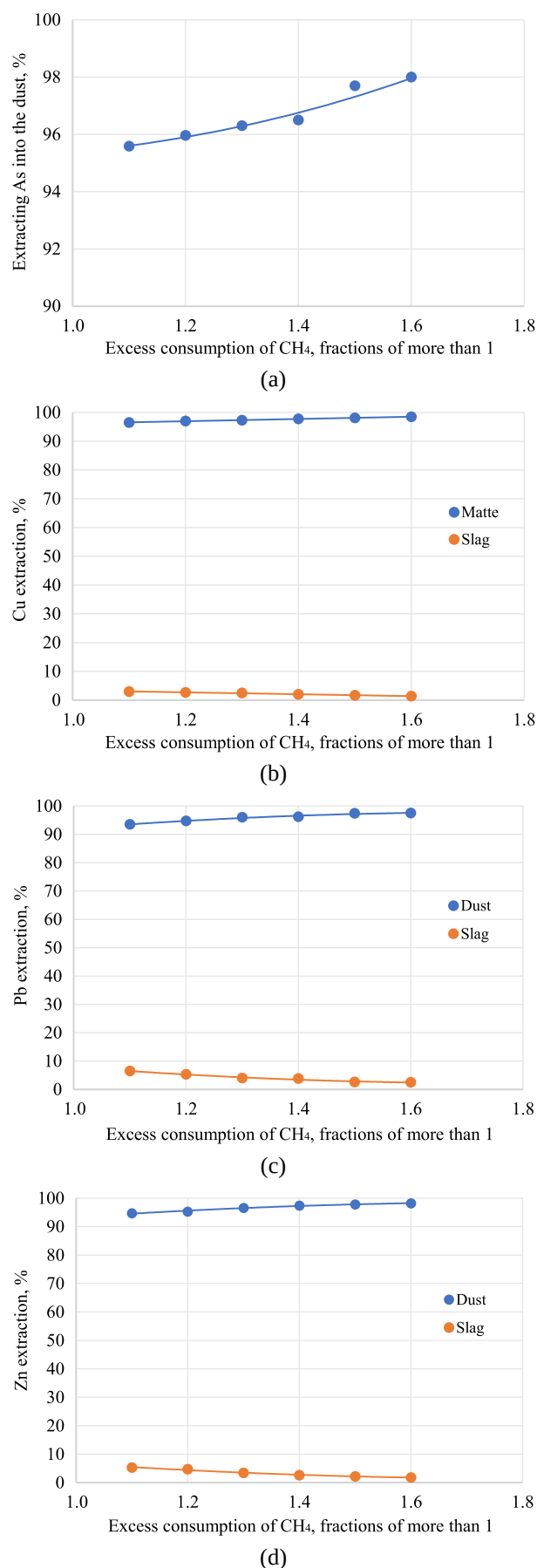


Figure 5. Distribution of elements between the products of reducing depletion of slags by natural gas in the presence of coal: (a) As; (b) Cu; (c) Pb; (d) Zn

The established optimal technological parameters of the process of reducing depletion of high-sulfur slags by natural gas in the presence of coal at a temperature of 1300°C meet the following criteria:

- The duration of slag purging with natural gas at the first stage is 10 minutes. The gas consumption exceeds by 10% its consumption from the SRA for the reduction of stable arsenic (V) oxide to its highly volatile compound, As_2O_3 ;

- The duration of the second stage of slag purging with natural gas in the presence of solid coal is 20 minutes:

- a) coal consumption is 10% more than the SRA for the reduction of lead oxide, zinc;

- b) the consumption of natural gas is 30% higher than the SRA for the recovery of magnetite and for intensive bubbling of the melt for coalescence of fine matte suspensions.

With the specified technological parameters, high technological indicators of the process of reducing depletion of slag have been achieved:

- the matte yield is 2.5% of the weight of the charge;
- the output of dust and gas is 10.4% of the weight of the charge;

- metal content in the recovered slag: copper – 0.35%; lead – 0.08%; zinc – 0.18%; arsenic – traces.

- extraction of metals: copper in matte – up to 98%; gold, silver in matte – 96%; arsenic in the dust – up to 99% (removed in the first stage of the technology by purging the slag with natural gas); lead, zinc in dust – up to 98% (removed in the second stage of the technology).

4. Conclusions

The results obtained indicate the high efficiency of the process of reducing depletion of slag, which makes it possible to extract valuable metals and remove harmful impurities. The low content of copper, lead and zinc in the recovered slag indicates a deep degree of depletion, which minimizes metal losses and reduces the environmental burden.

The high extraction of copper in matte, reaching 98%, demonstrates the efficiency of the process in terms of the concentration of copper in the valuable product. The extraction of gold and silver in matte at the level of 96% confirms the possibility of complex slag processing with the extraction of precious metals.

Selective removal of arsenic into dust, reaching 99%, is an important environmental advantage of the technology. The removal of arsenic in the first stage of the process using natural gas avoids its distribution to other products and reduces environmental pollution.

High extraction of lead and zinc into dust, reaching 98%, also helps to reduce the environmental burden and allows dust to be recycled to extract these valuable metals. The obtained indicators allow us to consider this technology as an effective and environmentally safe way of processing non-ferrous metallurgy slags.

Author contributions

Conceptualization: ND; Data curation: BS; Formal analysis: EZ; Funding acquisition: ND; Investigation: BS; Methodology: MK; Project administration: ND, BS; Resources: ND, BS; Software: EZ, MK; Supervision: ND; Validation: ND; Visualization: ND, EZ; Writing – original draft: ND, BS; Writing – review & editing: EZ, MK. All authors have read and agreed to the published version of the manuscript.

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Conflicts of interests

The authors declare no conflict of interest.

Data availability statement

The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

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Балқаш мыс балқыту зауыты жағдайында Ванюков пеші қождарының тотықсыздандырылған балқыту өнімдері арасында Cu, Pb, Zn және As-тың таралуы

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Андатпа. Балқаш мыс балқыту зауыты (БМЗ) жағдайында Ванюков пеші (ВП) қождарының тотықсыздандырылған балқыту өнімдері арасында Cu, Pb, Zn және As элементтерінің таралу нәтижелері келтірілген. 1300°C температурада көмірдің қатысуымен табиғи газды пайдалана отырып, қождарды екі сатылы тотықсыздандырып кедейлендіру процесінің оңтайлы технологиялық параметрлері анықталған. ВП қождарын екі сатылы тотықсыздандырып кедейлендіру жағдайында тотықсыздандырылған қождағы металдардың қалдық мөлшері: мыс – 0.35%; қорғасын – 0.08%; мырыш – 0.18%; мышьяк – іздік мөлшерде болатыны көрсетілген. Металдардың мақсатты өнімдерге жоғары дәрежеде алынуына қол жеткізді: мыстың штейнге алынуы – 98%-ға дейін; алтын мен күмістің штейнге алынуы – 96%; қожды табиғи газбен өңдеудің бірінші сатысында мышьяқтың шаңға алынуы – 99%-ға дейін, ал екінші сатыда қорғасын мен мырыштың шаңға алынуы – 98%-ға дейін. Алынған нәтижелер БМЗ-ның ағымдағы және жинақталған қождарын тауарлық өнімдер ала отырып кешенді, қалдықсыз өңдеудің инновациялық технологиялық платформасын әзірлеу барысында пайдаланылды.

Негізгі сөздер: ВП шлактары, тотықсыздандыру, көмір, табиғи газ, бөліп алу, бағалы металдар, газ және көмір шығыны.

Распределение Cu, Pb, Zn и As между продуктами восстановительной плавки шлаков печи Ванюкова в условиях Балхашского медеплавильного завода

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Аннотация. Приведены результаты по распределению Cu, Pb, Zn и As между продуктами восстановительной плавки шлаков Печи Ванюкова (ПВ) в условиях Балхашского медеплавильного завода (БМЗ). Установлены оптимальные технологические параметры процесса двухстадийного восстановительного обеднения шлаков природным газом в присутствии угля при температуре 1300°C. Показано, что в условиях двухстадийного восстановительного обеднения шлаков ПВ остаточное содержание металлов в восстановленном шлаке составляет: меди – 0.35%; свинца – 0.08%; цинка – 0.18%; мышьяка – следы. Достигнуто высокое извлечение металлов в целевые продукты: меди в штейн – до 98%; золота, серебра в штейн – 96%; мышьяка в пыль на первой стадии обработки шлака природным газом – до 99% и свинца, цинк в пыль во второй стадии – до 98%. Полученные результаты использованы при разработке инновационной технологической платформы комплексной безотходной технологии переработки текущих и накопленных шлаков БМЗ с получением товарных продуктов.

Ключевые слова: шлак ПВ, восстановление, уголь, природный газ, извлечение, ценные металлы, расход газа и угля.

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