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Metallurgy

Enlarged tests on the processing of copper-lead mattes obtained after reductive smelting of balanced feed charge

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ABSTRACT

The paper examines the behavior of copper, lead, zinc and arsenic during the oxidative blowing of intermediate copper-lead matte, which represents the second stage of the general technology for processing balanced raw materials for copper and lead production. The optimal parameters for the oxidative blowing of intermediate matte have been established: the time of blowing the melt with oxygen is 20 min; the oxygen consumption is 1.4 times higher than its consumption from the stoichiometrically required amount for the oxidation of zinc and iron sulfide; the temperature is 1250 °C. High indicators have been achieved for the complex selective extraction of metals into targeted products: lead into rough lead – 97.6%; copper into matte – 98.6%; zinc into slag – 56.8%, into matte – 1.7, into dust and gases – 41.5; arsenic and antimony into dust – up to 97.4% and 90%, respectively. A general process flowsheet has been developed for separate processing of balanced charges consisting of intermediate products of copper and lead production. The technology can be used for separate processing of multi-component raw materials of copper smelters and lead production of various types and compositions.

Keywords: copper-lead matte, copper, lead, zinc, arsenic, oxidizing blowing, melting, distribution.

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Introduction

The growth of volumes of substandard intermediate products and recycled materials of lead production necessitates the search for new technologies for their processing. At present, in lead and copper production, in connection with the involvement of complex multi-component raw materials in the processing, the yield of intermediate products with a high content of impurity metals, such as lead, zinc, arsenic, etc., is sharply increasing [[1], [2], [3], [4], [5], [6]]. The use of this type of raw material in processing, particularly in lead production plants, has resulted in an increase in the yield of intermediate products and recycled materials containing higher levels of arsenic, antimony, and their toxic compounds. Currently, none of the processes in the lead production technological chain achieve a sufficiently high level of sublimation of arsenic and antimony into dust. This shortcoming limits their removal from the main production cycle, leading to a significant accumulation within the facility. The existing technologies are no longer capable of effectively processing these materials, resulting in arsenic buildup, a notable rise in material costs, deterioration of technological performance, and an increase in health issues for communities living near metallurgical plants.

Special attention is drawn to the process of converting copper-lead matte, aimed at obtaining rough copper. The technology of converting copper matte is well developed in practice and is sufficiently fully covered in the technical literature. Nevertheless, the issues of increasing the extraction of copper into rough copper and improving the quality of copper-lead matte conversion products still remain open.

We did not have the task of conducting a detailed analysis of the conversion process of copper-lead matte produced at the Ust-Kamenogorsk Metallurgical Complex (UK MC) of Kazzinc LLP. The interest in considering the issue is caused by the fact that conversion can be considered as the final stage of processing an intermediate product – matte, obtained by separate processing of intermediates and recycled materials.

The processing of copper-lead matte at the UK MC is accompanied by a number of technological features, which we reported in previously published works. The conducted studies have established that the low extraction of copper into rough copper (~ 80%) is accompanied by an increased (up to 15%) distribution of copper into converter slag, and an insignificant (up to 5%) transition of it into dust.

In the process of converting copper-lead matte into rough copper, up to 1.5% of lead passes. The extraction of lead into dust is at a low level and is 40%. Up to 60% of the total amount of lead is concentrated in the converter slag. During conversion, galena present in matte is easily oxidized to form lead oxide and sulfurous anhydride. Lead oxide binds to silica to form lead silicate.

Zinc is distributed mainly between converter slag and dust: up to 80% of zinc is concentrated in converter slag.

The situation is somewhat different with the distribution of arsenic and antimony. During conversion, these impurities are distributed between converter slag, dust and rough copper. The main part of arsenic – up to 70%, passes into dust. 22% of arsenic is concentrated in the converter slag and ~ 7% of it is distributed into rough copper.

26.2% of antimony passes into rough copper, which is four times higher than that of arsenic. As a result of the low sublimation of antimony during conversion, its distribution into dust is insignificant, and amounts to only 40%. The remaining part of antimony - up to 36%, is concentrated in the converter slag.

From the results obtained, the following can be concluded. When converting copper-lead matte having a complex chemical and phase composition, it is not possible to achieve an optimal distribution of non-ferrous metals and related impurities (As, Sb) between the conversion products. In order to achieve optimal technological indicators for the distribution of metals during conversion, it is necessary either to significantly improve the quality of the matte before conversion, or already in the conditions of conducting the conversion process, to provide additional technological measures to improve the quality of products. In our opinion, the first option seems to be the most promising.

When organizing the technology of separate processing of intermediates and recycled materials, it is necessary to strive to obtain high-quality copper matte, ensuring a minimum content of Pb, Zn, As, Sb in it. In this regard, the existing technology at UK MC cannot be considered as a prospect for the future, and requires drastic changes.

One of the solutions to this problem is the development of technology for separate processing of copper- and lead-containing products, where various pyro- and hydrometallurgical processes are used [[7], [8], [9], [10], [11], [12], [13], [14], [15], [16], [17], [18]].

At KazNITU named after K.I. Satbayev, under the supervision of Professor Dosmukhamedov N.K., researchers have worked for several years with scientists from the Moscow Institute of Steel and Alloys (MISiS) and the Weizmann Institute of Science in Israel, Rehovot. They are conducting thorough research to develop new technology for processing intermediate products and recycled materials from copper and lead production. Based on the positive results of fundamental research, a comprehensive technology has been developed aimed at processing multi-component balanced raw materials and selective extraction of valuable metals into targeted products [[19], [20]].

The core *methodological principle* of the developed technology is a thermodynamic approach to modeling bubbling metallurgical processes. This approach ensures the accurate determination of the qualitative and quantitative characteristics of the resulting complex condensed (liquid) and vapor-gas phases, based on specific input parameters of the pyrometallurgical bubbling process. These parameters include the compositions and quantities of the initial materials, temperature conditions, oxidation-reduction potentials, and others.

The thermodynamic approach is grounded in the concept of reaching thermodynamic equilibrium (or a state near equilibrium) between the smelting products. This enables the application of chemical thermodynamics laws to predict the behavior of both the main and impurity components during the melting process. The basis for the possibility of using the equilibrium approach to describe processes occurring under conditions of intensive mixing of a multicomponent melt of complex composition are high rates of mass and heat transfer in them, and the conditions for the formation and separation of phases.

The basis for the developing of the general concept of the new bubbling technology is the separate **two-stage processing** of a balanced feed charge consisting of intermediate products and recycled materials from non-ferrous metallurgy.

At the first stage, selective separation of metals into targeted products is carried out in an electric furnace: copper into copper-lead matte; lead into rough lead; zinc into slag, and partial sublimation of rare, rare earth and dispersed metals into dust.

At the second stage, in an oxidation-reduction furnace with submerged tuyeres, a sufficiently deep extraction of lead (up to 98%) is carried out from copper-lead matte into rough lead, and copper is extracted into commercial copper matte with a high copper content (up to 70%). Zinc is concentrated in slag, and rare, rare earth and scattered metals pass into dust.

Studies on the behavior of non-ferrous and associated impurity metals during the smelting of a balanced charge in an electric furnace are covered in detail in [[19], [20]].

The aim of the present research is to study the distribution of copper, lead, zinc and arsenic between the products of oxidative blowing of intermediate copper-lead matte obtained after the reduction smelting of a balanced batch based on copper- and lead-containing products in an electric furnace.

Research methods

The object of the study is copper-lead matte obtained after reductive smelting of a balanced charge with natural gas of the following composition, % by weight: 54.6 Cu; 2.23 Pb; 12.4 Zn; 6.2 Fe; 0.19 As; 22.2 S.

Copper-lead matte was subjected to air blowing at different air consumption and time. Air consumption was calculated based on the condition of its consumption from the stoichiometric required amount (SRA) for complete oxidation of lead, zinc and iron sulfides.

In the experiments, the oxygen consumption varied within the range from 0.9 to 1.5 of the SRA, the melt blowing time was changed from 5 to 20 minutes. Based on the obtained results, the optimal process parameters were determined, ensuring high technological indicators for the extraction of non-ferrous metals into targeted commercial products.

The methodology for conducting the experiments was as follows.

A crucible containing an initial sample of 500 g was placed into a quartz reactor, which was then loaded into a furnace. Once the specified temperature of 1523 K was reached, the melt was held for 10 minutes to ensure homogeneity. Afterward, the melt was subjected to oxygen blowing for a specified duration. Oxygen consumption varied between 1 and 1.4 fractions of SRA for the oxidation of zinc and iron sulfides. The blowing time was set at 5, 10, 15, and 20 minutes. Once the oxygen blowing was completed, the tube was raised above the melt, and the furnace was allowed to cool. After cooling, the crucible with the sample was removed from the quartz reactor. The resulting smelting products, matte and slag, were separated and analyzed for metal content. The elemental composition of the melt products was determined using a scanning electron microscope equipped with a JED-2300 (JEOL) energy-dispersive X-ray spectrometer. The obtained diffraction data were processed, and the interplanar distances were calculated using the EVA software. Sample decoding and phase identification were performed with the Search/Match program, utilizing the PDF-2 powder diffractometric database for reference.

In order to obtain accurate data on the elemental composition of dust, an additional analysis was carried out using a D8 Advance spectrometer.

The dust yield in all experiments was determined by calculating the difference between the initial sample weight and the combined weight of the obtained smelting products.

Each experiment was repeated three times for reproducibility. The results of parallel experiments on the content of metals in the smelting products showed good convergence (error +/- 0.5% abs.).

Based on the average results of product yield and metal content in them, the material balance of the oxidative smelting of copper-lead matte was calculated.

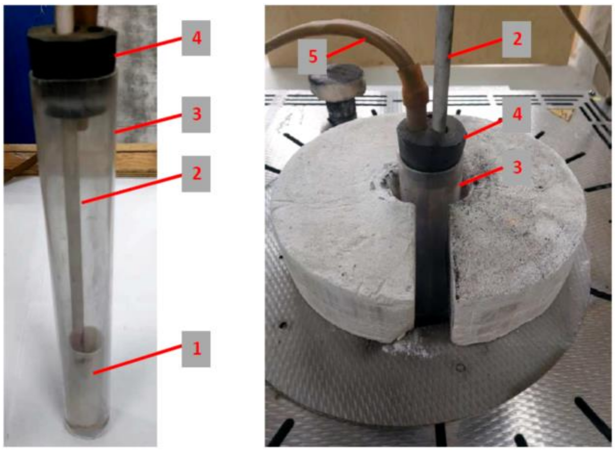
Results and discussion

The general view of the enlarged laboratory setup is shown in Fig. 1.

Table 1 shows the calculated material balance of the oxidative smelting of copper-lead matte with oxygen.

During oxidative smelting of matte with air, the yield of products was, % (of the total charge): matte – 49.8; slag – 12.1; dust, gases – 38.1.

The patterns of distribution of copper, lead, zinc and arsenic between smelting products under conditions of oxidative blowing of matte are shown in Fig. 2-5.



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

Figure 1 - General view of the installation for reaching final copper-lead matte

Table 1 – Material balance of oxidative smelting of intermediate matte with air at optimal parameters: oxygen consumption – 1.4 times exceeding its consumption from the SRA for oxidation of lead, iron and zinc sulfides; blowing time – 20 min.; T = 1250 °C

Products name	Quantity		Cu			Pb			Zn			Fe			As		
	g.	%	I	II	III	I	II	III	I	II	III	I	II	III	I	II	III
Loaded:																	
Matte	115.83	68.9	64.0	55.3	100.0	2.6	2.3	100.0	14.5	12.5	100.0	6.3	5.5	100.0	0.15	0.1	100.0
Air	44.53	26.5															
Flux	7.81	4.6															
Total:	**168.18**	**100.0**	**64.0**		**100.0**	**2.6**		**100.0**	**14.5**		**100.0**	**6.3**		**100.0**	**0.15**		**100.0**
Received:																	
Matte	83.84	49.8	63.9	76.3	99.9	0.8	0.9	30.0	0.4	0.5	3.0	0.2	0.2	3.0	0.05	0.1	30.0
Slag	20.34	12.1	0.1	0.1	0.1				2.8	13.6	19.0	6.2	30.3	97.0			
Dust. gases	64.00	38.1				1.8	2.9	70.0	11.3	17.7	78.0				0.10	0.2	70.0
Total:	**168.18**	**100.0**	**64.0**		**100.0**	**2.6**		**100.0**	**14.5**		**100.0**	**6.3**		**100.0**	**0.15**		**100.0**

Products name	Sb			S			O			N_2			SiO_2			Others			Total:
	I	II	III	I	II	III	I	II	III	I	II	III	I	II	III	I	II	III	
Loaded:																			
Matte	0.03	0.0	100.0	25.5	22.0	100.0										2.7	2.3	100.0	115.8
Air							9.4	21.0	100.0	35.2	79.0	100.0							44.5
Flux													5.8	74.8	100.0	2.0	25.2		7.8
Total:	**0.03**		**100.0**	**25.5**		**100.0**	**9.4**		**100.0**	**35.2**		**100.0**	**5.8**		**100.0**	**4.7**		**100.0**	**168.18**
Received:																			
Matte	0.03	0.03	100.0	17.1	20.4	67.0										1.3	1.6	28.9	83.8
Slag							2.9	14.1	30.6				5.8	28.7	100.0	2.7	13.1	57.0	20.3
Dust. gases				8.4	13.1	33.0	6.5	10.1	69.4	35.2	55.0	100.0				0.7	1.0	14.1	64.0
Total:	**0.03**		**100.0**	**25.5**		**100.0**	**9.4**		**100.0**	**35.2**		**100.0**	**5.8**		**100.0**	**4.7**		**100.0**	**168.18**

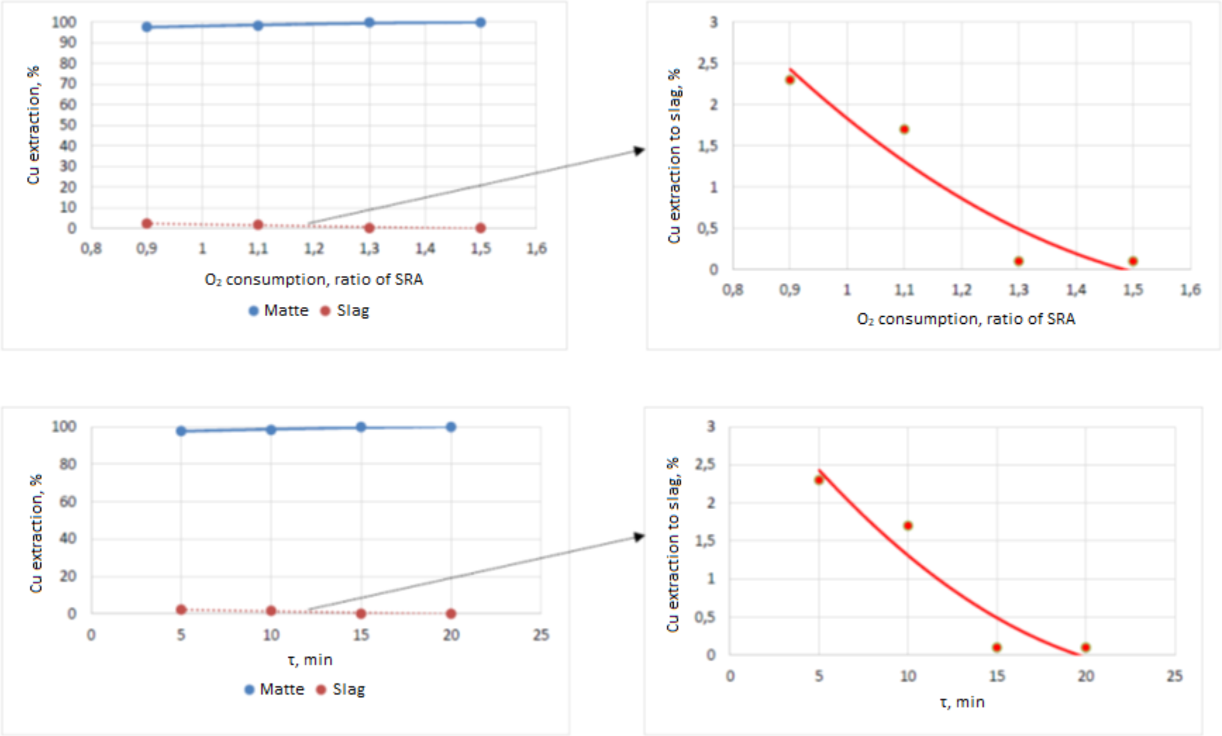


Figure 2 – Effect of oxygen consumption (ratio of SRA for oxidation of Pb, Zn, Fe sulfides) and the melt blowing time (τ, min) for copper extraction into smelting products

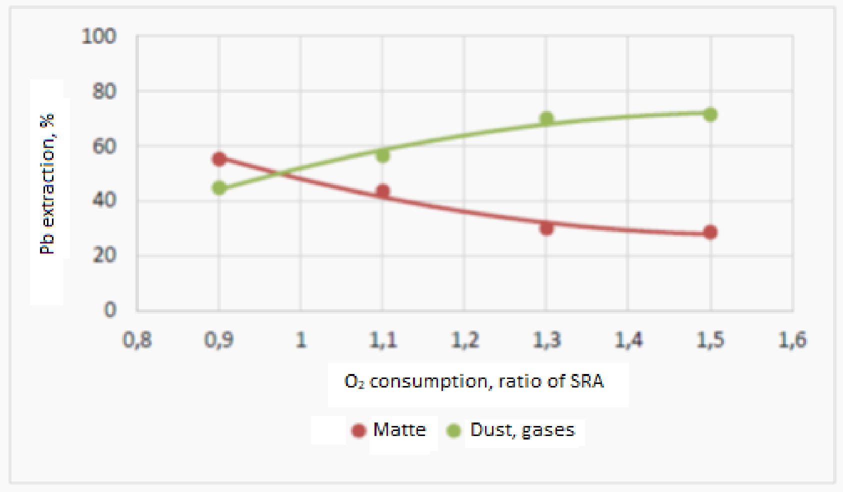


Figure 3 – Effect of oxygen consumption (ratio of SRA for oxidation of Pb, Zn, Fe sulfides) for the extraction of lead into smelting products

The best results for the complex selective extraction of copper, lead, zinc, and arsenic into the targeted smelting products were achieved with an oxygen consumption rate 1.4 times higher than the consumption from the SRA for the oxidation of zinc and iron sulfides, followed by their transfer as oxides into the slag. The matte blowing duration was set to 20 minutes. Under optimal process conditions, copper extraction into matte exceeded 98%, lead extraction into matte and dust was 30% and 70%, respectively, and zinc extraction into matte, slag, and dust was 3%, 19%, and 78%, respectively.

The results of the studies demonstrated the fundamental feasibility of separately processing copper-lead matte by intensively blowing it with air to produce commercial copper matte with a copper content of 76.3% (Table 1). These findings represent the final second stage in the overall technology for processing a balanced charge, which consists of copper-lead-containing intermediate products and recycled materials from copper and lead production. This process allows for the selective extraction of non-ferrous metals into targeted commercial products.

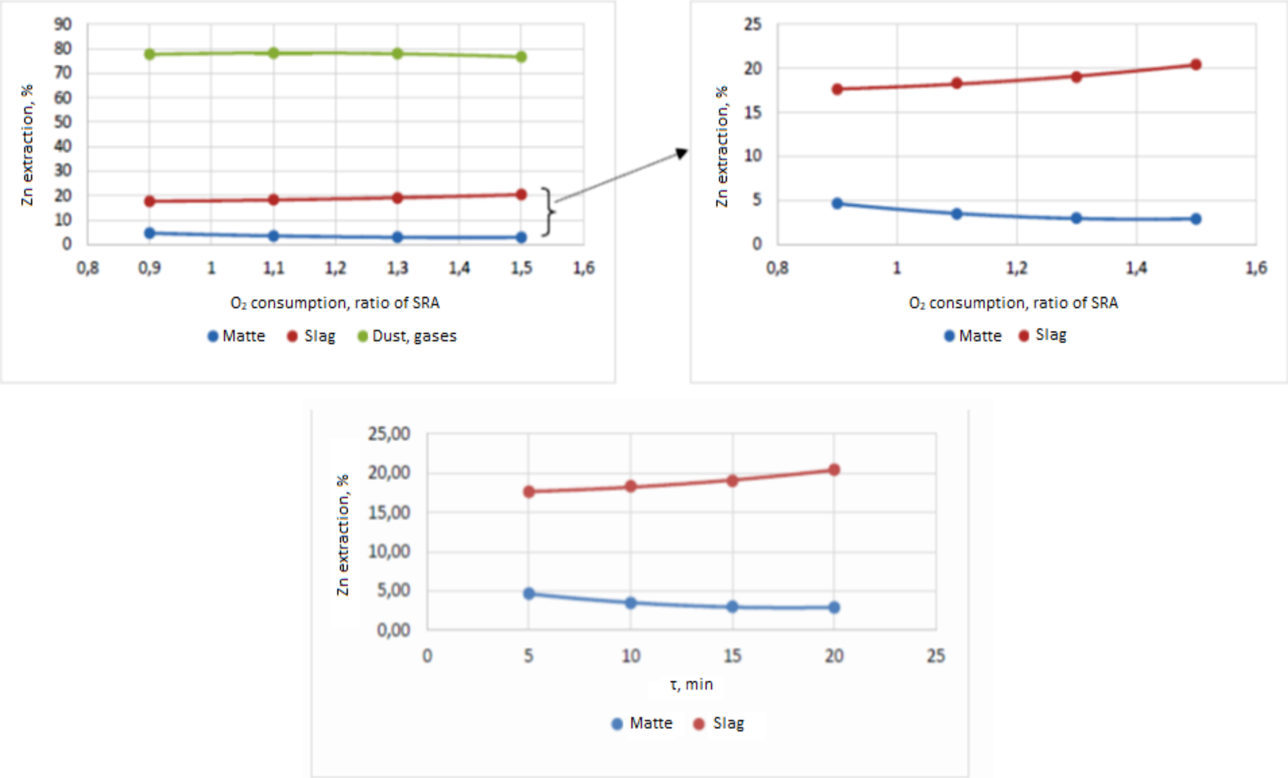


Figure 4 – Effect of oxygen consumption (ratio of SRA for oxidation of PbS, ZnS, FeS) and the melt blowing time (τ, min) for the extraction of zinc into the smelting products

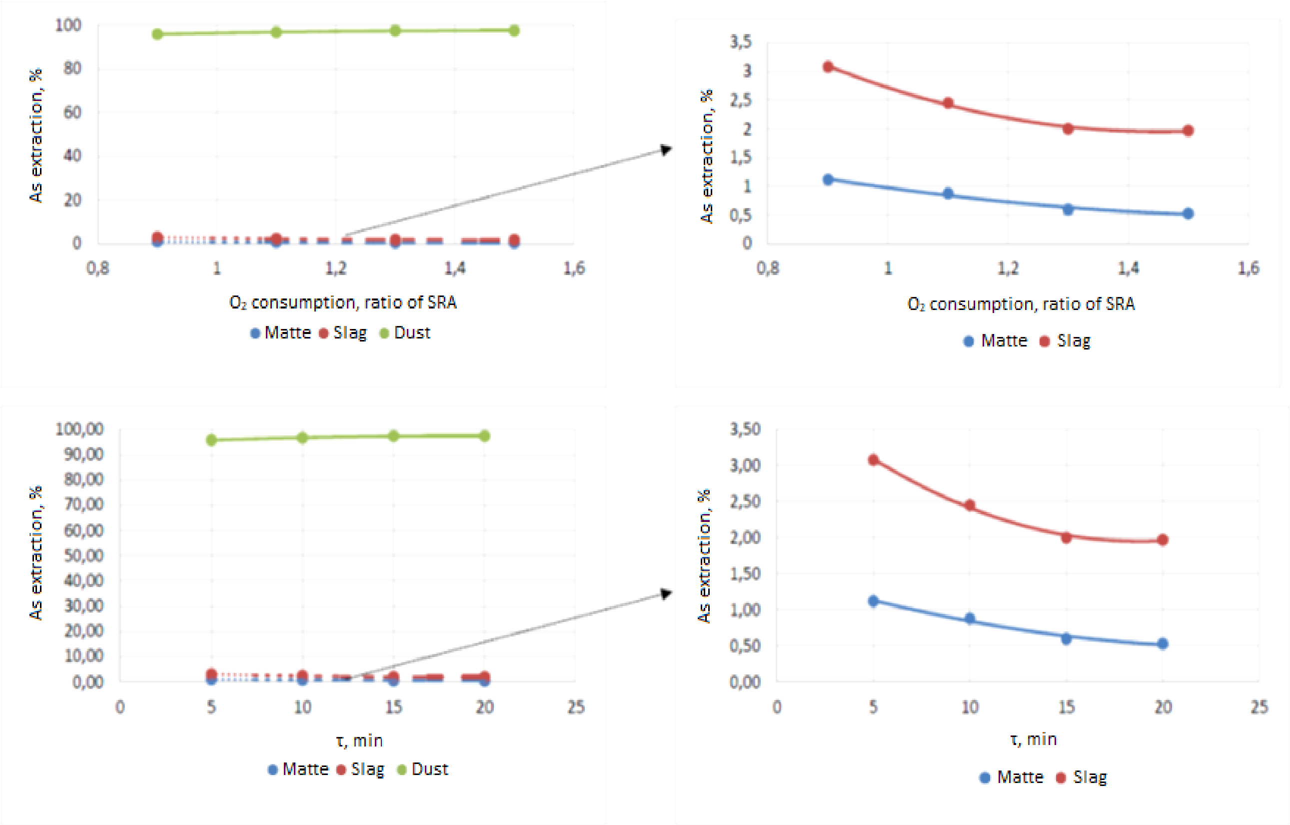


Figure 5 – Effect of oxygen consumption (ratio of SRA for oxidation of PbS, ZnS, FeS) and the melt blowing time (τ, min) for the extraction of arsenic into the melt products

For industrial implementation of the general reduction-oxidation technology for processing balanced charge the following parameters are recommended.

For the first stage – the recovery smelting of the balanced charge:

– melt blowing time:

- natural gas – 20 min;
- consumption of CH_4 is 1.7 times higher than its consumption from the stoichiometric required amount for the reduction of lead compounds;
- melting temperature – 1250 °C.

For the second stage – oxidative blowing of intermediate copper-lead matte:

- melt blowing time – 20 minutes;
- oxygen consumption is 1.4 times higher than its consumption from the stoichiometric required amount for the oxidation of zinc and iron sulfide;
- melting temperature – 1250 °C.

The implementation of the technology ensures the production of high-quality targeted products of the following compositions at the first stage:

- rough lead. % by weight: 99.34 Pb; 0.18 Cu; 0.08 Sb; other.
- Copper-lead matte, wt.%: 55.25 Cu; 2.25 Pb; 12.54 Zn; 5.48 Fe; 22.0 S; 0.13 As; 0.03 Sb; other.
- Slag, wt.%: 20.45 Fe; 18.58 SiO_2; 4.0 CaO; 8.33 Zn; 0.16 Cu; 1.11 Pb; others.

In the second stage:

- Copper -lead matte, wt.%: 76.3 Cu; 0.93 Pb; 0.52 Zn; 0.23 Fe; 20.4 S; 0.05 As; 0.03 Sb; other.
- Slag, wt.%: 30.3 Fe; 28.7 SiO_2; 6.0 CaO; 13.6 Zn; 0.19 Cu; others.

Total recovery of metals into targeted products:

– lead in rough lead – 97.6%;

– copper in matte – 98.6%;

- zinc:

into slag – 56.8%;

in matte – 1.7%;

into dust and gases – 41.5%;

– arsenic and antimony in dust – 97.4% and 90%, respectively.

The proposed general process flow diagram for the reduction-oxidation smelting of a balanced charge with the complex extraction of copper, lead and zinc into targeted commercial products is presented in Fig. 6.

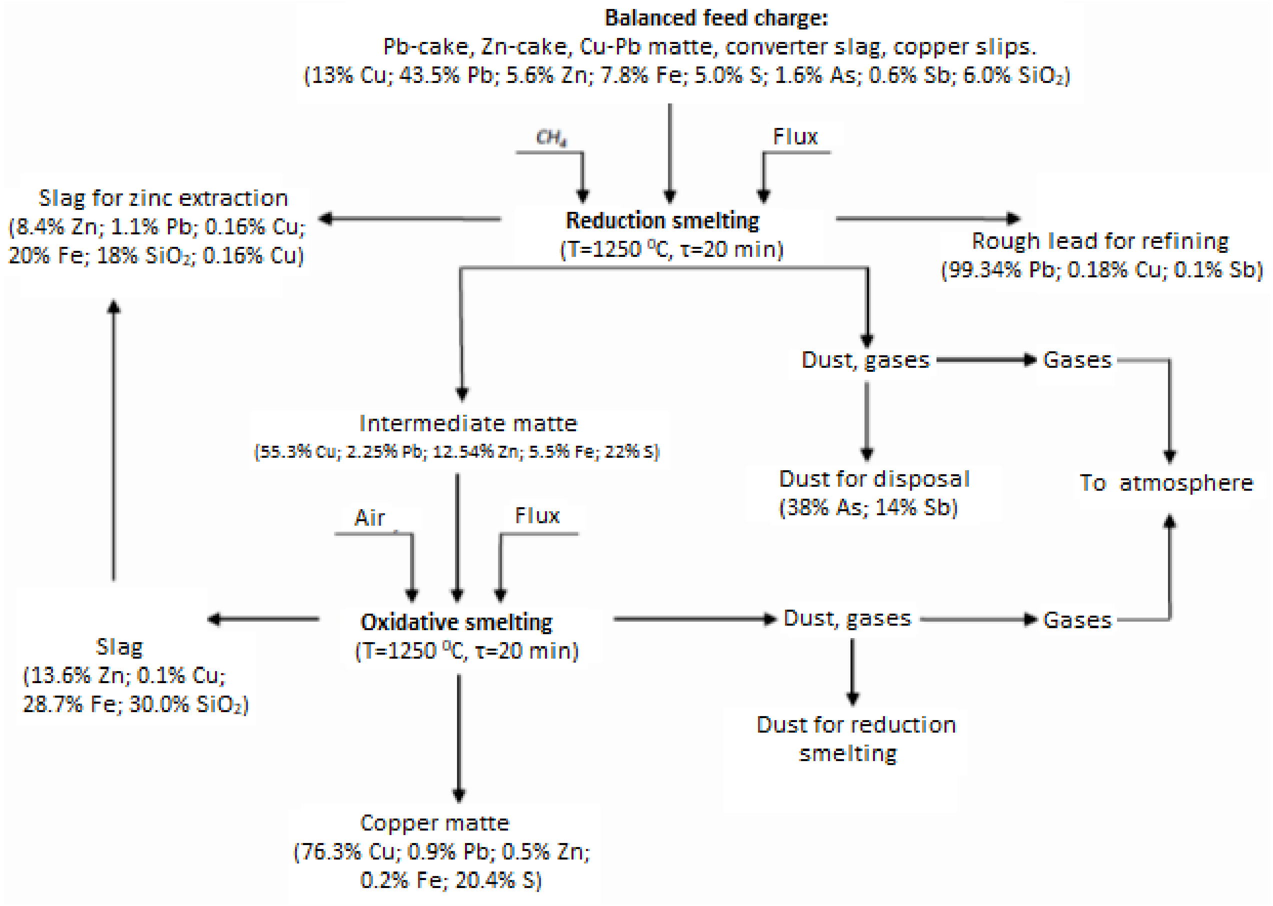


Figure 6 – Flow diagram of the technology for processing balanced charge, compiled on the basis of intermediate products and recycled materials of copper and lead production

The developed technology can be used for separate processing of balanced feed charge, composed on the basis of multi-component intermediate products and recycled materials of non-ferrous metallurgy of different types and compositions.

Conclusions

1. The feasibility of implementing reduction-oxidation smelting of a balanced charge has been demonstrated. The optimal technological parameters for oxidative blowing of intermediate matte have been established: the melt is blown with oxygen for 20 minutes, oxygen consumption is 1.4 times higher than its consumption from the SRA for the oxidation of zinc and iron sulfides, and the temperature is maintained at 1250 °C.

2. Under optimal process parameters, the following product yields were obtained as a percentage of the total charge: rough lead – 38.3%, copper matte – 15%, slag – 31.2%, and dust/gases – 15.5%. The copper matte produced had a high copper content (over 76%) with minimal impurities, including 0.93% Pb, 0.52% Zn, 0.23% Fe, 0.05% As, and 0.03% Sb.

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

4. Based on the collected data, a basic technological scheme for processing a balanced batch composed of a complex conglomerate of a mixture of intermediate products from copper and lead production was developed.

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

CRediT author statement: **N. Dosmukhamedov:** Supervision, Conceptualization; **E. Zoldasbay:** Investigation, Data curation; **A. Argyn:** Methodology, Writing-Original draft preparation; **Y. Icheva:** Investigation, Software, Validation; M**. Kurmanseitov:** Writing- Reviewing.

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Теңдестірілген шикіқұрамды тотықсыздандырып балқытқаннан кейін алынған мыс-қорғасын штейндерін қайта өңдеу бойынша ірілендірілген сынақтар

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ТҮЙІНДЕМЕ

Жұмыста мыс және қорғасын өндірісінің теңдестірілген шикізатын өңдеуге арналған жалпы технологияның екінші сатысындағы аралық мыс-қорғасын штейнін тотықтырып үрлеу кезінде мыс, қорғасын, мырыш және мышьяктың бөлініп таралуы зерттелді. Аралық штейнді тотықтырып үрлеудің оңтайлы параметрлері белгіленді: балқыманы оттегімен үрлеу уақыты – 20 мин; оттегі шығыны – мырыш пен темір сульфидін тотықтыру үшін СҚМ-рі шығыннан 1,4 есе артық; температура-1250 °С. Мақсатты өнімдерге металдарды кешенді селективті бөліп алу бойынша: қорғасынды тазартылмаған қорғасынға – 97,6%; мысты штейнге – 98,6%; мырышты қожға – 56,8%, штейнге – 1,7%, шаңға, газдарға – 41,5%; мышьяк пен сурьманы шаңға – сәйкес 97,4% және 90% дейінгі жоғары көрсеткіштерге қол жеткізілді. Мыс және қорғасын өндірісінің жартылай өнімдерінен тұратын теңдестірілген шикіқұрамды бөлек өңдеуге арналған жалпы технологияның сұлбасы әзірленді. Технологияны мыс балқыту және қорғасын өндірісінің көп компонентті шикізатының түрі мен құрамы бойынша бөлек өңдеу үшін пайдалануға болады.

Түйін сөздер: мыс-қорғасын штейні, мыс, қорғасын, мырыш, мышьяк, тотықтырып үрлеу, балқыту, бөлініп таралу.

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Укрупненные испытания по переработке медно-свинцовых штейнов, полученных после восстановительной плавки сбалансированной шихты

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АННОТАЦИЯ

В работе исследовано поведение меди, свинца, цинка и мышьяка при окислительной продувке промежуточного медно-свинцового штейна, представляющей вторую ступень общей технологии для переработки сбалансированного сырья медного и свинцового производства. Установлены оптимальные параметры окислительной продувки промежуточного штейна: время продувки расплава кислородом – 20 мин; расход кислорода – 1,4 раза превышающий его расход от СНК для окисления сульфида цинка и железа; температура – 1250 °C. Достигнуты высокие показатели по комплексному селективному извлечению металлов в целевые продукты: свинца в черновой свинец – 97,6%; меди в штейн – 98,6%; цинка в шлак – 56,8%, в штейн – 1,7, в пыль, газы – 41,5; мышьяка и сурьмы в пыль – до 97,4% и 90%, соответственно. Разработана общая технологическая схема для раздельной переработки сбалансированной шихты, составленной из полупродуктов медного и свинцового производства. Технология может быть использована для раздельной переработки различного по типу и составу многокомпонентного сырья медеплавильного и свинцового производства.

Ключевые слова: медно-свинцовый штейн, медь, свинец, цинк, мышьяк, окислительная продувка, плавка, распределение.

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