

UDC 669-1

TO THE CHOICE OF TECHNOLOGY FOR PROCESSING LEAD DUST OF COPPER PRODUCTION: ANALYSIS OF METHODS OF DUST RECYCLING

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Among the most important priorities for the innovative development of the mining and metallurgical industry of Kazakhstan the industrial development of technologies for processing lead converter dust from copper and lead production is included. An important prerequisite for the organization of a separate production for the processing of lead dusts of Kazakhmys Corporation LLP and Kazzinc LLP is their multicomponent nature and value, which, along with non-ferrous metals, contain dual-use metals such as rhenium, osmium, selenium and others. The content of rhenium in Zhezkazgan dust reaches 120 g/t and serves as a source of its extraction into commercial ammonium perchlorate. To organize production for the processing of arsenic-containing lead dust of Kazakhmys Corporation LLP with the production of targeted products from them, cost-effective and environmentally friendly technologies are needed that increase the technical level of production of marketable and intermediate products. Finding a rational technology for their processing is one of the key problems that need to be addressed. An increase in the content of arsenic in dust (up to 10%) makes a challenge to the development of new highly efficient technologies for processing dust with the extraction of valuable metals. In this paper, based on a brief analysis of the known pyro- and hydrometallurgical methods of dust processing, the main problems and possible solutions are shown. The review allows us to develop a general concept and outline new directions for the complex processing of dust.

Keywords: lead dust, pyrometallurgical methods, hydrometallurgy, arsenic, lead cake, rhenium, precious metals

The research was carried out within the framework of grant funding from the Science Committee of the Ministry of Science and Higher Education of the Republic of Kazakhstan for 2023-2025 in the priority area «Geology, mining and processing of mineral and hydrocarbon raw materials, new materials, technologies, safe products and designs» of the project AP19676951 «Development of resource saving, combined technology for the complex processing of multicomponent non-ferrous metallurgy dust with the production of marketable products.

One of the urgent tasks of the sustainable development of non-ferrous metallurgy in Kazakhstan is the organization of new industries (technologies) aimed at the complex processing of substandard intermediate products and recycling materials of copper and lead production with the maximum extraction of non-ferrous and associated valuable metals.

Among the most important priorities for the innovative development of the mining and metallurgical industry of Kazakhstan the industrial development of technologies for processing lead converter dust from copper and lead production is listed.

An important prerequisite for organizing a separate lead dust processing technology of Kazakhmys Corporation LLP is their multicomponent nature and value. Lead dust, along with non-ferrous metals (Pb, Zn, Cu, etc.), also contains a number of other strategically important dual-use valuable metals: rare metals, in particular, rhenium, osmium, selenium, tellurium, thallium, and others [1].

To organize production for the processing of lead dust of Kazakhmys Corporation LLP

with the production of commercial and targeted products from them, cost-effective and environmentally friendly technologies are needed that would improve the technical level of production of commercial and intermediate products obtained from lead dust, taking into account known trends and new areas of technology development.

The importance of the task of organizing technology for the processing of lead dust is pronounced by the fact that recently, due to the complication of their composition (increase in the content of copper, arsenic, antimony, etc.), large volumes of them began to accumulate on the territory of factories and enterprises without finding proper sales. The lack of a rational technology for their processing is one of the key problems that need to be addressed.

An analysis of literature sources shows that attempts to rationally solve this problem are in the focus of attention of a number of large research institutes and organizations. However, the issue of processing lead dust, both copper smelting and lead production, remains open to this day [2-4].

In this article, based on the analysis of the results of the most promising pyro- and hydro-metallurgical technologies for the disposal of dust from copper smelters and lead production, possible ways of their integrated use are shown, which have the greatest interest to the processing of lead dust of Kazakhmys Corporation LLP.

Pyrometallurgical methods of lead dust processing. Pyrometallurgical technologies for the processing of lead dust include the use of *smelting and roasting processes*.

Smelting processes. The most common is the method of melting dust in an electric furnace with sodium sulfate and soda in a reducing atmosphere [5]. For melting, a three-phase electric furnace with a power of 2300 kVA with a hearth area of 5 m² is usually used. The main smelting products are lead metal, cadmium fumes and sodium slag, accumulating zinc, arsenic, selenium and other trace elements. The average composition of the dust supplied for processing, %: 52-60 Pb; 11-19 Zn; 0.8-1.5 Cd; 5-6 S; Sb and As 0.1-1.5. The costs of soda and reducing agent are 20-25% and 8-12%, respectively. Exhaust gases (826-1026 °C) after the afterburner are fed to bag filters for cleaning.

Melting of granular dust from copper smelters is also carried out in electric furnaces in a reducing atmosphere at a temperature of 800–1000 °C [6]. The mixture contains, %: Na₂CO₃ – 1-5, coke – 4-8, slag – 10-25. Electrosmelting slag containing 25% Zn is processed by leaching. When processing a mixture of dust after copper removal (Pb – 60%; Zn – 15%) and converting dust (Pb – 80%, Zn – 4%), rough lead is obtained containing less than 5% impurities and slag (Pb – 5%) which has about 97% of the Zn contained in the initial dust. The efficiency of electrothermy can be increased mainly due to the high-quality preparation of the initial charge [7].

Fine lead dust from electric filters of the converter section of copper smelters in the Ural region, along with valuable components, contains a significant amount of arsenic [8]. For the processing of these dusts, a combined method is proposed, including their reduction smelting with sulfate lead cake, sodium sulfate, soda and coke. As a next step, arsenic is extracted from the matte-slag melt by aqueous leaching. Sulfide-alkaline aqueous leaching solutions are used as a reagent for the precipitation of arsenic from acid washing solutions of the sulfuric acid plant. When the solutions are combined, 98-99% As precipitates as As₂S₃. The advantages of this technology include: increasing the complexity of the use of raw

materials by increasing the extraction of lead, bismuth and precious metals into rough lead; the possibility of joint processing of converter dust and sulphate intermediate products (cake, sludge); favorable conditions for the removal of arsenic from the technological process of copper smelters in the form of a low-toxic compound – arsenic trisulfide (As₂S₃).

The work [9] presents the results of factory practice of Mitsui Kinzoku (Japan) processing zinc dust briquetted with coal in a shaft furnace. During smelting, slag and matte are obtained in the sump, and rough zinc oxide (Zn – 50%, Pb – 20%) is obtained in the condenser, which is mixed with zinc slag, rolled and roasted in a tube furnace. The cinder, containing up to 65% zinc, is reduced in a vertical retort and rough zinc and rimming are obtained. Both products are returned to blast smelting.

In Poland, a method was proposed [10] for processing lead-containing dust from shaft copper-smelting furnaces and converters. The melting of the briquetted charge is carried out in a shaft furnace with the addition of fine iron scrap (3-12% by weight of dust), lime (2-6%) and converter slag (10-12%). In this case, rough lead (Pb 92-94%), matte (Cu 10-12%, Pb 10-25%, S 28-30%) and slag (Pb < 5%, Cu ~1%) are obtained. The dust concentrates rhenium and other rare earth elements. Separation of liquid phases is carried out in a sump, and scrap iron is introduced to reduce the sulfur concentration in rough lead.

The technology for processing lead-containing dusts at the plant of Preussag AG Metal (Germany) [11] is an interesting one, which produces zinc by the New Jersey method. Processing is subjected to the dust of roasting sulfide zinc concentrates, %: 45-50 Pb; 10-12 Zn; 2.5-3.2 Cd; 9-10 S; 250-350 g/t Ag and Waelz process dust, %: 53-55 Pb; 13-15 Zn; 0.3-0.5 Cd; 5-6 S; 0.5-1 Cl; 80-120 g/t Ag. Processing is carried out in rotary kilns with the addition of barite concentrate or soda. In both cases, rough lead is produced, which accumulates silver, and dust, which contains most of the zinc and cadmium. This dust is also processed in a rotary kiln to produce clinker and separate dust which contains lead and cadmium. Lead- and cadmium-containing dust is processed in a rotary kiln to produce rough lead and dust containing up to 50% cadmium. The resulting dust is leached with sulfuric acid and cadmium is cemented from the sulfate solution with zinc powder. Further, vacuum refining produces zinc with a purity of 99.995%. A by-product of the process is zinc sulfate. Extraction of lead into rough lead is 96%, silver

into rough lead – 92%, zinc in clinker – 62%, in barite slag – 30%, in zinc sulfate – 1%. Extraction of cadmium is about 90%.

In Germany, a method has been implemented for processing sulfate cakes, sludges and dusts containing lead by reduction smelting in a rotary kiln. The material is mixed with lime (5-30%) and coke (5-20%) and loaded into a rotary kiln at 1000-1200 °C, which makes it possible to continuously produce rough lead containing: 0.25% Sb; 0.14% As; 0.047% Bi; 0.011% Cu [12].

The disadvantages of rotary kilns are:

- large dust loss, which significantly complicates the equipment scheme and reduces the environmental friendliness of the process;
- a large number of processing stages and products that require their further processing, and, associated with this, the loss of metal;
- high power consumption;
- difficulties of complex mechanization and automation of the process;
- the use of complex and expensive dust-collecting devices.

For the processing of lead converter dust from copper smelting production, composition, %: 60.5 Pb, 1.6 Cu, 3.35 As, 1.12 Zn, 1.77 Sb, 8.1 S, 121 g/t Ag, 87.5 g/t In prepare a charge containing NaOH and a reducing agent (coke). During the smelting in a short drum furnace, an extraction of lead into the rough metal is 97–99%. Alkaline slag after treatment with water contained up to 340 g/t In. The method is characterized by high extraction of indium (above 95%) and low consumption of reagents.

Roasting processes. The authors of [13] granulated lead dust from a copper smelter, and then coated the granules with a particulate refractory material (silicon oxide or high-silicon ore, or cement). Next, the mixture was roasted at a temperature of 550-700 °C for 0.5-2 hours. The consumption of coatings was 5-30% of the mass of dust. Roasting was carried out in a shaft furnace in a continuous mode, which significantly increased the productivity of the process and facilitated the solution of tasks for its mechanization and automation. Despite these advantages of the method, its implementation is possible with the addition of a refractory material, which significantly increases the cost of the technology.

In [14], lead dust from converting copper matte was mixed with crushed Fe-Si-Al alloy, which was added in an amount of 2-10% by weight of the dust. The initial mixture was rolled, dried and roasted at 500-750 °C, blowing air at a speed of 0.4-1 m/s. The method makes it possible to extract rhenium into the

gas phase at the level of 90-95%, to organize a continuous process, to use a calciner having a higher specific productivity with smaller dimensions. This technology is associated with the cost of the alloy, which leads to an increase in the cost of technology.

In [15], a highly dispersed product (converter dust) is mixed with coarse-grained material (part of crushed granules roasted at 550-700 °C), taken in an amount of 5-40% by weight of the charge. The initial mixture is granulated, the granules are dried and roasted at a temperature of 400-700 °C. The process fails to achieve a high extraction of arsenic into dust, gases, so the resulting lead cake containing arsenic experiences certain difficulties in further smelting process.

In [16], fine lead dust (0-63 μm) is mixed with coarse-grained material (0-2000 μm in size) in an amount of 5-20% by weight of the mixture, and the resulting mixture is granulated. After drying, the granules are roasted in a shaft-type reactor. Water is fed into the reactor at a temperature of 570-670 °C. This method fails to achieve a high removal of arsenic from the dust. The resulting cake, although it contains a high lead content, is heavily contaminated with arsenic, and therefore encounters difficulties with further processing. In addition, the supply of water to a hot reactor leads to the formation of elemental hydrogen, which makes the process explosive.

In [17], dispersed lead dust is granulated with calcium oxide in an amount of 15-40% by weight of the granules, and roasted at a temperature of 570-670 °C in the presence of water vapor. This technique reduces the volume of material flows at the stage of hydrometallurgical processing and increases the specific productivity of the equipment at this stage.

An analysis of the considered works shows that carrying out the dust roasting process without the addition of an oxidizing agent [14, 15] is inefficient, since the extraction of arsenic into the gas phase reaches only 85%. This is due to the fact that the higher sulfide and lower oxide of arsenic (As⁵⁺) are not oxidized to their volatile compounds – As³⁺.

Granulation of lead dust and sludge is carried out in order to filter the gas phase through the product layer in a laminar mode, reduce dust formation, which makes it possible to achieve high kinetic characteristics during the roasting process and increase the degree of extraction of arsenic, rhenium and osmium into the gas phase [14, 18, 19]. At the same time, to loosen the structure of the granules without destroying them, to increase the reaction surface

of the solid product, to expand the gas outlet channels inside the granules and improve the access of oxygen to the active sites, the addition of coarse-grained and coarse-grained materials to the fired material is required [16].

The addition of refractory substances to the roasting charge [14, 20, 21] has the following advantages: it prevents the granules from melting and sticking together; raises their melting temperature and thus allows distillation of arsenic, rhenium and osmium at high temperatures, which allows sufficiently complete extraction of rhenium and osmium into the gas phase.

Despite the advantages described above, pyrometallurgical technologies are characterized by the complexity of equipment design, high costs of energy, material and labor resources, as well as equipment investment. In addition, the use of pyrometallurgical technologies is accompanied by significant volumes of dust and gas mixtures that require their purification and disposal.

Hydrometallurgical processing methods of lead dust. Recently, in the world, special attention has been paid to hydrometallurgical technologies for the production of non-ferrous, rare and rare earth metals. Unlike pyrometallurgical methods, they are cost-effective, environmentally cleaner and include rational methods for processing technogenic raw materials and intermediate products.

There are various hydrometallurgical methods for processing lead dust. As a rule, the main operation in their processing by hydrometallurgical methods is leaching. Leaching is carried out in solutions of different acids (H_2SO_4 , HNO_3 , HCl), alkalis ($NaOH$, NH_4OH) or acidified salts ($FeCl_3$, $Fe_2(SO_4)_3$).

All of these methods are described in sufficient detail in the technical literature and do not require special commentary. Still it is important to mention essential basic shortcomings of each of them.

The disadvantages of alkaline leaching are:

- loss of expensive alkali;
- precious metals and copper, which remain in cakes from leaching, are not extracted into marketable products;
- high yield of semi-finished products and production wastes;
- the presence of effluents containing harmful impurities.

Sulphate hydrometallurgical schemes for the processing of lead dust are the most well studied, which is associated with the cheapness and availability of sulfuric acid. In addition, the lower solubility of lead sulfate compared to other non-ferrous sulfates provides greater se-

lectivity in hydrometallurgical schemes based on sulfuric acid. However, such schemes have significant drawbacks:

- there are problems associated with the need to utilize SO_4^{2-} ;
- regeneration of solutions is accompanied by significant losses of expensive reagents (up to 20% in each turn).

Perspective directions of development of lead dust processing technologies. Promising areas of lead dust processing include various options for bacterial leaching. In Iran, a method has been implemented for processing dust from copper plants containing, %: 36 Cu, 22.2 Fe, 12.2 S, obtained from dust collectors during the operation of converters and reverberatory furnaces. The experiments were carried out in an Erlenmeyer flask with a solution acidity of 1.8, a pulp density of 7%, a process temperature of 31°C, and a stirring speed of 150 rpm [22]. An increase in pulp density indicates an increase in bacterial growth in the initial phase of microorganism growth (lag phase), an increase in acid consumption, metal ion toxicity, copper concentration and tangential stress. As a result, the redox potential and copper recovery are reduced. According to the obtained curves, the maximum extraction of copper from biological conditions for densities of 2, 3, 4 and 7% was 42.2%, 45.9% and 83.1%, respectively. The obtained data indicate the possibility of recovering copper from copper-containing dust using natural mesophilic bacteria. This technology can be an alternative and promising process to cope with the problem of dust accumulation in enterprises.

The authors of [23] use the method of bacterial leaching with the help of *Acidithiobacillus ferrooxidans* and *Acidithiobacillus thiooxidans*, which oxidize iron and sulfur, to process lead dust from a copper smelter. Particle sizes less than 80 microns. The average content of copper in the dust is 30%. The basis of sulfide copper minerals is chalcocite, chalcopyrite, bornite and covelin. Since a significant amount of copper is in oxide form, chemical leaching with dilute sulfuric acid is carried out before the bioleaching process. The process is carried out in a 500 ml specialized vessel at a process temperature of 31°C and a stirring speed of 150 rpm. To increase the density of the pulp, special conditions were created with higher toxicity, tangential stress and a decrease in mass transfer, which, in turn, led to a slowdown in the process rate and copper recovery. To avoid this, much more microorganisms and a richer nutrient medium are needed, an increase in the percentage of solids in the pulp.

Under optimal conditions, up to 91% of copper is recovered from the dust.

Biotechnologies can be applied to solve environmental problems by converting hazardous materials into safe or valuable products as a powerful and cost-effective technology. This technology has a number of advantages over pyrometallurgical processes:

- relative simplicity;
- mild operating conditions;
- low capital costs;
- low energy consumption and environmental safety;
- the processing scheme is closed loop;
- circulating solutions after partial or complete regeneration are used as a nutrient medium for bacteria and a leaching solution.

Along with the listed advantages, bacterial leaching has a number of significant disadvantages: intensification of leaching is achieved by activating the vital activity of bacteria adapted to specific environmental conditions (type of ore, chemical composition of solutions, temperature, etc.). This requires a pH of 1.5-2.5, a high redox potential (Eh 600-750 mV), a favorable and stable chemical composition of the solutions, which is achieved by their regeneration and the mode of aerating and moistening (irrigation) of the ore. In some cases, nitrogen and phosphorus salts should be added, as well as bacteria grown on recycled solutions in regenerator ponds. The number of bacterial cells in the leaching solution and ore should be at least 106-107 per 1 ml or 1 g, respectively. The cost of 1 ton of copper obtained by bioleaching is 1.5-2 times lower than with conventional hydro- and/or pyrometallurgical methods.

The relevance of research on the search for a new technology for processing fine dusts of copper smelters is due to the following reasons:

- these products are valuable raw materials and must be subjected to self-processing, which is relevant for both economic and environmental reasons;
- utilization of copper smelter dust prevents potential damage to nature and human health and increases the complexity of the use of ore raw materials.

The disadvantages of pyrometallurgical schemes are the low quality of the products obtained, the need for purification and neutralization of gases. Products obtained from the processing of man-made waste in pyrometallurgical units, in most cases, require additional (more often hydrometallurgical) refinement, which significantly reduces the efficiency of pyrometallurgical schemes.

In the hydrometallurgical processing (leaching) of lead dust, solutions of acids, alkalis, salts, as well as organic solvents are used as solvents.

The use of acids is associated with an additional consumption of reagents for the selective separation of metals from solutions. In addition, it is necessary to create special acid-resistant equipment, often operating at elevated temperatures.

When using alkaline solvents for the extraction of copper, zinc and lead, difficulties arise with the recovery of solvents and their subsequent disposal, as well as the processing of the resulting products. Most often, they are contaminated with other heavy non-ferrous metals, which leads to the need for their further selective separation.

The use of amine-containing solvents makes it possible to achieve high selectivity in the extraction of metals into solution. Moreover, some solvents quite selectively affect lead compounds. Organic solvents are characterized by a high capacity for non-ferrous metals, as well as the possibility of their regeneration and return to the leaching stage. In addition, they do not require special construction materials.

Thus, the developed technology must comply with modern environmental requirements, be completely closed to sewage and solid waste, and fit into the overall production cycle of the enterprise.

Based on the analysis, it follows that the most effective technology for the processing of lead dust, which ensures its complex use, are combined methods that combine pyro- and hydrometallurgical methods. This approach will allow the selective extraction of valuable metals into marketable products with the removal of harmful impurities and their further utilization.

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