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PROCESSING OF NON-FERROUS METALLURGY WASTE SLAG FOR ITS COMPLEX RECOVERY AS A SECONDARY MINERAL RAW MATERIAL

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The article presents an overview of the methods of processing slag from Waelz process, and various approaches employed by scientists globally, aimed at processing such slags. Despite several listed methods of processing slag from Waelz process, none entails a sufficient complex nature of its processing. In addition, the slag processed from the Waelz process, found in the dumps, has not been used as a secondary raw material to date. The elemental chemical composition of the slag has been determined, represented by compounds of calcium, silicon, iron, aluminum, carbon, and heavy non-ferrous metals, namely zinc and lead. Thus, it has been established that these slags continue to pollute the environment for many years.

Keywords: industrial waste, Waelz process slag dumps, iron-containing additive, Portland cement clinker.

Lately, the content of metals in ores has been decreasing, due to the intensive extraction and processing of various ores rich in metals. At the same time, the annual amount of industrial waste containing both precious metals and other useful compounds is increasing [1]. Waste in dumps contains heavy non-ferrous metals, compounds of silicon, calcium, aluminum, iron, and affects the environment negatively [2], yet at the same time, they can serve as a secondary technogenic raw material. Thus, in Kazakhstan, in the process of extraction of non-ferrous metals at various enterprises, from the 1920s to the present, a significant amount of waste has been collected, namely Waelz process slags, which are stored in dumps, occupying plentiful lands, and polluting the environment. In this regard, the world experience of methods of complex processing of Waelz process slag dumps is considered.

During the Waelz process of natural and technogenic raw materials (oxide ores, cakes, slags), a clinker is formed, whose value depends on the type of raw materials processed. Thus, the clinker obtained from zinc cakes, in addition to 1 – 2% Zn, 1 – 3% Cu, and 0.5 – 0.8% Pb, contains a significant amount of silver and gold (300 – 600 g/ton). The clinkers obtained during the slag Waelz processing of the blast smelting of lead agglomerates contain Zn (0.9 – 1.0%), Pb (0.1 – 0.3%), Cu (0.5 – 1.0%), and precious metals. The clinkers of the Waelz processing of the Achisai ores contain small amounts of copper, precious metals (traces), 0.6 – 0.7% Zn, and 0.1 – 0.2% Pb. All clinkers also contain Si (11 – 12%), Fe (18 – 30%), and C (18 – 24%) [3].

Due to the different compositions of clinkers, it is necessary to analyze the existing methods of clinker processing. These are conventionally divided into two types:

– complex processing of the clinker of Waelz processing of zinc-containing materials with extraction of non-ferrous and precious metals, the carbon component using iron-containing material and non-metallic components;

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– the use of clinker of Waelz processing of zinc-containing materials as a charge material in various pyrometallurgical technologies.

In the copper production, a method of processing clinker with copper-zinc concentrates using agglomeration and blast smelting is well-known. The introduction of 18 – 24% clinker into the charge results in a low-grade matte. An increase in the clinker content in the agglomeration charge up to 29 – 34% raises the temperature of the exhaust gases and complicates the operation of turbo-exhausters [4]. This method has one more disadvantage, namely "smearing" of Zn and Pb on the processed products. Thus, when processing clinker with copper concentrate at the Karabash copper smelter, only 30% Zn and 60% Pb pass into sublimates [3].

On the industrial scale, tests were performed for processing in the oxygen-suspended electrothermal smelting unit of copper-zinc concentrates and clinkers from the current supply, and from dumps containing 2.24 – 5.76% Cu, 0.68 – 1.66% Pb, 0.8 – 1.38% Zn, 16.2 – 27.7% C, 26.56 – 30.48% Fe, 5.45 – 6.06% S, 14.1 – 15.01% SiO_2, and 1.42 – 2.38% CaO. With the introduction of 10% clinker, the extraction of Cu into the matte was 91.22%. Zinc sublimates contained 72.18 – 72.96% Zn, 4.75 – 5.11% Pb, 1.3 – 1.38% Fe, 0.99 – 1.2% (SiO_2 + CaO), 0.2 – 0.25% Cd, and 0.43 – 0.46% S. The processing occurred without any technological deviations with an electric power consumption of 2411.3 kWh per 1 ton of charge. However, despite this, the residual content of Zn in the slags was 9.46 – 10.68%, and that of Pb was 0.28 – 0.43% [5].

In Balkhashmed, zinc clinker was used instead of coal when smelting low-sulfur concentrates in PV [6, 7]. The elemental iron and carbon contained in the clinker function as energy reagents in the liquid phase, which, when interacting with oxygen, release the heat necessary for smelting. In the presence of clinker, the Cu content in the slag decreases, and Zn peroxidation is reduced. Replacement of coal with the clinker does not lead to technological complications in the operation of the PV and helps to reduce the temperature of the exhaust gases in the waste heat boiler and gas wastes (due to the reduction or complete absence, afterburning of volatile components of coal).

The method of smelting clinker [8] presents two cases consisting of a sufficient supply of oxygen-containing blast to the clinker layer and into the melt to ensure the predominant oxidation of the clinker solid carbon (in case 1), and the supply of an excess amount of metallic iron of the clinker to bind the clinker sulfur and dissolution in the matte phase (in case 2). This method has certain disadvantages, such as insignificant heat release in the melt due to oxidation of the excessive metallic iron of the clinker, necessitating the use of external heat sources to maintain the melt bath in working condition. Another disadvantage is the fundamental limitation on the maximum degree of enrichment of the resulting mattes for copper due to which obtaining conditioned mattes suitable for individual conversion becomes impossible during the processing of the clinker with a copper content of less than 2.5% (more than 50% of all produced and accumulated dumps). Thus, the possibilities of the industrial use of this method are limited significantly.

Another method for processing clinker from zinc production [9], containing carbon and metallic iron, includes smelting of clinker by feeding an oxidizing blast into the melt to obtain liquid and gaseous smelting products. This method also has some disadvantages, particularly, the unsatisfactory operational reliability of the smelting process due to the high tendency of melt baths to foaming, which results in irreversible procedural violations leading up to the destruction of the melting unit.

The clinker of the Ukrzinc plant was used as a fuel agent in the processing of low-grade lead smelting mattes. As a result of melting the charge in a shaft furnace, matte with 14 – 16% Cu and 9 – 10% Pb and sublimates with 17 – 25% Zn and 35% Pb were obtained. Nevertheless, Zn, Pb, and Cu in the slags amounted 2.5 – 2.9% in total [10].

A charge was developed for the processing of zinc-containing materials by melting with the use of clinker for the Waelz processing of zinc-containing materials, including 5 – 9 wt.% of carbonaceous reducing agent, 2 – 6 wt.% of calcium oxide, 3 – 10 wt.% of Waelz clinker, and the rest was zinc-containing material [11]. In this charge, clinker acts as a reducing agent and an iron-containing material at the same time. The use of such a charge enables reduction in the consumption of coke from 12 – 16% to 5 – 9%, an increase in the extraction of Zn by 0.8% (from 96% to 96.8%), reduction in the consumption of electricity by 50 – 100 kWh/t of zinc from 3700 up to 3650 – 3600 kWh/t, and an increase in the productivity by 10%.

The work of the Eastern Research Mining and Metallurgical Institute of Non-Ferrous Metals on the independent processing of clinkers from the Waelz zinc slag and Achisai ore is noteworthy.

The institute performed laboratory studies of enrichment of fresh clinker to obtain coal concentrate containing 51.5 – 60.2% of carbon, with its extraction being 86 – 95%. In addition, the magnetic separation of clinker was investigated. The most acceptable indicators for the production of high-quality coal concentrate and the maximum extraction of iron into the magnetic concentrate were obtained with a combination of flotation and magnetic separation. At the same time, the following products were obtained from the Achisai clinker:

– magnetic concentrate with an iron content of 56.3 – 60% and its extraction of 67 – 70%, which can be used in ferrous metallurgy;

– coal concentrate containing 56.1 – 58.3% of carbon, with 90.3 – 90.5% extraction;

– tailings with 55 – 59% output, with a content of up to 7% of iron and 2.3% of carbon, recommended for the production of building materials [3].

The possibilities of obtaining building materials, mineral wool from Achisi clinker [12], and its use in road construction [13] have been revealed. A similar work was performed with the clinkers of the Ust-Kamenogorsk lead-zinc plant (UKLZP) (Cu 1.73%, Zn 0.37%, and Pb 0.42%). After grinding, the clinker was subjected to wet magnetic separation. Up to 90% Fe and 85% Cu were extracted into the magnetic concentrate. From the non-magnetic fraction by flotation using pine oil, a coke concentrate was obtained with the extraction of up to 90% of carbon into it. Dump tailings of magnetic flotation beneficiation, containing in total up to 0.54% of non-ferrous metals (Zn, Pb, Cu), could be used in the production of building materials. The non-magnetic fraction, containing much less copper and more carbon, could be used for the agglomeration of lead concentrates [12].

The processing of Waelz clinker slags of mine lead smelting by magnetic separation to obtain (25 – 30%) ferromagnetic concentrate containing 75 – 89% Fe and 1 – 1.5% Cu, is well-known [14]. In this case, the magnetic concentrate was used in the fuming of slags, charge of agglomeration of lead production, and enrichment of oxidized copper ores (instead of cast iron shavings); from the non-magnetic fraction, a coal concentrate was obtained with 58% C, recommended for Waelz blowing into the furnace or granulation with recycled dust [15].

At the Electrozinc plant, an experiment was performed on blowing the clinker with compressed air (blowing off carbon) and supplying a coal-air mixture to the head of the Waelz kiln. However, despite an increase in productivity (by 10%) and a decrease in coke consumption, the experiment was terminated due to deterioration in the quality of Waelz oxide due to its contamination with ash and carbon [16].

The experience of processing rich clinker in Bulgaria is interesting [17]. Clinker containing 2.23% Cu, 1.31% Zn, 1.25% Pb, 19.1% C, 20.0% SiO_2, 4.47% S, as well as 200 g/ton Ag and 12 Au, was subjected to screening. The class of 16 mm and larger was shipped to copper smelters, and the rest (smaller than 16 mm) was separated in a heavy suspension, after which the heavy fraction was sent to the copper smelter, and the light fraction was used in Waelz furnaces. At the same time, the extraction of copper into products for metallurgical processing reached 93%.

For the processing of clinkers poor in precious metals, more complex technological schemes are used with a combination of flotation and magnetic separation. According to Unipromed, the extraction of Cu and Au in processed products can reach 91.7%, and that of Ag can reach 89.1%. At international enterprises, concentrates with 1.5% Cu and 515 – 620 g/ton Ag can be obtained with a Cu content of 0.05% in the non-magnetic fraction, 80% C (Peru, La Oroya plant) or with 1.6% Cu, 3.2 ton/g Au, and 544 ton/g Ag (Japan, Aizu plant) [18, 19].

Leading Research Institute of Chemical Technology provides a sequential selective separation of copper and zinc from the clinker by leaching with sulfuric acid at 60 – 80°C, extraction of gold and silver from the washed cake using sorption technology, followed by the separation of fine coke by flotation [20, 21]. Sorption technology includes pulp cyanidation, a disadvantage of the technology. Therefore, another method for the extraction of gold and silver using sodium bisulfite has been proposed. This technology entails the extraction of copper (up to 90 – 95%) in the form of copper powder, as well as gold and silver (up to 85 – 90% and 55 – 65%, respectively) into the Dore alloy and 95% of coal into a concentrate with up to 90% C.

To extract non-ferrous metals from Weltz clinkers, UKLZP and CEZP (Chelyabinsk Electrolytic Zinc Plant) in Gintsvetmet, developed a chloride sublimation method in a fluidized bed furnace [22 – 25]. The method was tested on a semi-industrial plant with an hourly consumption of 165 kg of raw materials. So, when processing UKLZP clinkers containing 1.89% Cu, 2.43% Zn, 0.87% Pb, 250 g/ton Ag, and 5 g/ton Au at 1223 – 1273 K, the degrees of chloride sublimation of the metals were 86.5% for Cu, 79% for Zn, 93.2% for Pb, 93.8% for Ag, and 88% for Au. Although Kavkaz-Giprotsvetmet developed a technical project for the construction of a clinker processing unit at the Electrozinc plant with a capacity of 100 thousand tons per year, the method has not been practically implemented. Disadvantages of the method include long duration of the process (5.5 hours), high consumption of a concentrated solution of $CaCl_2$ (30% of the mass of the ore), and a relatively high residual content of Zn (0.5%) and Cu (0.25%) in the cinder.

The Kazakh Institute of Chemical Technology has developed a chloride method for processing UKLZP clinkers in a tubular rotary kiln with the combination of chloride sublimation of non-ferrous metals in the kiln and formation of cement clinker [26]. At the preliminary stage of research, it was revealed that the carbon present in the clinker (20%) inhibited the chloride sublimation of non-ferrous metals. Therefore, the chloride sublimation was performed with a boosted charge (50% limestone, 35% clinker, 9% phosphogypsum, and 9.1% $CaCl_2$). At 1373 – 1423 K for 1 hour, the degrees of chloride sublimation for non-ferrous metals were 87.34% for Cu, 88.93% for Zn, 96.44% for Pb, and 90% for Ag. From the cinder, after grinding with 5% gypsum, cement grade M-300 was formed. The economic effect of the developed method is about 10 US dollars/ton of clinker; however, the method cannot be considered rational as it necessitates the processing of the charge with almost 55.9% of non-metallic components.

In the 1990s, Yuzhpolymetal started processing the Achisai clinker to produce magnetic concentrate and coke, which never became widespread at other enterprises, limiting itself to exploratory experiments. However, the technological indicators of this process (including the extraction of non-ferrous metals) are not described in the special literature.

In the 2000s, another attempt was made in the M. O. Auezov South Kazakhstan University (Kazakhstan) to comprehensively process the slag dumps from Waelz processing of zinc ores of the Achisai deposit. The technology enabled the removal of lead-zinc sublimates (up to 98%) and extraction of iron and silicon into an alloy (low-quality ferrosilicon). At the same time, the process was energy-consuming with the formation of a large amount of waste (more than 50%),

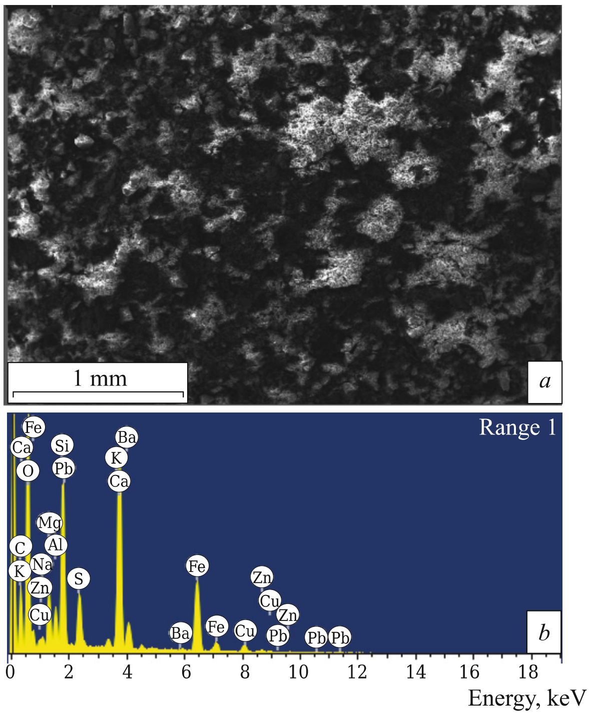

Fig. 1. Micrograph (*a*) and results of the elemental analysis of the metallurgical production waste (*b*).

which was proposed to be crushed into slag crushed stone. The studies have passed pilot testing, but have not been introduced into production [3, 27].

In view of the continuation of the work started and performed at the M. O. Auezov South Kazakhstan University, the authors of this article performed chemical and elemental analyses of the slag from Waelz (Fig. 1) with the aim of its reclamation [28 – 59] by the method of complex processing as iron-containing additive in the production of Portland cement clinker [60 – 63]. The chemical composition of Waelz slag in Achpolymetal was 14.87% CaO, 18.12% SiO_2, 2.81% MgO, 4.75% Al_2O_3, 26.98% Fe_2O_3, 0.94% Zn, 0.12% Pb, 0.11% Cu, 1.4% S, 18.6% C, 2.4% BaO, 8.9% others.

The results of the chemical composition analysis of the slag were studied, which coincide approximately with the earlier studies of its chemical composition with a predominance of iron content [3, 27, 34]. In addition, a sample of the Waelz slag was analyzed with a scanning electron microscope to assess the elemental composition. The research results showed (Fig. 1) that the real waste in the form of slag from Waelz processing contained calcium, silicon, oxygen, iron, aluminum (also confirmed by the previously performed chemical analyses [3, 27, 34]), necessary to obtain cement clinker.

Thus, based on the review, chemical, and electron microscopy analyses, the following conclusions can be drawn:

– despite the wide variety of methods for processing Waelz slags, large-scale processing of the Achisai slags has not been performed yet;

– over the years, slag dumps from the Waelz process at the Achpolymetal metallurgical plant continue to pollute the environment;

– the slag from Waelz processing, formed over many years of operation of the metallurgical plant Achpolymetal, is a valuable mineral raw material and can be used as an iron-containing additive in the production of Portland cement clinker.

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