

Carbon-Free Selective Extraction of Zinc and Lead from EAF-Dust

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Abstract—In electric-arc furnace dust, zinc content can reach 43%, lead and content can reach 4%. The ecotoxicant content such as dioxins and furans (D&F) adsorbed on dust particles achieve 500 ng/kg of dust. Generally, zinc and lead are reduced from their oxides by dint of carbon (an average of 500 kg/t dust). The results of thermodynamic calculations and experimental studies have shown that these metals can be extracted from dust without the participation of carbon or at its low content (less than 3%) [1]. Temperature of about 1400 K is required to extract lead, and about 2000 K is required for zinc extraction. The temperatures of their extraction depend on the dust composition, in particular, on the carbon content, chlorine and the O/C ratio [2]. They can also depend on phase and dispersed dust composition. A physicochemical analysis of the dust formation processes in electric arc furnaces (EAF) has been carried out, the composition and properties of dust have been investigated, and experimental studies of the zinc and lead selective extraction process in laboratory conditions have been led. A dust processing technology has been developed and the possible innovative potential of the expected results has been assessed. The approaches are based on the study of a continuous two-stage process of carbon-free and zinc and lead selective extraction from EAF dust of different composition. One of the main results of the work, along with the creation of a technology that ensures the selective zinc and lead extraction up to 99%, is the development of a process for neutralizing EAF dust from D&F to an environmentally safe level.

Keywords: ferrous metallurgy, non-ferrous metals, steel dust, electric-arc furnace dust (EAF dust, EAFD), carbon-free process, selective extraction, evaporation, zinc, lead, iron, secondary resources, resource saving, pollutants, hazard class, environmental safety

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INTRODUCTION

In the earth deposits of metallurgical enterprises, sufficient amount of waste has accumulated, in particular, steel-smelting dust that contains valuable components, which presently can become an additional resource of raw materials for a number of industries. At the same time, these wastes for many years have been sources of negative impact on the environment due to the presence of toxic substances in them [1]. In electric steel-smelting dust, the zinc content can reach 43%, the lead content is 4%, the content of such ecotoxicants as dioxins and furans (D&F) adsorbed on dust particles is 500 ng/kg of dust. The study of the physicochemical patterns of the dust component behavior for the purpose of resource-saving and environmentally safe extraction of zinc, lead, iron, and neutralization of chlorine and its organic compounds is an urgent task of sustainable industrial development.

Forecasts based on the dynamics of the formation of zinc-containing dust in EAF show that when it is processed by the method of carbon-free selective extraction, up to 128 thousand tons of zinc and 6.5 thousand tons of lead per year can be returned to

manufacture as secondary raw materials. In this case, the iron-containing residue (about 400 thousand tons/year with the iron oxide content of 65–70%) can be returned to the sintering plant.

The novelty of the work lies in the fact that the proposed process of selective extraction of non-ferrous metals is carbon-free, that is, it is carried out without the use of a reducing agent. Reducing industrial consumption of carbon is one of the main tasks of the BRICS countries, which is associated with the need to prevent climate change. At the same time, one of the main ways to reduce greenhouse gas emissions is a significant reduction in the greenhouse gas emission standards at the state level [2]. In this regard, transition to carbon-free technologies in metallurgy is necessary: saving carbon consumption up to 0.5 t/t of dust will prevent emissions of CO₂, the main greenhouse gas, by 1.1 million t/year.

It should be noted that the process of processing zinc-containing metallurgical waste (dust) is not the only way to involve secondary resources of zinc and lead to manufacturing. According to the authors, it is advisable to group scientific research in three direc-

tions, depending on the option for solving the dust processing problem. The weight of each of these areas is different, since the problems being solved have a different degree of complexity (in scientific terms).

The first option is based on the idea of preliminary preparation of charge materials before melting in EAF. If the waste of galvanized steel is preheated to a temperature of 1250 K before being fed into EAF, then zinc, with its boiling point reaching 1180 K, will evaporate [3]. The process temperature can be reduced to 850–900 K due to the use of reducing synthesis gas [4]. The extraction degree of zinc is more than 97% depending on the heating duration; the zinc content in the captured dust is more than 90%.

The second option is based on the fact that during the melting of the charge containing the waste of galvanized steel, volatile components, in particular zinc, evaporate first. If they are captured separately from the bulk of the dust, enriched zinc concentrate can be obtained. The extraction degree of zinc from the charge can reach 99.5 and 97.5% for lead [5]. In this case, special attention should be paid to the behavior of polychlorodibenzo-*p*-dioxins and polychlorodibenzofurans (D&F) due to their high content in the captured dust, i.e., 474 ng/kg of dust [6, 7]. To prevent the formation of D&F during the cooling of gases, quenching with water or cooled air is used in some cases [8, 9]. However, the processing efficiency with lime milk is assumed to be higher and, according to calculations performed using the Terra thermodynamic modeling program, will be 95.3% [10].

The third option, the main focus of the work, is based on the possibility of recycling zinc-containing dust accumulated at landfills. The reserve depletion of rich zinc-containing ores in the near future will lead to an increase in the market value of non-ferrous metals, which, in turn, will increase the economic efficiency of metallurgical dust processing [11]. The proposed option can be implemented in parallel with one of the first options or without.

In world practice, there are several directions for using EAFD—from dust neutralization (glass transition) and burial to the extraction of non-ferrous metals (reduction from oxides). They are mainly based on the environmental and economic requirements of environmental protection legislation. Along with hydro-metallurgical methods, thermal, including high-temperature heating sources (arc, plasma), are offered. For selective extraction of non-ferrous metals, rather complex multistage processes are proposed [12–16]. In recent years, the issue of PVC utilization during joint processing with EAFD has been widely discussed [17].

The problem of extracting non-ferrous metals from steel-smelting waste is not new. Despite the existence of a large number of waste recycling methods, more economical and safer methods are being searched all over the world [1]. The search for an optimal solution is complicated by the fact that the characteristics of the

EAFD differ significantly from each other, even within the same enterprise. Mainly, there are differences in the chemical and phase composition of different size particles, which significantly affect the processing method choice [18].

Research analysis on the problem of utilization of toxic zinc-containing dust shows that the main directions are carried out within the framework of traditional concepts—using reducing agents (carbon) to reduce the temperature of the reduction onset of zinc and other metals, which makes it impossible for their selective extraction. Even with the use of plasma and arc heating, this tendency persists, although high temperatures make it possible to change the process conditions, for example, to extract metals (zinc and lead) separately without using carbon.

In the paper, carbon-free selective method is studied for producing zinc, lead or their oxides. The results of thermodynamic calculations and experimental studies have shown that these metals can be extracted without carbon participation or its low content (less than 3%) [19]. The idea is based on the difference in the evaporation (decomposition) temperatures of zinc and lead oxides under weakly reducing conditions (at a certain C/O ratio in the system) [20]. Research in this area is new and quite relevant. When implementing the results of the work, it is expected to obtain a significant environmental effect by reducing the volume of the waste stored at landfills, reducing toxicity of dust, as well as emissions of greenhouse gas CO₂ into atmosphere.

THEORETICAL BACKGROUND

The object of study are EAFD samples obtained from the bag filters of several enterprises. According to the data provided by the enterprises, in the obtained dust samples, the content of components significant for study differs several times, % (by weight): zinc oxide 9.4–28.5; lead oxide 0.8–2.1; carbon 1.2–5.2; and chlorine 1.3–5.7. The total content of alkali metal oxides varies from 2 to 12%. The difference in the content of significant components is important from the viewpoint of research methods. It should be noted that the oxide composition of the dust presented by enterprises, as a rule, is based on stoichiometric recalculation from the element composition. It does not always correspond to the real composition, since there is sufficiently high content of chlorine and alkali metals in a number of samples, which suggests the presence of chlorides. In addition, thermodynamic calculations showed that some of the elements in the dust are in the composition of chlorides, carbonates, etc., which was confirmed experimentally [14, 18, 21, 22].

To identify the conditions that allow selective extracting zinc and lead from electric steel-smelting dust, it is necessary to determine the evaporation temperatures of zinc and lead compounds and the compo-

sition effect of EAFD on them. Thermodynamic modeling is carried out using the Terra software system for modeling phase and chemical equilibrium [23].

For the selective extraction of lead in the furnace, it is necessary to create a reducing atmosphere containing no more than 60% CO. An increase in the CO content will lead to joint reduction of lead, zinc and iron [24]. In this regard, it is necessary not only to create the required temperature zones, but also the redox conditions suitable for them. To quantify the redox conditions of the system, the ratio of oxygen and carbon, i.e., the ratio of the oxygen content to the carbon content, was estimated.

The extraction temperatures of lead (its oxides) and zinc (its oxides) were determined for the electric steel-smelting dust of PAO “Severstal” at oxygen-to-carbon ratio in the system from 5 to 25. The simulation result is shown in Fig. 1.

It is found that the most favorable conditions for selective extraction of zinc and lead are observed, when the ratio of oxygen to carbon is in the range of 12.7–25.0 (with carbon content of no more than 3%). In this case, zinc extraction (its oxide) proceeds at 1800–2050 K, extraction of lead (lead oxide) at 1350–1400 K. Zinc is selectively extracted from EAFD with carbon content in the range of 2–3%, while up to 5% zinc can pass into lead.

The temperature of the “carbon-free” extraction of zinc from the electric steel-smelting dust is quite high. Therefore, for this purpose, it is proposed to use plasma heating.

EXPERIMENTAL

Experimental studies of the behavior of zinc behavior and lead during high-temperature processing of dust were carried out in a laboratory plasma-arc furnace. The description of the installation and the experimental procedure are given in [25]. The attempt of fixing the moment of zinc-free lead extraction using single-section crucible was unsuccessful. Due to the high heating rates of dust up to temperatures of 2273–2473 K (170–340 K/s), they were removed almost simultaneously.

To solve the problem of separate extraction of zinc and lead in a laboratory plasma furnace, the anode assembly design was changed. A multi-section crucible was developed with different temperature heating zones in different sections. The samples after processing have been analyzed for the residual content of zinc and lead. At the same time, the residual content of chlorine and its compounds, including D&F, was analyzed according to the method described in [6, 7].

Since the removal of lead requires low temperatures, it was advisable to use a muffle furnace with a controlled temperature up to 1423 K (1150°C) for lead extraction. The behavior of all significant elements was also of interest. Their evaporation temperatures

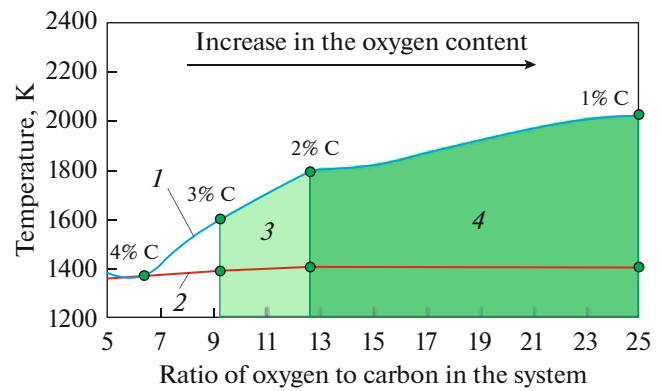


Fig. 1. Influence of the oxygen to carbon ratio in the system on the possibility of selective extraction of zinc and lead from EAF dust [20]: (1) Zn; (2) Pb; (3) zone providing selective extraction of Zn; (4) zone providing selective extraction of Zn and Pb.

and the degree of extraction (removal) were experimentally determined. Also possible chemical reactions that occurred at different temperatures and compositions of the system were studied.

The elemental composition of dust was determined by the methods of X-ray fluorescence spectrometry, atomic emission spectrometry with inductively coupled plasma (to determine the content of lead and zinc), combustion in a stream of oxygen followed by IR spectroscopy (to determine the content of sulfur and carbon), and gravimetry (determination of moisture) in “Analytical certification and environmental-analytical center OOO “Anserteko”. Since chlorine is of particular interest, its content in dust was additionally determined by a chemical method in the scientific and educational testing laboratory of physicochemistry of coals at NUST “MISIS”. The content of D&F adsorbed on dust was determined in the laboratory of analytical ecotoxicology at the A.N. Severtsov Institute of Ecology and Evolution of the Russian Academy of Sciences.

OBJECT OF STUDY

The object of study is the electric arc furnace dust of PAO “Severstal”. Content of elements, wt %: Fe—40.0; Zn—13.7; Ca—6; Na—2.8; Mn—2.6; Cl—1.8 (1.3); C—1.74; Mg—1.5; Si—1.3; K—1.0; Pb—0.8; S—0.47; Al—0.2; Cr—0.2; Cu—0.2; P—0.1; Ti—0.05; and W (moisture)—0.25. The rest of the sample is presumably oxygen. The total D&F content in the sample is 474 ng/kg of EAFD. There is an excess of the maximum permissible concentration of soil in populated areas 9.5 times; therefore, dust can be placed only at industrial landfills [26].

The calculated value of the ratio of the oxygen content to the carbon content (according to the elemental composition of EAFD) was $26.3/1.74 = 15.1$. Presum-

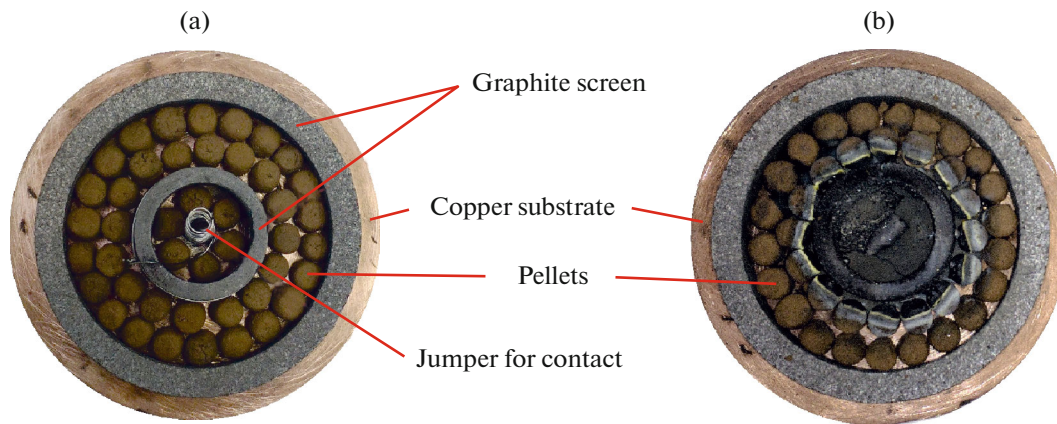


Fig. 2. Samples before (a) and after the experiment (b).

ably, required temperature for extraction of lead is 1408 ± 10 K, for zinc— 1816 ± 10 K (Fig. 1).

EXPERIMENTS IN A PLASMA-ARC FURNACE

Electric steelmaking dust is a finely dispersed powder, so it must be pre-pelleted to prevent the powder from blowing out by a plasma arc. The organization of different temperature zones is carried out by means of axisymmetric stacking of pellets ($d \approx 5$ mm) in accordance with Fig. 2.

Parameters of the experiment carried out in the plasma-arc installation: $U = 30$ V, $I = 117$ A, $P_{\text{chamber}} = 0.75\text{--}1.0$ atm, argon consumption—2 L/min, $t_{\text{heating}} = 40$ s. Each temperature zone is ring-shaped. Radii of the circles forming the zones, mm: zone 1—4 and 10; zone 2—13 and 19; zone 3—19 and 25. Calculated average surface temperature of samples, K: zone 1—2774; zone 2—1992; zone 3—1383 [27].

After plasma treatment, samples from different areas were analyzed for lead and zinc content. Based on the obtained data, the degree of extraction of zinc and lead was determined. The results are presented in Table 1.

In zone 1 with temperature near the sample surface of about 2500–3000 K, the extraction degree of zinc and lead exceeds 99%. In the other two zones, where

the arc temperature is approximately 1100–2300 K, lead is extracted more intensively than zinc: the extraction degree of metals from electric arc-furnace dust is 50 and 20%, respectively.

Studies in a plasma furnace have shown that the extraction degree of zinc and lead at high temperatures exceeds 99%. Although the lead extraction is higher than zinc extraction in low temperature zones, separate extraction has not been achieved. The use of a multisection crucible did not allow regulating temperature in the second section at the level of 1408 K; therefore, a laboratory muffle furnace SNOL 3/11-V was used to study lead behavior during heating of electric steel-making dust.

EXPERIMENTS IN THE MUFFLE FURNACE

Samples were heated in a muffle furnace at 573, 873, 1173, and 1423 K. Dust processing at a given temperature was carried out separately. The heating rate of the samples was 5 K/min, the initial temperature was 298 K, and the exposure time was 1 h. Heating and holding were carried out in an air atmosphere. The muffle furnace is equipped with a gas evacuation system.

For uniform heating of the dust along the height, the dust sample (150 g) was placed in six alundum crucibles ($D = 52$ mm; $d = 26$ mm; $H = 38$ mm) in equal layers (25 g each). The layer height was approximately

Table 1. Extraction degree of zinc and lead from EAF dust of PAO “Severstal” after plasma treatment

Zone	Before experiment			After experiment			$\beta_{\text{Zn}}, \%$	$\beta_{\text{Pb}}, \%$
	mass of sample, g	content, %		mass of sample, g	content, %			
		zinc	lead		zinc	lead		
1	4.02			1.72	0.23	0.0085	99.27	99.53
2	9.81	13.70	0.80	9.40	11.1	0.415	22.40	50.00
3	14.40			14.34	11.0	0.40	20.07	50.43

Table 2. Content of chemical elements in the samples (recalculation to the initial amount with account for losses)

Element	Content of elements, g				
	298 K	573 K	873 K	1173 K	1423 K
Pb	0.80	0.736	0.801	0.613	0.132
Zn	13.70	12.830	12.961	13.128	13.025
C	1.74	1.571	0.524	0.010	0.009
Na	2.80	1.508	1.617	1.028	0.000
Cl	1.80	1.790	1.583	1.361	0.000
K	1.00	0.995	0.890	0.875	0.189
D&F, ng/kg of dust	474	440	0.35	0.1	0.1

2.5–3.0 cm. After removing the dust samples from the muffle furnace, they were placed into a desiccator for cooling (for a day).

The actual weight loss of electric arc-furnace dust during heat treatment was: at $T = 573$ K—0.544%; at $T = 873$ K—1.060%; at $T = 1173$ K—2.753%; and at $T = 1423$ K—5.613%.

All samples before and after the experiment were sent to the laboratory to determine the chemical composition (Table 2). The degree of extraction of zinc and lead from EAFD after heating is shown in Fig. 3.

During the experiment, it is found that when the dust is heated to 1423 K, its content of C, Na, Cl is reduced to traces; the K content is reduced by 81%; Pb—by 83.5%. A decrease in the zinc content does not exceed 5%. The predominant presence of chlorine in inorganic compounds in the form of NaCl, KCl, along with an insignificant presence of ZnCl, PbCl and PbCl₂, was confirmed [21, 22]. Chloride evaporation can lead to contamination of extracted non-ferrous metals [28].

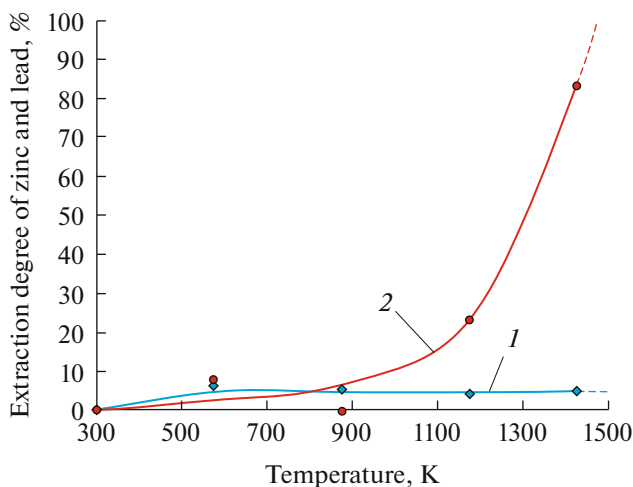


Fig. 3. Dependence of extraction degrees of lead and zinc from EAF dust on temperature of heating and holding: \blacklozenge —Zn; \bullet —Pb; (1) average Zn; (2) average Pb.

Extrapolation of the experimental data shows that the lead content in the sample under consideration decreases to traces, when the dust is heated to a temperature of about 1483 K (the maximum heating temperature of the samples in the muffle furnace was 1423 K).

It is important to note that the temperature ranges for the extraction of zinc and lead from EAFD depend on its content of carbon and oxygen. The oxygen content in the system (hence, the value of the oxygen-to-carbon ratio) accepted in the simulation may differ from the real one, which may lead to an error in the choice of the temperature regime for the selective extraction of lead and zinc for the considered dust composition. These questions and the observed partial extraction of zinc at low temperatures require further research.

According to experimental data, the most intensive D&F removal occurs in the temperature range of 473–873 K. Since D&F destruction occurs at temperatures above 1073–1123 K [29, 30], it can be assumed that desorption of dioxins and furans is observed from EAFD surface without destroying them. In this regard, the search for technologies aimed at reducing the D&F emission in the process of electric smelting becomes more significant.

CONCLUSIONS

Thermodynamic modeling of the process of selective extraction of zinc and lead from EAFD has been carried out. It has been established that the most favorable conditions are observed when the ratio of oxygen to carbon in dust is in the range of 12.7–25.0 (at the carbon content of no more than 3%). In this case, the zinc extraction (its oxide) proceeds at 1800–2050 K, extraction of lead (lead oxide) at 1350–1400 K.

Experiments in a laboratory muffle furnace have confirmed that when the dust is heated to 1423 K, its lead content decreases by 83.5%. The decrease in the zinc content does not exceed 5%. Extrapolation of the experimental data shows that the lead content in the sample under consideration will decrease to traces, when the dust is heated to a temperature of about 1483 K.

Experimental studies in a plasma furnace have shown that the degree of zinc extraction at high temperatures (2500–3000 K) exceeds 99%. Thus, the use of plasma technology for processing EAF dust, previously purified from lead, will allow obtaining pure zinc product, which is in high demand for the industry. This will increase both manufacturing and economic efficiency of electric steelmaking dust processing, as well as reduce the load on the ecosystem. In addition, the refusal to use an additional reducing agent will significantly reduce the emission of CO₂—the main greenhouse gas.

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