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Energy Efficient System for Galena Concentrate Processing

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ABSTRACT A new system of processing galena concentrates has been developed using the non-blast method of lead reduction from its sulphides by metallic iron. Excavated slag was used as flux agents to generate the stackable sulfur-containing products, instead of sulfuric acid. Based on this relatively modern technical concept of extreme energy saving, the new system of concentrate processing has been developed. The system includes an iron reduction and zinc sublimation system from excavated slag by soot-hydrogenic mixture, products of pyrolysis of natural gases, lead ousting in crude metal from its sulphides by metallic iron in the autogenous conditions of the rotary mixer, regenerative utilization of thermic wastes of the system, and neutralization of SO_2 gases by reagents obtained aboard (ZnO, Fe). The new system will reduce the effective specific equivalent fuel consumption by a factor of 3.77, and it will also expand the utilization factor of material waste products by a factor of 2.5–3 in comparison with the traditional methods of concentrate processing.

INDEX TERMS Inversion phase-rotary kiln, galena concentrate, excavated slag, mineral processing, soothydrogenic mixture.

I. INTRODUCTION

In the second half of the $20th$ century, non-ferrous metallurgy marked several technical milestones throughout the world [1]. During the same period, there were many technological advancements in the field of non-ferrous metallurgy in former USSR, however, due to various political and language barriers the development confined to the region itself [2], [3]. Scientists in USSR contributed to the improvement of existing technologies and development of new processes and equipment for treating complex metallurgical raw materials [4]. As such, the processes for torch-cyclone smelting, flash smelting, liquid bath smelting, fluid-bed smelting and other advanced technological processes were introduced, with the view of treating metal-rich raw materials [4]. Galena is the natural mineral form of lead (II) sulfide. Also known as galena glance, it is an abundant sulfide mineral that crystallizes in the form of octahydral structure. Most often, Galena concentrates are mixed with various impurities such as quartz and arsenic which makes the lead separation process complicated. It has been a known fact that the poorer the galena concentrate, the more energy consuming is the metal extraction process.

Nowadays, along with the traditional processes such as agglomeration (shaft smelting) fuming, new modern processes of lead smelting such as Kaldo in Scandinavian region, [5], [6], QSL in Germany [7], [8], Ausmelt/Isasmelt in Australia [5], [9], KIVCET in Kazakhstan [10], [11] and SKS in China [12], [13] have been industrially tested. The above listed processes work on the rich mixture of secondary raw materials and high-grade lead concentrates with 50 - 70% lead content. The limitations of raw material quality in such processes are determined by the intensively emanating gas jet into the bath of the lead rich slag and lead compounds of high volatility, which limits the temperature conditions [5]–[13]. The loss of the quality of raw material leads to the increase of lead content in the melted slag, which necessitates the increment in temperature. As a result, the process switches from the conditions of lead smelting to that of lead distillation into sublimates and dust; a process similar to the slag fuming.

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According to the research, reported in the literature [14], [15], for the Ausmelt process with 50% lead content in the raw material, the lead yield is 30 - 35% in the crude metal, 40-45% in the slag, and 20 - 30 % in the dust. In addition to that the modern processes of the lead raw materials processing keep the shortcomings of the traditional process such as enormous heat loss and environmental pollution during the water-stream granulation of the fuming slag; dumping with 2-3% of zinc, up to 1% of copper, 30% of iron and up to 60% of silicate part of slag. The storage and marketing of poor quality sulphuric acid pose many problems for the producer.

As such and the intensive exploitation of natural resources by the mankind, leads to their rapid depletion, which forces the scientific frontiers to search for new, more cost and resource effective technical solutions. There is also a demand for methods of waste-free processing of substandard and difficult-to-work-with minerals and secondary raw materials [16]. This study has been conducted in such backdrop to address the present day needs in this field.

II. METHODOLOGY

The proposed work is based on the results of experiments on reactor of inversion phase and a new method of scientific knowledge - method of extreme energy saving (MEES), [17], [18]. MEES describes the search of efficient variants of mineral processing in the following three directions: (I) selection of energy-saving melting equipment - main component of circuitry; (II) formation of waste-free, environmental friendly technologies; (III) development of thermal scheme, based on the proposed equipment and technology. As the proposed new method itself is a part of the outcomes of this study, the description is included under the section III below.

III. RESULTS AND DISCUSSION

The new method, the smelt layer of inversion phase, described below follows the above steps. In contrast to the technology of boiling layer of smelt [17], which works in conditions of ideal mixed layer, the newly proposed inversion phase layer method is a combination of ideal mixed and ideal displacement conditions [18]–[22]. It pave the way to develop the new unit 'reactor of inversion phase-rotary kiln' [18]. Figure 1-5 explains the process flow and principle of the new system.

Below is the process flow of the new mechanism;

- a. The precipitation of smelting was based on the reaction of lead ousting from its sulphide by metallic iron; $PBS + Fe = PB + FeS$
- b. The neutralization of $SO₂$ containing gases with metal iron and aqueous suspension of zinc sublimates; $SO_2 +$ $3Fe = FeS + 2FeO$; $SO_2 + ZnO + 2.5H_2O = ZnSO_3$. $2.5H₂O$
- c. The application of soothydrogenic mixture. This is one of the most effective reducing agents, extracted from the natural gas by its thermal decomposition.

For the calculation of thermal scheme, the following chemical composition of primary raw material and productions were considered in mass percentage:

FIGURE 1. A simplified thermal circuit of the non-waste galena concentrate processing system (pilot plant). 1 – reactor inversion phase with cylinder-separator, 2 – rotating part of the kiln, 3 – fixed part of the kiln with heat exchanger, for pyrolysis of natural gas, 4 – air heater, 5 – supersteam heater, 6 – additional natural gas input collector, 7 – slag loading tube, 8 – rotary kiln drive, 9 – densification of rotating part of kiln clearance, 10 – ejector scrubber, 11 – combustion chamber, NG (Π P Γ) – natural gas, SHM(CBC) – soot-hydrogenic mixture, CG ($\Gamma\Gamma$) – combustible gas, S (III) – slag, M(P) - melt.

FIGURE 2. Model of a pilot plant for processing of non-waste galena. A-Bunker, B-Heat exchangers, C-Rotary kiln, D-RIPh, E-Electric settler, F-Cupreous cast iron, G-Cast stone from silicate slag.

FIGURE 3. The Scheme of hourly consumption of iron according to the new system of lead production (7t) s, (1.5t) con: the amount of iron coming into the system with slag and concentrate, (2.5t) ss, (1.9t) ci: the amount of iron in the silicate slag and cast iron relatively.

a. Galena concentrate: P_B (45%-50%), Zn(4%-14%), Cu (up to 3%), S (15%-20%), Fe (4%-15%), CaO $(2\% - 10\%);$

FIGURE 4. The principle of the reactor of inversion phase. 1 oxidative chamber, 2 recovery chamber, 3 separation chamber, 4 partition, 5 burning devises.

- b. Slag from ''poor'' dumps: ZnO (3.5%-4.5%), PbO $(0.1\% - 0.15\%)$, Cu $(0.6\% - 1.0\%)$, FeO $(7\% - 8\%)$, Fe₂O₃ $(2\% - 3\%)$, Fe₃O₄(2\%-3\%), SiO₂(27\%-28\%), CaO, MgO (13%-14%), $Al_2O_3(7\% - 9\%)$, S (0.4%-0.5%)
- c. Rough cupreous cast iron: Fe (80%-85%), Cu (2%-3%), S (1%-1.5%), C (3%-3.5%), outlet of refined cast iron from rough one (85%-90%), matte outlet from consumable of sodium sulfide (85%-90%)
- d. Silicate melt: Fegen(5%-10%), CaO,MgO (35%-38%), $SiO₂(38\% - 39\%)$, $Al₂O₃(3\% - 4\%)$, suited to following technological conditions of acquisition of slag-wool: the acidity module of melt $M_K > 1.5$; viscosity module of melt $M_B < 1.2$; water resistances indices $\prod_B < 5$, melt viscosity at the exit from notches of reactor of inversion phase $\eta = 3.14$ Poise.
- e. Neutralization products of SO_2 -gases by zinc sublimates: $[ZnSO_3 \tcdot 2, 5H_2O, ZnSO_4 \tcdot 3(OH)_2 \tcdot 4H_2O,$ Zn $(OH)_2$] (80%-85%), [Pb SO₃, Pb SO₄] (5%-10%), $SiO₂ (5%-10%)$
- f. Lead refining from copper, tellurium, arsenic, antimony, silver, bismuth, and dross processing is omitted in circuitry, but their power consumption is taken into account in generalized type in Table; for the purification of lead from antimony and arsenic, the variant of bath air blow-off was considered; in this case, into floating passes the compounds of arsenic and antimony in trivalent condition and the air oxygen responds with lead according to the reaction $[Pb]+0.5O_2 = (PbO)$, then $3(PbO) + 2[Sb] = 3[Pb] + (Sb₂O₃), 3(PbO) +$ $2[As] = 3[Pb] + (As₂O₃)$; couplings formed after blowing the lead bath by air: $5PbO·Sb₂O₃$, $3PbO·Sb₂$ O_3 , PbO·Sb₂O₃, 5PbO· As₂O₃, 3PbO· As₂O₃, PbO· As2O3; content in melt floating - Sb (0.8%) and As (0.4%).

To overcome the above disadvantages of the traditional and modern systems of lead production, a new heat scheme of galena concentrate processing has been developed as shown in the simplified diagram in Figure 1. Figure 2 shows the simplified 3-D diagram of the pilot plant.

The conventions and symbols/signs used in the figure-5 and also in the text are: rotary kiln (RK), reactor of inversion phase (RIPh), electric settler (ES), electric furnace for copper and noble metal extraction from cupriferous cast iron into the copper-rich matte (EF), electric settler (ES), slag wool production (SWP), rotary mixer (M), high-temperature air heater (HTAH), reactor of soot-hydrogenic mixture (RSHM), radiation part of recovery boiler (RRB), mini electric power plant (MPP), chemical reactor of primary neutralization of $SO₂$ gases (ChR), convection part of recovery boiler (CRB), scrubber for secondary neutralization of $SO₂$ gases by aqueous suspension of zinc sublimates (Sc). L, S, Sm, SMS, SS, SW – limestone, solid slag, smelt, slag-metallic smelt, silicate slag, slag-wool, correspondingly; Pbg, Zng - lead and zinc gaseous, $Pb¹$ – liquid lead, Fe_{mat} - crushed matte rich in iron, Cumat – rich in Cu matte, CCI - Cu –contained cast iron, CI - cast iron, correspondingly; NG – natural gas, CG – combustible gases, SHM – soot-hydrogenic mixture, E – electric power, FW – feed water, OHS – overheated steam, DS – dead steam.

The operation principle of the system is as follows; The mixture of lead concentrate pellets and crushed limestone is fed into the rotary kiln (1) for pellet heating and limestone de-carbonization. The source of energy is the physical heat of the combustible gases from RIPh's recovery chamber (Figure 4) with temperature $t = 1500^{\circ}$ C. The charge heated up to $1000 \degree$ C is placed into the rotary mixer (6), where the de-copperized cast iron with temperature at 1400◦ C is filled from the electric settler (4a). In the mixer (6) the mixture of cast iron melt and the charge is intensively mixed. The precipitation melting process is based on the reaction of lead ousting from its sulphides with metallic iron contained in the cast iron, slag and matte formation and the exothermal reaction of CaO dissolution in the slag takes place. The source of energy is the extra heat of cast iron and the heat generation of scorification process in the mixer (6).

In the electric settler (4b), the separation of the melting product precipitation takes place, after which the rough lead is transferred to the refining process. The iron-rich slag and matte, after cooling and crushing together with poor solid slag, is continuously fed into the rotary kiln (2). The mixture of crushed matte and slag from rotary kiln (2), heated up to 900[°]C is directed into the RIPh's oxidizing melting chamber (1) (Figure 4), where melting and deep desulfurization takes place. The fuel is sulfur in the matte and natural gas, and the oxidizing agent is blast air with temperature at 800◦ C.

The production of oxide melt goes through the RIPh's oxidative chamber to the recovering chamber (2), where lead and zinc are sublimated to the gas phase. The reduced iron, in the form of cupriferous cast iron, is then transferred into the metallic phase. From RIPh (3) (as it is shown in Figures 1, 2 $\&$ 5) slag-metallic smelt flows into the electric settler (4), where they are divided into the cupriferous cast iron and silicate slag. After copper and noble metals are refined by sodium sulfide in the electric furnace (4a), the cupriferous cast iron is used as an iron-reagent in the

FIGURE 5. The complete thermal process flow diagram of the non-waste galena concentrate processing system.

rotary mixer (6) and for the purpose of primary neutralization of $SO₂$ gases in the chemical reactor (11).

There is a scheme of iron-reagent motion along the system (Figure 3). The matte, containing copper and noble metals, goes from the electric furnace (4a) for further processing, according to the described scheme. The silicate melt undergoes slag wool production (5) to produce the insulating materials. The combustible gases from RIPh's (3) recovery chamber, containing Pb^{g} , Zn^{g} , are distributed between the rotary kiln (1) and the radiation part of the recovery boiler (9), where compressor air is used as agent for oxidizing and injecting combustible gases (CG).

Then, the physical heat of CG is used in the rotary kiln (1) for concentrate-pellet heating and limestone de-carbonization. The CG before HT AH (7) and the reactor of SHM (8) are burnt up with the blast air. The waste gases from the RIPh's oxidizing chamber (1), containing SO2, will heat the slag and matte charge in the rotary kiln (2) and then will flow into the chemical reactor (11) for the primary neutralization of SO2. The reagent is the cast iron obtained in the system. The storable products of the chemical reactor FeO, FeS (vs sulphuric acid) can be used, for example, for the production of green vitriol. The waste gases of the chemical reactor (11), HT AH (7) and the reactor of SHM (8) are heated with blast air, natural gas and feed water in the convection part of the recovery boiler (CRB) (12). Then during the process of cooling they go to the scrubber (13) for the secondary neutralization of SO2 gases with zinc sublimates containing in the gases.

The cleansed gases after the scrubber are ejected into the atmosphere. Then the overheated steam from the radiation part of the recovery boiler (9) is directed into the mini power plant (10). The dead steam of is used for slag-wool production and thermal treatment of insulating materials in the polymerization chamber (5). The generated electricity of the mini power plant (10) is used inside the system. The system can also operate without dump slags, in which case, except for the matte, the cakes of zinc production, zinc-containing circulations, fluxes and the others can be fed into the rotary kiln.

The Table-1 depicts the results of the new system based on the process parameters of the Chymkent lead plant (ChLP), (Kazakhstan) during the period of commercial experiments.

The following data of the ChLP pertinent to the work in 1998 is taken into account:

Concentrate charges 1000 tons per 24 hours (41.66 tons per hour), burden material 840 tons per 24 hours (35 t per hour); the production of refined lead 135000 tons per year; operating period of lead smelting furnace 300 days per year. Comparison of the data in Table-1 shows that with the implementation of the new system, the specific natural gas consumption will increase by 3.5 times, the power energy by 1.3 times, while the total energy intake in term of conventional fuel will increase 1.64 times. Water rate will decrease 1.92 times and heat loss 2.06 times respectively. The waste of slag valuable components (P_B, Zn, Cu, Fe) on average will be reduced 3 times, the usage of power energy of sulfide sulfur will increase 8 times, the conversion of the silicate part of slag

TABLE 1. Comparison of the characteristics of the new system of lead production.

TABLE 1. (Continued.) Comparison of the characteristics of the new system of lead production.

into the useful product will increase 6.7 times. The need for the use of the process oxygen, small-sized coke and deficient and expensive coke will be eliminated. Instead of the imported components of burden (cakes, dust, clinker, fluxes) the own slags from poor zinc slag dumps will be used. As the subsidiary output $-ZnSO₃$, FeS, FeO, cupriferous cast iron, slag wool wares and power energy will be produced in a new system, the fuel saving versus production of substitutional aggregates with the same output is

$$
2388 \frac{K\Gamma y \cdot T}{\tau P_6}
$$

Taking the above into account, the effective specific equivalent fuel consumption will decrease 3.77 times in the new system and the saving of fuel will be

.

$$
960 \frac{kg \ c \cdot f}{tP_{\beta}}.
$$

The value of the specific economical effect by the application of the new system is 31.3 USD per ton of Pb (about 1 million USD per year), whereas the specific maximum capital investment is 85.5 USD per ton of Pb. These values are significantly less than the cost of operation of existing conventional systems. The conventional methods used at the same site of this study, for the galena concentrate processing cost more than twice that is required for the proposed method. Some other studies that proposed for similar processing operations; eg. Li *et al.* [23] where laboratory investigation has been done on the solidification of chromite ore processing

TABLE 2. Comparison of the specific fuel consumption in processing of lead concentrates.

residue (COPR) by blast furnace slag and calcium hydroxide, to find the enhancement effect of pre-reduction using zero-valent iron (ZVI) on the solidification treatment; have shown good potentials of cost reduction in the operations,

TABLE 2. (Continued.) Comparison of the specific fuel consumption in processing of lead concentrates.

however many of these studies have not done any attempts on conducting cost benefit analysis. A somewhat similar study done by Dikhanbaev *et al.* [24] also give prominence to the energy saving technique itself, which they have proposed, rather than concentering on the costing. The proposed method can be further improved by adopting some novel techniques introduced in the fields of metal extraction processes in removing heavy metals from waste sources [25], [26].

Computation of the specific fuel consumption in processing of lead concentrates:

To calculate the parameters of conventional system (CS), the following data of the Chimkent Lead Plant were taken:

- Consumption of concentrate 1000 tons/day (41.66 tons/hour), charge additives 840 tons/day (35 tons/hour);
- Productivity of refined lead 148,694 tons/year, for liquid slag 159,103 tons/year, for granulated slag 26897 tons/year;
- Working campaign of shaft lead smelting furnaces 324 days/year, fuming installation - 310 days/year.
- For the proposed system (PS), the concentrate consumption is 10 tons per hour, the consumption of ''poor'' zinc dump slag is 25 tons/h.

The outcomes of the calculations are depicted in Table-2. Specific consumption of reference fuel in the conventional (CS) and proposed (PS) systems:

$$
B_{red}^{CS} = \frac{B^{CS} - B_{sub}^{CS}}{P_{Pb}^{CS}}; \quad B_{red}^{PS} = \frac{B^{PS} - B_{sub}^{PS}}{P_{Pb}^{PS}}, \quad (1)
$$

 B^{CS} , B^{PS} - The fuel consumption in CS and PS; B_{red}^{CS} -reduced specific fuel consumption.

 $B_{\text{sub}}^{\text{CS}}, B_{\text{sub}}^{\text{PS}}$ – The conventional fuel consumption in "substituting'' products of the same quantity and quality as in the compared system, $B_{\text{red}}^{\text{CS}}, P_{\text{Pb}}^{\text{PS}}$ – refined lead productivity in CS and PS.

$$
B^{CS} = B_{NG} + B_C + B_E + B_{O_2} + B_S^{saw} + B_S^{con}
$$

\n= 9080 + 9565 + 11449 + 765 + 30 + 284
\n= 31173 kgc.f./h
\n
$$
B^{PS} = B_{ng}^{ox} + B_{ng}^{rec} + B_{ng}^{ref} + B_{ng}^{ref} + B_{settl}^{Ed} + B_{cu}^{Ed} + B_{sw}^{Ed}
$$

\n
$$
+ B_C^{E} + B_{dbm}^{E} + B_{pump}^{E} + B_S^{con}
$$

\n= 437 + 7813 + 154 + 65 + 295 + 360
\n+404 + 552 + 1878 + 35 + 68
\n= 12061 kgc.f./h
\n
$$
B_{sub}^{CS} = P_{Pb}^{sublim} \cdot B_{Pb} + P_{Cu} \cdot B_{Cu} + P_{Zn} \cdot B_{Zn}
$$

\n+ (G_{ss}·i_{ss})/q_{c.f.} + P<sub>H₂SO₄·B<sub>H₂SO₄
\n= 0, 35 · 580 + 0, 8 · 1290 + 2 · 912
\n
$$
- \frac{25000 \cdot 3175}{29330} + 16 \cdot 56
$$

\n= 666 l k s c f./h
\n(4)</sub></sub>

$$
B_{sub}^{PS} = P_{Cu} \cdot B_{Cu} + P_{Zn} \cdot B_{Zn} + P_{Fe}^{FeS} \cdot B_{Fe}
$$

+ $P_{Fe}^{FeO} \cdot B_{Fe} + P_{ci} \cdot B_{ci} + P_{sw} \cdot B_{sw} + W_{E} \cdot B_{E}$
= 0, 495 \cdot 1290 + 2, 351 \cdot 912 + 1, 368 \cdot 500
+2, 658 \cdot 500 + 170 \cdot 19 + 5007 \cdot 0, 5
= 10529 \text{kgc.f./h} (5)

where B_{Pb} , B_{Zn} , B_{Cu} , B_{Fe} , B_{ci} , $B_{H_2SO_4}$, B_{sw} , B_E – specific consumption of conventional fuel in the ''substituting'' installations, producing Pb, Zn, Cu, Fe, cast iron, sulfuric acid, slag wool, and electricity, (Reference data).

$$
B_{\text{red}}^{\text{CS}} = \frac{31173 - 6661}{18,75} = 1307 \text{kgc.f.}/\text{tPb}
$$
 (6)

$$
B_{red}^{PS} = \frac{12061 - 10529}{4,41} = 347 \text{kgc.f.}/\text{tPb}
$$
 (7)

One of the key energy efficiency criteria is the ratio of the specific fuel consumption in the compared systems:

$$
\frac{B_{\text{red}}^{\text{CS}}}{B_{\text{red}}^{\text{PS}}} = \frac{1307}{347} = 3,77
$$
 (8)

and practically achievable level of energy saving in the proposed system is

$$
\Delta B_{\text{prac}} = B_{\text{red}}^{\text{CS}} - B_{\text{red}}^{\text{PS}} = 1307 - 347 = 960 \text{ kgc.f.}/\text{tPb} \qquad (9)
$$

Thus, in the case of implementing the proposed system for lead production, the specific fuel consumption will decrease by 3-4 times compared to the conventional system; ''agglomeration - shaft smelting - fuming'', and the fuel savings from each produced ton of lead will be proportionate with the reduced fuel consumption for the production of one ton of zinc in sublimates $(B_{Zn}^{HP} = 912 \text{ kgc.f.}/tZn)$.

IV. CONCLUSSIONS

A new, non-waste, galena concentrate processing system with excavated slag used as the flux agent is developed. In the system, non-blast method of lead ousting from its sulphides by metallic iron has been employed. For iron reduction and zinc sublimation from excavated slag, the new melting unit reactor inversion phase has been selected. The system produces stackable sulfur-containing product, ZnSO3, FeS, instead of sulfuric acid. Implementation of the new system reduces the effective specific equivalent fuel consumption 3-4 times.

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