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DEVELOPMENT OF HYDROGEN-ENRICHED WATER GAS PRODUCTION TECHNOLOGY BY PROCESSING EKIBASTUZ COAL WITH TECHNOGENIC WASTE

In the dumps of metallurgical enterprises of Kazakhstan about 700 million tons of waste products are generated annually, and are polluting the atmosphere and the soil. The concentration of valuable components in waste products are no lower than in natural resources. The reserves of coal in the Ekibastuz basin are estimated to be more than a billion tons, and almost half of this is made up of ash. Every year, up to 30 million tons of ash-cinder waste is generated, which presents a serious threat to nature. Gallium and germanium concentrations in dumps are approximately 200 grams per ton, which is comparable to the content in coal before processing. The current research aims at creating a unit to obtain hydrogen-enriched water gas from Ekibastuz coal, with the production of zinc, gallium and germanium sublimates, copper-containing cast iron, slag wool and cast stone, through the joint processing of zinc-rich slag and ash-cinder wastes from thermal power plants. To achieve this, we used previous methods of extreme energy saving and a new method, the smelt layer with inversion phase. Experimental results from the "reactor inversion phase – rotary kiln" (RIPh) unit, which processed zinc-germanium contained slag, showed the potential to extract germanium from zinc sublimates, to reduce iron to the form of cupreous cast iron, and to obtain combustible gases and smelt suitable for slag-wool production. Calculations performed on the joint processing of Ekibastuz coal and zinc-rich slag using the proposed unit "reactor of inversion phase – rotary kiln – gas generator" showed it can obtain hydrogen-enriched water gas, along with the extraction of valuable components of primary raw material.

1. Introduction

In the dumps of metallurgical enterprises of Kazakhstan, billions of tons of waste products are accumulated, in which the content of valuable components is often higher than in natural ore deposits [1, 2]. The annual output of ash-cinder

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from thermal power plants, from only burning of Ekibastuz coal, that characterized by a high ash content (40–45%), is averaged 30 million tons. As well as the initial coal, ash emissions comprise up to 200 grams of gallium (Ga) and Germanium (Ge) per ton, which is wasted in dumps. By this time, more than 300 million tons of waste products are accumulated [3–5].

Previous research that was carried out examined the following: (1) ash-cinder utilization in road construction; (2) building material production, such as cell concrete and ash concrete, from waste products; (3) laboratory studies on the extraction of radioactive metals, alumina, and silica with the help of chemical reagents, from ash-cinder; (4) the extraction of an iron (Fe)-containing fraction using humid magnetic separation; (5) the separation of unburnt coal using the flotation technique; (6) ash utilization in the composition of reagents for the deep purification of sulphate-ions from sewage [6–10]. However, despite their importance, all the aforementioned research focuses on the processing of waste products from coal, following the principle they must be processed after they are produced. A future direction, in our opinion, would be the creation of technology that allows gasification of Ekibastuz coal with waste-free processing of the ash portion of coal, production of Ga and Ge sublimates, and smelt production with a composition suitable to produce construction materials.

Gas production from coal with high ash content in fluidized-bed generators, despite having high efficiency, has the following flaws:

- necessity of air or oxygen blowing;
- moderate calorific value of gas – 4200–4600 kJ/m^3 using steam-air blast, 8800–9200 kJ/m^3 using oxygen-steam blast;
- low efficiency in the utilization of the chemical energy of fuel – 50–54% (gasification efficiency);
- low degree of steam decomposition – 20–40%;
- high specific consumption of oxygen, which has high energy requirements;
- necessity of extreme humidity coal drying [11, 12].

The extraction of valuable components from metallurgy waste products using traditional methods requires two to three times greater specific fuel consumption in comparison to its production from natural raw material. Taking into account the fact that rich and valuable components of polymetallic ores in Kazakhstan are predicted to deplete in 30–35 years [1], the development of new forms of cheap fuel, the creation of energy-saving equipment and technology of complex processing of technogenic waste products, along with reducing specific fuel consumption (suitably CO_2 emissions), are urgent challenges that must be addressed [13].

2. Research methods

The current study uses basic techniques of extreme energy saving [15] and mathematical modeling of charge melting, zinc distillation, iron reduction from smelt to predict fuel consumption in the reactor inversion phase – rotary kiln (RIPh)

[16–18]. In addition, the chemical and physical properties of charge were calculated, to determine their suitability in the production of construction materials that require specific compositions [21]. The method of extreme energy saving is a search technique of specific ways and tools for development of heat technical systems, differing by the high technical and economic characteristics. Applied to mineral processing, the main directions of research are: the selection of technology based on waste free principles of process; development of science bases and creation of high efficiency melting equipment; development of energy-saving thermal circuitry implementing the given technologies and equipment.

3. Experiment results from the "reactor inversion phase – rotary kiln"

The RIPh is a new generation of melting equipment that uses the combination of two conditions, the ideal mixed and ideal displacement of smelt [19–24] (Fig. 1). The RIPh is composed of a smelting chamber, blow through lattice and combustion chamber for natural gas conversion (Fig. 2). The operating principle of RIPh is as follows. Natural gas and combustion air are supplied to mixing chambers through combustion chamber swirlers (3) and further to the burning chamber (4). Conversed gas comprising of reducing agents (CO, H_2), with a temperature of 1800–2000°C, at a rate of 500–600 m/s, pass through a lattice via nozzles (1), and is then fed under the layer of smelt in the smelting chamber for slag melting, zinc (Zn) and Ge sublimation, and iron reduction from oxides. Waste gases move through the separation chamber portion and are directed into the rotary kiln.

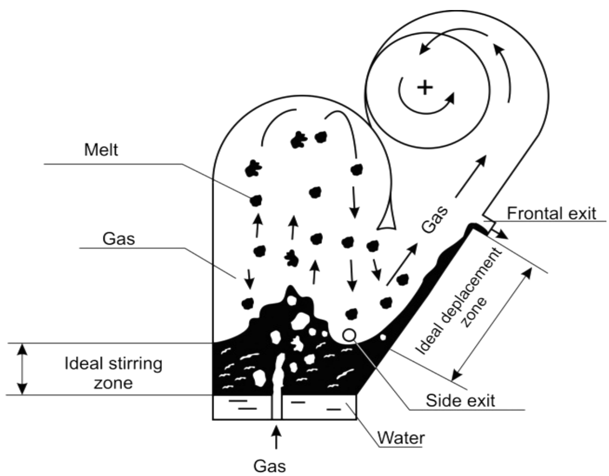

Fig. 1. Basic circuit of smelting chamber of reactor of phase inversion

The operating principle of the existing unit is as follows (Fig. 3): excavated slag from hoppers is constantly fed into the rotary kiln, where it is heated to 900°C by waste gases of reactor inversion phase (RIPh) and is routed into the same RIPh

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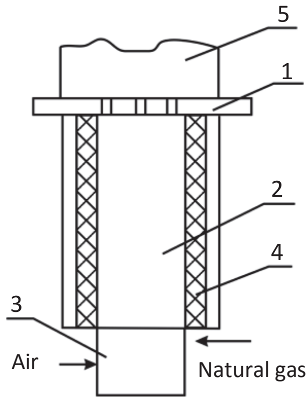

Fig. 2. Basic circuit of elements of combustion chamber and blow through lattice of reactor inversion phase. 1 – blow through lattice, 2 – burning chamber, 3 – mixing chamber, 4 – lining, 5 – smelting chamber of reactor inversion phase

for slag melting, Zn distillation and iron reduction in the form of cupriferous cast iron, at the smelt temperature of 1400–1450°C, and it is a continuous process. In the caissons of the reactor inversion phase, steam is generated with technical parameters. The chemical composition of excavated slag, wt%: ZnO (3.5–10); PbO (0.1–1.15); Cu (0.6–1.0); FeO (7–8); Fe_2O_3 (2–3); Fe_3O_4 (23–24); SiO_2 (27–28); CaO (13–14); Al_2O_3 (7–9); S (0.4–0.5).

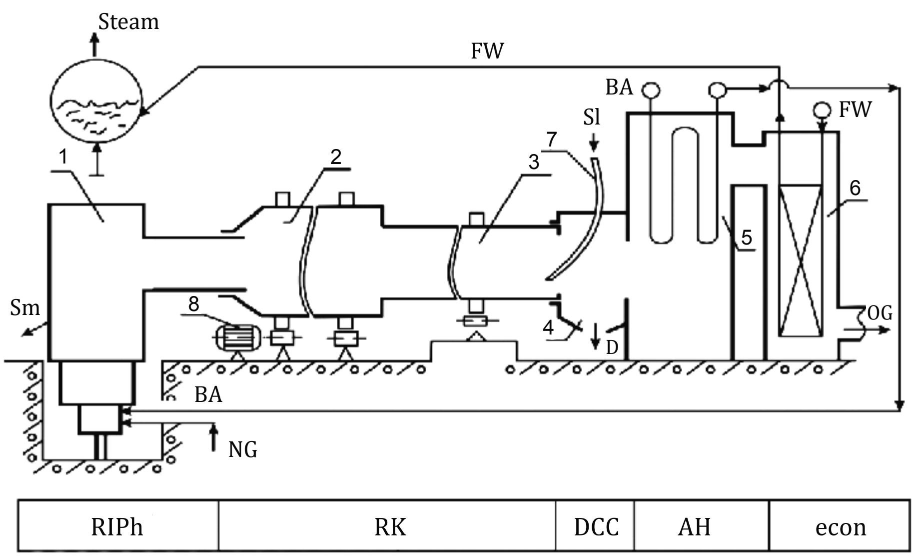

Fig. 3. Basic circuitry of existing pilot plant: 1 – reactor of inversion phase (RIPh), 2 – discharging and 3 – loading parts of rotary kiln, 4 – dust collector chamber (DCC), 5 – air heater (AH), 6 – economizer (econ), 7 – slag loading tube, 8 – drive of rotary kiln; Sl – slag, Sm – smelt, BA – blowing air, NG – natural gas, D – dust, FW – feed water, OG – off gases, RK – rotary kiln

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The combustible waste gases of RIPh, with temperature of 1450–1500°C, are transferred to heat slag in the rotary kiln and are further directed to the air heater for combustion air preheating. Following that, the combustible gases are cooled in the economizer to 200°C, and are directed into the scrubber for further cooling and catching of Zn and Ge sublimates in slurry. The combustible gases from the scrubber, during the experiment, are flared into the atmosphere.

The tests on the unit "reactor inversion phase – rotary kiln" (RIPh–RK) with output capacity 1.25 t/h, (fuel – natural gas, oxidizer – oxygen enriched up to 25% of the combustion air) showed that the reduction value of Zn and Ge from granulated slag in the reactor inversion phase amounted to approximately 73–75%, and the specific primary fuel, in comparable conditions, was two to three times lower than when processing the same sort of slag in the Waelz kiln of Leninigorsk polymetallic plant [25].

The results from examining the iron recovery from excavated slag, in the RIPh – RK unit showed the following:

- the percentage of zinc recovery in sublimates and iron in cupriferous cast iron – 70–75%;
- the composition of the silicate phase of smelt %: FeO (9–11), SiO_2 (40–42), CaO (23–24), Al_2O_3 (12–13), which may be used to produce slag wool or cast stone;
- calorific capacity of gas after rotary kiln – 7000–7500 kJ/m^3, heating power 1800–1900°C (for comparison, heating power of natural gas 2000–2020°C). The process had the following characteristics: output capacity on slag – 1000 kg/h; natural gas consumption – 600 m^3/h; coke dust – 320 kg/h, combustion air – 1400 m^3/h; oxygen – 430 m^3/h; electric power – 400 kWh; discharge correction factor of oxidizer – 0.4; reduction potential of gases in combustion chamber – 84–85% [25–27].

Based on results of the experiments, the recalculation of pilot plant characteristics, with a slag capacity of 1000 kg/h, was performed on an industrial sample with slag capacity of 31000 kg/h. After recalculating, the relation of specific fuel consumption in Waelz kiln of Leninigorsk polymetallic plant b_{Zn}^{wk} = 6000 kg of comparison fuel/ ton of Zn to equivalent specific fuel consumption b_{Zn}^{eq} = 1404 in unit RIPh–RK was 4.27 [26]. In the case of equivalent specific fuel consumption in RIPh–Rk, the emissions of CO_2 will be approximately 4 times lower than in Waelz-kiln of Leninigorsk polymetallic plant.

4. Results of calculated estimation of technological scheme of unit "reactor inversion phase – rotary kiln – gas generator"

The main component of the technological scheme explored is the unit comprised of existing melting equipment RIPh–RK and a standard gas generator (RIPh–RK–GG). The operating principle of the unit is as follows (Fig. 4). A continuous regime, granulated slag, crushed Ekibastuz coal, limestone and ash pellets from thermal power station dumps (the charge) are loaded into the rotary kiln (2), where they are heated up to 1000°C by reducing gases of RIPh. Here, the limestone decarbonization and the reduction of Fe from its oxides (FeO, Fe_3O_4, Fe_2O_3) takes place. The heated charge is supplied from the rotary kiln (2) to the gas generator (3) for coal gasification. The overheated steam is routed from the chamber (4) to the gas generator (3). The sensible heat of combustible gases of the rotary kiln heats up the saturated steam in the chamber (4) and the air in the air heater (5), and then the combustible gases are cooled down in the scrubber (6). The main stream of combustible gases after scrubbing, cleaning and compression, is injected into the reactor inversion phase for after-burning of carbon contained in ash-cinder of the gas generator (3), slag melting, iron reduction, and Zn, Ga and Ge sublimation. A portion of combustible gases are used for technological needs, for example the thermal treatment of cast stone or in the chambers of slag carpet polymerization. Zn, Ga and Ge sublimates, trapped by the scrubber in slurry form, are further processed using a well-known method.

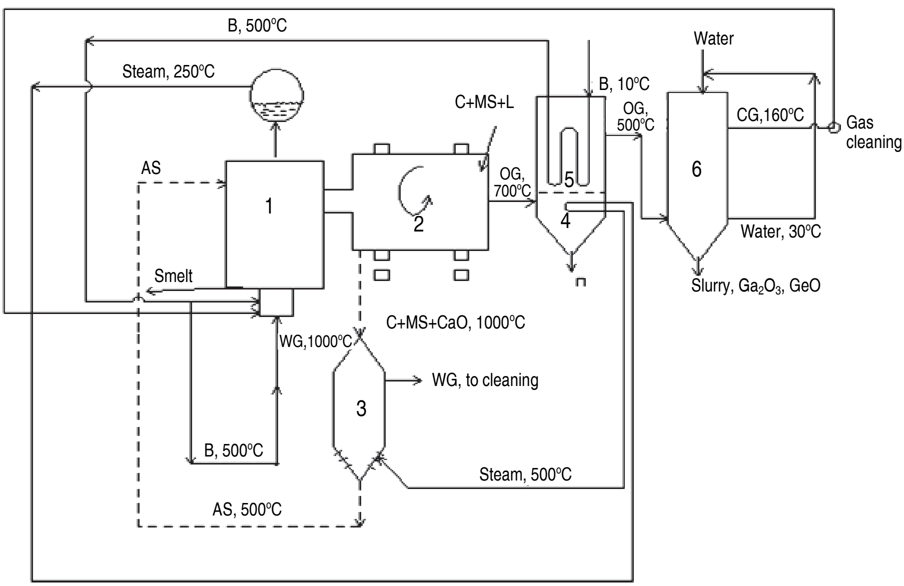

Fig. 4. Basic circuit of the proposed unit "reactor inversion phase – rotary kiln – gas generator": 1 – reactor inversion phase, 2 – rotary kiln, 3 – gas generator, 4 – after-burning chamber, 5 – air heater, 6 – scrubber; WG – water gas, B – oxygen enriched blast, MS – zinc-containing slag, L – limestone, П – dust, OG – outgoing gas, CG – combustible gas, AS – ash slag, C – coal

The slag-metallic smelt from the reactor is directed to the electric settler (omitted in Fig. 4) for separation of cupriferous cast iron from the silicate part of smelt. The latter is used to produce slag wool or cast stone. Water gas from (3) is channeled to cleaning, and then transported to consumers.

In the layer of the gas generator, endothermic and exothermic reactions occur:
Endothermic:

$$C + H_2O = CO + H_2 - 135.7 \ \text{kJ/mol}, \tag{1}$$

$$2Fe_3O_4 + H_2O = 3Fe_2O_3 + H_2 - 7.91 \ \text{kJ/mol}. \tag{2}$$

Exothermic:

$$2Fe + 3H_2O = Fe_2O_3 + 3H_2 + 61.8 \ \text{kJ/mol}. \tag{3}$$

Overall reaction for charge of any composition:

$$2C + (3 + 1.5n)H_2O + n\,Fe = C + 2H_2O + CO + 0.5n\,Fe_2O_3 + (1.5n)H_2\,, \tag{4}$$

where n is the mole amount of iron in excavated slag.

Eventually, the water gas is enriched by an additional mole of hydrogen. The moles' amount of hydrogen depends on the ratio of "coal/excavated slag" in the charge. The addition of charge components like Fe_2O_3 and Al_2O_3 would serve as catalysts to the gasification process, and Cu and S as modifiers to melt crystallization in the production of cast stone. The synthesis-gas produced, after the removal of harmful impurities, can be used for the production of industrial hydrogen and/or methanol. The possibility of methanol production from Ekibastuz coal with a billion-ton reserve, makes it possible to use it as an intermediate product in the production of synthetic motor fuel. For example, during the technological process "Mobile", the following cycle is performed: "coal-gasification → methanol → synthetic gasoline" [28].

The calculation of charge of the ash composition of Ekibastuz coal is, wt%: SiO_2 – 60; Al_2O_3 – 26; Fe_2O_3 – 8; CaO – 3; MgO – 1.5; (K_2O, Na_2O) – 1.5. The calculated composition of slag upon exiting the reactor inversion phase, wt%: FeO (7–8), CaO (34–35), MgO (3–5), Al_2O_3 (15–17), SiO_2 (41–43), K_2O, Na_2O (1.8–2.0). The slag wool produced from such a slag will satisfy the standard conditions stated in the brackets: acidity module of smelt – 1.88 > ($M_K \geqslant 1.5$); viscosity module of smelt 1.1 < (M_v < 1.2); water resistance index 3.95 < (Π_v < 5). Viscosity of homogeneous smelt for the stated composition, to temperature range 1400–1450°C – $\eta = 5.2 - 4.8$ poises, which makes it possible to release the smelt from notches in the RIPh.

For heat balance calculations of the RIPh–RK–GG unit, the basic reaction of gasification is used (5), with the condition that half of the unspent carbon in the gas generator is fed into the reactor inversion phase with the ash-slag, and 2/3 (66.6%) of the unreacted steam is transferred into water gas:

$$2C + 3H_2O = C + 2H_2O + CO + H_2\,. \tag{5}$$

The computational charge composition after the rotary kiln is as follows: carbon in coal – 30.76 kg; ash in coal – 30.76 kg; CaO – 7.7 kg; excavated slag– 30.76 kg; total – 100 kg. The ratio of "coal/excavated slag" is 2 : 1; iron percentage

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in excavated slag – 30%; amount of iron in excavated slag – $30.76 \cdot 0.3 = 9.228$ kg. For calculation of the heat balance, the exothermic effect of the reaction (3) in the gas generator was not taken into account.

The calculated calorific power by hydrogen enriched water gas after drying is $Q_{pH} = 11586$ kJ/m^3, which is close to its theoretical limit (11700 kJ/m^3). The gas high-temperature capacity is 2500°C, which is more than that of natural gas (2020°C), and that makes it possible to use in the melting – reduction processes.

Consequently, from Table 1, one can see that in the heat balance of the gas generator there is no requirement for the combustion of additional carbon, as is the case in traditional gas generators. The substantial contribution in balance is introduced by the heat from the charge, which is heated up to 1000°C (∼ 8%) and the steam is overheated up to 300°C (17%). Furthermore, the thermal effect of the total exothermic reaction – $2Fe + 3H_2O = Fe_2O_3 + 3H_2 + 61.791$ kJ/mol, which was not taken into account in the heat balance, provides an additional reserve of heat for the gasification process.

Table 1.

Heat balance of gas generator with 100 kg of charge

No	Input	%	No	Output	%
1	Sensible heat of charge $Q_s = 100$ kg $\cdot$ 1 kJ/kgK $\cdot$ 1000K = 100000 kJ	8.230	1	Sensible heat of ash-slag from gas generator $Q_s = 84.6$ kg $\cdot$ 1 kJ/kgK $\cdot$ 800K = 67680 kJ	5.583
2	Chemical heat of carbon provided by charge $Q_{ch} = 30.76$ kg $\cdot$ 29330 kJ/kg = 902191 kJ	74.253	2	Sensible heat of water gas $Qs = 84.6\rho_g \cdot 1.55 \cdot 1000$ $= 84.6 \cdot 0.9371 \cdot 1.55 \cdot 1000$ = 122869 kJ	10.136
3	Sensible steam heat from caissons of reactor inversion phase (P = 1.6 kg/sm^2; t = 300°C, $I_{pp} = 3075$ kJ/kg) $Q_s = 69,21$ kg $\cdot$ 3075 kJ/kg = 212820 kJ	17.515	3	Heat of endothermic reaction of water gas generation $Q_{end} = 15.38$ kg $\cdot$ 11308 kJ/kg = 173917 kJ	14.347
4	Total: $Q_{in} = 1215011$ kJ	100	4	Chemical heat of carbon in ash-slag $Q_{ch} = 15.38$ kg $\cdot$ 29330 kJ/kg = 451095 kJ	37.212
5	Imbalance: 0.23%		5	Total: $Q_{out} = 1212210$ kJ	100

5. Conclusions

In the unit RIPh–RK gas generator, the following flaws found in traditional gas generators are eliminated: the combustion of the coal portion for replenishment of the heat balance, heat loss at the processing stage of moist coal and loss of the

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coal portion with ash-slag waste. The computational calorific power of hydrogen-enriched water gas (11586 kJ/m^3) was more than on steam-air (4200–4600 kJ/m^3) and oxygen-steam blast (8800–9200 kJ/m^3), and approaches its theoretical limit (11700 kJ/m^3).

The joint processing of Ekibastuz coal and iron containing metallurgical slag makes it possible to obtain hydrogen-rich water gas, which can be utilized as a source in the production of technical hydrogen or methanol. The simultaneous production of several items (cupreous cast iron, Zn, Ga, Ge sublimates, slag wool/ cast stone) makes the water gas cheaper in comparison to the gas obtained using traditional methods. The energy-saving unit RIPh–RK gas generator has the potential for commercial applications, where huge dumps of metallurgical waste products and ash-cinder wastes of thermal power plants are accumulated and cheap fuel gas is not available.

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