Prospects of the Use of Ash-and-Slag Wastes from Fuel-Bed Firing of Brown Coal of the Kangalas Field of the Lena Basin

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Abstract

Granulometric and chemical composition of the ash-and-slag wastes (ASW) of fuel-bed firing of the brown coal from the Kangalas field of the Lena basin was investigated. The ash-and-slag material was separated by means of magnetic separation of the granulometric and hydrodynamic classification. The content of rare elements (RE) and rare-earth elements (REE) in the initial ash-and-slag and in separation products differing in particle size, density and magnetic properties was determined. It was shown that RE and REE content of the low-density products of hydrodynamic separation increases, while it decreases in the magnetic products. Manganese is also observed to get concentrated in the latter. In view of the low content of RE and REE in low-density products in comparison with the matter from ore deposits, it is concluded that the isolation of the metals from the investigated ASW is unreasonable. It was shown that the use of the investigated ASW in construction industry is promising; the compositions for obtaining nonfired bricks and building mortars of the grade strength were chosen.

INTRODUCTION

Coal from the Kangalas field (Yakutsk) relates to brown coal of B2–B3 grade on the basis of its genetic parameters and technological characteristics. Its properties are: vitrinite reflection index, 0.4–0.49; combustion heat value and the yield of volatiles in the fuel working state are 3660 kcal/kg and 49 %, respectively; maximal humidity is 30–33 %. The elemental composition of the dry ash-free coal (concentration, mass %) is: C 71.5, H 5.1, N 1.0, O 21.7 [1, 2]. The sulphur content of coal is low (S 0.6 %); ash content is 15 % [2]. According to the international coal classification (lignite, sub-bituminous, bituminous coal, anthracite), the Kangalas coal may be related to lignites on the basis of its metamorphism stage and heating capasity. In the Lena coal-bearing basin, the probable reserves of which exceed those of the Kuznetsk and Kansk-Achinsk basins, the Kangalas field is among the best proven ones and is considered as a reliable basis for obtaining power-plant fuel for long-term perspectives [2, 3]. Unlike other kinds of coal in Siberia, the ash-and-slag wastes (ASW) of the coal of Yakutia have not been investigated yet, as well as the directions of their use.

The modern approach to the problem of ASW provides the complex utilization, which includes in particular isolation of the microspherical components from the ash (due to their properties, these components are able to replace expensive synthetic microspheres) and the use of the major part of wastes (the silicate constituent) according to traditional or novel technologies in construction industry [4]. In the case of coal from the Kangalas field, attention is attracted to the discovered increased concentrations of rare elements (Sc, Sr, Ba, Nb, Zr, Y) and rare earths (RE and REE, respectively) [5, 6] and the possibility to concentrate these elements from volatile ash or its fine fractions, which may occur during coal combustion at thermal power plants [7, 8]. For instance, the data on concentrating such elements as Cd, Pb, Sb, Te, Ga, As, Zn, Ge, Sn, Bi in ash fractions with particle size less than 5 μm, due to the formation of volatile chemical compounds at burning temperature, followed by their condensation under cooling on the surface of ash particles, were reported in [7, 9, 10]. Redistribution of elements also occurs between the components of volatile ash with specific physical or chemical properties, for example such as magnetic microspheres. According to the data for 13 thermal power plants [11], in addition to iron, also Co, Ni, Sr are concentrated in them at a level exceeding that for the initial ash slag by a factor of 1.2–10, up to 28, and 1.1–8.5 times, respectively. At the same time, the concentration of REE in them in comparison with flue ash usually decreases: La and Ce by a factor of 1.3–4.7, Sm 1.3–6.2.

Extraction of the most valuable elements may increase the profitableness of ASW processing; however, as the world experience demonstrates, the high level of utilization of ash-and-slag from thermal power plants is achieved when these wastes are used in construction industry [12]. Broad variations of the composition of ash from different kinds of coal and different energy sources led to the elaboration of standards governing the quality of ash for application in this area. According to the ASTM C618 standard (the use of mineral mixtures for obtaining concrete) developed by the American Society of Tests and Materials, all the kinds of power plant ash were divided into two classes: class F (ash from the combustion of bituminous coal and anthracite) with CaO <8 %, $\Sigma(SiO_2 + Al_2O_3 + Fe_2O_3) > 70$ %, calcination loss (c. l.) <12 %, and class C (ash from the combustion of lignites and sub-bituminous coal) with CaO content 8–20 % (CI type) and >20 % (CH type), $\Sigma(SiO_2 + Al_2O_3 + Fe_2O_3) > 50$ %, c. l. <6 % [13]. The ash of class F is most widely used because the ash of class C has an unpredictable effect on the characteristics of final products due to the differences in the properties of Ca-containing phases, the composition of which is strongly dependent on the temperature conditions of their formation.

A number of All-Union State Standards (GOST) on the use of flue ash-and-slag from thermal power plants for obtaining concrete of different kinds and for preparing construction mortars exist in Russia (GOST 25818–91, 25592–91, 25485–89, 26644–85, 25820–83, 6133–99, 28013–98). According to these standards, acid ash, ash-slag mixtures, broken slag and sand with total $CaO_{tot} \leq 10$ % can be widely used as fillers or thermally stable additives, while basic ash ($CaO_{tot} \geq 30$ %) should be used as a binding agent. The use of ASW with CaO_{tot} equal to 10–30 % (the ASW of Kangalas coal belongs to this group, too) requires additional investigation of composition and tests in final products.

The goal of the present work was to study the composition of ASW from the combustion of coal from the Kangalas field, including RE and REE, to estimate the reasonableness of processing the wastes for the purpose of extracting these elements and microspheric components, and to evaluate the possibility of their use in the production of construction materials.

EXPERIMENTAL

The ash-and-slag waste material in the amount of about 100 kg was sampled from the ash tailing of the boiler house of Orbita station performing fuel-bed combustion of the brown coal of B3 grade from the Kangalas field in KVS-0.43 type boilers. According to GOST 23148–98, a representative sample (~10 kg) was collected from the ash and slag lot to study the characteristics and to carry out separation by means of the granulometric classification, magnetic separation and hydrodynamic separation. Size classification was performed with a vibration set-up equipped with a set of sieves with mesh size of 4.0, 1.0, 0.4, 0.2, 0.16, 0.1, 0.063, and 0.05 mm. Magnetic separation was carried out in the atmosphere of the air with an electromagnetic separator of 138T grade (Technical Specifications TU 24-8-1054–77) and in water medium using a hand-driven constant magnet. Hydrodynamic separation was carried out in a column with pulsating water upflow [14].

The granulometric composition of the material was determined by means of dry sieving according to GOST 18318–94, chemical composition and c. l. according to GOST 5382–91, packed density was measured by means of volumetric vessels, RE and REE content was determined by means of mass spectrometry with inductively coupled plasma (ELAN-9000 mass spectrometer of Perkin-Elmer Co., USA). Analysis conditions: ion source – argon inductively coupled plasma, resolution 0.7 a.m.u., power of the RF generator 1150 W, frequency 40 MHz, gas flow rate, l/min: cooling gas 15, auxiliary gas 1.2, spraying gas 0.94; Scott spraying chamber, solution flow rate 1 ml/min, weighed portion of the sample 0.2 g. After the acid treatment of the sample, the volume of the solution was brought to 200 ml (5 % HCl).

In order to obtain construction materials, the following materials were used: non-magnetic fraction of the ash-and-slag material >0.2 mm and aleurolitic clay ground to the specific surface equal to 2000 cm^2/g; sand with fineness module M_f = 3.82 with the packed density of 1550 kg/m^3 and clay particle content 7.5 %; portland cement of 400 grade. The properties of nonfired bricks were investigated with cylindrical samples (20 mm in diameter, 20–25 mm high) obtained by means of semi-dry pressing. Brick samples were kept for 7 days above water and up to 28 days in the air. The mortar samples were kept in molds without a bottom on the surface of clay brick for 1 day, under humid conditions up to 3 days, in the air up to 28 days. The properties of solutions were estimated with cubic samples sized 70 × 70 × 70 mm. Density, water absorption capacity, compression strength were determined according to the procedures described in [15].

RESULTS AND DISCUSSION

Investigation of the composition of ash-and-slag wastes of coal from the Kangalas field

According to the data on granulometric composition (Fig. 1), ASW is a material with the broad particle size range in which the fraction <0.2 mm mainly depicts the contribution from the ash component (yield: 56.4 %), while the fraction >0.2 mm depicts that from the slag component (yield: 43.5 %). For efficient magnetic and hydrodynamic separation, the ash

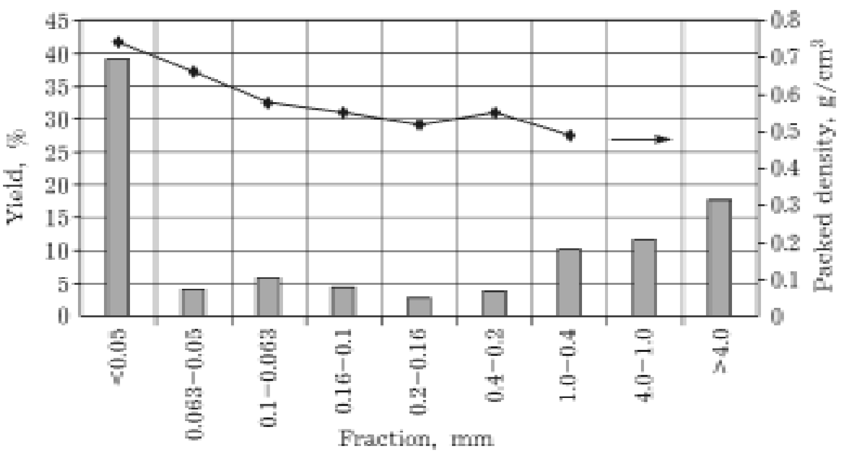

Fig. 1. Granulometric composition and apparent density of the narrow-sized fractions of ASW of the coal from the Kangalas field.

TABLE 1

Products of separation of the ash and slag material from the ash tailings of the boiler house of Orbita station (Yakutsk city)

Fraction, products	Yield, %	Packed density, g/cm^3	c. l., %	Content, g/t RE	
				Ba	Sr
Initial ash-and-slag from tailings	100	–	6.2	1949	1482
Fraction >0.2 mm:	43.5	–	7.6		
magnetic product (>0.2 mag.)	0.2	–	3.7	934	755
silicate product (>0.2 silic.)	43.3	–	7.6	1532	1399
Fraction 0.2–0.05 mm:	17.3	0.57	7.1		
Magnetic concentrate:	7.9	0.58	9.4		
magnetic product (0.2–0.05 mag.)	0.03	0.87	5.3	919	668
light product (0.2–0.05 light)	5.5	0.57	13.0	3427	2734
heavy product (0.2–0.05 heavy)	2.0	0.63	7.3	2827	2717
water-soluble product (0.2–0.05 sol.)	0.08	–	–		
Silicate product (0.2–0.05 silic.)	9.4	0.57	5.1	1669	1600
Fraction <0.05 mm:	39.1	0.65	6.6		
light product (<0.05 light)	36.0	0.77	8.1	1991	1852
heavy product (<0.05 heavy)	2.5	0.88	5.3	1363	1041
water-soluble product (<0.05 sol.)	0.6	–	–		

Note. All the fractions were obtained at the intermediate stages of he separation of narrow-sized products.

and slag material was sieved into three fractions, mm: >0.2 (43.5 %), 0.2–0.05 (17.3 %) and <0.05 (39.1 %) (Table 1). The fraction >0.2 mm was hand-separated into the magnetic (yield: 0.2 %) and silicate (43.3 %) products. From the fraction 0.2–0.05 mm, the silicate product and magnetic concentrate (yield: 7.9 %) were isolated; the latter was separated by means of the hydrodynamic procedure into the light and heavy products. The magnetic product was separated from the latter with the yield of 0.03 % (see Table 1). It was determined with the help of an optical microscope that this material is almost free from microspheres. The material of fraction <0.05 mm was separated using the hydrodynamic procedure on the basis of density into light and heavy products. We failed do isolate magnetic particles from the heavy product of this fraction.

So, the isolation of the magnetic fraction from ASW is unreasonable because its yield is less than 0.2 % and it is almost free from microspheric particles. The low yield and the absence of magnetic microspheres are likely to be due to the low iron content of the mineral part of coal (4.7 %) and the conditions of fuel-bed burning, including coarse grinding and low temperature which is insufficient for melting iron silicate eutectics.

Hydrodynamic separation applied firstly to concentrate heavy magnetic particles is less efficient for non-magnetic ash particles due to their close density. Nevertheless, underburnt particles are observed to get concentrated in the light products of hydroseparation for which c. l. increases by a factor of 1.8 and 1.5 for the fractions 0.2–0.05 and <0.05 mm, respectively (see Table 1). The following regularity is observed in the character of changes in c. l. value (Fig. 2, *a*): for the general trend of its decrease (from 7.6 to 6.6 %) with a decrease in fraction size (from >0.2 to 0.05 mm), lower c. l. values are characteristic of magnetic products (3.7 and 5.3 %) and higher for the light products of hydroseparation (13 and 8.1 %).

Ash and slag material prepared for hydrodynamic separation was preliminarily kept for more than 12 h in distilled water to remove water-soluble compounds. As the dispersity of the material increases from the size of 0.2–0.05 to <0.05 mm, their yield increases from 0.08 to 0.6 %. Solubility at room temperature was about burning the brown coal of B3 grade from the Kangalas field of the Lena basin

Sc	Cr	Ni	Co	Mn	Pb	Th	U	Nb	Zr	ΣREE
12	53	47	18	534	18	26	7	13	101	155
6	140	89	30	4943	72	12	4	9	47	79
8	50	38	13	379	12	26	7	10	127	136
7	70	64	23	6593	110	21	4	8	36	90
21	85	78	27	851	27	40	10	21	112	251
20	79	78	27	1108	17	35	9	20	137	230
11	58	59	15	436	46	23	6	13	73	147
13	68	60	18	584	34	36	8	15	82	219
8	54	56	11	523	35	27	6	10	74	153

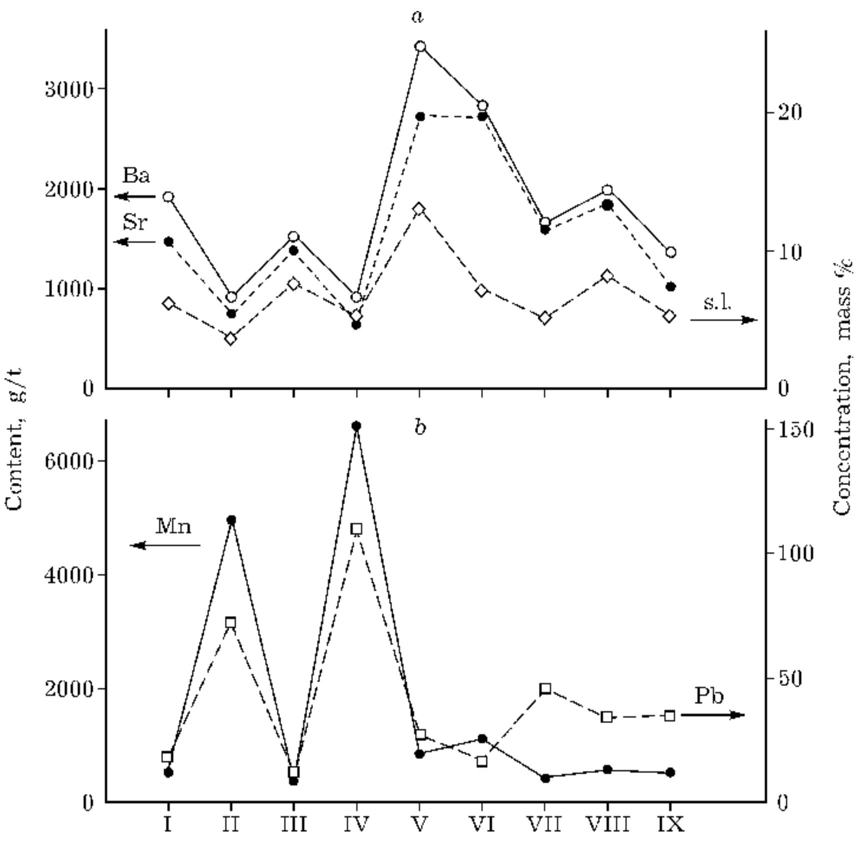

Fig. 2. Content of the products of separation of the ash and slag from the combustion of coal from the Kangalas field in different fractions. Here and in Fig. 4: Fractions, mm: I – initial ash-and-slag, II >0.2 (mag.), III >0.2 (silic.), IV 0.2–0.05 (mag.), V 0.2–0.05 (light), VI 0.2–0.05 (heavy), VII 0.2–0.05 (silic.), VIII <0.05 (light), IX –0.05 (heavy).

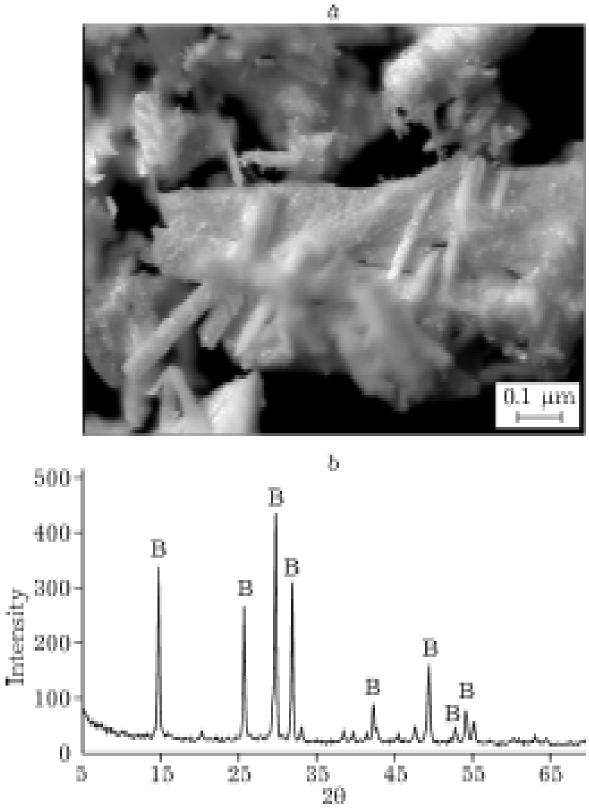

Fig. 3. Optical microscopic image (*a*) and diffraction patterns (*b*) of the dry residue of the water-soluble fraction of ash and slag of coal from the Kangalas field. B – bassanite $CaSO_4 \cdot 0.5H_2O$; *d* values: 5.97, 3.45, 3.00, 2.80, 2.14, 1.84, 1.73, 1.69.

3 g/l. After evaporation of the solutions and drying of the residuals at 100 °C, bassanite $CaSO_4 \cdot 0.5H_2O$ is crystallized (identification was carried out according to [16] (Fig. 3).

Investigation of microelements in the coal from the Kangalas field [1, 5] revealed increased concentrations of Sr, Ba, Nb, Sc, Y, and REE in the ash of coal samples taken from the marginal contact parts of a coal-bed. For the middle part of coal-bed, concentrations exceeding the clarke level were observed for Ba and Sr (by a factor of 3.4 and 9.7, respectively), while the levels comparable with the clarke ones were observed for Cr, Sc, Y, Yb, V, Co, Nb, and REE.

Analysis of the ASW from the combustion of the Kangalas coal showed that RE content in these products is close to that in the ash of coal samples taken from the middle part of a coal-bed. An exception is Ba and Sr, the content of which exceeds the clarke level by a factor of 4 and 5.5, respectively. The content of these elements in magnetic products decreases, while in the products of hydrodynamic separation it somewhat increases, especially for the fraction 0.2–0.05 mm (see Table 1, Fig. 2, *b*), and equals 3.4 and 2.7 kg/t for Ba and Sr, respectively. Magnetic fractions, with respect to the initial ash-and-slag, contain 9 and 12 times more manganese (up to 6.6 kg/t), lead 4–6 times (up to 0.1 kg/t) (see Table 1, Fig. 2, *a*). The concentrations of Cr, Ni, Co increase noticeably only in the magnetic product of the fraction >0.2 mm (by a factor of 2.6, 1.9, and 1.8, respectively).

The REE content of the initial ash and slag is close to that in the ash of coal samples taken from the middle part of coal-bed (ΣREE = 155 g/t). Similarly to barium and strontium, the concentrations of all the REE decrease in the magnetic products (Fig. 4); increased concentrations are observed in the products of hydrodynamic separation in the fraction 0.2–0.05 mm (ΣREE = 230–251 g/t) as well as in the light product in the fraction <0.05 mm (ΣREE = 219 g/t). In this case the concentrations of these elements are observed to correlate with c. l. values.

In spite of the limited amount of mined strontium and REE in Russia, their obtaining from the investigated ASW can hardly be considered as economically reasonable and competitive due to low concentrations in comparison with ore deposits, in particular Mazuevskoye deposit of strontium in the Perm Region (70 kg/t SrO) and Tomtorskoye deposit (niobium–rare earths) in Yakutia (about 80 kg/t Nb_2O_5 and 130–200 kg/t of the sum of REE oxides) [17].

Use of ash-and-slag wastes in the production of construction materials

In view of the low concentrations of magnetic microspheres, RE and REE, the main potential area of the application of ASW from the combustion of brown coal of the Kangalas deposit is construction industry. In the case of fuel-bed combustion, the ASW of the coal of the same grade may substantially differ in granulometric and phase composition, density, c. l. value. The chemical composition of ASW of the Kangalas coal sampled from the tailings (Fig. 5) is in good agreement with the known

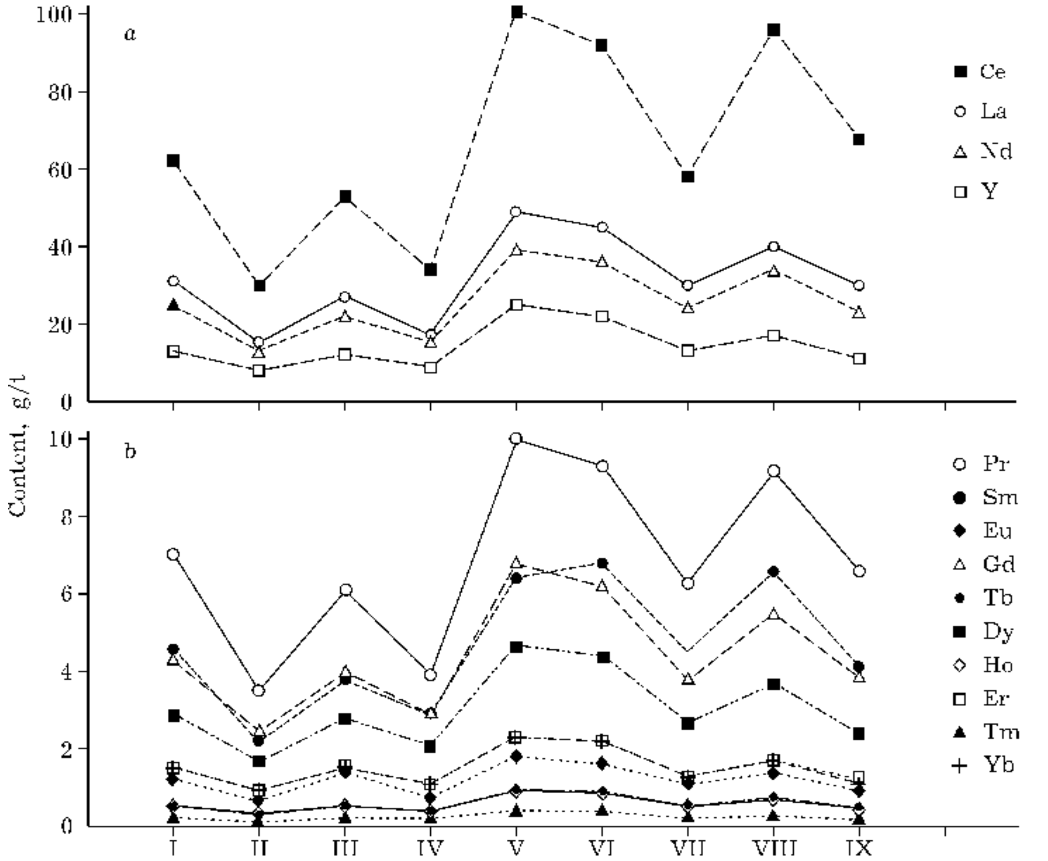

Fig. 4. REE content in the products obtained by separation of ASW of coal from the Kangalas field. For designations, see Fig. 2.

data on the ash residue of coal [2], except for the fact that ASW contains underburnt particles (c. l. 6–8 %). According to the classification on the basis of binding properties [18], the ash of the Kangalas coal belongs to latently active

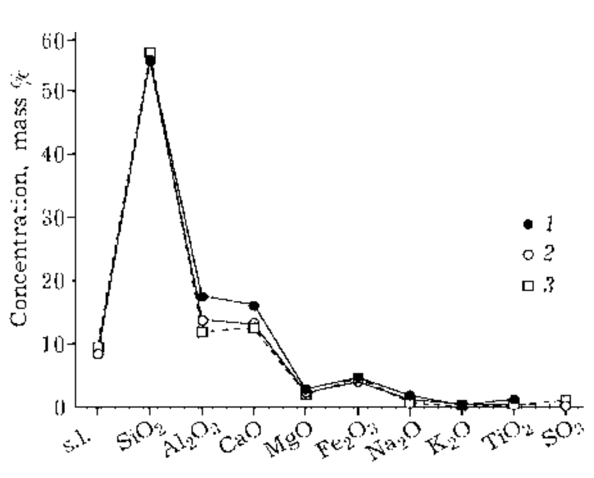

Fig. 5. Chemical composition of ash of brown coal from the Kangalas field of the Lena basin: *1* – ash of the initial coal, calculated for the sulphur-free mass according to the data of [2]; *2, 3* – ASW of coal (*2* – fraction >0.2 mm, *3* – <0.05 mm).

materials with the basicity module $M_b = (CaO + MgO + K_2O + Na_2O)/(SiO_2 + Al_2O_3)$ equal to 0.29; silicate module $M_s = SiO_2/(Al_2O_3 + Fe_2O_3)$ equal to 2.46, and the quality coefficient $K = (CaO + Al_2O_3 + MgO)/(SiO_2 + TiO_2)$ equal to 0.65. The use of this ash as a binder requires intensified hardening.

One of the promising directions is the technology of nonfired bricks by means of hyperpressing [19]; this method has some advantages over pressing at low specific pressure. In particular, the use of the former allows one to broaden the raw material basis of the initial substances and to decrease the consumption of binders in order to obtain the required grade strength of bricks.

In order to obtain non-fired bricks, the composition cement–ash–clay was used (Table 2). For the fixed consumption of cement (19 %), the effect on clay content was examined; on the basis of a set of indices, the ratio of ash to clay 1 : 1 was chosen. The concentration of ASW in the composition is 40.4 mass %. Press-

TABLE 2

Characteristics of construction products manufactured using the ASW of the Kangalas coal

Characteristic	Compositions	
	Cement : Clay : Ash 355 : 747 : 747 kg/m^3 (19.2 : 40.4 : 40.4 %)	Cement : Sand : Ash 370 : 780 : 780 kg/m^3 (19.2 : 40.4 : 40.4 %)
	Non-fired bricks	
Compression strength (MPa) at the age of, days:		
1	6.37	4.46
3	8.28	11.31
7	10.67	14.33
28	16.41	23.57
Mean density, kg/m^3	1850	1930
Water absorption, mass %	14	10.2
Grade	150	200
	Cement : Sand 310 : 1770 kg/m^3 (14.9 : 85.1 %)	Cement : Ash : Sand 250 : 25 : 1560 kg/m^3 (13.6 : 1.4 : 85.0 %)
	Construction masonry mortars	
Compression strength at the age of 28 days, MPa	9.12	7.81
Mean density, kg/m^3	2080	1840
Grade	75	75
Cement saving, kg/m^3	–	60 (19.4 %)

ing pressure was varied from 20 to 100 MPa; 40 MPa was chosen as an optimal value. Under these conditions, brick samples with the density of 1850 kg/m^3, water absorption 14.0 % and softening coefficient equal to 0.58 were obtained. According to GOST 379–95, the strength of such a brick corresponds to grade 150. The compositions with sand were investigated in which the clay component was replaced with sand at the ash to sand ratio of 1 : 1 (see Table 2). These bricks correspond to grade 200 in hardness; their density is 1930 kg/m^3. Long-term storage of brick samples for 70 days showed that an increase in strength is observed for the composition with sand within 16 to 20 %.

It is known that sand is often replaced in part with ash from thermal power plants in order to save cement in preparing construction mortars. The compositions of construction mortars based on the ASW composition with cement and sand were chosen (see Table 2). For cement consumption of 310 kg/m^3, masonry mortar with the strength corresponding to grade 75 was obtained (GOST 28013–98). The introduction of ash and slag material into the construction masonry mortar causes a decrease in the density of the mortar without changes in the grade strength and allows one to decrease the thermal conductivity of brickwork and decrease cement consumption by 10 %.

In addition, the ASW of this type can be used in the production of asphalt concrete (GOST 9128–97), dense concrete in the form of finely ground mixtures after the joint grinding of lime and fuel ash (GOST 25214–82), heat-insulating materials [20].

CONCLUSIONS

The granulometric composition of ash-and-slag wastes from the fuel-bed firing of brown coal from the Kangalas field was investigated. Its separation into the products differing in size, density and magnetic properties was carried out. The chemical composition of the initial material and the products of its separation was established, including RE and REE.

It was found that manganese and lead are concentrated in the magnetic products in the amount exceeding that fort in initial ash and slag by a factor of 9–12 and 4–6, respectively. In view of the low content of the magnetic fraction (<0.2 mass %), it was concluded that its separation from ASW as a target product is unreasonable.

It was shown that the concentrations of the major part of RE and REE investigated increase in the products of hydrodynamic separation and reach up to 3.4 kg/t for barium, 2.7 kg/t for strontium, 250 g/t for the sum of REE. In view of the low content of these elements in comparison with ore deposits, it is concluded that their separation from the investigated ASW is unreasonable.

The main area of utilization of ASW from the combustion of coal from the Kangalas field is construction industry. Compositions with the mass content of ASW about 40 % were chosen for the production of non-fired bricks of 150–200 grade with the mean density 1850 to 1930 kg/m^3, compositions of ASW with cement and sand for obtaining standard masonry mortar of 75 grade with a decrease in cement consumption by 19.4 mass % were also chosen.

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