PREPARATION OF COAL BRIQUETTES BASED ON NON-STANDARD KAZAKHSTAN COAL WITH VARIOUS ADDITIVES AND DETERMINATION OF THEIR QUALITY

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ABSTRACT

Briquetting of coal fines is one of the best solutions for the disposal of substandard coal and coal fines. The coals of the Central region of Kazakhstan were selected as the object of this study. When stored for a long time, the coals of this region dry out and turn into coal fines that are not suitable for consumption. It is established that mechanical properties of composite briquettes depend on the charge composition and some technological parameters of briquetting, including: the material composition of the briquetted mixture, moisture mixture, compacting pressure, mode of heat treatment of the briquettes, the type and flow rate of the binder component. To optimize the combustion of the coal mass, the pyrotechnic component or polymer additives were added to mixture with the initial coal. The pyrotechnic component produces an incendiary layer, which contains as a combustible component (coal sludge, cardboard), a magnesium igniter and oxidizing agents in the form of ammonium nitrate and barium chromate. The greatest influence on the duration of burning of brown coal is exerted by oxygen-containing compounds (90 %). It was found in the laboratory research to obtain high-quality fuel briquettes, the following characteristics are necessary: coal size 0 - 2.5 mm; coal humidity 10 - 11 %; pressing pressure 150.0 MPa; processing temperature 230° C; heat treatment time of 180 min. The introduction of incendiary composition in the briquette does not lead to a significant reduction in the size and destruction of aggregates, while a significant part of the pyrotechnic composition is located on the surface of the coal in the form of particles with sizes from 145.8 to 368.6 nm, mainly in the form of "coagulants".

Keywords: *coal, briquetting, binder, calorific value, incendiary composition.*

INTRODUCTION

Coal reserves in the world prevail over all other types of organic raw materials. Coal reserves in Kazakhstan are currently estimated at about 33 billion tons, which is about 4 % of the world reserves. Of these, more than 60 % are brown coal deposits, and just under 40 % are coal reserves [1 - 4]. In coal mining, 2/3 of coal can be lost as coal fines. A significant amount of coal fines is also lost during transportation. Coal fines are accumulated in large quantities at coal mining sites, at transshipment points and in most cases occupy useful areas, creating a threat of pollution of the surrounding area [5 - 7]. Kazakhstan is experiencing a shortage of cheap high-grade coal, which could be used for municipal needs [8 - 11]. Preparation of briquettes is one of the most appropriate solutions for the disposal of substandard coal and coal fines. Prospects and feasibility of fuel briquettes production are as following: briquettes in the production can be made more permeable than a monolithic piece of coal, in this case, the briquette burns completely, consequence 30 % fuel economy; heat treatment of briquettes can reduce the amount of moisture, while minimizing the loss of vaporization, consequence 5 % fuel economy; organic and mineral additives in the briquette can vary the properties of briquettes, improve their environmental friendliness and improve energy performance [12 - 15]. The technology of coal briquetting without binder additives seems more attractive at first glance, but at the same time electricity costs increase several times, the productivity and quality of the briquette decreases. To obtain durable briquettes, various cementing substances of organic and inorganic nature are added to the coal fines: for example, resin, dextrin, lime, asphalt, molasses, soluble glass, soda, clay, alum, cement [15 - 18]. One of the disadvantages of coal as a fuel is a significant smoke formation, when directly burning coal produces a large number of harmful emissions that load the environment: sulfur dioxide (SO_2), nitrogen oxides (NO), which form acids in contact with atmospheric moisture; carbon dioxide (CO_2), which accounts for 50 % of emissions into the atmosphere, creating a greenhouse effect [19 - 21]. With the aim of reducing harmful emissions, to eliminate the formation of smoke during combustion, for the manufacture of coal briquettes are introduced various additives, which are widely used camphor, naphthalene or nitrobenzene. The function of these binders is a molecular association, when the molecules of the binder decay, unsaturated compounds are formed, which easily react with the association of the aromatic component of coal [22 - 24]. As a result of recombination, heavier molecules are formed that are not capable of volatilization, and with further heating they break up again and as a result, smoke formation is significantly reduced [25 - 27].

The aim of this study was to obtain and characterize briquettes based on substandard raw materials of the Central region of Kazakhstan with different additives.

EXPERIMENTAL

In this work, for preparation of coal briquettes the coal fines from the Kazakhstan fields were applied [1, 2, 28]. The briquettes have been obtained by the well-known in literature technique [29 - 31], consisting of coal grinding, drying of coal to the desirable humidity, mixing coal with binders, heating the coal charge, pressing and cooling. Water was added before briquetting for necessary mixture plasticity and for dilution of the binder component. The strength of the briquettes has been defined after cooling the briquettes to ambient temperature by crushing on a press with fixing the maximum force that the prepared briquette can withstand [32, 33]. Mixtures of solid polymer residues were preheated, then after mechanical treatment, the fraction was separated to a particle size > 200 microns in a mixture with coal. Samples of coal with incendiary compounds were examined using SEM Ntegra Therma at an accelerating voltage of 10 - 30 kV.

Thermogravimetric studies were performed under the following experimental conditions: mass of a sample 0.3 - 0.03 g, analytical grinding, ceramic crucible with a lid 15 mm high and 5 mm diameter. The weight loss of the sample at a given temperature was determined according to the thermogravimetric curve. The rate of mass loss depends on the curve of differential thermogravimetry, from temperature to endo - or exo - effects - on the curve of differential thermogravimetry adsorption, in accordance with the standard procedure. The recorded instrument mass loss curves (TG curves) and mass loss rates (DTG curves) are recalculated per 1 g of the initial sample and expressed as temperature dependences, the nature of which is illustrated by the given thermograms. Based on the DTG curves, the temperatures (T_{max}) corresponding to the maximum mass loss rates were determined. In this paper, the mass loss is identical to the yield of volatile products, and the rate of mass loss is equivalent to the rate of release of volatile products when heated [32 - 34]. Thermogravimetric analysis was performed for samples of raw materials - solid polymer products and coal.

RESULTS AND DISCUSSION

Data of X-ray phase analysis of briquettes based on coal Oy-Karagay deposit after calcination (Fig. 1), show that the main phase is an amorphous phase of carbon with a mineral component in the form of iron oxide and SiO_2; also there are calcite Fe_2SiO_4 and feldspar in the

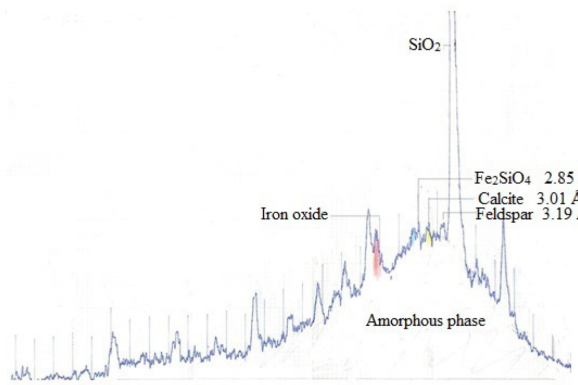

Fig. 1. XRD analysis of coal briquettes prepared on the base of brown coal from the Oy-Karagay deposit (Kazakhstan) after calcination.

composition [13, 35, 36]. Samples of coal briquette are almost X-ray amorphous, and weak peaks of SiO_2 and Co_3O_4 are also found. Peaks of 2.70 and 2.50 Å were detected, indicating the presence of Fe_2O_3 hematite (ASTM 33-664). There are also peaks at 3.87; 3.07; 2.34, 2.27 Å that show the presence of K_2O_2 (ASTM 32-827). Peaks: 4.26; 3.34; 2.45; 2.28; 2.23; 2.12; 1.81; 1.54 Å indicate SiO_2 quartz (ASTM 5-490). Peaks: 3.80; 3.01; 2.27; 1.191; 1.87 Å indicate calcite $CaCO_3$ (ASTM 5-586), [13, 37, 38].

During the heat treatment of coal, the mineral mass is heated to high temperatures, as a result of which the substances of the mineral mass undergo transformations, including decomposition and the formation of new substances at various interactions. In thermodynamic calculations, it is important to take into account not only the total content of mineral impurities in the coal, but also their chemical composition [39, 40].

The following indicators on the content of components were determined for brown coal of the Oy-Karagay deposit: sulfur 0.6 - 1.1 %; carbon 75 - 83 %; oxygen 15 - 37 %; hydrogen 3.0 - 6.5 %; nitrogen 0.7 - 1.2 %; humic acid up to 80 %. In the case of brown coal, it is important to take into account the distribution of heteroatoms in different functional groups, since the initial brown coals may have a high content of such components. The formation of a fragment of the organic mass of brown coal can be identified by the content of organosulphuric compounds with nitrogen and oxygen-containing substances [1, 13, 41 - 43].

In Figs. 2 and 3 the thermogravimetric decomposition curves of initial coal and coal with multipolymeric binding are presented. By means of the thermogravi-

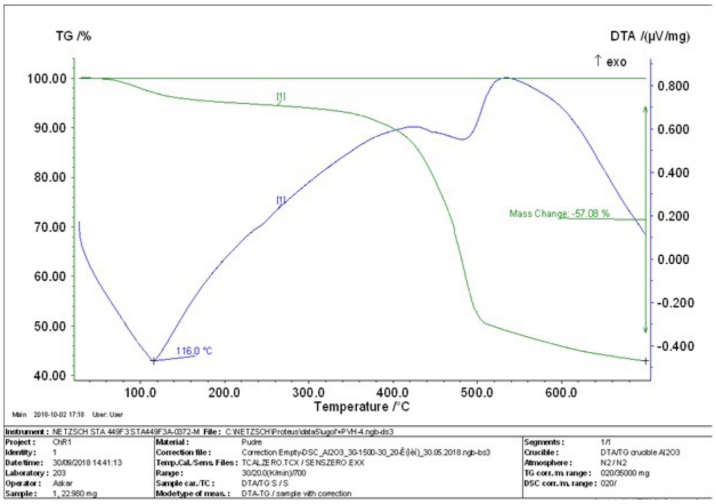

Fig. 2. Thermogravimetric curve of decomposition of initial coal.

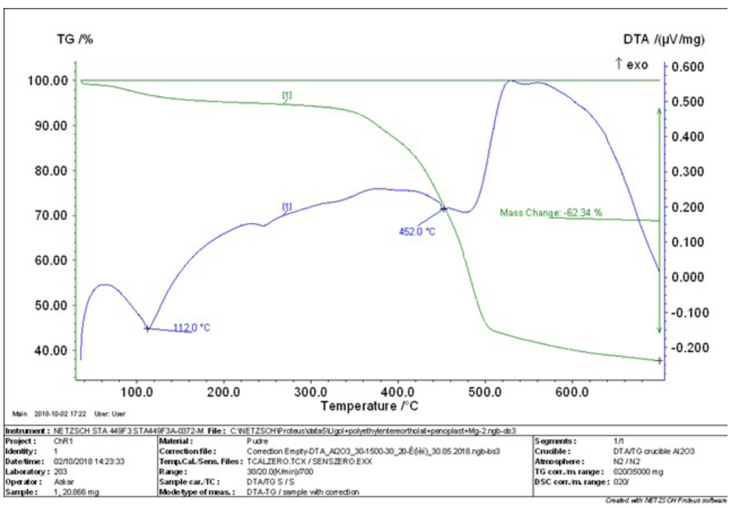

Fig. 3. Thermogravimetric curve of the decomposition of coal with a polymer binder.

metric analysis such parameters as temperature of the termination of burning of TG which is 700ºC and the maximum temperature of T_{max} 550ºC were found. The shape of the TGA curves primarily depends on such kinetic parameters as the reaction order, pre-exponential factor, and activation energy. These parameters are of paramount importance for elucidating the mechanism of thermal destruction of coal [31, 32]. It is known that the maximum of the differential curve of mass loss corresponds to the maximum speed of any process that occurs when exposed to temperature, and is equal to 50 % mass loss obtained using TGA [33]. For the binding component (Fig. 3) a differential curve of endothermic reactions is deflected down, while a differential curve of exothermic reactions is deflected up. The deviation value (peak of temperature) indicates a degree of temperature difference between the starting coal and the polymer binder, which is an indicator of the amount of the converted substance and the intensity of the reaction.

The coals of the Oy-Karagay deposit have an average technical strength. Commercial coal consists on average of 56 - 64 % of coal of the size class +13 mm and 30 % of the class less than 6 mm. The coals of this deposit dry up during long storage and turn into coal trifle, unsuitable for consumption. For this reason, it is necessary to briquetting coal fines [1, 11 - 13, 34].

The energy indicators of the Oy-Karagay coal deposit were established (Table 1): calorific value 6,700 kcal kg^{-1}; heat of combustion 3,600 kcal h^{-1}; yield of volatile products 27 - 45 %; humidity 7.8 %. Important factors that positively affect the quality characteristics of coal briquette are low ash content and high heat of combustion.

It is known that a significant role in the briquetting process is played by the preparation of coal charge. The sieve composition of coal and the distribution of grains of various sizes in the mixture must correspond to its

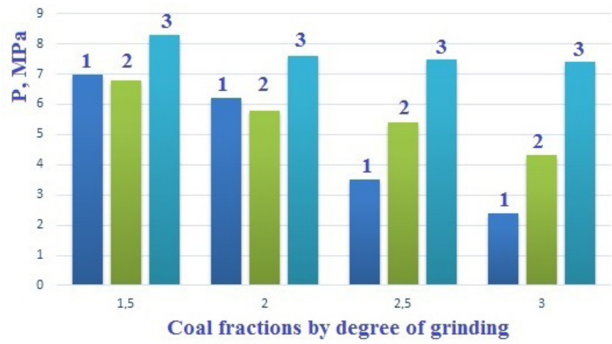

Fig. 4.Influence of degree of a grinding of coal on durability of briquettes, MPa: 1- coal tar, 2 -coal + technical carbon, 3 - coal + PET.

maximum compressibility, which provides the greatest strength of contacts between grains and high strength of briquettes with a minimum consumption of binder for briquetting [36 - 38]. Studies on the influence of the particle size distribution of coal on the mechanical properties of briquettes (Fig. 4) showed that an increase in compressive strength is especially noticeable in the briquette samples containing coal with the smallest size (coal class 1.5 mm). This is due to the fact that when briquetting fine coal, the number of interacting surface-active contact groups with the binder is as high as possible. The role of the so-called "active centers" on the solid surface increases, the adsorption interaction at the interface between the solid and liquid phases intensifies, the diffusion of binder maltenes into the pores and cracks of coal proceeds more efficiently, which inevitably leads to an increase in strength.

It was found that the strength of the compositions consisting of coal grains with a grain size of 3 mm is about 3 times lower than the strength of briquettes obtained from coal with a grain size of 1.5 at the same parameters of briquetting. This is due to the fact that

Table 1. Technological indicators of coal of the Oy-Karagay field.

Indicator, %			Content, %					Q_s^a, kcal / hour	q, kcal / kg
W^a	V^a	A^a	Sulfur (total)	Nitrogen	Carbon	Oxygen	Hydrogen		
7.8	27-45	12.5	0.6-1.1	0.7-1.2	75- 83	15-37	3.0-6.5	3,600	6,700

Note: W^a, V^a, A^a are moisture, volatile matter and ash content in an analytical state; is a high calorific value; q is caloric content.

when pressing the destruction of large coal grains and the formation of additional surfaces that are not wetted with a binder occurs.

Despite the fact that samples made only of coal dust (grinding degree 1, Fig. 4) and mixtures (grinding degree 6, Fig. 4) give higher compressive strengths, their use for briquetting is not rational in connection with the introduction of labor-intensive grinding and sieving operations into the technological cycle. Therefore, for further studies, coal with an optimal particle size of less than 2.5 mm was used.

The cohesion intensity of the briquetted coal particles increases significantly with increasing pressing pressure. Therefore, the effect of pressing pressure on the mechanical properties of briquettes on compositions containing tar from the Pavlodar refinery (Kazakhstan) as a binder was further studied (Fig. 5). Pressing was carried out with varying pressure from 50 to 200 MPa, the moisture content of the coal was optimal. It was found that the strength of the briquettes increases with increasing compression pressure to 150 MPa, then begins to decline. The optimum pressing pressure is set to 150 MPa.

Analysis of the results shows that the requirements of the State Standard 7299-84 on brown coal briquettes in terms of mechanical properties meet the following compositions made at a pressing pressure of 150 MPa and processed at a temperature of 230ºC for 180 min: 1) coal 90 wt. % + bitumen 10 wt. % (Pavlodar refinery), 2) coal 75 wt. % + tar 15 wt. % + sapropel 10 wt. %.

The following basic characteristics were determined for the developed briquette compositions: compressive strength, ash, volatiles yield, total sulfur content, total hydrogen content, water absorption, mass fraction of moisture, higher and lower calorific value. The main technical characteristics of brown coal briquettes are given in Table 2. The results of the research demonstrate that the compressive strength of briquettes made using modified tar and bitumen is higher than the normalized index by 15 - 35 %. Ash content ranges from 15 - 18 %, despite the fact that it is slightly higher in briquettes with modified tar, yet 27 % less than the normalized index. The sulfur content of the briquettes to below 7 to 15 times, water absorption is below 40 - 60 %.

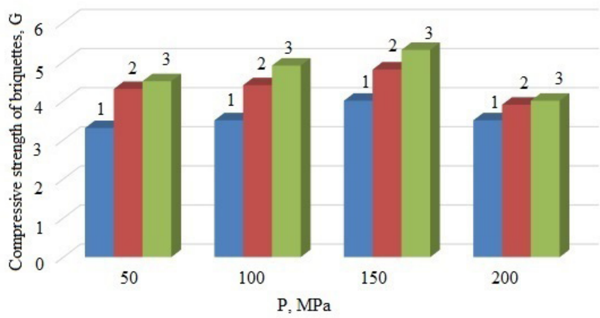

Fig. 5. Dependence of the strength of briquettes on the pressing pressure: 1 - coal + tar, 2 - coal + technical carbon, 3 - coal + PET.

To accelerate the combustion of the briquette, nitrates, manganese dioxide, copper oxide, vanadium oxide can be added in an amount of 0.03 - 0.06 %, and to slow down mineral fillers (chalk, kaolin, bentonite), which reduces production costs and significantly reduces

Table 2. Technical characteristics of brown coal briquettes.

No	Composition	$\sigma_{compression}$, MPa	A^d %	V_{daf} %	W %	Q^{daf}_s kcal / kg	Q^r_i kcal / kg
1	Coal + Tar	6.10	17	46	2.4	6,654	5,432
2	Coal + Technical Carbon	8.14	15	43	1.2	6,210	4,254
3	Coal + PET	10.4	14	40	2.4	6,862	5,020
4	Coal + Bentonite	12.5	19	37	2.5	6,952	5,352

Notes: $\sigma_{compression}$ is an ultimate strength in compression, MPa; A^d is dry ash content of fuel, %; V_{daf} is a yield of volatile substances, %; W is water absorption, %; Q_{daf} is the highest calorific value for the dry ashless state of fuel, kcal/kg; Qr_i is the lowest calorific value per fuel operating condition, kcal/kg.

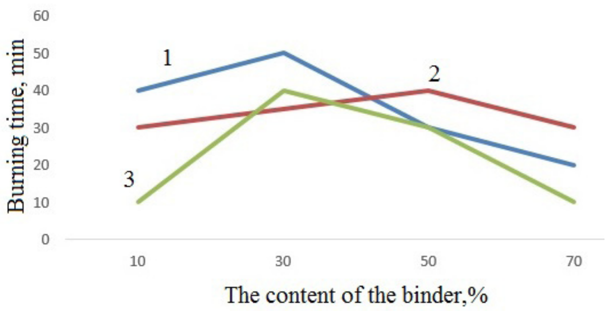

Fig. 6. Dependence of the burning time of a coal briquette from the content of the binder and pyrotechnic composition: 1- cardboard + coal, 2 -PET + coal, pyrotechnic composition + coal.

the burning rate fuel compared to lump coal [28 - 30, 39]. In this work, coal slurries (1 - 2 mm) of 30 - 60 %, with an ash content of 14.5 %, and humidity of up to 50 % were used as the test material. As a binder, paper waste in the form of cardboard 40 - 70 %, initial moisture content 5 % and epoxy resin 10 % were used. When choosing the temperature of briquette calcination, it is necessary, first of all, to keep in mind the physicochemical properties of the components of the briquette mixture and the results of exposure to the selected temperature. During the research on the production of briquettes from coal of the Oy-Karagay deposit, synthetic waste of polyethylene, polyethylene terephthalate, polypropylene were used as a binder.

Waste is a semi-solid viscous mass that retains its aggregate state at a temperature of 25 - 30°C, so when using them as a binder and obtaining a homogeneous charge composition, heating to a melting temperature of 120 - 130°C is required. The heating rate and final temperature are the main factors in the calcination process and determine the temperature field in the briquette. The temperature difference in the heating process by the volume of the briquette determines the uneven shrinkage, creates dangerous internal stresses and has a negative impact on the yield and quality of briquettes [7 - 10, 40, 41]. To optimize the combustion of the coal mass, the pyrotechnic component was added to mixture with the initial coal, thus producing an incendiary layer, which contains as a combustible component (coal sludge, cardboard), a magnesium igniter and oxidizing agents in the form of ammonium nitrate and barium chromate. Fig. 6 presents data on the duration of the burning of the briquette from the content of the incendiary composition. The greatest influence on the duration of burning of brown coal is exerted by oxygen-containing compounds (90 %). These data coincide with the literature data, because oxygen-containing groups really have a great influence on thermal stability, sorption activity, wettability, sintering, coking ability and other properties of coal [12, 42 - 45].

At the optimum amount of binder 30 % cardboard with 70 % of the coal slime is produced more amount of a long burning briquette coal with a lower calorific value. At the same time, coke formation occurs as a process of condensation of aromatic compounds, as well as the thermal decomposition of the organic part of the binder cardboard, accompanied by the decay of coal sludge molecules and volatilization of the lightest molecules.

Coal burning in the presence of a PET binder passes through a maximum at 50 % content, the burning time reaches 40 minutes. From the literature it is known that in organic compounds of PET, the bond of carbon with foreign atoms is usually weaker than the bonds between carbon atoms, which means that the escaping molecules contain a relatively lower proportion of carbon. As a result of atomic rearrangements, the most durable carbon-containing molecular structures accumulate in the non-volatile PET residue, while the less durable ones disappear. The presence of aromatic compounds in the binder provides a stronger coke, and therefore more durable briquettes. These considerations play a crucial role in the selection of organic substances as a binder for briquetting coal with its subsequent calcination [1, 13, 42 - 45]. The use of coal slime and paper waste in the form of cardboard allows to get an easy-to-manufacture and composition of the fuel briquette and simultaneously dispose of coal and paper waste. Coal slurries provide the calorific value of the fuel briquette. Waste paper in the core layer ensure reliable combustion. The presence of a hole in the briquette reduces emissions of harmful substances into the atmosphere.

Data from the SEM study showed that briquettes prepared on the basis of coal fines and formed in the form of cylinders with a diameter of 40 mm with an inner hole diameter of 12 mm and a length of briquettes 180 - 220 mm (Fig.7a). The formation of coal briquettes with a binder occurs as a result of adhesion of coal particles with the binder (Fig.7c). The introduction of incendiary composition in the briquette does not lead to a significant reduction in the size and destruction of aggregates,

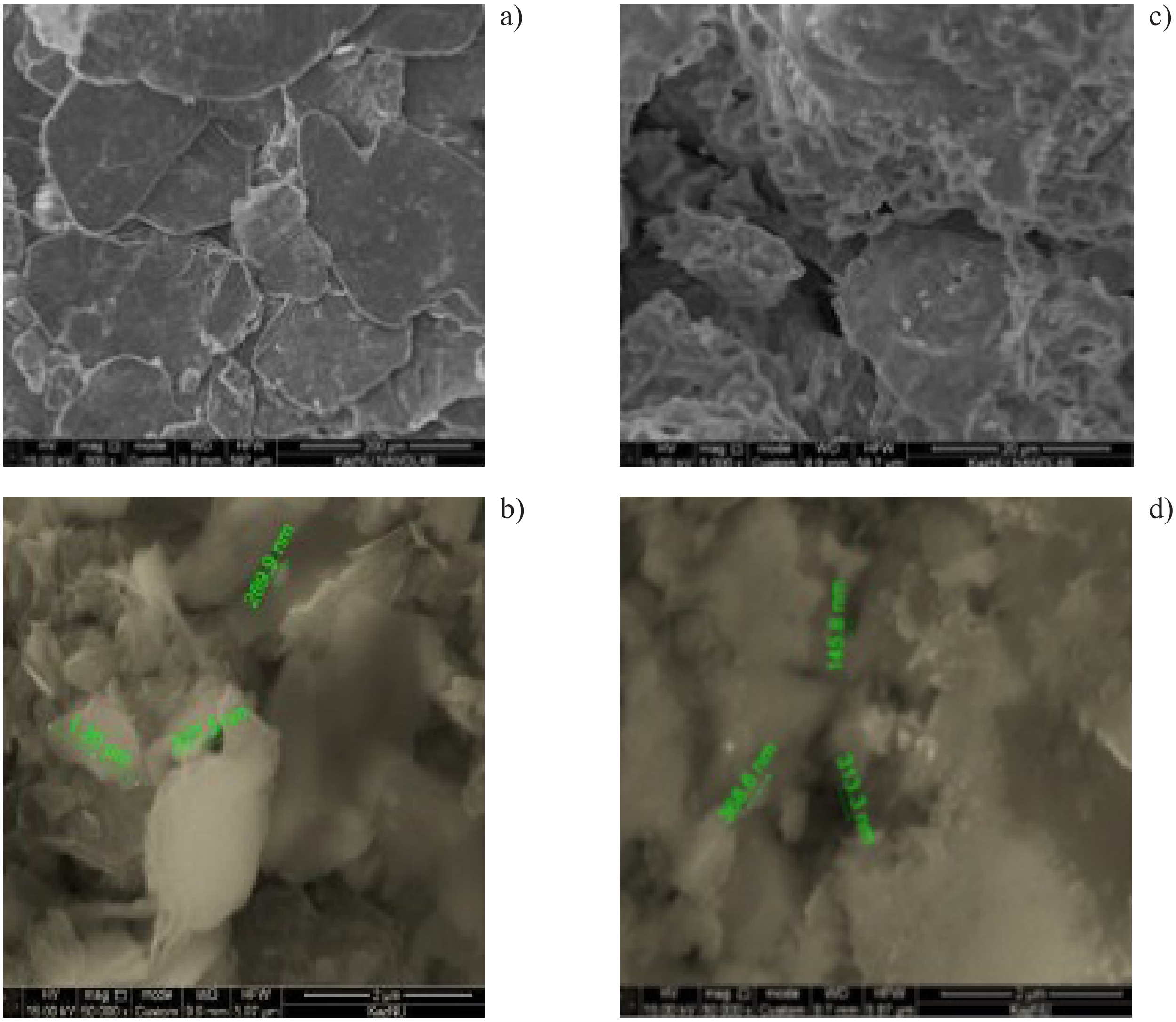

Fig. 7. SEM images of initial coal and coal briquettes: a - initial sample of coal (Oy-Karagay, Kazakhstan); b - coal briquette after calcination; c - coal briquette with PET at a melting temperature range of 120 - 130ºC; d - coal briquette with a pyrotechnic composition introduced into its composition.

while a significant part of the pyrotechnic composition is located on the surface of the coal in the form of particles with sizes from 145.8 to 368.6 nm, mainly in the form of "coagulants" (Fig.7d). The SEM data of coal with PET (Fig.7c) show that the organic component of coal is a mixture of various X-ray amorphous components, mechanical dispersion of coal briquette with PET does not lead to destruction of complexes.

CONCLUSIONS

Briquettes based on substandard raw materials of the Central region of Kazakhstan with different additives were obtained and characterized. It has been found that the mechanical properties of a composite briquette depend both on the composition of the mixture and the material composition of the briquetted mixture, moisture of the mixture, pressing pressure, heat treatment mode of the briquettes, type and consumption of the binder component. An increase in compressive strength is especially noticeable in briquette samples containing coal with the smallest size (coal class 1.5 mm). This is because when briquetting coal fines, the number of interacting surface-active contact groups with a binder is as high as possible. The role of the so-called active centers on the solid surface increases, the adsorption interaction at the interface between the solid and liquid phases is enhanced, the diffusion of binder maltenes into pores and cracks of coal occurs more efficiently, which inevitably leads to an increase in the strength of briquettes. The compressive strength of briquettes made using modified tar and bitumen is higher than the normalized index by 15 - 35 %. Ash content ranges from 15 - 18 %, despite the fact that it is slightly higher in briquettes with modified tar, yet 27 % less than the normalized index. The sulfur content of the briquettes to below 7 to 15 times, water absorption is below 40 - 60 %. The strength of the compositions consisting of coal grains with a grain size of 3 mm is about 3 times lower than the strength of briquettes obtained from coal with a grain size of 1.5 at the same parameters of briquetting. The use of coal sludge and paper waste in the form of cardboard makes it possible to obtain a fuel briquette that is easy to manufacture and compose and simultaneously dispose of coal and paper waste. Waste paper in the main layer provides reliable combustion. The presence of holes in the briquette reduces emissions of harmful substances into the atmosphere.

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