Ceramics Materials with Addition of Ashes from Combustion of Coal in Thermal Power Plants

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Summary

This article reviews results of an experimental study aimed to evaluate the possibility of using ashes - solid wastes from combustion of brown coal in fluid and granulation boilers as one of the ingredients in ceramics production.
Results of testing of the ashes from different combustion processes – combustion of slovak brown coal in granulation and fluid boilers and their usage as a components in dark ceramics showed partial usability in this field. These test consisted of firing the ashes in electric laboratory oven and in industrial fast firing oven. Sample (83) of brown coal fluid bed ash has low Fe content, material has light brown color at temperatures 1100 – 1190 °C after partial melting. Sample (84) with fluid fly ash has higher Fe content, resulting in dark brown color. Both of the samples from fluid combustion of coal require higher temperature for partial melting than 1190 °C. Sample (85) of stabilizate – product of desulphurization – was at the same temperature of 1100 °C completely molten. Sample (86) of granulation ash at the same temperature conditions expanded during firing.
Results of this testing practice have proven, that tested ashes have high content of coloring oxides, mainly iron oxide, which results in red up to brown color of the ceramic material. Because of high CaO content in fluid ashes they can only be used as an additive of porous ceramic materials.
Use of the tested ashes in compounds prepared using similar procedures as the industrial compounds resulted in higher suction capacity. Our tests have proven the possibility of using fluid bed ash as an opening material, fly ash as a partial replacement of opening material and melting ingredient, desulphurization product as a partial replacement of melting ingredient.
Laboratory tests confirmed usability of granulation ashes as an ingredient in dark colored tile manufacture.

Keywords: ceramics, fly ash, solid wastes, thermal power plant

Introduction

Operation technologies of combustion processes are carried out in granulation (temperature of 1 100 - 1 300°C) and fusion type chambers/boilers (temperature of 1 400 - 1 600°C) using powdered coal. In fluid boilers black and brown coal that does not have to be milled, only grinded to particle size of 1.6 – 6, max. 10 mm is combusted at temperature of 800 – 850°C.

Properties of individual fly ashes need to be determined in a first place. Burning of the same type of coal in different combustion chambers leads to formation of different mineral novelties. Knowledge of fly ash properties and their valuable components is essential for determining optimal separation and utilization methods [7].

Analysis

Burning of fossil fuels in different combustion boilers of thermal power plants leads to formation of large amount of wastes – fly ashes and slags. According to present law, wastes must be utilized in first place if possible, while primary area of utilization of these wastes is building industry [7,16].

60 – 70% of solid wastes in Slovakia consist from production of energy in thermal power plants / heating plants. Methods of their disposal depend on economical, legislative and ecological conditions.

Scholars and technicians are aware of possible methods and according conditions for fly ash utilization in building industry [1,18,19,20,21,22,23]. The problem is that each fly ash and slag is different (depending on coal type, combustion conditions and technology) and therefor it is not possible to directly apply common knowledge of fly ash disposal. Each type of solid waste from energetics must be individually analyzed for physical, chemical and mineralogical properties [6,9], radioactivity and technological properties. Consequently physical properties of final building materials must be determined.

Comment

Fly ashes can be used in these main areas of building industries: production of cement and porous concrete, light weight aggregates, wall components made of concrete with hydraulic binder, fine filler (replacement of fine aggregates), as a partial replacement of cement in concrete production, stabilization of bases in road building industries. Other utilization methods include mixed production of aluminum and cement (Grzymek's technology from 1934 was industrially realized in Poland, during and after World War II in Štamberk, Czech Republic), production of mortars, ceramics, terrazzo tiles, concrete roof tiles, bulk thermal insulation, production of insulation materials, mineral wool, polystyrene concrete, filler in epoxy, polyurethane and amine materials, polyester resins. Slag is often used as opening in production of building materials and concretes [4,6,13,15].

This work tests the possibility of ceramics production with addition of brown coal from fluid and granulation boilers.

Use of brown coal fly ash from granulation and fluid boilers in dark ceramics production

The possibility of utilization of brown coal fly ash is presented on sample of brown coal fly ash from ENO Nováky. Basic test were performed in the means of laboratory research on institutes of BERG faculty. Following works were done in factory Dlaždice, Michalovce [5], where effect of adding different amounts of fly ash on properties of ceramic tiles was investigated. High Fe content in fly ashes acts in ceramics as pigment (Fe_2O_3 oxidation). Basic compound (compound no. 5) used in this study as dominant ingredient – 90% wt. is a standard compound from which almost all industrial ceramics are produced.

Research was performed on representative samples of fly ashes produced in ENO Nováky. Following products were tested:

- Granulation fly ash I. and II. From combustion of coal in granulation boilers
- Fluid fly ash –light ash
- Fluid fly ash – bed ash I. and II.
- Desulphurization product
- Desulphurization tailings

Fig. 1 and 2 shows images of granulation and fluid ash morphology with extensive porosity of individual particles retaining the shape of original particles prior to combustion. Morphological properties of fly ashes and their porosity affects moisture content in tested compounds.

Surface areas of granulation fly ashes from ENO Nováky using Ströhlein Areameter ranged from 3.55 to 5.13 $m^2.g^{-1}$. 80% wt. are particles of grain size 0 – 0.071 mm, while 70% wt. are particles finer than 0.040 mm. Density raged from 2.01-2,20 $g.cm^{-3}$.

Surface area of fly ash – bed ranged from 4.06 to 5.1 $m^2.g^{-1}$. fly ash – light ash 5.51 to 5.85 $m^2.g^{-1}$ [1,6]. Density of fluid fly ash – bed was 2,81 $g.cm^{-3}$ and of fluid ash – light ash 2,66 $g.cm^{-3}$.

Tested brown coal fly ashes were sampled for chemical analyses and technological testing. Chemical analyses were done using standard silicate analysis method, where the contents of main components affecting the dominant properties of ceramics were analyzed: SiO_2, Al_2O_3, TiO_2, Fe_2O_3, CaO, MgO, Na_2O, K_2O and loss on ignition (LOI). Granulometric analysis was used to determine particle size distribution.

Red compound, in which effect of high Fe content acting as a pigment during firing is not considered negative, was chosen to practically evaluate possibility of using fly ashes in ceramic tiles production.

Because these materials are considered non plastic,

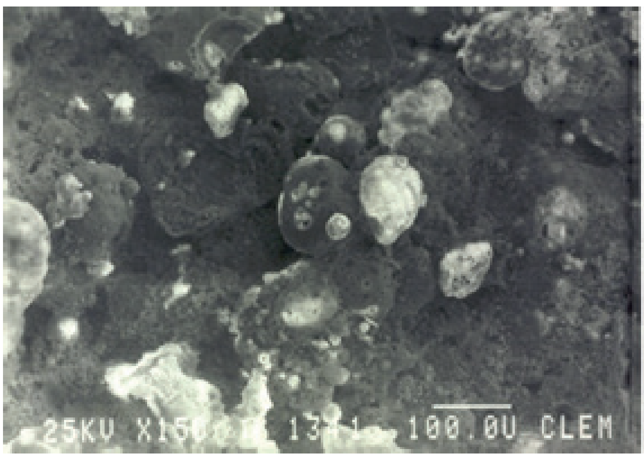

Fig. 1 Morphology of brown coal fly ash combusted in granulation boiler.

Rys. 1 Morfologia popiołów lotnych z węgla brunatnego spalanych w kotle rusztowym

Fig. 2 Morphology of brown coal fly ash combusted in fluid boiler.

Rys. 2 Morfologia popiołów lotnych z węgla brunatnego spalanych w kotle fluidalnym only used fly ashes were fired in electric laboratory furnace at 1 100°C for 12 hours and in industrial fast firing gas furnace at 1 195°C for 1 hour.

Composition of compacting granulate used in tests for red tiles production, factory labeled as "M5" is exactly defined. Weighted amount of granulate mixed with 10% wt. of fly ash were after water addition milled in laboratory mill. Suspension was then dried in laboratory dryer. Dry material was then crushed, sieved on 1.0 mm sieve, moisturized to 5% and compressed by laboratory compactor (Gabrielli, Italy) at 30 MPa.

Results of granulometric analyses (Tab. 1) and chemical composition of tested compounds (Tab. 2) are presented in following tables.

Tab. 1 Dry granulometry of tested fly ashes.

Tab. 1 Sucha granulometria badanych popiołów lotnych

	Mesh size [mm]					
	2.00	1.00	0.50	0.20	0.10	<0.10
Granulation ash I.	3.20	2.76	6.55	29.94	33.95	23.60
Granulation ash II.	4.70	2.77	6.13	28.51	34.14	23.75
Fluid ash - light	0.92	1.81	5.01	19.04	45.46	27.76
Fluid ash - bed	2.37	2.82	10.61	40.52	26.00	17.68
Desulphurization product	65.41	11.47	8.78	6.73	3.52	4.09
Desulphurization tailings	0.00	0.02	0.38	6.97	26.40	66.23
Fluid ash - bed II.	11.63	13.76	15.61	25.72	20.28	13.00

Tab. 2 Chemical composition of tested power plant ashes.

Tab. 2 Skład chemiczny popiołów pochodzących z elektrowni

		LOI.	SiO_2	Al_2O_3	TiO_2	Fe_2O_3	CaO	MgO	Na_2O	K_2O
SAMPLE	Granulation ash I.	4,42	56,26	19,03	0,62	**8,35**	6,16	2,40	0,94	1,82
	Granulation ash II.	3,87	56,28	19,18	0,62	**8,80**	8,06	0,24	0,96	1,96
	Fluid ash - light	11,96	36,46	9,93	0,42	**6,85**	24,64	7,52	0,76	1,36
	Fluid ash - bed	12,6	28,60	12,30	0,28	**3,22**	38,08	3,04	0,60	0,96
	Desulphurization prod.	16,32	37,20	15,59	0,51	**7,70**	17,92	2,40	0,76	1,60
	Desulphurization tail.	1,46	61,26	19,12	0,78	**8,80**	5,60	0,40	0,56	1,96
	Fluid ash - bed II.	9,49	32,82	15,37	0,37	**3,86**	31,70	4,16	0,56	1,42

For the comparison, results of EDX area microanalyses of granulation fly ash, fluid ash – bed and light ash in which analyzed chemical elements are present with content exceeding 0.1% are shown in Tab. 3 [6,10,15].

EDX area microanalyses of products of brown coal combustion products in granulation and fluid boilers [6,15] present qualitative definition of elements in mineral novelties and not their actual contents.

Remarkable on fly ash – light ash is its low sulphur content. Possible explanation is, that in this combustion product mineral novelties are probably not formed – anhydrite, hannebachite, gypsum, ettringite, thaumazite [4]. Only particle 4 is characteristic for minerals from desulphurization. Fe content is higher in light ash than in bed ash.

All the particles in fly ash – bed ash samples have high Ca content, ranging from 43.7 – 60.9 %. Character of the particles is typical for desulphurization process in excess of Ca. Sulphur content ranges from 3.6 – 27.3 %, magnesium from 2.5 – 7.1 %. Mineral novelty – calcium magnesium sulphate is typical product of desulphurization. Dominant elements in analyzed sample of fluid bed ash 2006 are: Si, Ca, Al, Fe, Mg and S.

Testing the possibility of using brown coal fly ashes in ceramics production

Fig. 3 shows results of ceramic potsherds produced using brown coal ashes from combustion of coal in fluid boilers and stabilizate – product of desulphurization.

Sample no. 83 is characteristic for testing of ceramic properties of fluid ash – bed, with only 3.22% and 3.86% Fe_2O_3 resp. – material has brown color. Temperature of 1100 – 1190°C was not high enough to melt inorganic component of ash, which contains 40 – 48% of alum silicates. Properties of sample no. 84 with fluid light fly ash are similar, although Fe content is higher - 6.86 % affecting the color to dark brown. Sample no. 85 where stabilizate – product of desulphurization is used was fully molten at temperature used what is a proof of low temperature eutectics content. Sample no. 86 was made using granulation ash that expanded during firing so much, that original matter remained only on the edges of form.

Results of chemical and technological analyses show high content of colorific oxides, especially iron oxide that

Tab. 3 Results of EDX area microanalyses of particles in granulation fly ash samples from ENO Nováky.

Tab. 3 Wyniki mikroanalizy EDX próbek ziaren granulowanych popiołu lotnego z ENO Nováky

	Content [%]								
EDX of particle	**Si K**	**Al K**	**K K**	**Ca K**	**Fe K**	**Mg K**	**S K**	**Ti K**	**Mn K**
331	56.7	20.2	5.5	1.0	13.3	2.5	0.0	0.8	0.0
332	58.5	19.6	3.5	3.9	11.7	1.6	0.0	0.8	0,4<
333	57.6	16.6	6.6	3.9	11.3	3.0	0.0	0.6	0,3<
321	48.4	19.1	7.6	4.3	16.9	0.9	1.7	0.8	0,3<
322	88.9	3.6	1.2	1.8	3.7	0.5	0.0	0,0<	0,2<
323	52.0	16.7	7.6	4.5	14.8	2.8	0.0	1.5	0,1<

Tab. 4 Shows EDX area microanalyses of fluid fly ash particles – bed ash and light ash.

Tab. 4 Mikroanaliza EDX ziaren popiołów lotnych z kotła fluidalnego – popiół denny i popiół lotny

Content of element [%]									
Particle no.	Si K	Al K	K K	Ca K	Fe K	Mg K	S K	Ti K	Mn K
Light 1	25,5	9,8	4,3	28,7	28,1	0,8<	1,0	1,8	0,0
Light 2	25,4	7,5	1,3	14,1	50,1	0,9	0,0	0,7	0,0
Light 3	52,5	17,5	4,3	10,1	13,0	1,8	0,0	0,4<	0,2<
Light 4	4,5	1,4	0,5<	61,5	3,9	2,3	25,9	0,0	0,0
Bed 1	18,1	5,6	0,5	53,3	7,6	7,1	7,0	0,4<	0,3<
Bed 2	26,2	9,3	0,5	43,7	10,3	5,3	3,6	0,8	0,2<
Bed 3	4,6	1,0	0,3<	60,9	2,7	2,5	27,3	0,7	0,2<

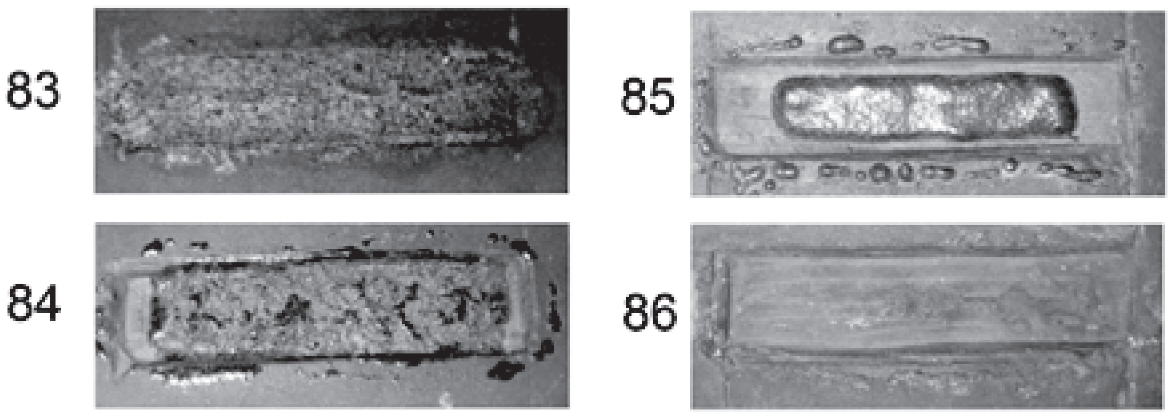

Fig. 3 Testing the possibility of utilization of brown coal ashes as components in ceramic production.

Rys. 3 Badanie możliwości utylizacji popiołów z węgla brunatnego jako składników do produkcji ceramicznej

SAMPLE no. 1	**SAMPLE no. 2**	**SAMPLE no. 3**	**SAMPLE no. 4**	**SAMPLE no. 5**
10 % weight of fluid fly ash – bed ash	10% weight of fluid fly ash – light ash	10 % weight of desulphurizatio n product	12 % weight of granulation fly ash	100 % weight of basic production compound
90 % weight of basic production compound	90 % weight of basic production compound	90 % weight of basic production compound	88 % weight of basic production compound	

Fig. 4 Composition of five different samples used in this study.

Rys. 4 Skład pięciu różnych próbek użytych w badaniach

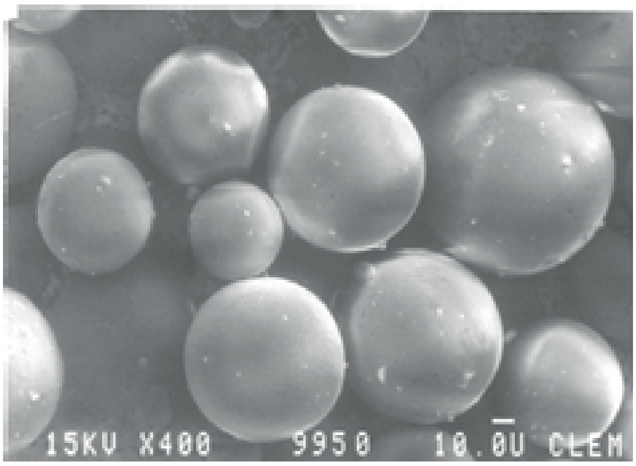

Fig. 5 Shape of microspheres from black coal fly ash combusted in fusion boilers of TEKO Košice

Rys. 5 Kształt mikrosfer pochodzących ze spalania popiołów lotnych z węgla kamiennego w kotłach TEKO Košice

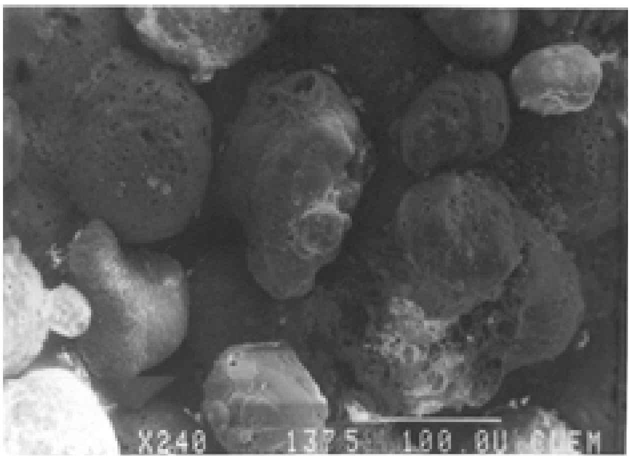

Fig. 6 Composition of particles of brown coal fly ash from granulation boiler

Rys. 6 Skład ziarnowy popiołów lotnych z węgla brunatnego pochodzących z kotła rusztowego

tones the ceramics red up to brown after firing – depending on conditions of firing. High Fe content has pigment like effect that does not negatively affects usability of fly ashes as one of the components in ceramics production, although it limits the use for production of ceramic tiles with dark potsherds, namely red, brown, dark brown or black color of material [5,16].

Results of research on possible utilization of fly ashes in testing compounds

Tested compounds were prepared using procedure similar to the one used to prepare compounds used in industrial production [5]. Weighted raw materials were milled in laboratory ball mills, after water and liquifiers addition, to max. 3% of material retained on the 0.063 mm sieve. In order to objectively consider effect of fly ash addition, industrial reference compound was prepared using the same procedure (sample no. 5).

Based on results of approval tests (Tab. 1 and 2) four laboratory testing compounds were chosen for red tiles production:

Negative effect of high Ca^{2+} ions content on rheological properties of suspension was observed already when wet mixing production compound. Typical industrial production suspension contains 45 – 48 % moisture. High Ca content excluded the possibility of preparing such concentrations of moisture in ceramics suspension.

Ashes with prevailing alumo-silicate content are naturally hydrophilic [6]. Notably higher specific surface area of granulation fly ashes is caused by high particle porosity, as apparent when comparing morphology of particles on Fig. 1 and Fig. 5 and their measured surface areas. Porosity of granulation fly ashes causes higher suction capacity of water. Capillarity cannot be neglected as well. Morphology of granulation fly ash particles with 3-5 $m^2.g^{-1}$ specific surface area, as in accordance with their roundness and porosity, affects moisture content of production ceramic compounds.

Comment: For the comparison: repeated surface area measurements of almost ideal spherical particles – microspheres – from black coal combustion in fusion boilers ranged from 0,50 to 0,76 $m^2.g^{-1}$ (Fig. 4) [6]. This difference in surface area of ashes from granulation boilers is caused by difference in porosity of particles (Fig. 1).

Parameters of compound were close to the ones achieved in industrial production only when granulation ash was used, although necessary solids content was not met. Higher water content in ceramic suspension causes higher energy consumption during the process of drying in spray drier.

Granulate was compacted using hydraulic compactor to 250 x 65 mm format. Press-works were after drying fired in roller furnace for 60 minutes at 1 190°C and industrial electric passage furnace for 52 hours at 1 100°C.

Comment: Modern approach in ceramics production is fast-firing, where high Fe_2O_3 content is contra productive, causing formation of reduction cores, product expansion up to its destruction. It is possible to improve properties of fly ashes for ceramics production by applying physical separation technologies – low intensity magnetic separation of Fe component [4,6]. However, this would negatively influence economical parameters of final product.

When slow firing method (within hours, not minutes) of ceramics production from compound with granulation fly ash content is used, final products are in compliance with quality standards of facing tiles, non-glazed tiles, etc. Products of firing – except compound no. 4 in which granulation fly ash was used had notably higher suction capacity when compared to reference production compound (no. 5), as expected because of higher Ca^{2+} content. Suction capacity of compound no. 4 – with granulation fly ash content was comparable to reference compound (without fly ash addition).

All the samples acquired during fast-firing in roller drier brown color and products were also deformed. Compound no. 4 in which granulation fly ash was used expanded during firing probably because of Fe_2O decomposition and gas formation.

Conclusion

If same type of coal is combusted in different types of boilers – granulation, fluid and fusion at different burning temperatures, different mineral novelties are formed despite the same chemical composition.

Utilization of brown coal fly ash from fluid boilers as a partial replacement of basic compound in dark ceramic production is limited because of high Ca^{+2} content. Calcium negatively affects liquification of ceramics suspension.

Utilization of brown coal fly ash from combustion of coal in granulation boilers as a possible component in dark ceramics production was in laboratory conditions proven as possible.

Based on firing test individual fly ashes can be utilized in different areas:

- fluid ash-bed as opening material
- light fly ash as a partial replacement of opening or smelting components,
- desulphurization product as a partial replacement of smelting component.

Result of laboratory tests of individual ashes point at possibility of using granulation ashes as component in dark ceramics production.

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Materiały ceramiczne z dodatkiem popiołów ze spalania węgla w elektrowniach termicznych

W artykule dokonano przeglądu wyników badania eksperymentalnego, mającego na celu ocenę możliwości wykorzystania popiołów - odpadów stałych ze spalania węgla brunatnego, w kotłach fluidalnych i granulacyjnych jako jednego ze składników w produkcji wyrobów ceramicznych.

Wyniki badań popiołów z różnych procesów spalania - spalanie słowackiego węgla brunatnego w kotłach granulacyjnych i fluidalnych, oraz ich użycie jako składników w ciemnych wyrobach ceramicznych, wykazały częściową użyteczność w tej dziedzinie. Test składał się z wypalania popiołów w elektrycznym piecu laboratoryjnym oraz w przemysłowym piecu do szybkiego wypalania. Próbki (83) popiołów z węgla brunatnego ze złoża fluidalnego mają niską zawartość Fe, materiał ma jasnobrązowy kolor w temperaturze 1100 - 1190°C po częściowym stopieniu. Próbki (84) popiołu lotnego ze złoża fluidalnego mają wyższą zawartość Fe, co w rezultacie daje ciemnobrązowy kolor. Obie grupy próbek z fluidalnego spalania węgla wymagają do ich częściowego stopienia temperatury wyższej niż 1190°C. Próbki (85) ze stabilizatu - produktu odsiarczania - były w temperaturze 1100°C całkowicie stopione. Próbki (86) popiołu z granulacji w tych samych warunkach temperaturowych, rozszerzyły się w czasie wypalania.

Wyniki tego badania wykazały że, w praktyce, badane popioły maja wysoką zawartość tlenków barwiących, głównie tlenku żelaza, co prowadzi do uzyskania czerwono-brązowego koloru materiału ceramicznego. Ze względu na wysoką zawartość CaO w popiele ze złoża fluidalnego, mogą być one używane tylko jako dodatku do porowatych materiałów ceramicznych.

Wykorzystanie badanych popiołów w związkach przygotowanych przy użyciu podobnych procedur jak w przypadku związków przemysłowych spowodowało wzrost zdolności zasysania.

Testy wykazały możliwość wykorzystania popiołu ze złoża fluidalnym jako materiału otwierającego, popiołu lotnego jako częściowo zastępującego materiał otwierający i topliwy, produkt odsiarczania jako częściowe zastąpienie substancji topliwej.

Badania laboratoryjne potwierdziły przydatność popiołów granulacyjnych jako składnika w produkcji płytek o ciemnym kolorze.

Słowa kluczowe: ceramika, popioły lotne, odpady stałe, elektrownia termiczna