Properties of Black and Brown Coal Combustion Products and Possibilities of Their Use

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Summary

This submission reviews the properties and possible utilization of solid wastes originating in the thermal power plants and heating plants as a by-product of brown and black coal combustion in the conditions of granulation, fusion and fluidized-bed fuel firing boilers. In order to choose appropriate utilization method of the ashes, knowledge of their petrologic composition, combustion method, as well as of their physical, chemical, mineralogical and technological properties is fundamental. Ashes are heterogeneous materials composite of particles with different properties affected by coal type and combustion temperature.

Physical properties of ashes from individual boilers include: particle size distribution, mass, volumetric and bulk density, hardness, compactibility, frost resistance, frost susceptibility, optic, electric and magnetic properties, thermal conductivity, fusibility, morphology.

Reactivity of ashes is affected by particle size distribution and surface area. Morphological properties of ashes depend primarily on combustion temperature, chemical composition and properties of coal, atmosphere in which the combustion takes place, combustion chamber construction and combustion process control. Black coal is combusted in fusion boilers at a temperature between 1400°C and 1600°C where ashes are partially up to fully molten. Morphology of inorganic particles is characteristic by its spherical shape and significantly lower surface area when compared to surface area of inorganic particles from granulation and fusion boilers. Brown coal is combusted in granulation boiler at temperatures 1100°C – 1300°C. Inorganic particles tend to be porous with higher surface area when compared to surface area of inorganic particles from combustion of black coal. Combustion temperature of coal in fluidized bed type boilers is 800°C – 850°C. Fluid ash particles from both black and brown coal preserve the shape of original coal particles, perforated structure prevail.

Surface areas of ashes from individual boilers and products of their processing (froth flotation and magnetic separation) ranges from 1 to 33 $m^2.g^{-1}$; densities range from 0.95 to 2.65 and 4.65 $g.cm^{-3}$ resp.

Keywords: ash, combustion, power plant, heating plant, boiler

Introduction

National Coal Council – NCC estimates the energy consumption to rise by more than 27% in the next 25 years, with coal combustion expected to be the major energy source especially in thermal power plants and heating plants.

Combustion of coal leaves a solid waste – by-product that needs to be utilized in industry.

In Slovak thermal power plants and heating plants, mostly the brown coal from "Hornonitrianske bane", Slovakia (70–75%) is burned. Black coal with its minor use is imported from Ukraine, Russia and partially also from Poland and Czech Republic – the "Ostrava" region.

1. Properties of ashes from different combustion regimes

Ashes are heterogeneous materials composite of particles with different properties affected by coal type and combustion temperature. Mineralogical composition of ashes and slags depends on mineralogical composition of burned coal and on combustion regime responsible for creation of mineral novelties. Occurrence of following minerals is most common in coal: kaolinite, halloysite, montmorillonite, pyrite, marcasite, pyrhotine, kalcite, dolomite, ankerite, siderite, halite, sylvite, quartz, gypsum, orthoclase, biotite, cyanite and apatite. Their presence varies from submicroscopic up to significant amounts (Rúžičková et al. 1983, Fečko et al. 2003, Michalíková et al. 2003, 2010a).

World-wide, with Slovakia not being an exception electrical energy is produced by burning of fossil fuels realized in various types of production units. These include fusion (1400–1600°C), granulation (1100–1300°C) and fluidized bed type (800–850°C) firing boilers.

Coal combustion temperature significantly influences morphology and mineralogical properties of ashes. Most important properties are considered to be the particle size distribution, surface area, density, content of crystallic and amorphous phase, presence of unburned coal residuals (UCR) characterized by loss on ignition (LOI), magnetite and non-magnetite phase content and content of major and minor chemical elements.

Prior to combustion in fusion type firing boilers it is necessary to mill the coal to 90% passing 0.2 mm mesh size, in granulation type boilers to achieve 0–2 mm size fraction and for combustion in fluidized-bed type boilers grinding the coal to 0–6.5–10 mm size fraction is satisfactory.

1.1. Physical properties

Physical properties include: particle size distribution, mass, volumetric and bulk density, hardness, compactibility, frost resistance, frost susceptibility, optic, electric and magnetic properties, thermal conductivity, fusibility, morphology (Kušnierová et al. 2011, Šimáčková et al. 2011).

1.1.1. Fly ash granularity

Reactivity of ashes is affected by particle size distribution and surface area. Deviation in particle size distribution and in percentage of individual size classes in ashes from brown coal and black coal combustion in fusion and granulation boilers is of minor significance. Size distribution of such ashes is in the range of 0–3 mm with high content (30–70%) of fine particles under 40 µm. Grain size of slags from fusion boilers is up to 30 mm and from granulation boilers up to 10 mm.

Particle size distribution of ashes from fluidized-bed type boilers is as follows:

- Fluid black coal ash – bed ash: 1–16 mm.
- Fluid black coal ash – light fly ash: 0–0.2 mm with 90% under 45 µm.
- Fluid brown coal ash – bed ash: 0–3 mm.
- Fluid brown coal ash – light fly ash: 0–0.4 mm 60% under 45 µm.

1.1.2. Surface area of fly ashes from different types of combustion boilers

Fusion boiler:

- **EVO Vojany power plant –** black anthracitic coal.
- Surface area of fly ashes from electrostatic dust separators depend on the content of UCR characterized by LOI: 0.68% LOI – 1.34 $m^2 \cdot g^{-1}$, 25.50% LOI – 6.82 $m^2 \cdot g^{-1}$.
- Flotation products – UCR
- Concentrate from first cleaning flotation – UCR: 88.22% LOI: 17.46 $m^2 \cdot g^{-1}$.
- Tailings from rougher flotation after magnetic separation: 0.61% LOI: 1.46 $m^2 \cdot g^{-1}$.
- **TEKO Košice heating plant** – black semi-anthracitic coal.
- Surface area of ash from settling pit: 4.39–4.79 $m^2 \cdot g^{-1}$. Pore volume: 0.0038–0.0068 $cm^3 \cdot g^{-1}$. 12–15% and more LOI.
- Surface area of fresh ash from accumulator: 5.57 $m^2 \cdot g^{-1}$. Pore volume: 0.0046 $cm^3 \cdot g^{-1}$. 23% LOI.
- Surface area of slag is less than 1 $m^2 \cdot g^{-1}$.

Granulation boiler:

- ENO Nováky power plant – brown coal.
- Surface area of ash: 4.16–5.62 $m^2 \cdot g^{-1}$.
- Surface area of slag: 26.47–29.82 $m^2 \cdot g^{-1}$.

Fluidized-bed boiler

- EVO Vojany power plant – black coal (year 2010).
- Fluid ash – light fly ash K5: 32.7265 $m^2 \cdot g^{-1}$. Pore volume: 0.0246 $cm^3 \cdot g^{-1}$
- K6: 31.7130 $m^2 \cdot g^{-1}$. Pore volume: 0.0240 $cm^3 \cdot g^{-1}$
- Fluid ash – bed ash K5 and K6: 1–3 $m^2 \cdot g^{-1}$
- ENO Nováky power plant – brown coal.
- Fluid ash – bed ash: 5.55–5.85 $m^2 \cdot g^{-1}$
- Fluid ash – light fly ash: 14–17 $m^2 \cdot g^{-1}$

1.1.3. Density

- Black coal ash from fusion type boilers: 1.95–2.15–2.35 $g \cdot cm^{-3}$,
- black coal ash from fluidized-bed boilers: 2.45–2.55 $g \cdot cm^{-3}$,
- brown coal ash from granulation boilers: 1.90–2.16 $g \cdot cm^{-3}$,
- brown coal ash from fluidized-bed boilers: 2.10–2.55 $g \cdot cm^{-3}$,
- Products of dressing the brown coal fly ash from fusion boilers:

– flotation concentrate – UCR: 0.95–1.49 $g.cm^{-3}$,
– Fe concentrate: 3.95–4–4.65 $g{\cdot}cm^{-3}$.

1.1.4. Hardness

Silicate content in fly ashes is about 50% of the weight thus making inorganic particles of the ash hard and fragile with hardness equal to quartz. Particles are likely to break on impact.

1.1.5. Compactibility

Compactibility of ashes along with their ***frost susceptibility*** and ***frost resistance*** is related to particle size and shape. Microscopic morphological analysis is crucial in order to explain the results of compaction tests – better ability of the porous and highly damaged grains to accept water (Ďurica and Krličková 1993). Frost resistance affinity is caused by the presence of porous unburned coal residuals with high surface area.

1.1.6. Fusibility

Fusibility of ashes is significant in ceramics burning, where ashes can be used as a replacement of natural aluminosilicates. Melting point (Batelka 1980, Vídeňská 1986) of easy melting ashes is 800–1040–1200°C, semi-difficult melting ashes 1200–1425°C and difficult melting ashes above 1425°C.

1.1.7. Morphology of particles

Temperature is a major factor affecting ash morphology. Morphological properties of ashes depend primarily on combustion temperature, chemical composition and properties of coal, atmosphere in which the combustion takes place, combustion chamber construction and combustion process control. Poly-component structure of ashes is closely related to combustion temperature which causes ashes to partially up to fully melt, while particles tend to conglomerate up to clinker.

Combustion of black coal in fusion boilers (in EVO Vojany, US Steel Košice and TEKO Košice) at a temperature between 1400°C and 1600°C where ashes are partially up to fully molten leads to formation of spherical particles and glasseous phase. Inorganic component of ashes is typical by spherical shape of microspheres (plerospheres and cenospheres). Magnetite novelties particles also have spherical shape. UCR particles are porous as confirmed by surface area measurements. These are of higher order than the surface areas of inorganic ash particles. Typical content of UCR in ash is 8–15% and higher.

Brown coal in granulation boiler is combusted at temperatures 1100°C – 1300°C while the upper bound is limited by ash particles melting point. Ash particles preserve the original shape of the coal particles, rounded because of partial melting, however perforated structure prevails. Spherical shape of the particles practically does not occur.

Coal combustion temperature in fluidized-bed type boilers (brow coal in ENO Nováky, black coal in EVO Vojany) is 800–850°C. Temperature cannot exceed 900°C as this leads to slag formation. Fluid brown coal as well as black coal ashes preserves the morphology of the original coal particles – mineral skelet with unburned coal residuals. Glasseous phase content is much lower when compared to its content in ashes from fusion and granulation boilers and is affected by the presence of easy melting phases meltable in the temperature range 800–850°C.

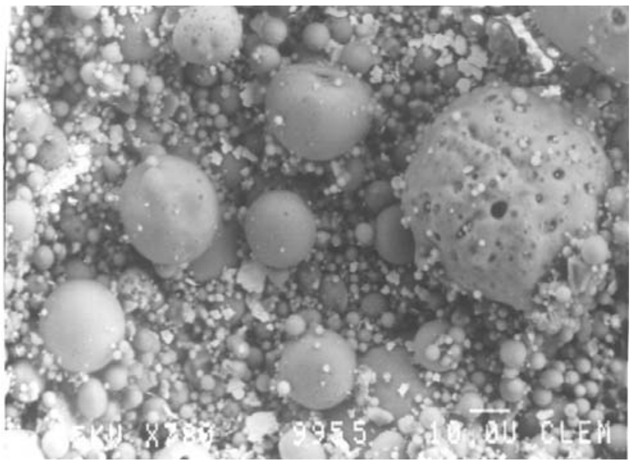


Fig. 1. Black coal ash morphology from – TEKO Košice (780× magnification)

Rys. 1. Morfologia próbki popiołu z węgla kamiennego – TEKO Košice (powiększenie 780×)

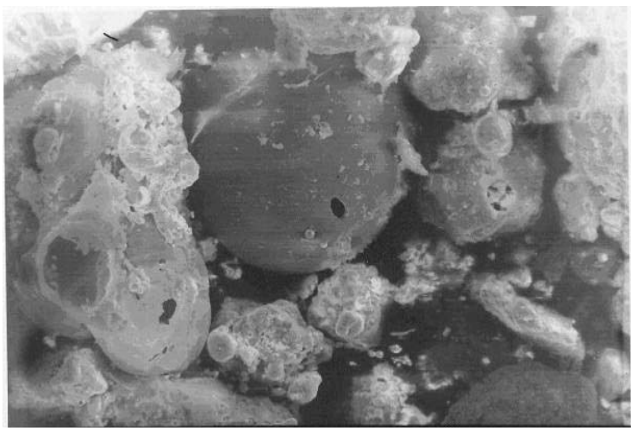


Fig. 2. Brown coal ash morphology – ENO Nováky (400× magnification)

Rys. 2. Morfologia próbki popiołu z węgla brunatnego – ENO Nováky (powiększenie 400×)

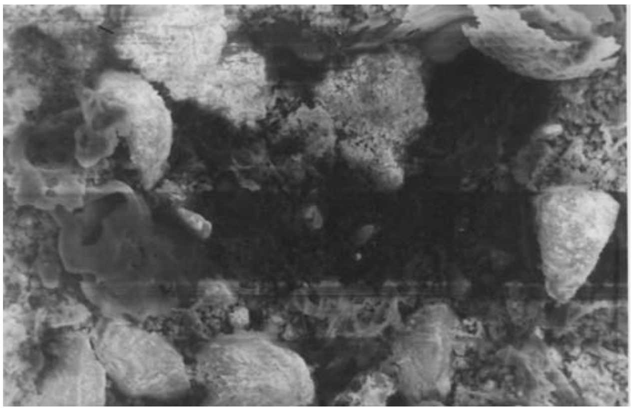


Fig. 3. Fluid light fly ash particle composition from EVO Vojany (550× magnification)

Rys. 3. Popiół fluidalny z EVO Vojany (powiększenie 550×)

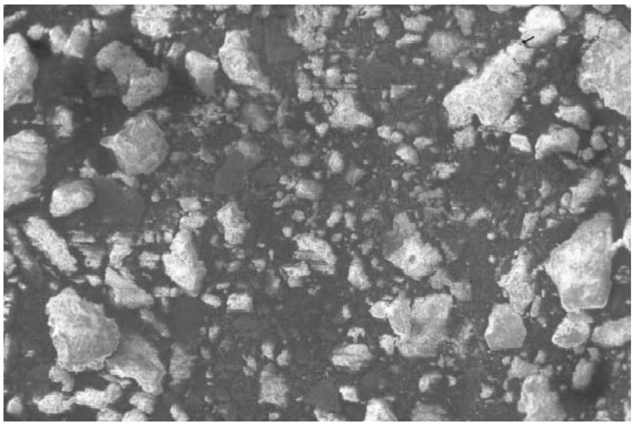


Fig. 4. Fluid light fly ash particle composition from ENO Nováky (110× magnification)

Rys. 4. Popiół fluidalny z ENO Nováky (powiększenie 110×)

Fig. 5. Fluid bed ash particle composition from EVO Vojany (90 x magnification)

Rys. 5. Popiół fluidalny z EVO Vojany (powiększenie 90×)

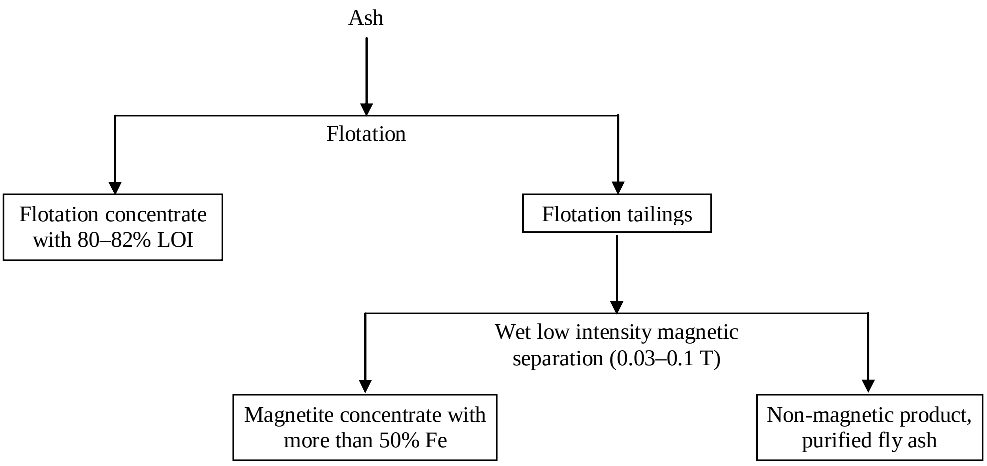


Fig. 6. Ideological flowsheet of black coal ash separation from fusion boilers

Rys. 6. Schemat ideowy flotacji popiołu lotnego

Morphology of black and brown coal ash from different combustion devices is shown in following figures (Fig. 1–5) – scanning electron photomicrographs:

Result of comparison of light fly ash and bed ash from fluidized-bed combustion of brown and black coal is as follows: there is no difference in appearance and morphology of particular ash particles from equal combustion conditions regardless of ash type and the only difference is size and composition – chemical elements content. This information can be used when monitoring pollution by dust. Contamination of area with solid wastes from coal burning in different combustion devices – fusion, granulation and fluidized-bed type boilers can be identified according to morphology of dust particles in particular distances from pollution source (Stárková 1997).

Morphology of particles affects also **sorption capacity**, which is different for particles of unburned coal residuals. Particles with 88.2% LOI and surface area of 17.46 $m^2 \cdot g^{-1}$ were identified in black coal ash flotation concentrates from fusion boilers. Tailings from rougher flotation and magnetic separation with 0.61% LOI has surface area of 1.46 $m^2 \cdot g^{-1}$, with almost ideally smooth surface of spherical particles and minimal sorption capacity.

1.1.8. Morphology of products from fly ash separation (Michalíková et al. 2003 monografia)

Black coal ash from fusion boilers contains four main valuable components. Applying separation methods it is possible to obtain products usable in different industrial areas (Hycnar 1987).

1. Flotation concentrate with high content of UCR, 78–88% LOI, micromorphology of unburned coal residuals particles with chemical elemental analysis in analysed particles (Fig. 7 and 8).
2. Magnetite concentrate with 50–55% Fe content, separated from ashes resulting from combustion of black coal in fusion boilers using low intensity magnetic separation and wet cleaning stage.
3. Ash cleared of unburned coal residuals and magnetite iron with 1–3% LOI.
4. Microspheres obtained as "on water surface floating particles" (Fig. 9).

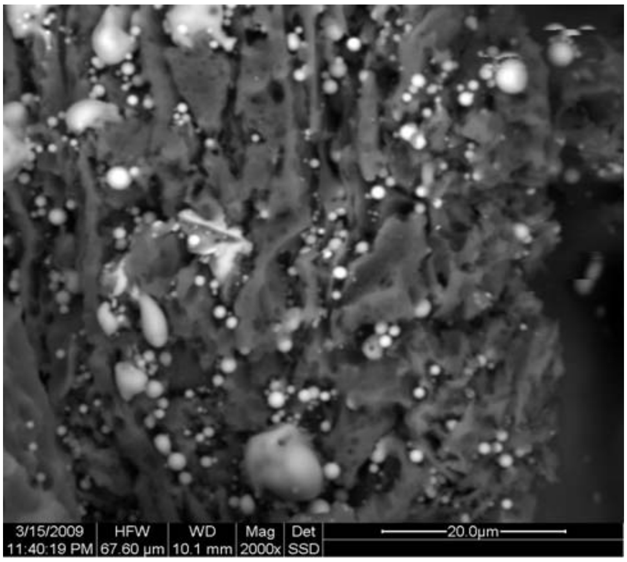
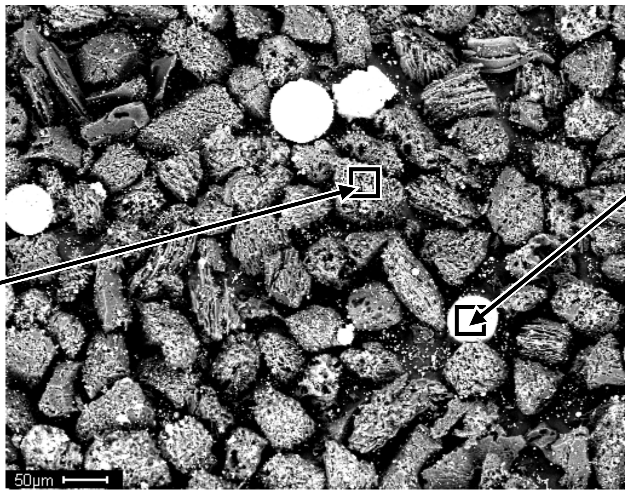


Fig. 7. Flotation concentrate particle of unburned coal residual

Rys. 7. Koncentrat flotacyjny z pozostałościami niedopału

Element	Wt%	At%
C	87.53	91.72
O	07.85	06.18
Al	01.60	00.75
Si	03.02	01.35



Element	Wt%	At%
C	43.13	66.42
O	16.84	19.46
Al	00.85	00.58
Si	01.52	01.00
Ca	00.36	00.17
Mn	01.37	00.46
Fe	35.93	11.90

Fig. 8. Micromorphology of particles of flotation concentrate

Rys. 8. Mikromorfologia cząsteczek koncentratu flotacyjnego

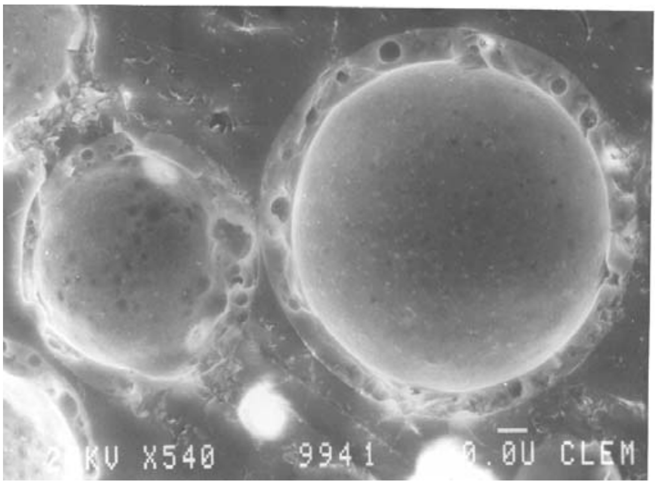


Fig. 9. Whet area of black coal fly ash from TEKO Košice (540× magnification)

Rys. 9. Powierzchnia przekroju popiołu lotnego z TEKO Košice (powiększenie 540×)

Separation of black coal ash from fusion boilers using following flowsheet resulted in products, that were further investigated using electron microscope with intention of determining their physical – mainly morphological and technological properties.

1.1.8.1. Morphology of unburned coal residuals

Research department of USSteel Košice conducted a micromorphological and EDX analysis of characteristic particles. Whet areas of particles with high carbon content were studied using the "Lucia" system image analysis on OLYMPUS optical microscope (40× immersion objective).

Fig. 7 shows unburned coal residuals with 82.50% loss on ignition.

Particles of partially coked coal samples – UCR – have higher internal porosity. In individual particles, fractions of inertinite, partially molten semifusinite to fusinite, integral grey areas of vitrinite, fractions of inertinite – micrinite and partially carbonated inertinite – semifusinite grains were detected. Particles were coked to maximum of 35%. UCR particles are of irregular shape with edges, some of the particles are porous, other are lamellar. Composition is formed by spherical inorganic particles with higher molecular density; other particles are composed of tiny spherical particles that form spherical conglomerate. In both types of particles, C, O, Si, Al, Fe, K, Mg, Ca and S, along with small amounts of Mn and other elements as well as particles with higher iron content were identified using EDX analyses. Samples contained also coked particles with thin-walled and thick-walled network structure, partially coked vitrinite, semifusinite and inertinite particles. Fig. 7 shows typical particle surface micromorphology under 2000× magnification – UCR with 0–5 μm microspheres with small inorganic spheres (cenospheres and plerospheres) wedged inside of pores. As previously stated, surface area of flotation concentrate is of about one order higher than the surface area of inorganic components.

Because of high inner and outer surface area (15–17 $m^2 \cdot g^{-1}$) have the UCR particles notable sorption ability (Michalíková et al. 2010b).

Fig. 8 shows UCR particles composition with EDX analyses of two selected areas. EDX of the typical UCR particle shows 87.53% carbon and EDX of inorganic particle shows 35.93% Fe content.

<u>*Morphology of microspheres*</u>

It is characteristic for the microspheres to have almost spherical shape and smooth surface. Most of them are hollow and porous, what can also be seen on the image of a whet area of black coal ash particle from TEKO Košice under 540× magnification. Low density of microspheres – starting from 0,3 $g \cdot cm^{-3}$ can be accounted to high shell porosity with porous inner part (Fig. 9).

Conclusion

Physical properties of ashes and acquired separation products significantly affect their usability especially in building industry. **Particle size distribution, morphology and surface area** affect their reactivity.

Hardness of ashes is lower than the hardness of quartz. Their comminution depends on their morphology; they can be easily disintegrated on impact; milling is very energy consuming because of silicate content. Our measurement of fusion boilers slag disintegration using mechanical and dynamical crushing showed, that calculated hardness coefficient is equal to harness coefficient of rocks such as shales, limestones, ankerite, gypsum, siderite, anthracite (Michalíková et al. 2003).

Compactibility, frost susceptibility and frost resistivity is affected by morphology and particle size of ashes.

Fusibility of ashes affects energy consumption in process of ceramics firing; melting point of individual ashes is in the range of 850–1425°C.

Morphological properties affect mixing properties of concrete mixtures. If ash is used **as an active ingredient in concrete and mortars, requirement of cement is decreased as it replaces its part**.

Knowledge of physical properties of ashes from fusion, granulation and fluidized bed type boilers make the choice of suitable application in building industry easier.

Results of our research of chemical and mineralogical properties of ashes will be discussed in next article.

This work was supported by the Scientific Grant Agency of the Ministry of Education of Slovak Republic VEGA under the grant No. 1/1222/12 and by the Slovak Research and Development Agency under the contract No. APVV- 0423-11.

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Właściwości produktów spalania węgla kamiennego i brunatnego oraz możliwości ich wykorzystania

W artykule przedstawiono właściwości oraz przegląd możliwości wykorzystania stałych produktów spalania węgla kamiennego i brunatnego pochodzących z elektrowni i elektrociepłowni spalających paliwo w kotłach rusztowych, fluidalnych i komorowych. Przedstawiono właściwości fizyczne, chemiczne, mineralogiczne i technologiczne popiołów.
Dla prawidłowego doboru metody utylizacji popiołów konieczna jest znajomość ich składu petrograficznego, metody spalania, oraz właściwości fizycznych, chemicznych, mineralogicznych.
Popioły są niejednorodnym materiałem kompozytowym z składającym się z cząstek o różnych właściwościach, wynikających z typu węgla oraz warunków (temperatury) spalania.
Właściwości fizyczne popiołów z poszczególnych kotłów to: rozkład wielkości cząstek, masa, objętość i gęstość nasypowa, twardość, zagęszczalność, mrozoodporność, wrażliwość na niskie temperatury, właściwości optyczne, elektryczne i magnetyczne, przewodnictwo cieplne, topliwości, i morfologia.
Reaktywność popiołu wpływa na rozkład wielkości cząstek i ich powierzchnię. Właściwości morfologiczne popiołów zależą przede wszystkim od temperatury spalania, składu chemicznego i właściwości węgla, atmosfery, w której następuje spalanie, konstrukcji komory spalania oraz sposobu sterowania procesem spalania.
Węgiel kamienny jest spalany w kotłach komorowych w temperaturze pomiędzy 1400°C a 1600°C, przy czym powstający popiół jest częściowo lub całkowicie stopiony. Cechą morfologiczną cząstek popiołu jest kulisty kształt i znacznie niższa powierzchnia w stosunku do powierzchni nieorganicznych cząstek powstających w kotłach rusztowych i fluidalnych Węgiel brunatny jest spalany w kotłach rusztowych w temperaturze 1100°C – 1300°C. Cząstki nieorganiczne są zazwyczaj porowate, mają większą powierzchnię, w porównaniu do powierzchni cząstek popiołu ze spalania węgla kamiennego. Temperatura spalania węgla w kotłach fluidalnych wynosi 800°C - 850°C. Cząsteczki popiołu lotnego zarówno z węgla kamiennego i brunatnego zachowują kształt pierwotnych cząstek węgla, z dominującą struktura porowatą.
Powierzchniach popiołów z poszczególnych kotłów i produkty ich przeróbki (flotacji pianowej i separacji magnetycznej) charakteryzują się powierzchnią od 1 do 33 $m^2 \cdot g^{-1}$ i odpowiednio gęstością w zakresie od 0,95 do 2,65 i 4,65 $g \cdot cm^{-3}$.

Słowa kluczowe: węgiel, popiół, spalanie, elektrownia, elektrociepłownia, kocioł