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Fly ash particles spheroidization using low temperature plasma energy

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Abstract. The paper presents the investigations on producing spherical particles 65–110 μm in size using the energy of low temperature plasma (LTP). These particles are based on flow ash produced by the thermal power plant in Seversk, Tomsk region, Russia. The obtained spherical particles have no defects and are characterized by a smooth exterior surface. The test bench is designed to produce these particles. With due regard for plasma temperature field distribution, it is shown that the transition of fly ash particles to a state of viscous flow occurs at 20 mm distance from the plasma jet. The X-ray phase analysis is carried out for the both original state of fly ash powders and the particles obtained. This analysis shows that fly ash contains 56.23 wt.% SiO_2; 20.61 wt.% Al_2O_3 and 17.55 wt.% Fe_2O_3 phases that mostly contribute to the integral (experimental) intensity of the diffraction maximum. The LTP treatment results in a complex redistribution of the amorphous phase amount in the obtained spherical particles, including the reduction of O_2Si, phase, increase of $O_{22}Al_{20}$ and Fe_2O_3 phases and change in Al, O density of $O_{22}Al_{20}$ chemical unit cell.

1. Introduction

Cenospheres are produced by thermal power plants (TPP) as a byproduct of coal combustion. A cenosphere is a lightweight, inert, hollow sphere that ranges in size from 50 to 500 μm. The continuous and nonporous cenosphere walls have the thickness ranging from 2 to 10 μm. The content of spherical particles is represented mostly by nitrogen and carbon dioxide [1-3].

The unique combination of the low density, small size, spherical shape, high hardness, melting temperature, and chemical inertness of cenospheres stipulates for their wide use in construction and other industries. Cenospheres have a mixed vitreous transparent structure comprising 76% glass, 22% mineral substance, and 2% impurities and are represented by aluminosilicate glass, silica, mullite, calcite, iron oxides, calcium silicates, and sulfates.

The aim of this work is to investigate the production of fly ash-based spherical particles with the size range between 20-100 μm using the low temperature plasma (LTP) treatment. The particle morphology is determined by the properties of fly ash (dispersiveness, chemical composition) and electrophysical properties of plasma generator [4]. Due to the heterogeneous chemical [5, 6] composition of fly ash, a number of preliminary transformations should be carried out to obtain particles with the predetermined properties.

The synthesis process includes three stages as presented in Figure 1. The first two stages relate to the preparation of fly ash for the subsequent production of spherical particles.

At Stage 1, the particles are grinded down in a planetary ball mill to submicrometer dimensions. At Stage 2, the spray drying technique is used to obtain agglomerated powders ranging in size from 30 to 120 μm. At Stage 3, agglomerated powders are subjected to a high-temperature plasma flow to produce spherical particles.

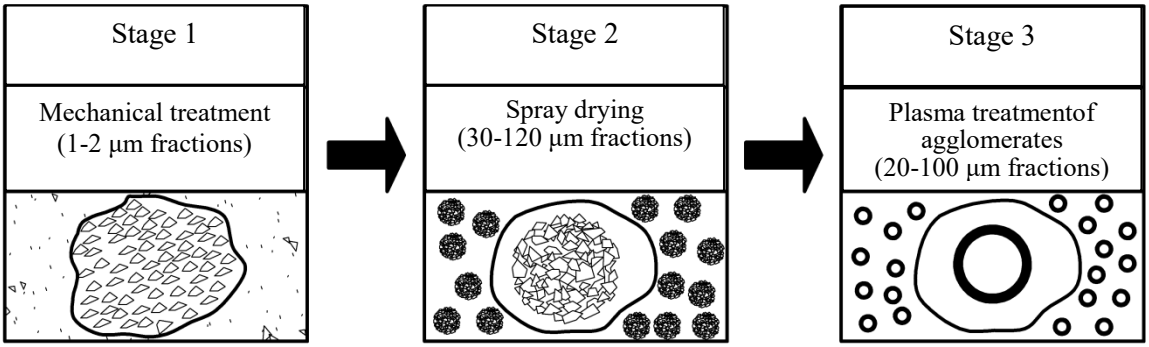

Figure 1. Flow-chart of producing fly ash-based spherical particles using plasma energy

2. Materials and Methods

Fly ash, produced from the combustion of coals was collected in dust collectors at Seversk TPP and used as the material to be treated. The results of the chemical composition analysis of fly ash are given in Table 1.

Table 1. Averaged chemical composition of fly ash collected at Seversk TPP

Oxide content	Composition, (wt.%)
SiO_2	51.16
Al_2O_3	35.07
Fe_2O_3	3.62
CaO	8.33
MgO	0.91
R_2O	0.23
Ignition losses	0.68
Viscosity modulus	5.23

Phase composition of the original specimens and obtained spherical particles was investigated on the XRD-6000 X-ray Diffractometer from Shimadzu which was modified for digitalsignalprocessing. The Rietveld refinement was used to quantify phases [7, 8] by modeling the amorphous phases dominating in the integral intensity of the diffraction maximum. The determination of amorphous phases was based on investigations of the original state of fly ash crystal lattices. Energy dispersive X-ray spectroscopy on the scanning electron microscope JSM-7500F (JEOL, Japan) was carried out to investigate the morphology, shape, and size of obtained spherical particles.

3. Experimental

The test bench was designed to carry out spheroidization of silicate particles. The structural scheme of producing spherical particles is presented in Figure 2.

The powder feeder is used to perform the fly ash delivery to the area of plasma jet. The delivery is provided with nitrogen as a carrier gas, which reduces the enthalpy and the rate of plasma jet when enters it together with the prepared mixture. As a result, the mixture discharges into the plasma jet mainly due to the inertial forces. While the particles are present in the high-temperature plasma flow,

they are heated and then transit to a viscous flow state. Owing to their surface tension force, the particles take on a spherical shape. These spheres travel to the collection unit represented by a box filled with water.

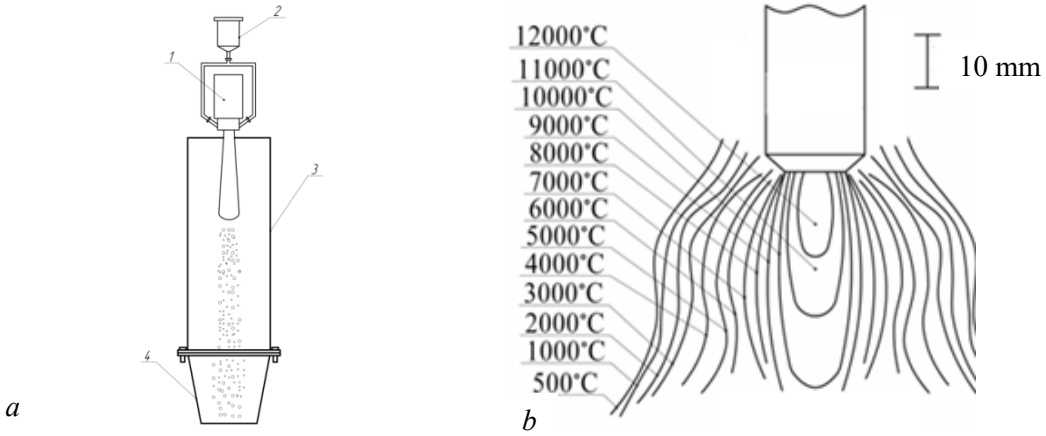

Figure 2. Structural scheme of producing spherical particles (*a*): *1* – plasma torch; *2* – powder feeder, *3* – processing chamber, *4* – particle collection unit. Distribution of plasma temperature field (*b*)

In Figure 2b, the distribution of plasma temperature field is shown. According to the data found in the literature, the melting temperature for fly ash is 1700 ºC. Thus, the conditions favorable for the particle transition to a viscous flow state are formed within the temperature range observed at 20 mm distance from the jet center. This fact proves the efficiency of the LTP treatment for the particle spheroidization.

4. Results and Discussion

4.1. X-ray phase analysis

After the LTP treatment, fly ash and spherical particles produced thereof were subjected to the X-ray phase analysis (XRP). The XRP analysis shows that such stable phases as SiO_2, Al_2O_3, Fe_2O_3, TiO_2, CaO, MgO are present in the original state of fly ash that is also supported by its chemical composition (see Table 1). Fly ash composition is represented by a complex mixture of SiO_2, Al_2O_3, Fe_2O_3 components. The amount of SiO_2 phase is ~56.2 wt.%, while that of Al_2O_3 and Fe_2O_3 phases is 14.41 and 17.55 wt.%, respectively. The XRD pattern of fly ash in its original state is plotted in Figure 3.

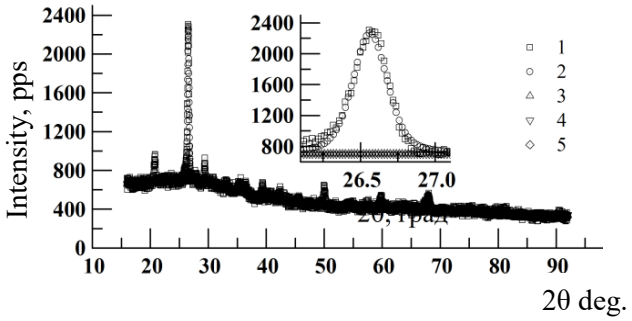

Figure 3. XRD pattern of fly ash in its original state: 1 – experimental; 2 - SiO_2 phases; 3 - Fe_2O_3 phases; 4 – TiO_2 phases.

The XRD pattern of fly ash is complex and characterized by both intensive and weak overlapping reflections. The Rietveld refinement allows determining the fly ash composition of 56.23 wt.% SiO_2, 20.61 wt.% Al_2O_3 and 17.55 wt.% Fe_2O_3 phases that make a dominating contribution to the integral (experimental) intensity of the diffraction maximums. The quantitative phase analysis (QPA) using the Rietveld method shows that the total phase content is 94.39 wt.%. This proportion is adequate to the results obtained for phase identification in fly ash.

The spherical particles produced as a result of LTP treatment, have an amorphous structure identified by the QPA by the Rietveld method. For this purpose, the model of amorphous SiO_2, Al_2O_3, Fe_2O_3 phases is created for their original state. The obtained atom density of these phases determines the size of the chemical unit cell with atoms of SiO_2, Al_2O_3, Fe_2O_3 phases. The XRD patterns of plasma-treated fly ash are presented in Figure 4.

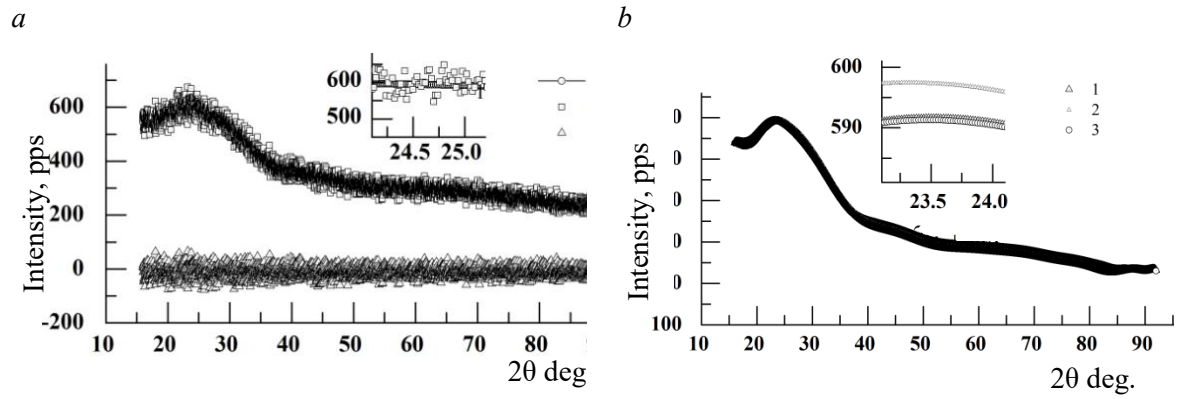

Figure 4.XRD patterns of fly ash particles after LTP treatment:
a – experimental (1); theoretical (2); b – amorphous phases: 1 – Fe_2O_3; 2 – $O_{22}Al_{20}$; 3 – SiO_2.

The optimum sizes of chemical unit cells and the equilibrium atom arrangement are detected at the following stage. The achieved amorphous state of $O_{22}Al_{20}$, Fe_2O_3 and O_2Si phases is characterized by the known space distribution of atoms in the chemical unit cell of the optimized size. In case of the primitive lattice of the space group, these phases can be identified by the chemical unit cells of $O_{22}Al_{20}$, Fe_2O_3 and O_2Si phases. The structural factors and, hence, the integral intensities of diffraction patterns can be obtained for optimized chemical unit cells of $O_{22}Al_{20}$ and O_2Si phases. The amorphous $O_{22}Al_{20}$, Fe_2O_3 and O_2Si model phases are used to determine the phase composition of fly ash. Figure 4 b contains the XRD patterns of fly ash particles analyzed by QPA by the Rietveld method. A superposition of the amorphous phase intensity is 93.13 wt.% of the XRD pattern shown in Figure 4a. The investigation results show that the spherical particles produced by the LTP treatment possess the amorphous structure. Additionally, the LTP treatment results in a complex redistribution of the amorphous phase amount in the obtained particles, i.e. the considerable decrease of O_2Si phase; increase of $O_{22}Al_{20}$ and Fe_2O_3 phases and change in Al, O density of $O_{22}Al_{20}$ chemical unit cell.

4.2. SEM Investigations

Figure 5a presents the scanning electron microscopy (SEM) image of the exterior view of the fly ash-based spherical particles obtained after the LTP treatment.

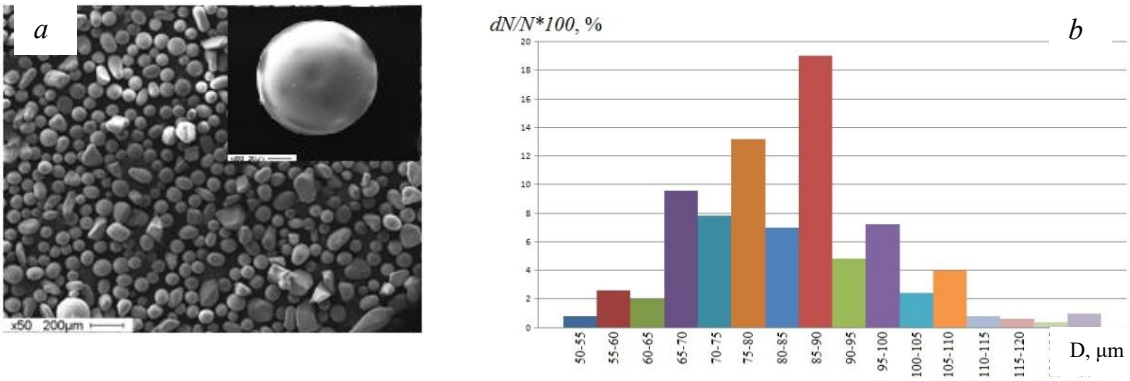

Figure 5. SEM image of spherical particles obtained after fly ash LTP treatment: *a* – exterior view, *b* – size distribution

According to Figure 5b, the greater number of spherical particles ranges in size from 65 to 110 μm. These particles have no defects and are characterized by a smooth exterior surface. The presence of coarse particles (130-300 μm) of irregular shape indicates the formation of big drops during the plasma treatment, which are then sintered but not able to melt.

5. Conclusions

These investigations allowed producing spherical particles having the size range of 65–110 μm based on fly ash generated from the coals combustion collected at Seversk TPP, Tomsk region, Russia. The spherical particles were obtained as a result of fly ash treatment with low temperature plasma energy. The exterior surface of the obtained spherical particles is smooth and without defects. The test bench was designed to carry out the experiments on spheroidization of fly ash-based particles. Thus, the conditions favorable for the particle transition to a viscous flow state were achieved within the temperature range observed at 20 mm distance from the jet center. The fly ash particles underwent the XRP analysis both grinded (original state) and plasma-treated. Using the Rietveld method, it was determined that fly ash contains 56.23 wt.% SiO_2, 20.61 wt.% Al_2O_3 and 17.55 wt.% Fe_2O_3 phases that make the dominating contribution to the integral (experimental) intensity of the diffraction maximums. The LTP treatment resulted in a complex redistribution of the amorphous phase amount in the obtained spherical particles, i.e. the considerable decrease of O_2Si phase; increase of $O_{22}Al_{20}$ and Fe_2O_3 phases and change in Al, O density of $O_{22}Al_{20}$ chemical unit cell.

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