

## Content of Various Size and Density Particles in Cenosphere Concentrates of Volatile Coal Combustion Ashes from the Kuznetsk Coalfield

S. N. VERESHCHAGIN<sup>1</sup>, L. I. KURTEEVA<sup>1,2</sup> and A. G. ANSHITS<sup>1,2</sup>

<sup>1</sup>*Institute of Chemistry and Chemical Technology, Siberian Branch of the Russian Academy of Sciences, Ul. K. Marksa 42, Krasnoyarsk 660049 (Russia)*

*E-mail: [snv@icct.ru](mailto:snv@icct.ru)*

<sup>2</sup>*Siberian Federal University, Pr. Svobodny 79, Krasnoyarsk 664041 (Russia)*

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### Abstract

Using an aerodynamic method with the subsequent particle size analysis of the fractions obtained a separation process was carried out for cenosphere concentrates from volatile ashes of the Kuznetsk field coal combustion at the Moscow Thermal and Electric Plant TETs-22, the Belovo State Regional Power Station as well as for several cenosphere concentrate fractions obtained at the Novosibirsk Thermal and Electric Plant TETs-5. As much as 90 fractions with the various particle size (70–250  $\mu\text{m}$ ) and bulk density (0.16–0.52  $\text{g}/\text{cm}^3$ ) have been isolated from the concentrate of the Moscow TETs-22. Irrespective of particle size the bulk density of fractions with the maximal yield is equal to 0.33–0.35  $\text{g}/\text{cm}^3$ , whereas the ratio of the apparent wall thickness to the particle diameter amounts to 0.042–0.043. The process of the aerodynamic separation of cenospheres from different concentrate sources can be satisfactorily described by theoretical dependences for the carryover of spherical particles; the deviations are caused, first of all, by a non-spherical shape of cenospheres.

**Key words:** volatile ashes, cenospheres, aerodynamic separation

### INTRODUCTION

One of the valuable components of volatile ashes from thermal power stations is presented by cenospheres those represent hollow thin-walled silica-alumina spherical entities. Owing to the morphological features of the structure, the chemical and mineral phase composition, they are widely used as components for grouting mortars, filling agents for polymeric and heat-insulating materials. For the last years it has been demonstrated that cenospheres can be used for obtaining some hi-tech products: porous matrices for the conditioning of liquid radioactive waste products and the wastes from hydrometallurgy [1–3]; sensitizers for emulsion explosives [4, 5]; carriers for obtaining encapsulated solid extraction agents and ion-exchange

materials (including zeolites) for the sorption of metal ions [6]; catalysts for hydrocarbon oxidation processes [7]; micro-spheric membranes for the diffusion separation of gases [8–11]. However, for the successful use of cenospheres in these fields it is necessary that they should exhibit certain chemical and mineral phase composition as well as stabilized physicochemical properties (particle diameter, wall thickness and morphology, bulk density, specific surface area, etc.).

Despite of a considerable number of the works devoted to the investigation of volatile ashes and their components (cenospheres), there are scarce data in the literature concerning the preset size particles content in cenosphere concentrates, the density, as well as the analysis of connection between physicochemical properties and morphological characteristics of particles.

So, the authors of [12] determined the grain-size composition of the components of carry-over ashes from various thermal power stations was determined using a laser diffraction micro-analyzer, allowing them carry out the express analysis of particles within the range of 0.16–1160  $\mu\text{m}$  in the aqueous media. They have determined the content of different size cenospheres in the isolated concentrates, characterized the fractions obtained using the methods of optical and scanning electron microscopy, established the bulk density and wall thickness values for the particles. The authors of [13] estimated the content of cenospheres with the density  $<2.2 \text{ g/cm}^3$  in ashes using a mixture of organic solvents and established the size distribution of ash particles ranging from 1 to 200  $\mu\text{m}$ . However, the products obtained in these studies represent a mixture of particles to a considerable extent heterogeneous in size, density and composition, which is caused by the use of simple single-phase separation methods those do not allow revealing possible internal interrelations between these parameters. It is obvious, that the experimental establishing of such dependences is of interest both from the standpoint of obtaining the cenospheres with the parameters required for their further practical application, and from the point of understanding the process of microsphere formation under coal combustion conditions.

Much more complete separation of the components of volatile ashes into narrow fractions according to their physicochemical properties could be achieved using complex multi-stage separation methods with the combination of magnetic and electromagnetic separation, grain-size classification, gravity separation with the help of static methods (within the liquid media of various density) or dynamic ones (in an upstream or downstream liquid/gas flow). So, according to [14], the gravity separation of a cenosphere concentrate in the environment of organic solvents combined with magnetic separation and particle size sieve analysis has allowed the authors to obtain about 50 products differing in the content of iron oxide, in the size and bulk density.

However this method is laborious, low-productive, it requires for high expenses, for toxic, highly inflammable and expensive solvents and results in the pollution of cenospheres.

Moreover, the method of particle fractionation, based on the hydrostatic principle, cannot provide fine separation of the concentrate due to a limited set of liquids with various density values needed.

Much more flexible adjustment of the properties of fractions obtained could be achieved using dynamic classifiers in the liquid or gas phase, based on the carryover of particles with certain characteristics in a moving stream of working medium [15–17].

The purpose of the present work consisted in determining the content of cenospheres with certain size and bulk density in the cenosphere concentrates of volatile coal combustion ashes from the Kuznetsk coal field, adjusting the complex procedure of cenosphere mixture separation into narrow fractions with respect to size and density as well as determining the interrelation between these parameters.

## EXPERIMENTAL

In this work we used the cenosphere concentrates from volatile ashes obtained in the process of Kuznetsk coal combustion at the Moscow TETs-22 (M), the Belovo State Regional Power Station (B) and the Novosibirsk TETs-5 (N). Figure 1 displays the distribution of cenospheres throughout the sizes in the initial concentrates sampled at the Moscow TETs-22 and the Belovo power station. The size of the most part of particles entering into their composition varies within the range of 0.03–0.2 mm. The bulk density of the initial non-separated concentrates of M and B cenospheres is equal to 0.36 and 0.42  $\text{g/cm}^3$ , respectively. From the concentrate of N cenospheres we the way sieve separations obtained and used in the experiments the fraction of 0.125–0.1 mm with the bulk density of 0.39  $\text{g/cm}^3$ .

For quality monitoring of the fractions obtained (the absence of splinters, the occurrence of damages due to aerodynamic separation, sphericity of particles) and the estimation of their distribution throughout the size we used the method of optical microscopy (a Biolam microscope equipped with a computer system for image registration). An automatic size measurement was carried for 2000–4500 particles with the use of specially developed Msphere software.

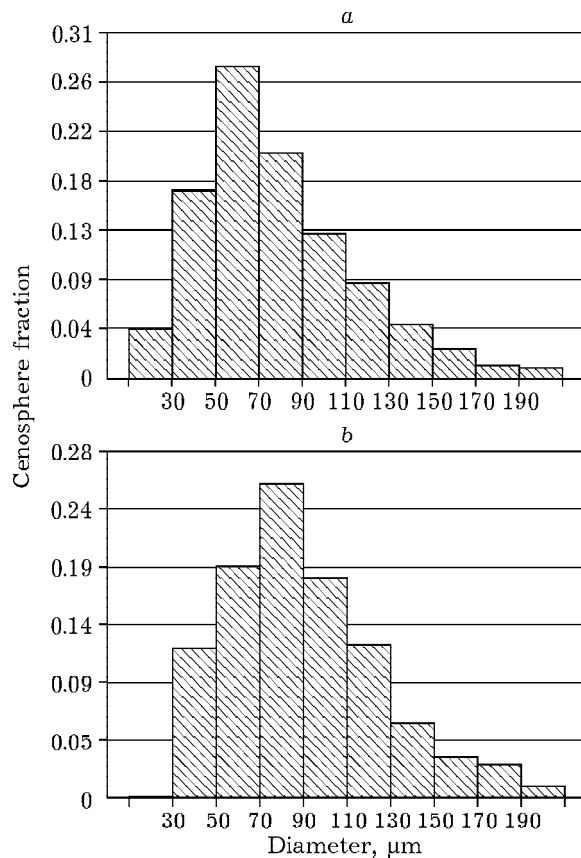


Fig. 1. Size distribution of cenospheres (the fraction of the total number of particles) in the initial concentrate of the Moscow TETs-22 (a) and the Belovo State Regional Power Station (b).

The area and perimeter of individual particles were calculated using ImageJ software [18].

The installation for the aerodynamic separation of cenospheres (Fig. 2) consisted of a batching feeder 1, a wind tunnel 2 (32 mm internal diameter, 320 mm length), containing two gas-distribution gratings positioned at a distance of 40 and 90 mm from the bottom of the tunnel, as well as a cyclone 3 with a collector 4. The flow rate of air was set with the help of a regulator and was measured by a rotameter 5. We used the values of air flow rate corresponding to linear velocities of 0.38, 0.34, 0.30, 0.26, 0.21, 0.17, 0.13 and 0.085 m/s recalculated for the wind tunnel cross-section taking into account pressure variation in the tunnel during the separation of cenospheres.

Compressed air was supplied into the bottom part of the wind tunnel from a compressor through the regulator and rotameter. In the course of one cycle, as much as 20 cm<sup>3</sup> of cenospheres were fed into the air flow between the

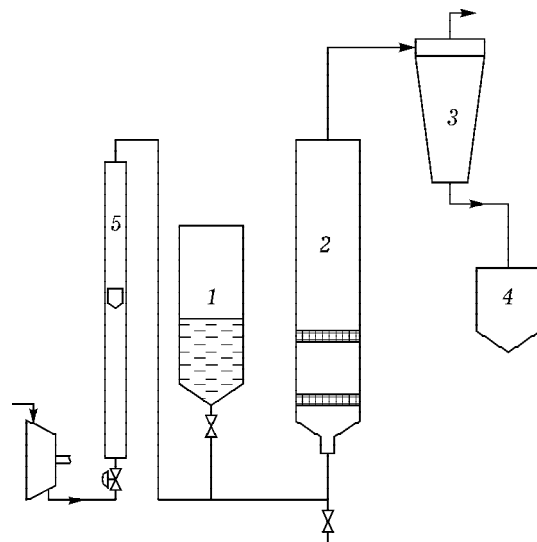


Fig. 2. Schematic diagram of the set-up for the separation of cenospheres in the upstream airflow: 1 - cenosphere batching feeder, 2 - wind tunnel, 3 - cyclone, 4 - collector, 5 - rotameter.

rotameter and the wind tunnel, which cenospheres moved by the air current towards the wind tunnel and were then blown therein during 10 min. Light cenospheres were therewith carried away by the air flow into a cyclone and were accumulated within its bottom part, whereas heavier particles stayed in the wind tunnel. After completing the cycle the air supply was stopped, and heavy cenospheres were then removed from the apparatus through an orifice in the bottom part of the wind tunnel. The portion of the initial concentrate (150 cm<sup>3</sup>) was separated at the increasing values of air flow rate, since the minimal one. Light cenospheres were separated into fractions, whereas heavy particles were used as an initial material for the subsequent separation at a higher air flow rate. For the separation of large heavy cenospheres those required for higher linear air flow velocities, we additionally used a wind tunnel with the internal diameter of 22 mm. In this case we loaded as much as 10 cm<sup>3</sup> of cenospheres were loaded, and the linear air velocity was ranged from 0.11 to 0.62 m/s.

Thus, each separated fraction of cenospheres was characterized by certain size of particles and flow velocity. For example, the fraction with the particle size  $-0.25+0.2$  mm and the linear air velocity of 0.34–0.38 m/s contains more than 90 % of cenospheres 0.2–0.25 mm

in diameter those are removed at the velocity of 0.38 m/s, but are staying at the velocity of 0.34 m/s.

The mass loss value in the process of aerodynamic separation did not exceed 5 % for the complete separation of the concentrate with a consecutive use of all the mentioned velocity values. The mass loss in the course of grain-size sieve classification did not exceed 2 %.

## RESULTS AND DISCUSSION

The procedure of cenosphere concentrate fractionation by size and bulk density is based on the process of aerodynamic separation of particles in the upstream air flow and the subsequent grain-size classification.

According to the laws used in chemical technology [19], the carryover of spherical particles occurs at gas flow velocity value ( $W$ ) which is determined as

$$W = Ar\mu/(d_{eq}\rho(18 + 0.575\sqrt{Ar})) \quad (1)$$

Here  $Ar = d_{eq}^3\rho g(\rho_1 - \rho)/\mu^2$  is the Archimedean criterion;  $d_{eq}$  is the equivalent diameter of a particle, m;  $\mu$  is the viscosity of medium,  $N \cdot s/m^2$ ;  $g = 9.807 \text{ m/s}^2$  is the free fall acceleration;  $\rho_1$  is the apparent density of cenospheres,  $kg/m^3$ ;  $\rho$  is the density of medium,  $kg/m^3$ .

One can see that at equal diameter of particles the possibility of carryover for spherical particles of identical size is determined by their density, whereas at equal density this value is determined by particle diameter. When there are particles of different size and density in the mixture (as it is in the case of cenosphere concentrate), then there will be the carryover at the preset gas flow velocity could be observed only for those particles, whose proportion between  $d_{eq}$  and  $\rho_1$  satisfies the expression (1). In this connection after the aerodynamic separation of the initial concentrate at different airflow velocities we carried out a sieve separation of the fractions obtained to isolate the products containing particles close to each other both in size, and in the apparent density.

The apparent density of particles was estimated from the assumption that all cenospheres are of spherical shape. It is known, that free space fraction in the layer of close-sized spherical particles poured randomly amounts to about

0.4 [20]. Hence, the bulk density of a cenosphere layer ( $\rho_b$ ) can be determined as

$$\rho_b = 0.6\rho_1 \quad (2)$$

Using the method of aerodynamic separation for the concentrate of cenospheres and the subsequent grain-size classifications of the fractions obtained from the initial concentrate sampled at the Moscow TETs-22, we have succeeded in isolation of about 90 products with various bulk density and particle size. As one would expect, at low airflow velocities there is the carryover of the finest and lightest particles occurring, whereas at high velocity values the ablation of large and heavy particles is observed. The maximum yields of narrow cenosphere fractions depend on the airflow velocity and on the size of particles. So, the maximum number of cenospheres with the size  $-0.105+0.07$  mm were blown within the flow velocity range of 0.13–0.21 m/s, whereas the maximum yield of particles with diameters ranging within  $-0.2+0.165$  mm was obtained at the velocity values of 0.34–0.38 m/s. The bulk density of the products obtained (Fig. 3) varies within the range of 0.15–0.52  $g/cm^3$ , the particles with the density of 0.3–0.4  $g/cm^3$  prevailing in all the fractions. Their content depends on the size and ranges from 2 to 6.5 mass % with respect to the initial concentrate. The content of cenospheres with the bulk density  $0.45 < \rho_b < 0.2 \text{ g/cm}^3$  was much less than 1 %.

The fact that in all the fractions, irrespective of particle size, the particles with the maximal yield are characterized by close values of

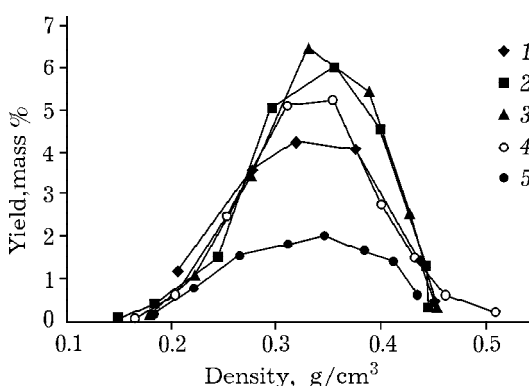


Fig. 3. Content of particles with different bulk density and size. Fraction, mm:  $-0.105+0.070$  (1),  $-0.135+0.105$  (2),  $-0.165+0.135$  (3),  $-0.200+0.165$  (4),  $-0.250+0.200$  (5).

bulk density ( $0.33\text{--}0.35\text{ g/cm}^3$ ) (see Fig. 3) is the most interesting and unexpected result of separation.

When taken into account that each particle represents a sphere with diameter ( $D$ ) and wall thickness ( $h$ ) formed from a material with density ( $\rho_w$ ) and it has no defects such as gas bubbles and extraneous inclusions, it follows from simple geometrical reasons in view of the expression (2) that

$$\rho_b = 0.6\rho_w(1 - (1 - 2h/D)^3) \quad (3)$$

So, basing on the  $\rho_b$  value experimentally obtained for certain fraction one could calculate the wall thickness to the particle diameter ratio value  $h/D$ , and to determine the average particle wall thickness  $h$  from  $D$  value. It is obvious, that the calculated value of  $h$  is the apparent value since a real particle wall is always to some extent containing heterogeneities in the form gas inclusions, pinholes of various nature crystal phases, unburned carbon particles, etc. Nevertheless, the equality of the apparent density values for cenospheres of different size corresponds to the constancy of the  $h/D$  ratio for these particles. Thus, from the results presented in Fig. 3, and from the expression (3) it follows that for the cenospheres whose content is maximal in this fraction, there is statistically significant correlation between particle size and wall thickness observed in the cenosphere concentrate under investigation. Using the value of  $\rho_w = 2.45\text{ g/cm}^3$  (densimetric wall material determination [12]) one can easily calculate that for the bulk density of  $0.33\text{--}0.35\text{ g/cm}^3$  the ratio  $h/D = 0.042\text{--}0.043$ . Establishing the nature of the law revealed requires for special studies, however it is evident that this law is not casual and could be most likely connected with the mechanism of cenosphere formation in the process of coal combustion.

According to optical microscopy (Fig. 4), all the products represent a mixture of particles whose shape is mainly close to spherical one; there are also separate non-spherical irregular-shaped particles and fusion-spliced aggregations of several cenospheres. On the surface of some cenospheres one can observe roundish flowed entities oozing out, whose size varies from several micrometers to  $20\text{--}30\text{ }\mu\text{m}$ .

Since cenospheres in the products isolated are far from ideal spheres in shape, it is of interest to compare experimental values of gas flow velocity providing the carryover of parti-

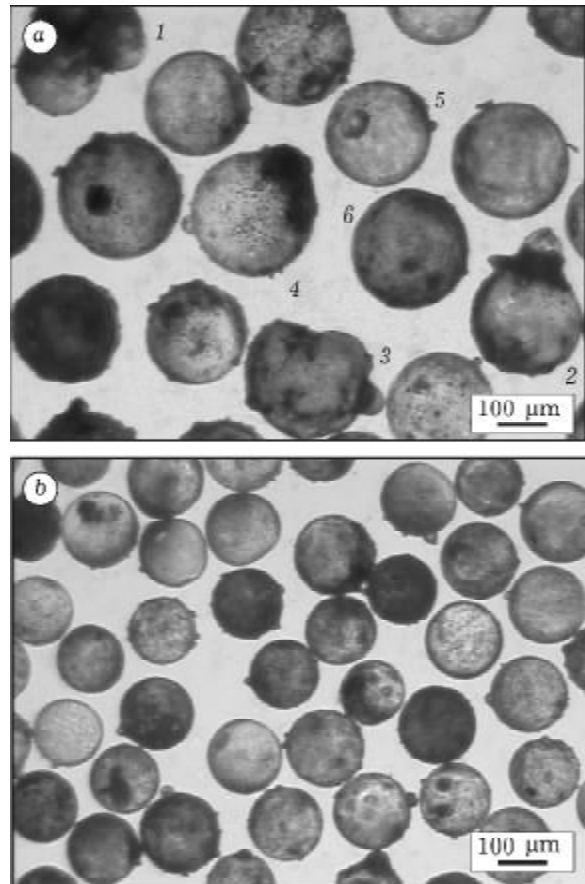


Fig. 4. Photomicrographs for cenospheres obtained in the process of aerodynamic separation of ash concentrate from the Moscow TETs-22 in transmitted light: a - fraction of  $-0.25+0.2\text{ mm}$ , linear airflow velocity of  $0.34\text{--}0.38\text{ m/s}$ ; b - fraction of  $-0.165+0.135\text{ mm}$ , linear airflow velocity of  $0.26\text{--}0.30\text{ km/s}$ ;  $\alpha = 0.75$  (1),  $0.77$  (2),  $0.78$  (3),  $0.86$  (4),  $0.92$  (5),  $0.98$  (6).

cles of certain size and density with theoretical values calculated from the equation (1). For the estimation of the equivalent diameter  $d_{eq}$  calculation required for particle size distribution has been determined for all the separated fractions basing on optical microscopy data. The histograms of particle size distribution represented almost symmetric bar graphs whereof the fraction sizes have been determined, *i.e.* the size range including more than 90 % of particles.

For polydisperse mixtures (*i.e.* the mixtures containing particles of different size) the equivalent diameter  $d_{eq}$  is usually determined as

$$d_{eq} = \left( \sum \frac{x_i}{d_i} \right)^{-1} \quad (4)$$

Here  $x_i$  is a mass fraction of the  $i$ -th fraction;  $d_i$  is the average diameter of particles in the  $i$ -th fractions.

In the case under our consideration it is difficult to perform such calculation since to determine the mass percentage of the particles with certain size in the fraction is impossible. In this connection an arithmetic-mean size  $d_1$  was used as the equivalent diameter value. Taking into account the fact that the type of particle size distribution is almost symmetric, the distribution itself exhibits an insignificant width, and the difference in density of separate particles within the fraction caused by the method of obtaining is insignificant. The calculated difference between  $d_1$  and  $d_{eq}$  does not exceed 5–8 % (under the assumption that there is equality in the particle density within the fraction).

Figure 5 displays the field of correlations for the experimentally obtained value of gas flow velocity  $W$  resulting in the carryover of the fraction with the average size  $d_1$  and density  $c_i$ , and the theoretical velocity value calculated with the use of size  $d_1$  and  $\rho_1$  from the equation (1). One can see that the experimental points obtained from the separation of sample M, are concentrated near the line of regression with unit slope ratio (the correlation coefficient amounting to 0.95). Additional experiments concerning the separation of cenosphere concentrates obtained from the ashes of other sources (samples B and N) have also demon-

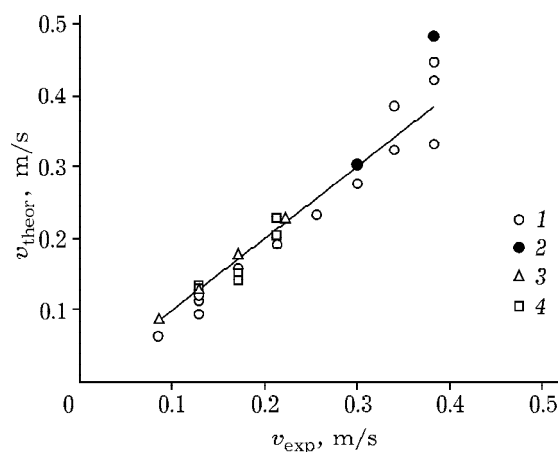


Fig. 5. Comparison of theoretical and experimental values of airflow velocity obtained in the aerodynamic separation of the cenosphere concentrate from the Moscow TETs-22 (1, 2), the Belovo Power Station (3), and the Novosibirsk TETs-5 (4).

strated good agreement between the theoretical and experimental values of gas flow velocity. Thus, for the majority of fractions the process of aerodynamic separation can be quite adequately described by the expression (1).

The greatest point deviation from the line of regression is observed for the highest gas flow velocity of 0.38 m/s (see Fig. 5). To all appearance, it is connected with the fact that in the case of high velocities the airflow in the apparatus with small diameter is much less uniformly distributed throughout the cross-section. Similar picture could be also observed when the shape of particles differs from spherical one, since in this case will change both the particle carryover velocity and the density of their packing would change. Accordingly, the calculated value of apparent density should change too, which would result in changing the calculated  $W$  value.

It is known that being the particles of irregular shape, the carryover velocity to differ from the values calculated according to the equation (1). Usually for irregular-shaped particles the  $d_{eq}$  value is supposed to be equal to the diameter of a conditional sphere whose volume  $V$  is equal to the volume of this irregular-shaped body [19], according to the expression

$$d_{eq} = \sqrt[3]{\frac{6}{\pi} V} = 1.24 \sqrt[3]{\frac{M}{\rho}} \quad (5)$$

Here  $V$ ,  $M$ ,  $\rho$  are the volume, mass, density of a particle, respectively.

It is obvious that to perform a quantitative account particle nonsphericity within the framework of suggested approach is impossible. However, basing on the microscopic studies one could perform a semiquantitative evaluation of the content of various shape particles (see Fig. 4). A spherical cenosphere in the photomicrographs should look like a circle, whereas irregular-shaped particles (lengthened, with the ledges, fusion-spliced agglomerates and so on) non-parallel oriented with respect to the optic axis of a microscope, will look like irregular-shaped figures. The fact whether a particle image is close to the circle can be judged the parameter  $\alpha$  which is determined as

$$\alpha = 4\pi S/P^2 \quad (6)$$

Here  $S$  is the area of the particle image;  $P$  is the perimeter of the circle whose area is equal to the area of the particle image.

For all spherical particles  $\alpha = 1$ , and any shape distortion will result in the reduction of this parameter. The dependence of parameter  $\alpha$  on particle shape is presented in Fig. 4, *a*. So, for the cenospheres 1–6 values of  $\alpha = 0.75, 0.77, 0.78, 0.86, 0.92, 0.98$ , respectively. One can see that at  $\alpha > 0.92$  the deviation from sphericity is either absent, or insignificant; for irregular-shaped particles  $\alpha \leq 0.86$ .

The parameter  $\alpha$  have been calculated for the particles entering into the composition of the two samples designated by solid points in Fig. 5: sample I – the fractions of cenospheres corresponding to the point of the greatest deviation (particle size  $-0.25+0.2$  mm, the gas flow velocity of  $0.34-0.38$  m/s); sample II – the fractions of cenospheres satisfying to the linear law ( $-0.165+0.125$  mm, the gas flow velocity of  $0.26-0.30$  m/s).

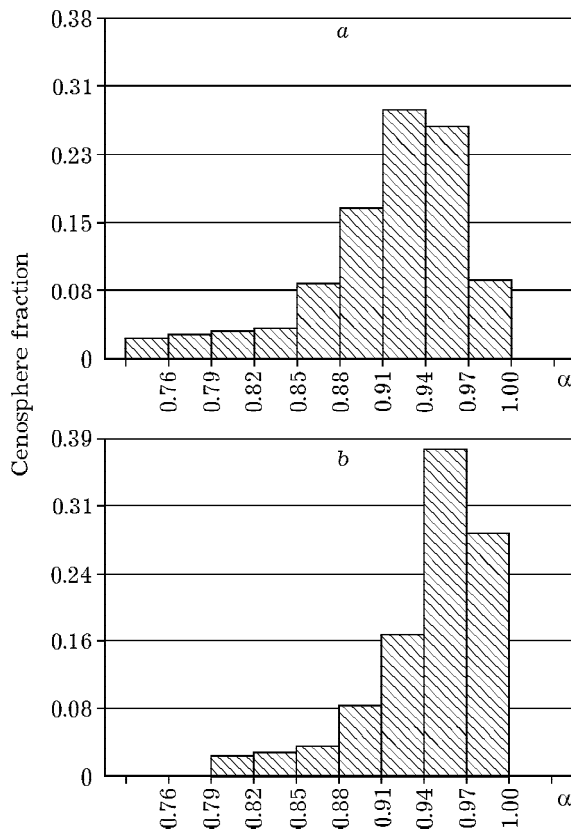


Fig. 6. Parameter  $\alpha$  distribution for cenospheres obtained in the aerodynamic separation the ash concentrate from the Moscow TETs-22: *a* – fraction of  $-0.25+0.2$  mm, linear airflow velocity of  $0.34-0.38$  m/s; *b* – fraction of  $-0.165+0.135$  mm, linear airflow velocity of  $0.26-0.30$  m/s.

Figure 6 displays the  $\alpha$  value frequency distribution for the chosen samples of cenospheres. One can see that for the sample I (see Fig. 6, *a*) the distribution exhibits a higher width, the distribution median is accounted for 0.926, whereas for the sample II (see Fig. 6, *b*) a narrower distribution is observed with the median value of 0.954. The differences observed for the distributions correspond to a much greater content of irregular-shaped cenospheres in the sample I as compared to the sample II. So, one can observe  $\alpha < 0.92$  for 42 % of particles, and  $\alpha < 0.94$  for 61 % of particles for the sample I. For the sample II one can observe  $\alpha < 0.92$  for 21 % of particles, and  $\alpha < 0.94$  for 31 % of particles. Thus, the discrepancies found out between experimental and calculated gas flow velocity values required for the carryover of preset-sized cenospheres with certain value of the apparent density, could be most likely caused by deviations from the spherical particle shape.

## CONCLUSION

An approach is developed for determining the yield of cenospheres with different size and bulk density entering into the concentrate composition. Correlation between the content, size and bulk density has been determined for cenosphere concentrate from volatile ashes by-produced in the Kuznetsk coal combustion at the Moscow Thermal Electric Power Plant TETs-22. It has been demonstrated that for the particles of any size the maximal yield is observed for the cenospheres with the density of  $0.33-0.35$  g/cm<sup>3</sup> and the apparent wall thickness to particle diameter ratio  $h/D = 0.042-0.043$ .

It is shown that the process of aerodynamic separation is satisfactorily described by theoretical dependences for the ablation of spherical particles, whereas the deviations are caused, first of all, by non-spherical shape of cenospheres.

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