Study of SiO\textsubscript{2} nanopowder produced by plasma arc method

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Abstract. The work is devoted to the study of the plasma arc method of obtaining nanosized silicon oxide from natural raw materials from Russian fields, such as diatomite of Kamyshlovsk deposit (Sverdlovsk region), the quartzite of Chupinsk deposit (Republic of Karelia), quartz sand of Tugansk deposit (Tomsk region). The method of transmission electron microscopy was used to study the morphology. Brunauer-Emmett-Teller method was used to study the surface. The main structural and morphological characteristics of the obtained nanoparticles were determined. The particles of nanoscale SiO\textsubscript{2} obtained by the plasma arc method spherical shape, the size distribution 10–300 nm, the specific surface area 37–71 m\textsuperscript{2}\textperg.

1. Introduction

Today, one of the urgent tasks of the nuclear fuel cycle is the immobilization of liquid radioactive waste of different chemical composition and activity [1–6]. Technologies for their safe and long-term storage are being developed, among which the possibility of using nanoscale materials to form a ceramic matrix containing radionuclides is being studied. As such a matrix can be used, for example, SiO\textsubscript{2} nanopowder [7]. In this regard, demand for new methods of obtaining it.

This paper studies the structural and morphological characteristics of silicon dioxide powder produced by the plasma-arc method [8], which makes it possible to use available and environmentally friendly natural high-silica materials as raw materials for SiO\textsubscript{2} nanopowder production. This method is based on physical processes of melting and evaporation of raw materials under the action of plasma energy of an electric arc discharge, followed by condensation of the gas phase in the form of nanoscale particles. The aim of the work was to study the structural and morphological characteristics of nano-sized silicon dioxide, obtained by the plasma arc method from high-silica natural raw materials.

2. Materials and methods

Available high-silica materials of natural origin from Russian deposits (quartzite from the Chupinsk deposit in the Republic of Karelia – 99.96 wt.% SiO\textsubscript{2}, enriched quartz sand from the Tugansk deposit in the Tomsk region – 98.15 wt.% SiO\textsubscript{2}, diatomite of the Kamyshlovsk deposit in the Sverdlovsk region –
80.40 wt.% SiO₂) were used as raw materials for the obtaining of SiO₂. Some impurities of Al₂O₃, Fe₂O₃, CaO, MgO, R₂O are contained in raw due to natural origin of materials.

To obtain a SiO₂ nanopowder, a unique atmospheric pressure DC arc plasma installation (figure 1) developed at the Department of Applied Mechanics and Materials Science at the Tomsk State University of Architecture and Building was used.

![Scheme of plasma unit for SiO₂ production.](image)

The principle of operation of the installation is based on the physical processes of melting and evaporation of raw materials with the subsequent condensation of the target nanopowder from the gas phase.

The thermal characteristic of plasma unit are shown in the table 1.

| N  | Power (kW) | Heat Flux Rate (MW·m⁻²) | Arc temperature (K) |
|----|------------|--------------------------|---------------------|
| 1  | 24±0.5     | 1.0±5×10⁻⁴               | 2800±30             |
| 2  | 37±0.6     | 1.5±6×10⁻⁴               | 3400±32             |
| 3  | 56±0.8     | 2.5±8×10⁻⁴               | 4600±40             |
| 4  | 59±0.7     | 3.0±7×10⁻⁴               | 4900±45             |

The second mode (Power 37 kW, Heat Flux Rate 1.5 MW·m⁻²) was used in this research to avoid impurities in the product [9].

The obtained powders were investigated to determine the main structural and morphological characteristics [10, 11].

Shape and size of the particles were characterized by Transmission Electron Microscopy method by using CM 12 microscope (Philips, The Netherlands), 120 kV. Samples were prepared by dispersion of the powders in ethanol using ultrasonic bath. Then the colloids obtained were placed by drops onto copper grid with carbon thin film and dried to remove the liquid.

Particles size distributions were obtained using iTEM Software (Olympus, Japan). Diagrams were constructed for the grain-size distribution of not less than 1000 nanoparticles.

A Brunauer-Emmett-Teller (BET) surface area analysis [12] of the nanopowder was performed on the Quantachrome Instruments NOVA 2200 surface area analyzer (USA). Nitrogen was used as the adsorbate
gas. The bath temperature is 77 K. The measurement was carried out using a 5 point method. For the analysis of the selected sample weighing 0.5 g.

3. Results and discussion

TEM images of nano-sized silica particles and their size distribution diagrams are presented in figures 2–4.

![Figure 2](image-url)

**Figure 2.** TEM images and distribution diagrams of silica nanoparticles obtained by the plasma arc method from quartzite (Chupinsk deposit, Russia).

The size distribution of SiO$_2$ nanoparticles obtained by plasma processing of quartzite is close to normal. The nanopowder contains particles with sizes of 10–300 nm, most of them (94–96 %) are in the range of up to 100 nm, 73–75% of particles are in the range of 11–50 nm, 18–20% of particles are in the range of 51–100 nm. The distribution peak (26–28%) accounts for particle sizes of 21–30 nm, and the average particle size is 30–40 nm.

Nanoparticles of SiO$_2$ powder obtained by the plasma arc method from diatomite have a size distribution in the range of 10–300 nm, the largest number of them (80–84%) have sizes up to 100 nm, there are 35–40% of particles in the range of 11–40 nm, 40–44% of particles are in the range of 41–100 nm. The peak of the distribution (12–14%) falls on particles 11–20 nm in size. In this case, a more uniform size distribution is observed – for every 10 nm, the proportion of particles of this size decreases on average by 1–5 %. The average particle size is 60–70 nm.

So, according to the images obtained by the method of transmission electron microscopy, it was established that the particles of the spherical samples under study are polydisperse, and their diameter lies in the size range from 10 to 300 nm. The images show significant agglomeration of nanoparticles.

Investigation of the specific surface of the samples of the obtained nanoparticles by the BET method showed that the specific surface of the products ranges from 37 to 71 m$^2$g$^{-1}$. These data are consistent with the established particle size distributions according to TEM data.
Figure 3. TEM images and distribution diagrams of silica nanoparticles obtained by the plasma arc method from enriched quartz sand (Tugansk deposit, Russia).

The SiO$_2$ nanopowder obtained by plasma processing of quartz sand consists of particles in the size range from 10 to 200 nm, most of them (93–95%) are in the range up to 100 nm. 76–78% of particles are in the range of 11–50 nm, and 15–19% of particles are in the range are in the range of 51–100 nm. The distribution peak (30–34%) accounts for particle sizes of 11–20 nm, the average particle size is 30–40 nm.

Figure 4. TEM images and distribution diagrams of silica nanoparticles obtained by the plasma arc method from diatomite (Kamyshlovsk deposit, Russia)

4. Conclusion
It was established that the particles of nanoscale SiO$_2$ obtained by the plasma arc method have the following structural and morphological characteristics: the shape of the particles is spherical, the size distribution is in the range from 10 to 300 nm, the specific surface area is from 37 to 71 m$^2$.g$^{-1}$. 
It was found that the content of 0.04–2% impurities in natural high-silica material does not lead to an increase in the size of nanoparticles of average size (30–40 nm). An increase of impurities up to 20% in natural high-silica material leads to an increase in the average particle size to 60–70 nm. This can be explained by the fact that the presence of impurities in raw expands the temperature range and lowers the boundary value of temperature compared to pure substances. Consequently, the processes of evaporation and condensation occur faster and cause a more intensive growth of SiO₂ nanoparticles per unit of time.

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