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Plasmadynamic synthesis in the Si-C-N-O system

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Abstract. The present work is aimed at studying the possibility of synthesizing nanodispersed particles in the Si-C-N-O system and then studying the resulting synthesis products. Three series of experiments were carried out under different atmospheres of the reactor chamber: air, air + argon, argon. The possibility of synthesis of particles of the ternary Si-C-N system was considered. In the course of the study, it was found that, in the air atmosphere of the reactor chamber, the production of silicon carbonitride particles is not feasible due to the oxidation of precursor phases. The result was the production of nanodispersed particles of cubic silicon carbide, which were obtained by the method of plasmadynamic synthesis in a hyper-velocity jet silicon-carbon plasma. The obtained products were subjected to a thermal analysis. During the thermal analysis, the most optimal annealing temperature range was found to be 600-700 ˚C. Annealing at this temperature in air allows the synthesis product to be eliminated from the unreacted carbon phase.

1. Introduction

In the last decade, the problem of obtaining materials of a new generation is urgent. The development of the industry is increasing demand for materials with high properties. One of the materials that corresponds modern production's needs is silicon carbide. High refractoriness, chemical resistance and high hardness make it possible to use silicon carbide in the production of abrasive materials [1-3]. A wide-bandgap along with high operating temperatures, gives silicon carbide great prospects in power electronics [4, 5]. Silicon carbide ceramics have remarkable thermal properties, which makes it possible to use SiC for production of refractories with high thermal stability and strength [6, 7]. The presence of such characteristics stimulates the development of scientific directions related to the production of silicon carbide. The problem of synthesizing nanodispersed materials has particular importance in recent years. This is because the substance with nanosized particles is capable of exhibiting unique combinations of mechanical, thermal, electrical, and other properties, different from the substance consisting of coarse-grained particles [8, 9]. Another important developing scientific direction is the synthesis in the ternary system. The Si-C-N system has some advantages with binary Si-C system: mechanical strength and high hardness, resistance to chemical influences, and high thermal conductivity [10]. Presumably, such properties are acquired due to the presence of a bond between all three atoms of the system.

There are many ways to produce nanopowders, but they are inefficient due to a high cost of used precursors, large grain size, long process time, etc. [11-17]. The method of direct dynamic synthesis in a hypersonic silicon-carbon plasma jet is used in the work. Generation of the plasma jet is implemented by a coaxial magnetoplasma accelerator (CMPA) with graphite electrodes [18].

2. Experimental

In the present work, influence of the composition of the gas atmosphere on nanosized silicon carbide powder are carried out. The possibility of synthesis silicon carnitride particles is also considered, so series of experiments with the air atmosphere of the reactor chamber are conducted.

The series of experiments were conducted with a different gas atmosphere of the reactor-chamber (series 1 – air, series 2 – argon + air, series 3 – argon). All series of experiments were carried out with the expiration of the plasma jet in free space without barriers. Power to the accelerator was supplied from a capacitive energy storage with a charge energy U=3 kV and a capacity C=6 mF.

The precursors in experiments were carbon black and silicon powder, which were mixed in ratio Si:C = 3:1, because stoichiometric equilibrium is established at a given precursor ratio [19]. Then mixture was placed into plasma formation zone at the beginning of the accelerating channel. The results of experiments were the production of powdered products, which were studied by X-ray diffractometry (Shimadzu XRD 6000 (CuKα – radiation)) and transmission electron microscopy (Philips CM 12).

3. Results and Discussion

3.1. Influence of the reactor chamber's gas atmosphere

Diffractions were analyzed in the software package PowderCell 2.4 using the PDF4 + database. The X-ray diffractions are presented in figure 1. The results of quantitative XRD analysis are presented in table 1. The product obtained in an air atmosphere consists of only silicon oxide (space group-F4-1 3 2 {210}), which gives one amorphous reflex on the X-ray diffraction. In experiments with an atmosphere of air and argon: the product includes the phase of cubic silicon carbide (β-SiC) and cubic silicon in addition to the predominant oxygen oxide. The product obtained in an argon atmosphere is characterized by a high content of β-SiC phase (over 90%).

Table 1. The results of quantitative XRD analysis.

Gas atmosphere of the reactor chamber	Phase composition (mass%)			
	SiC	Si	C	SiO_2
Air	-	-	-	100
Air and argon	12.2	11.8	-	76
Argon	92.6	4.9	2.5	-

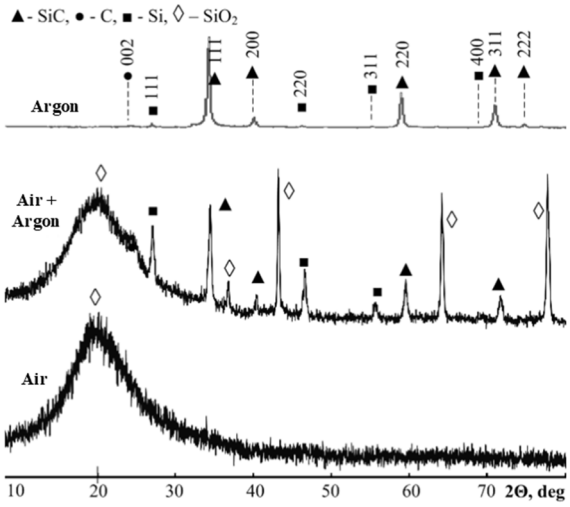

Figure 1. Diffraction patterns of synthesized powders.

The results of XRD analysis are confirmed by the results of transmission electron microscopy. Figure 2 shows the accumulation of particles of the product obtained when the plasma jet flows into air reactor chamber's atmosphere. In this case, only silicon oxide particles, which have a spherical shape characteristic of the amorphous material, are formed. When a plasma jet flows into a mixture of air and argon, there are particles of silicon carbide in addition to the spherical particles of silicon dioxide.

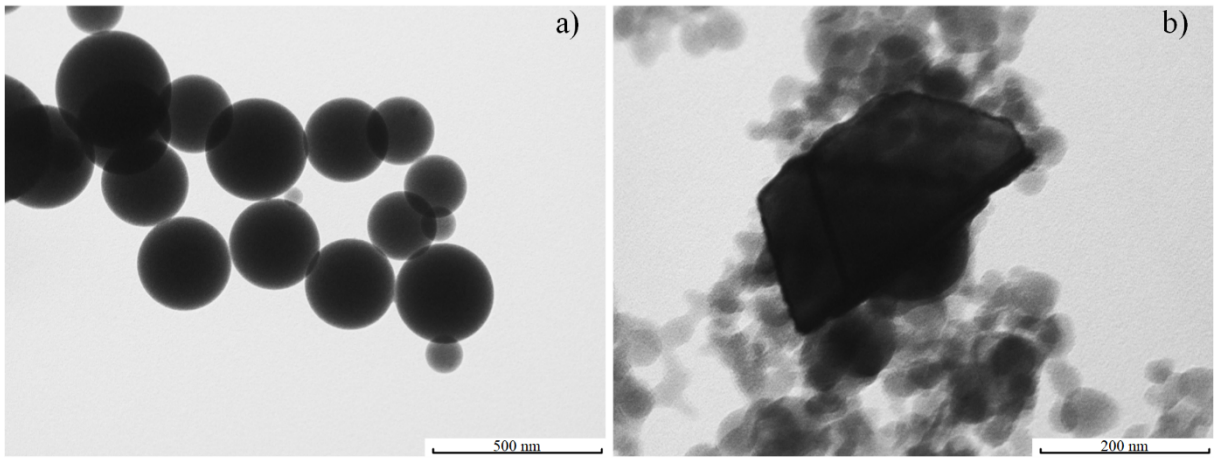

Figure 2. TEM-images of the product, obtained in series 1 (a) and series 2 (b).

3.2. Thermal analysis of the synthesized product

The series of experiments were carried out to investigate the possibility of purification. The studies consisted of thermogravimetric analysis (TGA) by synchronous thermal analyzer SDT Q600. Due to the large content of silicon carbide, the products obtained in the third series of experiments were investigated. The analysis was carried out in the temperature range from 45 to 1200°C in the air. The thermogravimetric (TG) curves shown in figure 3 were constructed using experimental data.

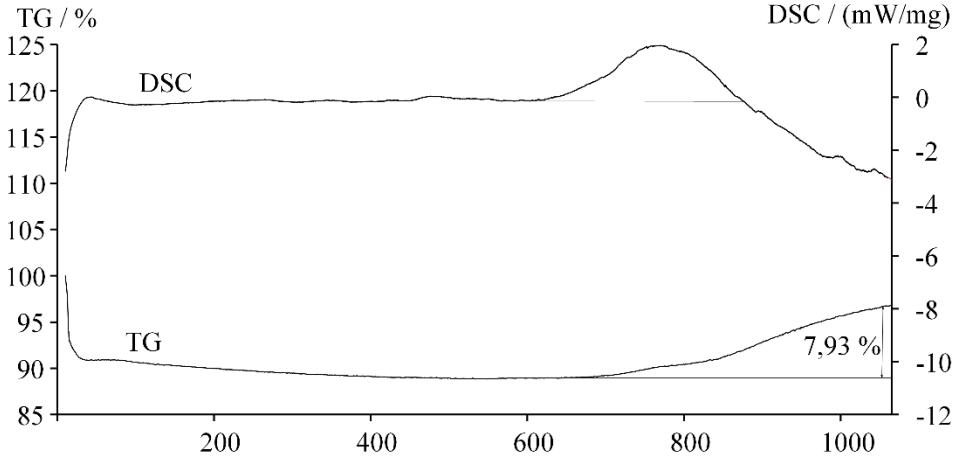

Figure 3. TGA curves.

When the temperature reaches 600°C, an exothermic reaction occurs, which is revealed by the increase in the intensity of the DSC curve (differential scanning calorimetry). Together with DSC, TG slight increases. This is due to the fact that there is a reaction of carbon-oxygen interaction with the formation of carbon oxide (IV) in the range of 600-700°C. Heating up to this temperature range allows

achieving complete elimination of the carbon phase, which is confirmed by X-ray diffraction data. A further increase in temperature leads to a reaction between silicon and oxygen. The resulting silicon dioxide is visible on the X-ray diffractions obtained in the form of an amorphous peak (figure 4). The growth of the TG curve also characterizes the appearance of silicon dioxide, which precipitates, thereby increasing the mass.

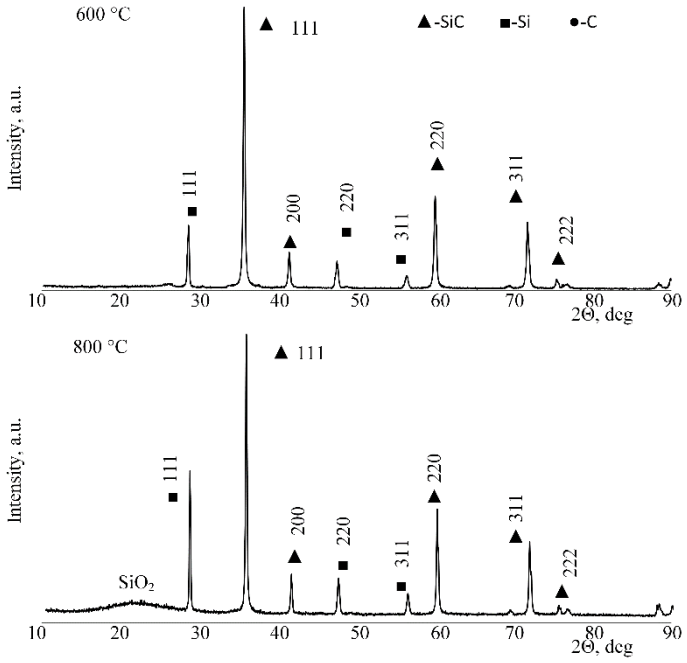

Figure 4. X-ray diffractions from annealing.

4. Conclusions

By the method of direct plasmadynamic synthesis the possibility of synthesizing the products of the Si-C-N-O system was investigated. Nanodispersed products which contains of cubic silicon carbide were obtained. The possibility of synthesis of the Si-C-N triple system's particles was studied. It was found precursor phases of carbon and silicon are oxidized, when the plasma jet flows into the air atmosphere of the reactor chamber, so that the particles of both silicon carbide and silicon carbonitride are not formed in the head shock of the plasma jet. Thermal analysis of powdered products showed that when the product is annealed in the temperature range 600-700°C, the reaction of precursor carbon with oxygen from the air takes place. In view of this, the nanodispersed powder is purified from the carbon phase.

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