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Synthesis of silicon carbide nanopowders in free flowing plasma jet with different energy levels

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Abstract. Silicon carbide (SiC) nanopowders were produced by the synthesis in an electrodischarge plasma jet generated by a high-current pulsed coaxial magnetoplasma accelerator. The present work focuses on the experiments where the obtained hypersonic plasma jet flew into space of the reactor chamber without impact on a target. The energy level of experiments was changed from ~10.0 to ~30.0 kJ. Four experiments were carried out at different energy levels. The powder products synthesized by the plasmadynamic method were studied by such well-known methods: X-ray diffraction (XRD), transmission electron microscopy (TEM). All the powders mainly contain cubic silicon carbide (β-SiC) particles with clear crystal structures and triangular shapes. SiC content reaches its maximum value 95% at the energy level 21.0 kJ, then SiC content is decreased to 70% the energy level 27.8 kJ. The powder crystallites in different experiments have approximately the same average crystallite size because quasistationary time, which allows growing powder crystallites, is absent.

1. Introduction

Silicon carbide (SiC) is one of the most important non-oxide materials and it is a very attractive material for a wide range of industrial applications. This compound has many excellent physical and chemical properties due to strong covalent bonding: superhardness, low density, high mechanical strength, Young's modulus, thermal and corrosion resistance, radiation hardness [1–4].

Silicon carbide is a promising ceramics for various industrial applications even at temperatures above 1000 °C including externally stable ceramics, high temperature semiconductor devices, grinding powder, brake pads, modern nuclear reactors and etc. [5, 6].

Nanomaterials are known to have unique and higher properties than conventional materials. Various silicon carbide nanostructures are widely used as ceramics reinforcement, for creation of nanostructured ceramics, micro- and nanoelectromechanical systems (MEMS and NEMS) [7–9]. It is well known that carbothermal reaction, physical vapor transport (PVT), self-propagation high-temperature synthesis (SHS), chemical vapor deposition (CVD), sol-gel methods, gas-phase reaction method are the most commonly method used to synthesize SiC materials [10–13]. The cost, time and energy expenses are comparatively high in all above synthesized methods. There is a big problem to produce nanosized silicon carbide particles in large volumes.

Nano-SiC synthesis can be realized in the hypersonic plasma of 11–28 kJ. In this work, the synthesis of nanosized silicon carbide powders was implemented by plasmadynamic method. The possibility of SiC nanopowders production by plasmadynamic method was shown in [14, 15]. The aim of this work is to implement nano-SiC synthesis in a free flowing hypersonic plasma jet and to evaluate the effect of plasma energy on nanosized product characteristics.

2. Experimental Conditions

The plasmadynamic method has been already successfully applied for obtaining nanopowders of the Si-C system. This method is based on the use of a coaxial magnetoplasma accelerator invented in Tomsk Polytechnic University. This original installation was used to generate a plasma jet. The schematic diagram of the experimental system is shown in Figure 1. Only the type of a coaxial magnetoplasma accelerator with graphite electrodes [3] could be applied for obtaining silicon carbide nanopowders without different extraneous phases (not Si-C system) associated with electric erosion of electrodes. The fundamentals of the applied synthesis technique are discussed in detail for synthesis of Si-C phases in [14], [15]. The present work focuses on the experiments where the obtained hypersonic plasma jet flew into space of the reactor chamber. Such the experiments are significantly different from the previous works [15] where another form of the flowing plasma jet was used. These works include experiments with the impact of the plasma jet on a copper target installed at a distance of 23 mm from acceleration channel.

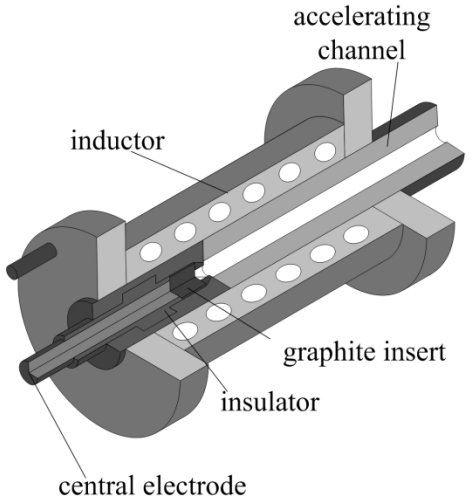

Figure 1. Schematic diagram of the experimental system based on a coaxial magnetoplasma accelerator.

Pulsed power supply of a magnetoplasma accelerator was carried out by a capacitive energy storage with the maximum stored energy W_{max} = 360 kJ. The present study includes the experiments which were carried out at the different levels of accumulated energy W_c and charging voltage U_{ch} from 2.0 kV to 3.5 kV. It was assumed that the variation of plasma jet energy should lead to changing main characteristics of the synthesized product such as the phase composition (percentage content of silicon carbide and attendant phase) and the product dispersion. The capacity of a capacitive energy storage remained constant C = 6 mF.

A mixture of solid precursors in the form of silicon (averaged sizes of particles ~50 μm) of and carbon (amorphous powder) in an approximately stoichiometric ratio was placed to a plasma formation zone. The discharge between the central electrode and the coaxial electrode (accelerated channel) lead to transition of a precursor mixture to a plasma jet. The generated plasma jet was accelerated by the magnetic field of an accelerating channel and an inductor (a solenoid). Table 1 shows the basic electrical and energy parameters of the experiments: operating current I_m, the voltage on the electrodes of magnetoplasma accelerator U_m, the discharge power P_m, the released energy W. The above values were determined by typical oscillograms of working current and voltage on electrodes obtained by a Tektronix TDS1012 oscilloscope. The space of the reactor chamber was filled with inert gas (argon).

The powder product synthesized by the plasmadynamic method was collected from the wall of the reactor chamber after precipitation of suspended particles. The collected powders were studied without any additional preparation by such well-known methods as X-ray diffraction (XRD) (Shimadzu XRD

6000 diffractometer, CuKα-radiation, λ = 0.15406 nm), transmission electron microscopy (TEM) (Philips CM 12 microscope). Qualitative X-ray analysis was performed using PowderCell 2.4 software package based on Rietveld method using PDF4+ database.

Table 1. The average size of nanoparticles and aggregates in suspensions.

Experiments	Electrical and energy parameters				
	U_c [kV]	I_m [kA]	U_m [kV]	P_m [MWt]	W [kJ]
1	2.0	2.5	3.0	3.5	27.8
2	65	76	98	111	21.0
3	12.0	18.8	27.0	36.8	18.8
4	10.5	13.0	19.0	29.7	12.0

3. Results and discussion

The results of the product study by X-ray diffractometry are shown in Figure 2 in the forms of the obtained X-ray diffraction patterns (for two experiment with the released energy $W_1 = 21$ kJ and $W_2 = 12$ kJ). According to the nature of peaks, the product consists of several crystalline phases. The full-profile phase and structural analysis of the X-ray diffraction spectrums of the products are presented in Table 2. All the products include phases corresponding to the following structural models: cubic silicon carbide β-SiC, space group (SPGR) – F-43m {216}; cubic silicon cSi, SPGR – F-43/d-32/m {227}; graphite gC, SPGR – P6-3mc {186}.

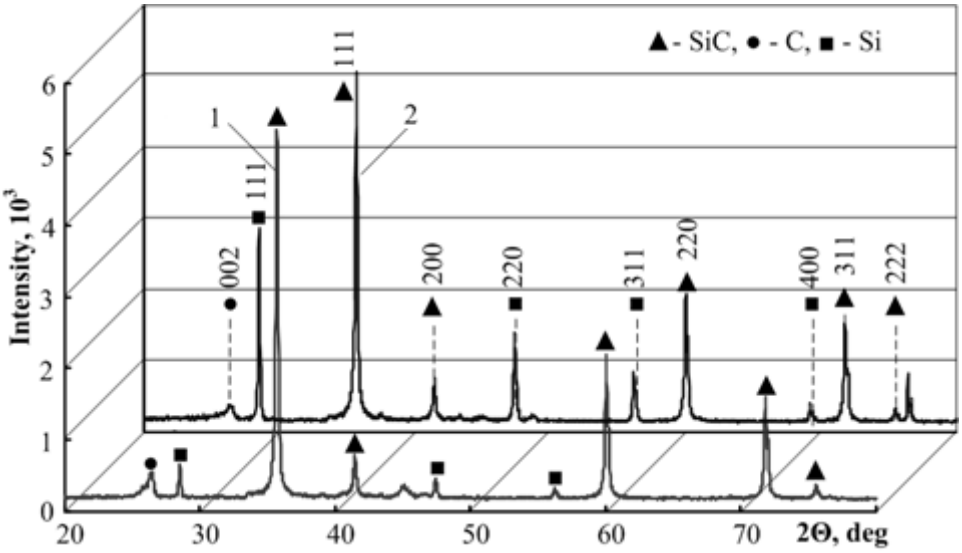

Figure 2. Diffraction patterns of the synthesized powders.

In general, phase composition of the products which were synthesized in the experiments with free flowing plasma jet is similar to the powders obtained by impact of the plasma jet on a copper target [4]. Comparison of the quantitative analysis results allows saying that there is an approximate equality of the silicon carbide content in the products synthesized at the same energy levels. However another tendency appears in relation to the effect of plasma jet energy on phase composition. SiC content reaches its maximum value (ω = 95%) at the energy level W = 21 kJ, then SiC content is decreased down to ω = 70% the energy level W = 27.8 kJ. This tendency can be explained by increase of pT-parameters due to increase of the energy level and increasing wear of the surface of a carbon accelerating channel at the greatest energy. Contamination of the final product by eroded carbon is confirmed by the calculated data presented in Table 2: the main impurity in the product of the experiment at the energy level W= 27.8 kJ is carbon phase, graphite. Increase of the energy level leads to changing the ratio of the silicon and carbon impurities to increase the amount of carbon.

Table 2. The full-profile phase and structural analysis of the X-ray diffraction spectrums of the products.

Phase *W* [kJ]		SiC	Si	C
1. 12.0	Content [%]	65,0	19.0	16.6
	CSA [nm]	60	40	20
2. 18.8	Content [%]	80.0	6.6	15.7
	CSA [nm]	60	35	10
3. 21.0	Content [%]	95.0	0.6	6.3
	CSA [nm]	50	40	10
4. 27.8	Content [%]	70.0	4.0	24.5
	CSA [nm]	70	80	10

A slight change of the main phase of coherent scattering areas (CSA) within 20 nm should be noted. This phenomenon is related to the form of a flow of hypersonic plasma jet from an accelerating channel. When a plasma jet impacts a copper target, pT-parameters increase in the shock wave induced in the vicinity of the copper target, so the size of silicon carbide crystallites increase. When a plasma jet flows into a reactor chamber freely without any impact on a target, a quasistationary time which allows growing powder crystallites is absent. In the experiments of the last case, powder crystallites have approximately the same average crystallite size, characterized by the value of CSA.

The results of the study by X-ray diffraction are confirmed and complemented by the results of the analysis by transmission electron microscopy (TEM) presented in Figure 3. Figure 3a shows the general bright-field TEM-image typical for of the experiments carried out by free flowing plasma jet. The present image includes the accumulation of crystalline particles. Significant quantity of typical triangular shaped particles in are presented in Figure 3a. The intensive Debye rings corresponding to cubic silicon carbide proves the predominant content of this phase in the product. The selected area electron diffraction (SAED) is presented in Figure 3b. The most striking and the main diffraction reflexes relate to reflecting planes of cubic silicon carbide. Dark-field TEM-image (Figure 3c) was obtained at the diffracted light beam on the packets plane SiC (111). The glow of reflecting planes of particles can be seen. It proves the particles of this fraction relate to silicon carbide phase.

4. Conclusions

Silicon carbide (SiC) nanopowders were produced by the synthesis implemented in an electrodischarge plasma jet generated by a high-current pulsed coaxial magnetoplasma accelerator. The present work focuses on the experiments where the obtained hypersonic plasma jet flew into space of the reactor chamber without impact on a target. The energy level of experiments was changed from ~10.0 to ~30.0 kJ. Four experiments were carried out at different energy levels. The powder products synthesized by the plasmadynamic method were studied by such well-known methods as X-ray diffraction (XRD), transmission electron microscopy (TEM). All the powders mainly contain cubic silicon carbide (β-SiC) particles with clear crystal structures and triangular shapes. SiC content reaches its maximum value 95% at the energy level of 21.0 kJ, then SiC content is decreased to 70% at the energy level of 27.8 kJ. The powder crystallites in different experiments have approximately the same average crystallite size because quasi-stationary time—which allows growing powder crystallites—is absent.

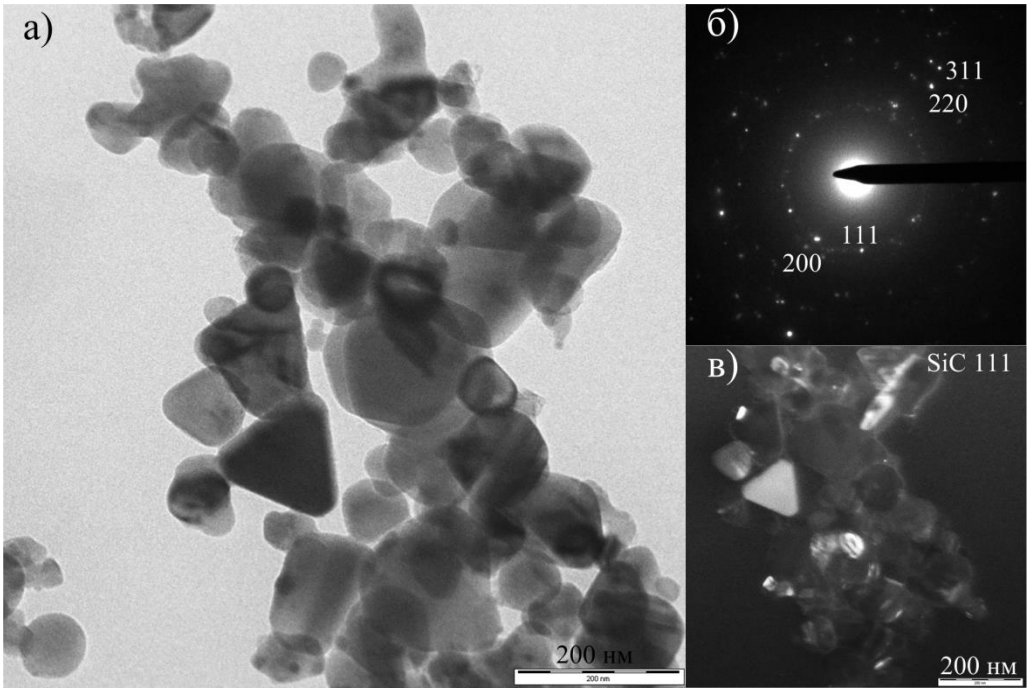

Figure 3. Bright-field TEM-images of the plasmadynamic synthesis product (W = 29.7 kJ).

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Acknowledgments

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