# Self-Propagating High-Temperature Synthesis of Silicon Carbide and Silicon Nitride Nanopowders Composition using Sodium Azide and Halides

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#### Abstract

Regularities of self-propagating high-temperature synthesis (SHS) or combustion synthesis (CS) by using "silicon - sodium azide - ammonium hexafluorosilicate - carbon - aluminum" powder mixture in the nitrogen atmosphere were investigated. The thermodynamic analysis of the combustion synthesis was performed. Experimental investigation of the combustion process: the measurement of linear rates of the combustion front propagation and the maximum combustion temperatures was conducted in a laboratory reactor with working volume 4.5 liters. The influence of the components ratio in the initial mixture on the combustion temperature, combustion rate and composition of reaction product was studied. The phase composition of the product synthesized was determined with an X-ray diffractometer. It was disclosed that the SHS product consists of the composition (mixture) of silicon carbide nanopowder with silicon nitride whiskers and a final halide. Investigation of surface topography and morphology of the product particles was carried out with a scanning electron microscope. Optimal mixture for the synthesis of nanoscale composition based on silicon carbide was determined: " $^{14}Si+6NaN_{3}+(NH_{4})_{2}SiF_{6}+15C+Al$ ". In this case, the SHS product consists of four phases: silicon carbide ( $\beta$ -SiC) – 48.57 wt.%,  $\alpha$ -silicon nitride ( $\alpha$ -Si<sub>3</sub>N<sub>4</sub>) – 27.04 wt.%,  $\beta$ -silicon nitride ( $\beta$ -Si<sub>3</sub>N<sub>4</sub>) - 5.83 wt.%, and sodium hexafluoroaluminate (Na<sub>3</sub>AlF<sub>6</sub>) - 18.56 wt.%. The average particle size of the composition was in the range of 70–130 nm. It was shown that the composition of the silicon carbide with silicon nitride and the final halide Na<sub>3</sub>AlF<sub>6</sub> playing a role a flux can be used as a modifier of castable aluminum alloys and as a reinforcing phase of aluminomatrix composites.

### Introduction

Carbide compounds are generally known as very hard ceramics with outstanding chemical properties. Silicon carbide is one of the non-oxide ceramics which has various industrial applications in the form of powder, whisker etc. High melting point, high thermal conductivity, high oxidation resistance, high mechanical strength and good chemical properties are some of its most important characteristics [1]. In particular, SiC particles are widely used as reinforcing phase in aluminomatrix composites [2]. In order to elaborate nanostructured ceramics like SiC monolith, different techniques are required to produce the starting SiC nanomaterials such as plasma, combustion, CVD-based methods, etc. These uncommon methods are expensive and arduous because they require a thorough control of complex processes. It is assumed that ideal starting powder should be very small particle sized (<100 nm) associated with a narrow size range and a high purity. Both  $Si_3N_4$  and SiC themselves exhibit good chemical stability and high temperature shock resistance [3].  $Si_3N_4$ /SiC composites also possess very desirable properties such as high hardness, high strength and high fracture toughness.

The well-known process of self-propagating high-temperature synthesis (SHS) or combustion synthesis (CS), discovered by A.G. Merzhanov, I.P. Borovinskaya, and V.M. Shkiro, provides considerable possibilities to obtain nanopowders of variable high-melting compounds [4, 5]. Azide technology of SHS designated as SHS-Az applying sodium azide NaN<sub>3</sub> as solid nitriding agent instead of gaseous nitrogen allows us to produce a wide range of ceramic powders of nitrides and composition on their basis including Si<sub>3</sub>N<sub>4</sub>–SiC [6–9]. The use of halides, low combustion temperature and presence of gaseous and condensed by-products inhibit the recrystallization and agglomeration of product grains and make

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it possible to obtain weakly sintered nanopowders and nanofibers of the SHS-Az products. This report presents results of attempts to synthesize nanopowders of silicon carbide SiC through silicon nitride  $Si_3N_4$  in a manner like [10, 11] but using sodium azide and halides. In [11] the SHS was used for the synthesis of high-dispersed silicon carbide powder and the "silicon nitride – silicon carbide" powders composition by burning in a nitrogen atmosphere the powders mixture of silicon and carbon. During the synthesis, a mixture of very small particles with large particles of the product was obtained.

#### **Materials and Methods**

The composition for obtaining of silicon nitride by the SHS-Az technology is known: " $14Si+6NaN_3+(NH_4)_2SiF_6$ " [7]. For the synthesis of silicon carbide through silicon nitride, it is convenient to use the following equation [8]:

$$14\text{Si} + 6\text{NaN}_3 + (\text{NH}_4)_2\text{SiF}_6 + y\text{C} = y\text{SiC} + 6\text{NaF} + 4\text{H}_2 + \frac{(15-y)}{3}\text{Si}_3\text{N}_4 + \frac{2y}{3}\text{N}_2.$$
(1)

It is known that this initial mixture allows us to synthesize "silicon carbide – silicon nitride" composition with a molar ratio of phases 50/50 [8]. To obtain silicon carbide only, it is proposed to take the excess of carbon. In this case, the equation will look as follows:

$$19Si + 6NaN_3 + (NH_4)_2SiF_6 + 20C = 20SiC + 6NaF + + 10N_2 + 4H_2.$$
(2)

To reduce the probability of occurrence of the silicon nitride in the reaction products and improve the energy of mixture for silicon carbide obtaining, it is proposed to add aluminum and reduce the amount of sodium azide in the initial mixture. Then equations (1) and (2) can be written as:

 $14Si + 3NaN_3 + (NH_4)_2SiF_6 + 15C + Al = 15SiC +$  $+ Na_3AlF_6 + 5N_2 + 4H_2;$ (3)

 $19Si + 3NaN_3 + (NH_4)_2SiF_6 + 20C + Al = 20SiC +$  $+ Na_3AlF_6 + 5N_2 + 4H_2.$ (4)

To establish the possibility of initial components mixture to burn, thermodynamic calculations using the computer program "Thermo" developed at the Institute of Structural Macrokinetics and Materials Science RAS (Chernogolovka, Moscow region) were performed.

Experimental investigation of the combustion process: the measurement of linear rates of the combustion front propagation and the maximum combustion temperatures was conducted in a laboratory reactor with working volume 4.5 liters. The inner diameter of the reactor space is 0.147 m, the height is 0.255 m. The methodology of synthesis, measurement of linear velocities and the maximum combustion temperatures are described in detail in [5-9].

The phase composition of the product synthesized was determined by "ARL X'TRA" X-ray diffractometer. Shooting of the X-ray spectrum were performed using Cu-radiation with a continuous scanning in the interval of  $2\theta$  angles from 20 to 80 degrees at a step of 2 deg./min [12]. The qualitative phase analysis was performed by comparing the set of experimental interplanar distances d/n with barcode radiographs database of the International Centre for Diffraction Data ICDD PDF2. The quantitative phase analysis was performed by the Rietveld method with the program PDXL 1.8.1.0 using open crystallographic database (COD).

Investigation of surface topography and morphology of the product particles were conducted by the "Jeol JSM-6390A" scanning electron microscope with the "Jeol JED-2200" attachment.

## **Results and Discussions**

Figures 1 and 2 show results of the thermodynamic calculations: adiabatic temperature, formation enthalpy and combustion product composition for " $14Si+6NaN_3+(NH_4)_2SiF_6+yC$ " system with different carbon content.



Fig. 1. Dependence of the adiabatic temperature and enthalpy of the "14Si–6NaN<sub>3</sub>–(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>– yC" system from the carbon amount.

Increasing the carbon content in the mixture from 1 to 15 moles leads to a decrease of 1280 K in the adiabatic reaction temperature. If the carbon content is 1 mole, the reaction products are presumably contain silicon carbide, silicon nitride, sodium fluoride, nitrogen, hydrogen, and free silicon. With the carbon content increasing up to 15 moles, the synthesis products are silicon carbide, sodium fluoride and gaseous nitrogen and hydrogen.

Thermodynamic calculations allow us to suggest that " $14Si+6NaN_3+(NH_4)_2SiF_6+15C$ " system is optimal for the synthesis of silicon carbide because the combustion of this system brings about the formation of the target silicon carbide and gaseous by-products: nitrogen, hydrogen and sodium fluoride, which are easily removed and do not pollute the final product. Adiabatic combustion temperature and the products formation enthalpy are sufficient for the formation of silicon carbide in the process of self-propagating high-temperature synthesis using sodium azide.



Fig. 2. Dependence of the combustion products composition of the " $14Si-6NaN_3-(NH_4)_2SiF_6-yC$ " from the carbon amount.

Table 1 shows the results of thermodynamic analysis of the combustion of the " $19Si+6NaN_3+$  ( $NH_4$ )<sub>2</sub>SiF<sub>6</sub>+20C" mixture. The table shows that the silicon carbide (SiC) and sodium fluoride (NaF) form as a result of the combustion. The temperature and reaction enthalpy are sufficient for the application of the SHS process.

Table 1Results of thermodynamic analysis of the" $19Si+6NaN_3+(NH_4)_2SiF_6+20C$ " mixture combustion

Parameter	Value	
Volume of gas products, l	46.41	
Pressure of gas products, atm.	40.00	
Temperature, K	1563.24	
Gas products amount, mole	14.01	
Products enthalpy, kJ	-2554.26	
H <sub>2</sub> , mole	4.00	
N <sub>2</sub> , mole	10.00	
NaF, mole	6.00	
SiC, mole	20.00	

The results of the investigation of dependence of the combustion temperature and front velocity from the carbon content in the "14Si+6NaN<sub>3</sub>+  $(NH_4)_2SiF_6+yC$ " system are presented in Table 2. The carbon content y = 15 moles is stoichiometric. With increasing of carbon content in the initial mixture, the combustion temperature decreases, which agrees with the results of the thermodynamic calculations. The combustion front velocity decreases as well. The results of X-ray phase analysis shows that the maximum yield of silicon carbide is observed in the system with 15 moles of carbon, that is consistent with the results of thermodynamic analysis. But the free silicon is found in combustion products of the "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+15C" mixture. To increase the content of desired product of SiC and remove the unreacted silicon, the experiments were conducted, in which carbon was taken in excess: y > 15 moles. The increase of carbon in the mixture leads to decrease in the combustion temperature and reaction rate. At the content of carbon y = 22 moles, the combustion becomes unstable, the sample becomes not completely burned.

#### Table 2

Dependence of the combustion temperature and front velocity of the "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+yC" system from the carbon amount (y)

Carbon amount, y, mole	Combustion temperature, °C	Front velocity, cm/s
5	1850	0.83
10	1700	0.68
15	1650	0.52
16	1650	0.50
18	1600	0.20
20	1450	0.07
22	1150	0.03

To favour the combustion by an increase the energy value of the system, the experiments were performed with the addition of the aluminum powder in the amount of 5 and 10 wt.% in the "14Si+6NaN<sub>3</sub> +(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+15C" mixture. Combustion temperature and front velocity were 2050 °C and 0.6 cm/s, 1850 °C and 0.48 cm/s respectively.

The combustion parameters for the "14Si+3NaN<sub>3</sub>+  $(NH_4)_2SiF_6+15C+Al$ ", "19Si+3NaN<sub>3</sub>+ $(NH_4)_2SiF_6$ +5C", "19Si+6NaN<sub>3</sub>+ $(NH_4)_2SiF_6+20C$ ", "19Si+3NaN<sub>3</sub>+ $(NH_4)_2SiF_6+20C+Al$ " mixtures are presented in Table 3.

Figure 3 shows the results of X-ray phase analysis of the combustion product of the "14Si+6NaN<sub>3</sub> +  $(NH_4)_2SiF_6+5C$ " mixture. The product is composed of four phases:  $\alpha$ -silicon nitride ( $\alpha$ -Si<sub>3</sub>N<sub>4</sub>),  $\beta$ -silicon nitride ( $\beta$ -Si<sub>3</sub>N<sub>4</sub>), sodium fluoride (NaF), free silicon (Si). Based on the ratio of peak heights, it can be assumed for the content of the products: NaF > $\beta$ -Si<sub>3</sub>N4> $\alpha$ -Si<sub>3</sub>N<sub>4</sub>> Si.

 Table 3

 Dependence of the combustion temperature and front velocity from mixture composition

Mixture composition	Combustion	Front
	temperature,	velocity,
	°C	cm/s
14Si+3NaN <sub>3</sub> +(NH <sub>4</sub> ) <sub>2</sub> SiF <sub>6</sub> +15C+A1	1570	0.79
$19Si+3NaN_3+(NH_4)_2SiF_6+5C$	1720	0.82
19Si+6NaN <sub>3</sub> +(NH <sub>4</sub> ) <sub>2</sub> SiF <sub>6</sub> +20C	1690	0.74
19Si+3NaN <sub>3</sub> +(NH <sub>4</sub> )2SiF <sub>6</sub> +20C+A1	1750	0.87



Fig. 3. XRD results of the "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+ 5C" mixture combustion products:  $\bullet - \beta$ -Si<sub>3</sub>N<sub>4</sub>;  $\bullet -$  NaF;  $\bullet -$  Si;  $\bullet - \alpha$ -Si<sub>3</sub>N<sub>4</sub>.

Figure 4 shows the results of X-ray phase analysis of the combustion products of the "14Si+6NaN<sub>3</sub> +(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+10C" mixture. The products after a water washing consist of three phases:  $\alpha$ -Si<sub>3</sub>N<sub>4</sub>,  $\beta$ -Si<sub>3</sub>N<sub>4</sub> and  $\beta$ -SiC. The ratio of peak heights shows:  $\beta$ -Si<sub>3</sub>N<sub>4</sub>> $\alpha$ -Si<sub>3</sub>N<sub>4</sub>>SiC. An increase of 5C leads to SiC formation. The NaF is easy removed from the combustion products by water washing. Below the content of the combustion products is indicated without water washing.



Fig. 4. XRD results of the "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+ 10C" mixture combustion washed products:  $\blacklozenge -\beta$ -Si<sub>3</sub>N<sub>4</sub>;  $\blacklozenge -$ SiC;  $\blacktriangle -\alpha$ -Si<sub>3</sub>N<sub>4</sub>.

Figure 5 shows the results of X-ray phase analysis of the combustion products of the "14Si+6NaN<sub>3</sub> +(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+15C" mixture. The products are composed of four phases:  $\beta$ -SiC,  $\alpha$ -Si<sub>3</sub>N<sub>4</sub>, NaF, and Si, with NaF>Si>SiC> $\alpha$ -Si<sub>3</sub>N<sub>4</sub>. The increase of carbon content in the initial mixture leads to the increase of the silicon carbide content in the product.



Fig. 5. XRD results of the "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+ 15C" mixture combustion products:  $\bullet$  – SiC;  $\bullet$  – NaF;  $\bullet$  – Si;  $\blacktriangle$  –  $\alpha$ -Si<sub>3</sub>N<sub>4</sub>.

Figure 6 shows the results of X-ray phase analysis of the combustion products of the "19Si+3NaN<sub>3</sub> +(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+5C" mixture with an increased content of silicon: silicon nitride ( $\beta$ -Si<sub>3</sub>N<sub>4</sub>) – 67.3%, silicon nitride ( $\alpha$ -Si<sub>3</sub>N<sub>4</sub>) – 24.7%, silicon carbide ( $\beta$ -SiC) – 6.3 % and free silicon (Si) – 1.7%.



Fig. 6. XRD results of the " $19Si+3NaN_3+(NH_4)_2SiF_6$ +5C" mixture combustion washed products: -SiC;  $-\alpha-Si_3N_4$ ;  $-\beta-Si_3N_4$ ; -Si.

Figure 7 shows the results of X-ray phase analysis of the combustion products of the "19Si+6NaN<sub>3</sub> +(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+20C" mixture composed of four phases:  $\beta$ -SiC,  $\alpha$ -Si<sub>3</sub>N<sub>4</sub>,  $\beta$ -Si<sub>3</sub>N<sub>4</sub>, NaF.

Figure 8 shows the results of X-ray phase analysis of the combustion products of the "19Si+3NaN<sub>3</sub> +(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+20C+Al" mixture with an addition of aluminum. The products are composed of four phases: silicon nitride ( $\alpha$ -Si<sub>3</sub>N<sub>4</sub>) – 51%, silicon carbide ( $\beta$ -SiC) – 23%, silicon nitride ( $\beta$ -Si<sub>3</sub>N<sub>4</sub>) – 12.6%, and sodium hexafluoroaluminate (Na<sub>3</sub>AlF<sub>6</sub>)– 13%. In this case, we can see an increased content of  $\beta$ -SiC and formation of Na<sub>3</sub>AlF<sub>6</sub> instead of NaF in the combustion products.



Fig. 7. XRD results of the "19Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+ 20C" mixture combustion products:  $\bullet$  – SiC;  $\bullet$  – NaF;  $\blacktriangle$  –  $\alpha$ -Si<sub>3</sub>N<sub>4</sub>;  $\bullet$  –  $\beta$ -Si<sub>3</sub>N<sub>4</sub>.



Fig. 8. XRD results of the "19Si+3NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+ 20C+Al" mixture combustion products:  $\bullet$  – SiC;  $\blacktriangle$  –  $\alpha$ -Si<sub>3</sub>N<sub>4</sub>;  $\bullet$  –  $\beta$ -Si<sub>3</sub>N<sub>4</sub>;  $\bullet$  – Na<sub>3</sub>AlF<sub>6</sub>.

Figure 9 shows the results of X-ray phase analysis of the combustion products of the "14Si+6NaN<sub>3</sub> +(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+15C+Al" mixture. A method of corundum numbers with using of several single peaks allowed us to determine proportion of phases in the product: silicon carbide ( $\beta$ -SiC) – 48.6%, silicon nitride ( $\alpha$ -Si<sub>3</sub>N<sub>4</sub>) – 27.0%, silicon nitride ( $\beta$ -Si<sub>3</sub>N<sub>4</sub>)– 5.8%, sodium hexafluoroaluminate (Na<sub>3</sub>AlF<sub>6</sub>) – 18.6%. This content of  $\beta$ -SiC is largest for all initial mixtures investigated by us in this work. Free silicon was not revealed.



Fig. 9. XRD results of the "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub> +15C+Al" mixture combustion products:  $\bullet$  – SiC;  $\blacktriangle$  –  $\alpha$ -Si<sub>3</sub>N<sub>4</sub>;  $\bullet$  –  $\beta$ -Si<sub>3</sub>N<sub>4</sub>;  $\bullet$  – Na<sub>3</sub>AlF<sub>6</sub>.

Figure 10 shows the results of X-ray phase analysis of the combustion products of the "14Si+3NaN<sub>3</sub> +(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+15C+Al" mixture with a decreased content of sodium azide (NaN<sub>3</sub>). The products are composed of five phases:  $\beta$ -SiC,  $\alpha$ -Si<sub>3</sub>N<sub>4</sub>,  $\beta$ -Si<sub>3</sub>N<sub>4</sub>, Na<sub>3</sub>AlF<sub>6</sub>, and Si.



Figure 11 shows the surface topography and morphology of the powder particles synthesized from "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+5C" mixture. Combustion products are composed of threadlike crystals of silicon nitride with a diameter of 100–200 nm and particles of sodium fluoride of an irregular shape.



Fig. 11. Surface topography and morphology of the powder particles synthesized from " $14Si+6NaN_3+(NH_4)_2SiF_6$ + 5C" mixture: a) ×10000; b) ×20000.

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Figure 12 shows the surface topography and morphology of the powder particles synthesized from "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+15C" mixture. Combustion products are composed of spherical particles of silicon carbide with a diameter of about 150 nm, threadlike crystals of silicon nitride with a diameter of 100–300 nm and particles of sodium fluoride of an irregular shape.



Fig. 12. Surface topography and morphology of the powder particles synthesized from "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub> +15C" mixture: a)  $\times$ 5000; b)  $\times$ 10000.

Figure 13 shows the surface topography and morphology of the powder particles synthesized from "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+15C+AI" mixture. Combustion products are composed of spherical particles of silicon carbide with a diameter of 80–120 nm, threadlike crystals of silicon nitride with a diameter of about 100 nm and particles of sodium fluoride of an irregular shape.





Fig. 13. Surface topography and morphology of the powder particles synthesized from " $14Si+6NaN_3+(NH_4)_2SiF_6$ +15C+Al" mixture: a) ×1000; b) ×50000.

Figure 14 shows the surface topography and morphology of the powder particles synthesized from " $14Si+3NaN_3+(NH_4)_2SiF_6+15C+Al$ " mixture. Combustion products are composed of spherical particles of silicon carbide with a diameter of 70–100 nm, threadlike crystals of silicon nitride with a diameter of about 100 nm and particles of sodium hexafluoroaluminate of an irregular shape.



Fig. 14. Surface topography and morphology of the powder particles synthesized from " $14Si+3NaN_3+(NH_4)_2SiF_6$ +15C+Al" mixture: a) ×100; b) ×25000.

Figure 15 shows the surface topography and morphology of the composition (mixture) of particles of silicon nitride and silicon carbide synthesized from "19Si+3NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+5C" mixture. Combustion products are composed of spherical particles of silicon carbide with a diameter of about 100 nm and threadlike crystals of silicon nitride with a diameter of about 100 nm coated by sodium fluoride.



Fig. 15. Surface topography and morphology of the powder particles synthesized from " $19Si+3NaN_3+(NH_{4)2}SiF_6$ +5C" mixture: a) ×1500; b) ×10000.

Figure 16 shows the surface topography and morphology of the composition of particles based on silicon carbide synthesized from " $19Si+6NaN_3+$ ( $NH_4$ )<sub>2</sub>SiF<sub>6</sub>+20C" mixture. Combustion products are composed of spherical particles of silicon carbide with a diameter of about 100 nm and thread-like crystals of silicon nitride with a diameter of 100–130 nm coated by sodium fluoride.



Fig. 16. Surface topography and morphology of the powder particles synthesized from " $19Si+3NaN_3+(NH_4)_2SiF_6$ +20C" mixture: a) ×1000; b) ×25000.

Figure 17 shows the surface topography and morphology of the composition of particles based on silicon carbide synthesized from "19Si+3NaN<sub>3</sub>+ (NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+20C+AI" mixture. Combustion products are composed of spherical particles of silicon carbide with a diameter of 70–100 nm and threadlike crystals of silicon nitride with a diameter of about 100 nm coated by sodium hexafluoroaluminate.



Fig. 17. Surface topography and morphology of the powder particles synthesized from " $19Si+3NaN_3+(NH_4)_2SiF_6$ +20C+Al" mixture: a) ×5000; b) ×25000.

As we could see, the final product of the SHS-Az process with the largest content of  $\beta$ -SiC (in the case of the "14Si+6NaN<sub>3</sub>+(NH<sub>4</sub>)<sub>2</sub>SiF<sub>6</sub>+15C+Al" initial mixture) contains along with a target silicon carbide the following by-products: silicon nitride and sodium hexafluoroaluminate. The presence of Na<sub>3</sub>AlF<sub>6</sub> may be a positive factor when this SHS-Az product is used for example to modify the aluminum alloys, as Na<sub>3</sub>AlF<sub>6</sub> is well wetted and absorbed by aluminum melt and can facilitate the entry of the micro and nanopowders of SiC and AlN in the molten aluminum during obtaining aluminomatrix composites reinforced with particles of refractory compounds of AlN and SiC [13]. Sodium hexafluoroaluminate  $(Na_3AlF_6)$  plays the role of flux by the introduction of the reinforcing particles in the molten aluminum (as well as sodium fluoride). Our preliminary experiments have shown that the obtained compositions (mixtures) of nanopowders enter easily into molten aluminum and are distributed there, providing a good modifying effect by refining the grains of aluminum alloy and reducing its shrinkage during solidification. The composition based on silicon carbide ( $\beta$ -SiC – 48.6%,  $\alpha$ -Si<sub>3</sub>N<sub>4</sub>–27.0%,  $\beta$ -Si<sub>3</sub>N<sub>4</sub>–5.8%, Na<sub>3</sub>AlF<sub>6</sub>–18.6%) was used to obtain a modifier (master alloy) for aluminum alloys. The efficiency of the modifier was estimated by modifying of industrial aluminum alloy Al-6% Si-2% Cu (Russian brand AK6M2). Figure 18 shows the microstructure of Al-6%

Si-2% Cu alloy prior to and after modification. The modification of Al-6% Si-2% Cu alloy by Cu-10% SiC master alloy reduces the grain size of  $\alpha$ -Al dendrites by 2.5 times. The Brinnell hardness is increased by 20%, the percent elongation is increased by 3 times, the tensile strength is increased by 20% relative to properties of the non-modified alloy.



Fig. 18. Microstructure of Al-6% Si-2% Cu alloy prior to and after modification: a, b – prior to modification, c, d – after modification.

### Conclusions

Optimal mixture for the synthesis of nano scale composition based on silicon carbide is the "14Si+  $6NaN_3+(NH_4)_2SiF_6+15C+Al$ ". During combustion of this mixture reached the maximum content of silicon carbide ( $\beta$ -SiC- 48.6%) in the synthesized composition of silicon carbide nanopowder (spherical particles with a diameter of 70–130 nm), silicon nitride threadlike nanocrystals (a diameter of about 100 nm), and the flux of sodium hexafluoro-aluminate. This composition can be used as an effective inoculant in cast aluminum alloys and as a reinforcing phase in aluminomatrix composites.

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