PREPARATION OF Si_3N_4-SiC POWDER COMPOSITION USING THE SHS AZIDE TECHNOLOGY WITH POLYTETRAFLUOROETHYLENE*

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Ceramic compounds silicon nitride Si_3N_4 and silicon carbide SiC have unique physical, mechanical and chemical properties, due to which they are used in various fields, including as high-temperature materials for structural purposes. The combination of silicon nitride and carbide in the Si_3N_4—SiC composite material makes it possible to use the advantages of each of these single-phase ceramics, especially in the submicron and nanostructured state, and obtain composite ceramics with significantly improved properties. However, such composite materials have a high cost and are difficult to manufacture, which is due to the high cost of ceramic nanopowders and the practical impossibility of their homogeneous mechanical mixing. Therefore, chemical methods of direct synthesis of ceramic powders inside the desired composition materials from inexpensive reagents are preferred. Among the well—known methods for the synthesis of submicron and nanosized powder Si_3N_4-SiC compositions, the method of self-propagating high-temperature synthesis (SHS), based on the combustion of inexpensive starting reagents, stands out for its simplicity and energy efficiency [1]. The use of such a type of SHS as azide SHS using powders of silicon Si, carbon black C, sodium azide NaN_3 as a nitriding reagent and the halide salt NH_4F as an activating gasifying additive allowed us the preparation of highly dispersed Si_3N_4–SiC compositions with different ratios of nitride and carbide phases, differing from those, synthesized earlier using the combustion process, as more high dispersion and high content of the α-phase Si_3N_4 [1]. However, the experimental composition of the synthesized powder compositions differed markedly from the calculated theoretical composition with a significantly higher content of silicon nitride and a significantly lower content of silicon carbide, as well as the presence of an impurity of free silicon.

In this regard, in this work, the use of polytetrafluoroethylene C_2F_4 in the azide SHS method as the initial carbonizing and activating reagent instead of carbon black and NH_4F salt was investigated, because C_2F_4 can promote the formation of silicon carbide in large quantities, and in the form of nanofibers and nanoparticles [2, 3]. The initial reaction equations for the synthesis of target Si_3N_4–SiC compositions with a molar phase ratio from 1:4 to 4:1 had the following form:

$$8Si+4NaN_3+C_2F_4=2Si_3N_4+2SiC+10NaF+2N_2 \quad (1)$$
$$5Si+4NaN_3+C_2F_4=Si_3N_4+2SiC+4NaF+4N_2 \quad (2)$$
$$7Si+8NaN_3+2C_2F_4=Si_3N_4+4SiC+8NaF+10N_2 \quad (3)$$
$$14Si+4NaN_3+C_2F_4+N_2=4Si_3N_4+2SiC+4NaF+10N_2 \quad (4)$$

The results of thermodynamic calculations showed for these reactions, that the adiabatic temperatures (2770-2957 K) are high enough to realize a self-sustaining combustion regime, and the composition of reaction products corresponds to the right-hand sides of equations (1)-(4). The experimental study was carried out by burning the initial powder mixtures in bulk in a 4.5 liter reactor in an atmosphere of gaseous nitrogen with a pressure of 4 MPa. Combustion temperatures and velocities, structure and phase composition of combustion products were determined. The study showed that the synthesized compositions are agglomerates of powders from particles ranging in size from 140 to 290 nm, the phase composition of which includes impurities of free carbon from 4.5 to 13 wt.%, and the content of the SiC phase is large and close to the theoretical composition according to equations (1)–(4), in contrast to the results of [1].

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