An investigation of SHS products in titanium, carbon (black carbon) and aluminum powder mixtures.

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Abstract. Metal matrix composite powders "TiC-Al binder" were synthesized in the wave combustion mode and investigated. Combustion concentration limits of Ti-Al-C powder mixtures were founded. Phase and elemental composition of self-propagating high temperature synthesis (SHS) products, morphology of composite powders and the dependence of the carbide phase size in the structure of the composite on the reaction mixture composition were investigated. The SHS composite powders have a lumpy, predominantly equiaxed shape, favourable for good flowability. The carbide inclusions size in the aluminium matrix decreases monotonically with a content increase of the of thermally inert aluminium binder.

1. Introduction
Combustion synthesis products in titanium, carbon and aluminum powder mixtures in the wave combustion and thermal explosion modes have been studied in papers [1-7]. It has been established, that with high aluminum content in the reactive mixtures SHS products consist of two main phases: titanium carbide and unbound aluminum. The main aims in presented studies were to determine the phase composition of the synthesized products and the titanium carbide lattice parameter. The structure of the synthesized products, which can be used for the preparation of metal matrix composite powders, has not been studied.

"TiC-metal binder" composite powders [8, 9] have been successfully used for electron beam surfacing [10, 12] and plasma sputtering [11] of wear-resistant coatings. It is known, that phase composition and structure of composite powders (dispersity and morphology of carbide inclusions and volume fraction of metal binder) affect the properties of coatings.

The purposes of this work were: a) to determine the concentration limits of wave combustion mode synthesis in Ti-Al-C powder mixtures; b) to specify phase and elemental composition of SHS products; c) to investigate the morphology of composite powders obtained by products crushing and sieving; d) to investigate the dependence of the carbide phase size in the structure of the composite on the reaction mixture composition.

2. Materials and experimental procedures
Reaction mixtures were prepared from titanium (TPP-8, 99,5 %, <160 µm.), carbon black (P-803, 300 nm average particles size) and aluminum (PA-4, 98 %, <100 µm.) powder mixtures. The powders
were mixed for 4 hour and compacted to 35-38% porosity Ø 20×25 mm cylindrical samples. Composition of reaction mixtures and target volume content of the aluminum binder (provided under assumption, that other phases rather than aluminum and titanium carbide are absent in the synthesized products) are presented in table 1. The ratio of titanium and carbon in the reaction mixtures corresponded to equiatomic composition of titanium carbide.

Synthesis was carried out in a sealed reactor under 0.5 bar excess argon pressure. The combustion reaction was initiated by heating of an igniting pellet by molybdenum wire coil. A resultant porous cakes were crashed and sieved to <125 microns granules of composite powders.

Structure studies were carried out at the Centre of collective usage “NANOTECH” of Institute of strength physics and materials science of Siberian branch of Russian Academy of Sciences (ISPMS SB RAS) and National Research Tomsk State University (TSU) by X-ray diffraction (XRD-6000, CuKα) and scanning electron microscopy (LEO EVO 50, Zeiss, Germany and Philips SEM 515).

### Table 1. Reaction mixtures composition and targeted phase structure.

| Target phase composition of synthesized products | Reaction mixtures proportion (mass %) | C | Ti | Al |
|------------------------------------------------|--------------------------------------|---|----|----|
| TiC+10 mass.%Al                                |                                      | 18| 72 | 10 |
| TiC+20 mass.%Al                                |                                      | 16| 64 | 20 |
| TiC+30 mass.%Al                                |                                      | 14| 56 | 30 |
| TiC+40 mass.%Al                                |                                      | 12| 48 | 40 |
| TiC+50 mass.%Al                                |                                      | 10| 40 | 50 |

### 3. Results and discussion

#### 3.1. Phase composition of synthesized products

Synthesis in all compacts from the powder mixtures indicated in table 1 was carried out in a stationary wave combustion mode.

| Target phase composition | Relative phase content (%) | TiC (a, nm) | Al | Al:Ti |
|--------------------------|-----------------------------|-------------|----|-------|
| TiC+10 mass.%Al          |                             | 83 (0.4324) | 12 | 5     |
| TiC+20 mass.%Al          |                             | 80 (0.4326) | 15 | 5     |
| TiC+30 mass.%Al          |                             | 61 (0.4327) | 35 | 4     |
| TiC+40 mass.%Al          |                             | 58 (0.4327) | 38 | 4     |
| TiC+50 mass.%Al          |                             | 57 (0.4327) | 40 | 3     |

X-ray patterns of SHS powders are shown in figure 1, and the relative content of phases in the synthesized products, determined from the sum of the areas under the peaks of the individual phases – in table 2. The lattice parameters of all phases, the size of coherent-scattering region and crystal lattices elastic displacement (∆d/d) are also determined from the results of X-ray diffraction evaluation. Dependences of the phase structural characteristics on the reaction mixtures composition remained constant within the measurement accuracy. The titanium carbide lattice parameter turned out to be slightly less than the reference value (0.43274 nm [6]) for the equiatomic composition titanium carbide. The pronounced lattice parameter dependence on the aluminium content in the reaction powder mixtures was not found, in contrast to the data of [6], according to which the lattice parameter of titanium carbide in SHS composites of TiC+Al was found to be much smaller and monotonically decreased from 0.4322 nm at 10% Al binder content to 0.4312 nm at 40%. A possible reason for the decrease in the carbide lattice parameter in [6] is the contamination of the reaction mixtures metal components with oxygen during a long (5 hours) treatment in a planetary mill. In addition to carbon
deficit [8], oxygen or nitrogen dissolution in the lattice [13] the reason for the change in the carbide lattice parameter in SHS composites can be dissolution of the metal binder components in the carbide lattice [9, 14].

As the aluminium content in the reaction mixtures increases, its content in the synthesis products increases as well, but the content of titanium carbide, respectively, decreases. As for other phases, the intermetallic compound Al₃Ti is detected in the SHS products. Its content is small and differs little within studied compositions.

**Figure 1.** X-ray diffraction patterns (CuKα) of SHS powders with different target Al binder content (wt. %): 1 – 10%; 2 – 20%; 3 – 30%; 4 – 40%; 5 – 50%.

**Figure 2.** Morphology of SHS powders with different content of Al binder (mass. %): a) 10%; b) 20%; c) 30%; d) 40%; e) 50%.
3.2. Morphology of SHS powders and the dispersity of the carbide phase

The granules of the composite powder (figure 2) have a predominantly lumpy shape without sharp angles and edges. This form of granules resulted from the crushing is due to the composite structure of the SHS product (figure 3). The plastic aluminum binder compensates a zero plasticity of the carbide phase. So the granules have rounded and close to the equiaxed shape, that promotes good flowability, which is necessary to ensure a stable, constant feed rate from the feeders used in the surfacing and spraying technologies.

The carbide particles size was estimated from the high magnification images of the granules surface (figure 3). As expected, the dispersion of the carbide phase decreases monotonically as the content of the thermally inert aluminium powder increases in the reaction mixtures (figure 4). Such dependence is typical for SHS products representing the titanium carbide base metal matrix composites [8, 9, 15], including composites with an aluminium binder [5, 6].

![Microstructure of SHS powders with different targeted binder content. Mass.% Al: a) 10%; b) 20%; c) 30%; d) 40%; e) 50%.](image)

![Dependence of the carbide inclusions average size in SHS composites on the content of aluminum binder in the reaction mixtures.](image)
4. Conclusion
1. The SHS composite powder obtained by crushing of SHS products synthesized from the reaction mixtures of titanium, carbon black, and aluminum has a lumpy, predominantly equiaxed shape, favorable in terms of good flowability.
2. The size of carbide inclusions in the aluminum matrix decreases monotonically with an increase of thermally inert aluminum powder content in the powder mixtures and reaches a minimum value of 0.8 μm.

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