Synthesis and properties of the materials obtained by SHS mode for radiation protection

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Abstract. The article shows the process of protective composite materials manufacturing. Also, the analysis of experimental results concerning the composite materials protective properties is given. The advantages of SHS method are considered in comparison with traditional materials. The uniqueness of SHS obtained products based on combination of nuclear-physical properties and parameters is presented.

1. Introduction

Nuclear facilities for various purposes are powerful sources of integrated ionizing radiation which are based on the fission of heavy isotopes stimulated by neutrons. Inside such devices and their surrounding structures, a lot of neutron-stimulated reactions take place causing damage to various structural materials.

A range of research works [1-3] indicated that the creation of a fine-grained structure in the sintering process leads to the increased resistance characteristics of ceramics, which supports the idea that such ceramic materials including metal-ceramics are very promising for use in nuclear and energy engineering in terms of their radiation resistance characteristics.

Now, materials obtained in the mode of self-propagating high temperature synthesis (SHS) are finding increasing application in various fields of science and technology. The advantages of the SHS technology include simplicity of hardware implementation, short time periods of the synthesis course, relatively low energy consumption [3]. In addition, the synthesis is characterized by a unique feature of the existence of a high-temperature medium during the time of reaction, which allows different types of additional external influences, which make it possible to regulate the structure and properties of final products [4]. From this perspective, it can be described as a directed synthesis, whose preparation and implementation of the regimes deliver/provide with the material with the desired combination of properties.

For the protection from the integrated flow of ionizing radiation it is advisable to use materials which include various elements [5, 6]. In case of high-energy neutron radiation flow, it is advisable to use a mixture of light and heavy elements. Heavy protection component must, on the one hand, have a high cross section for inelastic scattering of neutrons of fission spectrum and the low threshold of the inelastic neutron scattering (tungsten, niobium), on the other – to have a high slowed-down neutron capture cross section while emitting low-energy capture gamma-ray emission (indium, hafnium, tungsten). Lightweight component should effectively slow down the fast neutrons in the process of elastic scattering (beryllium, carbon, boron) and have a high capture cross section of low-energy neutrons (boron). Consequently, for protection against ionizing radiation flows of nuclear power plants it is possible to use materials that contain materials such as boron, tungsten, indium, and others.

Borides of metals of a high atomic number with high concentrations of a heavy nuclear component, for example, can be attributed to such materials.

Thus, it can be assumed that a mixture of tungsten and light elements can be considered as promising materials for the protection from the flow of fission neutrons and gamma rays.

This paper presents the results of theoretical and experimental studies on the production and use of new materials used as a protection from the fields of ionizing radiation.

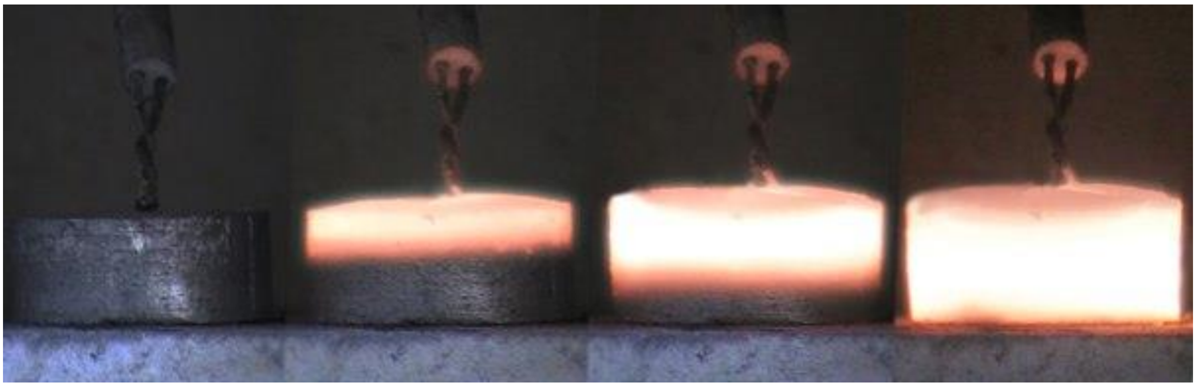
2. Experiment

The synthesis of materials for the following reactions were carried out:

$$W + B \rightarrow WB, \quad (1)$$

$$4B + C \rightarrow B_4C. \quad (2)$$

In the experiments were used powders of boron (chemical purity - 93%), tungsten (99%), carbon (98%), nickel (99%), aluminum (99%), which were mixed in a planetary ball mill AGO-2c for 10 minutes at a frequency of 50 Hz. The pressing of initial blend is performed by a hydraulic press model P338 (pressure is 3.5 MPa). Metal molds are made from tool steel. The inner diameter of the matrix is 30 mm. Duration of compression was 30 minutes. The resulting samples were placed in the furnace, where the synthesis process was carried out the air atmosphere, as shown in Figure 1. Control of combustion temperature was performed by spaced tungsten-rhenium thermocouples (TBP A-2) with an output to a memory storage device for temperature measurement and temperature control.



Different times of synthesis of the sample
0 s 2 s 4 s 6 s

Figure 1. Propagation of the combustion wave at the time of the synthesis process of the sample.

The analysis of qualitative and quantitative composition of the synthesized samples was carried out on X-ray diffractometer Shimadzu XRD6000.

Experimental study on the protective properties of boron-containing materials from neutron flows was carried out with the setup which is shown in Figure 2.

The graphite prism was composed out of 18 blocks of graphite each sized 200x200x600 mm with through-holes of a diameter of 65 mm. In the openings of all blocks (except the one where the source IBN-10 type (fast neutrons flow $1.02 \cdot 10^7$ 1/s), the protective sample and detector (BDKN-03) were located), there were cylindrical graphite rods of the same length in the sleeves.

Experiments were carried out with variation of the thickness of protective shields made of boron-containing materials B_4C and WB. Safety properties of the synthesized materials depending on fast neutron flow were studied and compared to the analogous properties of the traditional material of nuclear technology - graphite.

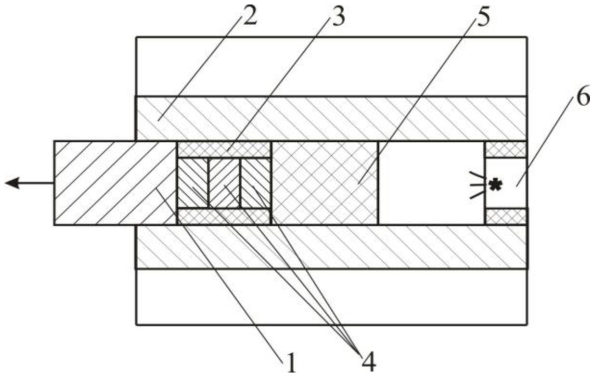
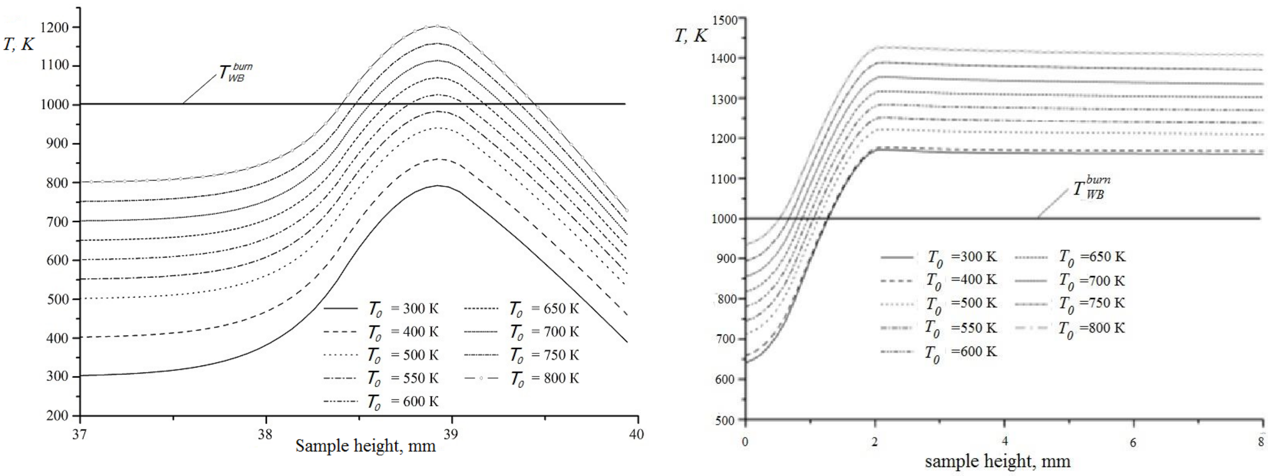


Figure 2. Experimental graphite prism: 1 – face detector of fast neutrons (BDKN-03); 2 – block of graphite prism; 3 – composing graphite sleeve; 4 – inlaid shield of ceramic samples; 5 – a removable protective shield of graphite; 6 – Pu-Be source of fast neutrons.

3. Results and discussion

Figure 3 shows the temperature distribution in the initial (a) and final (b) the time for the W-B system, obtained with the implementation of experimentation results.



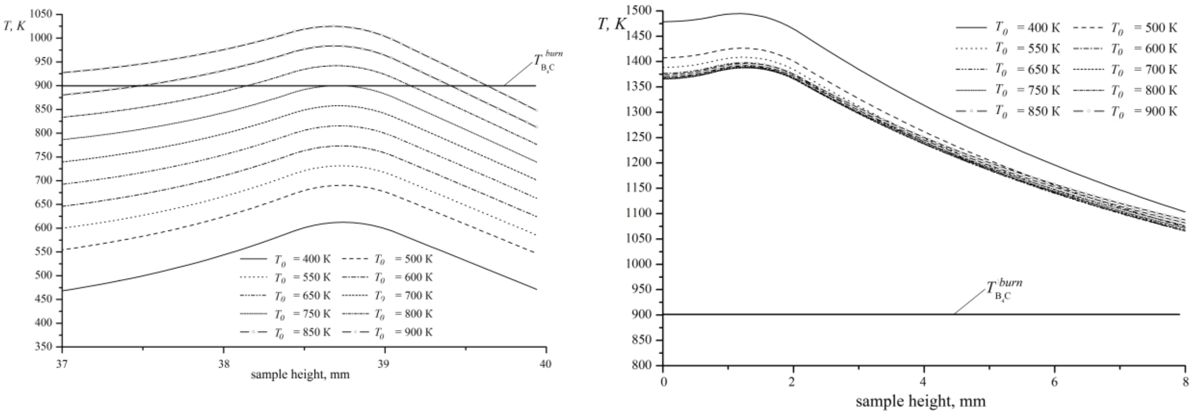
a) b)

Figure 3. Distribution of temperature in the initial (a) and final (b) the time for the W-B system after the initiation of the fusion reaction.

From the distributions obtained it can be seen that with the values of the initial temperature up to 600 K, the maximum temperature of the process in the initial period is lower than the initiation temperature of the tungsten-boron system, then it can be assumed that the propagation of the combustion wave would be unstable, or the synthesis process would not be observed. When the initial combustion temperature is 700 K and above, the minimum phase formation is observed in this sample. Similarly, the temperature distribution was obtained for the system B-C (Figure 3).

There is a weak dependence of the process temperature on initial pre-heating temperature. Thus, at values of from room temperature to 700 K the temperature at the initial moment of time has a value lower than the B-C system combustion temperature value. It can be assumed that the distribution of the combustion wave will be unstable.

X-ray analysis of obtained materials in the course of synthesis was carried out in order to determine their phase composition. Investigations of the phase composition revealed that the structure of the obtained materials, beyond those required, also contains a large number of other phase components. This is due to the staging of the process of combustion in these SHS systems where each step corresponds to a certain temperature. Because of the rapidity of the SHS process and performed during burning inhomogeneity of the temperature field over the volume of the sample synthesized, in the structure of the sample local areas were formed, where the formation of the final product stopped at one stage or another.



a) b)

Figure 4. The distribution of temperature in the initial (a) and final (b) time for the B-C system after the initiation of the fusion reaction

Obtaining necessary compounds required for the synthesis of temperature regimes throughout the sample volume can be achieved by increasing the thermal effect of the combination of reactions occurring in the system. For this purpose, the initial blend of reagents has been extended by introducing additional components entering into an exothermic reaction, thereby increasing the total heat effect. Reactive mixture of nickel and aluminum are used as these additives. With their introduction to the process temperature is increased from 1500-1700 K to 2500 K.

Figure 5 shows the fragments of an X-ray picture of a material based on W-B compounds, characterizing its phase structure without any additional reagents (Figure 5a) and in the presence of these additives (Fig. 5b). It is evident that the introduction of reactive Ni-Al mixtures, leading to increased exothermicity of the SHS systems, allowed to avoid multiphase. In this case the material was a nearly two-phase system of the required phase components. A downward shift of α-WB phase quantity was observed, an upward shift of tungsten diboride phase, which makes them competitive on the content. Phase of tungsten oxide WO_3 and intermetallic compounds of nickel are present at the level of background.

In the experiments shown in Fig. 6 the dependence of the attenuation of fast neutrons on the geometric and mass thickness of the boron-containing materials and graphite was determined. Approximation was performed by the method of least squares (precision 7%).

Experiments have shown that the use of boron-containing SHS-ceramics for the protection from fast neutrons provides gains with regard to dimension indicators. This is illustrated by the dependences shown in Fig. 4. For example, the weakening of the multiplicity of neutrons is equal to 1.3 is provided by a graphite screen media 45.6, ceramic WB - 7 Ceramic B_4C - 2 g/cm^2 thick. The optimal dimensions and weight of the material for protection against fast neutrons is B_4C, WB and then, finally, C.

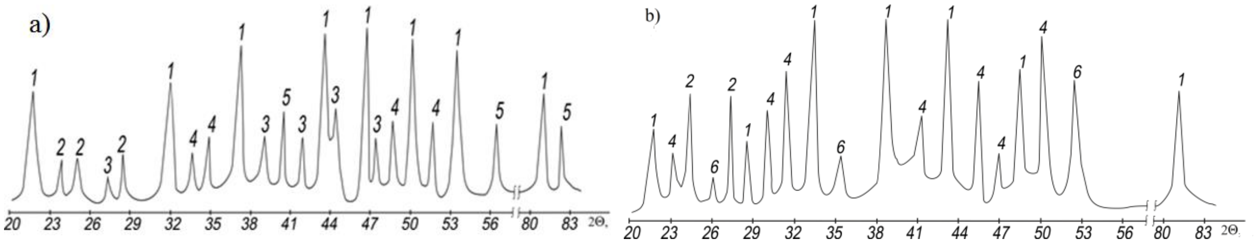


Figure 5. Fragments of an X-ray picture of a material based on tungsten boride (a) and boride of tungsten with a nickel-aluminum modifier (b), characterizing multiphase SHS product: 1 – tungsten boride α-WB phase; 2 – tungsten oxide WO_3 phase; 3 – phase connection W_2B_4; 4 – tungsten diboride phase; 5 – tungsten boride β-WB phase; 6 – nickel aluminide Ni_3Al phase.

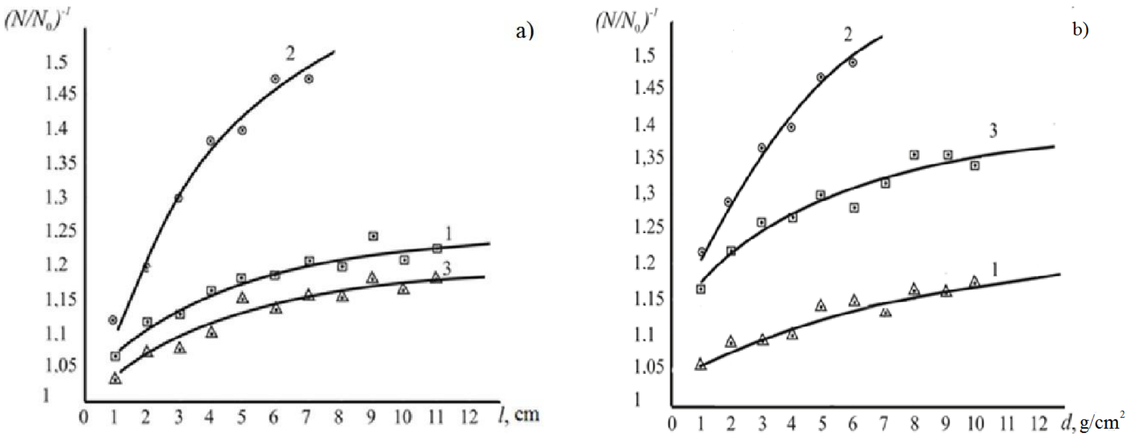


Figure 6. Dependence of the attenuation coefficient $(N/N_0)^{-1}$ on the geometric (a) and the mass thickness (b) of the shielding material: 1 – graphite, 2 – boron carbide, 3 – tungsten boride.

4. Conclusion

Composite materials intended for the manufacturing of protective elements from the neutron flows were synthesized in the process of combustion. Analysis of the results of instrument experiments on protective properties of composite materials, resulting from the combustion process, allows to formulate the following conclusions:

- products obtained in the SHS mode, provides a unique combination of nuclear-physical properties and parameters of weight and size, which allows them to use for effective protection against fast-neutron fluxes;
- SHS method allows to obtain materials that provide advantages in mass and size parameters for the protection of the fluxes of fast neutrons compared with traditional materials.

References

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