

On a Peculiarity of Structure Formation in Combustion of High-Caloric Metallothermic Compounds under Microgravity Conditions

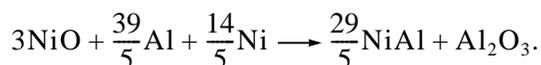
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Gravity strongly affects the combustion of high-caloric metallothermic compounds. In these processes, high-temperature heterogeneous melts composed of mutually insoluble components are produced as the products of combustion. Usually, these melts contain a metallic (heavy) and oxide (lighter) phases. In the gravity field, the phase separation takes place on the macroscopic level (the heavy phase precipitates and the lighter phase rises to the surface) and a two-layer (or multilayer) cast product is eventually formed. This phenomenon was used in investigating the processes of the self-propagating high-temperature synthesis in centrifugal separators, in which the gravity effect of phase separation was enhanced [1–3]. Of no less interest is the opposite formulation of the problem: to study how these processes proceed in the weightless state, i.e., without gravity. Earlier, a marked effect of microgravity on combustion processes and structure formation was noted in combustion of element-containing mixtures [4–7].

In this paper, we present the results of the pioneering experiments on the “liquid-flame” combustion of metallothermic compounds, which were carried out at the orbital space station “Mir”.

In the preliminary investigations on the earth, we conducted a search for an initial system for realizing a “liquid” flame under the microgravity conditions. Based on the results of the thermodynamic analysis and experiments, the composition containing 60% of the thermite mixture $3\text{NiO} + 5\text{Al}$ and 40% of the mixture of elements $\text{Ni} + \text{Al}$ was chosen as the model system. The overall scheme of the chemical transformation in this mixture is of the form:



The model mixture satisfies the principal requirements of the space “liquid-flame” experiment:

a high temperature (2650 K) is attained in combustion; at this temperature, all the condensed substances (initial, intermediate, and final) are in the liquid-phase state, while the concentration of gaseous products (vapors and suboxides) is low;

the combustion of the mixture proceeds within the steady regime with a low rate (~ 0.5 cm/s), the substance loss caused by dispersing the melt is less than 1%, and the combustion products (aluminum oxide and nickel aluminide) take the cast form with a distinct separation of layers.

The major set of experiments on realizing the “liquid-flame” combustion under the microgravity conditions at the “Mir” station¹ and the comparative terrestrial experiments were carried out using the setup “Optizon” at the air pressure of 0.4 atm [8]. For the synthesis, we used the powders of NiO , Ni , and Al with the particle sizes less than $10 \mu\text{m}$. After mixing, the initial mixtures were pressed in the form of tablets with mass of 4.5 g, 8.5 mm in diameter, and 20 mm in height. The tablets were placed in quartz cups of 13 mm in diameter and 80 mm in height. The tablets were ignited by locally heating the upper face of a tablet employing focused beams from three halogen lamps. Upon completion of the experiments at the “Mir” station, the samples were transported to the earth for investigation. A visual analysis of the combustion products revealed that, under both the microgravity and terrestrial conditions, the metallic and oxide phases were separated. In both cases, the combustion products took the cast form. The comparison between the characteristics of combustion products produced under the microgravity conditions and natural terrestrial conditions showed that they differ only slightly (see table). The X-ray phase analysis has shown that the combustion products obtained both at the “Mir” station and under the terrestrial conditions are identical. In both cases, the metallic phase represents nickel aluminide (NiAl) with the b.c.c. lattice, and the oxide phase is aluminum oxide with the corundum lattice. Thus, we can conclude that the grav-

¹ The gravity acceleration at the “Mir” station is $10^{-2}g$, where g is the free fall acceleration.

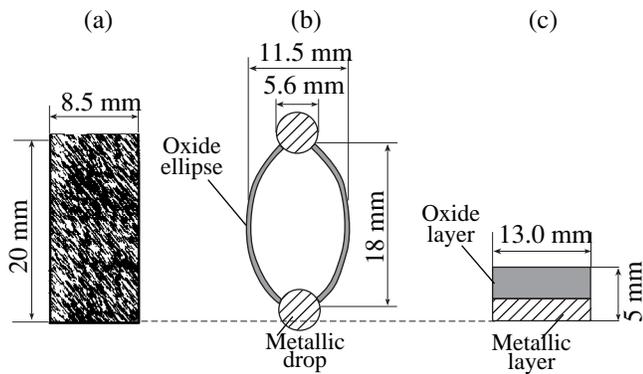


Fig. 1. Effect of microgravity on the macrostructure of cast combustion products: (a) initial tablet; (b) space experiment; and (c) terrestrial experiment.

ity only weakly affects the processes of dispersion, the formation of chemical and phase compositions of the combustion products, and also the completeness of the phase separation between the metallic and oxide phases.

Contrastingly, the macrostructures of the cast products obtained under the space and terrestrial conditions are dramatically different. Under the microgravity conditions, the oxide phase is formed in the shape of a thin-wall shell (a prolate spheroid). At the poles of the shell, metallic particles of spherical form and of approximately equal weight ($m \sim 1.5$ g, Fig 1b) are located. Smaller-sized metallic spherical particles ($m \sim 0.2$ – 0.3 g) were found in the quartz cup. The total height of the object obtained is approximately equal to the height of the initial tablet. The combustion products obtained under terrestrial conditions were in the form of two dense cylindrical layers with a distinct separation between the metallic and oxide layers (Fig. 1c). The total height of the layers is four times less than the height of the initial sample, and the diameter of the layers is equal to the diameter of the quartz cup.

The microanalysis revealed that the samples obtained under the space conditions are dramatically distinct in the microstructure of the oxide phase and are identical in the microstructure of the metallic phase

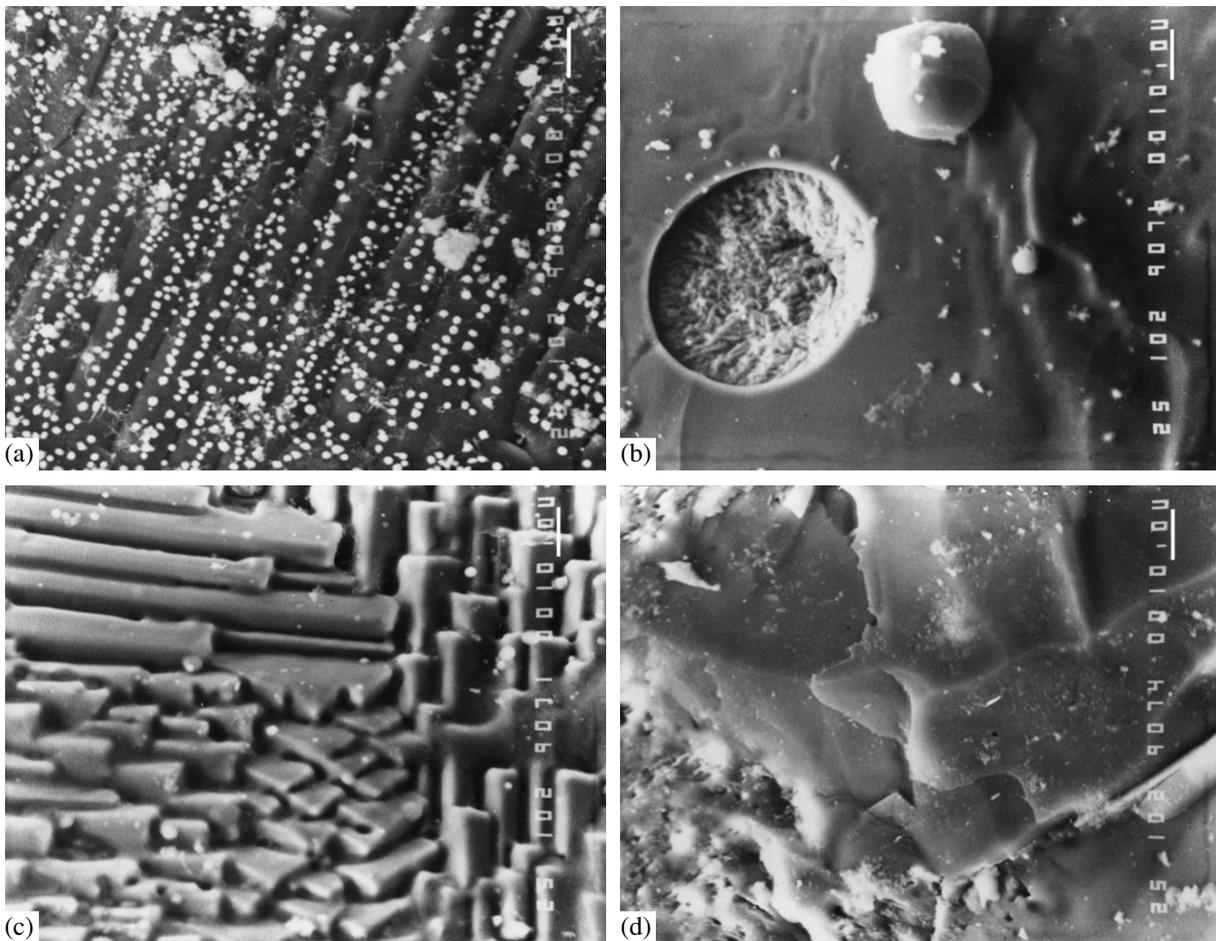


Fig. 2. Effect of microgravity on the microstructure of cast combustion products: (a) and (b) space experiments; (c) and (d) terrestrial experiments; (a) inner surface of the oxide shell; (b) outer surface of the oxide shell; (c) free surface; (d) surface adjacent to the metallic ingot. 400 \times .

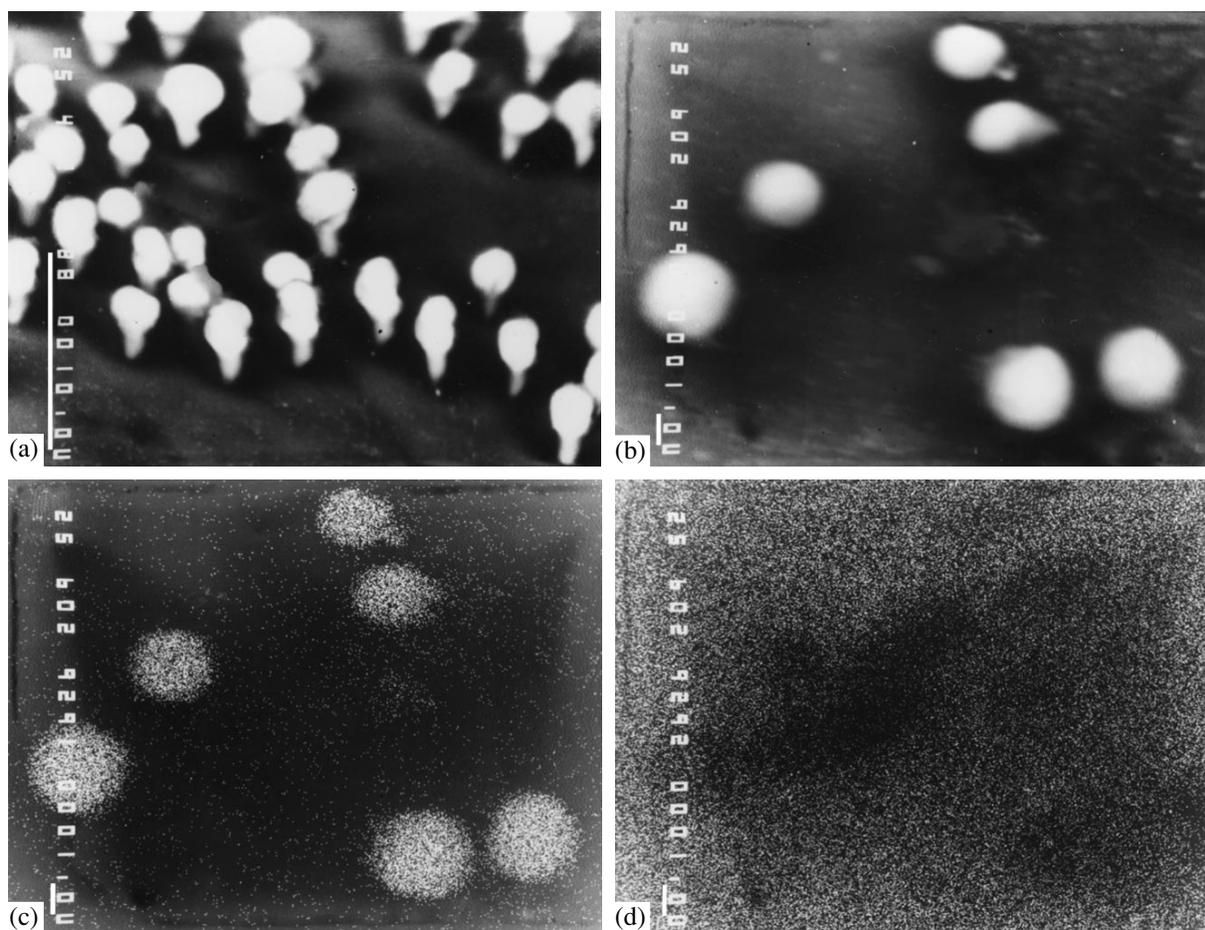


Fig. 3. Distribution of elements in metallic particles at the surface of the oxide shell; (a) and (b) surface element; (c) distribution of Ni; (d) distribution of Al in particles. (a) 4000 \times ; (b), (c), and (d) 6000 \times .

from that obtained under the terrestrial conditions. Moreover, an appreciable difference between the microstructures of the outer and inner surfaces of the oxide phase of the combustion products (the shell) obtained under the microgravity conditions was revealed (Figs. 2a and 2b). Namely, at the inner surface of the oxide sphere, the alternation of metallic and oxide bands was found, the metallic bands having a discrete structure. The metallic particles forming the rows are mushroom-shaped, contain Ni and Al (Fig. 3), and, according to the X-ray analysis, are intermetallic compounds (NiAl). On the outer surface, there are no metallic "rows," and the major area of this surface is smooth. The analysis of the oxide-film cross section revealed the multitude of channels of the rounded cross section, which came out into the inner cavity of the sphere.

On the surface of the "terrestrial" samples, there are virtually no metallic particles, and the surface has a pronounced relief (Figs. 2c and 2d).

The results obtained enable us to conclude that gravity strongly affects the formation of macrostructure and microstructure of cast combustion products.

It is known that the process of phase separation in the combustion products for the thermite-type systems proceeds in two stages under the terrestrial conditions:

Effect of microgravity on the substance loss in combustion and characteristics of the phase separation

Parameters	Experimental conditions		
	$a = 10^{-2}g$	$a = 1g$	calculated values
Initial mass of a tablet, g	4.5	4.5	4.5
Final mass of products, g	4.0	3.9	4.5
Final mass of the metallic phase, g	3.43	3.30	3.73
Final mass of the oxide phase, g	0.57	0.60	0.77
Depth of dispersion, mass %	11	13	—
Completeness of the metal yield, mass %	92	88	100
Completeness of the oxide yield, mass %	74	78	100

the stage of forming small-sized (primary) drops of the metallic phase caused by the forces of surface (interphase) tension and the stage of precipitating the drops under the action of gravity forces and forming an ingot. In the space experiments, the second stage was absent. Both of these processes, i.e., the formation of the primary drops and the following separation between the metallic and oxide phases, proceed only by the action of the forces of interphase tension without participation of the gravity forces. A whimsical shape of the space products is associated with a weak gassing. Under the terrestrial conditions, the gravity ejects rapidly the gas bubbles from the melt. Under the space conditions, a gas inflates the liquid product and it forms a large bubble.

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