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# Self-Propagating High-Temperature Synthesis: First Space Experiments

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Self-propagating high-temperature synthesis (SHS), i.e., the frontal propagation of a locally initiated reaction (combustion) through a porous medium with the formation of solid products [1], is often gravity-sensitive. Investigating the gravity effect on SHS and its products is one of the important lines of inquiry [2, 3]. After the first experiments under microgravity conditions during a parabolic flight of a laboratory aircraft [4], experiments under prolonged space zero-gravity conditions are of great interest.

In this work, the results of the first space experiments performed aboard the Mir orbital space station were presented, which have suggested a new insight into SHS processes.

A reaction mixture was prepared from nickel-coated aluminum particles of primarily spherical shape. The characteristic size of clad particles was 100–150 μm (Fig. 1). The thickness of the nickel coating was chosen so that the Ni-to-Al ratio in every clad particle was equiatomic, which corresponds to the optimum composition of the mixture. This ensures the well-studied simplest SHS reaction to form nickel monoaluminide [5]:

$$\mathrm{Ni} + \mathrm{Al} \rightarrow \mathrm{NiAl} + Q.$$

Below are some characteristics: the nickel melting point $T_m(\mathrm{Ni}) = 1728$ K, the aluminum melting point $T_m(\mathrm{Al}) = 933$ K, the nickel monoaluminide melting point $T_m(\mathrm{NiAl})$ equals the adiabatic combustion temperature $T_C = 1911$ K (the product is partially molten at the maximum temperature of the SHS process), and the heat of reaction $Q = 118.5$ kJ/mol. Clad particles are convenient for experiments, since the contact between the reactants (nickel and aluminum) is reliable independently of the interparticle distance [6].

Samples of two types, produced under laboratory conditions, were examined: cylindrical samples compacted from the clad-particle powder with a density of 3.45 g/cm$^3$ and a porosity of 32%, and uncompacted powder samples with a density of 2.37 g/cm$^3$ and a porosity of 53% (under normal gravity). The latter samples were powders in evacuated sealed quartz ampules, where the ratio of the void volume (of pores and the free space within an ampule) to the particle volume in the ampule was 70 : 30. The gas release upon the reaction is insignificant, since both reactants (nickel and aluminum) are in the condensed (solid or liquid) state. The equilibrium aluminum vapor pressure under synthesis conditions was 2 mm Hg at the combustion temperature 1640°C. The impurity gas release (of water vapor, for the most part) in a similar ampule under terrestrial conditions did not exceed 0.00019 mol/g.

Terrestrial and space (aboard the Mir orbital space station) experiments were carried out with an Optizon-1 zone-melting setup designed at Nauchnyi Tsentr Research Institute [7]. The ignition was performed locally by a radiant beam from three halogen lamps, which was focused on a 1.5–2.0-mm-wide annular region. Features of the radiant-beam ignition of SHS systems have previously been explored in [8]. The design of the reaction chamber allowed one to conduct experiments in space vacuum. Unfortunately, the com-

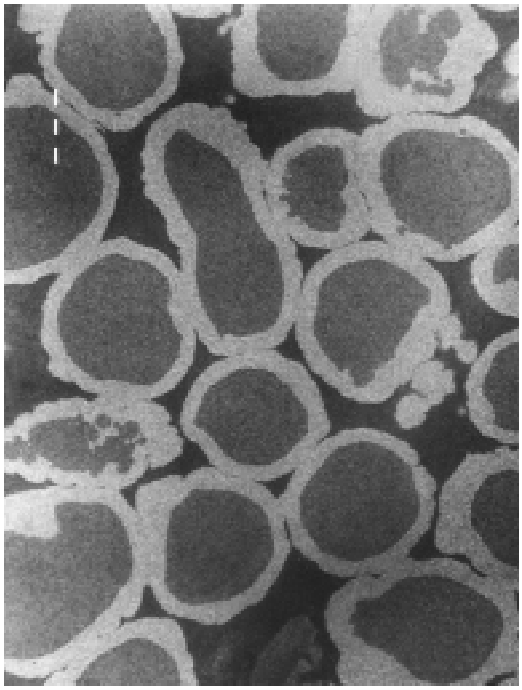

**Fig. 1.** Microstructure of the initial clad particles (polished cross-section).

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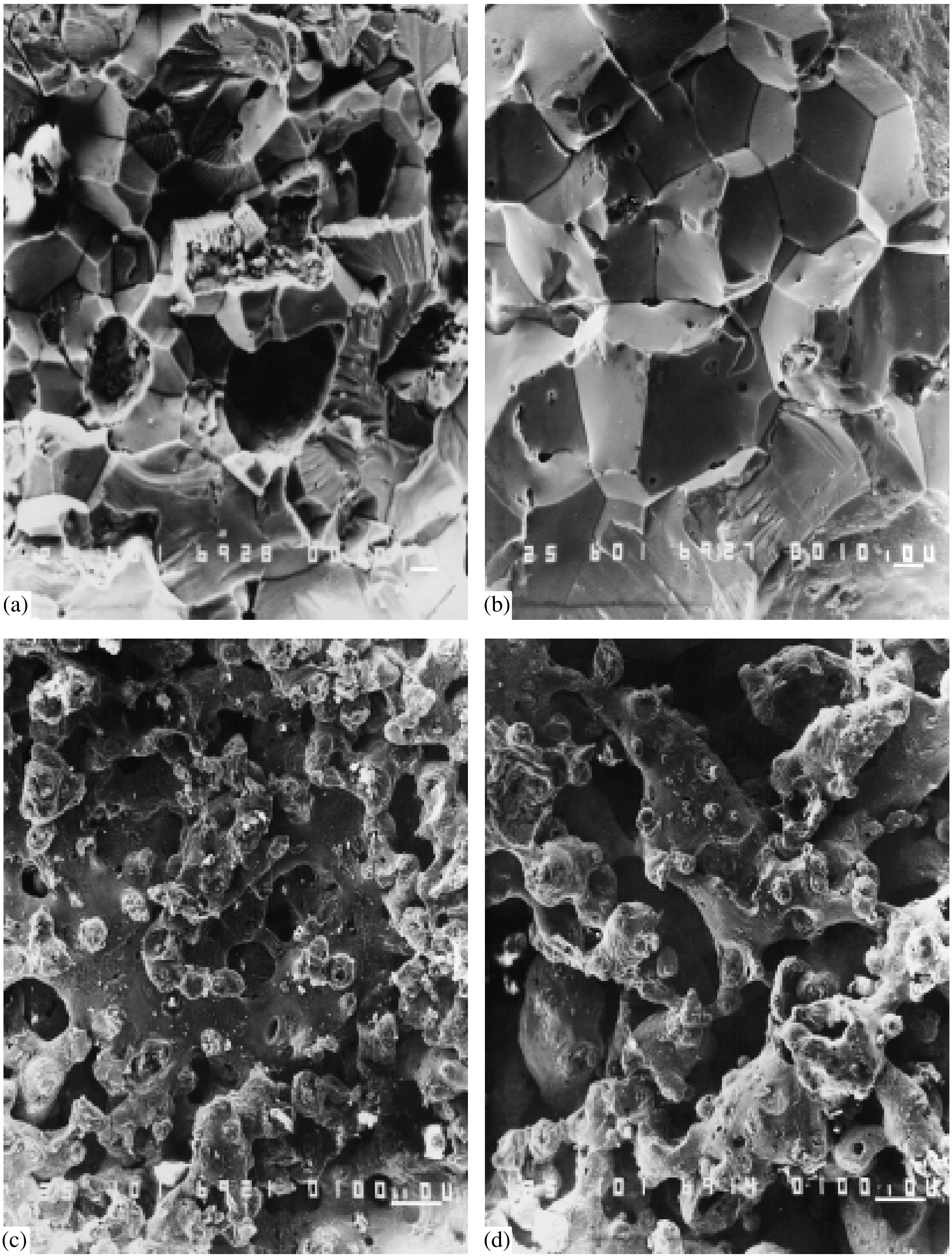

**Fig. 2.** Microstructures of fractures: (a) Earth-burnt compacted sample (600×), (b) space-burnt compacted sample (600×), (c) Earth-burnt uncompacted sample (100×), and (d) space-burnt uncompacted sample (100×).

bustion velocity in space experiments was not measured. After SHS experiments aboard the Mir orbital space station, the samples were returned to Earth.

Compacted samples burnt in terrestrial and space experiments retained their cylindrical shape and sizes. The surface of a sample burnt in space is smoother, has the specular metallic luster, and is free from cracks and cavities observed on the surface of a sample burnt on Earth. In the upper part of the samples, where they were ignited, melt drops formed, probably, due to the excess of heat brought by an igniting radiant beam. Under zero-gravity conditions, several round drops 1–3 mm in diameter were detached from the upper face of the sample. Electron-probe and X-ray diffraction analyses showed that the chemical and phase compositions of the Earth- and space-burnt samples are identical and correspond to the end product NiAl. Diffraction peaks in the X-ray diffraction pattern of the space-burnt sample are about half as narrow again as those for the Earth-burnt sample. As is seen from the microstructures of the fractures in Figs. 2a and 2b, nickel monoaluminide grains formed under zero-gravity conditions are larger. In the space-burnt sample, the intergranular fracture dominates, whereas in the Earth-burnt sample, the fraction of the brittle intragranular fracture is large, which is indicated by the characteristic river structures in the fracture pattern. The structure of the space-burnt sample is more homogeneous, and the portion of large anisotropic pores in it is greater. The open porosity is higher in the Earth-burnt sample, and the closed porosity is higher in the space-burnt sample.

Thus, zero-gravity influence on compacted samples leads to the formation of larger and more perfect crystalline grains of NiAl, probably, owing to the zero-gravity effect on the crystallization.

Figure 3 displays the uncompacted samples after terrestrial and space experiments, and also the quartz ampule filled with the initial clad-particle powder. In both terrestrial and space experiments, SHS was initiated at the central part of the quartz ampule (Fig. 3a). Under normal gravity, the uncompacted powder layer had burnt without change in its volume; and neither expansion of the layer, nor its shrinkage occurred. Only a characteristic scar in the reaction initiation zone is observed, which can be explained by the excess of heat brought by an igniting radiant beam. The combustion product formed under zero-gravity conditions is much more porous (the density is 1.51 g/cm$^3$, and the porosity is 70%) and has the shape of the internal space of the ampule. For both the Earth- and space-burnt samples, the product is NiAl. Figures 2c and 2d present the microstructures of the samples. The main structural component is irregularly shaped globules bearing traces of melting, which form a porous spatial network. In the space-burnt sample, this network has an openwork-like structure, globules are bound weakly, and pieces of the sample are readily crushed to powder upon pressing.

The results of combustion of the uncompacted samples are of greatest interest. Since no expansion occurs in combustion of the uncompacted powder, one can assert that, as early as before the commencement of combustion, particles under zero-gravity conditions have uniformly occupied the entire space within the ampule and formed a stable cloud similar to a gas-solid suspension, but without gas-in vacuum. This is also corroborated by the direct visual observations made by Cosmonaut A.Ya. Solov'ev. Particles in the cloud slowly move relative to each other, sometimes colliding with one another.

(a) (b) (c)

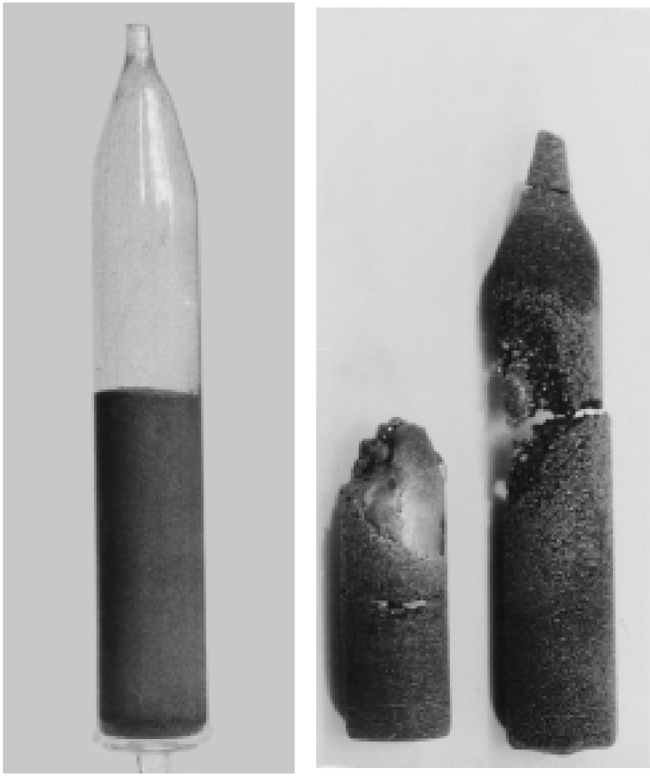

**Fig. 3.** Photographs of (a) the experimental ampule, and the uncompacted samples burnt under (b) normal gravity and (c) microgravity.

One could expect that a sample after burning will remain particulate and, after having been returned to the Earth to normal gravity conditions, will form a powder of the reaction product (NiAl). However, in our case, the combustion product has the highly porous connected skeleton (network) structure (Fig. 2d). This is explained by features of combustion of Ni–Al clad particles. Figure 4 depicts the microphotographs of separate particles burnt (under normal gravity conditions) without contact with neighboring particles. Since aluminum melts at lower temperature than nickel, it breaks the nickel coating and flows out. Therefore, particles acquire a complex gnarled shape with snags, which intersect snags of other particles, thus forming a spatial openwork-like network.

The heat transfer from burnt particles to the initial ones, which ensures the autowave process, can be due to both radiation, and contact heat conduction upon particle collisions, and should be scrutinized in detail. In many SHS systems, combustion is well known to remain stationary even if the density of a reaction mixture is as low as its bulk density. It is natural to assume that, at densities below the bulk density, combustion is also possible. Currently, gasless combustion in this density range is virtually unstudied: the combustion modes in this range are unknown; the lower combustion

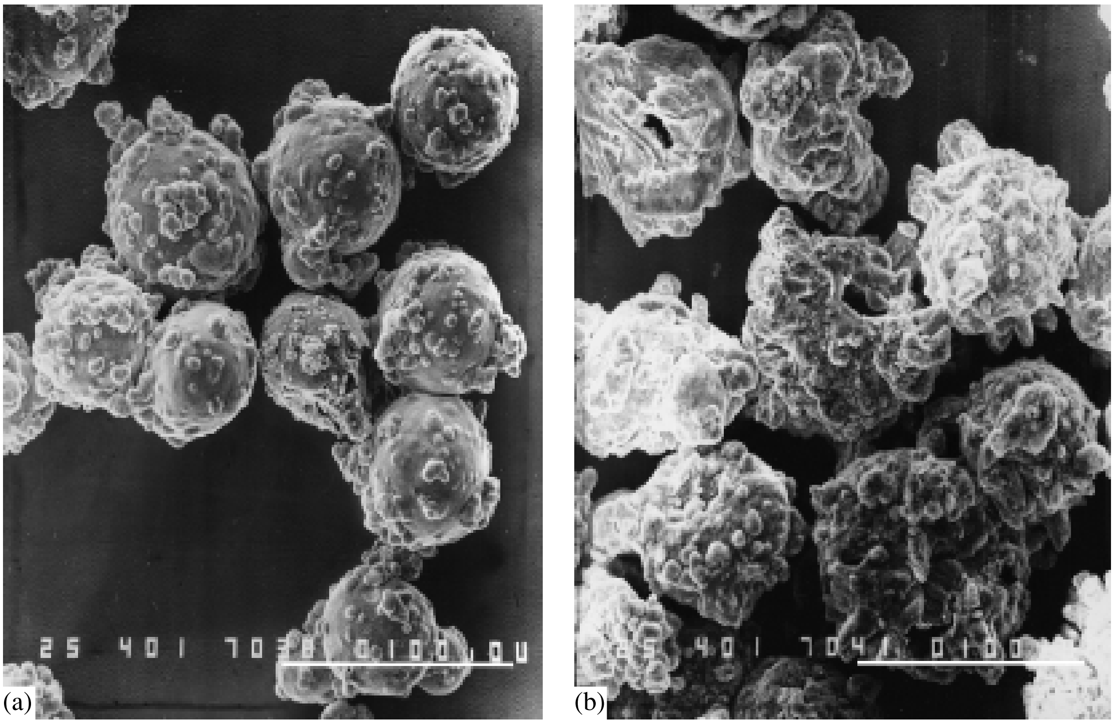

**Fig. 4.** Microstructure of the (a) initial and (b) burnt clad particles (400×).

limits versus density are undetermined; and even the possibility itself for gasless combustion in a system of particles that are weakly bound to one another is doubtful. One can obtain a density below the bulk density under gravity conditions by, e.g., blowing an inert gas through a particle layer (fluidized bed), or using other techniques for suspending particles (levitation): electrostatic traps, acoustic waves, etc. However, all these methods strongly deviate combustion. Under prolonged space zero-gravity conditions aboard the Mir orbital space station, a pure experiment has been carried out: particles have been distributed in space before the onset of combustion.

The results of this experiment are of great theoretical and practical significance.

First, SHS has been performed in a system of initially noncontacting particles (“noncontact” SHS). The possibility of such experiments has been mentioned in [9]. However, they have not been conducted under terrestrial conditions. Zero gravity provides the best conditions to investigate such systems.

Second, the new approach for manufacturing highly porous materials was implemented. Previously, to synthesize such materials, gasifying agents [3, 4, 10] or thermally expanding graphite [11] were added to a reaction mixture. In the heating zone, they caused a great increase in the volume of the initial reaction mixture, and combustion proceeded in the low-density medium (the density is usually below the bulk density). In our case, another mechanism takes place, namely the formation of a highly porous structure in the zone (or the products) of combustion owing to the changes in the shape and the sizes of particles in combustion. This mechanism has certain advantages and can be competitive: e.g., it enables one to produce controlled-shape samples (articles), since the particle cloud uniformly occupies the space of any shape.

The experiments described suggest the main conclusion that space zero-gravity experiments on SHS in systems whose density initially equals the bulk density are quite promising and can give fundamentally new results. Of particular interest is performing SHS in clouds of unlike particles (e.g., an Al–Ni powder mixture) for revealing the role of gas-transport reactions in the gasless combustion mechanism [9].

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