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1. Personnel

Co-Principal Investigator (1)

Dr. Paul J. McGinn, Department of Chemical Engineering, University of Notre Dame.

Post-Doctoral Research Associates (3)

The following post-doctoral research associates participated in the research program, and were supported by the grant:

Dr. Hu-Chun Yi (July 94 - Jun 95); Dr. Alexander Rogachev (Apr.-Sept. 95); Dr. Alexander Mukasyan (Nov 95 - Jan 97).

Ph.D. Student (1)

Aleksey Pelekh, a third-year graduate student, has been working on the experimental aspects of this project as part of his doctoral research.

2. Publications Acknowledging this Grant

Varma A., Yi H.C., and McGinn P.J., "The Effect of Gravity on the Combustion Synthesis of Ni-Al and Ni_3Al-TiB_2 Composites from Elements", *Third International Microgravity Combustion Workshop,* Cleveland, Ohio, 1995.

Yi H.C., Varma A., Rogachev A.S., and McGinn P.J., "Gravity-Induced Microstructural Nonuniformities during Combustion Synthesis of Intermetallic-Ceramic Composite Materials", *Ind. Eng. Chem. Res.*, **35**, 2982-2985 (1996).

Mukasyan A.S., Pelekh A.E., Varma A., Rogachev A.S., and Jenkins A., "Effects of Gravity on Combustion Synthesis in Heterogeneous Gasless Systems, *AIAA Journal*, (1997, in review).

Mukasyan A.S., Pelekh A.E., and Varma A., "Combustion Synthesis in Gasless Systems under Microgravity Conditions", *J. Mater. Synth. & Proc.*, (1997, in review).

Varma A., Mukasyan A.S., and Pelekh A.E., "The Effects of Gravity on Combustion and Structure Formation during Combustion Synthesis in Gasless Systems", *Fourth International Microgravity Combustion Workshop,* Cleveland, Ohio, (1997, in press).

3. Presentations Acknowledging this Grant

"The Effect of Gravity on the Combustion Synthesis of Ni-Al and Ni_3Al-TiB_2 Composites from Elements", *Third International Microgravity Combustion Workshop,* Cleveland, OH, April 1995.

"The Effects of Gravity on Combustion and Structure Formation during Combustion Synthesis in Gasless Systems", *Fourth International Microgravity Combustion Workshop,* Cleveland, OH, May 1997.

"The Effects of Gravity on Combustion and Structure Formation of Heterogeneous Systems", *4-th International Symposium on SHS,* Toledo, Spain, October 1997.

"The Role of Gravity on the Mechanism of Combustion Synthesis of Advanced Materials", *AIChE Annual Meeting*, Los Angeles, CA, November 1997.

4. Summary of Results

For reasons of brevity, only selected examples of results are described below. Additional results and details are available in the publications listed in section 2.

The main goal of our research was to study the influence of gravity on the combustion characteristics of heterogeneous gasless systems. In addition, some aspects of microstructure formation processes which occur during gasless combustion synthesis (CS) were also studied. Four directions for experimental investigation were explored: (i) the influence of gravity force on the characteristic features of heterogeneous combustion wave propagation (average velocity, instantaneous velocities, shape of combustion front); (ii) the combustion of highly porous mixtures (with porosity greater than that for loose powders), which cannot be obtained in normal gravity; (iii) the effect of gravity on sample expansion during combustion, in order to produce highly porous materials under microgravity conditions; and (iv) the effect of gravity on the structure formation mechanism during the CS of poreless composite materials.

4.1 Systems Under Investigation

A number of systems were chosen to investigate different aspects of the influence of gravity on combustion synthesis process. They all have the common feature that at least one liquid phase exists in the reaction front. Owing to this feature, gravity can be expected to play a role in the mechanism of combustion and structure formation.

All experiments were conducted under otherwise identical conditions in both normal ($g=10\ m/s^2$) and microgravity (μG, $g=10^{-4}\ m/s^2$ during 2.2 s free-fall in Drop Tower and $g=10^{-1}\ m/s^2$ in ~20 s parabolic trajectories), to make a direct comparison and to establish the effects of gravity on combustion and structure formation in the investigated systems.

4.2 Effect of Gravity on Combustion Velocity

The results on combustion velocity for different systems show that the average velocity value was somewhat larger in terrestrial conditions. More importantly, in all cases, the root mean square (RMS) deviations of calculated velocities, characterizing the steadiness of front propagation, were also greater in terrestrial conditions than in microgravity.

We believe that convection of inert gas, taking place in the reaction chamber during the combustion process in terrestrial conditions, leads to instability of combustion front propagation along the sample. These effects can be important not only for fundamentals of structure formation, but also for direct production of net-shape articles by the combustion synthesis method. Indeed, instability of combustion wave propagation usually results in undesired non-uniform properties in the produced material.

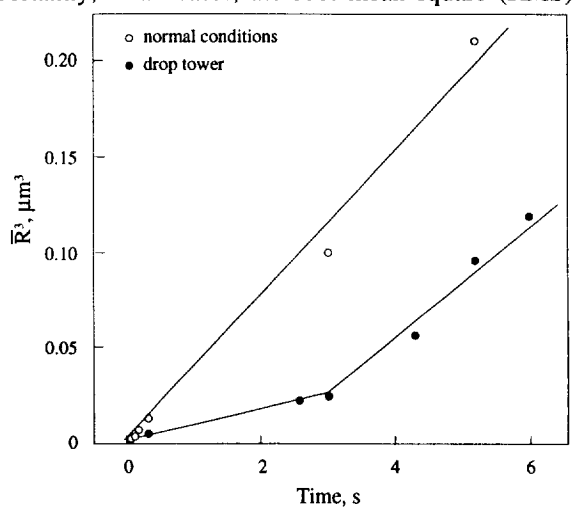

Figure 1: Evolution of size of TiB_2 particles during combustion synthesis of Ni_3Al-TiB_2 composite in different gravity conditions.

4.3 Effect of Gravity on Particle Growth

It was observed that gravity influences not only propagation of the combustion wave, but also ***microstructure*** of the final product. The results obtained on quenched samples showed that this difference is related to the process of grain growth during CS. For the (3Ni+Al)-(Ti+2B) reaction system, the characteristic dependencies of average TiB_2 particle size as a function of liquid phase existence duration are shown in Figure 1, for normal and microgravity conditions. In both cases, very fine particles (~0.1 μm) of TiB_2 formed immediately in the reaction front. However, the rate of particle growth in μG condition

(during the first 2 s) appears to be lower than at normal conditions. We believe that enhanced mass transfer, owing to melt convection and buoyancy in normal gravity, promotes the grain growth.

4.4 Effect of Gravity on Sample Expansion

The data on the average measured combustion front velocity (U_c) and relative linear final expansion (l_f/l_0) of several systems were obtained. For all cases, final expansion and U_c appeared to be higher in μG than in normal conditions. The dependencies of relative sample length (l/l_0) as a function of time are presented in Figure 2. It should be noted that in both cases, the expansion velocities (U_{ex}) remain constant as the combustion wave propagates along the sample; however, the value of U_{ex} is greater under μG conditions.

It is interesting that gravity does not influence combustion velocity measured relative to the unreacted part of the sample: $U_c'=U_c-U_{ex}$. This implies that the region of sample expansion is located behind the reaction front.

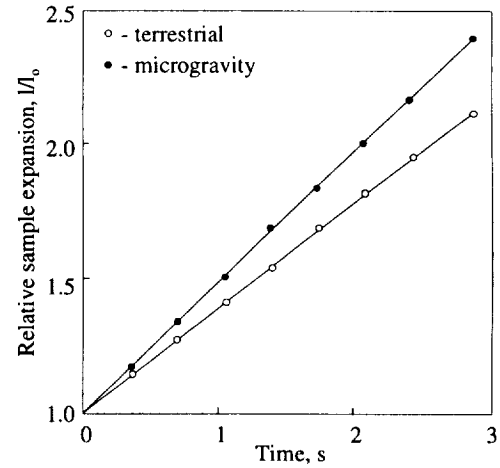

Figure 2: Relative sample expansion as a function of time for different gravity conditions (Ti-C-Ni system).

4.5 Effect of Gravity on Combustion of High Porous Mixtures

To investigate the role of gravity on the mechanism of heat conduction in heterogeneous media during combustion, several systems were studied. We expected to find a change in the behavior of reaction front propagation due to the decrease of contact between particles in μG conditions. For this purpose, combustion of loose powder mixtures was examined. The quartz tube was half-filled with reactant mixture, which was ignited from the bottom. For loose (porosity = 0.8) Ni+Al mixture in normal gravity, the combustion front propagated relatively steady, with an average velocity 1.6 cm/s. The average front velocity in μG was about 5 cm/s. This dramatic increase in combustion velocity may be explained by a change in the primary mechanism of heat transfer in the reaction medium. Since space is available for expansion in a partially filled tube, in microgravity conditions, the decrease of optical density of reaction medium increases the radiation contribution, thus increasing the combustion velocity. On the other hand, owing to settling of particles, this effect is suppressed in normal gravity conditions.

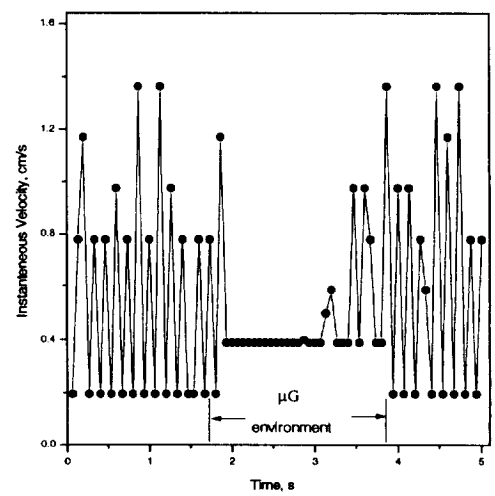

Figure 3. Instantaneous velocities as a function of time for combustion of Ni/Al cladded particles.

The dependence of instantaneous velocity as a function of time obtained during combustion of loose (porosity = 0.55) Ni/Al cladded particles in the drop tower experiment is shown in Figure 3. The quartz tube half-filled with loose reactant mixture was ignited from the bottom in normal gravity conditions. After about 2 s of reaction, the chamber was dropped and during the next 2 s combustion occurred in μG. Then, the CS process continued under terrestrial condition once again. A relatively narrow velocity distribution was observed for combustion in microgravity, as compared to normal conditions. Usually, the former type of distribution exists when a quasi-homogeneous mechanism of combustion occurs. This may take place in the following cases of gasless combustion synthesis:

(i) high sample density, where heat conduction through the essentially poreless medium controls the wave propagation process;
(ii) very high porous mixtures, where radiation becomes the main mechanism of heat transfer.

We believe that the latter condition was realized under μG in the experiments discussed above. However, more detailed investigations should be carried out to clarify the nature of the observed phenomena.

4.6 Effect of Gravity on Phase Separation

Several experiments have been conducted to investigate phase separation effect during CS in the CoO_4-Cr-Mo-Al system. The following reaction occurs in the combustion wave:

$$3CoO_4 + 8Al \rightarrow 3Co + 4Al_2O_3$$

The Co-based alloy is a biomaterial used for production of orthopedic devices, and the role of Cr and Mo is to impart the desired microstructure and properties. In this system, the high combustion temperature (T_{ad} >3000 K) leads to the full melting of reaction mixture in the combustion front. Due to the large difference in densities of Al_2O_3 (ρ=2.8 g/cm^3) and Co (ρ=7.9 g/cm^3), we expected to obtain quite different materials in normal and microgravity conditions: relatively full separation of phases in normal gravity, and composite materials with relatively uniform distribution of Al_2O_3 phase over the Co-Cr-Mo matrix in μG experiments.

The experimental results show that (1) gravity does not influence combustion velocity in this system (U_c= 2.5±0.4cm/s in μG; U_c=2.7±0.5cm/s in normal gravity); and (2) relatively complete separation (~80% of terrestrial) was achieved in μG condition. However, microstructures of the alloys were quite different. Dendrite-type microstructures characterize terrestrial samples, while globular uniform structures were synthesized in microgravity. The effect of relatively complete phase separation in microgravity conditions can be explained by the role of surface forces in the case when melted phases (~ 50vol.% of Co-Cr-Mo and ~50vol.% of Al_2O_3) form a continuous network.