ELECTRIC FIELD EFFECTS IN SELF-PROPAGATING HIGH-TEMPERATURE SYNTHESIS UNDER MICROGRAVITY CONDITIONS

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INTRODUCTION

Self-propagating high-temperature synthesis (SHS) has been used to form many materials [1]. SHS generally involves mixing reactants together (e.g., metal powders) and igniting the mixture such that a combustion (deflagration) wave passes though the mixture. The imposition of an electric field (AC or DC) across SHS reactants has been shown to have a marked effect on the dynamics of wave propagation and on the nature, composition, and homogeneity of the product (e.g., [2-5]). The use of an electric field with SHS has been termed "field-assisted SHS".

Combustion wave velocities and temperatures are directly affected by the field, which is typically perpendicular to the average wave velocity. The degree of activation by the field (e.g., combustion rate) is related to the current density distribution within the sample, and is therefore related to the temperature-dependent spatial distribution of the effective electrical conductivity of reactants and products. Furthermore, the field can influence other important SHS-related phenomena including capillary flow, mass-transport in porous media, and Marangoni flows. These phenomena are influenced by gravity in conventional SHS processes (i.e., without electric fields) [6-9]. As a result the influence of the field on SHS under reduced gravity is expected to be different than under normal gravity. It is also known that heat loss rates from samples, which can depend significantly on gravity, can influence final products in SHS (e.g., [10]).

This research program is focused on studying field-assisted SHS under reduced gravity conditions. The broad objective of this research program is to understand the role of an electric field in SHS reactions under conditions where gravity-related effects are suppressed. The research will allow increased understanding of fundamental aspects of field-assisted SHS processes as well as synthesis of materials that cannot be formed in normal gravity.

RESEARCH EFFORTS

The research will investigate the following topics.

(a) The effect of the field on the dynamics of SHS waves in the absence of gravitationally induced buoyancy and phase separation effects. This aspect of the work includes the effect of the field on wave velocity and temperature, on the mode of propagation (e.g., planar or spinning) and its transition, and on the conversion profile. Implied in this part of the research is a significant effect of the field on interfacial energies, capillary spreading, mass transport in porous media, and electrically-induced Marangoni flows.

(b) The effect of the field on phase formation and structure evolution in SHS. This research has practical implications as well as fundamental implications. We plan to investigate the role of the field on product formation (nature of the product, composition of phases, homogeneity of the phase(s), and grain size).

The work includes both terrestrial and reduced gravity experiments. The latter will be carried out in parabolic flights and in drop towers. The parabolic flight experiments will use existing facilities (COSYM™, developed by Guigne International, Ltd.) in NASA's KC-135 aircraft. Experiments will also performed at the NASA Glenn Research Center 2.2 Second Drop Tower.

The research will employ systems that produce varied amounts of liquid. For example, the systems Si + C and Mo + Si_2 allow investigation of the case where one reactant melts and the other does not, the system Ta + C allows investigation of a system where neither the reactants not the products melt, and the systems Fe + Al or Ti + Al allow investigation of the case where both reactants and products melt with high field activation. These systems are expected to respond to the gravity level in different ways, e.g., through liquid transport effects.

NORMAL GRAVITY EXPERIMENTS

Normal-gravity experiments have been performed to elucidate the effects of gravity by using different sample orientations for ignition: bottom, top or side. In all cases, reactant powder mixtures were pressed into pellets. The pellets were ignited at one end using a tungsten coil that was heated with current. In addition, a layer of chemical igniter material was sometimes placed on the end of a sample. This layer was composed of Ti, B and Fe_2O_3 and was typically about 3.2 mm thick. Sample pellets had dimensions of 25.5 × 6.5 × 15.5 mm. Samples were generally ignited in ultrahigh-purity argon environments at 1 atm.

Investigation of the Si + C system with a high-speed camera (HSC) and a long-range microscope allowed us to examine microscale phenomena associated with combustion waves. Reactions with pulsating combustion fronts showed droplets of liquid silicon forming on the surface before the reaction and enlarging as the reaction wave pauses before the next pulse. Most of these droplets disappeared with the next pulse. All ignition orientations showed this behavior. In bottom ignition samples, reactant outgassing usually blocked the view of the wave front in the HSC because of buoyant flows. This was not observed in top ignition because the wave propagation was in the opposite direction of the convective flow.

The Ti + Al system was investigated with and without the use of an igniter layer for both top and bottom ignition orientations. The wave dynamics were investigated using Tracker 3, which is an Object Tracking and Image Processing software package supplied by NASA [11]. Tracker 3 is able to track the wave front position, which enables us to obtain wave velocity data. In the case of coil-only ignition (no igniter layer) at low applied fields, higher reaction temperatures and velocities existed for the bottom ignition orientation compared with top ignition (figure 1). This is likely a result of buoyant flows that would have been caused by the igniter coil in addition to the heated sample surface and flows inside the porous samples. These flows would have preheated the entire sample before ignition, causing higher wave velocities. This difference decreased as the applied field was increased (figure 1). It is hypothesized that this is because Joule heating within samples was dominant at higher field levels.

Ignition with an igniter layer is more robust and reliable then ignition achieved with just the tungsten coil. Therefore experiments were performed with the igniter layer, which showed similar effects at low applied fields. Velocity magnitudes converged at lower fields relative to results without the igniter layer with increasing fields. At high fields, the results from top ignition experiments showed increased velocity magnitudes relative to bottom ignition (figure 2).

Although the reactants are intrinsically electrically conducting, the green pellets were electrically insulating due to the low reactant density, which was about 59% of the theoretical maximum.

Figure 3 shows sample resistance data that were calculated using voltage and current data obtained during the experiments. The resistance falls quickly with formation of a molten phase and formation of the reaction zone. The resistance data show interesting results that can be related to changes in velocity at different fields. Resistances in certain ignition orientations tend to cluster together (figure 3). At low fields, resistances for the top ignition experiments had lower values than for bottom ignition. As the field was increased, resistances for both orientations became similar. However, at high fields, resistance of the top ignition experiments exceeded the bottom, similar to the velocity measurements (figure 3). These effects are believed to be due to the dominance of melting of the reactant (aluminum) at low fields and product at high fields.

Other efforts have involved quenching studies and development of gas flow visualization systems. Quenching experiments are performed by turning off the electric field during the combustion process, which causes the deflagration wave to quench under certain conditions, essentially freezing the structure of the wave. Wave structure will be analyzed using XRD, SEM and EPMA for both ignition orientations. For flow visualization, Schlieren and shadowgraph systems have been constructed at UC Davis using small, low-power components that can be employed in the reduced-gravity experiments.

We are also pursuing modeling studies. Computational efforts involve extending previous numerical models [3,12,13] to account for phenomena such as melting, capillary flows, buoyant flows and 3-d effects. We are evaluating use of the FlexPDE commercial finite element code [14] that may have the capability to model 3-d combustion with complex geometries. The asymptotic studies will be concerned with determining the influences of electric fields on stability and extinction of field-assisted SHS waves.

REDUCED GRAVITY EXPERIMENTS

Development of hardware for the reduced gravity experiments is proceeding. It is anticipated that parabolic-flight and drop-tower experiments will be performed during the current year of this cooperative agreement. These experiments will provide data on wave velocities, sample temperatures and sample microstructures for comparison with the normal-gravity experiments that have been performed.

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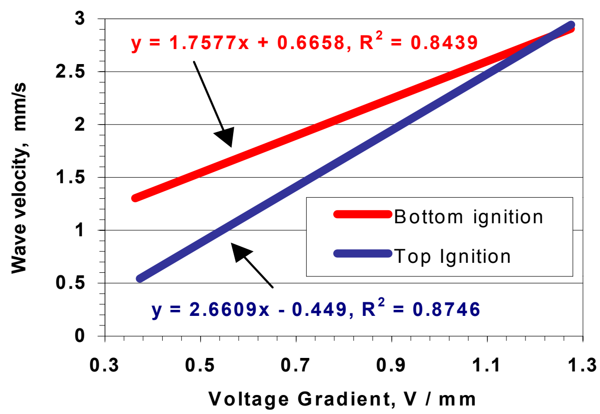

Figure 1. Wave velocity vs. field with coil-only ignition (no igniter layer) in the Ti + Al system.

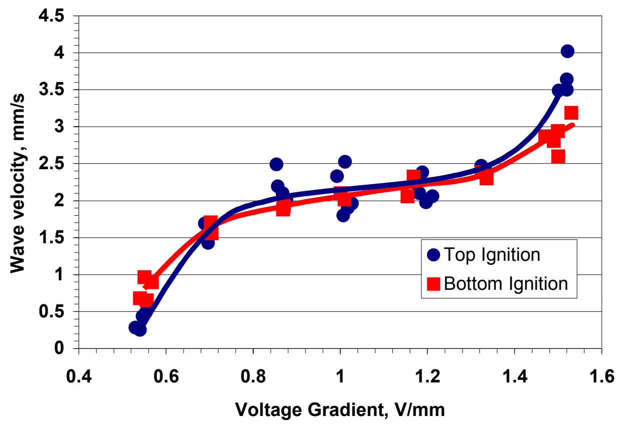

Figure 2. Wave velocity vs. field with coil and igniter layer in the Ti + Al system.

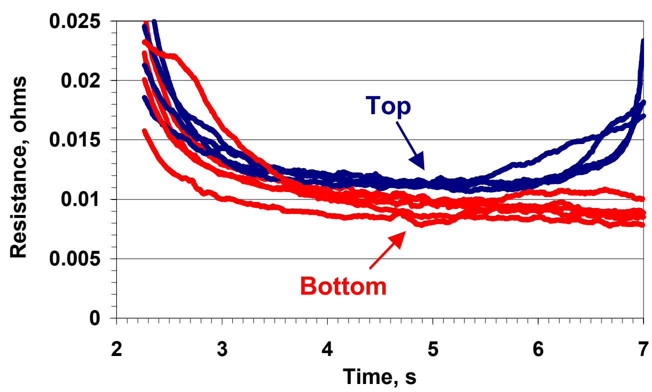

Figure 3. Comparison of resistances for bottom and top ignition at high field strengths (Ti + Al, voltage gradient ≈1.5 V/mm).