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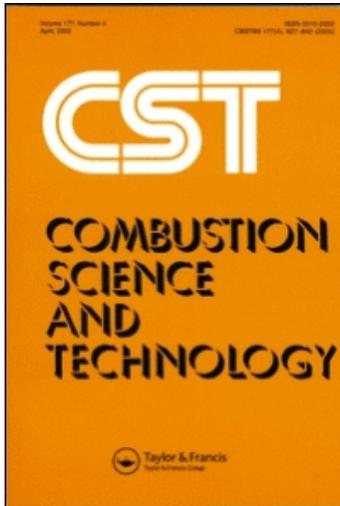
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THE EFFECT OF ADDED Al_2O_3 ON THE PROPAGATION BEHAVIOR OF AN Al/CuO NANOSCALE THERMITE

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Three types of experiments were performed on an Al/CuO nanoscale thermite to understand the effect of adding a diluent (40 nm Al_2O_3 particles) to the mixture: the constant volume pressure cell, the unconfined burn tray, and the instrumented burn tube. The addition of Al_2O_3 decreased the pressure output and reaction velocity in all three experiments. Burn tube measurements showed three reaction velocity regimes: constant velocity observed when 0% (633 mls) and 5% (570 mls) of the total weight is Al_2O_3 , constant acceleration observed at 10% (146 mls to 544 mls over a distance of 6 cm) and 15% (69 mls to 112 mls over a distance of 6 cm) Al_2O_3 , and an unstable, spiraling combustion wave at 20% Al_2O_3 . The pressure measurements correlated to these three regimes showing a dropoff in peak pressure as Al_2O_3 was added to the system, with relatively no pressure increase observed when 20% of the total weight was Al_2O_3 . Equilibrium calculations showed that the addition of Al_2O_3 to an Al/CuO mixture lowered the flame temperature, reducing the amount of combustion products in the gas phase, thus, hindering the presumed primary mode of forward heat transfer, convection.

Keywords: Burning rate; Diluent; Nano-aluminum; Thermite

INTRODUCTION

A thermite reaction is defined as a reaction between a metal and a metal oxide (Wang et al., 1993). This work was focused on the metal-metal oxide reaction, not intermetallics (another name for metal-metal reactions). Using aluminum as the metal fuel with different oxidizers creates highly exothermic reactions that have applications in many areas associated with self-propagating high temperature

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synthesis (SHS) (Wang et al., 1993) and energetic materials (Naud et al., 2003; Pantoya et al., 2004).

Burning velocities for these materials have been shown to be highly dependent on the particle size (Pantoya and Granier, 2005), whereby smaller particles lead to faster burning rates. Thermites with particle sizes on the nanoscale, also referred to as metastable interstitial composites (MICs), can exhibit burning rates up to 1000 m/s (Bockmon et al., 2005). The high burning rates and exothermicity makes these materials of great interest to the combustion community.

One aspect of MIC materials that is not well understood is the mode by which energy is transferred ahead of the reaction front to sustain the propagation, or the propagation mechanism. Five propagation mechanisms may be considered when examining the reaction propagation: radiation, conduction, acoustics, compaction, and convection (Asay et al., 2004). Solid energetic materials are controlled by conduction, when deflagrating, which can be enhanced by radiation (Son and Brewster, 1995; Begley and Brewster, 2007). Acoustics (shock processes) and compaction become important when a reaction produces pressures sufficient to induce volume changes in the material. This occurs during detonation or the transition to detonation. Convection is possible if the material is porous and hot interstitial gas, reactants, or products can be propelled forward through the material by high reaction zone pressures (Kuo et al., 1978; Ershov et al., 2001).

Nanoscale thermites exhibit combustion velocities (~ 1000 m/s) approximately four orders of magnitude greater than that of the micron-scale thermites (~ 0.1 m/s) (Bockmon et al., 2005 and Wang et al., 1993). The drastic increase in velocity is due to the extremely small time scales associated with mass diffusion and reaction rates brought about by the small particle sizes. This does not allow time for any heat loss or depressurization within the reaction zone leading to high pressure, hot gases that can be propelled ahead of the front. Therefore, these systems are thought to be controlled by a convective propagation mechanism (Asay et al., 2004).

Because the controlling propagation mechanism is convection, both gas production and temperature, should be important factors when optimizing for the fastest burning rate. Sanders et al. (2006) observed that for four different metal oxides (Bi_2O_3 , MoO_3 , CuO , and WO_3), the burning rate on a burn tray was maximized at the stoichiometry that also produced the highest peak pressure in the pressure cell. Moreover, equilibrium calculations showed that all of the optimum stoichiometric ratios were related to the gas production and phase of the products. This optimum stoichiometry was found to be fuel rich (~ 1.4) for all of the metal oxides except copper oxide, which optimized at an equivalence ratio close to 1. This difference was attributed to the fact that one of the main products, copper, has a relatively high boiling point of 2835 K and needed the high temperature of a stoichiometric reaction to keep it in the gas phase. This work focused on the reaction of nano-aluminum (nAl) with copper oxide. The global reaction is



where ΔH_r is the heat of reaction based on the mass of the reactants.

Similar to varying the stoichiometry, adding a diluent into the system will decrease the overall temperature of the reaction. Adding the end product, particularly Al_2O_3 , as a diluent was a common practice in micron-scale thermite SHS in order to reduce combustion temperatures and change the mechanical properties of the products (Varma et al., 1992; Munir and Anselmi-Tamburini, 1989). Moreover, the decrease in combustion temperature gave way to slower reaction velocities and decreasing amounts of gaseous species (Wang et al., 1993).

This work focused on the effects of dilution on the combustion properties of the Al/CuO nano-scale thermite. Alumina nano-particles were added in given percentages and their effect on pressure and reaction velocity was studied. A similar study was performed with an Al/MoO₃ system by Foley et al. (2006) from which comparisons are made. The global reaction for this thermite system is given in Eq. (2).



The authors hypothesized that the dilution will lower combustion temperatures, which will decrease gas production and, thus hinder the convective propagation mechanism.

EXPERIMENT

Three experiments were performed to characterize the effects of adding a diluent (Al_2O_3 nano-particles) to an Al/CuO nanoscale thermite or MIC. The pressure cell gave pressure traces for a constant volume explosion, the burn tray yielded a two-point velocity of the reaction propagating through the unconfined material, and the burn tube provided information about the pressure and reaction velocity in confined conditions. Brief descriptions are given here, but detailed experimental descriptions can be found in Sanders et al. (2006).

Materials and Material Preparation

Note: Care should be exercised and small amounts of material used when handling the formulated composites because of their sensitivity to impact, spark, and friction.

Nano-aluminum was purchased from *Nanotechnologies Inc.* (currently, Novacentrix Inc.) and had a nominal particle size of 80 nm with 88% active aluminum (Mang et al., 2006). Particles were assumed to all have a spherical geometry. Copper oxide particles were purchased from Technologies Inc. with all particles assumed to have cylindrical geometry with dimensions of 21 nm × 100 nm. Assumptions on particle geometries are from SEM analysis of the Al and CuO particles (Fig. 1). The alpha-aluminum oxide (Al_2O_3) particles were purchased from *Nanotechnologies Inc.* and had a nominal particle size of 40 nm.

All composites had a ratio of 22% nAl and 78% CuO by mass which is based on the optimization from Sanders et al. (2006). Alumina was added to this system in increments to produce the desired dilution (increments are labeled as percentage of *added* alumina, thus the overall alumina percentage will be higher due to the inherent

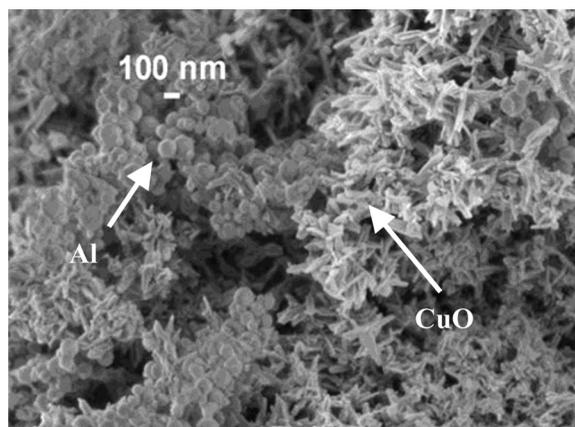


Figure 1 SEM of Al/CuO nanoscale thermite at 25K \times magnification.

alumina shell on the nAl). Mixtures were combined in glass vials and slurried in ~ 12 ml of hexanes. The slurry was sonicated for a total of 1 minute in 0.5 s intervals (50% duty-cycle) at 200 W using a *Heat Systems XL 2020* sonicator. It was then placed in a steel pan and dried on a hot plate at $\sim 48^\circ\text{C}$ for ~ 10 minutes (until material appeared dry). The material was then sieved through a 355 μm mesh to break up any large agglomerates.

Pressure Cell

A modified Parr bomb was used to acquire constant volume pressure traces (Perry et al., 2004). Ignition was achieved by pulsing a 1064 nm Nd:YAG laser (~ 9 mJ) onto the material in the cell via an optical fiber. The free volume of the cell was 13 cm³ and the appropriate amount of material was placed in a cup resting at the bottom of the cell. Pressure measurements were taken at the wall of the cell using a *PCB Piezotronics* piezoelectric pressure transducer with a *PCB Piezotronics* signal-conditioner (model 482A20). Data were recorded at 10 MHz with a *National Instruments* PCI-6115 data acquisition board (DAQ). The q-switch from the laser was used to trigger the data acquisition system.

The mass of MIC material placed in the cup remained a constant value of 17.5 mg irrespective of the amount of added diluent in the system to keep the energy content constant. For example, if an experiment was being performed on a sample with 5% added Al₂O₃ nano-particles, the amount of MIC would be 17.5 mg (22% by mass nAl including oxide passivation, and 78% CuO), but the total mass of material in the cup would be 18.42 mg.

Burn Tray

A loose sample of material (50 mg) was lined up on a metal tray to measure an unconfined reaction velocity (Perry et al., 2004). Two holes, ~ 1 mm in diameter, were 20 mm apart on the base of the metal tray, on top of which the material was

placed. Two optical fibers were placed in these holes and attached on their other end to a *Thorlabs* DET-210 photodiode to detect light emission from the reaction at the beginning and end of the line of material. The distance and time between these two light signals were used to find a two-point velocity. The light from the first photodiode was also used as a trigger for the DAQ system. The data were collected by the same DAQ system described in the previous section. Material ignition was achieved by piezoelectric discharge.

Burn Tube

The burn tube experiment originally designed and used by Bockmon et al. (2005) was used in this experiment with some modifications. This experiment gave a means of measuring the reaction velocity in a confined, more one-dimensional, cylindrical geometry. An acrylic tube used to hold the material, with length of 8.9 cm, inner diameter of 0.32 cm, and outer diameter of 0.64 cm, was placed in a polycarbonate block. Six pressure transducers and optical fiber ports were located on each side of the block at 1 cm intervals.

Materials were loaded into acrylic tubes using a *Cleveland* vibrating block to assure uniform powder density. Packing densities were approximately 6% of the theoretical maximum density (TMD), which corresponded to 250 mg per experiment or 0.36 g/cm^3 . Initiation of the reaction was achieved by means of an exploding bridge wire (EBW), which was placed at one end of the tube and fired by a *Cordin* 640 Pulsor at 1.7 kV. A *Stanford Research Systems* pulse generator was used to manually trigger the Pulsor and DAQ system. The signal from the pulse generator corresponds to $t = 0$ in the experiments. For these experiments two *Tektronix* digital oscilloscopes (models 754D and 7054) with sampling rates of 5 MHz (0.2 μs resolution) were used to acquire the data.

A *Phantom 7.0* high-speed video camera was used to view the luminosity from the reaction wave propagating down the tube. A frame rate of 110,000 frames per second was used at a pixel resolution of 256×32 and exposure time of 1 μs . An appropriate aperture was chosen depending on the experiment to view the reaction without saturation. The pulse generator triggered the camera as well.

RESULTS

Three tests were performed to characterize the effects of added Al_2O_3 nanoparticles on the burning rate of an Al/CuO nanoscale thermite: the pressure cell, the burn tray, and the burn tube. Both the pressure cell and the burn tray gave information about an unconfined burn while the burn tube was in a confined setup. Confinement effects were expected to play a significant role because the convective mode of heat transfer was the controlling propagation mechanism.

Pressure Cell and Burn Tray

A single pressure trace was recorded in the pressure cell for each experiment. Typical results are shown in Figure 2 for varying weight percent of Al_2O_3 . Below 5% added Al_2O_3 , ringing was seen in the record. This was interpreted as shock waves

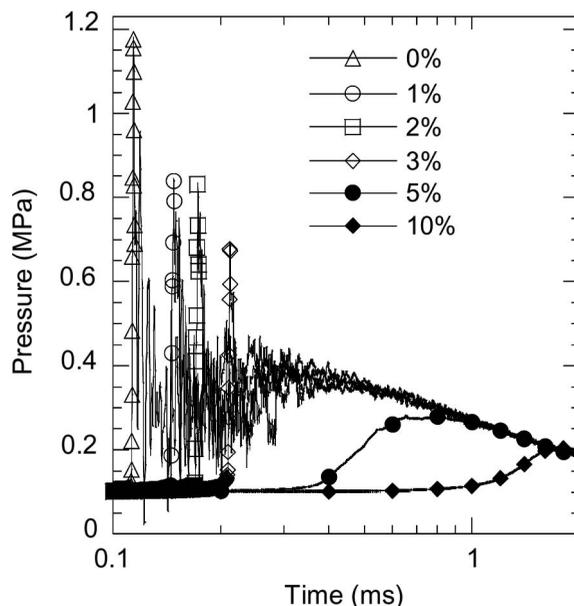


Figure 2 Typical pressure traces from the pressure cell for increasing amounts of Al_2O_3 . Data labels shown only on initial pressure rise for each trace.

reflecting off the walls inside the cell. A drastic change occurred in the pressure traces when 5% Al_2O_3 was added to the mixture whereby the peak pressure and rate of pressure rise (dP/dt) significantly decreased and the induction time (τ_i) significantly increased.

The rate of pressure rise was defined as the difference between the peak pressure and atmospheric pressure divided by the difference between the time of peak pressure and the time where the pressure first rises above atmospheric conditions. Induction time was defined as the time from when the ignition energy was delivered to when the pressure rises above 10% of the peak pressure. Moreover, above 5% added Al_2O_3 there was no ringing in the pressure trace indicating the reaction was slowed to the point that a shock wave was not produced in the gas surrounding the sample.

The Al/ MoO_3 system used in Foley et al. (2006) showed a similar drastic change in behavior for the rate of pressure rise and induction time at 20% added Al_2O_3 (compared to 5% for the Al/CuO system), which indicated that the Al/CuO CuO system was more sensitive to the addition of Al_2O_3 . The peak pressure for the Al/ MoO_3 system using 19 mg, however, was relatively low (0.33 MPa) compared to the Al/CuO system (1.17 MPa) for the sharp rising pressure traces (low percentage of Al_2O_3).

The pressure traces were fairly repeatable for each Al/CuO/ Al_2O_3 case, except for the 4% case indicating the system was on the threshold of a change. The 4% case was not shown due to its extremely high variability in results. Specifically, some results in this range showed a sharp pressure rise with ringing and some resulted in a more monotonic rise in pressure similar to conditions where more Al_2O_3 was

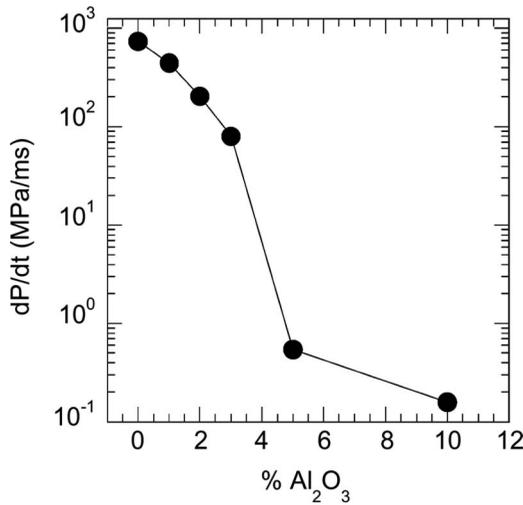


Figure 3 Rate of pressure rise in the pressure cell with increasing weight percentages of Al₂O₃.

added. This distinct transition was indicative of a change in the mode of reaction propagation occurring.

Three experiments were performed for each weight percentage of Al₂O₃ in the pressure cell and values of peak pressure, dP/dt and τ_i were averaged. Figures 3 and 4 show the trends of dP/dt and τ_i , respectively. The slope of the pressure rise dropped two orders of magnitude when the Al₂O₃ weight percentage increased from 3% to 5%, showing two different regimes. The induction time also showed a

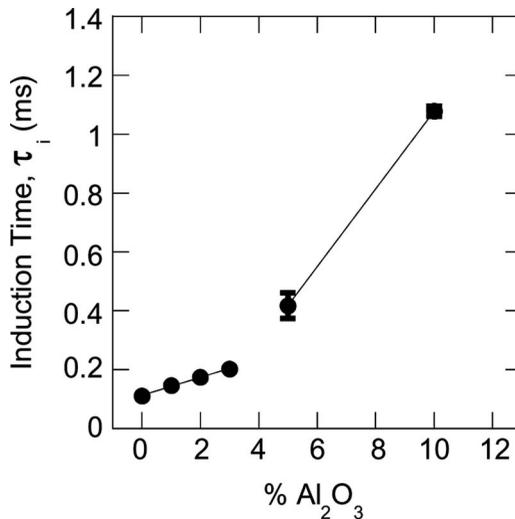


Figure 4 Induction time (τ_i) for the pressure rise in the pressure cell with increasing weight percentage of Al₂O₃.

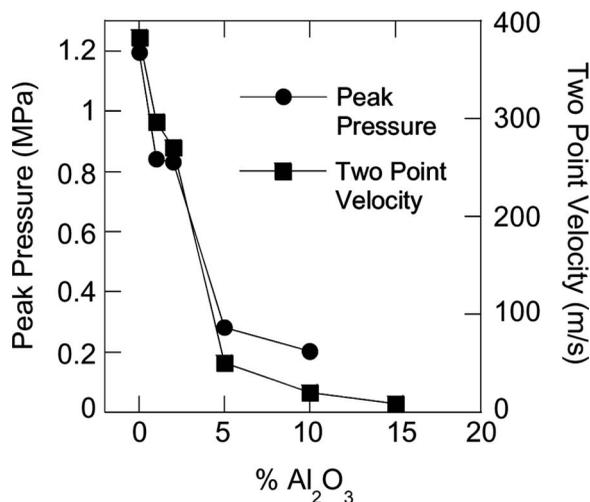


Figure 5 Peak pressure in Parr bomb and open tray velocity result. There is a clear correlation between peak pressure in the pressure cell and two-point velocity on the burn tray with increasing weight percentages of Al₂O₃.

bimodal nature with a significant increase at 5%. Error bars representing a 95% confidence interval are present for all points, although some are hidden by their respective label.

The trend for the peak pressure, when varying the weight percent of Al₂O₃, was similar to that for the two-point velocity in the burn tray experiment as shown in Figure 5. For both, there was an initial drop of ~27% in either peak pressure or velocity with only 1% Al₂O₃ added and then a drastic dropoff of ~75% at 5% Al₂O₃. In contrast, Foley et al. (2006) observed a linear decrease in peak pressure and a change of regimes for velocity at 20% Al₂O₃ (Al/MoO₃ system). Error bars representing a 95% confidence interval are present, but smaller than the data point labels. The correlation between the pressure and velocity was also shown in Sanders et al. (2006) for various stoichiometric ratios. This gave further evidence that the propagation mechanism was closely related to the effects of pressure or gas production.

There was a limit to the amount of Al₂O₃ that could be added to the system for each test. The material in the pressure cell would not ignite with 15% by weight of Al₂O₃ by laser, nor in the burn tray with 20% Al₂O₃ using a piezo-igniter. In contrast, the Al/MoO₃ from Foley et al. (2006) was ignitable with 50% Al₂O₃ using the same ignition systems. The burn tray experiment had a higher threshold for Al₂O₃ because the ignition systems were different and more material was used and less was in contact with its container promoting more heat generation and less heat losses.

Burn Tube

As was shown with the pressure cell and burn tray, this material was particularly sensitive to the addition of Al₂O₃. An addition of only 5% by weight of

Al_2O_3 in the mixture drastically changed the pressure output and propagation behavior. Both of these tests were unconfined, allowing some of the interstitial and combustion product gases to leave the system. The burn tube, however, confined the material laterally so all of the gas remained in the system, with some being propelled forward through the interstitial spaces. The photodiodes detected the light given off by the reaction while the pressure transducers detected the pressure change caused by the heating of the interstitial gases and the gas produced by the reaction.

Light measurements. Six photodiodes recorded the light production in the burn tube each separated by 1 cm. Figure 6 shows a typical sequence of images and light traces from a single experiment. Knowing the distance of each light detector and the time of arrival for the light trace, a position versus time plot and, thus a velocity could be acquired for each experiment. Multiple experiments were performed and averaged for each Al_2O_3 weight percentage. Both the position and time were zeroed at the first position since this study was not concerned with ignition effects. Figure 7 shows a typical graph of position vs. time for each Al_2O_3 percentage. The 0% and 5% experiments were nearly linear indicating a constant velocity; while the 10% and 15% had a second order polynomial fit indicating a non-steady velocity or constant acceleration.

The average velocities for the 0% and 5% cases were $633\text{ m/s} \pm 7\%$ and $570\text{ m/s} \pm 5\%$, respectively. The average accelerations for the 10% and 15% cases were $2,798,400\text{ m/s}^2 \pm 30\%$ and $76,686\text{ m/s}^2 \pm 17\%$, respectively. The initial and final velocities (from the fit) for the 10% added Al_2O_3 case, were 146 m/s and 544 m/s , respectively. Likewise, for the 15% added Al_2O_3 case, the initial and final velocities (from the fit) were 69 m/s and 112 m/s , respectively. The error represents a 95% confidence interval for a small sample. The Al/MoO_3 system first showed non-steady velocity at 50% Al_2O_3 (Foley et al., 2006).

For the experiment with 20% Al_2O_3 not all of the photodiodes recorded light during the experiment. This indicated that the reaction did not pass through the material covering that particular fiber optic and, therefore, the front was not continuous, but consisted of various discrete fronts, or fingers, propagating through

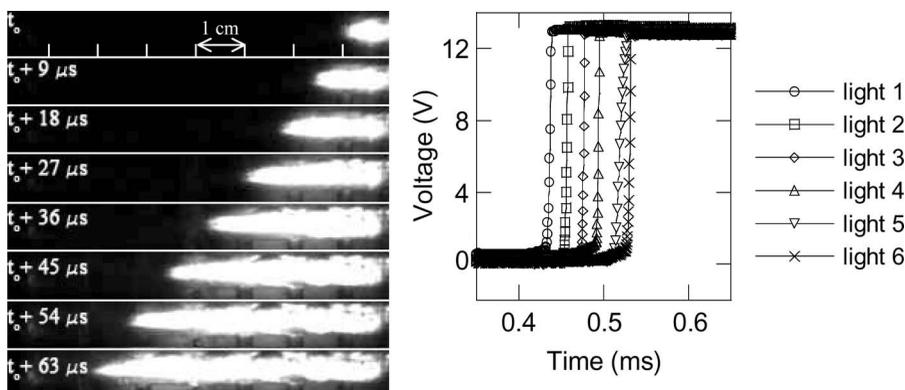


Figure 6 Typical sequence of images and light traces from an experiment with 5% Al_2O_3 .

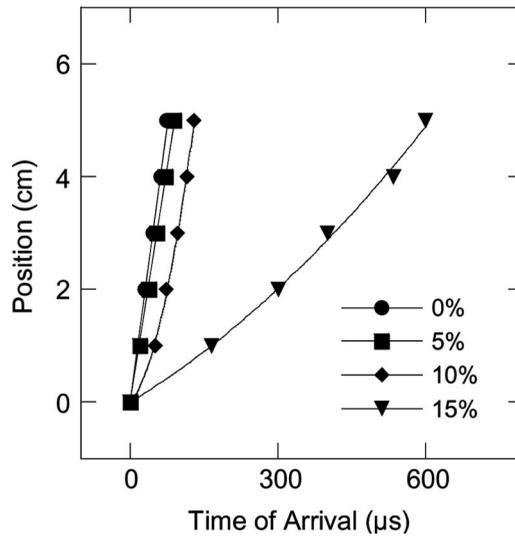


Figure 7 Typical position versus time graphs for reaction waves in the burn tube with various weight percentages of Al_2O_3 .

the material. In fact, images of this experiment in Figure 8 show a completely different flame structure spiraling through the material similar to, but not exactly the same as what was observed by Munir and Anselmi-Tamburini (1989) for diluted classical thermites.

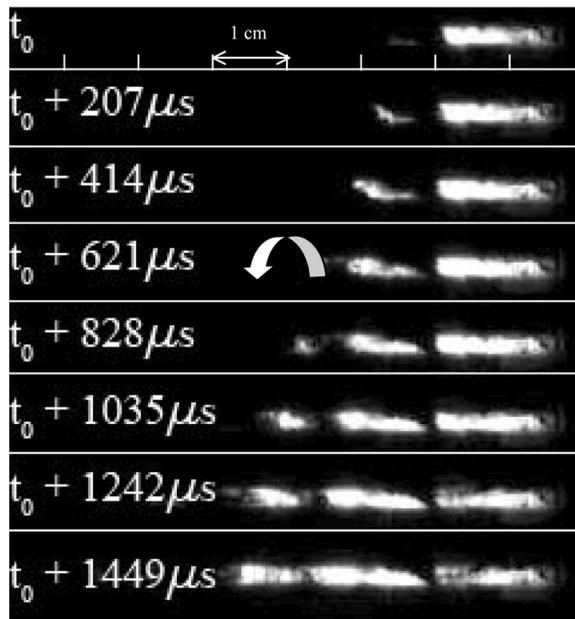


Figure 8 Spiraling combustion instability shown.

This instability was attributed to the diluent decreasing the flame temperature and causing a thermal instability. The spiraling instability observed by Munir and Anselmi-Tamburini (1989) was different in that most of the material was consumed. Moreover, the samples were packed pellets as opposed to loose powder in a tube and the constituents used were not Al/CuO. Preceding this spiraling combustion, a flat and steady, but relatively slow front was observed to have speeds of ~ 5 m/s. This will be discussed further after examining the pressure traces for these unstable experiments.

Pressure measurements. The pressure transducers measured the change in pressure at a particular point due to the reaction forming hot gases and heating up the interstitial air. Ideally, these traces would be flat until the reaction reached the face of the transducer, and would then show a sharp pressure rise with a relatively slow decay as was shown with other experiments (Bockmon et al., 2005; Foley et al., 2006; Sanders et al., 2006). For this experiment, however, at 0% and 5% Al_2O_3 , the traces were not as ideal. Even though most traces showed the discrete pressure rise, some had oscillations prior to the sharp pressure rise. Drops in the pressure to approximately -2 MPa (Fig. 9) were seen, which is due to equipment malfunction (a pressure of -2 MPa relative to atmospheric pressure indicates an overall negative pressure).

Some traces did not have a sharp pressure rise at all and just oscillated around zero with an amplitude of approximately $\pm 2 - 7$ MPa. These were usually found at the 5th and 6th pressure transducer positions and maybe associated with effects from the open end of the tube. The time scale for these drastic pressure drops and

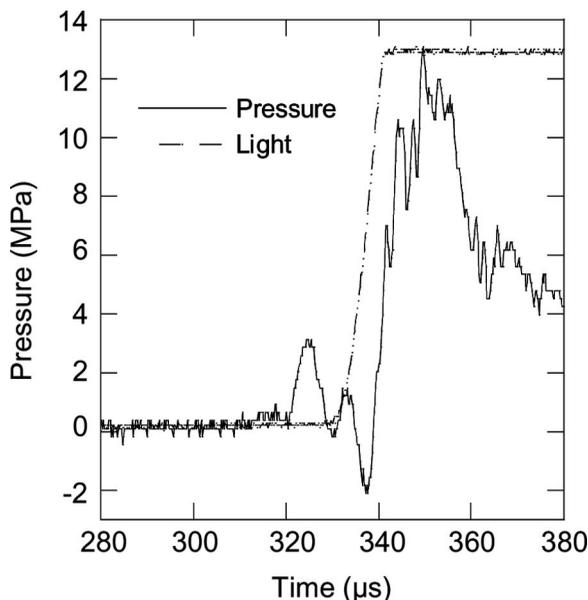


Figure 9 Example of pressure rise before light trace and extreme negative pressure oscillation.

oscillations was $\sim 5\mu\text{s}$, which was too fast to attribute to thermal effects on the pressure transducers. Therefore, this could only be due to mechanical or electrical effects. This issue has not yet been resolved. Because of these unknown inconsistencies in the data, the pressure traces were not used to find a particular reaction velocity, but rather as a measure of the gas production in the system and heating of the interstitial air.

The experiments with 0% and 5% added Al_2O_3 gave peak pressures of $\sim 10\text{ MPa}$ and $\sim 9\text{ MPa}$, respectively, for most positions down the tube. However, at 10% added Al_2O_3 , where the reaction wave velocity became unsteady (constant acceleration), most of the positions showed relatively low peak pressures, $\sim 2\text{ MPa}$, and only a few positions showed pressures between 6 and 8 MPa. Figure 10 shows pressure traces from the 2nd and 5th position of a particular experiment with 10% Al_2O_3 demonstrating this bimodal behavior.

In this particular experiment, the pressure, or gas production, showed a drastic increase at the end of the tube giving an explanation for the accelerating front. The slow reaction in the beginning stages built up gases that would eventually propel the reaction forward and give it this constant acceleration. Presumably, it would eventually reach a steady propagation rate. Similar trends for the pressure were seen for 15% added Al_2O_3 with fewer positions showing pressures of $\sim 8\text{ MPa}$. For 20% added Al_2O_3 , pressure rises of $\sim 2\text{ MPa}$ occurred at all pressure locations for all experiments indicating a significantly lower temperature reaction and more condensed phase combustion products. Much lower gas production for this experiment could be the reason for the slow and highly unstable reaction front.

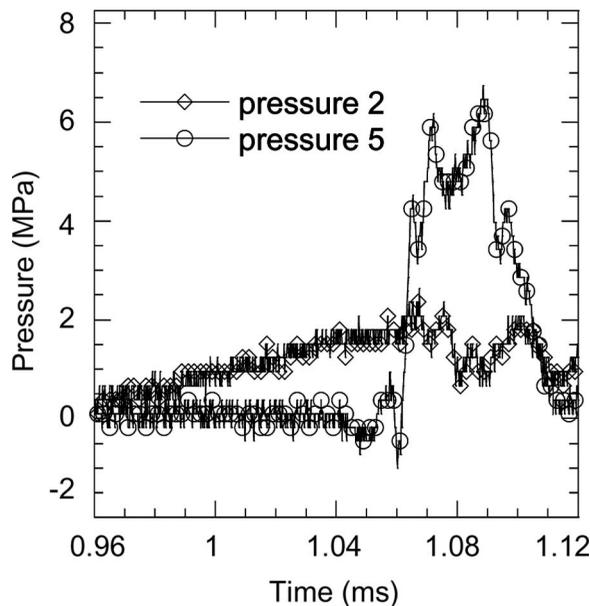


Figure 10 Pressure traces at position 2 and 5 for an experiment with 10% Al_2O_3 showing the bi-modal characteristic with the high peak pressures at the later positions down the tube.

As Al_2O_3 was added to the system, the reaction in the burn tube changed from a steady rate (0% and 5% by weight), to an accelerating regime (10% and 15%), and finally, to an unstable regime (20%) where a relatively slow burning velocity was observed, which eventually transitioned to a spiraling combustion wave. The pressure traces gave indication that the gases being produced by the hot reaction, such as vaporized aluminum, unstable intermediates such as AlO or Al_2O or stable products such as Cu gas, were condensing or not even forming due to the lower adiabatic flame temperatures. Therefore, this did not allow for hot gases to be propelled forward to drive the reaction down the tube convectively. This all occurred at lower levels of Al_2O_3 compared to the Al/MoO_3 system.

Thermodynamics

Equilibrium calculations were performed using CHEETAH 4.0 (Fried et al., 2004) with the JCZS product library developed by Hobbs and Baer (1999). Because of the localized pressure rises seen when the reaction passes a transducer, the system was thought of as small cross-sections of material burning in a nearly constant volume. Therefore, the calculations were performed assuming a constant volume. The calculations included the Al_2O_3 from the passivation layers and the interstitial air, but did not include the effect of heat loss through the walls.

It was reasonable to neglect the heat loss because the time scale for conduction through the acrylic tube, τ_{loss} , was much greater than the time for the reaction to propagate through all of the material, τ_r . The time scale for heat loss was defined as $L^2/\alpha_{\text{acr}} = 80$ s, where L is the length scale for conduction defined as the wall thickness of the tube, 3.2 mm, and α_{acr} is the thermal diffusivity of acrylic, $0.137 \text{ mm}^2/\text{s}$. The time for the reaction to propagate through all of the material was $L_{\text{tube}}/v_r = 127 \mu\text{s}$, where L_{tube} is the length of the tube (8.9 cm) and v_r is the velocity of the reaction wave ($\sim 650 \text{ m/s}$).

Figure 11 shows the calculations and averaged experimental values for peak pressure as the weight percent of Al_2O_3 increased. There was a fair agreement between calculations and experiments, which gave a first order validation to these calculations and allowed them to be used to help interpret some results. Figure 12 shows the adiabatic flame temperature as a function of the weight percentage of Al_2O_3 . As expected, adding Al_2O_3 decreases the adiabatic temperature.

To understand the primary mechanism for propagation, the calculated product concentrations and their phases were examined. The concentrations of the primary products as a function of Al_2O_3 concentration are shown in Figure 13. Even though most of the products were in the condensed phase, the depletion of the gas phase products (bottom half of Fig. 13) was what most effected the propagation. Examination of the total gas phase products showed an 82% decrease to a mole fraction of 0.004 when increasing from 0% to 20% Al_2O_3 .

Furthermore, at values above 15% Al_2O_3 , the equilibrium results suggest that the nAl reacted with the interstitial air to form solid AlN , thus removing some of the initial interstitial gas from the system. These results further suggest that the addition of Al_2O_3 lowered the flame temperature, which demoted gas production, as well as the overall energy release rate, and began to remove interstitial gases from the system, lowering the pressure and, thus, hindering the convective mode of heat transfer.

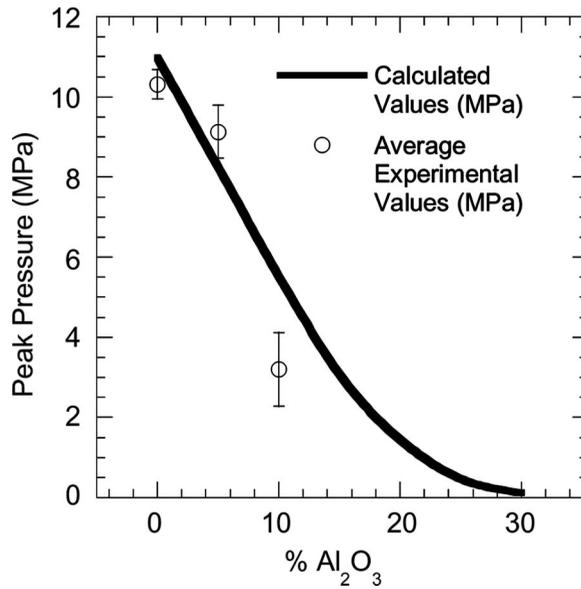


Figure 11 Comparison of average experimental peak pressure values in the burn tube to the calculations for the peak pressure with added Al_2O_3 percentage for constant volume explosion.

Since convection was the primary propagation mechanism for this system, the reaction velocity decreased drastically and eventually became unstable at 20% Al_2O_3 where gaseous species were almost non-existent.

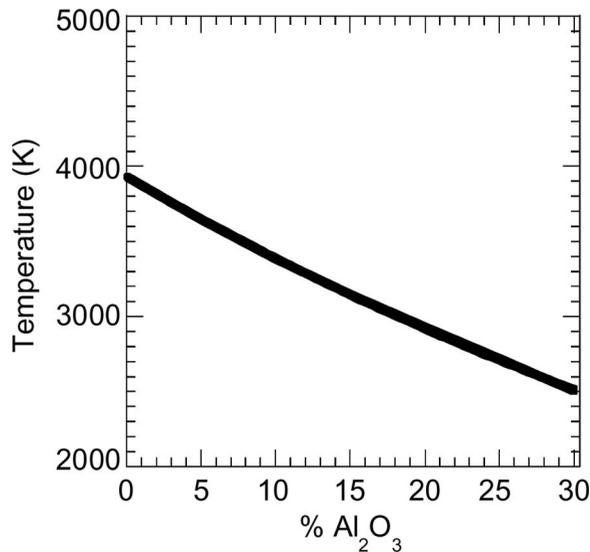


Figure 12 Calculated adiabatic flame temperature as a function of added Al_2O_3 percentage for constant volume explosion calculations.

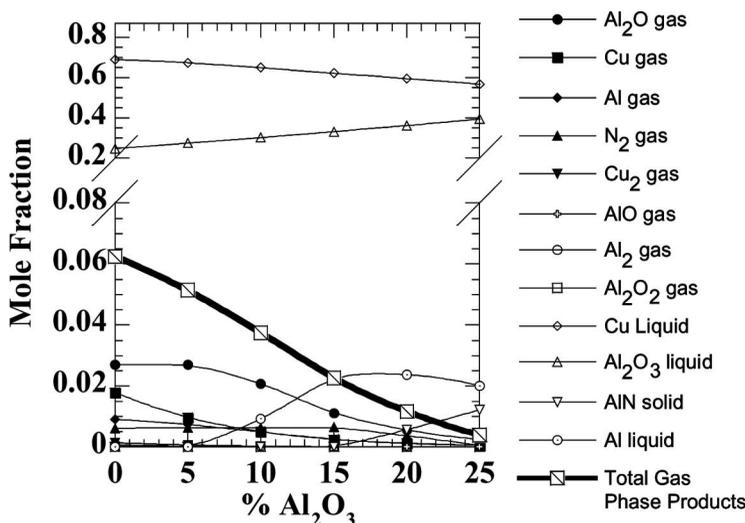


Figure 13 Product concentrations versus Al₂O₃ percentage for constant volume calculations.

CONCLUSIONS

This work focused on the effects of dilution of an Al/CuO nano-scale thermite with Al₂O₃ nano-particles. These solid phase end-products acted as a heat sink, which reduced combustion temperatures. This, in turn, hindered gas production and decreased the overall effect of the convective propagation mechanism. Three tests were performed to examine how propagation velocities and pressure output varied as a function of diluent (Al₂O₃ nano-particles) concentration. The pressure cell and burn tray were unconfined tests while the burn tube confined the material laterally.

The pressure cell showed a dramatic drop in peak pressure and rate of pressure rise when only 5% Al₂O₃ was added to the system. Moreover, the induction time drastically increased when the Al₂O₃ concentration exceeded 5%. These trends suggested that the addition of Al₂O₃ nanoparticles hindered the gases produced by combustion. The peak pressure trend corresponded directly with the two-point velocities found on the burn tray, which gave evidence that hindered gas production decreased the reaction velocity.

In the instrumented burn tube, the peak pressures were recorded as the reaction wave propagated through the material. These pressure traces demonstrated a clear dropoff as Al₂O₃ concentration increased, eventually showing relatively no pressure increase at 20% Al₂O₃. Furthermore, the propagation velocity changed from a steady rate of ~650 m/s for 0% and 5% Al₂O₃ to an unsteady velocity (constant acceleration) regime for 10% and 15% Al₂O₃, and finally became unstable at 20% Al₂O₃.

The unstable regime at 20% Al₂O₃ initially showed an extremely slow propagation that eventually transitioned into a spiraling propagation through the material. Similar spiraling phenomena were observed with micron-scale thermites when a diluent was added to the system (Munir and Anselmi-Tamburini, 1989).

This change in propagation behavior and pressure output in the burn tube with added diluent was interpreted utilizing the results from equilibrium calculations. The peak pressures predicted by constant volume calculations matched well with the peak pressures recorded experimentally in the burn tube for each diluent concentration. Therefore, these types of calculations were used throughout to explain other phenomena.

Calculations showed the total gas production dropping 82% to a mole fraction of 0.004 when increasing from 0% to 20% Al₂O₃. This drastic depletion in gaseous species predicted by the calculations at 20% Al₂O₃ correlated well with the experimental propagation behavior. The reaction front drastically decreased in velocity and eventually demonstrated a spiraling combustion instability. This instability was a direct result of the primary mode of heat transfer, convection, being prohibited.

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