Combustion of Metal Particles in Gaseous Detonation Products

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1 Introduction

The addition of reactive metals (such as aluminum, magnesium, etc.) to condensed explosives in order to increase the total energy release of the explosive is now common practice. Although the metals (typically in powdered form with diameters ranging from submicron to millimetres) do not react quickly enough to contribute to the detonation front itself, they can react in the products of the condensed-phase explosive or in the surrounding atmosphere, significantly contributing to the strength of the blast wave. The energy release of metal combustion with air (20-30 kJ per g of metal) compared to the energy release of an explosive can be realized if the reaction of the metal particles can be organized to occur sufficiently fast to contribute to the blast wave.

Many outstanding questions remain in such systems. For example, it is not clear when and where the particles ignite, whether they react with shock-heated air or detonation products, etc. It is difficult to obtain detailed quantitative data related to this phenomenon because of the nature condensed-phase detonations. The approach taken here to gain some insight and understanding into the phenomenon is to simulate the high-explosive detonation products with gaseous detonation products. Metal particles are injected into a gas detonation tube and their behaviour can be monitored. In this way more quantitative data can be obtained.

Previous work [1] had suggested that aluminum particles in the present experimental conditions may be reacting in the kinetic regime rather than the diffusive regime. For this reason, the aim of the present study is to estimate the combustion temperature of the aluminum particles to determine in which regime the chemical reaction takes place. In the diffusive regime, the particles should be at a constant temperature, significantly higher than the flow, while in kinetic regime, the particle temperature should follow quite closely the flow temperature.

2 Experimental Setup

The experimental apparatus consists of a 6.35-cm-inner-diameter steel tube, 60-cm-long. One end of this tube is closed with a blind flange and equipped with a high-voltage weak spark igniter. The igniter is powered by a 30 kV trigger module. The tube is instrumented with two PCB pressure transducers for characterization of the detonation process and determination of time zero.

The other end of the tube is sealed by a pneumatically actuated gate valve (see Fig. 1). Opposite the gate valve is a clear acrylic test section approximately 1.6 m long with its far end open. The detonation tube is first evacuated and then filled to 1 atmosphere with a stoichiometric mixture of hydrogen and oxygen. The mixture is prepared by an in-line mixing technique with choked orifices to maintain the flow rate constant irrespective of

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the downstream pressure. The mass flow rates of hydrogen and oxygen are measured with two FVL-1600 type mass flow meters (Omega Eng. Inc.). Metering valves that serve as critical orifices allow precise adjustment of the gas flow rates. The oxygen and hydrogen flows are mixed inside a 150 mL stainless steel cylinder filled with $\frac{1}{4}$ inch stainless steel balls. The quality of the flow control and mixing were verified by measuring oxygen concentration with a NOVA 370 (Nova Analytical Systems Inc., Hamilton, ON) oxygen analyzer.



Figure 1. Schematic of the experimental apparatus. A gate valve separates the steel driver section on the right from the clear acrylic test section on the left.

A sample of approximately 50 mg of metal powder is placed on a knife blade located in the test section, 50 mm from the gate valve. The gate valve is opened (the opening time is less than a second). Upon reaching its fully open position, the gate valve activates a micro switch that triggers the ignition system. After a short run distance, a detonation is formed and propagates through the driver section. Upon arrival of the detonation at the air interface, a blast wave is transmitted into the test section.

A Canon reflex camera is focused on the test section located 120 cm away. The distance between the point of acquisition and the gate valve was varied in the study. Light from the test section image is collected and transferred to a miniature Ocean Optics USB-4000 spectrometer via a 5-m long, 100-µm optical fiber (see Fig. 1). The estimated spectral resolution of the spectrometer-fiber system is approximately 2 nm. The field of view of the optical system at 120 cm is approximately 3 mm. While the spectrometer's linear CCD array is sensitive in the range of 370-1100 nm, the practical spectral interval is limited to approximately 400-850 nm due to the strong absorption of the acrylic tube in the UV and near IR ranges. All experiments discussed in the present study were done with 100 ms electronic exposure time. This is essentially an open-shutter technique since the duration of the luminous event is approximately 3-5 ms.

Figure 1 is a schematic of the experimental apparatus illustrating the gate valve, the knife blade and the optical system. The aluminum powder used was H50 obtained from Valimet Inc.



Figure 2. Typical spectrum of H50 aluminum powder obtained at 50 cm from the gate valve.



Figure 3. Typical spectrum of aluminum oxide powder obtained at 50 cm from the gate valve.

3 Spectrometer Calibration

The spectral sensitivities of the spectrometer's CCD and optical system are complex functions of wavelength. The cumulative sensitivity function was derived using the factory provided spectral emissivity of an Ocean Optics LS-CAL calibration lamp (tungsten halogen with Teflon diffuser). The spectral transparency of the acrylic tube was determined from the calibrated lamp with and without the acrylic tube. All results were corrected for the spectral aberrations introduced by the acrylic tube.

4 Results

In all experiments discussed, a steady CJ detonation was observed with the pressure transducers. The detonation velocities recorded varied between 2740 and 2790 ms ($V_{CJ} = 2834$ m/s).

Figure 2 shows a typical spectrum of H50 aluminum powder. The spectrum shows a well developed system

of AlO bands in the range of 450-550 nm superimposed on a strong continuum background. Also visible are non-resolved sodium doublets (589.0 and 589.6 nm) and partially resolved potassium lines (766.5 and 769.9 nm).

For comparison, Fig. 3 shows a typical spectrum obtained with 50 mg of pure (99.9%) submicron (< 0.1 μ m) aluminum oxide powder (γ -Al₂O₃). Aluminum oxide serves as an inert control experiment. The aluminum oxide spectrum shows a very weak continuum background that is difficult to separate from strong lines and bands. Therefore, it cannot be used to estimate temperature. The spectrum also shows the sodium and potassium lines.

Figure 4 shows the spectrum of a control experiment with no powder on the blade. The spectrum shows no continuous background. Again, sodium and potassium lines are clearly visible.



Figure 4. Typical spectrum of H50 aluminum powder obtained at 50 cm from the gate valve.

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Figure 5. Intensity of the continuous background at 650 nm (region clean of lines and bands) normalized by the intensity of the peak potassium line.

Experiments were performed with aluminum powder, inert aluminum oxide powder and without powder with the spectrometer focused at 11, 50 and 100 cm downstream of the gate valve. Figure 5 shows the intensity of the background at 650 nm (region with no lines), normalized with the peak intensity of the potassium line (visible on all spectra). In this way, a qualitative comparison of temperature can be obtained between cases with no powder, aluminum powder and aluminum oxide powder. The relative intensity is always higher in the aluminum case as compared to the oxide. This is to be expected as a result of chemical reaction of the aluminum powder with the water vapour. The relative intensity of the aluminum oxide case is also higher than the no powder case where the intensity at 650 nm is negligible, presumably because there are no condensed emitters. Both the aluminum and oxide cases show peak intensity near 50 cm. This is probably due to the fact that it takes a certain time to heat up the particles. It is possible to estimate the temperature with the

spectra obtained with aluminum powder by polychromatic fitting to Planck's law. Planck's law of black body radiation can be rearranged in the following form:

$$\ln\left[\frac{i(\lambda,T)\lambda^{5}}{\varepsilon(\lambda,T)2hc^{2}}\right] = -\frac{hc}{kT}\frac{1}{\lambda}$$

where ε is the emissivity of the condensed emitter. In the case of aluminum combustion, the main condensed emitter is likely to be the nano-size (≤ 100 nm) aluminum oxide formed as a result of further oxidation of gaseous aluminum suboxides (AlO, AlO₂, etc.). For such small particles (smaller than the wavelength), the emissivity is inversely proportional to the wavelength (Rayleigh limit [2]). Furthermore, the absoption index of molten aluminum oxide is also inversely proportional to the wavelength in the 400-1000 nm range [3], suggesting that the spectral emissivity of the of oxide particles is inversely proportional to λ^2 [4]. If this is the case, a good linear fit should be obtained when plotting $\ln(i\lambda^7)$ as a function of $1/\lambda$. The slope of this straight



Figure 6. Close to grey character (straight line) of the continuous spectrum in the 550 to 770 nm spectral interval.

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Figure 7. Temperature estimate by polychromatic fitting of the continuous spectrum to Planck's law (H50 Al at 50 cm from the gate).

line is inversely proportional to temperature.

Indeed, it is found in all cases with aluminum powder that a good linear fit can be obtained in the range of 630 to 760 nm (region clean of lines and bands). Figures 6 and 7 are typical spectra fit to Planck's law. This procedure was done for every experiment with aluminum powder. It should be noted that because of the strong dependence of the spectral intensity with temperature, the calculated temperature can be attributed to the maximum temperature of the condensed emitter during the time of acquisition of the spectrometer.

Figure 8 shows the temperatures estimated from the aluminum combustion spectra as a function of distance along the test section. While there is considerable scatter, the results show a clear trend of decreasing temperature with increasing distance. The estimated temperatures decrease by approximately 500-1000 K over 100 cm of test section. Also appearing on this figure are two data points (open squares) representing spectra obtained outside of the test section (the data



Figure 8. Temperatures estimated by polychromatic fitting to Planck's law as a function of distance along the test section.

point at 100 cm was obtained with a 75 cm long test section, and the data point at 126.5 cm was obtained with a 120 cm long test section). It appears that further cooling occurs beyond the exit of the tube but there is no dramatic change of trend.

The adiabatic flame temperature was calculated using NASA's equilibrium code CEA [ref]. This value (4020 K) is indicated on Fig. 8 by a horizontal dashed line. The temperatures estimated with the spectra are significantly below this temperature (500-1500 K).

Finally, Solver II [ref], an unstructured Euler CFD code was used to simulate the flow field. The trajectory of a single 50 µm particle was then calculated in this flow field. The solid blue curve on Fig. 8 represents the flow temperature experienced by that particle. The flow starts out near the CJ temperature (3673 K), and then cools as it expands into the test section. Note that the expansion is only planar and therefore not that severe. It appears that the aluminum combustion temperatures estimated by spectroscopy follows the flow temperature: both appear to cool by 500-1000 K over the 100 cm of test section.

5 Discussion and Conclusions

Metal particles can react in two distinct regimes: kinetic or diffusive. In the diffusive regime, the particle temperature is significantly higher than the flow temperature, the kinetics can be assumed to be infinitely fast and the reaction rate is limited by the diffusion of oxidizer to the particle. In the kinetic regime, the diffusion of oxidizer and heat are essentially infinitely fast and therefore, the reaction rate is limited by the particle temperature (chemical kinetics) which is close to the flow temperature. Small particles and large relative velocity between the flow and the particle increase the convective rates and promotes the kinetic regime. While 50 μ m particles may not be considered small for typical experiments of aluminum combustion in quiescent (or near quiescent) flow (reflected shock tubes, flat flame burners, etc.), the very high-speed flow (~ 1000 m/s) may lead the particles to react in the kinetic regime. The fact that the measured temperatures agree more with the flow temperature suggest that the particles may be reacting in the kinetic regime in the present experiment.

Previous work where light emitted by the reacting particles was observed showed that it is difficult to find an ignition event. Also, the combustion times that were estimated from these results did not agree with the d^2 law for combustion in the diffusive regime. All these findings also tend to suggest that particles in the present experiment may be reacting in the kinetic regime, rather than the more familiar diffusive regime.

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