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Heat transfer effects in nano-aluminum combustion at high temperatures

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ABSTRACT

Recent measurements of nano-aluminum combustion in which burning time and peak particle temperature are measured simultaneously have suggested that heat transfer models currently used for burning nanoparticles may significantly overestimate heat losses during combustion. By applying conventional non-continuum heat transfer correlations to burning nano-aluminum particles, the observed peak temperatures, which greatly exceed the ambient temperature, should only be observable if the burning time were very short, of the order of 1 μs, whereas the observed burning time is two orders of magnitude larger. These observations can be reconciled if the energy accommodation coefficient for these conditions is of the order of 0.005, which is the value suggested by Altman, instead of approximately unity, which is the common assumption. Experimental data obtained in the heterogeneous shock tube under a wide array of conditions are compared with basic heat transfer models, and the agreement of both peak temperature values and emission intensity traces for low energy accommodation coefficients supports the hypothesis of Altman and co-workers.

1. Introduction

Nano-aluminum combustion is an active area of research due to the potential to improve performance in propellants and explosives [1,2]. The mechanism of nano-aluminum combustion remains poorly understood. For larger particles burning in the diffusion limit, a fair understanding of the ignition and combustion characteristics has been demonstrated [3] such that predictive simulations are possible. However, for particle sizes approaching the micron scale under most conditions, many of the trends observed in large particle combustion no longer apply. Burning rates begin to deviate from a d^2 law, with exponents curiously observed to be less than unity [4–6]. The pressure dependence of the burning rate becomes significant [7], and there is evidence that the relative oxidation efficiencies of CO_2 and H_2O change [4]. Peak combustion temperatures begin to decrease, and ignition temperatures are also markedly lower [8]. For nano-scale Al, a significant ambient temperature dependence on the burning rate emerges [9].

Several modeling efforts on n-Al combustion have occurred [8,10,11] and some observations have been reconciled. However, a robust model capable of simulating combustion kinetics over a wide range of conditions has not yet been achieved. A common assumption in particle combustion is that as particle size decreases, particle combustion transitions from a gas-phase diffusion limit to a mode of combustion limited by surface reaction or solid-state diffusion. In this classic latter limit, there is no gas-phase combustion, and species concentration and thermal gradients approach zero. Due to rapid heat transfer of small particles, the particle temperature does not significantly exceed that of the ambient gas. Indeed for some conditions, e.g., n-Al burning in CO_2, negligible temperature overshoots were observed in previous work. However, under other conditions with more efficient oxidizers at higher pressures, significant rises in particle temperature were measured [9].

For nano-scale particles in most environments, Knudsen number effects on heat transfer cannot be neglected since $Kn \sim .001–1$. These effects are considered in laser induced incandescence (LII) experiments to determine particle size distributions of nano-particles in flows [12] and must also be considered when determining the transient thermal profile of a combusting nano-aluminum particle. However, use of non-continuum heat transfer expressions requires estimation of the energy accommodation coefficient (EAC). For LII experiments involving carbon particles, values of 0.4 have been measured [13], and thus it is common to use similar values (or even a value of 1) for other materials under similar conditions. However, theoretical and experimental work performed by Altman suggests that at high particle and gas temperatures, certain metal and metal oxide nano-particles have very small energy accommodation coefficients [14,15]. Radiation consequently becomes a more significant pathway for heat transfer in the low accommodation coefficient regime. The experimental work

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by Altman et al. was performed using laser irradiation to heat up nanoparticles generated in a flame. The energy accommodation coefficient was found to be near 0.005 which agreed nicely with their theoretical upper limit [14]. Nano-aluminum particle combustion may potentially experience a similar thermal isolation effect in many applications. With such low accommodation coefficients the heat transfer from the particle via collision with gas molecules becomes inefficient, leading to particle temperatures much higher than those expected using EAC $\sim$ 1.

In this work we perform experiments on nano-aluminum combustion to monitor the particle temperature, burn rate, and emission spectra. High particle temperatures and longer burn times are expected for a transition to the free molecular regime accompanied by a low energy accommodation coefficient. For ultrafine particles, classical theory predicts that rapid heat transfer results in combustion temperatures that only minimally exceed the ambient temperature, even when common Knudsen number correlations are used for Nusselt number calculation [12]. Prediction of the particle temperature requires specification of the reaction rate (i.e., heat release rate) in addition to the heat transfer coefficient. In this research the experimental data on burning time and temperature are supported by a simple model of nano-aluminum combustion that employs as few limiting assumptions as possible, focusing only on the energy balance leading to particle temperature rise. Multiple heat transfer models are considered to determine the predicted transient particle temperature and to see if the nano-aluminum particle experiences thermal isolation from the surrounding gas.

2. Experimental set-up

The nano-aluminum particles were investigated using a heterogeneous shock tube described in detail in a previous publication [16]. The shock tube is capable of producing controlled high temperature and high pressure environments with various gas compositions. Temperatures greater than 4000 K and pressures above 30 atm are achievable behind the reflected shock with test times near 2 ms. Low temperatures were used for this study to test nano-aluminum combustion which has been shown to ignite at temperatures below 2000 K [9]. The combustion of the nano-aluminum particles was monitored behind the reflected shock in order to achieve the high pressures desired. The pressure was varied between 3.5–20 atm to determine the effect of the oxidizer concentration on the particle temperature and burn time.

The shock tube has an 8.4 m driven section and an 8.9 cm internal diameter. A converging dual diaphragm system separates the high pressure helium gas from the oxidizing environment. The driver to driven pressure ratio is controlled to produce the desired shock strength. The velocity of the shock is measured using four piezoelectric pressure transducers at different axial locations. The ambient temperature and pressure of the gases trailing the incident and reflected shock are calculated with the Gordon-McBride equilibrium code [17] using the known initial driven pressure, composition, and measured shock velocity. The test time of the shock tube can also be found from the pressure transducer traces and is typically near 2 ms.

A schematic of the shock tube operation with radial injection and the end section with fiber optic view ports is shown in Fig. 1. This end section allows for photodiode access at multiple axial locations. The end wall has a sapphire view port for further optical access. The fiber optic section is described in further detail in a previous publication [9]. Each test was run with three photodiodes monitoring different axial locations centered at the location of aluminum particle stagnation behind the reflected shock. A nano-aluminum particle has a very small Stokes number, and therefore the particle accelerates quickly behind the incident shock and stagnates within a few microseconds behind the reflected shock. For this reason particle motion behind the reflected shock is neglected.

Particles are injected radially into the test gas prior to diaphragm rupture using a pneumatically driven piston. The particles become entrained in the gas flow behind the incident shock and are swept towards the end-wall until the reflected shock stagnates them and they combust. Four particle classes were chosen to vary particle diameter while measuring burn time, temperature, and emission spectra.

A Hitachi S-4700 high resolution scanning electron microscope (SEM) was used to accurately characterize the particle size distribution of each sample. Over 100 particle diameter measurements were made from each sample in order to obtain a distribution. Table 1 shows the number average and mass average particle diameters of each nominal sample powder. The SkySpring 18 nm particles were not characterized because the resolution required

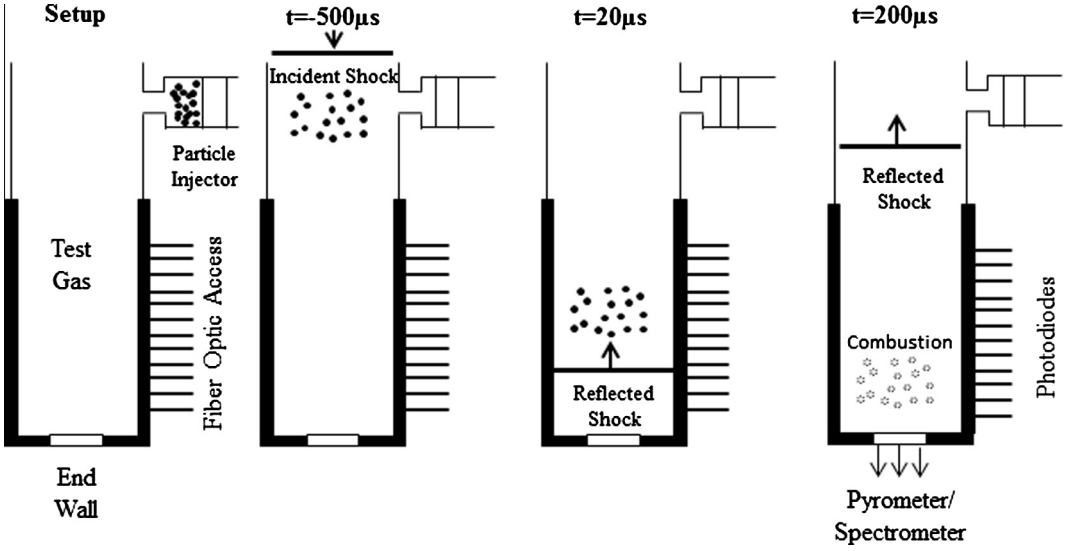

Fig. 1. Schematic of shock tube test section with fiber optic access.