An experimental study of the combustion of an aerosol of coarse magnesium particles in microgravity is reported. Particles with sizes between 180–250 μm were aerosolized in a 0.5-L combustion chamber and ignited in a constant-pressure, microgravity environment. Two flame images were produced simultaneously using interference filters separating adjacent MgO and black body radiation bands at 500 and 510 nm, respectively. The characteristic MgO radiation was used as an indicator of the gas-phase combustion. Comparison of the two filtered flame images showed that preheat and combustion zones can be distinguished in the flame. Experiments have also shown that in microgravity the flame speed depends on the initial particle speeds varied in the range of 0.02–0.4 m/s. This dependence is, most likely, due to the role the moving particles play in the heat transfer processes. Product analyses showed an oxide coating on the surfaces of particles collected after experiments in which the flame speeds were higher than 0.1 m/s. No oxide coating was detected in the products collected after experiments in which a slower flame propagation was observed. However, the particles collected after such experiments contained significant amounts of dissolved oxygen. Strong MgO radiation and production of dense MgO smoke clouds were observed in all the experiments, including those with the slowly propagating flames. Therefore, it has been suggested that the MgO produced in the vapor-phase flame is not the primary source of the MgO coating found on the burnt particle surfaces. An alternative mechanism of forming the oxide coating is, consistent with the earlier single metal particle combustion studies, via the formation of a metal–oxygen solution followed by a phase separation occurring within the burning particles. © 2000 by The Combustion Institute

INTRODUCTION

Mechanisms of combustion and flame propagation in two-phase systems and, in particular, in metal aerosols are of considerable interest because of numerous practical implications for propulsion, explosives, pyrotechnics, incendiaries, flame synthesis, and fire suppression techniques. Combustion of single particles of condensed fuels, e.g., hydrocarbons, coals, plastics, and metal particles has been extensively studied and modeling of the two-phase combustion system is usually based upon the single particle combustion mechanisms. However, some intriguing questions about combustion in the two-phase systems remain: (1) Do particle–particle and particle assembly–flame interactions affect and alter the single particle combustion processes in aerosol flames? (2) What are the specific particle–particle and particle–flame interaction effects that are significant in combustion of a two-phase system?

The microgravity environment can be exploited to address experimentally these questions. Specifically, a two-phase system using particles large enough to be observable, can be produced and ignited. In normal gravity, large particles would quickly fall down and the lifetime of such a two-phase system would have been very short. However, in microgravity, flame propagation through such an unusual two-phase system can be established, and the single particle combustion and the bulk two-phase system combustion mechanisms can be correlated with each other using experimental observations. The combustion can be analyzed adopting techniques developed for single particle combustion studies. With this approach in mind, an experimental apparatus has been designed and built and microgravity experiments...
have been conducted with aerosolized coarse metal powders burning at constant pressure [1–3]. Magnesium was used in the first experiments because Mg single particle combustion has been extensively studied and because Mg particles ignite readily, making the microgravity experiments simpler and more reliable.

For an aerosol of coarse magnesium particles, particle motion due to drag forces was found to change the particle number density (and, thus, local equivalence ratio) in the unburnt aerosol during the constant pressure flame propagation experiments [2]. Interestingly, a similar observation has been reported for microgravity combustion of aerosolized iron particles [4]. Two luminous zones could be distinguished in the Mg aerosol flame propagating in microgravity based on the radiation spectra and intensity profiles [2]. Red color dominated in the radiation of a luminous flame zone adjacent to the unburnt aerosol, and green color dominated in a more bright zone adjacent to the burnt aerosol. Because the peak of the characteristic MgO radiation lies in the green part of the spectrum, it has been suggested in Ref. 2 that the two zones can be identified as the preheat and combustion zones.

This paper presents further experimental results characterizing combustion of a magnesium aerosol in microgravity. In addition to optical diagnostics, combustion products collected after microgravity experiments have been analyzed. Results of this work are compared with the recently reported experiments on single Mg particle combustion [5] that have indicated that oxygen dissolution and phase changes occurring in a burning particle affect the single particle combustion scenario and product composition and morphology.

**EXPERIMENTAL**

The microgravity experiments are conducted using the 2.2-s NASA Glenn Research Center drop tower. Aerosol is produced and ignited in a 0.5-liter, rectangular combustion chamber with a narrow, 19-mm-wide central section through which the flame propagates. The chamber is built of aluminum and is equipped with two Lexan windows, three gas-ports, and two electrical feedthroughs. The chamber’s floor is a silicone rubber membrane which aerosolizes the powder using acoustic oscillations transmitted from an acoustic exciter. The chamber is mounted on top of the acoustic exciter and is connected to a 3-gal gas reservoir tank to prevent pressure build-up upon aerosol ignition. A detailed description of the experimental apparatus is given elsewhere [2].

Metal powder is placed in the chamber prior to the microgravity experiment. When the experimental rig is released in the drop tower, the acoustic exciter is turned on for a 0.4-s interval that results in the aerosolizing particles in the combustion chamber. The aerosolized particles move with randomly directed velocities of about 0.5 m/s due to the particle collisions with the vibrating silicone floor and walls of the chamber. After the acoustic exciter is turned off, a time delay of 0.1–1.2 s is provided so that the particle velocities decrease due to aerodynamic drag. The longer the time delay before ignition, the smaller the particle velocities become. The delay between the time when the chamber floor membrane stopped oscillating and when the aerosol was ignited varied in these experiments to investigate the effect of the initial particle velocity on the flame propagation. After the delay, the tungsten wire igniter is rapidly heated and the aerosol ignites. The entire aerosol combustion event occurs within 0.4–0.8 s. After a time delay of 2.1 s measured from the beginning of the test, the combustion chamber is flushed with an inert gas to quench all postcombustion reactions and rapidly cool the condensed combustion products.

Similar mass loads of 1.1 g of magnesium particles (Aldrich Chemical) with sizes between 180 and 250 μm were used in all of the magnesium combustion experiments described in this paper. The flame structure and details of individual particle combustion are visualized using both high-speed movie and regular-speed video cameras placed at different sides of the combustion chamber. An optical setup including two mirrors tilted slightly relative to each other and two interference filters with the wavelengths of 500 and 510 nm (half bandwidth of 8 nm for each filter) separating the MgO [6] and black body radiations respectively, was used to produce simultaneously two filtered images of the
flame on each video-frame. Because the filter wavelengths were very close to each other, the color sensitivity of the video camera is nearly the same for both of the filtered flame images. That made it possible to compare directly the radiation profiles measured at the MgO characteristic wavelength with those of the black body radiation. The MgO radiation intensity determined from these measurements was used as an indicator of the gas-phase reaction.

Combustion products were collected after each test and analyzed after the microgravity experiments were completed. Samples of the combustion products were embedded into epoxy and cross-sectioned for the internal phase analyses. A Philips XL30 field emission scanning electron microscope (SEM), a Cameca SX50 X-ray electron probe microanalyzer (EPMA), and a Siemens D500 X-ray diffractometer (XRD) using Cu Kα1 radiation were used for analyses of the collected postexperiment powders.

The high-speed movies recorded during the experiments were transferred to SVHS video format. Both high-speed (transferred) and regular-speed videos were digitized and used to characterize flame radiation and determine the flame propagation speed as a function of the ignition delay. NASA Glenn Research Center Tracker software was used to track (utilizing the threshold tracking procedure) the flame front position in time.

RESULTS AND DISCUSSION

Flame Speed Measurements

Three typical examples of flame front position versus time plots are shown in Fig. 1. For each such set of data, an average flame speed was determined as a slope of a linear fit; a plot of the average flame speed as a function of the time delay is shown in Fig. 2. The error bars shown signify the standard deviations for the slope of the found linear fit. It is interesting that a clear trend to higher flame velocities at the shorter time delays is observed. Because the residual particle speed (i.e., the initial speed of particles at the ignition moment) is the only parameter affected by the time delay, the observed trend indicates that the particle residual speed (that always is comparatively low, on the order of 0.1 m/s) noticeably affects the flame propagation. To understand better the nature of this correlation, the particle speed can be estimated as

\[ v = v_0 \exp\left(-t/\tau\right) \]  

(1)

where \( v_0 \) is the initial particle speed, and \( \tau \) is the relaxation time that can be found as

\[ \tau = \left(\rho d^2 C_\varepsilon\right)/(18 \eta) \]  

(2)

where \( \rho \) and \( d \) are the particle density and diameter, respectively, \( \eta \) is gas viscosity (\( \eta = 1.8 \times 10^{-5} \) kg/[m·s] for room temperature air), and \( C_\varepsilon \) is Cunningham slip correction factor [7]. For reference, a velocity history estimated for a 200-μm magnesium particle with \( v_0 = 0.5 \) m/s.

![Fig. 1. Magnesium flame front positions determined using threshold tracking for experiments with different time delays between the end of the silicone floor oscillation and ignition.](image1)

![Fig. 2. Average speed of the magnesium flame propagation as a function of time delay. Estimated particle and flame velocities are also shown.](image2)
(a typical experimental particle residual velocity immediately after the acoustic oscillations stop) is plotted in Fig. 2. For such low particle velocities, turbulence effects are virtually nonexistent. The rate of particle combustion, however, is slightly affected by the particle motion due to convection effects on the rates of the heat and mass transport processes. Therefore, two-phase flame propagation models that are based on single particle combustion models predict a weak dependence of the burning velocity (and thus, also flame speed) on the particle speed. For example, according to the Ballal’s model [8], burning velocity

\[ S_u = \frac{\delta}{t_c} \]  

(3)

where \( \delta \) is the thickness of the reaction zone, and \( t_c \) is the particle combustion time.

\[ t_c = \frac{d^2}{K} \]  

(4)

where \( K \) is the evaporation constant depending on the particle velocity (forced convection) according to the Försling correction [9]:

\[ K = (8/p)(\lambda/C_p) \ln(1 + B)(1 \\
+ 0.276Re^{1/2}Pr^{1/3}) \]  

(5)

where \( \lambda \) and \( C_p \) are the gas mixture heat conductivity and heat capacity, respectively, \( Re \) and \( Pr \) are the Reynolds and Prandtl numbers, respectively, and \( B \) is the transfer number [9] and is defined as:

\[ B = \frac{Q_o Y_{\text{O}_2}/(\gamma + C_p(T_\infty - T_s))/L} \]  

(6)

where \( Q_o \) is the energy released in combustion per mole of oxygen consumed, \( Y_{\text{O}_2} \) is the oxygen mole fraction in the mixture, \( T_\infty \) and \( T_s \) are the ambient and particle surface temperatures, respectively, \( \gamma \) is the stoichiometric factor, and \( L \) is the specific metal vaporization energy. To provide a simple estimate, the reaction zone thickness, \( \delta \) which is a function of the heat conductivity and the temperatures in the preheat and combustion zones, can be taken as a constant equal to the optically determined thickness of the combustion zone [1–3]. Substituting it and Eqs. 4, 5, and 6 into Eq. 3 and computing particle velocities using Eq. 1, the changes in the burning velocity \( S_u \) were estimated for delay times \( t \) in the range of 0 to 1.2 s.

This resulting \( S_u(t) \) curve is also plotted in Fig. 2. Comparison of the estimated \( S_u(t) \) curve versus the changes in the experimental flame speed shows that the change in the estimated burning velocity is significantly less than that observed experimentally. One can also see that for the small time delays, predicted burning velocity is less than the experimental flame speed, and this trend reverses for longer delays. It is also interesting to note that the estimated particle residual speed becomes equal to the experimental flame speed at the same delay time at which the experimental flame speed coincides with the computed burning velocity.

This observed effect of particle residual speed on flame propagation speed can be rationalized considering the contribution of particles into the heat transfer mechanisms. Indeed, Fig. 2 shows that when low time delays (\( t < 0.6 \) s) were used, the particle velocities were higher than the flame propagation velocity, therefore, particles could have served as the heat transfer agents. This effect can be significant when particle velocities are appreciably different from the gas velocity. Microgravity could have been essential in observing this effect since the buoyant flows developing in normal gravity would significantly intensify the heat transport and mask the contribution of the moving particles.

Finally, it should be noted that the flames ignited after long delay times (\( t > 0.9 \) s) extinguished after propagation over a length of 20–30 mm rather than propagated through the entire combustion chamber, as the flames ignited after shorter delay times. Because the constant pressure apparatus was used, a flow of cold outside air entered the combustion chamber as soon as the flame extinguished. This flow rapidly cooled the partially burnt aerosol in the combustion chamber.

**Flame Radiation**

Two optically filtered images of the flame were simultaneously acquired in each video frame using tilted mirrors and interference filters, as described above. Comparisons of the radiation profiles measured along the same line in each of the two filtered images of the magnesium flame showed, consistently with the previous observations using color differential images [2], that a
zone dominated by the black body radiation exists ahead of the flame whereas MgO radiation dominates within the brighter flame zone. An example of the two intensity profiles illustrating this observation is shown in Fig. 3. When comparing the intensities of the two profiles, a correction for black body emissivity difference between the two wavelengths used should be taken into consideration. Using Planck’s equation for the black body emission, the ratio of the radiations at the two wavelengths $\lambda_1$ and $\lambda_2$ is

$$\frac{E_{\lambda_1}}{E_{\lambda_2}} = \left( \frac{\lambda_2}{\lambda_1} \right)^5 \frac{e^{c_2/\lambda_2 T} - 1}{e^{c_2/\lambda_1 T} - 1}$$

(7)

where $c_2 = 1.436$ cm·K, and $T$ is temperature. For $\lambda_1 = 510$ nm and $\lambda_2 = 500$ nm, the ratio varies approximately from 1.6 to 1.1 as the temperature increases from 1000 to 2500 K. Therefore, a radiation signal filtered through a 510-nm filter should noticeably exceed the signal filtered through a 500-nm filter at low temperatures and that difference should become almost negligible as the temperature increases. That agrees with the higher signal measured through the 510-nm filter ahead of the flame front (cf. Fig. 3). In the combustion zone, the intensity measured for the 500-nm radiation significantly exceeds that for the 510-nm radiation, indicative of the strong MgO band radiation. A combustion zone dominated by 500-nm radiation was observed to form in all the microgravity combustion experiments, indicating development of a vapor-phase flame in which the MgO products were produced.

Combustion Product Size Distribution

A dense cloud of airborne smoke was observed in the chamber after each experiment. This smoke, produced usually in a vapor-phase flame, was not analyzed and simply vented prior to collection of the powder from the chamber floor. Powders collected from the chamber after combustion experiments were examined using an optical microscope. Feret diameter defined as the diameter of a circle having the same area as the object and computed as $d_F = (4 \cdot \text{area}/\pi)^{0.5}$, was determined for each particle with image analyzing software (UTHSCSA ImageTool) and used to produce particle size distributions. The size distributions of both unburnt Mg particles (size-classified using sieves with nominal sizes of 180 and 250 $\mu$m), and the burnt powders are shown in Fig. 4. Even though magnesium burns actively in the vapor phase, the burnt particle sizes do not show a decrease. Instead, a slight shift towards larger particle size is observed for the burnt powder as compared to the original magnesium. This shift is most likely due to a thick and porous oxide coating produced on burnt particles (see below).
Combustion Product Surface Morphology and Elemental Compositions

An SEM was used to compare the surface morphology of unburnt and burnt metal particles. Electron dispersive spectroscopy (EDS) was used to compare qualitatively oxygen contents in surface layers of different samples. General views of various magnesium particles are shown in Fig. 5 and close-ups of the particle surface are shown in Fig. 6. Five particle samples chosen for the SEM and EDS analyses were unburnt (virgin) particles and particles collected after runs 53, 56, 57, and 60, in which ignition delays of 0.4 s (a typical delay used previously [1, 2]), 1.05 and 0.95 s (long delays), and 0.1 s (a short delay), respectively, were used. “Virgin” magnesium particles have clean surfaces with small-size, white particles attached (Fig. 6a). EDS analyses using standardless calibration showed that these white particles consist of stoichiometric MgO. Surfaces of particles collected after experiments in which different ignition delays were used, were markedly different. Particles collected after both runs #53 and 60 with the ignition delays of 0.4 and 0.1 s, respectively, are coated with a fibrous layer (Figs. 6b and 6d). This surface layer is similar to the oxide coating observed on particles quenched in single magnesium particle combustion experiments [5]. EDS measurements showed that these surface layers contained high amounts of oxygen, generally comparable to that detected in the small MgO particles on the surface of virgin magnesium. The surfaces of the particles collected after runs 56 and 57, in which long delays of 1.05 and 0.95 s, respectively, were used, were indistinguishable from the surfaces of the unburnt particles. However, EDS showed significant oxygen presence in surface layers (the depth of the electron beam penetration is about 2–3 μm) for most of these particles (Table 1). Surface morphology and oxygen content detected for particles collected after runs 56 and 57 are very different from any of the quenched single magne-
sium particle samples analyzed in an earlier research [5].

The internal elemental compositions were analyzed using EPMA for three cross-sectioned combustion product samples. These were the same combustion product samples for which surface elemental analyses were conducted using EDS: samples 53, 56, and 60 for which the ignition delays in microgravity experiments were 0.4, 1.05, and 0.1 s, respectively. EPMA oxygen content measurements were calibrated using a commercial MgO standard sample by C. M. Taylor. Magnesium content measurements were calibrated using cross-sections of

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**TABLE 1**

Summary of Electron Microprobe Analyses of Magnesium Combustion Products

<table>
<thead>
<tr>
<th>Sample</th>
<th>Time Delay Used in Microgravity Test to Suppress Particle Motion, s</th>
<th>Maximum Oxygen Content Measured in Cross-section (WDS), atomic %&lt;sup&gt;a&lt;/sup&gt;</th>
<th>Oxide Layer (yes/no)</th>
<th>Surface Oxygen/Magnesium Peak Ratio (EDS)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MgO</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>0.42</td>
</tr>
<tr>
<td>Mg</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>0.05</td>
</tr>
<tr>
<td>Run 60</td>
<td>0.1</td>
<td>0.47</td>
<td>Yes</td>
<td>0.42</td>
</tr>
<tr>
<td>Run 53</td>
<td>0.4</td>
<td>0.49</td>
<td>Yes</td>
<td>0.30</td>
</tr>
<tr>
<td>Run 57</td>
<td>0.95</td>
<td>—</td>
<td>No&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.20&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>Run 56</td>
<td>1.05</td>
<td>3.26</td>
<td>No</td>
<td>0.20</td>
</tr>
</tbody>
</table>

<sup>a</sup> Oxygen detection limit is approximately 0.3 atomic %.

<sup>b</sup> Several particles coated with oxide were detected. These particles were not used for surface oxygen/magnesium peak ratio measurements.

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**Fig. 6.** SEM images of surfaces of: (a) “virgin” Mg particles; (b) combustion products, run 53, time delay 0.4 s; (c) combustion products, run 56, time delay 1.05 s; (d) combustion products, run 60, time delay 0.1 s.
the 99% pure (Aldrich Chemical) magnesium particles employed in our experiments. Consistent with the previous observations of the particle surfaces, a thick oxide layer was present on the surfaces of particles quenched after runs 53 (see example in Figs. 7a and 7b) and 60, but not on the particles collected after run 56 (in which a long ignition delay was used, see Fig. 8). The cross-sections revealed the highly porous structure of the oxide layer as shown in Fig. 7b. The particle interiors were metallic and, according to our analyses, contained essentially pure magnesium for most of samples 53 and 60. Local regions with detectable oxygen content inside the particles were also observed; the maximum measured oxygen contents are shown in Table 1. Significantly higher oxygen contents were measured in most of the particles from sample 56, for which the longest ignition delay was used in the microgravity experiment and a slow flame propagation was observed (cf. Fig. 2). This higher oxygen content is consistent with the high oxygen/magnesium peak ratios found for these particles from EDS, also shown in Table 1. Note, that several oxygen-free particles that could not be distinguished from the unburnt Mg were also observed in this sample.

**Combustion Products Bulk Compositions**

Combustion products of magnesium collected after the microgravity experiments were analyzed using XRD. Four samples collected after runs 53, 56, 57, and 60 were used (the same samples that were selected for SEM and EPMA analyses). For reference, XRD spectra for coarse magnesium powders used in our experiments were also collected. Silicon powder was used as an internal standard in these analyses.
The measured XRD spectra are shown in Fig. 9. XRD spectra for runs 53 and 60 showed both magnesium and magnesium oxide peaks. This result supports the electron microscopy analyses in which oxide layers were detected on surfaces of particles recovered after these runs. As shown in Fig. 9, only magnesium peaks appeared in the XRD spectra for runs 56 and 57, for which longer ignition delays were used and measured flame velocities were also very low. The sensitivity of these measurements for MgO is about 1%. The absence of oxide in products collected after runs with long ignition delays determined from XRD analyses is, also, consistent with the electron microscopy results that showed no oxide layers on particles collected after runs 56 and 57 (“long” delays).

CONCLUDING REMARKS

Observations of the luminous flame structure using interference filters have shown that the preheat and combustion zones can be optically distinguished in magnesium aerosol flames in microgravity, consistent with the earlier results based on the comparison of the green and red portions of the video-signal [2].

An effect of particle motion prior to ignition on the flame propagation speed in microgravity has been observed. It appears that this can be attributed to the role the moving particles play in the heat transfer through a steady gas. When the initial particle speeds were lower than 0.1 m/s, the observed flame propagation speeds became less than predicted by a simplified model [8]. Such slow moving flames were observed to extinguish before propagating through the entire combustion chamber. More detailed modeling appears to be necessary to interpret these observations fully.

Radiation measurements showed strong MgO emission for all the flames, indicative of vapor-phase Mg combustion and MgO formation. In addition, a dense smoke (MgO) cloud, usually produced by the vapor-phase flame, was observed after all experiments. Despite this visible and spectroscopic similarity between the “slow” and “fast” flames, the surfaces of the combustion product particles are markedly different. Powders collected after the experiments in which the flame velocities were low (i.e., those with long time delays) had no MgO layer and were indistinguishable by SEM from those of the unburnt powder (Figs. 6 and 8). In addition, MgO was not detected for these “slow” flame products by XRD measurements (Fig. 9). In “fast” flames, the SEM and XRD measurements (Figs. 6, 7, 9) clearly show MgO coating on particles. It thus seems clear that the deposition of the smoke oxide produced in the gas-phase flame on the surface of burning particles was inefficient for the slow flames. It also appears reasonable to suggest that the oxide smoke deposition process might not be the primary source of the thick MgO layer found on the surfaces of particles collected after the experiments with shorter time delays and higher observed flame speeds.

A higher dissolved oxygen content was detected inside the oxide-free particles collected from the “slow” flames. Since in these experiments flame extinguished before propagation through the entire chamber, it is suggested that
these combustion products represent partially burned particles that were rapidly quenched due to flame extinguishment. The difference between these particles and those collected from “fast” flames is, therefore, in the completeness of their combustion and quenching rate. The presence of the thick oxide layer on the surfaces and absence of oxygen in the interiors of particles collected from “fast” flames is consistent with the combustion mechanism including the formation of an Mg-O solution inside burning particles that saturates and undergoes a phase change producing the MgO phase, as was previously proposed based on single particle combustion experiments [5].

The absence of a surface oxide layer and the presence of dissolved oxygen inside the particles collected from the “slow” flames indicate that the phase transition has not occurred for these particles, i.e., that the particles were quenched before the solution has saturated. The results of this work indicate that the MgO formation mechanism via the phase separation from Mg-O solution could contribute more significantly to the formation of an oxide coating than the MgO deposition from the vapor-phase flame.

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