COMBUSTION OF AEROSOLIZED SPHERICAL ALUMINUM POWDERS AND FLAKES IN AIR

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Combustion rates and completeness of aerosolized spherical aluminum powders and flakes are compared using constant volume explosion experiments. The comparison of particles and flake sizes was made based on their specific surface areas determined using the Brunauer–Emmett–Teller (BET) method and respective “BET diameters.” It is observed that the rates of pressure rise and respective rates of flame propagation were higher for spherical aluminum powders with BET diameters of about 2 to 5 μm compared to aluminum flakes for which the respective BET diameters were under 1 μm. In agreement with the flame propagation rates, the overall completeness of combustion was also higher for spherical powders compared to flakes. It is suggested that aerosolized flakes could be agglomerated in gas flows more than spherical particles causing their inferior combustion performance.

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INTRODUCTION

Aluminum powders are widely used as additives in energetic formulations for propellants, explosives, and pyrotechnics (e.g., Price and Sigman, 2000). Combustion processes of aluminum particles have been extensively

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studied (see, for example, recent reports by Beckstead et al., 2001; Bucher et al., 2000; Dreizin, 1996, 1999; Rossi et al., 2001). Particle size and surface morphology are important parameters affecting both the rate of combustion additives as well as hydrodynamic particle behavior in the ignition zone and flame area. A number of recent investigations deal with an accelerated burn rate that is possible to achieve using reduced particle sizes and, ultimately, nanosized aluminum particles, produced using state-of-the-art technologies (e.g., Mench et al., 1998; Pranda et al., 2000). At the same time, in many practical applications, e.g., in pyrotechnic formulations, aluminum flakes with submicron thickness that are readily available commercially are used instead of regular powders (Tadahiro et al., 1997, Kosnake et al., 2000). It is expected that because of the high specific surface of flakes, they could ignite easier than spherical or spheroid particles of similar mass. An accelerated ignition should result in an acceleration of the overall burn rate of aerosolized aluminum flakes as compared to regular powders. At the same time, gas dynamics of the flakes should be remarkably different from that of regular powders, which could be important for combustion in multiphase systems. We were unable to find systematic comparisons of ignition and combustion behaviors of aluminum flakes versus spherical or other “three-dimensional” aluminum particles in the literature. Therefore, no guidance is currently available for selection of either regular aluminum powders or flakes as a fuel or fuel additive for applications in explosives, propellants, or incendiaries. Current interest in comparing combustion parameters of aluminum powders and flakes is stimulated, in particular, by development of fuel–air explosives and thermobaric explosives for which metal aerosol serves as a fuel and air as an oxidizer. Therefore, combustion rates of aerosolized spherical aluminum powders and flakes in air are compared in this work. A constant volume explosion experimental technique is used to assess the burn rates based on the measured rates of pressure rise. In addition, combustion products are collected and analyzed.

MATERIALS

A set of commercially available spherical aluminum powders with particle sizes in the 1–50-μm range was compared to a set of commercially available aluminum flakes. Aluminum powders were acquired from Alfa Aesar, Atlantic Equipment Engineers, and Toyal America. Representative scanning electron microscope (SEM) images of aluminum
particles are shown in Figure 1. One can see that most of the particles have spherical or nearly spherical shapes. Particles by Atlantic Equipment Engineers are somewhat less spherical than other powders; however, the difference in the shape does not appear to be significant. Particle sizes were measured by low-angle laser light scattering (LALLS) using a Coulter LS 230 enhanced laser diffraction particle size analyzer. The resulting size distributions are shown in Figure 2. Respective mean particle diameters based on both sample volume and surface area are shown in Table 1. The flakes were acquired from Iowa Pyro Supply; representative SEM images are shown in Figure 3. To initially assess the reference particle size for the flakes, the SEM images were analyzed so that typical dimensions were determined for flakes positioned parallel to the image plane. These dimensions measured from the SEM images generally agreed with those implied by the mesh sizes specified by the flake suppliers. In several images, flakes positioned normally to the image plane were observed as shown, for example, in Figure 4. Several flake thickness measurements were made. The observed thickness varied from 20 to 200 nm.

Figure 1. SEM images of aluminum powders used.
Specific surface area of the flakes, $A$, was determined by nitrogen absorption employing the Brunauer–Emmett–Teller (BET) isotherm (multipoint analysis, using a high-speed gas sorption analyzer, NOVA 3000). Based on these measurements, equivalent BET diameters for the flakes were estimated as $d_{BET} = \frac{6}{\rho A}$, where $\rho$ is the aluminum density, 2.7 g/cm$^3$ (Winter, 2003). To provide a fair comparison between the combustion behavior of flakes and particles, specific surface areas of particles were also measured using a BET isotherm and equivalent BET diameters were computed and are shown in Table 1. These BET diameters for both flakes and powders were used as a reference size to

Figure 2. Size distributions for the aluminum powders obtained using LALLS.
<table>
<thead>
<tr>
<th>Type of A1</th>
<th>ID per manufacturer</th>
<th>Mean diameter, ( \mu m ) based on volume distribution, by LALLS</th>
<th>Mean diameter, ( \mu m ) based on area distribution, by LALLS</th>
<th>Specific surface area, m(^2/g) by BET</th>
<th>Equivalent BET diameter, ( \mu m )</th>
</tr>
</thead>
<tbody>
<tr>
<td>FLAKES</td>
<td>− 325 mesh</td>
<td>N/A</td>
<td>N/A</td>
<td>7.75</td>
<td>0.30</td>
</tr>
<tr>
<td></td>
<td>− 20 + 325 mesh</td>
<td>N/A</td>
<td>N/A</td>
<td>4.78</td>
<td>0.47</td>
</tr>
<tr>
<td></td>
<td>− 20 + 120 mesh</td>
<td>N/A</td>
<td>N/A</td>
<td>3.20</td>
<td>0.71</td>
</tr>
<tr>
<td></td>
<td>− 90 mesh</td>
<td>N/A</td>
<td>N/A</td>
<td>3.16</td>
<td>0.72</td>
</tr>
<tr>
<td>POWDERS</td>
<td>3–4.5 ( \mu m )</td>
<td>7.18</td>
<td>5.20</td>
<td>1.16</td>
<td>1.93</td>
</tr>
<tr>
<td></td>
<td>1–5 ( \mu m )</td>
<td>9.38</td>
<td>5.39</td>
<td>0.80</td>
<td>2.81</td>
</tr>
<tr>
<td></td>
<td>X-65</td>
<td>10.87</td>
<td>7.40</td>
<td>0.46</td>
<td>4.92</td>
</tr>
<tr>
<td></td>
<td>10–14 ( \mu m )</td>
<td>20.33</td>
<td>16.28</td>
<td>0.41</td>
<td>5.41</td>
</tr>
</tbody>
</table>
Figure 3. SEM images of aluminum flakes used.

Figure 4. High-magnification-SEM image of aluminum flakes showing their thickness.
compare the measured combustion parameters. Note that the BET equivalent diameters for the flakes are about an order of magnitude smaller than those of powders. This is generally consistent with the very small flake thickness as observed using SEM (cf. Figure 3).

**EXPERIMENTAL**

A schematic diagram of the experimental apparatus is shown in Figure 5. It was designed after the Bureau of Mines constant volume explosion apparatus (Cashdollar, 1994; Hertzberg et al., 1992), and employs a 9.2-L explosion vessel previously described by Schoenitz et al. (2003). Prior to each experiment, a powder charge of known mass was placed in a pipe elbow under the nozzle, and the explosion vessel was closed and evacuated to about one-third of an atmosphere. Only air was used as oxidizer in this work; therefore, no additional purging of the explosion vessel was required. A reservoir was filled with air to a pressure of ca. 60 psi. The powder charge was introduced to the explosion vessel with a gas blast from the reservoir through a solenoid valve and the open reservoir valve (cf. Figure 5). The pressure in the reservoir tank was selected so that

![Figure 5. Schematic diagram of the constant volume explosion apparatus.](image-url)
during the time when the solenoid valve was opened (e.g., 0.2 s) the pressure in the explosion vessel reached 1 atm. Following a delay of 0.3 s provided to reduce the turbulence in the explosion vessel, the heated wire igniter was activated. A 8-mil (203.2-µm-diameter) tungsten wire heated up with an AC voltage served as an igniter. The voltage was selected to melt the wire during 60 ms, which limited duration of the ignition energy input; the estimated total electrical energy supplied to the igniter was 30 J. A pressure trace measured using a transducer by Schaevitz Sensors was recorded using a Rapid Systems four-channel digital oscilloscope board. Simultaneously, an ignition pulse was recorded for time reference. Final pressure in the explosion vessel, reduced due to the consumption of oxygen, was recorded after the combustion was completed and after a delay of several minutes to allow the gas in the vessel to cool to room temperature. Using a vent valve and a purge valve (cf. Figure 5) the vessel pressure was brought to 1 atm, the vessel was opened, and the condensed products were collected for further analysis. Phase compositions of the combustion products were analyzed using x-ray diffraction. A Philips X’pert MRD x-ray diffractometer system with Cu Ka radiation (1.5438 Å) was used at 45 kV and 40 mA.

After the collection of combustion products and cleaning the interior of the vessel, the nozzle was removed and the vessel was again closed and evacuated. A gas blast from the reservoir through the solenoid valve was repeated to clean the pipe elbow and remove residue of the original powder charge that was not dispersed during the combustion experiment. The powder blasted in the vessel during this postexperiment procedure was collected and weighed. The weight of this powder residue varied from 3 to 20% of the original charge. Thus, the mass of powder charge was corrected to estimate the equivalence ratio of the air/fuel mixture in each experiment. In most experiments, an equivalence ratio in the range 1.1–1.5 was used; however, for some powders a wider range of equivalence ratios was explored.

**DATA PROCESSING**

The equivalence ratio $\psi$ was computed for each run using the amount of oxygen in the vessel prior to explosion based on the respective experimental pressure in the vessel and the loaded powder mass corrected by the experimentally determined amount of residual powder. Each recorded pressure curve was time differentiated and the value of the maximum
rate of pressure rise, \( \frac{dP}{dt_{\text{max}}} \), was found. A theoretical treatment of the constant volume explosion experiment makes a direct connection between the maximum rate of pressure rise, \( \frac{dP}{dt_{\text{max}}} \), and the rate of combustion (Glassman, 1996). Therefore, the maximum rate of pressure rise is commonly used to gauge different combustible powders (Cashdollor, 2000). The processing of the pressure traces in this work was the same as was done by Cashdollor (1994, 2000) and Schoenitz et al. (2003). The error of the \( \frac{dP}{dt_{\text{max}}} \) found for each individual run is estimated to be less than \( \pm 3\% \), and so the main experimental error was caused by the poor reproducibility of the experiments. This, in turn, was primarily caused by minor differences in the amounts of the powder being dispersed by the gas blast, as was obvious from the variation in the mass of the powder residue collected after the experiment (see earlier discussion). The experimental values of \( \frac{dP}{dt_{\text{max}}} \) were plotted for each material type (different size spherical Al powders and flakes) as a function of the experimental equivalence ratio, as illustrated in Figure 6. To compare materials to one another, the effect of somewhat different equivalence ratios in different runs should be minimized. Thus, a linear fit for the experimental points was found for each material type and the respective values of \( \frac{dP}{dt_{\text{max}}} \) corresponding to the equivalence ratio of 1 were

![Figure 6.](image-url)  
**Figure 6.** Experimental data collected in the constant volume explosion tests for the maximum rate of pressure rise for different types of aluminum powders as a function of experimental equivalence ratio. The equivalence ratio was changed by varying initial powder mass load and by to slight changes in the initial pressure in the vessel.
identified, as shown by thin vertical lines in Figure 6. The error of such \(dP/dt_{\text{max}}\) was estimated as a standard deviation of the experimental \(dP/dt_{\text{max}}\) values from those implied by the produced linear fit at all the respective equivalence ratios. Characterizing each powder with its respective values of \(dP/dt_{\text{max}}\) found at \(\psi = 1\) enabled us to compare directly the maximum rates of pressure rise and, therefore, the rates of flame propagation for different powders.

**RESULTS AND DISCUSSION**

The values of \(dP/dt_{\text{max}}\) extrapolated from the experimental trends of \(dP/dt_{\text{max}}\) (Figure 6) for the equivalence ratio \(\psi = 1\), for all the aluminum powders investigated in this project, are shown in Figure 7. For spherical aluminum powders, a trend of a significantly higher rate of flame propagation for finer particle sizes is clear for all but the finest powder. A small decrease in the rate of flame propagation \((dP/dt_{\text{max}})\) was observed for the Al powder with nominal diameters 3–4.5 \(\mu\)m by Alfa Aesar for which the BET diameter was found to be smaller than that for an Atlantic Equipment Engineers powder with nominal diameters of

![Graph showing maximum rates of pressure rise inferred for equivalence ratio of one for different aluminum powders (particles) and flakes tested in this work.](image)

**Figure 7.** Maximum rates of pressure rise inferred for equivalence ratio of one for different aluminum powders (particles) and flakes tested in this work.
1–5 μm (cf. Table 1). As noted earlier, the BET diameters for all the Al flake types were significantly smaller than those for Al powders; however, the measured rates of pressure rise were relatively low. The change in the rate of pressure rise was also small between the different types of flakes. Large error bars, especially for the flakes of nominal size −325 mesh, are indicative of a poor repeatability of the pressure traces between the experiments with the same type of flakes (cf. Figure 6). Repeating experiments did enhance statistical data processing but did not eliminate vast discrepancies between the measured values of $dP/dt_{\text{max}}$ from run to run.

Comparison of spherical powders versus flakes shows that generally higher rates of pressure rise and, thus, of flame propagation are achieved with spherical powders with nominal sizes in the general range 1–10 μm. This observation was supported by the measurements of the final pressure in the explosion vessel. A ratio of the final pressure in the vessel over the initial pressure, or the pressure ratio, was inversely proportional to the oxygen consumption in an explosion. Analogous to the date processing for the $dP/dt_{\text{max}}$ measurements, a linear fit for the experimental pressure ratio as a function of equivalence ratio $\psi$ was found for each type of material. The resulting values of the pressure ratios implied by the linear fit at $\psi = 1$ are shown in Figure 8 for both flakes and spherical particles with the error bar showing the standard deviation of experimental points from the found linear fit. For experiments conducted at $\psi = 1$ or greater, all the available oxygen should be consumed in case of complete combustion. For air containing 21 mol% of O$_2$ and used as the only oxidizer in these experiments, the minimum final pressure ratio to the initial pressure should, therefore, be equal to $(1 - 0.21)/1 = 0.79$. Comparing the experimental pressure ratios shown in Figure 8, one can see that indeed the combustion of finer aluminum powders was more complete than that of flakes. Also in agreement with very low values of $dP/dt_{\text{max}}$ for the coarser aluminum powder, the respective pressure ratio is fairly high, showing a rather incomplete combustion.

Combustion product phase analysis based on x-ray diffraction patterns was conducted for selected samples of each aluminum powder. Representative examples of the measured x-ray diffraction patterns are shown in Figure 9. The observed patterns were compared to reference patterns from the PDF-2 database (Powder Diffraction File, Joint Committee for Powder Diffraction, Standards International Centre for Diffraction Data, Newtown Square, Pennsylvania, USA).

In addition to diffraction peaks of residual Al, peaks roughly consistent with spinel-like aluminum oxides or oxynitrides are observed in
Figure 8. Ratios of the final pressures in the explosion vessel cooled after explosion experiment over the initial pressures inferred for equivalence ratio of 1 for different aluminum powders (particles) and flakes tested in this work. The dashed line shows the minimum possible ratio for air expected if all the available oxygen is consumed in the explosion.

Figure 9. X-ray diffraction patterns of two representative combustion products. Aluminum peaks are marked with solid diamonds; vertical dashed lines show the peak positions of δ-Al₂O₃ (PDF-2 database, reference card 46-1131).
the diffraction patterns. For comparison purposes, the reference pattern of $\delta$-Al$_2$O$_3$ is shown in Figure 9. Although a number of structurally similar oxides or oxynitrides could be matched against the measured patterns with equal success, a perfect match was not achieved. It is expected that the combustion products consist of disordered oxide phases with at least traces of nitrogen. A more detailed analysis is needed to determine structure and composition of the oxide products of Al combustion in the present experiment. To assess the degree of conversion in the combustion, the ratio of integrated intensities of Al and Al oxide diffraction peaks was used as a semiquantitative measure. In the absence of detailed structural information, the diffraction peaks in the vicinity of the 100% -reflection of $\delta$-Al$_2$O$_3$ were summarily integrated from 65.5$^\circ$ to 68.5$^\circ$ 2$\Theta$ and set in proportion to the 220 peak of Al at 65.19$^\circ$ 2$\Theta$. The results of this comparison are shown in Figure 10; as expected, these estimates mirror well the ratios of initial to final pressures shown in Figure 8.

The observed trend of the less complete and slower combustion for the aluminum flakes as compared to the regular, micron-sized spherical powders is somewhat unexpected. As noted earlier, very thin flakes with a highly developed specific surface should react faster during

![Graph](image)

**Figure 10.** Semiquantitative comparison of combustion completeness estimated via integrated intensities of Al and Al oxides obtained from x-ray diffraction patterns of combustion products.
heterogeneous reactions at relatively low temperatures typical of aluminum ignition. Based on the measured BET diameters, it was also expected that, upon ignition, molten droplets formed from individual flakes would be smaller than molten aluminum particles. Therefore, faster ignition and combustion rates for the flakes would seem reasonable. However, the preceding arguments do not consider dramatic differences in the behavior of spherical particles and flakes in gas flows. Although detailed analysis of these fluid mechanics problems is outside the scope of this project, it is clear that flakes could readily induce local swirls and create their own flow patterns, whereas fine particles tend to follow the gas flow. Producing local swirling patterns could result in significant agglomeration of the flakes prior to their ignition. Such an agglomeration could be one of the reasons for relatively slow and incomplete combustion observed in this work. Additional experiments or theoretical analyses would be needed to further elucidate the effects of such flake–gas flow interaction on the rate of their combustion.

CONCLUSIONS

Combustion rates of spherical aluminum particles and flakes aerosolized in air are measured and compared to each other. The constant volume explosion technique was used for combustion experiments. Even though the BET diameters of all the tested aluminum flake samples were less than respective BET diameters for spherical aluminum particles, both the combustion rates and reaction completeness were higher for the particles with volume mean diameters of up to 10 μm. This conclusion is based on comparisons of the rates of pressure rise, consumption of oxygen from the air in the vessel, and completeness of the aluminum conversion to oxide based on the analyses of the condensed combustion products. It is speculated that the relatively slow rate of combustion and incomplete reaction observed for flakes are due to agglomeration of flakes before their ignition within the small-scale eddies produced as a result of flakes’ interaction with the gas flows induced by the propagating flame.

REFERENCES


