Research Article

Uranium Dust Cloud Combustion: Burning Characteristics and Absorption Spectroscopy Measurements

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1. Introduction

Uranium metal is used in a variety of applications in which combustion can occur, either as a desired effect or as a potential hazard. For example, combustion of metal from armor-piercing, depleted uranium projectiles can cause significant damage [1]. In general, limited work has been published on the combustion of uranium [2–6].

Due to its pyrophoric nature, uranium powder exposed to air at room temperature forms an oxide layer primarily comprising UO2 [4, 7–9]. Calculations performed using NASA CEA [10] with constant enthalpy and pressure (and φ = 1) indicate that the adiabatic flame temperature of uranium in air and oxygen are 3773 K and 4843 K, respectively. Actual flame temperatures of clouds of uranium particles will likely vary spatially and temporally and may be significantly lower in general.

Mouradian and Baker Jr. created a model for determining uranium and zirconium burning temperatures in air with respect to sample size, geometry, and airflow whilst making some allowance for the oxide layer [3]. From this work, for a spherical particle under natural convention and a size of less than 0.01 cm and decreasing, the burning temperature approaches about 3200 K. Due to the high boiling point of uranium (∼4400 K) as compared to its oxide (∼3800 K) and burning temperature at atmospheric pressure, it is expected to burn heterogeneously limited by the amount of oxide [11–13]. As in the case of Zr and Ti combustion, the suboxide UO is also expected to form a range of liquid solutions with the metal [14, 15], and therefore behave similarly in combustion.

Dust explosions involve the combustion of particulate dispersed in air and require the presence of several components: fuel, oxidation, confinement, mixing, and ignition source [16]. Dust explosions have often been studied in the context of worker safety and the prevention of accidental occurrences in environments such as coal mines [17]. Thermobaric weapons also operate in this manner, wherein an aerosol or dust-like fuel is dispersed and subsequently detonated [18]. According to electrical safety standards, a
Figure 1: Uranium powder size distribution [38].

Figure 2: ESEM image of uranium powder particles [38].

Figure 3: Higher resolution ESEM images of singular uranium particle. Small flecks and fuzzy irregularities on surface are likely due to a uranium oxide layer [38].
uranium dust cloud is considered potentially hazardous and electrically conductive and has a low ignition temperature [19]. A model has also previously been created to simulate aerosolized uranium dispersion in the context of potential health and safety effects [20]. These circumstances may be present when uranium is processed as fuel for nuclear reactors, and in other scenarios where uranium is combusting [21]. Thus, dust combustion of uranium at various scales, in single particles and in variously sized clouds, is of interest.

Various experiments have been performed and models set up to study and emulate high-temperature plasma-like behavior and oxidation phenomena, which might occur in a nuclear fireball. Species that may evolve during uranium excitation, such as atomic uranium and oxides of uranium,
have been investigated by various groups. Many uranium and uranium monoxide transitions were determined by the work of Kaledin et al., in which vapor created from the heating of uranium samples was characterized using laser-induced fluorescence (LIF) [22]. They further characterized uranium monoxide bands using pulsed laser deposition in a supersonic expansion [23]. Laser-induced breakdown spectroscopy has been used as an analog to fireball conditions to study uranium kinetics in the presence of oxygen [24–28]. A reaction mechanism has been determined and calculations performed based on these experimental results [29, 30]. Koroglu et al. observed the condensation of gas phase species into particulate from a plasma flow reactor in the presence of oxygen whilst cooling from 5000 K to 1000 K, including the formation of uranium oxides [31]. Work has also been completed regarding the oxidation of uranium, where a diffusion rate law was determined for the oxidation of uranium at lower temperature (nonplasma) regimes [7, 32–34]. The plasma-forming experiments provide a different pathway to uranium product formation, condensing from much higher temperatures, whereas the experiments discussed in this document are more similar to that performed by Hertzberg et al., wherein a selection of metal powders were ignited to determine such characteristics as explosibility and burn velocity [35].

In this work, the dynamics, temperature, and gas phase environment of particulate combustion of natural uranium is characterized using absorption spectroscopy. Beneficially, absorption spectroscopy measurements can be used to quantitatively determine temperature and concentration of species with the knowledge of energy state information of targeted transitions [36, 37]. Few measurements are available on particulate combustion of U, which occurs after fragmentation from impact. Herein, high-speed imaging and...
Figure 7: Entire dust cloud setup schematic for absorption measurements [38].

Figure 8: Top view schematic for metal powder burn time experiments [38].

Table 1: Powder characterization for burn time testing [15, 38, 48–59].

| Element | Mass (mg) | Metal $\Delta H_{\text{vap,298K}}$ (kJ/mol) | Oxide $\Delta H_{\text{vap,298K}}$ (kJ/mol) | Oxide volatilization temperature (K) | Metal boiling point (K) | Source |
|---------|-----------|-------------------------------------------|------------------------------------------|-----------------------------------|------------------------|--------|
| U       | 6         | 3.6 2.6 4.1 3.6 5.4                      | 477                                      | 621                               | ~3800                  | 4404   | Reduced from oxide |
| Al      | 2.6       | 2.2 2.2 2.2 2.8 2.2                      | 284                                      | 1860                              | 4000                   | 2791   | Alfa aesar |
| B       | 2.4       | 2.3 2.3 2.3 2.3 2.3                      | 499                                      | 360                               | 2340                   | 4139   | Alfa aesar |
| Fe      | 3.3       | 3.5 3.2 3.2 3.2 3.3                      | 354                                      | 610                               | 3400                   | 3133   | Alfa aesar |
| Hf      | 3.9       | 2.8 2.4 2.8 2.4 2.4                      | 648                                      | 1461                              | ~4350                  | 4575   | Alfa aesar |
| Mo      | 3.8       | 3.7 3.4 3.4 3.6 3.4 3.6                 | 617                                      | 333                               | 900                    | 5833   | Alfa aesar |
| Si      | 2.6       | 2.7 2.7 2.7 2.7 2.7 2.7                 | 383                                      | 353                               | 2633                   | 3177   | Sigma aldrich |
| Ta      | 2.2       | 3.7 3.3 3.6 2.4 2.4                    | 753                                      | —                                 | 1200                   | 5693   | Alfa aesar |
| Ti      | 3.2       | 3.3 3.3 3.3 2.4 2.4                    | 427                                      | 1890                              | 3300                   | 3631   | Alfa aesar |
| W       | 2.3       | 3.8 3.8 3.1 3.1 3.1                   | 774                                      | 452                               | 1970                   | 6203   | Alfa aesar |
| Zr      | 2.2       | 3.7 3.7 3.7 3.7 3.7                   | 591                                      | 920                               | 4280                   | 4703   | Alfa aesar |
broadband measurements of burn time provide additional insight into U combustion phenomena as well as that of other metal powders in order to establish comparisons between previously published data and observed uranium combustion behavior.

2. Materials and Methods

In order to ensure the validity of the uranium dust cloud combustion experiments described herein, the uranium powder was characterized using various methods prior to testing. Uranium powder was handled with precautions typically taken to avoid heavy metal inhalation and was on testing. Uranium powder was characterized using various methods prior to combustion experiments described herein, the uranium In order to ensure the validity of the uranium dust cloud combustion behavior.

Table 2: Information used to calculate Boltzmann temperature of Fe I from Kurucz database [38, 43].

| \( \lambda \) (nm) | \( I_l \) | \( I_n \) | \( E_{Bol} \) (cm\(^{-1}\)) | \( A_{10} \) (s\(^{-1}\)) |
|-------------------|--------|--------|-----------------------------|--------------------|
| 381.584           | 4      | 3      | 11976.24                    | 1.31E + 08          |
| 382.0425          | 5      | 4      | 6928.268                    | 6.67E + 07          |
| 382.4444          | 4      | 3      | 0                           | 2.83E + 06          |
| 382.5881          | 4      | 3      | 7376.764                    | 5.98E + 07          |
| 382.7822          | 3      | 2      | 12560.93                    | 1.05E + 08          |
| 383.4222          | 3      | 2      | 7728.059                    | 4.52E + 07          |
| 384.0437          | 2      | 1      | 7985.784                    | 4.70E + 07          |
| 384.9966          | 1      | 0      | 8154.713                    | 6.05E + 07          |
| 385.6371          | 3      | 2      | 415.933                     | 4.64E + 06          |
| 385.9911          | 4      | 4      | 0                           | 9.69E + 06          |
| 386.5523          | 1      | 1      | 8154.713                    | 1.55E + 07          |
| 387.2501          | 2      | 2      | 7985.784                    | 1.05E + 07          |

In this publication; however, the oxide layer exhibits a distinct texture. The oxide thickness was measured using ImageJ at regular intervals along the edges of many particles and a distribution was determined as shown in Figure 5. The average oxide layer thickness was about 43.3 nm, with a standard deviation of 28.6 nm. The oxide layer can significantly affect particle combustion behavior and is discussed further later on.

A dust cloud chamber was created in order to facilitate the confined dispersal and combustion of \( \sim \)0.025 g of uranium powder (Figure 6). Powder was loaded into a small conical cavity with two tungsten electrodes on opposite sides. The 5 kV discharge of a 1 uF capacitor generates a local plasma in the cavity that rapidly heats, ignites, and disperses the powder into the chamber above the cavity (see inset Figure 6). The internal volume of the chamber was 2.62E – 3 m\(^3\). The chamber is capable of pressurization with a gas mixture of choice as well as vacuum down to a few torr (Figure 6).

Timing of the system was controlled with a pulse generator (Quantum Composers 9514) which delayed the acquisition of the event with respect to the firing of the dust cloud. An additional pulse generator (Quantum Sapphire 9214) was used to trigger line scan camera acquisition with a duration of 359.5 \( \mu \)s (Hamamatsu C11165-02). A digital oscilloscope (Picoscope 4424) and silicon photodiode were used for burn time measurement (Figure 7).

A Xenon flash lamp and custom-built 1.5 m spectrometer with 2400 gr/mm grating served as a broadband source to acquire an absorption spectrum of the gaseous combustion environment. The flash lamp has a duration of 7 \( \mu \)s full width at half maximum (FWHM) in this region. The system has a resolution of 0.0029 nm/pixel [42]. The spectral range targeted was between 381 and 387 nm due to an abundance of ground state U I and U II transitions [43]. Prior to the experiment, a reference spectrum of the flash lamp was taken and divided out from the spectrum acquired during the experiment, which would be taken at various delay times from the initial spark. The pathlength of the system was 5.08 cm. A Chronos 1.4 high-speed camera was also used to image uranium combustion at 4436.754 fps and 220.63 \( \mu \)s exposure. For the tests pertaining to this document, the ambient environment was air at atmospheric pressure.

Metal powder combustion has been studied extensively in the past, and burn time is a commonly used metric for its evaluation [44–47]. Burn time has strong dependence on particle size, temperature, and oxygen availability. Further
Figure 9: Selected photodiode traces used to determine burn times for various metals tested.
tests to characterize burn times of various similarly sized (<10 µm) metal powder (Ta, W, Zr, Hf, Al, B, Ti, Fe, Mo, and Si) dust clouds in air for comparison with uranium were conducted using a setup as shown in Figure 8.

Two photodiodes (Torbials PDA100A2) measured the intensity of broadband emissions from the combustion of ∼3 mg of various metal powders as well as uranium. These photodiodes measured light from one of the chamber windows, which was split using a beamsplitter and attenuated using neutral density (ND) filtration to ensure that each test would have at least one signal without saturation. Uranium tests were also repeated for comparison and to confirm results from prior testing. Most of the powders were obtained commercially with ≥99% purity from vendors summarized in Table 1. The uranium powder was reduced from the oxide. Powders were then sieved using the Gilson Autosiever Sonic Sifter (GA-6) and 3-inch diameter 10 µm sieve (GAA-83). Uranium powder was sifted by hand in a glovebox due to the high toxicity of inhalation. The Chronos 1.4 high speed color camera was used to image combustion as in previous uranium experiments, taking images every 300 µs.

These tests focused on both burn time acquisition and imaging to observe the occurrence of the exploding particle effect in an attempt to draw further conclusions about the nature of uranium particle combustion. Many of these powders and their burn characteristics have been studied in the past, and drawing commonalities between these and uranium can lead to a better understanding of the
mechanisms through which uranium burns. Table 1 shows the data used to apply Glassman’s criterion to data interpretation, where if the volatilization temperature of the oxide exceeds the boiling point of the metal, the metal shall combust in the vapor phase [58, 59]. Variation in mass used for each test is due to difficulty in measuring the minute quantities of powder necessary for these experiments. Charge masses were chosen to be within the expected flammability limits for the given chamber size, while providing reliable ignition using the existing experimental setup [41].

Since Fe transitions were prevalent in the spectra obtained from the uranium dust cloud. Table 2 shows the energy level information obtained from the Kurucz atomic spectral database used to calculate Boltzmann temperature for Fe I [43]. Iron has a lower temperature threshold for vapor phase formation, and the signatures resulted from impurities present in the test chamber.

### 3. Results and Discussion

Burn time data was obtained for several metal powders that had been sifted to <10 μm in diameter and was quantified as the time between 10% and 90% of the area under the curves obtained from photodiode readouts (Figure 9). This methodology was chosen to eliminate the effect from the large initial spike seen in most of the data as well as to quantify when the bulk of the powder had been combusted. Repeatability is often an issue with metal combustion. Prior work indicates variability in combustion even in single particles of the same size, so repeated measurements should lead to a burn time distribution representative of a given
material [60]. These experiments were designed to provide relative information on uranium combustion kinetics in the test configuration. All tests were performed in 1 atm air. Burning time data based on luminosity traces for Al, B, Hf, Fe, Mo, Si, Ta, Ti, W, Zr, and U dust clouds were obtained. It is important to note that burning powders cannot necessarily be distinguished from other hot particles (i.e., oxide products) that may be present in the observed window, and that higher temperatures will dominate emission. Due to this, the calculation of these burn times involves the

![Figure 13: Emission from uranium dust cloud combustion in 1 atm air environment.](image1)

![Figure 14: Absorbance at 1 ms delay from spark in 1 atm air. The red peaks are Fe I transitions modeled at the Boltzmann calculated temperature of 3428 K.](image2)

| Element | Air (K) | O₂ (K) |
|---------|---------|--------|
| Al      | 3668.93 | 3968.53|
| B       | 4058.79 | 5543   |
| Fe      | 3014.59 | 3383.55|
| Hf      | 4120 [35]| —      |
| Mo      | 3191.98 | 4109.48|
| Si      | 3072.56 | 4899.67|
| Ta      | 3778.5  | 5062.85|
| Ti      | 3595.08 | 4461.72|
| W       | 3494.22 | 4374.91|
| Zr      | 4000.69 | 5118.31|
| U       | 3773.32 | 4843.2 |
assumption that all light emitted is due to burning and not other emissions. The Al burn times were comparable to those reported by Gill et al. for similar diameters [60]. B particle burn times were slightly longer than those previously reported for single-particle measurements in acetylene flames, but were similar to those from laser ignition [61]. Fe burn time was also comparable [62]. Titanium burn times were similar to those reported by Badiola and Dreizin for laser spark ignition, and Zr burn times were slightly longer [47]. Similar data for Hf, Mo, Si, Ta, and W were not found in the literature for particles of similar size.

There is some scatter in these measurements, possibly due to slight differences in masses tested or condensed phase emissions. Measurements in which photodiodes readings were saturated with light were excluded from this dataset since they would skew the calculation of burn time. The two photodiodes were measuring the same field of view with different amounts of neutral density (ND) filtration, so that if one saturated during a given test, time data might still be acquired by the other. Small particle explosions cause momentarily brighter emissions, causing spikes to appear in some of the data traces. This phenomenon is discussed later. Photodiode readings may also vary depending on locations of particles, as although the field of view was maximized, parts of the chamber were obscured from view of the photodiodes. There is much particle movement from the initial upward jetting, impacts with the chamber surfaces, and subsequent falling and mixing.

As for uranium, some preliminary measurements can be found in Read’s thesis [41]. Those burn times were much shorter than those reported in this document on average, possibly due to the much lower ambient pressures, different neutral density filtration (to prevent detector saturation) and larger masses tested (Figure 10).

Multiple of these materials were found to burn heterogeneously in the literature. Based on the previous work using this system, it is unlikely that temperatures over 4000 K were reached [41]. Burn times for each material tested are summarized in Figure 11. For some species, there is considerable variation in measurements.

These results on average correlate to relative burn time data in which Zr had shorter burn times than Ti for particles of roughly the same size [47] and results in which Al had longer burn times than Ti powders [63]. In tests of small amounts of high explosive materials doped with Al and B, B exhibited the slower burn rate [64]. Since these

Figure 15: Aluminum dust cloud storyboard (2.2 mg).
data correlate with relative burn times found in other publications, it may be reasonable to conclude that U burn times are on average longer than that of Zr, Ti, and Al, and shorter than that of other species tested for particles of the same size.

Burn behavior was also observed for each material tested. At the temperature range tested, uranium is expected to burn heterogeneously, since its oxide volatilization temperature is lower than the metal boiling point. A storyboard of uranium dust cloud combustion is shown in Figure 12 in a 1 atm air environment.

Additionally, there is an exploding particle agglomerate effect at later times. This phenomenon is similar to that seen by Zepper et al. in aluminum combustion and by Dreizin in refractory materials such as zirconium and titanium [65–69]. As zirconium combusts in the presence of air, liquid ZrO$_2$N$_y$ intermediates form and ZrO$_2$ precipitates out with gaseous nitrogen contained in voids. Newly exposed Zr burns with gaseous oxygen as the previously contained nitrogen escapes [66, 69]. A similar process occurs in titanium combustion, wherein a liquid solution of Ti, O, and N containing gaseous nitrogen contain voids simultaneously forms stable titanium oxides and an unreacted Ti surface whilst gaseous nitrogen escapes [70]. It stands to reason that this sequence of events could also be occurring in the exploding particles of uranium based on the reported U-O and U-N phase diagrams [14, 15, 71, 72].

An appendix is provided with images from the combustion of other metal powders. Rather interestingly, powders that produced more robust clouds tended to have higher ignition temperatures in dust cloud environments as reported in the literature [73], but all ignition temperatures were still much lower than those reached by the system. W, Fe, and Mo were considered weakly explosible in comparison to other materials tested as determined by rate of pressure rise after ignition of the powder dust clouds, whereas Al, U, Ti, and Zr were considered severe [35, 73].

Spectral measurements were taken from uranium combustion to determine what emission or absorption features would appear in these conditions of excitation. Emission from a uranium dust explosion in a 1 atm air environment is shown in Figure 13. Continuum emission was subtracted out.

Fe I signatures from trace amounts of particulate present dominate the spectrum, indicating that the temperature of
combustion is insufficient to excite uranium transitions. Figure 14 shows the absorbance spectrum from combustion of uranium at a 1 ms delay from initial combustion with an overlaid model at a Boltzmann method calculated temperature of 3428 K. Assuming complete dispersal of the powder, the maximum concentration of uranium in the chamber should be 9.53 g/m³ or $2.41 \times 10^{22}$ atoms/m³.

The concentration of iron in the chamber should be much lower than that of the uranium present, but iron transitions are excited at much lower temperatures than that of uranium. The iron concentration along the path measured was determined to be $1.976 \times 10^{17}$ atoms/m³ which corresponds to about 18.24 μg. The total amount of uranium powder used in this test was 25 mg. Using the Beer-Lambert Law, and with the assumption of constant attenuation coefficient at a given moment along the optical path through the dust cloud, the detection limit of uranium was $2.781 \times 10^{16}$ atoms/m³ along the path from correlation to the ground state Fe I transition at 385.9911 nm. This region of the electromagnetic spectrum, ∼370–395 nm, has an abundance of ground state U I and U II transitions, which are highly likely to appear in absorption if vapor phase uranium is detected. This further corroborates the conclusion that uranium is not burning in the vapor phase at this temperature.

Table 3 shows the adiabatic flame temperatures as calculated in NASA CEA in Air and O₂ for all powders tested with the exception of Hf, which was found in a Snyder [10, 35]. Of these elements, Fe had the lowest adiabatic flame temperature in Air, which corroborates its appearance in the vapor phase. Since vapor phase uranium signatures were not detected, it is likely that temperature in the system did not exceed ∼3773 K, which is supported by temperatures gleaned from emission and absorption measurements of the uranium dust cloud, consistently lower than that in Figure 14. The amount of uranium and oxygen present in the system would constitute a fuel lean mixture, and thus would burn at a lower maximum temperature, certainly lower than the volatilization temperature of uranium oxide and the boiling point of uranium. Badiola and Dreizin noted that NASA CEA code does not take into account nitride formation [47]. The observed temperature is somewhat higher than that theorized by Mouradian and Baker for uranium and zirconium spheres of this size regime burning in air [3].
Not much information was found dealing primarily with the burning of uranium aside from the prediction of burn temperatures and rates for uranium of various sizes and geometries [3], but similarities can be drawn from work on Zr. Like uranium, Zr behaves refractorily, burns heterogeneously, and exhibits the exploding particle effect described earlier. Combustion of micron regime Zr and Ti yields spectra reminiscent of blackbodies with no unique discernible atomic transitions (aside from the iron impurity). Similarly, uranium transitions are absent from Figures 13 and 14. Uranium has a relatively thick protective oxide layer that further prevents the particle from entering the vapor phase, and it oxidizes readily [5, 39, 40, 74]. This oxide layer builds quickly, and that increased oxide layer thickness leads to increased emissivity [75].

Figure 18: Hafnium dust cloud storyboard (3.9 mg).
Figure 19: Molybdenum dust cloud storyboard (3.4 mg).
Figure 20: Silicon dust cloud storyboard (2.6 mg).
Figure 21: Tantalum dust cloud storyboard (shot 2, 2.8 mg).
Figure 22: Tantalum dust cloud storyboard 2 (shot 1, 2.2 mg).
Figure 23: Titanium dust cloud storyboard (3.0 mg).
Figure 24: Tungsten dust cloud storyboard (3.0 mg).
4. Conclusions

From comparison to other metal powders, inferences were made about the dust cloud combustion of micron-sized uranium powder. It was determined that the burn temperature was limited to the volatilization temperature of uranium dioxide per Glassman’s criterion, and that combustion behavior was similar to that of zirconium and other refractory metals in terms of burn time and exploding particle phenomena. From these similarities, it is conjectured that uranium also goes through the sequence of phase transitions, oxide precipitation, and gaseous nitrogen void formation during combustion that leads to this occurrence. Calculation of a detection limit from the Fe I signatures observed led to the conclusion that less than $\sim 2.781E+16$ atoms/m$^3$ U I was present in the path measured. This is due to the low temperatures achieved in the system ($\sim 3400$ K) as compared to uranium oxide volatilization, as well as the strong containment of the uranium metal by the oxide, which inhibits vapor formation. Burn times yielded in this study seem consistent with relative burn times for different species from other publications; however, it may be beneficial to conduct a study on burn times of singular uranium particles to obtain a more accurate burn time. Similar work could be performed under conditions of varying pressure and O$_2$ content, as well as shock tube heating of uranium powder at controlled temperature to observe change in spectra produced.

Appendix

Additional dust cloud storyboard images. (see Figures 15–25).

Data Availability

Data can be made available upon request from Nick Glumac: glumac@illinois.edu.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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