A STUDY OF IGNITION PHENOMENA OF BULK METALS BY RADIANT HEATING

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Introduction

Early research work on combustion of metals was motivated by the knowledge of the large heat release and corresponding high temperatures associated with metal-oxygen reactions. The advent of space flight brought about an increased interest in the ignition and combustion of metallic particles as additives in solid rocket propellants (Refs. 1 and 2). More recently, attention has been given to the flammability properties of bulk, structural metals due to the number of accidental explosions of metal components in high-pressure oxygen systems (Ref. 3).

The following work represents a preliminary study that is part of a broader research effort aimed at providing further insight into the phenomena of bulk metal combustion by looking at the effects of gravity on the ignition behavior of metals. The scope of this preliminary experimental study includes the use of a non-coherent, continuous radiation ignition source, the measurement of temperature profiles of a variety of metals and a qualitative observation of the ignition phenomena at normal gravity.

The specific objectives of this investigation include: a) a feasibility study of the use of a continuous radiation source for metal ignition, b) testing and characterization of the ignition behavior of a variety of metals and c) building a preliminary experimental database on ignition of metals under normal gravity conditions.

The advantage of using a continuous radiation light source lies primarily on its non-intrusive character and on the nature of its spectral distribution. A short-arc lamp provides broadband radiation in the ultraviolet, visible and near-infrared regions (in the 200-1200 nm range); since the absorptivity of metals increases significantly with decreasing wavelength within this region, this feature will in turn result in a decreased input power requirement. The flexibility, ruggedness and low-weight properties of these lamps make them also very attractive for future experiments on high-speed centrifuges (for elevated gravity environments) and zero-gravity simulators (for reduced gravity experiments).

The metals tested in this investigation, iron (Fe), 1018 carbon steel, titanium (Ti), deoxidized copper (Cu) and aluminum (Al) were selected because of their importance as elements of structural metals and their simple chemical composition (pure metals instead of multi-component alloys to avoid complication in future morphology studies). These samples were also chosen to exemplify the two different combustion modes experienced by metals: heterogeneous, surface oxidation (Fe, 1018 carbon steel, Ti and Cu) and

homogeneous, gas phase reaction (Al). In addition, these specimens exhibit a wide variety of melting temperatures, heats of combustion, oxide formation processes and adiabatic flame temperatures. Copper was selected since no reported cases of copper rod burning were found in the literature. 1018 carbon steel was chosen for comparison purposes since it has an almost identical composition as iron except for a 0.18% carbon content difference (and less than 1% of other elements). The experimental approach consisted in providing temperature profiles, ignition temperature values and a visual record of the heating, ignition and combustion stages of the metal specimen.

Experimental Apparatus and Procedures

The experimental system is shown in Figure 1. The radiation source consisted of a 1000 W xenon short-arc lamp with 250 W of broadband output radiation power (with approximately 42% of output power between 800-1200 nm and the rest between 200-800 nm).

The high-intensity, non-coherent light comes out of the lamp in a collimated beam (4° half-angle) and is then intercepted by an aspheric lens (8 cm diam., 6 cm f.l.) which provides the maximum light collection efficiency for a given focal length under these conditions. The beam is then focused down to a 5 mm spot on the top surface of a metal sample providing approximately a 1 MW/m^2 power density (corrected for transmitting medium absorption). The cylindrical metal specimen is 5 mm in diameter and 5 mm high and rests on top of a circular refractory brick base which isolates the sample from heat conduction to its pedestal. These components are located inside a 4.5 liter stainless steel cylindrical chamber which permits input radiation and visual access through a 10 cm diam., 2 cm thick quartz window. An oxygen environment (99.5% pure O_2) was used in all cases at absolute pressures between 0.4 and 4.0 atm. The chamber pressure was monitored by a solid-state piezoresistive pressure transducer (0-20 atm range). The specimen surface temperature was measured by a Pt-Pt 10% Rh (0.1 mm diam.) thermocouple located at approximately 0.3 mm below the surface. Visual observations were recorded with a black and white Panasonic camera with a #5-density aluminized mylar filter.

Prior to the experimental run, the metal sample was thoroughly cleaned with acetone and placed on top of the refractory firebrick. Care was taken to assure that the thermocouple made good contact with the metal. After closing the combustion chamber, three evacuation and filling cycles were executed to assure a pure oxygen atmosphere inside. The lamp was turned on to full power and the beam was focused on top of the metal specimen. The experiment was monitored throughout by temperature and pressure displays and the video camera. After complete combustion, the sample was allowed to cool down and the final pressure was recorded to quantify oxygen consumption. The chamber was opened and the solid combustion products were stored for future analysis. A minimum of 5 samples of each metal were tested to ensure consistency and experimental repeatability.

Results and Discussion

Initial testing of the short-arc lamp was performed with simple heating and melting experiments with the different metals in air at atmospheric conditions. Melting was successfully achieved in all cases in time periods ranging from 15 to 25 seconds. The lamp was then tested for radiant ignition of metal samples in a pure oxygen atmosphere. All metals except for aluminum exhibited ignition and combustion within the same time period as with the melting tests. The results obtained are summarized in the next three sections.

I) Qualitative observations

A visual record of all successful experimental runs revealed the existence of three important and distinctive phases:

a) The heating period included an initial sudden thermal expansion of the top surface of the specimen after radiation was applied. Surface structure changes were also detected by the variation of the radiation characteristics of the sample. These surface changes are related to the allotropic nature of metals in the solid phase, the oxide layer formation and the solid-liquid phase transition.

b) The ignition phase was characterized by the formation of a liquid metal pool on the top surface of the sample, followed by rapid oxidation and a noticeable increase in energy release evidenced by the intense radiation emitted from the reaction zone.

c) The combustion event, i.e., the self-sustained propagation of the reaction zone was accompanied (in the case of iron, 1018 carbon steel and titanium) by a violent exothermic reaction including outward expelling of small particles that collected in the walls and window of the chamber. The detection of intense luminosity is due to the high temperature (3000-4000 K) obtained in the reaction zone. A marked difference in reaction intensity was observed between the iron and the 1018 carbon steel samples, where the latter exhibited a much higher flux of ejected hot particles probably due to its carbon content. The behavior of copper in the combustion phase had very different characteristics as explained in the Special cases section.

II) Temperature profiles

Temperature records also revealed uniquely the existence of the heating, ignition and combustion stages. Figure 2 shows the temperature histories of titanium, copper, iron and 1018 carbon steel.

All samples showed an increase in temperature with asymptotic approach to a steady-state value in the first few seconds of radiative heating. In the preliminary melting experiments in air the steady-state temperature value (not shown) was fixed by the melting point of the metal due to the constant-temperature phase change. In contrast, a point of inflection was reached on the temperature vs. time plots corresponding to ignition experiments in pure oxygen (see Figure 2). This transition point is identified as the temperature at which the heat generated by the metal oxidation first exceeds the heat lost through conduction, convection and radiation. This temperature is defined in this investigation as the critical ignition temperature of the metal sample. Beyond this point, the metal specimen is driven into a thermal runaway region as a consequence of the exponential dependence of the reaction rate on temperature. The acceleration of the reaction rate ultimately reaches the catastrophic oxidation level which defines the exothermic, self-propagating combustion zone. At this stage, the temperature drastically increases to the vaporization temperature of the metal oxide (approx. 3000-4000 K depending on the metal type) where it stays throughout the combustion phase. As shown in Figure 2, the thermocouples were unable to follow the temperature increase in this last stage because their maximum operating temperature is around 2000K.

Table 1 shows the heat of combustion, melting temperature, ignition temperature, and adiabatic flame temperature of the different metals tested. In the case of iron, 1018 carbon steel, copper and titanium the ignition temperature is always below or in the range of the melting temperature of the metal. This result is typical of metals exhibiting heterogeneous reaction with non-protective oxide layers. In the case of iron and 1018 carbon steel, the temperature profiles and ignition temperatures are very similar due to the almost identical chemical composition. For the titanium run shown in Figure 2, the thermocouple broke before the temperature curve reached the point of inflection corresponding to the critical value for ignition (approx. 1720 K).

III) Special cases

The following cases represent two noteworthy examples of different behavior from the previously mentioned results.

a) Aluminum. All ignition tests performed on aluminum were unsuccessful. This metal proved to be impossible to ignite due to the formation of a thick, extremely resistant, insulating oxide layer that prevented the molten metal inside to come into contact with the surrounding oxygen atmosphere. The existence of this protective aluminum oxide layer has been confirmed by other studies (Refs. 4 and 5). The ignition temperature of aluminum is much higher than the melting temperature of the metal and it actually corresponds to the melting point of the metal oxide (Al_2O_3). Different techniques (including forced mechanical stressing and prolonged radiation heating) were used to induce ignition in the metal sample, but all proved unsuccessful.

b) Copper. A very interesting and unexpected result of this investigation was the ignition and combustion of copper samples. Previous work (Ref. 6) has classified copper as a non-flammable metal due to the sudden extinction of the rod after ignition on upward propagation studies. However, under our experimental configuration, clear evidence of ignition and subsequent indefinite downward propagation of copper specimens was obtained at 0.84 atm. In contrast with iron, 1018 carbon steel and titanium, copper exhibited a smooth combustion phase characterized by a slow regression rate with a low-luminosity reaction zone. This behavior may be due to the low heat of combustion which retards the melting and subsequent oxidation of the metal in the molten surface. This same argument may explain the difficulty of burning copper in the upward propagation mode. Under this configuration, the detachment of the first molten ball from the vertical rod removes the thermal mass that preheats the solid metal in front of it and causes a rapid cooling of the reaction zone (where the low heat of reaction is unable to overcome heat losses) with extinguishment following afterwards. Several tests with different specimen lengths (up to 30 mm) were made to provide unmistakable evidence of self-propagation and complete oxidation of the sample. In addition, all tests showed a noticeable decrease in chamber pressure (2-5%) providing further indication of complete metal oxidation.

Conclusions

This study has demonstrated the effectiveness of using a non-coherent, continuous radiation source for metal ignition. In addition, ignition and combustion tests were performed on different metal specimens, including iron, 1018 carbon steel, titanium and copper. Qualitative observations and temperature profiles have revealed the existence of the heating, ignition and combustion stages. These zones are characterized by well-defined surface structure changes, surface temperature behavior, luminosity and chemical activity. Values of ignition temperatures (defined here as the critical point of inflection in the temperature vs. time curve) were obtained and compared with results of previous investigations (Refs. 4,8,9). Good agreement (+/- 10%) was found considering the differences on ignition temperature definition and experimental technique used among the various studies.

The experimental work is being complemented by modeling studies which include the effects of conduction, convection and radiation heat transfer in the solid specimen and the surrounding gas. The computational model explores the effects of gravity (0.0 g to 10.0 g) and pressure (0.15 atm to 10 atm) on the heating phase of the metal sample under non-oxidizing conditions. Preliminary results indicate that for a constant input heat flux on the specimen, there is an increase in convection heat transfer that asymptotically approaches a constant value both for increasing gravity and for increasing pressure. This limit in convection heat transfer imposes a limit on the minimum steady state specimen temperature

obtained. The heat transfer studies are designed to help evaluate the energy balance on the specimen during heating prior to ignition.

Acknowledgements

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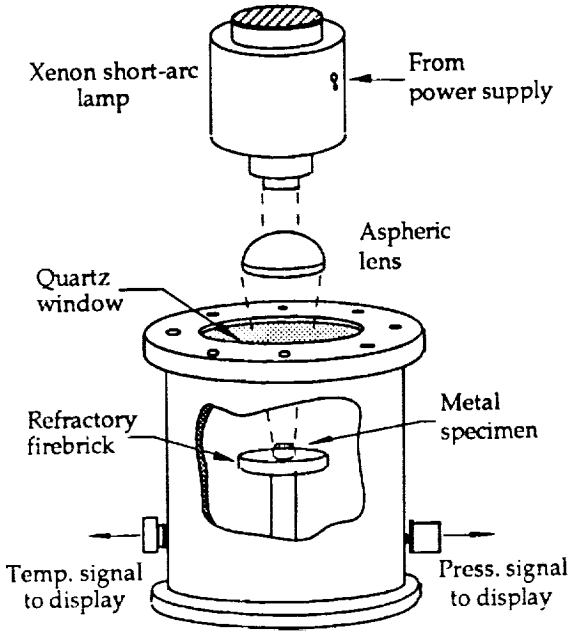

Figure 1. Experimental Apparatus

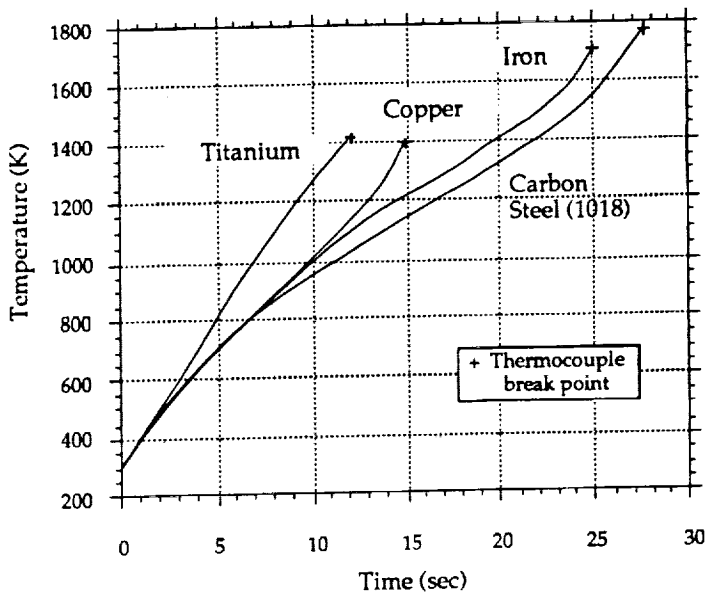

Figure 2. Temperature vs. time behavior of iron, 1018 carbon steel, copper and titanium

Metal Specimen	Heat of Combustion (cal/g)	Melting Temperature (K)	Ignition Temperature (K) [a]	Adiabatic Flame Temp. (K)
Aluminum (Al)	7425	930	2300	3800
Titanium (Ti)	4710	1963	1720	3300
Iron (Fe)	1765	1812	1490	3000
Carbon Steel (1018)	1765	1812	1450	3000
Copper (Cu)	585	1356	1060	1635

[a] From this investigation, except for aluminum from Brzustowski, et. al. [7]

Table 1. Heats of combustion, melting, ignition and adiabatic flame temperatures for the different metal specimens used in this investigation.

COMMENTS

Question (John Moore, Colorado School of Mines): I noticed that you said you did not ignite the aluminum, yet you quote an ignition temperature for aluminum of 2300 K - this is the approximate melting point of aluminum - there must be some connection. Can you please comment?

Answer: In the present investigations of the ignition of metals by a continuous radiation source we were unable to ignite the aluminum specimens. We feel that this is due to the limitation of the power of the radiant ignition source used. In previous studies using a CO_2 laser for ignition (Reference 4 in the paper) we were able to ignite specimens of Al 6061. In those studies the aluminum specimen formed a solid aluminum coating on the surface with molten aluminum in the core of the specimen. The ignition occurred when the oxide coating approached its melting temperature and weakened sufficiently to crack or otherwise expose the molten aluminum to the oxygen. Therefore, there is a connection between the melting temperature of aluminum and the ignition process in the aluminum specimens.