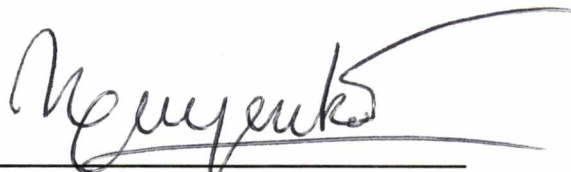


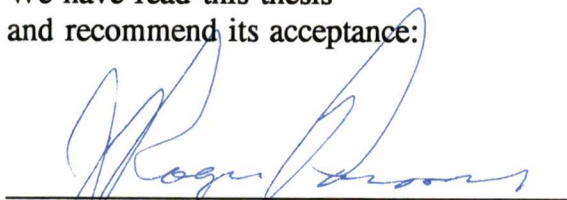
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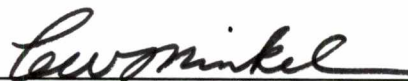


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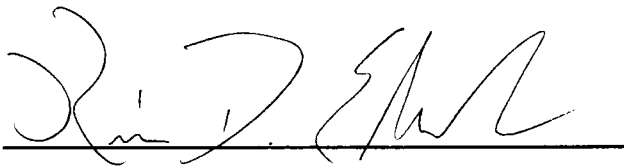
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**EFFECTS OF INITIAL DIAMETER AND AMBIENT PRESSURE
ON THE IGNITION AND COMBUSTION
OF LASER-IGNITED ALUMINUM AGGLOMERATES**

A Thesis

Presented for the

Master of Science

Degree

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Kevin Dean Edwards

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ABSTRACT

An experimental investigation was conducted to determine the effects of initial diameter and ambient pressure on the ignition and burning characteristics of aluminum agglomerates. Agglomerates with initial diameters of 535, 835, and 1000 μm were ignited by a CO_2 laser pulse in an oxygen/nitrogen (20/80) environment at pressures ranging from 0.1 to 4.3 MPa. The combustion event was recorded using high-speed cinematography. A two-color pyrometer and broad-band radiometer were used to measure the surface temperature of and the radiation emitted by the burning droplet. Data from these three sources was used to determine the ignition delay time and burning time of the aluminum agglomerates.

Analysis of the combustion residue showed that the combustion process at atmospheric pressure varies greatly from that at elevated pressures. The products from experiments conducted at atmospheric pressure consisted of hollow spheres of oxide. Experiments conducted at elevated pressures resulted in products consisting of a small core of unburned aluminum surrounded by an opaque layer of oxide.

The ignition delay time and burning time were found to be dependent upon ambient pressure. The ignition delay time and burning time initially decrease with increasing pressure. However, a pressure limit is eventually reached beyond which increasing pressure no longer has an effect. Power law analysis showed the ignition delay time to be proportional to the initial agglomerate diameter raised to the power of 1.28. The burning time was found to be directly proportional to the diameter.

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CHAPTER 1

INTRODUCTION

The use of slurry fuels in air-breathing propulsion systems has recently attracted considerable interest due to the advantages of such fuels. Slurry fuels consist of micron-sized solid fuel particles and a liquid fuel carrier. This mixture provides the high energy content of a solid fuel while maintaining the ease of injection of liquid fuels. As a result, the use of slurry fuels in aircraft propulsion systems can provide a greater thrust and range without requiring an increase in the volume dedicated to fuel storage.

Studies on coal-oil and carbon-black slurries [1-4] have shown that combustion of slurry fuels occurs in two stages. In the first stage, the liquid component is rapidly vaporized and consumed leaving behind the powdered solid fuel which coalesces into porous agglomerates. In the second stage, the hot oxidizing gases heat the agglomerate to ignition and full-fledged combustion.

Unfortunately, the use of slurry fuels presents some serious problems. The combustion of the agglomerates requires far more time than the original micron-sized particles. Since residence time within the combustion chamber is typically less than 100 ms, the combustion process might not be complete when the fuel leaves the chamber. This results in reduced efficiency in the combustor and expansion nozzle and could even cause serious erosion problems in compact combustors and in expansion nozzles.

Metals such as aluminum, boron, and magnesium have been considered as possible solid components of slurry fuels because of their high volumetric energy content and high flame temperature. Aluminum has an advantage over other metals in that it primarily burns in the vapor phase allowing for a higher reaction rate than for a metal such as boron which primarily reacts at the droplet surface. Theoretically, the flame temperature is approximately equal to the boiling temperature of aluminum oxide (~ 3800 K). The high flame temperature increases the vaporization rate of aluminum thus accelerating the combustion rate. However, there are some inherent problems in the use of aluminum as the solid component due to the high ignition temperature and the presence of condensed products. Since the oxide layer which forms around the particle must be melted before the aluminum core can be ignited, the ignition temperature is approximately equal to the melting temperature of aluminum oxide (2315 K). In addition, condensed aluminum oxide produced at the flame front is convected inward and collects on the surface of the molten aluminum droplet. The oxide forms caps on the surface of the droplet and inhibits the diffusion of aluminum vapor sometimes resulting in flame extinction.

The combustion processes of single aluminum particles has undergone considerable study; however, study of aluminum agglomerate combustion has been undertaken only recently. Turns, Wong, and Ryba [5] studied the ignition and combustion characteristics of aluminum slurry agglomerates with initial diameters of 300 to 800 μm . The agglomerates were ignited by exposure to the hot product gases from a flat flame burner. The study investigated the effects of oxygen concentration,

ambient gas temperature, and initial diameter on the burning time of the aluminum agglomerates. However, in this study, the ambient pressure was limited to atmospheric whereas typical combustor pressures range from 0.1 to 2.0 MPa. Meftah [6] conducted a study to determine the effects of ambient pressure on the ignition delay time and burning time of aluminum agglomerates. The agglomerates were ignited with a CO₂ laser pulse in a quiescent oxygen/nitrogen (20/80) environment with ambient pressures ranging from 0.1 to 4.3 MPa. However, Meftah's study was limited to agglomerates with an average initial diameter of 535 μm .

The objective of this study is to investigate the effects of both ambient pressure and initial diameter on the ignition and combustion characteristics of aluminum agglomerates. Agglomerates with average initial diameters of 835 and 1000 μm were ignited using a CO₂ laser pulse in a quiescent oxygen/nitrogen (20/80) environment at ambient pressures ranging from 0.1 to 4.3 MPa. In addition, results from the previous study by Meftah [6] using 535 μm diameter agglomerates at the same pressures were incorporated into the analysis.

The agglomerates were supported on the tip of SiC fibers and mounted inside a high-pressure combustion chamber. Ignition of the agglomerates was achieved by a 300 ms pulse from a CO₂ laser beam. The beam's power was maintained at approximately 35 W. The flame structure was studied using high-speed cinematography. A fast-response, near-infrared, two-color pyrometer and a broadband radiometer were used to measure the surface temperature of the burning droplet

and the intensity of the emitted radiation, respectively, during the combustion process. Results from the pyrometer and radiometer along with the cinematographic records were used to determine the ignition temperature, ignition delay time, and burning time.

In this study, the ignition point is defined as the point at which the flame first appears in the cinematographic record. By comparison with the cinematographic records, the ignition point was found to be characterized by a sharp peak on the intensity plot. Once the ignition point is determined, the ignition delay time is taken as the time elapsed between the beginning of laser irradiation of the agglomerate and the ignition point. Once ignited, the droplet undergoes full-fledged, self-sustained combustion. Using the cinematographic records, the end of the combustion process was found to correspond to a sudden transition to an exponentially decaying cooling curve on the intensity plot. The burning time can then be determined as the time elapsed between ignition and the end of self-sustained combustion.

In addition, it is hoped to determine whether the combustion rate of the aluminum agglomerates is controlled by mass diffusion or chemical kinetics. As Mulcahy and Smith [7] point out, if the combustion rate is diffusion controlled, the burning time is independent of ambient pressure. For kinetics-controlled reactions, however, an increase in pressure should result in a decrease in burning time. Results from this study are expected to confirm Meftah's results [6] by showing that as pressure is increased, the combustion of aluminum transitions from a kinetics to a diffusion-controlled reaction.

Chapter 2 is a survey of the available literature on previous studies of the ignition and combustion processes of single aluminum particles and agglomerates. Chapter 3 gives a more detailed description of the experimental apparatus and procedure used in this study. An analysis of the results from the current study is presented in Chapter 4. Finally, Chapter 5 presents conclusions from this study.

CHAPTER 2

LITERATURE SURVEY

The majority of research into the combustion characteristics of aluminum has been concentrated upon the burning of single aluminum particles. However, investigations of the burning of coal-oil and carbon-black slurry fuels [1-4] have shown that the combustion of such fuels occurs in two stages. First, the liquid fuel carrier vaporizes and is consumed leaving behind the powdered solid fuel. The solid fuel particles then coalesce into porous agglomerates. In the second stage, the agglomerates are heated and ignited. Few studies have been undertaken to determine the burning characteristics of these porous aluminum agglomerates.

This chapter gives an overview of the available literature on studies of the combustion characteristics of both single aluminum particles and porous agglomerates. Section 2.1 presents previous studies of the combustion of single aluminum particles while Section 2.2 reviews studies on the combustion characteristics of aluminum agglomerates.

2.1 COMBUSTION OF SINGLE ALUMINUM PARTICLES

As pointed out by Davis [8], adding particles of powdered aluminum to solid fuel propellants will theoretically increase the specific impulse and available thrust. In practice, however, the increase in specific impulse never achieves the full

theoretical potential and under some conditions it even decreases. Davis postulates that the two principal causes for this phenomena are incomplete combustion due agglomeration of the aluminum particles and the inability of the condensed products to reach thermal and kinetic equilibrium during the expansion process.

In an effort to determine the optimum conditions to increase the thrust efficiency, aluminum particles ranging in size from 53 to 103 μm were ignited at atmospheric and elevated pressures. Experiments at atmospheric pressure were conducted by injecting the aluminum particles into the center of the premix flame of a Meker-type burner. A pressurized bomb was used to burn a mixture of ammonium perchlorate and paraformaldehyde containing aluminum particles in a nitrogen environment at elevated pressures.

Using photographic records of the atmospheric tests, Davis discovered that the flame radius was larger than the radius of the initial aluminum particles. A plume of oxide was present in the wake of the burning particles. These two phenomena are indicative of vapor-phase oxidation. In addition, hollow spheres of oxide of the same approximate size as the initial particles were discovered in the combustion products. In the later stages of combustion, Davis discovered that the burning droplets exhibited periodic luminosity suggesting that only a portion of the droplet was burning and that it was spinning rapidly. At oxygen concentrations less than 32%, the combustion event ended gradually with the flame intensity slowly decreasing. At oxygen concentrations above 32%, however, the combustion event ended dramatically when the burning droplet violently fragmented into several smaller burning droplets.

At atmospheric pressures, Davis found that the ignition delay time increased as the particle diameter increased. Oxygen concentration was found to have little effect upon the ignition delay time. However, ignition delay time was found to be strongly dependent upon ambient gas temperature. In general, the ignition delay time decreased as the gas temperature was increased. The opposite was found for the burning time of the aluminum particles. Gas temperature had little effect upon the burning time while increasing the oxygen concentration resulted in a decrease in burning time.

At elevated pressures, Davis discovered that the burning time decreased with increasing pressure up to a point after which increasing pressure had no effect upon the burning time. The pressure at which this transition occurred was determined to vary with the composition of the ammonium perchlorate and paraformaldehyde mixture. Davis also found that the burning time was dependent upon the particle size. As would be expected, larger particles exhibited a longer burning time. At pressures above 6.87 MPa, the burning time was found to be proportional to the diameter raised to 1.8 power.

Davis concluded that, in order to achieve efficient combustion, the aluminum particles should be as small as possible to assure complete combustion before the particles leave the combustor. In addition, the propellant-matrix temperature should be sufficiently high to provide a short ignition delay time. Finally, the operating pressure should be in excess of 6.87 MPa to promote the complete combustion of the aluminum particles.

Friedman and Maček [9], investigated the effects of oxygen concentration, ambient gas temperature, and particle size on the ignition and combustion of aluminum particles. Particles with diameters less than $100\text{ }\mu\text{m}$ were ejected into the hot product gases of a flat flame burner. The burner could be controlled to vary the temperature and composition of the product gases. Long-exposure still photographs were used to determine the ignition delay time and the burning time of the particles.

Friedman and Maček discovered that the ignition delay time was proportional to the square of the particle diameter. The ignition limit for the aluminum particles was found to be $\sim 2300\text{ K}$ and was only slightly affected by oxygen concentration. Varying the oxygen concentration from 8% deficient to 36% excess resulted in a variance in ignition temperature of only 2360 to 2200 K as shown in Figure 2.1. Similarly, the particle size was found to have little effect on the ignition temperature.

In an attempt to explain their results, Friedman and Maček developed a model for the ignition of the aluminum particles based on heat transfer by conduction to a sphere of uniform temperature. However, their model showed that the ignition temperature limit should drastically decrease when either the oxygen concentration or particle diameter were increased which conflicted with their experimental observations. Since the ignition temperature limit found experimentally was approximately the same as the melting temperature of Al_2O_3 (2315 K), they proposed that the oxide layer surrounding the particles prohibits the diffusion of aluminum vapor. As a result, the oxide layer has to be melted before the aluminum vapor can diffuse through and react with the oxygen, thus igniting the particle.

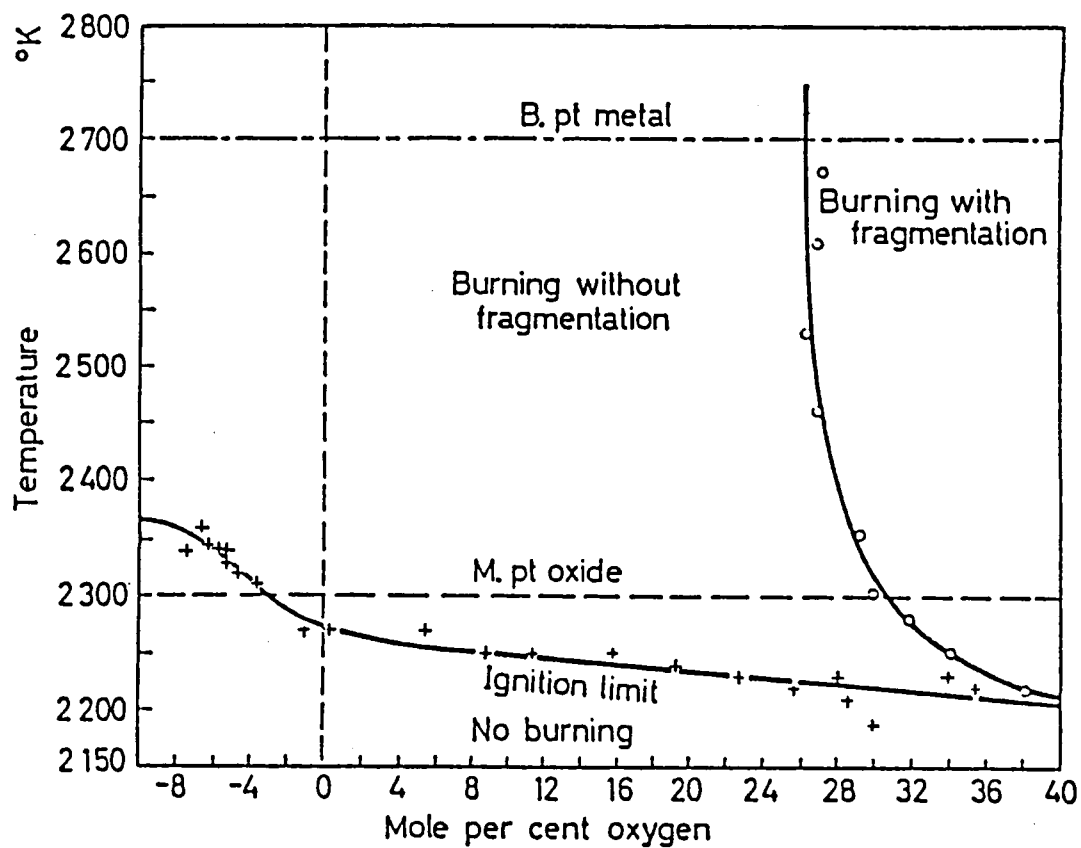


Figure 2.1 Experimental ignition and fragmentation limits for 35 μm diameter aluminum particles as observed by Friedman and Maček [9].

After ignition, Friedman and Maček discovered that two modes of combustion existed characterized by whether or not fragmentation of the burning droplet occurred. For conditions where fragmentation did not occur, hollow shells of oxide were found to form on the surface of the burning droplets. They postulated that heat transfer from the flame front increased the temperature of the molten droplet above the boiling point of aluminum at various hot spots on the droplet's surface. As a result, the vapor pressure inflated the oxide layer covering the droplet thus forming the hollow spheres of oxide. In addition, the droplets were found to experience asymmetric combustion and, hence, to spin periodically.

The burning time of the particles were found to be proportional to the particle diameter raised to the 1.5 power. As the authors point out, since spherical diffusion-flame theory gives burning time to be proportional to the square of the diameter and models with diffusion limited by a molten oxide film give burning time proportional to the diameter, the actual case must lie somewhere between.

Friedman and Maček found that for oxygen concentrations greater 38%, the droplets fragmented shortly after ignition. The oxygen concentration at which fragmentation occurred was found to decrease slightly when the ambient gas temperature was increased as shown in Figure 2.1. The particles which underwent fragmentation had a shorter burning time and were more likely to undergo complete combustion.

In a second paper, Friedman and Maček [10] further investigated the ignition and combustion characteristics of aluminum particles with emphasis on the effects of

water vapor. Particles were again injected into the hot product gases of a flat flame burner. The burner could be controlled to vary the composition and ambient temperature of the product gases.

Friedman and Maček found that the combustion process for the particles burning in a low-moisture environment was similar to those from the previous study at high moisture content with three exceptions. First, whereas in high-moisture environments hollow spheres of oxide were found to grow from the surface of the burning droplets, in low-moisture environments the droplets steadily reduced in size with no evidence of the formation of hollow oxide spheres. The oxide products were found to consist of solid particles of varying size and shape which were smaller than the original aluminum particles. Second, the combustion process was completed much more quickly in low-moisture environments. Third, in photographs taken of experiments conducted at low moisture content, the burning droplets were surrounded by a more diffuse luminosity than in high moisture content experiments which indicates the increased presence of aluminum vapor adjacent to the particle.

Friedman and Maček concluded that the ignition process was unaffected by moisture content. They proposed that the ignition process is controlled only by the diffusion of aluminum vapor through the oxide layer which coats the particle. Ignition is therefore possible only when this oxide layer is melted with no regards to ambient gas composition. However, they concluded that moisture content has a large effect upon the combustion process of the particles. It was found that diffusion and combustion take place in the vapor-phase when little to no water is present.

However, when large concentrations of water is present, the aluminum vapor is forced to diffuse through a layer of condensed oxide slowing the combustion process.

Brzustowski and Glassman [11] developed a theoretical model to describe the combustion processes of aluminum and magnesium droplets in an attempt to explain the difference in results obtained using experimental techniques and previous theoretical models such as the one proposed by Friedman and Maček [9]. The model was based on the typical combustion model for hydrocarbons; however, several changes in the assumptions and approximations used had to be made to account for the differing characteristics of the vapor-phase combustion of metals. For example, at high burning rates, the flame temperature is fixed at the boiling temperature of the oxide. As a result, the oxide produced will be in various phases with at least some in condensed form. The diffusion of the oxidizer toward the flame is inhibited by this condensed oxide. In addition, the condensed oxide cannot diffuse away from the flame front, but instead, must be convected away by the bulk motion of the gases. The condensed oxide increases the importance of radiation in the heat transfer from the flame to the molten droplet and the surroundings. Finally, the evaporation rate of the metal from the molten droplet's surface may be considerably lower than the diffusion rate of the metal vapor to the flame front.

Brzustowski and Glassman's model consists of a spherical droplet of fuel surrounded by a thin concentric flame. The flame and droplet are separated by a stagnant film of gases through which the metal vapor must diffuse. The surroundings consist of another stagnant gas region which stretches to infinity. The oxidizer

diffuses toward the flame front from the surroundings. Both fuel and oxidizer are assumed to reach the flame in stoichiometric proportions and to be consumed completely. The fuel is vaporized and heated to the flame temperature by conduction and radiation of heat from the flame front. The additional energy from the combustion process is transferred to the surroundings. Convection of condensed oxide particles opposes the diffusion of the oxidizer to the flame front. The pressure of the system is assumed uniform and constant and the entire process is assumed to be quasi-steady. Analysis of the model was focused toward the diffusion rates of the oxidizer and fuel to the flame front, the heat transfer rate to the droplet and the surroundings, and the evaporation rate of the fuel at the droplet surface.

Brzustowski and Glassman applied the energy equation to the boundaries of the stagnant films to develop the heat transfer relations. The diffusion and evaporation relations were developed using Fick's Law and a modified form of the Knudsen-Langmuir equation, respectively, for a two species, ideal-gas system. The solution to the resulting governing equations provides a great deal of information about the flame structure and the burning rate. However, gross trends cannot be discerned and the numerical method of solution requires prior knowledge of the values and trends of the variables involved. In an attempt to rectify this problem, Brzustowski and Glassman developed a simplified model in which the flame temperature is assumed constant (at the boiling point of the oxide) and the evaporation rate is assumed to always be sufficient to keep up with the diffusion of vapor to the flame front. The simplified solution allowed the determination of the gross effects of radiation from the flame

zone, identification of dimensionless parameters used to characterize the combustion process, and illustration of the differences between combustion of hydrocarbons and metals. In addition, the simplified model provided a basis upon which the numerical solution to the full-blown governing equations could be constructed.

Results showing the effects of certain parameters on the burning rate and flame stand-off radius based on Brzustowski and Glassman's simplified model are shown in Figures 2.2 and 2.3. The varied parameters include the dimensionless oxidizer flux, βD_o ; the transfer number, B ; and a dimensionless radiation parameter, $\sigma_{B,A}$. The transfer number for the evaporation of fuel is primarily dependent upon and inversely proportional to the latent heat of evaporation for the fuel. The dimensionless radiation parameter is essentially a ratio of the heat transfer rate due to radiation to that due to conduction.

Figure 2.2 shows the effects of these parameters on the ratio of flame radius to particle radius, r_B/r_A . As can be seen, as the transfer number is increased, the flame radius also increases. This is expected since a lower latent heat of evaporation requires a low temperature gradient at the surface of the particle and hence a larger flame radius. The flame radius was also found to increase with increasing $\sigma_{B,A}$. In other words, an increase in the heat transfer rate due to radiation without changing the total rate of energy transfer allows for a lower temperature gradient. Since the temperature at the flame and the surface are fixed at the boiling points of the oxide and aluminum, respectively, a decrease in the temperature gradient requires an increase in the flame radius. Increasing the oxidizer flux, βD_o , was found to cause a

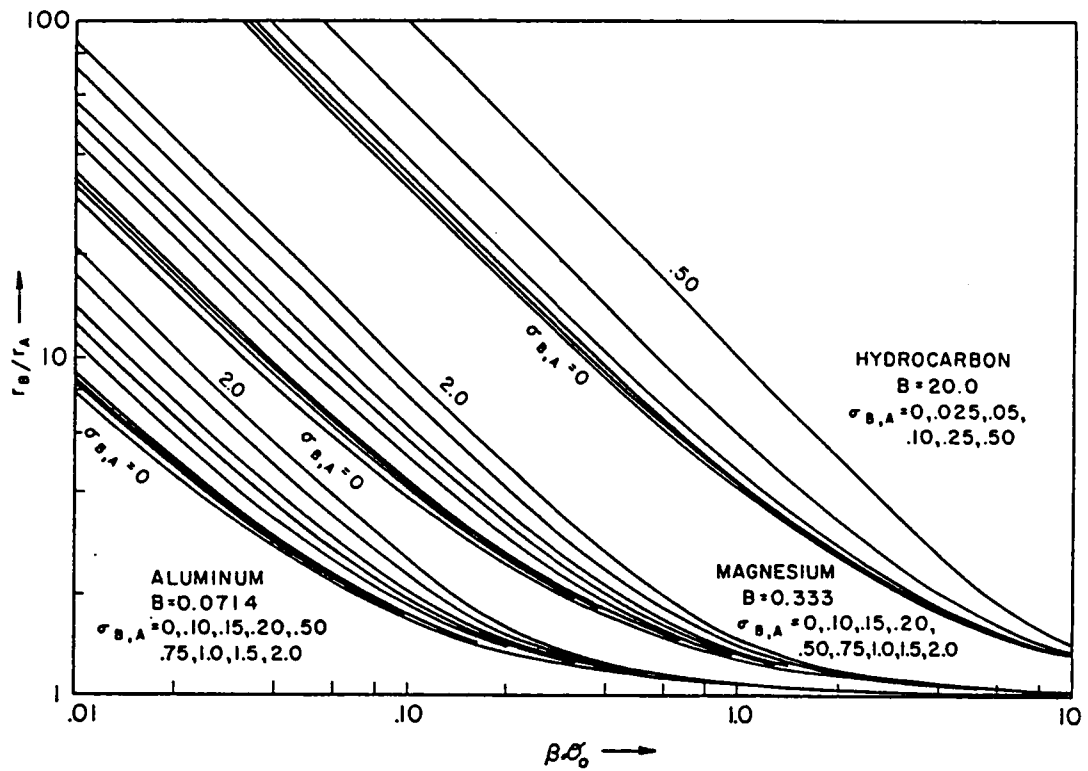


Figure 2.2 Effects of the dimensionless oxidizer flux, βD_o ; transfer number, B ; and radiation parameter, $\sigma_{B,A}$, on the flame stand-off ratio using the analytical model proposed by Brzustowski and Glassman [11].

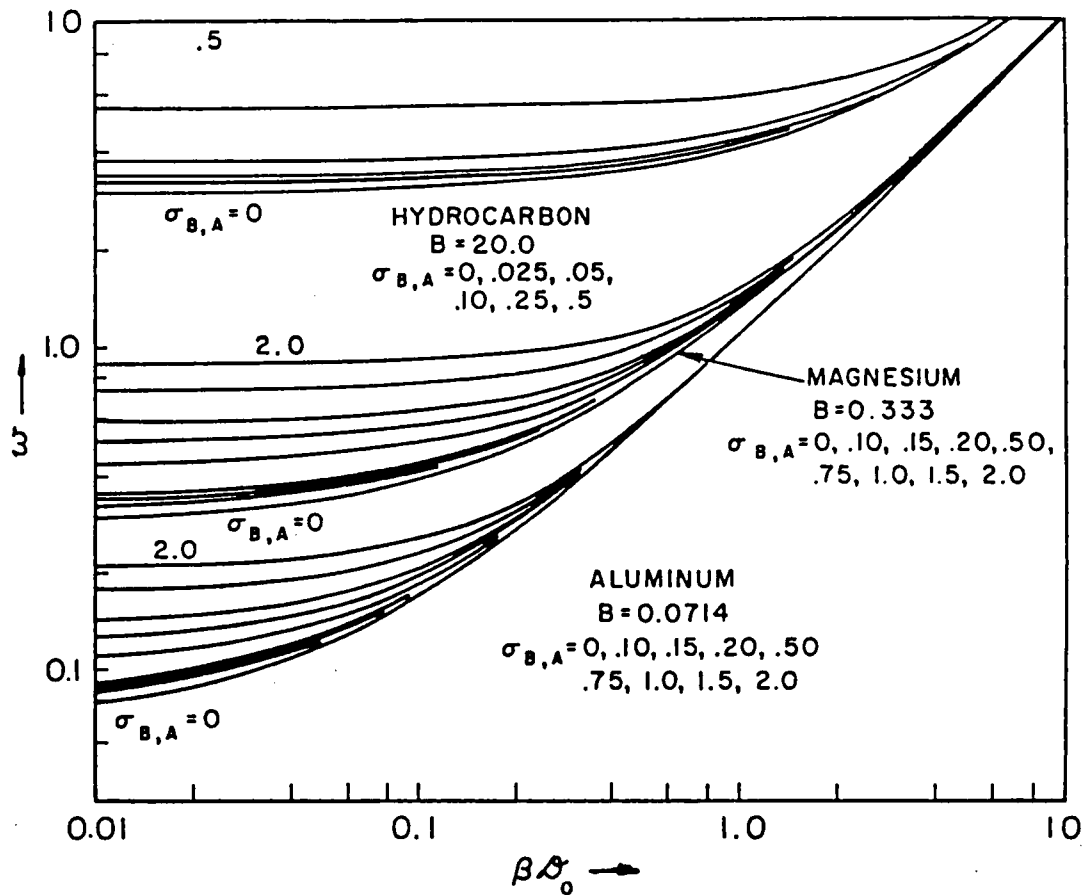


Figure 2.3 Effects of the dimensionless oxidizer flux, βD_0 ; transfer number, B ; and radiation parameter, $\sigma_{B,A}$, on the dimensionless burning rate, ω , using the analytical model proposed by Brzustowski and Glassman [11].

decrease in the flame radius. This suggests that a high oxidizer flux requires a high concentration gradient of fuel vapor. As a result, vapor-phase flames can be expected to be very close to the droplet surface unless the diffusion flux of the oxidizer is very low.

The effects of the parameters on the non-dimensional burning rate, ω , are shown in Figure 2.3. As can be seen, the burning rate increases with an increase in the transfer number. This is reasonable since a lower heat of vaporization results in a higher transfer number. The burning rate was also found to increase with increasing oxidizer flux. As Brzustowski and Glassman point out, the flame zone acts as a sink for the oxidizer, and the diffusion rate of the oxidizer to this sink is proportional to r_B/r_A . At high values of βD_o , the burning rate is almost proportional to the oxidizer flux, and the flame is almost contiguous to the surface of the particle. At lower values, however, the burning rate increases much more slowly. The effects on the burning rate due to the radiation parameter, $\sigma_{B,A}$ is directly related to its effect on r_B/r_A . As $\sigma_{B,A}$ increases, the flame radius, and thus the size of the oxidizer sink, increases which, in turn, results in an increase in the burning rate.

While Brzustowski and Glassman's model provides an indication of the gross trends associated with the combustion process, it does have several limitations. For example, the model assumes that the combustion process is quasi-steady whereas experimental studies have shown evidence of numerous transient phenomena such as jetting, spinning, and fragmenting. Experimental studies have also shown that combustion often occurs only in preferred locations as opposed to occurring over the

entire surface as this model assumes. Finally, the model neglects the convection of condensed oxide from the flame front to the droplet surface. Any effects this condensed oxide has on the burning process is therefore unaccounted for by the model.

Bartlett, Ong, Fassell, and Papp [12] proposed an opposing view to the combustion model developed by Brzustowski and Glassman. In their model, the ignition process begins with the melting of the oxide layer covering the aluminum particle. Vapor from the inner aluminum core, now a molten droplet, begins to diffuse through the oxide coating and react with the oxidizer at the outer surface of the oxide shell thus initiating combustion. The additional heat from the combustion process begins to vaporize the inner aluminum core and the vapor pressure causes the oxide layer to expand. Thus a layer of aluminum vapor is formed between the molten droplet and the concentric liquid oxide shell. The aluminum vapor must diffuse through the oxide shell and then react with the oxidizer at the outer surface of the shell. Bartlett et al. postulated that fragmentation of the burning particle occurs when the oxide shell expands too rapidly. They further postulated that when fragmentation does not occur, the aluminum core is completely consumed. Without heat generated by the reaction or transferred from hot ambient surroundings, the oxide layer quickly cools and solidifies resulting in the hollow oxide spheres found in numerous experimental investigations. The hollow spheres generally range in size from less than 1 μm to slightly larger than the original particle size and sometimes contain unburned aluminum.

Drew, Gordon, and Knipe [13] conducted experiments in an attempt to determine which of the two above mentioned models proved more accurate. A blast burner was supported in an inverted position and aluminum particles were injected into the flame. Since particles were dropped through the flame region as opposed to being convected upward by the bulk gas motion, larger particles could be studied. The inverted orientation also allowed for easier collection of the combustion products by simply placing a porcelain dish underneath the burner. In addition to the photographic method generally used to observe the combustion process, Drew, Gordon, and Knipe used quenching methods to further study the combustion process at its various stages. A collecting plate was passed through the reaction region at a given level. When the burning particles impacted with the collecting plate, they were instantly quenched with no discernable disturbance to their appearance. In this manner, quenching was performed at 0.25 to 1.0 in increments along the path of the burning particles to gain an understanding of the events taking place throughout the combustion process.

While Friedman and Maček [10] concluded the ignition of aluminum particles was unaffected by the presence of water vapor, other investigations such as those by Blackburn and Gulbransen [14] have shown that water accelerates the ignition process. It was postulated that water vapor is responsible for oxidation at the surface of the particle prior to ignition. Drew, Gordon, and Knipe found that particles quenched during the early stages of ignition were expelling a stream of small oxide particles approximately $5\text{ }\mu\text{m}$ in diameter. In addition, particles quenched at a

slightly later stage showed evidence of small surface imperfections of the same size as the oxide particles being expelled. They speculated that this phenomena was the result of surface oxidation by water vapor. In the late stages of the ignition process, an oxide cap was found to form on the droplet surface. Drew, Gordon, and Knipe felt the cap was formed by agglomeration of the molten oxide which had constituted the protective oxide layer prior to ignition.

Studies of particles quenched during self-sustained combustion showed that the flame and the plume of products contained sub-micron-sized oxide spheres. Occasionally particles would impact in such an orientation that a cross-section of the flame-structure was formed as shown in Figure 2.4. The quenched particle in Figure 2.4 clearly shows an asymmetrical burning pattern. Half of the particle is covered with an oxide cap which prevents combustion from occurring. A near-parabolic flame structure surrounds exposed metal half of the particle.

Drew, Gordon, and Knipe found that the residue collected from particles which were not quenched consisted of some unburned aluminum particles, numerous small solid oxide spheres, and several hollow oxide spheres which were larger than the original particles as shown in Figure 2.5. Particles quenched during the combustion process showed signs of the hollow oxide spheres being formed on the half of the particle surface which was covered by the oxide cap. Figure 2.6 shows signs of a hollow oxide sphere growing from the burning particle in the lower half of the photograph. In the upper right hand corner of Figure 2.7, a hollow oxide sphere is shown adjacent to a burning particle which is believed to be its parent. These



Figure 2.4 Photograph of a 150 μm diameter aluminum particle quenched during self-sustained combustion showing the cross-section of the flame structure from Drew, Gordon, and Knipe [13].

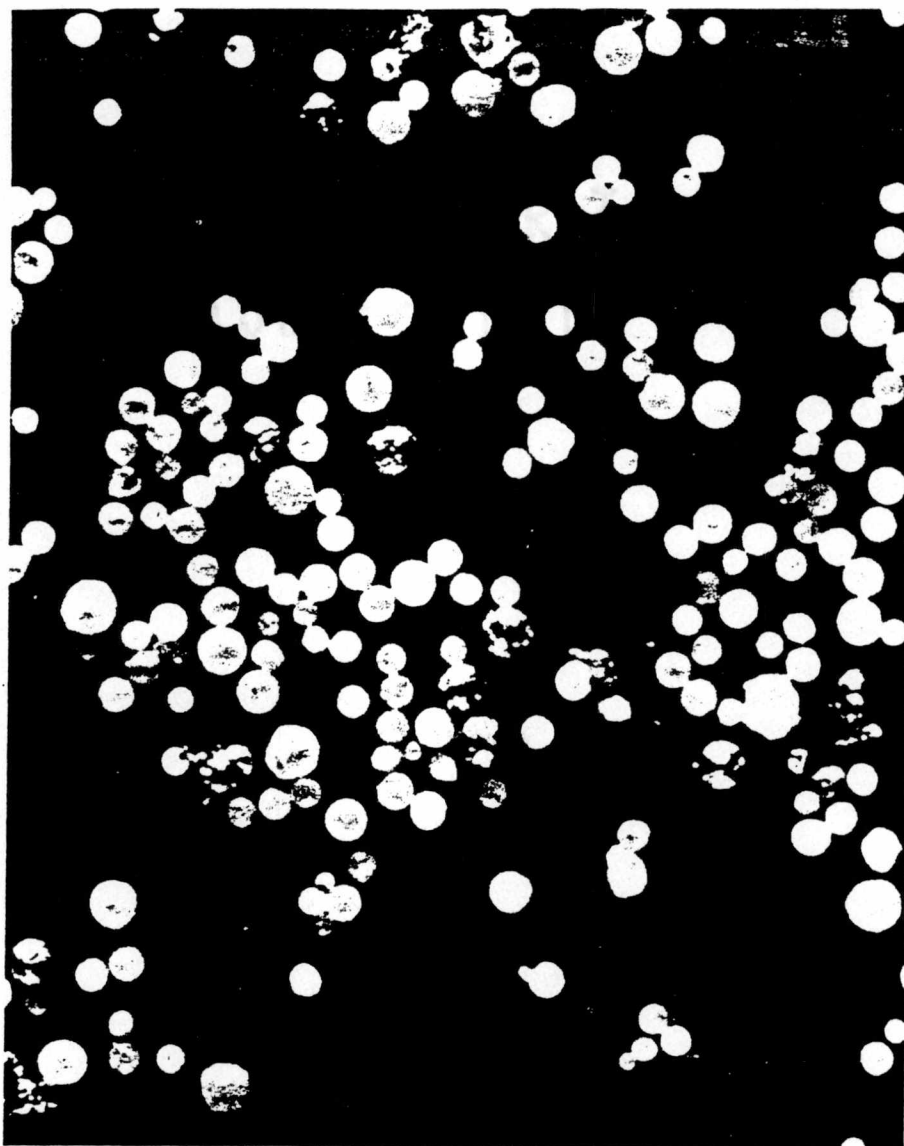


Figure 2.5 Photograph of combustion residue from 70 μm diameter aluminum particles consisting of large hollow oxide spheres, large unburned aluminum particles, and small solid oxide spheres from Drew, Gordon, and Knipe [13].



Figure 2.6 Photograph of 150 μm diameter aluminum particles quenched during self-sustained combustion showing the formation of a hollow oxide sphere (lower half) on the surface of the oxide cap from Drew, Gordon, and Knipe [13].



Figure 2.7 Photograph of 150 μm diameter aluminum particles quenched during self-sustained combustion showing a fully developed hollow oxide sphere (upper right) just separated from its parent droplet from Drew, Gordon, and Knipe [13].

observations seem to confirm conclusions drawn by Friedman and Maček [10] that the hollow spheres were the result of ballooning of the oxide layer. They also showed that the hollow spheres were not present in the residue from particles burned in dry environments. This lead them to believe that hydrogen could be responsible for ballooning of the oxide layer.

Drew, Gordon, and Knipe concluded that their observations tend to support the model proposed by Brzustowski and Glassman and to discount the model proposed by Bartlett et al. However, the burning geometry of the aluminum particles is not as simple as for hydrocarbon droplets due to the presence of the condensed oxide on the droplet surface.

Prentice [15] studied the combustion of aluminum particles ignited by a xenon flash discharge tube and a Nd-glass pulse laser. Unlike previous studies, use of these techniques allowed the particles to be burned in room temperature environments. Experiments were conducted in environments containing various concentrations of oxygen, argon, and nitrogen. The aluminum particles were formed from squares of Reynolds household aluminum foil which were precisely cut so as to form droplets of known diameter when melted. The xenon flash discharge technique involved dropping the particles through a glass pipe surrounded by a helical quartz xenon flash discharge tube. When the particle reached the area enclosed by the xenon pulse lamp, the lamp was discharged igniting the particle. The combustion process was studied by quenching and long-exposure still photographs. For the pulse-laser experiments, particles were placed inside a rectangular stainless steel combustion chamber and

irradiated by a Nd-glass laser pulse. The combustion event was recorded using high speed cinematography.

Prentice found that convection of condensed oxide to the surface of the burning droplet seemed to be the deciding factor as to which mode of combustion the particles undergo. When condensed oxide formed at the flame front was convected to and deposited upon the droplet surface, spinning, jetting, and fragmenting were likely to occur whereas these phenomena did not occur when no accumulation of condensed oxide on the droplet surface was observed.

Quenched particles burned in oxygen/argon environments were shown to have no oxide accumulated upon the droplet surface. As a result, they did not fragment, spin, or jet oxide and termination of the combustion process consisted of the droplet steadily burning to completion. In these environments, Prentice proposed that Brzustowski and Glassman's model would closely approximate the combustion process. Particles ignited using both techniques in oxygen/argon (20/80) environments exhibited burning times which were proportional to the initial diameter of the particle.

Prentice discovered that the presence of nitrogen resulted in the accumulation of the condensed oxide on the droplet surface leading to spinning, jetting, and fragmenting. The oxide was found to collect and form a cap on the droplet surface covering as much as 60% of the droplet surface. Previous studies described the cap as being lens-shaped and totally impervious to the diffusion of aluminum vapor. However, using a scanning electron microscope, Prentice discovered the cap to be

more indicative of a fumarole. Indications of smoke patterns around quenched particles seemed to indicate that aluminum vapor was diffusing through the fumarolic-shaped oxide cap.

In experiments conducted in dry air environments, the time until the burning droplet fragmented was found to be proportional to the ambient pressure. Prentice also found that, at ambient pressure, the time until fragmentation occurred was proportional to the particle diameter. In addition, the droplet fragmentation observed at the cold ambient temperatures of this study was less violent than that observed in hot ambient conditions. Prentice found that adding as little as 5% nitrogen to an oxygen/argon (20/80) environment resulted in spinning and fragmenting. It was thus concluded that the presence of nitrogen and, hence, a condensed oxide cap on the droplet surface were the key factors promoting the violent burning characteristics commonly associated with aluminum combustion.

Wilson and Williams [16] utilized the results of Prentice in an attempt to verify the Brzustowski-Glassman combustion model. Prentice had found that aluminum particles burned in oxygen/argon environments did not accumulate oxide on the droplet surface and burned symmetrically and steadily to completion. Prentice theorized that, under these conditions, the combustion process would closely follow Brzustowski and Glassman's model. Wilson and Williams attempted to verify this by burning aluminum particles ranging from 24 to 90 μm in diameter in oxygen/argon environments. The particles were suspended on glass fibers by electro-static and van der Waals attraction and ignited by a laser pulse. Upon ignition the particles

separated from the glass fibers and, therefore, the combustion process was unaffected by their presence. The combustion event was recorded using high-speed cinephotomicrography.

Wilson and Williams had hoped to verify that aluminum particle combustion followed the " d^2 -law." However, they only confirmed results from previous studies which had shown that the burning time is proportional to d^N with experimentally determined values of N widely scattered between 1 and 2. Wilson and Williams did confirm that the burning droplets were surrounded by a detached flame indicative of vapor-phase combustion. The stand-off ratio of the flame was constant throughout the combustion process but was found to increase with increasing oxygen concentration. An increase in oxygen concentration was also found to result in an increase in burning rate. No ignition occurred at oxygen concentrations below 4%, and at oxygen concentrations above 20%, oxide caps on the droplet surface and irregular burning patterns were observed despite the absence of nitrogen. In addition, Wilson and Williams observed an energy flux threshold below which ignition was delayed and oxide droplets accumulated in the flame region. This energy flux threshold increased with increasing pressure.

In later work, Prentice [17] studied the effects of water vapor on the combustion process of aluminum particles in oxygen/nitrogen, oxygen/argon, and oxygen/carbon dioxide environments. Particles were formed from discs of aluminum foil precisely cut to form droplets with diameters ranging from 250 to 400 μm . The particles were ignited by a laser pulse in wet and dry environments.

Prentice found that burning rate was greater in wet environments for all gas mixtures investigated and that combustion occurred fastest in wet oxygen/carbon dioxide environments. Previous studies had shown that condensed oxide did not accumulate on the droplet surface in dry oxygen/argon environments. However, Prentice found that this was not the case in the presence of water vapor. In fact, combustion in all gas mixtures led to oxide accumulation and fragmentation when water vapor was present. In addition, the oxide accumulated on the droplet surface in wet environments was found to differ from that accumulated in dry environments. In wet environments the oxide appeared more transparent and wet the droplet allowing the surface to be completely covered with an oxide shell.

Prentice also wrote a critique of previous attempts at modelling the combustion process of aluminum particles and called for a more accurate model. Prentice felt that previous models were practically useless due to inappropriate attempts to extrapolate models based on the combustion of hydrocarbon droplets. These models neglected a great amount of experimental evidence which contradicts the principal assumptions. Such models predict that burning time is proportional to the square of the initial particle diameter. Prentice dismissed this " d^2 -law" because of the assumptions it entailed. For example, a steady state combustion process is assumed despite the fact that aluminum combustion is characterized by spinning, jetting, and fragmenting. Wilson and Williams' carefully controlled experiments in dry oxygen/argon environments produced a more steady combustion process, but as Prentice points out, these results are interesting but unproductive as the conditions

tested have no practical application. In addition, the models are based on the single step irreversible reaction $\text{Al}_{(\text{g})} + \text{O}_{2(\text{g})} \rightarrow \text{Al}_2\text{O}_{3(\text{g})}$ which is not thermodynamically feasible. The models also assume spherical symmetry whereas it is generally agreed that combustion of aluminum occurs asymmetrically. While it has been shown that some of the condensed oxide formed at the flame front is convected inward and deposited upon the droplet surface, the models only account for the loss of mass from the droplet surface. The asymmetry has been shown to be due to the presence of the oxide cap on the droplet surface and reactions occurring on the cap which contradicts the assumption that no surface reactions occur. In addition, the presence of nitrogen, carbon dioxide, and water vapor have been shown to cause surface reactions which lead to fragmentation. Finally, while most models neglect radiation heat transfer, its effects have proven to be significant.

In a further attempt to disprove the " d^2 -law," Prentice took data from his current study and attempted to determine the relation between burning time and diameter. Assuming burning time was proportional to d^N , Prentice found the value of N to vary from 1.3 to 2.5 and to be dependent upon oxygen concentration and oxidizer type. The value was found to be insensitive to the accumulation of oxide on the droplet surface or even fragmentation of the particle.

Law [18] developed an analytical model for the combustion of metal particles shown schematically in Figure 2.8 which incorporates the accumulation of condensed oxide on the droplet surface. The model is capable of representing four different modes of combustion which have been observed experimentally. Model A allows for

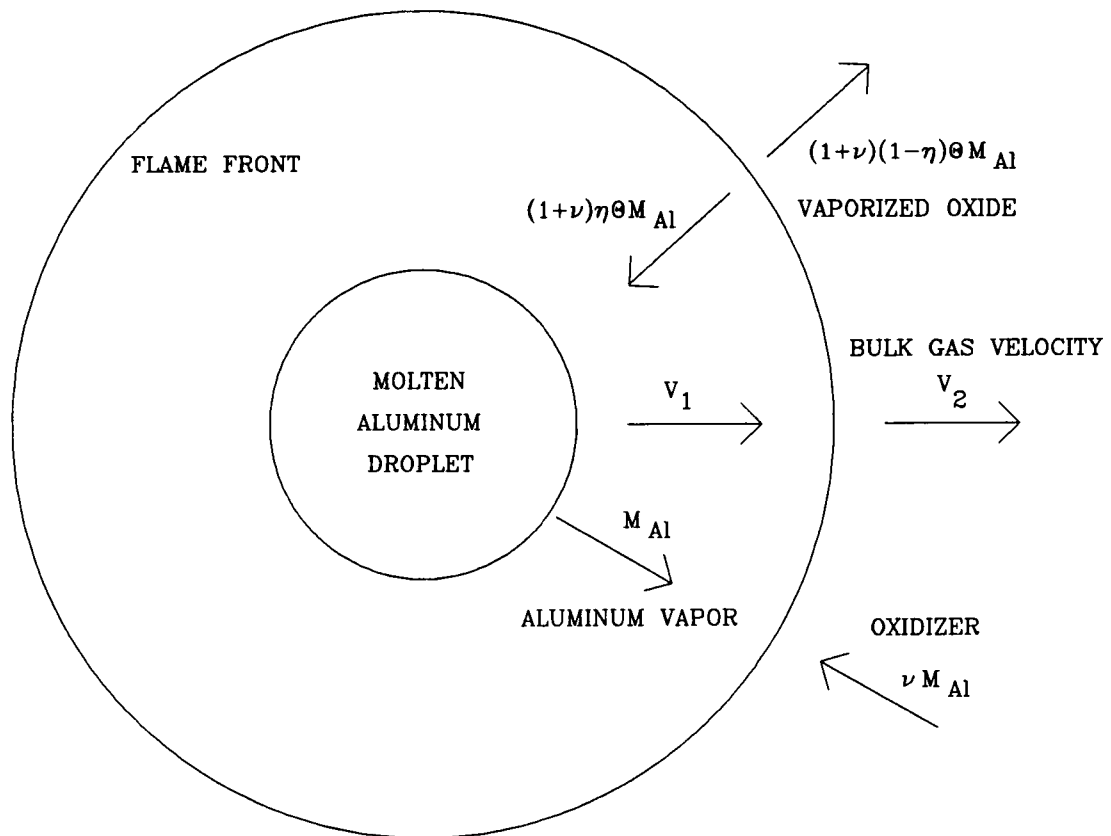


Figure 2.8 Schematic of the model proposed by Law [18] for the combustion of aluminum particles incorporating the accumulation of condensed oxide on the droplet surface.

the accumulation of oxide on the droplet surface while Model B prohibits oxide accumulation. In these two models, the flame temperature is held constant at the boiling point of the oxide. Model C represents combustion in dilute atmospheres with all oxide formed at the flame front in the condensed phase and the flame temperature falling below the oxide boiling point. Model D represents combustion in enriched atmospheres with all oxide being vaporized and the flame temperature increasing above the oxide boiling point.

In most respects Law's model resembles the standard hydrocarbon droplet model. For example, standard assumptions include steady state combustion, spherical symmetry, constant pressure, and negligible heat transfer by radiation. The temperature of the burning droplet is assumed to be at the boiling point of aluminum. Evaporation of the fuel and diffusion through condensed oxide on the surface is assumed to occur very rapidly. The fuel and oxidizer are assumed to react instantaneously and completely at the flame front in stoichiometric proportions.

In order to maintain the flame temperature, a fraction, θ , of the oxide formed at the flame front is vaporized. A fraction, η , of the vaporized oxide diffuses inward and condenses on the droplet surface. The remainder of the oxide formed is in the condensed phase and must be convected away by the bulk gas motion. Law proposed four possible modes for the transport of the condensed oxide based on the bulk gas velocities V_1 and V_2 in the region between the flame and droplet and in the region outside the flame, respectively. In the first mode, $V_1 > 0$, $V_2 > 0$, and the condensed oxide is convected away towards infinity. For the second mode with

$V_1 > 0$ and $V_2 < 0$, the oxide is trapped at the flame front. In the third mode, $V_1 < 0$ and $V_2 < 0$ resulting in the oxide being swept inward and depositing on the droplet surface. The fourth mode in which $V_1 < 0$ and $V_2 > 0$ was shown to be irrelevant since combustion could not be supported with the bulk gas motion in both regions moving away from the flame front.

The accumulation of oxide on the droplet surface is expected to slow the diffusion of aluminum vapor. Uneven distributions of the oxide leads to spinning and jetting which violates the steady state assumption. Significant accumulation of oxide can so limit the diffusion of aluminum vapor that vapor-phase combustion is replaced by surface burning. Even greater accumulations of oxide can result in extinction of the combustion process. All of these cases violate key assumptions in Law's model leading to deviations from the predicted "d²-law." For these conditions, the burning time has often been shown to be proportional to d^N with N ranging from 1 to 2. Law's model was found to accurately describe pure vapor-phase combustion. The accuracy of the solution was severely compromised when oxide accumulation reached the levels described above. However, the model was capable of determining the conditions for which oxide accumulation becomes a serious problem.

Results from Law's model are shown in Figures 2.9, 2.10 and 2.11. Figure 2.9 shows the effects of oxygen mole fraction, Y_{O_∞} , and ambient temperature, T_∞ , on the nondimensional mass burning rate, M_F , and flame stand-off ratio, $\hat{r}_f$, for Models A, C, and D. As can be seen, the mass burning rate increases almost linearly with increasing oxygen mole fraction. In addition, the ambient temperature is shown

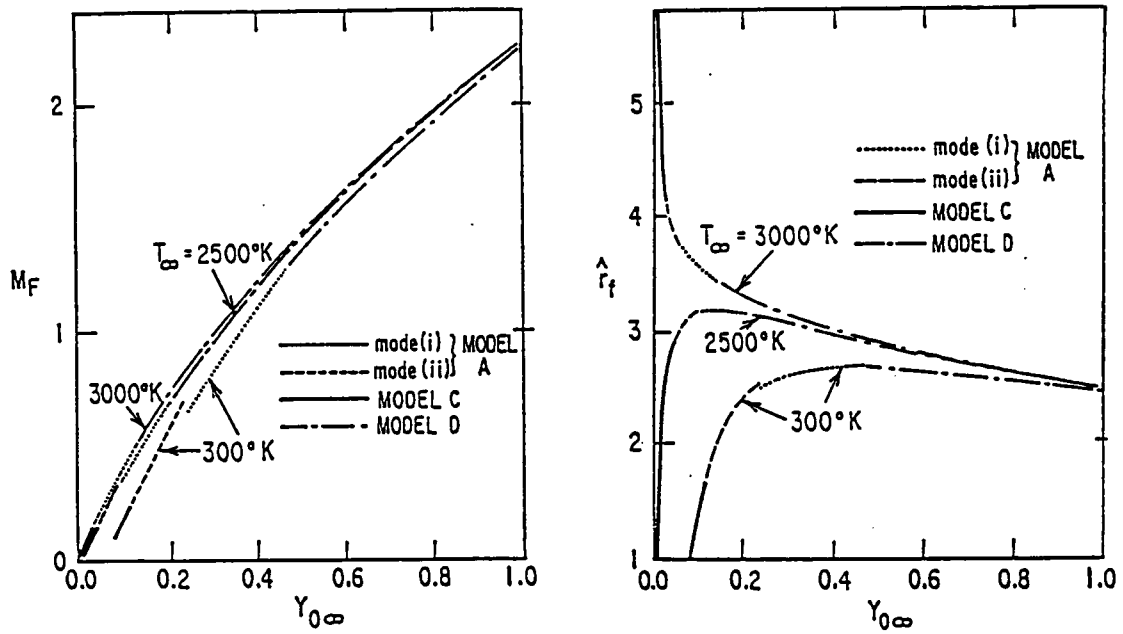


Figure 2.9 Effects of oxygen mole concentration, $Y_{O_{\infty}}$, on the nondimensionalized mass burning rate, M_F , and flame stand-off ratio, $\hat{r}_f$, for aluminum particles at various ambient temperatures, T_{∞} , based on the analytical model allowing surface accumulation of oxide proposed by Law [18].

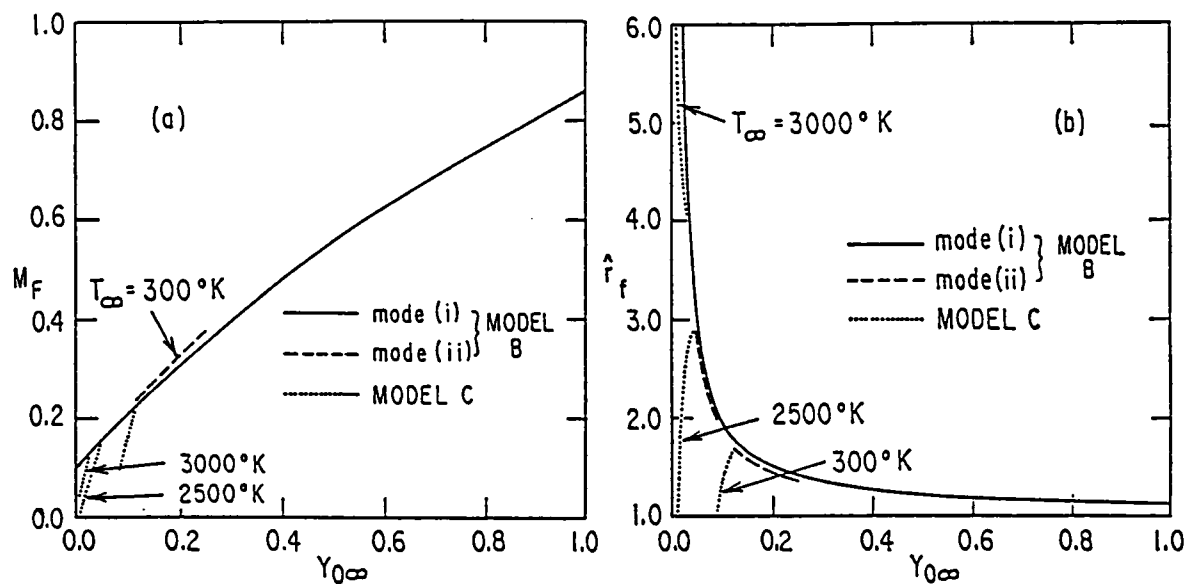


Figure 2.10 Effects of oxygen mole concentration, $Y_{O_{\infty}}$, on the nondimensionalized mass burning rate, M_F , and flame stand-off ratio, $\hat{r}_f$, for aluminum particles at various ambient temperatures, T_{∞} , based on the analytical model preventing surface accumulation of oxide proposed by Law [18].

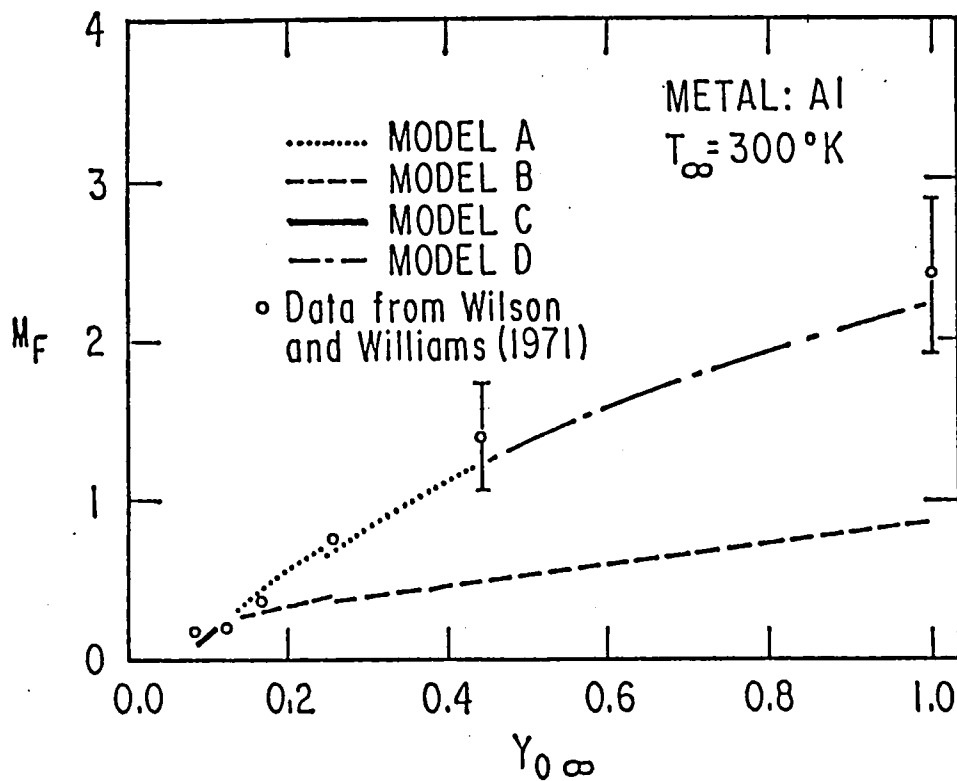


Figure 2.11 Comparison of the experimentally observed effects of oxygen mole concentration, $Y_{O_{\infty}}$, on the nondimensionalized mass burning rate, M_F , of aluminum particles with that predicted by the analytical model proposed by Law [18].

to have little effect on the mass burning rate. Figure 2.9 also shows that for ambient temperatures below the surface temperature of the droplet, the flame stand-off ratio increases dramatically with increased oxygen mole fraction when the oxide is trapped at the flame front but tends to decrease slightly as Y_{O_∞} is further increased. When the ambient temperature is higher than the surface temperature, the stand-off ratio was shown to decrease rapidly from its initial value at infinity and then decrease more slowly as oxygen mole fraction is increased. At high oxygen mole fractions the stand-off ratio is only slightly affected by ambient temperature.

Figure 2.10 shows the effects of oxygen mole fraction and ambient temperature on the mass burning rate and flame stand-off ratio for Models B and C where oxide accumulation is inhibited. A comparison of Figure 2.10 with Figure 2.9 shows that the lack of oxide accumulation greatly reduces the mass burning rate. For example, the mass burning rate is reduced by a factor of 2.5 at an oxygen mole fraction of 0.5. The stand-off ratio is shown to be close to one at all but the lowest values of oxygen mole fraction and ambient temperature.

Figure 2.11 shows a comparison of experimental results obtained by Wilson and Williams with results from Law's model. Results from Models A, C, and D were found to agree well with Wilson and Williams' data. However, Model B results were much lower. This result is somewhat surprising in that Wilson and Williams' study was performed in an oxygen/argon environment which prevented oxide accumulation. Thus it would seem that results would correspond better to Model B than to Models A, C, and D which allow oxide accumulation.

2.2 COMBUSTION OF ALUMINUM AGGLOMERATES

Wong and Turns [19] studied the ignition process of aluminum-based slurry droplets with JP-10 as the liquid carrier. Droplets ranging from 500 to 1100 μm in diameter were supported by 100 μm SiC fibers and ignited by exposure to the hot product gases from a flat flame burner. Observations were made using high-speed cinematography. Experiments were performed in both wet and dry environments with oxygen concentration varied from 10% to 25%. The ambient temperature ranged from 1250 to 1800 K. All of the experiments were performed at atmospheric pressure.

Wong and Turns developed a model to describe the ignition process of the slurry droplets. The ignition process was found to involve distinct stages as shown schematically in Figure 2.12. In the first stage, the JP-10 liquid carrier is consumed. The solid particles then coalesce into porous agglomerates which are heated to the melting point of aluminum in the second stage. The protective oxide layer maintains the integrity of each particle during the melting process. However, the agglomerate continues to heat up and eventually reaches the melting point of the oxide. The individual particles of the agglomerate then coalesce into a single molten droplet which is immediately ignited. Combustion of the droplet occurs in the vapor-phase. Wong and Turns defined the ignition time as the time elapsed between the introduction of the particle to the hot product gases and the beginning of vapor-phase combustion of the molten aluminum droplet.

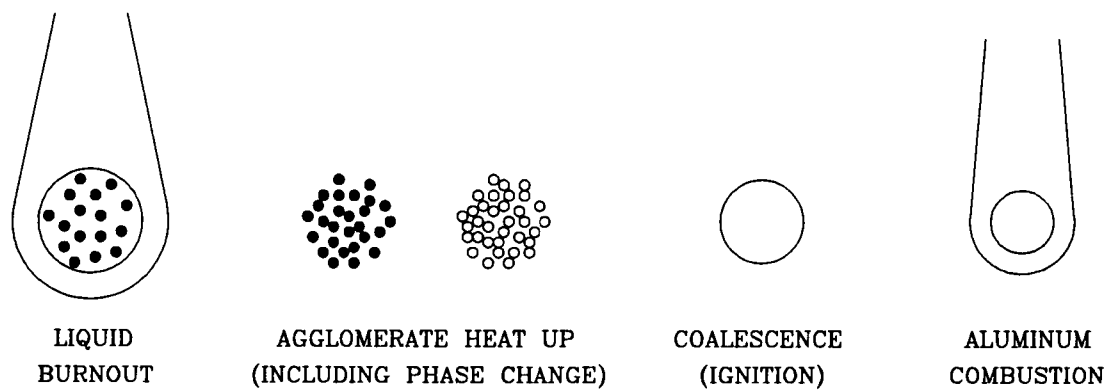


Figure 2.12 Ignition and combustion model for an aluminum-based slurry droplet proposed by Wong and Turns [19].

The analytical model used to predict ignition time involved the modelling of the liquid burnout and the agglomerate heat-up. For the analytical model the ignition time was taken as the time until the agglomerate reached the melting point of the oxide. Wong and Turns assumed that the droplets and agglomerates were spherical with a uniform temperature distribution. The heating of the agglomerate was assumed to depend on convection from the surroundings, generation by surface reactions, and radiation to the surroundings.

Theoretical and experimental results from Wong and Turns' study are shown in Figures 2.13 through 2.15. Figure 2.13 shows the effects of initial droplet diameter on the ignition time. The ignition time was found to be proportional to the diameter raised to the power of 1.28 ± 0.18 . As can be seen, results from the analytical model agree quite well with the experimental results. The burnout of the liquid carrier was shown to occupy the majority of the ignition time, especially at high temperatures. Figure 2.13 also shows that pure droplets of JP-10 of the same size as the slurry droplets burn in approximately the same amount of time required for the agglomerates to ignite. Figure 2.14 shows the effects of ambient gas temperature on the ignition time. As would be expected, increasing the ambient gas temperature results in a higher heat transfer rate to the agglomerate and hence a significantly shorter ignition time. Again, the analytical model is seen to predict the observed behavior rather well. The cusp in the curves below 1500 K is a result of an increase in flow velocity due to the method used to control the gas temperature. Wong and Turns proposed that below the ignition limit, heat loss due to radiation and convection

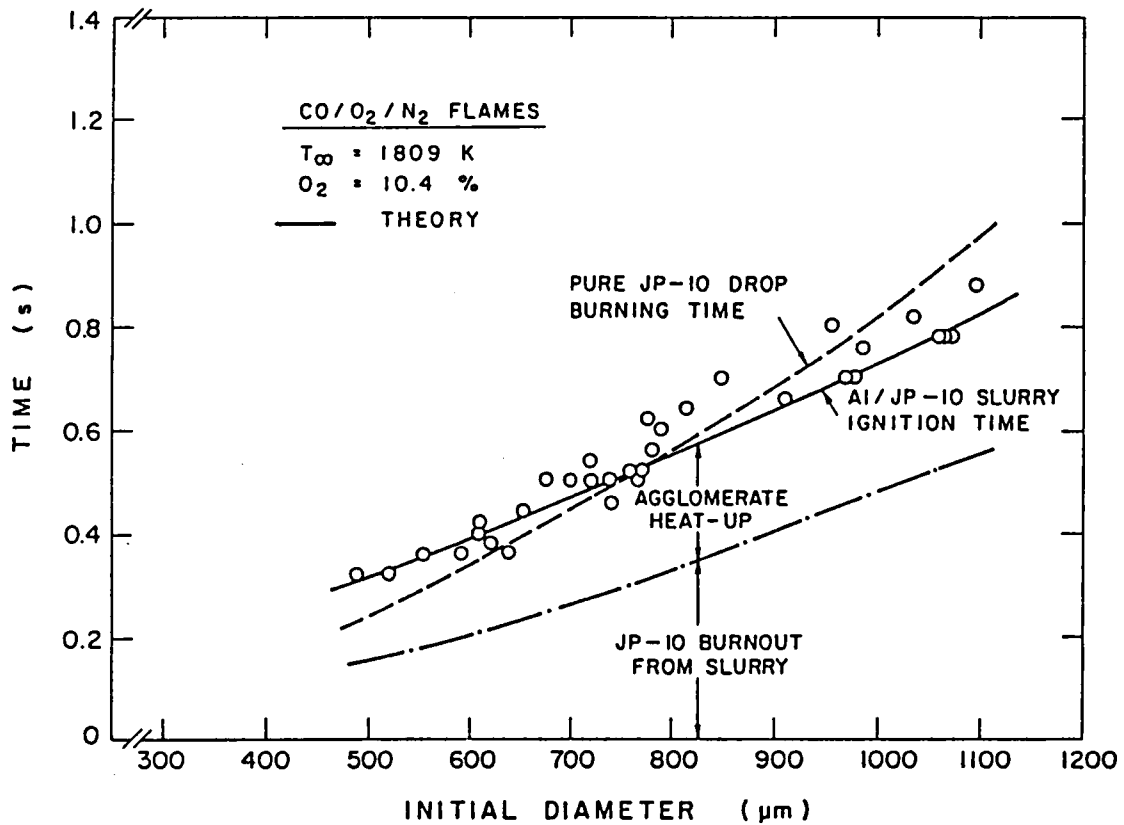


Figure 2.13 Effects of initial droplet diameter on the ignition time of aluminum/JP-10 slurry droplets as observed by Wong and Turns [19].

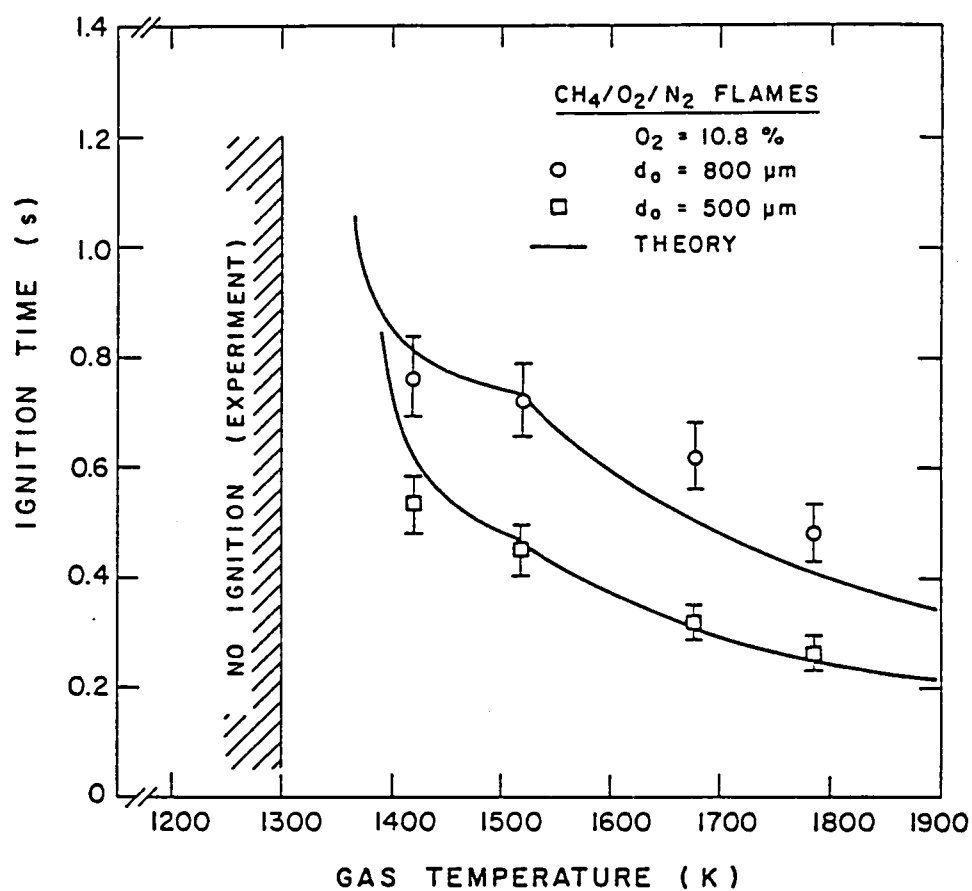


Figure 2.14 Effects of ambient gas temperature on the ignition time of aluminum/JP-10 slurry droplets as observed by Wong and Turns [19].

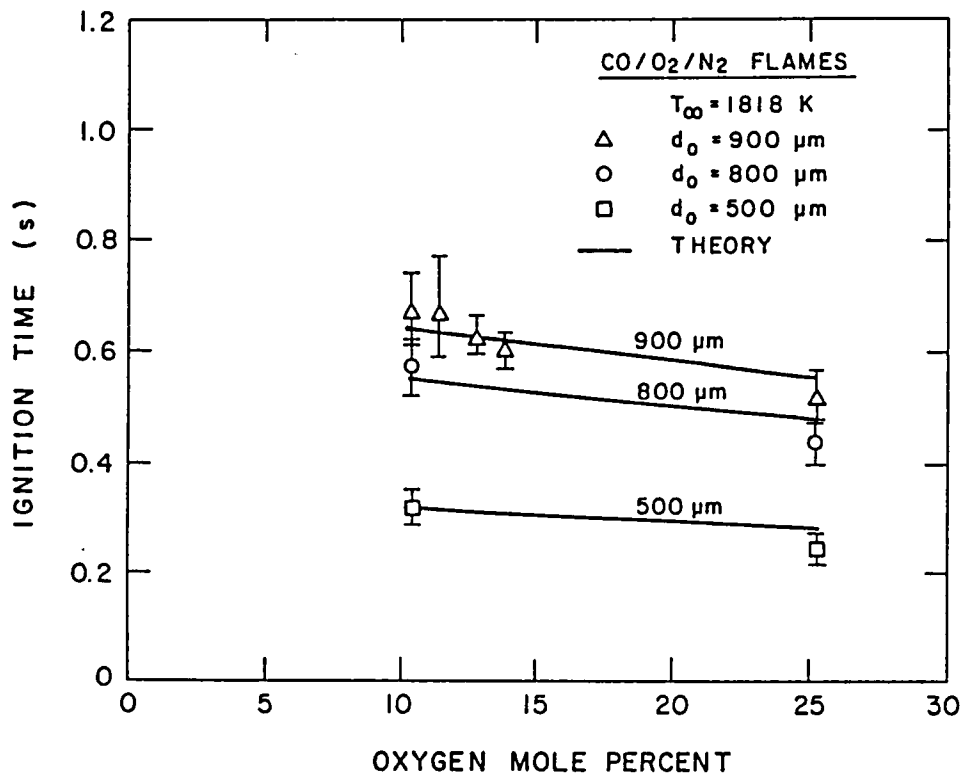


Figure 2.15 Effects of ambient oxygen mole fraction on the ignition time of aluminum/JP-10 slurry droplets as observed by Wong and Turns [19].

cannot be compensated for by heat release from the chemical reaction. Figures 2.15 shows the effects of oxygen mole fraction on the ignition time. Experimental observations showed that an increase in the oxygen mole fraction results in an approximately equal shortening of both the liquid burnout and the agglomerate heat-up. However, as shown in Figure 2.15, a large increase in oxygen mole fraction results in only a slight decrease in ignition time. From these results, Wong and Turns concluded that while chemical reactions have some effect upon the ignition time, thermal effects are the dominating factor.

In a second paper, Turns, Wong, and Ryba [5] investigated the combustion process of the aluminum agglomerates formed during the burning of aluminum/JP-10 slurry droplets. Droplets ranging in size from 500 to 1100 μm were supported on 125 μm SiC fibers and ignited by exposure to the hot product gases of a flat flame burner. The agglomerates formed in the combustion process ranged in size from 300 to 800 μm in diameter. Experiments were performed in environments with oxygen concentrations ranging from 10% to 25% and ambient gas temperatures ranging from 1500 to 1800 K. High-speed cinematography was used to record the combustion process.

As mentioned previously, and shown in Figure 2.12, the combustion of the slurry droplets began with the burnout of the liquid carrier. The solid fuel particles then coalesced into porous agglomerates. The agglomerates were then heated to the melting point of the oxide at which point the oxide layer surrounding each particle could no longer maintain the integrity of individual particles. The particles then

coalesced to form a molten droplet of aluminum as indicated in cinematographic records by a sharp decrease in diameter. As soon as coalescence was complete, ignition was found to occur characterized by the appearance of a detached vapor-phase diffusion flame surrounding the droplet. It was observed that the combustion process was generally characterized by a nearly spherical flame structure with a plume of oxide smoke convected away in the particle's wake. An oxide cap was found to accumulate on the leeward side of the molten droplet inhibiting combustion in that area. Shifting of the oxide cap's position resulted in an asymmetric smoke trail. Occasionally, small oxide particles were seen to be jetting from the surface of the burning droplet. Near the end of the combustion event, the flame was observed to collapse to the droplet surface. The droplet continued to react at the surface for a short period of time with no trace of oxide smoke. The period of surface burning was followed by flame extinction and gradual cooling of the particle. Turns, Wong, and Ryba chose to define the burning time as the time elapsed between ignition and the disappearance of the oxide smoke.

An analytical model shown schematically in Figure 2.16 was developed by using experimental observations to expand the vapor-phase combustion model developed by Law. In addition to Law's general assumptions, Turns, Wong, and Ryba assumed that the flame temperature and surface temperature of the droplet were maintained at the boiling points of the oxide and aluminum, respectively. The model accounted for the increase in burning rate and the asymmetrical flame geometry due to forced convection from the hot product gases. The accumulated oxide was

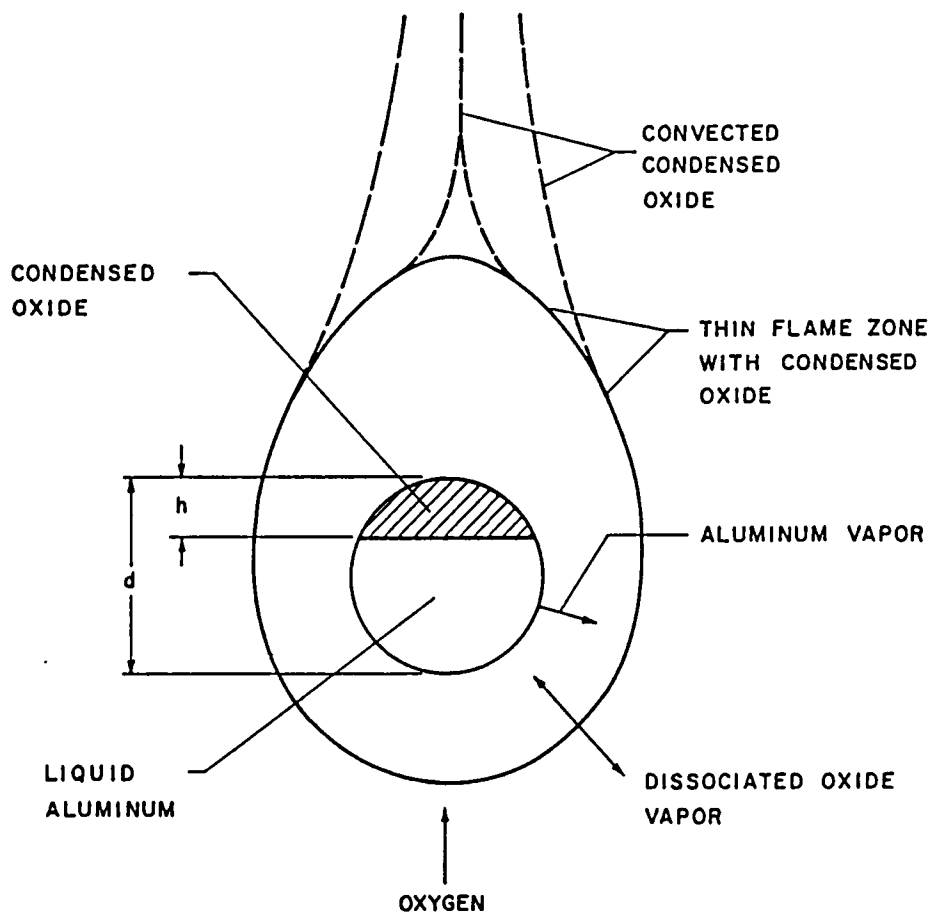


Figure 2.16 Combustion model for aluminum agglomerates accounting for the accumulation of oxide on the surface as proposed by Turns, Wong, and Ryba [5].

assumed to form a spherical segment which continued to grow throughout the combustion process. It was assumed that combustion did not occur over the solid angle occupied by the oxide cap. Finally, the effects of radiation heat transfer were neglected because they were found to affect the burning rate only slightly. In addition, the theoretical radiative losses often became so large as to prevent the dissociation of oxide at the flame front and, hence, the accumulation of oxide on the droplet surface which was observed experimentally.

Typical results from the analytical model showing the life history of the burning aluminum agglomerates are shown in Figure 2.17. In Figure 2.17, zero time represents the ignition of the aluminum agglomerate and d_m is the diameter of the molten droplet immediately after coalescence. As combustion progresses, a fraction of the oxide produced at the flame front accumulates on the droplet surface increasing the size of the oxide cap. When the combustion process is complete, the mass of the accumulated oxide is over 40% of the mass of the original agglomerate, and, assuming a nonporous product, the final particle size is about 65% of the original droplet size. Experimental results showed that the final particles were somewhat porous and ranged in size from 60% to 80% of the original droplet size.

Theoretical and experimental results from Turns, Wong, and Ryba's study are shown in Figures 2.18 through 2.21. The effects of diameter on the burning time of the aluminum agglomerates are shown in Figure 2.18. As can be seen, the experimental values essentially fall within the region bracketed by the two theoretical curves. The upper curve ($F=0$) is based on the assumption that the oxide cap

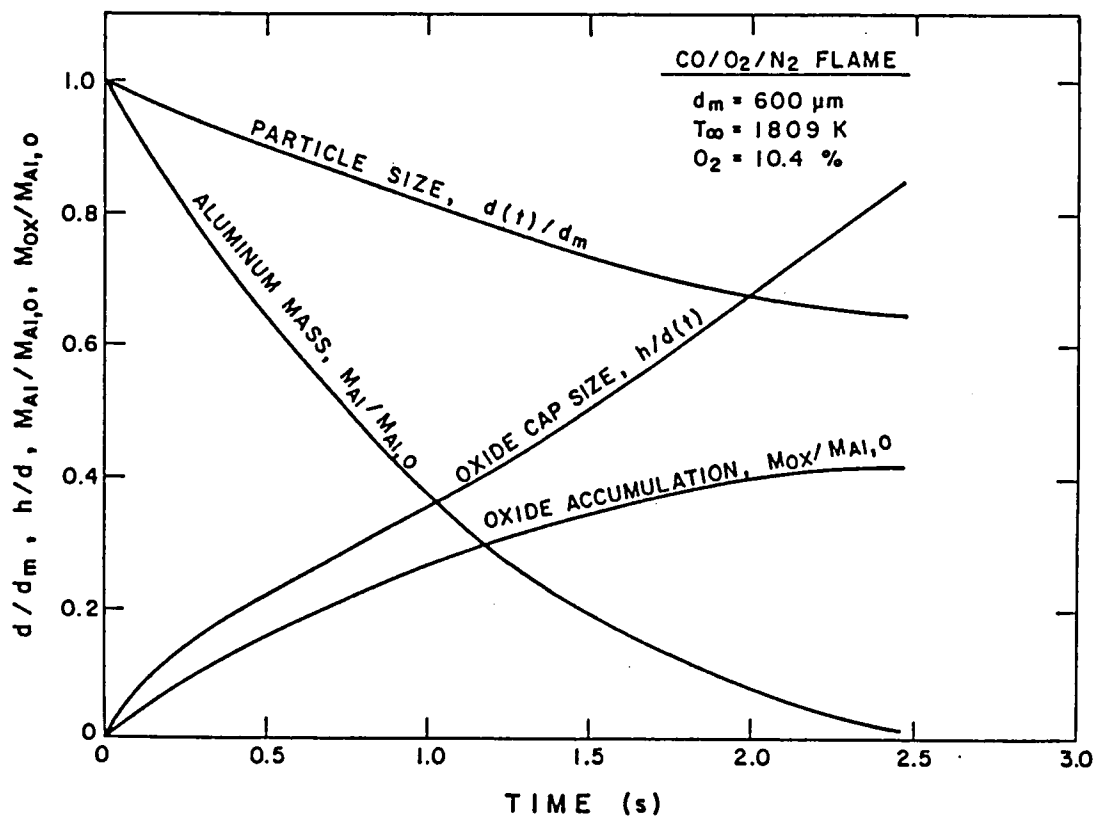


Figure 2.17 Life history of an aluminum agglomerate after ignition as predicted by the analytical model proposed by Turns, Wong, and Ryba [5].

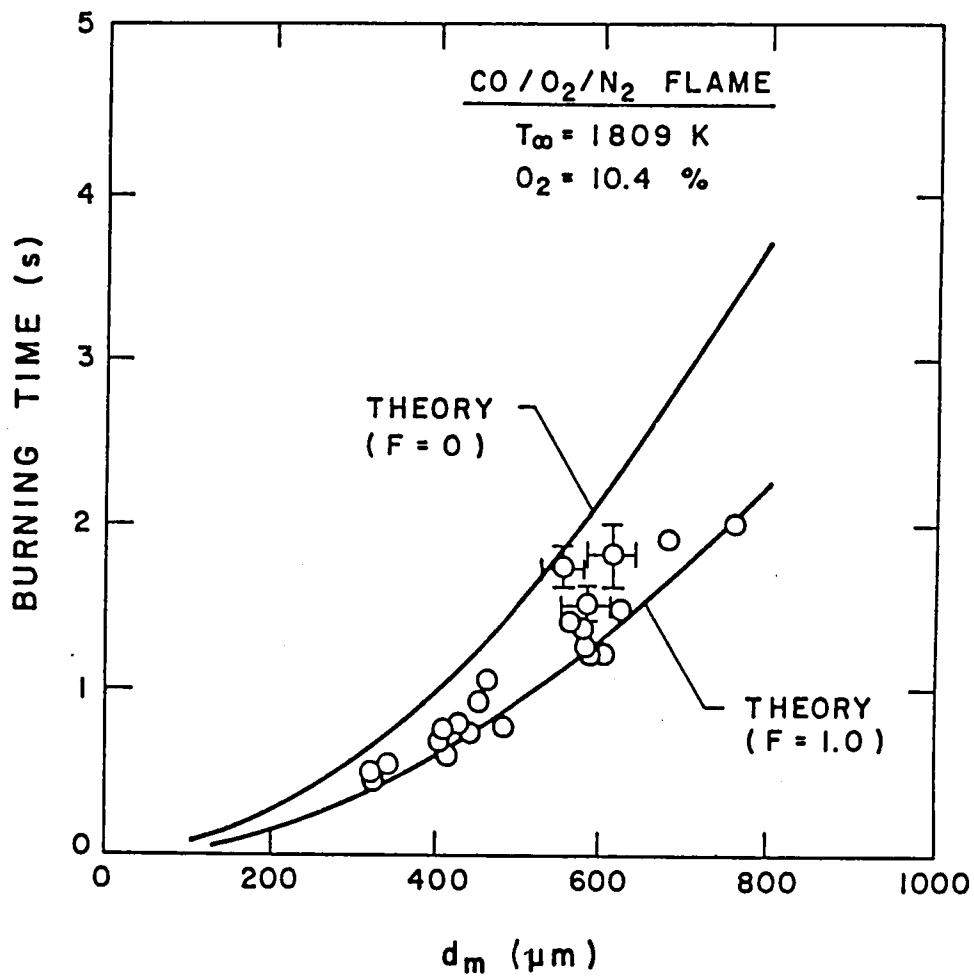


Figure 2.18 Effects of the initial molten droplet diameter on the burning time of aluminum agglomerates as observed by Turns, Wong, and Ryba [5].

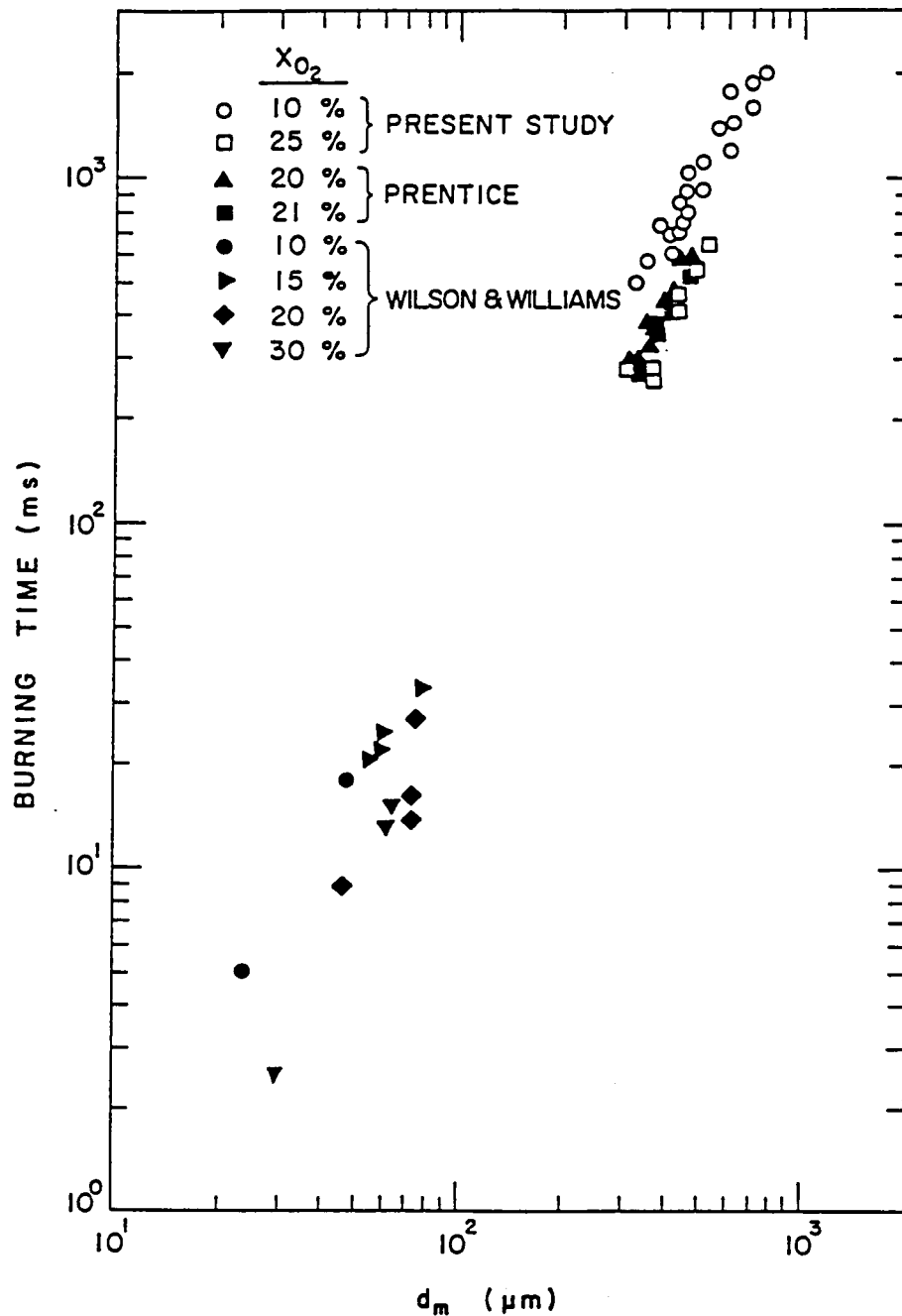


Figure 2.19 Comparison of the burning time of aluminum agglomerates observed by Turns, Wong, and Ryba [5] to the burning time of single aluminum particles from previous studies.

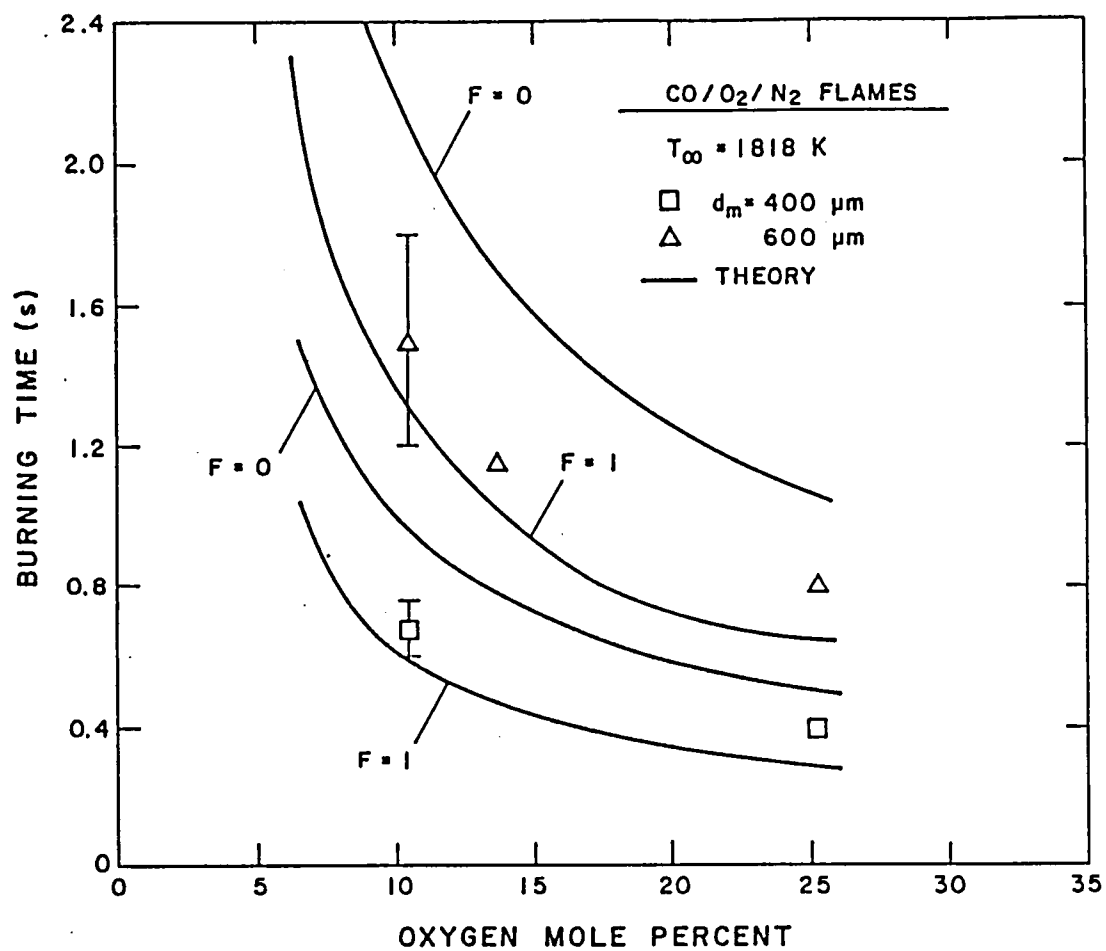


Figure 2.20 Effects of ambient oxygen mole fraction on the burning time of aluminum agglomerates as observed by Turns, Wong, and Ryba [5].

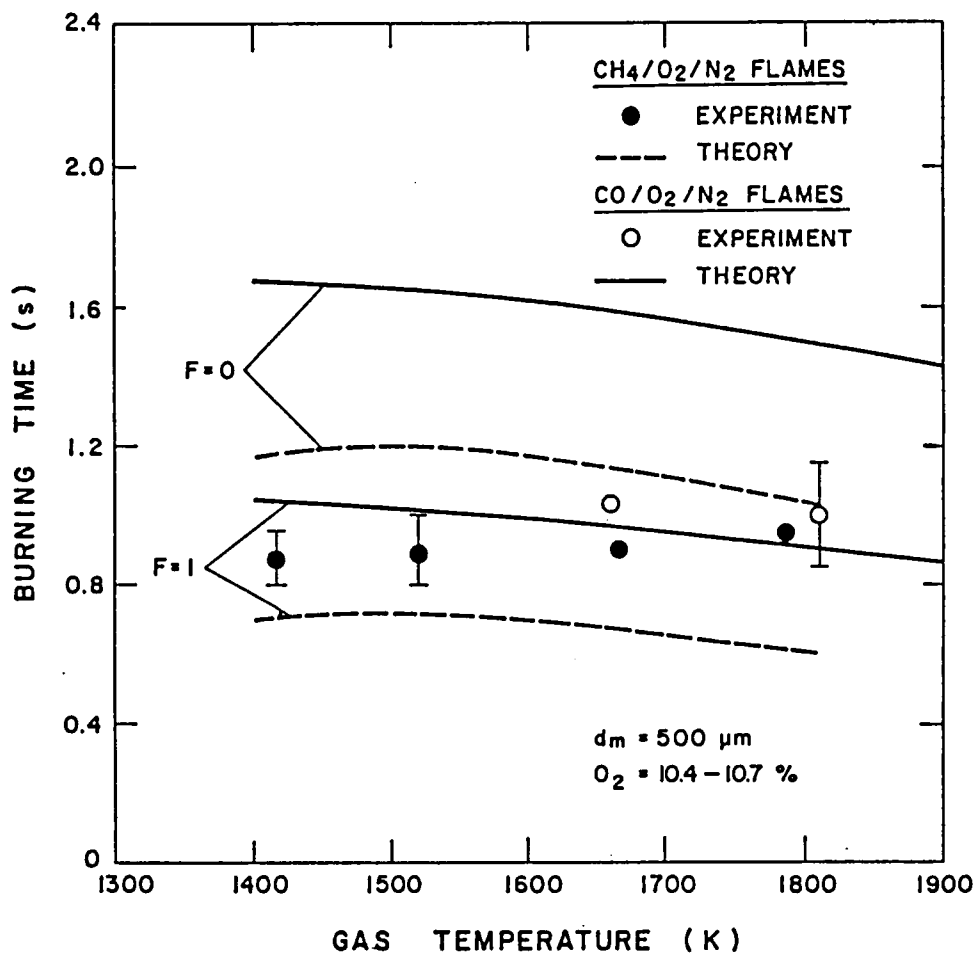


Figure 2.21 Effects of ambient gas temperature on the burning time of aluminum agglomerates as observed by Turns, Wong, and Ryba [5].

completely inhibits the diffusion of aluminum vapor. The lower curve ($F=1$) assumes that cracks or fumaroles are present to allow diffusion of aluminum vapor through the oxide cap as observed by Prentice [15]. The experimental results seem to agree more with the lower curve. The burning time was found to be proportional to the diameter raised to the power of 1.77 ± 0.20 . Figure 2.19 shows that experimental values from this study seem to correspond well with those from the studies conducted by Prentice [15] and Wilson and Williams [16] using single aluminum particles. It is reasonable that the burning time for the agglomerate would correspond to that for a single particle since the agglomerate coalesces into a single molten droplet prior to ignition.

The effects of oxygen mole fraction and ambient gas temperature on the burning time of the aluminum agglomerates are shown in Figures 2.20 and 2.21, respectively. As shown in Figure 2.20, the experimental results are again bracketed by the theoretical curves and again agree more with the fumarolic cap theory. Increasing the oxygen mole fraction is shown to result in a decrease the burning time. This effect is more pronounced at lower oxygen mole fractions. As shown in Figure 2.21, the ambient gas temperature was found to have little effect upon the burning time of the agglomerates. However, the ambient temperature was found to affect the degree of completion of the combustion process. At high flame temperatures (above 1650 K) all aluminum was found to be consumed. However, at low flame temperatures (below 1550 K) unburned aluminum was found in the combustion products.

Meftah [6] studied the effects of ambient pressure on the ignition and combustion processes of aluminum agglomerates. Whereas previous studies were performed using flat flame burners at atmospheric pressure, Meftah's study used a CO₂ laser-pulse to ignite aluminum agglomerates supported inside a high-pressure combustion chamber. By this method he was able to create pressure environments similar to those found in typical combustors.

Meftah's study involved the combustion of aluminum agglomerates of initial average diameter of 535 μm in a oxygen/nitrogen (20/80) environment at pressures ranging from 0.1 to 4.3 MPa. The experimental method is identical to the method used in the present study and described in the following chapter.

Meftah found no particular trend to the effects of ambient pressure on the ignition delay time. However, at low pressures (below 2.2 MPa) the burning time and flame stand-off ratio were both found to decrease dramatically with increasing pressure. At high pressures, further increases in pressure were found to have little effect on either the burning time or stand-off ratio. Furthermore, at high pressures the flame was nearly congruous to the surface of the burning droplet.

CHAPTER 3

EXPERIMENTAL APPARATUS AND PROCEDURE

This chapter will present a brief description of the apparatus and procedure used in this experimental study. Section 3.1 gives an overview of the experimental method and procedure. Section 3.2 describes the test conditions for the experiments. The method used to produce the aluminum agglomerates is detailed in Section 3.3. Section 3.4 describes the equipment used in this study. The methods of data acquisition and analysis are outlined in Section 3.5.

3.1 OVERVIEW OF EXPERIMENTAL METHOD AND PROCEDURE

In this study, aluminum agglomerates with initial average diameters of 835 and 1000 μm were burned in a quiescent oxygen/nitrogen (20/80) environment at ambient pressures ranging from 0.1 to 4.3 MPa. The agglomerates suspended at the tip of SiC fibers were mounted inside a high-pressure combustion chamber. Ignition was achieved using a 35 W, 300 ms pulse from a CO_2 laser. Cinematographic records were taken with a high-speed camera. The surface temperature and intensity of the radiation emitted throughout the combustion process was recorded with a fast-response, near-infrared, two-color pyrometer and a broad-band radiometer, respectively. Figure 3.1 shows a schematic of the equipment set-up used in this experimental investigation.

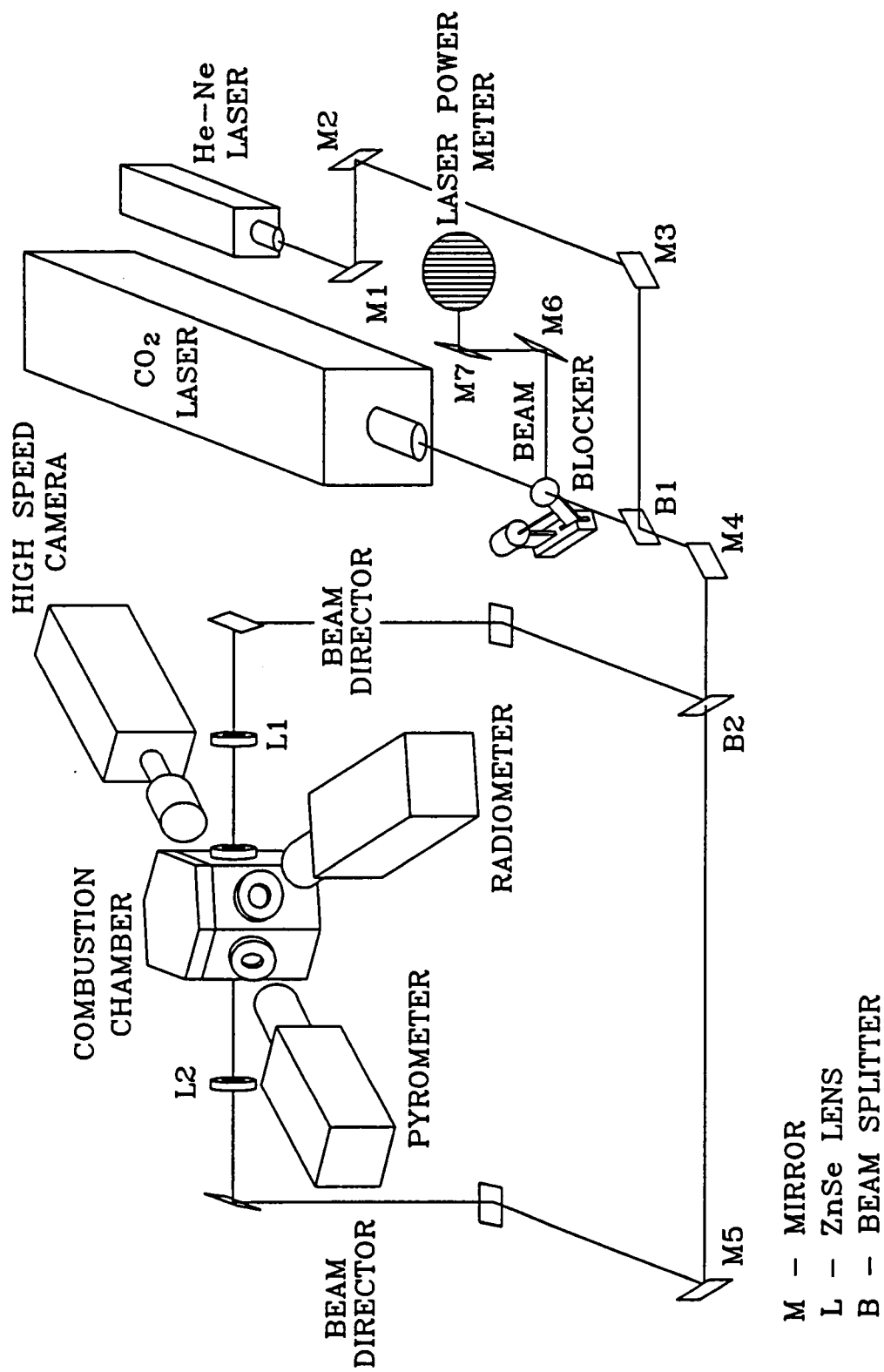


Figure 3.1 Schematic of experimental set-up.

The experimental procedure began by turning on the CO₂ laser. First, the flow of cooling water through the cooling jacket was initiated. Next, a vacuum pump was used to evacuate the plasma tube. Valves were then opened allowing the ionizing gases (He, N₂, CO₂) to be drawn into the plasma tube by the vacuum pump. Next, the CO₂ laser power supply was turned on with voltage set to half and current set to full (10 kV, 100 mA). As soon as the laser became active, the current was decreased to 50 mA. The ZnSe flat mirror at the output end of the CO₂ laser was aligned to produce the brightest, most uniform beam possible. This could be judged either by using the power meter or by placing a Zirconia fire brick in the beam's path. The laser's power could also be adjusted by varying the gas flow rates.

In order to prevent preferred ignition points, the CO₂ laser beam was focused on either side of the agglomerate. This was accomplished using a Ge (55/45) beam splitter to split the CO₂ laser beam into two separate beams. The two beams were directed by a series of mirrors and focused onto the agglomerate by ZnSe plano-convex lenses with 250 mm focal lengths.

Coincidence of the two CO₂ laser beams and alignment with the agglomerate was checked using a He-Ne laser introduced into the same optical path. A plastic strip was placed inside the combustion chamber and irradiated by a 40 ms pulse from the CO₂ laser. When the two beams were coincident, they produced a single circular hole. If this was not the case, the mirrors in each path were adjusted until the beams became coincident. The He-Ne laser was then adjusted so that the beam passed through the center of the hole to assure proper alignment.

The fiber-supported aluminum agglomerates were then mounted vertically inside the high-pressure combustion chamber. The combustion chamber was mounted on an adjustable platform which could be moved in the vertical direction and either horizontal direction to align the agglomerate with the laser beam using the He-Ne laser beam. The fields of view of the pyrometer, radiometer, and camera were then centered on the agglomerate.

A vacuum pump was used to evacuate the combustion chamber and piping system. The desired amounts of oxygen and nitrogen were introduced into the chamber from pressurized tanks. A desiccant was used to remove any moisture present in the gases since previous studies have shown that water vapor has a considerable effect upon the ignition and combustion processes.

The power of the CO₂ laser beam was measured by the power meter. The laser's power was adjusted by varying the flow rate of the gases and alignment of the internal mirrors. The duration of the CO₂ laser pulse could be varied from 40 to 1000 ms using a solenoid-activated beam blocker shown in Figure 3.2. The beam blocker could be activated either directly by the experimenter or by an electrical pulse sent by the high-speed camera. The beam blocker was connected to the data acquisition system to trigger data collection by the pyrometer and radiometer.

The combustion event was recorded by three instruments. A fast-response, near-infrared, two-color pyrometer measured the surface temperature of the aluminum droplet. The pyrometer operates in two narrow regions centered around 0.8 and 1.0 μm . The intensity of the radiation emitted by the burning droplet was measured

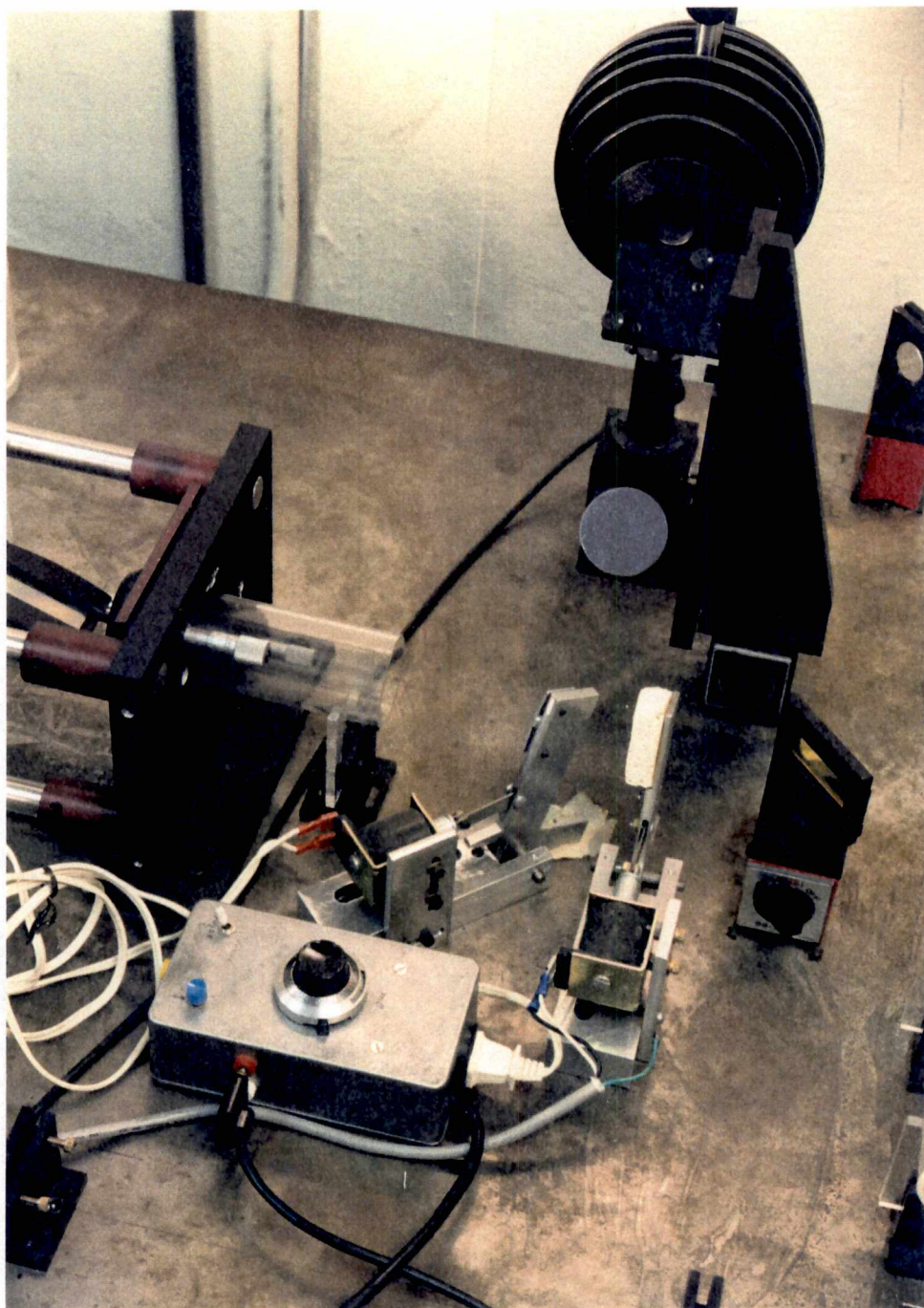


Figure 3.2 Photograph of solenoid-activated beam blocker assembly and timer used to direct the CO₂ laser beam to the power meter and to control the duration of the laser pulse.

by a broad-band radiometer. The radiometer is sensitive to radiation emitted in the spectral region from 0.3 to 1.1 μm . Data from the pyrometer and radiometer were collected by computer. Cinematographic records were taken of several experiments for each diameter at each pressure using a high-speed 16-mm motion picture camera.

3.2 TEST CONDITIONS

In this study, aluminum agglomerates with initial diameters of $835\ \mu\text{m} \pm 7\%$ and $1000\ \mu\text{m} \pm 5\%$ were burned in an oxygen/nitrogen (20/80) environment. The effects of ambient pressure was studied by conducting experiments at pressures of 0.1, 0.5, 0.8, 1.5, 2.2, 3.0, 3.6, and 4.3 MPa. The gases were desiccated to prevent the results from being affected by the presence of water vapor.

The laser pulse was maintained at 300 ms and $35.19 \pm 0.23\ \text{W}$ for the primary experiments. Additional experiments were performed to determine the effects of laser power and pulse duration on the ignition delay time and burning time of the agglomerates.

3.3 ALUMINUM AGGLOMERATE PRODUCTION TECHNIQUE

The aluminum agglomerate production technique has been described elsewhere by Meftah [6], only a brief discussion is given here. The aluminum agglomerates used in this study were produced using ninety-nine percent pure powdered aluminum

with an average particle size of 3 μm . Alcohol was added to the powder to form a mixture. A hypodermic syringe was used to place a drop of the mixture on the tip of a 150 μm diameter SiC fiber. Once the alcohol evaporated, the agglomerates were viewed under a microscope to assure that they were spherical. The agglomerates were allowed to dry for at least one week before being measured with the microscope and used in experimentation. Figure 3.3 shows a photomicrograph of a typical agglomerate with an initial average diameter of 1000 μm .

3.4 DESCRIPTION OF EXPERIMENTAL APPARATUS

This section gives a brief outline and description of the experimental apparatus used in this study. A more detailed account is given by Zaidi [20].

The high-pressure combustion chamber as shown in Figure 3.4 consists of a main chamber body and cap flange both machined from 316 stainless steel. The chamber body is six-sided with a 1" window in each side. It is 12 cm tall with a 6.5 cm inner diameter and a minimum wall thickness of 2.4 cm. The internal volume of 0.4 L is sufficiently large enough to prevent pressurization during combustion.

The cap flange is secured to the main chamber using six 3/8-16 Cr-alloy bolts. An O-ring is used to seal the gap between the flange and the main chamber. A clamping mechanism is used to secure the fiber-supported agglomerates to the cap flange. The flange is marked to insure proper installment such that the agglomerate can be positioned in the laser beam's path.

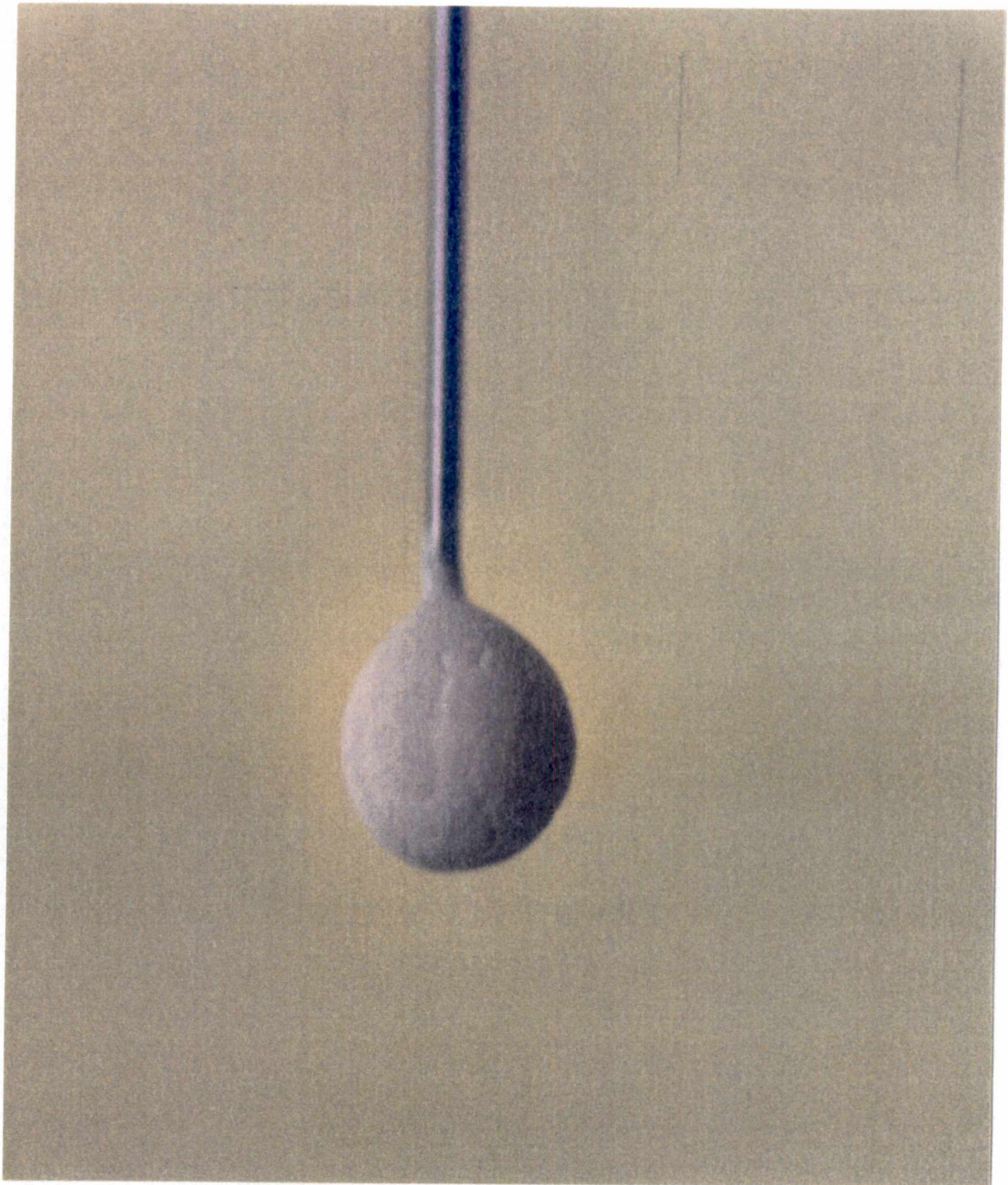


Figure 3.3 Photomicrograph of typical aluminum agglomerate with initial average diameter of $1000\ \mu\text{m}$.

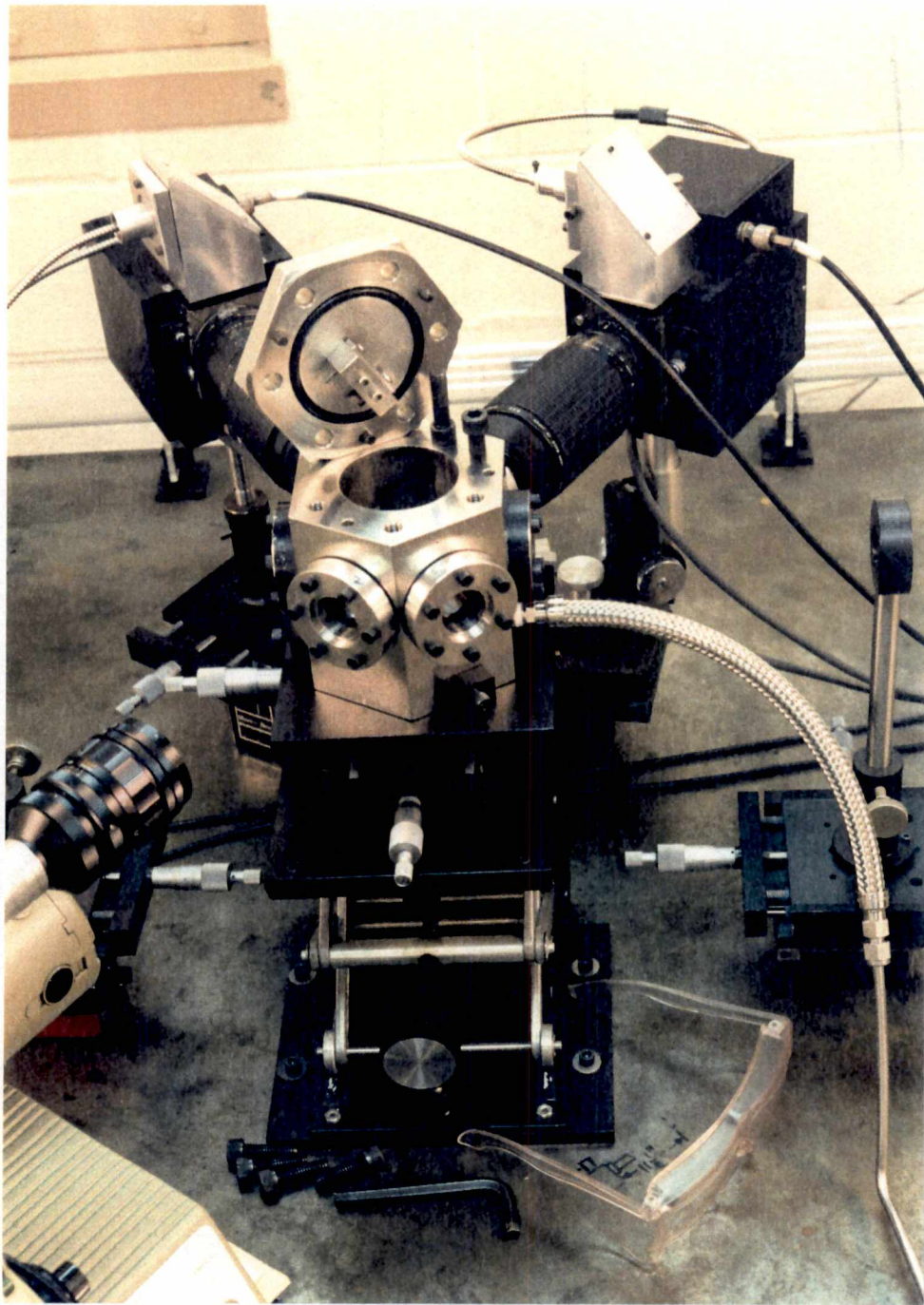


Figure 3.4 Photograph of high-pressure combustion chamber showing lab jack used to align the agglomerate with the CO₂ laser beam and orientation of the radiometer (left background), pyrometer (right background), and high-speed camera (left foreground).

The main chamber is equipped with six 1" windows held in place by flanges. Each stainless steel flange is secured by six 10-32 Cr-alloy bolts. Two opposing 1/2" thick ZnSe windows are used to transmit the CO₂ laser pulse. The remaining four windows are made of 1/2" thick glass. Three of these windows are reserved for the pyrometer, radiometer, and high-speed camera to record the combustion event. The remaining window is used as an observation window.

The oxygen/nitrogen mixture was introduced to the combustion chamber from pressurized tanks using a piping system shown schematically in Figure 3.5. A vacuum pump was used to evacuate the system. A desiccant was used to remove any moisture in the gases. After each experiment the combustion chamber was depressurized by venting to the atmosphere. The proper concentration of gases was achieved by controlling the partial pressure of each gas. A low pressure gage capable of measuring vacuum pressures was used to control the evacuation of the system and pressurization with oxygen. This gage was also used to control the pressurization with nitrogen for experiments at or below 0.8 MPa. For experiments with a higher total ambient pressure, a gage with a larger range had to be used. The least count of the low pressure gage is 2 psi (2 in Hg in vacuum) while the high pressure gage has a least count of 5 psi. This combination allowed the concentration of oxygen to be specified as $20\% \pm 1.6\%$ at an ambient pressure of 0.5 MPa. As pressure increases, the error in oxygen concentration decreases to a value of 0.2% at 4.3 MPa. For experiments with an ambient pressure of 0.1 MPa, a vacuum gage with a least count of 1 in Hg was used to reduce the error in oxygen concentration to an acceptable level.

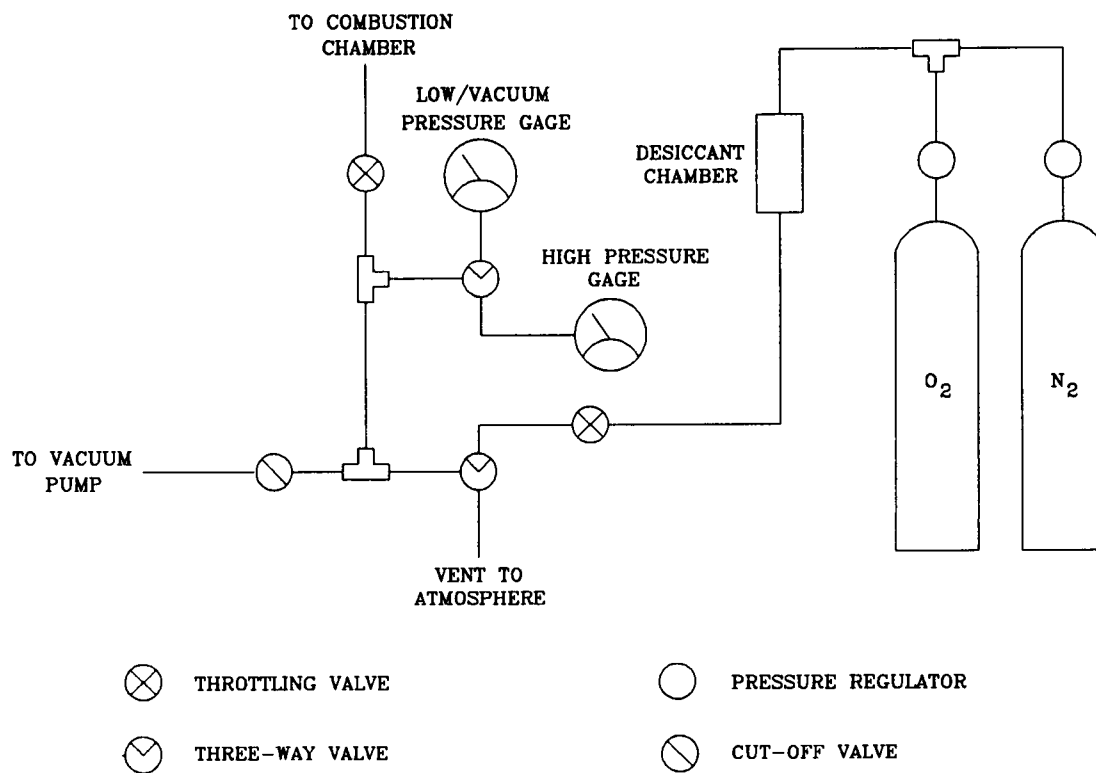


Figure 3.5 Schematic of piping system used to evacuate and pressurize the combustion chamber.

The CO₂ laser system is shown schematically in Figure 3.6. The laser optical resonator consists of a gold coated spherical concave 100% reflective copper mirror with a 10 m radius of curvature and a ZnSe flat 35% reflector. The separation distance of the two mirrors is approximately 2.5 m which is less than the radius of curvature of the concave spherical mirror. The resonator is therefore referred to as a quasi-hemispherical resonator. The ZnSe partially-reflective flat mirror allows power to be coupled out of the laser by zero-order reflection.

A four rod structure supports the laser plasma tube and the mirror assemblies. The laser plasma tube is constructed of Pyrex and has an 18 mm inner diameter and a 38 mm diameter water cooling jacket. Two anodes are located at either end and the single cathode is located in the center. Gases enter through a single inlet located in the center of the plasma tube and exit through outlets at either end. This construction ensures uniform gas flow throughout the amplifying section. A rectangular Zn-Se window is mounted on either end at a Brewster angle of 67.4°. This prevents reflection of the transmitted electric field in the plane of incidence. The output of this design, therefore, is linearly polarized in the vertical direction.

The gases are premixed in a separate chamber and drawn through the laser plasma tube by a vacuum pump. Optimum operation of the laser is achieved with a mixture of 80% He, 15% N₂, and 5% CO₂. The gas mixture is excited by a 20 kV, 100 mA Universal Voltronics power supply. A discharge current of 70 mA has been found to produce the maximum laser power.

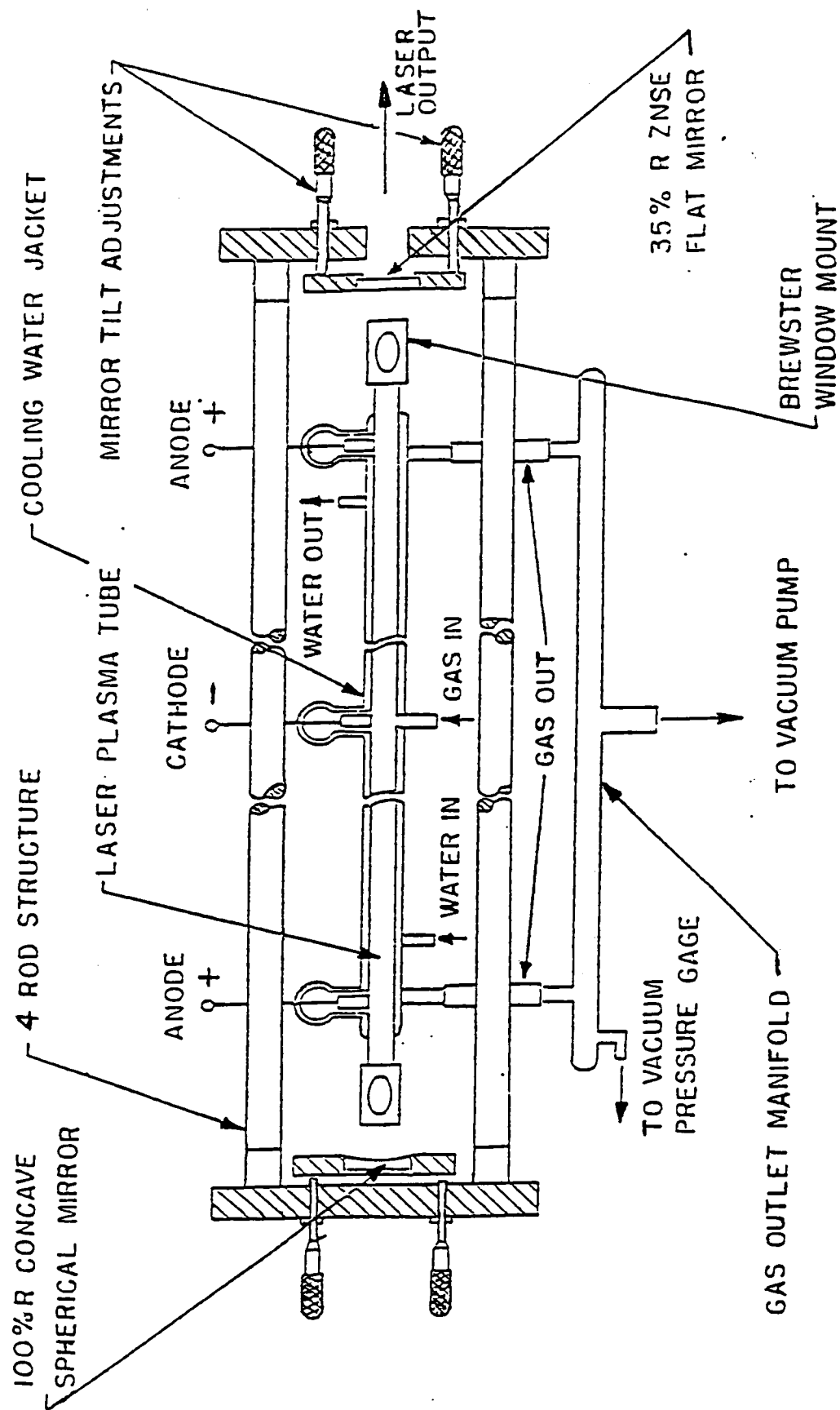


Figure 3.6 Schematic of the laser optical resonator, laser plasma tube, and mechanical structure of the CO₂ laser.

A fast-response two-color pyrometer operating in the near-infrared was used to measure the surface temperature of the burning droplet. It has a temperature range of approximately 1400 to 4000 K and a maximum time resolution of 25 μs . A Sigma macro zoom lens was attached to magnify the image of the agglomerate. The pyrometer is equipped with a Nikon Kellner 10x eyepiece which was used to center the agglomerate in the field of view prior to each experiment. A mirror was then moved into position to reflect the image to a bifurcated fiber-optic cable which splits the image into two beams. Each beam passes through a separate interference filter (one centered at 0.8 μm and the other at 1.0 μm) before reaching separate detectors.

A broad-band radiometer was used to record the intensity of the radiation emitted during the combustion event. The design and operation of the radiometer is similar to that of the pyrometer. However, the radiation focused by the zoom lens is reflected by a mirror onto a non-bifurcated fiber-optic cable and transmitted directly to a single detector. Since no interference filters are used, the radiometer is sensitive to a broad spectral region between 0.3 and 1.1 μm .

A Hycam high-speed 16-mm motion picture camera was used to take a cinematographic record of the combustion event for select experiments. At least two films were taken for each diameter at each pressure. Film speed was set at 1000 frames per second. A timer was used to produce a flash of light inside the camera at a frequency of 100 Hz. This resulted in a bright spot at the edge of every tenth frame of the film which could be used to accurately correlate the events on the cinematographic record with those recorded by the pyrometer and radiometer. When

the camera was used during an experiment, it was connected to the trigger for the beam blocker. After approximately 25 ft of the 100 ft roll of film had been exposed, the camera would send an electrical signal to trigger the solenoid-activated beam blocker which would in turn activate the data acquisition system. This allowed the camera to reach a steady film speed and to pass the first few layers of film exposed during loading before the combustion event began.

3.5 DATA ACQUISITION AND ANALYSIS

The data acquisition system consisted of an IBM-compatible 486 computer and a Metrabyte Dash 16 A/D board. The A/D board is capable of sampling up to sixteen channels at a maximum sampling rate of 50,000 Hz.

The data acquisition system was activated by the beam blocker's trigger mechanism. When the trigger was activated, an electrical signal was sent to the A/D board. The electrical signal was used to instruct the A/D board to begin collecting data from the pyrometer and radiometer. Since the beam blocker requires some time to swing completely out of the laser's path, there was a lag between the initiation of the data record and the irradiation of the agglomerate. This lag was found to be approximately 30 ms.

A computer program was used to reduce the data and store it on the hard drive. A calibration equation was used to convert the voltage signals from the pyrometer to surface temperature values. The signals from the radiometer were

normalized to give the relative intensity of the radiation emitted by the burning droplet. The data was plotted and used to determine the ignition delay time and burning time using methods which will be described later.

At least ten experiments were performed for each diameter at each pressure. In some experiments either the agglomerate failed to undergo complete combustion or the fiber was partially consumed. This data was eliminated from analysis.

CHAPTER 4

RESULTS AND DISCUSSION

This chapter consists of a presentation and discussion of the results obtained from the present experimental study of the effects of ambient pressure and initial diameter on the ignition and combustion characteristics of laser-ignited aluminum agglomerates. Section 4.1 describes the definition and determination of the ignition delay time and burning time. A discussion of the elimination of "bad" data due to incomplete combustion or fiber consumption is also presented. Section 4.2 presents general observations from this study based on the examination of the cinematographic records and combustion products. Section 4.3 addresses the effects of ambient pressure and initial diameter on the ignition delay time and burning time of the aluminum agglomerates. Finally, determination of the rate controlling factors for the reaction process is discussed in Section 4.4.

4.1 ANALYSIS OF EXPERIMENTAL RESULTS

In the present study, the ignition point of the aluminum agglomerate is defined as the point at which the flame first appears in the cinematographic record. With this definition, the ignition delay time is the time elapsed between the beginning of laser irradiation and the ignition point. Once ignited, the agglomerate enters a period of self-sustained combustion followed by a period of gradual cooling. The burning time

is taken as the time elapsed between the ignition point and the end of self-sustained combustion stage.

The ignition delay time and burning time of the aluminum agglomerates were first determined from cinematographic records. The cinematographic records were then compared to the intensity and temperature plots in order to develop a method for determining the ignition delay time and burning time from these plots. As explained earlier, there was a lag of approximately 30 ms between the initiation of the data record and laser irradiation of the agglomerate. This was due to the beam blocker needing time to physically swing out of the beam's path. The ignition point was found to correspond to the presence of a sharp peak on the intensity plot. The termination of self-sustained combustion was found to be characterized by a sudden transition to an exponentially decaying cooling curve on the intensity plot.

Use of the temperature curve was less effective in determining the ignition delay time and burning time. The molten droplet apparently moved out of the limited field of view of the pyrometer on numerous occasions. The ignition temperature was confirmed to be ~ 2300 K which corresponds to the melting temperature of Al_2O_3 . The ignition point was occasionally represented by a sharp peak on the temperature curve, especially at low pressures. The termination of self-sustained combustion was difficult to define on the temperature plot but tended to correspond to a transition towards a somewhat parabolic down-slope in the curve. However, due primarily to droplet movement, neither of these trends were found to be as reliable as those represented by the intensity plot.

In order to obtain a good statistical average value for the ignition delay time and burning time, at least ten successful experiments were performed for each diameter at each pressure. A large number of experiments were deemed "unsuccessful" due either to excessively incomplete combustion or significant consumption of the SiC fiber. The burning times of the experiments deemed "unsuccessful" due to incomplete combustion were roughly half as long as for "successful" experiments. Products from "successful" experiments conducted at elevated pressures typically consisted of a small inner core of unburned aluminum surrounded by a thick layer of opaque oxide. Experiments deemed "unsuccessful" due to incomplete combustion typically resulted in products consisting almost entirely of unburned aluminum surrounded by a thin layer of oxide as shown in Figure 4.1. While experiments deemed "unsuccessful" and "successful" both proved to involve incomplete combustion of the aluminum agglomerate, the size of unburned aluminum core from successful experiments was found to be significantly less than the size of the original agglomerate. For unsuccessful experiments, however, the unburned aluminum residue was of approximately the same size as the original agglomerate. The oxide product from successful experiments was opaque and cream-colored whereas unsuccessful experiments produced a more transparent oxide.

In some residues from successful experiments, a small portion of SiC from the supporting fiber was discovered. Turns, Wong, and Ryba [5] also observed consumption of the supporting SiC fiber. They concluded that consumption of the SiC fiber had little influence on the combustion process since an analysis of the

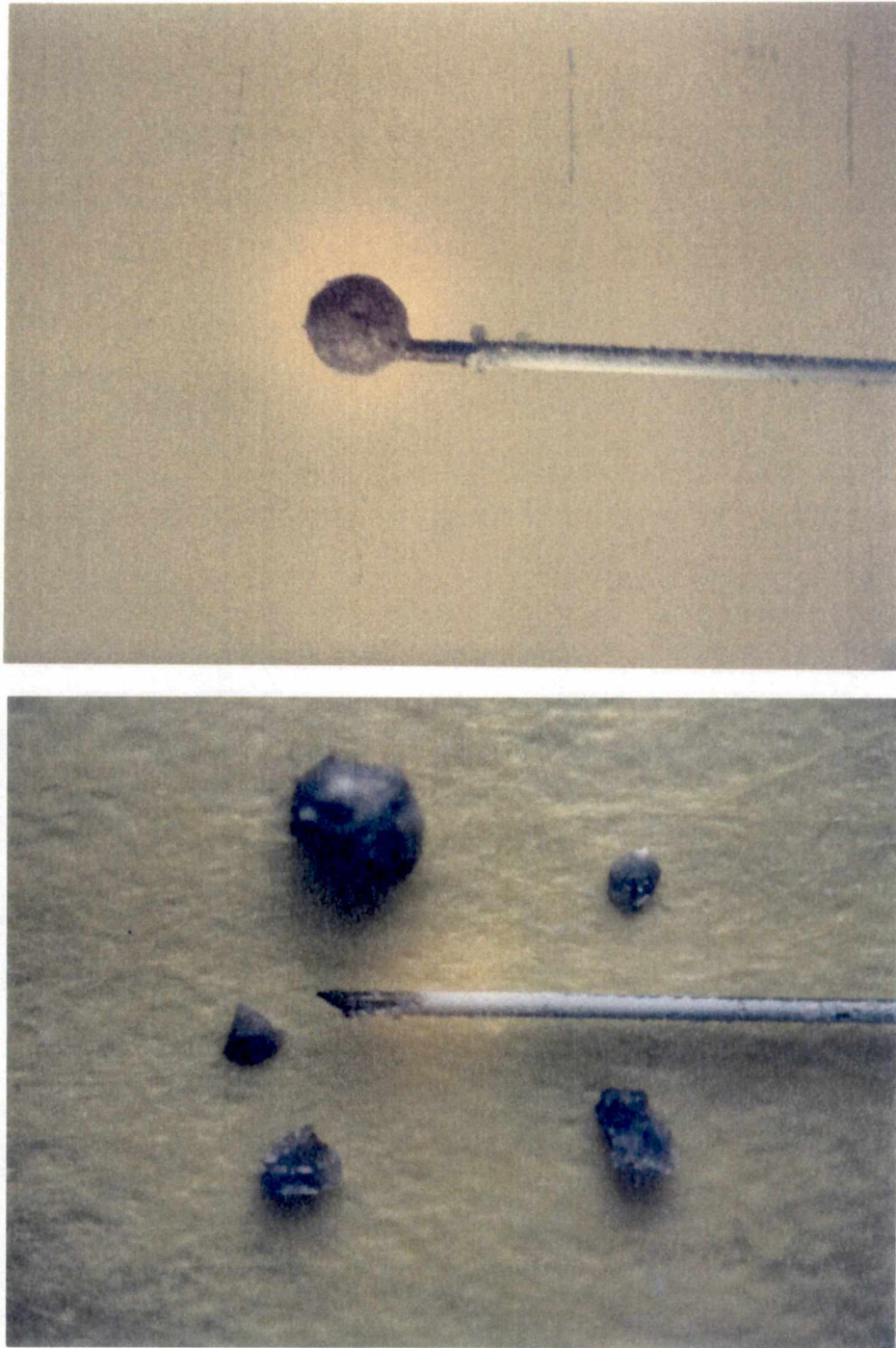


Figure 4.1 Photomicrographs of combustion residue from experiments with 835 μm diameter agglomerates at elevated pressures for which incomplete combustion was observed.

products from their experiments showed no signs of carbon compounds and only trace amounts of β -SiC.

In the early stages of the combustion process, the droplets were observed to change position from being centered on the supporting fiber to being offset to one side and slightly higher. This repositioning occurred while the laser pulse was still incident upon the droplet and the surface temperature of the droplet was at its highest. It is likely, therefore, that any consumption of the fiber occurred at this point. Thus, it is suggested that the repositioning of the droplet is either the effect or the cause of fiber consumption. That is to say that the portion of the fiber covered by the droplet may have been consumed forcing the molten droplet to move upward for support. On the other hand, the repositioning could be due to the molten droplet seeking the maximum support available by surface tension. In this case, once the droplet had repositioned itself, a portion of the fiber would be exposed both to the laser pulse and direct heat transfer from the flame zone possibly resulting in fiber consumption.

In this study, experiments were disregarded only when the fiber consumption was significant enough that the burning droplet moved out of the field of view of the pyrometer, radiometer, and camera. When this occurred, it was impossible to determine the burning time of the agglomerates.

An analysis of the occurrence of unsuccessful experiments indicates a dependence upon both initial diameter and ambient pressure. Table 4.1 shows the percentage of experiments which were deemed successful for 835 and 1000 μm diameter agglomerates at each pressure (experiments in which significant fiber

Table 4.1 Percentage of experiments deemed successful for each diameter at each pressure. Experiments in which significant fiber consumption was observed were not included in the analysis.

Pressure, MPa	Percentage of Successful Experiments	
	835 μm DIA	1000 μm DIA
0.1	62.5	52.6
0.5	55.6	41.7
0.8	76.9	35.5
1.5	83.3	40.0
2.2	66.7	50.0
3.0	71.4	76.9
3.6	66.7	62.5
4.3	90.9	84.6

consumption was observed were not included in this analysis). As can be seen, the likelihood of obtaining complete combustion generally increases as initial diameter is decreased. An increase in ambient pressure is also seen to roughly increase the likelihood of complete combustion.

There are numerous possible explanations for the incomplete combustion of these agglomerates. Larger agglomerates were found to be less likely to burn to completion. This could be a result of the larger mass, especially at low pressures since a larger mass of aluminum would require a greater amount of oxidizer to react to completion. Since an increase in pressure results in an increase in the likelihood of the experiment being successful, it is possible that at low pressures there is not enough oxidizer to react with all of the aluminum or, more likely, that the diffusion rate of the oxidizer is insufficient to maintain the reaction. However, there is another possible contribution. Three different pressure gages were used to measure the partial pressure of the oxygen and nitrogen. The gage used for experiments conducted at 0.1 MPa has a readability of 0.5 in of Hg. This only allows the oxygen concentration to be specified as $20\% \pm 2.0\%$. A low pressure gage was used to monitor the evacuation of the combustion chamber and piping system and the pressurization of the combustion chamber with oxygen for all experiments and nitrogen for experiments at or below 0.8 MPa. This gage has a readability of 1 in of Hg in vacuum and 1 psi in gage allowing the oxygen concentration to be specified as $20\% \pm 0.6\%$ at 0.5 MPa. Experiments at higher pressures required the use of gage with a higher range and a readability of 2.5 psi to monitor the pressurization of the system with nitrogen. This

combination of low and high pressure gages allows the oxygen concentration to be specified as $20\% \pm 0.7\%$ at 1.5 MPa. The error decreases with increasing pressure so that at 4.3 MPa the oxygen concentration can be specified as $20\% \pm 0.2\%$. While the errors appear small, it is possible that low oxygen concentration due to the readability of the pressure gages was partially responsible for the incomplete combustion. The fact that higher pressures have less error in oxygen concentration and higher success rates tends to support this possibility. Additionally, high oxygen concentrations could be partially responsible for fiber consumption since experiments at 30% oxygen often resulted in fiber consumption.

While all the above explanations are at least reasonably plausible, none sufficiently explain the phenomenon. It is more likely that a combination of these factors lead to incomplete combustion. Other factors were addressed which could have possibly led to incomplete combustion of the agglomerate. Several agglomerates were weighed and measured in an attempt to determine if the porosity of the agglomerates produced varied; however, no variance could be discerned. While the laser was carefully aligned, there is some possibility that the laser did not always irradiate the entire agglomerate. The laser power was maintained within a small range of 35.19 ± 0.23 W thus making it unlikely that variance in laser power had any effect upon the success of the experiment.

It seems then that incomplete combustion of the aluminum agglomerates is more of a consequence of large agglomerate size and low ambient pressure than test conditions. For this reason, it appears that the agglomerate size should be kept small

and the ambient pressure kept high in order to assure that the majority of the aluminum burns to completion.

4.2 GENERAL OBSERVATIONS

This section describes observations from the current study of the ignition and combustion of aluminum agglomerates with initial average diameters of 835 and 1000 μm at ambient pressures ranging from 0.1 to 4.3 MPa. The observations were made by analyzing the cinematographic records, intensity and temperature plots, and combustion products. Increasing the agglomerate diameter was found to result in an increase in the ignition delay time and burning time. Otherwise, initial agglomerate diameter was found to have no effect upon the ignition and combustion process. The ambient pressure, however, was found to have a dramatic effect on the combustion process. Specifically, the combustion process at atmospheric pressure was found to be significantly different than the combustion process at elevated pressures. In addition, agglomerates burned in a 30% oxygen environment displayed more violent combustion involving jetting and fragmenting. This section is further divided into three subsections. Section 4.2.1 describes observations from experiments conducted at atmospheric pressure. Observations from experiments conducted at elevated pressures are discussed in Section 4.2.2. Section 4.2.3 gives an overview of observations from experiments conducted in a 30% oxygen environment at 0.34 MPa.

4.2.1 OBSERVATIONS FROM TESTS AT ATMOSPHERIC PRESSURE

Results from experiments conducted at 0.1 MPa showed the combustion process to be quite different than at higher pressures. The ignition delay time and burning time were found to be considerably longer than at elevated pressures. In addition, the combustion residues were found to be very different from those formed at elevated pressures. Figure 4.2 shows a typical intensity and temperature plot for the combustion of a 1000 μm diameter agglomerate at 0.1 MPa.

The ignition and combustion of the aluminum agglomerates are characterized by three stages. The first stage consists of the heating of the agglomerate to the ignition point. The agglomerate is irradiated on both sides by the CO_2 laser pulse causing both halves to glow. As the beam continues to heat the agglomerate, the glowing area grows until the entire agglomerate is glowing dull orange. As observed by Wong and Turns [19], the protective oxide layer surrounding the individual aluminum particles which constitute the agglomerate, maintains the integrity of each particle until the surface temperature of the agglomerate reaches the melting point of Al_2O_3 (2315 K). At this point the oxide layer surrounding each molten droplet of aluminum can no longer maintain the integrity of the individual droplets and the agglomerate coalesces into a single molten droplet of aluminum. The oxide layer then cracks in numerous areas allowing aluminum vapor to diffuse through and react with oxygen thus igniting the droplet. The reaction zone quickly spreads to envelop the entire droplet and develop into a concentric diffusion flame zone separated from the

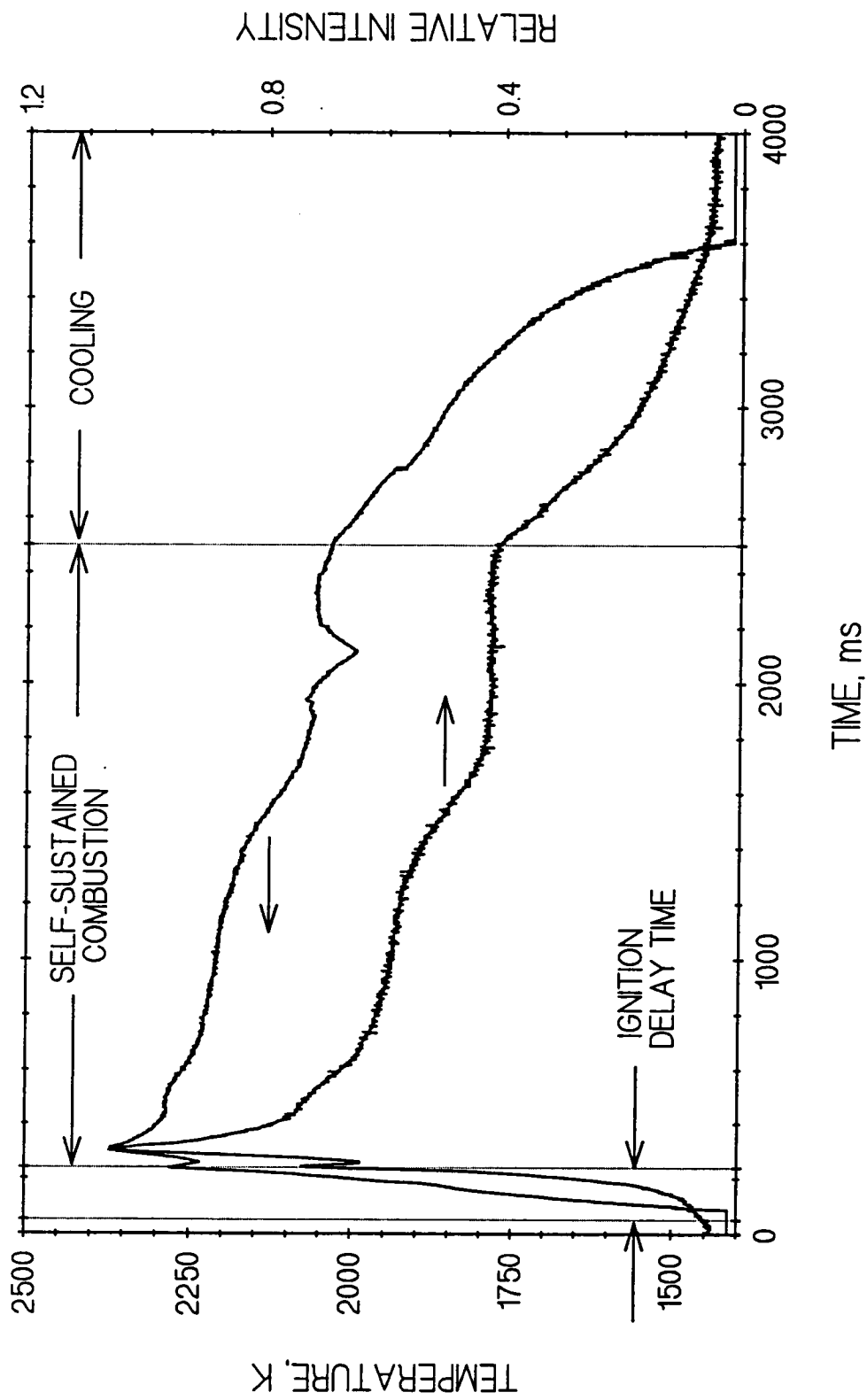


Figure 4.2 Typical temperature and relative intensity plot for a 1000 μm diameter aluminum agglomerate at 0.1 MPa.

surface of the droplet. The rapid growth in the flame size and intensity was found to reach a peak and then diminish slightly. This occurrence results in a sharp peak being formed on the intensity plot at the ignition point. The temperature plot occasionally shows a similar peak as in Figure 4.2. Using the temperature plot, the surface temperature of the droplet at the ignition point was found to be approximately equal to the melting point of the oxide (2315 K).

The second stage of the combustion process involves the self-sustained combustion of the molten aluminum droplet. The onset of this stage is characterized by the full development of the flame. At atmospheric pressures, the flame forms some distance from the surface of the droplet as typified by vapor-phase combustion. The flame structure consists of a bright white inner zone and a blue-green outer envelope concentric to the droplet. Spectroscopic investigations of the flame zone by previous investigators [21, 22] have shown that the blue-green region is indicative of the presence of AlO. A large plume of oxide smoke is convected upward from the reaction zone depositing a thick coating of condensed oxide on the SiC fiber and the top of the combustion chamber.

Once ignited, the droplet transitions to a period of steady combustion which occupies a significant portion of the total burning time. During this period, the droplet is steadily consumed and reduced in size. The flame stand-off ratio initially remains rather constant for some time before gradually decreasing until the flame collapses to the droplet surface. After this happens, the reaction takes place at the droplet surface for the remainder of the combustion process until eventually

extinguishment occurs. During the period of surface burning, the droplet glows bright white and gradually diminishes in intensity and changes color to a dull orange. As the combustion process draws to an end, the droplet begins to solidify rather uniformly.

The final stage of the combustion process consists of the cooling of the combustion residue. As mentioned previously, the cooling of the residue is characterized on the intensity plot by an exponentially decaying curve. At low pressures, the cooling process requires a considerable amount of time as shown in Figure 4.2. Also, the transition from self-sustained combustion to cooling is quite prominent.

At atmospheric pressure, the combustion residue consists of a hollow sphere of oxide and some unburned aluminum as shown in Figure 4.3. The outer surface of the sphere is rough and seems to consist of small solid spheres of oxide. The inner surface is smooth and appears to be coated with a thin layer of unburned aluminum.

In previous studies at atmospheric pressure such as those by Friedman and Maček [9,10] and Drew, Gordon, and Knipe [13], hollow oxide spheres were also found in the combustion products. However, these experiments were conducted in wet environments whereas dry environments, such as in the case of the present study, were found not to produce the hollow oxide spheres. In addition, the hollow spheres from these studies were transparent with a smooth outer surface. Quenching studies by Drew et al. showed that the hollow oxide spheres were formed by ballooning of the oxide cap resulting in a bilobal structure to the burning droplet. Cinematographic

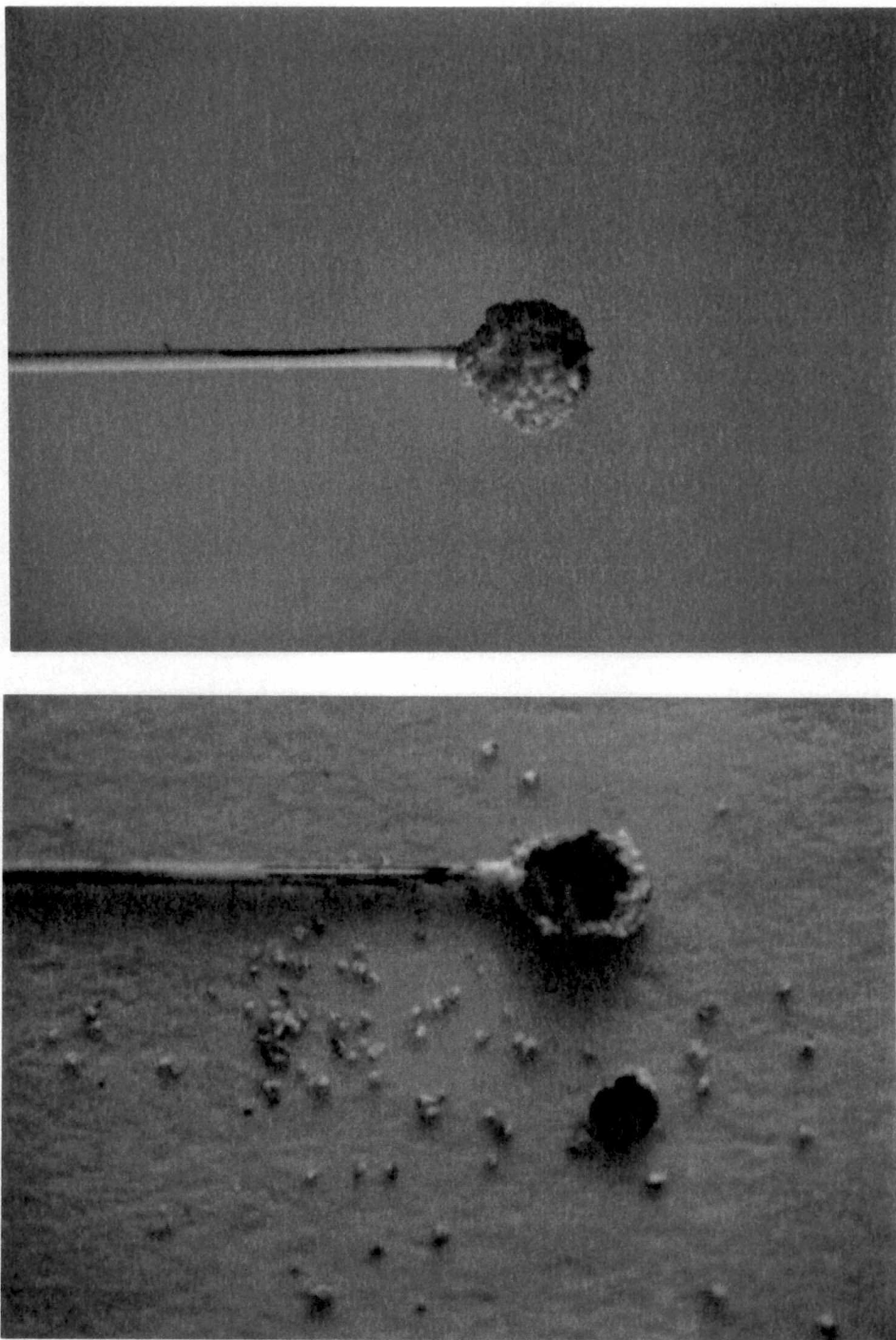


Figure 4.3 Photomicrographs of typical combustion residue from 1000 μm diameter aluminum agglomerates at 0.1 MPa.

records from the current study show no evidence of such a bilobal structure. In truth, the current study actually seems to support the much criticized and dismissed combustion model proposed by Bartlett, Ong, Fassell, and Papp [12]. In their model, the molten oxide layer surrounding the aluminum droplet is inflated by the vapor pressure of the aluminum and forms a concentric shell around the droplet. Once the aluminum has been consumed, Bartlett et al. proposed that the oxide shell cools and solidifies forming the hollow oxide sphere. However, Drew, Gordon, and Knipe suggest that since the boiling temperature of aluminum is higher than the melting temperature of the oxide, as cooling occurred the aluminum vapor would condense thus allowing the oxide bubble to collapse before it could solidify. It is possible that the condensation of the aluminum vapor on the inner surface in some way prevents the collapse of the oxide bubble.

4.2.2 OBSERVATIONS FROM TESTS AT ELEVATED PRESSURES

At first glance, the combustion process of the aluminum agglomerates at elevated pressures seems similar to the process at atmospheric pressure. However, analysis of the combustion products and a closer analysis of the cinematographic records show that the process is dramatically different. Figure 4.4 shows a typical temperature and intensity plot for an 835 μm diameter agglomerate at 2.2 MPa.

The combustion process can again be characterized by three distinct stages. The heating and ignition of the agglomerate at elevated pressures is similar to that at

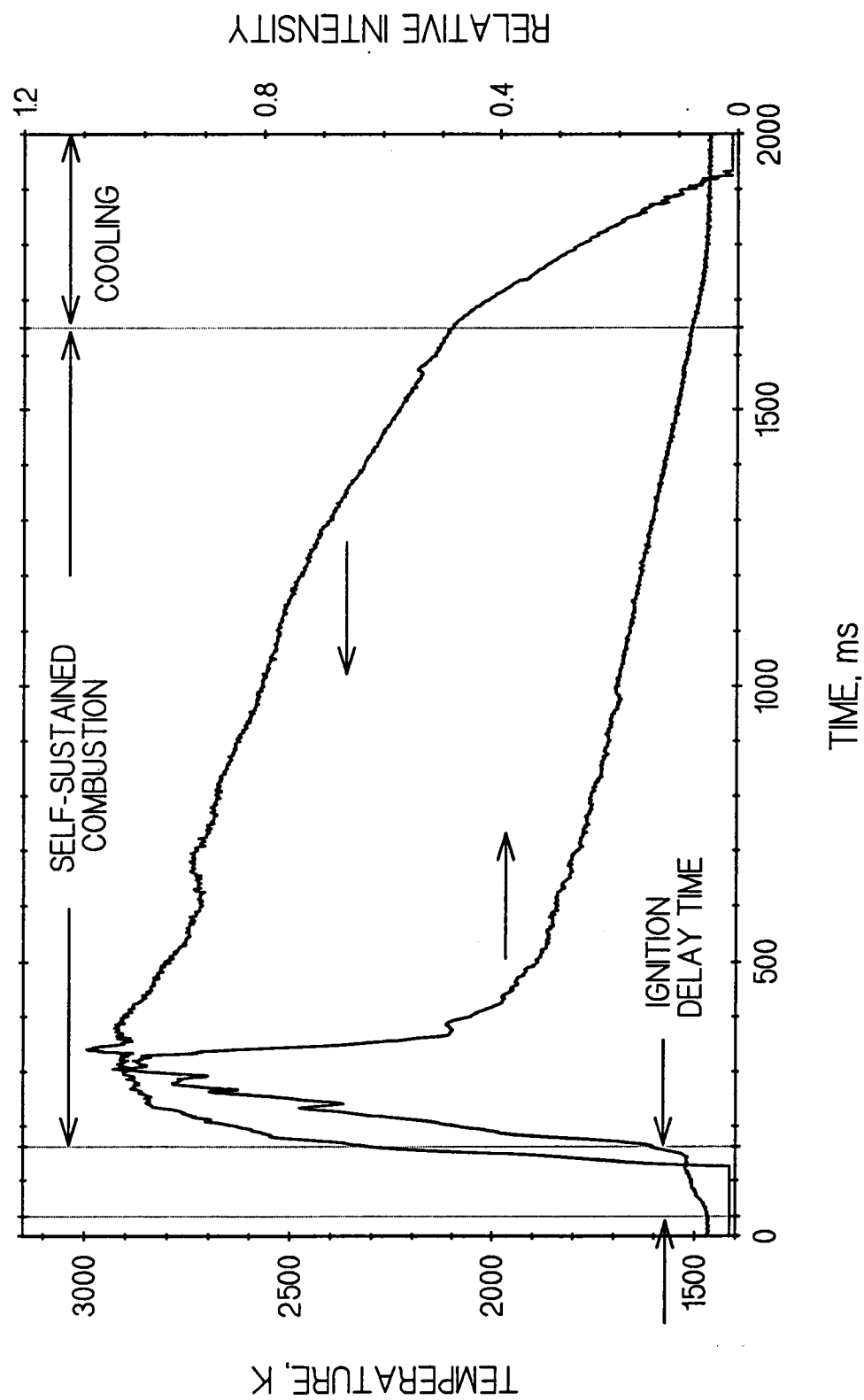


Figure 4.4 Typical temperature and relative intensity plot for an 835 μm diameter aluminum agglomerate at 2.2 MPa.

atmospheric pressure. The ignition delay time, however, was found to decrease with increasing pressure. Also, the flame forms closer to the droplet surface as pressure is increased. As a result, the development of the flame is less dramatic and the characteristic ignition peak on the intensity curve becomes less prominent.

Once the flame has fully developed, the droplet enters the self-sustained combustion stage. The flame structure is essentially the same as at atmospheric pressure and consists of an inner white zone surrounded by a concentric blue-green region. The flame stand-off ratio was found to decrease with increasing pressure. The plume of oxide also decreased in size as pressure was increased.

At the beginning of the self-sustained combustion, the droplet undergoes a short turbulent period during which the droplet changes position from being centered on the SiC fiber to being offset to one side. As mentioned previously, this phenomena is believed to be associated with either surface tension or fiber consumption. This change of position causes the flame region to wave and flicker rather violently. At 0.5 MPa, the turbulent period is quite long. Initially the droplet changes position; however, the flame region continues to wave and flicker for some time after the droplet has moved. As pressure is increased, the length of the turbulent period decreases. At and above 3.6 MPa the turbulent period lasts only long enough for the droplet to change position. On rare occasions burning particles were observed to be sporadically ejected from the droplet during the turbulent period at all elevated pressures. The turbulent period transitions into a period of steady burning followed by a period of surface burning just as at atmospheric pressure

At 0.5 MPa the period of steady burning occupies the majority of the burning time, and the flame stand-off ratio is considerably less than at atmospheric pressure. Eventually, the flame collapses to the surface, the plume of oxide disappears, and the reaction begins to take place on the droplet surface. As pressure is increased, the period of steady burning becomes shorter and the stand-off ratio decreases. At 2.2 MPa, the steady and surface burning periods are of approximately equal length. As pressure is further increased, the surface burning accounts for the majority of the burning time. At and above 3.6 MPa, the flame is formed almost at the surface of the droplet. The turbulent period lasts only long enough for the particle to change position and the period of steady combustion is almost non-existent. The reaction occurs at the surface of the droplet for almost the entire self-sustained combustion stage. Only a small plume of oxide is observed, and, as a result, very little oxide accumulates on the SiC fiber.

Typical residues from experiments conducted using 835 and 1000 μm diameter aluminum agglomerates at pressures of 3.0 and 0.8 MPa, respectively, are shown in Figure 4.5. As can be seen, the residue primarily consists of opaque oxide which was typically found to surround a core of unburned aluminum which is much smaller than the original agglomerate. A small amount of unburned aluminum was occasionally found to protrude to the surface of the particle forming a small spherical droplet as in the case shown in Figure 4.5.

Apart from the presence of the unburned aluminum core, residues from experiments at elevated pressures seem to correspond to the agglomerate combustion

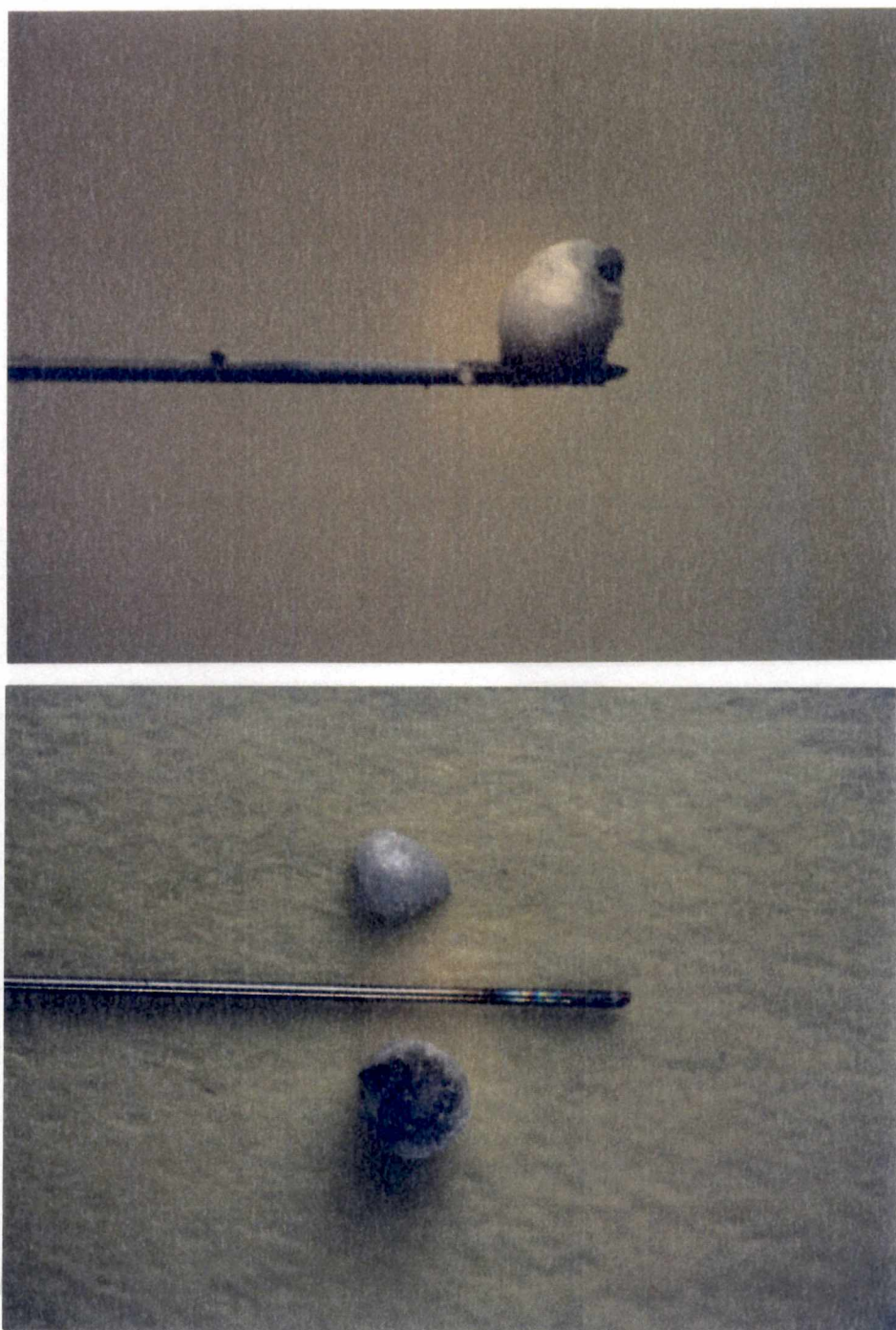


Figure 4.5 Photomicrographs of typical combustion residues from aluminum agglomerates with initial diameters of 835 and 1000 μm burned at pressures of 3.0 and 0.8 MPa, respectively.

model proposed by Turns, Wong, and Ryba [5]. In their model, the oxide cap formed on the surface of the burning droplet continues to grow throughout the combustion process due to accumulation of condensed oxide convected inward from the flame zone. In their study, the oxide residues were found to be porous and about 60% to 80% as large as the initial agglomerate. The residue shown in Figure 4.5 from the current study appears non-porous and is approximately 50% as large as the initial agglomerate. In some of the experiments conducted by Turns, Wong, and Ryba, the oxide layer was found to completely surround the burning droplet. In the current study, the oxide layer was found to be thicker on one side of the residue. This suggests that the oxide layer formed a cap on one side and then expanded during the combustion process until the entire surface of the droplet was covered. It is probable that this oxide coating then inhibited the diffusion of aluminum vapor to such a degree that combustion was no longer possible resulting in incomplete combustion of the agglomerate. For cases where a small amount of unburned aluminum was found to protrude to the residue surface, it is believed that the oxide layer grew to cover all but a small region of the droplet surface. The molten aluminum then flowed through this hole to react with oxygen at the surface. Eventually the heat loss to the surroundings would become greater than the heat produced by the reaction. As a result, the molten aluminum droplet would cool and the evaporation rate of the aluminum would slow until it was no longer able to keep up with the reaction rate. This would again result in incomplete combustion of the agglomerate. This observation is supported by cinematographic records. During the later stages of

surface burning, the cooling of the droplet was observed to commence at one end and then spread until the majority of the droplet was dull orange with a small area, presumably burning aluminum, glowing bright white.

4.2.3 OBSERVATIONS FROM TESTS IN A 30% OXYGEN ENVIRONMENT

Several experiments were conducted in which aluminum agglomerates with an initial average diameter of 1000 μm were burned in an oxygen/nitrogen (30/70) environment at 0.34 MPa. In these experiments, the combustion process was very violent with burning particles constantly being ejected from the droplet. The SiC supporting fiber was rapidly consumed and the droplets always moved out of the field of view of the pyrometer, radiometer, and camera often within 500 ms of ignition. As a result, burning time could not be determined, and, thus, further study of the ignition and combustion processes of aluminum agglomerates in 30% oxygen environments was not attempted.

4.3 EFFECTS OF AMBIENT PRESSURE AND INITIAL DIAMETER

This section discusses the results obtained from the current experimental study on the effects of ambient pressure and initial diameter on the ignition delay time and burning time of aluminum agglomerates. In the present study the ignition delay time is defined as the time elapsed between the beginning of laser irradiation and the

ignition of the aluminum agglomerate. The burning time is defined as the time elapsed between ignition and the end of the self-sustained combustion event. Previous studies have shown that the ignition delay time of aluminum agglomerates is considerably shorter than the burning time. However, these studies have also shown that the ignition delay time of the aluminum agglomerates is approximately the same as the burning time for a similar size droplet of pure liquid fuel. Thus, consideration of both the ignition delay time and the burning time of the aluminum agglomerates is important when determining whether the combustion process for a slurry fuel will be complete when the fuel leaves the combustor.

Experiments were performed at eight different ambient pressures ranging from 0.1 to 4.3 MPa using agglomerates with initial average diameters of 835 and 1000 μm . In addition, results obtained by Meftah [6] in a previous study using aluminum agglomerates with an initial average diameter of 535 μm at the same ambient pressures were incorporated into the analysis. The agglomerates were burned in a quiescent oxygen/nitrogen (20/80) environment at room temperature. The gases were desiccated to prevent the results from being affected by the presence of water vapor. Ignition was achieved with a 300 ms pulse from a 35 W CO_2 laser beam.

Section 4.3.1 presents the effects of ambient pressure and Section 4.3.2 discusses the effects of initial agglomerate diameter on the ignition delay time and burning time. In addition, Section 4.3.3 gives a brief discussion of the effects of laser power and pulse duration on the ignition delay time and burning time of the aluminum agglomerates.

4.3.1 EFFECTS OF AMBIENT PRESSURE

In the present study the ambient pressure was found to have a dramatic effect upon both the ignition delay time and the burning time of the laser-ignited aluminum agglomerates. Table 4.2 shows the ignition delay time for 535, 835, and 1000 μm diameter laser-ignited aluminum agglomerates as a function of ambient pressure. Figure 4.6 presents the same information in graphical form. In both instances the associated error bands represent twice the standard deviation of the mean.

As can be seen, the initial study by Meftah using agglomerates with an initial diameter of 535 μm found no correlation between ambient pressure and ignition delay time. The ignition delay time was found to vary from 55 to 80 ms with no obvious trend associated with pressure. However, the present study showed a strong association between ignition delay time and ambient pressure. At atmospheric pressure, the ignition delay time was found to be quite long, 200 ms, for both 835 and 1000 μm diameter agglomerates. As pressure was increased the ignition delay time was initially found to decrease rapidly. Above 1.5 MPa for 835 μm diameter agglomerates and above 2.2 MPa for 1000 μm diameter agglomerates, however, the ignition delay time remained relatively constant at approximately 100 ms for 835 μm diameter agglomerates and 125 ms for 1000 μm diameter agglomerates.

Since decreasing the pressure also decreases the mass of oxygen present, it is reasonable to expect that the time required for ignition to occur will increase. Furthermore, when pressure reaches zero, the agglomerate would obviously never

Table 4.2 Effects of ambient pressure on the ignition delay time of 535, 835, and 1000 μm diameter aluminum agglomerates.

Pressure, MPa	Ignition Delay Time, ms		
	535 μm DIA	835 μm DIA	1000 μm DIA
0.1	74	200 ± 20	200 ± 25
0.5	77	165 ± 10	195 ± 10
0.8	63	150 ± 20	175 ± 10
1.5	66	110 ± 20	165 ± 10
2.2	78	115 ± 10	140 ± 10
3.0	80	100 ± 10	130 ± 15
3.6	60	105 ± 15	130 ± 10
4.3	55	90 ± 10	120 ± 10

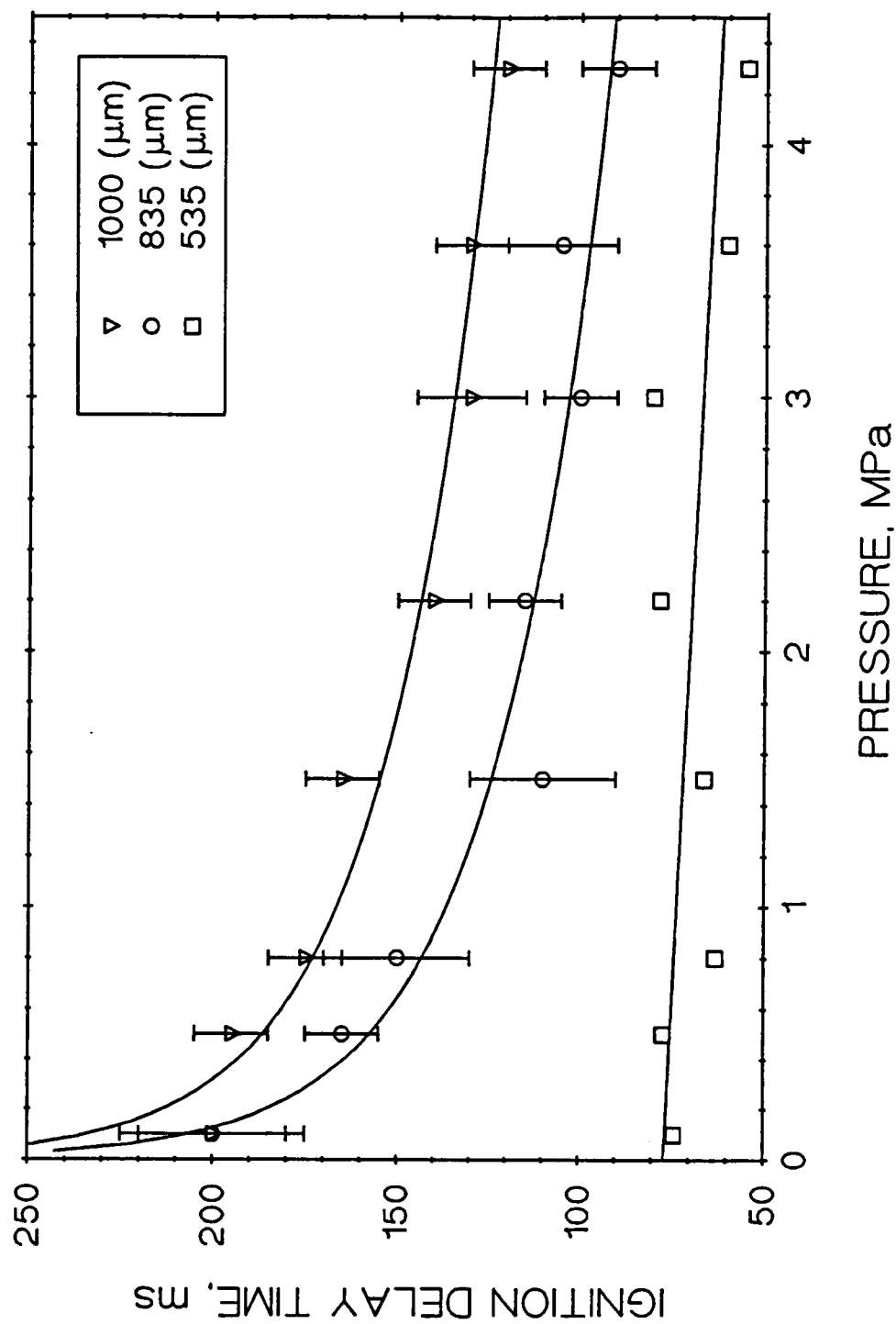


Figure 4.6 Effects of ambient pressure on the ignition delay time of aluminum agglomerates with initial average diameters of 535, 835, and 1000 μm .

ignite and thus the ignition delay time would approach infinity just as indicated in Figure 4.6. As pressure is increased, however, eventually a point is reached beyond which increasing pressure does not result in a decrease in ignition delay time. The ignition of aluminum is primarily dependent upon there being enough heat transferred to the agglomerate to melt the oxide layer. Increasing the pressure also results in a greater heat loss from the agglomerate to the surroundings. The observation that ignition delay time is relatively constant at high pressures seems to indicate that a balance is reached between these two effects.

It should be noted that the ignition delay time will be dependent upon the method used to ignite the agglomerates. In practical applications, ignition would be achieved by heating of the agglomerates by the hot ambient gases at an elevated pressure. In the experiments conducted by Wong and Turns [19], ignition was achieved by exposure to the hot oxidizing gases of a flat flame burner at atmospheric pressure. In the current study, ignition was achieved in a cold ambient environment at various pressures by irradiation from a CO₂ laser pulse. This method would seem to provide greater radiant heat transfer than convection from hot ambient surroundings even though there would be some heat loss to the cold ambient surroundings.

At atmospheric pressure, agglomerates from the current study were found to have a slightly shorter ignition delay time than that found by Wong and Turns which seems to confirm the assumption that the laser irradiation provides more effective heat transfer than convection from hot ambient gases. However, the difference is only slightly larger than the experimental uncertainty. The ignition delay times found by

Wong and Turns appear more accurate due to ignition being achieved by heat transfer with the hot surroundings. However, the current study has shown that at elevated pressures the ignition delay time is reduced. Thus, it is predicted that for practical applications, with ignition achieved by exposure to hot ambient gases at elevated pressures, the ignition delay time will be longer than the values found in the current study but shorter than that predicted by Wong and Turns for atmospheric pressure.

Table 4.3 presents the burning time of laser-ignited aluminum agglomerates with initial average diameters of 535, 835, and 1000 μm as a function of ambient pressure. The same information is presented in graphical form in Figure 4.7. Again, the associated error bands represent twice the standard deviation of the mean.

At atmospheric pressure, the burning time was found to be 1338, 1860, and 2280 ms for 535, 835, and 1000 μm diameter agglomerates, respectively. As can be seen, for all diameters the burning time initially decreases rapidly as pressure is increased. However, a pressure limit is soon reached beyond which further increases in pressure have little effect upon the burning time. This pressure limit seems to occur at approximately 2.2 MPa for 535 and 835 μm diameter agglomerates and 1.5 MPa for 1000 μm diameter agglomerates. Above these limits the burning time remains relatively constant at approximately 950, 1570, and 1830 ms for 535, 835, and 1000 μm diameter agglomerates. As will be discussed later, it is believed that this trend in burning time indicates that the combustion rate of aluminum transitions from being controlled by chemical kinetics to being controlled by mass diffusion as ambient pressure is increased.

Table 4.3 Effects of ambient pressure on the burning time of 535, 835, and 1000 μm diameter aluminum agglomerates.

Pressure, MPa	Burning Time, ms		
	535 μm DIA	835 μm DIA	1000 μm DIA
0.1	1338	1860 $\pm$ 95	2280 $\pm$ 45
0.5	1248	1665 $\pm$ 100	1975 $\pm$ 125
0.8	1080	1665 $\pm$ 130	1875 $\pm$ 115
1.5	1005	1675 $\pm$ 120	1840 $\pm$ 110
2.2	950	1560 $\pm$ 140	1830 $\pm$ 70
3.0	949	1575 $\pm$ 140	1790 $\pm$ 110
3.6	959	1590 $\pm$ 80	1815 $\pm$ 40
4.3	895	1555 $\pm$ 85	1865 $\pm$ 60

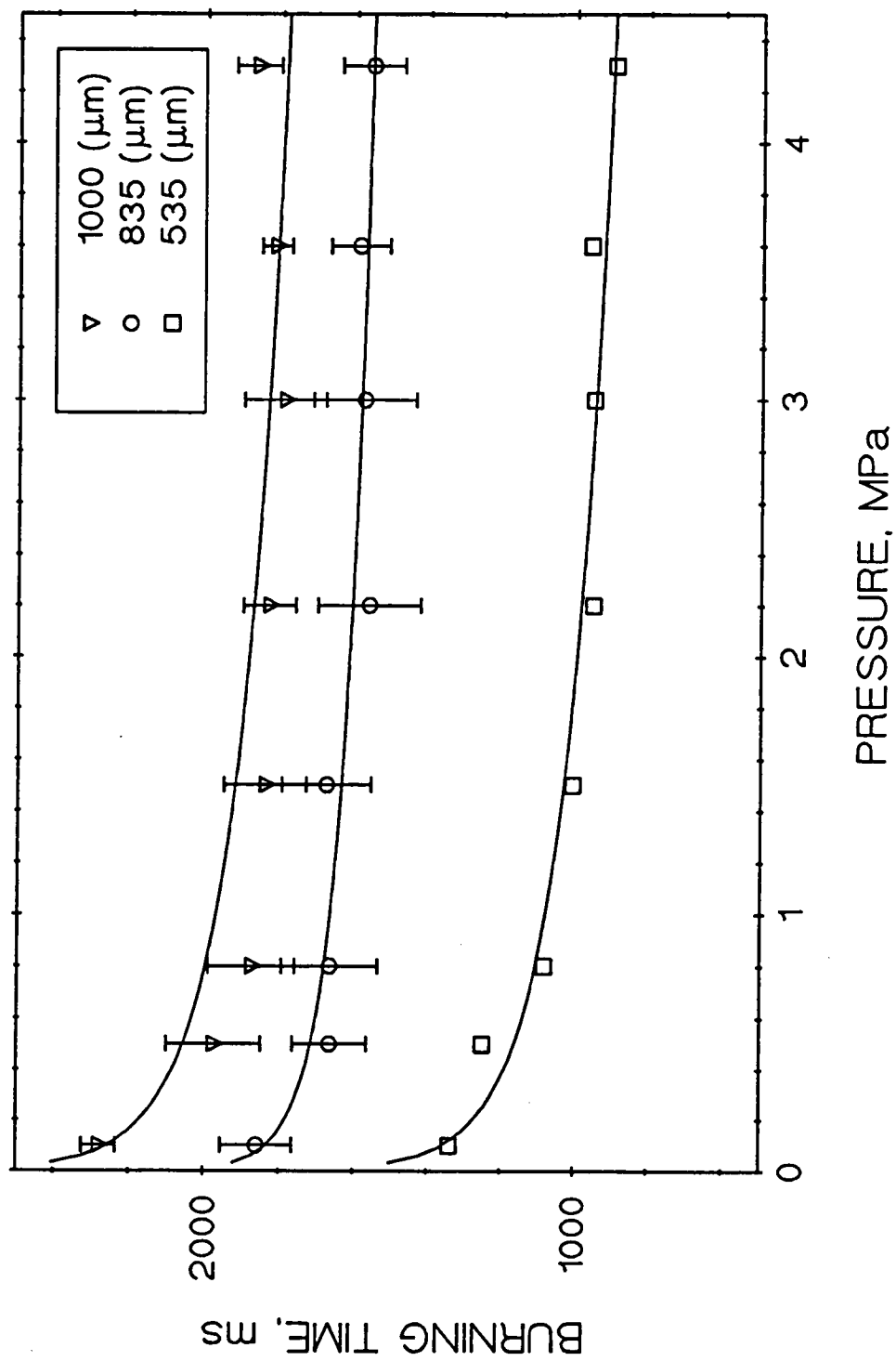


Figure 4.7 Effects of ambient pressure on the burning time of aluminum agglomerates with initial average diameters of 535, 835, and 1000 μm .

4.3.2 EFFECTS OF INITIAL DIAMETER

Both the ignition delay time and the burning time were found to increase with increasing agglomerate diameter as should be expected. Since ignition of the aluminum agglomerates is dependent upon melting of the oxide layer surrounding the agglomerate, it is reasonable to expect that an increase in diameter would result in a longer ignition delay time as a larger agglomerate would require a greater amount of energy to heat it to the melting point of the oxide. Similarly, it is reasonable to expect that the larger mass of aluminum in a larger agglomerate would require more time to be consumed.

At each pressure, the ignition delay time and burning time were fit to a rate equation of the form $t = kd_0^N$. Table 4.4 gives the values of N for both ignition delay time and burning time at each pressure obtained from this analysis. For ignition delay time, the value of N was found to range from 0.713 to 1.66 with an average value of 1.28. No definite trend is apparent; however, the higher values of N seem to correspond more to lower pressures. The average value of N from this study corresponds precisely to the value of 1.28 ± 0.18 found by Wong and Turns in their study of the ignition process of aluminum agglomerates formed during the combustion of aluminum-based slurry droplets at atmospheric pressures [19].

For the burning time, the value of N was found to range from 0.713 to 1.18 with an average value of 0.964. The value of N was found to increase with increasing pressure.

Table 4.4 Exponent values obtained by curve-fitting ignition delay time and burning time to a rate equation of the form $t = k_0^N$.

Pressure, MPa	Value of Exponent, N	
	Ignition Delay Time	Burning Time
0.1	1.66	0.825
0.5	1.49	0.713
0.8	1.65	0.892
1.5	1.38	0.993
2.2	0.904	1.05
3.0	0.713	1.03
3.6	1.21	1.03
4.3	1.20	1.18

The burning time obviously does not correspond to the "d²-law" but, instead, seems to be proportional to the initial diameter of the aluminum agglomerate. As Friedman and Maček [9] point out, this is an indication that the diffusion of the aluminum vapor is limited by the presence of molten oxide film which corresponds to experimental observations from the present study. These results, however, do not correspond to those found in previous studies. Studies of the combustion of single aluminum particles at atmospheric pressure [9, 16] have shown the value of N to vary from one to two with the majority of the data lying closer to two. In their study of the combustion of aluminum agglomerates at ambient pressure, Turns, Wong, and Ryba [5] found the value of N as 1.77 ± 0.20 .

4.3.3 EFFECTS OF LASER POWER AND DURATION

Several experiments were conducted at various pressures to determine the effects of laser power and duration on the ignition delay time and burning time of the aluminum agglomerates. There was found to exist a lower limit dependent upon both laser power and duration below which ignition would not occur. For example, for 1000 μm diameter agglomerates at an ambient pressure of 0.5 MPa, a laser power of 32.63 W and a pulse duration of 300 ms was found to be insufficient to ignite the agglomerate. However, increasing either the laser power to 34.96 W or the laser pulse duration to 400 ms was found to result in ignition of the aluminum agglomerates.

Large variances in laser power were not attempted as the primary objective was to determine if small variations in laser power would significantly affect the experimental results. Within the range of laser power tested, 30.30 to 37.29 W, no effect upon ignition delay time could be discerned as the values remained within the range of the experimental uncertainty. Likewise, laser power was found to have no discernable effect upon the burning time of the aluminum agglomerates. Experiments were conducted using 1000 μm diameter agglomerates at 2.2 MPa with laser pulse duration of 1000 ms. The burning time of these agglomerates were found to be of the same order as those ignited by a 300 ms laser pulse. In addition, some of the agglomerates failed to burn to completion just as with a 300 ms laser pulse. Thus, it would appear as though laser pulse duration has no effect upon burning time.

4.4 DETERMINATION OF RATE CONTROLLING FACTORS

Results obtained from the current study of the combustion characteristics of aluminum agglomerates were used in an attempt to determine what factors control the burning rate of the agglomerates. The effects of both ambient pressure and initial diameter on the burning time of the agglomerates were used in this analysis.

Power-law analysis of the burning time for the aluminum agglomerates indicated that the burning time is directly proportional to the initial diameter of the agglomerate at all pressures. Such a relation is typically indicative of a reaction which is controlled by reactions occurring at the droplet surface as a result of the

molten oxide layer inhibiting the diffusion of aluminum vapor. Examination of the combustion residue seems to support this conclusion.

However, previous studies on the combustion of single aluminum particles at atmospheric pressure [9, 16] have led to values of the exponent in the rate equation which were widely scattered from one to two where an exponent of two is indicative of pure vapor-phase diffusion. Turns, Wong, and Ryba's [5] study of the combustion of aluminum agglomerates resulted in an exponent value of 1.77 ± 0.20 . Obviously, there is a large amount of discrepancy in the reported values of the burning rate exponent resulting in perhaps an even larger discrepancy in the predicted mode of combustion.

As pointed out by Prentice [17], the use of rate equations to determine the controlling factors in the combustion process of aluminum is therefore rather pointless. His own experiments produced a wide range of values for the rate equation exponent. In some cases the results made sense. For example, at low oxygen concentrations large amounts of oxide were observed to accumulate on the droplet surface and the exponent value was found to be 1.3. As the oxygen concentration was increased less oxide accumulated on the droplet surface and the exponent approached two. Under some conditions, however, the burning droplet was found to have large amounts of accumulated oxide on the surface and to fragment violently prematurely ending the combustion event. The exponent value for these experiments was found to be two which, as mentioned above, is indicative of a slow burnout due to vapor-phase diffusion controlled combustion.

Prentice found the value of the exponent to be dependent upon the oxygen concentration and oxidizer type but to be independent of the amount of oxide accumulated upon the droplet surface. This observation directly contradicts the assumption that an exponent of one indicates that diffusion is inhibited by the presence of a molten oxide layer whereas a value of two indicates that diffusion is uninhibited by the presence of an oxide layer.

As Mulcahy and Smith [7] have pointed out, a reaction which is controlled by chemical kinetics will show a decrease in burning time as pressure is increased. In addition, the burning time is proportional to the initial diameter of the droplet. When the reaction is diffusion controlled the burning time is proportional to the square of the initial diameter.

Analysis of the results obtained in the current study seem to indicate that the reaction rate is kinetics controlled at low pressures. As pressure is further increased, however, the reaction seems to transition to a more diffusion-controlled process. Increasing pressure was found to result in a decrease in the burning time up to approximately 1.5 MPa for 1000 μm diameter agglomerates and 2.2 MPa for 535 and 835 μm diameter agglomerates. However, as the pressure nears these limits, the resulting decrease in burning time becomes smaller. Above the pressure limit, the burning time is unaffected by an increase in pressure suggesting the process is diffusion controlled. However, the burning time was found to be proportional to the initial diameter whereas the burning time of a diffusion-controlled reaction is proportional to the square of the initial diameter. It is expected that this low effect of

initial diameter is the result of the presence of condensed oxide on the droplet surface which inhibits the diffusion of aluminum vapor and leads to incomplete combustion of the aluminum agglomerates.

CHAPTER 5

CONCLUSIONS

The purpose of this investigation was to observe the ignition and combustion processes of laser-ignited aluminum agglomerates. Special attention was focused on determining the effects of ambient pressure and initial diameter on the ignition delay time and burning time. In addition it was hoped to determine the rate controlling factors for the combustion of the aluminum agglomerates.

It was determined that ignition of the aluminum agglomerate depends upon the melting of the oxide layer surrounding each of the individual particles which constitute the agglomerate. The ignition temperature was found to be approximately 2300 K which corresponds to the melting temperature of Al_2O_3 . Once enough energy has been supplied to the agglomerate by the laser to heat the agglomerate to this temperature, the oxide layer surrounding each particle melts allowing the particles to coalesce into a single droplet of molten aluminum. As this occurs, the oxide layer cracks in several areas allowing aluminum vapor to diffuse through and react with oxygen from the surroundings. The reaction area quickly spreads to envelop the entire droplet thus igniting the droplet.

Once ignition has occurred, the droplet undergoes full-fledged, self-sustained combustion. The flame region was found to be separated from the droplet surface seemingly confirming that the combustion of aluminum occurs in the vapor-phase. The flame structure was found to consist of an inner bright white region surrounded

by a concentric blue-green envelope indicative of the presence of AlO. The presence of AlO indicates that the reaction consists of multiple steps which contradicts the assumption generally made in analytical models that the reaction is a one-step irreversible process of the form $Al_{(s)} + O_2 \rightarrow Al_2O_3$. A plume of oxide smoke is convected upwards from the flame region and condenses upon the SiC fiber and the top of the combustion chamber.

The self-sustained combustion stage was found to consist of three distinct periods. The droplet first undergoes a period of turbulent combustion during which it shifts position from being centered on the supporting SiC fiber to being off-set to one side and moves slightly higher on the fiber. It is suggested that this phenomenon is caused by either surface tension or consumption of the SiC fiber. During the turbulent period the flame was observed to wave and flicker, and, at elevated pressures, small burning particles were occasionally ejected from the droplet. This turbulent period in the combustion process was not observed in experiments conducted at atmospheric pressure.

The turbulent period is followed by a period of steady burning characterized by the droplet being surrounded by a relatively stationary flame. During the period of steady combustion, the diameter of the agglomerate is steadily decreased and the flame stand-off ratio remains relatively constant. At low pressures this period of steady combustion accounts for the majority of the burning time; however, as pressure is increased, the length of the steady combustion period decreases until, at and above 3.6 MPa, it was observed to last for only a brief period of time.

At the end of the period of steady burning, the flame collapses to the surface of the droplet. The resulting period of surface burning was found to be relatively short at low pressures. As pressure is increased, however, the length of the period was found to increase until, at and above 3.6 MPa, it accounts for the majority of the burning time.

Combustion residue from experiments conducted at atmospheric pressure were found to consist of hollow spheres of oxide with a rough outer surface and a smooth inner surface coated with unburned aluminum. These hollow spheres did not appear to be the result of ballooning of the oxide cap as observed by Friedman and Maček [9,10] and Drew, Gordon, and Knipe [13] since no indication of a bilobal structure was observed during the combustion process in the cinematographic records. Also, their experiments were conducted in wet environments whereas experiments conducted in dry environments, such as the present study, have not been observed to produce the hollow spheres. The hollow spheres actually seem more indicative of the model presented by Bartlett, Ong, Fassell, and Papp [12] in which the molten oxide layer is inflated by the vapor pressure of the aluminum.

Residues from experiments conducted at elevated pressures showed a much different geometry consisting of a small core of unburned aluminum surrounded by thick layer of cream-colored oxide. Small protrusions of unburned aluminum were occasionally found on the surface of the residue particle. The general structure of the residue seems to suggest the model proposed for the combustion of aluminum agglomerates by Turns, Wong, and Ryba [5]. In their model an oxide cap is formed

on the droplet surface which grows in size during the combustion process due to the accumulation of oxide convected inward from the flame region. It is believed that in the current study, these oxide caps grow to completely surround the droplet leading to flame extinction and incomplete combustion.

Both the ignition delay time and burning time were found to be dependent upon the ambient pressure. As the ambient pressure approaches zero, both the ignition delay time and burning time were found to approach infinity. Increasing pressure initially resulted in a dramatic decrease in both; however, eventually a limit was reached beyond which the ignition delay time and burning time remained relatively constant despite further increases in pressure. This limit is felt to indicate a balance between an increase in burning rate due to an increase in the diffusion rate of oxidizer and a decrease in burning rate due to increased heat loss to the surroundings.

The ignition delay time was found to be proportional to the initial diameter raised to the power of 1.28 which is precisely the result obtained by Wong and Turns in their study of the ignition of aluminum agglomerates [19]. However, the burning time was found to be directly proportional to the initial diameter of the agglomerate whereas previous studies [5, 9, 16, 17] have shown the burning rate to be proportional to the diameter raised to some power ranging widely from one to two.

Since the burning time of the aluminum agglomerates was found to initially decrease and then remain essentially constant as the ambient pressure was increased, this study seems to indicate that the combustion of aluminum transitions from a kinetics-controlled reaction to a reaction controlled by mass diffusion. The low order

of the diameter effect upon the burning time is suspected to be a direct consequence of the presence of a layer of molten oxide which inhibits the diffusion of aluminum vapor to the flame front leading to incomplete combustion of the agglomerates.

This study does indicate that high ambient pressure and small initial agglomerate size are the key factors in obtaining complete combustion. The use of slurry fuels should be restricted to situations where these two criteria can be assured. Otherwise, incomplete combustion of the aluminum will result in poor efficiency in the combustor and expansion nozzle. In addition, the unburned aluminum particles and large oxide residue particles could result in serious erosion and wear of the combustion chamber and expansion nozzle.

REFERENCES

REFERENCES

- 1 Miyasaka, K. and Law, C.K., (1980), "Combustion and Agglomeration of Coal-Oil Mixtures in Furnace Environments," *Combustion Science and Technology*, Vol. 24, pp. 71-82.
- 2 Szekely, Jr., G.A. and Faeth, G.M., (1982a), "Combustion Properties of Carbon Slurry Drops," *AIAA Journal*, Vol. 20, pp. 422-429.
- 3 Szekely, Jr., G.A. and Faeth, G.M. (1982b), "Reaction of Carbon-Black Slurry Agglomerates in Combustion Gases," *Nineteenth Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, p. 1077-1085.
- 4 Szekely, Jr., G.A., Turns, S.R., and Faeth, G.M., (1984), "Effects of Carbon-Black Properties on the Combustion of Carbon-Black Slurry Agglomerates," *Combustion and Flame*, Vol 58, pp. 31-43.
- 5 Turns, S.R., Wong, S.-C., and Ryba, E., (1987), "Combustion of Aluminum-Based Slurry Agglomerates," *Combustion Science and Technology*, Vol. 54, pp. 299-318.
- 6 Meftah, K., (1992), "Ignition and Combustion Phenomena of Laser-Ignited Aluminum Agglomerates at Elevated Pressures," Master's Thesis, University of Tennessee, Knoxville.
- 7 Mulcahy, M.F.R. and Smith, I.W., (1969), "Kinetics of Combustion of Pulverized Fuel: A Review of Theory and Experiment," *Reviews of Pure and Applied Chemistry*, Vol. 19, pp. 81-107.
- 8 Davis, A., (1962), "Solid Propellants: The Combustion of Particles of Metal Ingredients," *Second Rocket Propulsion Symposium*, Cranfield, pp. 359-367.
- 9 Friedman, R. and Maček, A., (1962), "Ignition and Combustion of Aluminum Particles in Hot Ambient Gases," *Combustion and Flame*, Vol. 6, pp. 9-20.
- 10 Friedman, R. and Maček, A., (1963), "Combustion Studies of Single Aluminum Particles," *Ninth Symposium (International) on Combustion*, Academic Press, New York, pp. 703-712.

- 11 Brzustowski, T.A. and Glassman, I., (1964), "Vapor-Phase Diffusion Flames in the Combustion of Magnesium and Aluminum: I. Analytical Developments," *Heterogeneous Combustion*, AIAA Progress in Astronautics and Aeronautics, Vol. 15, Academic Press, New York, pp. 75-115.
- 12 Bartlett, R.W., Ong, Jr., J.N., Fassell, Jr., W.M., and Papp, C.A., (1963), "Estimating Aluminum Particle Combustion Kinetics," *Combustion and Flame*, Vol. 7, pp. 227-234.
- 13 Drew, C.M., Gordon, A.S., and Knipe, R.H., (1964), "Study of Quenched Aluminum Particle Combustion," *Heterogeneous Combustion*, AIAA Progress in Astronautics and Aeronautics, Vol. 15, Academic Press, New York, pp. 17-39.
- 14 Blackburn, P.E. and Gulbransen, E.A., (1960), "Aluminum Reactions With Water Vapor, Dry Oxygen, Moist Oxygen, and Moist Hydrogen Between 500 and 625°," *Journal of the Electrochemical Society*, Vol. 107, pp. 944-950.
- 15 Prentice, J.L., (1970), "Combustion of Pulse-Heated Single Particles of Aluminum and Beryllium," *Combustion Science and Technology*, Vol. 1, pp. 385-398.
- 16 Wilson, Jr., R.P. and Williams, F.A., (1970), "Experimental Study of the Combustion of Single Aluminum Particles in O₂/Ar," *Thirteenth Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, pp. 833-844.
- 17 Prentice, J.L., (1972), "Combustion of Laser-Ignited Aluminum Droplets in Wet and Dry Oxidizers," Paper # 74-146, AIAA Twelfth Aerospace Sciences Meeting.
- 18 Law, C.K., (1973), "A Simplified Model for the Vapor-Phase Combustion of Metal Particles," *Combustion Science and Technology*, Vol. 7, pp. 197-212.
- 19 Wong, S.-C. and Turns, S.R., (1987), "Ignition of Aluminum Slurry Droplets," *Combustion Science and Technology*, Vol. 52, pp. 221-242.
- 20 Zaidi, W.H., (1991), "Ignition and Combustion Phenomena of Laser-Ignited Boron Agglomerates at Elevated Pressures," Master's Thesis, University of Tennessee, Knoxville.
- 21 Rautenberg, T.H., and Johnson, P.D., (1960), "Light Production in the Oxygen-Aluminum Reaction," *Journal of the Optical Society of America*, Vol. 50, pp. 602-606.

22 Brzustowski, T.A. and Glassman, I., (1964), "Spectroscopic Investigation of Metal Combustion," *Heterogeneous Combustion*, AIAA Progress in Astronautics and Aeronautics, Vol. 15, Academic Press, New York, pp. 41-73.

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