

COMBUSTION OF LIQUID FUELS AND POLLUTANT FORMATION: A REVIEW

PART I. LIQUID FUEL COMBUSTION PROCESS

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Abstract—In Part I of a two-part contribution, recent advances in understanding the combustion process of liquid fuels are reviewed. The main objective of the review is to provide an introduction to the subject and to stimulate concerns on the issues related with the liquid fuel combustion processes.

INTRODUCTION

The combustion of sprays of liquid fuels is of significant practical importance and currently accounts for a considerable share of the world energy use. Although the propagation of flames supported by gaseous fuels has been studied extensively over the past two centuries, investigations into the combustion of liquid fuel spray have been afforded much less attention. In recent years the wide use of spray injection has found applications in many practical systems ranging from furnaces to gas turbines and rocket engines. These two latter applications provided an impetus for a deeper understanding of the processes which govern the combustion of liquid fuel sprays.

Recently, the awareness of producers and consumers alike has been sharpened not only to the finite nature of energy resources and the pressing need to conserve them but also to the damaging effects on the environment when these resources are consumed with insufficient care. Efficient handling and combustion of fuels are therefore essential from the aspects of energy conservation and environmental protection. There is increasing agreement that safeguards should be based on logically applied knowledge and judgement rather than emotional pressures. In general terms, the design and operation of equipment for fuel-handling and combustion have a greater influence on combustion-generated emissions than the nature of the fuel itself. Nevertheless, significant steps can be taken to control both the components and the contaminants of fuel to minimize problems from emissions [Levy(1983)].

In this two-part paper, recent advances in studies on the combustion of liquid fuels and pollutant formation

from the process are reviewed. The purpose of the review is to provide an introduction to the subject and to stimulate concerns on issues related to combustion-generated air pollutants complemented with a bibliography pertinent to the topics covered.

COMBUSTION OF LIQUID FUELS

Liquid fuels are injected into combustion chambers through atomizers to facilitate the disintegration of the liquid into a spray of droplets. When the fuel vapor is ignited in the vicinity of the droplets this results in an increased rate of evaporation. Reaction zones will be formed partly surrounding each droplet or in a region where fuel/air ratios are within limits of inflammability. In most practical systems the combustion process is very complicated, and the sequence of physical and chemical processes involved can be represented by the scheme shown in Fig 1.

To obtain a full description of the processes involved in spray combustion it is necessary to have complete knowledge of the mechanism of combustion of the individual droplets, any interaction between the droplets, and the statistical description of the droplets with regard to size and spatial distribution. It is very difficult to obtain detailed information on the mechanism or rate of burning by direct studies of spray combustion. Due to these experimental difficulties a number of experimental techniques have been developed to study the combustion of isolated single droplets under carefully controlled conditions. Theoretical analyses and mathematical models are also greatly assisted if only single isolated droplets are considered.

To apply results from single droplet studies to multi-

droplet problem of spray combustion is not an easy matter and remains a challenging problem fundamental to spray combustion.

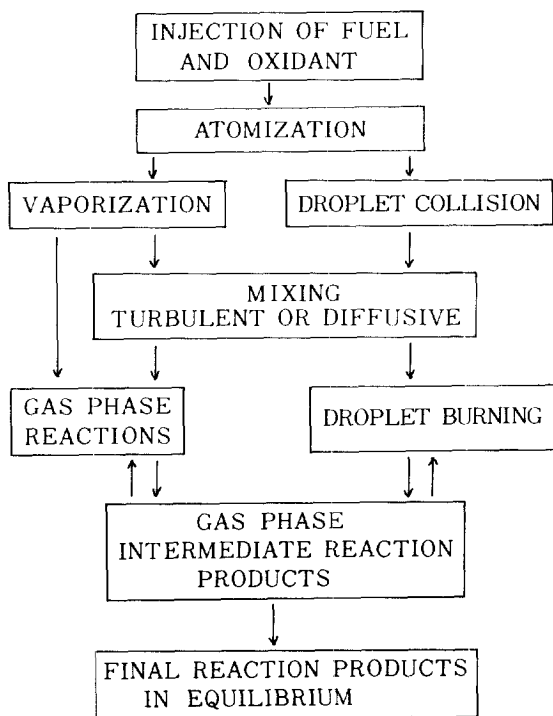


Fig. 1. Combustion processes in a liquid fuel combustion chamber.

2.1 Atomization

The injection of liquid fuel into combustion chambers through atomizers facilitates the disintegration of the liquid into a spray of droplets to enhance the transfer processes and the uniform distribution of the liquid over large areas. Most practical liquid fuel spray have a size distribution over a wide range of droplet sizes with a mean droplet diameter between 75 and 130 μm [Bolle and Moureau(1982)].

The main processes which make up the atomization of the spray are [Stambuleanu(1976)]:

- (i) development of liquid stream;
- (ii) disintegration of the stream by collision and friction;
- (iii) disintegration caused by surface tension;
- (iv) disintegration caused by turbulent motion;
- (v) coalescence and breaking up of droplets by collision.

The atomization process proceeds stepwise, and each step is a time dependent process. The resulting droplets may be unstable in the sense that they may coalesce or break up. This instability in the formation process of the droplets may become the source of oscillation in the

combustion chamber and may possibly lead to the flame instability [Petela and Tysowska(1982)].

Comprehensive treatments on the analyses and design of atomization system can be found in several monographs and reviews [Beér and Chigier(1972), Stambuleanu(1976), Chigier(1976), Jones(1977), Reed(1978), Lefebvre(1980), Singer(1981)].

2.2. Vaporization

The combustion of liquid fuels takes place in the gas phase, and is limited by the rate of mass transfer from the liquid to the vapor phase. The rate of mass transfer is proportional to the interfacial area, which is obtained through atomization process. In order to sustain high combustion intensities and obtain reasonable combustor residence times, rapid vaporization is an absolute necessity.

Fuel injected into a furnace is heated to reduce its viscosity enough to permit adequate atomization. The fuel droplets must undergo further heating until they attain their initial boiling point after which they dissipate most of the heat received as latent heat of vaporization. Because of the preferential vaporization of light components, the boiling point of the fuel will increase, requiring sensible heat to be conducted into the droplet such that the liquid may sustain continued vaporization. Due to the increased liquid temperature and reduced surface area the mass rate of vaporization for a particular droplet is reduced. For a poly-dispersed spray, the small droplets will vaporize completely much sooner than the large droplets. Thus, even though all droplets diminish in size, the mean droplet size and the overall rate of vaporization will initially increase and later diminish.

Mass and energy transport in the gas phase of a vaporizing droplet is influenced by its velocity and the environment through which it travels. This interaction of the droplet with its environment is system specific; the only generalization on the vaporization rate of the droplet is the increased transport rates that result from boundary layer thinning by the velocity of the droplet and its environment.

In the liquid phase there is less certainty as to the transport processes taking place. Whether circulation within the droplet is important depends upon particle size, viscosity, velocity and vaporization rate, and the importance of liquid phase reactions is subject to fuel composition and the contribution of radiation to the heat transfer to the droplet.

2.3 Mixing

Intimate contact between the vaporized fuel and oxidizing gas must be accomplished for combustion to proceed, and thus adequate space must be provided in the combustion chamber for mixing. This mixing process is usually the rate limiting step of heterogeneous combustion and involves a complex interaction of molecular dif-

fusion and the flow field in the combustor. Macroscale mixing is accomplished by the turbulent fluid motion whereas microscale mixing is governed by molecular diffusion.

2.4 Chemical Reaction

The energy released by the reaction of fuel and oxidant is the desired product of a combustor. The reaction takes place in the gas phase creating combustion products that may be innocuous or undesirable. In the liquid phase polymerization and cracking reactions may occur resulting in the formation of coke.

Hydrocarbon combustion is a very complex process. In spite of the fact that the theoretical and experimental investigations in this field are fairly numerous [Westley(1976), Kaufman(1983)], the complex character of the problem has prevented the disclosure of the true mechanism of the reaction. A characteristic specific to the mechanism of combustion of hydrocarbon with branched reactions is that branching is due not to the reactions of radicals with fuel or oxygen molecules, but to the presence of a relatively stable intermediate product formed by a non-branching process and which is able to react in the system, to give either products or radicals, which can initiate the primary chain leading to the formation of more intermediates. This process is known as degenerate branching [Semenov(1935)].

The oxidative mechanisms that dominate the combustion process would ideally result in the formation of carbon dioxide and water only. However, the additional pyrolysis reactions that a high molecular weight hydrocarbon undergoes inadequate mixing of fuel and oxidant, and the inclusion of nitrogen, sulfur, and other materials in the fuel results in the formation of pollutants of which those of current concern are:

- (i) carbonaceous particulates;
- (ii) unburnt hydrocarbons;
- (iii) polycyclic aromatic hydrocarbons;
- (iv) carbon monoxide;
- (v) sulfur oxides;
- (vi) nitrogen oxides;
- (vii) volatile inorganics, *e.g.* lead and arsenic compounds.

The liquid phase temperature of a fuel droplet is regulated by vaporization of material from the surface. For heavy fuel oils, the vaporization occurs at temperatures high enough to cause heavy fuel components to undergo pyrolysis reactions. Among the pyrolysis products, carbonized residues, which may form as cenospheres, are of concern [Hottel, *et al*(1955)]. In an oil-fired utility boiler plant carbonized residues, possibly with a certain amount of soot in combination with oxides of sulfur from the sulfur in the fuel, give rise to the problem of acid smuts [Nettleton (1979)].

COMBUSTION OF LIQUID FUEL DROPLETS

An essential prerequisite for any understanding of spray combustion and its application to the design of an efficient combustion system is knowledge of the combustion of droplets. The topic has been the subject of a number of reviews [Williams(1973). Krier and Foo (1973), Faeth(1977), Law(1982)].

3.1 Historical Outline

The science of droplets began when Archimedes of Syracuse (287-212 B.C.) established the relationship between the volume and the radius of a sphere [Struik (1967)]. More than 2,000 years had to pass, however, before light was shed on the science of flow around droplets. In 1851, Stokes succeeded in calculating the viscous drag of a slow-moving sphere. Soon afterward, the thermodynamic behavior of capillary equilibrium was elaborated by Sir W. Thomson (later Lord Kelvin) in 1869, Gibbs in 1878, and Helmholtz in 1886, leading to a formula for the size of the smallest droplet that is thermodynamically stable in a given environment. The first treatises bearing on the evaporation of drops was Maxwell's analysis(1878) of diffusion in dense mercury in a vacuum. The latter line of research was later pursued by Langmuir (1913, 1916, 1932) and by Knudsen(1915), who laid down the concepts describing the interaction of single gaseous molecules with liquid or solid surface.

In the 1930's great advances were made in the theory of heat and mass transfer. The basic analogy between these processes was pointed out by Schmidt (1929).

Aerodynamic concepts, like boundary layer and similarity theory, began to be exploited for the prediction of heat and mass transfer to spheres in streaming media, as first done by Frössling(1938) and later by Ranz and Marshall(1952). The study performed until the mid-1950's on evaporation and growth of droplets in continuum environments are summed up in a treatise by Fuchs(1959).

The foundation of the present understanding of droplet combustion was prepared in the late nineteenth century during studies on low temperature evaporation of droplets [Maxwell(1878)]. However, analogies with the coal particle combustion during the 1920's [Nusselt (1924)] led to the concept of heterogeneous combustion and it was not until extensive studies in the 1940's that the present day model of diffusion flame combustion was accepted [Williams(1973)].

The basic droplet combustion model was formulated by Varshavskii(1945), Godsave(1953), Spalding(1953), Goldsmith and Penner(1945), and Wise, *et al.*(1955) for an isolated, pure-component droplet burning in a quiescent oxidizing medium. This model has since been named the " d^2 -law" because it predicts that the square

of the droplet diameter (*i.e.* the surface area) decreases linearly with time. In subsequent years research concentrated on verifying this model and extending it to include other effects.

In the past few years there has been increasing interest on multicomponent fuel combustion, which necessitates detailed investigation of liquid-phase transport processes, and on what can be termed as spray effects, which account for the fact that in realistic situations the droplet is placed in the spray interior; therefore the environment it experiences can be quite different from that of the isolated droplet [Law(1982)].

The recent advances in computational and experimental facilities brought about an enhanced level of understanding of the droplet combustion process.

3.2 Spherico-Symmetric Quasi-Steady State Models

The basis of most droplet combustion theories is the spherico-symmetric model developed by Godsave (1951, 1953) and Spalding(1953). All subsequent studies have only produced minor refinement. Godsave obtained the following expression for the mass rate of vaporization of a single component droplet under steady state conditions with spherical symmetry:

$$\dot{m} = \frac{2\pi kd \ln[1 + C_p(T_f - T_s)/(L - R_a)]}{C_p(1 - d/d_f)} \quad (1)$$

where \dot{m} = mass rate of vaporization; k = thermal conductivity; d = droplet diameter; C_p = specific heat capacity at constant pressure of gas phase; T_f = flame temperature; T_s = temperature at droplet surface; L = specific latent heat of vaporization; R_a = absorbed radiant energy per unit mass vaporized; d_f = flame sheet diameter. No consideration of flame position, d_f from gas phase composition is included in Eq. (1) and thus must be found experimentally. However, for the case of a droplet vaporizing in a hot atmosphere without combustion, $d_f \rightarrow \infty$ and the parenthetical term in the denominator may be ignored.

Spalding(1953) postulated that combustion occurs in a boundary layer surrounding the droplet, and that the thickness of the layer may be predicted by analogy with heat and mass transfer coefficients. By solving the transport equations for heat and oxidant surrounding the droplet, Spalding obtained expressions for T_f , d_f and \dot{m} , the last being

$$\dot{m} = \frac{2\pi kd}{C_p} \ln(1 + B) \quad (2)$$

where B denotes a transfer number,

$$B = \frac{Q_{\infty} Y_{\infty}}{L\mu} + C_p \left(\frac{T_{\infty} - T_s}{L} \right) \quad (3)$$

Since the only time dependent term of Eqs. (1) and (2) is d , by expressing \dot{m} in terms of d , a relationship for

the temporal size variation is found:

$$d_0^2 - d^2 = \lambda t \quad (4)$$

In Eq. (4), λ is termed the vaporization rate constant, and is related to the mass rate of vaporization, \dot{m} , by the equation

$$\lambda = \frac{4\dot{m}}{\pi\rho_l d} \quad (5)$$

3.3. Non-Steady State Models

While Eq. (4) has been found adequate for simple fuels burning at near atmospheric pressure, the vaporizing droplets are actually in a transient state which is evidenced by changes in the ratio of flame to droplet diameters that, according to the quasi-steady analysis of Goldsmith and Penner(1954), should be constant. Kotake and Okazaki(1969) undertook a numerical analysis of these effects, and Krier and Wronkiewicz (1972) were able to express the d^2 -law, Eq. (4) in terms of d_f/d . Rosner and Chang(1973) studied the non-steady effects of droplet vaporization at near critical conditions. Crespo and Linan(1975) performed an asymptotic analysis of the non-steady effects in the gas phase. Hubbard, *et al.*(1975) considered in a numerical study the effects of transients and variable properties. The results showed that temporal storage of mass species, energy, and radial pressure variations in the gas phase to be negligible, the initial transient behaviour being due to sensible heat accumulation in the droplet and related variations in the surface pressure. The transient evaporation is also claimed to be independent of the initial size, but to follow d^2 -law behaviour. Law(1976) also found liquid phase heating to be the primary cause of transients following a lumped parameter analysis of the droplet. Wise and Abolow(1957) derived temperature distributions within the droplet as a function of $\lambda/8\alpha$ (α = thermal diffusivity) and found that liquid phase temperature profiles to be significant if $\lambda/8\alpha$ is greater than unity.

3.4 Influence of Convection

Generally, it is known that convection increases the vaporization rate, that the thermal flux is not uniform over the droplet surface, that a boundary layer of finite thickness exists, and that flame shape is not spherical and that the flame may even become detached and burning the wake of the droplet.

The usual method of considering the effect of convection is to assume similarity to solid sphere models and empirically fit constants to a correction factor for the quiescent case [Frössling(1938), Ranz and Marshall (1952)];

$$\dot{m}_c = \dot{m}(1 + a\text{Re}^b \text{Sc}^c) \quad (6)$$

where the constants a, b and c are taken to be $a = 0.3$, $b = 0.5$, and $c = 1/3$ [Ranz and Marshall(1952)]. Further

discussions on the physical meaning of the exponents b and c are presented by Ruckenstein(1981).

There have been a number of analytical studies on the boundary layer convective evaporation and combustion of droplets. Examples are the forced convective evaporation by Prakash and Sirignano(1978), the forced convective burning by Krishnamurthy and Williams (1974), the natural convective burning by Wu, *et al.*(1982), and the mixed, forced and free- convective gasification, ignition, steady burning and extinction by Fernandez-Pello and Law(1983).

3.5 Interaction Between Droplets

Studies on interference between droplets during combustion were initiated by Rex, *et al.*(1956), who experimentally determined the burning rates of suspended droplet arrays arranged in some simple geometric configurations. Their results showed that within experimental scatter the d^2 -law established for isolated droplet burning still holds. For the simplest configuration investigated, two horizontally aligned droplets, they observed that the burning constant exhibited a local maximum when it was plotted against the initial droplet separation distance. This was attributed to the competing factors of reduced oxygen supply and heat loss.

There have been several experimental studies [Nuruzzaman, *et al.*(1971), Sangiovanni and Kesten(1977a), Labowsky and Sangiovanni(1980)] on the interference effects. Also a number of theoretical analysis [Labowsky(1976), Ray and Davis(1980), Bellan and Cuffel(1982)] for the combustion of interacting droplets have been developed providing valuable information for the understanding of interference effects. One of the most advanced models is the analysis of Labowsky (1976) who studied the nearest neighbor interactions on the evaporation rate of particle clouds. Labowsky used modified method of images to determine the vapor concentration field in arrays of upto nine quasi-steady state vaporizing particles. The method employed is unwieldy and does not account for unsteady behaviour. However, Labowsky empirically correlates the efficiency of evaporation for an array of N particles having interdrop spacing, S , and diameters, d :

$$\eta_A = \tanh\left\{\left(\frac{S}{d}\right)^{0.025(N-1)+0.287} \ln\left\{5.71/(N-1)^{0.562}\right\}\right\} \quad (7)$$

Ray and Davis(1980) present a generalized treatment of combined heat and mass transport between an assemblage of particles and their environment. The unsteady state diffusive transport is shown to depend upon size and positions of the particles, and the rate processes are found to differ markedly from single particle rates due to particle interactions. The expression obtained for equivalent to Eq. (7) is

$$\eta_A = \frac{2 - \frac{N}{(N-1)} \frac{S}{d} - \frac{1}{(N-1)} \sum_{n=1}^{N-2} \frac{S}{S_n}}{N \left\{ \frac{d}{S} - \frac{1}{(N-1)} \frac{S}{d} - \frac{1}{(N-1)} \sum_{n=1}^{N-2} \frac{S}{S_n} \right\}} \quad (8)$$

where S_n is the distance of the n th particle to the end of the array.

3.6 Finite Rate Kinetics

The discussions of the previous sections are limited to either pure vaporization in which chemical reaction rate is everywhere vanishingly slow compared with the diffusion rate, or to flame-sheet combustion in which reaction is again frozen everywhere except at the infinitesimally-thin flame where the reaction rate is infinitely fast

There are, however, certain phenomena in which the chemical reaction rate can be of the same order as the diffusion rate in certain parts of the flow field, and therefore can only be described by allowing for finite rate kinetics. One pertinent example to droplet studies is the ignition of droplets. The ignition is a transient phenomenon involving the rapid transition from evaporation and chemically controlled processes to diffusional controlled near-steady-state combustion. As was discussed by Rah, *et al.*(1982) the ignition of droplets is closely related to soot and NO_x formation.

3.7 Ignition of Liquid Fuel Droplets

Although there have been numerous studies of fuel droplet combustion, the fuel droplet ignition process is still poorly understood. When a droplet is injected into a hot gas environment, it undergoes vaporization forming a fuel vapor/oxidizer mixture of combustible proportion around the droplet. Both the critical conditions necessary for ignition to occur and the physical and chemical processes involved should be studied.

The ignition delay time is defined as time to ignition measured from the time a droplet of liquid fuel is introduced into a high temperature oxidizing medium. Most of the experimental studies on liquid fuel droplet ignition are concerned with the measurements and empirical correlations on the ignition delay time [Mullins (1949, 1955), Nishiwaki(1955), Masdin and Thring (1962), El-Wakil and Abdou(1966), Faeth and Olson(1968), Wood and Rosser(1969), Sangiovanni and Kesten(1977a, 1977b), Saitoh, *et al.*(1982), Rah, *et al.*(1982)]. It has been a common practice among researchers to represent the temperature effect on the ignition delay time as an Arrhenius-type function. However, it is unlikely that this representation is very accurate owing to the subjective and indefinite nature of the criterion by which the ignition delay time is estimated. Other possible complications are discussed by Mullins(1955).

The idea of studying the droplet ignition by examining the conditions of a sudden jump in mass burning rate for changes in parameters was initiated by Polymeropoulos and Peskin(1969). They integrated the balance equations for the steady state combustion of liquid fuel droplets in an oxidizing atmosphere for various fuel-oxygen systems numerically. They obtained a continuous description of the droplet behavior from the low mass burning rate kinetic controlled region to the high mass burning rate diffusion controlled region. Ignition conditions were obtained as a function of ambient oxygen concentration for various reaction rates, reaction orders, and stoichiometric ratios.

Sangiovanni and Kesten(1977b) solved the equations describing the vaporization and transport processes which occur during the ignition period when a single, isolated fuel droplet is entrained in a combustion gas stream. They used the ignition criterion first suggested by Faeth and Olson(1968). The criterion is based on the premise that ignition takes place when the concentration of a critical chain-branching intermediate chemical species(unidentified) reaches a critical value in the gas phase. Although the criterion is claimed to have an advantage in terms of the simplicity of application, it is the arbitrariness of selecting an unidentified species concentration that brings about doubts on the validity of the model.

Law(1975) analyzed the quasi-steady diffusion flame structure in droplet by using the technique of large activation energy asymptotics [Williams(1971), Buckmaster and Ludford(1982)] for a one-step Arrhenius reaction in the gas phase. He produced the characteristic ignition-extinction S-shaped curve for various combustion regimes and determined the critical Damköhler numbers for ignition and extinction. Law (1978) later applied these results to the analysis of the droplet ignition process by coupling it with an analysis of droplet heating process.

The quasi-steady state theory simplifies the mathematics involved and is useful in studying the droplet burning process. However, the processes involved in fuel droplet ignition are inherently unsteady, and the quasi-steady state theory may not be appropriate for the analysis of the droplet ignition process. The problem should be treated as a "parametric sensitivity" problem of transient equations rather than a "stability" problem of the steady state solutions of balance equations. The numerical analyses of Blushenko, *et al.*(1973) and Niioka, *et al.*(1980) seem to lead to the right direction of approach by treating the system of partial differential equations, which describes the unsteady behavior of the processes.

In a study by Rah(1984), an attempt was made to analyze the problem of liquid fuel droplet ignition with

the reasoning based on the accumulated knowledge on the thermal theory of ignition of gaseous fuels [Frank-Kamenetskii(1969), Merzhanov and Averson(1971), Gray and Sherrington(1977)]. Balance equations were set up and nondimensionalized. The strategy of treating the moving phase boundary and the ignition criterion were explained. The system of partial differential equations was solved numerically and the calculation results were compared with the experimental data. The study was extended to the case of real fuel droplet. Following the line of approach taken for the case of single component droplet, it was shown that the ignition process of multicomponent real fuel droplet could be treated in a similar way.

3.8 Experimental Studies

Experimental studies of droplet combustion have employed the following methods:

- (i) suspended droplets [Godsave(1951, 1953)];
- (ii) porous sphere with liquid fed to its interior [Spalding(1953)];
- (iii) single droplets or droplet array in free flight [Chang(1941), Hottel, *et al.*(1955), Nuruzzaman, *et al.*(1971), Sangiovanni and Kesten(1977a), Hanson (1982), Rah, *et al.*(1982), Rah(1984)].

Among these methods free droplet experiments offer the advantages of small sizes, non-interference from suspension fiber, and capability of using volatile fuels. However, the method is generally more complex and delicate. Furthermore, since the droplets are not stationary, it is usually more involved to obtain detailed photography. Their free fall motion, together with their continuously diminishing size, also imply that the intensity of forced convection continuously changes.

Experiments have been performed to determine ignition times, to confirm the validity of the d^2 -law, and the variation of the burning rate with temperature, pressure, and oxygen concentration, and to obtain empirical correlations for the effects of convection.

Several techniques have been employed at minimizing or eliminating buoyancy due to the influence of gravity [Kumagai and Isoda(1957), Kumagai, *et al.*(1971), Knight and Williams (1980), Miyasaka and Law(1981), Okajima and Kumagai (1975, 1983)]. Recent interest to conduct scientific experiments on board the Space Shuttle may provide an opportunity to study droplet combustion under a convection-free and stationary environment.

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