

DROPLET COMBUSTION AND SOOT FORMATION IN MICROGRAVITY†

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1. Introduction

One of the most complex processes involved in the combustion of liquid fuels is the formation of soot. A well characterized flow field and simplified flame structure can improve considerably the understanding of soot formation processes. The simplest flame shape to analyze for a droplet is spherical with its associated one-dimensional flow field. It is a fundamental limit and the oldest and most often analyzed configuration of droplet combustion.

Spherical symmetry in the droplet burning process will arise when there is no relative motion between the droplet and ambience or uneven heating around the droplet periphery, and buoyancy effects are negligible. The flame and droplet are then concentric with each other and there is no liquid circulation within the droplet. An understanding of the effect of soot on droplet combustion should therefore benefit from this simplified configuration. Reviews by Haynes and Wagner (1981), Howard and Kausch (1981), Smith (1981), and Glassman (1988) provide a thorough account of our state of understanding on soot formation in a wide variety of flaming configurations. Soot formed during spherically symmetric droplet combustion, however, has only recently drawn attention and it appears to be one of the few aspects associated with droplet combustion which have not yet been thoroughly investigated. For this review, the broad subject of droplet combustion [e.g., Williams 1973; Faeth 1977; Law 1982; Sirignano 1983] is narrowed considerably by restricting attention specifically to soot combined with spherically symmetric droplet burning processes that are promoted by microgravity.

2. Visualization of Soot in Droplet Combustion

Figure 1a is a photograph [Jackson and Avedisian 1993] that shows the basic configuration of near-spherical symmetry that is the focus of this review, and fig. 1b is a schematic drawing. A heptane droplet is shown which is levitated by microgravity (the droplet and ambience are in free fall at nearly the same rate). The droplet is almost concentric with a spherical soot pattern or "shell" (the dark ring) and outer luminous zone. The term "spherically symmetric droplet combustion" (SSDC) that is used throughout this review refers to a situation like that shown in fig. 1 which has the outward appearance of spherical symmetry.

Visualizations of soot formation in SSDC are limited and qualitative compared to the larger body of work reported on sooting processes in droplet flames at normal gravity [e.g., Sangiovanni and Liscinsky 1984; Kadota and Hiroyasu 1984; Randolf and Law 1986]. This is due in part to the greater difficulty of creating the microgravity environment for experimentation. It is not a simple matter to do testing in microgravity because the laboratory must be physically dropped and that creates problems in design and operation. The droplet must be formed and ignited, kept near motionless during its burning, photographed with the proper lighting to visualize simultaneously the droplet, soot, and outer luminous zone, and enough time must be available to observe the entire period of burning.

3. Creating a Microgravity Environment

The purpose of creating a microgravity environment for droplet combustion is to remove the influence of buoyancy. The goal is to create a situation in which the evaporation induced (Stefan) velocity is much larger than the relative velocity between the droplet and ambience, created either by buoyancy or a forced motion. An order of magnitude estimate of the Stefan velocity can be obtained from the quasi-steady state theory for spherically symmetric droplet combustion [e.g., Williams 1973] and using the droplet diameter, D , as a characteristic length scale by convention [Law and Williams 1972].

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Williams (1980) and it was later used as a basis for a simplified analysis of the process [Jackson et al. 1992; Choi et al. 1993]. Because of the symmetric flow field and temperature gradient around the droplet, the soot particles will ideally reside in an apparent shell-like structure as shown in fig. 1, hence the name soot shell. The shell will have a finite thickness because of variations in the aggregate sizes that get trapped by this force balance. The precise composition of the soot shell is not known. However, 1) it is highly porous, 2) its shape will be spherical under ideal conditions, 3) its diameter is time dependent, and 4) it resides between the droplet and flame.

The ability to photograph the soot shell and outer luminous zone of a flame requires proper lighting. Back-lighted images can totally obscure the soot patterns if the lighting is intense enough. Simultaneous measurement of D , D_s (the mean soot shell "diameter"), and D_f can be difficult or impossible to measure if soot image intensity dominates; and, the soot aggregates and droplet may not be visible if flame luminosity dominates (e.g., as observed by Jackson et al. 1991). The finite rate of oxidation of the aggregates could, for example, place them well past the flame and lead to the primary reaction zone (the flame) being confused with the luminous zone [Randolf and Law 1986]; the flame may be better defined by a peak in a radical species like OH. Differences in visibility are most likely the result of lighting variations among the studies reported. For example, photographs of free heptane droplets burning in microgravity [Kumagai et al. 1971; Avedisian et al. 1988; Yang and Avedisian 1988] showed no soot shell. On the other hand, microgravity experiments reported by Okajima and Kumagai (1975), again for heptane, clearly revealed the shell structure for heptane (apparently the first to do so) probably because a different lighting arrangement was used. However, surprisingly, almost no commentary was offered about the soot patterns that were clearly visible in the photographs shown. Later photographs of free decane [Shaw et al. 1988a; Choi et al. 1992] and heptane [Jackson and Avedisian 1993] droplets burning in microgravity showed soot shells surrounding the droplets. Soot patterns have also been photographed for methanol/toluene mixtures [Jackson et al. 1991] and chlorinated hydrocarbon droplets [Jackson et al. 1992, 1993] again burning in microgravity.

The largest aggregates in the shell can be observed using conventional back-lighted photographs, though thermophoretic sampling [Megarides and Dobbins 1987] and light scattering techniques [e.g., Santoro et al. 1987] are more quantitative. Soot aggregates collected from microgravity droplet flames [Jackson et al. 1992] show no distinctive features compared with particles recovered in other droplet combustion configurations (e.g. Nakanishi et al. 1981; Bonczyk and Sangionvanni 1984; Dobbins and Megarides 1990). Some evidence has been reported that primary particle size is dependent on the gravitational level, being larger when formed in microgravity [Greenberg et al. 1994].

The time dependence of D_s (the shell "diameter" really has a finite thickness) reflects the temporal variation of D_f during burning. Measured values of the evolution of D_s/D are shown in fig. 2 for n-heptane [Jackson and Avedisian 1993] and are compared with measurements of the outer luminous zone, D_f/D , which is here taken to be the flame diameter. That both D_s/D and D_f/D track together, with $D_s/D < D_f/D$ as shown in figs. 1 and 2, is consistent with the fact that soot precursors form on the fuel rich side of the flame [e.g., Dobbins 1994].

5. Parametric Effects on Soot Formation in Spherical Droplet Flames

5.1 Introduction

The parameters pressure, droplet composition, ambient gas composition, initial droplet diameter and their effect on soot formation in SSDC narrows considerably the available experimental results. Very few studies have been reported on how these parameters influence soot formation in SSDC, due most likely to the greater difficulty of carrying out systematic SSDC experiments in microgravity.

5.2 Initial Droplet Diameter

The ideal spherically symmetric quasi-steady droplet burning process should not be influenced by the initial droplet diameter. However, the actual spherically symmetric burning process may not be quasi-steady (see fig. 2) because it exhibits features not accounted for in the ideal case such as soot formation.

An effect of initial diameter, D_0 , on the droplet burning rate, defined as $K = -d(D^2)/dt$, has been measured [Jackson and Avedisian 1993]. K is obtained directly from measured droplet diameters by plotting the variation of (D/D_0^2) with t/D_0^2 since from the quasi-steady theory of droplet combustion, $D^2 = D_0^2 - Kt$ [e.g., Williams 1973]. Figure 3a shows a typical variation for heptane (Jackson 1994) which is the most extensively studied hydrocarbon for microgravity droplet flames. The linearity between K and t/D_0^2 is apparent and there is a slight difference in K for the two D_0 values shown which is more clearly revealed by using a computer-based data analysis system to analyze the motion picture movie frames [Jackson 1994]. Fig. 3b shows the resulting variation of K with D_0 for heptane for the range of D_0 considered [Jackson and Avedisian 1993]. The line is drawn to suggest trends. An explanation for an effect of D_0 on K has been offered based on speculations about sooting tendencies of droplets with different initial diameters. The only available evidence for SSDC is circumstantial and based on visual comparisons of image intensity. In a different droplet configuration, experiments on monodispersed droplet streams have shown that soot formation increases as the droplet spacing decreases such that a single flame surrounds the group [Sangiovanni and Liscinsky 1984]. If the combustion of a group of closely spaced droplets is roughly equivalent to that of a single larger droplet (Labowsky and Rosner 1978), this result is also consistent with proportionally increased sooting with increasing droplet diameter. A connection between fuel molecule residence time and droplet diameter has been offered as a reason for an influence of D_0 on soot formation and the droplet burning rate [Jackson et al. 1992]. The temperature of the droplet flame should also effect the propensity for sooting [Glassman 1987] but it is not known how large the variation of flame temperature with D_0 is for the range of D_0 examined.

Burning rate measurements reported by Kumagai et al. (1971), Okajima and Kumagai (1975), and Hara and Kumagai (1990, 1994) show that K is nearly independent of D_0 , or at most increases slightly. These data were recently criticized [Choi et al. 1990] on the grounds that spherical symmetry was not attained initially in the experiments because of various levels of convection which went undetected. Convection was thought to be caused by the method of igniting the droplets. With spark ignition and the force it can create on a droplet [Shaw et al. 1988b], ignition symmetry can be a factor in setting up convective currents around a droplet that could persist [Monaghan et al. 1968] throughout burning.

Additional reasons for the differences are speculated. Convection can sweep soot precursors or aggregates away from the droplet and through the flame, to open the flame, or curtail the period that a shell would stay intact during burning. The heat transfer to the droplet could then be reduced and lead to a burning rate that is more indicative of a lightly, or no, sooting limit which (presumably) would be higher than when a shell forms early in burning and remains intact throughout the droplet lifetime. These effects may be reduced as the droplet diameter decreases since sooting is proportionally less pronounced for small droplets. That K would not depend on D_0 for a sooting fuel would be consistent with a burning condition in which soot does not influence the process. This could arise if soot aggregates are swept away by convection and are never trapped between the droplet and flame. Figure 3b shows the calculated burning rate in this limit where sooting is not a factor for heptane [Jackson and Avedisian 1994]. In any event, it appears that K is somewhat sensitive to the soot patterns formed and the duration that these patterns remain intact throughout burning.

The results of Kumagai et al. (1971), Okajima and Kumagai (1975) and Hara and Kumagai (1994) showed burning rates for heptane that are higher than those shown in fig. 3 and reported elsewhere [Choi et al. 1990]. The predicted quasi-steady soot-free SSDC burning rate of heptane is about $.86\text{mm}^2/\text{s}$ [Jackson and Avedisian 1994] in atmospheric air, which compares with measured values of around $.78\text{mm}^2/\text{s}$ [Hara and Kumagai 1990, 1994], independent of D_0 , and the results of fig. 3 for the smallest droplets studied. For $D_0 \approx 1\text{mm}$, burning rates measured by Jackson and Avedisian (1993) are lower, averaging about $.6\text{mm}^2/\text{s}$. An additional heptane burning rate for $D_0 \approx 1.12\text{mm}$ was reported as $.5\text{mm}^2/\text{s}$ [Choi et al. 1990] for a droplet with the lowest relative droplet/gas velocity (and presumably burning with the greatest degree of symmetry) from among those they studied which is consistent with the trends shown in fig. 3.

5.3 Composition

Few studies have examined the influence of composition on droplet combustion in microgravity (e.g., Yang et al. 1990; Mikami et al. 1993), and one specifically addresses soot for droplets burning in microgravity [Jackson et al. 1991] for a mixture of methanol and toluene. In that study, a strong effect of methanol addition to toluene on soot image intensity was found (for roughly the same lighting conditions). For example, 5% toluene in methanol burns essentially the same as pure methanol, including the observation of extinction [Cho et al. 1990, 1992; Yang et al. 1990; Jackson et al. 1991]. The appearance of soot particles was not evident even for a 25% toluene mixture (though particles could have gone undetected due to the optical arrangement used). Higher toluene concentrations create more luminous flames signifying more soot formation. Only for a 50% toluene mixture was significant soot aggregate and shell formation observed. Some of the aggregates formed were carried through the flame and oxidized (the Stefan force on the particles evidently being able to overcome the thermophoretic force).

Figure 4 is a photographic sequence that shows the development of a soot shell and outer luminous zone for a toluene/methanol mixture droplet that contains initially 50% toluene. An early period of droplet burning (for $t < 0.15$ s in fig. 4) is characterized by no visible soot formation and relatively low flame luminosity. Because methanol is vaporized more rapidly than toluene from the surface, the droplet surface becomes richer in toluene as burning proceeds and the toluene fractional mass vaporization increases. Furthermore, a toluene flame should be larger than a methanol flame so that the residence time will be increased, and with time more soot would be produced. It should be noted that even for a single component fuel there will be a delay time for soot to form which, of course, cannot be attributed to preferential vaporization.

6. Disruptive Burning of Single Component Fuels

An interesting phenomenon has occasionally been observed during combustion of single component fuels in microgravity, namely that the droplet will sometimes burst or explode near the end of combustion. This phenomenon has only been observed when soot forms. Originally observed by Knight and Williams (1980) in microgravity experiments on free n-decane droplets, and termed "flash extinction", it has been more prominent in the observations reported by Shaw et al. (1988a) for near spherically symmetric n-decane droplet combustion. Disruptive burning for a single component fuel droplet has also been observed for butyl alcohol and furfural alcohol droplets burning in a convective environment [Kesten et al. 1980]. Three arguments have been offered for disruptive burning of single component fuels.

The first assumes that low volatility soot precursor species get absorbed into the droplet during combustion [Shaw and Williams 1990]. Once absorbed, the droplet is no longer a single component fuel and the mechanism for disruption can then be much the same as speculated for a multicomponent fuel droplet (e.g., Law 1982; Avedisian and Glassman 1981). The second conjectures that hot soot agglomerate particles are driven toward the droplet by a dominant thermophoretic force after which the particles will contact the droplet sometime during combustion [Kesten et al. 1980] and cause rapid boiling of the liquid. The third assumes explosive ignition of soot particles if the flame is very close to the droplet surface [Choi et al. 1993].

Some of these mechanisms require an inward motion of soot agglomerates, or soot precursor species, toward the droplet surface, which can be provided by the thermophoretic force acting on the particles as discussed previously. Others, require that the aggregates are driven away from the droplet (and toward the flame). The location where forces on aggregates balance depends on the aggregate size and O_2 concentration in the ambience [Jackson and Avedisian 1992; Choi et al. 1993]. If the aggregates are driven away from the droplet, they can pass through the flame where they can ignite, perhaps explosively. Further work will be required to identify more precisely the correct mechanism for this unusual behavior for single component (sooting) fuels.

7. Modeling Soot Formation in Spherically Symmetric Droplet Flames

Modelling soot formation process for SSDC has not been accomplished because of the complexities involved. Complete combustion chemistry and soot formation dynamics are aspects of that must be coupled with the conservation equations. The problem is compounded by the fact that both the chemical kinetic mechanism for many fuels and the rate equations for soot formation in droplet flames are not known with certainty. Some of the building blocks needed for SSDC

modelling have been developed in past research, for example the influence of flame radiation [Saitoh et al. 1993; Lage and Rangle 1993] and complete chemistry for several fuels [Cho et al. 1990, 1992; Jackson and Avedisian 1994]. Models for soot production rates in flames [Kennedy et al. 1990; Frenklach et al. 1990] have also been developed. Concerning the soot shell, the only aspect thus far addressed has been to show with simplified models that the forces acting on soot particles can in fact lead to locations between the droplet and flame where they balance [Jackson et al. 1992; Choi et al. 1993]. The effect of the shell on heat and mass transfer between the droplet and flame has not yet been addressed in analysis.

8. Summary and Needs

Two broad research needs to further understand soot formation during SSDC are the following. Firstly, there is a dearth of experimental observations on soot formation in SSDC. This fact is tied to the great difficulty of forming a droplet, igniting it, and keeping it motionless during combustion, and all in a buoyancy-free environment. Preferably, experiments should be done with free or unsupported droplets. If suspended droplets are used, care should be used in interpreting the results, especially the effect of the fiber on droplet shape and attraction of soot particles.

Secondly, analyses of droplet combustion processes should begin to include formation of a soot shell and its effect on burning. The analyses will probably have to be numerical as it is unlikely that a single, or a few, reaction steps will be capable of capturing the pathways to soot formation. The difficulty of this undertaking is compounded by the uncertainty of both the rate equations involved with soot formation and the chemistry of combustion. Work should continue developing validated chemical kinetic models (complete chemistry) of hydrocarbon fuels, especially those that are liquid at room temperature, and fundamental experiments are needed to quantify the steps in the formation of soot that will be important inputs to SSDC models.

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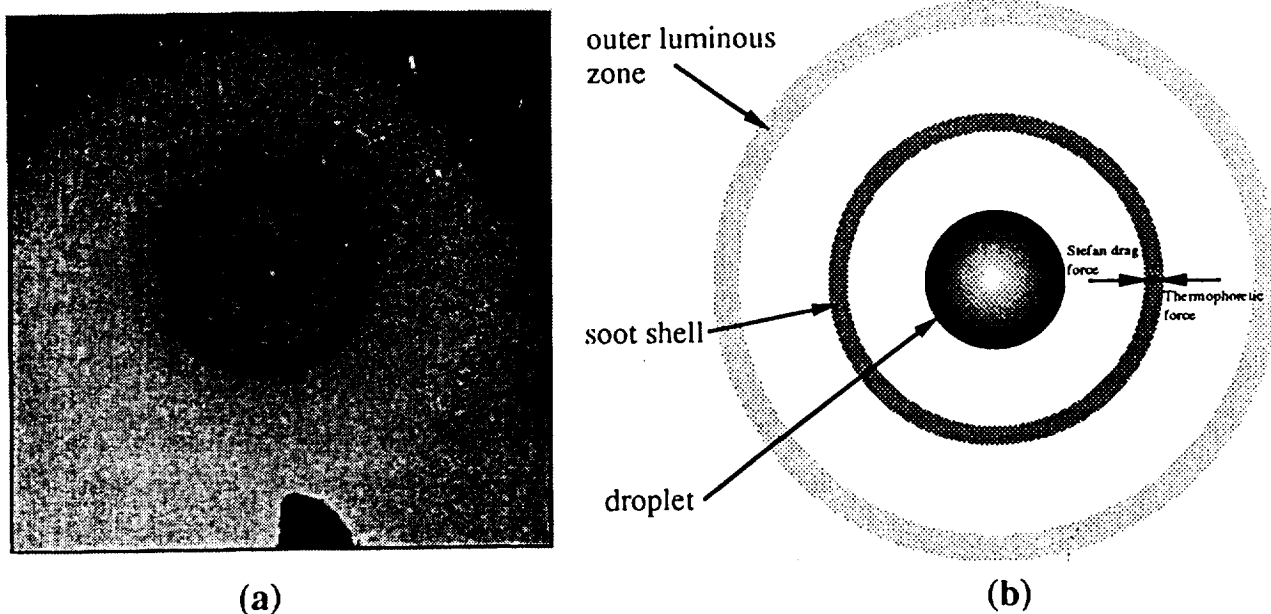


Figure 1: Basic configuration of spherically symmetric combustion of a stationary and unsupported heptane droplet. In the photograph, $D_0 \approx 0.7$ mm

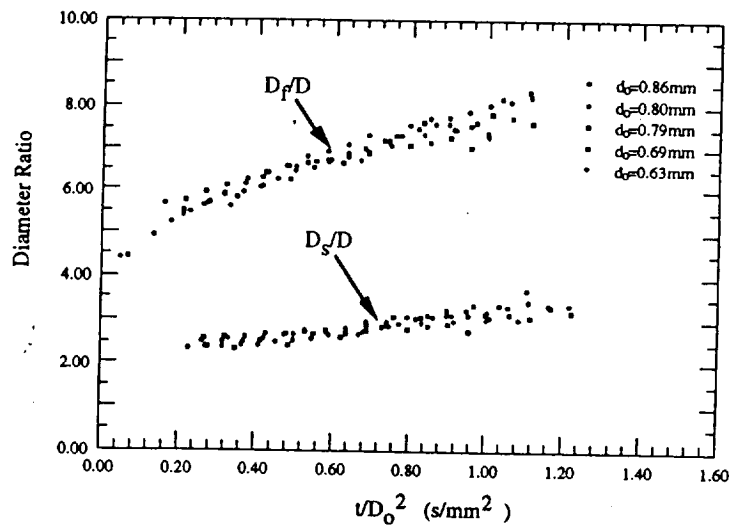


Figure 2: Variation of luminous zone and soot shell diameters (divided by the instantaneous droplet diameter) with t/D_0^2 .

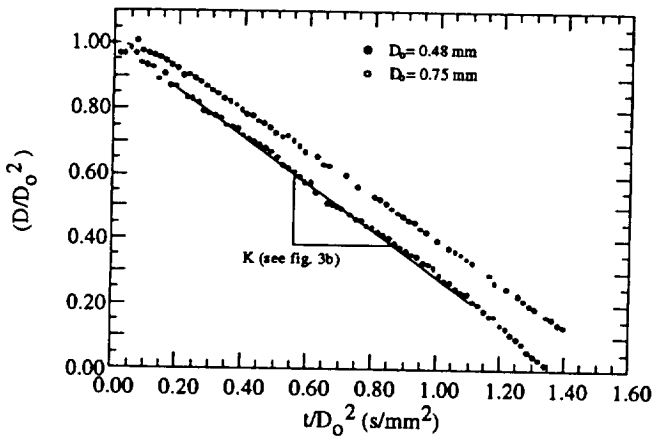


Figure 3a: Variation of $(D/D_0)^2$ with t/D_0^2 for heptane droplets burning in room temperature air at atmospheric pressure.

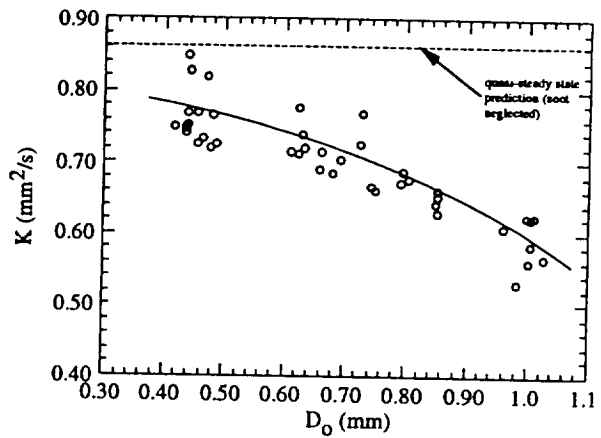


Figure 3b: Burning rates of heptane in room temperature air at atmospheric pressure in microgravity.

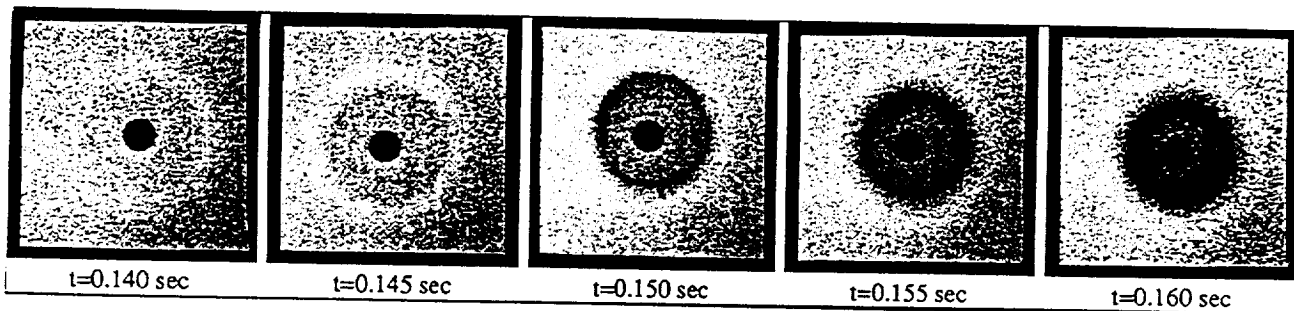


Figure 4: Development of soot shell for combustion in microgravity of a methanol/toluene mixture droplet with $D_0 \approx 0.49$ mm. Droplet was initially an equivolume mixture of methanol and toluene. Time after ignition is shown beneath each picture.