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Numerical investigation of soot formation from microgravity droplet combustion using heterogeneous chemistry

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ABSTRACT

The use of isolated droplets as idealized systems is an established practice to get an insight on the physics of combustion, and an optimal test field to verify physical submodels. In this context, this work examines the dynamics of soot formation from the combustion of hydrocarbon liquid fuels in such conditions. A detailed, heterogeneous kinetic mechanism, describing aerosol and particle behavior through a discrete sectional approach is incorporated. The developed 1-dimensional model accounts for (i) non-luminous and luminous radiative heat losses, and (ii) incomplete thermal accommodation in the calculation of the thermophoretic flux. The combustion of droplets of *n*-heptane, i.e., the simplest representative species of real fuels, was investigated as test case; an upstream skeletal reduction of the kinetic mechanism was carried out to limit calculation times. After checking the performance of the reduced mechanism against gas-phase experimental data, the transient evolution of the system was analyzed through a comprehensive study, including fiber-suspended ($D_0 < 1$ mm) as well as free ($D_0 > 1$ mm) droplets.

The different steps of soot evolution were quantified, and localized in the region between the flame front and the soot shell, where particle velocity is directed inwards because of thermophoresis, and residence times are much higher than what usually found in diffusion flames. As a result, growth, coalescence, and aggregation steps are significantly enhanced, and soot accumulates in the inner shell, with an evident modification of the particle size distribution, if compared to what observed in conventional combustion conditions. The model exhibits a satisfactory agreement with experimental data on flame temperature and position around the droplet, while for larger droplets an increasing sensitivity to the radiation model was observed. It is found that the latter has a significant impact on the production of soot, while scarcely affecting the location of the soot shell. On the other side, the inclusion of incomplete thermal accommodation in the thermophoretic law brought about more accurate predictions of both volume fractions and shell location, and highlighted the primary role of thermophoresis in these conditions, as already found in literature through more simplified approaches.

1. Introduction

Nowadays, one of the major concerns raised by the combustion of liquid fuels is related to the formation of soot, toward which the interest of the scientific community is mostly motivated by its impacts on combustion efficiency [1], global warming [2,3], and human health [4–6]. Indeed, the use of a liquid feed in the form of a spray, and the resulting heterogeneous combustion create favorable conditions for particle nucleation from Polycyclic Aromatic Hydrocarbons (PAH) and their subsequent growth [7]. The significant amount of soot is the macroscopic outcome of the interaction among several processes affecting real devices, including fuel evaporation and combustion, convection, and radiation. The timescale overlapping between the phenomena at stake results in the impossibility to decouple them, and makes the direct investigation of such systems a very challenging task.

In the attempt to gain a deeper understanding of the dynamics of droplet combustion and soot formation, as well as to develop and test submodels before their implementation in the Computational Fluid Dynamic (CFD) codes, in the latest decades significant experimental and modeling effort has been directed towards the combustion of simpler, idealized systems like spherical droplets in microgravity conditions [8–11]. Although their size is significantly larger than those found in real systems – millimeters instead of tens of microns – and they do not consider buoyancy-related effects, they provide fundamental information about the intricate

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Nomenclature

Roman Symbols

$\hat{h}_R$	Enthalpy of formation [J/kg]
c_p	Constant-pressure specific heat [J/kg/K]
dp	Soot primary particle diameter [m]
fv	Soot volume fraction [-]
I	Radiation intensity [W/m^2]
I_b	Blackbody intensity [W/m^2]
j	Flux [kg/m^2/s]
K	Burning rate [mm^2/s]
k	Thermal conductivity [W/(m•K)]
N_d	Soot number density [1/m^3]
q_R	Heat of radiation [W/(m^2•K)]
r_d	Droplet radius [mm]
v	Velocity [m/s]
$V_{th,r}$	Thermophoretic diffusivity [-]

coupling between the involved physical and chemical processes. As a matter of fact, they represent an optimal trade-off between the complexity of the problem, which is reduced to one spatial dimension because of spherical symmetry, and its comprehensiveness, since most phenomena involved in spray combustion are still considered, e.g. evaporation- and diffusion-induced transport, heterogeneous properties, radiation, and aerosol chemistry. In this way, the physical submodels representing each of them can be benchmarked in relatively "simplified" conditions. Conversely, assessing theoretical models of the governing phenomena from the interpretation of multidimensional experiments would be much more complicated, because of the resulting physical (and therefore, numerical) complexity.

For this reason, large experimental activity on soot formation in spherical droplets was carried out via reduced-gravity experiments. According to the droplet size, such conditions were attained either through drop towers or in the outer space. Most experimental campaigns date back to around 1990s: the shell-like structure of soot, placed between the flame layer and the droplet surface, was first observed by Shaw et al. [12]. The reason behind such a peculiarity was later attributed by Jackson and Avedisian [13] to the competition between the convective - or Stefan - flow, due to evaporation, and the thermophoretic flow acting on particles. They correlated the decrease in the burning rate with droplet size to soot production because of a combination of barrier and radiation effect, affecting the total heat transferred to the droplet. The structure of the flame enclosing the droplet was then studied by Mikami et al. [14] by using hooked thermocouples, and showing that the maximum temperature region, i.e. the reaction zone, is located outside of the yellow luminous zone, whose color is due to radiation from soot. The transient evolution of soot volume fractions was quantified in later works [15–17]: the maximum values were found to be between 10 and 100 ppm, i.e. more than one order of magnitude higher than what observed in gas-phase diffusion flames with the same fuels [18,19]. Finally, experiments on space laboratories [11,20,21] allowed an extensive investigation on large-sized droplets (super-millimetric), whose combustion dynamics cannot be followed in drop towers because of their insufficient length. In this way, light could be shed on radiative extinction of large droplets [20] and cool flame burning due to the low-temperature chemistry of *n*-alkanes [21,22].

From a modeling standpoint, a detailed description of the key phenomena affecting spherical droplet combustion is not straightforward, and it is often not compatible with an acceptable computational effort. The formation of soot adds substantial complexity to the modeling of droplet dynamics, which has been the subject of extensive research for long time [23–26]. First of all, the transient evolution of soot is strongly correlated to the gas-phase chemistry, from which soot precursors (PAH) are generated. Therefore, a detailed kinetic description is a necessary requirement to be predictive on soot formation. Second, the presence of soot has an impact on heat transfer and especially on luminous radiation, which on turn affects the burning rates and the overall combustion process. As a result, modeling activity has often been based on one or more assumptions to simplify the size of the problem and make it computationally viable: Chang and Shier [27] used a one-step reaction mechanism to parametrically study the correlation between soot production, flame radiance and droplet burning rates. Similarly, Baek et al. [28] analyzed soot/radiation interaction through global chemistry and a simplified two-equation model [29] describing the evolution of soot volume fraction and number density. On the other side, Jackson and Avedisian [30] were among the first to incorporate detailed chemistry in droplet calculations: coherently with experimental observations, they could find an increase in acetylene concentration (a key species among soot precursors) with the droplet diameter, but they could not analyze PAH evolution into soot particles because the adopted kinetic model described only gas-phase combustion. Later studies using detailed chemistry [8,31] neglected soot formation and the related effects on the combustion behavior.

In the light of the previous modeling efforts, in this work the dynamics of soot formation in the combustion of spherical droplets is investigated through a comprehensive approach, which incorporates a detailed kinetic mechanism representing soot formation from the underlying gas-phase. An existing mathematical model of droplet evaporation and ignition is extended towards a detailed description of the key processes involving soot: besides the description of aerosol dynamics coupled to gas-phase kinetics, nongray radiation effects are considered for both gas-phase species and solid particles, and thermophoretic effect on solid particles is accounted for. The main features of the mathematical model are recalled in Section 2, with an emphasis on the submodels of major relevance for the purposes of this work. The development of the kinetic mechanism of soot formation is then separately outlined in Section 3. In Section 4, the model is leveraged to shed light on the transient evolution of the two-phase system, and the role of soot in such dynamics: numerical predictions are compared against the experimental data collected so far, and *n*-heptane is used as case study since it has been the most studied fuel in microgravity conditions. Conclusions are drawn in Section 5.

2. Mathematical model

The mathematical model describes the combustion of an isolated droplet in a gas-phase environment, idealizing the experimental conditions of drop towers and outer space. The core framework is fully described in [32,33] and is based on the following assumptions:

- Spherical symmetry and absence of gravity;
- Constant pressure;
- Absence of reactions in liquid phase;
- Thermodynamic equilibrium at the liquid/gas interface.

In particular, spherical symmetry allows to consider the problem as 1-dimensional. Considering a monocomponent fuel, the conservation equations for energy and velocity in the liquid phase are:

$$\rho_L c_{p,L}\left(\frac{\partial T_L}{\partial t}+v_L\frac{\partial T_L}{\partial r}\right)=\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 k_L\frac{\partial T_L}{\partial r}\right) \tag{1}$$

$$\frac{\partial \rho_L}{\partial t}+\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\rho_L v_L\right)=0 \tag{2}$$