Vaporization and gas-phase combustion of electrosprayed heptane in a small chamber

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A B S T R A C T

A model is formulated of the vaporization and gas-phase combustion of a dilute spray of electrically charged droplets of heptane with a coflow of preheated air in a mesoscale combustion chamber. The model includes an Eulerian/Lagrangian treatment of the gas and the droplets, an equilibrium vaporization submodel, and a single step mechanism for the combustion of heptane vapor. The effects of the inlet air temperature, the fuel flow rate and the overall equivalence ratio are investigated. Two combustion modes are described for globally lean systems. When rapid vaporization leads to a kernel of high fuel vapor concentration, combustion may begin before the fuel diffuses in the air coflow, in a layer of intense reaction that locally depletes the oxygen and leaves behind a region of high temperature fuel vapor surrounded by a diffusion flame. When the vaporization rate and the maximum fuel vapor concentration are decreased by increasing the size of the droplets or the inlet velocity of the air, combustion occurs in a lifted lean premixed flame after the fuel has fully vaporized and mixed with the air coflow. Ignition and extinction conditions are discussed. The degree of accuracy that can be expected from the single step reaction mechanism is also discussed.

1. Introduction

The energy density of liquid hydrocarbons is about 100 times larger than that of the currently available batteries, which makes power-generation systems based on the combustion of these fuels a promising technology in the quest for better power sources for portable equipment and other applications [1,2]. Many conventional techniques and operation principles of macroscopic combustion systems become impractical for miniaturized system, although the extent of the modifications required is smaller for devices with overall dimensions of several centimeters intended to generate powers of 10–100 W, referred to as mesoscale devices, than for smaller microdevices. Difficulties are also smaller when the liquid fuel is vaporized and burned under steady conditions in a chamber coupled to a direct energy conversion module such as a thermoelectric or thermovoltaic generator, or to a Stirling engine, than when conventional engines using classical thermodynamic cycles are miniaturized. Only mesoscale, steady combustion devices are considered here.

Reduction or disappearance of turbulent mixing, heat transfer, and flame wrinkling in small systems are important differences with their macroscopic counterparts, though Wu et al. [3] were

able to stabilize a non-premixed flame in a mesoscale combustor by means of a vortex flow that extends the concept of whirl combustion of macroscopic combustors. Increased heat losses due to the augmented surface-to-volume ratio is another effect of miniaturization, which increases the vaporization and chemical times, and may compromise combustion efficiency. A third effect of miniaturization is the reduction of the residence time, which should be larger than the vaporization and chemical times in order to achieve complete combustion and prevent the emission of pollutants such as CO and unburned hydrocarbons.

The notion of heat recuperation or excess enthalpy for miniature devices, whereby part of the thermal enthalpy of the combustion products is used to preheat the reactants, was first studied by Weinberg and coworkers [4–6] and later extended to many meso- and micro-scale combustors to achieve steady combustion under conditions of large heat losses or lean mixtures, which would otherwise result in extinction.

With exceptions [7], the liquid fuel must be atomized into small droplets that have to disperse and vaporize in the short residence time available. Flow-blurring atomizers relying on aerodynamic forces have been used by Sadasivuni and Agrawal [8] to atomize kerosene in an annular heat recuperator surrounding a cylindrical combustion chamber, where the droplets vaporize and the vapor mixes with the air before entering the chamber. Gomez and coworkers used electrosprays to atomize JP8 into electrically

charged droplets in their mesoscale combustor [9–12]. Electro-sprays have also been used in combination with flame-attaching grids in narrow tubes [13,14].

Catalytic combustion may improve combustion stability and reduce pollutant emission in the conditions of short residence times and low temperature typical of small scale combustors. Catalytic combustion in a series of catalyst-coated grids at the outlet of a chamber where fuel droplets vaporize in a coflow of preheated air has been successfully used by Gomez and coworkers [9–12]. Despite the favorable properties of catalytic combustion, gas-phase combustion may still retain some advantages in excess enthalpy mesoscale systems, stemming from their volumetric rather than superficial character, provided heat recuperation overcomes heat losses to a sufficient extent to make gas-phase combustion possible.

The purpose of this paper is to numerically study the feasibility of gas-phase combustion in a small chamber similar to those used by Deng et al. [11] in their experiments with multiplexed electro-spray sources and catalytic grids. A two-way coupled model of the dispersion, vaporization and gas-phase combustion of a single electro-spray from a multiplexed source in a coflow of hot air has been developed which is based on the model proposed in Ref. [15] to study the heating and vaporization of a spray of heptane droplets in a chamber. This model, which in turn builds on a model of Arumugham-Achary et al. [16], consists of a combination of an Eulerian mesoscale description of the gas and a Lagrangian description of the spray of droplets, carried out with a method of particles. The flow in the chamber is laminar (Reynolds number in the range 100–500), which makes the results free from the uncertainties of turbulence modeling. The model relies on the validation of Ref. [16] (see also references therein) against an individual electro-spray source, and on the previous computations in Ref. [15], which were qualitatively compared to the experiments of Refs. [9–12] to the limited extent permitted by the available data. The previous model is extended here to account for the combustion of heptane vapor by means of a single step mechanism calibrated to reproduce the ignition delay times measured for heptane–air mixtures in conditions similar to those found at ignition in the chamber. The degree of accuracy that can be expected from this drastic simplification of the heptane–air kinetic mechanism depends on the structure of the computed solutions and is discussed in Section 3. The model of Ref. [15] is also improved by using a more realistic droplet heating and vaporization submodel.

2. Formulation

Kyritsis et al. [9] and Deng et al. [11] used two-dimensional periodic arrays of identical electro-spray sources to inject small, nearly monodisperse, electrically charged fuel droplets through one of the bases of a cylindrical chamber. To approximately model this system, the invariance of the array under certain discrete angle rotations is replaced here by axial symmetry. A single electro-spray source of the periodic array is considered that injects droplets and a coflow of air through a circular orifice of radius r_h at one of the bases of a cylindrical chamber, and symmetry conditions (see Eq. (16) below) are applied at a distance R from an axis through the center of the orifice, as sketched in Fig. 1. The source generates Φ droplets per unit time with radius a_0 , electric charge e_0 , temperature T_0 and velocity v_{pI} , and the air enters the chamber with velocity v_{gI} and temperature $T_1 > T_0$ through the same orifice. The droplets heat up and vaporize in the chamber due to the heat transferred by conduction from the hot air, generating fuel vapor that reacts with the oxygen of the air. The combustion products leave the chamber through the opposite base, at a distance L from the injection orifice.

Fig. 1. Schematic of the investigated configuration with indication of relevant dimensions. The grey area represents the vaporizing spray. The heptane vapor burns in a combustion region represented by a red line in the figure. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

A dilute spray is considered. The mean distance between droplets, ℓ_d , is large compared to the radius of a droplet and small compared to the size of the chamber; i.e. $a_0 \ll \ell_d \ll L$. In these conditions, an Eulerian, mesoscale description of the gas flow [17] combined with a Lagrangian, particle-in-cell description of the droplets [18,19] can be used.

2.1. Droplets

The droplets interact mainly through the surrounding gas and through the electric field induced by the electric charges they carry. Direct interactions between droplets are rare events when $a_0 \ll \ell_d$.

The mass, momentum and energy conservation equations for droplet i are

$$\begin{aligned} \frac{d\mathbf{x}_i}{dt} &= \mathbf{v}_i, & m_i \frac{d\mathbf{v}_i}{dt} &= \mathbf{F}_i + e_i \mathbf{E}_i, \\ \frac{dm_i}{dt} &= -\dot{m}_i, & m_i c \frac{dT_i}{dt} &= q_{g_i} - \dot{m}_i L_v, \end{aligned} \quad (1)$$

where $\mathbf{x}_i(t)$ and $\mathbf{v}_i(t)$ are the position and velocity of the center of the droplet, $m_i = \frac{4}{3}\pi\rho_l a_i^3$ with ρ_l the density of the liquid and $a_i(t)$ the radius of the droplet, $T_i(t)$ is the temperature of the droplet, which is approximately uniform because the thermal conductivity of the liquid is large compared to the thermal conductivity of the gas, and $e_i(t)$ is the electric charge of the droplet, whose time evolution is computed as described below. In these equations $\mathbf{F}_i$ is the force exerted by the gas on the droplet, q_{g_i} is the heat flux reaching the droplet, and $\dot{m}_i$ is the vaporization rate. These quantities depend on the radius, velocity and temperature of the droplet, and on the local velocity, $\mathbf{v}_{g_i}$, temperature, T_{g_i} , and composition of the gas seen by the droplet. Finally c and L_v are the specific heat and the latent heat of vaporization of heptane, and $\mathbf{E}_i$ is the electric field at the position of the droplet.

As a case representative of the conditions of interest (see Section 2.3), the vaporization time of a droplet of heptane of initial radius $a_0 = 8.75 \mu\text{m}$ injected with initial velocity $v_{pI} = 10 \text{ m/s}$ and temperature $T_0 = 293 \text{ K}$ in quiescent air at a pressure of 3 atm and various temperatures has been computed. The model of Abramzon and Sirignano [20], the correlations of Clift et al. [21] for the drag coefficient, Nusselt and Sherwood numbers, and the 1/3 rule for the average gas properties have been used to evaluate the right-hand sides of equations (1) without electric force. The simple power laws $\mu_g/\mu_{g0} = k_g/k_{g0} = (T_g/T_0)^\kappa$ with $\kappa = 0.75$ are assumed for the viscosity and thermal conductivity of the gas, together with constant values of the Prandtl and Lewis numbers. The results, shown by the solid curve in Fig. 2(a), are very similar to the vaporization time for a droplet at rest with the same initial and gas

Fig. 2. (a) Vaporization time of a droplet of heptane of initial radius $a_0 = 8.75 \mu\text{m}$, temperature $T_0 = 293 \text{ K}$ and velocity $v_{p0} = 10 \text{ m/s}$ as a function of the temperature of the surrounding air, computed with the model of Abramzon and Sirignano [20] (solid curve), and leaving out the effect of the slip flow (dashed curve). (b) Vaporization time of the same droplet at rest, computed using (2) and (3) in the last two equations (1) (solid, coincides with the dashed curve of (a)), neglecting the Stefan flow (dashed), and in the asymptotic limit $L_v/R_f T_b \rightarrow \infty$ (dotted).

conditions (dashed curve), which is due to the rapid deceleration of the small droplet considered. On the basis of this comparison, the effect of the slip flow on q_{g_i} and $\dot{m}_i$ is neglected in what follows. In these conditions, the stationary energy and species conservation equations around the droplet can be solved in terms of the Spalding mass transfer numbers (see, e.g. Sánchez et al. [22]) to give

$$\lambda_i \equiv \frac{\dot{m}_i}{4\pi a_i \bar{k}_{g_i}/c_p} = \frac{1}{\text{Le}_f} \ln \left(\frac{1 - Y_{f_i}}{1 - Y_{f_{sl}}} \right) \quad (2)$$

$$\frac{q_{g_i}}{4\pi a_i \bar{k}_{g_i} (T_{g_i} - T_i)} = \frac{\lambda_i}{e^{\lambda_i} - 1},$$

where the Clausius-Clapeyron relation and the 1/3 rule for the average thermal conductivity of the gas,

$$Y_{f_{sl}} = \frac{W_f}{W} \exp \left(\frac{L_v}{R_f T_b} - \frac{L_v}{R_f T_i} \right) \quad (3)$$

$$\bar{k}_{g_i} = k_g(\bar{T}_i) \text{ with } \bar{T}_i = \frac{1}{3} (2T_i + T_{g_i}),$$

are used. In these equations, W_f and T_b are the molecular mass and boiling temperature of the heptane, R_f and Le_f are the gas

constant and Lewis number of the heptane vapor, $Y_{f_{sl}}$ and Y_{f_i} are the mass fractions of heptane in the gas at the surface of the droplet and away from the droplet, and c_p and W are the specific heat and the mean molecular mass of the gas at the surface of the droplet. This latter quantity is approximately computed using the simplified expression $W_f/W = Y_{f_{sl}} + (1 - Y_{f_{sl}})W_f/W_{N_2}$, where W_{N_2} is the molecular mass of nitrogen. This approximation accounts for the large difference of the molecular masses of fuel and nitrogen, while taking the molecular masses of all other species equal to that of nitrogen [22]. The specific heat c_p is assumed to be constant.

The effects of additional simplifications of (2) and (3) (not used in what follows) are illustrated in Fig. 2(b). The dashed curve in this figure shows the droplet vaporization time when the Stefan flow is neglected, which reduces (2) to $\lambda_i = Y_{f_{sl}} - Y_{f_i}$ and $q_{g_i}/4\pi a_i \bar{k}_{g_i} (T_{g_i} - T_i) = 1$, and the dotted curve gives results pertaining to the asymptotic limit $L_v/R_f T_b \rightarrow \infty$ [23], in which vaporization occurs at $T_i = T_b$ and is preceded by a stage of droplet heating without vaporization. As can be seen, the errors incurred by using these approximations are moderate in many cases. The error of the first approximation is largest at high temperatures, and the error of the second approximation is largest when the gas temperature approaches the boiling temperature of the fuel.

The Reynolds number of the slip flow, $\text{Re}_i = \rho_{g_i} |\mathbf{v}_i - \mathbf{v}_{g_i}| a_i / \mu_{g_i}$, is moderately small during most of the droplet evolution. In addition, consideration of the Stefan flow leads to a modification of the Stokes law that is small when $\lambda_i = O(1)$ [24]. Neglecting this correction, the force of the gas on the droplet is taken to be

$$\mathbf{F}_i = -c_{f_i} (\mathbf{v}_i - \mathbf{v}_{g_i}), \quad c_{f_i} = 6\pi \bar{\mu}_{g_i} a_i \quad (4)$$

where $\bar{\mu}_{g_i} = \mu_g(\bar{T}_i)$

The maximum electric charge that a droplet of radius a_i can bear is given by the Rayleigh limit $e_{Ri} = 8\pi (\epsilon_0 \sigma a_i^3)^{1/2}$ [25], where σ is the surface tension of the liquid and ϵ_0 is the electric permittivity of the gas. This limit is reached when the droplet vaporizes keeping its electric charge, and then the droplet undergoes a Coulomb explosion which, for liquids of high electric conductivity, consists in the rapid emission of a large number of very small daughter droplets that carry a significant fraction of the electric charge of the parent droplet with only a very small fraction of its mass [26]. The results of Gomez and Tang [27] suggest that this is the mode of Coulomb explosion that occurs for the moderate conductivities usually achieved with doped heptane, with the droplet losing about 30% of its charge in the process. Thus, Coulomb explosions are idealized here as instantaneous events in which the mass of the droplet does not change while

$$e_i \text{ decreases to } (1 - \alpha)e_i \text{ whenever } e_i \geq e_{Ri}, \quad (5)$$

with $\alpha = 0.3$. The daughter droplets vaporize rapidly, and the electric charges lost by the droplets that underwent explosions soon reappear as a population of charged nanometer-size residues (solvated ions) whose charge density, ρ_r , obeys the conservation equation

$$\frac{\partial \rho_r}{\partial t} + \nabla \cdot [\rho_r (\mathbf{v}_g + K\mathbf{E})] = S_r, \quad (6)$$

where $\mathbf{v}_g$ is the velocity of the gas, K is the mobility of the residues, and S_r is the charge generated by Coulomb explosions per unit volume and time, given by (13) below. Charged residues reach the metallic bases of the chamber, where they are absorbed.

Droplets are continuously injected, Φ per unit time, at the injection orifice with a radial Gaussian distribution with standard deviation $r_c < r_h$, truncated at the orifice radius r_h . The injection conditions read therefore

$$\left. \begin{aligned} x_i &= 0, \quad r_i = \text{rand}[G(r)], \quad \mathbf{v}_i = v_{p_i} \mathbf{i} \\ a_i &= a_0, \quad T_i = T_0, \quad e_i = e_0 \end{aligned} \right\} \text{ at } t = \frac{i}{\Phi}, \quad (7)$$

with the cumulative distribution function

$$G(r) = \frac{1 - \exp(-r^2/r_c^2)}{1 - \exp(-r_h^2/r_c^2)} \text{ for } r < r_h,$$

$$G(r) = 1 \text{ for } r > r_h.$$

Here x and r are distances along and normal to the symmetry axis, with $x = 0$ at the orifice, and $\mathbf{i}$ is a unit axial vector. Upon injection, the droplets evolve according to equations (1) until they disappear when they fully vaporize or hit one of the bases of the chamber.

2.2. Gas

Conservation equations for the mesoscale description of the gas [17] can be derived from the principles of conservation of mass, momentum and energy applied to the gas in elementary control volumes of size large compared to ℓ_d but small compared to L . These volumes contain many droplets exchanging mass, momentum and energy with the gas, whose effect appears as distributed source terms in the gas equations. For a low Mach number mesoscale flow, leaving out radiation and using Fick laws for the diffusion of the fuel vapor and the oxygen, these equations are

$$\frac{\partial \rho_g}{\partial t} + \nabla \cdot (\rho_g \mathbf{v}_g) = S, \quad (8)$$

$$\rho_g \left(\frac{\partial \mathbf{v}_g}{\partial t} + \mathbf{v}_g \cdot \nabla \mathbf{v}_g \right) = -\nabla p + \nabla \cdot \boldsymbol{\tau}' - \mathbf{F} + \rho_r \mathbf{E}, \quad (9)$$

$$\rho_g \left(\frac{\partial Y_j}{\partial t} + \mathbf{v}_g \cdot \nabla Y_j \right) = \nabla \cdot \left(\frac{k_g/c_p}{Le_j} \nabla Y_j \right) + (1 - Y_j) S_j - s_j w, \quad (10)$$

$$\rho_g c_p \left(\frac{\partial T_g}{\partial t} + \mathbf{v}_g \cdot \nabla T_g \right) = \nabla \cdot (k_g \nabla T_g) - S_h + q w, \quad (11)$$

plus the equation of state $\rho_g T_g = \rho_{g0} T_0 \bar{W}/W_{air}$. Here ρ_g , T_g , $\mathbf{v}_g$ and Y_j are the mesoscale-averaged gas density, temperature, velocity and species mass fractions (with $j = F, O$ denoting fuel and oxygen hereafter); $\bar{W}/W_{air}$ is the mean molecular mass of the gas scaled with the mean molecular mass of the inlet air, which reduces to $\bar{W}/W_{air} = [1 + (18/7)Y_F]^{-1}$ with the approximation mentioned in Section 2.1 and $W_F/W_{N_2} = 25/7$; $\boldsymbol{\tau}'$ is the viscous stress tensor given by the Navier–Poisson law; c_p and Le_j are the specific heat of the gas and the Lewis numbers of the species, assumed to be constant; and ρ_{g0} is the density of the gas at the inlet temperature of the droplets, T_0 .

Combustion in the gas phase is modeled with a drastically simplified single step mechanism. The mass of fuel consumed per unit volume and time is given by the Arrhenius law $w = \rho_g B \exp(-T_0/T_g) Y_F Y_O$, where the frequency factor and activation temperature are chosen to approximately reproduce the ignition delay times measured by Ciozki and Adomeit [28] in shock tube experiments with heptane at about 3 atm. This is also the chamber pressure considered in what follows, so that the single step mechanism is expected to give a fair account of the autoignition of gaseous heptane–air mixtures in the chamber, which, as will be seen, is relevant to determine the ignition conditions. Specifically, the slope of the upper curve in Fig. 3 of Ref. [28] at about the “line of pressure variation” and an ignition delay time of 10 ms at 1000 K are chosen to determine $B = 2.2 \times 10^6 \text{ s}^{-1}$ and $T_0 = 10^4 \text{ K}$. In addition, for heptane, $s_F = 1$, $s_O = 3.52$ and $q = 39,974 \text{ kJ/kg}$.

The laminar flame speed computed with the single step mechanism and the simple power-law transport coefficients introduced in Section 2.1 is compared to the flame speed computed with the detailed mechanism of Jerzembeck et al. [29] and full multicomponent transport properties. Results for a gas pressure of

Fig. 3. Laminar speed of a heptane–air flame as a function of the fresh gas temperature for two values of the equivalence ratio, $\phi = 0.35$ (lower curve of each pair) and 0.7. Solid curves are for the detailed mechanism of Jerzembeck et al. [29], dashed curves are for the single step mechanism described in the text [heptane consumption rate $w = \rho_g B \exp(-T_0/T_g) Y_F Y_O$ with $B = 2.2 \times 10^6 \text{ s}^{-1}$ and $T_0 = 10^4 \text{ K}$], and dotted curves are for the Westbrook and Dryer single step mechanism [30].

3 atm and the two values $\phi = 0.35$ and 0.7 of the equivalence ratio are shown in Fig. 3 as functions of the fresh gas temperature. When $\phi = 0.35$, the flame speed of the single step mechanism (lower dashed curve) is surprisingly close to the flame speed of the detailed mechanism (lower solid curve) in a wide range of fresh gas temperatures. When $\phi = 0.7$, the single step mechanism underpredicts the flame speed (upper dashed and solid curves), but the agreement can be restored by multiplying the frequency factor B by 4.84. For comparison, the flame speed computed with the single step mechanism of Westbrook and Dryer [30] strongly overpredicts the flame speed at both values of the equivalence ratio (dotted curves in Fig. 3).

The ability of the single step mechanism to reproduce the laminar flame speed in wide ranges of gas conditions could be improved by the method proposed by Fernández-Tarrazo et al. [31] (see Giusti et al. [32] for a recent extension to the combustion of alcohols), which is based on using values of the heat of reaction, the activation temperature and the frequency factor that depend on the local equivalence ratio and are chosen to fit the correct laminar flame speed and adiabatic flame temperature across the whole flammability range. This refinement could improve the prediction of combustion characteristics in nonuniform mixtures (see Section 3.3 below) but it has not been used here.

The mesoscale-averaged electric field satisfies

$$\mathbf{E} = -\nabla \varphi, \quad \nabla^2 \varphi = -\frac{\rho_e + \rho_r}{\epsilon_0}. \quad (12)$$

The source terms on the right-hand sides of Eqs. (6) and (8)–(12) are [15,22]

$$\left. \begin{aligned} S_r &= \alpha \frac{\sum (e_i |\delta t|)}{\delta t \delta V}, \quad S = S_F = \frac{\sum \dot{m}_i}{\delta V}, \quad S_O = 0 \\ \mathbf{F} &= \frac{\sum [\mathbf{F}_i + \dot{m}_i (\mathbf{v}_{g_i} - \mathbf{v}_i)]}{\delta V} \\ S_h &= \frac{\sum [q_{g_i} + \dot{m}_i c_p (T_{g_i} - T_i)]}{\delta V}, \quad \rho_e = \frac{\sum e_i}{\delta V} \end{aligned} \right\} \quad (13)$$

where the sums are over the droplets present in a control volume δV centered at point $\mathbf{x}$ at time t , and $\sum (e_i |\delta t|)$ means sum of the charges of the droplets in δV that have undergone a Coulomb explosion in the time interval δt around t . The limits $\delta V \rightarrow 0$ and $\delta t \rightarrow 0$ are understood in the mesoscale description of the gas.

Boundary conditions for the gas equations, already specialized for an axisymmetric flow, are

$$\left. \begin{aligned} \mathbf{v}_g &= v_{g,i} \mathbf{i}, \quad T_g = T_1, \quad \varphi = 0 \\ \rho_g u_g Y_f - \frac{k_g/c_p}{Le_g} \frac{\partial Y_f}{\partial x} &= 0 \\ \rho_g u_g Y_o - \frac{k_g/c_p}{Le_o} \frac{\partial Y_o}{\partial x} &= \rho_{g1} u_{g1} Y_{o0} \end{aligned} \right\} \text{ for } r < r_h \quad (14)$$

$$\left. \begin{aligned} \mathbf{v}_g &= 0, \quad \frac{\partial Y_j}{\partial x} = 0 \\ T_g &= T_1, \quad \varphi = 0 \end{aligned} \right\} \text{ for } r_h < r < R$$

at $x = 0$, the zero-gradient conditions

$$v_g = \frac{\partial \omega_g}{\partial x} = \frac{\partial Y_j}{\partial x} = \frac{\partial T_g}{\partial x} = \varphi = 0 \quad (15)$$

at the chamber outlet, $x = L$, and

$$\frac{\partial u_g}{\partial r} = v_g = \frac{\partial Y_j}{\partial r} = \frac{\partial T_g}{\partial r} = \frac{\partial \varphi}{\partial r} = 0 \quad (16)$$

at $r = R$. Here u_g and v_g are the axial and radial components of the gas velocity, and ω_g is the only nonzero component of the vorticity. The hyperbolic equation (6) needs a boundary condition ($\rho_r = 0$) only in regions of the boundary where $\mathbf{v}_g + K\mathbf{E}$ points inward.

2.3. Characteristic values and orders of magnitude

The nondimensional form of problem (1)–(16) used for the numerical computations is given in the Appendix.

Values of the parameters are chosen for the simulations to be representative of some of the configurations investigated by Gomez and coworkers [9–12]. The chamber length is $L = 38.1$ mm, with a mean spacing between electrospray sources leading to $R = 5.33$ mm, an injection orifice of radius $r_h = 3.8$ mm, and $r_c = 0.8$ mm. The fuel is heptane doped with an electric conductivity enhancer, with physical properties $\rho_l = 679.5$ kg/m³, $c = 2241$ J/K kg, $\sigma = 2.01 \times 10^{-2}$ N/m, $T_b = 371.6$ K and $L_v = 3.65 \times 10^5$ J/kg. A base case is considered in which a flow rate $Q_b = 1.5$ ml/h is atomized per electrospray source into droplets of diameter $2a_{ob} = 17.5$ μ m, carrying an electric charge $e_{ob} = 1.08 \times 10^{-13}$ C, which are injected with velocity $v_{p1} = 10$ m/s and temperature $T_0 = 293$ K. The mass of heptane per droplet is then $m_{ob} = \frac{4}{3}\pi\rho_l a_{ob}^3 = 1.907 \times 10^{-12}$ kg, and the number of droplets injected per source per unit time is $\Phi_b = Q_b/(\frac{4}{3}\pi a_{ob}^3) = 1.49 \times 10^5$ s⁻¹. The size and charge of the droplets change when the flow rate of liquid atomized changes. The scaling laws for an electrospray in the cone-jet mode (see, e.g., [26]) provide the relations $a_0/a_{ob} = (e_0/e_{ob})^{1/2} = \Phi_b/\Phi = (Q/Q_b)^{1/2}$. Air ($Y_{o0} = 0.23$) preheated to a temperature $T_1 > T_0$ is injected in the chamber together with the droplets, with a velocity v_{g1} such that the overall equivalence ratio $\phi = 3.52 \rho_l Q/(0.23 \pi r_h^2 \rho_{g1} v_{g1})$ has a chosen value. The three main control parameters whose effect will be analyzed are the scaled flow rate of liquid Q/Q_b , the scaled inlet gas temperature T_1/T_0 , and the overall equivalence ratio ϕ .

The characteristic stopping or adaptation time of the droplets, determined from the balance of inertia and aerodynamic drag in the second equation (1), is $t_s = m_0/c_{f1}$ with $c_{f1} = 6\pi\mu_{g1}a_0$. The characteristic heating and vaporization times are $t_h = t_s(T_b - T_0)/(T_1 - T_0)$ and $t_v = t_s L_v/c_p(T_1 - T_b)$, from analogous balances in the fourth and third equations (1), where order unity factors have been left out. These three times are of the same order because $(T_b - T_0)/(T_1 - T_0)$ and $L_v/c_p(T_1 - T_b)$ are of order unity, though heating and vaporization may be very much enhanced if they occur in regions where combustion has increased the gas temperature well above T_1 . The characteristic penetration distance of the spray in the chamber is $\ell_s = v_{p1}t_s$; the characteristic number density of droplets is $n_0 = \Phi/(\pi r_c^2 v_{p1})$; and the characteristic mass loading of the spray is $\mathcal{M} = m_0 n_0/\rho_{g1}$.

The characteristic electric field induced by the charge of the droplets in the region of the spray is $E_s = e_0 n_0 \ell_s/\epsilon_0$, from the Poisson equation (12). The ratio of electric to aerodynamic forces in the second equation (1) is of order $\Pi = e_0 E_s/c_{f1} v_{p1} = m_0 n_0 e_0^2/\epsilon_0 c_{f1}$.

The drag of the droplets induces a motion of the gas in the region of the spray with characteristic velocity $v_{gs} = \mathcal{M}^{1/2} v_{p1}$ determined from the balance of inertia and momentum exchange terms in the momentum equation (9); i.e. $\rho_{g1} v_{gs}^2/\ell_s \sim n_0 c_{f1} v_{p1}$, which is a relevant balance insofar as the Reynolds number $Re = \rho_{g1} v_{gs} \ell_s/\mu_{g1}$ is large. The ratio of this velocity to the inlet gas velocity is $v_{gs}/v_{g1} = 6.53 \times 10^{-2} (r_h/r_c)^2/(\mathcal{M}^{1/2} \phi)$, where the factor 6.53×10^{-2} is the stoichiometric fuel-to-air mass ratio. The characteristic velocity of the gas downstream of the spray is $v_{gc} = (r_h/R)^2 v_{g1}$ in the absence of combustion, and larger than this by a factor $6.53 \times 10^{-2} q\phi/c_p T_1$, due to the thermal expansion of the gas, when combustion occurs. The ratio of the residence time of the gas in the chamber, $t_{gc} = L/v_{gc}$, to the chemical time $t_{ch} = (Be^{-T_0/T_1} Y_{o0})^{-1}$ is a Damkohler number, t_{gc}/t_{ch} , which must be larger than a certain critical value for ignition to occur.

In terms of the three control parameters of the problem,

$$t_s = 6.5 \times 10^{-4} \frac{Q/Q_b}{(T_1/T_0)^\kappa} \text{ s}, \quad n_0 = \frac{7.42 \times 10^9}{(Q/Q_b)^{1/2}} \text{ m}^{-3},$$

$$\frac{v_{g1}}{v_{p1}} = \frac{7.8 \times 10^{-3}}{\phi} \frac{Q}{Q_b} \frac{T_1}{T_0}, \quad \frac{v_{gs}}{v_{p1}} = 0.11 \left(\frac{Q}{Q_b} \frac{T_1}{T_0} \right)^{1/2},$$

leading to the dimensionless parameters

$$\left. \begin{aligned} \frac{\ell_s}{L} &= 0.17 \frac{Q/Q_b}{(T_1/T_0)^\kappa}, \quad \mathcal{M} = 1.16 \times 10^{-2} \frac{Q}{Q_b} \frac{T_1}{T_0} \\ Re &= 4.83 \times 10^2 \frac{(Q/Q_b)^{1/2}}{(T_1/T_0)^{1/2+\kappa}}, \quad \Pi = 6.33 \times 10^{-9} \frac{(Q/Q_b)^{3/2}}{(T_1/T_0)^\kappa} \\ \frac{t_{gc}}{t_{ch}} &= \frac{4.86 \times 10^5 \phi}{(Q/Q_b)(T_1/T_0)} \exp\left(-\frac{T_0}{T_1}\right), \quad \frac{T_0}{T_1} = 34.13, \quad \frac{q}{c_p T_0} = 1.35 \times 10^2. \end{aligned} \right\} \quad (17)$$

Thus, for values of the control parameters of order unity, the inlet velocity of the droplets is large compared to the inlet velocity of the gas, and the latter is somewhat smaller than the velocity of the gas induced by the drag of the droplets. The mass loading is fairly small, implying that fuel vapor saturation is unlikely even in the absence of combustion, and the Reynolds number of the gas flow is fairly large.

The small value of Π implies that electric forces do not play an important role in the motion of the droplets while their velocity is of the order of the inlet velocity v_{p1} . However, radial electric forces become important when the droplets slow down due to the aerodynamic drag, and lead to a characteristic mushroom shape of the spray plume which, as will be seen, has a significant effect on the reactive flow.

2.4. Numerical methods

The numerical treatment of the problem is analogous to that of Refs. [15] and [33]. As in those works, axisymmetric solutions are sought.

Given the large number of droplets present in the chamber, a method of particles [18,19] is used in which each particle represents a parcel of the one-droplet phase space containing a constant number of droplets. Each particle obeys the same equations (1)–(5) and (7) as the droplets it represents, but there may be many fewer particles in the discretization than droplets are in the real spray, the number of droplets per particle being a free parameter of the method. Each particle counts for as many droplets as it represents in the sums of equations (13). The statistical axisymmetry of the spray in the conditions of interest becomes exact axisymmetry for the method of particles formulation by assuming that a particle injected at a certain r amounts to $2\pi\xi$ identical droplets equispaced on a circumference of radius r [18]. The time between particle injections is then $2\pi\xi/\Phi$; see Refs. [15] and [33] for details.

Fig. 4. Mixing temperature at the chamber outlet scaled with T_0 as a function of the inlet gas temperature scaled with T_0 for $Q/Q_b = 1$, $\phi = 0.7$ (0); $Q/Q_b = 1$, $\phi = 0.35$ (1); $Q/Q_b = 1.69$, $\phi = 0.35$ (2); $Q/Q_b = 2.25$, $\phi = 0.35$ (3); and $Q/Q_b = 2.25$, $\phi = 0.35$ with satellite droplets (3', dashed). The upper branch of curve 3' is the short dashed segment at the right-hand side of the figure; its lower branch coincides with the lower part of curve 3.

The decomposition $\rho_g \mathbf{v}_g = \nabla \varphi_g + \nabla \times (\psi_g \mathbf{e}_\phi)$, with $\mathbf{e}_\phi$ a unit azimuthal vector, is carried to (8) and the curl of (9) to eliminate the pressure and obtain equations for the functions φ_g and ψ_g . These equations are discretized together with (6) and (10)–(12) using second order finite differences, and marched in time along with (1) using second order schemes until the conditions at the chamber outlet stabilize (the flow is never strictly time-independent because discrete particles are continuously injected into the chamber). A Crank–Nicolson method is used for the viscous, diffusion and conduction terms, and an Adams–Bashforth method is used for the rest. A nonuniform 240×240 grid, finer around the symmetry axis and the injection orifice, and a dimensionless time step in the range 5×10^{-4} – 2×10^{-3} are used.

The toroidal volumes generated by rotating the grid cells around the symmetry axis are used for δV , and the time step is used for δt in (13). The sums in these equations (actually in the equivalent dimensionless equations (A.14) for the particles of the particle method) are evaluated using a linear weighting particle-in-cell method [18].

The value of the numerical parameter η in (A.14), which is the number of droplets per particle scaled with $n_c L^3$ (see Appendix) is chosen for the mean number of particles in the computational domain to be in the order of 10^5 , as numerical tests show that the results are then independent of this number and of the grid.

Upon convergence, the magnitudes pertaining to the droplets defined in (A.19) are averaged over 10^4 additional time steps.

3. Results and discussion

The mixing temperature at the chamber outlet, defined as

$$T_m = \frac{\int_0^R \rho_g u_g T_g r dr}{\int_0^R \rho_g u_g r dr} \quad \text{at } x = L,$$

is shown in Fig. 4 as a function of the inlet air temperature, T_1 , for various values of the flow rate of fuel and the overall equivalence ratio. The results show frozen (lower) and intense combustion (upper) branches that end at ignition and extinction points, except for curve 3, which displays a fast but continuous transition with increasing T_1 . The dotted lines in the figure are not intended to represent intermediate branches, which are unstable and cannot be computed with the numerical methods used here; they are in-

Fig. 5. Results for $Q/Q_b = 2.25$, $\phi = 0.35$, and $T_1 = 523$ K (a), 723 K (b), 743 K (c) and 763 K (d). Shown are the dimensionless droplet number density (5 black contours equispaced between 500 and 2000 plus 4 contours equispaced between 10 and 300), some isotherms of the gas (red; dashed contours are for $T_g < T_1$ and solid contours for $T_g > T_1$; $\theta_g = (T_g - T_0)/(T_1 - T_0)$ is between 0.89 and 1 in (a), between 0.88 and 1.27 in (b), between 0.88 and 4.45 in (c), and between 0.88 and 5.13 in (d)), Y_r (10 grey levels between 0 (white) and 3.68×10^{-2} in (a), 5.57×10^{-2} in (b), 5.96×10^{-2} in (c), and 6.36×10^{-2} in (d)), the gas-phase reaction rate (green, in (c) and (d)), the local equivalence ratio (dotted, contours $3.52 Y_r / Y_0 = 0.25$, 0.5 and 0.75, increasing toward the symmetry axis, in (c) and (d)), and gas velocity arrows. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

cluded just to connect upper and lower branch pairs. The ignition and extinction points shift towards lower inlet air temperatures when the equivalence ratio increases (compare curves 0 and 1). They shift towards higher inlet air temperatures, and the range of multiplicity narrows and even disappears, when the flow rate of fuel increases (sequence of curves 1–3). The mixing temperature in the upper branch increases with the equivalence ratio. For complete combustion and in the absence of heat losses, this temperature should be independent of the flow rate of fuel; the small differences shown in Fig. 4 will be discussed below.

3.1. Structure of the flow

Figure 5 shows the structure of the flow in a plane through the axis of the chamber for $Q/Q_b = 2.25$ and $\phi = 0.35$ (curve 3 of Fig. 4) at various values of T_1 . The contours of constant droplet number density in the lower part of the figures show that the spray has a mushroom shape and its size increases when the inlet air temperature decreases. The increase of size is due to the increase of the vaporization time. The mushroom appearance is an effect of the electric repulsion of the charged droplets already reported in Ref. [15]. In the absence of vaporization and air motion, the droplets would stop at a distance $v_{p1} m_0 / c_{g1}$ from the injection orifice. Radial electric forces become important when the velocity of the droplets becomes small compared to v_{p1} , and lead to a localized increase of the spray cross-section. Another effect of the electric forces is the path reversal of some of the droplets at low inlet air temperatures, a phenomenon named fly back by Deng and

Fig. 6. Results in the upper branch for $Q/Q_b = 1.69$, $\phi = 0.35$, and $T_1 = 623$ K (a and b) and 773 K (c and d). Shown in (a) and (c) are the dimensionless droplet number density (7 black contours equispaced between 200 and 3000), some isotherms of the gas (red; dashed contours are for $T_g < T_1$ and solid contours for $T_g > T_1$; $\theta_g = (T_g - T_0)/(T_1 - T_0)$ is between 0.88 and 8.43 in (a), and between 0.93 and 6 in (c)), Y_r (10 grey levels between 0 (white) and 4.82×10^{-2} in (a) or 1.94×10^{-1} in (c)), and gas velocity arrows. Shown in (b) and (d) are Y_o (10 dashed blue contours equispaced between 0 and 0.23), the local equivalence ratio (green, contours $3.52 Y_r/Y_o = 0.2, 1$ and 2 , increasing toward the symmetry axis, which act as markers of the diffusion flame), and Y_r (same grey levels as in (a) and (c)). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Gomez [34]. Fly back is marked in Fig. 5(a) by the large droplet number density and the large fuel vapor concentration away from the symmetry axis in the lower part of the chamber. Fly back is due to the increase of the number of droplets in the spray when the inlet air temperature decreases and the vaporization last longer. The downward pointing electric field induced by the charge of these droplets around the injection orifice causes some newly injected droplets to fly back towards the base of the chamber.

Autoignition of the gaseous mixture generated by the vaporization of the spray first occurs at the top of the chamber. This leads to a lifted premixed flame that stabilizes at some distance above the spray, in a region where the velocity and temperature of the fresh gas vary smoothly with streamwise distance. Vaporization in this combustion mode occurs without any effect of the heat released by the flame. The flame shifts downward, to a point of higher gas velocity, when the inlet temperature of the gas increases, because this increases the final combustion temperature and the burning velocity of the flame. Eventually the flame reaches the vaporizing spray and combustion transitions to a different mode that is described below.

Some results for $Q/Q_b = 1.69$ and $\phi = 0.35$ (curve 2 of Fig. 4) are shown in Fig. 6. The size of the droplets and the inlet air velocity are smaller than in the case just discussed, and the droplets decelerate and vaporize faster. Only the lower part of the chamber is shown in Fig. 6. Fly back occurs in the lower branch at lower inlet air temperatures than before (results not displayed). Autoignition of the gas mixture also occurs at a lower air temperature than before, owing to the smaller velocity of the gas. In contrast with the previous case, autoignition now leads to a jump to a state of intense combustion. The combustion mode found before for very high values of T_1 exists now on the whole upper branch of curve 2. The structure of the flow in this mode consists of a lower region of intense reaction that locally depletes the oxygen and leaves

behind a ball of high temperature fuel vapor surrounded by a diffusion flame where this fuel burns with oxygen from the colder air stream that did not go through the lower reaction region. A weak lean wing flame exists in some cases that extends towards the air side from the head of the diffusion flame, but this wing is very short and does not seem to play an important role.

Combustion in the lower region occurs at the same time as vaporization, before the fuel vapor has had time to diffuse into the air stream. This fact accounts for the depletion of the oxygen in a globally lean system. The heat released by the chemical reaction increases the vaporization rate, which is faster than in the lower branch for the same value of the inlet air temperature.

The height of the lower reaction region above the injection orifice increases, and the size of the ball of high temperature fuel behind this region decreases, when the inlet air temperature decreases.

Extinction in this combustion mode occurs before the lower reaction region detaches from the spray. Figure 7 shows some variables along the symmetry axis for various values of the inlet air temperature. The reaction rate in the lower reaction region, shown in the lower part of Fig. 7(a), decreases with the inlet air temperature. However, this is not due to a decrease of the final combustion temperature, which increases slightly when the inlet air temperature decreases, but to a decrease of the fuel mass fraction in this region; see Fig. 7(b). The decrease is caused by a geometrical effect. As the inlet air temperature decreases and the lower reaction region shifts upward, it locates at a point where the cross-section of the spray is larger. Since the mass flux of fuel is a constant, the mass fraction attained upon vaporization decreases, until the reaction region cannot withstand the gas velocity and extinction occurs. The radial extent of the lower reaction region increases in the process, involving an increasing fraction of the air stream, and thus leaving behind less and less fuel. The radial

Fig. 7. Distributions of variables along the axis of the chamber for $Q/Q_b = 1.69$, $\phi = 0.35$, and $T_1/T_0 = 1.86, 1.96, 2.13, 2.30$ and 2.47 , increasing from right the left in each figure. (a) Temperature of the gas (solid) and reaction rate (dashed, to be read off the scale at the right-hand side of the figure). (b) Fuel vapor mass fraction (solid) and oxygen mass fraction (dashed).

divergence of the spray is an effect of the electric repulsion of the droplets, as was mentioned before.

The height of the lower reaction region above the base of the chamber is controlled by the vaporization of the liquid fuel. This height is very small when $Q/Q_b = 1$ and $\phi = 0.35$ (results not displayed) because the size of the droplets and the inlet air velocity are then very small. In these conditions, the lower reaction region loses heat to the base of the chamber, which accounts for the slight defect of mixing temperature in the upper branch of curve 1 of Fig. 4.

To further assess the influence of the electric forces, computations have been carried out assuming that the droplets are neutralized before they enter the chamber, but they are not otherwise modified. In the dimensionless formulation of the Appendix, this amounts to setting $\mathcal{P} = 0$ without changing other dimensionless parameters. Figure 8 compares results with and without droplet charge for a given set of parameter values. The lifted flame that exists with charged droplets changes to the other combustion mode when the charge is removed. The reason is that the radial dispersion of the spray is very small in the absence of charge, leading to a large concentration of fuel vapor that can sustain the locally rich lower reaction region.

Transition between the two modes can also be controlled by changing the radius of the injection orifice. The inlet velocity of the gas increases when this radius is decreased, which may lead to a

Fig. 8. Comparison of results for charged droplets (a) and uncharged droplets (b). Values of the control parameters are $Q/Q_b = 2.25$, $\phi = 0.35$ and $T_1 = 753$ K. Shown are the dimensionless droplet number density (5 black contours equispaced between 300 and 3500 plus 2 contours at 5 and 100), some isotherms of the gas (red; dashed contours are for $T_g < T_1$ and solid contours for $T_g > T_1$; $\theta_g = (T_g - T_0)/(T_1 - T_0)$ is between 0.88 and 4.76 in (a), and between 0.97 and 6.29 in (b)), Y_r (10 grey levels between 0 (white) and 6.16×10^{-2} in (a) or 2.38×10^{-1} in (b)), and the gas-phase reaction rate (green). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

lifted flame even for small values of the flow rate of fuel. Thus, numerical results for an injection orifice of radius $r_h = 1.9$ mm, which is half the radius considered above and leads to a four fold increase of the inlet air velocity for given values of the flow rate of fuel and the overall equivalence ratio, show (results not displayed) that the single premixed flame of Fig. 8(a) is already obtained for $Q/Q_b = 1$.

3.2. Effect of satellite droplets

Electrospray sources working in the cone-jet mode are known to generate droplets with narrow distributions of size [26], which justifies the assumption of a monodisperse population used in the injection condition (7). However, depending on the physical properties of the liquid, an equal number of satellite droplets may be generated together with the primary droplets in the atomization process. The results of Tang and Gomez [35] and

Deng and Gomez [34] indicate that this may be the case with electrosprays of heptane, the ratios of satellite-to-primary droplet radius and electric charge being about $1/3$ and $1/3^{3/2}$, respectively. Due to their small size, satellite droplets carry only small fractions of the spray mass and charge, and they decelerate and vaporize faster than primary droplets. Additional computations have been carried out with a modified form of the injection condition (7) that accounts for satellite droplets. Denoting by a_0 and e_0 the initial radius and charge of the primary droplets, and taking into account that the atomization process in an electrospray generates a sequence of alternate primary and satellite droplets, the number of droplet pairs injected per unit time is Φ' given by the volume conservation condition $\frac{4}{3}\pi[a_0^3 + (a_0/3)^3]\Phi' = Q$, and the injection condition replacing (7) is

$$\left. \begin{aligned} x_i &= 0, \quad r_i = \text{rand}[G(r)], \quad v_i = v_{pi} \mathbf{i} \\ a_i &= a_0, \quad T_i = T_0, \quad e_i = e_0 \quad \text{if } i \text{ is odd} \\ a_i &= \frac{1}{3}a_0, \quad T_i = T_0, \quad e_i = \frac{1}{3^{3/2}}e_0 \quad \text{if } i \text{ is even} \end{aligned} \right\} \quad \text{at } t = \frac{i}{(2\Phi')}.$$

Some results computed with this condition for $Q/Q_b = 2.25$ and $\phi = 0.35$ are shown in Fig. 4 (curve 3'). This curve features ignition and extinction points, and a short range of multiplicity. The increased fuel vapor concentration due to the early vaporization of the satellite droplets favors the presence of the lower reaction region of the second combustion mode. This mode is obtained in the upper branch of curve 3' for T_1 higher than about 735 K, where transition occurs to the combustion mode with a lifted premixed flame. This flame shifts upwards when T_1 is further decreased, until blow off, which is the mechanism of extinction in this combustion mode, occurs at about $T_1 = 715$ K. When $Q/Q_b = 1.69$, $\phi = 0.35$, vaporization of satellite droplets reduces the height of the lower reaction region above the injection orifice, but it does not lead to significant changes in curve 2.

3.3. Effect of the simplified kinetics

The single step mechanism used in this work is a strong simplification of any realistic kinetics and cannot be expected to give correct results in the wide range of conditions displayed by the numerical computations. This range spans autoignition of the gas mixture downstream of the spray (at the ignition or rapid transition points of Fig. 4), premixed flame propagation [in part of the high temperature region of curves 3 and 3', and in parts of the upper branches of curves analogous to 1 and 2 for smaller injection orifices (not shown)], and vaporization and combustion coexisting in a region of the chamber (in the second combustion mode described above). Therefore it is appropriate to discuss to what extent the computed results may be affected by the simplified kinetics.

The numerical results show that at ignition the chemical reaction first becomes important at the top of the chamber, where the spray has already vaporized and a heptane-air mixture has been formed. Since the frequency factor and activation energy of the Arrhenius reaction are determined from ignition delay time experiments at the pressure of the chamber (3 atm), the single step mechanism is expected to correctly describe the autoignition that occurs in the chamber.

Since the laminar flame speed of the single step mechanism is in good agreement with the speed computed with a detailed mechanism when $\phi = 0.35$ (cf. Fig. 3), the results of the single step mechanism are also expected to be reasonably accurate for the mode of combustion with a lean premixed flame lifted above the vaporizing spray. This includes the prediction of extinction, when it occurs in this mode through blow off of the premixed flame.

The single step mechanism cannot give quantitatively correct results for the second combustion mode, for which vaporization and combustion coexist, and more realistic mechanism would significantly increase the numerical burden. However, this mode

of combustion appears to be robust, in the sense that the overall burning rate is determined to a large extent by the rates of vaporization and mixing, and combustion can be expected to occur whenever the chemistry is sufficiently fast to keep pace with these processes and supply the necessary heat. To test these ideas, additional computations have been carried out replacing the Arrhenius law introduced in Section 2.2 with the single step mechanism of Westbrook and Dryer for heptane [30], which largely overpredicts the laminar flame speed at the pressure and temperatures of interest. The results show that when the original mechanism leads to the second combustion mode (e.g., in the upper branches of curves 0–2 of Fig. 4), the same occurs with the modified mechanism, with only small changes in the flow field. The modified mechanism extends the upper branches of these curves to smaller values of the inlet air temperature, revealing an uncertainty in the location of the extinction point that cannot be resolved with single step mechanisms. Similar results are obtained when the upper branch of curve 0 is recomputed with the frequency factor B multiplied by 4.84, which is the factor that brings the laminar flame speed of the single step mechanism to agree with the flame speed of the detailed mechanism [29] for $\phi = 0.7$. In summary, the results of the previous sections for the second combustion mode are expected to be reasonable away from the extinction point.

4. Conclusions

Steady gas-phase combustion in a mesoscale chamber fed with electrosprayed heptane and a coflow of preheated air through an injection orifice has been simulated numerically. The simulation includes the dispersion and vaporization of a dilute spray of small, electrically charged droplets of heptane and the combustion of the vaporized heptane. An Eulerian/Lagrangian method has been used that relies on the disparity of scales between the size of the droplets, the mean distance between droplets, and the size of the spray. Simple models have been used for the aerodynamic drag of the droplets, the conduction heat flux reaching their surfaces, their vaporization rate, and the loss of electric charge in Coulomb explosions. Combustion in the gas-phase has been modeled with a single step mechanism.

The effects of the inlet gas temperature (T_1), the fuel flow rate (Q) and the overall equivalence ratio (ϕ) have been analyzed. The mixing temperature evaluated at the chamber outlet as a function of the inlet air temperature features regions of frozen and intense combustion, with ignition and extinction values of T_1 that depend on Q and ϕ .

Two different combustion modes have been found. In one mode, combustion begins immediately upon fuel vaporization, in a layer of intense reaction that locally depletes the oxygen and leaves behind a region of high temperature fuel vapor surrounded by a diffusion flame. In the other mode, combustion occurs in a lean premixed flame located well above the spray, where the fuel has fully vaporized and mixed with the air. The first combustion mode depends on rapid vaporization of the fuel to generate a region of excess fuel vapor. The distance of the intense reaction layer to the injection orifice increases with the flow rate of fuel (which also increases the size of the droplets) until transition to the second combustion mode occurs at a certain value of this parameter. This transition can be controlled acting on the electric charge of the droplets, which determines the divergence of the spray and thus the maximum fuel vapor concentration, or on the radius of the injection orifice, which determines the inlet velocity of the gas for given values of Q and ϕ . The presence of small satellite droplets in the spray favors the first combustion mode.

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Appendix A. Dimensionless problem

Problem (1)–(16) can be written in dimensionless form scaling distances (x, r) with L , times with $t_c = m_0/c_{f_0}$, where $c_{f_0} = 6\pi\mu_{g_0}a_0$, velocities with $v_c = L/t_c$, droplet masses, radii and charges with m_0, a_0 and e_0 , the vaporization rates $\dot{m}_i$, forces $\mathbf{F}_i$ and heat fluxes q_{g_i} with $c_{f_0}, c_{f_0}v_c$ and $c_{f_0}c_p(T_1 - T_0)$, the electric field and electric potential with E_c and $E_c L$, where $E_c = \epsilon_0 n_c L / \epsilon_0$ with $n_c = \Phi / (v_c L^2)$, the gas density and pressure with ρ_{g_0} and $\rho_{g_0} v_c^2$, and introducing the scaled droplets and gas temperatures $\theta_i = (T_i - T_0) / (T_1 - T_0)$ and $\theta_g = (T_g - T_0) / (T_1 - T_0)$. Denoting the dimensionless variables with the same symbols used before for their dimensional counterparts, the problem takes the form

• Particles:

$$\frac{d\mathbf{x}_i}{dt} = \mathbf{v}_i, \quad a_i^3 \frac{dv_i}{dt} = \mathbf{F}_i + \mathcal{P} e_i \mathbf{E}_i \quad (\text{A.1})$$

$$\frac{da_i^3}{dt} = -\dot{m}_i, \quad a_i^3 \frac{d\theta_i}{dt} = \frac{c_p}{c} (q_{g_i} - \dot{m}_i \mathcal{L}_v) \quad (\text{A.2})$$

$$e_i \text{ decreases to } (1 - \alpha)e_i \text{ whenever } e_i \geq e_{r_i} = \text{Ra } a_i^{3/2} \quad (\text{A.3})$$

with

$$\begin{aligned} \dot{m}_i &= \frac{2/3}{\text{Pr}} \bar{k}_{g_i} a_i \lambda_i, \quad q_{g_i} = \frac{\dot{m}_i (\theta_{g_i} - \theta_i)}{e^{\lambda_i} - 1}, \quad \mathbf{F}_i = -\bar{k}_{g_i} a_i (\mathbf{v}_i - \mathbf{v}_{g_i}) \\ \lambda_i &= \frac{1}{\text{Le}_f} \ln \frac{1 - Y_{f_i}}{1 - Y_{f_{si}}}, \quad Y_{f_{si}} = \frac{25}{\exp\left(\beta_b \gamma \frac{\theta_b - \theta_i}{1 + \gamma \theta_i}\right) + 18} \end{aligned}$$

$$\bar{k}_{g_i} = (1 + \gamma \bar{\theta}_i)^\kappa, \quad \bar{\theta}_i = \frac{2\theta_i + \theta_{g_i}}{3} \quad (\text{A.4})$$

where, as before, $v_{g_i}, \theta_{g_i}, Y_{f_i}$ and $\mathbf{E}_i$ are the gas properties and the electric field interpolated to the position $\mathbf{x}_i$ of particle i .

The injection conditions

$$\begin{aligned} x_i &= 0, \quad r_i = \text{rand}[G(r)], \quad \mathbf{v}_i = v_{p_i} \hat{\mathbf{i}} \\ a_i &= 1, \quad \theta_i = 0, \quad e_i = 1 \end{aligned} \quad \text{at } t = i\eta. \quad (\text{A.5})$$

are used, with

$$G(r) = \frac{1 - \exp(-r^2/r_c^2)}{1 - \exp(-r_h^2/r_c^2)} \quad \text{for } r < r_h,$$

$$G(r) = 1 \quad \text{for } r > r_h,$$

where $\eta = 2\pi c_{f_0} \xi / m_0 \Phi$ is the number of droplets per particle scaled with $n_c L^3$.

• Gas, charged residues and electric potential:

$$\frac{\partial \rho_g}{\partial t} + \nabla \cdot (\rho_g \mathbf{v}_g) = \mathcal{F} S, \quad (\text{A.6})$$

$$\rho_g \frac{D\mathbf{v}_g}{Dt} = -\nabla p + \frac{1}{\mathcal{R}} \nabla \cdot \boldsymbol{\tau}' - \mathcal{F} \mathbf{F} + \mathcal{F} \mathcal{P} \rho_r \mathbf{E}, \quad (\text{A.7})$$

$$\rho_g \frac{DY_f}{Dt} = \frac{1}{\mathcal{R} \text{Pr} \text{Le}_f} \nabla \cdot (k_g \nabla Y_f) + (1 - Y_f) \mathcal{F} S - w, \quad (\text{A.8})$$

$$\rho_g \frac{DY_o}{Dt} = \frac{1}{\mathcal{R} \text{Pr} \text{Le}_o} \nabla \cdot (k_g \nabla Y_o) - \frac{88}{25} w, \quad (\text{A.9})$$

$$\rho_g \frac{D\theta_g}{Dt} = \frac{1}{\mathcal{R} \text{Pr}} \nabla \cdot (k_g \nabla \theta_g) - \mathcal{F} S_h + \frac{\chi}{\gamma} w, \quad (\text{A.10})$$

$$\frac{\partial \rho_r}{\partial t} + \nabla \cdot [\rho_g (\mathbf{v}_g + \mathcal{K} \mathcal{P} \mathbf{E})] = \mathcal{F} S_r, \quad (\text{A.11})$$

$$\nabla^2 \varphi = -\rho_e - \rho_r \quad \text{with } \mathbf{E} = -\nabla \varphi, \quad (\text{A.12})$$

with $D(\cdot)/dt = \partial(\cdot)/\partial t + \mathbf{v} \cdot \nabla(\cdot)$,

$$\begin{aligned} \rho_g (1 + \gamma \theta_g) [1 + (18/7) Y_f] &= 1, \quad \boldsymbol{\tau}' = k_g [\nabla \mathbf{v}_g + (\nabla \mathbf{v}_g)^T], \\ k_g &= (1 + \gamma \theta_g)^\kappa, \quad w = \rho_g \mathcal{D} \exp\left(\frac{\beta \theta_g}{1 + \gamma \theta_g}\right) Y_f Y_o \end{aligned} \quad (\text{A.13})$$

and

$$\begin{aligned} S &= \eta \frac{\sum \dot{m}_i}{\delta V}, \quad \mathbf{F} = \eta \frac{\sum [\mathbf{F}_i + \dot{m}_i (\mathbf{v}_{g_i} - \mathbf{v}_i)]}{\delta V}, \quad S_r = \eta \alpha \frac{\sum (e_i \delta t)}{\delta t \delta V}, \\ S_h &= \eta \frac{\sum [q_{g_i} + \dot{m}_i (\theta_{g_i} - \theta_i)]}{\delta V}, \quad \rho_e = \eta \frac{\sum e_i}{\delta V}, \end{aligned} \quad (\text{A.14})$$

where the sums are now over the particles of the particle method (see Section 2.4).

The boundary conditions

$$\left. \begin{aligned} v_g &= v_{g_i} \hat{\mathbf{i}}, \quad \theta_g = 1, \quad \varphi = 0 \\ \rho_g u_g Y_f - \frac{k_g}{\mathcal{R} \text{Pr} \text{Le}_f} \frac{\partial Y_f}{\partial x} &= 0 \\ \rho_g u_g Y_o - \frac{k_g}{\mathcal{R} \text{Pr} \text{Le}_o} \frac{\partial Y_o}{\partial x} &= \rho_{g_i} u_{g_i} Y_{o_i} \end{aligned} \right\} \text{ for } r < r_h$$

$$\left. \begin{aligned} v_g &= 0, \quad \frac{\partial Y_f}{\partial x} = 0, \quad \frac{\partial Y_o}{\partial x} = 0 \\ \theta_g &= 1, \quad \varphi = 0 \end{aligned} \right\} \text{ for } r_h < r < R$$

at $x = 0$,

$$v_g = \frac{\partial \omega_g}{\partial x} = \frac{\partial Y_f}{\partial x} = \frac{\partial Y_o}{\partial x} = \frac{\partial \theta_g}{\partial x} = \varphi = 0 \quad (\text{A.16})$$

at $x = 1$, and

$$\frac{\partial v_g}{\partial r} = v_g = \frac{\partial Y_f}{\partial r} = \frac{\partial Y_o}{\partial r} = \frac{\partial \theta_g}{\partial r} = \frac{\partial \varphi}{\partial r} = 0 \quad (\text{A.17})$$

at $r = R$, are imposed.

Apart from the numerical parameter η , the problem contains the 24 dimensionless physical parameters

$$\left. \begin{aligned} \mathcal{P} &= \frac{m_0^2 \epsilon_0^2 \Phi}{\epsilon_0 v_c^3 L^3}, \quad \mathcal{F} = \frac{m_0^2 \Phi}{\rho_{g_0} c_{f_0} L^3}, \quad \mathcal{R} = \frac{\rho_{g_0} c_{f_0} L^2}{m_0 \mu_{g_0}}, \\ \text{Pr} &= \frac{\mu_{g_0} c_p}{\lambda_{g_0}}, \quad \text{Le}_f = \frac{\lambda_{g_0}}{c_p \rho_{g_0} D_{f_0}}, \quad \text{Le}_o = \frac{\lambda_{g_0}}{c_p \rho_{g_0} D_{o_0}}, \\ \gamma &= \frac{T_1 - T_0}{T_0}, \quad \frac{c_p}{c}, \quad Y_{o_0}, \quad \kappa, \quad \beta_b = \frac{T_b}{T_0}, \quad \mathcal{D} = \frac{m_0 B}{c_{f_0}} e^{-\beta_b}, \quad \chi = \frac{q}{c_p T_0}, \\ \beta_b &= \frac{L_v}{K_f T_b}, \quad \mathcal{L}_v = \frac{L_v}{c_p (T_1 - T_0)}, \quad \theta_b = \frac{T_b - T_0}{T_1 - T_0}, \\ v_{p_i} &= \frac{m_0 v_{p_i}}{c_{f_0} L}, \quad v_{g_i} = \frac{m_0 v_{g_i}}{c_{f_0} L}, \quad R = \frac{R^*}{L}, \quad r_h = \frac{r_h^*}{L}, \quad r_c = \frac{r_c^*}{L}, \\ \alpha, \text{ Ra} &= \frac{8\pi (\epsilon_0 \sigma a_0^3)^{1/2}}{e_0}, \quad \mathcal{K} = \frac{c_{f_0} K}{e_0}. \end{aligned} \right\} \quad (\text{A.18})$$

The asterisks in $v_{p_i}^*, v_{g_i}^*, R^*, r_h^*$ and r_c^* are used to temporarily denote dimensional magnitudes. The flow rate of heptane injected in the chamber is $Q = \frac{4}{3} \pi a_0^3 \Phi$. The dimensionless inlet gas velocity can be written in terms of the overall equivalence ratio ϕ as

$$v_{g_i} = \frac{88}{25\pi} \frac{(1 + \gamma) \mathcal{F}}{Y_{o_0} r_h^2 \phi}.$$

For the base case of Section 2.3, the values of these parameters are $\mathcal{P} = 5.064 \times 10^{-4}$, $\mathcal{F} = 2.715 \times 10^{-6}$, $\mathcal{R} = 1.538 \times 10^5$, $\text{Pr} = 0.7$, $\text{Le}_f = 2.6$, $\text{Le}_o = 1$, $c_p/c = 0.446$, $Y_{f_0} = 0.23$, $\kappa = 0.75$, $\beta_b = 34.13$, $\mathcal{D} = 2.205 \times 10^{-12}$, $\chi = 134.72$, $\beta_b = 11.82$, $\mathcal{L}_v = 1.246/\gamma$, $\theta_b = 0.2676/\gamma$, $v_{p_i} = 0.17$, $v_{g_i} = 0.247(1 + \gamma)/\phi$, $R = 0.14$,

$r_h = 0.1$, $r_c = 0.021$, $\alpha = 0.3$, $Ra = 2.548$, and $\mathcal{K} = 16.345$. The variation of the dimensionless parameters with Q/Q_b follows immediately from the scaling laws of Section 2.3.

The mean number density, radius, density, velocity and temperature of the droplets are

$$\begin{aligned} n_d &= \eta \frac{\sum 1_i}{\delta V}, & a_d &= \frac{1}{n} \eta \frac{\sum a_i}{\delta V}, \\ \rho_d &= \eta \frac{\sum a_i^3}{\delta V}, & \mathbf{v}_d &= \frac{1}{\rho_d} \eta \frac{\sum a_i^3 \mathbf{v}_i}{\delta V}, & \theta_d &= \frac{1}{\rho_d} \eta \frac{\sum a_i^3 \theta_i}{\delta V}, \end{aligned} \quad (\text{A.19})$$

where $\sum 1_i$ is the number of particles in δV .

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