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Published online: 14 Jun 2013.

To cite this article: John Sisti & Paul E. DesJardin (2013): A semi-analytical, multizone model of droplet combustion with varying properties, Combustion Theory and Modelling, DOI:10.1080/13647830.2013.791725

To link to this article: http://dx.doi.org/10.1080/13647830.2013.791725

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A semi-analytical, multizone model of droplet combustion with varying properties

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(Received 30 October 2012; final version received 4 February 2013)

A multizone droplet burn model is developed to account for changes in the thermal and transport properties as a function of droplet radius. The formulation is semi-analytical – allowing for accurate and computationally efficient estimates of flame structure and burn rates. Zonal thermal and transport properties are computed using the Cantera software, pre-tabulated for rapid evaluation during run-time. Model predictions are compared to experimental measurements of burning n-heptane, ethanol and methanol droplets. An adaptive zone refinement algorithm is developed that minimizes the number of zones required to provide accurate estimates of burn time without excess zones. A sensitivity study of burn rate and flame stand-off with far-field oxygen concentration is conducted with comparisons to experimental data. Overall agreement to data is encouraging with errors typically less than 20% for predictions of burn rates, stand-off ratio and flame temperature for the fuels considered.

Keywords: droplet combustion; variable properties; multizone; n-heptane; ethanol; methanol

Nomenclature

\( B_q \) Thermal transport number [–]
\( C_P \) Mass weighted specific heat [J kg\(^{-1}\) K\(^{-1}\)]
\( D \) Diffusivity coefficient [m\(^2\) s\(^{-1}\)]
\( K \) Gasification rate [mm\(^2\) s\(^{-1}\)]
\( h \) Enthalpy [J kg\(^{-1}\)]
\( \Delta h_C \) Heat of combustion [J kg\(^{-1}\)]
\( k \) Thermal conductivity [W m\(^{-1}\) K\(^{-1}\)]
\( L \) Fuel–Oxidiser concentration factor [–]
\( L'_{\text{vap}} \) Effective latent heat of vaporisation [J kg\(^{-1}\)]
\( \text{Le} \) Lewis number [–]
\( m \) Mass [kg]
\( \dot{m} \) Mass flow rate [kg s\(^{-1}\)]
\( N \) Number of zones [–]
\( \dot{q}'' \) Heat flux [W kg\(^{-1}\) m\(^{-2}\)]
\( \dot{Q} \) Heat flow rate [W]
\( r \) Radius [m]
\( r^* \) Surface normalised radius [–]

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1. Introduction

The topic of droplet combustion has a rich history. Reviews and texts on droplet combustion include those of Faeth [1, 2], Sirignano [3, 4], Williams [5, 6], Law [7], Kuo [8], Turns [9] and Glassman [10]. Early theories of droplet burning came from Spaulding [11], Godsave [12] and Lorell et al. [13, 14] and were based on either single or two-zone descriptions with constant properties within each zone. While these early theories are elegant and relatively simple to formulate, they are also well known to greatly over-predict the flame radius and under-predict burn rates [7, 15]. Since flame radii and burn rates are a major source of data from droplet experiments, it is important to have a droplet burn model that is sufficiently accurate to predict these quantities. The major source of the modelling assumption error leading to the exaggerated flame radius is the dependence of thermal and transport properties on radius, stemming from changes in composition and temperature. Many enhancements to these early theories have improved the description of property variation either through improved analytics [16, 17] or direct numerical integration [18].

For the current effort, it is highly desirable the droplet model be computationally efficient since its intended use is in a subgrid scale (SGS) model for large eddy simulations of
turbulent reacting, multiphase flows in a dilute limit [19, 20]. Current models for droplet burning usually fall into two categories. Models in the first category are detailed numerical integration, where the governing equations are numerically discretised, linearised and solved, e.g. [18, 21–24]. These computations are often demanding since sufficient resolution is required to approximate the gradients of temperature and composition. While these models are valuable for understanding quasi-steady burning behaviour of isolated droplets, they contain far too many degrees of freedom for incorporation into a larger spray calculation for which millions of droplets (or groups of droplets) are transported.

Models in the second category are entirely analytical. This class of model relies on layers of approximations to obtain a closed form solution. This category includes the model developed by Law and Law [17] to account for variable transport properties. In their model, the transport properties are first assumed separable in terms of their temperature and composition dependence. The dependence of each function is assumed to follow a particular form (often linear). The functional form of the mixing rules for transport properties also has to be chosen with care to allow a tractable result. At the end of the analysis, numerical integration is still required for the most general description.

A key finding from these previous studies is the importance of accounting for changes in thermal and transport properties to predict burning rate, flame stand-off and flame temperature accurately. The objective of the present study is to devise a hydrocarbon droplet model that allows for a general description of property variation, yet is computationally efficient so it may be used in larger scale spray simulations. The approach is based on a newly developed multizone, semi-analytical approach where local convection–diffusion solutions are used to describe the variation of temperature and composition (species mass fractions) within each zone. Analytical results are derived to determine the flame temperature and stand-off ratio in the context of a flame-sheet approximation which allows for the use of an adaptive, flame-fitted zone ‘mesh’. The solution algorithm for the resulting eigenvalue problem is straightforward, requiring no matrix inversion and therefore is computationally efficient. The multizone description can also potentially be dynamically adjusted so as to reduce the degrees of freedom to the limit of a two-zone model where accurate local estimates of burn rates are not required.

The remainder of this study begins with the mathematical formulation of the multizone model. Results are then presented with comparison to experimental data. Finally, conclusions are drawn and a summary of findings is presented.

2. Model formulation

The particle is assumed to be spherically symmetric with an infinitely thin flame sheet located at \( r = r_f \) (see Figure 1). The single step reaction mechanism is simply \( m_F + v_O m_O \rightarrow \sum v_p m_p \). Assuming the species only diffuse from concentration gradients (Fickian diffusion), the system of ordinary differential equations describing the steady-state flow in the inner and outer regions of the flow are

\[
Y_i \dot{m} - 4\pi r^2 \rho D_i \frac{dY_i}{dr} = \dot{m}_i \tag{1a}
\]

\[
\sum h_i \dot{m}_i - 4\pi r^2 k \frac{dT}{dr} = \dot{Q} \tag{1b}
\]
where \( \dot{m}_i \) is the mass flow of the \( i \)th species, \( \dot{m} \) is the overall mass flow rate and \( \dot{Q} \) is the overall energy flow rate (to be defined). The diffusion coefficient \( D_{i-m} \) represents the diffusion of the \( i \)th species in the mixture. Note, Equation (1a) doesn’t automatically satisfy overall mass conservation, i.e. \( \sum \dot{m}_i \neq \dot{m} \), which serves as a constraint for determining the diffusion coefficients. Various approximations exist for constraining \( D_{i-m} [25, 26] \). Unfortunately, none of the analytical methods allow for each species to have its own diffusion coefficient and still satisfy overall mass conservation. The simplest approximation is to assume all species have equal diffusion coefficients. The current study assumes this to be the case. The variation in transport properties due to composition and temperature changes are accounted for by decomposing the radial distribution of properties into an arbitrary number of zones, as shown in Figure 1. Zones 1 through \( f \) reside in the inner region, between the droplet surface and the flame. Zones \( f+1 \) through \( \infty \) reside in the outer region, between the flame and the far-field. The outer and inner boundaries of each zone, or shells, are defined as \( r_k \) and \( r_{k-1} \) where \( k \) is the zone number. At the droplet surface, \( k = 0 \equiv s \) (i.e. \( r_0 = r_s \)). For the inner region, \( s < k \leq f \) and \( D_{i-m} = D_{F-m} \), where \( D_{F-m} \) is the diffusivity of the fuel into the rest of the mixture. For the outer region, \( f < k \leq \infty \) and \( D_{i-m} = D_{O-m} \), where \( D_{O-m} \) is the diffusivity of the oxidiser (\( O_2 \)) into the rest of the mixture.

Within each zone, the thermal and transport properties are assumed constant. The term \( \sum h_i \dot{m}_i \) in Equation (1b) may be expressed as \( \dot{m} \sum \epsilon_i C_{P,i} T = \dot{m} \Lambda P T \), where \( \epsilon_i \equiv \dot{m}_i/\dot{m} \) is a mass flow rate ratio, and \( \Lambda P = \sum \epsilon_i C_{P,i} \) is a mass flow-rate weighted specific
heat. Note, \( \Lambda_p \) is not equal to the commonly used mass fraction weighted definition, i.e. \( \Lambda_p \neq C_p = \sum Y_i C_{p,i} \). For closure of the eigenvalue problem, \( \epsilon_i \) is specified as a constraint for all species in the inner and outer regions. Equations (1a) and (1b) can be readily integrated to determine the composition and temperature distributions within each zone:

\[
\frac{Y_{i,k} - \epsilon_{i,k}}{Y_{i,k} - \epsilon_{i,k}} = \exp \left[ -\frac{\dot{m}}{4\pi} \frac{1}{\Gamma_{i,k}^*} \left( \frac{1}{r_{k-1}^*} - \frac{1}{r_k^*} \right) \right] \quad (2a)
\]

\[
\frac{\Lambda_{p,k} T_{k-1} - \dot{Q}_k}{\Lambda_{p,k} T_k - \dot{Q}_k / \dot{m}} = \exp \left[ -\frac{\dot{m}}{4\pi} \frac{\Lambda_{p,k}}{k_k^*} \left( \frac{1}{r_{k-1}^*} - \frac{1}{r_k^*} \right) \right], \quad (2b)
\]

where \( \Gamma_{i,k} = \rho D_{l,m,k} \). The subscripts \( k \) or \( k - 1 \) indicate shell (\( r, Y_i, T \)) or zone (\( \Gamma, \Lambda_p, k, \epsilon_i, \dot{Q} \)) properties. Zone \( k \) is bounded by shells \( k - 1 \) and \( k \).

The quantities \( \epsilon_{i,k} \) and \( \dot{Q}_k / \dot{m} \) are the mass and heat flow rates for each zone. They are constant in each region (i.e. \( \epsilon_{i,k} = \epsilon_{i,in} \) in the inner region). From overall mass conservation, \( \dot{m} = \dot{m}_{F,s} = 4\pi r_s^2 \dot{m}_{F,s} \) is constant for all zones and can be shown to be equal to the fuel burn rate, via a mass balance at the fuel surface. From species conservation, \( \epsilon_{F,k} = 1 \) for the fuel and \( \epsilon_{i,k} = 0 \) for all other species in the inner region since the product and inert species are assumed insoluble in the fuel (i.e. single-fuel droplets and water dilution for alcohols is neglected). In the outer region, \( \epsilon_{F,k} = 0 \) (thin flame assumption) and \( \epsilon_{i,k} = \nu_i \) for the remaining reacting species, where \( \nu_i \) is the amount of species produced or consumed per unit mass of fuel. For the inert species (\( N_2 \)), \( \epsilon_{i,k} = 0 \), for both the inner and outer regions.

From an energy balance at the fuel surface, \( \dot{Q}_1 / \dot{m} = \Lambda_{p,1} T_k - L_vap \), where \( L_vap = L_vap - \dot{q}_v'' / \dot{m}'' \) is the effective latent heat of vaporisation with \( \dot{q}_v'' = -k_l dT/dr|_{l,s} \) being the heat flux to the surface from the liquid. Neglecting heat losses from thermal radiation and finite-rate chemistry, for the inner regions, \( \dot{Q}_1 = \dot{Q}_2 = \cdots = \dot{Q}_k = \dot{Q}_{in} \), where \( \dot{Q}_{in} \) represents the energy flow from the ‘fuel side’ of the flame. Employing an energy balance across the flame surface then \( \dot{Q}_{f+1} / \dot{m} = \dot{Q}_f / \dot{m} + \Delta h_C = \Lambda_{p,1} T_s - L_vap + \Delta h_C \), where \( \Delta h_C \) is the heat of combustion. Similarly for the outer regions, \( \dot{Q}_{f+1} = \dot{Q}_{f+2} = \cdots = \dot{Q}_{out} \), where \( \dot{Q}_{out} \) is the flow of energy from the ‘oxidiser side’ of the flame.

Taking multiple ratios of Equation (2a) for the fuel through the inner zones and multiple ratios for the oxidiser through the outer zones, the following relations can be derived:

\[
1 - Y_{F,s} = \exp \left\{ -\frac{\dot{m}}{4\pi r_s} \left[ \frac{1}{\Gamma_{F,f}} \left( \frac{1}{r_{f-1}^*} - \frac{1}{r_f^*} \right) + \sum_{k=1}^{f-1} \frac{1}{\Gamma_{F,k}} \left( \frac{1}{r_{k-1}^*} - \frac{1}{r_k^*} \right) \right] \right\} \quad (3a)
\]

\[
\frac{\nu_O}{\nu_O + Y_{O,\infty}} = \exp \left\{ -\frac{\dot{m}}{4\pi r_s} \left[ \frac{1}{\Gamma_{O,f+1}} \left( \frac{1}{r_f^*} - \frac{1}{r_{f+1}^*} \right) + \sum_{k=f+2}^{\infty} \frac{1}{\Gamma_{O,k}} \left( \frac{1}{r_{k-1}^*} - \frac{1}{r_k^*} \right) \right] \right\} \quad (3b)
\]
where \( r^* = r/r_s \) is the normalised radial distance. Taking the ratio of these expressions and solving for \( r_f^* \) provides an expression for the flame stand-off ratio,

\[
r_f^* = \frac{L}{\Gamma_{O,f+1}} + \frac{1}{\Gamma_{F,f}} \frac{1}{r_f^*} - \frac{1}{r_s^*} + \sum_{k=1}^{f-1} \left( \frac{1}{\Gamma_{F,k} r_{k-1}^*} - \frac{1}{r_k^*} + \frac{1}{\sum_{j=2}^{f} \Gamma_{O,K} \left( \frac{1}{r_{k-1}^*} - \frac{1}{r_k^*} \right)} \right),
\]

(4)

where the factor \( L \equiv \ln(1 - Y_{F,s})/\ln[v_O/(v_O + Y_{O,\infty})] \) depends only on the concentrations of the fuel at the surface and oxidiser at the far-field.

Taking the ratio of Equations (2a) and (2b) provides a relationship between a given species and the temperature within a zone:

\[
\frac{Y_{i,k-1} - \epsilon_{i,k}}{Y_{i,k} - \epsilon_{i,k}} = \left( \frac{\Lambda_{P,k} T_{k-1} - \dot{Q}_k/m}{\Lambda_{P,k} T_k - \dot{Q}_k/m} \right)^{\text{Le}_{i,k}},
\]

(5)

where \( \text{Le}_{i,k} \) is Lewis number for the \( i \)th species in the \( k \)th zone. Using the equal diffusivity species assumption, \( \text{Le}_{i,k} = \text{Le}_{F,k} = k_k / (\rho \Lambda_P D_{F-m})_k \) for all species in the inner region and \( \text{Le}_{i,k} = \text{Le}_{O,k} = k_k / (\rho \Lambda_P D_{O-m})_k \) for all species in the outer region. Taking multiple ratios of Equation (5) through the zones in the outer region with \( i = O \), the following relation is obtained:

\[
\frac{v_O}{v_O + Y_{O,\infty}} = \prod_{k=f+1}^{\infty} \left( \frac{\Lambda_{P,k} T_{k-1} - \dot{Q}_{out}/m}{\Lambda_{P,k} T_k - \dot{Q}_{out}/m} \right)^{\text{Le}_{O,k}},
\]

(6)

which is used to solve for the flame temperature explicitly:

\[
T_f = \left( \frac{\Lambda_{P,f+1} T_{f+1} - \dot{Q}_{out}/m}{\Lambda_{P,f+1}} \right)^{1/\text{Le}_{O,f+1}} \times \left[ \frac{1 + Y_{O,\infty}}{v_O} \right]^{-1/\text{Le}_{O,f+1}} \prod_{k=f+2}^{\infty} \left( \frac{\Lambda_{P,k} T_{k-1} - \dot{Q}_{out}/m}{\Lambda_{P,k} T_k - \dot{Q}_{out}/m} \right)^{-\text{Le}_{O,k}/\text{Le}_{O,f+1}} \frac{\dot{Q}_{out}/m}{\Lambda_{P,f+1}},
\]

(7)

In the limit of two-zones, the properties in the inner and outer zones are equal, respectively, therefore \( \Gamma_{F,k} = \Gamma_{F,in}, \Gamma_{O,k} = \Gamma_{O,out}, \Lambda_{P,k} = \bar{\Lambda}_{P,in}, \Lambda_{P,k} = \bar{\Lambda}_{P,out}, \text{Le}_{F,k} = \bar{\text{Le}}_{F,in} \) and \( \text{Le}_{O,k} = \bar{\text{Le}}_{O,out} \), where the \( \bar{\text{()}} \) notation represents an average over either the inner or
outer regions. Making these substitutions, Equations (4) and (7) reduce to

\[ r_f^* = 1 + \frac{\ln \left(1 - Y_{F,s}\right)^{-\frac{1}{\ln \left(1 + Y_{O,\infty}/v_O\right)}}}{\ln \left(1 + Y_{O,\infty}/v_O\right)} \]  

(8a)

\[ T_f = \frac{T_{\infty}}{(1 + Y_{O,\infty}/v_O)^{1/\bar{L}_{\text{e},\text{out}}}} + \frac{\Delta h_C - \tilde{\Lambda}_{P,\text{in}} T_s + L'_\text{vap} \left[1 - \frac{1}{1 + Y_{O,\infty}/v_O}\right]}{\tilde{\Lambda}_{P,\text{out}}} \],

(8b)

which is the same as previous two-zone model descriptions [7, 16].

3. Numerical implementation

Equations (2), (4) and (7), along with a relation for the fuel vapour-pressure curve, represent a coupled system of equations for the solution of \(Y_i, T\) and \(r_f^*\). The solution of these equations employs an adaptive zone mesh that is described in terms of the number of zones in the inner region (\(N_{\text{in}}\)) bounded between \(r_s^*\) and \(r_f^*\). The number of zones in the outer region (\(N_{\text{out}}\)) is bounded between \(r_f^*\) and \(r_s^*\). The last shell at \(r_s^*\) is set to an arbitrarily large number (\(N_{\text{two-zone}} \times 10^{30}\)) corresponding to the ‘far-field’. The algorithm for determining the eigenvalue \(\dot{m}\) starts with the initialization of \(T, Y_i\) and the zone properties from a two-zone model estimate in which \(N_{\text{in}} = N_{\text{out}} = 1\). The number of zones needed in the inner and outer regions are first determined from the two-zone model by specifying a maximum temperature variation within a zone, \(\Delta T_{\text{max}}\). \(\Delta T_{\text{max}} \geq |T_k - T_{k-1}| = \Delta T\) when the two-zone model is divided up into \(N_{\text{in}}\) inner zones and \(N_{\text{out}}\) outer zones and \(\Delta T\) is constant in each region. An adaptive zone mesh is used to position the shells. In this approach, a normalised radial distance \(r^*\) is mapped to a regularised temperature space \(\eta\). In the inner region, \(r_{\text{in}}^* = (r^* - r_s^*)/(r_f^* - r_s^*)\) and \(\eta_{\text{in}} = (T - T_s)/(T_f - T_s)\). In the outer region, \(r_{\text{out}}^* = (r^* - r_s^*)/(r_f^* - r_s^*)\) and \(\eta_{\text{out}} = (T_f - T)/(T_f - T_\infty)\). The motivation for mapping with respect to \(r^*\) instead of \(r\) is because \(r^*\) is a strong function of \(r_f^*\) (in \(\eta\)-space) but \(r^*\) is not. \(r^*\) only needs to be calculated once. Shell locations are defined in \(\eta\)-space as \(\eta_k = k \Delta \eta\), where \(k\) is the shell number and \(\Delta \eta = 1/N\). The position of \(r_{\text{in}}^*\) is determined by substituting the definitions of \(r^*\) and \(\eta\) into Equation (2b) and using the properties of the two-zone estimate, resulting in the following expressions for the inner zones:

\[ r_{\text{in},k}^* = \frac{1}{r_f^* - r_s^*} \left[ \frac{\tilde{\Lambda}_{P,\text{in}} \left[ \eta_{\text{in},k} \left( T_f - T_s \right) + T_s - \dot{Q}/\dot{m} \right]}{\bar{\Lambda}_{P,\text{in}} T_f - \dot{Q}/\dot{m}} \right]^{-1} - r_s^* \]

(9)

and for the outer zones:

\[ r_{\text{out},k}^* = \frac{1}{r_f^* - r_s^*} \left[ \frac{\tilde{\Lambda}_{P,\text{out}} \left[ \eta_{\text{out},k} \left( T_\infty - T_f \right) + T_f - \dot{Q}/\dot{m} \right]}{\bar{\Lambda}_{P,\text{out}} T_f - \dot{Q}/\dot{m}} \right]^{-1} - r_f^* \]

(10)

Once \(r^*\) is calculated, \(r^*\) is determined using the definition of \(r^*\), i.e. \(r_{\text{in}}^* = r_f^* + (r_f^* - r_s^*)r_{\text{in}}^*\). Figure 2 illustrates the use of the adaptive mapping showing (a) the allocation of

\[ \text{(Continued on next page…)} \]
zones in $r^*$ space and (b) the mapping of $r^{*\in}$ in terms of $\eta_{\text{in}}$. As shown in Figure 2(a), the shells are near the surface where the temperature gradient is higher. In the outer region, the mapping results in shells being placed near the flame.

Figure 3 is a flowchart of the overall algorithm for solving the eigenvalue problem. A three-level nested iterative method is used to solve for composition, temperature field, droplet burn rate, flame stand-off ratio and flame temperature. From outer to inner, the loops are the $Y_{F,s}$-loop (the Illinois algorithm [27, 28]), the properties-loop (the Jacobi iteration) and the $r^{*f}$-loop (the secant method). The initial state of the $Y_{F,s}$-loop, properties-loop and $r^{*f}$-loop are supplied by the solution of the two-zone model (i.e. $Y_{F,s}$, $r^{*f}$ and the properties) and $r^*$ calculated from the adaptive mesh. The surface temperature is computed from vapour pressure relations given by Ewing and Sanchez Ochoa [29] for $n$-heptane and Antoine [30] for ethanol and methanol (coefficients from NIST [31]). As indicated by Equation (4), the flame stand-off $r^{*f}$ is a weighted sum of the the properties and locations of the remaining zones. The zone locations, $r^*_k$, in turn, depend on the flame stand-off ratio, therefore iteration is required using Equation (4) to compute $r^{*f}$. Once the $r^{*f}$-loop has converged, the mass flow rate $\dot{m}$ may be determined by using either Equation 3(a) or Equation 3(b) to provide a value for $\dot{m}$:

$$
\dot{m} = \left\{ \begin{array}{ll}
-4\pi r_s \frac{\ln(1 - Y_{F,s})}{\sum_{k=1}^{f}(1/r^{*e}_k - 1/r^{*\in}_k)/\Gamma_{F,k}} \\
4\pi r_s \frac{\ln(1 + Y_{O,\infty}/\nu_O)}{\sum_{k=f+1}^{\infty}(1/r^{*e}_k - 1/r^{*\in}_k)/\Gamma_{O,k}}
\end{array} \right.
$$

(11)

$T_f$ is determined using Equation (7) and an estimate of droplet surface temperature. The temperature field is decoded using Equation (2b), sweeping from $r^{*f}$ to both $r^{*e}$ and $r^{*\in}$. The species are decoded using Equation (2a), sweeping from $r^{*f}$ to $r^{*e}$ and $r^{*\in}$ for $i = F$ and $i = O$, respectively. The remaining species are swept from $r^{*\in}$ to $r^{*e}$.

Once the revised temperature and species distributions are known, the mixture weighted thermal and transport properties are updated using a tabulated database. The database is constructed a priori using the Cantera software [32]. Species properties (i.e. $C_p$, $k$, $D_{ij}$, etc.), are tabulated for each temperature and pressure between 300 and 3000 K and 0.5 and 30 atm. XML formatted tables are created by reading CHEMKIN formatted mechanism files with thermal and transport data. The $n$-heptane data comes from the Combustion Research Group at the University of California, San Diego [33]. The ethanol and methanol data comes from Lawrence Livermore National Laboratory [34]. The size of the tables are dynamically sized such that interpolation errors are less than 0.1% over the entire range of temperature and pressure. Ideal gas mixture rules are used to compute thermal properties. Mixing rules of Mathur et al. [35] and Bird et al. [36] are used to determine bulk diffusion and thermal conductivity properties.

Using the updated properties, $r^{*f}$ and $r^*$ must be calculated again using the $r^{*e}$-loop. The property-loop is iterated until $r^*$ is no longer changing. The error is determined using an $\ell^2$-norm of $r^*$ between the most recently calculated values and the values from the beginning of the most recent property-loop iteration.

Lastly, a revised value of $Y_{F,s}$ is determined using Equation (5):

$$
Y_{F,s} = 1 - \prod_{k=1}^{f} \left( \frac{\Lambda_{p,k} T_{k-1} - \dot{Q}_{\text{in}}/\dot{m}}{\Lambda_{p,k} T_k - \dot{Q}_{\text{in}}/\dot{m}} \right)^{\Lambda_{F,k}}.
$$

(12)
Figure 2. Illustration of mesh adaptivity showing (a) shell positions and (b) $r^*_m$ versus $\eta_m$ for $\Delta T_{\text{max}} = 100 \text{ K}$.
which is compared to the initial guess of $Y_{F,s}$ and iterated until an error tolerance of $1 \times 10^{-8}$ is satisfied. Further step-by-step details on the iterative solution procedure are summarised in Appendix A.

4. Results and discussion

The burning of $n$-heptane, ethanol and methanol droplets over a range of oxygen environments is explored. These fuels are selected because of the wealth of experimental data available in the literature on burn rates ($K$) and flame stand-off ratio ($r_f^*$). For each fuel, a zone refinement study is conducted to ensure the final reported results are zone independent.
Figure 4. Error in $K$, $r_f^*$ and $T_f$ versus $\Delta T_{\text{max}}$ and $N$ for (a) $n$-heptane, (b) ethanol and (c) methanol. For all cases, $T_\infty = 298$ K, $P_\infty = 1$ atm and $Y_{O,\infty} = 0.23$. 
Table 1. Burning rates, stand-off ratios, flame and surface temperatures for selected fuels at selected states. The two-zone calculation is denoted by parentheses ( ).

<table>
<thead>
<tr>
<th>Fuel</th>
<th>( Y_{O,\infty} )</th>
<th>( T_{\infty} (K) )</th>
<th>( K ) (mm²s⁻¹)</th>
<th>( r_f^* )</th>
<th>( T_f (K) )</th>
<th>( T_s (K) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( n )-Heptane</td>
<td>0.23</td>
<td>298</td>
<td>0.53 (0.99)</td>
<td>0.64 [37]</td>
<td>11.0 (20.0)</td>
<td>2247 (2286)</td>
</tr>
<tr>
<td></td>
<td>0.23</td>
<td>1200</td>
<td>0.64 (1.24)</td>
<td>1.02 [49]</td>
<td>9.2 (18.4)</td>
<td>2859 (2889)</td>
</tr>
<tr>
<td>Ethanol</td>
<td>0.23</td>
<td>298</td>
<td>0.45 (0.76)</td>
<td>0.54 [38, 39]</td>
<td>6.9 (11.0)</td>
<td>2139 (2200)</td>
</tr>
<tr>
<td>Methanol</td>
<td>0.23</td>
<td>298</td>
<td>0.46 (0.75)</td>
<td>0.54 [42]</td>
<td>5.3 (8.2)</td>
<td>2062 (2139)</td>
</tr>
</tbody>
</table>

Figure 4 shows the error in \( K, r_f^*, T_f \) as a function of \( \Delta T_{\text{max}} \) and total number of zones for (a) \( n \)-heptane, (b) ethanol and (c) methanol. In general, the error in computing \( T_f \) is much less than that for \( r_f^* \) and \( K \) for the same number of zones. The reason for this behaviour is that \( r_f^* \) and \( K \) depend on the gradients of \( Y_i \) and \( T \) and therefore are more sensitive to the resolution near the droplet surface in the inner region and near the flame in the outer region. For each fuel, the rate of convergence for \( r_f^* \) and \( K \) using the adaptive mesh (square) is much higher than using a uniform mesh (diamond). With an adaptive mesh, approximately 200, 80 and 50 zones are needed to keep the error in \( K \) under 1% for \( n \)-heptane, ethanol and methanol, respectively, compared to the 10,000+ zones required for the uniform mesh to achieve only 10% accuracy. For 5% error, approximately 40, 20 and 15 zones are needed using the adaptive mesh, which is very reasonable for use in large scale spray calculations where millions of drops are being solved for.

Also shown on Figure 4, on the right axis, are the relative runtimes (square) for each case normalised by the time to run at the \( \Delta T_{\text{max}} = 0.1 \) case. For each fuel, as the order of magnitude of \( N \) increases, so does the normalised time, \( \tau \), with a slope of roughly unity. The overhead time can somewhat be seen for the lower values of \( N \) as the difference between cases is slightly less than one order of magnitude of \( \tau \).

Table 1 shows a comparison of the multizone model predictions to experimental data and estimates using a two-zone model (shown in parenthesis). A two-zone model consistently over-estimates burn rates and stand-off ratios when compared to experimental data. The errors in burn rates are 55, 22 and 39% for \( n \)-heptane, ethanol and methanol, respectively. The multizone model error is much less: 17, 17 and 15%. Estimates of flame stand-off are still lower in error than the two-zone model. The equal-diffusivities assumption accounts for the underestimation of burning rate and the over estimation of \( r_f^* \).

Law and Law [16] state \( r_f^* \) is a strong function of diffusion in the outer region. They suggest increasing the diffusivity in the outer region will increase burn rate, decrease stand-off ratio and increase flame temperature. Of the species in the outer region, \( O_2 \) may not be the only leading order rate-limiting species. Other estimates of diffusivity in the outer region could be explored.

Figures 5, 6 and 7 show flame structure for \( n \)-heptane, ethanol and methanol, respectively. In (a), these figures show the temperature and species mass fractions versus \( r^* \) and (b) shows the distribution of \( \text{Le}, \Lambda_P, C_P, k \) and \( \Gamma \) as a function of \( r^* \). As shown, step discontinuities in \( \Lambda_P, \Gamma \) and \( \text{Le} \) are observed across the flame due to the selection of \( \epsilon_i \) and \( D_{i,m} \) in the inner region versus the outer region. For each of the three fuels, \( \Lambda_P \) is much larger than \( C_P \). By using \( \Lambda_P \), the assumption of non-unity Lewis number is consistent throughout the model. If \( \Lambda_P \) is replaced by \( C_P \) (not shown), burn rates and stand-off ratios are greatly over-predicted – consistent with a unity Lewis number assumption.
Figure 5. *n*-Heptane droplet flame structure showing (a) $T$, $Y_i$ and (b) Le, $\Lambda_P$, $C_P$, $k$ and $\Gamma$. For this case, $\Delta T_{\text{max}} = 1$ K, $T_\infty = 298$ K, $P_\infty = 1$ atm and $Y_{O,\infty} = 0.23$. 
Figure 6. Ethanol droplet flame structure showing (a) $T$, $Y_i$ and (b) $\text{Le}$, $\Lambda_P$, $C_P$, $k$ and $\Gamma$. For this case, $\Delta T_{\text{max}} = 1$ K, $T_{\infty} = 298$ K, $P_{\infty} = 1$ atm and $Y_{O_2,\infty} = 0.23$. 
Figure 7. Methanol droplet flame structure showing (a) $T$, $Y_i$ and (b) $\text{Le}$, $\Lambda_R$, $C_P$, $k$ and $\Gamma$. For this case, $\Delta T_{\text{max}} = 1 \text{ K}$, $T_\infty = 298 \text{ K}$, $P_\infty = 1 \text{ atm}$ and $Y_{O,\infty} = 0.23$. 
Figure 8. \(n\)-Heptane droplet flame structure showing \(T, Y_F\) and \(Y_O\) \cite{18, 21}. For this case, \(\Delta T_{\text{max}} = 1 \, \text{K}, T_\infty = 298 \, \text{K}, P_\infty = 1 \, \text{atm}\) and \(Y_{O,\infty} = 0.23\).

Figure 8 shows the flame structure of \(n\)-heptane with simulation data from Manzello et al. \cite{21} and Puri and Libby \cite{18}. Manzello et al. use a finite rate chemistry mechanism. Puri and Libby use a single-step reaction at a flame sheet, similar to this study, but with the addition of a water–gas shift equilibrium reaction. Consequently, the temperature and concentrations of fuel and oxidiser of this study match very well with Puri and Libby. The two flame sheet studies vary from those of Manzello et al. because of finite-rate chemistry effects.

Figures 9, 10 and 11 show the sensitivity of (a) \(K\) and (b) \(r_F^*\) as a function of far-field oxygen concentration for \(n\)-heptane, ethanol and methanol, respectively. For \(n\)-heptane, comparisons are made to the experimental data of Lee et al. \cite{37}, Manzello et al. \cite{21}, Okajima and Kumagai \cite{38}, and Hara and Kumagai \cite{39}. For ethanol, comparisons are made to the experimental data of Okajima and Kumagai \cite{38}, Hara and Kumagai \cite{39} and Yozgatligil et al. \cite{40}. For methanol, comparisons are made to the experimental data of Marchese et al. \cite{41}, Hicks et al. \cite{42}, Yang et al. \cite{43}, Lee and Law \cite{44}, Dietrich et al. \cite{45} and Okai et al. \cite{46}.

As \(Y_{O,\infty}\) increases, \(K\) appears to increase logarithmically and \(r_F^*\) decreases exponentially. Overall, the agreement of the multizone model predictions to the data is very satisfactory with errors less than 20%. In general, model predictions appear to consistently under-predicted the burn rate and over-predict flame stand-off when compared to the data. While the remaining differences could be attributed to physical processes not accounted for in the present model (e.g. soot and radiation \cite{40, 47}, dilution of fuel from water vapour \cite{41, 44}, finite-rate chemistry \cite{14}, unsteady heating \cite{13, 15}, multi-dimensional effects.
Figure 9. $n$-heptane (a) burn rate and (b) flame stand-off ratio versus $Y_{O,\infty}$ [21, 38, 39]. For all cases $\Delta T_{\text{max}} = 1\,\text{K}$, $T_{\infty} = 298\,\text{K}$ and $P_{\infty} = 1\,\text{atm.}$
Figure 10. Ethanol (a) burn rate and (b) flame stand-off ratio versus $Y_{O,\infty}$ [38–40]. For all cases $\Delta T_{\text{max}} = 1 \text{ K}$, $T_\infty = 298 \text{ K}$ and $P_\infty = 1 \text{ atm}$. 
Figure 11. Methanol (a) burn rate and (b) flame stand-off ratio versus $Y_{O,\infty}$ [41–46]. For all cases $\Delta T_{\text{max}} = 1$ K, $T_{\infty} = 298$ K and $P_{\infty} = 1$ atm.
Figure 12. Methanol burn rate versus $X_{CO_2,\infty}$ [42]. For all cases, $\Delta T_{\text{max}} = 1 \text{ K}$, $T_\infty = 298 \text{ K}$, $P_\infty = 1 \text{ atm}$ and $X_{O,\infty} = 0.21$.

...it is difficult to discern these effects given the apparent wide range of reported burn rates for the same pressure, temperature and oxygen environment.

Figure 12 shows the effectiveness of the multizone model to account for properties based on composition of the gas. The oxidiser far-field concentration is kept at a constant mole fraction, $X_{O}$, of 0.21. The increasing concentration of CO$_2$ displaces the nitrogen in the far-field. By keeping the oxidiser concentration constant, the flame is not being starved by increasing the CO$_2$ concentration and the effects are purely because of the property variations. The current study matches gasification rate, $K$, very well to the experimental data of Hicks et al. [42]. The gasification rate decreases slightly with the increase in CO$_2$ concentration.

In order better to understand the reasons why the two-zone model grossly over-predicts the burn rates and flame stand-off ratio, Equation (5) is substituted into Equation (4) so the flame stand-off ratio may be expressed in terms of a single ‘effective’ Lewis number, $L_{\text{eff}} \equiv (\bar{k}_{\text{in}}/\bar{A}_{P,\text{in}})/(\Gamma_{O,\text{out}})$, in the limit of a two-zone description [7]:

$$\frac{r_f}{r_s} = r_f^{*} = 1 + \frac{\ln[(1 + B_q)^{L_{\text{eff}}}] \ln[1 + Y_{O,\infty}/\nu_O]}{\ln[1 + Y_{O,\infty}/\nu_O]},$$

where $B_q \equiv \bar{A}_{P,\text{in}}(T_f - T_s)/L_{vap}'$ is the thermal transfer number. As discussed by Law and indicated by Equation (13), individual Lewis numbers of the inner and outer regions are actually irrelevant for defining the flame stand-off ratio in a two-zone model [7]. Rather, the leading order effect on flame stand-off is a combination of inner and outer transport variables defining the effective Lewis number. Based on their variable property model and
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5. Conclusions

A multizone hydrocarbon droplet burn model is developed that accounts for variations in gas-phase thermal and transport properties. The appeal of this semi-analytical approach is that good estimates of burn rates can be determined at a fraction of the cost compared to direct numerical integration approaches, but also without incurring the errors of simplified two-zone descriptions. The use of the adaptive mesh shows excellent error convergence rates and a means to balance desired levels of accuracy with computational cost. Model predictions of burn rates and flame stand-off are in favourable agreement with experimental data for the fuels and oxygen environments considered, with errors less than 20%.
Acknowledgements

Support for this work has been provided by NAVAIR through the STTR phase II program under contract N68335-10-C-0418.

References

Appendix A. Solution of the eigenvalue problem

Equations (2), (4) and (7), along with a relation for the fuel vapour pressure curve, represent a coupled system of equations for the solution of the radial distributions of $Y_i$, $T$ and the location of $r_f^*$. The solution procedure for solving these equations is summarised in the following steps along with a flowchart (Figure 3).
1. Initialize $T$ and $Y_i$ at shell locations using two-zone model.
2. Solve for $r_s^{**}$ using Equations (9) and (10).
3. Calculate $r_s^*$ from $r_s^{**}$.
4. Guess a value of fuel vapour at droplet surface $Y_{F,s}$ (beginning of $Y_{F,s}$-loop).
5. Compute the partial pressure of the fuel at the surface, $P_{F,s}$, and $T_s$ using vapour-pressure curve.
6. Guess $r^*$, $T_k$, $Y_{i,k}$, $\Lambda_{P_s}$, $k$, $\Gamma_{F,k}$, $\Gamma_{O,k}$, $L_{eF,k}$ and $L_{eO,k}$ (beginning of property-loop).
7. Guess $r^*$ (beginning of $r_s^*$-loop).
8. Solve for $r_s^*$ using Equation (4).
9. Calculate $r_s^*$ from $r_s^{**}$.
10. Comparing the initially guessed value of $r_s^*$ from step 7 to that from step 8 provides an error function that is driven towards zero using a Jacobi method until an error criterion of $1 \times 10^{-13}$ is satisfied (end of $r_s^*$-loop). Return to step 7 with new guess of $r^*$, if necessary.
11. Calculate $\dot{m}$ using Equation (11).
12. Calculate $T_f$ using Equation (7).
13. Decode $T$ using new value of $T_f$ and Equation (2b).
   a. Inner shells of $T$ are decoded by sweeping from $r_s^*$ to $r_s^*$:
      $$T_{k-1} = \frac{1}{\Lambda_{P_s}} \left\{ \frac{Q_{\text{in}}}{\dot{m}} + \left( \Lambda_{P_s} T_k - \frac{Q_{\text{in}}}{\dot{m}} \right) \exp \left[ -\frac{\dot{m}}{4\pi r_s} \frac{\Lambda_{P_k}}{k} \left( 1/r_{k-1}^* - 1/r_k^* \right) \right] \right\}.$$
   b. Outer shells of $T$ are decoded by sweeping from $r_s^*$ to $r_s^*$:
      $$T_k = \frac{1}{\Lambda_{P_s}} \left\{ \frac{Q_{\text{out}}}{\dot{m}} + \left( \Lambda_{P_s} T_{k-1} - \frac{Q_{\text{out}}}{\dot{m}} \right) \exp \left[ -\frac{\dot{m}}{4\pi r_s} \frac{\Lambda_{P_k}}{k} \left( 1/r_{k-1}^* - 1/r_k^* \right) \right] \right\}.$$
14. Decode $Y_i$ fields on new mesh, using Equation (2a).
   a. Inner shells of $Y_{F,k}$ are decoded by sweeping from $r_s^*$ to $r_s^*$:
      $$Y_{F,k-1} = \epsilon_{F,k} + (Y_{F,k} - \epsilon_{F,k}) \left[ -\frac{\dot{m}}{4\pi r_s} \frac{\Lambda_{P_k}}{\Gamma_{F,k}} \left( 1/r_{k-1}^* - 1/r_k^* \right) \right].$$
   b. Outer shells of $Y_{O,k}$ are decoded by sweeping from $r_s^*$ to $r_s^*$:
      $$Y_{O,k} = \epsilon_{O,k} + (Y_{O,k-1} - \epsilon_{O,k}) \left[ -\frac{\dot{m}}{4\pi r_s} \frac{\Lambda_{P_k}}{\Gamma_{O,k}} \left( 1/r_{k-1}^* - 1/r_k^* \right) \right].$$
   c. Outer shells of remaining species are decoded by sweeping from $r_s^*$ to $r_s^*$:
      $$Y_{i,k-1} = \epsilon_{i,k} + (Y_{i,k} - \epsilon_{i,k}) \left[ -\frac{\dot{m}}{4\pi r_s} \frac{\Lambda_{P_i}}{\Sigma_{i,k}} \left( 1/r_{k-1}^* - 1/r_k^* \right) \right].$$
   d. Inner shells of remaining species are decoded by sweeping from $r_s^*$ to $r_s^*$:
      $$Y_{i,k-1} = \epsilon_{i,k} + (Y_{i,k} - \epsilon_{i,k}) \left[ -\frac{\dot{m}}{4\pi r_s} \frac{\Lambda_{P_i}}{\Sigma_{i,k}} \left( 1/r_{k-1}^* - 1/r_k^* \right) \right].$$
15. Compute mixture weighted properties of $\Lambda_F$, $k$, $\Gamma_F$, $\Gamma_O$, $Le_F$ and $Le_O$ at each zone location using an arithmetic average of temperature and composition from neighbouring shells. Mixture weighted transport properties are determined using Cantera.

16. Compute the $\ell^2$-norm error between $r^*$ from step 6 to that from step 9. Using a Jacobi method, iterate until an error criterion of $1 \times 10^{-12}$ is satisfied (end of property-loop). Return to step 6 with new guess of $r^*$, if necessary.

17. Calculate $Y_{F,s}$ from Equation (12).

18. Comparing the initially guessed value of $Y_{F,s}$ from step 4 to that from step 17 provides an error function that is driven towards zero using the Illinois algorithm until an error criterion of $1 \times 10^{-8}$ is satisfied (end of $Y_{F,s}$-loop). Return to step 4 with new guess of $Y_{F,s}$, if necessary.