Droplet Autoignition in a Reactive Oxidant/Fuel–Vapor/Inert Environment

Xianguo Li*

University of Victoria, Victoria, British Columbia V8W 3P6, Canada

The effects of one-step reaction orders have been studied on the ignitability of a fuel droplet in a fuel-lean, oxidant-lean, and both fuel- and oxidant-lean environments with finite flame location. The matched asymptotic analysis of large activation energy is employed with the Arrhenius kinetics and arbitrary fuel and oxidizer reaction orders. The variable thermophysical properties are taken to depend on both temperature and composition of the gas mixture with distinct binary diffusion coefficients and nonunity Lewis numbers. It is found that the ignition Damköhler number varies significantly with the fuel and oxidant reaction orders, and decreases as the ambient fuel and oxidant concentration are increased. Autoignition is prevented if the ambient oxidant concentration is sufficiently low. It is also found that the mixture Lewis number should be defined appropriately, depending on the ambient fuel and oxidant concentration if equal binary diffusion coefficients are used. The variable thermophysical properties affect ignition phenomenon partly through the droplet mass evaporation rate and partly through the thermal driving force for evaporation. By using an appropriate mixture Lewis number, the frozen evaporation rate and the thermal driving force, ignition event can be predicted accurately from the analysis with constant properties with equal binary diffusion coefficients.

Nomenclature

B^*	= pre-exponential factor in Arrhenius law
c_p	= $c_p^*/c_{p,\infty}^*$
c_p^*	= specific heat of fuel vapor at constant pressure
$c_{p,\infty}^*$	= specific heat of fuel vapor evaluated at T_∞^*
D	= Damköhler number
D_{ij}	= $D_{ij}^*/D_{ij,\infty}^*$
D_{ij}^*	= mass diffusion coefficient of species pair i and j
E^*	= activation energy
H	= H^*/Q^*
H^*	= effective latent heat of evaporation
h	= h^*/Q^*
h^*	= specific enthalpy
$Le_{ij,\infty}$	= $(\lambda_\infty^*/c_{p,\infty}^*)/(\rho^* D_{ij,\infty}^*)$
$\dot{m}$	= $(\rho^* v^* r^{*2})/(r_s^* \lambda_\infty^*/c_{p,\infty}^*)$, mass evaporation rate
n_i	= overall reaction order with respect to species i
n	= $n_F + n_O$
p^*	= pressure
Q^*	= heat of combustion per unit mass of fuel
R^{0*}	= universal gas constant
r	= r^*/r_s^*
r^*	= radial coordinate
r_s^*	= instantaneous radius of the droplet
T	= $c_{p,\infty}^* T^*/Q^*$
T^*	= temperature
T_a^*	= activation temperature, E^*/R^{0*}
V^*	= diffusion velocity
v^*	= velocity
W^*	= molecular weight
$\bar{W}^*$	= average molecular weight of the mixture
W_i	= $W_i^*/\bar{W}_\infty^*$
X	= mole fraction

α	= ambient fuel concentration parameter, see Eq. (25)
β	= $\Sigma_i \gamma_{i,f} h_i(T_\infty) - \Sigma_i \gamma_{i,f} h_i(T_s) + H$, thermal driving force for droplet evaporation
Γ	= perturbation in mass flux fraction
γ	= mass flux fraction
$\Delta, \Delta_1, \Delta_2$	= reduced system Damköhler number
δ_{ij}	= $\delta_{ij} = 1$ if $i = j$; $\delta_{ij} = 0$ if $i \neq j$
ε	= T_∞^2/T_a, a small perturbation parameter
η	= inner stretching variable
θ	= perturbation in temperature
κ	= ambient oxidant concentration parameter, Eq. (35)
λ	= $\lambda^*/\lambda_\infty^*$
λ^*	= thermal conductivity
ν	= stoichiometric molar coefficient
ρ	= ρ^*/ρ_∞^*
ρ^*	= density
χ	= perturbation in molar concentration

Subscripts

F	= fuel
f	= frozen state
I	= ignition
i	= 1 fuel, 2 oxidizer, >2 inert gases
O	= oxidizer
P	= products
s	= droplet surface
∞	= ambience

Superscript

$*$	= dimensional quantity

Introduction

T HE study of droplet autoignition in sprays is of fundamental and practical importance in the modeling of spray combustion, which is widely used in many power generation and propulsion devices such as diesel engines and gas turbine combustors. The ignition of droplets in sprays directly relates to the flame propagation, and strongly influences the ignition delay and flame stabilization in practical spray combustion systems. Therefore, prediction of ignition event is necessary for

the simulation of spray combustion in practical energy conversion systems. Although many experimental measurements and numerical simulations of the single droplet ignition process have been carried out,[1-5] the ignition processes of droplets in various reactive environments remain far from well understood. Even with recent advances in computational facilities,[6] the detailed numerical simulation of individual droplet vaporization and ignition in practical spray combustion modeling is beyond the current computational capability because of the large number of droplets present in such sprays. The ignition of the droplets in realistic sprays is extremely complex, affected by the physical and chemical properties of the liquid fuel, the prevaporized vapor concentration in the interdrop space, the droplet size and interdrop distance, and the flow effects associated with convection and turbulence. So far most knowledge of ignition characteristics associated with droplets in sprays has been obtained from studies for single droplets, and usually a simple criterion for the occurrence of droplet autoignition is derived, which incorporates all of the physical and chemical parameters of the given system. Such a simple criterion is very useful as a submodel for realistic spray combustion modeling,[7] and is the objective of the present study. Therefore, an analytical approach will be employed instead of detailed numerical simulation, such as the work of Botros et al.[2] and Samuel and Karim.[5]

The quasisteady basic droplet model, sometimes known as the d^2-law, has been studied both experimentally and theoretically.[8,9] This approach simplifies the theory and mathematics involved and is useful for analyzing the vaporization, ignition, and combustion of droplets. Based on a quasisteady theory, Law[10] derived an ignition criterion for a single fuel droplet burning in an unbounded oxidizing environment through the use of a matched asymptotic technique in the limit of large activation energy. This criterion is valid only for unity reaction orders and unity Lewis numbers for both fuel and oxidizer. As Aggarwal[11] showed, the use of a one-step reaction with unity exponents is inadequate as a rigorous kinetics representation for the ignition of droplets in sprays. Hence, Mawid and Aggarwal[12] extended Law's analysis[10] to include the effect of arbitrary reaction orders with respect to the fuel and oxidizer, but the unity Lewis number for both fuel and oxidizer was again employed in their analysis. Further, the constant thermophysical properties and equal binary diffusion coefficients have been assumed in the previous ignition analysis. As a result, the success of their results, to a certain degree, depends on the judicious selection of the types of species, their relative concentrations and reference temperature at which all the assumed constant properties are evaluated. Recently, the work of Law[10] and Mawid and Aggarwal[12] has been extended by Li and Renksizbulut[13] to include distinct binary diffusion coefficients and variable thermophysical properties with concentration and temperature dependence, arbitrary Lewis numbers, and arbitrary reaction orders with respect to the fuel and oxidizer.

In all previous analyses, the ignition event is assumed to occur at an infinitely large distance away from the droplet surface. However, experimental observations and theoretical studies[2,5,14] show that ignition occurs only a few droplet diameters away from the droplet surface. Further, the fuel vapor is almost always present in the environment prior to droplet ignition in practical sprays. Considering these two conditions, Law and Chung[15] derived a modified ignition criterion for droplets in sprays. However, an assumption was still made of unity reaction orders and Lewis numbers, as well as constant properties and equal binary diffusion coefficients. On the other hand, the majority of the recent works assume finite location of the flame, and several of the recent numerical studies assume distinct binary diffusion coefficients, variable thermophysical properties, arbitrary Lewis numbers, and single or multiple reaction steps for the droplet ignition within reactive environments of fuel and air (e.g., Samuel and Karim[5]).

Therefore, the present study will use the large activation energy asymptotics to investigate the droplet ignition in reactive environments and with the occurrence of an ignition event at a finite distance from droplet surface, to examine the effects of chemical kinetics on the ignition phenomena and to develop simple criteria for the prediction of ignition event. The analysis will include distinct binary diffusion coefficients and variable properties along with arbitrary reaction orders and Lewis numbers. Therefore, the present study may be regarded as an extension of both Law and Chung's[15] and Li and Renksizbulut's[13] study. Since the purpose of the present work is to examine the effects on the ignition criterion of chemical kinetics and variable thermophysical properties with a weakly reactive environment at a finite distance, the emphasis will be placed on the procurement of the reduced and ignition (critical) Damköhler numbers.

For droplet combustion, the diffusion flame-sheet model of the constant thermophysical properties from Godsave[8] has been considerably extended to account for the effects of variable thermophysical properties and general nonunity Lewis numbers.[16-22] A summary with discussion in this aspect has been given by Williams.[23] It is beyond the scope of the present article to review the theory of droplet combustion. However, the variable transport property formulation of droplet combustion given by Law and Law,[20,21] and that of droplet vaporization by Law,[24] will be incorporated into the present analysis of droplet ignition, as Li and Renksizbulut did.[13]

Formulation

Consider a single-component fuel droplet vaporizing in a quiescent and hot environment located at r_∞^*. The droplet is characterized by the instantaneous surface temperature T_s^* and radius r_s^*. The environment consists of known concentrations of an oxidizer, fuel vapor, and noncondensable inert gases with the ambient temperature T_∞^* and pressure p_∞^* given. All of the gas phase processes are assumed to be quasisteady, isobaric, and spherically symmetric. Effects caused by gravity and thermal radiation are neglected, and the heat of combustion is taken to be constant. Species diffusion arising only from concentration gradients is assumed with negligible thermal diffusion (Soret effect). Further, the species binary diffusion coefficients are regarded as distinct, and all of the thermophysical properties are considered to be dependent on both temperature and species concentrations with arbitrary Lewis numbers.

As for chemical kinetics representation, Aggarwal[11] examined the droplet ignition with various chemical reaction schemes to take into account partial oxidation, and recommended the use of the overall one-step global reaction with nonunity reaction orders for droplet ignition analysis. In particular, the reaction order of $n_F = 0.25$ for fuel concentration and $n_O = 1.5$ for oxidant concentration, as recommended by Westbrook and Dryer,[25] was found to be adequate, and has been frequently adopted in ignition analyses of droplets and sprays.[12,26-30] Single-step chemical reaction has also been used in a recent numerical simulation of droplet ignition and combustion.[5] Therefore, in this study, overall one-step irreversible chemical reaction is assumed between the fuel and oxidizer with arbitrary reaction orders with respect to each of the reactant concentrations. Hence, the chemical reaction may be represented by

$$\nu_F F + \nu_O O + \text{Inerts} \xrightarrow{k} \nu_P P + \text{Inerts} - \nu_F W_F^* Q^*$$
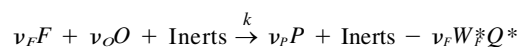
where the term $\nu_F W_F^* Q^*$ represents the heat of combustion, and the reaction rate constant k is described by the Arrhenius law of reaction rate with constant B^*. The overall reaction orders with respect to the fuel and oxidizer, denoted by n_F and n_O, respectively, are not necessarily the same as the stoichiometric coefficients ν_F and ν_O, as discussed earlier.

With all of the previous assumptions, the governing equations for the gas-phase conservation of species (such as oxi-

dizer O and fuel F) and energy, as well as species diffusion, can be expressed as follows[13,23]:

$$\frac{\dot{m}}{r^2} \frac{d\gamma_i}{dr} = -\frac{\nu_i W_i}{\nu_F W_F} \dot{\omega} \tag{1}$$

$$\frac{\dot{m}}{r^2} \frac{d}{dr} \left(\sum_i \gamma_i h_i \right) - \frac{1}{r^2} \frac{d}{dr} \left(r^2 \lambda \frac{dT}{dr} \right) = \dot{\omega} \tag{2}$$

$$r^2 \frac{dX_i}{dr} = \dot{m} \sum_j \frac{Le_{ij,\infty}}{f_D(T)} \left(\frac{\gamma_j}{W_j} X_i - \frac{\gamma_i}{W_i} X_j \right) \qquad (i = 1, 2, \ldots, N) \tag{3}$$

where γ_i is the mass-flux fraction of chemical species i, and $\Sigma_i \gamma_i \equiv 1$. But the value of γ_i is not necessarily limited to the range between the values of 0 and 1. Equation (3) is the one-dimensional form of the Stefan–Maxwell equation governing the multicomponent species diffusion, and

$$Le_{ij,\infty} = \frac{\lambda_\infty^*}{c_{p,\infty}^*(\rho_\infty^* D_{ij,\infty}^*)} \tag{4}$$

is the Lewis number. The summation sign here is to be performed over all of the species present in the system, unless otherwise stated. In deriving Eq. (3), the diffusion coefficients D_{ij}^* have been taken as $D_{ij}^* = f_D^*(T^*)(d_{ij}^* T^*/p^*)$ and $f_D(T) = f_D^*(T^*)/f_D^*(T_\infty^*)$.[13,21,31] The chemical reaction term $\dot{\omega}$ is given by

$$\dot{\omega} = D(1/T)^n X_F^{n_F} X_O^{n_O} \exp[-(T_a/T)] \tag{5}$$

where the system Damköhler number is

$$D = \frac{c_{p,\infty}^*}{\lambda_\infty^*} \nu_F W_F^* B^* \left(\frac{p^* c_{p,\infty}^*}{R^{0*} Q^*} \right)^n r_s^{*2} \tag{6}$$

The boundary conditions for Eqs. (1–3) are

$$\text{as} \quad r = r_\infty: X_i = X_{i,\infty} \tag{7}$$

$$T = T_\infty \tag{8}$$

$$\text{at} \quad r = 1: \gamma_i = \delta_{iF} \tag{9}$$

$$\lambda_s \frac{dT}{dr} = \dot{m}H \tag{10}$$

$$T = T_s = \text{const} \tag{11}$$

where $\delta_{iF} = 0$ for $i \neq F$, and $\delta_{iF} = 1$ for $i = F$.

Analysis

Frozen Solution

In this regime, all of the chemical reactions are frozen because of the infinitely large value of the activation energy (i.e., $T_a \to \infty$). Then, following that of Law and Law[21] and Li and Renksizbulut,[13] the mass-flux fraction, species concentration, and temperature distribution can be determined for this frozen state as follows:

$$\gamma_{i,f} = \delta_{iF} \tag{12}$$

$$X_{i,f}(\zeta) = \sum_k K_{i,k} \exp(\Lambda_k \zeta) \qquad (i = 1, 2, \ldots, N) \tag{13}$$

$$\int_{T_\infty}^{T_f} \frac{f_\lambda(T_f')}{f_D(T_f')} \frac{dT_f'}{\sum_i \gamma_{i,f} h_i(T_f') - h_\infty + \beta} = -\int_0^\zeta \frac{d\zeta'}{\sum_i a_i X_{i,f}(\zeta')} \tag{14}$$

where

$$\zeta = \int_r^{r_\infty} \frac{\dot{m}_f}{r'^2 f_D[T_f(r')]} \, dr' \tag{15}$$

is a new independent spatial variable, Λ_k are the eigenvalues of the coefficient matrix in Eq. (3) with the mass-flux fraction given in Eq. (12), and $K_{i,k}$ are the constants of integration determined from

$$\sum_k K_{i,k} \Lambda_k = -\sum_j Le_{ij,\infty} \left(\frac{\gamma_{i,f}}{W_j} X_{i,\infty} - \frac{\gamma_{i,f}}{W_i} X_{j,\infty} \right)$$
$$(i = 1, 2, \ldots, N) \tag{16}$$

and the thermal driving force for the evaporation of fuel droplet is given by

$$\beta = \sum_i \gamma_{i,f} h_i(T_\infty) - \sum_i \gamma_{i,f} h_i(T_s) + H \tag{17}$$

It should also be pointed out that in obtaining Eq. (14), the simple linear molar mixing rule, originally suggested by Kennard,[32] is adopted here following Law and Law[20,21] in droplet combustion, Law[24] in droplet evaporation, and Li and Renksizbulut[13] in droplet ignition. The accurate expressions for mixture thermal conductivity, like the Tondon and Saxena formula,[33] depend inseparably on the temperature and species concentrations, rendering the analytical solution of the temperature distribution almost impossible except for a binary system.[24] As shown by Law[24] for pure droplet vaporization, results from the linear molar mixing rule and the accurate expression of the Tondon and Saxena[33] formula agree within 1%. Further, from the kinetic theory of gases,[23] it is reasonable to assume that the thermal conductivity of all the pure gas species has the same temperature dependence with a different proportionality constant a_i for each individual species. Hence, the mixture thermal conductivity used in the previous analysis is in dimensionless form:

$$\lambda = \sum_i a_i X_i f_\lambda(T)$$

Since the distribution of temperature and species concentrations is known, the frozen droplet evaporation rate $\dot{m}_f$ can be obtained by evaluating Eq. (14) at the droplet surface, where $T_f = T_s$ and $\zeta = \zeta_s$, which corresponds to $r = 1$. By comparing with the constant property results of Law and Chung,[15] it is clear that $\dot{m}_f$ is affected by the specific dependence of the thermophysical properties on the concentration and temperature distributions. Further, the results of Li and Renksizbulut[13] can be recovered by allowing $r_\infty \to \infty$ in the previous frozen solutions.

Ignition in a Fuel-Lean Environment

For ignition analysis, which follows the work of Law,[10] Law and Chung,[15] Mawid and Aggarwal,[12] and Li and Renksizbulut,[13] the activation energy is assumed to be large but finite, and the chemical reaction is therefore expected to occur near the hot environment at r_∞.[15] As a result, the flowfield may be divided into two regions: 1) an outer, convective–diffusive region that is free from the chemical reactions, and 2) an inner diffusive–reactive region in the neighborhood of the hot environment where all of the chemical reactions occurring in the system are concentrated.

In the outer, convective–diffusive region away from the hot environment, the mixture temperature decreases, and all of the chemical reactions are practically frozen because of the strong temperature dependence of Arrhenius kinetics. But the mass flux fraction, species concentration, and temperature distribu-

tions differ from their frozen solution by a small amount of order $\varepsilon = T_\infty^2/T_a$ because of convection and diffusion effects. The lengthy presentation of the perturbation terms is avoided here since they are not prerequisite for the following analysis. The detailed process is very similar to the previous studies.[10,12,13,15] The mass evaporation rate is also expanded in the powers of ε

$$\dot{m} = \dot{m}_f + \varepsilon\dot{m}_1 + \mathbb{O}(\varepsilon^2) \qquad (18)$$

where $\dot{m}_1$ is the first-order perturbation to the droplet vaporization rate.

For the inner, diffusive–reactive region next to the hot environment, the species concentrations and temperature are also perturbed, because of the initiation of the chemical reactions, from their frozen solutions by a small amount of the order of ε, such that

$$T_{in} = T_\infty + \varepsilon[\theta(\eta) - \beta\eta] + \mathbb{O}(\varepsilon^2) \qquad (19)$$

$$\gamma_{i,in} = \gamma_{i,f} + \Gamma_i(\eta) + \mathbb{O}(\varepsilon) \qquad (20)$$

$$X_{i,in} = X_{i,\infty} + \varepsilon\left[\sum_k K_{i,k}\Lambda_k\eta + \chi_i(\eta)\right] + \mathbb{O}(\varepsilon^2) \qquad (21)$$

where η is defined as

$$\eta = (\dot{m}_f/r_\infty^2)[(r_\infty - r)/\varepsilon] \qquad (22)$$

Note that although the chemical reactions have small effects on the temperature and concentrations as assumed in Eqs. (19) and (21), they exert significant influences on the mass flux fraction as shown in Eq. (20), which will be confirmed later on.

As $r \to r_\infty$ ($\varepsilon \to 0$), Eq. (15) becomes, to the leading-order term, $\zeta = \eta\varepsilon$. By the inner expansion given in Eqs. (19) and (21), it can be seen that $f_D(T) = 1 + \mathbb{O}(\varepsilon)$ and $f_\lambda(T) = 1 + \mathbb{O}(\varepsilon)$. Therefore, in terms of the inner stretching variable and with the help of Eq. (16), the energy equation, Eq. (2), becomes, to the leading-order term

$$\frac{d^2\theta}{d\eta^2} = -\Delta\left(\alpha + \eta + \frac{\chi_F}{\sum_j Le_{Fj,\infty}X_{j,\infty}/W_F}\right)^{n_F} \exp(\theta - \beta\eta) \qquad (23)$$

where

$$\Delta = \frac{r_\infty^4}{\dot{m}_f^2}(D\varepsilon^{n_F+1})\left(\frac{1}{T_\infty}\right)^n \left(\frac{1}{W_F}\right)^{n_F} X_{O,\infty}^{n_O} \exp\left(-\frac{T_a}{T_\infty}\right)$$
$$\times \left(\sum_j Le_{Fj,\infty}X_{j,\infty}\right)^{n_F} \qquad (24)$$

is the reduced system Damköhler number regarded as $\Delta = \mathbb{O}(1)$, and

$$\alpha = \frac{X_{F,\infty}}{\varepsilon}\left(\sum_j Le_{Fj,\infty}X_{j,\infty}/W_F\right)^{-1} \qquad (25)$$

represents the fuel concentration at the hot environment.

Similarly, Eq. (1) can be shown to take the following form:

$$\frac{d\Gamma_i}{d\eta} = \frac{\nu_i W_i}{\nu_F W_F}\Delta\left(\alpha + \eta + \frac{\chi_F}{\sum_j Le_{Fj,\infty}X_{j,\infty}/W_F}\right)^{n_F} \exp(\theta - \beta\eta)$$
$$(i = 1, 2, \ldots, N) \qquad (26)$$

which confirms that the inner expansion of the mass flux frac-

tion as assumed in Eq. (20) is correct in order of the magnitude in terms of ε.

For the species concentrations, the left-hand side (LHS) of Eq. (3) is expressed in terms of the inner variable η; then the derivative of both sides of the equation is taken with respect to η. Taking account of Eq. (26), while keeping in mind that $\Delta = \mathbb{O}(1)$, the distribution of the species molar fraction in the inner region is derived as follows:

$$\frac{d^2\chi_i}{d\eta^2} = -\sum_j Le_{ij,\infty}(\nu_j X_{i,\infty} - \nu_i X_{j,\infty})\frac{\Delta}{\nu_F W_F}\left(\alpha + \eta\right.$$
$$\left. + \frac{\chi_F}{\sum_j Le_{Fj,\infty}X_{j,\infty}/W_F}\right)^{n_F} \exp(\theta - \beta\eta) \quad (i = 1, 2, \ldots, N) \qquad (27)$$

Application of the local Shvab–Zeldovich formulation to Eqs. (23) and (27) would result in explicit expressions for the species concentrations. Particularly, for $i = F$, we have

$$\theta + \frac{\chi_F}{\sum_j Le_{Fj,\infty}X_{j,\infty}/W_F} = 0 \qquad (28)$$

when considering the finiteness of θ and χ_i as $\eta \to \infty$, and the boundary conditions at $\eta = 0$. Substituting Eq. (28) into Eq. (23) yields

$$\frac{d^2\theta}{d\eta^2} = -\Delta(\alpha + \eta - \theta)^{n_F}\exp(\theta - \beta\eta) \qquad (29)$$

and the appropriate boundary conditions are

$$\theta = 0 \quad \text{at} \quad \eta = 0 \qquad (30)$$
$$\frac{d\theta}{d\eta} = 0 \quad \text{as} \quad \eta \to \infty$$

where the second boundary condition is obtained by matching the inner and the outer solution as $\eta \to \infty$.

It is clear that Eq. (29) is the only equation that needs to be solved for the present problem. After the temperature is obtained, the species concentrations can be determined from Eq. (28) and the mass-flux fractions can be obtained similarly through the application of the local Shvab–Zeldovich formulation to Eqs. (23) and (26). The first-order perturbation term to the droplet evaporation rate can be found from $\dot{m}_1 = \theta(\infty)/\beta$. In the previous ignition analysis it has been assumed that $X_{F,\infty} \leq \mathbb{O}(\varepsilon)$ and $X_{O,\infty} = \mathbb{O}(1)$.

Ignition in an Oxidant-Lean Environment

For $X_{F,\infty} = \mathbb{O}(1)$ and $X_{O,\infty} \leq \mathbb{O}(\varepsilon)$, the equations governing the distribution of temperature, mass-flux fractions and species concentrations in the inner reactive–diffusive region become

$$\frac{d^2\theta}{d\eta^2} = -\Delta_1(\kappa - \theta)^{n_O}\exp(\theta - \beta\eta) \qquad (31)$$

$$\frac{d\Gamma_i}{d\eta} = \frac{\nu_i W_i}{\nu_F W_F}\Delta_1(\kappa - \theta)^{n_O}\exp(\theta - \beta\eta) \qquad (32)$$

$$\frac{d^2\chi_i}{d\eta^2} = -\sum_j Le_{ij,\infty}(\nu_j X_{i,\infty} - \nu_i X_{j,\infty})$$
$$\times \frac{\Delta_1}{\nu_F W_F}(\kappa - \theta)^{n_O}\exp(\theta - \beta\eta) \qquad (33)$$

where $i = 1, 2, \ldots, N$, and

$$\Delta_1 = \frac{r_\infty^4}{\dot{m}_f^2} (D\varepsilon^{n_O+1}) \left(\frac{1}{T_\infty}\right)^n \left(\frac{v_O}{v_F W_F}\right)^{n_O} X_{F,\infty}^{n_F} \exp\left(-\frac{T_a}{T_\infty}\right)$$

$$\times \left(\sum_j Le_{Oj,\infty} X_{j,\infty}\right)^{n_O} \tag{34}$$

$$\kappa = \frac{X_{O\infty}}{\varepsilon} \frac{v_F W_F}{v_O} \left(\sum_j Le_{Oj,\infty} X_{j,\infty}\right)^{-1} \tag{35}$$

It is clear that the mass-flux fractions and species concentrations can be determined through an application of the local Shvab–Zeldovich formulation, provided that the temperature field is known. Therefore, Eq. (31) is the only equation that needs to be solved, and the boundary conditions are exactly the same as those given in Eq. (30).

Ignition in a Fuel/Oxidant-Lean Environment

When both fuel and oxidant concentrations are low in the ambience, or $X_{F,\infty} \leq \mathbb{O}(\varepsilon)$ and $X_{O,\infty} \leq \mathbb{O}(\varepsilon)$, the same analysis procedure as given previously leads to the following equation governing the ignition phenomenon:

$$\frac{d^2\theta}{d\eta^2} = -\Delta_2(\alpha + \eta - \theta)^{n_F}(\kappa - \theta)^{n_O} \exp(\theta - \beta\eta) \tag{36}$$

where the reduced system Damköhler number is given by

$$\Delta_2 = \frac{r_\infty^4}{\dot{m}_f^2} (D\varepsilon^{n+1}) \left(\frac{1}{T_\infty}\right)^n \left(\frac{1}{W_F}\right)^{n_F} \left(\frac{v_O}{v_F W_F}\right)^{n_O}$$

$$\times \exp\left(-\frac{T_a}{T_\infty}\right) \left(\sum_j Le_{Fj,\infty} X_{j,\infty}\right)^{n_F} \left(\sum_j Le_{Oj,\infty} X_{j,\infty}\right)^{n_O} \tag{37}$$

and the parameters α and κ are given in Eqs. (25) and (35), respectively. The boundary conditions are exactly the same as those given in Eq. (30).

Clearly, as $\kappa \to \infty$, the previous equations reduce to Eqs. (29) and (30), the case of droplet ignition in a fuel-lean environment. On the other hand, the limit of $\alpha \to \infty$ recovers the case of droplet ignition in oxidant-lean ambience. Thus, the present case may be regarded as a general representation of the droplet ignition problem. Normandia and Buckmaster[34] have studied the case of limited ambient oxidizer mass fraction $Y_{O\infty} = \mathbb{O}(\varepsilon)$ and vanishing ambient fuel mass fraction $Y_{F\infty} = 0$ with infinite flame location $r_\infty = \infty$, and have shown that for sufficient ambient oxidizer concentration, the combustion of a fuel droplet will display proper ignition and extinction phenomena. This is indeed the case for the present situation, as the results show in the next section.

Results and Discussion

Equations (29), (31), and (36) have been solved, subject to Eq. (30), by linearizing the highly nonlinear terms on the right-hand side of the equations with finite difference representation for the second-order differential term on the LHS of the equations. Figure 1 shows the resulting temperature distribution within the inner reactive–diffusive zone as a function of $X = \beta\eta$ for the case of droplet ignition in the fuel-lean environment. The results shown are for $\alpha = 0$, $\beta = 0.5$, $n_F = 0.25$, and several values of Δ. It is seen that the temperature increases quickly and then approaches an asymptotic value, and there exist double solutions for suitable values of Δ. When the maximum temperature $\theta(\infty)$ is plotted as a function of Δ for given values of α, β, and n_F, as shown in Fig. 2, the resulting curves exhibit the lower, ignition branch of the classical S-shaped ignition–extinction curve.[35] Therefore, the vertical tan-

gents to these curves yield the critical ignition Damköhler number Δ_I. Droplet ignition occurs when Δ, given in Eq. (24), exceeds the value of Δ_I.

Figure 3 presents the ignition Damköhler number as a function of β for droplet ignition in a fuel-lean environment. The solid curves represent the results for the fuel reaction order of $n_F = 0.25$ and the dashed curves for $n_F = 1.0$, which is the case studied by Law and Chung.[15] It is seen that the ignition Damköhler number increases rapidly with β and decreases as the ambient fuel concentration increases. The latter may be understood by the fact that the ambient fuel concentration, as
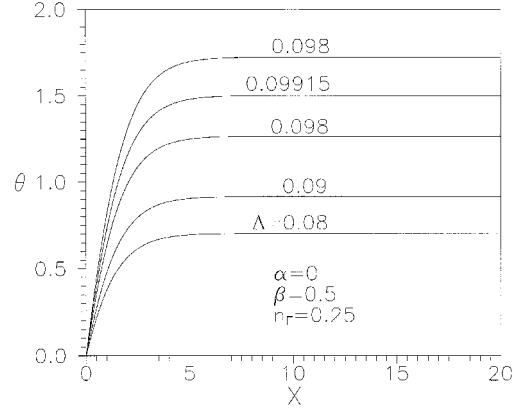

Fig. 1 Temperature distribution within the inner reactive–diffusive zone as a function of the stretching variable $X = \beta\eta$ for the case of droplet ignition in a fuel-lean environment. $\alpha = 0$, $\beta = 0.5$, $n_F = 0.25$, and Δ as shown.
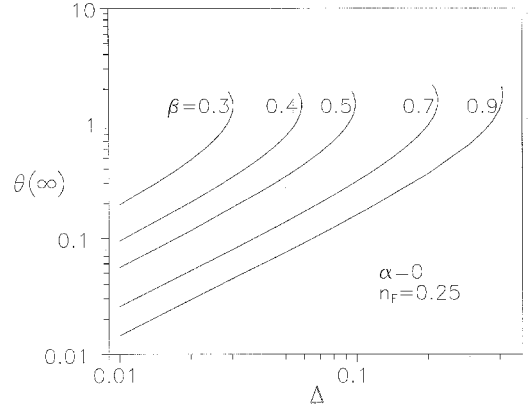

Fig. 2 Maximum perturbed temperature $\theta(\infty)$ as a function of Δ for droplet ignition in a fuel-lean environment. $\alpha = 0$, $n_F = 0.25$, and β as shown.
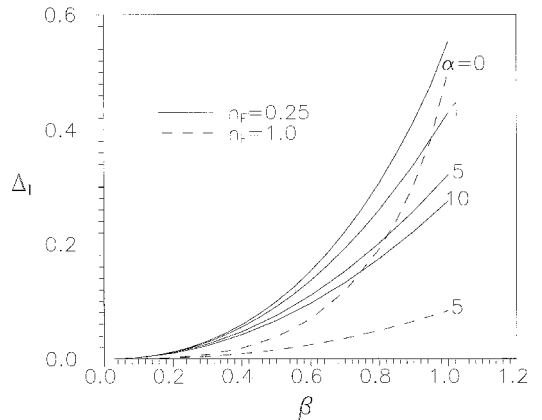

Fig. 3 Δ_I as a function of β for droplet ignition in a fuel-lean environment. Solid curves, $n_F = 0.25$; dashed curves, $n_F = 1.0$.

seen in Eq. (29), and the heat release caused by the reaction of the ambient fuel, contribute to enhancing chemical reactions and self-acceleration leading to the occurrence of an ignition event. It is also clear that for a given value of β, the ignition Damköhler number for the nonunity reaction order ($n_F = 0.25$) is larger than the corresponding value for the unity reaction order. This might be expected, since, for the same fuel concentration, the smaller reaction order yields a smaller rate of reaction and heat release, hence, a smaller temperature rise in the reaction zone. To have the same temperature increase needed for autoignition to occur, higher ambient fuel concentration is required for smaller reaction orders. However, as pointed out by Mawid and Aggarwal,[12] this does not necessarily imply that droplet ignition delay time would be longer for the nonunity reaction order of $n_F = 0.25$, as n_F also appears in Δ. The effect of the reaction order has to be examined for specific cases.

The results presented in Fig. 3 (i.e., for $n_F = 0.25$) is correlated into a single expression for easy application in spray combustion modeling. The following relation correlates the present numerical results within 20% for $0.02 \leq \beta \leq 1$:

$$\Delta_I = \frac{0.4627}{(1 + \alpha/31.18)^{1.85}} \beta^{f(\alpha)}, \quad f(\alpha) = 2 + \frac{0.27}{(1 + \alpha/15.11)^2} \quad (38)$$

For droplet autoignition in a purely oxidizing environment (i.e., $\alpha = 0$), the previous correlation is accurate within 2% for $0.02 \leq \beta \leq 0.4$.

Results are given in Fig. 4 for droplet ignition in an oxidant-lean environment for the oxidant reaction order of $n_O = 1.0$ and 1.5. It is clear that $\Delta_{1,I}$ decreases with κ, because higher ambient oxidant concentration, as Eq. (31) would suggest, produces more heat release, which would accelerate the fuel oxidation process toward the ignition event. For given values of κ, $\Delta_{1,I}$ is proportional to β^2. It is also seen that the ignition Damköhler number is smaller for a higher oxidant reaction order of $n_O = 1.5$, as compared with the case of $n_O = 1.0$. This is because for the same given conditions the higher value of n_O would result in faster chemical reactions with more heat release. Therefore, for given values of β, larger n_O needs a lower ambient oxidant concentration for the chemical reactions to culminate in autoignition when compared with the case of smaller n_O. However, for κ below a certain critical value (approximately between the value of 5–6 for the present case), the curves of $\theta(\infty)$ become single-valued for given values of Δ, indicating that ignition will not occur for sufficient low ambient oxidant concentration. This result is consistent with the findings of Normandia and Buckmaster.[34] On the other hand, the case of $\kappa \rightarrow \infty$ is equivalent to droplet in a premixed burning environment; this guarantees the occurrence of droplet ignition under such conditions.[5] The numerical results for
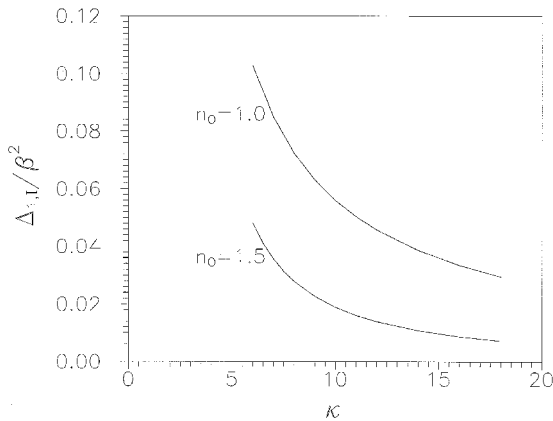
$n_O = 1.5$ can be correlated, within 1%, by the following expression:

$$\Delta_{I,1} = \frac{0.4830\beta^2}{(\kappa - 1.336)^{1.5}} \quad (39)$$

Figure 5 shows the ignition Damköhler number for droplet ignition in both fuel- and oxidant-lean environments for $n_F = 0.25$, $n_O = 1.5$, $\kappa = 10$, and two values of α. It is apparent that the ignition Damköhler number increases with β and decreases with α, as would be expected from the previous discussion. The case of $\alpha = 0$ is commensurable with the study of Normandia and Buckmaster.[34] As discussed earlier, ignition will not occur if κ is sufficiently small.

On the other hand, when comparing Eqs. (29), (31), and (36) with the results of Li and Reksizbulut,[13] it becomes clear that the ignition phenomenon is governed by different equations when an ignition event is considered to occur at a finite distance from the droplet surface. The reduced Damköhler number for the present case is inversely proportional to the square of the droplet evaporation rate, whereas it is directly proportional to the square of $\dot{m}$ for a droplet autoigniting in an unbounded oxidizing environment.[13] Further, the dependence of Δ, Δ_1, and Δ_2 on $\varepsilon = T_\infty^2/T_a$ is also significantly different for the two cases. This is understandable because the two situations differ physically. Therefore, it would lead to an unknown uncertainty in predicting the ignition delay time by using the ignition criterion developed for droplets in an unbounded environment. However, a comparison of Eqs. (29) and (31), with its counterpart given in Law and Chung,[15] reveals that the ignition criterion is governed by similar equations, whether the thermophysical properties are variable or assumed constant. The definition of β, as shown in Eq. (17), together with the frozen solution for the mass-flux fraction given in Eq. (12), suggests that the appropriate c_p relevant to ignition phenomenon should be that of the fuel vapor rather than that of the inert gases, as frequently used.

The present analysis also indicates that to the leading-order term, the specific dependence of the thermal conductivity and binary diffusion coefficients on temperature and composition does not affect the ignition Damköhler number. However, their effects do show up if the analysis is performed to the next (second) order. Therefore, they only make a minor contribution to Δ_I, $\Delta_{1,I}$, and $\Delta_{2,I}$. The main influences of variable thermophysical properties on the ignition phenomena are through the droplet evaporation rate $\dot{m}_f$, which appears in the reduced Damköhler number Δ, Δ_1, and Δ_2 as the square factor, and β, which increases the ignition Damköhler number under all of the conditions investigated, as shown in Figs. 3–5. Therefore, as expected, ignition will be more difficult to achieve for drop-
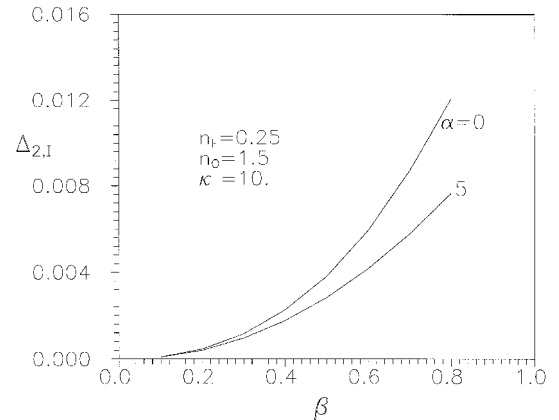

Fig. 4 $\Delta_{1,I}$ as a function of κ for droplet ignition in an oxidant-lean environment with $n_O = 1.0$ and 1.5.

Fig. 5 $\Delta_{2,I}$ as a function of β for droplet ignition in both fuel- and oxidant-lean environments with $n_F = 0.25$, $n_O = 1.5$, $\kappa = 10$, and α as shown.

lets with liquid-phase transient heating and large latent heat of vaporization, such as heavy hydrocarbon fuels.

The nonunity Lewis numbers influence directly the reduced system Damköhler number as shown in Eqs. (24), (34), and (37), and indirectly affect the ignition Damköhler number through parameters α and κ, as shown in Figs. 3–5. If the present results are compared with those corresponding to the case of equal binary diffusion coefficients, it is found that the reduced system Damköhler numbers for both cases are identical, provided that a mixture Lewis number is defined as

$$Le_\infty = \sum_j Le_{Fj,\infty} X_{j,\infty} \tag{40}$$

for droplet ignition in a fuel-lean environment, and is defined as

$$Le_\infty = \sum_j Le_{Oj,\infty} X_{j,\infty} \tag{41}$$

for droplet ignition in oxidant-lean surroundings. However, for ignition in an ambience that is lean in both fuel and oxidant, the mixture Lewis number should be determined from the following relation:

$$Le_\infty = \left[\left(\sum_j Le_{Fj,\infty} X_{j,\infty} \right)^{n_F} \left(\sum_j Le_{Oj,\infty} X_{j,\infty} \right)^{n_O} \right]^{1/n} \tag{42}$$

Therefore, Eqs. (40–42) indicate that the frequently invoked assumption of the equal binary diffusion coefficients is acceptable for the prediction of ignition phenomenon, provided that an appropriate mixture Lewis number is evaluated according to the previous equations.

Finally, it should be pointed out that if the droplet surface temperature is not a constant, but rather, is perturbed by the amount of, say, εT_{s1}, then it can be shown that the value of the maximum $\theta(\infty)$ in the inner diffusive–reactive region is increased by a constant amount proportional to T_{s1}, whereas the ignition Damköhler number is not affected. Hence, the results of the present ignition analysis remain the same. However, droplet heating will influence the ignition delay time, e.g., as shown by Mawid and Aggarwal.[12] The results of the present work, particularly Eqs. (38) and (39), can be easily incorporated, as a submodel for droplet ignition, into an overall spray combustion model, as shown by Huang and Chiu.[36]

Conclusions

This study analyzes the ignitability of a fuel droplet in a reactive oxidant/fuel-vapor/inert environment simulating droplets in the spray interior. The matched asymptotic analysis of large activation energy is employed, and an overall one-step irreversible chemical reaction is considered with the Arrhenius kinetics and arbitrary fuel and oxidizer reaction orders. The variable thermophysical properties are taken to depend on both temperature and species composition of the gas mixture with distinct binary diffusion coefficients and nonunity Lewis numbers. It is found that the ignition Damköhler number increases with the thermal driving force for evaporation and increases significantly with a reduction in the fuel and oxidant reaction orders, and it decreases as the ambient fuel and oxidant concentration are increased. Further, ignition will not occur if the ambient oxidant concentration is sufficiently low. It is also found that the specific heat should be that of the fuel rather than that of the inert gases, and the mixture Lewis number should be defined appropriately, depending on the ambient fuel and oxidant concentration as given in Eqs. (38–40), if equal binary diffusion coefficients are used. The variable thermophysical properties affect ignition phenomenon partly through the droplet mass evaporation rate and partly through the thermal driving force for evaporation. The former affects the re-

duced Damköhler number, and the latter influences the ignition Damköhler number. If proper care is taken of the mixture Lewis number, the frozen evaporation rate of the droplet, and the thermal driving force, the ignition phenomenon can be predicted accurately from the analysis with constant properties with equal binary diffusion coefficients.

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