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HYDRODYNAMIC STABILITY OF MULTICOMPONENT DROPLET
GASIFICATION IN REDUCED GRAVITY

I. Aharon and B. D. Shaw
Mechanical and Aeronautical Engineering Department
University of California
Davis, CA 95616

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ABSTRACT

This investigation addresses the problem of hydrodynamic stability of a two-component droplet undergoing spherically-symmetrical gasification. The droplet components are assumed to have characteristic liquid species diffusion times that are large relative to characteristic droplet surface regression times. The problem is formulated as a linear stability analysis, with a goal of predicting when spherically-symmetric droplet gasification can be expected to be hydrodynamically unstable from surface-tension gradients acting along the surface of a droplet which result from perturbations. It is found that for the conditions assumed in this paper (quasisteady gas phase, no initial droplet temperature gradients, diffusion-dominated gasification), surface tension gradients do not play a role in the stability characteristics. In addition, all perturbations are predicted to decay such that droplets were hydrodynamically stable. Conditions are identified, however, that deserve more analysis as they may lead to hydrodynamic instabilities driven by capillary effects.

INTRODUCTION

Influences of volatility differences and liquid-phase transport on the vaporization and combustion behaviors of multicomponent droplets have been the subject of numerous investigations because of their potential importance to the efficient use of fuel sprays as well as their fundamental interest. Many of these investigations have focused upon

spherically-symmetric low Reynolds-number evaporation and combustion of multicomponent droplets. These investigations have demonstrated that liquid-phase species transport may be important when surface regression is fast compared to species transport, and when significant volatility differences exist between liquid components. Landis and Mills [1], for example, report results of numerical analyses concerning spherically-symmetric evaporation of miscible binary droplets, demonstrating the effects of volatility differences and slow liquid-phase species transport. Law et al. have presented numerical and theoretical analyses for spherically-symmetric binary fuel evaporation with or without combustion [2-4]. Experimental work in this area has been active [see 5-12 for example]. Law et al. [12,13] have also reported results regarding evaporation of methanol or other liquid droplets in humid air; a focus of these studies was the effects of water condensation on droplet evaporation. Reduced-gravity experiments on methanol droplet combustion where combustion-generated water was reasoned to have condensed onto the liquid droplet have been reported by Choi et al. [14].

Some of these investigations have demonstrated, for binary droplets with sufficiently different liquid component volatilities, that an initial transient period exists during which the more volatile component is preferentially evaporated. During this time, as the droplet volume decreases, the fraction of the less volatile component in the droplet is increased relative

to the initial state. Surface regression, when sufficiently rapid compared to liquid species transport, causes a "concentration boundary layer" to develop near the droplet surface, where species profiles exhibit sharp spatial changes; the less volatile component is "swept up" in a thin layer because it cannot diffuse away from the droplet surface rapidly enough. It has been observed that under appropriate conditions (e.g., with species transport sufficiently slow compared to surface regression rates), a state may be attained where changes in droplet compositions occur slowly with respect to changes in droplet size [1,2]. This state, which occurs after the initial transients have decayed, typically exhibits a concentration boundary-layer structure.* For sufficiently small diffusion rates, this boundary-layer structure may persist for most of the remaining droplet lifetime.

Based on this observation, Law and Law [11] presented an asymptotic analysis concerning spherically-symmetric vaporization and combustion behaviors of multicomponent droplets for the case where liquid-phase species diffusion is slow compared to droplet surface regression rates. In their analysis, Law and Law focused upon liquid-phase behavior for the case where the boundary-layer structure changes slowly, following decay of initial transients. A conclusion of this work is that once the boundary-layer composition profiles have been established in a droplet, the mass-flux fraction of any species off the droplet surface is equal to the initial liquid-phase mass fraction of that species prior to vaporization. The analysis of Law and Law [11] was extended by Shaw [15], who demonstrated that following decay of initial transients, the mass-flux fraction of any species off the droplet surface is instead approximately equal to the volume-averaged mass fraction of that species in the droplet interior.

If temperature or species concentration gradients exist at the surface of a liquid droplet (e.g., because of small perturbations in the environment or within the droplet), it is

* Here, "boundary layer" refers to the species concentration boundary layer near the droplet surface.

possible for surface-tension gradients to exist. If sufficiently intense, these surface-tension gradients can cause significant motions of the liquid-gas interface, influencing transport of species and energy in the gas and liquid phases. It is capillary effects such as these that this paper is concerned with. Theoretical studies will be performed to determine the influence of capillary convection on droplet hydrodynamic stability, with a goal of determining how capillary convection may compromise the attainment of spherical symmetry. The goal of this research is to perform a linear stability analysis to determine whether the spherically-symmetrical state is stable with respect to small perturbations in the flow variables. Viscous effects as well as the effects of surface tension gradients will be allowed. The analysis will consider only the portion of the droplet history where the spherically-symmetrical species profiles may be considered quasi-steady. Earlier portions of the droplet history associated with transient liquid heating and the initial buildup of the species concentration profiles will not be addressed.

It is noted that there have been many theoretical and experimental studies dealing with the motions of droplets and bubbles resulting from capillary effects. A literature review has revealed, however, that most previous studies of droplet gasification have generally neglected effects of surface-tension gradients, though a few studies have appeared. Higuera and Liñán [16] have presented a linear stability analysis of an unsupported and stationary droplet vaporizing in a hot stagnant atmosphere. Chai et al. [17] have also presented results pertaining to stability of evaporating droplets vaporizing in a stagnant environment. Lozinski and Matalon [18] presented analytical studies of spinning droplets with thermocapillary effects. Niazmand et al. [19] have recently presented numerical calculations for vaporizing droplets that are moving relative to their environment. The calculations of Niazmand et al. showed that capillary flows can significantly affect droplet internal velocity and temperature fields of vaporizing octane and methanol droplets, even when droplet initial Reynolds numbers

(relative to the gas phase) are as large as 50. It is noted that following the initial publications of Niazmand et al. [19], Shih and Megaridis [20] presented computational results on the effects of thermocapillary stresses and flows for nearly the same conditions as those considered by Niazmand et al. None of these previous studies have considered droplets with more than liquid species, which is the primary subject of this paper.

ESTIMATES OF THE IMPORTANCE OF CAPILLARY EFFECTS

Estimates of the importance of capillary effects may be made by considering a droplet of radius R . A temperature or species concentration gradient along the liquid surface will produce a surface-tension gradient that will be balanced by surface shear stresses in the gas and liquid phases. For these estimates, gas-phase shear stresses will be assumed negligible relative to liquid shear stresses such that surface-tension gradients are primarily balanced by liquid shear. Neglect of gas-phase shear in surface force balances is justified if the gas-phase viscosity is small relative to the liquid viscosity, a condition that is generally satisfied for subcritical conditions, and if liquid velocities relative to the ambient gas are small, i.e., there is not a thin momentum boundary layer in the gas phase. Liquid-phase shear will be assumed to be characterized by $\mu_L u/R$, where μ_L is the liquid viscosity, u a velocity characteristic of the flow induced in the droplet and u/R represents a characteristic velocity gradient at the droplet surface. Use of R as a characteristic distance should be appropriate since for hydrocarbon and alcohol fuels of interest, the ratio of the evaporation constant k (based on the droplet diameter, that is, $k = -4d(R^2)/dt$, where t is time) to the liquid kinematic viscosity ν_L is of order unity or less (i.e., $k/\nu_L \leq 1$). Physically, if $k/\nu_L \leq 1$, momentum changes at the droplet surface should have sufficient time to influence the droplet interior before the droplet evaporates. A balance between liquid shear stress and

surface tension gradients gives $u \sim \Delta\sigma/\mu_L \sim r/t_c$ where $\Delta\sigma$ is a characteristic difference in surface tension from one side of the droplet to the other, and t_c is a characteristic time for a fluid particle to be convected halfway around the droplet periphery (surface-tension gradients are characterized by $\Delta\sigma/R$, and gas-phase shear is neglected assuming that the liquid-phase viscosity is much larger than the gas-phase viscosity). If we define $t_b = R^2/k$ as a characteristic gasification time of a droplet, we may expect that capillary effects are negligible if $t_b/t_c = R\Delta\sigma/(k\mu_L) \ll 1$. Conversely, if $R\Delta\sigma/(k\mu_L) \geq 1$, capillary effects may cause significant convective flows over the lifetime of a droplet.

The magnitude of $\Delta\sigma$ depends upon gradients of temperatures and species concentrations along the droplet surface. Let us first consider surface temperature gradients alone. For this case we may write $\Delta\sigma = |\partial\sigma/\partial T| \Delta T$, where T is temperature, σ surface tension, and ΔT is the temperature difference from one side of the droplet to the other. If this relation is used in the expression for t_b/t_c , we may solve for $\Delta T \sim k\mu_L/(R|\partial\sigma/\partial T|)$ as being required for $t_b/t_c \sim 1$. For a 1 mm diameter hydrocarbon or alcohol droplet undergoing combustion, estimates suggest that $\Delta T \sim 0.01$ K is required to make $t_b/t_c \sim 1$. These estimates are based upon the characteristic values $k = 10^{-6}$ m²/s, $\mu_L = 4 \times 10^{-4}$ kg/(m s), and $|\partial\sigma/\partial T| = 10^{-4}$ N/(m °K), which are appropriate for near-atmospheric pressure conditions and droplet gasification in a hot environment or combustion. Since the ratio t_b/t_c scales linearly with ΔT , increases in ΔT produce increases in t_b/t_c . It thus appears that even very small temperature differences may induce significant convective flows.

We may also consider the effects of species concentration gradients along the droplet surface, neglecting temperature gradients. For simplicity, attention will be restricted to

droplets composed of two miscible liquid components. For this case, we may write $\Delta\sigma \sim |\partial\sigma/\partial x_1| \Delta x_1$ where x_1 is the mole fraction of component "one" at the liquid surface, and Δx_1 is the difference in x_1 from one side of the droplet to the other. Use of this result in the expression for t_b/t_c yields $\Delta x_1 \sim k\mu_L/(R |\partial\sigma/\partial x_1| h)$ for $t_b/t_c \sim 1$.

Consider a methanol droplet of about 1 mm diameter. It is known that methanol (or other alcohol) droplets may absorb water from the gas phase. The characteristic values $|\partial\sigma/\partial x_1| = 0.03 \text{ N/m}$, $k = 10^{-6} \text{ m}^2/\text{s}$, and $\mu_L = 4 \times 10^{-4} \text{ kg/(m s)}$ are appropriate for making order-of-magnitude estimates for methanol-water mixture droplets undergoing combustion at near atmospheric pressure. Use of these values provides the result that $\Delta x_1 \sim 10^{-5}$ for $t_b/t_c \sim 1$. Hence, very small variations in water concentration along the surface of a methanol droplet may induce significant convective flows. Since the ratio t_b/t_c scales linearly with Δx_1 , larger values of Δx_1 produce increases in t_b/t_c . Significant capillary flows will negate the possibility of attaining liquid-phase spherical symmetry by enhancing mixing in an asymmetric and unknown manner. Estimates similar to those presented above for methanol-water mixtures have also been made for miscible binary hydrocarbon mixture droplets. These estimates suggest that for droplet components that vary significantly in volatility, e.g., heptane-hexadecane, small differences in surface mole fractions of one component ($\Delta x \sim 10^{-4}$) may cause $t_b/t_c \sim 1$. Such droplets (with significant volatility differentials) have been the subject of a number of reduced-gravity experimental efforts which are striving for spherical symmetry.

Based upon the above estimates, it appears that small perturbations in temperature or species profiles along the surface of a binary miscible droplet may induce significant convective flows. If sufficiently intense, these convective flows will seriously

compromise spherical symmetry, making interpretation of reduced-gravity experimental results (which are presumed to be spherically symmetrical) difficult. For these reasons, theoretical studies of the influences of capillary phenomena on droplet gasification and combustion are presented below. These studies will allow the influences of capillary phenomena to be delineated with greater precision, allowing for better interpretation of reduced-gravity experimental results, and also for increased understanding of droplet gasification phenomena in general. For simplicity, the analyses are restricted to evaporation in a hot environment; effects of gas-phase combustion will be considered at a later time. However, it is noted that the results presented here should be valid for combustion of droplets when the flame lies a significant distance away from the droplet surface, for example when droplets are burned in environments with low oxidizer concentrations.

In the following, the basic unperturbed solution for quasisteady droplet evaporation in a hot environment will first be presented. This solution, which is presented for the convenience of the reader, is a summary of an analysis presented by Shaw [15]. After presentation of this solution, the viscous-flow governing equations will be perturbed and linearized. These equations will be solved and stability criteria will be developed.

THE UNPERTURBED SOLUTION

Liquid-Phase Analysis

With the assumption of a constant liquid species diffusivity D_L and also assuming that liquid densities do not vary with time, liquid-phase species conservation may be taken to be described by the partial differential equation and boundary conditions (where y_i represents the mass fraction of species i)

$$\frac{\partial y_i}{\partial t} = \frac{D_L}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial y_i}{\partial r} \right) \quad (1)$$

$$\left(D_1 \frac{\partial y_1}{\partial r} + y_1 \frac{dR}{dt}\right)_{r=R} = \delta_1 \frac{dR}{dt}, \quad \left(\frac{\partial y_1}{\partial r}\right)_{r=0} = 0$$

and the initial condition $y_1(r,0)=y_{10}$. Here, y_{10} is taken for simplicity to be constant. The variable δ_1 is the mass-flux fraction of species 1 from the droplet surface i.e., $\delta_1 = (dm_1/dt)/(dm/dt)$, where m is the droplet mass, m_1 the mass of component 1 in the droplet, and t the time. The droplet surface is regressing, so the location of the liquid-phase boundary is time dependent. In the following, the droplet surface is fixed at unity with the transformation $x=r/R$. Then with $\varepsilon = D_L/[R(dR/dt)]$, liquid phase species conservation is described by the initial value problem

$$\frac{\partial y_1}{\partial \tau} = \frac{\varepsilon}{x^2} \frac{\partial}{\partial x} \left(x^2 \frac{\partial y_1}{\partial x} \right) - x \frac{\partial y_1}{\partial x} \quad (2)$$

$$\left(\varepsilon \frac{\partial y_1}{\partial x} - y_1\right)_{x=1} = -\delta_1, \quad \left(\frac{\partial y_1}{\partial x}\right)_{x=0} = 0$$

$$y_1(x,0) = y_{10}.$$

The physical time t and the nondimensional

time τ are related by $t = \int_0^\tau d\tau [d \ln(R_0/R)/d\tau]$.

When d-square law surface regression holds, i.e., where the droplet surface area decreases linearly with time, $\varepsilon = 8D_L/k$, where $k = 8R(dR/dt)$ is the evaporation constant. For this case it then follows that $t/t_b = 1 - e^{-2\tau}$, where $t_b = 4R_0^2/k$ is the droplet evaporation lifetime and R_0 is the initial droplet radius.

The situation of interest here is when liquid-phase species transport is slow relative to droplet surface regression. Consider the second component to have a volatility that is low relative to the first component. The more volatile component will be initially preferentially vaporized, leaving the less volatile component behind in the droplet. The droplet will become richer in the less volatile component relative to the initial state, with most of the lower-volatility component left

behind being collected within a boundary layer at the droplet surface. As surface regression proceeds, the surface mass fraction of the less volatile component builds, and a state will eventually be attained where vaporization of the less volatile component becomes significant. Since both species will then vaporize vigorously, and liquid-phase diffusion is slow, further changes in droplet species profiles will occur slowly. Only that part of the combustion history where both components are vaporizing vigorously with slowly-changing species profiles will be analyzed here. Analyses of transient boundary layer buildup have been given elsewhere [21].

Inspection of Eq. (2) suggests that once boundary layer growth has slowed considerably and surface conditions change slowly, further changes in species profiles over the droplet radius will occur over times $\Delta\tau$ which may be of order $(1/\varepsilon)$. The time period $\Delta\tau \sim O(1/\varepsilon)$ may comprise the major portion of the remaining droplet lifetime in physical time, since $\varepsilon \ll 1$ is expected, as argued below. Because characteristic changes in y are of order unity or less, the magnitude of the partial derivative $\partial y_1/\partial \tau$ for these times may be estimated as $(\partial y_1/\partial \tau) \lesssim O(\varepsilon)$. Hence, it may be assumed that the boundary-layer structure which has evolved once vigorous vaporization of both components begins will be adequate for evaluating δ_1 . These observations are qualitatively supported by available numerical results [2], where the boundary-layer structures were observed to persist for most of the droplet lifetime, until near the end, when the more volatile component was vaporized much faster than the less volatile component.

Analysis of the boundary layer structure proceeds by considering Eq. (2). Because D_L is generally small compared to characteristic magnitudes of $R(dR/dt)$, it may be considered that $\varepsilon \ll 1$. The "stretched" coordinate $z = (1-x)/\varepsilon$ may then be inserted into Eq. (2). By using techniques of matched asymptotic

expansions, it may be shown [15] that the mass-flux fraction δ_i may be expressed as

$$\delta_i = y_{i0} + 3\varepsilon(y_{i1} - y_{i0}) + O(\varepsilon^2) \quad (3)$$

while the asymptotic boundary-layer structure is provided by

$$y_i = y_{i0} +$$

$$(y_{i1} - y_{i0})\exp(-z)[1 + \varepsilon(3z + z^2/2) + \dots] \quad (4)$$

To complete the formulation of the model, the gas phase and the gas/liquid coupling must be analyzed, as presented below.

Gas Phase Analysis

For analysis of the gas phase, the simplifications of unity Lewis number, quasisteadiness i.e., neglect of the transient terms in the governing equations, negligible body forces, negligible radiant heat flux, equal and constant specific heats and binary diffusion coefficients, and that diffusion obeys Fick's law will be employed. In the following, the subscript + denotes that conditions are evaluated on the gas side of the gas-liquid interface.

Assuming spherical symmetry, the energy and species conservation equations for the gas phase can respectively be written as

$$\rho v \frac{c_p}{\lambda} \frac{dT}{dr} = \frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{dT}{dr} \right)$$

$$v \frac{dY_i}{dr} = \frac{D_g}{r^2} \frac{d}{dr} \left(r^2 \frac{dY_i}{dr} \right)$$

where ρ is density, c_p is specific heat, λ is thermal conductivity (assumed constant), T is temperature, r is the radial distance outward from the droplet center, D_g is the gas-phase species diffusivity (assumed constant and equal for all species), Y_i the mass fraction of species i , and v is the gas velocity. These equations are to be integrated subject to the conditions that $T = T_+$ and $Y_i = Y_{i+}$ at the

droplet surface ($r = R$) as well as $T = T_\infty$ and $Y_i = Y_{i\infty}$ at infinity. The solutions to these equations can be written as

$$\frac{T - T_\infty}{T_+ - T_\infty} = \frac{Y_i - Y_{i\infty}}{Y_{i+} - Y_{i\infty}} = \frac{1 - \exp(-\phi/r)}{1 - \exp(-\phi/R)} \quad (5)$$

where the assumption of unity Lewis number has been used and $\phi = r^2 \rho v (c_p/\lambda)$. By applying the conditions for conservation of energy and species at the droplet surface ($\rho_+ v_+ L = -\lambda (dT/dr)_+$ and $\delta_i = Y_{i+} - (D_g/v)(dY_i/dr)_+$) it may be shown that $\phi = R \ln(1 + B_T) = R \ln(1 + B_i)$ where $B_T = c_p(T_\infty - T_+)/L$ and $B_i = (Y_{i+} - Y_{i\infty})/(\delta_i - Y_{i+})$ are transfer numbers, δ_i is the mass-flux fraction of species i in the gas phase, and $L = \sum \delta_i L_i$ is the heat of vaporization of the liquid mixture at the droplet surface. It is noted that B_T and B_i are required to have the same numerical values.

From conservation of mass principles, it can also be shown that $k = 8\lambda \ln(1+B)/(\rho_L c_p)$ where $B = B_T = B_i$. It may be shown that the surface mass fraction of a droplet component is calculated using $Y_{i+} = (B\delta_i + Y_{i\infty})/(1 + B)$, while ε is related to B by

$$\varepsilon = (D_L \rho_L c_p)/[\lambda \ln(1+B)] \quad (6)$$

Gas/Liquid Coupling Analysis

To complete the formulation, the coupling between the gas and liquid phases must be considered. For simplicity, in this analysis phase equilibrium and ideal solution behavior will be assumed to describe vaporization. Mole fractions (X) and mass fractions are related by the identity $X_i = (Y_i/W_i)/(\sum Y_j/W_j)$. Gas and liquid mole fractions will be related using Raoult's law and the Clausius-Clapeyron relation with constant vaporization enthalpy, giving

$$X_{i-} = X_{i+} \exp[L_i(1/T_+ - 1/T_{b_i})/R_i] \quad (7)$$

In Eq. (7), T_{b_i} is the boiling temperature of component i at the ambient pressure, and R_i is the gas constant for species i . The phase equilibrium relations used here are not essential for the development of this model. Nonideal mixtures may be modeled, for example, with the use of activity coefficients.

To solve for the spherically-symmetrical evaporation state, we first specify y_{10} . Once this is done, values for the surface composition in the gas phase are assumed (subject to $\sum Y_{1+} = 1$). The surface temperature is then iterated until $\sum \delta_i = 1$ is attained for the gas phase. The gas-liquid coupling relations are then used to calculate the surface composition on the liquid side of the gas-liquid interface. The parameter ϵ is then calculated using Eq. (6), and a value for y_{10} based upon the values found above is finally calculated from Eq. (3). If this value is not sufficiently close to the initially-specified value of y_{10} , the above procedure is repeated with new values for the surface composition in the gas phase. Calculations for various cases are presented by Shaw [15], where it is shown that the $O(\epsilon)$ correction to δ_i present in Eq.(3) is dominant when y_{10} is sufficiently small.

The spherically-symmetrical model has now been completely formulated. The results presented above are required for the perturbation analysis developed in the next section.

LINEARIZED STABILITY EQUATIONS

Liquid Phase

The conservation equations for the liquid are assumed to be the incompressible constant-property relations appropriate for viscous fluids with negligible viscous dissipation. These equations are listed below.

$$\nabla \cdot \vec{v} = 0 \quad (\text{Continuity})$$

$$\frac{d\vec{v}}{dt} = -\nabla p + \mu \Delta \vec{v} \quad (\text{Momentum})$$

$$\frac{dT}{dt} = \alpha_l \Delta T \quad (\text{Energy})$$

$$\frac{dy_i}{dt} = D_i \Delta y_i \quad (\text{Species})$$

It is to be noted that ∇ represents the gradient operator while Δ represents the Laplacian operator. In addition, \vec{v} is the velocity vector, p is pressure, T is temperature, and α_l is the thermal diffusivity of the liquid. In addition,

$\frac{d}{dt} = \frac{\partial}{\partial t} + \vec{v} \cdot \nabla$ is the Lagrangian derivative. Since the droplet surface is regressing, it is convenient (as outlined above) to define the normalized radial coordinate $x = r/R$. When this is done, the Lagrangian derivative is transformed to the form

$$\frac{d}{dt} = \frac{\partial}{\partial t} + \frac{\vec{v}}{R} \cdot \nabla^* - x \frac{1}{R} \frac{dR}{dt} \frac{\partial}{\partial x}$$

where ∇^* is the dimensionless gradient operator expressed in terms of x .

We will now perturb the liquid-phase variables by assuming that $\vec{v} = \vec{v}_0 + \vec{v}'$, $p = p_0 + p'$, $T = T_0 + T'$, $y_i = y_{i0} + y_i'$ where the subscript "o" denotes the solution for the spherically-symmetrical state and the superscript "prime" denotes the perturbation quantity. It is assumed that $\vec{v}_0 = 0$, $p_0 = \text{constant}$, $T_0 = \text{constant}$, and that y_{i0} is predicted by Eq. (4). The assumption of constant T_0 is consistent with the idea that early transients associated with initial droplet heat-up have decayed.

Substitution of the perturbed variables presented above into the liquid-phase governing equations produces evolution equations for the perturbation variables (note that "primes" will be omitted for brevity). In

deriving these equations, the dimensionless variables $\tilde{V}_L = (8R/k)\tilde{V}$, $P_L = 8p'R^2/(k\mu_L)$ and $T_L = T/T_\infty$ are introduced, where T_∞ is the ambient temperature and the subscript "L" denotes liquid. The perturbation equations are linearized by neglecting products of the perturbation quantities or their derivatives, and terms involving the spherically-symmetrical base solutions cancel. In addition, the dimensionless time τ defined earlier is introduced, though τ is rescaled as described below (note that $\exp(-\tau) = R/R_0$). The timescale for liquid-phase species diffusion is generally much greater than that for diffusion of energy. A characteristic time τ_0 for species diffusion across the thickness e of the boundary layer is simply $\tau_0 = e$ (on the τ timescale). For analysis of the equations, we therefore introduce the rescaled time $\tilde{\tau} = \tau/e$ to produce the final form of the equations, which are shown below.

$$\nabla \cdot \tilde{V}_L = 0 \quad (\text{Continuity}) \quad (8)$$

$$\Delta \tilde{V}_L - \nabla P_L = 0 \quad (\text{Momentum}) \quad (9)$$

$$\frac{1}{e} \frac{\partial T_L}{\partial \tilde{\tau}} + x \frac{\partial T_L}{\partial x} = e Le \Delta T_L \quad (\text{Energy}) \quad (10)$$

$$\frac{1}{e} \frac{\partial y_i}{\partial \tilde{\tau}} + x \frac{\partial y_i}{\partial x} + \tilde{V} \cdot \nabla y_i = e \Delta y_i \quad (\text{Species}) \quad (11)$$

In deriving Eqs. (8) - (11), it has been assumed that the velocity field in the liquid phase will be a "creeping" flow (i.e., dominated by viscosity) such that fluid acceleration can be neglected in the first approximation. The variable Le is the liquid-phase Lewis number; Le is assumed to be large relative to unity and the product eLe is treated as $O(1)$.

Solutions to Eqs. (8) - (11) will be sought assuming that each scalar perturbation

quantity ($Z(x, \theta, \phi)$) can be expressed in terms of a normal mode decomposition based upon expansions in terms of spherical harmonics

$$Z = \sum_{n=0}^{\infty} \sum_{m=-n}^n Z_n(x) Y_n^m(\theta, \phi) e^{\beta_n \tilde{\tau}} \quad (12)$$

where Z_n depends only on the transformed radial coordinate x and $Y_n^m(\theta, \phi)$ is the spherical harmonic function. The function Z_n represents a spatial amplitude of the perturbation mode n . A normal-mode analysis is appropriate here since the unperturbed solution is quasisteady in the time variable τ . A final goal of the analysis will be to calculate the growth factor β_n , which will determine whether a perturbation will grow, decay, or remain steady.

If we multiply the radial component of the momentum equation (V_x) by x and then twice take the curl, we obtain the equation

$$\left(\frac{d^2}{dx^2} + \frac{2}{x} \frac{d}{dx} - \frac{n(n+1)}{x^2} \right)^2 (x V_{Lx}) = 0 \quad (13)$$

where V_{Lx} is the amplitude of the amplitude of the velocity perturbation in the x -direction for the n^{th} mode (for brevity the subscript "n" is dropped from all equations involving amplitudes). In deriving this equation, use was made of the identity

$$\Delta [f(x) Y_n^m(\theta, \phi)] = Y_n^m(\theta, \phi) \left[\frac{d^2 f}{dx^2} + \frac{2}{x} \frac{df}{dx} - \frac{n(n+1)}{x^2} f \right]$$

which is applicable for spherical harmonics and where $f(x)$ is an arbitrary function of x . Equation (13) is to be solved subject to the condition that the solution is finite at $x = 0$. Integration yields

$$V_{Lx} = a_1 x^{n-1} + a_2 x^{n+1} \quad (14)$$

where a_1 and a_2 are integration constants that will be determined by considering the solutions at the gas-liquid interface ($x = 1$). Interface conditions will be discussed later.

The solution to the energy equation (Eq. (10)) may be developed by noting that we may reasonably expect that $\epsilon \ll 1$. To maintain a balance between dominant terms in Eq. (10), it can be assumed that time derivatives are at most $O(1/Le)$ in magnitude, which implies that the perturbed temperature field may be considered quasisteady in the first approximation. We will thus drop the transient term from Eq. (10), allowing us to focus upon the effects of species perturbations on stability. Effects of temperature perturbations were studied by Higuera and Liñán [16], who found that instabilities from temperature perturbations were important only when the base solution exhibits significant temperature gradients at the droplet surface; spatial temperature gradients are assumed to have decayed to negligible levels for the conditions under study here. If we now assume a solution of the form of Eq. (12), the resulting ordinary differential equation for the amplitude T_{Lx} is

$$\frac{d^2 T_{Lx}}{dx^2} + \left(2 - \frac{x}{\epsilon Le}\right) \frac{dT_{Lx}}{dx} - \frac{n(n+1)}{x^2} T_{Lx} = 0. \quad (15)$$

The point $x = 0$ of Eq. (15) is a regular singular point, and it is possible to develop a Frobenius series solution about $x = 0$. When this is done, it is found that the infinite-series solution actually converges very rapidly, and as a result the truncated expansion

$$T_{Lx} = a_3 \left[x^n + \frac{1}{\epsilon Le} \frac{1}{16(2n+3)} x^{n+2} \right] \quad (16)$$

may be used with little loss in accuracy (a_3 is a constant of integration). Applicability of Eq. (16) is based upon the condition that ϵLe is not small relative to unity; this condition is typically well-satisfied for droplet vaporization problems. Though not shown here, numerical solutions of Eq. (15) compare very well with Eq. (16) for the assumed conditions. In developing Eq. (16), the condition $T_{Lx} = 0$ was applied at the droplet center.

Substituting Eq. (12) into Eq. (11) yields the evolution equation for the amplitude y_{ILx} of the liquid-phase species perturbation

$$\begin{aligned} & \beta_n y_{ILx} + \epsilon x \frac{dy_{ILx}}{dx} \\ & + (a_1 x^{n-1} + a_2 x^{n+1}) (y_{I-} - y_{I0}) e^{(x-1)/\epsilon} \\ & = \epsilon^2 \left(\frac{d^2 y_{ILx}}{dx^2} + \frac{2}{x} \frac{dy_{ILx}}{dx} - \frac{n(n+1)}{x^2} y_{ILx} \right). \end{aligned} \quad (17)$$

In developing this equation, the leading-order solution for droplet species profiles was used (see Eq. (4)). An asymptotic solution to Eq. (17) will be sought assuming that $\epsilon \ll 1$, which is generally met for droplet gasification in hot environments. Examination of Eq. (19) indicates that a boundary-layer structure may be expected. The boundary layer will exist close to the droplet surface, and it is found that an appropriate scaling to allow analysis of the boundary-layer is provided by the transformation $x = 1 - \epsilon z$, where $z \leq O(1)$ in the boundary layer. Using this transformation and retaining only the largest terms provides the leading-order inner problem

$$\begin{aligned} & \frac{d^2 y_{ILx}}{dz^2} + \frac{dy_{ILx}}{dz} - \beta_n y_{ILx} = \\ & (a_1 + a_2) (y_{I-} - y_{I0}) \exp(-z). \end{aligned} \quad (18)$$

The solution to Eq. (18) may be written as $y_{ILx} = a_{4I} \exp(s_4(1-x)/\epsilon) + a_{5I} \exp(s_5(1-x)/\epsilon) - (a_1 + a_2)(y_{I-} - y_{I0}) \exp[(x-1)/\epsilon]/\beta_n$, where a_{4I} and a_{5I} are constants and $s_4 = [-1 - (1 + 4\beta_n)^{1/2}]/2$ and $s_5 = [-1 + (1 + 4\beta_n)^{1/2}]/2$. The leading-order outer solution to Eq. (17) is simply $y_{ILx} = 0$. To match the inner and outer solutions we set $a_{5I} = 0$, because for $\text{Re}(\beta_n) > 0$ the inner solution would grow exponentially fast for increasing z . The solution to Eq. (20) may therefore be expressed as

$$\begin{aligned} & y_{ILx} = a_{4I} \exp[s_4(1-x)/\epsilon] - \\ & (a_1 + a_2) (y_{I-} - y_{I0}) \exp[(x-1)/\epsilon]/\beta_n. \end{aligned} \quad (19)$$

Gas Phase

For analysis of the gas phase, we will introduce the simplifying assumptions that density changes, which are a result of temperature changes, may be neglected in the perturbation equations. We will also assume that the vaporization is slow such that $\phi/R = \ln(1+B) \ll 1$ (but where $\epsilon = 8D_1/k$ is also small relative to unity). This allows simplified analytical solutions to be found for the gas-phase perturbation variables. For analysis of the case where $\phi/R \gtrsim 1$ and where density perturbations cannot be neglected, numerical integration of the perturbation equations is needed; this analysis is now in progress and will be presented elsewhere.

When $\phi/R \ll 1$, it is easily shown that a Reynolds number based upon the gas velocity and gas properties at the droplet surface is small relative to unity. Under these conditions, we may neglect the convective acceleration terms in the momentum equation. We will perturb the spherically-symmetrical solution by assuming that $\vec{v} = \vec{v}_0 + \vec{v}'$, $p = p_0 + p'$, $T = T_0 + T'$, and $Y_i = Y_{i0} + Y_i'$ where the subscript "o" denotes the solution for the spherically-symmetrical state and the superscript "prime" denotes the perturbation quantity. If we further let $x = r/R$ and define the dimensionless perturbed variables $\vec{V}_G = (\rho_G/\rho_L)(8R/k)\vec{v}'$, $T_G = T'/T_\infty$, $Y_{iG} = Y_i'$ and $P_L = 8p'R^2(\rho_G/\rho_L)/(k\mu_L)$, the linearized conservation equations can be written as

$$\nabla \cdot \vec{V}_G = 0 \quad (\text{Continuity}) \quad (20)$$

$$\Delta \vec{V}_G - \nabla P_G = 0 \quad (\text{Momentum}) \quad (21)$$

$$\Delta T_G = O(\phi/R) \quad (\text{Energy}) \quad (22)$$

$$\Delta Y_{iG} = O(\phi/R) \quad (\text{Species}) \quad (23)$$

where the subscript "G" denotes the gas phase. Solutions to Eqs. (20) - (23) can be developed in the same fashion as was done for the liquid-phase equations. The solutions for the amplitudes of the n^{th} mode of each perturbation variable are listed below, where the condition that perturbations should vanish as $x \rightarrow \infty$ was applied and also where terms of order ϕ/R or smaller were neglected.

$$V_{Gx} = b_1 x^{-n} + b_2 x^{-n-2} \quad (24)$$

$$T_{Gx} = b_3 x^{-n-1} + O(\phi/R) \quad (25)$$

$$Y_{iGx} = b_{4i} x^{-n-1} + O(\phi/R) \quad (26)$$

The amplitude in Eq. (24) is for the radial velocity component. In addition, b_1 , b_2 , b_3 and b_{4i} are integration constants.

INTERFACE CONDITIONS

The solutions for the perturbation quantities contain nine unknown constants, which can be determined by imposing appropriate jump conditions at the liquid-gas interface. Written in dimensionless form, equations describing the interface conditions in terms of the perturbation amplitudes are listed below.

$$V_{Lx} = V_{Gx} \quad (27)$$

$$\frac{d}{dx}(x^2 V_{Lx}) = \frac{\rho_L}{\rho_G} \frac{d}{dx}(x^2 V_{Gx}) \quad (28)$$

$$\left[\frac{d^2}{dx^2} + 2 \frac{d}{dx} - 2 + n(n+1) \right] V_{Lx} = \gamma \left[\frac{d^2}{dx^2} + 2 \frac{d}{dx} - 2 + n(n+1) \right] V_{Lx} + M_T n(n+1) T_{Lx} + M_Y n(n+1) Y_{1Lx} \quad (29)$$

$$T_{Lx} = T_{Gx} \quad (30)$$

$$\frac{dT_{Gx}}{dx} = \frac{\lambda_L}{\lambda_G} \frac{dT_{Lx}}{dx} + \frac{\phi}{R} \frac{L_\infty}{c_p T_\infty} V_{Gx} \quad (31)$$

$$\frac{\phi}{R} \left(y_{1Lx} - \epsilon \frac{dy_{1Lx}}{dx} + y_{1-} V_{Lx} \right) = \frac{\phi}{R} (Y_{1Gx} + Y_{1G0} V_{Gx}) - \frac{dY_{1Gx}}{dx} \quad (32)$$

$$\frac{\phi}{R} \left[y_{2Lx} - \epsilon \frac{dy_{2Lx}}{dx} + (1 - y_{1-}) V_{Lx} \right] = \frac{\phi}{R} (Y_{2Gx} + Y_{2G0} V_{Gx}) - \frac{dY_{2Gx}}{dx} \quad (33)$$

$$y_{1Lx} = \Gamma_1 Y_{1Gx} - \frac{y_{1-} L_1 T_\infty}{R_1 T_{0+}^2} T_L \quad (34)$$

$$y_{2Lx} = \Gamma_2 Y_{2Gx} - \frac{(1 - y_{1-}) L_2 T_\infty}{R_2 T_{0+}^2} T_L \quad (35)$$

Equation (27) describes overall mass conservation across the interface, while Eq. (28) equates tangential velocities. Equation (29) describes the balance between the gas and liquid shear stresses and the surface tension gradients that occur from temperature and species gradients where $M_T = 8R\sigma_T/(T_\infty k\mu_L)$ and $M_Y = 8R\sigma_Y/(k\mu_L)$ are Marangoni numbers and $\gamma = (\mu_G/\mu_L)(\rho_L/\rho_G)$. Equations (30) equates gas and liquid temperatures, and Eq. (31) is an energy balance across the interface. Equations (32) and (33) are the interface species conservation equations, while Eqs. (34) and (35) are the linearized versions of the equilibrium condition (Eq. (7)) where $\Gamma_1 = \exp[L_1(1/T_{0+} - 1/T_{b1})/R_1]W_G/W_L$ and $\Gamma_2 = \exp[L_2(1/T_{0+} - 1/T_{b2})/R_2]W_G/W_L$ where L_1 and L_2 , R_1 and R_2 , and T_{b1} and T_{b2} are the enthalpies of vaporization, gas constants and boiling temperatures of species 1 and 2, respectively, and W_G and W_L are average molecular weights for the gas and liquid phases. In deriving Eqs. (34) and (35), perturbations in gas and liquid average molecular weights were neglected.

Substituting the gas and liquid solutions into the interface relations produces nine equations relating the nine unknown constants. The equations can be expressed in matrix form as $[A][B] = [0]$, where $[B] = [a_1, a_2, a_3, a_{41}, b_1, b_2, b_3, b_{41}, b_{42}]$ is a column vector and $[A]$ is shown on the last page of this paper. In this matrix, $\Omega_1 = (n - 1)/n + M_Y(y_{1-} - y_{10})/(2\beta_n)$, $\Omega_2 = (n + 2)/(n + 1) + M_Y(y_{1-} - y_{10})/(2\beta_n)$, $\Omega_3 = 1 + [16\epsilon L_0(2n + 3)]^{-1}$, $\Omega_4 = (\lambda_L/\lambda_G)(n + 2)/[16\epsilon L_0(2n + 3)]$, $\psi_1 = y_{1-} L_1 T_\infty/(R_1 T_{0+}^2)$ and $\psi_2 = (1 - y_{1-}) L_2 T_\infty/(R_2 T_{0+}^2)$.

STABILITY PREDICTIONS/DISCUSSION

The stability of two-component droplet gasification can be evaluated by setting the determinant of $[A]$ equal to zero and solving for the growth factor β_n . Because of the large number of terms in the matrix, this was accomplished using a symbolic calculation software package [22]. The variable β_n was found to depend upon a large number of terms (roughly 100), and for brevity the dispersion relation is not listed here.

Surprisingly, it was found that the dispersion relation did not depend upon either of the Marangoni numbers defined earlier (M_A and M_Y); they cancelled out of all calculations in the determinant. This was verified directly by calculating the minors associated with the terms Ω_1 , Ω_2 , $-M_A\Omega_3/2$ and $-M_Y/2$ in $[A]$; the minors for $-M_A\Omega_3/2$ and $-M_Y/2$ were identically zero, while the minors for Ω_1 and Ω_2 were exactly equal. In addition, the dispersion relation predicted that droplets should be absolutely stable, that is, $\beta_n < 0$ in general. In retrospect, this result might have been expected. To clarify why this is so, we should recall that the leading-order solution ($y_1 = y_{10} + (y_{1-} - y_{10})\exp(-z)$) was used for the unperturbed state, while Eq. (3) shows that the leading-order expression for the mass-

flux fraction of species i leaving the droplet is simply $\delta_i = y_{i0}$, i.e., δ_i is dependent only upon the initial mass fraction of species i and is independent of variations in other variables. As a result, in this diffusion-controlled limit for liquid species transport, small perturbations in liquid-phase velocity and species profiles do not influence the mass-flux fraction of a species leaving a droplet, and δ_i will be unchanged over the surface of a droplet as will the liquid-phase composition at the surface. By Eq. (5), the gas-phase solution will therefore be unchanged, and small perturbations in the liquid phase will not significantly alter gas-phase species or temperature fields so long as $\delta_i = y_{i0}$ is an adequate approximation. Under these conditions, all perturbations die out from viscous decay of velocity fields.

It should be noted, however, that these conclusions are not expected to be valid for all cases. For example, it is possible for the $O(\epsilon)$ correction in Eq. (3) to be important, and when this is so the analysis presented here may not be valid. This can happen, for example, when species i is initially present in very small amounts and is of low volatility relative to the other liquid species so that it must build to appreciable levels at the droplet surface before it can vaporize. Another possibility is when species i was not even present initially in the droplet, but was first absorbed and then gasified from the droplet surface (e.g., water condensation onto alcohol droplets). For analysis of the stability characteristics of droplets under these conditions, further analysis is necessary. These analyses are being performed, and will be published at a later date.

Even though these results were derived for slow gasification rates ($\phi/R \ll 1$), it is expected that they will be valid for rapid gasification ($\phi/R \geq 1$) as well as for droplet combustion. So long as the liquid species transport is diffusion controlled and $\delta_i = y_{i0}$ is a good approximation for the liquid phase, small perturbations in liquid-phase variables (e.g., velocities) will not significantly affect

δ_i . As a result, small perturbations in liquid-phase variables will not induce surface temperature or species gradients, and velocity perturbations will decay from viscous effects.

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1	1	0	0	-1	-1	0	0	0
$\frac{\rho_0}{\rho_L}(n+1)$	$\frac{\rho_0}{\rho_L}(n+3)$	0	0	n-2	n	0	0	0
Ω_1	Ω_2	$-\frac{M_T}{2}\Omega_3$	$-\frac{M_Y}{2}$	$-\gamma\frac{n-1}{n}$	$-\gamma\frac{n+2}{n+1}$	0	0	0
0	0	Ω_3	0	0	0	-1	0	0
0	0	Ω_4	0	$\frac{\phi}{R}\frac{L}{c_p T_{\infty}}$	$\frac{\phi}{R}\frac{L}{c_p T_{\infty}}$	n+1	0	0
$\frac{\phi}{R}y_{1-}$	$\frac{\phi}{R}y_{1-}$	0	$\frac{\phi}{R}(1+s_4)$	$-\frac{\phi}{R}Y_{100}$	$-\frac{\phi}{R}Y_{100}$	0	$-n-1-\frac{\phi}{R}$	0
$\frac{\phi}{R}(1-y_{1-})$	$\frac{\phi}{R}(1-y_{1-})$	0	$-\frac{\phi}{R}(1+s_4)$	$-\frac{\phi}{R}Y_{200}$	$-\frac{\phi}{R}Y_{200}$	0	0	$-n-1-\frac{\phi}{R}$
$\frac{y_{1-}-y_{10}}{\beta_n}$	$\frac{y_{1-}-y_{10}}{\beta_n}$	$-\psi_1\Omega_3$	-1	0	0	0	Γ_1	0
$\frac{y_{1-}-y_{10}}{\beta_n}$	$\frac{y_{1-}-y_{10}}{\beta_n}$	$\psi_2\Omega_3$	-1	0	0	0	0	Γ_2

The Matrix [A]

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