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## HYDRODYNAMIC STABILITY OF MULTICOMPONENT **DROPLET GASIFICATION IN REDUCED GRAVITY**

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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 **]\_yd.rodynamically**unstable **from** surfacetension 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, diffusiondominated 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** liquidphase transport **on** the vaporization and combustion behaviors of multlcomponent droplets have been the **subject** of numerous tnvestlgattons 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-vmmetric low Reynolds-number<br>evanoration and combustion of evaporation and combustion of<br>multicomponent droplets. These multicomponent droplets. The 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 llqutd components. Landis **and** Mills [1], for example, report results of numerical analyses<br>concerning spherically-symmetric concerning *spherically-symmetric* evaporation of miscrole binary dropped demonstrating the effects of volatility differences **and** *slow* liquid-phase **species** transport. Law et al. have presented numerical and theoretical **analyses** for *sphertcaUy-symmetrie* binary fuel evaporation with or without combustion [2-4]. **Experimental** work In thts **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 eombustlon 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 volation 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 loss 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 change<sub>3</sub> in droplet compositions **occur slowly** wtth 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 multtcomp0nont **droplets** for the case where liquid-phase **species** diffusion **ts** *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 eoncluslov of this work Is that **once** the boundary-layer composition profiles have been established in a droplet, the mass-flux • **fraction of ahy species** off the droplet surface is equal to the initial liquid-phase mass fraction of that species prior to vaporization. **The analyists of** Law and **Law [11]** was extended **by** Shaw **[15]:** who demonstrated that following decay of initial transients, the mass-flux **fraotlon** 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 droplel **(e.g.,** because **of** small perturbations m the environment **or within** the **droplet),** it **is** **possible** for surface-tension gradients to exist. If sufficiently intense, these surfacetension gradients can cause significant motions of the liquid-gas interface, influencing transport of *speetes* and energy tn the gas and liquid phases. It Is capillary effects such **as** these that this paper Is concerned with. **Thcoretleal** *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 sphericallysymmetrical 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 **w111**consider **only** the portion **of** the droplet history where the spherically*symmetrical species* **profiles** may oe considered quasi-steady. **Earlier** portions of the droplet history associated with transient liquid heating **and** !he initial buildup of the **spectes 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 Linan [16] have presented a hnear 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 thermocaptllary effects. N\_azmand et al. **1191** have recently presented numerical calculations for vaporizing droplets that are moving **relat:ve** to their environment. The calculations of Ntazmand etal. showed that capillary flows **dan** significantly affect droplet **interna|** velocity **and** temperature **fields** of vaporizing octane and methanol droplets, even when droplet initial **Reynolds** numbers

**<sup>\*</sup>** Here, **"boundary** layer" **refers to** the **species concentration** boundary **layer'**near the **dropletsurface.**

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(relative **to** the gas phase) arc as large **as** 50. **It** is noted that following the **initial** publications **of Nlazmand et al.** [19], Shih **and** Megafldis **[20]** presented computational **results on** the effects **of** thermocaplllary stresses and flows for nearly **the** same **conditionsas thoseconsidered**by Niazmand et **al.**None of **these** previous studieshave **considered** droplets with more than liquid species, which is **the** primary *subject* of **thts** paper.

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### **ESTIMATES** OF THE IMPORTANCE **OF** CAPILLARY EFFECTS

Estimates of **the importance** of capillary effectsmay be made by **consideringa 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 **velodfic.s relative to** the **ambient gas am** 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  $\mu_L$  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 dropl\_t 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(R2)/dt,** where **t is time) to** the liquid kinematic viscosity  $v_L$  is of order unity or less (i.e.,  $k/v_L \le 1$ ). Physically, if  $k/v_L \le 1$ , momentum changes at the droplet surface should have sufficient **time** to **tnfluenc\_ the** droplet interior before the droplet evaporates. A balance between liquid shear stress and

surface tension gradients gives  $u - \Delta \sigma / \mu_L$ 

 $r/t_c$  where  $\Delta\sigma$  is a characteristic difference in surface**tensionfrom** one **side** of **the** droplet to\_e othex,**and** to**isa characteristictlm¢** for .a **fluid** particle**to** be **convccted** halfway around the droplet periphery (surface-tension **gradientsare characterized**by **AcffR,and** gashase shear **is** neglected **assuming that the** " **quid-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  $t_b/t_c = R\Delta\sigma/(k\mu)$  << 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 **Ao** depends **upon gradients** of **temperatures** and species **concentrations alonjg the** droplet surface. **I.\_t us first considersurface**temperature gradientsalone. For this case we may write  $\Delta \sigma = |\partial \sigma / \partial T|$ **AT, where T is temperature,** o surface **tension, and AT is** the **temperature difference from one side of the droplet to the other. If this relation is used in the** expression **for trite,** we may solve for  $\Delta T \sim k\mu_L/(R \theta \sigma/\partial T)$  as **being required** for  $t_b/t_c \approx 1$ . For a 1 mm **diameter** hydrocarbon **or alcohol droplet** undergoing **combustion,** estimates suggest **that**  $\Delta T \sim 0.01$  K is required to make  $t_b/t_c$ **1. These** estimates **are based upon the characteristic** values  $k = 10^{-6}$  m<sup>2</sup>/s,  $\mu_L = 4 \times 10^{-6}$  $10^{-4}$  kg/(m s), and  $\partial \sigma / \partial T$  =  $10^{-4}$  N/(m °K), which **are appropriate for near.atmospherlc** pressure **conditions and droplet gasification** m **a** hot environment **or combustion.** Since the ratio  $t_b/t_c$  scales linearly with  $\Delta T$ , increases in  $\Delta T$  produce increases in  $t_b/t_c$ . It **thus** appearsthat **even** very small **tcmpcrature** differencesmay **induce** significant**convective** flows.

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

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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 ltqutd *surface,* and  $\Delta x_1$  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  $\Delta x_1$  ~  $k\mu_1/(R\bar{\omega}\sigma/\partial x_1)$  for  $t_b/t_c \sim 1$ .

Consider a methanol droplet of about 1 mm diameter,Itisknown **that**methanol (orother alcohol) droplets may absorb water from the gas phase. The characteristic values  $\partial \sigma/\partial x_1$  $= 0.03$  N/m,  $k = 10^{-6}$  m<sup>2</sup>/s, and  $\mu_L = 4 x$ 10 -4 k\_(m **s) are appropriate** for making order-of-m\_gnttude estimates **for** methanolwater 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 tn water **concentration along** the *surface* of **a** methanol droplet may **induce** *significant* **convective** flows. Since the ratio  $t_b/t_c$  scales linearly with  $\Delta x_1$ , larger values of  $\Delta x_1$  produce increases in  $t_b/t_c$ . Significant capillary flows will negate the possibility of **attaining** liquid-phase spherical symmetry by enhancing mixing tn .an **asymmetric and** unknown nianner. **Estimates** similar to those resented **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<sub>b</sub>/t<sub>c</sub>  $\sim 1$ . Such droplets (with significant volatility differentials) have been the subject of a number of reducedgravity experimental **efforts** which **are**

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

striving **for** spherical symmetry.

.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 studtes .will allow the **influences** of capillary phenomena to be delineated with greater preelsion, allowing for better **Interpretation** of reduced-gravity experimental results, and<br>also for increased understanding of droplet also for mcreased anderstanding or dropped simplicity, the analyses **are** restricted to evaporation in a hot environment; effects of **aS-ph.aso**combustion willbe **considered**at**a** later time. However, it is noted that the .\_sults presented here **should** be valid **for** combustion of dropletswhen the **flame** lies**a sil\_nificant** distance **away from** the droplet surface, for **example** when droplets **are** burned **in** environments wtth low oxidizer concentrations.

In the following, the basic unperturbed **solution** for **qu\_lsteady** 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 viscoustlow governing equations will be perturb **and** llnearlzed.These equations will be solved **and** *stability* **criteria** will be developed.

## THB UNPERTURBED SOLUTION

## Liquid-Phase Analysis

With the **assumption** of **a constant** liquid .spe\_?esdfffuslvityDr.**and alsoassuming that** hquid densitiesdo not vary with **time,**liquidphase species conservation may be taken to be described by the partial differential **equation and** boundary **condltions**(where Yl represents the mass fraction of species i)

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

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\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  $\delta_i$  is the mass-flux fraction of species

1 from the droplet surface i.e.,  $\delta_1$  = (dml/d0l(dm/d0, where m **is** the droplet mass, m<sub>1</sub> the mass of component i 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  $e=$ -D<sub>L</sub>/[R(dR/dt)], liquid phase species conservation is described by the tnttial value problem

$$
\frac{\partial y_i}{\partial \tau} = \mathcal{L} \frac{\partial}{\partial x} \left( x \frac{2 \partial y_i}{\partial x} \right) - x \frac{\partial y_i}{\partial x}
$$
(2)  

$$
\left( \mathcal{L} \frac{\partial y_i}{\partial x} - y_i \right)_{x=1} = -\delta_i, \left( \frac{\partial y_i}{\partial x} \right)_{x=0} = 0
$$
  

$$
y_i(x,0) = y_{i0}.
$$

The physical time t and the nondimensional

time  $\tau$  are related by  $t = \int_0^t d\tau/[\text{dln}(R_0/R)/\text{d}t]$ . 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  $Ut_b=1-e^{-2\tau}$ , where  $t_b = 4R_0^2/k$  is the droplet evaporation lifetime and  $\overline{R}_0$  is the initial droplet radius.

**The situation** of Interest here Is when liquid**phase** species transport ts **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 vglatile component builds, **and** a state will eventually be **attained** where vaporization **of the** less volatile **component** becomes significant. Since both species will \_en **vaponr\_ vigorously, and** liquid-phase diffusion is *slow,* further changes **in** droplet species profll\_ will **occur slowly,** Only that **part of** the **combustion** history where both components **are vaporizing vigorously** with slowly-changing speetes profiles will be analyzed here. *Analyses* of transient boundary layer butldup 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** speetes **profiles** over the droplet radius will occur over times **Ax** which may be of order (l/e). The time period  $\Delta \tau \sim O(1/\epsilon)$  may comprise the major **portion of** the remaining droplet lifetime **in** physical time, stnee **e<<l** Is expected, **as argued** below. Because eharaeteflstie changes in **y are** of order unity or less, the magnitude of the partial derivative  $\frac{\partial y}{\partial t}$  for these times may be estimated as  $(\partial y_i / \partial \tau) \leq O(\epsilon)$ . 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  $\delta_i$ . 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** eomponenL

**Analysis of** the boundary layer *structure* proceeds by considering **Eq.** (2). Because DL is generally small **compared to characteristic** magnitudes **of R(dR/d0, it** may be considered that e << 1. **The** "stretched" **coordinate** z=(lx)/e may then be inserted **into** Eq. (2). By using techniques of matched **asymptotic** expansions, it may be shown [15] that the mass-flux fraction  $\delta_i$  may be expressed as

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

while the asymptotic boundary-layer structure is provided by

$$
y_1 = y_{i0} +
$$
  
(y<sub>1</sub> - y<sub>i0</sub>)exp(-z)[1 + e(3z + z<sup>2</sup>/2) + ...]. (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

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

$$
v \frac{dY_1}{dr} = \frac{D_g}{r^2} \frac{d}{dr} \left( r^2 \frac{dY_1}{dr} \right)
$$

where  $\rho$  is density,  $c_p$  is specific heat,  $\lambda$  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 integrated subject to the conditions that  $T = T_{+}$  and  $Y_1 = Y_{1+}$  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

$$
\frac{T - T_{\infty}}{T_+ - T_{\infty}} = \frac{Y_1 - Y_{1\infty}}{Y_{1+} - Y_{1\infty}} = \frac{1 - \exp(-\phi/r)}{1 - \exp(-\phi/R)}
$$
(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  $B_T =$  $c_p(T_{\bullet\bullet}-T_+)/L$  and B<sub>i</sub> =  $(Y_{i+}-Y_{i\bullet})/(\delta_i-Y_{i+})$ are transfer numbers,  $\delta_i$  is the mass-flux fraction of species i in the gas phase, and  $L =$  $\Sigma \delta_l L_i$  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  $B = B_T = B_l$ . It may be shown that the surface mass fraction of a droplet component is calculated using  $Y_{1+} = (B\delta_i + Y_{i-})/(1 + B)$ , while  $\epsilon$  is related to  $B$  by

$$
\varepsilon = (D_{L}\rho_{L}c_{p})/[\lambda \ln(1+B)]. \tag{6}
$$

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  $X_i = (Y_i/W_i)/(ZY_i/W_i)$ . Gas and liquid mole fractions will be related using Raoult's law and the Clausius-Clapeyron relation with constant vaporization enthalpy, giving

$$
X_{i} = X_{i+} exp[L_{i}(1/T_{+} - 1/T_{b_{i}})/R_{i}] . \qquad (7)
$$

In Eq. (7),  $T_{b_1}$  is the boiling temperature of component i at the ambient pressure, and **Ri** is the **gas oonstan! for** species **i. The** phase equilibrium **relattons** 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<sub>10</sub>. Cite this **Is done,** values **for the surface** composition **in** the gas phase **are assumed** (subject to  $\Sigma \dot{Y}_{1+} = 1$ ). The surface **temperature** is then iterated until  $\Sigma \delta_1 = 1$  is attained **f6r the gas phase. The gas-liquid** couplingr01aflons **are then used to** calculate the surface **composition on** the **liquid** side of **the** gas-liquid **interface.** *The* parameter *e* is then **calculated** using **Eq. (6),** and **a** value **fo** r **y\_o based** upon **the values found.above is finally calculated from Eq.** (3). If thts **value is** not sufficiently close to the initially-specified value of y<sub>10</sub>, the above procedure is repeated with new values for the surface composition **in the gasphase. Calculations for various cases** are presented **by** Shaw **[15].** where **it is**  $s$ hown that the  $O(e)$  correction to  $\delta_i$  present in Eq.(3) **is dominant** when Ylo **is** sufficiently smaU.

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

## LINEARLZED STABILITY **EQUATIONS**

#### Liquid Phase

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

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

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

$$
\frac{d\Gamma}{dt} = \alpha_1 \Delta T
$$
 (Bnergy)  

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

It **is to** bc noted that V **represents** the gradient operator while A **represents the Laplacian** operator. In addition,  $\vec{v}$  is the velocity vector, p is pressure, **T** is temperature, and  $\alpha_i$  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** V\* **is the dimensionless gradient operator cxpressed** in **terms** of **x**.

We will **now** perturb the liquid-phase variables by assuming that  $\vec{v} = \vec{v}_0 + \vec{v}'$ ,  $p = p_o + p'$ ,  $T = T_o + T'$ ,  $y_i = y_{io} + y_i'$ where **the** subscript "o" **denotes** the solution **for the** spherically-symmetrical state and the superscript "prime" **denotes** the perturbation quantity. It is assumed that  $\vec{v}_0 = 0$ ,  $p_0 =$ constant, To **=** constant, and **that Yio** is predicted by Eq. (4). The assumption **of** constant To **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** equattons for the perturbation variables (note **that** "primes" **will** be **omitte.d** for **brevity).** In deriving these equations, the dimensionless variables  $\vec{V}_L = (8R/k)\vec{v}$ ,  $P_L = 8p'R^2/(k\mu_L)$ and  $T_L = T/T_{\bullet}$  are introduced, where  $T_{\bullet}$  is the amblent temperature and the subscript "L" denotes liquid. The perturbation equations are linearized by noglecting products of the perturbation quantities or their derivatives, and terms involving the sphericallysymmetrical base solutions cancel. In addition, the dimensionless time  $\tau$  defined earlier is introduced, though  $\tau$  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  $\tau_0$  for species diffusion across the thickness  $\varepsilon$ of the boundary layer is simply  $\tau_c = e$  (on the  $\tau$  timescale). For analysis of the equations, we therefore introduce the rescaled time  $\tau = \tau/e$  to produce the final form of the equations, which are shown below.

$$
\nabla^{\bullet} \cdot \vec{V}_{L} = 0 \quad \text{(Continuity)} \tag{8}
$$

$$
\Delta^{\bullet} \vec{v}_{L} \cdot \nabla^{\bullet} P_{L} = 0 \quad (\text{Momentum}) \tag{9}
$$

$$
\frac{1}{\varepsilon} \frac{\partial T_L}{\partial \tilde{\tau}} + x \frac{\partial T_L}{\partial x} = \varepsilon L \varepsilon \Delta^T T_L \quad \text{(Energy)} \quad (10)
$$

$$
\frac{1}{\varepsilon} \frac{\partial y_i}{\partial \tilde{x}} + x \frac{\partial y_i}{\partial x} + \tilde{v} \cdot \nabla^* y_{i0}
$$
  
=  $\varepsilon \Delta^* y_i$  (Specles) (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 liquidphase 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  $(\mathbb{Z}(x,\theta,\phi))$  can be expressed in terms of a normal mode decomposition based upon expansions in terms of spherical harmonics

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

where Z<sub>n</sub> 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  $\tau$ . A final goal of the analysis will be be to calculate the growth factor  $\beta_n$ , which will determine whether a perturbation will grow, decay, or remain stoady.

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

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

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

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

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

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

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. (I0)) may be developed by noting that we may reasonably expect that *e* << 1. To maintain a balance between dominant terms in dEq. (10), tt can be assumed that time erivatives are **at** most O(1/Le) In magnitude, which implies that the perturbed temperature field may be considered qunslsteady In the first **approximation.** We will thus drop the *transient* term from Eq. (10), **allowtng** us to **focus up0n** the **effects** of *species* perturbations on stability. Effects of temperature perturbations were studied by Higuera and Linan [16], who found that instabilities from temperature perturbations were **impotent** only when the base *solution* **exhibits** signtficant **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 TLa **is**

$$
\frac{d^{2}T_{Lx}}{dx^{2}} + \left(\frac{2}{x} - \frac{x}{E Lc}\right) \frac{dT_{Lx}}{dx}
$$
  
-\frac{n(n+1)}{x^{2}} T\_{Lx} = 0. \t(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

$$
T_{Lx} = a_3 \left[ x^n + \frac{1}{e Lc} \frac{1}{16(2n+3)} x^{n+2} \right] \qquad (16)
$$

**may** be used with little loss **in accuracy** (as **is a** constant of **integration). Applicability** of Eq. (16) is based upon the condition that eLe **is** not **small** relatlve 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. **(1** 1) **yields** the evolution equation for the **amplitude** Yita **of** the liquid-phase *species* perturbation

$$
\beta_0 y_{iLx} + \varepsilon \times \frac{dy_{iLx}}{dx} + (a_1 x^{n-1} + a_2 x^{n+1}) (y_{i-}y_{i0}) e^{(x-1)/\varepsilon}
$$
 (17)  
=  $\varepsilon^2 \left( \frac{d^2 y_{iLx}}{dx^2} + \frac{2}{x} \frac{dy_{iLx}}{dx} - \frac{n(n+1)}{x^2} y_{iLx} x \right).$ 

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  $\epsilon < 1$ , which is **generally** met for droplet gasification in hot environments. Examination of Eq.  $(19)$ tndleates 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 analysts of the boundary-layer is **provided** by the transformation  $x = 1 - \varepsilon z$ , where  $z \le O(1)$  in the boundary layer. Using this transformation **and** retaining only the largest terms provides the leading-order inner problem

$$
\frac{d^2y_{1Lx}}{dz^2} + \frac{dy_{1Lx}}{dz} - \beta_n y_{1Lx} =
$$
\n
$$
(a_1 + a_2) (y_1 - y_{10}) \exp(-z).
$$
\n(18)

The **solution** to Eq. (18) may be written as  $y_{\text{ILX}} = a_{41} \exp(s_4(1 - x)/\epsilon) + a_{51} \exp(s_5(1 - x)/\epsilon)$ x)/e) -  $(a_1 + a_2)(y_1 - y_{10})$  exp[(x - 1)/ $\varepsilon$ ]/ $\beta_n$ , where  $a_{41}$  and  $a_{51}$  are constants and  $s_4 = [-1 (1 + 4\beta_n)^{1/2}$ ]/2 and s<sub>5</sub> = [-1 + (1 + 413n)la]/2. The leading-order outer **solution** to Eq. (17) is simply  $y_{iLx} = 0$ . To match the inner and outer solutions we set  $a_{5} = 0$ , 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**

$$
y_{iLx} = a_{4i} exp[s_{4}(1 - x)/\epsilon] -
$$
  
(a<sub>1</sub> + a<sub>2</sub>) (y<sub>1</sub> - y<sub>i0</sub>) exp[(x - 1)/\epsilon]/\beta<sub>n</sub>. (19)

## Oas **Plisse**

**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 wfllalso **assume that the vaporization is slow such that**  $\phi/R =$ In  $(1 + B) \ll 1$  (but where  $\varepsilon = 8D<sub>1</sub>/k$  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  $\phi/R \ge 1$  and where density perturbations cannot be neglected, numerical *,integration* of **the** perturbation equations is needed; this analysis is now in **progress and** will be pre\_nted *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** ts **small** relative **to unit,/,. Under these** conditions, **we may neglect the convective acceleration terms in** the momeatum 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**sphericaIly-symmetrlcal**stateand the** superscript "prime" denotes the perturbation quantity. If we further let  $x = r/R$  and define the dimensionless perturbed varlablos  $\vec{V}_G = (\rho_O/\rho_I)$  (8R/k) $\vec{v}$ ,  $T_G = T/T_{\infty}$ ,  $Y_{1G} =$  $Y_i'$  and  $P_L = 8p'R^2(\rho_G/\rho_L)/(k\mu_L)$ , the **lineaflzed** *conservation* equations can be written **as**

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

$$
\Delta^{\bullet}\vec{V}_{Q} - \nabla^{\bullet}P_{Q} = 0 \quad (Momentum) \tag{21}
$$

$$
\Delta^{\bullet}T_{\mathbf{G}} = \mathbf{O}(\phi \mathbf{R}) \quad (\text{Energy}) \tag{22}
$$

$$
\Delta^{\dagger} Y_{1G} = O(\phi R) \quad \text{(Specics)} \tag{23}
$$

0

where **the** subscript "O" 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 **vamsh** as  $x \rightarrow \infty$  was applied and also where terms **of order** ¢/R **or smaller were** neglected.

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

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

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

**The** amplitude **in** Eq. (24) is for **the** radial **velocity** component. In addition,  $b_1$ ,  $b_2$ ,  $b_3$ and b<sub>41</sub> 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** am listed below.

$$
V_{Lx} = V_{Gx} \tag{27}
$$

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

$$
\left[\frac{d^{2}}{dx^{2}} + 2\frac{d}{dx} - 2 + n(n+1)\right]V_{Lx} =
$$
\n
$$
\gamma \left[\frac{d^{2}}{dx^{2}} + 2\frac{d}{dx} - 2 + n(n+1)\right]V_{Lx}
$$
\n(29)  
\n+ M<sub>T</sub> n(n+1)T<sub>Lx</sub> + M<sub>Y</sub> n(n+1)<sub>y</sub><sub>Lx</sub>

$$
T_{Lx} = T_{Gx} \tag{30}
$$

 $\bullet$ 

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

$$
\frac{\phi}{R} \Big( y_{1Lx} - \varepsilon \frac{dy_{1Lx}}{dx} + y_1. V_{Lx} \Big) =
$$
\n
$$
\frac{\phi}{R} \Big( Y_{1Gx} + Y_{1G0} V_{Gx} \Big) - \frac{dY_{1Gx}}{dx}
$$
\n(32)

$$
\frac{\phi}{R} \Big[ y_{2Lx} - e \frac{dy_{2Lx}}{dx} + (1 - y_{1-}) V_{Lx} \Big] =
$$
\n
$$
\frac{\phi}{R} (Y_{2Gx} + Y_{2G0} V_{Gx}) - \frac{dY_{2Gx}}{dx}
$$
\n(33)

$$
y_{1Lx} = \Gamma_1 Y_{1Gx} - \frac{y_1 L_1 T_{xx}}{R_1 T_{0+}^2} T_L
$$
 (34)

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

**Equation** (27) describes **overall** mass conservation **across** the interface, while **Eq.** (28) equates tangential velocities. Equation (29) descnbes the balance between the gas **and** llqutd **shear** stresses **and** the *surface* tension gradients that occur from temperature **and** *species* **gradients** where Mr =  $8R\sigma_T/(T_m k\mu_L)$  and  $M_Y = 8R\sigma_{Y_1}/(k\mu_L)$  are Marangoni numbers and  $\gamma = (\mu_0/\mu_L)(\rho_L/\rho_0)$ . 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  $\Gamma_2 =$  $exp[L_2(1/\Gamma_{0+} - 1/\Gamma_{b2})/R_2]$ W<sub>G</sub>/W<sub>L</sub> where L<sub>1</sub> and La,Rt and R2, and Tbt and Tb2 are**the** enthalpies of vaporization, gas constants and boiling **temperatures** of *species* 1 and **2,** respectively, and We and WL **are** average molecular weights for the gas and liqui 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  $[B] =$ [at, a2, as a4t, hi, b2, b3, b41, b4a] **is** a column vector and [A] is *shown* on the last page of this paper. In this matrix,  $\Omega_1 = (n - \frac{1}{2})$  $1/n + M_Y(y_1 - y_{10})/(2\beta_n), \Omega_2 = (n + 2)/(n + 1)$ 1) +  $M_Y(y_1 - y_{10})/(2\beta_n)$ ,  $\Omega_3 = 1 +$  $[16\varepsilon\text{Lo}(2n + 3)]^{-1}$ ,  $\Omega_4 = (\lambda_1/\lambda_0)(n + \ln 1)$  $2)/[16\varepsilon$ Le(2n + 3)] $], \psi_1 = y_1$ . L<sub>1</sub>T., $/(R_1T_{0+}^2)$ and  $\Psi_2 = (1 - y_1) L_2 T_{\bullet} / (R_2 T_{0+}^2)$ .

#### STABILITY PREDICTIONS/DISCUSSION

The **stability** of two-component droplet gaslflcatlon can bo evaluated by *setting* the determinant of [A] equal to zero and solving for the growth factor  $\beta_n$ . Because of the large number of terms in the matrix, this was accomplished using **a** symbolte calculation software package [22]. The variable  $\beta_n$  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  $(M_A \text{ and }$ My); they cancelled out of **all** calculations in the determinant. This was verified directly by calculating the minors **associated** with the terms  $\Omega_1$ ,  $\Omega_2$ ,  $-M_A\Omega_3/2$  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  $\Omega_1$  and  $\Omega_2$  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 n 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-

**",. °**

 $\mathcal{A}_\bullet$ 

flux **fraction of** species i leaving **the droplet** is  $\delta_i = y_{i0}$ , i.e.,  $\delta_i$  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 liqutd-phase velocity** and species **profiles do not** influence the **mass-flux fraction of** a species **leaving a droplet,** and **51** 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 **gasphase species or** temperature **fields** so **long as**  $\delta_i$   $\approx$   $y_{i0}$  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(e)** correction **in Eq. (3)** to be **important, and** when **this is so. the analysis presented** here may not **be vahd. This** can happen, **for example,** when species **t is. initially present in vary small amounts** and **ts of low volatility** relative **to the other liquid speeles** so **that** It **must** build **to** appreciable **levels** at the **droplet surface before tt** can **vaporize. Another possibility is when** species **i was not even present initially in the droplet, but** was **first** absorbed **and** then gasifiedfrom **the droplet** . surface (e.g., water *condensation* **onto** alcohol **droplets). For** analysts **of the** stability charactoristtcs **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  $(\phi/R \ge 1)$  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 liqu!d-phaso variables (e.g., velocities)** will **not significantly affect** .° \_51.As. **a result,** *small* perturbations in liquidphase.variables will not **induce** surface temperature **or** species gradients, **and velocity perturbations will decay from viscous** effects.

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