

UNSTEADY VAPORIZATION OF LIQUID DROPLET*

BY

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Abstract. The development of the temperature and concentration profiles in the vicinity of a vaporizing liquid droplet exposed to a hot gas, when Soret and Dufour numbers are finite, is examined under linearized theory consisting of two coupled diffusion equations.

Time-dependent solutions for the temperature and concentration, each consisting of the principal and auxiliary modes, are obtained for arbitrary Soret, Dufour and Lewis numbers and four different types of boundary conditions.

The effective diffusivity of the principal modes of the temperature solution is found to increase, and that of the concentration solution to decrease, when Soret and Dufour numbers increase. For the case of fixed surface temperature and surface concentration, the numerical analysis indicates that the fuel vapor diffusion is adversely affected by the Soret effect, whereas heat conduction from the hot gas to the liquid droplet increases as the Dufour number increases.

The present method can be applied to a system of linearized equations of heat and mass transfer including more than two independent transport processes. The method is also applicable to the problems of multidiffusion.

I. Introduction. The aero-thermochemical aspects of heterogeneous combustion of liquid fuel in rocket and air breathing propulsion systems has been of recent theoretical and experimental interest. The existing theoretical research on droplet burning, summarized in the book by Williams [1], has been directed toward the prediction of the flame shapes, temperatures and concentration profiles, and also the burning rate of the droplet.

More recently, attention has been focused on the problems in which some of the assumptions made in the previous works have been relaxed. Examples are the inclusion of the effect of the relative motion of the droplet with respect to the surrounding gas by Fendell [2] and Fendell et al. [3], and the effect of the droplet deformation by Chiu [4]. In all of the existing work, however, the Soret effect (thermal diffusion), and Dufour effect (diffusion thermo-effects) are neglected because the Soret and Dufour numbers are small. Thermal diffusion is a transport of matter caused by a local temperature gradient (see for example Chapman and Cowling [5]). The diffusion thermo-effects, a process reciprocal to thermal diffusion, is a flow of thermal energy caused by a concentration gradient.

While these two coupled transport processes are indeed negligible in steady-state combustion, their effects may be considerable in unsteady or transient state combustion.

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For example, a sudden exposure of a cold evaporable liquid droplet to the hot gas will establish an initial steep temperature gradient which will enhance thermal diffusion. This situation occurs when cold droplets are sprayed into the hot combustion chambers or pass through a shock wave, as in Lu and Chiu [6] and Panton and Oppenheim [7].

Details of the heat and mass transfer process in this transient period are of practical interest in determining the propellant characteristics prevailing in the pre-ignition phase. The unsteady vaporization of the droplet is treated by Williams [8], yet the analysis of the transient droplet vaporization, including the Soret and Dufour effects, has not yet been reported. It was pointed out by von Kármán [9] that the thermal diffusion and diffusion thermo-effect are designated as complicated phenomena whose effects cannot be determined by elementary analysis. Detailed knowledge of the Soret and Dufour effects and their bearing on nonsteady combustion is still largely unavailable. It is the purpose of this paper to present an unique method of obtaining the general solution of the equations governing heat and mass transfer including the Dufour and Soret effects. By doing so, we hope to deduce the salient features of the combined effects of thermal diffusion and diffusion thermo-effect in a thermo-chemical system. In particular, an attempt is made to develop the temperature and the concentration profiles in the vicinity of the evaporable liquid droplet exposed to a hot gas with Soret and Dufour effects duly taken into account.

In Sec. 2 the governing equations for the linearized theory of heat and mass transfer and the boundary conditions, presented in the book by Luikov and Mikhailov [10], are recapitulated. Luikov and Mikhailov solved these equations for various boundary conditions for some simple geometries including one- and two-dimensional Cartesian coordinates, and cylindrical and spherical coordinates with preserved symmetry. Unfortunately, their method of solution is extremely complicated and is not applicable to nonsymmetric problems.

We demonstrate in Sec. 3 that the difficulties encountered in Luikov and Mikhailov's analysis can be removed by decoupling the systems of coupled parabolic equations. The decoupling is achieved by applying the method of irreducible operators developed recently by Chiu [11]. It is then shown that the solutions of the initial vaporization of a liquid droplet consist of two fundamental modes. Each mode is the solution of the uncoupled heat conduction equation, and each of these two uncoupled equations possesses different effective thermometric conductivity, which depends on the Soret and Dufour numbers, among others.

The solutions for the temperature and the concentration of vaporizing species, corresponding to four different kinds of boundary conditions, are presented in Sec. 4. The expression for the rate of the vaporization of the droplet is given in Sec. 5. The numerical results of the initial development of the temperature and concentration and the rate of vaporization corresponding to the case of the fixed surface temperature and concentration are presented.

II. Equations and boundary conditions of heat and mass transfer. The linearized equations of the heat and mass transfer developed by Luikov and Mikhailov [10] for a binary gas mixture consisting of the vaporizing species and the surrounding hot gas that is at rest with respect to the droplet are given in the following nondimensional form:

$$\frac{\partial \theta}{\partial t} = \left(1 + \frac{Du}{Le} So\right) \nabla^2 \theta + \frac{Du}{Le} \nabla^2 n_1 + Q_1, \quad (1a)$$

$$\frac{\partial n_1}{\partial t} = \frac{1}{\text{Le}} \nabla^2 n_1 + \frac{\text{So}}{\text{Le}} \nabla^2 \theta + Q_2, \quad (1b)$$

where Q_1 and Q_2 are distributed heat and mass sources and Du , Le , and So are the Dufour, Lewis, and Soret numbers, respectively. θ and n_1 are the nondimensional temperature and the molar concentration of the vaporizing species. Note that in the absence of the Dufour and Soret effects, the system of equations degenerates into two uncoupled equations of parabolic type. For a finite Du and So , the equations are coupled. The effect of coupling becomes important as the Dufour and the Soret numbers increase. Following the linearized theory of Luikov and Mikhailov, we assume that the Dufour, Soret and Lewis numbers are constant. This assumption is not expected to change the qualitative picture of the intercoupling provided that the range of the concentration variation is not too large.

Further assumptions made in the analysis are that the convective term is small compared with the temporal change in the local temperature and concentration. The assumption is justified for a vanishing Péclet number and a slow rate of droplet vaporization. The change in the droplet size is assumed to be small within the time interval considered. This assumption is consistent with that of slow vaporization. The coupled system of equations (1a) and (1b) are to be solved under prescribed initial and boundary conditions.

Three different conditions proposed by Luikov and Mikhailov [10] and another kind of boundary condition which is used in the steady-state burning of the droplet will be discussed in the remainder of this section.

We consider the following general boundary conditions given by Luikov and Mikhailov [10]:

$$\frac{\partial \theta}{\partial r} + B_{11} q_a + B_{12} q_m = 0, \quad (2a)$$

$$\frac{\partial n_1}{\partial r} + \text{So} \frac{\partial \theta}{\partial r} + B_{22} q_m = 0, \quad (2b)$$

where $B_{i,j}$ are constants which depend on the properties of the gas mixture and the liquid droplet at the interface. Eq. (2a) represents the conservation of the energy on the droplet surface. The first term in (2a) is the heat conduction from the gas to the droplet and the second term represents transfer of heat other than that represented by the first term (for example, the heat conduction from the droplet surface toward the interior of the droplet when the temperature is nonuniform). The third term expresses the heat required for the vaporization of the liquid fuel at the surface.

Eq. (2b) is the conservation of mass of the vaporizing species. The first term represents the diffusion by the concentration gradient, the second term expresses the thermal diffusion and the third term is the mass flux of the vaporizing species.

We shall first discuss the third boundary condition. This is to specify q_a and q_m , appearing in Eqs. (2a) and (2b), as functions of the surface temperature and concentration [10] as follows:

$$q_a(t) = \gamma_a[\theta(t) - \theta_p(t)], \quad (3a)$$

$$q_m(t) = \gamma_m[n_1(t) - n_p(t)], \quad (3b)$$

where θ_p is the surface temperature of the liquid droplet and n_p is the equilibrium con-

centration corresponding to the temperature of the droplet surface. γ_m and γ_q are the coefficients of the mass and heat exchange at the interface.

The boundary condition of the second kind assigns the time dependence of q_a and q_m . Hence this boundary condition amounts to prescribing the temperature and the concentration gradients as functions of time on the interface.

The boundary condition of the first kind prescribes the temperature and concentration as functions of time on the interface.

By the application of the method of the irreducible operator it can be shown that problems with boundary conditions of either the first or second kind can be reduced to initial and boundary value problems similar to those of heat conduction. The third and fourth boundary value problems can also be reduced to a heat conduction problem in which explicit initial conditions are obtained from coupled integral equations.

A slight modification of the boundary condition of the third kind leads to the following condition customarily used in the literature [2]:

$$\frac{\partial \theta}{\partial r} = \frac{Sc \operatorname{Re} L U_r}{Le D}, \quad (4a)$$

$$\frac{\partial n_1}{\partial r} + So \frac{\partial \theta}{\partial r} = -\frac{Sc \operatorname{Re} U_r}{D} (1 - n_1), \quad (4b)$$

where Sc is the Schmidt number, Re is the Reynolds number and L is the latent heat of the vaporization. Note that the molecular weights of both species are assumed to be approximately the same and that U_r is a known function of time.

For the sake of convenience, Eqs. (4a) and (4b) may be called the boundary conditions of fourth kind.

III. Fundamental modes of the linearized equation of heat and mass transfer. In the analysis of the system of equations (1a) and (1b), Luikov and Mikhailov [10] adopted the method of elimination by which one of the two dependent variables is eliminated. This results in a differential equation involving fourth-order derivatives of spatial coordinates and second-order derivatives with respect to time. In cases where pressure diffusion should be retained, the highest order of the spatial derivatives become six and that of the time derivative becomes three, and so on.

The treatment of these higher-order differential equations is extremely complicated and laborious. In the present paper we adopt the method of irreducible operators [11]. The basic idea underlying this method is to decouple two equations through the linear transformation of the dependent variables in conjunction with the application of a complementary operator. The decoupled equations are of second order.

The system of equations (1a) and (1b) are the simplest type of coupled equations which admits a simple decoupling described in the following. To begin with we write (1a) and (1b) as

$$I \frac{\partial \psi}{\partial t} = K \nabla^2 \psi + Q, \quad (5)$$

where I is a 2-by-2 unit matrix, ψ and Q are column vectors given by

$$\psi = \begin{bmatrix} \theta \\ n_1 \end{bmatrix}, \quad Q = \begin{bmatrix} Q_1 \\ Q_2 \end{bmatrix}, \quad (6a)$$

and K is a 2-by-2 matrix given by

$$K = \begin{bmatrix} 1 + \frac{Du}{Le} \frac{So}{Le} & \frac{Du}{Le} \\ \frac{So}{Le} & \frac{1}{Le} \end{bmatrix}. \quad (6b)$$

The general boundary conditions (2a) and (2b) are also expressed in the following matrix form:

$$A \frac{\partial \psi}{\partial r} + Bq = 0 \quad (7)$$

where

$$A = \begin{bmatrix} 1 & 0 \\ So & 1 \end{bmatrix}, \quad B = \begin{bmatrix} B_{11} & B_{12} \\ 0 & B_{22} \end{bmatrix}, \quad q = \begin{bmatrix} q_a \\ q_m \end{bmatrix}. \quad (8)$$

The decoupling of the matrix equation (5) is obtained by applying the simple transformation of the dependent variables

$$\psi = \alpha \chi, \quad (9)$$

where α is a 2-by-2 nonsingular matrix with the elements α_{ij} , $i, j = 1, 2$, yet undetermined.

The new dependent variable χ , which is a two-component column vector, is related to ψ by the inverse transformation

$$\chi = \alpha^{-1} \psi, \quad (10)$$

where α^{-1} is the inverse matrix of α .

Substituting (9) into (5), and applying the complementary operator [11], which is simply the inverse of matrix α for the present system of equations, yields

$$I \frac{\partial \chi}{\partial t} = \alpha^{-1} K \alpha \nabla^2 \chi + \alpha^{-1} Q. \quad (11)$$

The decoupling of Eq. (11) is achieved by choosing the elements α_{ij} so that the matrix $\alpha^{-1} K \alpha$ is diagonal.

The proper choice of α_{ij} is given by

$$\alpha_{11} = \alpha_{22} = 1, \quad (12a)$$

$$\alpha_{12} = \frac{1}{2 Le So} \left\{ 1 - \frac{1}{Le} + \frac{Du}{Le} So - \left[\left(1 - \frac{1}{Le} + \frac{Du}{Le} So \right)^2 + \frac{4 Du So}{Le^2} \right]^{1/2} \right\}, \quad (12b)$$

$$\alpha_{21} = -\frac{1}{2 Le Du} \left\{ 1 - \frac{1}{Le} + \frac{Du}{Le} So - \left[\left(1 - \frac{1}{Le} + \frac{Du}{Le} So \right)^2 + \frac{4 Du So}{Le^2} \right]^{1/2} \right\}. \quad (12c)$$

When the absolute values of the Dufour and Soret numbers are smaller than unity, and if the Lewis number is different from unity, α_{12} and α_{21} are given by

$$\alpha_{12} \simeq -\frac{Du}{Le - 1}, \quad (13a)$$

$$\alpha_{21} \approx \frac{So}{Le - 1}. \quad (13b)$$

In the limiting case of small Du and So , α_{12} and α_{21} approach zero as is expected. The special case of $Le = 1$ is obtained by replacing Le by unity in Eqs. (12b) and (12c).

By choosing $\alpha_{i,i}$ to be those given by (12a), (12b), (12c), Eq. (11) becomes

$$\frac{\partial \chi_1}{\partial t} = L_{11} \nabla^2 \chi_1 + R_1, \quad (14a)$$

$$\frac{\partial \chi_2}{\partial t} = L_{22} \nabla^2 \chi_2 + R_2, \quad (14b)$$

where

$$L_{11} = \frac{1}{2} \left\{ 1 + \frac{1}{Le} + \frac{Du So}{Le} + \left[\left(1 - \frac{1}{Le} + \frac{Du So}{Le} \right)^2 + \frac{4 Du So}{Le^2} \right]^{1/2} \right\}, \quad (15a)$$

$$L_{22} = \frac{1}{2} \left\{ 1 + \frac{1}{Le} + \frac{Du So}{Le} - \left[\left(1 - \frac{1}{Le} + \frac{Du So}{Le} \right)^2 + \frac{4 Du So}{Le^2} \right]^{1/2} \right\}, \quad (15b)$$

$$R_1 = (\det \alpha)^{-1} (Q_1 - \alpha_{12} Q_2), \quad (16a)$$

$$R_2 = (\det \alpha)^{-1} (-\alpha_{21} Q_1 + Q_2), \quad (16b)$$

and

$$(\det \alpha)^{-1} = \frac{1}{2} \left\{ 1 + \frac{1 - \frac{1}{Le} + \frac{Du So}{Le}}{\left[\left(1 - \frac{1}{Le} + \frac{Du So}{Le} \right)^2 + \frac{4 Du So}{Le^2} \right]^{1/2}} \right\}. \quad (16c)$$

Eqs. (14a) and (14b) are uncoupled equations of parabolic type. The operators $(\partial/\partial t) - L_{11} \nabla^2$ and $(\partial/\partial t) - L_{22} \nabla^2$ are the irreducible operators. The solutions χ_1 and χ_2 of the irreducible operators are called the fundamental modes.

The temperature and the concentration solutions are constructed from the fundamental modes as $\theta = \chi_1 + \alpha_{12} \chi_2$, $n_1 = \alpha_{21} \chi_1 + \chi_2$. In the limiting case of $Du = So = 0$, both α_{12} and α_{21} vanish identically; hence $\theta = \chi_1$ and $n_1 = \chi_2$.

This limiting consideration suggests that the temperature solution consists of the principal mode χ_1 and the auxiliary mode χ_2 , whereas the concentration solution consists of the principal mode χ_2 and the auxiliary mode χ_1 .

The coefficients of diffusion L_{11} and L_{22} appearing in two irreducible operators are positive for any Du and So numbers which may be positive or negative.

The numerical values of L_{11} and L_{22} are plotted for some Dufour, Lewis and Soret numbers in Fig. 1.

IV. Temperature and concentration solutions. In this section we present the method of construction of the temperature and concentration solutions from the fundamental modes for four different boundary conditions.

Throughout the analysis the inhomogeneous terms Q_1 and Q_2 will be taken to be zero, and the initial distributions of the temperature and the concentration of the vaporizing species are assumed to be given by:

$$\theta(r, 0) = f_1(r), \quad (17a)$$

$$n_1(r, 0) = f_2(r). \quad (17b)$$

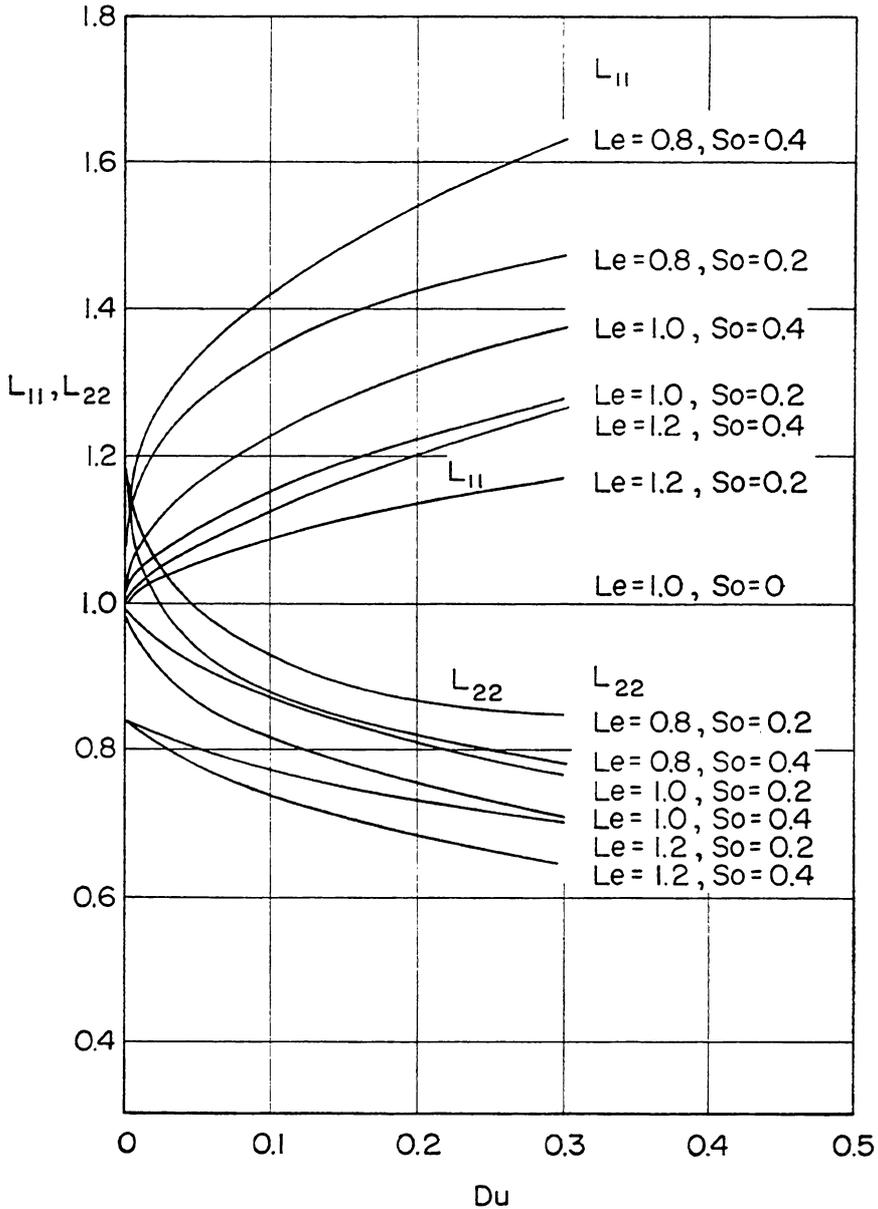


FIG. 1. Variation of the diffusivities of fundamental modes.

Case 1. Boundary value problem of the first kind. The first boundary conditions for θ and n_1 are

$$\theta(1, t) = g_1(t), \tag{18a}$$

$$n_1(1, t) = g_2(t). \tag{18b}$$

From the transformation (10) we obtain the corresponding initial and boundary con-

ditions for the fundamental modes.

$$\chi_1(r, 0) = (\det \alpha)^{-1}[f_1(r) - \alpha_{12}f_2(r)] = \mathfrak{F}_1(r), \quad (19a)$$

$$\chi_2(r, 0) = (\det \alpha)^{-1}[-\alpha_{21}f_1(r) + f_2(r)] = \mathfrak{F}_2(r), \quad (19b)$$

$$\chi_1(1, t) = (\det \alpha)^{-1}[g_1(t) - \alpha_{12}g_2(t)] = \mathfrak{G}_1(t), \quad (20a)$$

$$\chi_2(1, t) = (\det \alpha)^{-1}[-\alpha_{21}g_1(t) + g_2(t)] = \mathfrak{G}_2(t). \quad (20b)$$

The solutions $\chi_1(r, t)$ and $\chi_2(r, t)$ satisfying the above initial and boundary conditions are well known (see for example Carslaw and Jaeger [12]). From the $\chi_1(r, t)$ and $\chi_2(r, t)$, which will not be reproduced hereafter, we obtain the temperature and concentration as follows:

$$\begin{aligned} \theta(r, t) = & \frac{1}{2r\sqrt{(\pi L_{11}t)}} \int_1^\infty r' \mathfrak{F}_1(r') \{e^{-(r-r')^2/4L_{11}t} - e^{-(r+r'-2)^2/4L_{11}t}\} dr' \\ & + \frac{2}{r\sqrt{\pi}} \int_{(r-1)/2\sqrt{(L_{11}t)}}^\infty \mathfrak{G}_1 \left[t - \frac{(r-1)^2}{4L_{11}\mu^2} \right] e^{-\mu^2} d\mu \\ & + \alpha_{12} \left\{ \frac{1}{2r\sqrt{(\pi L_{22}t)}} \int_1^\infty r' \mathfrak{F}_2(r') \{e^{-(r-r')^2/4L_{22}t} - e^{-(r+r'-2)^2/4L_{22}t}\} dr' \right. \\ & \left. + \frac{2}{r\sqrt{\pi}} \int_{(r-1)/2\sqrt{(L_{22}t)}}^\infty \mathfrak{G}_2 \left[t - \frac{(r-1)^2}{4L_{22}\mu^2} \right] e^{-\mu^2} d\mu \right\}; \end{aligned} \quad (21a)$$

$$\begin{aligned} n_1(r, t) = & \frac{1}{2r\sqrt{(\pi L_{22}t)}} \int_1^\infty r' \mathfrak{F}_2(r') \{e^{-(r-r')^2/4L_{22}t} - e^{-(r+r'-2)^2/4L_{22}t}\} dr' \\ & + \frac{2}{r\sqrt{\pi}} \int_{(r-1)/2\sqrt{(L_{22}t)}}^\infty \mathfrak{G}_2 \left[t - \frac{(r-1)^2}{4L_{22}\mu^2} \right] e^{-\mu^2} d\mu \\ & + \alpha_{21} \left\{ \frac{1}{2r\sqrt{(\pi L_{11}t)}} \int_1^\infty r' \mathfrak{F}_1(r') \{e^{-(r-r')^2/4L_{11}t} - e^{-(r+r'-2)^2/4L_{11}t}\} dr' \right. \\ & \left. + \frac{2}{r\sqrt{\pi}} \int_{(r-1)/2\sqrt{(L_{11}t)}}^\infty \mathfrak{G}_1 \left[t - \frac{(r-1)^2}{4L_{11}\mu^2} \right] e^{-\mu^2} d\mu \right\}. \end{aligned} \quad (21b)$$

In particular, for uniform initial temperature distribution $f_1(r) = \theta_A$ and uniform initial concentration $f_2(r) = n_{1A}$, with constant surface temperature $g_1(t) = \theta_1$ and constant surface concentration $g_2(t) = n_{1e}$, Eqs. (21a) and (21b) reduce to

$$\theta(r, t) = \theta_A - \frac{1}{r} \left\{ V_1 \operatorname{erfc} \frac{r-1}{2\sqrt{(L_{11}t)}} + \alpha_{12} V_2 \operatorname{erfc} \frac{r-1}{2\sqrt{(L_{22}t)}} \right\}, \quad (22a)$$

$$n_1(r, t) = n_{1A} - \frac{1}{r} \left\{ \alpha_{21} V_1 \operatorname{erfc} \frac{r-1}{2\sqrt{(L_{11}t)}} + V_2 \operatorname{erfc} \frac{r-1}{2\sqrt{(L_{22}t)}} \right\}, \quad (22b)$$

where

$$V_1 = \mathfrak{F}_1 - \mathfrak{G}_1, \quad (23a)$$

$$V_2 = \mathfrak{F}_2 - \mathfrak{G}_2, \quad (23b)$$

The time-dependent temperature and concentration profiles for $Le = 1.0$, $So = 0.2$, and $Du = 0.1$ are presented in Fig. 2. The initial temperature distribution in the atmosphere is unity and the surface temperature is held at 0.2. The initial fuel vapor concentration is zero throughout the atmosphere and is 0.2 on the droplet surface.

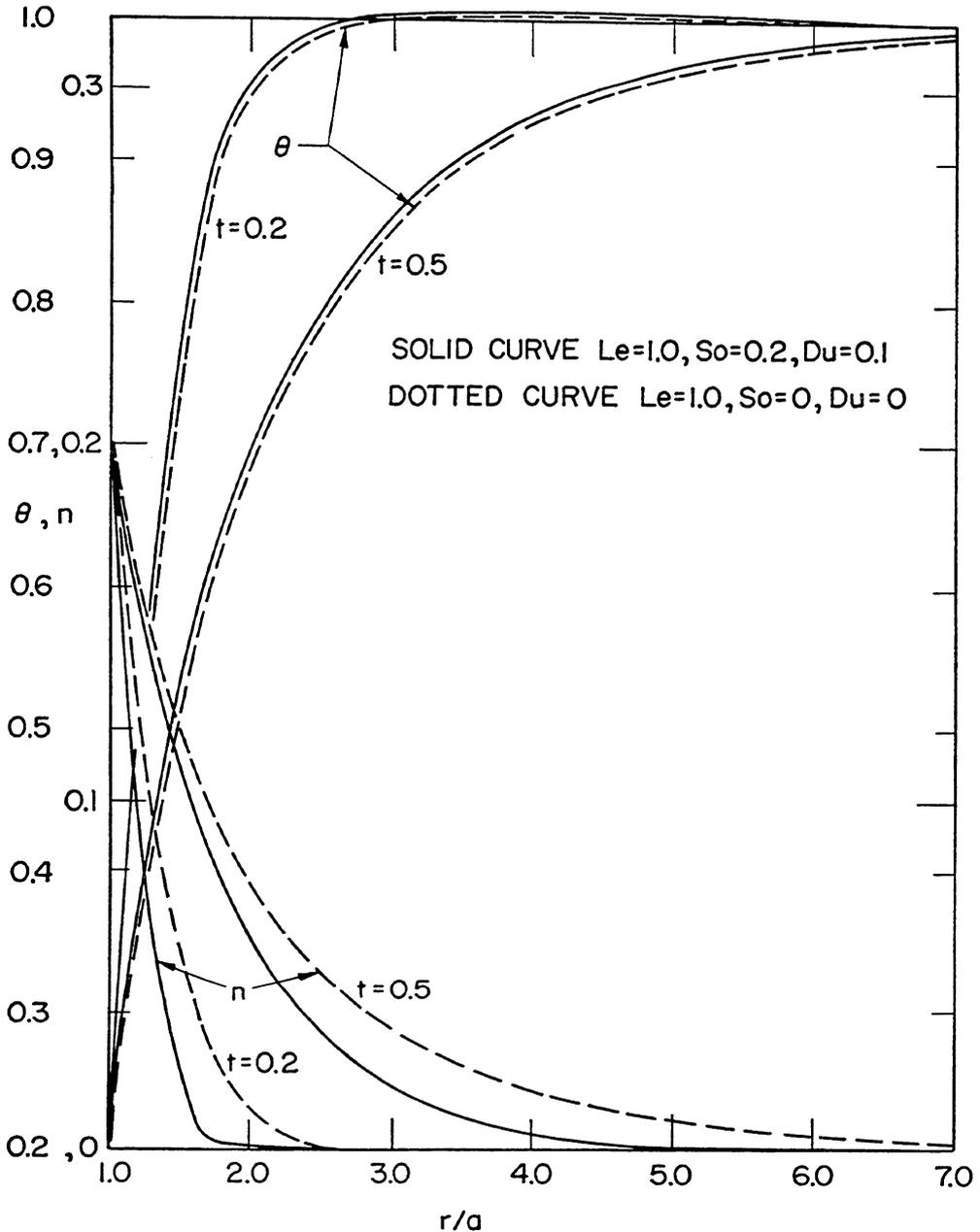


FIG. 2. Distributions of temperature and concentrations.

It is noted that at $t = 0.2$, $r = 1.5$, the fuel vapor concentration with $So = 0.2$, $Du = 0.1$ is approximately one-third of that obtained for the case of vanishing Dufour and Soret numbers. The reduction in the concentration is attributed to the Soret effect which tends to diffuse the fuel vapor from the high temperature zone back to the colder zone, i.e. the region occupied by the droplet.

The decrease in the temperature difference between the droplet and the atmosphere reduces the heat conduction. This results in a decrease in the effect of thermal diffusion, as is shown in Fig. 3.

Case 2. Boundary value problem of the second kind. In the second kind of boundary condition, q_m and q_a are given as functions of time. The boundary condition on the droplet surface is

$$(\partial\theta/\partial r)_{r=1} = -B_{11}q_a(t) - B_{12}q_m(t), \tag{24a}$$

$$(\partial n_1/\partial r)_{r=1} = -B_{11}S_0q_a(t) + (B_{12}S_0 - B_{22})q_m(t), \tag{24b}$$

We first define variables $\Omega_i = r \chi_i$ for $i = 1, 2$. The boundary conditions for $\chi_{1,2}$ become

$$\left(\frac{\partial\chi_1}{\partial r}\right)_{r=1} = \left(\frac{\partial\Omega_1}{\partial r} - \Omega_1\right)_{r=1} = (\det \alpha)^{-1} \left(\frac{\partial\theta}{\partial r} - \alpha_{12} \frac{\partial n_1}{\partial r}\right)_{r=1}, \tag{25a}$$

$$\left(\frac{\partial\chi_2}{\partial r}\right)_{r=1} = \left(\frac{\partial\Omega_2}{\partial r} - \Omega_2\right)_{r=1} = (\det \alpha)^{-1} \left(-\alpha_{21} \frac{\partial\theta}{\partial r} + \frac{\partial n_1}{\partial r}\right)_{r=1}. \tag{25b}$$

Now define new variables u_i for $i = 1, 2$

$$u_i(r, t) = (\partial\Omega_i(r, t)/\partial r) - \Omega_i(r, t). \tag{26}$$

It can be shown that the u_i 's satisfy the following equations:

$$\partial u_1/\partial t = L_{11} \partial^2 u_1/\partial r^2, \tag{27a}$$

$$\partial u_2/\partial t = L_{22} \partial^2 u_2/\partial r^2. \tag{27b}$$

The initial and boundary conditions for the u_i 's are given by

$$u_1(r, 0) = (\det \alpha)^{-1} \left\{ r \left(\frac{\partial\theta}{\partial r} - \alpha_{12} \frac{\partial n_1}{\partial r} \right) + (1-r)(\theta - \alpha_{12} n_1) \right\}_{t=0} = r\tilde{\mathcal{F}}_1(r), \tag{28a}$$

$$u_2(r, 0) = (\det \alpha)^{-1} \left\{ r \left(-\alpha_{21} \frac{\partial\theta}{\partial r} + \frac{\partial n_1}{\partial r} \right) + (1-r)(-\alpha_{21}\theta + n_1) \right\}_{t=0} = r\tilde{\mathcal{F}}_2(r), \tag{28b}$$

$$u_1(1, t) = -(\det \alpha)^{-1} \{ B_{11}(1 + \alpha_{21} S_0)q_a(t) + [B_{12}(1 + \alpha_{12} S_0) - \alpha_{12}B_{22}]q_m(t) \} = \tilde{\mathcal{G}}_1(t) \tag{29a}$$

$$u_2(1, t) = (\det \alpha)^{-1} \{ B_{11}(\alpha_{21} + S_0)q_a(t) + [B_{12}(\alpha_{21} + S_0) - B_{22}]q_m(t) \} \equiv \tilde{\mathcal{G}}_2(t). \tag{29b}$$

The solutions for $u_1(r, t)$ and $u_2(r, t)$ may be found, for example, in Carslaw and Jaeger [12].

These $u_i(r, t)$ are then substituted into Eq. (26) to find $\Omega_i(r, t)$. The temperature and concentration are finally calculated to be

$$\begin{aligned} \theta(r, t) = & -\frac{1}{2r\sqrt{(\pi L_{11}t)}} \int_0^\infty e^{-\eta} \int_1^\infty r'\tilde{\mathcal{F}}_1(r') \{ e^{-(r+\eta-r')^2/4L_{11}t} - e^{-(r+\eta+r'-2)^2/4L_{11}t} \} dr' d\eta \\ & - \frac{2}{r\sqrt{\pi}} \int_0^\infty e^{-\eta} \int_{(r+\eta-1)/2\sqrt{(L_{11}t)}}^\infty \tilde{\mathcal{G}}_1 \left[t - \frac{r+\eta-1}{4L_{11}\mu^2} \right] e^{-\mu^2} d\mu d\eta \\ & - \alpha_{12} \left\{ \frac{1}{2r\sqrt{(\pi L_{22}t)}} \int_0^\infty e^{-\eta} \int_1^\infty r'\tilde{\mathcal{F}}_2(r') \{ e^{-(r+\eta-r')^2/4L_{22}t} - e^{-(r+\eta+r'-2)^2/4L_{22}t} \} dr' d\eta \right. \\ & \left. + \frac{2}{r\sqrt{\pi}} \int_0^\infty e^{-\eta} \int_{(r+\eta-1)/2\sqrt{(L_{22}t)}}^\infty \tilde{\mathcal{G}}_2 \left[t - \frac{r+\eta-1}{4L_{22}\mu^2} \right] e^{-\mu^2} d\mu d\eta \right\}; \end{aligned} \tag{30a}$$

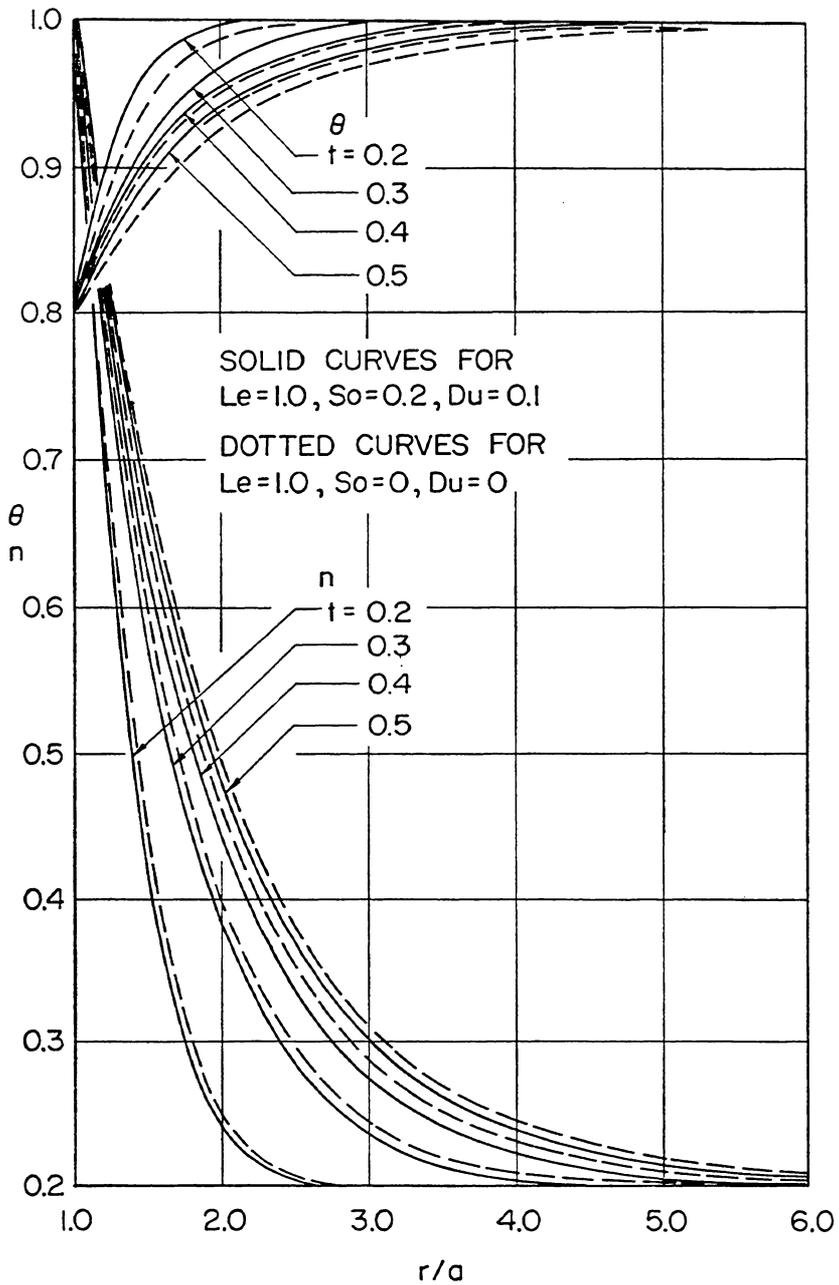


FIG. 3. Distributions of temperature and concentrations.

$$\begin{aligned}
 n_1(r, t) = & -\frac{1}{2r\sqrt{(\pi L_{22}t)}} \int_0^\infty e^{-\eta} \int_1^\infty r' \tilde{\mathfrak{F}}_2(r') \{e^{-(\tau+\eta-r')^2/4L_{22}t} - e^{-(\tau+\eta+r'-2)^2/4L_{22}t}\} dr' d\eta \\
 & - \frac{2}{r\sqrt{\pi}} \int_0^\infty e^{-\eta} \int_{(\tau+\eta-1)/2\sqrt{L_{22}t}}^\infty \tilde{\mathfrak{G}}_2 \left[t - \frac{r+\eta-1}{4L_{22}\mu^2} \right] e^{-\mu^2} d\mu d\eta \\
 & - \alpha_{21} \left\{ \frac{1}{2r\sqrt{(\pi L_{11}t)}} \int_0^\infty e^{-\eta} \int_1^\infty r' \tilde{\mathfrak{F}}_1(r') \{e^{-(\tau+\eta-r')^2/4L_{11}t} - e^{-(\tau+\eta+r'-2)^2/4L_{22}t}\} dr' d\eta \right. \\
 & \left. + \frac{2}{r\sqrt{\pi}} \int_0^\infty e^{-\eta} \int_{(\tau+\eta-1)/2\sqrt{L_{11}t}}^\infty \tilde{\mathfrak{G}}_1 \left[t - \frac{r+\eta-1}{4L_{11}\mu^2} \right] e^{-\mu^2} d\mu d\eta \right\}
 \end{aligned} \tag{30b}$$

Cases 3 and 4. Boundary value problems of the third and fourth kinds. The third boundary condition may be rewritten in the following matrix form:

$$I(\partial\chi/\partial r) + \alpha^{-1}A^{-1}B\gamma\alpha\chi - \alpha^{-1}A^{-1}B\gamma C = 0, \tag{31a}$$

where γ is a 2-by-2 matrix and C is a column vector given by:

$$\gamma = \begin{bmatrix} \gamma_a & 0 \\ 0 & \gamma_m \end{bmatrix}, \quad C = \begin{bmatrix} \theta_p \\ n_p \end{bmatrix}. \tag{31b}$$

Replacing χ_i by Ω_i/r in Eq. (31a) gives

$$I(\partial\Omega/\partial r) + (\alpha^{-1}A^{-1}B\gamma\alpha)\Omega - \alpha^{-1}A^{-1}B\gamma C = 0. \tag{31c}$$

By redefining the matrix B in Eq. (31a), one can obtain the boundary condition of the fourth kind described in Eqs. (4a) and (4b):

$$I(\partial\chi/\partial r) + \alpha^{-1}A^{-1}B\gamma\alpha\chi - \alpha^{-1}A^{-1}B\gamma C + \alpha^{-1}A^{-1}E = 0 \tag{31d}$$

where

$$B = \begin{bmatrix} 0 & 0 \\ 0 & B_{22} \end{bmatrix}, \quad E = \begin{bmatrix} E_1 \\ 0 \end{bmatrix}, \tag{31e}$$

$$B_{22} = -(\text{Sc Re } U_r)/D, \quad E_1 = -(\text{Sc Re } U_r L)/DLe. \tag{31f}$$

Since the third and fourth terms contain no dependent variables θ and n_1 explicitly, these two boundary conditions may be written as

$$I(\partial\chi/\partial r) + \alpha^{-1}A^{-1}B\gamma\alpha\chi + S = 0, \tag{31g}$$

where $S = \alpha^{-1}A^{-1}E - \alpha^{-1}A^{-1}B\gamma C$. Note that S_i 's are given functions of t .

We now consider the solutions of Eqs. (14a) and (14b) subject to the boundary condition (31g).

Define the following two new dependent variables u_1 and u_2 ,

$$u_1(r, t) = (\partial\Omega_1/\partial t) - (1 + h_{11})\Omega_1, \tag{32a}$$

$$u_2(r, t) = (\partial\Omega_2/\partial t) - (1 + h_{22})\Omega_2, \tag{32b}$$

where

$$h_{ij} = -(\alpha^{-1}A^{-1}B\gamma\alpha)_{ij}, \tag{32c}$$

and u_1 and u_2 satisfy Eqs. (27a) and (27b).

The initial and boundary conditions for u_1 and u_2 are

$$u_1(r, 0) = (\det \alpha)^{-1} \left\{ \left(r \frac{\partial \theta}{\partial r} + [1 - r(1 + h_{11})] \theta \right) - \alpha_{12} \left(r \frac{\partial n_1}{\partial r} + [1 - r(1 + h_{11})n_1] \right) \right\} = r \hat{\mathfrak{F}}_1(r), \quad (33a)$$

$$u_2(r, 0) = (\det \alpha)^{-1} \left\{ -\alpha_{21} \left(r \frac{\partial \theta}{\partial r} + [1 - r(1 + h_{22})] \theta \right) + \left(r \frac{\partial n_1}{\partial r} + [1 - r(1 + h_{22})n_1] \right) \right\} = r \hat{\mathfrak{F}}_2(r), \quad (33b)$$

$$u_1(1, t) = \frac{\partial \Omega_1}{\partial r} - (1 + h_{11})\Omega_1 = h_{12}\Omega_2 + S_1 = h_{12}\hat{\mathfrak{G}}_2 + S_1, \quad (34a)$$

$$u_2(1, t) = \frac{\partial \Omega_2}{\partial r} - (1 + h_{22})\Omega_2 = h_{21}\Omega_1 + S_2 = h_{21}\hat{\mathfrak{G}}_1 + S_2. \quad (34b)$$

Note that the initial conditions for u_1 and u_2 are given in terms of the unknown functions $\Omega_1(1, t) = \hat{\mathfrak{G}}_1(t)$ and $\Omega_2(1, t) = \hat{\mathfrak{G}}_2(t)$ respectively. Hence prior to the calculation of the solutions, two unknown functions $\hat{\mathfrak{G}}_1(t)$ and $\hat{\mathfrak{G}}_2(t)$ are to be determined. This proceeds as follows.

Since $\Omega_1(r, t)$ and $\Omega_2(r, t)$ are bounded in the domain, the proper solutions of Eqs. (32a) are

$$\Omega_1(r, t) = - \int_0^\infty u_1(r + \eta, t) e^{-(1+h_{11})\eta} d\eta, \quad (35a)$$

$$\Omega_2(r, t) = - \int_0^\infty u_2(r + \eta, t) e^{-(1+h_{22})\eta} d\eta, \quad (35b)$$

where $u_1(r, t)$ and $u_2(r, t)$ are formally given by Carslaw and Jaeger [12].

$$u_1(r, t) = \frac{1}{2\sqrt{(\pi L_{11} t)}} \int_1^\infty r' \hat{\mathfrak{F}}_1(r') \{ e^{-(r-r')^2/4L_{11}t} - e^{-(r+r'-2)^2/4L_{11}t} \} dr' + \frac{2}{\sqrt{\pi}} \int_{(r-1)/2\sqrt{L_{11}t}}^\infty \left\{ h_{12}\hat{\mathfrak{G}}_2 \left[t - \frac{r-1}{4L_{11}\mu^2} \right] + S_1 \left[t - \frac{r-1}{4L_{11}\mu^2} \right] \right\} e^{-\mu^2} d\mu \quad (36a)$$

$$u_2(r, t) = \frac{1}{2\sqrt{(\pi L_{22} t)}} \int_1^\infty r' \hat{\mathfrak{F}}_2(r') \{ e^{-(r-r')^2/4L_{22}t} - e^{-(r+r'-2)^2/4L_{22}t} \} dr' + \frac{2}{\sqrt{\pi}} \int_{(r-1)/2\sqrt{L_{22}t}}^\infty \left\{ h_{21}\hat{\mathfrak{G}}_1 \left[t - \frac{r-1}{4L_{22}\mu^2} \right] + S_2 \left[t - \frac{r-1}{4L_{22}\mu^2} \right] \right\} e^{-\mu^2} d\mu. \quad (36b)$$

Substituting (36a) and (36b) into (35a) and (35b), the following two relations are obtained on the surface of the droplet:

$$\Omega_1(1, t) = \hat{\mathfrak{G}}_1(t) = \phi_1(t) - \frac{2h_{12}}{\sqrt{\pi}} \int_0^\infty \int_{\eta/2\sqrt{L_{11}t}}^\infty \hat{\mathfrak{G}}_2 \left[t - \frac{\eta}{4L_{11}\mu^2} \right] e^{-\mu^2 - (1+h_{11})\eta} d\mu d\eta, \quad (37a)$$

$$\Omega_2(1, t) = \hat{\mathfrak{G}}_2(t) = \phi_2(t) - \frac{2h_{21}}{\sqrt{\pi}} \int_0^\infty \int_{\eta/2\sqrt{L_{22}t}}^\infty \hat{\mathfrak{G}}_1 \left[t - \frac{\eta}{4L_{22}\mu^2} \right] e^{-\mu^2 - (1+h_{22})\eta} d\mu d\eta, \quad (37b)$$

where $\phi_1(t)$ and $\phi_2(t)$ are known functions of t given by

$$\begin{aligned} \phi_1(t) = & \frac{-1}{2\sqrt{(\pi L_{11}t)}} \int_0^\infty \int_1^\infty r' \hat{\mathfrak{F}}_1(r') \{ e^{-(1+\eta-r')^2/4L_{11}t} - e^{-(1+\eta+r')^2/4L_{11}t} \} e^{-(1+h_{11})\eta} dr' d\eta \\ & - \frac{2}{\sqrt{\pi}} \int_0^\infty \int_{\eta/2\sqrt{(L_{11}t)}}^\infty \mathcal{S}_1 \left[t - \frac{\eta}{4L_{11}\mu^2} \right] e^{-\mu^2 - (1+h_{11})\eta} d\mu d\eta, \end{aligned} \tag{38a}$$

$$\begin{aligned} \phi_2(t) = & \frac{-1}{2\sqrt{(\pi L_{22}t)}} \int_0^\infty \int_1^\infty r' \hat{\mathfrak{F}}_2(r') \{ e^{-(1+\eta-r')^2/4L_{22}t} - e^{-(1+\eta+r')^2/4L_{22}t} \} e^{-(1+h_{22})\eta} dr' d\eta \\ & - \frac{2}{\sqrt{\pi}} \int_0^\infty \int_{\eta/2\sqrt{(L_{22}t)}}^\infty \mathcal{S}_2 \left[t - \frac{\eta}{4L_{22}\mu^2} \right] e^{-\mu^2 - (1+h_{22})\eta} d\mu d\eta. \end{aligned} \tag{38b}$$

Eqs. (37a) and (37b) are coupled linear integral equations which can be decoupled by the method of elimination as follows:

$$\begin{aligned} \hat{\mathfrak{G}}_1(t) = & \phi_1(t) - \frac{2h_{12}}{\sqrt{\pi}} \int_0^\infty \int_{\eta/2\sqrt{(L_{11}t)}}^\infty \phi_2 \left[t - \frac{\eta}{4L_{11}\mu^2} \right] e^{-\mu^2 - (1+h_{11})\eta} d\mu d\eta \\ & + \frac{4h_{12}h_{21}}{\pi} \int_0^\infty \int_{\eta/2\sqrt{(L_{11}t)}}^\infty \int_0^\infty \int_{\eta'/2\sqrt{(L_{22}(t-\eta/4L_{11}\mu^2))}}^\infty \hat{\mathfrak{G}}_1 \left[t - \frac{\eta}{4L_{11}\mu^2} - \frac{\eta'}{4L_{22}\mu'^2} \right] \\ & \cdot e^{-\mu'^2 - (1+h_{22})\eta' - \mu^2 - (1+h_{11})\eta} d\mu' d\eta' d\mu d\eta, \end{aligned} \tag{39a}$$

$$\begin{aligned} \hat{\mathfrak{G}}_2(t) = & \phi_2(t) - \frac{2h_{21}}{\sqrt{\pi}} \int_0^\infty \int_{\eta/2\sqrt{(L_{22}t)}}^\infty \phi_1 \left[t - \frac{\eta}{4L_{22}\mu^2} \right] e^{-\mu^2 - (1+h_{22})\eta} d\mu d\eta \\ & + \frac{4h_{12}h_{21}}{\pi} \int_0^\infty \int_{\eta/2\sqrt{(L_{22}t)}}^\infty \int_0^\infty \int_{\eta'/2\sqrt{(L_{11}(t-\eta/4L_{22}\mu^2))}}^\infty \hat{\mathfrak{G}}_2 \left[t - \frac{\mu}{4L_{22}\mu'^2} - \frac{\eta'}{4L_{11}\mu'^2} \right] \\ & \cdot e^{-\mu'^2 - (1+h_{11})\eta' - \mu^2 - (1+h_{22})\eta} d\mu' d\eta' d\mu d\eta \end{aligned} \tag{39b}$$

Exact solutions for $\hat{\mathfrak{G}}_1(t)$ and $\hat{\mathfrak{G}}_2(t)$ are rather difficult to obtain, except for cases in which either h_{12} or h_{21} is zero. For example, when $h_{12} = 0$ we have

$$\hat{\mathfrak{G}}_1(t) = \phi_1(t), \tag{40a}$$

$$\hat{\mathfrak{G}}_2(t) = \phi_2(t) - \frac{2h_{21}}{\sqrt{\pi}} \int_0^\infty \int_{\eta/2\sqrt{(L_{22}t)}}^\infty \phi_1 \left[t - \frac{\eta}{4L_{22}\mu^2} \right] e^{-\mu^2 - (1+h_{22})\eta} d\mu d\eta. \tag{40b}$$

If h_{12} and h_{21} are such that $4h_{12}h_{21}/\pi = \lambda \ll 1$, the approximate solutions for (39a) and (39b) may be given by the following series solution of ascending power of λ :

$$\hat{\mathfrak{G}}_1(t) = \hat{\mathfrak{G}}_1^{(0)}(t) + \lambda \hat{\mathfrak{G}}_1^{(1)}(t) + \lambda^2 \hat{\mathfrak{G}}_1^{(2)}(t) + \dots + \lambda^n \hat{\mathfrak{G}}_1^{(n)}(t) + \dots, \tag{41a}$$

$$\hat{\mathfrak{G}}_2(t) = \hat{\mathfrak{G}}_2^{(0)}(t) + \lambda \hat{\mathfrak{G}}_2^{(1)}(t) + \lambda^2 \hat{\mathfrak{G}}_2^{(2)}(t) + \dots + \lambda^n \hat{\mathfrak{G}}_2^{(n)}(t) + \dots, \tag{41b}$$

$$\hat{\mathfrak{G}}_1^{(0)}(t) = \phi_1(t) - \frac{2h_{12}}{\sqrt{\pi}} \int_0^\infty \int_{\eta/2\sqrt{(L_{11}t)}}^\infty \phi_2 \left[t - \frac{\eta}{4L_{11}\mu^2} \right] e^{-\mu^2 - (1+h_{11})\eta} d\mu d\eta, \tag{42a}$$

$$\hat{\mathfrak{G}}_1^{(1)}(t) = \iiint \hat{\mathfrak{G}}_1^{(0)} \left[t - \frac{\eta}{4L_{11}\mu^2} - \frac{\eta'}{4L_{22}\mu'^2} \right] e^{-\mu'^2 - (1+h_{22})\eta' - \mu^2 - (1+h_{11})\eta} d\mu' d\eta' d\mu d\eta, \tag{42b}$$

$$\hat{\mathfrak{G}}_1^{(n)}(t) = \iiint \hat{\mathfrak{G}}_1^{(n-1)} \left[t - \frac{\eta}{4L_{11}\mu^2} - \frac{\eta'}{4L_{22}\mu'^2} \right] e^{-\mu'^2 - (1+h_{22})\eta' - \mu^2 - (1+h_{11})\eta} d\mu' d\eta' d\mu d\eta. \tag{42c}$$

Similarly, $\mathcal{G}_2^{(n)}(t)$ are given by

$$\mathcal{G}_2^{(0)}(t) = \phi_2(t) - \frac{2h_{21}}{\sqrt{\pi}} \int_0^\infty \int_{\eta/2}^\infty \phi_1 \left[t - \frac{\eta}{4L_{22}\mu^2} \right] e^{-\mu^2 - (1+h_{22})\eta} d\mu d\eta, \tag{43a}$$

$$\mathcal{G}_2^{(1)}(t) = \iiint \mathcal{G}_2^{(0)} \left[t - \frac{\eta}{4L_{22}\mu^2} - \frac{\eta'}{4L_{11}\mu'^2} \right] e^{-\mu'^2 - (1+h_{11})\eta' - \mu - (1+h_{22})\eta} d\mu' d\eta' d\mu d\eta, \tag{43b}$$

$$\mathcal{G}_2^{(n)}(t) = \iiint \mathcal{G}_2^{(n-1)} \left[t - \frac{\eta}{4L_{22}\mu^2} - \frac{\eta'}{4L_{11}\mu'^2} \right] e^{-\mu'^2 - (1+h_{11})\eta' - \mu - (1+h_{22})\eta} d\mu' d\eta' d\mu d\eta. \tag{43c}$$

The solutions for the temperature and concentration are finally obtained as follows:

$$\theta(r, t) = -\frac{1}{r} \int_0^\infty u_1(r + \eta, t) e^{-(1+h_{11})\eta} d\eta - \frac{\alpha_{12}}{r} \int_0^\infty u_2(r + \eta, t) e^{-(1+h_{22})\eta} d\eta, \tag{44a}$$

$$\eta_1(r, t) = -\frac{\alpha_{21}}{r} \int_0^\infty u_1(r + \eta, t) e^{-(1+h_{11})\eta} d\eta - \frac{1}{r} \int_0^\infty u_2(r + \eta, t) e^{-(1+h_{22})\eta} d\eta. \tag{44b}$$

V. Rate of vaporization. The droplet vaporization corresponding to the third boundary condition can be calculated from Eq. (36b) together with (44b), whereas the rate of vaporization is prescribed in the cases of second and fourth boundary conditions.

According to Eq. (2a) the rate of vaporization corresponding to the first boundary condition with the assumption of $B_{11}q_a = 0$ is given by

$$(\partial\theta/\partial r) + B_{12}q_m = 0. \tag{45}$$

With the proper choice of the reference rate of vaporization, say $m = (4\pi k_a T_A)/L$, B_{12} becomes unity and the dimensionless rate of vaporization is given by

$$q_m(t) = \frac{1}{\sqrt{(\pi L_{11} t)}} \left\{ \frac{1}{2} \int_1^\infty \frac{r'(r' - 1)}{L_{11} t} \mathfrak{F}_1(r') e^{-(r'-1)^2/4L_{11}t} dr' - \mathfrak{G}_1(t) \right\} + \frac{\alpha_{12}}{\sqrt{(\pi L_{22} t)}} \left\{ \frac{1}{2} \int_1^\infty \frac{r'(r' - 1)}{L_{22} t} \mathfrak{F}_2(r') e^{-(r'-1)^2/4L_{22}t} dr' - \mathfrak{G}_2(t) \right\}. \tag{46a}$$

In particular, for a step type initial temperature and concentration distribution, the rate of vaporization is obtained from (23a) as follows:

$$q_m(t) = V_1 + \alpha_{12} V_2 + \frac{1}{\sqrt{(\pi t)}} \left(\frac{V_1}{\sqrt{L_{11}}} + \frac{\alpha_{12} V_2}{\sqrt{L_{22}}} \right) = \theta_A - \theta_i + \frac{(\det \alpha)^{-1}}{(\pi t)} \left\{ \frac{\theta_A - \theta_i - \alpha_{12}(n_A - n_i)}{\sqrt{L_{11}}} + \frac{\alpha_{12}[-\alpha_{21}(\theta_A - \theta_i) + (n_A - n_i)]}{\sqrt{L_{22}}} \right\}. \tag{46b}$$

In the limit of large t , the rate of vaporization is given in the following dimensional form:

$$\lim_{t \rightarrow \infty} q_m^*(t) \simeq \frac{4\pi k a}{L} (T_A - T_i). \tag{46c}$$

This result agrees with that obtained in the steady state theory (see Williams [1]) for the limiting case of slow vaporization,

$$q_m^*(t) = \frac{4\pi k a}{C_p} \ln \left[1 + \frac{C_p(T_A - T_i)}{L} \right] \simeq \frac{4\pi k a}{L} (T_A - T_i).$$

It is shown in Fig. 4 that the burning rate at $So = 0.2, Du = 0.2$ at $t = 2$ is approximately 20% greater than the burning rate with $So = Du = 0$.

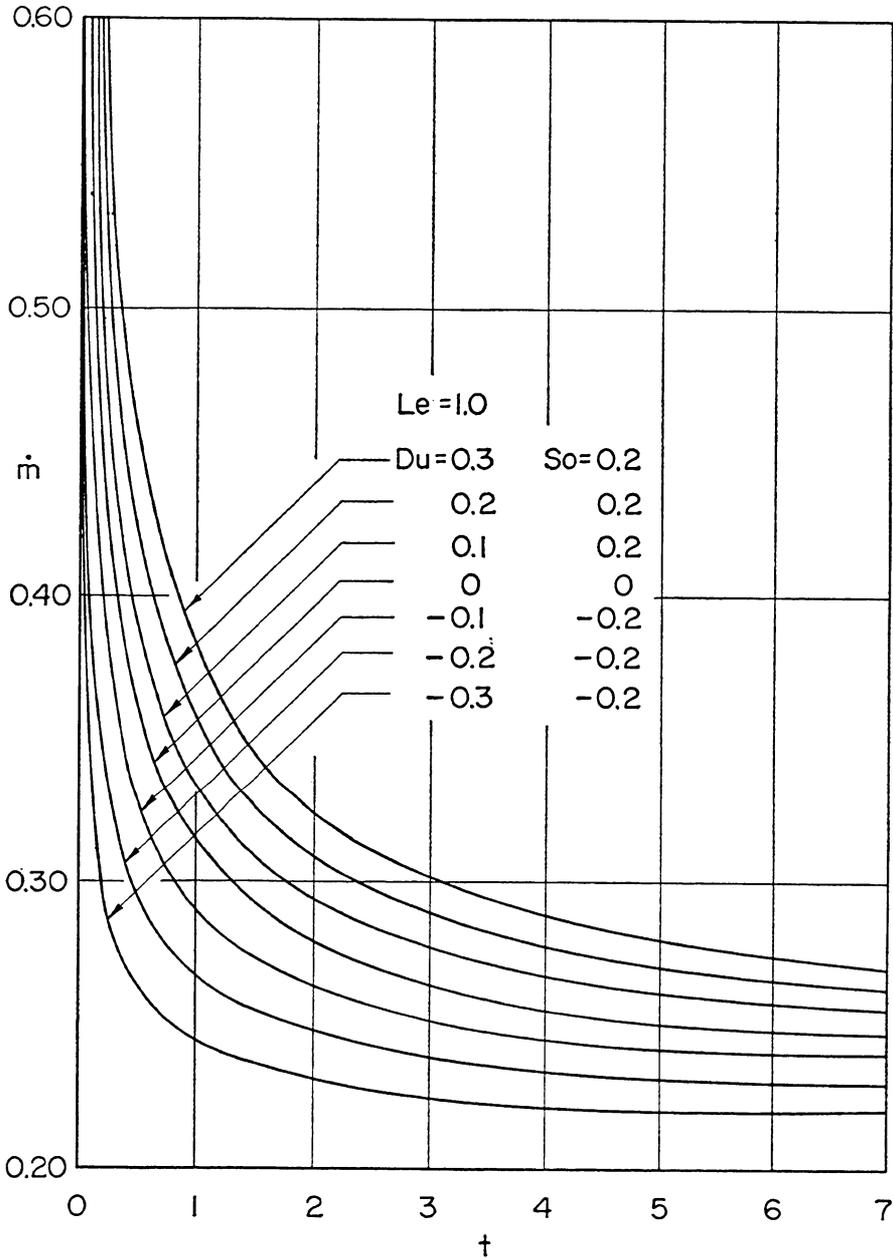


FIG. 4. Rate of droplet vaporization.

VI. Concluding remarks. An unique analytical method for predicting the development of the temperature and concentration profiles in the vicinity of an evaporable

liquid droplet exposed to a hot gas, including the Soret and Dufour effects, has been presented.

It is found that the fuel vapor diffusion is adversely affected by the Soret effect if the initial temperature of the droplet is lower than that of the surrounding hot gas. The fuel concentration is in general lower than the case of no Soret effect.

For a fixed surface and ambient gas temperatures, the Dufour effect tends to raise the gas temperature. This results in the increase of the rate of vaporization.

It is to be pointed out that the heat transfer inside the droplet tends to lower the rate of vaporization. Hence heat conduction inside the droplet should be taken into account together with the mass and heat transfer in the ambient gas. A preliminary investigation based on matching of the interior and exterior solutions reveals that the Dufour and Soret effects result in a rapid vaporization but slow mass diffusion. The substantial accumulation of the fuel vapor gives rise to higher concentration in the vicinity of the droplet, but lower concentration in the region far from the droplet.

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