

# Nonequilibrium Evaporation from a Heated Liquid Layer

S. P. Lin\* and M. Hudman†

Clarkson University, Potsdam, New York 13699-5725

The nonequilibrium evaporation or condensation under weightless conditions across an interface between a viscous gas and a viscous liquid, which is heated from below at a finite depth, is analyzed. An exact solution of the problem involving a moving boundary between a liquid and a gas–vapor mixture is obtained. Numerical results are used to demonstrate the dependence of the evaporation rate on the ratios of density, pressure, temperature, and the molecular weights of the two phases separated by the interface. It is shown that in a nonequilibrium evaporation process, heat transfer across the interface is not merely to supply the heat of evaporation as assumed in equilibrium evaporation, but also to effect the change in the internal energy of the gas–vapor mixture and to provide the flow work and kinetic energy required for the removal of vapor by a nondiffusive process. A numerical example is given to show the magnitude of the error arising from the equilibrium assumption. Based on the obtained exact solution, a conceptual experiment for the determination of the evaporation coefficient devoid of natural convection effect at microgravity is suggested.

## I. Introduction

**R**APID evaporation or condensation occurs in many applications, including liquid fuel combustion,<sup>1,2</sup> powder metallurgy,<sup>3</sup> and film cooling and film coating.<sup>4–7</sup> The phase changes across a liquid–gas interface are usually observed to take place in temperature and mass concentration fields with steep gradients. Yet it is commonly assumed that the Clapeyron equation (the thermodynamic equilibrium equation), can be adequately applied to describe the effect of phase change. The inadequacy of this assumption has been recognized,<sup>8,9</sup> and alternatives have been pursued.<sup>10–13</sup> Recently, a constitutive equation for evaporation/condensation has been found to have important applications.<sup>14,15</sup> However, in these applications, the evaporation coefficient is treated as a free parameter. On the other hand, quantitative knowledge of the evaporation coefficient is necessary to assess the impact of phase change on the fluid dynamics of many important practical problems, including atomization.<sup>16–22</sup> Attempts to evaluate the evaporation coefficient on Earth<sup>10</sup> have suffered from the lack of an exact theoretical solution because of the mathematical difficulty associated with gravity. This makes the comparison of tentative approximate solutions with experiments problematic. Moreover, natural convection on Earth masks the pure physical effect of evaporation and condensation.

An exact solution to the problem of rapid evaporation from a heated liquid layer into an inert gas–vapor mixture at zero-gravity has been discovered. This exact solution coupled with an experiment at microgravity will enable one to determine the functional form as well as the numerical value of the evaporation coefficient. A theory that can be used as the basis of such an experiment is given.

## II. Governing Equations

Consider the evaporation of a liquid layer, which is heated from below, into a gas–vapor mixture as shown in Fig. 1. The gas and the liquid are both Newtonian, nonreactive, and in-

compressible. The governing equations in a zero-gravity environment are

$$\nabla \cdot (V_i) = 0 \quad (1)$$

$$\rho_i \frac{dV_i}{dt} = \nabla \cdot \tau_i \quad (2)$$

$$\frac{dT_i}{dt} = \kappa_i \nabla^2 T_i \quad (3)$$

$$\frac{dC_i}{dt} = D_i \nabla^2 C_i, \quad (i = 1, 2) \quad (4)$$

where the subscript  $i$  stands for the liquid phase if  $i = 2$ , and the gas phase if  $i = 1$ ,  $\rho$  is the density,  $V$  is the velocity,  $d/dt$  stands for the substantial derivative,  $p$  is the pressure,  $\tau$  is the stress tensor,  $T$  is the temperature,  $\kappa$  is the thermal diffusivity,  $D$  is the mass diffusivity of the volatile component, and  $C$  is its mass concentration. Since the fluids considered are Newtonian and incompressible,

$$\tau_i = p_i \delta + \mu_i [\nabla V_i + (\nabla V_i)^T] \quad (5)$$

where  $\delta$  is the identity matrix, the superscript  $T$  stands for transpose, and  $\mu$  is the dynamic viscosity. Density is not con-

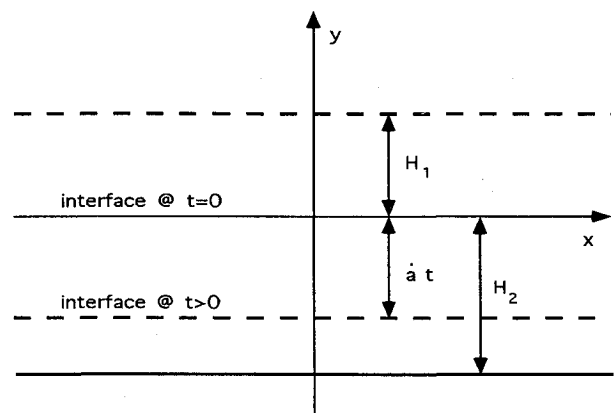


Fig. 1 Definition sketch.

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\*Professor, Mechanical and Aeronautical Engineering Department. Associate Fellow AIAA.

†Research Assistant, Mechanical and Aeronautical Engineering.

stant, although fluids are considered incompressible. All other physical properties will be considered constant. The previous governing equations must be supplemented by the equation of state, which relates density to temperature and pressure. For an ideal gas, e.g.,

$$\rho_1 = p_1 M / RT_1 \quad (6)$$

$$M = (p_{1v}/p_1)M_\beta + (1 - p_{1v}/p_1)M_\alpha \quad (7)$$

where  $R$  is the ideal gas constant and  $M$  is the molecular weight of the gas-vapor mixture,  $p_{1v}$  is the vapor pressure, and  $M_\alpha$  and  $M_\beta$  are, respectively, the molecular weights of the dry gas and the vapor.

The corresponding boundary conditions at the liquid-gas interface  $S = y - h(x, z, t) = 0$  include the following:

Kinematic:

$$|\nabla S| U \cdot \mathbf{n} - \partial_t h = 0 \quad (8)$$

Mass balance of mixture, and the volatile component:

$$[\rho(V - U) \cdot \mathbf{n}]_2 \equiv [\rho_1(V_1 - U) \cdot \mathbf{n} - \rho_2(V_2 - U) \cdot \mathbf{n}] = 0 \quad (9)$$

$$[\rho C(V - U) \cdot \mathbf{n}]_2 + [\rho(-D\nabla C) \cdot \mathbf{n}]_2 = 0 \quad (10)$$

Momentum:

$$[\rho V(V - U) \cdot \mathbf{n} - \boldsymbol{\tau} \cdot \mathbf{n}]_2 = -\sigma \nabla \cdot \mathbf{n} \quad (11)$$

Energy:

$$[\rho(V - U)(\frac{1}{2}V \cdot V + e - gy) \cdot \mathbf{n} - V \cdot \boldsymbol{\tau} \cdot \mathbf{n} - k\nabla T \cdot \mathbf{n} + (\rho D\nabla C)(e_\alpha - e_\beta) \cdot \mathbf{n}]_2 + \sigma(\nabla \cdot \mathbf{n})U \cdot \mathbf{n} = 0 \quad (12)$$

where  $\mathbf{n}$  is the unit normal vector pointing from the liquid to the gas at the interface,  $\nabla$  is the gradient operator,  $\partial_t$  denotes partial time differentiation,  $\sigma$  is the surface tension,  $e$  is the specific internal energy,  $-gy$  is the potential energy per unit mass that is negligible at microgravity,  $U$  is the interfacial velocity, and  $e_\alpha$  and  $e_\beta$  are, respectively, the specific internal energy of the host component and the evaporating phase. In addition to the previous interfacial conditions, one must also specify the temperature and the vapor concentration at the interface. Usually it is assumed that the gas and liquid phases are in thermal equilibrium and the vapor concentration is prescribed,<sup>1,22</sup> i.e.,

$$T_1 = T_2 \quad (13)$$

$$C_1 = C_1(T_1, p_{1v}) \quad (14)$$

where  $p_{1v}$  is the pressure of the vapor above the interface. Then the interface cannot be in thermodynamic equilibrium, since, in general,  $p_2 \neq p_1$  at the interface and thermodynamic equilibrium requires both mechanical and thermal equilibrium.<sup>23</sup> On the other hand, there is no reason why the liquid and the gas phases should have the same temperature at the interface.<sup>24</sup> This is borne out by the exact solution to be presented. An alternative to conditions (13) and (14) is to prescribe the liquid phase and the gas phase temperatures, i.e.,

$$T_2 = T_2(p_2, \rho_2) \quad (15)$$

$$T_1 = T_1(p_1, \rho_1) \quad (16)$$

where  $p_2$  may be taken to be equal to  $p_1$ , so that the interface is also in mechanical equilibrium. Then  $C_1$  cannot be prescribed. Another alternative is to use Eq. (15), but replace Eq.

(16) with the relation between the vapor flux and the temperature change across the interface<sup>15</sup>:

$$\rho_1(V_1 - U) \cdot \mathbf{n} - \rho_1 D_1 \nabla C_1 \cdot \mathbf{n} = E \left( \frac{M_\beta}{2\pi R} \right)^{1/2} \left( \frac{p_{2v}}{\sqrt{T_2}} - \frac{p_{1v}}{\sqrt{T_1}} \right) \quad (17)$$

where  $E$  is the evaporation coefficient,  $M_\beta$  is the molecular weight of the liquid,  $p_{2v}$  is vapor pressure at  $T_2$ , and  $p_{1v}$  and  $T_1$  are, respectively, the pressure and temperature of the vapor above the liquid. However,  $E$  depends on the heat and mass fluxes across the interface. The functional dependence of  $E$  on these fluxes is not known. It appears that what constitutes an appropriate temperature condition at an interface with a phase change is an unresolved question of physics.

In general,  $p_{1v} \neq p_{2v}$  and  $T_1 \neq T_2$  in Eq. (17), and no thermodynamic equilibrium is assumed across the interface. Hence, the set of boundary conditions (15) and (17) has wider applications than the other two sets of boundary conditions. Despite its important recent applications,<sup>7,14,15</sup>  $E$  has not yet been determined from comparing experimental measurements with exact solutions of transport equations. Attempts on Earth to compare experiments with the corresponding approximate theories for the determination of  $E$  suffer from uncertainty arising from the unavoidable interference from gravity.<sup>10</sup> The microgravity environment offers an opportunity to determine  $E$ , without ambiguity, from experiments in comparison with an exact solution. Such an exact solution is given next.

### III. Solution

A flat interface can be maintained even at microgravity if the pressure change across the interface just balances the net momentum flux at the interface, according to Eq. (11). For given  $T_{20}$ ,  $p_{20}$ ,  $T_{10}$ ,  $p_{10}$ , and  $C_{10}$ , where the subscript 0 denotes the constant interfacial value of the physical variable to which it is assigned, it is easily verified that the following fields of velocity, temperature, concentration of vapor, pressure, and density satisfy Eqs. (1-6) exactly:

$$V_1 = jv_1, \quad U = j\dot{a} \quad (18)$$

$$T_1 = T_{10} \quad (19)$$

$$C_1 = C_{10} + B[\exp(D_1 n^2 t + n(y - v_1 t)) - 1] \quad (20)$$

$$n = v_0/D_1, \quad v_0 = v_1 - \dot{a} \quad (21)$$

$$p_1 = p_{10} \quad (22)$$

$$p_{1v} = p_{10} C_1 / [C_1 + (M_\beta/M_\alpha)(1 - C_1)]$$

$$\rho_1 = p_{10} M / RT_1 \quad (23)$$

$$V_2 = 0, \quad p_2 = p_{20}, \quad C_2 = 1 \quad (24)$$

$$T_2 = T_{20} - A[\exp(\kappa_2 m^2 t + my) - 1] \quad (25)$$

$$m = -\dot{a}/\kappa_2 \quad (26)$$

$$\rho_2 = \rho_{20}[1 + \alpha(T_2 - T_{20})] \quad (27)$$

in which  $j$  is a unit vector in the  $y$  direction, and  $\alpha$  is the thermal expansion coefficient of the liquid. The boundary conditions (8) determine the interfacial position to be  $h = \dot{a}t$ , and the four unknown constants  $A$ ,  $B$ ,  $v_1$ , and  $\dot{a}$  are determined from the boundary conditions (9-12). They are found to be

$$A = -[v_1 p_{10} + \rho_{10} v_0 (e_{10} + v_1^2/2) + \rho_{20} \dot{a} e_{20} + (e_{\alpha 1} - e_{\beta 1})(\rho_{20} \dot{a} + \rho_{10} C_{10} v_0)] / \rho_{20} C_{p2} \dot{a} \quad (28)$$

$$B = C_{10} + (\rho_{20}/\rho_{10})\dot{a}/v_0 \quad (29)$$

$$\dot{a} = \mp[\rho_{10}(p_{20} - p_{10})/\rho_{20}(p_{20} - \rho_{10})]^{1/2} \quad (30)$$

$$v_1 = \pm(p_{10} - p_{20})/\rho_{20}\dot{a} \quad (31)$$

where  $e_{10} = e_1(T_{10})$ ,  $e_{20} = e_2(T_{20})$ ,  $e_{\alpha 1} = e_\alpha(T_{10})$ ,  $e_{\beta 1} = e_\beta(T_{10})$ ,  $\rho_{10}$  is evaluated from Eq. (6) with  $p_1 = p_{10}$  and  $T_1 = T_{10}$ , and the upper and lower signs in Eqs. (30) and (31) represent evaporation and condensation, respectively. Note that  $\dot{a} \neq 0$  in Eq. (31). It is seen from Eq. (30) that  $\dot{a} = 0$  when  $p_{10} = p_{20}$ . In this case  $v_1 = 0$ . By use of the relation

$$e_{10} = C_{10}e_{\beta 1} + (1 - C_{10})e_{\alpha 1} \quad (32)$$

the expression of  $A$  can be simplified to

$$A = \{\rho_{20}\dot{a}[e_{\beta 1}(T_{20}) + e_{\beta 1}(T_{10}) - e_{\beta 1}(T_{20})] - p_{1v} - \rho_{10}v_0v_1^2/2 - (\rho_{10}v_0 + \rho_{20}\dot{a})e_{\alpha 1}(T_{10})\}/\rho_{20}C_{p2}\dot{a} \quad (33)$$

where

$$e_{\beta 1}(T_{20}) = e_{\beta 1}(T_{20}) - e_{20}(T_{20}) \quad (34)$$

is the internal energy increase required for the change of phase from liquid to vapor at  $T_{20}$ . The second and third terms in the square bracket of Eq. (33) represent the additional internal energy change because of the temperature jump across the interface. The rest of the terms in Eq. (33) represent the nonequilibrium effect because of the motion of the fluid and interface.

The constant rate of evaporation/condensation cannot be achieved unless the heat flux  $Q$  and the mass flux  $J$  are held constant at the interface. These fluxes are given by

$$Q = -\rho_{20}C_{p2}\kappa_2\nabla T_2 = -\rho_{20}C_{p2}\dot{a}A \quad (35)$$

$$J = -\rho_{10}D_1\nabla C_1 + \rho_{10}C_{10}v_0 = -\rho_{20}\dot{a} \quad (36)$$

where  $C_{p2}$  is the constant pressure specific heat of the liquid. Note that the flux is positive if it is directed from the liquid toward the interface. Although the fluxes are constant, the temperature and concentration fields are unsteady. This is necessitated by the motion of the interface and the gas. It is seen from Eqs. (33) and (35) that heat is transferred not only for providing the heat of evaporation, but also for supplying the kinetic energy and the flow work to the convecting gas-vapor mixture. It is also seen that evaporation can take place either at  $T_{10} > T_{20}$  or  $T_{20} > T_{10}$ . The value of  $A$  for the two nonequilibrium cases is different, however. The previous exact solution shows how the heat can be transferred without heating up the gas phase.

The obtained exact solution can now be substituted into Eq. (17) to determine the evaporation coefficient  $E$ . In Eq. (17),  $p_{2v}$  may be taken as the saturated pressure at  $T_{20}$ . If the dry gas-vapor mixture is treated as an ideal gas mixture,  $p_{1v}$  and  $\rho_{10}$  are related to  $C_{10}$  by

$$p_{1v} = p_{10}C_{10}/[C_{10} + (M_\beta/M_\alpha)(1 - C_{10})] \quad (37)$$

$$\rho_{10} = [p_{1v}M_\beta + (p_{10} - p_{1v})M_\alpha]/RT_{10} \quad (38)$$

To facilitate a conceptual experiment, we write Eq. (17) in a dimensionless form with the aid of Eqs. (6), (10), (37), and (38),

$$E = \rho_r(M/M_\beta)[- \dot{a}(2\pi M_\beta/RT_{10})^{1/2}] \cdot \{(p_{2v}/p_{10})(T_1)^{1/2} - C_{10}/[C_{10} + M_r(1 - C_{10})]\}^{-1} \quad (39)$$

where

$$\rho_r = \rho_{20}/\rho_{10}, \quad T_r = T_{20}/T_{10}, \quad M_r = M_\beta/M_\alpha \quad \text{and}$$

$$(M/M_\beta) = (p_{1v}/p_{10}) + (1 - p_{1v}/p_{10})/M_r$$

Note that  $p_{1v}/p_{10}$  is a function of  $C_{10}$  and  $M_r$  according to Eq. (37). Thus, for a given liquid evaporating at  $T_{20}$  and  $p_{20}$  in a known gas at a different set of  $T_{10}$  and  $p_{10}$ ,  $C_{10}$  can be varied and  $\dot{a}$  measured to determine  $E$  as a function of  $\rho_r$ ,  $T_r$ ,  $C_{10}$ , and another dimensionless parameter  $[- \dot{a}(2\pi M_\beta/RT_{10})^{1/2}]$ . Of course, a different set of fluids must be used in the experiments to yield the dependence of  $E$  on  $M_r$ . The value of  $\dot{a}$  can also be calculated from Eq. (30) and compared with the measured value to confirm the theory. The tabulated values of  $E$  thus obtained can be used to estimate the instantaneous value of  $\dot{a}$  from Eqs. (17) and (10) when exact solutions are not easily obtained in various practical problems.

#### IV. Numerical Results

The values of transport properties including the mass diffusivity of water vapor in air, and specific heat and conductivity of water are taken from Raznjevic,<sup>25</sup> Haar et al.,<sup>26</sup> and Edward et al.<sup>27</sup>

It follows from Eqs. (30), (31), (37), and (38) that

$$v_1/\dot{a} = 1 - g_1/[p_{1v}/p_{10} + (1 - p_{1v}/p_{10})/M_r]$$

$$g_1 = \rho_{20}RT_{10}/p_{10}M_\beta, \quad M_r = M_\beta/M_\alpha$$

The ratio  $v_1/\dot{a}$  as a function of  $M_r$  for given  $T_{10} = 185^\circ\text{C}$ ,  $T_{20} = 180^\circ\text{C}$ ,  $p_{10} = 1.5538 \text{ MPa}$ ,  $p_{20} - p_{10} = 10^{-5} \text{ N/m}^2$ , and  $p_{2v} = 1.0021 \text{ MPa}$ , are plotted in Fig. 2 for various water vapor concentrations  $C_{10}$ . Note that  $v_1/\dot{a} < 0$ , regardless of whether  $\dot{a} > 0$  or  $\dot{a} < 0$ . That is to say  $v_1$  is always opposite to  $\dot{a}$  in both evaporation and condensation. All curves intersect at  $(M_r, v_1/\dot{a}) = (1, g_1 - 1)$ . For a dry gas whose molecular weight is smaller than that of water, the convective velocity of the mixture relative to the interfacial velocity decreases with an increase in the vapor concentration. For a dry gas whose molecular weight is larger than that of water, the converse is true. For  $C_{10} = 1$ ,  $-v_1/\dot{a}$  reaches the asymptotic value of  $g_1 - 1$ , which is the ratio for the case of evaporation into vacuum. The interfacial velocity corresponding to  $-v_1/\dot{a}$  given in Fig. 2 can be calculated from Eqs. (30), (37), and (38) for various values of  $C_{10}$  and  $M_r$ . The results are given in Fig. 3.

The evaporation coefficient  $E$  corresponding to the constant  $C_{10}$  curves in Fig. 3 are obtained from Eq. (17) and are given in Fig. 4. Note that the left side of Eq. (17) is simply equal to  $-\rho_{20}\dot{a}$  according to Eq. (11). It is seen that the evaporation

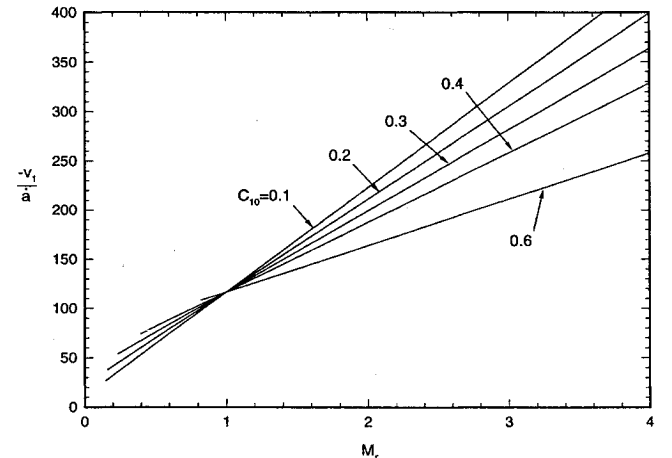


Fig. 2 Ratio of convective velocity to interfacial velocity.

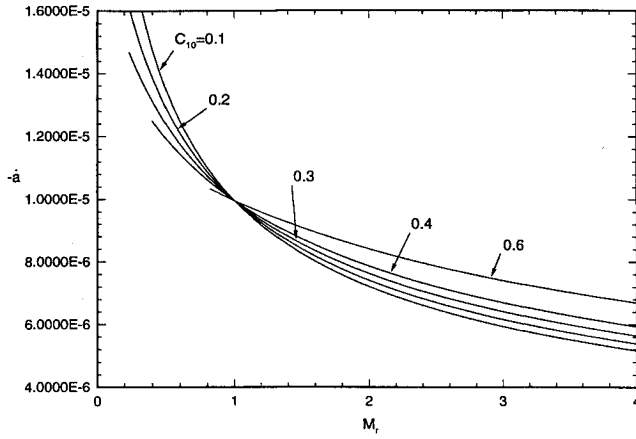


Fig. 3 Interfacial velocity.

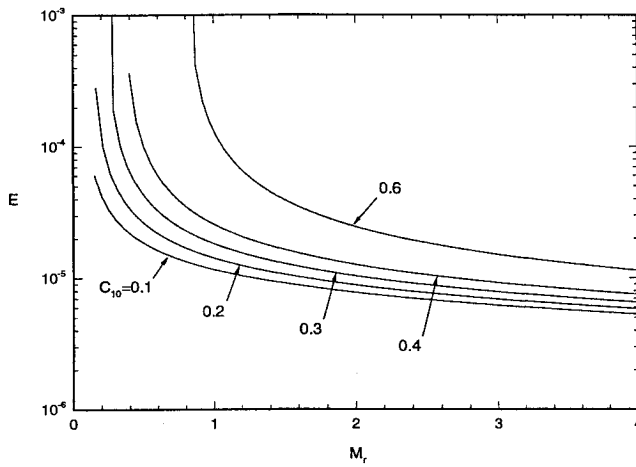


Fig. 4 Evaporation coefficient.

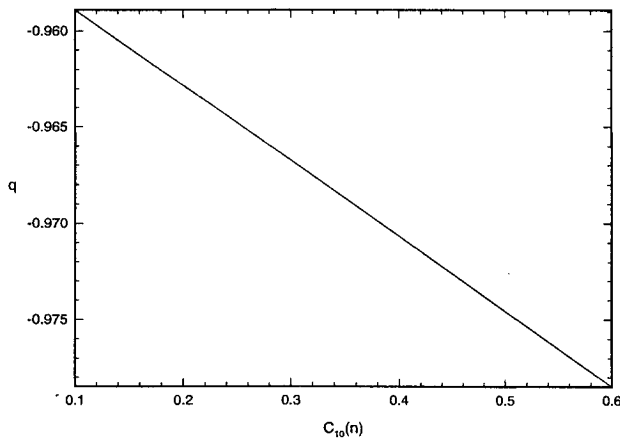


Fig. 5 Heat flux required for constant evaporation.

coefficient increases with  $C_{10}$  for all  $M_r$  in Fig. 4. The increase in  $E$  is more dramatic for  $M_r < 1$ .

It follows from Eqs. (35) and (33) that the dimensionless heat flux  $q$  is given by

$$q = Q/\rho_{20}C_{p2}T_{20}\dot{a} = q_1 + q_2 \quad (40)$$

$$q_1 = -[e_{f8}(T_{20}) + e_{B1}(T_{10}) - e_{B1}(T_{20})]/C_{p2}T_{20} - [(v_1/\dot{a} - 1)/\rho_r + 1][e_a(T_{10})/C_{p2}T_{20}] \quad (41)$$

$$q_2 = +[p_{10}v_1/\rho_{20}\dot{a} + (\rho_{10}/\rho_{20})(v_1/\dot{a} - 1)v_1^2/2]/C_{p2}T_{20} \quad (42)$$

where  $C_{p2}$  is the constant pressure specific heat of the liquid evaluated at  $T_{20}$ , and  $q_1$  is the heat flux used to raise the specific thermal energy in the phase transition across the interface. The first and second terms in the expression of  $q_2$  represent, respectively, the flow work and the kinetic energy increase associated with the phase change. As a function of  $C_{10}$  and  $M_r$ ,  $q$  is plotted in Fig. 5 for the case of evaporation. For the case of condensation, the sign of  $q$  is simply reversed. For a given  $M_r > 1$ ,  $q$  decreases with increasing  $C_{10}$ . This is because of the fact that the rate of increase of  $Q$  is faster than that of  $\dot{a}$  as  $C_{10}$  is increased when  $M_r > 1$ . The converse is true for  $M_r < 1$ . Figure 6 shows the fraction of heat flux that is spent in producing the mechanical energy required to convect the water

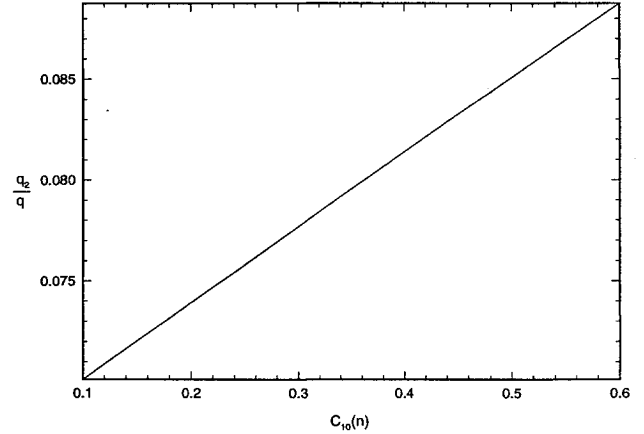


Fig. 6 Ratio of mechanical to thermal energy in evaporation.

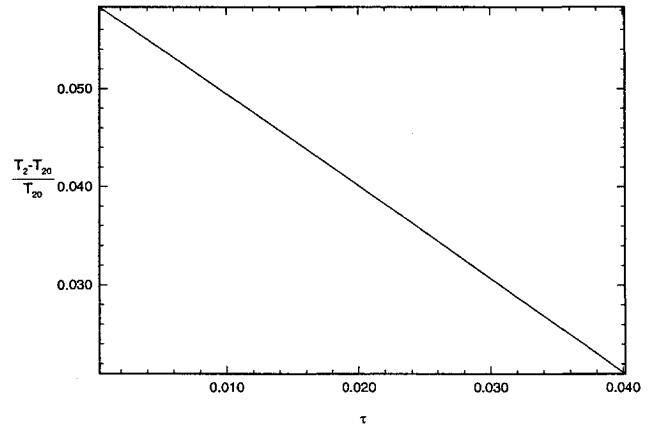


Fig. 7 Temperature variation required for constant evaporation.

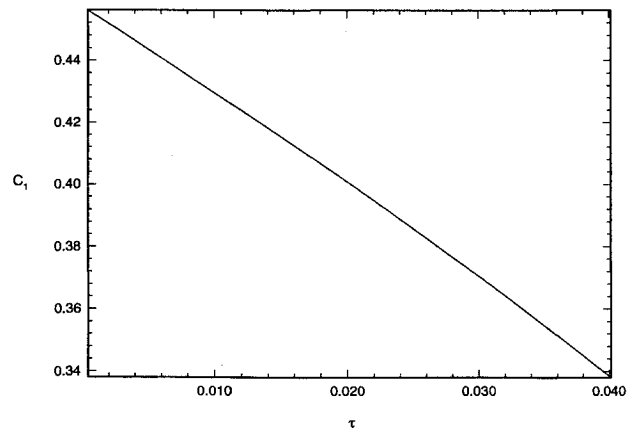


Fig. 8 Concentration variation required for constant evaporation.

vapor away from the air–water interface. This fraction becomes more significant when evaporation takes place at a higher rate.

Figures 7 and 8 give, respectively, the temperature variation at  $y = H_2 = -0.1$  cm, and the concentration variation at  $y = H_1 = 0.1$  cm with time, which are required to maintain the constant  $\dot{a}$  and  $v_1$  at  $C_{10} = 0.6$  and  $M_r = 0.621$ , with the other interfacial conditions remaining the same. The dimensionless time  $\tau$  in these figures is defined by

$$\tau = (d^2/\kappa_1)t$$

The maximum  $\tau$  in the figure corresponds to a dimensional time of approximately 60 s. The effect of the parameter  $g_1$  is simply to shift the curves in Figs. 2–6 upward when  $g_1$  is increased. It should be pointed out that the ideal gas Eq. (5) is adequate for the temperature and pressure ranges used in the present examples.<sup>28</sup>

## V. Discussion

An exact solution for the temperature field in the liquid, and the fields of velocity and vapor concentration in a constant temperature inert gas above a moving liquid–gas interface are obtained for the case of zero gravity. The interfacial velocity and the vapor–gas mixture velocity associated with the constant rate evaporation/condensation are obtained as functions of interfacial pressure, temperature, concentration, and the molecular weights of the liquid and gas. Numerical results are given for the case of water evaporating or condensing in an inert gas. The heat and mass fluxes required for various rates of evaporation/condensation are evaluated. The obtained exact solution clearly demonstrates that in nonequilibrium evaporation, neither thermal equilibrium nor mechanical equilibrium at the liquid–gas is required, i.e., the evaporation may take place with temperature and pressure jumps across the interface. The first term in the expression of  $q_1$  in Eq. (40) is heat of evaporation, and the rest of the terms represent the change of internal energy associated with the temperature jump across the interface. The first and second terms in the expression of  $q_2$  in Eq. (41) represent, respectively, the flow work and the kinetic energy associated with the convection of vapor–gas mixture. The sum of  $q_1$  and  $q_2$  is the total heat transfer required for evaporation. Thus, the commonly used interfacial condition that the heat transfer across an evaporating interface is only to supply heat of evaporation is adequate only for equilibrium evaporation without temperature jump and without vapor convection.

Figure 6 gives an example that an equilibrium interfacial energy condition may lead to an error of 10%. The ratio of the convective velocity of the gas–vapor mixture to the interfacial velocity may increase or decrease with increasing interfacial vapor concentration, depending on if the molecular weight of the dry gas is larger or smaller than that of the liquid. It follows from Eq. (42) that the error is even greater if  $M_\beta > M_\alpha$ . It is demonstrated that the temperature and concentration fields in nonequilibrium evaporation are by nature highly unsteady because of the movement of the interface, even if it is moving at a constant speed. In fact, the temperature at a given distance  $H_2$ , below the initial interfacial position and the concentration at  $y = H_1$  must follow the prescribed time variations depicted in Figs. 7 and 8 to maintain the constant evaporation rate. This temperature and concentration variation must be controlled in an experiment in space to determine the evaporation coefficient according to this theory. The concentration control is probably more challenging than the temperature control in experiments. The maintenance of  $T_{10}$ ,  $p_{10}$ ,  $T_{20}$ ,  $p_{20}$ , and  $C_{10}$  in the conceptual experiment described at the end of Sec. III is relatively easy to achieve. However, it should be pointed out that the obtained exact flow may be unstable. The surface temperature may vary along the interface because of uneven evaporation as a consequence of instability. Since the surface

tension increases with decreasing temperature, the temperature variation will cause local surface traction. This traction may be sufficiently large to overcome the viscous resistance and drags the liquid below the interface to create Marangoni convection if the liquid layer is sufficiently thin.<sup>5</sup> The liquid layer being heated from below may also become unstable because of buoyancy force, and form Rayleigh cells<sup>5</sup> even at microgravity if the liquid layer is sufficiently thick. The critical depth beyond which the liquid layer is unstable with respect to Rayleigh convection is probably of the order  $g/g_m$  where  $g_m$  is the gravitational acceleration at microgravity. Hence, the liquid layer in the proposed experiment must be sufficiently thick to prevent Marangoni instability, and sufficiently thin to prevent Rayleigh instability. To determine the stable range of thickness for the experiments, we must carry out stability analysis, using the obtained exact solution as the basic state. The stability analysis will be carried out in the near future.

## VI. Conclusions

Nonequilibrium evaporation or condensation is a highly unsteady process, even for the case of constant rate of phase change. The commonly used energy boundary condition that, the heat transfer across an evaporating interface is solely to provide heat of evaporation, is adequate only for the case of equilibrium evaporation. In nonequilibrium evaporation, in addition to the heat of evaporation, a significant portion of the heat transfer is used to generate the flow work and the kinetic energy associated with the removal of vapor by nondiffusive processes and to effect change in the internal energy of the gas–vapor mixture. The convective velocity of the gas–vapor mixture predicted in this theory is purely because of evaporation and is not affected by natural convection. Hence, the present theory provides a basis for a space experiment to determine the evaporation coefficient without the masking of the natural convection effect on Earth.

## Appendix: Error Analysis

The exact solution given in this work is based on the assumptions that the fluid is incompressible and the fluid properties are constant. The validity of these assumptions must be examined. The incompressibility of the gas phase requires that

$$\partial_t \rho_1 + v_1 \partial_y \rho_1 = e_1 = 0 \quad (A1)$$

Substitution of Eq. (23) into Eq. (43) yields:

$$e_1 = -\left(\frac{p_{10}M}{RT_1^2}\right)(\partial_t T_1 + v_1 \partial_y T_1) + \left(\frac{p_{10}}{RT_1}\right)(\partial_t M + v_1 \partial_y M)$$

Substituting Eqs. (19), (6), (7), (22), and (20) into this equation, we have

$$e_1 = \rho_1 B (\dot{a}^2/D_1) [1 - (v_1/\dot{a})^2] M_\alpha M_r (M_r - 1) / [M_r + (1 - M_r)C_1]^2 \cdot M \exp[(\kappa_1/D_1)(\tau - \dot{a}y/\kappa_1)] \quad (A2)$$

Eq. (44) gives the total time rate of change of the gas density at  $(y, \tau)$ , the maximum of which occurs at  $y = H_1$ . Thus, the ratio of the change in density up to time  $\tau$  to the density itself is

$$\tau e_1/\rho_1 = E_1(\tau)$$

$E_1(\tau)$  is a reasonable estimate of the error incurred by the assumption on gas incompressibility. The values of  $E_1(\tau)$  corresponding to the case of evaporation depicted in Figs. 7 and 8 are given in Table A1, together with the values of  $C_1(H_1, \tau)$ .

**Table A1 Error associated with incompressibility assumption**

| $\tau \times 100$ | $100 \times [T_2(-H_2, \tau) - T_{20}]/T_{20}$ | $C_2(H_2, \tau)$ | $E_1 \times 10^3$ |
|-------------------|------------------------------------------------|------------------|-------------------|
| 0.0               | 5.870                                          | 0.4571           | 0.0               |
| 0.8112            | 5.121                                          | 0.4349           | 0.823             |
| 1.6225            | 4.365                                          | 0.4118           | 1.629             |
| 2.4337            | 3.604                                          | 0.3878           | 2.416             |
| 3.2450            | 2.836                                          | 0.3628           | 3.183             |
| 4.0156            | 2.101                                          | 0.3380           | 2.676             |

It is seen that the error is less than five-hundredths of 1%.

Similarly, the total time rate of change of liquid density is

$$\frac{d\rho_2}{dt} = e_2 \quad (\text{A3})$$

The substitution of Eqs. (27) and (25) into (A3) gives,

$$e_2 = -\alpha\rho_{20}A(\dot{a}^2/\kappa_2)\exp[\tau - (\dot{a}/\kappa_2)y] \quad (\text{A4})$$

The maximum value of  $e_2$  occurs at  $y = \dot{a}$ . Hence, the ratio of the liquid density change to the liquid density itself  $E_2$ , is

$$E_2 < -\alpha\rho_{20}A\tau_m \exp(\tau_m - \dot{a}^2/\kappa_2)/\rho_2(H_2, 0)$$

where  $\tau_m$  is the maximum time involved in the evaporation process, and  $\rho_2$  is evaluated at  $y = -H_2$  and  $\tau = 0$ , where  $\rho_2$  is the minimum. The value of  $E_2$  corresponding to the case of Figs. 7 and 8 is found to be  $1.6 \times 10^{-8}$ , using  $\alpha = -2.64 \times 10^{-4}$ .

Had we not assumed that the conductivity is constant, we would have  $(\rho_2 C_{p2})^{-1} \nabla \cdot (k_2 \nabla T_2)$  instead of  $(\rho_2 C_{p2})^{-1} k_2 \nabla^2 T_2$  for the heat conduction term in the energy equation. Thus, the ratio of the neglected term to the retained term is

$$E_3 = \nabla k_2 \cdot \nabla T_2 / k_2 \nabla^2 T_2 = \left( \frac{dk_2}{dT_2} \right) \left( \frac{dT_2}{dy} \right)^2 / k_2 \left( \frac{d^2 T_2}{dy^2} \right)$$

Substituting Eq. (25) into the previous equation, we have

$$E_3 = \left( \frac{dk_2}{dT_2} \right) \left( \frac{A}{k_2} \right) \exp \left( \frac{\dot{a}^2 t}{\kappa_2} - \frac{\dot{a} y}{\kappa_2} \right) \quad (\text{A5})$$

The maximum  $E_3$  occurs at  $y = 0$ . This maximum value corresponding to the case depicted in Figs. 7 and 8 is found to be 0.038. This error is the maximum local error at a given  $(y, t)$ . The error associated with integration of  $\nabla^2 T$  over a distance of  $H_2$ , assuming the thermal diffusivity to be constant, can be estimated from

$$E_4 = \left[ \kappa_{22} \int \int \nabla^2 T_{22} dy dy - \kappa_{21} \int \int \nabla^2 T_{21} dy dy \right] / \kappa_{21} \int \int \nabla^2 T_{21} dy dy \quad (\text{A6})$$

where  $\kappa_{22}$  and  $\kappa_{21}$  are the thermal diffusivity evaluated, respectively, at  $y = -H_2$  and  $y = 0$ , and  $T_{22}$  or  $T_{21}$  is the temperature field evaluated, respectively, with  $\kappa_{22}$  or  $\kappa_{21}$  held constant. Substituting Eq. (25) into Eq. (A6) we have,

$$E_4 = (\kappa_{21}/\kappa_{22}) \exp[-(1 - \kappa_{21}/\kappa_{22})(\dot{a}^2 t/\kappa_{21} - \dot{a} y/\kappa_{21})] - 1$$

The maximum value of  $E_4$  in the time and space domain covered by Figs. 7 and 8 is found to be  $4.8 \times 10^{-6}$ .

Had we not assumed that the mass diffusion coefficient  $D_1$  is not constant, we would have  $\nabla \cdot (\rho_1 D_1 \nabla C_1)$  instead of

$\rho_1 D_1 \nabla^2 C_1$  for the mass diffusion term. Hence, the ratio of the neglected term to the retained term is

$$E_5 = D_1 \nabla C_1 \cdot \nabla \rho_1 / \rho_1 D_1 \nabla^2 C_1 \quad (\text{A7})$$

Although  $D_1$  is constant, this local relative error may not be zero because of the variation of  $\rho_1$  and  $C_1$  with  $y$ . Substituting the expressions for  $C_1$  and  $\rho_1$  into Eq. (A7) we have,

$$E_5 = M_r(M_\beta - M_\alpha)/M[C_1 + M_r(1 - C_1)]^2 B \exp[(\kappa_2/D_1) \times (1 - v_1/\dot{a})(\dot{a}^2/\kappa_2 - y\dot{a}/\kappa_2)] \quad (\text{A8})$$

The maximum value of  $E_5$  increases with  $t$ , and occurs at  $y = H_1$ . This maximum value corresponding to the case of Figs. 7 and 8 is found to be 0.17. This relatively large error is because of the fact that the particular case of evaporation is considered to have taken place at the interfacial vapor pressure 1.002 MPa, which is considerably less than the saturation vapor pressure  $p_{1s} = 1.1227$  MPa at  $T_{10} = 185^\circ\text{C}$ . When the vapor pressure is increased close to the equilibrium saturation pressure, the relative error is considerably reduced. The error also decreases as the molecular weight of the evaporating liquid approaches that of dry gas. In the limit of  $M_\alpha = M_\beta$ , this error vanishes as can be seen from Eq. (A8).

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