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Diffusion and Evaporation of a Liquid Droplet

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The process of evaporation and diffusion of a spherical liquid droplet in an atmosphere of noncondensable gas is studied theoretically. An equation for the shrinkage of the radius of the droplet is derived on the basis of continuity and momentum equations. Further, a conjugate problem consisting of the energy and mass balance for the gaseous environment is formulated. An approximation of thin thermal and diffusion boundary-layers is introduced to simplify the analysis. Results are presented for methanol-nitrogen, ammonia-nitrogen, and sodium-argon systems. It has been observed that the droplet of highly viscous fluid exhibits rapid contraction.

Nomenclature

a = thermal diffusivity of the vapor-gas mixture, cm^2/s
B = collision parameter
C_A = mass concentration in the vapor-gas mixture, g-moles/cm^3
C_{A0} = mass concentration in the vapor-gas mixture at the droplet surface, g-moles/cm^3
$C_{A\infty}$ = mass concentration in the vapor-gas mixture in the bulk flow, g-mole/cm^3
D = mass diffusivity of the vapor-gas mixture, cm^2/s
h = Lagrange space coordinate, cm^3
J = ratio of molecular weight of liquid to that of gas
k = thermal conductivity of the vapor-gas mixture, cal/cm·s·K
L = latent heat of vaporization, cal/g
M_a = solute molecular weight
M_b = solvent molecular weight
NA = mass transfer rate, g-moles/s·cm^2
p' = total pressure, dyne/cm^2
p = static pressure, dyne/cm^2
p_R = static pressure at the bubble surface, atm
p_V = vapor pressure at temperature T, atm
p = initial uniform static pressure, atm
R = radius of the droplet, cm
R_0 = initial droplet, cm
$\dot{R}$ = velocity of the droplet surface, cm/s
$\ddot{R}$ = acceleration of the droplet surface, cm/s^2
r = radial coordinate, cm
T = temperature of the vapor-gas mixture, K
T_b = boiling temperature of liquid, K
T = uniform initial temperature, K
t = time, s
U = radial velocity of the vapor-gas mixture, cm/s
U_I = temperature variable defined by Eq. (36), K
V_I = concentration variable defined by Eq. (22b)
μ = dynamic viscosity of the liquid, cm/s
μ_b = dynamic viscosity of the solvent, cm/s
ρ = density of liquid, g/cm^3
σ = surface tension between liquid and its vapor, cp
σ_a = molecular diameter
τ_I = time variable defined by Eq. (22a), s
τ = Lagrange time coordinate, s
ψ = association parameter

Introduction

THE presence of working fluid in the reservoir of a hot reservoir gas-controlled heat pipe is always undesirable. However, it may not always be true that a hot reservoir without wick also has no fluid or vapor. Most heat pipes contain excess liquid and it is quite possible that circumstances may place this liquid in the reservoir. For example, in a spacecraft thermal control application, the excess liquid may develop in the reservoir due to vibration of the vehicle during launch. Under such conditions, when the heat pipe starts operation, the liquid in the reservoir will vaporize and establish a vapor pressure corresponding to the reservoir temperature. This forces the noncondensable gas (NCG) out. Since the evaporator is acting similarly, the gas will be compressed to a relatively small volume in the condenser. This results in comparatively higher operating temperature and pressure conditions than desired. The only mechanism to remove this liquid is diffusion. In the following study the process of diffusion and evaporation in the nonwicked reservoir is considered.

One of the earliest investigations of evaporation in stagnant gases was made in 1877 by Maxwell,[1] who considered constant temperature evaporation from a wetted sphere in an attempt to describe the phenomenon of wet bulb temperature. Maxwell simplified the system by neglecting the convective effects and assuming equilibrium of liquid and vapor at the interface. He solved the conservation equations of mass and energy in the gas phase. Newbold and Amundson[2] improved the model to describe accurately the evaporation of a multicomponent droplet especially near its boiling point. It was concluded from the analysis that the augmentation of diffusive mass transfer by Stefan flow from the droplet plays an essential part in the model. Navon and Fenn[3] studied the interfacial mass and heat transfer during evaporation both theoretically and experimentally and showed that the overall evaporation rate is governed primarily by the surface temperature and the diffusional resistance of the gas. A comprehensive review of the evaporative convection is presented by Berg et al.[4]

In the present study, we treat the evaporation of single droplets of working fluid in an atmosphere of NCG. The process is considered as a diffusion and convection-dominating phenomenon.

For modeling ease, we postulate the following assumption for the mathematical description of the process: 1) the droplet is perfectly spherical; 2) the temperature and concentration of the droplet are uniform; 3) the droplet is situated on the reservoir wall under the saturated condition, and 4) the working fluid is incompressible.

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Mathematical Model

Equation of Motion

The liquid present in the nonwicked reservoir is supposed to be a spherical droplet of radius R_0 in a NCG at uniform temperature T. With the assumption of spherical symmetry, the continuity and momentum equations for an incompressible fluid outside the droplet are:

$$\frac{\delta}{\delta r}(r^2 u) = 0 \tag{1}$$

$$\frac{\delta u}{\delta r} + u\frac{\delta u}{\delta r} = -\frac{1}{\rho}\frac{\delta p'}{\delta r} + v\left[\frac{1}{r^2}\frac{\delta}{\delta r}\left(r^2\frac{\delta u}{\delta r}\right) - 2\frac{u}{r^2}\right] \tag{2}$$

From Eq. (1), it follows that

$$\frac{\delta u}{\delta r} = -2\frac{u}{r} \tag{3}$$

and

$$\frac{\delta}{\delta r}\left(r^2\frac{\delta u}{\delta r}\right) = \frac{\delta}{\delta r}(-2ur) = 2u \tag{4}$$

The viscous factor of Eq. (2) is therefore equal to zero. Equation (2) then becomes

$$\frac{\delta u}{\delta t} + u\frac{\delta u}{\delta r} = -\frac{1}{\rho}\frac{\delta p'}{\delta r} \tag{5}$$

where p' is the sum of all the normal stresses and is given by the static pressure plus the normal friction stresses:

$$p' = p + 2\mu\frac{\delta u}{\delta r} = p - 4\mu\frac{u}{r} \tag{6}$$

Replacing Eq. (6) in Eq. (5) and using relation (3), we obtain

$$\frac{\delta u}{\delta t} - 2\frac{u^2}{r} = -\frac{1}{\rho}\frac{\delta p}{\delta r} - 12\frac{\mu}{\rho} \tag{7}$$

If R is the radius of the contracting liquid drop at time t, the equation of continuity (1) requires:

$$u = -(R^2/r^2)R' \tag{8}$$

or

$$\frac{\delta u}{\delta t} = -\frac{\ddot{R}R^2 + 2R\dot{R}^2}{r^2} \tag{9}$$

Replacing Eqs. (8) and (9) into Eq. (7), it follows:

$$-\frac{\ddot{R}R^2 + 2R\dot{R}^2}{r^2} - 2\frac{R^4\dot{R}^2}{r^5} = -\frac{1}{\rho}\frac{\delta p}{\delta r} + 12\frac{\mu}{\rho}\frac{R^2}{r^4}\dot{R} \tag{10}$$

Integrating Eq. (10) between r and $r = \infty$, we have

$$-\frac{\ddot{R}R^2 + 2R\dot{R}^2}{r} - \frac{1}{2}\frac{R^4}{r^4}\dot{R}^2 = \frac{p(T) - p_\infty}{\rho} + 4\frac{\mu}{\rho}\frac{R^2\dot{R}}{r^3} \tag{11}$$

and at the surface of the droplet, we have

$$R\ddot{R} + \frac{5}{2}\dot{R}^2 + 4\frac{\mu}{\rho}\frac{\dot{R}}{R} = -\frac{P_R(T_S) - p_\infty}{\rho} \tag{12}$$

The vapor pressure $p_R(T_S)$ of the NCG at the droplet boundary is given in terms of pressure $p_V(T_S)$ of the liquid vapor within the droplet by

$$p_R(T_S) = p_V(T_S) - (2/R)\sigma(T_S) \tag{13}$$

Replacing $p_R(T_S)$ from Eq. (13) into Eq. (12) we obtain

$$R\ddot{R} + \frac{5}{2}\dot{R}^2 + 4\frac{\mu}{\rho}\frac{\dot{R}}{R} = -\frac{1}{\rho}\left[p_V(T_S) - p_\infty - 2\frac{\sigma}{R}\right] \tag{14}$$

The temperature at the droplet surface is T_S and it is assumed that the liquid in the droplet has uniform density and pressure and is in thermodynamic equilibrium at the temperature T_S. A similar equation without the viscous and pressure terms was obtained originally by Lord Rayleigh[5] for vapor-bubble growth. Plesset and Zwick[6] introduced the pressure term and Donne and Ferranti[7] introduced the viscous term in the study of the problem of the growth of vapor bubbles in a superheated liquid. In the study of the vapor growth problem, the right side of Eq. (14) takes a positive sign and the coefficient of the second term on the left side of Eq. (14) is replaced by 3/2. This difference comes by the negative velocity of the droplet contraction. Equation (14) is a second-order differential equation. For the complete mathematical description of the process of the receding of the droplet radius, the following initial conditions are prescribed:

$$R(0) = R_0 = \dot{R}(0) \tag{15}$$

Diffusion Equation

The liquid droplet is supposed to have uniform concentration and pressure. After the evaporation of the droplet, the vapor is diffused into the environment of NCG. Under these circumstances, we need to consider the phenomenon of diffusion outside the droplet surface. The diffusion equation in spherical coordinates in the presence of spherical symmetry is given by

$$\frac{\delta C_A}{\delta t} + u\frac{\delta C_A}{\delta r} = \frac{D}{r^2}\frac{\delta}{\delta r}\left(r^2\frac{\delta C_A}{\delta r}\right) \quad R(t) \leq r \leq \infty \tag{16}$$

The boundary conditions are:

$$r \to \infty, \quad C_A = C_\infty \tag{17}$$

$$r = R(t), \quad C_A = C_{A0} \tag{18}$$

The initial condition is

$$t = 0, \quad C_A = C_{A\infty} \tag{19}$$

The diffusion equation (16) is complicated by the presence of the convective term on the left-hand side. This term can be formally eliminated by introducing a transformation to the Lagrange coordinate h, τ with the position:

$$\left.\begin{aligned} h &= \tfrac{1}{3}[r^3 - R^3(t)] \\ \tau &= t \\ V(h,t) &= C_A(r,t) \end{aligned}\right\} \tag{20}$$

On substitution, Eq. (16) becomes

$$\frac{\delta V}{\delta t} = D\frac{\delta}{\delta h}\left(r^4\frac{\delta C_A}{\delta h}\right) \tag{21}$$

Further introducing a time variable τ_1, defined as

$$\tau_1 = \int_0^\tau R^4(\tau)\,\mathrm{d}\tau \tag{22a}$$

and a new concentration variable V_1 given by

$$V_1(h,\tau_1) = \int_h^\infty [C_{A\infty} - V(h',\tau)]\mathrm{d}h' \qquad (22b)$$

we obtain

$$\frac{\delta V_1}{\delta \tau_1} = D\left(1+\frac{3h}{R^3}\right)^{4/3} \frac{\delta^2 V_1}{\delta h^2} \qquad (23)$$

Here we introduce a diffusion boundary-layer approximation that the concentration gradient is appreciable only in a thin layer about the droplet. Thus

$$\frac{\delta V_1}{\delta \tau_1} = D\frac{\delta^2 V_1}{\delta h^2} \qquad (24)$$

The boundary conditions are

$$C_A\Big/_{r=R(t)} \Rightarrow C_A\Big/_{h=0} \Rightarrow \frac{\delta V_1}{\delta h}\Big/_{h=0} = C_{A0} \qquad (25)$$

$$V_1\Big/_{\tau\to 0} = 0, \quad V_1\Big/_{h\to\infty} = 0 \qquad (26)$$

The system of Eqs. (24-26) are tackled by the Laplace transform.[8] The solution can be written as

$$V_1 = -2C_{A0}(D\tau)^{1/2}\mathrm{i}\ \mathrm{erfc}(h/2\sqrt{D\tau}) \qquad (27)$$

where the integral of the error function (ierfc) is defined as

$$\mathrm{ierfc}(h/2\sqrt{D\tau}) = \int_{(h/2\sqrt{D\tau_1})}^\infty \mathrm{erfc}\xi \mathrm{d}\xi \qquad (28)$$

From Eq. (27), it follows that

$$V(h,\tau) - C_{A\infty} = C_{A0}\mathrm{erfc}(h/2\sqrt{D\tau}) \qquad (29)$$

From Eq. (29), it follows:

$$\frac{\delta V}{\delta h}\Big/_{h=0} = -\frac{C_{A0}}{\sqrt{\pi D \tau_1}} \qquad (30)$$

which can be rewritten in the original coordinate as

$$\frac{\delta C_A}{\delta r}\Big/_{r=R(t)} = -\frac{R^2(t)}{\sqrt{\pi D\tau_1}}C_{A0} \qquad (31)$$

The mass transfer at the interface is

$$N_A\Big/_{r=R(t)} = -D\frac{\delta C_A}{\delta r}\Big/_{r=R(t)} = C_{A0}\sqrt{\frac{D}{\pi\tau_1}}R^2(t) \qquad (32)$$

and the evaporation rate is given by

$$\frac{\mathrm{d}V_A}{\mathrm{d}t} = \frac{N_A S}{CA}\Big/_{r=R(t)} = 4\sqrt{\frac{\pi D}{\tau_1}}R^2(r) \qquad (33)$$

Energy Equation

With the assumption of spherical symmetry, the equation for temperature distribution outside the droplet surface in spherical coordinates can be written as

$$\frac{\delta T}{\delta t} + u\frac{\delta T}{\delta r} = \frac{a}{r^2}\frac{\delta}{\delta r}\left(r^2\frac{\delta T}{\delta r}\right), \quad R(t) \leqq r \leqq \infty \qquad (34)$$

The rate of heat loss by evaporation at the droplet surface[9] is given by

$$-LJB\mu\frac{\delta C_A}{\delta r} \qquad (35)$$

At the droplet surface, the heat transfer to it by conduction balances the heat loss by evaporation, thus

$$\left(-k\frac{\delta T}{\delta r}\right) - LJB\mu\frac{\delta C_A}{\delta r} = 0, \quad r = R(t) \qquad (36)$$

In addition, we assume that the temperature of the vapor-gas mixture is equal to the temperature of NCG at a large distance from the droplet surface, i.e.,

$$T = T_\infty, \quad r\to\infty \qquad (37)$$

For the initial condition, we suppose,

$$T = T_\infty, \quad t = 0 \qquad (38)$$

The convective type of energy equation (34) is coupled by the diffusion equation at the droplet boundary. The coupling at the boundary in Eq. (36) can be eliminated by substituting the concentration gradient term from the previous section. Proceeding as earlier, we have

$$\frac{\delta U_1}{\delta \tau_1} = a\left(1+\frac{3h}{R^3}\right)^{4/3}\frac{\delta^2 U_1}{\delta h^2} \qquad (39)$$

where

$$U_1 = \int_h^\infty [T_\infty - U(h',\tau_1)]\mathrm{d}h' \qquad (40)$$

Here we will also introduce a thermal boundary layer approximation[6] that the temperature gradient is appreciable only in a thin layer about the droplet surface. Thus Eq. (39) becomes

$$\frac{\delta V_1}{\delta \tau_1} = a\frac{\delta^2 U_1}{\delta h^2} \qquad (41)$$

The boundary conditions are

$$k\frac{\delta^2 U_1}{\delta h^2}\Big/_{h=0} = -LJB\mu\frac{C_{A0}}{\sqrt{\pi D\tau_1}} \qquad (42)$$

and

$$U_1\Big/_{h=\infty} = 0, \quad U_1\Big/_{\tau\to 0} = 0 \qquad (43)$$

By the application of Laplace transform, the system of Eqs. (41-43) can be solved. The solution is expressed as

$$U_1 = -2\frac{LJB\mu}{k}C_{A0}(a\tau_1)^{1/2}\mathrm{ierfc}(h/2\sqrt{a\tau_1}) \qquad (44)$$

From Eq. (44) it follows:

$$U(h,\tau) - T_\infty = \frac{LJB\mu}{k}C_{A0}\mathrm{erfc}(h/2\sqrt{a\tau_1}) \qquad (45)$$

The Numerical Treatment

We have to solve Eq. (14) for motion of the droplet for R under the initial conditions of Eq. (15). The value of R is further integrated to get another time variable τ_1. Once the droplet radius and the new time variable τ_1 are determined at a particular time instant, we calculate the concentration profile from Eq. (29), the evaporation rate from Eq. (33), and the temperature profile from Eq. (45). For the present study, all of the thermodynamic and fluid-dynamic properties of the system are considered constant. In practice, to do so we replace the second-order differential equation (14) in a set of two differential equations of the first order:

$$\dot{R}=Y$$

$$\dot{Y}=-\left[\frac{1}{\rho R}\left\{p_V(T_S)-p_\infty-2\frac{\sigma}{R}\right\}+4\frac{\mu}{\rho}\frac{Y}{R^2}+\frac{5}{2}\frac{Y^2}{R}\right] \tag{46}$$

The time variable τ is expressed in the form of the first-order differential equation as

$$\dot{\tau}_1=R^4(t) \tag{47}$$

The system of Eqs. (46) and (47) are integrated by a library subroutine DVERK available at the University of Stuttgart computer center which is a Runge-Kutta method based upon Verners fifth- and sixth-order pair of formulas.[10]

Results and Discussions

Figures 1-4 show the results of the calculations performed for the present study. The values of the thermodynamic and fluid dynamic parameters used in the sample calculation are presented in Table 1. The coefficient of diffusivity for the vapor-gas system is calculated from the expression[11]

$$D=\frac{9.76(10^{-8})(M_b\psi)^{1/2}T}{\mu_b\sigma_a 1.8} \tag{48}$$

We shall also approximate the relation between the equilibrium vapor pressure and the temperature by a relation

$$p_V(T_S)=p_V(T_\infty)-\frac{p_V(T_\infty)-p_\infty}{T_\infty-T_b}(T_\infty-T_S) \tag{49}$$

Figure 1 shows the history of evaporation of a droplet, i.e., the relation of the droplet radius vs time, for various combinations of working fluids and control gases. The results are obtained on the assumption of the constant concentration [Eq. (18)] and hence the temperature at the droplet surface. In this way the contraction of the droplet radius is unaffected by the change in surface conditions.

It is noted from Fig. 1 that the features of evaporation are almost same for the working fluids NH_3, CH_3OH, and Na considered for the sample calculation. The evaporation curves are always convex at the beginning and later become concave. This is because the evaporation rate is a decreasing funtion of time at the beginning (Fig. 2). This is in accordance with the physics of the phenomenon. The evaporation of the droplet is caused by the diffusion and the convection of the vapor. The

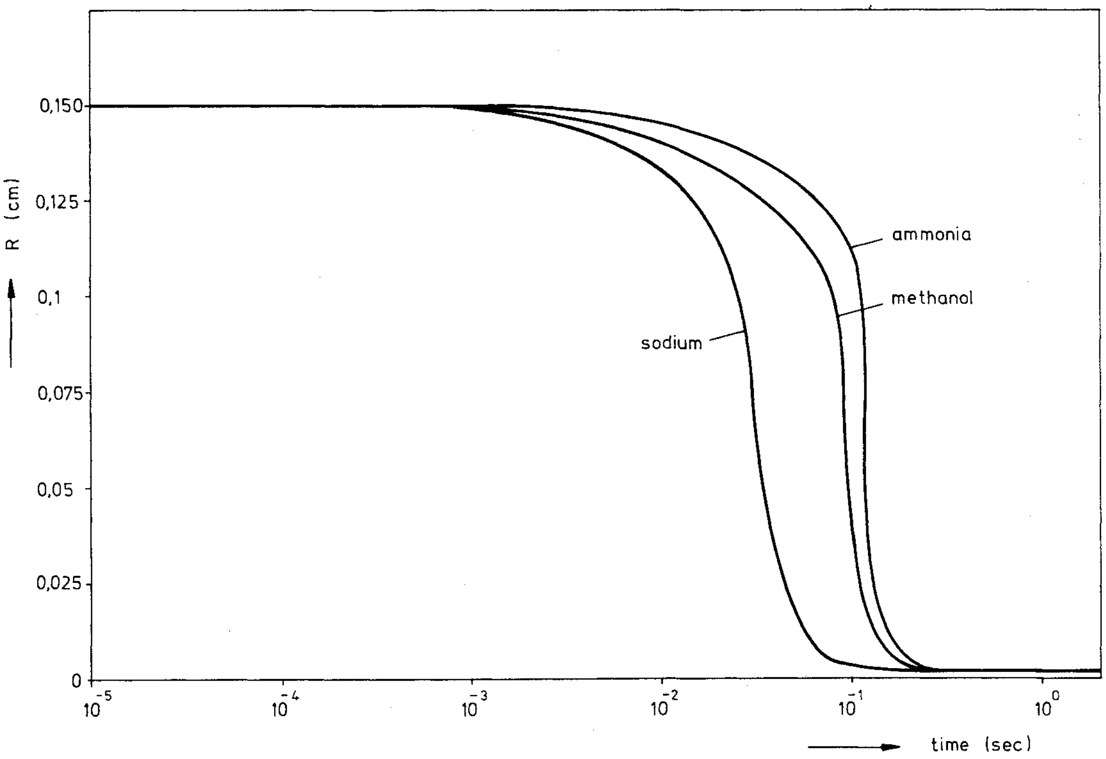

Fig. 1 Evaporation history of liquid droplet.

Table 1 Thermodynamic and fluid dynamic parameters

	T_∞	T_b	σ	σ_a	μ	M_a	M_b	ρ	ψ	p_∞	$p_V(T_b)$
Methanol-nitrogen	303	337.8	0.0218	31.2	0.00545	28	32	0.782	1.9	0.25	1.3
Ammonia-nitrogen	233	239.7	0.031	31.2	0.0027	28	17	0.6904	1.0	0.76	1.0
Sodium-argon	1000	1153	0.0104	29.9	0.016	39.94	22.997	0.7254	1.0	2.81	7.54

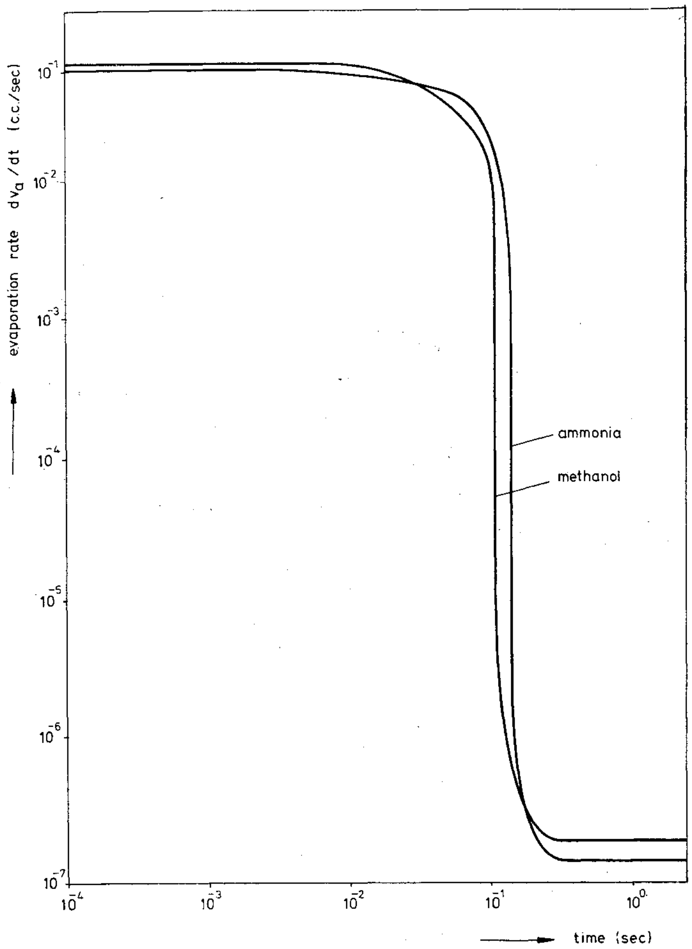

Fig. 2 Evaporation rate vs time.

process of diffusion is dominant very near the droplet surface and the process of convection is dominant further out. The rate of evaporation is proportional to the difference of the surface temperature and the mainstream temperature (temperature of the NCG). This process continues until the surface is at a temperature equal to that of the mainstream. The mainstream temperature increases due to convection currents induced by the temperature gradient near the droplet surface which results from the rate of evaporation being a decreasing function of time. In the latter stage the mainstream has the same temperature as that at the surface and the droplet evaporates at a constant rate of slow diffusion. Kotake and Okazaki[12] also observe a similar feature of evaporation, especially of methanol and ethanol in a quiescent air environment.

The process of the contraction of the droplet radius is primarily influenced by the viscous factor. The sodium droplet exhibits a more rapid contraction than the ammonia because the latter is less viscous.

The assumption of boundary layer to simplify the process of calculation is fairly well justified. This is confirmed from the temperature and the concentration profile obtained in Figs. 3 and 4. The diffusion boundary layer is thinner than the thermal boundary layer.

Conclusions

The process of evaporation and diffusion of a liquid droplet in an atmosphere of noncondensable gas is studied theoretically. The analysis is simplified by the introduction of the thermal and diffusion boundary layer. The boundary-layer approximation to study the mechanism is well justified. Furthermore, the droplet of highly viscous fluid exhibits rapid contraction.

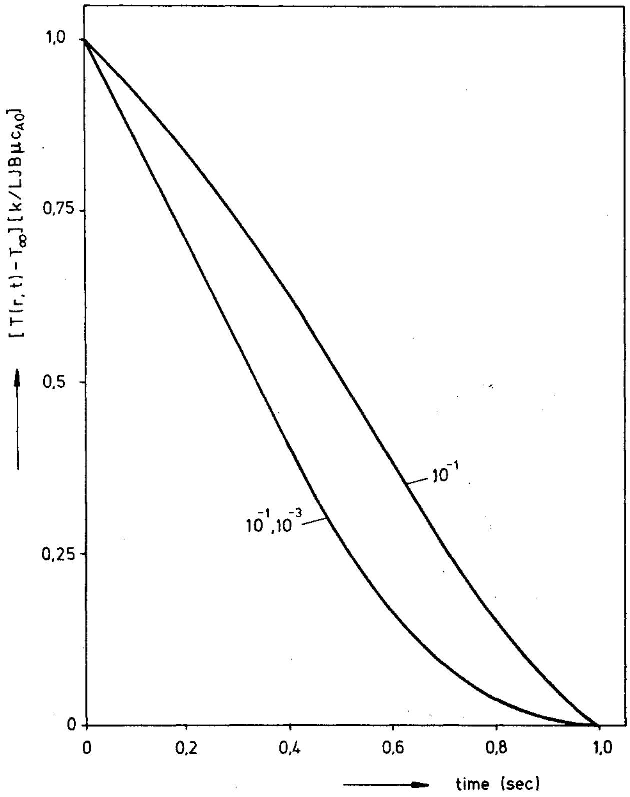

Fig. 3 Temperature distribution in thermal boundary layer.

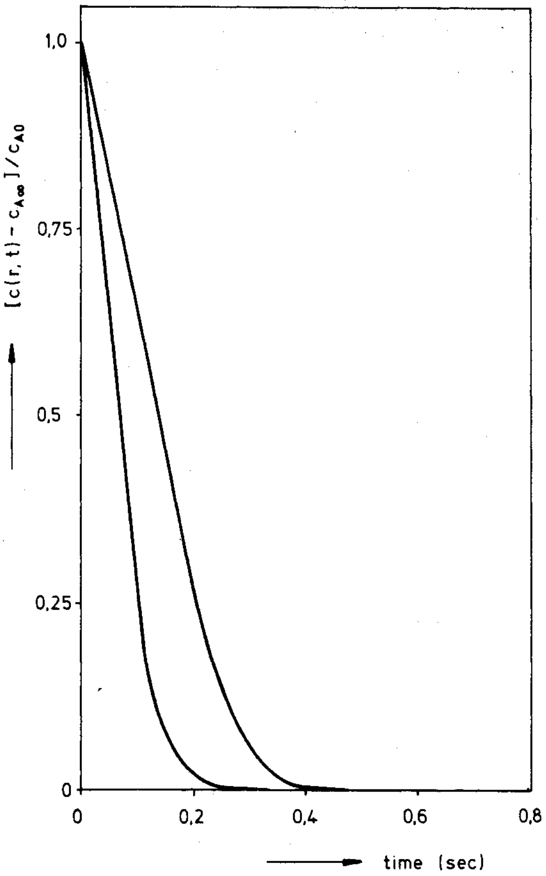

Fig. 4 Concentration distribution in diffusion boundary layer.

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