APPENDIX B

TRANSPORT PROCESSES AND INTERFACIAL
PHENOMENA IN AN EVAPORATING MENISCUS

by

CONF-910739--2

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DE91 005561

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EXTENDED ABSTRACT

for

1991 ASME/AICHE National Heat Transfer Conference
(AIAA Sponsored Sessions)
July 28-31, 1991 Minneapolis, MN

When a liquid film wets a solid surface, a contact line region is formed where the vapor, liquid and solid phases are in close proximity. The film thickness in this region varies from about 10 μm (Capillary Meniscus) to less than about 100nm (Adsorbed film). In addition to being functions of temperature and pressure (as for a bulk phase), the thermodynamic properties (e.g., chemical potential) of these thin films depend on their shape (curvature) and thickness due to surface forces [1,2]. The coupled transport processes and interfacial phenomena occurring in this microscopic region are also controlled by these surface forces.

The objective of this paper is to report experimental data which complement earlier analytical models of this region [3,4]. Figure 1 shows the system geometry which consists of a flat silicon plate partially immersed at a small angle, θ , in a pool of liquid. The plate is in a closed cell and a spreading

liquid (1,1,2- Trichlorotrifluoro ethane), in equilibrium with its own vapor, forms a zero contact angle with the plate. The plate can be electrically heated at the upper end by supplying power to a thin, rectangular platinum heater which is painted on the backside of the silicon wafer. The meniscus thickness profile, which is related to the effective pressure in the liquid, was used as a probe for understanding the sensitivity of the meniscus to the non-equilibrium effects associated with evaporation/condensation mechanisms.

A long working distance microscope was used to view the naturally occurring interference fringes in the liquid. Coherent Light ($\lambda = 546 \text{ nm}$) was used at normal incidence and the reflected interference patterns were viewed and captured using a CCD video camera. The data acquisition process was automated by using an image processing system which facilitated improved data resolution and enhanced data analysis. A continuous film thickness profile from the capillary meniscus region down to the thickness of the adsorbed film was obtained. This continuous profile, consisting of data at every pixel, was amenable to accurate differentiation schemes. Thus, after an image had been captured, all the relevant information, i.e., the thickness, slope, curvature and curvature gradient profiles, could be automatically obtained and used in understanding the phenomena. Transient data was captured by continuously recording on a high quality, U-Matic, Sony, VO-9600 VCR. The high resolution VCR was connected through a Sony FCG-700 Frame Counter, which superimposed each frame with a permanent identification number. Thus, the oscillatory motion of the contact line, at a high power input, could be studied.

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THEORETICAL BACKGROUND [3,4]

The following equation models the conservation of mass in the meniscus

$$\rho_l \frac{\partial \delta(x,t)}{\partial t} = - \frac{\partial \Gamma}{\partial x} - m_e \quad (1)$$

where,

$$\Gamma = - \frac{\delta^3}{3\gamma} \frac{d}{dx} [\Delta P_g] + \frac{\delta^2}{2\gamma} \sigma' \quad (2)$$

$$\Delta P_g = - \sigma K - \Pi + \rho_l gH \quad (3)$$

$$K = \frac{\delta''}{[1 + (\delta')^2]^{3/2}} \quad (4)$$

$$\Pi = \frac{-B}{\delta^n} \quad (5)$$

$$m_e = m_{e, \Delta T} + m_{e, \Delta P} = a (T_{iv} - T_v) + b (\Delta P_g) \quad (6)$$

$$b = C_1 \left[\frac{M}{2\pi R T_l} \right]^{0.5} \left[\frac{V_l P_v}{R T_{lv}} \right] \quad (7)$$

$$a = b \left[\frac{\Delta h}{T_v} \right] \quad (8)$$

RESULTS AND ANALYSIS

At equilibrium, there is perfect isothermality i.e., all three phases and the two interfaces are at the same temperature. At equilibrium, $\Delta P_g = 0$ for the entire meniscus. Also $\partial\delta/\partial t = 0$ and hence, $\Gamma = 0$, $\Gamma' = 0$, $\Delta T = 0$, $\sigma' = 0$ and $M_e = 0$. However, equilibrium is difficult to maintain and a small transfer of mass between the liquid and vapor phases (evaporation or condensation) resulting from small temperature gradients and/or vibrations has a large effect on the pressure field. Figure 2 shows three "near-equilibrium" profiles in which a small amount of evaporation from the adsorbed film causes it to thin down from its equilibrium values. A positive curvature gradient (Figure 3) which causes fluid flow from the capillary meniscus to the adsorbed film is highest in magnitude for the case when the adsorbed film is least thick. The results confirm the sensitivity of the system and help to characterize (determine the disjoining pressure) the system.

The heat transfer studies were conducted by supplying power ,P, to the platinum heater. The fringe separation decreased and the change in the pressure field was manifested as a dramatic change in the film thickness profile (Figure 4). Fluid flowed towards the heated end to replace the fluid lost by evaporation and to maintain a steady meniscus. Consistent with theoretical models [5], the curvature increased very rapidly from a value of zero in the adsorbed film to a high value and then decreased rapidly to a thickness of about 1 μm . The location and magnitude of the maximum curvature were found to be a function of the power input (Figure 5).

At higher power inputs, the two terms on the right hand side of Eq. (1) could not balance each other and the meniscus started to oscillate. The oscillations involved a translation of the meniscus to an "advanced" position; a short steady period (for about 1 sec) at the advanced position; then translation

of the meniscus back to a "receded" position; a short steady period at the receded position and then a repeat of the cycle. The separation of the fringes was very low throughout the cycle, but the fringes were visibly closer (i.e., the thickness, curvature and curvature gradient profiles were steeper) at and near the receded position. The amplitude of the meniscus translations increased and the time period of the oscillations decreased with an increase in power input. Figure 6 shows oscillating profiles taken at intervals of 15 frames (1/2 second). The "advanced" and "receded" film thickness profiles are shown in Figure 7 and the corresponding curvature profiles are shown in Figure 8. The profiles are compared with a constant slope profile which is for a horizontal meniscus (i.e. slope = $\tan \theta$). The build-up of curvature is quite pronounced for the "receded" profile. Therefore, the curvature gradient which pumps liquid into the thin film region is quite large. A preliminary understanding of the oscillation process as related to the balance between the pumping capacity of the fluid and the evaporative flux caused by the heated solid is obtained. Surface tension gradients may also cause recirculation.

Microcomputer based image processing technology has enabled us to investigate these complex phenomena in equilibrium, heated and oscillating meniscii. The results demonstrate that the processes of change-of-phase heat transfer and fluid flow in thin films are intrinsically connected because of their common dependence on the intermolecular force field and gravity.

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NOMENCLATURE

a	Property value defined by Eq. (8)	$\text{kg/m}^2 \text{ sec } \kappa$
B	Force constant defined by Eq. (5) when $n = 4$, $B = B$ (London - van der Waals constant)	J - m
	when $n = 3$, $B = \bar{A} = \frac{A}{6\pi}$	J
b	Property value defined by Eq. (7)	sec/m
C_i	Evaporation coefficient [Eq. (7)]	[]
g	Acceleration due to gravity	m/sec^2
Δh	Enthalpy of vaporization per unit volume	J/m^3
H	Height above pool	m
K	Curvature	m^{-1}
M	Molecular Weight	kg mol/kg
M_e	Interfacial evaporative mass flux	$\text{kg/m}^2 \text{ sec}$
P	Pressure	N/m^2

	or Power Input	J/sec
R	Universal Gas Constant	J/kg mol K
T	Temperature	K
t	Time	sec
V	Molar Volume	m ³ /kg mol
x	Distance along the plate	m

GREEK

Δ	Indicates Difference	
δ	Film Thickness	m
θ	Angle	Radians
Π	Disjoining Pressure	N/m ²
σ	Liquid Vapor Surface Tension	N/m
ρ	Density	kg/m ³
λ	Wavelength of Light in Vacuum	m
ν	Kinematic Viscosity	m ² /sec
Γ	Mass flowrate per unit width	kg/m sec

SUBSCRIPTS

g	Including effect of gravity
i	Average
l	Liquid
lv	Liquid-Vapor

v Vapor

ΔT Due to temperature jump

ΔP Due to pressure jump

SUPERSCRIPTS

Indicates Differentiation

Figure 1 Completely Wetting Liquid on an Inclined Plate

Figure 2 Film thickness profiles for R113 on Silicon with a surface oxide, $\theta \approx 6.5^\circ$.

Figure 3 Curvature (K) versus Film Thickness (δ) for R113 on Silicon with a surface oxide, $\theta \approx 6.5^\circ$.

Figure 4 Film thickness profiles for Heat Transfer Studies; R113 on Silicon with a surface oxide, $\theta \approx 6.5^\circ$.

Figure 5 Curvature (K) versus Film Thickness (δ) for Heat Transfer Studies; R113 on Silicon with a surface oxide, $\theta \approx 6.5^\circ$.

Figure 6 Film thickness profiles for Oscillating Meniscus; Frames 0, 15, 31, 45; R113 on Silicon with a surface oxide, $\theta \approx 6.5^\circ$; $P = 600\text{mW}$.

Figure 7 Film thickness profiles for Oscillating Menisci of R113 on Silicon with a surface oxide, $P = 600\text{mW}$, $\theta \approx 6.5^\circ$.

Figure 8 Curvature (K) versus Film Thickness (δ) for Oscillating profiles of R113 on Silicon with a surface oxide, $P = 600\text{mW}$; $\theta \approx 6.5^\circ$.

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06 / 07 / 91