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THE ROLE OF ADSORPTION IN DETERMINING THE MINIMUM FILM BOILING TEMPERATURE

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Abstract — A minimum film boiling mechanism is suggested, based on the adsorption characteristics of the system. The wetting process is characterized by a precursor, non-evaporating film. As the surface temperature increases the film thickness decreases, with a sharp decrease at a temperature threshold determined by the heat of adsorption. The minimum film boiling temperature is assumed to be the temperature at which only one monolayer exists. The predicted values are in fair agreement with experimental results for different systems.

NOMENCLATURE

a,	constant;
A,	Avogadro's number;
b,	constant;
c_p,	specific heat;
d,	molecular dimension;
k,	thermal conductivity;
k_1,	non-dimensionless number;
M,	molecular weight;
n,	constant;
P,	pressure;
P_0,	saturation pressure;
Q_a,	heat of adsorption;
R,	gas constant;
S_{ls},	spreading coefficient of liquid on bare surface;
S_{lsv},	spreading coefficient of liquid on adsorbed film;
T,	temperature;
T_{cr},	thermodynamic critical temperature;
T_{mfb},	minimum film boiling temperature;
T_{hn},	homogeneous nucleation temperature;
V_l,	molecular volume;
x,	thickness of adsorbed film.

Greek symbols

α,	contact angle;
β,	dimensionless parameter;
Γ,	number of adsorbed molecules per unit area;
Γ_0,	number of adsorbed molecules per unit area which form a monolayer;
γ,	surface tension;
ε,	field potential of solid surface;
θ,	non-dimensional occupancy;
π,	film pressure;
ρ,	density;
τ,	residence time of molecule in adsorbed state.

Subscripts

l,	liquid;
v,	vapor;
s,	surface.

INTRODUCTION

ONE OF the problems of interest with respect to nuclear reactor safety is the assessment of the minimum temperature to sustain film boiling. At this temperature, usually termed the minimum film boiling temperature (T_{mfb}), the vapor film separating the hot surface from the liquid becomes unstable and the regime changes to transition boiling.

Berenson [1] following earlier analyses by Chang [2] and Zuber [3] used the concept of a continuous horizontal vapor film, subject to Taylor instability, in formulating a model for the minimum film boiling temperature. However, contacts of liquid–solid well into the film boiling region were recorded by Bradfield [4], Swanson *et al.* [5] and Yao and Henry [6]. Henry [7] recognized the importance of surface properties, in view of frequency of random contacts near T_{mfb}, by introducing the interface temperature upon contact and evaporation from a residual liquid microlayer in order to correct the Berenson equation. The resulting empirical equation agrees fairly well with a variety of experimental data points, including four for sodium–tantalum. Gunnerson and Croneberg have recently presented a more detailed correction model for the Berenson analysis [8].

Baumeister and Simon [9] developed a correlation for T_{mfb} based on the maximum superheat of the liquid derived from a thermodynamic analysis. They also introduced the thermal properties of the surface, and in addition, the surface energy which is related to wetting characteristics. The importance of wetting effects was shown in the liquid–liquid minimum film boiling experiments conducted by Henry *et al.* [10]. The differences between the experimental results and the solid–liquid correlation were attributed to the difference in wetting characteristics. Other expressions for T_{mfb} based on thermodynamic maximum superheat arguments have been presented [11, 12].

A hydrodynamic component was introduced to the wetting problem in a set of experiments conducted by Waldram *et al.* [13] and Ochiai and Bankoff [14], in which liquid drops fell onto a hot liquid surface, and the occurrence of film boiling or coalescence was recorded. In modeling these experiments, Bankoff *et al.* [15] found that the film thickness at coalescence time is at least one order of magnitude larger than that required for a coalescence mechanism, such as film instability due to short range forces [16] or free molecule heat conduction [17]. Hence, it was suggested that coalescence is a two-stage process. In the first stage, the vapor is squeezed due to induced hydrodynamic forces or inherent instabilities, and depends mainly on Weber number. Tongues of liquid may then reach the hot surface and contact is established. The stage in which contact is sustained is the second stage in the coalescence process and it depends mainly on the temperatures and physicochemical properties of the two surfaces. The significance of the second stage was demonstrated in dropping experiments [13] in which droplets of diethyl ether and *n*-pentane contacted hot glycerol oil and exhibited heterogeneous nucleation, indicating a contact angle configuration. When those droplets were used in a rising-droplet type of experiments in a glycerol column [18], or when they fell onto a hot silicone oil surface, they nucleated homogeneously, indicating perfect wetting. This difference in behaviors suggests that the second stage in the wetting process is characterized by the removal of a noncondensable adsorbed gas layer from the surface and is controlling [13].

In this paper we discuss the role of the adsorption phenomenon in the second stage and how it relates to the prediction of T_{mfb}.

STATIC CONFIGURATION

Any attempt to analyze the wetting mechanism by continuum theory encounters theoretical difficulties, since the local molecular forces acting on the contact line are the controlling forces. However, in static situations, the contact angle and the spreading coefficient are the classical means of describing the balance of surface tension forces and hence the tendency for wetting.

The system described in Fig. 1 consists of a liquid film in contact with a solid surface covered with an adsorbed film. Initially, when the surface is still clean, the spreading coefficient is defined as the change in free energy per unit area for a liquid spreading over a bare surface:

$$S_{ls} = \gamma_s - (\gamma_{sl} + \gamma_{lv}), \tag{1}$$

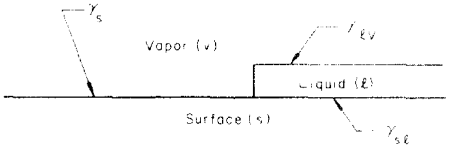

FIG. 1. Initial configuration for wetting.

where γ_s, γ_{sl} and γ_{lv} are the surface tensions of the clean surface, the surface–liquid interface and liquid–vapor interface, respectively. As adsorption proceeds on the exposed surface, the interfacial tension at the solid–vapor, γ_{sv}, is no longer identical to γ_s, but decreases according to the Gibbs equation [19]:

$$d\gamma = -\Gamma RT d \ln P \tag{2}$$

or

$$\pi = -\int d\gamma = RT \int \Gamma d \ln P \tag{3}$$

where Γ is the surface excess per unit area and π is the film pressure.

If S_{ls} is initially negative, equilibrium can be achieved only by change of the configuration of the system to one involving a contact angle such that

$$\gamma_{lv} \cos \alpha = \gamma_{sv} - \gamma_{sl}. \tag{4}$$

Another situation exists when S_{ls} is initially positive. The adsorption may proceed to form a condensed layer (or a complete monolayer) which results in γ_{sv} approaching a limiting value asymptotically ($\gamma_{sv} = \gamma_{sl} + \gamma_{lv}$, which corresponds to a duplex film [20]). The adsorption isotherm in this case is described in Fig. 2. The vapor–surface interface will be unstable with respect to the liquid–surface interface, and complete wetting with positive S_{ls} will occur. However, when the two states (bulk liquid and thin film) are able to coexist, a contact angle is established. In this case the adsorption isotherm must cross the saturation pressure (P_0) line and have an unstable region. A Gibbs integration to the first crossing gives

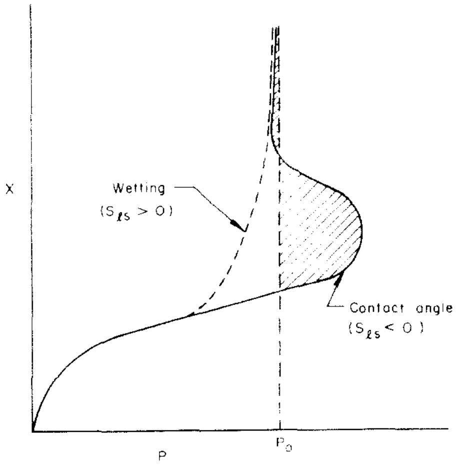

FIG. 2. Variation of adsorbed film thickness with pressure.

$$\pi = \gamma_s - \gamma_{sv} = RT\int_{\Gamma=0}^{\Gamma_0} \Gamma \mathrm{d}\ln P, \tag{5}$$

while that to the limiting condition of an infinitely thick film (liquid state) gives

$$\gamma_s - \gamma_{sl} - \gamma_{lv} = RT\int_{\Gamma=0}^{\infty} \Gamma \mathrm{d}\ln P. \tag{6}$$

The difference between the two integrals is represented by the net shaded area in Fig. 2 and gives the spreading coefficient for the liquid on an adsorbed film:

$$S_{lsv} = \gamma_{sv} - (\gamma_{sl} + \gamma_{lv}) = RT\int_{\Gamma_0}^{\infty} \Gamma d\ln P \tag{7}$$

or

$$\gamma_{lv}(\cos\alpha - 1) = RT\int_{\Gamma_0}^{\infty} \Gamma d\ln P. \tag{8}$$

Following the analysis given by Adamson and Ling [21] the adsorption isotherm (Fig. 2) can be represented by the equation

$$RT\ln\frac{P_0}{P} = \frac{\varepsilon}{(x/d)^3} - b\mathrm{e}^{-ax} \tag{9}$$

and consequently

$$\cos\alpha = 1 + \frac{\varepsilon}{\gamma_{lv}V_l^{2/3}x_0^2}\left(\frac{1}{2} - \frac{1}{ax_0}\right) \tag{10}$$

where ε, a and b are empirical temperature-dependent constants, d is a molecular dimension, V_l is the molecular volume and x is the thickness of the adsorbed film (x_0 corresponds to the first crossing of the P_0 line by the adsorption isotherm). For a positive value of b, a discontinuity is possible and a film of finite thickness x_0 can be in equilibrium with the bulk liquid at $P = P_0$. For this case a finite contact angle is predicted if ax_0 is less than two. When b is negative, wetting always occurs.

DYNAMIC CONFIGURATION

As early as 1936 it was observed that during the spontaneous spreading of a liquid drop on a solid surface, a sensible film of liquid (primary film) spreads from the drop and may be followed by the spreading of the drop itself [22]. It was suggested that the primary film pulls the liquid drop out to form a secondary film which is followed by the bulk liquid. The secondary film motion was explained by the surface tension gradient generated from differences in thickness. The primary film may be formed by deposition of liquid from the vapor alone [22], or by surface diffusion from the drop edge as well as by deposition from the vapor [23].

In another study conducted by Bascom *et al.* [24], the spontaneous spreading of non-polar liquids on smooth, clean metal surfaces was found to involve an unseen protruding lip of liquid, less than 1000 Å in thickness, which is followed by a thicker film. Here also the movement of the secondary film was assumed to result from a surface tension gradient across the transition zone between the primary and secondary films.

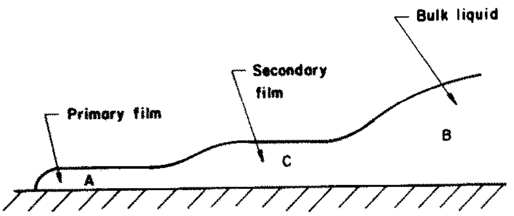

FIG. 3. Illustration of spreading mechanism.

Contrary to the spontaneous spreading experiments described above, Ghiradella *et al.* [25] conducted a forced spreading experiment in which liquid spreading was induced by hydrodynamic forces. The same precursor film was found to exist. Its length varied directly with spreading velocity and could reach distances of the order of a few millimeters ahead of the bulk liquid. Spreading experiments with silicone fluids showed [26] that the precursor film also depends on the contact angle, extending further ahead of the spreading fluid the smaller the contact angle is.

From the above investigations, the picture of spreading is illustrated in Fig. 3. Shortly after the liquid contacts the surface the strong dispersive forces acting on the contact line has caused a thin layer of liquid (A) to move out from the bulk of the liquid (B). Since the effectiveness of the short range forces cannot extend above the surface more than a very few molecular diameters, the layer (A) is quite thin and probably is not a fluid in the normal dimension. Following (A) is a region of liquid (C) sufficiently thick to have complete liquid properties.

The same general configuration, as well as the application of equations (1)–(8), can be extended to non-isothermal systems where cold liquid contacts a hot surface [27]. In the analysis performed by Potash and Wayner [28] and Wayner *et al.* [29] on evaporation from a two-dimensional meniscus, the meniscus was divided into separate regions (Fig. 4) which can be readily identified with the zones described in Fig. 3: the thin film region whose thickness can vary from a monolayer to approximately 20 Å and because of dispersion forces is not evaporating; an evaporating thin film whose thickness ranges approximately from 20 Å to 10^{-5} m, where fluid flow results from very large pressure gradients due to curvature; the bulk liquid.

MODEL

Based on the experimental observations described

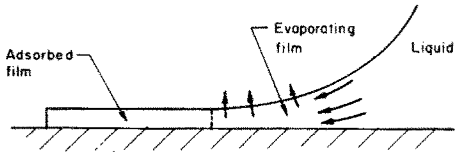

FIG. 4. Illustration of transport processes in an evaporating meniscus.

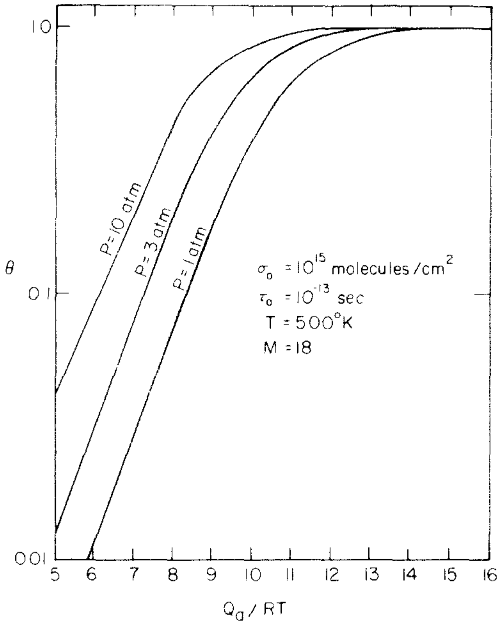

FIG. 5. Adsorption isobars according to Langmuir model.

before we propose that wetting in a non-isothermal system is controlled by a thin film (the primary film, at least monolayer in thickness) that pulls some liquid to form an evaporating meniscus which transfers the energy from the hot surface. As long as the temperature allows the formation of at least one monolayer of liquid molecules on the surface, wetting is possible. When the temperature is increased above a specific value, no continuous monolayer (or closed packed patches of adsorbate) can be formed, and initial spreading will be discontinued. This temperature is postulated to correspond the minimum film boiling.

The simplest temperature dependence of adsorption can be represented by the Langmuir monolayer adsorption model [30], in which the fraction of adsorption sites which are occupied is given by

$$\theta \equiv \frac{\Gamma}{\Gamma_0} = \frac{n\tau}{\Gamma_0 + n\tau} \tag{11}$$

where $n = AP(2\pi MRT)^{1/2}$ is proportional to the mean thermal velocity in the gas phase, and $\tau = \tau_0 \exp(Q_a/RT)$ is a residence time in the adsorbed state. Thus,

$$\theta = \frac{\exp(Q_a/RT)}{k_1 + \exp(Q_a/RT)} \tag{12}$$

where

$$k_1 \equiv \frac{(2\pi MRT)^{1/2}\Gamma_0}{AP\tau_0}. \tag{13}$$

Adsorption isobars are shown in Fig. 5 where the exponential dependency of θ on temperature is clear.

The temperatures which correspond to highly adsorbed surface ($\theta = 0.9$) were calculated from equation (12) and compared with experimental data for solid–liquid and liquid–liquid systems. The results for several solid–liquid systems are presented in Table 1 where the experimental T_{mfb} are the solid–liquid interface temperatures calculated by

$$T_{mfb} = \frac{T_s + \beta T_l}{1 + \beta} \tag{14}$$

where

$$\beta = \left[\frac{(k\rho c_p)_l}{(k\rho c_p)_s}\right]^{1/2} \tag{15}$$

and T_s is the surface temperature. The trend exhibited is clearly correct and the predictions are in fair agreement with the experimental data.

Minimum film boiling temperatures of various liquids in contact with water are given in Table 2. The reduced temperature T_{mfb}/T_{cr} calculation for most liquids are in good agreement with the value 0.9 observed experimentally for perfect wetted liquid–liquid systems.

Table 2. T_{mfb} for liquid–liquid systems*

Cold liquid	T_{cr} (K)	Q_a (Kcal/mole) [35]	T_{mfb}/T_{cr}
Methane	191	1.8	0.906
Ethylene	282	2.6	0.886
Carbon dioxide	304	3.0	0.949
Ethane	305	2.4	0.757
Propane	369	3.1	0.808
i-butane	407	3.8	0.898
n-butane	425	4.6	1.04

*$k_1 = 20$.

Table 1. Comparison between predicted and experimental T_{mfb} (solid–liquid systems)*

System	Q_a (Kcal/mole)	Predicted T_{mfb} (K)	Experimental T_{mfb} (K)
Platinum–water	15.22 [31]	610	640 [32]
Aluminum–water	11.32 [31]	453	490 [9]
Copper–nitrogen	3.5 [30]	120	105 [33]
Any surface–helium	0.1 [30]	4	3.5 [34]

*$k_1 = 3.6 \times 10^4$.

DISCUSSION

It is known that dispersive forces are the controlling forces in adsorption, evaporation, and liquefaction. Hence, it is not surprising that the adsorption potential of gas on various surfaces runs more or less parallel to the ease with which it is condensed to liquid or the difficulty in evaporating the liquid. For the condensation process it was shown that the first step in nucleation process is adsorption of vapor on a solid particle [31]. Ordering materials by their heat of adsorption values agrees with the experimentally determined order of nucleation abilities. Considering the minimum film boiling mechanism, it was shown here that ordering surface–liquid systems by their heat of adsorption agrees with the experimentally determined order of wetting ability.

As the interface temperature exceeds the liquid homogeneous nucleation temperature (T_{hn}), the high rate of nucleation in the bulk liquid, resulting from density fluctuations, will cause immediate termination of contact along the liquid–surface interface, regardless of the adsorption characteristics. This is equivalent to Baumeister and Simon's minimum film boiling correlation in which T_{mfb} approaches T_{hn} when the surface energy is very large.

Under most operating conditions the Leidenfrost temperature for discrete liquid drops is equal to T_{mfb}. However, in the absence of hydrodynamic instabilities the probability of liquid tongues reaching the surface is remote. Thus, it is not surprising that drops may still be in film boiling near the saturation temperature when the surface is cooled down rapidly. However, any mechanical instability which exists in the system (e.g. the method of introducing the drop, tapping, etc.), will immediately induce contact and, consequently, wetting.

In a recent paper Bankoff and Chung [36] proposed that adsorption dependency on temperature may explain the large effective contact angle exhibited when a liquid front is advancing into a hot dry spot. Their argument is based on equation (8) which indicates that Γ decreases (and hence α increases) as the temperature increases at constant system pressure, with a sharp decrease at a temperature threshold determined by the heat of adsorption.

In view of equation (10) a more vigorous explanation may be given. For a static configuration we can evaluate the temperature-dependence of the contact angle from equation (10) provided a good knowledge about the temperature dependence of the parameters ε, a, V_l, and γ_{lv} is obtained. These parameters can be evaluated at various temperatures. Among these quantities, the molecular volume, V_l, is conceivably least affected by temperature. The dispersion potential of the solid surface, ε and γ_{lv} are expected to be linearly dependent on the temperature. If a is also assumed to be weakly dependent on temperature, then in equation (10) the thickness of the adsorbed layer, x_0, can be dominant in determining the temperature effect at high interface temperatures. At some threshold temperature the adsorbed layer becomes a monolayer resulting in very large contact angle, corresponding to a non-wetting system. This result is consistent with the assumption made above regarding the surface coverage at T_{mfb}.

It is recognized that the Langmuir adsorption equation as well as the coverage criteria used to determine a condensed monolayer are highly simplified. Actually, the heat of adsorption for a specific system is a complicated function of surface coverage and temperature, the adsorption isotherms do not always follow the Langmuir equation and the maximum monolayer coverage may be as low as 0.3 [37] due to adsorbent structural perturbation. At present only few experimental values of the adsorption parameters for pertinent systems and temperature regions are all known, and so we regard this model as tentative and subject to experimental verification.

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LE ROLE DE L'ABSORPTION DANS LA DETERMINATION DE LA TEMPERATURE MINIMALE DE L'EBULLITION EN FILM

Résumé—On suggère un mécanisme de l'ébullition en film minimale, basé sur les caractéristiques d'absorption du système. Le processus de mouillage est caractérisé par un film précurseur sans évaporation. Quand le température de la surface augmente, l'épaisseur du film décroît avec une diminution rapide au seuil de température déterminé par la chaleur d'adsorption. La température d'ébullition en film minimale est supposée être celle pour laquelle il existe une couche monomoléculaire. Ces valeurs estimées sont en bon accord avec les résultats expérimentaux pour différents systèmes.

DIE ROLLE DER ADSORPTION BEI DER BESTIMMUNG DER NIEDRIGSTEN TEMPERATUR DES FILMSIEDENS

Zusammenfassung—Vorgeschlagen wird ein Modell, welches das Filmsieden im Leidenfrost-Punkt auf der Grundlage der Adsorptionseigenschaften des Systems beschreibt. Der Benetzungsvorgang wird durch einen vorläufig nicht verdampfenden Film charakterisiert. Mit zunehmender Oberflächentemperatur nimmt die Dicke des Films ab. Besonders ausgeprägt ist die Abnahme der Filmdicke dann, wann die Temperatur erreicht ist, bei der die Adsorptionsenthalpie frei wird. Es wird angenommen, daß bei der kleinsten Temperatur, bei der noch Filmsieden stattfindet, nur noch eine einmolekulare Schicht besteht. Die berechneten Werte stimmen gut mit den an verschiedenen Systemen gemessenen Werten überein.

РОЛЬ АДСОРБЦИИ В ОПРЕДЕЛЕНИИ МИНИМАЛЬНОЙ ТЕМПЕРАТУРЫ ПРИ ПЛЕНОЧНОМ КИПЕНИИ

Аннотация — Предложен механизм кризиса пленочного кипения, в основу которого положены адсорбционные характеристики системы. Процесс смачивания характеризуется наличием неиспаряющейся пленки. По мере возрастания температуры поверхности пленка утоншается, особенно резко при пороговой температуре, определяемой теплотой адсорбции. За минимальную температуру пленочного кипения принимается температура, при которой существует только один монослой. Расчетные значения хорошо согласуются с экспериментальными данными для различных систем.