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ON THE ROLE OF INERT GAS IN INCIPIENT BOILING LIQUID METAL EXPERIMENTS

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Abstract—In experimental studies which measure the amount of liquid superheat required to nucleate the liquid alkali metals, a considerable amount of conflicting and for the most part, unexplained data have been presented. The incipient superheat has been shown or postulated to be affected by the pressure-temperature history, liquid velocity, pressure, heat-flux, liquid purity, dissolved gas content, surface conditions, particle radiation, heating method, length of a time of operation, and perhaps others. This paper presents a theoretical study of the effect of inert gas on incipient superheats and illustrates that among other things, the gas diffusion from surface cavities into the liquid solution accounts for the so-called "heat-flux effect". On the basis of this study, it was determined that almost all of the published data do not include sufficient information to adequately characterize the experimental systems. The non-specification of the inert gas partial pressure in the gas cover blanket, for example, can cause an uncertainty in the incipient superheats as large as 100°C.

NOMENCLATURE

A,	constant appearing in Henry's Law, equation (28);
B,	constant appearing in Henry's Law, equation (28);
C_A,	molar concentration of inert gas in liquid solution;
C_{A1},	molar concentration of inert gas in liquid solution at start of heating;
C_A^*,	molar concentration of inert gas in surface cavities;
$\overline{C_A^*}$,	volumetric average of C_A^*;
C_B,	molar concentration of liquid in liquid solution;
C_B^*,	molar concentration of vapor in surface cavities;
D,	dimensionless diffusivity ratio, $D = D_{AB}/\alpha$;
D_{AB},	mass diffusivity coefficient of inert gas in liquid solution;
D_{AB}^*,	mass diffusivity coefficient of inert gas in vapor;
E,	dimensionless parameter, $E = M_B Z_1/RAT_0 Z \rho_B$;
F,	dimensionless parameter, $F = B/T_0$;
h_{fg},	latent heat of vaporization;
k,	thermal conductivity of liquid;
$K(T)$,	Henry's Law "constant" evaluated at T;
M_B,	molecular weight of liquid;
P_A,	partial pressure of inert gas;
P_{AO},	partial pressure of inert gas in surface cavities when P'_L occurred;
P_{A1}, P_{AG},	partial pressure of inert gas in gas blanket;
P_{A1}^*,	partial pressure of inert gas in surface cavities at start of heating;
$P_A^*, P_A^*(t)$,	partial pressure of inert gas in surface cavities at time t;
P_L,	pressure of liquid;
P'_L,	maximum liquid pressure experienced prior to nucleation;

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$P_v(T)$,	vapor pressure of liquid evaluated at T;
q_w,	wall heat flux;
q'_w,	equivalent wall heat flux, $q'_w = Q'Z$;
Q,	dimensionless wall heat flux, $Q = q_w Z/kT_0$;
Q',	internal energy generation rate;
R,	ideal gas constant;
R_e,	equivalent radius of largest unflooded cavity;
T,	liquid temperature;
T_c,	temperature of gas and vapor in surface cavities;
T'_L,	temperature of liquid when P'_L occurred;
T_{sat},	saturation temperature corresponding to P_L;
T_w, $T_w(t)$,	wall temperature at time t;
T_0	initial liquid temperature;
t,	time;
x,	vertical coordinate;
x_A,	mole fraction of inert gas in liquid solution;
Z,	liquid metal pool depth;
Z_1,	nominal surface cavity depth;
Z_2,	length defined as $Z_2 = Z + Z_1$.

Greek symbols

α,	thermal diffusivity;
ζ,	dimensionless coordinate defined in equation (22a);
θ, $\theta(\zeta, \tau)$,	dimensionless temperature difference defined in equation (22c);
ρ_B,	density of liquid;
σ,	surface tension evaluated at T;
τ,	dimensionless time defined in equation (22b);
ϕ, $\phi(\zeta, \tau)$,	dimensionless concentration difference defined in equation (22d).

INTRODUCTION

THE STUDY of boiling alkali metals has been primarily directed toward safety studies of liquid-metal-cooled, fast-breeder reactors ("LMFBR"). In these reactor systems, the normally highly subcooled liquid metal could reach boiling temperatures during postulated accidents of loss of coolant flow or sudden reactor power increases. The boiling may cause serious problems since certain regions of the reactor have positive void coefficients of reactivity, i.e. the introduction of voids (i.e. vapor) can cause local increases in reactor power, and possibly result in fuel melting.

Therefore, in order to properly design instrumentation for initiating reactor shutdown to avoid damage, or to postulate realistic theoretical models which predict the extent of damage, it is necessary to be able to predict the conditions under which boiling will occur.

In the past several years, a number of experiments have been reported in which incipient boiling superheats of sodium and postassium were measured. However, the data are conflicting in certain aspects and are also widely disparate, ranging from about 5 to 820°C. A large number of parameters were determined or postulated to affect the incipient boiling superheat among which are the following: (1) pressure-temperature history, (2) pressure, (3) heat-flux, (4) liquid purity, (5) dissolved gas content, (6) surface conditions, (7) nuclear radiation, (8) heating method, (9) length of time of operation, (10) liquid velocity, and perhaps others. The roles of these parameters will be now briefly discussed.

(1) *Pressure-temperature history*

A detailed study of this effect has been presented in [1] and qualitatively confirmed by experiments reported in [2–4]. The model describing this effect asserts that a liquid metal would penetrate surface cavities to a depth determined by the system pressure and temperature; furthermore, scale and/or oxide coatings on the surfaces of the cavity will be cleaned by the liquid metal up to the penetration depth, thus creating a wetted condition. Beyond this depth, the cavity will remain non-wetted.

Thus, if the maximum pressure experienced by the system was P'_L at a temperature of T'_L, all cavities with radii greater than R_e will be flooded and wetted, where

$$R_e = \frac{2\sigma(T'_L)}{P'_L - P_v(T'_L) - P_{A0}}. \quad (1)$$

Nucleation must now occur from cavities with radii less than R_e, so that the maximum excess vapor pressure in a cavity required for nucleation is

$$P_v(T_w) - P_L = \frac{2\sigma(T_w)}{R_e} - P_A^*. \quad (2)$$

The maximum superheat requirements may be directly calculated from the excess pressure by using vapor pressure data or the Clausius-Clapeyron equation. Using the latter approximate approach, one can obtain the following relationship from equation (2), (see, for example, [5]):

$$T_w - T_{sat} \cong \frac{2R(T_{sat})^2\sigma}{P_L h_{fg} R_e} \quad (3)$$

for $P_A^* = 0$ and constant physical properties.

It is immediately obvious from equations (1) and (2) that a highly pressurized system (prior to nucleation) will require larger superheats than will a lower pressurized system.

(2) *Pressure*

As the system pressure at nucleation, P_L, is increased, the incipient superheat should decrease, as illustrated by equation (3). This result has been observed in almost all of the available experimental data.

(3) *Heat flux*

Data have been presented indicating that the incipient superheat may be decreased [16], increased or decreased [7], increased [2] or unaffected [8, 9] by an increase in the heat flux. None of these reports have presented a satisfactory explanation for this effect, however, it will be shown in the present paper that the heat flux effect results from inert gas leaving surface cavities during heating. Also, an erroneous effect may be observed experimentally, caused by very slight errors in thermocouple location. This possibility is discussed in the Appendix.

(4) *Purity and dissolved gas content*

The extremely large superheat values reported in [8] were attributed primarily to a very efficient purification of the liquid metal prior to testing. The importance of the dissolved gas content will be explored in the present paper. It must be pointed out that most reported experiments do not include information on either the liquid purity or the dissolved gas content. This information is quite essential if the data from various experiments are to be realistically compared or applied.

(5) *Surface conditions*

The effect of surface roughness on incipient boiling superheats has been studied [10] and it was concluded that roughening a surface will lower the superheat. Also, the spread in the data reported in [9] is explained as being compatible with the surface condition of the experimental tube. However, the equivalent cavity model [1] states that the available nucleation sites are determined by the pressure–temperature history rather than surface roughness. This has been experimentally confirmed in a preliminary report [17].

(6) *Nuclear radiation*

In a recent theoretical study of this possible effect [11] it was concluded that it would be highly unlikely that, at saturation pressures near one atmosphere, any nuclear radiation encountered would induce nucleation in the bulk of the liquid sodium before boiling would occur at surface cavities. The only experimental data known to the present authors, [17], confirms this conclusion at thermal neutron fluxes up to 10^{11} neutrons/cm^2 s.

(7) *Heating method*

Boiling liquid metal experiments have been conducted using two general types of heating methods: (1) indirect (conduction through a channel wall to the liquid), and (2) direct (Joule heating of the channel wall and liquid metal). The latter technique has resulted in somewhat larger superheats than the former at equivalent heating rates which can be explained in part by larger inert gas solubilities caused by the inverted liquid temperature profile (i.e., direct heating results in the liquid temperature being larger than the wall temperature while indirect heating causes the opposite condition). This phenomenon is discussed further in this paper.

(8) *Length of time of operation*

Several experimenters have observed that the incipient superheat generally increases with the length of time of operation of the apparatus [8, 9] while others have not reported any such variation [6, 12]. This so-called aging effect may be caused by the gradual wetting of the larger nucleation sites, leaving smaller and smaller cavities available for nucleation. If this explanation is correct, it appears that the equivalent cavity model of [1] should be able to predict this behavior by noting the pressure-temperature history during operation.

(9) *Liquid velocity*

The results of several experiments have been recently published [18, 19] in which the forced convection incipient boiling conditions have been measured. The preliminary indications are that the superheat decreases markedly with increasing velocity, approaching zero superheat at about 2 m/s. The mechanism responsible for this effect is not at all clear, but might be explained by the presence of minute inert gas bubbles in the liquid stream. These bubbles could originate either in the heat exchanger of a loop where the decreasing liquid temperature tends to drive inert gas out of solution or from entrainment from the gas blanket region. If these bubbles could then reach the heated section of a loop, they would tend to re-dissolve as the liquid temperature is increased. However, the time available for dissolution is the transit time of the fluid in the heated section; thus, as the liquid velocity is increased, the probability of gas bubbles existing at the heater exit (where nucleation is most likely to occur) is increased. The presence of these bubbles will, of course, tend to reduce the incipient superheat to zero.

Even though these speculations seem to explain the reported results, additional experimental data is required before any final conclusions can be reached on this important point for nuclear reactor safety studies.

In this paper, the role of inert gas in liquid metal, pool boiling experiments will be theoretically studied, and it will be shown that the heat flux effect results from inert gas diffusion. It will be also shown that not only the heat flux, but the initial system temperature and inert gas partial pressure must be specified in order to characterize an experimental system.

MODEL DEVELOPMENT

In order to predict the conditions under which a liquid metal will nucleate following some prescribed pressure-temperature history, a value of T_w must be chosen so that equation (2) is satisfied. However, during a heating transient, not only does T_w vary, but also P_A^* (the partial pressure of inert gas in surface cavities) must vary as gas leaves or enters surface cavities. In order to determine the variation of P_A^* with time, the solubility of inert gas in the liquid metal must be known as a function of pressure and temperature and the mass diffusion coefficient of the inert gas in the liquid metal must also be available. However, data are extremely sparse for the former and non-existent for the latter property. Some solubility data for argon in liquid sodium at temperatures up to 530°C are available [13] and are extrapolated for use in the present work. The diffusivity of argon in liquid sodium was estimated using an empirical relationship based on the Stokes–Einstein equation [14].

Since the partial pressure of gas in equilibrium with a dilute solution of the gas in a liquid is related to the concentration in the liquid by

Henry's Law, the concentration of the gas must be known as a function of time in order to determine the gas partial pressure. Thus, a solution of the coupled heat- and mass-transfer problem in a liquid metal pool during a heating transient must be found.

To this end, a mathematical description of a simplified physical model is formulated, the solution of which yields the transient wall temperature and the inert gas concentration and partial pressure. A sketch of the physical model is shown in Fig. 1.

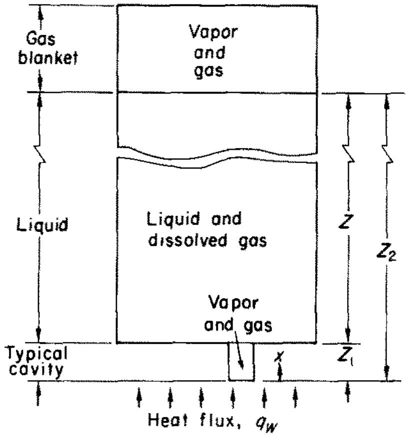

FIG. 1. Simplified physical model.

The conservation equations for the three zones may be written assuming a one-dimensional system with molecular diffusion the only mode of heat and mass transfer:

Gas Blanket

$$P_{AG} = P_{A1} \text{ (Constant).} \tag{4}$$

Liquid solution

$$\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial x^2} \tag{5}$$

$$\frac{\partial C_A}{\partial t} = D_{AB} \frac{\partial^2 C_A}{\partial x^2}. \tag{6}$$

Vapor cavity

$$T_c = T(L_1, t) \tag{7}$$

$$\frac{\partial C_A^*}{\partial t} = D_{AB}^* \frac{\partial}{\partial x}\left[(C_A^* + C_B^*)\frac{\partial}{\partial x}\left(\frac{C_A^*}{C_A^* + C_B^*}\right)\right] \tag{8}$$

where the simplified form of Fick's Law was used in equation (6) but not in equation (8) since $C_A^* + C_B^*$ is not constant.

Furthermore, the partial pressure of the inert gas is related to the mole fraction of the dissolved gas in the liquid at the vapor–liquid interface by Henry's Law.

$$x_A = K(T)\,P_A \tag{9}$$

where x_A is the mole fraction of A in the solution. Thus, the concentration of the gas at $x = Z_2$ is

$$\frac{C_A(Z_2, t)}{C_A(Z_2, t) + C_B(Z_2, t)} = K[T(Z_2, t)]\,P_{A1}. \tag{10}$$

However, $C_B(Z_2, t) \gg C_A(Z_2, t)$ and noting that

$$C_B = \rho_B / M_B. \tag{11}$$

equation (10) can be simplified to

$$C_A(Z_2, t) = \rho_B P_{A1} K[T(Z_2, t)]/M_B. \tag{12}$$

Also, the partial pressure of the inert gas in the cavity can be related to the concentration of the gas in the liquid at $x = Z_1$,

$$P_A^* = \frac{C_A(Z_1, t)\,M_B}{\rho_B K[T(Z_1, t)]}. \tag{13}$$

Finally, the initial and boundary conditions to be used with equation (5) through (8) describing the transient following a step-change in heat flux are

$$t = 0: \quad T(x, 0) = T_0; \quad C_A(x, 0) = C_{A1}; \quad P_A^* = P_{A1}^* \qquad \text{(14a, b, c)}\dagger$$

$$x = Z_2: \quad \frac{\partial T(Z_2, t)}{\partial x} = 0; \quad C_A(Z_2, t) = \rho_B P_{A1} K[T(Z_2, t)]/M_B \qquad \text{(15a, b)}$$

$$x = Z_1: \quad \frac{\partial T(Z_1, t)}{\partial x} = -\frac{q_w}{k}; \quad D_{AB}\frac{\partial C_A}{\partial x}$$

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† Where $C_{A1} = \rho_B P_{A1}^* (K(T_0)/M_B$.

$$= D^*_{AB}(C^*_A + C^*_B)\frac{\partial}{\partial x}\left(\frac{C^*_A}{C^*_A + C^*_B}\right) \qquad (16a, b)$$

$$x = 0: \quad \frac{\partial}{\partial x}\left(\frac{C^*_A}{C^*_A + C^*_B}\right) = 0. \qquad (17)$$

The mathematical description of the physical model is complete and well posed and a method of solution must now be sought.

METHOD OF SOLUTION

Since typical surface cavities are extremely small relative to other system dimensions, it would be more meaningful to work with the volumetric average concentration of the inert gas in the cavity rather than local values. This volumetric average can be defined as

$$\overline{C^*_A}(t) = \int_v C^*_A \, \mathrm{d}V \Big/ \int_v \mathrm{d}V = \frac{1}{Z_1}\int_0^{Z_1} C^*_A(x, t)\, \mathrm{d}x. \qquad (18)$$

Integration of equation (8) with respect to x over $(0, Z_1)$ and use of the conditions (16b) and (17) results in

$$\frac{\partial \overline{C^*_A}}{\partial t} = \frac{D_{AB}}{Z_1}\left(\frac{\partial C_A}{\partial x}\right)_{x = Z_1} \qquad (19)$$

If the inert gas in the cavity is assumed to be ideal, then

$$\overline{C^*_A} = \frac{P^*_A}{RT(Z_1, t)} \qquad (20)$$

where P^*_A is given by equation (13), and equation (19) can be transformed into

$$\frac{\partial C_A}{\partial x} = \left(\frac{M_B Z_1}{R\rho_B D_{AB}}\right)\frac{\partial}{\partial t}\left[\frac{C_A}{TK(T)}\right] \text{at } x = Z_1. \qquad (21)$$

Finally, if the following non-dimensional quantities are defined as

$$\zeta = \left(\frac{Z_2 - x}{Z}\right), \quad \tau = \frac{\alpha t}{Z^2}, \quad \theta(\zeta, \tau) = \left(\frac{T - T_0}{q_w Z/k}\right), \quad \phi(\zeta, \tau) = \frac{C_A - C_{A1}}{C_{A1}} \qquad (22a, b, c, d)$$

the partial differential system describing the transient heat and mass transfer in a pool of liquid metal and dissolved inert gas following a step change in the wall heat flux is

$$\frac{\partial \theta}{\partial \tau} = \frac{\partial^2 \theta}{\partial \zeta^2}, \quad \frac{\partial \phi}{\partial \tau} = D\frac{\partial^2 \phi}{\partial \zeta^2} \qquad (23a, b)$$

$$\tau = 0: \quad \theta = \phi = 0 \qquad (24a, b)$$

$$\zeta = 0: \quad \frac{\partial \theta}{\partial \zeta} = 0, \quad \phi = \exp.\left(\frac{QF\theta}{1 + Q\theta}\right) - 1 \qquad (25a, b)$$

$$\zeta = 1: \quad \frac{\partial \theta}{\partial \zeta} = 1 \qquad (26)$$

$$\zeta = 1: \quad \frac{\partial \phi}{\partial \zeta} = -\left(\frac{E}{D}\right)\frac{\partial}{\partial \tau}\left[\left(\frac{1 + \phi}{1 + Q\theta}\right)\exp.\left(\frac{F}{1 + Q\theta}\right)\right] \qquad (27)$$

where Henry's Law "constant", $K(T)$, has been expressed as [12]

$$K(T) = A\exp.\left(-\frac{B}{T}\right) \qquad (28)†$$

and the symbols are defined in the Nomenclature.

An observation of equation (23a) through (27) reveals that although the dimensionless concentration, $\phi(\zeta, \tau)$, depends upon the dimensionless temperature, $\theta(\zeta, \tau)$, the system describing $\theta(\zeta, \tau)$ can be solved independently of $\phi(\zeta, \tau)$. An expression for $\theta(\zeta, \tau)$ can be obtained through a straightforward application of the Laplace transform and the details of which will not be shown here.‡ The form chosen for most rapid convergence for small values of τ is the following:

$$\theta(\zeta, \tau) = 2\sqrt{\left(\frac{\tau}{\pi}\right)}\sum_{n=0}^{\infty}\left\{\exp.\left[-\frac{(2n + 1 - \zeta)^2}{4\tau}\right]\right.$$

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† In the argon-sodium system for 603°K < T < 803 K, $A = \exp(-4{\cdot}9045)\ \text{atm}^{-1}$ and $B = 10\,458.$°K.

‡ See, for example, [20].

$$+ \exp.\left[-\frac{(2n+1+\zeta)^2}{4\tau}\right]\Bigg\}$$

$$- \sum_{n=0}^{\infty}\left[(2n+1-\zeta)\operatorname{erfc}\left(\frac{2n+1-\zeta}{2\sqrt{\tau}}\right)\right.$$

$$\left. + (2n+1+\zeta)\operatorname{erfc}\left(\frac{2n+1+\zeta}{2\sqrt{\tau}}\right)\right] \quad (29)$$

where erfc (x) is the complimentary error function,

$$\operatorname{erfc}(x) = \frac{2}{\sqrt{\pi}}\int_x^{\infty} e^{-u^2}\,du. \quad (30)$$

The problem which now remains is the solution of equations (23b), (24b), (25b), and (27), using the expression for $\theta(\zeta, \tau)$ from equation (29). An analytical solution to this system could not be obtained and a numerical technique was used. The technique chosen was the Method-of-Lines (or differential-difference) used in conjunction with a fourth-order Runge–Kutta. The results of the numerical computation was a tabulation of the function $\phi(\zeta, \tau)$ for specified values of the parameters D, E, F, and Q†. This function could then be used with the nucleation model to predict incipient superheats.

CALCULATION OF INCIPIENT BOILING SUPERHEATS

In this paper, the equivalent cavity model is used to predict the maximum cavity size available for nucleation. Thus, P_L', T_L', and P_{AO} must be specified in order to determine R_e from equation (1). Then the wall heat flux, q_w, the initial temperature, T_0, the pool depth, Z, and the nominal cavity depth, Z_1, must be specified in order to determine the parameters D, E, F, and Q.† Knowing these parameters, the functions $\theta(\zeta, \tau)$ and $\phi(\zeta, \tau)$ can be computed and the wall temperature and argon concentration at the liquid-vapor interface in the cavity can be obtained using equations (22c) and (22d) as follows:

$$T_w(t) = T_0 + \frac{q_w Z}{k}\theta(1, \tau) \quad (31)$$

$$C_A(Z_1, t) = C_{A1}[1 + \phi(1, \tau)] \quad (32)$$

and the transient partial pressure of inert gas in the cavity can be computed using equations (13), (32), and (31):

$$P_A^*(t) = \frac{M_B C_A(Z_1, \tau)}{\rho_B K[T_w(t)]}. \quad (33)$$

Using these relations, (31), (32) and (33), the incipient boiling criterion, from equation (2), can be expressed as

$$P_v[T_w(t)] - P_L = \frac{2\sigma[T_w(t)]}{R_e} - P_A^*(t). \quad (2a)$$

Calculations are performed for increasingly larger values of time, t, until equation (2a) is satisfied. At this time, the value of T_w is known, and the superheat required for boiling inception is directly calculated as $T_w - T_{sat}$.

In summary, the following nine quantities must be prescribed in order to predict an incipient boiling superheat: P_L', T_L', P_{AO}, q_w, T_0, Z, Z_1, P_{A1}^*, and P_L. It is noted here that usually only P_L and q_w are specified in data presented in the literature, leaving a serious problem in forming a complete specification of the experimental conditions.

DISCUSSION OF RESULTS

Since the pressure-temperature history effect on incipient boiling superheats has been previously discussed [1], all calculations presented in this paper were performed for a fixed value of R_e (i.e. by specifying P_L', T_L, and P_{AO}). There still remains six parameters which can be varied; however, the pool depth, Z, and the nominal surface cavity depth, Z_1, were held constant, and the four remaining parameters (q_w, T_0, P_{A1}^*, P_L) were systematically studied.

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† Sodium physical properties were based on [15] and the mass diffusion coefficient was calculated using the information in [14]. These parameters are defined in the Nomenclature.

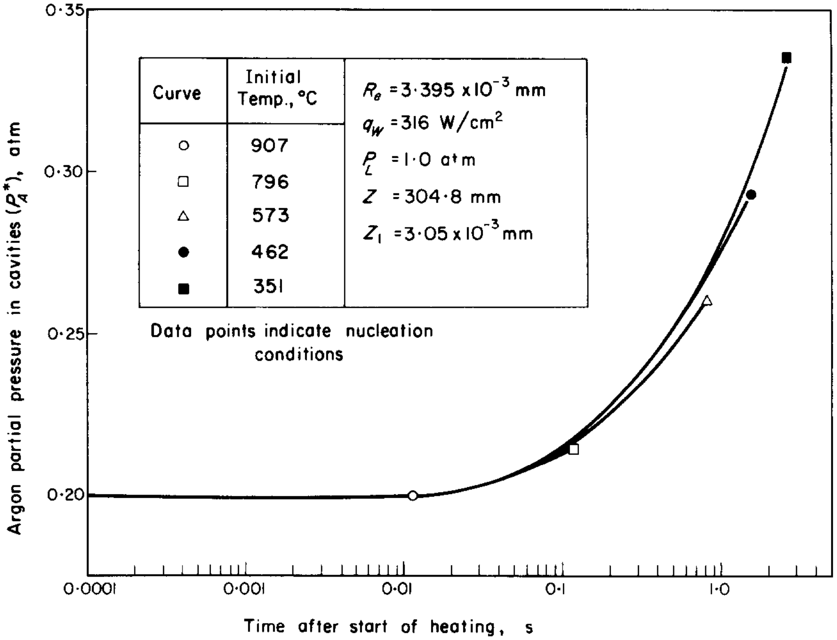

FIG. 2. Effect of initial temperature on the argon partial pressure in surface cavities for q_w = 316 W/cm^2.

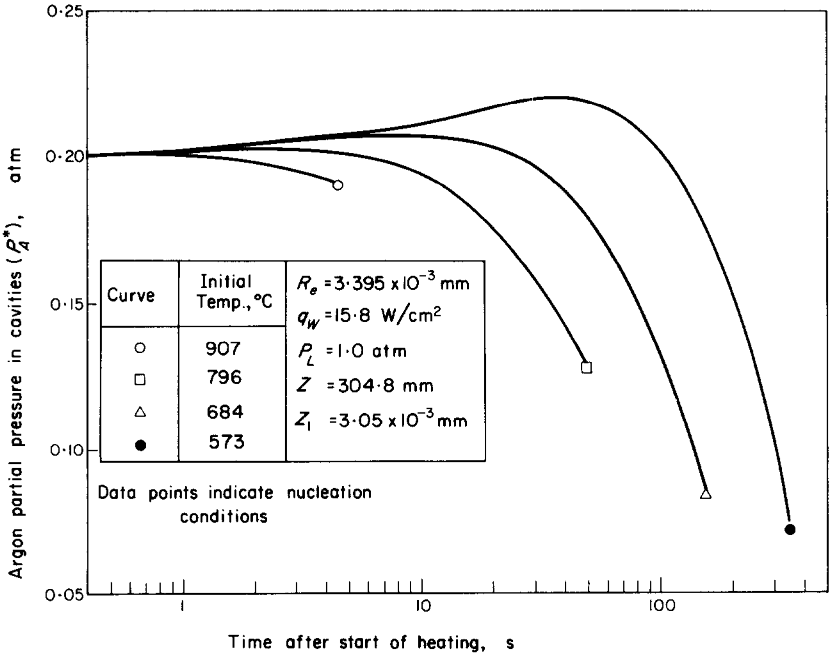

FIG. 3. Effect of initial temperature on the argon partial pressure in surface cavities for q_w = 15·8 W/cm^2

The two primary results of the analysis are the transient inert gas partial pressure in the surface cavities, $P_A^*(t)$, and the incipient boiling superheat requirement $(T_w - T_{sat})$, and these are discussed in some detail. The calculations were performed over the following parameter ranges: $15·8 \leqslant q_w \leqslant 316$ W/cm^2, $350 \leqslant T_0 \leqslant 910$°C, $0 \leqslant P_{A1}^* \leqslant 0·90$ atm, and $0·2 \leqslant P_L \leqslant 2·0$ atm.

The effect of the initial liquid temperature, T_0, on the transient inert gas partial pressure in the surface cavities, $P_A^*(t)$, is illustrated in Figs. 2 and 3. The high flux behavior ($q_w = 316$ W/cm^2) is shown in the former figure, while the low heat flux behavior ($q_w = 15·8$ W/cm^2) is shown in the latter figure. The data points in both figures indicate the conditions at which nucleation occurred and the calculations terminated.

In Fig. 3, P_A^* increases at first from its initial value of 0·20 atm, and then decreases until nucleation occurs. This behavior can be explained by examination of the mechanisms which act to change P_A^*; the increase in the gas temperature tends to increase P_A^* (see equation 20) and the solution of the gas into the liquid tends to decrease P_A^*. These two mechanisms are in opposition to one another, and whether there is a net increase or decrease in P_A^* depends primarily upon the length of time available between the start of heating and nucleation. This length of time is essentially controlled by the heat flux and the initial temperature. Thus, it is seen that the primary effect of decreasing the initial temperature is to increase the length of time available for the two aforementioned mechanisms to operate. For large heat fluxes, the decrease of the initial temperature results in an increase in the value of P_A^* at nucleation, while for small heat fluxes, a decrease of T_0 results in a decrease in the value of P_A^* at nucleation.

Using the computed values of $P_A^*(t)$ in conjunction with $T_w(t)$, the incipient boiling superheat can be evaluated using equation (2a). The results are shown in Figs. 4 and 5 for the low and high heat flux conditions at system pressures of 1·0 and 0·2 atm, respectively. These results reflect the behavior of $P_A^*(t)$ discussed previously and exhibit an increase, a decrease, or an increase followed by a decrease in the incipient superheat

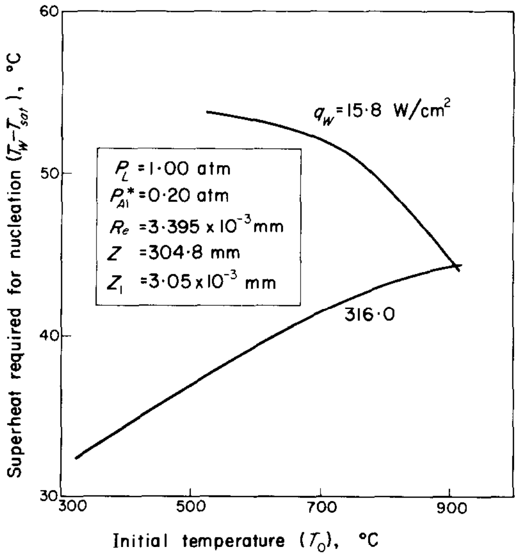

FIG. 4. Effect of initial temperature on the incipient boiling superheat for sodium at a system pressure of 1·00 atm.

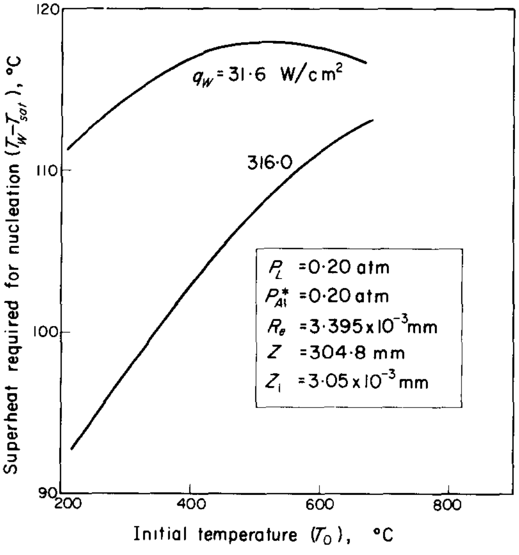

FIG. 5. Effect of initial temperature on the incipient boiling superheat for sodium at a system pressure of 0·20 atm.

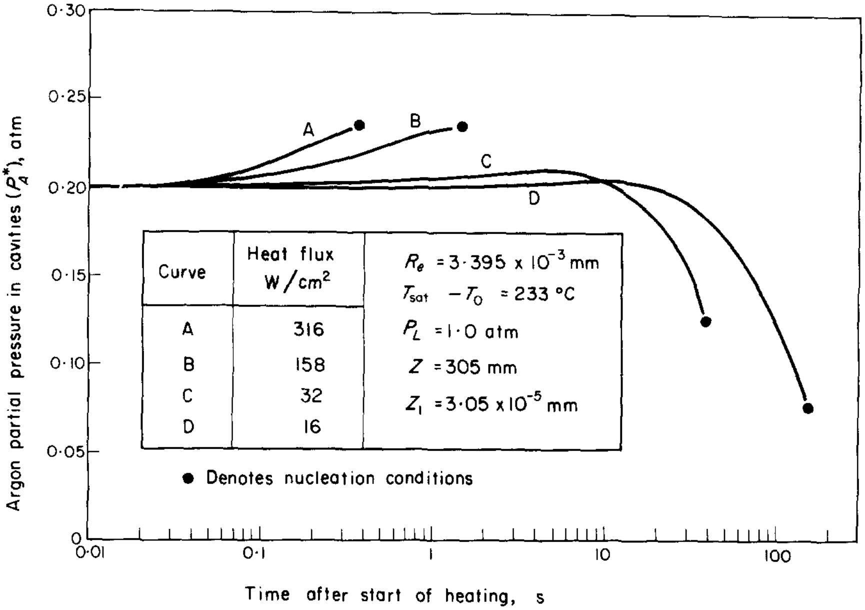

FIG. 6. Effect of heat flux on the argon partial pressure in surface cavities.

as the initial temperature is increased, depending upon the values of the heat flux and the system pressure.

The effect of the heat flux, q_w on the transient partial pressure of inert gas in the surface cavities, is illustrated in Fig. 6. The general variation of P_A^* with the heat flux is similar to its dependence on the initial temperature, except in this case, a smaller value of q_w results in a longer length of time between the start of heating and nucleation. Thus, the value of P_A^* at nucleation decreases with decreasing heat flux. This results in the incipient superheat decreasing with increasing heat flux as shown in Fig. 7.

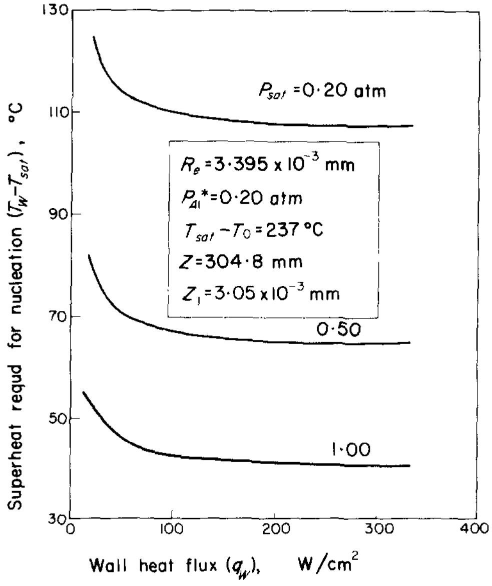

FIG. 7. Effect of heat flux on incipient boiling superheats for sodium.

This heat flux dependency agrees qualitatively with the experimental data published in [16]; a quantitative comparison could not be made since the experimental conditions in [16] were not sufficiently specified. The experimental data reported in [2] and [3] indicated an increase in the incipient superheat with an increase in the heat flux, which is in apparent conflict with the theoretical results presented in this paper. However, the data in [2] and [3] were limited to very low heat fluxes ($q_w \lesssim 7$ W/cm²) and significant natural convective flow resulted during the heating transients as indicated by relatively large (~10–30°C) temperature oscillations prior to boiling. Since the model presented in this paper does not take into account natural convection effects, the present analysis cannot be directly compared to these low heat flux data.

Another factor to be aware of in the comparison of the present analysis to experimental results is the importance of the length of time between successive incipient boiling runs. If this time is short relative to the length of time for the experimental system to reach mass and thermal equilibrium (i.e. for the inert gas to diffuse back into the surface cavities), then the initial inert gas partial pressure, P^*_{A1}, would tend to decrease from experiment to experiment in an uncontrolled manner. Therefore, in a series of experiments in which only the heat flux is externally varied, and the time between runs is short in the sense described previously, the incipient boiling superheat would tend to increase; this increase would occur with either an increase or decrease in the heat flux.

The importance of including the variation of P^*_A in calculating incipient boiling superheats is further illustrated in Fig. 8. The ratio of the superheat calculated using equation (2a) with the variable $P^*_A(t)$ to that calculated using P^*_A constant at its initial value is plotted vs. the wall heat flux for several different system pressures. It is clear that a 20 to 30 per cent error in the incipient superheat can result in the heat flux range of about 10 to 30 W/cm^2 if a constant P^*_A is assumed. For large heat fluxes ($q_w \gtrsim 200$ W/cm^2), the error approaches 10 per cent, and for an intermediate heat flux range ($50 \gtrsim q_w \gtrsim 100$ W/cm^2), the error is only about 5 per cent.

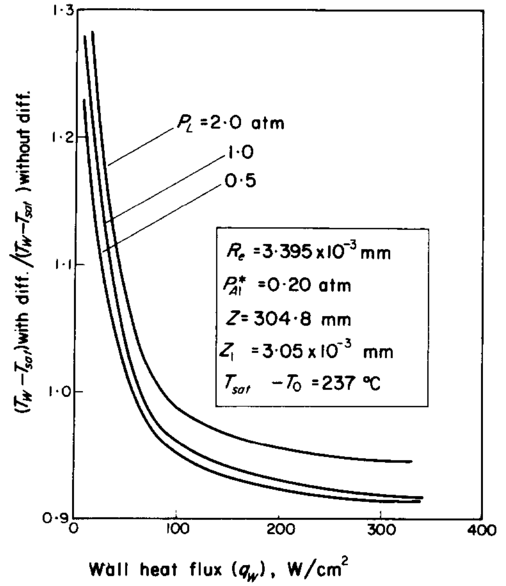

FIG. 8. Effect of heat flux on the ratio of the superheat with argon diffusion to that with no argon diffusion for several system pressures.

From the analysis, it is clear that the initial value of the inert gas partial pressure should affect the incipient boiling superheat. Indeed, it is an extremely important parameter as shown in Fig. 9. Unfortunately, previously published experimental work does not include a value of this parameter. For the case of no inert gas present ($P^*_{A1} = 0$), the superheat is independent of q_w (and also all other parameters with the exception of P'_L, T'_L, and P_L) as shown in Fig. 9. For sufficiently large values of P^*_{A1}, boiling inception is shown to occur at negative superheats (below the saturation temperature), however, in

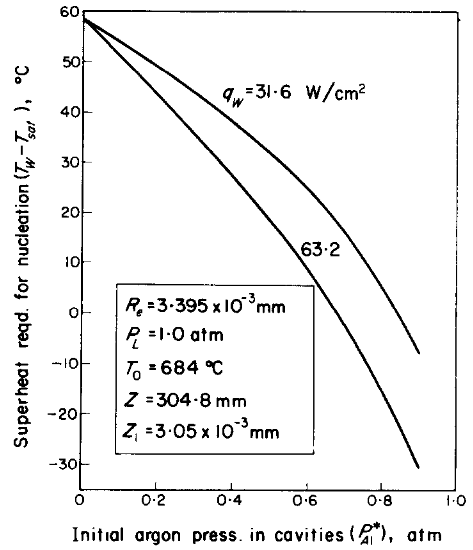

FIG. 9. Effect of the initial argon partial pressure in surface cavities on the incipient boiling superheat for sodium.

practice, boiling would probably occur at zero superheat.

The system of equations describing the dimensionless inert gas concentration (equations 23b, 24b, 25b, and 27) indicate that $\phi(\zeta, \tau)$ depends upon the temperature distribution in the liquid. Thus, since the mode of heating (direct or indirect) will affect the temperature field, it should also affect the incipient boiling superheat. For the purposes of comparison, the transient temperature field resulting from a sudden application of a uniform internal energy source (direct heating) is derived in the Appendix. Using this transient temperature in conjunction with the mass diffusion equations and the incipient boiling criterion (equation 2a), the incipient superheat resulting from direct heating can be compared to that resulting from indirect heating. These results are shown in Fig. 10 for a wide range of heat fluxes. The predicted behavior is in qualitative agreement with available data in that the incipient superheat will be larger in a system that is heated directly.

Perhaps a different relationship between the internal energy generation rate and an equivalent wall heat flux than is usually used can be postulated in order to account for the liquid temperature variation and to more closely compare data from experiments using these two heating techniques.

An attempt was made to compare the theoretical predictions of the present model to the experimental results presented in [7]. During the course of these calculations, an interesting effect caused by the thin liquid layer above the heater ($Z \cong 60$ mm) was noted. It was found that a very slight change in the initial inert gas partial pressure (P^*_{A1}) could cause a large change in the incipient boiling superheat. An observation of the transient cavity pressure, $P^*_A(t) + P_v(T_w)$, as shown in Fig. 11, clearly illustrates the reason for this phenomena.

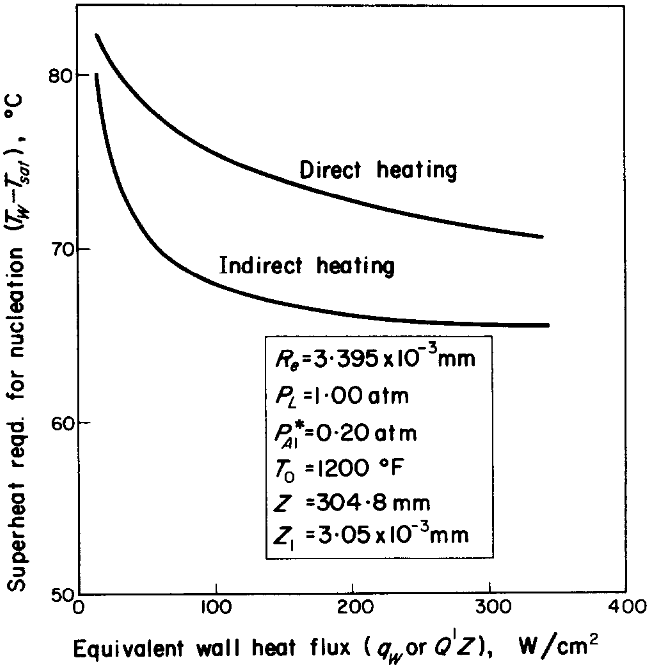

FIG. 10. Effect of the method of heating on the incipient boiling superheat for sodium.

It has been shown previously that the inert gas partial pressure in the cavities, P_A^*, increases at first due to the increasing gas temperature, and then decreases due to solution of the gas in the liquid, while the vapor pressure, $P_v(T_w)$, steadily increases. For very thin liquid layers (small Z), the gas temperature would increase more rapidly than for thick layers at the same heat flux, resulting in a more rapid increase in P_A^*. Thus, if the initial value of P_A^* was sufficiently large, nucleation could occur during this initial rise; however, if P_A^* was slightly smaller, nucleation would be delayed until the vapor pressure had increased sufficiently, resulting in a much larger incipient superheat.

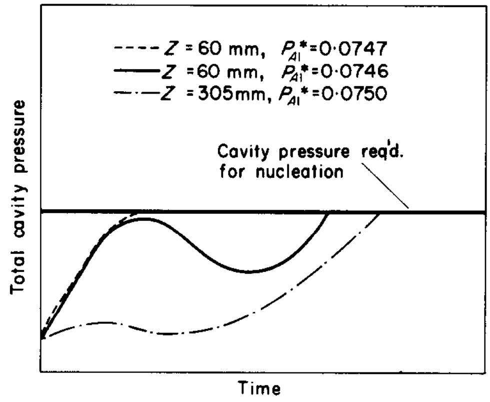

FIG. 11. Transient total cavity pressure.

Due to this extreme sensitivity to the initial inert gas partial pressure and the lack of this information as well as the pressure–temperature history in [7], any quantitative comparison was not felt to be meaningful.

SUMMARY AND CONCLUSIONS

In this paper, the role of inert gas diffusion during incipient boiling experiments with liquid metals has been theoretically analyzed. Detailed calculations have been presented for the argon–sodium system for a wide variety of parameters. It has been shown that the initial liquid temperature, heat flux, initial inert gas partial pressure, and heating technique can significantly affect the incipient boiling superheat. The second and last of these effects have been experimentally observed and this paper presents the first theoretical basis for their existence. The effect of heat flux on the incipient boiling superheat has been shown to be caused by inert gas diffusion out of surface cavities; the analysis clearly indicates a decrease in superheat with an increase in heat flux for a series of experiments in which *only* the heat flux is varied. However, it was also shown that the opposite behavior can result (i.e., an increase in superheat with an increase in heat flux) if the series of experiments are conducted in such a manner that the proper initial conditions are not attained prior to each run.

On the basis of this paper, it is urged that all future incipient boiling data include sufficient information to characterize the experiment, i.e. the following minimum number of parameters must be specified: P'_L, T'_L, P_{AO}, P_L, T_0, P^*_{A1}, q_w, Z and Z_1. The concentration of dissolved impurities should also be given. Furthermore, extreme caution must be used in the interpretation of reported effects on the incipient boiling superheats of liquid metals.

Finally, data on the solubility of inert gases in liquid metals at temperatures larger than 600°C and the diffusion coefficient in similar systems must be obtained in order to quantitatively predict the incipient boiling superheat.

ACKNOWLEDGEMENTS

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APPENDIX

Calculation of Transient Temperature for Directly Heated Liquid

If the liquid is heated directly by an internal energy source of strength Q' W/cm^3, the transient temperature is described by

$$\alpha k \frac{\mathrm{d}T}{\mathrm{d}t} = Q'; \qquad T(0) = T_0$$

which has the solution

$$T = T_0 + \frac{Q't}{\alpha k}.$$

Q can be related to an equivalent wall heat flux, q'_w, by

$$q'_w = Q'Z$$

so that in terms of the dimensionless quantities previously defined,

$$\theta(\tau) = \tau.$$

Prediction of Diffusivity Coefficient of Argon in Liquid Sodium

The empirical relationship used for calculating D_{AB} was proposed by Wilke and Chang [14] and is valid for small concentrations of A in B:

$$D_{AB} = 7{\cdot}4 \times 10^{-8} \frac{(\psi_B M_B)^{\frac{1}{2}} T}{\mu \tilde{V}_A^{0{\cdot}6}}$$

where $\tilde{V}_A$ is the molar volume of A in cm^3/g-mole as liquid at its normal boiling point, μ is the viscosity of the solution in centipoises, ψ_B is an "association parameter" taken to equal 1·0 for an unassociated solvent, M_B is the molecular weight of B, and T is the absolute temperature in °K. It is claimed that this equation is accurate within ±10 per cent for dilute solutions of nondissociating solutes, however, there apparently are no experimental data on the argon–sodium system to compare with this prediction.

Therefore, several calculations were performed to determine the effect of the parameter $D = D_{AB}/\alpha$ on the incipient superheats. Letting D be the predicted value, the superheat was calculated using $D/10$, D, and $10D$; the results are shown in Table 1.

The values of the other parameters were un-

Table 1. Effect of D on the incipient boiling conditions

D_{used}/D	P_A^* at nucleation (atm)	$T_w - T_{sat}$ (°C)
0·1	0·2159	42·54
1·0	0·1258	49·44
10·0	0·0568	54·55

changed for these three calculations and were: $R_e = 3{\cdot}395 \times 10^{-3}$ mm, $P_L = 1{\cdot}00$ atm, $T_0 = 684$°C, $P_{A1}^* = 0{\cdot}20$ atm, $q_w = 31{\cdot}6$ W/cm², $Z = 304{\cdot}8$ mm and $Z_1 = 3{\cdot}05 \times 10^{-3}$ mm. As would be expected, increasing the diffusion coefficient increases the rate at which inert gas leaves the surface cavities, resulting in a lower value of the inert gas partial pressure in the cavities at nucleation and subsequently a larger incipient superheat. Therefore, the absolute values of the superheats presented in this paper must be treated with some caution, however, the predicted trends can be accepted with reasonable confidence.

Apparent Heat Flux Effect Caused by Thermocouple Positioning Errors

It is the purpose of these comments to indicate a possible misinterpretation of a heat flux effect resulting from experiments in which the temperature of the heated surface is determined by extrapolation.

Referring to Fig. 12, the actual wall temperature required for nucleation at the wall-sodium interface is $T(0) \equiv T_w$. But, if the wall temperature is measured at $x = b$ and its exact position is uncertain by the amount c, an error in the extrapolation of $T(b)$ to $T(0)$ can result in an apparent heat flux effect. Noting that

$$T(x) = T_w + \frac{q_w x}{k}$$

and if the thermocouple actually measures the temperature at $x = b$ while the distance $b + c$ is used for extrapolation purposes, the calculated wall temperature will be

$$T_{w,calc.} = T(b) - \frac{q_w b}{k}$$

while the exact wall temperature is

$$T_{w,exact} = T(b) - \frac{q_w(b + c)}{k}$$

so that

$$T_{w,calc.} - T_{w,exact} = \frac{q_w c}{k}.$$

If for the moment, it is assumed that $T_{w,exact}$ is independent of the heat flux, it is clear that $T_{w,calc.}$ will increase with increasing q_w. This apparent effect is illustrated by the following numerical example, where $c = 0{\cdot}25$ mm, $k \cong 0{\cdot}2$ W/cm°C, and $T_{w,exact} - T_{sat} = 100$°C, then

q_w, W/cm²	10	50	100	200	300
$T_{w,calc.} - T_{sat}$, °C	101	106	113	125	138

It is clear that identical reasoning will show a decreasing superheat with increasing heat flux if the extrapolation error is of opposite sign, i.e. if $c < 0$. Therefore, one must be extremely cautious in the interpretation of experimental results in which a heat flux dependency of the incipient boiling superheat is claimed at moderate to high heat fluxes.

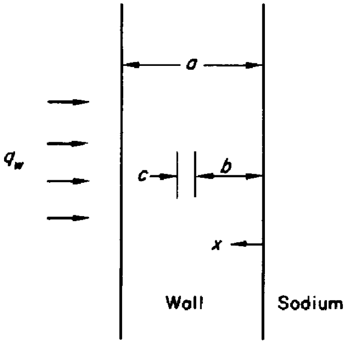

FIG. 12. Thermocouple location in a heated wall.

Résumé—Dans les études expérimentales où l'on mesure la quantité de surchauffe de liquide nécessaire pour nucléer les métaux alcalins liquides, une quantité considérable de résultats contradictoires et, pour la plus grande partie, inexpliqués a été présentée. On a démontré ou postulé que la surchauffe de début est affectée par l'histoire de la pression et de la température, la vitesse du liquide, la pression, le flux de chaleur, la pureté du liquide, la quantité de gaz dissous, les conditions de la surface, les rayonnements particulaires, la méthode de chauffage, la durée de l'opération et peut-être par d'autres facteurs. Cet article présente une étude théorique de l'effet d'un gaz inerte sur les surchauffes de début et illustre que, parmi d'autres choses, la diffusion du gaz à partir des cavités de la surface vers la solution liquide rend compte de l'effet appelé "effet du flux de chaleur". Sur la base de cette étude, on a établi que presque tous les résultats publiés ne contiennent pas assez d'informations pour caractériser de façon adéquate les systèmes expérimentaux. La non-spécification de la pression partielle du gaz dans la couverture gazeuse, par exemple, peut causer une incertitude dans les surchauffes de début aussi grande que 100°C.

Zusammenfassung—In experimentellen Untersuchungen, wobei die Grösse der Überhitzung gemessen wurde, die zur Blasenbildung bei flüssigen Alkali-Metallen notwendig ist, wurden eine beträchtliche Menge sich widersprechender und zum grossen Teil unerklärliche Messergebnisse vorgelegt. Es wurde gezeigt, oder angenommen, dass die beginnende Überhitzung von folgenden Einflüssen abhängt: Druck- und Temperaturvorgeschichte, Geschwindigkeit der Flüssigkeit, Druck, Wärmestromdichte, Reinheit der Flüssigkeit, Gehalt an gelöstem Gas, Oberflächenbeschaffenheit, Teilchenstrahlung, Art der Heizung, Länge der Versuchszeit und wahrscheinlich weiteren Einflüssen. Diese Arbeit beschreibt eine theoretische Untersuchung des Einflusses von Inertgas auf die beginnende Überhitzung und veranschaulicht, dass neben anderen Dingen die Gasdiffusion von den Kavitäten der Overfläche in die flüssige Lösung für den Einfluss der Wärmestromdichte verantwortlich ist. Auf Grund dieser Untersuchung wurde festgestellt, dass nahezu alle veröffentlichten Daten keine ausreichenden Angaben enthalten, um die experimentellen Systeme genügend genau zu beschreiben. Wenn zum Beispiel der Partialdruck des Inertgases in der bedeckenden Gasschicht nicht angegeben ist, bedeutet dies eine Unsicherheit der beginnenden Überhitzung bis zu 100°C.

Аннотация—В экспериментальных исследованиях по определению степени перегрева жидкости, необходимого для начала пузырькового кипения в жидких щелочных металлах, имеется значительное количество противоречивых и большей частью необъясненных данных. Показано или постулируется, что на требуемый начальный перегрев влияет изменение температуры и давления, скорость жидкости, давление, тепловой поток, чистота жидкости, содержание постороннего газа, состояние поверхности, излучение частиц, способ нагрева, продолжительность работы и, возможно, другие факторы. Данная работа представляет теоретическое исследование влияния инертного газа на начальный перегрев. В ней также показывается, что диффузия газа из полостей на поверхности в раствор жидкости учитывает так называемый «эффект теплового потока». На основе этого исследования установлено, что почти все опубликованные данные не дают достаточной информации для удовлетворительной характеристики экспериментальных систем. Например, если не учитывать парциального давления инертного газа в газовой оболочке, ошибка расчета начального перегрева может достигнуть 100°С.