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NOTES ON THE LIQUID-METAL BOILING PHENOMENON

A. I. Krakoviak

ABSTRACT

A brief review of boiling for ordinary fluids with emphasis on the wall superheat required for bubble initiation from different surfaces at varying pressures is presented. Calculations based on an equation valid for normal fluids indicate that superheats from two to eight times that for water are required to initiate a vapor bubble in some of the alkali metals. This high superheat and the high thermal conductivity peculiar to liquid metals are combined to explain the unstable liquid-metal boiling phenomenon in natural- and forced-circulation loops.

Forced-convection boiling-potassium heat-transfer coefficients correlated by the Lockhart-Martinelli two-phase flow parameter are in reasonable agreement with the forced-convection boiling-water data of Dengler and Addoms.

Some recommendations on bubble initiation sites and future work with liquid metals are also included.

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NOTES ON THE LIQUID-METAL BOILING PHENOMENON

A. I. Krakoviak

INTRODUCTION

A brief review of the boiling phenomenon in ordinary fluids (water, alcohol, etc.) with emphasis on the superheat required to initiate a bubble from a surface may be useful in explaining the erratic behavior observed in the natural- and forced-circulation boiling-potassium loops at the Oak Ridge National Laboratory.

The characteristic boiling curve similar to the one shown in Fig. 1 was first developed by Nukiyama[1] in 1934, who boiled water from an electrically heated horizontal platinum wire submerged in a pool of water at saturation temperature. In boiling regime I, as the wall temperature is raised above saturation temperature, convection currents circulate superheated liquid and vapor is produced by evaporation at the free liquid surface. In regime II, a further increase in wall temperature results in the formation of vapor bubbles which rise from favored spots on the heated surface and condense before reaching the free surface. In regime III, more numerous bubbles form at the heated surface and rise to the free liquid surface and into the vapor space. This article will be limited to these three regimes.

Heidrich[2] in 1931, using distilled degassed water in a clean glass container heated by a solution of calcium chloride, investigated the temperature distribution and heat flow rates for regime I where vapor is produced by evaporation from the free liquid surface without the formation of bubbles at the heated surface. The curves in Fig. 2 show that near the surface the temperature gradient is linear, indicating a stagnant layer through which heat is transferred by conduction alone. At the higher values of liquid superheat, larger temperature gradients through thinner stagnant layers accompanied higher evaporation rates, w/A. Below the stagnant layer, convection currents tend to maintain a constant temperature with depth.

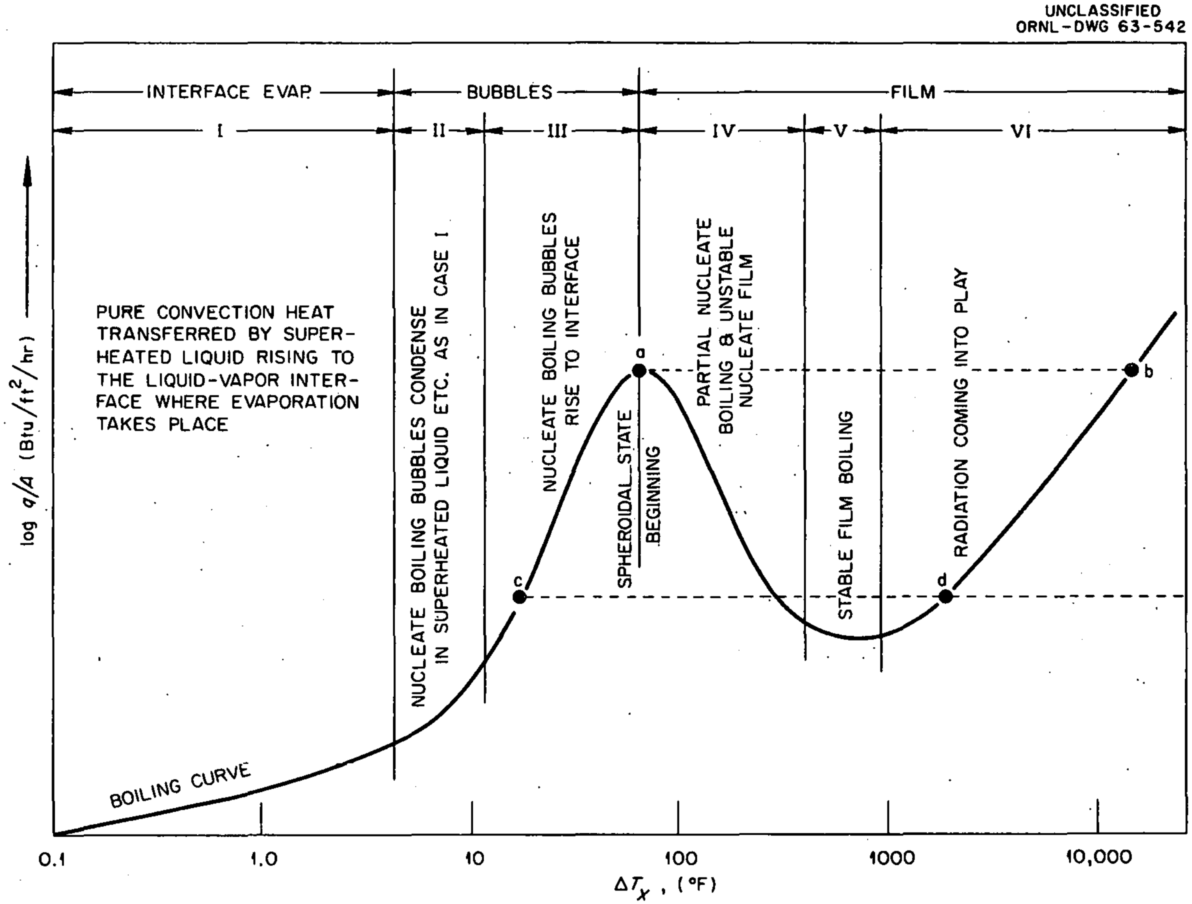

Fig. 1. Typical Boiling Curve for Water.

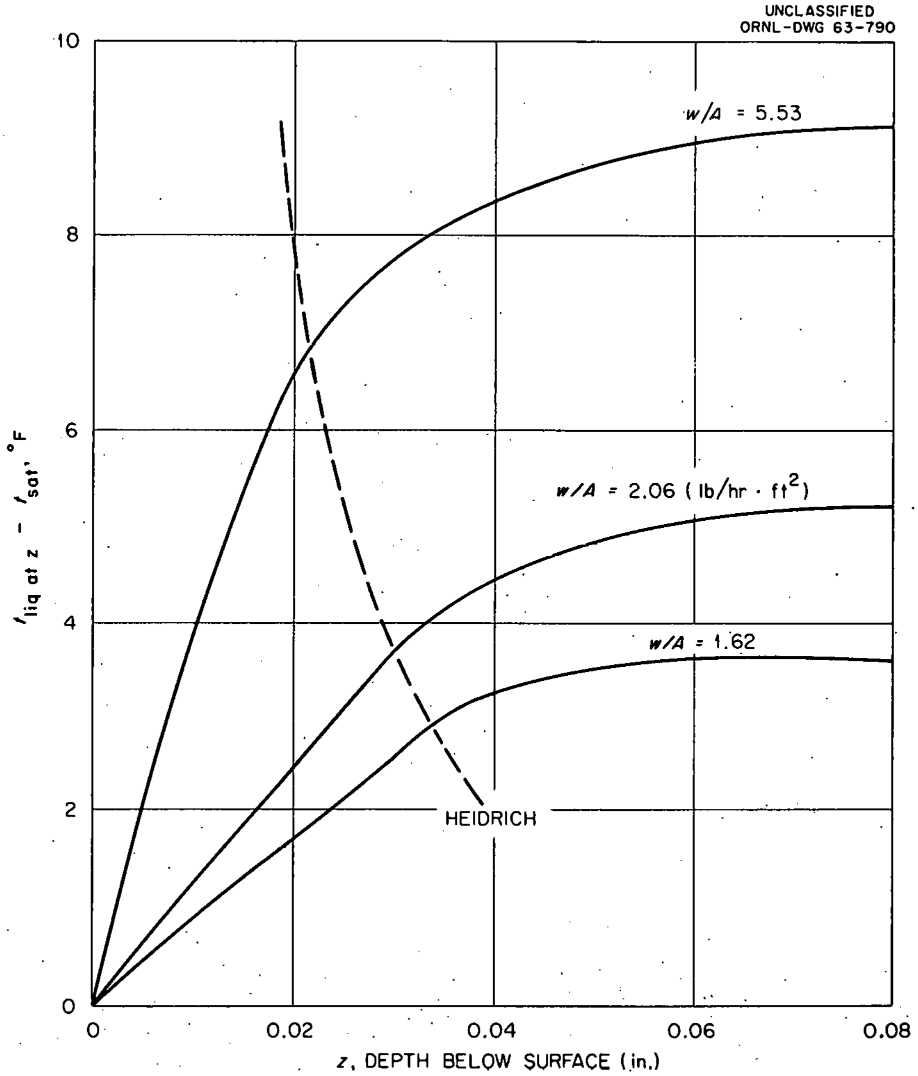

Fig. 2. Liquid Superheat as a Function of Depth and Boiling Rate in a Pool of Water.

In regimes II and III (where vapor bubbles are formed), the Laplace equation relates the pressure, P_v, of a vapor inside a spherical bubble and the external pressure on the liquid, P_ℓ, to the radius of the bubble, r, and the surface tension of the liquid, σ, as follows:

$$P_v - P_\ell = \frac{2\,\sigma}{r} \,. \tag{1}$$

This equation shows that for bubble growth, P_v must be greater than P_ℓ, and for evaporation the temperature of the liquid must be greater than the temperature of the vapor; thus for boiling to exist, the liquid must be superheated with respect to the vapor in the bubble as well as to itself.

Ellion[3] in 1954 combined the Clausius-Clapeyron equation with the equation of Laplace for a thermodynamic evaluation of the nucleation process with the following equation by assuming the specific volume of the vapor is large compared with that of the liquid and that it can be found by the gas law, $V_g = (R\,T_{sat})/P_\ell$:

$$T_v - T_{sat} = \frac{2\,R\,T_{sat}^{2}\,\sigma}{h_{fg}\,P_\ell\,r} \,, \tag{2}$$

where

T_v is the absolute temperature of the vapor within a spherical bubble,

T_{sat} is the absolute temperature of the saturated liquid,

P_ℓ is the external pressure on the liquid,

h_{fg} is the latent heat of vaporization of the liquid, and

V_g is the specific volume of the vapor.

This equation predicts the approximate liquid superheat required for equilibrium of a bubble of radius r.

Wallis and Griffith[4] in 1959 carried out nucleation experiments by immersing in water a copper sheet containing a conical cavity made by pressing into the surface of the copper a phonograph needle having an apex angle of 18°. Using the cavity radius at its mouth as r, their data agreed quite well with Eq. (2) as shown in Fig. 3. They showed that the superheat required to maintain ebullition at a rate from 0.2 to 5 bubbles/sec was inversely proportional to pressure.

Berenson[5] using a heated copper surface showed that higher wall superheats were required at a given heat flux to boil pentane from smoother surfaces. The curves in Fig. 4, from left to right, represent the wall superheats required for surfaces which had been finished with progressively finer emery cloth. The smoother surface has smaller cavities and hence the radius of curvature of the vapor bubble is smaller than for the rough surface and from Eq. (2) requires a greater wall superheat for bubble growth.

Farber and Scorah[6] using Chromel C wire submerged in water showed that a smaller wall superheat at a given heat flux is required for boiling as pressure is increased. Their data in Fig. 5 are also consistent with Eq. (2).

From a photographic study of the velocities and paths of bubbles in pool boiling experiments, Jakob[7] noticed that bubbles appeared to initiate at favored spots on the heated surface and that the frequency of bubble formation depends on the size of the bubbles at the moment of break-off. His data in Fig. 6 show frequency to be a hyperbolic function of the diameter of a sphere having the same volume as the bubble at the moment of break-off. He also observed that in the heat flux range up to 18,000 Btu/hr·ft^2 an increase of 700 Btu/hr·ft^2 activated one more cavity to produce a steam column.

Any surface has a cavity size distribution peculiar to that particular surface and may be represented by the curve in Fig. 7. As heat is applied to the surface, the larger cavities become activated first, and as heat flux is increased more cavities and hence smaller cavities become activated requiring greater wall superheats in accordance with Eq. (2). Rohsenow[8] postulates that the slope of the q/A vs ΔT curve expresses the change in ΔT necessary to increase the number of activated cavities sufficiently to accommodate the new heat flux. Although the slope is predominantly in the neighborhood of three, data are available showing slopes varying from unity for contaminated surfaces to slopes

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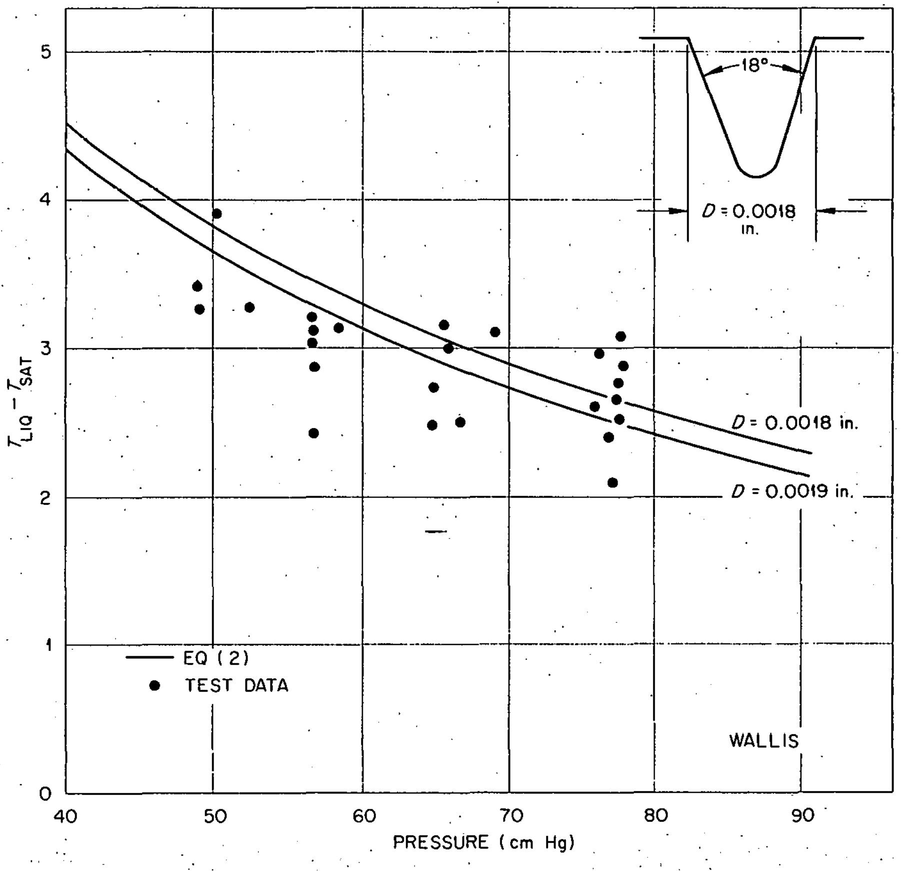

Fig. 3. Measured Superheat Compared with Those Calculated.

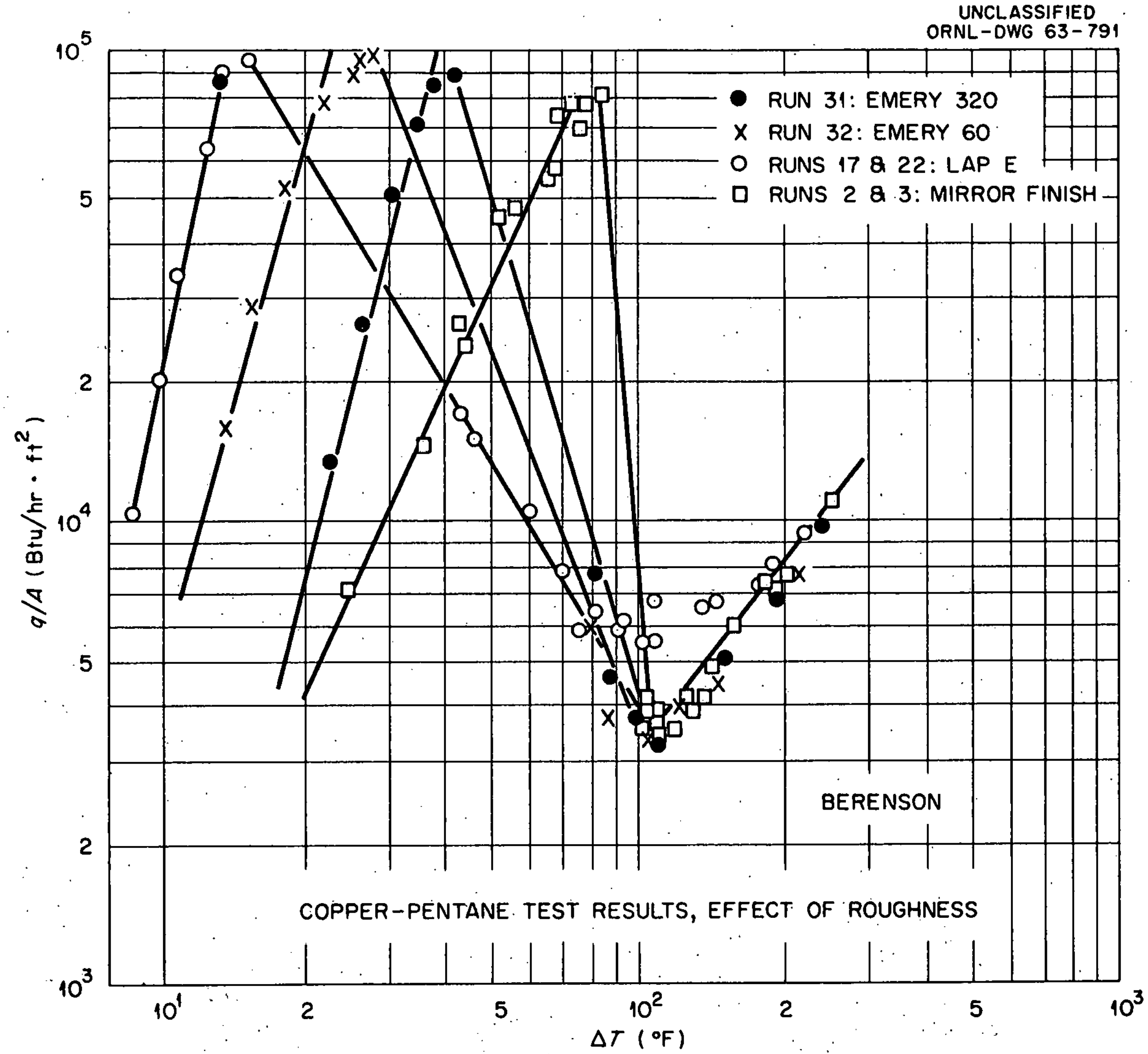

Fig. 4. Effect of Surface Roughness on Superheat.

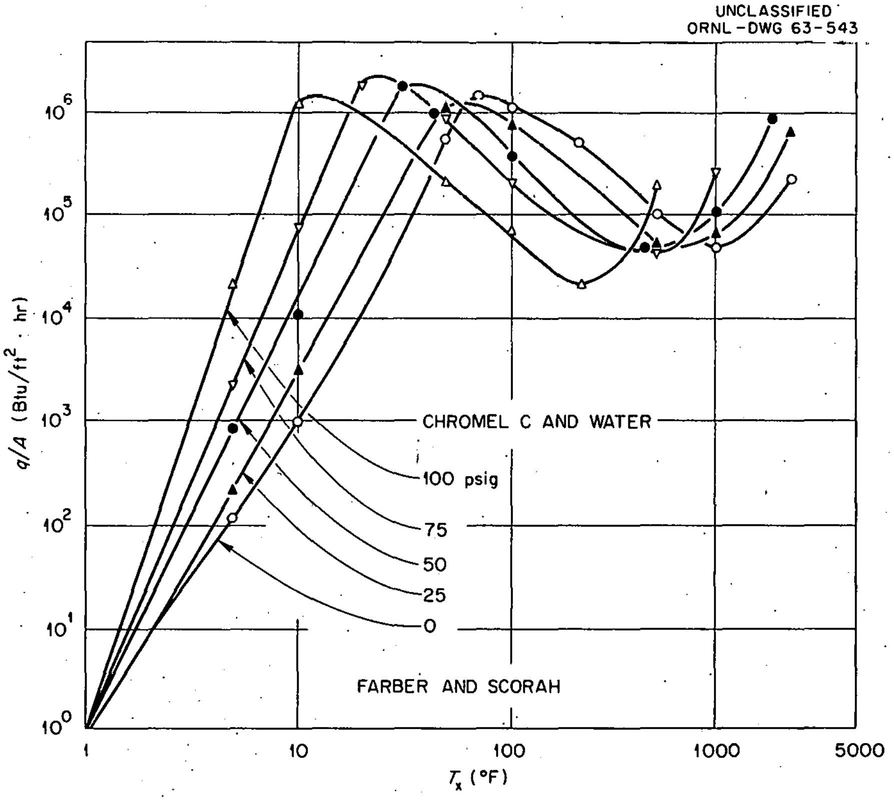

Fig. 5. Effect of Pressure on Superheat.

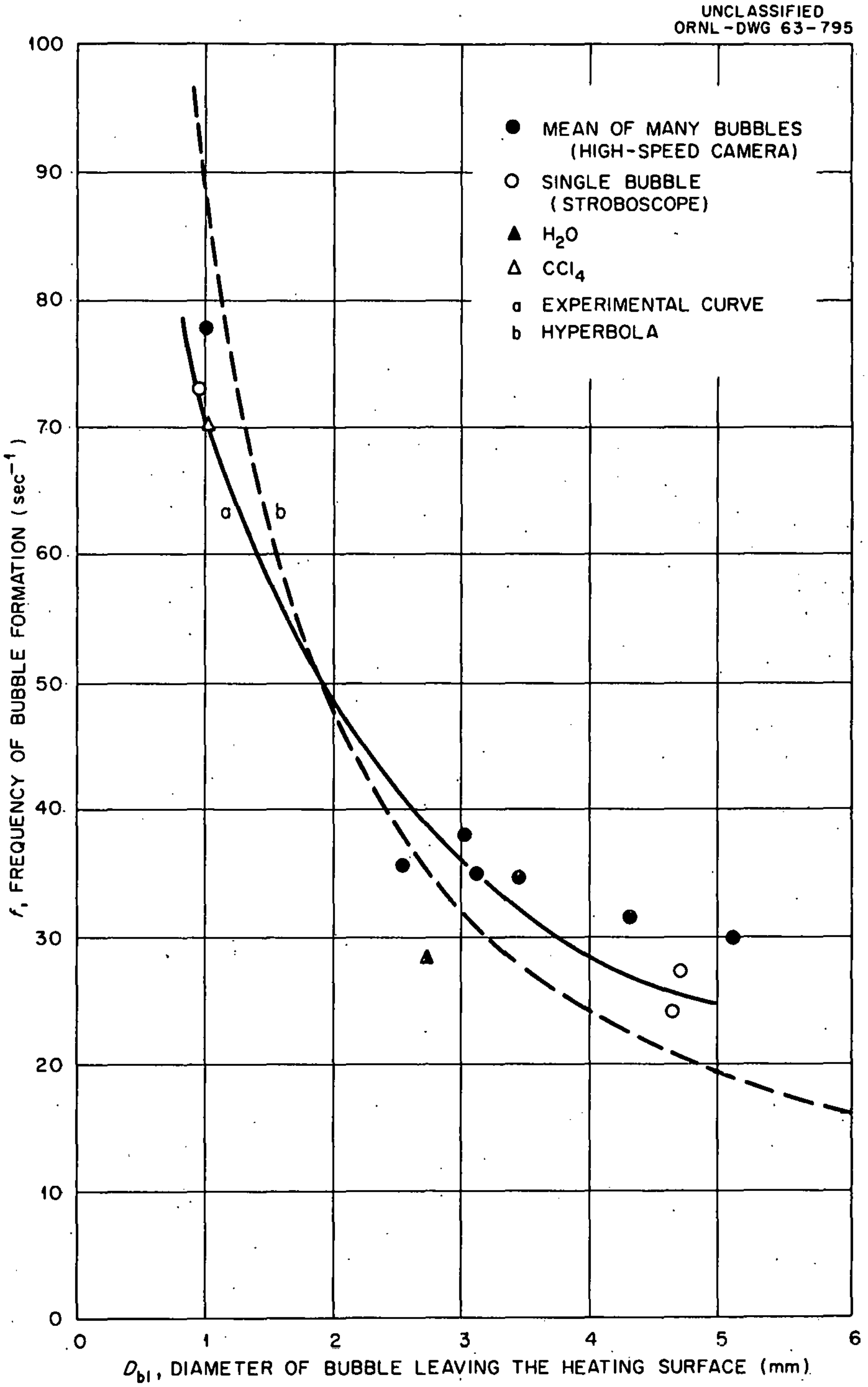

Fig. 6. Bubble Formation Frequency as a Function of Bubble Diameter.

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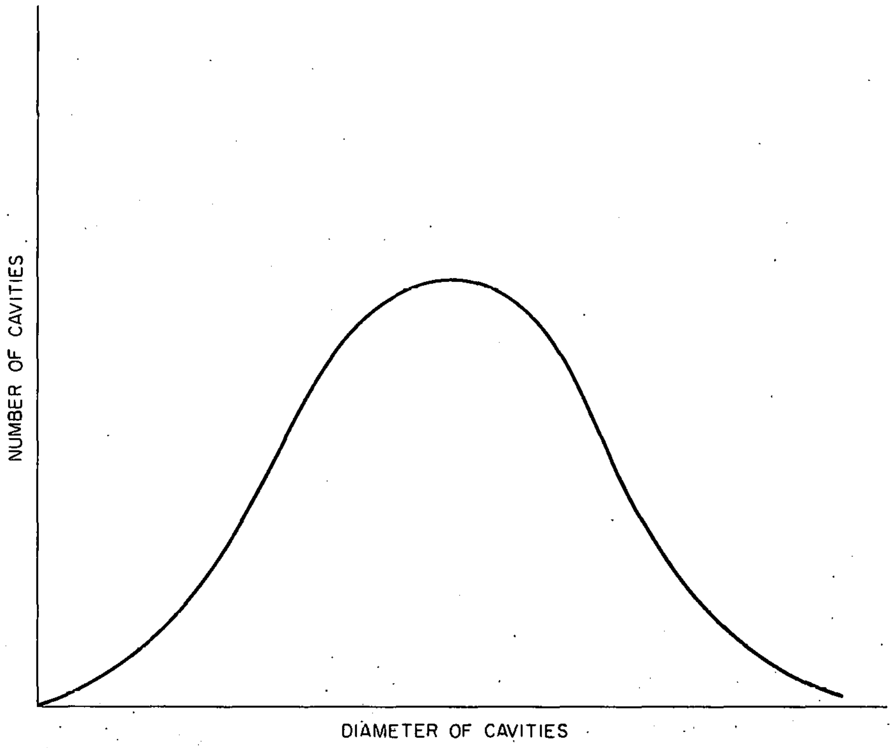

Fig. 7. Normal Cavity Size Distribution Curve.

of 25 for very clean surfaces. It is his postulate that the actual slope depends upon the uniformity of the size and shape distribution of the cavities in a particular surface.

Wallis and Griffith[4] have counted nucleation spots as a function of wall superheat for three different fluids (ethanol, methanol, and water) boiling from the same surface and obtained the curves shown in Fig. 8. Rearranging Eq. (2) and applying the ideal gas law equation, they presented Eq. (2) in the following form:

$$r = \frac{2 \sigma T_w V_{fg}}{h_{fg} (T_w - T_{sat})} , \qquad (3)$$

where

V_{fg} is the specific volume change of vaporization, and
T_w is the absolute temperature of the wall.

Using the observed superheat values from Fig. 8 in Eq. (3), the calculated cavity radius, r, produced one line common to the three fluids as shown in Fig. 9, thus characterizing that particular surface for boiling heat transfer.

Corty and Foust[9] reported a hysteresis loop for the q/A vs ΔT plot as shown in Fig. 10. When a particular surface is immersed in a liquid and as heat is applied to the surface, the boiling curve may trace any line between abdc and abfc to point C, depending on the amount of inert gas trapped in the cavities of the surface. When the heat flux is reduced after having boiled at point C for quite some time, the boiling curve follows the path cdba where boiling ceases. A subsequent increase in heat flux may cause the boiling curve to follow the path abe before boiling is initiated and returns to point C. Griffith and Wallis reported that a surface which produced a boiling curve following path abgc with tap water would follow curve abedc if the water was degassed before starting. Apparently, the degassed water dissolved all of the gas from the cavity before boiling began; consequently, the cavity filled with liquid and was no longer active.[4] These investigators also showed that conical cavities such as shown in Fig. 11a are not stable, whereas the

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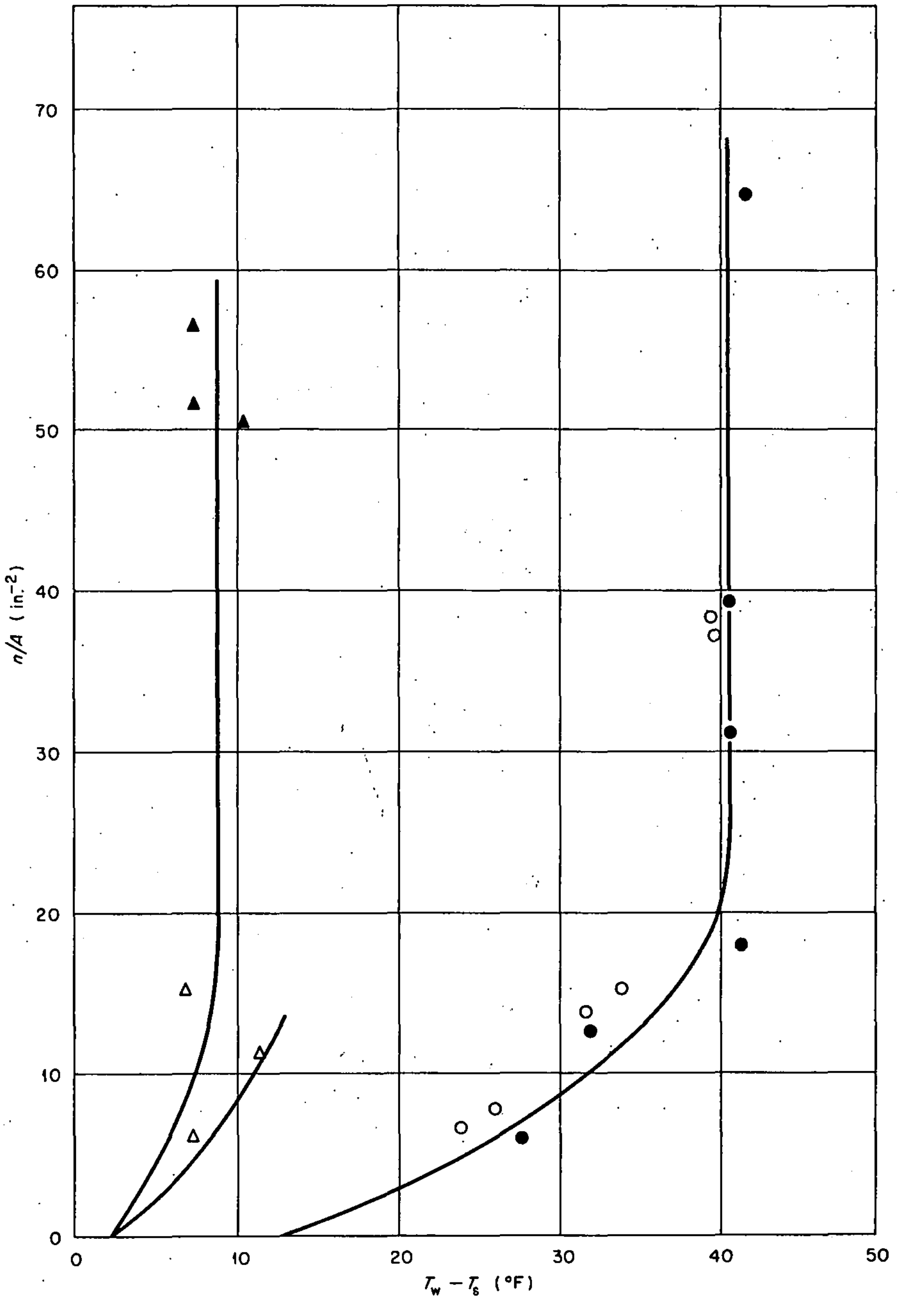

Fig. 8. Number of Active Spots per Unit Area Vs Wall Superheat.

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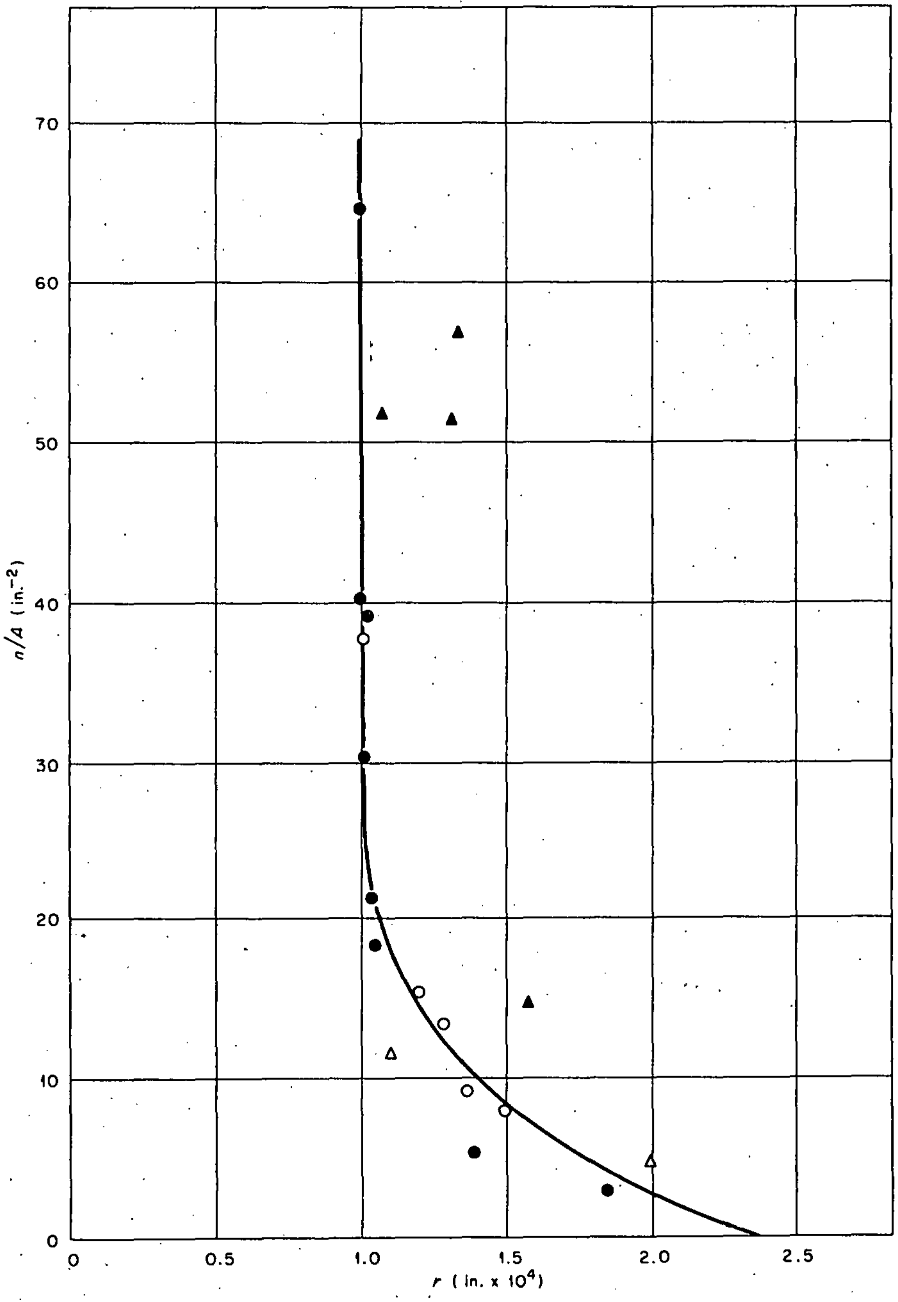

Fig. 9. Data From Fig. 8 as a Function of r as Calculated from Eq. (3).

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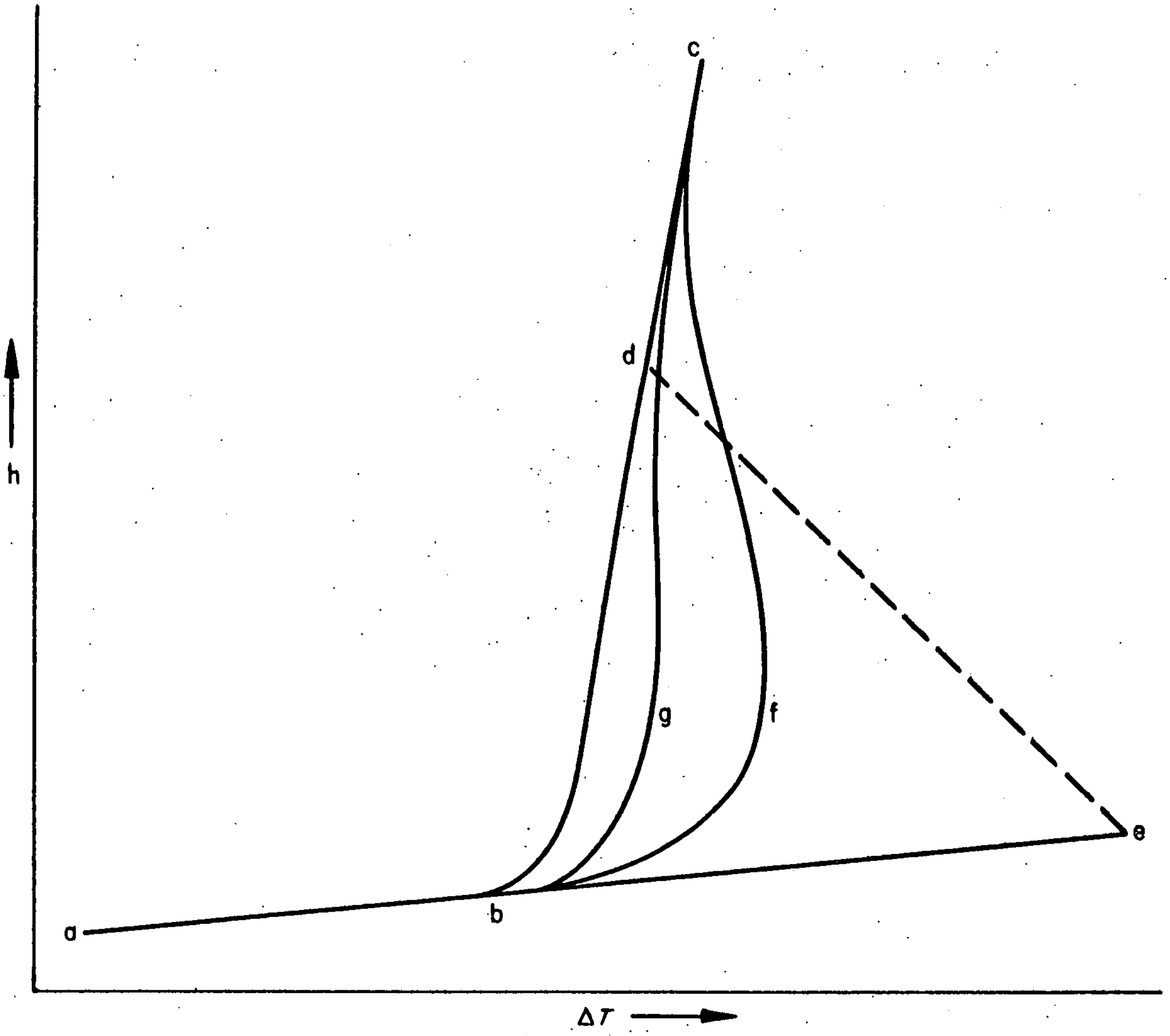

Fig. 10. Typical Hysteresis Loop for Pool Boiling of Water.

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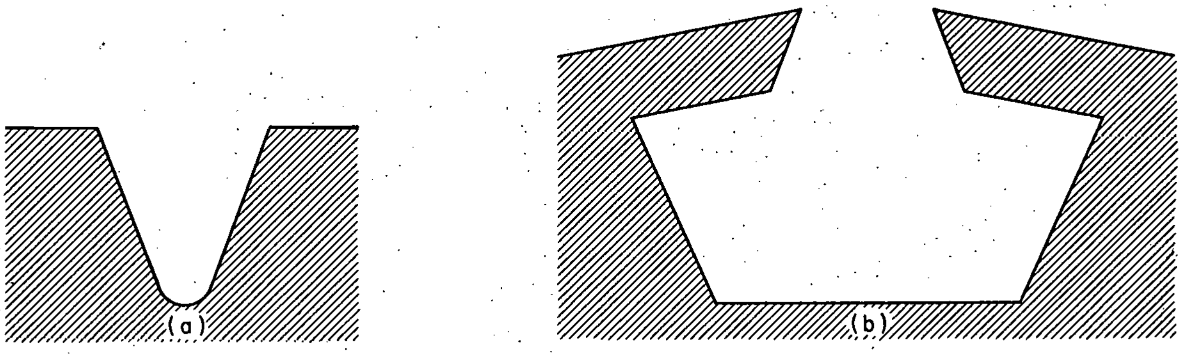

Fig. 11. Conical and Re-entrant-Type Cavities.

re-entrant-type cavities shown in Fig. 11b are very stable in that the liquid must be subcooled to deactivate this type of cavity. Wallis and Griffith also reported that a sudden increase in pressure during boiling would deactivate a cavity and cause the boiling curve to follow path abed in Fig. 10 before boiling could be re-established.

BOILING AS APPLIED TO LIQUID METALS

Since Eq. (2) is substantiated by the experimental data of several investigators working with more than one fluid, one may calculate, by comparison with water, the superheat required to initiate a vapor bubble from a surface immersed in a liquid metal with the following equation:

$$\frac{T_{v_w} - T_{sat_w}}{T_{v_x} - T_{sat_x}} = \frac{\sigma_w \, T_{v_w} \, V_{fg_w} \, h_{fg_x} \, r_x}{\sigma_x \, T_{v_x} \, V_{fg_x} \, h_{fg_w} \, r_w} , \qquad (4)$$

where the subscripts w and x refer to water and a liquid metal, respectively.

Using the physical properties (Table 1) of water, sodium, potassium, rubidium, and cesium at their respective normal boiling points and assuming that 30°F is required to sustain vapor bubbles from a particular surface with a cavity of radius r immersed in water at atmospheric pressure, the superheats required to sustain vapor bubbles from the same surface immersed in the above alkali metals (at one atmosphere) were calculated. It was found that superheats of 30, 125, 258, 101, and 67°F were required to sustain vapor bubbles from a given cavity when in H_2O, K, Na, Rb, and Cs, respectively. In comparison with water, this means that the superheat required to initiate bubble formation must be increased by a factor of 4.2 for K, 8.6 for Na, 3.4 for Rb, and 2.2 for Cs. The boiling potassium loops of interest are those boiling in the absence of inert gases. These then should be compared with distilled, degassed water boiling from a clean surface, where superheats of ~90°F are required to initiate a vapor bubble as was discussed for the hysteresis loop in Fig. 10. Comparable superheats calculated to initiate vapor bubbles in K, Na, Rb, and Cs at atmospheric pressure are then 375, 774, 203, and 201°F,

Table 1. Physical Properties of Water and Liquid Metals Evaluated at their Respective Normal Boiling Points

Liquid	Normal Boiling Point, °F	Spec. Vol. Vapor ft^3/lb	Density of Liquid, lb/ft^3	Latent Heat of Vaporization, Btu/lb	Thermal Conductivity, Btu/hr·ft·°F	Surface Tension, lb/ft
Water	212	26.80	59.83	970	0.393	0.00403
Potassium	1400	32.47	41.58	851	18.1	0.0043[a]
Sodium	1618	60.6	46.4	1609	30.1	0.0077[b]
Rubidium	1270	16.67	82.15	347.5	11.8	0.003[c]
Cesium	1260	10.42	104.7	213.8	10.6	0.002[c]

[a] Estimated from preliminary data of J. W. Cooke, Oak Ridge National Laboratory.

[b] Estimated from data of J. W. Taylor, AERE M/R 1247.

[c] Estimated from data of S. W. Strauss, "Surface Tension of Liquid Metals at their Melting Points," Nuclear Science and Engineering, 8: 362–363 (1960).

respectively.

The foregoing assumptions and calculations coupled with the high thermal conductivity of potassium may explain the observed boiling instabilities and anomalies in the natural- and forced-circulation loops at ORNL. In the forced-circulation loop operated by the author (when potassium entered the boiler subcooled), wall temperatures on the boiler increased relatively slowly (~20 sec) to approximately 150 to 250°F above the average operating temperature. At this temperature (not necessarily constant), there was an extremely rapid (10^{-1} sec) decrease in boiler temperature with a simultaneous and equally rapid increase in loop pressure and condenser temperature.

To illustrate the widely different temperature profiles and thus the different boiling behavior between water and liquid metals, imagine an infinitely long 1-in.-OD solid rod surrounded by an infinitely long clamshell-type heater. After a sufficient length of time with energy radiating to the rod at a constant rate of, say, 30,000 Btu/hr·ft^2 (maximum heat flux of all but one boiling-potassium loop at ORNL), the temperature profile within the rod will assume the paraboloid of revolution shown in Fig. 12a. Paths a and b represent the temperature gradients in a rod of thermal conductivities equivalent to potassium (18.1 Btu/hr·ft·°F) and water (0.393), respectively. Now if this rod is replaced with a 1-in.-ID tube filled with water with 1 atm of pressure above it, then when the wall temperature rises to 30°F (depending on the cavity size of heated surface) above saturation temperature, tiny "explosions" of liquid into vapor take place at the wall and grow very rapidly in the 0.005-in. superheated annulus depicted in Fig. 12b before moving toward the center of the tube where they either grow more slowly or collapse, depending on whether the fluid is at saturation temperature or subcooled. If subcooled, the fluid would eventually reach saturation temperature and remain there with T_w ~30°F higher than saturation temperature and boil with no great pressure perturbations. Now if this rod is replaced with a 1-in.-ID tube filled with potassium with 1 atm of pressure above the potassium, then the wall temperature would rise 375°F above saturation temperature before bubble nucleation at the wall. However, because of the conductivity of potassium,

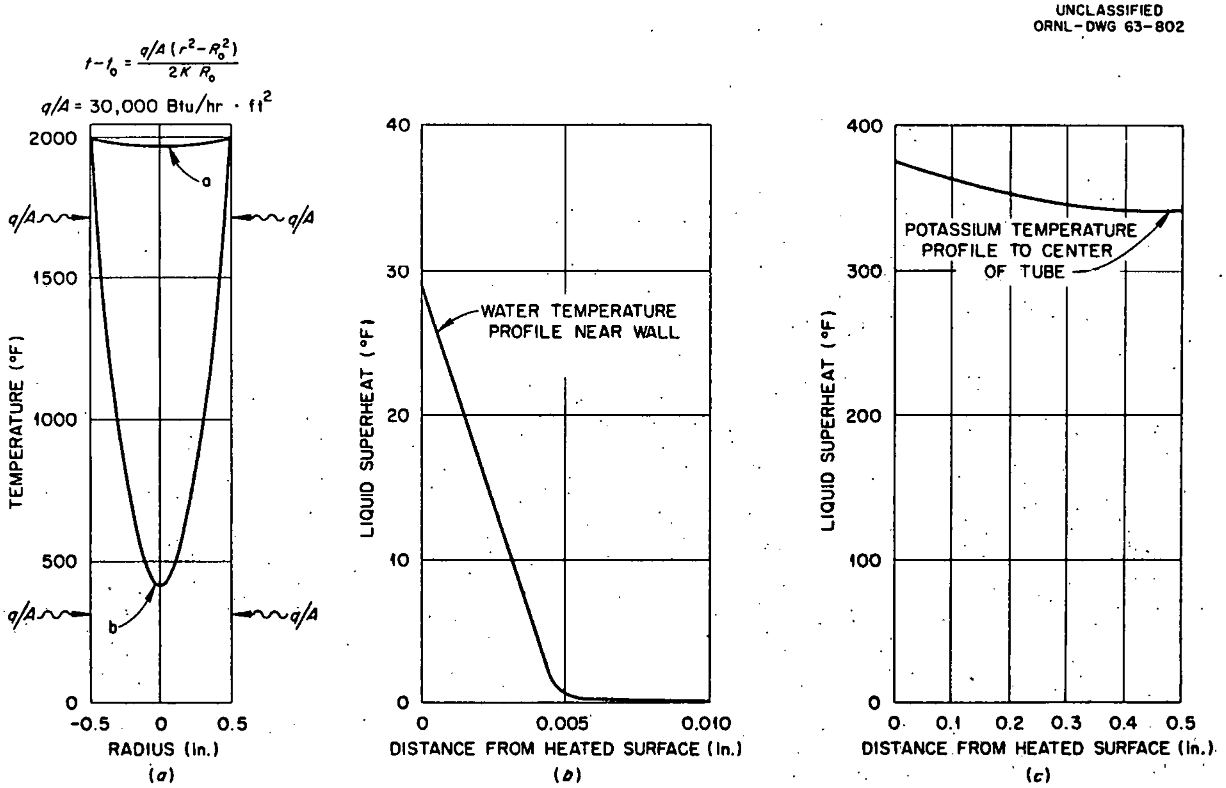

Fig. 12. Temperature Profile Within a Rod Under Conditions Discussed on Page 22.

the entire volume of potassium in the tube would be at a superheat of not less than 340°F (Fig. 12c). Now when a bubble nucleates, the energy of superheat from the entire volume is released in a matter of microseconds in a violent "explosion". The energy of superheat in a unit volume of potassium when released under these conditions produces a vapor quality of ≈7.6 wt %, resulting in a momentary high-pressure surge and supplies a large amount of vapor to the condenser, thus increasing the condenser temperatures. This cycle repeats itself at a frequency depending on the heat flux and thermal gradient in the bubble-producing cavity. If the heated surface contains a re-entrant-type cavity with a sufficiently high thermal gradient to maintain a liquid-vapor interface at the cavity mouth, then stable boiling without excessive liquid superheat can be expected.

If Eq. (2) is used to calculate the superheat required to initiate a bubble from a surface (a surface which requires a superheat of 30°F to maintain a steam bubble) and Fourier's conduction equation is used to calculate the heat transfer across a liquid (assuming no convection contribution), the curves in Fig. 13 indicate that a 2.5-in. thickness of sodium at 1618°F will sustain a heat flux of 1×10^5 Btu/hr·ft^2 without nucleating a vapor bubble at the heated surface; the same thickness of potassium will sustain a heat flux of 3.3×10^4 Btu/hr·ft^2 without nucleation. Increased pressure at the heated wall due to static liquid head and convection currents (neither of which were considered in this analysis) would increase the sustainable heat flux appreciably.[10] Consequently, all liquid-metal pool boiling data (q/A vs ΔT) taken without the aid of a contact microphone or other means to detect ebullition should be questioned.

BEHAVIOR OF NATURAL- AND FORCED-CIRCULATION LOOPS

In the case of natural-circulation loops of the closed U-tube manometer type, nucleation and concomitant release of the energy of superheat (evidenced by a pressure surge and boiler temperature decay) take place in a matter of microseconds, whereas the temperature increase or superheat buildup portion of the cycle takes 20 to 60 sec, depending on heat flux and heat capacity of the fluid and boiler. During this temperature

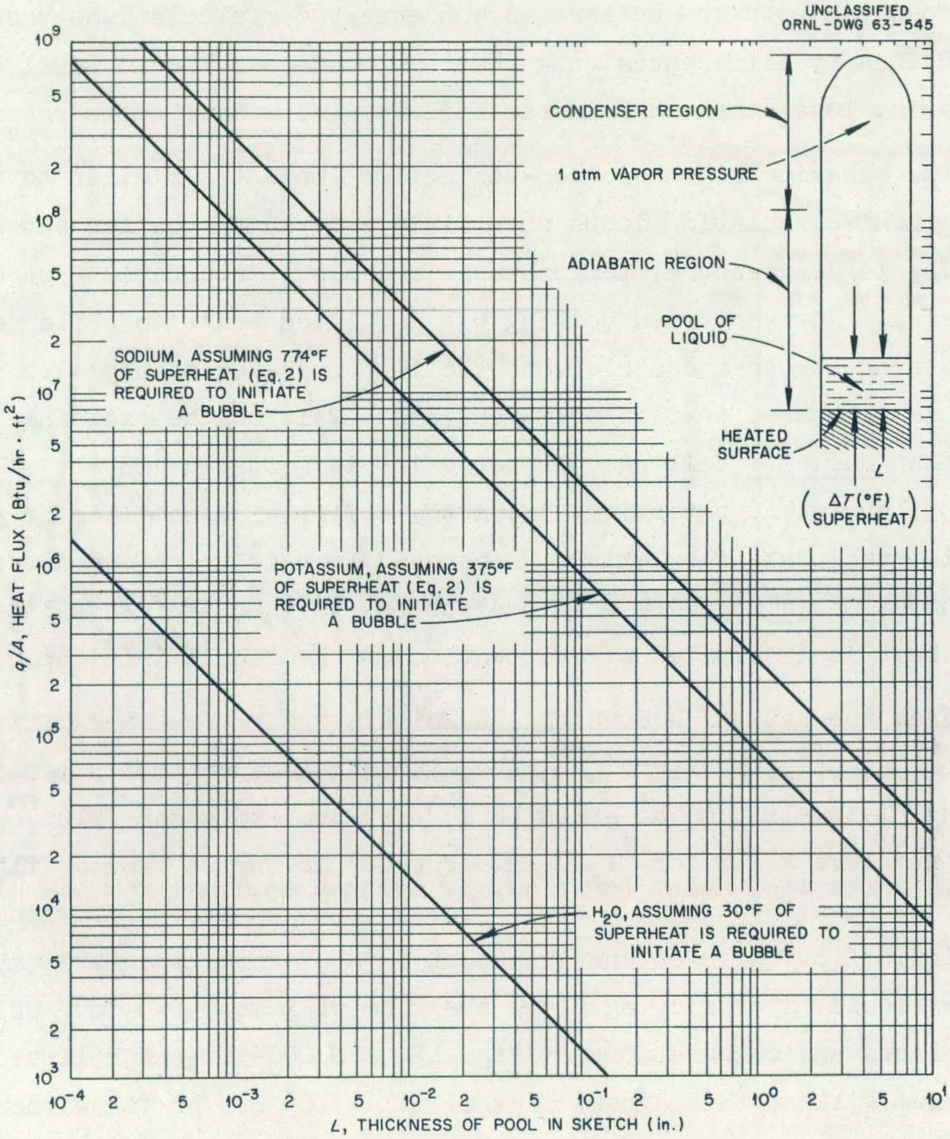

Fig. 13. Heat Conduction as a Function of Δt and Thickness.

buildup portion of the cycle, the liquid level in the boiler and condenser does not fluctuate, and vapor is produced only by evaporation at the liquid-gas interface in the boiler. This is supported by the fact that sharp lines of demarcation (delineating the liquid-vapor interfaces) were found in both the boiler and condenser.[11] Calculations based on liquid density differences show that the condenser liquid level should be ~1 1/4 in. lower than that in the boiler; ~1-in. difference was found.

The behavior of the forced-convection loops was similar to that in the natural-circulation loops when fluid entered the boiler subcooled; that is, large pressure, temperature, and flow perturbations were encountered. However, when boiling was initiated prior to boiler entrance by flashing across a restriction, the loop operated stably with no pressure, temperature, or flow perturbations. This can be explained by the fact that nucleate boiling does not exist in a forced-convection potassium boiler, where potassium enters the boiler with some finite quality; that is, two-phase flow exists. Instead, heat is transferred primarily by conduction (along with some convection) to the liquid-gas interface where heat is dissipated by evaporation of the liquid into vapor.

From the data of Radovcich and Moissis[12] for air-water mixtures flowing concurrently in a vertical tube, it is estimated that potassium flowing in a 1-in.-ID tube at atmospheric pressure and a mass flux of 1×10^5 lb/hr·ft^2 (0.6×10^5 for 0.320-in.-ID tube) is in the annular flow regime at a vapor quality of ≥ 0.4 wt %. Then, assuming zero slip with all of the liquid flowing in an annulus about a vapor core, the maximum thickness of the liquid layer flowing along the tube wall is 0.040 in. Using the assumptions and calculations of Fig. 13, a 0.040-in.-thick liquid film of potassium will sustain a heat flux of 2.1×10^6 Btu/hr·ft^2 without nucleation at the heated surface (7×10^6 for sodium). Thus nucleate boiling is "suppressed" because the high thermal conductivity of the annulus of liquid potassium and vaporization at the liquid-gas interface preclude the development of sufficient wall superheat to initiate bubble formation at the liquid-solid interface.

FORCED-CONVECTION HEAT TRANSFER

Dengler and Addoms,[13] working with a water system heated by steam jackets, have reported forced-convection nucleate boiling at low qualities in the first half of their boiler and forced-convection surface evaporation in the latter half of their boiler where high qualities and very thin liquid annuli existed. Their forced-convection nucleate boiling data are presented as the solid dots in Fig. 14.

Schrock and Grossman[14] in 1961, working with water in forced-convection electrically heated test sections, correlated their results to both the Lockhart-Martinelli two-phase flow parameter, X_{tt}, and to a boiling number, B_o, first described by Mumm.[15] The boiling number, $B_o = (q/A)/G\ h_{fg}$, is the ratio of radial heat flux to longitudinal heat flow. In Fig. 15 they point out that heat-transfer coefficients with high boiling numbers (where nucleate boiling predominates) are independent of the Lockhart-Martinelli parameter, whereas for the case of low boiling numbers (where nucleate boiling is precluded and surface evaporation takes place), the heat-transfer coefficients are strongly dependent on the two-phase parameter. They correlated all of their data with B_o and X_{tt} as shown in Fig. 16.

A Dengler-Addoms type of correlation is shown in Fig. 17 for potassium where h/h_ℓ is the ratio of the heat-transfer coefficient with boiling (h) to that which would be obtained without boiling (h_ℓ) at the same inlet mass flow and is a measure of the improvement in heat transfer which accrues from the boiling process. For the low flows of this investigation (Peclet moduli < 100), the nonboiling coefficient was based on the equation $N_{Nu} = 0.047\ N_{Pe}$, as developed from the data of Johnson, Hartnett, and Clabaugh[16] and from preliminary results with potassium at ORNL. The runs are grouped in series of three to five runs; there were equipment repairs and delays between series. Although there is wide scatter in the data, the slope of each series is consistent with each of the others and to that for water. The scatter among series is attributable to a faulty thermocouple controlling the adiabatic region near the boiler exit thermocouple well. This was noticed after the boiler was disassembled.

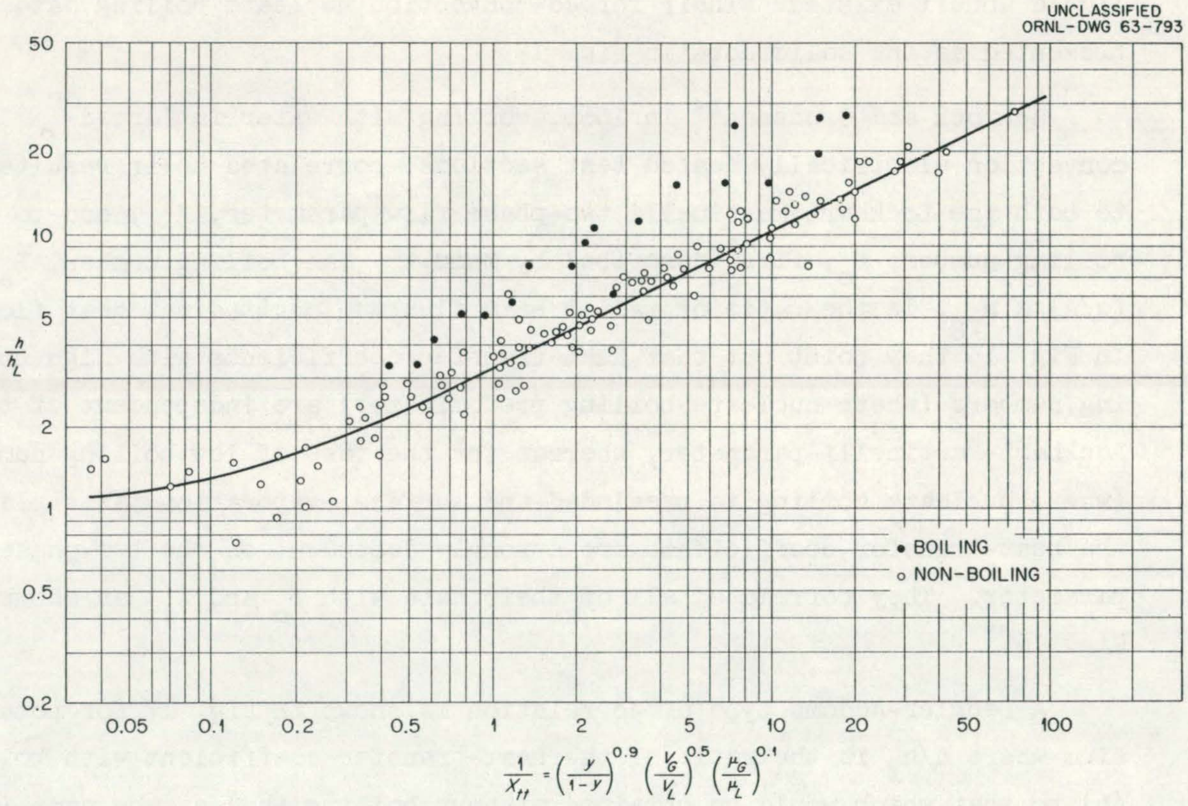

Fig. 14. Ratio of Heat-Transfer Coefficient as a Function of the Lockhart-Martinelli Two-Phase Parameter.

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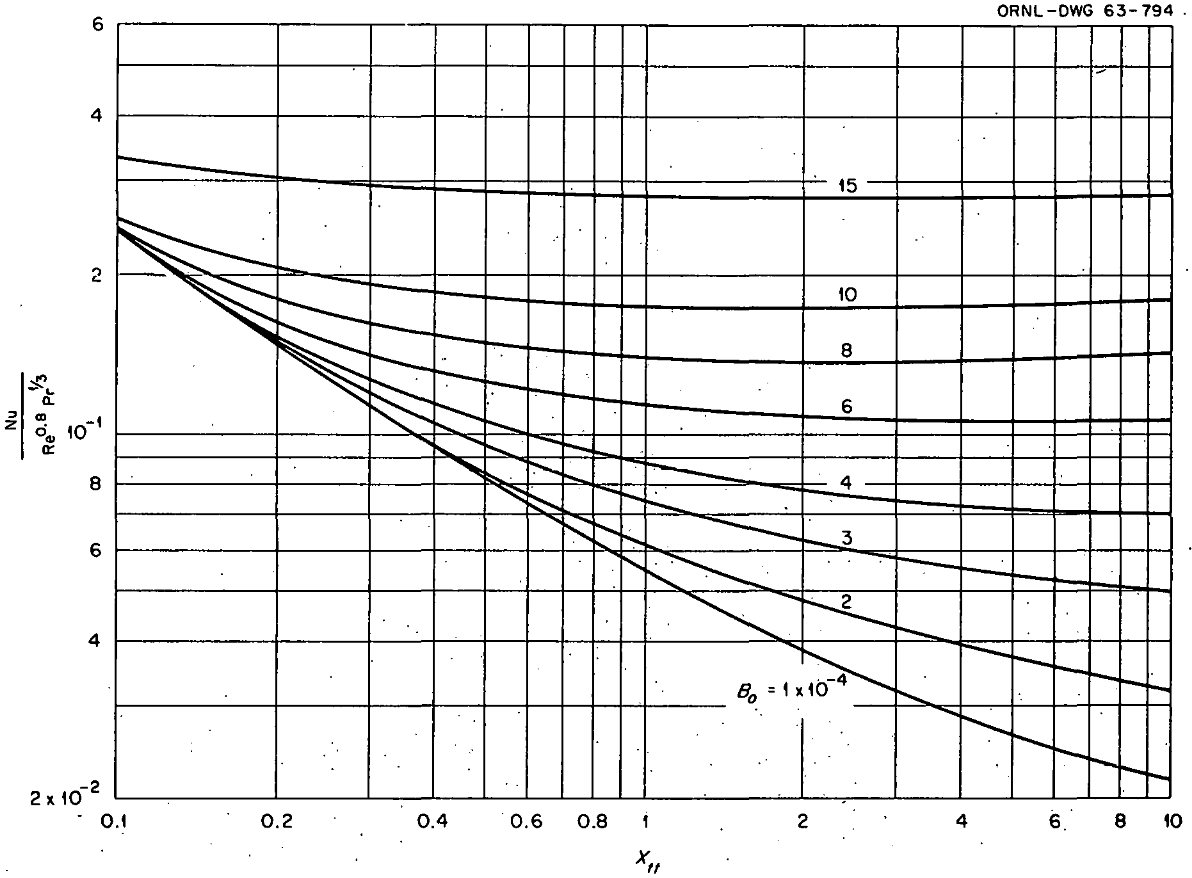

Fig. 15. Effect of Boiling Number and the Lockhart-Martinelli Two-Phase Parameter on Heat Transfer.

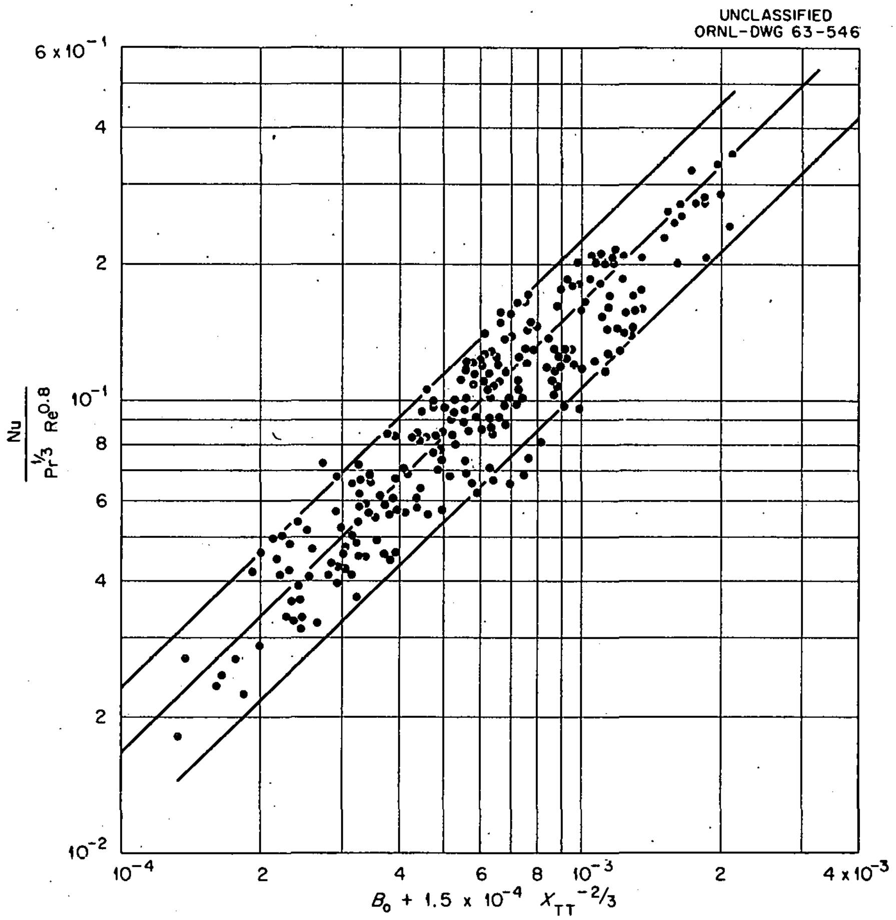

Fig. 16. Heat-Transfer Rate as a Function of Boiling Number and the Two-Phase Parameter of Lockhart and Martinelli.

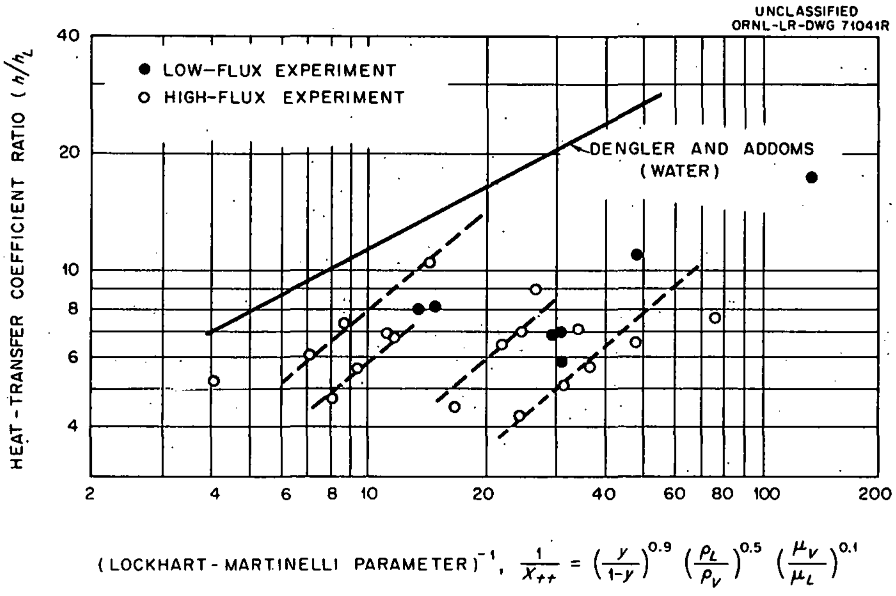

Fig. 17. Ratio of Boiling to Nonboiling Coefficients as a Function of the Two-Phase Flow Parameter.

Since, under corresponding conditions, heat-transfer coefficients for nonboiling potassium are much greater than those for nonboiling water, it is not surprising that for the liquid metal the gain in the heat-transfer coefficient due to boiling is only one-third that found with water.

CAVITY SIZE CALCULATIONS

If a superheat of 10°F is desirable in a boiling-liquid-metal system, then by Eq. (3) the calculated cavity radius to sustain boiling from a surface submerged in K, Na, Rb, and Cs is 0.94, 1.87, 0.77, and 0.52×10^{-3} in., respectively. Although a cavity of the re-entrant type shown in Fig. 11b is desirable, an alternate cavity such as that shown in Fig. 18c may be readily made by drilling a hole of the desired diameter to a depth of ≥ 0.008 in. and "sandblasting" with stainless steel particles to peen the mouth. A deeper cavity would take advantage of the temperature gradient through the heated wall and tend to offset Professor Bankoff's[4] apprehension that liquid capillary pressure tends to deactivate a cavity during the interval between bubble release and the formation of a new one.

RECOMMENDATIONS

In view of the above analysis, it is recommended that:

1. The sheath of rod-type heaters immersed in potassium be drilled by Elox or other methods to a depth of ≥ 0.008 in. (diameter ~0.004 in.) and "sandblasted" to peen the mouth to a diameter of 0.002 in. (Fig. 18c). These holes should be drilled ~1/4 in. downstream from the beginning of the heated length.

2. In tubular boilers heated by clamshell-type heaters, holes can be drilled through the wall and subsequently welded and "sandblasted" as shown in Fig. 18a; and alternate cavity is shown in Fig. 18b. Preferably, the cavities should be in the small-diameter tube section of the preheater to establish two-phase flow prior to boiler entry. Nucleation should be initiated in a small-volume section of the loop, thus minimizing the pressure perturbations caused by nucleation.

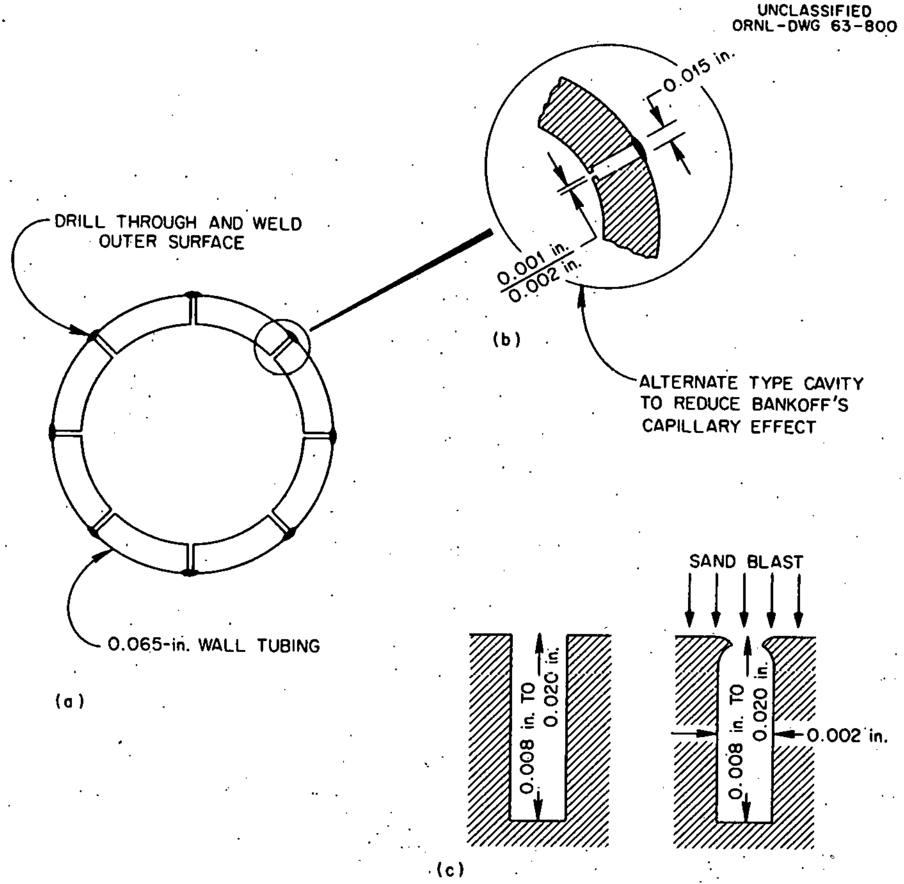

Fig. 18. Recommended Cavity Sizes and Shapes for Nucleation in a Boiling Potassium System.

3. Delineate for potassium a curve similar to that in Fig. 2 and regime III portion of Fig. 4.

4. A program of evaluating the superheat required to initiate boiling and to test Eq. (3) with potassium would be of interest in the study of liquid-metal boiling.

The assumptions and correlations presented here are admittedly oversimplified; they are primarily presented to emphasize the difference between two-phase liquid-metal heat transfer and two-phase heat transfer for ordinary fluids. It is hoped that this paper will provoke more thought and effort for a more quantitative study of boiling-liquid-metal heat transfer.

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