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FILM BOILING OF R-11 ON LIQUID METAL SURFACES

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An interesting problem is the effect of an immiscible liquid heating surface on the process of film boiling. Such surfaces raise questions concerning interface stability to disturbances, effects of gas bubbling, and vapor explosions in layered systems. The specific motivation for this study was to investigate film boiling from a liquid surface with application to cooling of molten reactor core debris by an overlying pool of reactor coolant. To investigate this phenomenon, an apparatus consisting of a nominal six-inch diameter steel vessel to hold the liquid metal and boiling fluid was constructed; coolant reservoirs, headers, controllers, and allied instrumentation were attached. A transient energy balance was performed on the liquid metal pool by a submerged assembly of microthermocouples in the liquid metal and an array of thermocouples on the wall of the test vessel. The thermocouple data were used to determine the boiling heat flux as well as the boiling superheat. On an average basis, the deviation between the prediction of the Berenson [4] model and the experimental data was less than one percent when Berenson was corrected for thermal radiation effects. Evidence from visualization tests of R-11 in film boiling over molten metal pools to superheats in excess of 600 K supports this conclusion.

1. Introduction

An interesting problem is the effect of a liquid heating surface on the process of film boiling. Such surfaces raise questions concerning the nature of nucleation sites, surface stability to the effects of disturbances originating in the boiling liquid, effects of gas bubbling on boiling fluxes, and the mechanisms of vapor explosions in layered systems. The first step in investigating such processes was to look at the liquid-liquid boiling process in the absence of such effects. Several experimental studies have appeared in the literature concerning the phenomenon of one liquid boiling

over another more dense hot liquid. Most have been concerned with modest heater superheats which have limited the investigations to nucleate boiling [1,2]. One investigation relied on visual determination of mass balance to infer the film boiling heat flux [3].

2. Background

Recently, film boiling from solid surfaces has attracted considerable attention and the literature is quite extensive. Most studies have treated boiling from wires, spheres, or plates. In spite of its application to cooling of molten reactor core debris, mechanisms of vapor explosions, and molten core-concrete erosion, only a few have concerned themselves with boiling from a liquid surface.

One of the first such investigations was performed by Gordon et al. [1]. In their experiments, they measured the heat flux for water, methanol, and ethanol in nucleate boiling over shallow layers of reagent grade triple distilled mercury. They were only able to achieve superheats from 4 to 57 K in their experiments and did not achieve the critical heat flux for these fluid pairs, i.e., the heat flux continued to monotonically increase with increasing mercury temperature. They reported that the mercury surface exhibited violent oscillations which increased the actual interfacial surface area for heat transfer as ΔT_{sat} increased, resulting, at the highest heat fluxes, in a violent churning motion of the mercury surface with mercury drops being entrained into the lighter fluid, then falling back. They concluded that higher maximum heat flux was possible for liquid-liquid nucleate boiling than liquid-solid nucleate boiling due to the surface area enhancement effect.

Novakovic et al. [2] reproduced Gordon's experiments, improving on the previous experimental techniques. They investigated nucleate boiling once again with saturated ethanol and water boiling over shallow layers of triple distilled mercury. In their tests, they only achieved boiling superheats up to 45 K, once again limiting themselves to the region of nucleate boiling less than the critical heat flux. They claimed that their improved apparatus and experimental technique eliminated spurious

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contributions to the boiling heat flux, resulting in lower measured boiling heat flux than Gordon for the same superheat; they concluded that required surface superheats for a specific heat flux were larger for boiling on a mercury surface than for boiling on other metallic surfaces. Nevertheless, neither Gordon nor Novakovic investigated the film boiling regime.

Henry et al. [3] performed an experimental study of minimum film boiling for liquid-liquid systems. Quantitative measurements and visual observations were made of liquid-liquid film boiling of thin layers of saturated R-11, R-22 and water on a hot mercury pool. The cooling rate of the mercury due to the film boiling quench as well as visual observations enabled determination of when the lower limit of film boiling was achieved. The temperature of the mercury pool, as well as a mass balance on the boiling fluid from photographic data, enabled an estimation of the boiling heat flux and superheat. Visual observations of the minimum temperature for these fluids in liquid-liquid film boiling appeared to compare well with the calculated results from Berenson [4]. The investigation did produce the first measured film boiling curves on a liquid surface to superheats over 200 K. The data agreed reasonably well with earlier solid surface measurements by Hosler and Westwater [5] and Lao [6] using Freon 11 as the boiling liquid.

The purpose of the present study was to investigate liquid surface film boiling and, in particular, to design an apparatus which would operate at high surface superheats to examine surface stability effects and to measure film boiling curves under these high temperature conditions. This latter aspect has received little attention from other investigators. It was intended to extend the range of boiling superheat from a maximum of 200 K achieved by Henry et al. to at least 600 K. Also, to augment the sparse existing data base, quantitative direct measurements of liquid-liquid film boiling were to be made.

3. Experimental Apparatus

To investigate the phenomenon of liquid-liquid film boiling, the experimental apparatus described below was constructed [7]. It consisted of a nominal six-inch, schedule 10, stainless-steel cylindrical vessel, 45 cm high, to hold the boiling fluid pair, coolant reservoirs, heaters, controllers, and allied instrumentation. The cylinder was open to the atmosphere and the tests were performed at ambient atmospheric pressure. The boiling vessel, reservoirs, and piping were insulated with Fibrefrax insulation to minimize heat losses. The reservoir had a solenoid operated valve for release of a prescribed volume of coolant through a downcomer and into the boiling vessel. The bottom of the downcomer had a splash plate to prevent penetration of the coolant flow into the liquid metal surface. The test vessel was welded above a track along which a thermo-

statically controlled heater was mounted. The heater could be slid away and a four-inch thick base of insulation moved into place to insulate the test section pool. The vessel was insulated radially with a three-inch thick cylinder of insulation as well.

A vertical assembly consisting of eight calibrated thermocouples (diameter = 0.16 cm) was bolted to a vertical traversing mechanism aligned along the centerline axis of the test section. Seven of the thermocouples were immersed into the liquid metal pool, one immersed in the boiling coolant layer. Four of the seven thermocouples were concentrated in the top one centimeter of the pool, the other three at depths of three, five, and seven centimeters below the liquid metal pool surface. The thermocouples extended nearly 2.5 centimeters from the thermocouple assembly horizontally along the isothermal plane in order to minimize conduction errors. The liquid metal pool was measured to be essentially isothermal, with a maximum measured temperature difference across the pool of approximately 10 K, essentially eliminating conduction errors in the thermocouples. An electrical continuity probe was mounted to the thermocouple assembly. The assembly was lowered into the liquid metal until the continuity probe detected the pool surface. At this time, the thermocouples were precisely positioned at predetermined locations in the pool below the surface. A schematic of the experimental apparatus is shown in Figure 1.

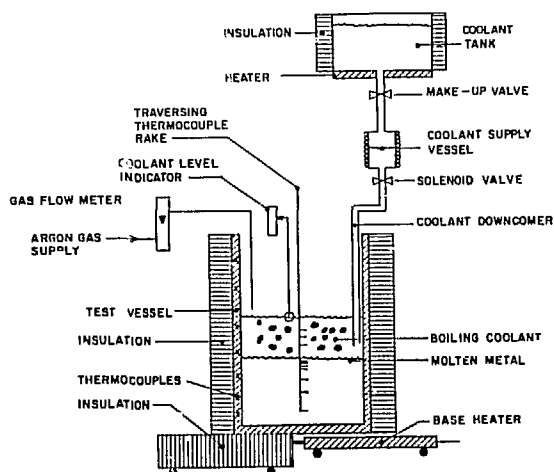


Fig. 1 Schematic of Experimental Apparatus

A float-type device was lowered into the test vessel for determination of the depth of the boiling fluid. As the coolant was boiling, the float device indicated the coolant level, and fluid was introduced to the test section at a controlled rate from a secondary reservoir to replenish the boiled-off coolant. This maintained a constant depth of the boiling fluid

for the duration of the experiment.

The metals used for the liquid metal pool were Wood's metal, bismuth and lead. Temperature-dependent thermophysical properties for these liquid metals were curve-fitted and stored in the data analysis software. Preliminary analysis of experimental data revealed that the temperature distribution near the top surface of the liquid metal pool could be approximated by an exponential function of depth below the boiling interface while the bulk of the liquid metal pool was well-mixed and nearly isothermal.

4. Experimental Procedure

The experiment was begun by first placing the heater under the vessel bottom and heating the liquid metal to a specified initial temperature. An argon cover gas was injected into the test vessel to minimize surface oxidation of the liquid metal during the heat-up phase. The thermocouple assembly was installed and lowered into the metal pool until an electrical continuity probe indicated the correct position in the melt. Once all test conditions had been satisfied, the heater was removed and the base insulation was slid into position and raised to the vessel bottom by cam rollers. The liquid level device was then installed, the thermocouples connected to the appropriate data acquisition channels, and data acquisition initiated.

Data were acquired by routing the thermocouple output through a 150 F Rycal reference junction to a Series 620 Neff Multiplexer. The Neff data channels were individually calibrated to $\pm 5 \mu\text{V}$ over the range 0-50 mV. The data were then stored on disk on a Hewlett Packard Series 1000 minicomputer and on magnetic tape for further data analysis. The Neff measurement channels were individually filtered at 10 Hertz by low pass filters and data acquisition usually performed at 25 Hertz.

For the first five seconds, data were acquired without boiling to get a measurement of the enthalpy change of the liquid metal pool due to boundary heat losses through the insulation. Subsequently, the solenoid valve was actuated, releasing an initial charge of two liters of saturated coolant through the downcomer in the test vessel and onto the liquid metal surface. This initial fill process usually took five seconds. Boiling of the coolant and cooling of the liquid metal proceeded as a transient quench process from the initial prescribed temperature until the pool began to solidify. Once solidification began, data acquisition was terminated. The data analysis was applicable only to the portion of the quench data during which time the entire pool was molten, usually a period of approximately ten minutes.

The thermocouple raw data were integrated over a variable time interval to numerically smooth the temporal fluctuations in the data.

The thermocouples in the liquid metal were associated with a finite volume of the melt and were used to perform a central difference lumped-parameter energy balance on the liquid metal to determine the pool-averaged heat flux per unit surface area over the specified time interval, q'' . The surface superheat, ΔT_{sat} , was likewise calculated by extrapolating the concentration of thermocouples at the surface to the boiling interface. The quantities q'' and ΔT_{sat} were used to construct the boiling curve during the quench. The major parameters used in the experiments are shown below.

Experimental Parameters

Melt Composition	Bismuth, lead, Wood's metal
Melting Temperature	545 600 345 K
Coolant	R11
Melt Depth	8 cm
Coolant Depth	10 cm
Melt Temperature	Up to 900 K
Coolant Temperature	Saturation
Data Rate	25 Hz
Pressure	Ambient
Pool Diameter	15 cm
Cover Gas	Argon

5. Discussion of Experimental Results

The results to be discussed include both visual/photographic observations of the experiments performed as well as quantitative data in the form of heat flux vs. superheat boiling curves. These will be discussed in detail below.

5.1 Visual/Photographic Observations of the Liquid-Liquid Boiling Interface

Initial experiments performed were photographic visualization studies of the boiling interface between the liquid metal melt and the boiling R-11 coolant. The test section for these tests was a 7.5 cm diameter quartz glass vessel. The test section was heated from below, insulated at the sides, and open at the top. A pool of liquid metal 8 cm deep was formed and raised to as high as 900 K. The test section was left on the heater and a layer of R-11 of equal depth was deposited over the metal. Visual and photographic observations of the ensuing boiling interactions were conducted. It was found that a very stable, quiescent interface was observed between the liquids with no apparent interfacial disturbances or mixing. This was in spite of the vigorous hydrodynamics in the R-11 above, induced by the film boiling process. There was no evidence of any liquid-liquid contacts across the boiling interface. A low amplitude rocking motion at the top of the liquid metal was observed, indicating that the pool was still molten. In these visualization tests, no differences from film boiling on solid surfaces was observed, suggesting that the film boiling heat flux of R-11 on liquid metal melts may be well-characterized by classical film boiling on a flat, horizontal surface [4,8].

5.2 Quantitative Liquid-Liquid Film Boiling
Data and Comparison to Berenson's Model [4]

Fully-instrumented liquid-liquid film boiling tests with R-11 and the three liquid metals specified were subsequently conducted in the cylindrical steel test vessel previously discussed. For the tests to be presented, initial temperatures of the melts were as high as 900 K, representing a boiling superheat of up to 600 K. The results of the experiments with Wood's metal, bismuth and lead are shown in Figure 2. In these data, a single data point represents a time-integration over a period of 30 seconds. In all these cases, the R-11 coolant was saturated. These data are believed to be the first liquid-liquid boiling data to go to superheats in excess of 600 K. In Figure 2, the data are compared to the prediction of the classical Berenson film boiling model [4], in which the film boiling heat transfer coefficient is given as,

$$h_B = 0.425 \left[\frac{3 k_{vg} \rho_v (\rho_l - \rho_v) h_{fg}^*}{\mu_v \Delta T (\sigma/g\Delta\rho)} \right]^{1/4} \quad (1)$$

The modified latent heat of vaporization, h_{fg}^* , is given by $h_{fg}^* = h_{fg} + 0.4 C_p \Delta T$.

A film boiling model similar to that of Reference [4] was developed by Hamill and Baumeister [8]. The form of the film boiling heat transfer coefficient developed in [8] differed from Equation 1 only in the magnitude of the prefactor constant, 0.41, and a higher modified latent heat of vaporization, $h_{fg}^* = h_{fg} + 0.95 C_p \Delta T$. Comparative analyses were performed with the film boiling models of [4] and [8] in order to determine the deviation between the two models over the temperature range of the present experiments for R-11. It was determined that over the range of boiling superheat of 300-600 K, the Hamill-Baumeister film boiling heat transfer coefficient exceeded that of Berenson by 5-8%, respectively, not including any correction for radiation heat transfer. On the basis of the degree of agreement evident, the data will only be compared to the Berenson model [4] in this discussion. However, the observations of this comparison should be applicable to the Hamill-Baumeister model [8] as well, within the limits of deviation just discussed.

In the comparison presented in Figure 2, the calculated film boiling heat transfer was augmented by a radiation correction which assumed gray body radiation between parallel surfaces. In this fashion, the calculated film boiling heat transfer is given by,

$$q''_{calc} = h_B \Delta T_{sat} + \frac{3}{4} q''_{rad} \quad (2)$$

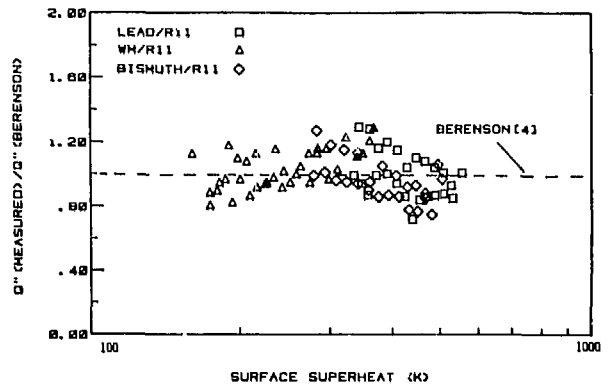


Fig. 2 Measured Liquid-Liquid Film Boiling Heat Flux of R-11

In these calculations, it was found that the radiation correction accounted for approximately 4% of the total heat flux at $\Delta T_{sat} = 200$ K and approximately 13% of the total heat flux at $\Delta T_{sat} = 500$ K.

The data were found to be in excellent agreement with the Berenson film boiling model when corrected for radiation effects. On an average basis, the data for the three liquid metals and R-11 had a deviation of less than one percent from the Berenson model prediction, although the point-by-point scatter in the data is as much as $\pm 20\%$.

5.3 Satisfaction of Minimum Film Boiling Requirements

The visual observations as well as the excellent comparison of the experimental data to Reference [4] suggest that all the experiments were in the stable film boiling regime. Berenson derived an expression for the minimum temperature difference to support film boiling by simply dividing a solution by Zuber [9] for the minimum heat flux by his film boiling heat transfer coefficient.

Recognizing that Berenson's formulation was rigorously applicable only for an isothermal surface, Henry [10] proposed a correlation of available experimental data to extend the applicability of Berenson's formulation by including the effects of transient surface wetting and evaporation of a microlayer of fluid which may be left behind after liquid-liquid contact. The minimum film boiling temperature difference thus correlated was,

$$\frac{T_{min} - T_{min,i}}{T_{min,i} - T_l} = 0.42 \left[\frac{k_{\rho c_l}}{k_{\rho c_w}} \right]^{1/2} \left(\frac{h_{fg}}{c_p (T_{min,i} - T_l)} \right)^{0.6} \quad (3)$$

where $T_{min,i}$ refers to the isothermal surface minimum film boiling temperature derived in Reference [4], T_l is the liquid temperature, and T_{min} is the minimum film boiling temperature

allowing for the non-isothermal effects just specified.

Baumeister and Simon [11] developed a prediction technique for the Lienedrost temperature which they found to also apply to prediction of the minimum film boiling temperature for pool boiling of saturated liquids on smooth, clean surfaces as well. Their model included the effects of the solid-liquid surface energy ratio, the thermodynamic critical temperature, and heat conduction in the heater surface. In this analysis, it is assumed that their variable, A, is the atomic number of the heater surface material and not the atomic mass. Their equation has not been included here because it appears to contain dimensional constants. The reader is referred to Reference [11] for the details.

Calculation of the minimum superheat required to support film boiling of R-11 in these experiments by the models of Berenson [4], Henry [3], and Baumeister and Simon [11] resulted in a predicted minimum wall superheat of 90K, 125K, and 150K, respectively. These requirements were satisfied in the experiments performed, as seen in Figure 2, in which the minimum wall superheat achieved was greater than 160K.

3.4 Correlation of Experimental Data

Recently, Klimenko et al. [12,13] have proposed a general correlation for film boiling heat transfer. The form of the correlation is such that the Nusselt number is presented as a function of the Archimedes number, Prandtl number, and Jacob number (see [12] for a definition of these quantities). The experimental data reported in the present paper satisfy the conditions that the Archimedes number is less than 10^8 and the Jacob number is greater than 0.7. For this regime, Klimenko recommended the following correlation:

$$Nu = 0.19 Ar^{1/3} Pr^{1/3} \quad (4)$$

The present experimental data were analyzed in a form consistent with Klimenko's correlation and the authors arrived at the following correlation:

$$Nu = 0.177 Ar^{1/3} Pr^{1/3} \quad (5)$$

The latter relation is recommended on the basis of less scatter in the data base. The form of the correlation suggests that the film boiling heat flux is linearly proportional to the surface superheat. Upon investigation, it was found that the present data do indeed exhibit the linear relationship inherent in Equation 5, over the range of superheat investigated. Klimenko's correlation, however, also predicts dependencies upon $\Delta T_{sat}^{1/3}$ and $\Delta T_{sat}^{1/2}$ in other regimes of Archimedes and Jacob number. If it is desired to resort to a correlation, Equation 5 is recommended for $Ar < 10^8$ and $Ja > 0.7$. However, we find our data agree well with the established film boiling models [4,8] when corrected for the radiation contribution to the heat transfer, and

we recommend that these models be used for application to other fluids.

5.5 Comparison to Existing Data

In addition to the experimental data taken in this study, there exist several additional data sets in the literature with which to compare these results. Two of these were restricted to film boiling of R-11 on solid surfaces, namely Hosler and Westwater [5] and Lao et al. [6]. The third study specifically addressed liquid-liquid film boiling, that of Henry et al. [3]. These data are shown in Figure 3 along with the present data and the Berenson model corrected for thermal radiation effects.

Examination of the data reveals that all four data bases are in general agreement with the trend of the Berenson film boiling model. The present data, in addition to their good agreement with the referenced film boiling models, have been extended to higher superheats than previous measurements. The only other liquid-liquid film boiling data [3] lie somewhat higher in magnitude with greater scatter than the present data. In spite of these observations, the data of Reference [3] are encouraging with respect to the reasonable agreement in magnitude with both the present data and Reference [4], as well as the available minimum film boiling correlations that have been referenced [4,9,10,11].

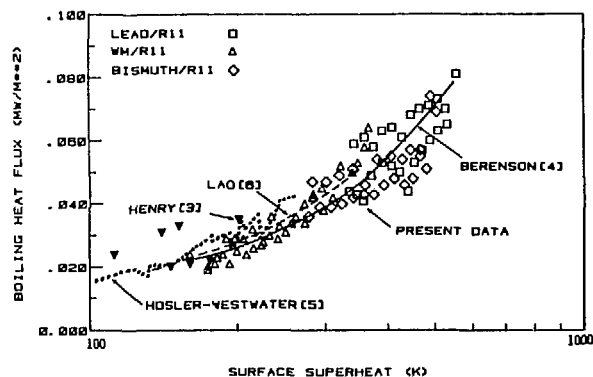


Fig. 3 Comparison of Measured Film Boiling Data (R-11) to Berenson Film Boiling Model [4]

6. Conclusions

Film boiling heat transfer between unmixed and immiscible pairs of overlying liquid layers has been investigated. In this paper, the results of R-11 in film boiling over molten liquid metals have been reported and the following observations and conclusions can be made:

- (1) Visual observations indicate that R-11 boils stably on deep molten metal pools

with no interfacial liquid-liquid contacts or disturbances [7].

- (2) The present liquid-liquid film boiling data agree well with the liquid-liquid data of Henry et al. [3] and the film boiling models of Berenson [4] and Hamill and Baumeister [8] when corrected for thermal radiation.
- (3) The data agree well with other stable film boiling data and exceed the required minimum superheats of both isothermal and non-isothermal surface minimum film boiling criteria [4,9,10,11].
- (4) Under no circumstances were conditions approaching explosive or even violent boiling interactions observed, as has been observed by the authors with water under similar conditions [7]. Understanding the reasons why may help to explain the mechanisms of vapor explosions.

Future work on this subject will concentrate on the effects of oxidized melts, alternate boiling fluids, porous solid surfaces, and non-condensable gas injection.

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