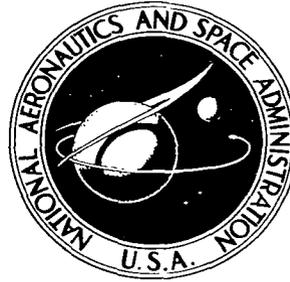


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METASTABLE LEIDENFROST STATES

*by Kenneth J. Baumeister, Robert C. Hendricks,
and Thomas D. Hamill*

*Lewis Research Center
Cleveland, Ohio*





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SUMMARY

Experiments demonstrate that the lower limit of the plate temperature necessary to sustain Leidenfrost film boiling of a water drop is very near the saturation temperature of the liquid. Starting with a water drop on a hot plate in the stable film-boiling regime, cooling the plate slowly, and taking care to eliminate gross perturbations of the system, the drop can remain in the film-boiling state for plate temperatures as low as the saturation temperature. For plate temperatures in the neighborhood of the saturation temperature, the drop collapses onto the plate surface without nucleation.

This observation corroborates theoretical considerations and modifies the current concept of the so-called Leidenfrost point prevalent in the literature. The present cool-down experiments indicate that the Leidenfrost phenomenon can terminate over a range in temperatures from those reported in the literature down to the saturation temperature depending on the characteristics of the heater surface and system perturbations. This perhaps explains the wide range of values reported for the so-called Leidenfrost point by different investigators. This may also explain the rather annoying anomaly that the same piece of equipment can yield different values of the Leidenfrost point.

In essence, the so-called Leidenfrost point of a liquid drop is a function of the environmental vibrations (natural or induced) and surface conditions (roughness and thermal diffusivity). Thus, for a smooth surface isolated from surrounding vibrations, Leidenfrost film boiling of liquid drops can exist for plate temperatures very near the saturation temperature of the liquid.

INTRODUCTION

If a quantity of liquid is placed on a sufficiently hot plate, the bottom surface of the liquid will evaporate so quickly that the liquid will "float" on a cushion of its own vapor. This state of affairs is referred to either as film boiling or the Leidenfrost phenomenon.

Film boiling falls into two broad categories (see fig. 1):

- (1) Bulk film boiling, characterized by a continuous amount of liquid resting on the plate (fig. 1(f))
- (2) Leidenfrost film boiling, characterized by a discrete amount of liquid resting on a plate

The discrete regime includes drops and small puddles of liquid, as shown in figures 1(a) to (e).

This report is concerned solely with the Leidenfrost boiling of drops (figs. 1(a) to (c)) whose geometric characteristics are now discussed. For very small volumes (fig. 1(a)) the shape of a liquid drop is nearly spherical. With larger volumes (fig. 1(b)) a drop tends to flatten out into a disk. For further increases in volume, the drop thickness tends toward an asymptotic value (fig. 1(c)). This large drop, called an extended drop, is essentially a small puddle. In addition, for very large volumes, bubble breakthrough occurs (figs. 1(d) and (e)).

According to reports in the literature (refs. 1 to 3), a minimum plate temperature exists below which Leidenfrost boiling terminates. This minimum plate temperature is called the Leidenfrost point of the liquid. A certain amount of ambiguity exists as to the exact experimental meaning of the Leidenfrost point. In some experimental studies, the Leidenfrost point is taken to be that plate temperature below which it is impossible to sustain film boiling when a drop is placed on the plate. This definition is poor because the method of delivering the drop onto the plate affects the results. Different experimenters will measure different Leidenfrost points for the same liquid, since each experimenter has his own unique way of depositing the drop on the plate.

A more meaningful way of observing the termination of Leidenfrost film boiling is to start with a drop already in a stable Leidenfrost state and then to cool the plate slowly. This technique was mentioned in reference 4. The rate of cooling is a quantity that can be defined precisely, thereby eliminating the vagaries associated with the method of placing the drop on the plate. However, even the cool-down approach will not give a unique value for the Leidenfrost termination plate temperature unless care is taken to eliminate gross perturbations of the system. Examples of gross perturbations are very rough plate surfaces, plate vibrations induced by machines in the immediate surroundings, specks of dirt entrained in the drop, large circulation currents within the drop, and drop oscillations.

Theoretical considerations to be discussed in the next section indicate that the lower limit of the Leidenfrost phenomenon is the saturation temperature of the liquid. The purpose of this study was to verify theoretical observations and to measure the Leidenfrost termination temperature of water at atmospheric pressure.

The experiments indicated that the so-called Leidenfrost point is not a unique thermodynamic or state property of the liquid since the termination or collapse of steady-

state film boiling can occur over a wide range of plate temperatures. A motion-picture film supplement C-244 that illustrates the foregoing results is available on loan. A request card and a description of the film are included at the back of this report.

SYMBOLS

C_p	specific heat of vapor at constant pressure, J/(g)(°C)
g	acceleration of gravity, cm/sec ²
k	thermal conductivity of vapor, W/(cm)(°C)
r_o	maximum radius of drop, cm
T_p	temperature of hot plate, °C
T_s	saturation temperature of liquid, °C
v	drop volume, cc
δ	gap thickness, cm
ϵ	characteristic roughness height, cm
λ	heat of vaporization, J/g
λ^*	modified heat of vaporization, $\lambda^* = \lambda \left[1 + \frac{7}{20} C_p (T_p - T_s) / \lambda \right]$, J/g
μ	absolute viscosity of vapor, g/(cm)(sec)
ρ	vapor density, g/cm ³
ρ_ℓ	liquid density, g/cm ³

THEORETICAL PREDICTION OF THE LEIDENFROST POINT

The concept that there need be no lower limit to the Leidenfrost phenomenon other than the saturation temperature is the subject of this discussion. In reference 5, a theoretical analysis results in an expression for the gap thickness beneath a liquid drop in film boiling. For the case of a perfectly smooth surface, the steady-state gap thickness beneath a liquid drop is given by

$$\delta = \left[\frac{3\pi k (T_p - T_s) \mu r_o^4}{2\rho\rho_\ell v g \lambda^*} \right]^{1/4} \quad (1)$$

where the relation of r_o to v for a water drop is given in reference 6.

Figure 2 presents the calculated steady-state gap thickness for water as a function of the temperature difference $T_p - T_s$ for drop volumes of 1 and 0.1 cubic centimeter. Clearly, the theoretical calculations presented in this figure show that the gap thickness remains finite even for temperature differences as low as 1°C . Thus, within the limits of the analysis, no a priori reason exists to explain why film boiling could not occur for all plate temperatures greater than the saturation temperature of the liquid.

In real systems, however, film boiling can terminate because of surface roughness or system perturbations. Consider figure 2. All real surfaces have some roughness associated with them; that is, the perfectly smooth surface assumed in the derivation of equation (1) does not exist in reality. In addition, the derivation of equation (1) was based on the assumption that the liquid is in an environment in which there are no surrounding vibrations or system perturbations.

The experimental results of reference 7 indicate how system perturbations and surface roughness could effect the Leidenfrost termination point of a drop. Photographs in reference 7 of liquid drops in film boiling lead to the following conclusions that are germane to this analysis.

For a sufficiently smooth surface (e.g., a chromium surface polished to a mirror finish), a gap thickness with no liquid-solid contact between the drop and the plate can exist; although for rough surfaces (e.g., a rough aluminum plate) an intermittent local liquid-solid contact occurs at various points beneath the drop. Furthermore, if system perturbations such as vibrations can occur, intermittent local liquid-solid contact between the drop and the plate will exist even on a very smooth surface.

Consider the analytical results displayed in figure 2. If the plate temperature is decreased, the vapor gap shrinks in accordance with equation (1). For temperature differences in the neighborhood of 1°C , the computed gap thickness is approximately 0.0025 centimeter. If all surface protrusions ϵ due to surface roughness are much less than this value, it seems reasonable that film boiling can be sustained even at this very low temperature difference. This theoretical observation is shown to be correct in the EXPERIMENTAL RESULTS section of this report.

APPARATUS AND EXPERIMENTAL PROCEDURE

The experimental procedure consisted of placing about 5 cubic centimeters of water on a slightly tapered plate at about 300°C . This plate temperature is hot enough to ensure stable film boiling. The gas fire, which heated the plate, was then extinguished, and cooling water was introduced at the base of the hot plate. When stable coolant flow had been established, the drop was permitted to evaporate to less than 2 centimeters in diameter. While the temperature difference $T_p - T_s$ was greater than 140°C , drop

vibration, internal circulation, oscillation, and rotation were damped out mechanically by inserting a wire into the drop. In some cases, these random drop motions damped out naturally.

A schematic drawing of the stainless-steel hot plate used in these experiments is shown in figure 3. The plate was surrounded by a concentric transite ring and heated from below by a gas flame. A polished 2° conical surface was machined on the center of the heated surface to facilitate drop retention. A water-cooled coil was attached to the base of the stainless-steel plate to control the surface cooling rate.

A 30-gage Chromel-Alumel open-ball-type thermocouple was placed 0.08 centimeter below the heater surface. The leads emerged from a 0.116-centimeter pilot hole in the center body and were contained in a cement-covered slot. The reference junction was at the temperature of melting ice.

The thermocouple signal was recorded on a self-balancing strip-chart potentiometer. The system was calibrated by using hot water, a thermometer, and a second thermocouple. The measurements appear to be accurate within 2° C.

The surface temperature was obtained by linear extrapolation. For example, consider a typical drop that "wetted" the surface. With an average coolant temperature beneath the plate of 14° C, and a thermocouple temperature of 88° C, the temperature gradient was 137° C per centimeter. The temperature difference between the thermocouple and the surface was calculated to be 11° C. Within the assumption of steady uniform cooling and the accuracy of the thermocouple, the surface temperature of 99° C ($88^{\circ} + 11^{\circ}$) is the fluid saturation temperature for the ambient conditions at which this experiment was performed.

Drop excursions were monitored manually and recorded by a two-camera system with a common time base. One camera was used to photograph drop motion and the other to monitor the surface temperature history on a strip-chart recorder.

EXPERIMENTAL RESULTS

In order to illustrate the difference between currently accepted concepts of the Leidenfrost phenomenon and those discussed herein, a brief review of the conventional boiling and evaporation curves will be presented. In most drop evaporation studies, a known volume of liquid is placed on a hot plate at a fixed temperature and the total time of evaporation is measured. Experiments are repeated for the same given volume but at different plate temperatures. Such results are plotted as shown in figure 4, that is, total evaporation time as a function of the driving temperature difference. This curve, called the evaporation curve, can be interpreted as the inverse of the conventional boiling curve observed in bulk boiling studies.

As shown in figure 4, two relative extremums are on the curve. The minimum time of evaporation corresponds to the maximum (burnout) heat flux, whereas, the relative maximum evaporation time is the so-called Leidenfrost point. Under normal conditions (placing the drop on a plate at the indicated temperature), the drop will be in one of the following modes of heat transfer for a given plate temperature:

- (1) Film boiling for plate temperatures higher than the Leidenfrost point
- (2) Transition boiling for plate temperatures between burnout and the Leidenfrost point
- (3) Nucleate boiling for plate temperatures between burnout and the boiling incipience
- (4) Free convection and conduction for plate temperatures below that necessary for the incipience of nucleate boiling

Figure 5 is the conventional boiling curve with heat flux plotted against the driving temperature difference. On this curve is plotted an extension of the fully established film-boiling line. Points on this dashed line, hereinafter referred to as the metastable Leidenfrost line, were achieved experimentally in this study by the quasi-steady cool-down technique. Clearly, if a drop in the metastable Leidenfrost condition becomes unstable, the terminal state can be one of the following:

- (1) Transition boiling
- (2) Nucleate boiling
- (3) Free convection and conduction

Furthermore, if a drop is brought quickly into a metastable state and is held there, a vaporization-time curve, depicted by the dashed line in figure 4, is possible.

Motion-picture sequences of the termination of metastable Leidenfrost boiling into the three states just mentioned are shown in figures 6 to 8. In all cases, the drop was placed on a high temperature (550° C) plate and the plate was allowed to cool down to the temperature where termination occurred. The photographic sequence consists of four consecutive frames of the film showing the quiescent Leidenfrost state a few instants before termination and then transition to a final terminal state. The time between pictures is 0.02 second.

Transition-Boiling Termination

The violent reaction characteristic of a drop undergoing a change from the Leidenfrost mode to the transition boiling mode is indicated in figure 6. A probable path of transition is shown in figure 5. In figure 6(a), the drop on a plate at 180° C is relatively quiescent in the Leidenfrost state. Figure 6(b) shows the drop contacting the surface. As more of the surface is contacted, a violent spattering and hissing of the drop takes place (fig. 6(c)). Fragmentation of the drop into small liquid particles can be seen in

figure 6(d). The transition boiling termination has all the characteristics of a small explosion.

Nucleate-Boiling Transition

The transition from the Leidenfrost state to the nucleate-boiling mode is illustrated in figure 7. A 0.94-centimeter-diameter drop in the Leidenfrost state just prior to touchdown is shown in figure 7(a). The surface temperature (127°C) is falling at 2.5°C per minute. If the liquid were wetting the surface with the same temperature difference, nucleate boiling would occur. Figure 7(b) shows the drop starting to wet the surface and vapor pockets forming beneath the drop. Figures 7(c) and (d) illustrate the drop in the terminal nucleate-boiling state.

Free-Convection and Conduction Termination

A transition from the Leidenfrost state directly to a free-convection mode is depicted in figure 8. The top view of a 0.76-centimeter-diameter drop in the Leidenfrost state is shown in figure 8(a). The surface temperature is falling at the rate of 7.5°C per minute. Figure 8(b) shows the drop just prior to touchdown with the surface temperature within 1°C of the saturation temperature. In figure 8(c), the drop falls toward the plate wetting the periphery. The drop continues to wet and comes to equilibrium as shown in figure 8(d). It is important to note that the terminal condition is a pool of liquid in which no nucleate boiling was observed during the transition. Thus, metastable Leidenfrost states can exist very near the saturation temperature.

Persistence of Metastable States

In one particular experiment, the plate was allowed to come to a steady-state temperature a few degrees above the liquid saturation temperature. This was accomplished by shutting off the coolant flow beneath the plate. Under these conditions, the drop stayed in the metastable state for over a minute. The experiment was terminated by resuming the coolant flow beneath the plate.

Mechanically Induced Termination

The Leidenfrost explosion can be induced by impacting or piercing a drop in the Leidenfrost state with a wire. There appears to be some relation between the temperature difference and mechanical impact required to terminate the film-boiling state. To illustrate this point, consider the sequence of photographs of figure 9. Prior to this sequence of pictures a probe piercing the drop and touching the plate failed to induce a Leidenfrost explosion; the plate temperature was 206°C . When the same probe pierced the drop (figs. 7(a) and (b)) at a plate temperature of 200°C , however, the surface became wetted as seen in figure 7(c). Thereafter, the usual reactions (fig. 7(d)) followed, as discussed in the transition-boiling-termination section and shown in figure 6.

A motion-picture film supplement C-244 that illustrates the foregoing results is available on loan. A request card and a description of the film are included at the back of this report.

DISCUSSION OF RESULTS

The present experimental results indicate that the lower limit of the plate temperatures for which Leidenfrost film boiling can exist is very near the saturation temperature of the liquid. However, if oscillations or other system perturbations are allowed to occur, the probability of liquid-solid contact is increased. A momentary liquid-solid contact, induced by some system perturbation, may drain a significant amount of heat from a localized area of the plate. If the thermal diffusivity of the metal is low, the temperature of the cooled area may never recover to its initial value. This leads to a nonuniform evaporation rate under the drop along with a nonuniform pressure (see symmetric pressure field of ref. 5). The nonuniform pressure field aggravates any initial oscillation, thereby escalating the number of liquid-solid contacts. The net result of this chain reaction manifests itself as a termination of quiescent film boiling.

CONCLUSIONS

Using a heated plate and a cool-down technique demonstrated that

1. The lowest plate temperature sufficient to support Leidenfrost film boiling occurs very near the saturation temperature of the liquid.
2. The steady-state film boiling of liquid drops can terminate over a wide range of plate temperatures, depending on the magnitude and nature of the system disturbances.

Three cases are possible on termination of steady-state film boiling:

- a. The drop collapses and wets the plate at a temperature below that required for incipence of nucleate boiling
 - b. The drop goes into nucleate boiling
 - c. The drop goes into transition boiling
3. Film boiling can be terminated artificially under certain circumstances by mechanically perturbing the system.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, January 3, 1966.

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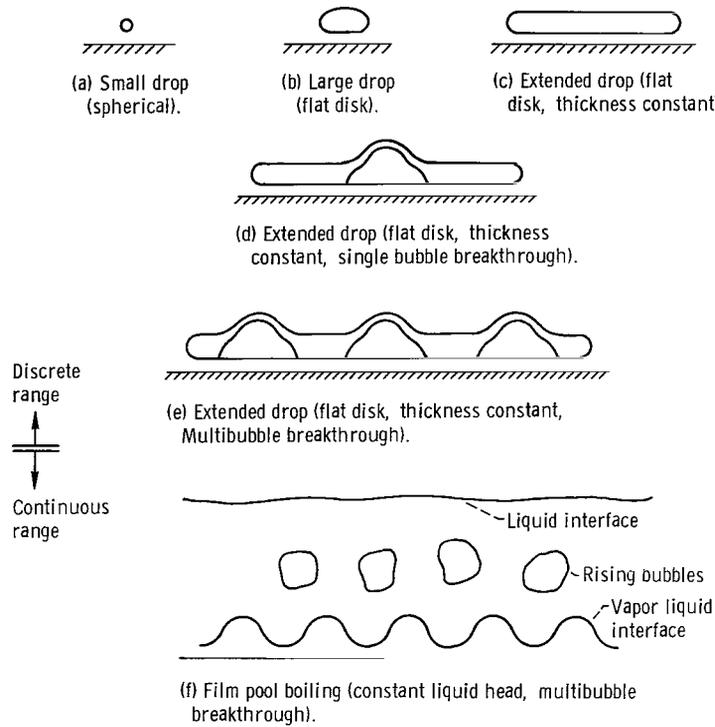


Figure 1. - Film-boiling states of liquid masses.

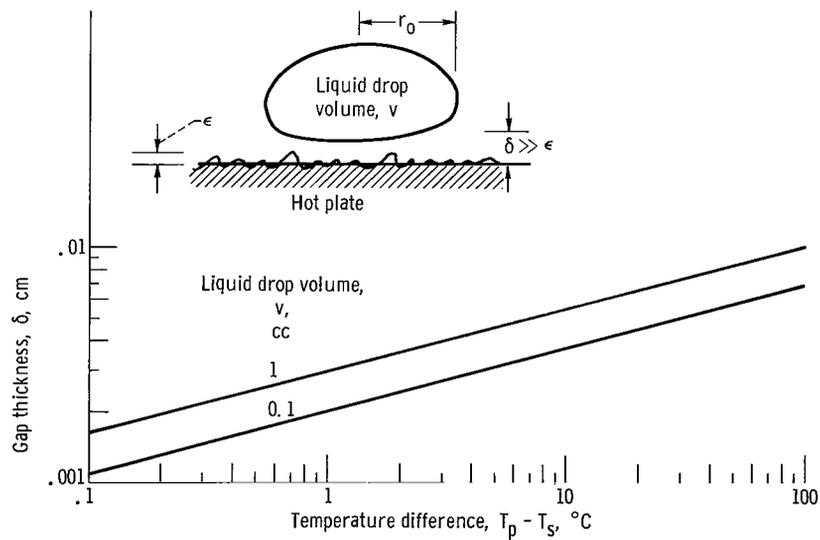


Figure 2. - Gap thickness as function of temperature difference between plate and drop.

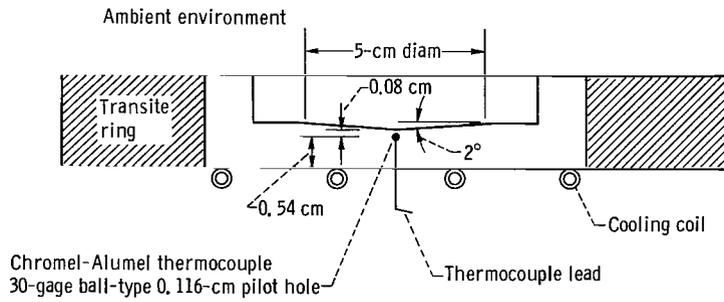


Figure 3. - Stainless-steel hot plate.

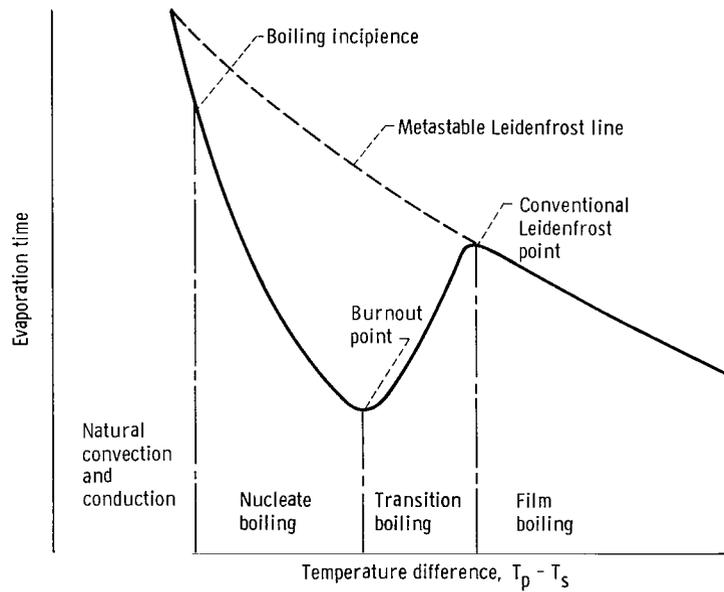


Figure 4. - Evaporation-time curve.

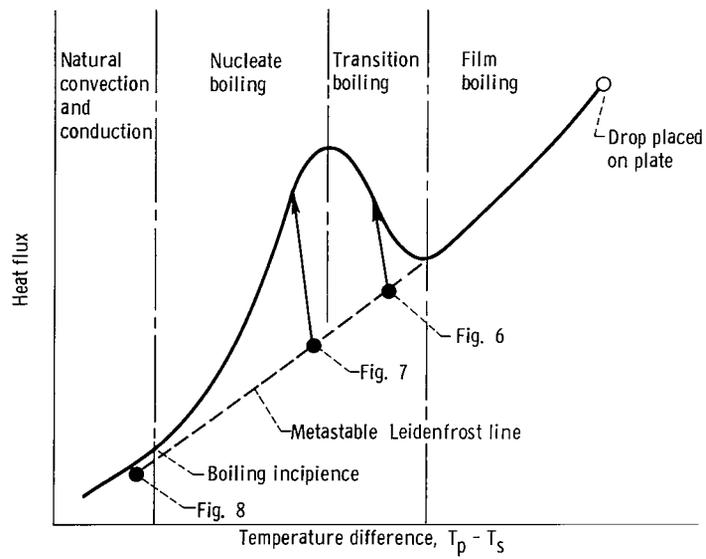
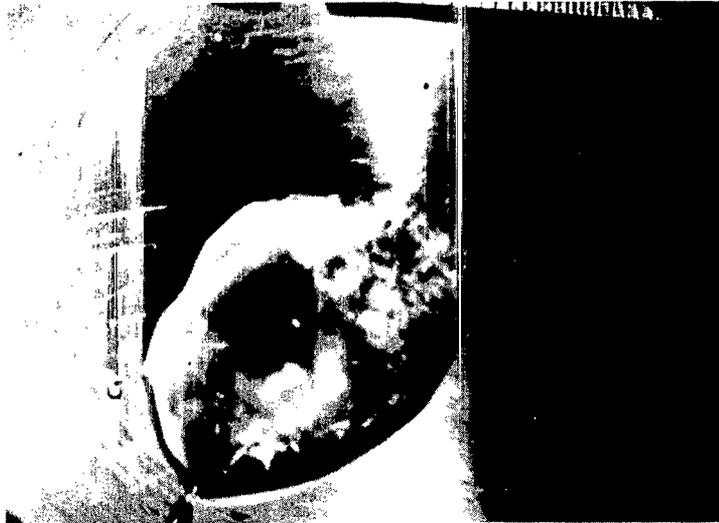
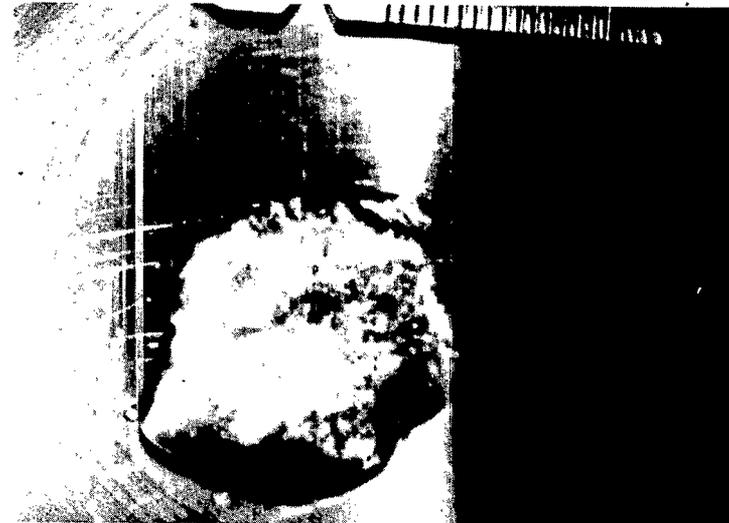


Figure 5. - Conventional boiling curve illustrating termination of metastable Leidenfrost film boiling.



(a) Leidenfrost boiling.



(b) Onset of transition boiling.



(c) Termination of Leidenfrost boiling.



(d) Transition boiling.

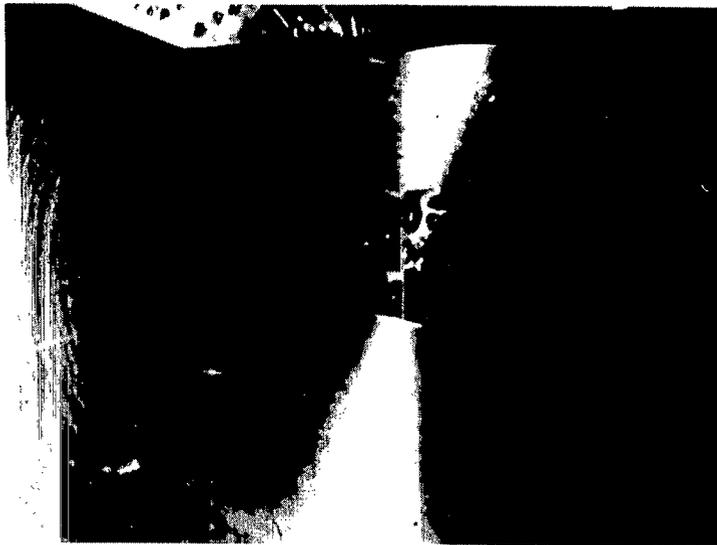
Figure 6. - Termination of steady-state Leidenfrost film boiling in transition-boiling state (hot-plate temperature, 180°C).



(a) Leidenfrost boiling.



(b) Onset of nucleate boiling.

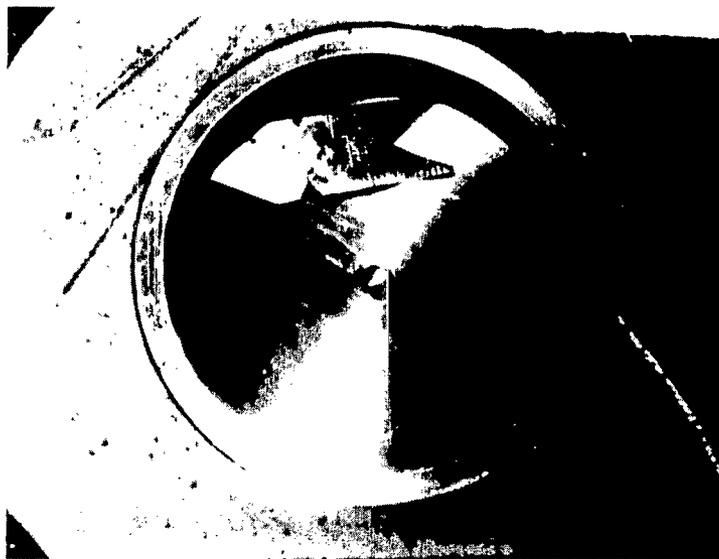


(c) Termination of Leidenfrost boiling.



(d) Nucleate boiling.

Figure 7. - Termination of steady-state Leidenfrost film boiling in nucleate-boiling state (hot-plate temperature, 127° C).



(a) Leidenfrost boiling.



(b) Onset of wetting.



(c) Termination of Leidenfrost boiling.



(d) Liquid "wetting" surface.

Figure 8. - Termination of steady-state Leidenfrost film boiling in free-convection state (hot-plate temperature, $\sim 100^{\circ}\text{C}$).



(a) Leidenfrost boiling.



(b) Onset of transition boiling.



(c) Termination of Leidenfrost boiling.



(d) Transition boiling.

Figure 9. - Mechanically induced termination of steady-state Leidenfrost film boiling.

A motion-picture film supplement C-244 is available on loan. Requests will be filled in the order received. You will be notified of the approximate date scheduled.

The film (16 mm, approximately 16 min, color, sound) shows that metastable Leidenfrost film boiling of water drops can exist for plate temperatures very close to the fluid saturation temperature. Further, through schematic illustrations and selected motion-picture sequences, metastable Leidenfrost boiling is shown to terminate in (1) transition boiling, (2) nucleate boiling, or (3) natural convection-conduction.

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