Abstract

An overview of liquid-vapor nucleation is given. As is demonstrated, a reasonably good qualitative understanding exists of nucleation and incipient boiling, but a general quantitative description is still lacking.

The result of thermodynamic equilibrium across curved liquid-vapor interfaces is presented. The extension of this to include the interaction with idealizations of surface cavities is made to demonstrate how superheat requirements for nucleation will be affected by surface roughness, flow velocity and buoyancy. A successful description of the nucleate boiling process must include a quantitative understanding of nucleation.

Experimental measurements involving high liquid superheats and nucleation delay times are presented to show examples of homogeneous nucleation. Examples of nucleation and boiling on smooth glass substrates and on metal surfaces with various surface roughnesses are presented, with fluids including H₂O, Freon 113, LN₂ and LH₂, at a/g = 1 and a/g = 0.

An appropriate physical description or characterization of heating surfaces for nucleation purposes is still lacking.
Nucleation

Classifications

Pure Substance

- Homogeneous (within bulk)
- Heterogeneous (on container walls)

Mixtures

- Homogeneous (vapor phase out of solution)
- Heterogeneous (gas trapped in container walls)

Empirical Categories -- Boiling Points

- Cryogenic
- "Normal"
- Liquid Metals
With equilibrium (unstable)

Given:

\[ P_t \rightarrow T_s \]

\[ T_v > T_s \]

\[ r^* = \frac{25/P_t}{\frac{(T_v - T_s)h_l}{l_f} - 1} \]

If \( r < r^* \rightarrow \text{collapse} \)

If \( \frac{26}{r^*P_t} \leq 41 \)

If \( r > r^* \rightarrow \text{growth} \)

\[ r^* = \frac{25RT_vT_s}{P_t h_l (T_v - T_s)} \]

\( \Delta T_{s,\text{sup.}} \uparrow \rightarrow r^* \downarrow \)

\( \Delta T_{s,\text{sup.}} \)

271
A - CROSS-SECTIONAL AREA

Lumped model.

Distributed model - Lagrangian coordinates.
Comparison of distributed analysis with experimental run No. 307 - Complete decompression transient, long pipe.
Comparison of distributed analysis with experimental run
No. 307 - Initial decompression transient, long pipe.
\[ T_v - T_s = \frac{2 \sigma RT_s^2}{P \cdot h_{fp} \cdot r} \]

\[ T_s (r = \infty) \]

\[ T_e (y, t) \]

Heating Wall
Transient superheat $\Delta T$ versus time $\tau$ with different heat-fluxes $Q''_{ss}$ (system thin-film/Freon 113, $T_s = 320$ K, $p = 1$ bar). Inception points are marked with (IB).

<table>
<thead>
<tr>
<th>$Q''_{ss}$ [kW/m$^2$]</th>
<th>126</th>
<th>129</th>
<th>133</th>
<th>145</th>
<th>153</th>
<th>195</th>
</tr>
</thead>
</table>

Transient superheat $\Delta T$ versus time $\tau$ with different heat-fluxes $Q''_{ss}$ (system platinum-wire/water, $T_s = 373$ K, $p = 1$ bar). Inception points are marked with (IB).

| $Q''_{ss}$ [kW/m$^2$] | 624 | 496 | 174 | 290 | 267 |
In an attempt to incorporate the influence, on nucleation time, of system variables such as properties and microgeometry of the heated surface and convection effects, a “nucleation factor” is introduced in the equations of the homogeneous nucleation theory, defined by

$$F = (A_{het}/A_{hom})$$  \hspace{1cm} (1)

$A_{het}$ is the work required to activate a nucleation site in a heterogeneous circumstance. The variation in $F$ then corresponds to a boiling inception temperature $T_{IB}$ ranging from $T_{SSI}$ to $T_{hom}$.

For a liquid at a pressure $P_L$ and a superheat temperature $T$, which produces a “normal” saturation pressure $P_s$ (for a vapor bubble $r = \infty$), for thermodynamic equilibrium the critical spherical bubble size $r_c$ is given by

$$\frac{P_s}{P_L} = 1 + \frac{2\sigma}{r_c P_L}$$  \hspace{1cm} (2)

The only assumption in equation (2) is that the system state is far enough from the thermodynamic critical state so that $\nu \gg \nu_L$. A further product of this assumption is that $P_s = P_v$, the vapor pressure within the bubble. For nucleation to occur the bubble formed must exceed the critical bubble size given by equation (2) by at least one molecule. The expression of Fisher [11] for the homogeneous nucleation rate of bubbles of critical size, neglecting the term for the free energy of activation for the motion of an individual molecule of liquid past its neighbors into or away from the bubble surface, is given by

$$J = \frac{nkT}{h} e^{-(A_{hom}/kT)}$$  \hspace{1cm} (3)

where

$$A_{hom} = \frac{4}{3} \pi \sigma r_c^2$$  \hspace{1cm} (4)

Nucleation will occur when a bubble of critical size forms per unit volume in a reasonable time, say $\tau$. This $\tau$ will be termed the nucleation time, and neglects the time required to achieve a steady state of embryo distribution in any transient process. In equation (3) then, $J \propto 1/\tau$, and for our present purposes the proportionality will be taken as an equality. For the heterogeneous nucleation case, replacing $A_{hom}$ in equation (3) by $A_{het}$ of equation (1), with equations (2, 4) and the Clausius-Clapyron equation, and $\nu \gg \nu_L$ the resulting expression relates the nucleation time $\tau$, the nucleation factor $F$, and the superheated liquid temperature $T$

$$\frac{T - T_s}{T_s} = \ln \left( \frac{16\pi \sigma^3 F}{P_c^2 3kT \ln \frac{nkT\tau}{h}} \right)^{1/2} + 1 - 1 \right\}^{-1}$$  \hspace{1cm} (5)
Nucleation factor $F$ versus dimensionless inception temperature for Freon 113 and water ($T = T_S$, $p = 1$ bar)
Comparisons of nucleate pool boiling of LH₂ at atmospheric pressure, a/g = 1.

Schematic of test surface mounting technique.
Heat Flux Decreases With Successive Data Points

- Run 111—Polished Stainless Steel
- Run 122—600 Grit Stainless Steel
- Run 125—280 Grit Stainless Steel

Effect of roughness on heat transfer to liquid hydrogen from horizontal stainless steel surfaces.
Heat Flux Decreases With Successive Data Points

- Run 12°—600 Grit Stainless Steel Surface Heating Upwards
- Run 169—600 Grit Stainless Steel Surface Heating Vertically

Effect of orientation on heat transfer to liquid hydrogen from 600 grit stainless steel surfaces.
Polished Stainless Steel Surface

Heat Flux Decreases With Successive Data Points Except for Solid Symbols Which are for Increasing Heat Flux

- Run 108—Liquid Hydrogen, With Convection Shield
- Run 94—Liquid Nitrogen, With Convection Shield

Effect of liquid on heat transfer from a horizontal polished stainless steel surface.
<table>
<thead>
<tr>
<th>No.</th>
<th>Surface</th>
<th>Orientation</th>
<th>Observations</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Polished S.S.</td>
<td>Upward</td>
<td>6</td>
</tr>
<tr>
<td>2</td>
<td>Polished S.S.</td>
<td>Downward</td>
<td>4</td>
</tr>
<tr>
<td>3</td>
<td>600 Grit S.S.</td>
<td>Upward</td>
<td>3</td>
</tr>
<tr>
<td>4</td>
<td>600 Grit S.S.</td>
<td>Downward</td>
<td>5</td>
</tr>
<tr>
<td>5</td>
<td>Polished Cu</td>
<td>Upward</td>
<td>1</td>
</tr>
<tr>
<td>6</td>
<td>600 Grit S.S.</td>
<td>Upward</td>
<td>3</td>
</tr>
<tr>
<td>7</td>
<td>Polished S.S.</td>
<td>Upward</td>
<td>7</td>
</tr>
<tr>
<td>8</td>
<td>Polished S.S.</td>
<td>Vertical</td>
<td>4</td>
</tr>
<tr>
<td>9</td>
<td>280 Grit S.S.</td>
<td>Upward</td>
<td>3</td>
</tr>
<tr>
<td>10</td>
<td>280 Grit S.S.</td>
<td>Vertical</td>
<td>3</td>
</tr>
<tr>
<td>11</td>
<td>Teflon</td>
<td>Upward</td>
<td>4</td>
</tr>
<tr>
<td>12</td>
<td>Polished Cu</td>
<td>Upward</td>
<td>4</td>
</tr>
<tr>
<td>13*</td>
<td>600 Grit S.S.</td>
<td>Vertical</td>
<td>3</td>
</tr>
<tr>
<td>14</td>
<td>600 Grit Cu</td>
<td>Upward</td>
<td>5</td>
</tr>
</tbody>
</table>

*Different surface than 3, 4, and 6

Average $q/A$ and $\Delta T_b$

Range of $\Delta T_b$

Individual Observations Fell Along the Natural Convection Correlation Shown. The range of Heat Fluxes Observed is not Shown.

$Nu = 0.14(GrPr)^{1/3}$

Initial vapor formation conditions.
Combining, equations (1) and (2), the minimum vapor superheat necessary for the hemispherical vapor bubble to grow is

$$\Delta T_{sat} = T_v - T_{sat} = \frac{2\sigma T_{sat}}{r_c h_{fg} P_g}$$

Approximating the vapor temperature by the heater surface temperature, and considering that for a given surface the value and distribution of $r_c$ are constant, the ratio of the heater surface superheats necessary for the initial formation of vapor of the two fluids is given, from equation (3) as

$$\left(\frac{\Delta T_{sat}}{h_{fg} P_g}\right)_{N_2} = \left(\frac{\sigma T_{sat}}{h_{fg} P_g}\right)_{N_2} / \left(\frac{\sigma T_{sat}}{h_{fg} P_g}\right)_{H_2}$$

The calculated quantity is listed in Table 2 with the measured values from Fig. 10. The comparison is reasonably good, especially for the polished copper surface.
1. Camera
2. Sprocket & Chains
3. Spherical Reflector
4. Mercury Arc Lamp
5. Light Source Housing
6. Thread Rods
7. Lighting Lens
8. Test Surface
9. Relay Mirror
10. Mirror Holder
11. Cross Bar
12. Drop Package
13. Viewing Window
14. "O" Ring
15. Test Tube
16. Inside Dewar
17. Outside Dewar
18. Flange
19. Camera Rail
20. Relay Mirror
21. Camera Lens

Test vessel and package
Note: Symbol "B" Indicates Calibration Point Immediately Prior to Boiling Test. "A" Indicates Immediately Following.

Calibration shift with time (PCG-15) in LN₂
Transient surface temperature, F113, vertical, a/g=1 and = 0

$\text{TSAT (NOMINAL)} \approx 117 ^\circ F$
Transient surface temperature, F113, horizontal-up, a/q=1 and = 0
Transient surface temperature, LN₂, vertical, a/g=1 and =0

\( T_{\text{Sat}} \) (Nominal) = 139°K
<table>
<thead>
<tr>
<th>(Q/A)T</th>
<th>a/g</th>
<th>PCG</th>
</tr>
</thead>
<tbody>
<tr>
<td>7,200</td>
<td>1</td>
<td>29</td>
</tr>
<tr>
<td>15,000</td>
<td>1</td>
<td>29</td>
</tr>
<tr>
<td>28,300</td>
<td>1</td>
<td>29</td>
</tr>
<tr>
<td>15,100</td>
<td>0</td>
<td>29</td>
</tr>
<tr>
<td>28,500</td>
<td>0</td>
<td>29</td>
</tr>
</tbody>
</table>

$T_{Sat \ (Nominal)} = 139^\circ R$

**Transient surface temperature, LN$_2$, horizontal-up, a/g=1 and = 0**
Comparison of transient surface temperature of a vertical surface in F113 and LN₂, a/q=1

\[
T_{\text{Sat}}(\text{Nominal}) = \begin{cases} 
\text{F113} = 117^\circ F \\
\text{LN}_2 = 139^\circ R
\end{cases}
\]
BIBLIOGRAPHY


5.


Robert Hendricks/Lewis Research Center:

In your first set of slides, where you showed a rapid drop in pressure, that rapid drop of pressure is mimicked by the rapid drop in temperature when you did your boiling study. We have done quite a bit on that and found that the temperature beneath the growing bubble drops extraordinary rapidly and recovers in much the same way as the pressure does.

Merte:

Are you talking about the pipe venting problem?

Hendricks:

Yes, that's right.

Merte:

The temperature will not drop until vapor is formed.

Hendricks:

Your temperature didn't drop; your pressure dropped. In our case when we did the nucleate boiling studies, our temperature dropped, but our pressure was constant.

Merte:

The temperature dropped as soon as the vapor bubbles were formed.

Hendricks:

I see; you are in equilibrium.

Merte:

We had thermocouples inserted in the liquid too, and you could see that the vapor formed around the thermocouples eventually. The temperature did indeed drop, because once the bubbles are formed, of course, it is the consumption of the super heat by the growing vapor that drops the temperature of the liquid, which is, I think, what you are referring to.

Hendricks:

Right! When you got your sonic velocities from the experimentation, did you compare that with the equation of state, Martin-Powell, or something like that?

Merte:

Yes.

Hendricks:

Was the comparison good?
Merte:

Yes! Now, this is for the liquid state only; details of this comparison can be found in the cited reference.

Hendricks:

I thought that Hsu's criteria had dual roots for radii?

Merte:

I was looking at the more general problem. He dealt with the specific description of the transient temperature distribution in the boundary layer. He took a given thickness and then treated it as a semi-infinite solid where you ultimately had a step change in the surface temperature. Pete Griffith did work treating the boundary layer as a semi-infinite solid. Rosenhow and Furgel did some work with forced convection using a heat transfer coefficient to describe the temperature gradient at the surface.

Hendricks:

What was the reduced temperature and pressure of the Freon versus water when you did the transient heating?

Merte:

I don't remember that. Do you mean relative to the critical state?

Hendricks:

Yes, because they won't compare unless they are on the same reduced temperature and pressure basis.

Merte:

We did it in terms of the homogeneous superheat; that is divided by the homogeneous superheat.

Hendricks:

I agree, but unless the thermodynamic states are corresponding states, they won't agree.

Merte:

I don't remember what they were.

Hendricks:

We generated a great deal of data with a ribbon heater and liquid hydrogen. I think that the data would probably fall in the center of your curve, but those were for ribbon heaters which may be a little different from the wires. I would like to suggest, for your Q versus Delta T plot, showing the effect of roughness; the initiation of nucleation is of the right order, but when you go up into the high fluxes, the functional relationship crosses back over again, because
you are probably blanketing the surface with vapor, which is equivalent to giving you a higher resistance.

Merte:

Obviously, there is going to be some point when we get to high enough heat fluxes where you get interference between the nucleating sites.

Shigeo Nakanishi/Analex Corporation--Lewis Research Center:

The heating rate appears to influence the level of super heat that the liquid can sustain before incipient boiling begins. Is that correct?

Merte:

Yes, that is correct.

Nakanishi:

In that event, since the available zero-g test time is very short, could all the results tend to be biased toward the high heating rate? If you could compare the very slow heating rate results with a sustained or long term zero-g case, you could possibly have a difference between the short-term and the long term zero-g tests?

Merte:

That was what moved the points on the curve. For example, with the Freon 113, you noticed there was a line, and the data had covered the various points of the line because we varied the heating rate. I agree with what you are implying there; the nucleation point is indeed a function of more than just the surface temperature. It has got to be a function of the temperature distribution in the liquid boundary layer. That is why we are in the process of setting up to make some measurements of the temperature gradient which weren't made previously. I think it is a combination of the temperature distribution in the liquid near the surface and the surface temperature together that govern when this nucleation takes place.