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Onset of Boiling in a Metastable Free Jet

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ABSTRACT

In a thermodynamic metastable fluid jet of superheated water the process of boiling was studied. In this case the onset of boiling was not influenced by nuclei of the wall. The jet is generated in specially designed short nozzles with small diameters by the expansion of water from a pressure of above 1 bar and a temperature above 100 °C to the pressure of the ambient air (approx. 1 bar). During the expansion the fluid suddenly becomes metastable or superheated and single vapor bubbles are formed in the jet. In this experimental arrangement the onset of boiling, the bubble growth and bubble frequency are investigated as functions of the fluid state before the pressure drop in the nozzle. In the superheated fluid two different mechanisms of bubble formation are observed. In the first case the formation process is activated by nuclei in the superheated fluid; this mechanism depends on the superheat and the overexpansion in the nozzle, the gas content and the number of activated particles in the fluid. However, it is independent of the nozzle shape and the jet velocity. The second mechanism depends on the flow conditions, the jet velocity, shape and diameter of the nozzle.

INTRODUCTION

The onset of boiling is investigated very often. However, the problem is still unsolved because it depends on many parameters such as: the properties and the thermodynamic state of the fluid, the micro size nuclei statistically distributed on the heated solid surface, and the fluid conditions before boiling. An additional problem is, that in normal boiling after the first boiling onset, the virgin state of the surface is lost and in subsequent experiments the conditions on the surface are changed. Therefore these experiments are normally very time consuming

In superheated liquid bubbles are formed by the local breakdown of the meatastable state; this process is the onset of boiling. From thermodynamic and statistical considerations three mechanisms for such processes are known:

- the homogeneous nucleation
- the heterogeneous nucleation
- the activation of nuclei.

The homogeneous nucleation is defined as a statistical process induced by molecular fluctuations in the bulk of a superheated liquid. If a nuclei of critical radius r_c is formed by these fluctuations a viable bubble is generated. From thermodynamic stability (if $\rho_l \gg \rho_v$) the critical radius follows as:

$$r_c = \frac{2\sigma}{\Delta p} \tag{1}$$

with σ the surface tension and Δp the isothermal pressure difference between the saturation pressure p_s and the liquid pressure p_∞, $\Delta p = p_s - p_\infty$, called later the overexpansion. To produce an inital large bubble with r_c in a reversible isothermal way the critical activation energy Δ G is necessary, which results from volume and surface tension work on a bubble:

$$\Delta G = \frac{16 \cdot \pi\, \sigma^3}{3 \cdot \Delta p^2} \tag{2}$$

The homogeneous theory assumes, that the critical activation energy can only be produced by the molecular fluctuations, which form a void defect in the liquid lattice structure with radius r_c. The probability J that such a process takes place is given by the Boltzmann law:

$$J = C \cdot \exp(-\Delta G/kT) \tag{3}$$

with k the Boltzmann constant and T the temperature. The coefficient C is calculated by Volmer /1/ using statistical mechanics (see/2, 3/. Measured superheating temperatures in boiling experiments are much lower compared with calculated ones. This is due to the idealistic assumptions in this theory, such as ideal pure fluid, no impurities, and no effects by the solid surface. In reality, however, the liquid is surrounded by walls and in direct contact with the heated surface. The surface itself - ideal plane or rough - in connection with the wetting behavior of the liquid can act as a catalyst and reduce the nucleation energy required for the formation of bubbles. This fact is considered in the heterogeneous nucleation theory.

The thermodynamic stability for bubble formation does not require the formation of a full spherical bubble of size r_c, only the curvature of the critical radius r_c between vapor and liquid is required. If the bubble is generated in a cavity or on a plane surface with a finite wetting angle between vapor, liquid and the solid wall, less volume and surface work is necessary as in the case of a spherical bubble with the same radius, the reduced nucleation energy is:

$$\Delta G_{het} = \phi \cdot \Delta G \qquad (4)$$

This can be shown easily with figure 1; the surface is an ideal plane, however when the wetting angle is 90°, the nucleation energy is reduced by $\phi = 0.5$. In the second sketch the bubble is nucleated in a cavity. The necessary energy depends on the geometry of the cavity and the wetting angle and can be calculated /2, 3/ by the geometrical consideration only. The values for ϕ are between unity and zero.

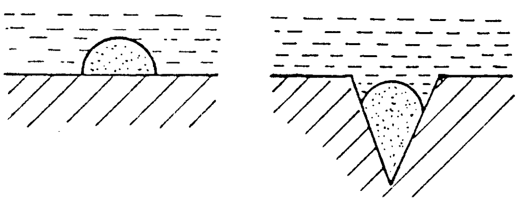

Fig. 1 Reduction of nucleation energy, on a plane surface and in a cavity.

However, in reality ϕ cannot be calculated for a technical surface, because the cavities are of micro size ($< 10^{-6}$m), their geometry and distribution on the surface is unknown and due to the very fast process of nucleation, the dynamic wetting angle has to be assumed. In heterogeneous nucleation the assumption is that all cavities on the surface are flooded with liquid and completely wetted. The bubbles of critical size are formed in the cavities by statistical fluctuation only.

The process of boiling onset is different, if the small cavities are filled with vapor or gas, which is normally the case on technical surfaces. Here, the nuclei still exist and can be activated by changing the fluid state, which is done by superheating the fluid layer or by pressure reduction. The stability criteria for vapor inclusions in cavities was theoretically investigated by several authors /3, 4, 5, 6, 7/; it follows that boiling starts when the vapor inclusion in a cavity reaches a radius equal to the radius s of the aperture of the cavity. Note that the wetting angle of the liquid and the aperture angle of the cavity do not influence this process. Therefore vapor nuclei in cavities are activited if the nucleus radius r_n is equal to the aperture radius s, as:

$$r_n = s = 2\sigma / \Delta p \qquad (5)$$

In the following this nuclei activation is investigated in a superheated jet of water. The great advantage of this method is, that the fluid state can be easily changed and all the bubbles formed in the jet are activated by nuclei.

PROCEDURE

In the pressure temperature diagram (figure 2) the expansion to produce a superheated jet is shown. Subcooled pressurized water of state A is expanded in a specially designed nozzle to state B. The sudden pressure drop from the initial pressure p_o to the ambient pressure p_∞ crossing the saturation pressure p_s causes a superheated liquid state which is maintained and collapses only locally by the formation of bubbles. The liquid water in one phase state can be regarded as an incompressible liquid for which the isotropic expansion is almost isothermal; the initial temperature $T_o = T_A$ is the temperature at point B $T_B = T_A$. The isobaric superheating of the liquid is $\Delta T = T_B - T_{s,\infty}$ and the isothermal overexpansion is $\Delta p = p_s - p_\infty$. The superheat can be changed by varying the initial temperature, and the flow conditions can be adjusted by varying the boiler pressure p_o and by using different shaped nozzles.

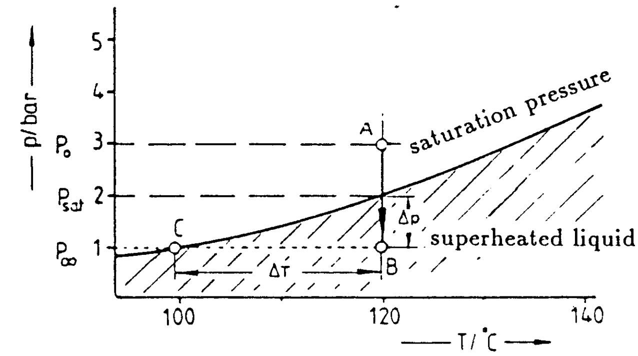

Fig. 2 Expansion in the nozzle in a phase diagram

The process of bubble formation in the superheated jet is shwon in figure 3. Where bubbles develop in the jet the metastable state changes locally to a stable two phase state. Most of the bubbles are not created in the center of the jet, as shown here, we observed in a jet of 1mm diameter that they can grow to five times the size of the jet diameter. As soon as the bubbles have grown too far beyond the jet diameter, they burst and create sequential bubbles. Just after the nozzle the jet surface is cooled by evaporation, however, by unsteady state heat transfer calculation it can be demonstrated that only a very thin layer of the jet surface is cooled to saturation temperature according to the ambient pressure, the bulk liquid in the center is kept at the initial level of superheating. The bubbles in the jet are formed locally where nuclei in form of micro particles flow within the jet liquid.

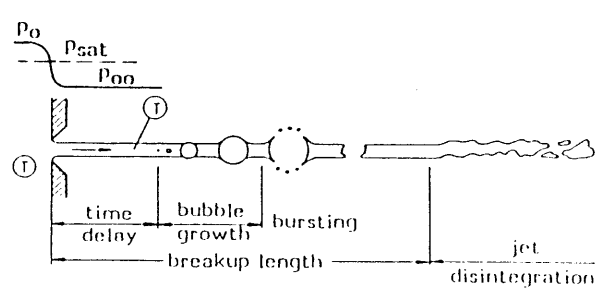

Fig. 3 Sketch of developments in a superheated jet

EXPERIMENTAL TECHNIQUES

In a boiler pure demineralized and filtered water is electrically heated above 100 °C, however, beyond the saturation temperature corresponding to the boiler pressure, for details see /8, 9/. The boiler pressure can be adjusted by a pad of nitrogen. It is ascertained that this nitrogen does not influence the gas content of the water jet, which originates from the lower section of the boiler. From the boiler the water flows in wide tubes into a vessel which acts as a calm region before the nozzle. In a bypass the gas content of the water can be measured. It can be adjusted by boiling the water in a boiler to degas it or by gas injection to saturate the water with gas. From the calm vessel the water flows through a short brass nozzle with a diameter between 1 to 2 mm. Two different shapes of nozzles were tested: nozzles with round inlet and jet contraction of 0.98 and nozzles with a sharp edged inlet with jet contraction of 0.80. The jet velocity could be calculated by the initial pressure in the calm vessel and the jet contraction. The calculations were verified by high-speed photography of the jet.

The number of bubbles in the jet was counted using a light scatter method. A laser beam crosses the free jet at an angle of 45 degrees, see figure 4, the light reflected back from the bubbles is registered by a photomultiplier and determines the bubble frequencies. The jet without bubbles scatters the light in all directions. All measured signals are processed by a control computer. With a high-speed drum camera and a nanolight flash, pictures were taken of the jet loaded with bubbles so that the different bubble formations and bubble growth could be registered.

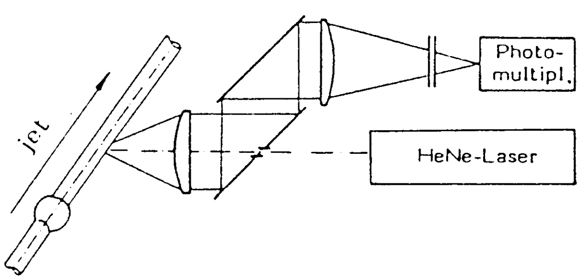

Fig. 4 Light-scatter method to determine the bubble frequency

RESULTS

Within the breakup length of the jet three mechanisms of boiling formation could be distinguished, identified as:

- particle boiling
- surface boiling
- wall boiling.

In all three cases homogeneous nucleation can be excluded, as the degree of superheating or of pressure drop is too small to trigger it off. In figure 5, the regions in which the typical boiling mechanisms occur are shown as functions of the initial boiler pressure p_o and the degree of superheat ΔT. The region of particle boiling is independent of the shape of the nozzles, while the surface regions and wall boiling could be shifted to higher superheat if sharp-edged nozzles are used.

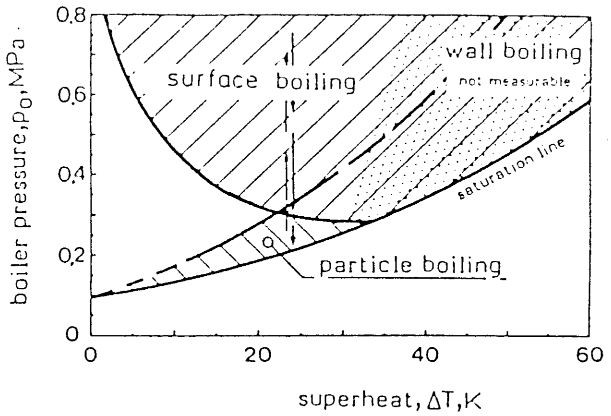

Fig. 5 Boiling regions for the nozzle with 1mm diameter and round inlet.

Particle boiling

A typical high-speed photograph series of bubbles from particle boiling is seen in figure 6. The bubbles are formed within the bulk liquid of the jet; they are spherical and may increase to a size 5 times of the jet diameter. The origin of these bubbles are steam and gas inclusions in the pores of minute floating particles suspended in the liquid. They are activated by the pressure drop. Even though the water is filtered before being filled into the boiler, these small particles are unavoidable. By a process of boiling in the boiler or the injection of gas, it is possible to activate the nuclei in the particles after the pressure drop. By increasing the initial pressure, the nuclei can be deactivated as well.

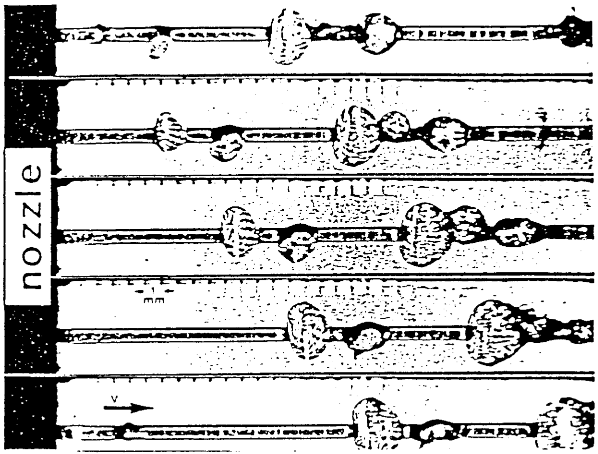

Fig. 6 High-speed photographs of bubbles in the jet. ($\Delta T = 20.3$ K, $v_f = 14.4$m/s, $d_f = 1$mm picture sequences 0.26ms)

Looking at the figure 5 we see that at a superheat of $\Delta T < 20K$ if the boiler pressure p_o is increased with a constant ΔT above the region of particle boiling the nuclei density goes to zero; no bubbles are observed in the white area. If the pressure is reduced from the white area to the region of the particle boiling, bubbles are still not formed in the jet, the nuclei are deactivated by flooding. This behavior indicates that the bubbles are formed by activation of nuclei and not by homogeneous or heterogeneous nucleation. In this last case the pressure before the expansion has no influence on the nucleation process.

The upper limits of the particle boiling region could be determined varying the rate of superheat ΔT and the initial boiler pressure p_o. This limit was recorded in many experiments:

$$p_o - p_s = p_s - p_\infty$$

or:

$$\Delta p_o = \Delta p \qquad (5)$$

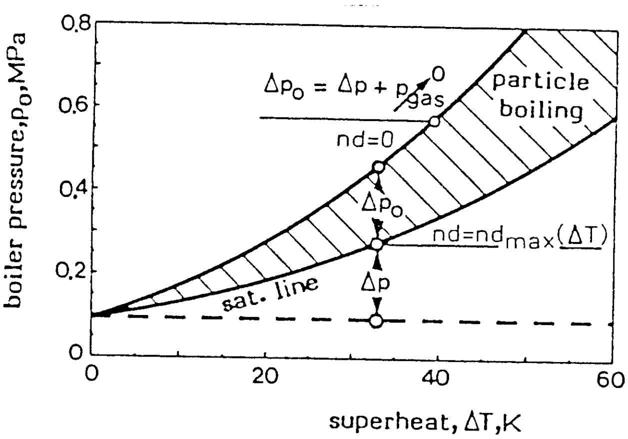

Fig. 7 Limits for particle boiling.

These results are summarized in figure 7. The lower limit of particle boiling region is the saturation line. By the expansion from saturation pressure to ambient pressure, the nuclei density reaches maximum values, which means that most nuclei are activated. By increasing the pressure the nuclei density is reduced and zero at the upper limits nd = 0. The measurements with gas content in the boiler water shows an increase of nuclei density till the water is saturated with the gas. More gas- injection does not increase the nuclei density. The upper limit of particle boiling is shifted to higher initial pressure (index g is the partial gas pressure).

$$p_{og} - p_s = \Delta p_{og} = \Delta p + p_g \qquad (6)$$

according to the pressure in the bubble:

$$p_b = p_s + p_g$$

which yields the stability condition:

$$\frac{2\sigma}{p_o - (p_s + p_g)} = \frac{2\sigma}{\Delta p} \qquad (7)$$

Relation 7 is valid up to the saturation of dissolved gas in the liquid, which depends on pressure and temperature.

The mechanism of nuclei activation can be explained by figure 8.

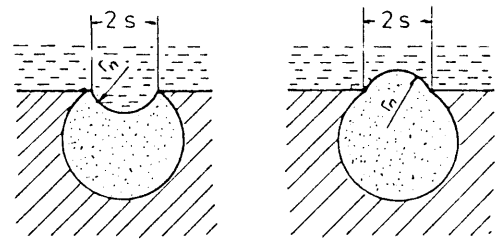

Fig. 8 Cavity model for nucleus activation. Left: liquid pressure $p_L > p_s$ Right: liquid pressure $p_L < p_s$

On the left side the liquid pressure p_L is above the saturation pressure p_s, vapor is enclosed in a cavity with pressure p_s, according to the temperature of the system. The concave radius between liquid and vapor of this nucleation is given by:

$$r_n = \frac{2\sigma}{p_L - p_s} \qquad (8)$$

The vapor nucleus is stable as long as the nucleus radius r_n is larger than the aperature radius s of the cavity. If the pressure increases to a value $r_n = s$, the surface tension is no longer able to stabilize the phase boundary and the nucleus will be flooded with liquid and can not be activated by decreasing pressure. If the liquid pressure p_L is increased to a value $r_n \geqslant s$ the nucleus will remain stable. By decreasing the pressure r_n will increase to a plane surface when $p_L = p_s$.

If the liquid pressure falls below saturation, r_n becomes convex and decreases with decreasing pressure. With $r_n = s$ an unstable situation is reached, then the nucleus is activated and a bubble is formed. This is the point of boiling onset. As experimentally observed and expressed by equation 5, the initial pressure difference Δp_o exceeds the overexpansion Δp all nuclei are flooded and can not be activated by a pressure drop.

Gas injection

The influence of gas injection on nuclei density was studied in the following manner: The water was first degased and pressure was increased to a value $\Delta p_o > \Delta p$, so all nuclei are flooded and no bubbles are observed. At several superheating temperatures ΔT air was injected into the boiler and the content of oxygen was measured. As it can be seen in figure 9, the nuclei density increased with the O_2-content in the liquid till the saturation in a gas solution is reached. More gas can not be solved and the nuclei density did achieve a constant value at each superheated temperature. This observation indicates that gas can activate the nuclei in pores and that at each temperature a saturation point for nuclei density is achieved.

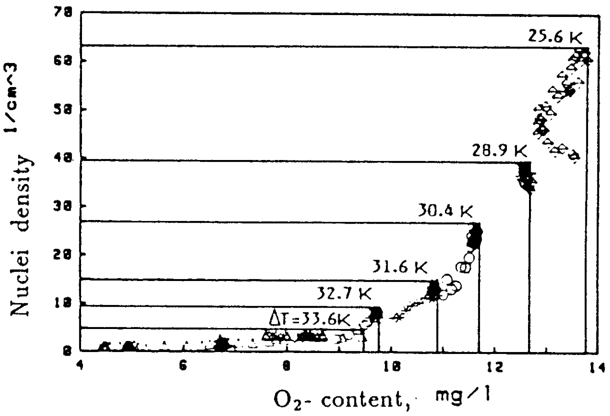

Fig. 9 Nuclei density as function of O_2- content, and at ΔT, saturation of nuclei density is reached at the gas saturation.

Surface boiling

When the pressure in figure 5 is increased above the region of particle boiling in the white area no bubbles at first could be observed. With increasing pressure, the jet velocity is increased and a second area of boiling is detected. The bubbles of this boiling regime are only on the surface of the jet, see figure 10.

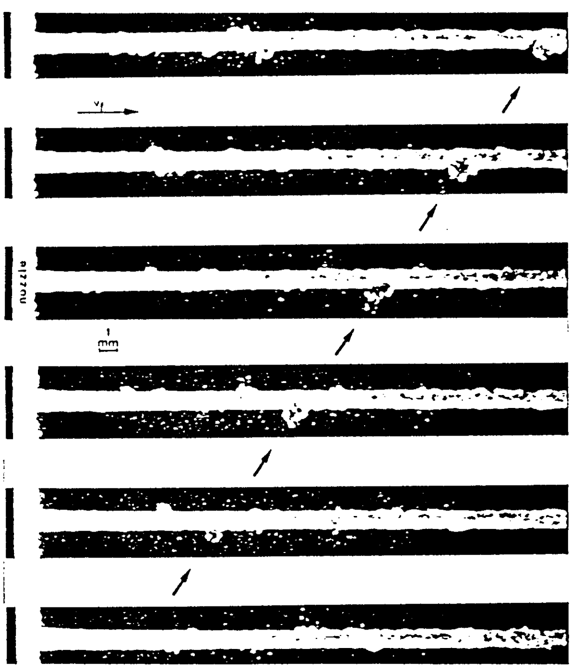

Fig. 10 Surface bubbles in the jet ($\Delta T = 22.6$ K, $v_f = 36$ m/s, picture sequence 0.12 ms)

They do not have the typical spherical shape and look more like scars on the surface, so that they are distinguishable from bubbles formed by the nuclei of particles. With increasing pressure the amount of generated bubbles, the nuclei density is increasing. With decreasing pressure the nuclei also decrease and is zero at the same pressure at which boiling starts. The onset of boiling is shifted to higher pressure and superheat for nozzles with greater jet concentration, (sharped-edged inlet) and smaller jet diameters. From high-speed pictures the locus of the origin of the bubbles could be formed. The bubble frequency, figure 11, versus the delay time for bubble formation has a typical Gaussian distribution, with a maximum value, which is shifted to the nozzle at higher pressures. The influence of gasification on this boiling is weak. The real physical origin of this kind of boiling is not yet clear. The high jet velocity can induce turbulent fluctuations, which can locally increase the overexpansion. However, it can be estimated that this turbulent pressure drop is not large enough to induce homogeneous nucleation. An other explanation could be as follows: on the edges of the nozzle inlet, small vortices are produced which detach and flow with the jet. Outside of the nozzle they continue their rotation and absorb gas or vapor beyond the surface, which may act as nuclei for further bubble formation just below the surface. Another influence could be the feedback pressure fluctuations from the oscillating frequency of the jet disintergration.

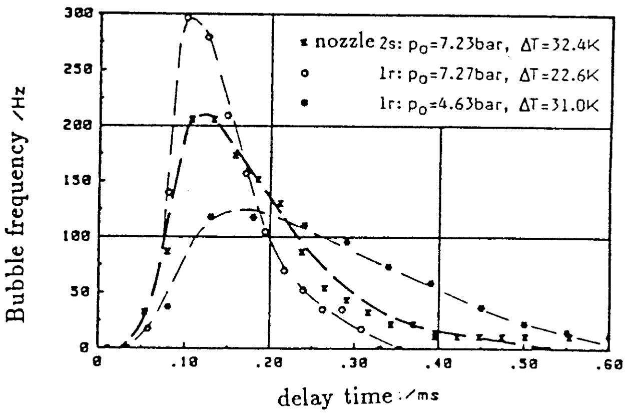

Fig. 11 Bubble frequency versus the delay time for formation after the nozzle.

None of these arguments can alone explain the observed behavior. However, it is clear that this boiling is influenced by the flow and the flow conditions just before the liquid is released from the wall of the nozzle. For the onset limit of this regime an emprical correlation could be developed:

$$\mathrm{Re} = (57.2 - 8.5\Delta p_n) \cdot 10^{\ 3} \qquad (9)$$

For the Re-number a semi-spherical flow towards the nozzle with average velocity v_s is assumed, which can be calculated for an incompressible liquid for a semi-sphere with a diameter d_n:

$$v_s = \frac{v_f}{2}\mu^2 \qquad (10)$$

with v_f being the jet velocity, μ the jet contraction, and d_n the nozzle diameter; with kinematic viscosity as follows:

$$\mathrm{Re} = \frac{v_s d_n}{\nu} = \frac{v_f \cdot \mu^2 d_n}{2\nu} \qquad (11)$$

Δp_n is a modified overexpansion; the defined overexpansion $\Delta p = p_s - p_\infty$ is reduced by the pressure difference before and after the jet contraction, for Δp_n follows from the Bernoulli equation:

$$\Delta p_n = \Delta p - \frac{\rho}{2} \cdot v_f^2 \cdot (1 - \mu^4) \qquad (12)$$

Equation (9) describes reasonably well the onset limit of surface boiling as indicated in figure 12.

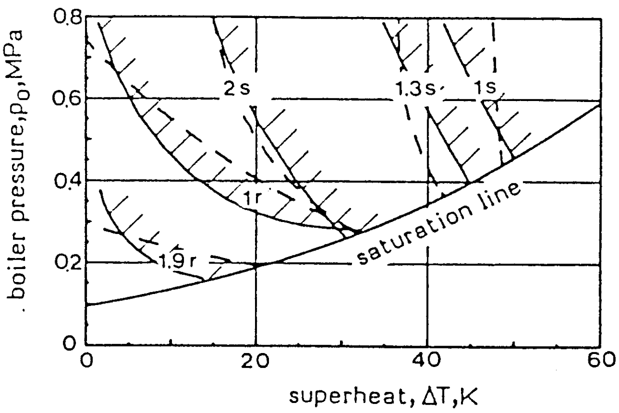

Fig. 12 Comparison of equation 9 dashed line, with the experimental results of different nozzles, r: round nozzle, s: sharp-edged, the number is the diameter of the nozzle.

Wall boiling

With the term wall boiling we understand the normal nucleate boiling on solid surfaces where boiling is generated by nuclei on the wall. If the wall of the nozzle is in contact with the superheated liquid caused by the pressure drop and the pressure in the liquid decreases below the saturation pressure, then wall boiling can even occur in a nozzle. However, in the case of short nozzles we used here, the overexpansion necessary for bubble formation takes place in the jet contraction region where the wall is no longer in contact with the liquid. A pressure calculation of the pressure drop in the nozzle shows, that the saturation pressure is achieved beyond the point of wall conduct. In the case described here, wall boiling would occur in a region where the mechanisms are too active to allow for a separation of the three described mechanisms, see fig. 5. With longer nozzles and greater diameters, the regions of surface and wall boiling will be shifted toward to and finally into the nozzle.

SUMMARY

In this paper three mechanisms of boiling are described, which are observed in superheated free jets. The mechanisms of nuclei activation could be studied by the variation of pressure, temperature and gas content. Each bubble in the jet is a "first one" created in the bulk isothermal superheated liquid. The liquid parts between the bubbles are still metastable superheated. The deactivation of nuclei by increasing pressure supports the theory of nuclei activation, which can here be clearly distinguished from heterogeneous or homogeneous nucleation. In these experiments only water was used as a test fluid. Water has a high surface tension, therefore a high pressure is needed to deactivate the nuclei, which means flooding the cavities. With other liquids and generally at higher saturation conditions, it is not excluded that heterogeneous or even homogeneous nucleation could be achieved. However, that investigation would need a closed loop. At higher jet velocities a boiling mechanism just below the surface of the jet could be observed. This mechanism behaves quite differently in comparison to the first one. Some explanations of the origins of surface boiling are given, however, none of them is individually conclusive. By the specially designed nozzles the normal nucleate boiling mechanism on the wall of the nozzles was excluded, this facilitates the investigation and study of the other mechanisms.

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