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MECHANISM OF THE INITIAL STAGE OF BUBBLE GROWTH IN A LIQUID CLOSE TO THE SUPERHEAT LIMIT

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If a superheated liquid is not in equilibrium with a vapor separated from it by a plane liquid–vapor interface, it is thermodynamically stable with respect to a vapor bubble of certain dimensions because of the role of surface energy. At any degree of superheat, equilibrium with the bubble is unstable, the increase and decrease in the bubble dimensions being of the same probability. Such a bubble is known as the critical nucleus. Its radius is given by the equation

$$R_* = 2\sigma/(p_s - p''), \tag{1}$$

where σ is the surface tension, and p_s is the pressure within the bubble, p'' is the pressure in liquid. The condition of the mechanical equilibrium is reduced to the same equation.

The concept of the critical nucleus is the basis of the homogeneous nucleation theory [1–6] which enables the upper limit of a superheat to be calculated. Only nucleus formation is considered. It is as if nuclei that reach the critical dimensions are removed from the system and replaced by an appropriate mass of liquid. The theory does not give any information concerning the dynamics of bubble growth either. Nevertheless, some researchers use the concept of the critical nucleus as the initial condition in a description of the bubble growth in superheated liquid. A bubble begins to grow after the disturbance of the balance, given by Eq. (1), when the pressure difference exceeds the surface tension. According to this scheme, the initial bubble growth results from the fact that the vapor pressure in the bubble is higher than that in the surrounding liquid.

In the case of unactivated boiling [7] the process takes place at the existing centers which are vapor–gas bubbles in the superheated liquid or any cavities at the rough solid surfaces that are in contact with the liquid. Under these conditions it is not necessary to surmount any potential barrier to form a bubble. The growth process starts when the radius of curvature of the vapor–liquid interface exceeds that of the critical nucleus. The radius of the critical nucleus is equal to that of the orifice of the cavity [8], which is the center of the evaporation and is also given by Eq. (1). Superheats in the case of the existing centers are significantly lower than the superheat limit. Thus, application of the concept of critical nucleus to the description of the bubble growth process in the case of unactivated boiling is justified by the presence of gas bubbles either in the liquid or in the cavities of the heater.

By contrast, in the case of spontaneous activated boiling, when the superheat almost attains its limit, the use of the expression for critical nucleus as given by (1) in an effort to explain the initial stage of bubble growth has no such clear physical basis as in the previous case of unactivated evaporation. It has already been mentioned in [3] that an advent of the critical nucleus concept "does not answer the question of the new phase formation, but just transfers it from large-sized objects to those of extremely small size."

The homogeneous nucleation theory assumes that the vapor phase formation manifests itself as the emergence of tiny vapor bubbles that are known as the critical nuclei. These nuclei come into existence spontaneously, as a result of fluctuations. However, it was shown in [9] that in the vicinity of the spinodal the critical fluctuation has little in common with the critical nucleus. The vapor–liquid interface is absent, and the liquid density changes smoothly and at every point is greater than that of the vapor phase at equilibrium. The closer the superheat to its limit, the larger the characteristic size of the density change. At the spinodal this size

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approaches infinity; that is, the surface tension concept itself that explains the existence of the metastability region in the context of the classical homogeneous nucleation theory (along with the general principles of thermodynamics) loses its meaning under these conditions. Based on these results, a conclusion was made in [10] that Eq. (1) should not be used in a description of the critical nucleus. Some other researchers [11, 12] were also doubtful whether the surface tension concept was applicable to the nucleation process analysis.

If the use of the Eq. (1) at superheats that are close to the limit is improper, the following question arises: What is the mechanism of bubble growth start-up and how does growth proceed during the initial stage? In the experimental investigation [13] of the dynamical effects that occur during vapor bubble growth within a single droplet of butane at the superheat limit at atmospheric pressure, it was found that the bubble surface was extremely rough and the growth rate is shown to be constant and two orders of magnitude greater than would be predicted by conventional bubble-growth theories. Explorations of this phenomenon, carried out in [14, 15], made it apparent that at high evaporation rates the vapor recoil impulse was of the major importance in the bubble growth process. Based on the results of [14, 15], the following probable mechanism is offered which describes the initial stage of bubble growth in a liquid heated to the superheat limit. All the estimates and reasoning which follow are given in the context of the approximations of the Hertz–Knudsen evaporation theory.

For the majority of organic liquids at high superheats the effective heat of evaporation [7] is negative: $L_* < 0$, $L_* = L-(c'_{ps}T_s - c'_pT'')$, where L is the heat of evaporation, T_s is the temperature of metastable liquid, T'' is the saturation temperature, which corresponds to the liquid pressure p'', and c'_{ps} and c'_p are the specific heats of the liquid at temperatures T_s and T'', respectively. For instance, for butane at a superheat of 105 K and at atmospheric pressure $L_* = -2 \cdot 10^5$ J/kg. What this means is that the energy stored in metastable liquid is sufficiently great to evaporate the liquid completely without an additional heat supplement from an external source. For water at high superheats the effective heat of evaporation, though positive, is an order of magnitude lower than the heat of evaporation: at atmospheric pressure and at superheat of 586 K [7] $L_* = 1.1 \cdot 10^5$ J/kg. With this great amount of energy accumulated, a liquid rupture at any weak point (for example, at a Frenkel "island") initiates the process of rapid evaporation into the formed cavity. This causes the liquid to be acted upon by the vapor recoil impulse whose value can be estimated from the equation

$$\Delta p = j^2/\rho''_s, \tag{2}$$

where the vapor mass flux is given by the Hertz–Knudsen equation

$$j = \beta\left(\frac{RT_s}{2\pi M}\right)^{1/2}(\rho''_s - \rho''). \tag{3}$$

Here β is the accommodation coefficient; R is the universal gas constant; M is the molecular mass; ρ'' is the vapor density at the temperature of saturation T''; ρ''_s is the vapor density at the temperature T_s. For water under conditions previously discussed and at $\beta = 1$ Eq. (2) gives $\Delta p = 24 \cdot 10^5$ Pa. This value is comparable with the pressure amplitudes which were measured within the near-surface water layer during the explosive boiling caused by radiation-stimulated heating which was close to the threshold values [16, 17]. For butane, calculations of the vapor recoil impulse carried out using Eqs. (2) and (3) give the value $3 \cdot 10^5$ Pa.

The vapor recoil impulse gives rise to growth of a vapor cavity during the initial stage of spontaneous boiling. For the stationary movement the liquid velocity resulting from the vapor recoil impulse can be estimated using the following equation [14]:

$$v = j\left(\frac{2}{\rho'_s\rho''_s}\right)^{1/2}, \tag{4}$$

where ρ'_s is the liquid density at the temperature T_s. For the above values of the vapor recoil impulse the liquid velocity equals 84 m/sec for water and 37 m/sec for butane, which is roughly an order of magnitude higher than that of liquid evaporation (j/ρ'_s) under these conditions.

The value of the vapor recoil impulse depends on the temperature of the liquid near the vapor–liquid interface, while the duration of its main effect upon the liquid depends on the temperature decrease rate.

ɔr organic liquids the energy stored is sufficient for their complete evaporation, some decrease of the temperature of the liquid still occurs near the interface due to the nonuniform energy distribution between molecules. Only the "hottest" molecules escape from the liquid phase. The jetting-driven bubble growth continues until the temperature of the liquid near the bubble surface reaches a value of saturation that corresponds to the pressure in the liquid. Further growth depends on the process of heat transfer through the liquid to the bubble surface. If the bubble surface loses its stability [13–15], then again, the increase of the disturbance amplitude is governed by the vapor recoil impulse and, for the stationary process, the disturbance growth rate in superheated liquid can be evaluated from Eq. (4). Increase of the pressure within the system leads to suppression of the interface instability [18, 19].

In such a manner, while in the case of unactivated boiling the initial growth of a bubble is caused by the difference between pressures within the nucleus and in the surrounding liquid, in the case of spontaneous boiling it proceeds owing to the effect of the vapor recoil impulse upon the liquid during evaporation into the cavity formed as a result of the liquid rupture.

The proposed hypothesis can be tested in experiments of boiling-up when only a small volume of the liquid reaches a high level of metastability while the bulk has a temperature below that of saturation. Such conditions can be attained, for example, during radiation-stimulated heating of a liquid within the area of a laser beam focus or by rapid warming of a heater placed in the cold liquid. In the case of spontaneous boiling under such conditions the vapor pressure within the bubble will be lower than that in the liquid at every stage of its life. The initial bubble growth will be conditioned by the influence of the vapor recoil impulse. Surface tension and pressure in the surrounding liquid counteract the bubble growth.

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