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DESTABILIZATION OF FILM BOILING BY MEANS OF A THERMAL RESISTANCE

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Abstract—Described is the type of vaporisation which takes place when a thermal resistance, consisting in a film of a substance of low heat conductivity, is placed between the surface of a quenched sample and the cooling liquid. This type of vaporisation, larvate boiling, is characterised by an alternate wetting/non-wetting of the solid surface.

Two conditions are necessary for larvate boiling: thermal resistance and surface effusivity.

Substituting larvate boiling for film boiling allows the heat flux between a solid surface at high temperature and the cooling liquid to be greatly increased.

When a metallic object at high temperature is dipped in a volatile liquid, two successive modes of evaporation are observed: film boiling and nucleate boiling.

The determination of the limiting surface temperatures in the presence of these two modes of vaporisation, enables one to predict the mode of vaporisation which takes place on the surface (Chevrier *et al.* 1972).

Previous studies on film boiling (Bradfield 1965; Semeria & Martinet 1965; Moreaux 1973) have shown that there is no wetting of the surface while there exists a film of vapor, and the cooling rate of the sample remains low.

In order to increase the heat flux density from the surface of the sample, the liquid must wet the solid surface as in nucleate boiling. It is known, however, that this process of vaporisation can only be obtained at relatively low surface temperatures, lower than a critical temperature L_s known as the transition temperature (Chevrier *et al.* 1972).

Some authors (Sato 1932; Cowley *et al.* 1962; Chevrier & Beck 1970) have shown that by coating the surface of the metallic sample by a substance of low thermal conductivity, it is possible to increase the heat flux transferred in the range of temperatures where film boiling normally exists. This increase is due to a particular mode of vaporisation, larvate boiling, which is an intermediate mode between film and nucleate boiling (Moreaux & Beck 1969; Beck & Chevrier 1971).

1. CONDITIONS OF APPEARANCE OF LARVATE BOILING

Larvate boiling is obtained when an appropriate thermal resistance is placed between the metallic sample and the cooling liquid. This process of larvate boiling substitutes film boiling. It has been studied in the case of two liquids: water and liquid nitrogen.

1.1 *Larvate boiling with water*

Layers of zirconium oxide of thickness varying from 25 to 1000 μm have been deposited on the surface of a cylindrical nickel sample. The cooling of the sample at an initial temperature of 850°C quenched in boiling water is described in figure 1.

When the thickness of zirconium oxide coating is increased, the time it takes to cool the sample to the water temperature decreases. The thermal resistance acts to destabilize the film boiling. In fact, the limiting temperature of film boiling increases with the thickness of the coating. The mode of *evaporation* which appears next is larvate boiling. The heat flux densities are increased when coating thickness augments from 0 to 400 μm. At the same time, when the

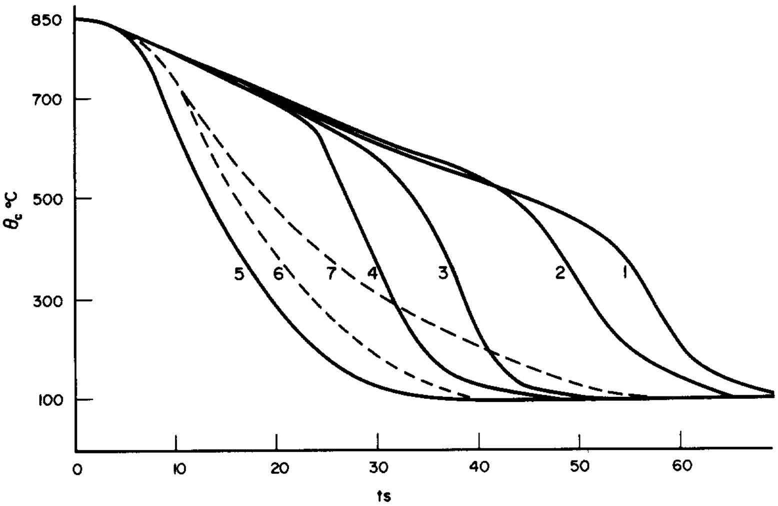

Figure 1. Cooling rates in the center of a cylindrical nickel sample (D = 16 mm, H = 48 mm) coated with zirconium oxide and quenched from 850°C in water at 100°C. (1) e = 25 μm. (2) e = 50 μm. (3) e = 100 μm. (4) e = 200 μm. (5) e = 400 μm. (6) e = 600 μm. (7) e = 1000 μm.

coating thickness is larger than 400 μm, the screen effect slows the cooling down. Then, nucleate boiling takes place.

1.2 *Liquid nitrogen*

Samples have been quenched starting at ambient temperature. The thermal resistance consists of a layer of polymer resin (Chevrier & Beck 1971). Figure 2 describes the entire cooling process as a function of the thickness of the coating, showing the same phenomena as in the case of pure water. There exists a certain critical thickness where the cooling is fastest.

1.3 *Saline solutions*

When a metallic sample is quenched in a boiling aqueous solution of 10% NaCl, salt precipitated on the surface is observed at the end of the film boiling regime (Moreaux & Beck 1969). This coating provides a thermal resistance which alters the cooling of the sample (figure 3). One observes a mode of evaporation different from film boiling, which precedes the onset of nucleate boiling (parts AB of the curve of the figure). This is larvate boiling.

The case here is different from the preceding ones in that the thermal resistance forms during the process of film boiling which it destabilizes. It is the existence of larvate boiling which explains the effect of accelerated cooling obtained by the addition of thermally stable salt to the water used for quenching. (Zavarine 1935; Bigot & Faivre 1956).

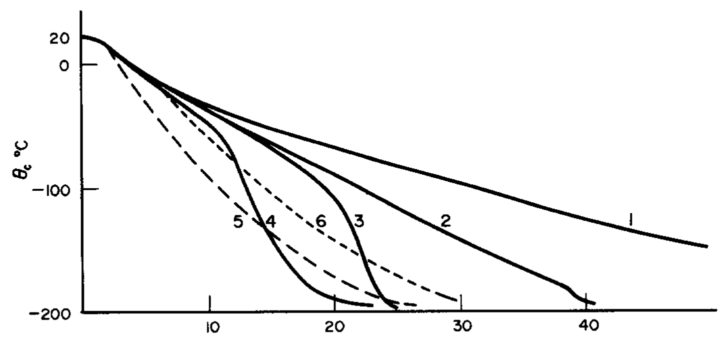

Figure 2. Cooling rates in the center of a cylindrical nickel sample (D=10 mm, H = 10 mm) coated with polymeric resin and quenched from θ_0 = 20°C in liquid nitrogen. (1) Without coating. (2) e = 2 μm. (3) e = 60 μm. (4) e = 90 μm. (5) e = 200 μm. (6) e = 500 μm.

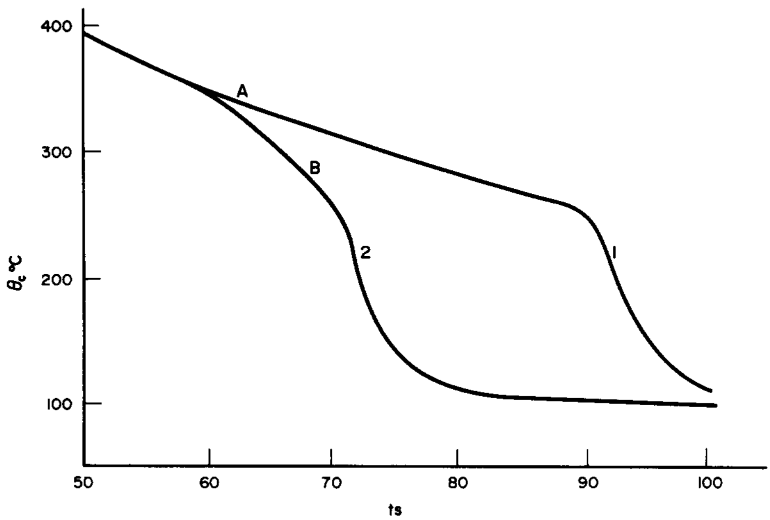

Figure 3. Cooling rates in the center of a cylindrical nickel sample ($D = 16$ mm, $H = 48$ mm). (1) Quenched from $\theta_0 = 700$°C in water at 100°C. (2) Quenched from $\theta_0 = 700$°C in an aqueous solution of 10% NaCl at 101°C (the cooling curve between 700°C and 390° has not been transcribed).

2. EXPERIMENTAL STUDY OF LARVATE BOILING

Larvate boiling, as observed by means of ultra-fast cinematography, presents itself in the form of a continuous vapor film, similar to that in film boiling. The liquid–vapor interface is the site of amplitude instabilities sufficient to cause wetting of the surface. At each point of the sample, the thickness of the vapor film oscillates between zero and a very large value. This "respiration" of the vapor film is perturbed by the upward displacement of the vapor under the effect of buoyancy. Still, the cinematographic observation of the larvate boiling enables one to detect the pulsed movement of the vapor film, since the frequency of the pulsations exceeds 50 Hz.

The experimental study of the phenomena required also measurement of the temperature variations at the surface of the insulator. It is known, however, that it is actually impossible to determine the superficial temperature of an insulator when it varies rapidly. A technique consisting of studying the electric conduction between a point on the surface of the sample and the liquid permitted to establish the existence of direct solid–liquid contacts, and hence wetting of the outer surface.

A zirconium oxide-coated nickel sample was used in these experiments. A 0·3 mm diameter silver wire was passed through a cavity in the interior of the sample and extended to the surface through a hole of the same diameter. The wire was insulated from the mass of the sample. After putting on the zirconium oxide film, the silver wire was cut flush with the surface. Thus, a very tiny portion of the surface was made conductive.

The silver wire, connected to the negative pole of a d.c. generator acted as a cathode. The water was made slightly conductive by adding to it 1% of NaCl. A platinum electrode served as the anode. The variations of current circulating through the assembly was recorded on a fast recorder or on an oscilloscope.

During film boiling, this setup allowed to verify the absence of any contact between the water and the surface of the sample. As soon as larvate boiling began, brief contacts of 0.5×10^{-2} to 2×10^{-2} sec duration appeared at an average frequency between 100 and 50 Hz, which increased as nucleate boiling was approached. Just before the onset of the latter, the duration of the contacts was the shortest: 0.5×10^{-2} sec.

The ultra-fast cinematography showed that the different points on the surface of the sample are not simultaneously wetted, but that the surface is swept by a wetting front moving from bottom to top at an average velocity between 1 and 5 m/sec.

3. MECHANISM OF LARVATE BOILING

The necessary condition for larvate boiling to replace film boiling is the existence of a thermal resistance between the solid and the liquid, as shown schematically in figure 4. In fact, the heat flux across the thermal resistance depends on the value of the latter, since

$$\varphi / s = -\lambda(\theta_s - \theta_d)/e.$$

However, at the initial instant of quenching, the heat radiated by the sample is enough to prevent any wetting of the surface (Moreaux & Beck 1972). As a result, the temperature θ_d remains quite close to the initial quenching temperature. The layer where film boiling exists, consisting of water vapor, absorbs a major part of the energy radiated from the outer surface of the sample. The evaporation of the liquid being slowed down, the vapor film is not replenished and some primary wetting of the surface takes place, resulting in a considerable decrease in θ_d. This results in an increase of heat flux through the coating. Note that there is no way to measure the decrease in θ_d or to calculate it. One can, however, consider that the decrease in θ_d is stronger as the effusivity of the outer surface is lower. Also, the short flux of heat transferred by the insulating film causes the formation of a new layer of vapor which stops the brief wetting of the surface. This results again in an increase of θ_d which, by blocking again heat conduction in the coating, breaks the dynamic equilibrium by which film boiling is maintained. The disappearance of the vapor film allows a new contact between the liquid and the insulator. Thus, an alternating effect of wetting and non-wetting of the surface is obtained, as had been observed above.

This alternating effect results in fluctuations of θ_d between two extreme values, the lower corresponding to the wetted state, the higher to the non-wetted state of the surface. Meanwhile, θ_s decreases monotonically, while the temperature fluctuations are localized in the outer layers of the coating.

The practical interest in larvate boiling results from the possibility of wetting an outer surface whose temperature exceeds the critical temperature above which film boiling begins. The allowed heat flux density may therefore be increased by a factor as high as ten.

4. CONDITIONS FOR EXISTENCE OF LARVATE BOILING

The appearance of larvate boiling requires thermal resistance of the coating, and superficial effusivity.

4.1 *Thermal resistance*

The cooling curves represented in figures 1 and 2 show a progressively increasing cooling rate as the thermal resistance of the insulating film increases. This increase in heat flux density is

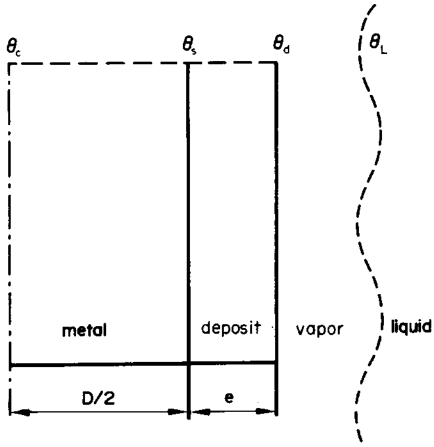

Figure 4. Scheme of a sample with coating of thickness e. Notation of different temperature.

made possible by the larvate boiling replacing film boiling or the more premature nucleate boiling.

When the thermal resistance reaches its critical value, an evaporation process similar to that of nucleate boiling takes place from the very initial instant of quenching. The cooling rate reaches its highest value, and the evaporation process is the most efficient. Any further increase of thermal resistance, which slows down the conduction in the solid, lowers the heat flux density from the solid to the liquid.

These effects are illustrated in figure 5 which shows the influence of thermal resistance R of the coating on the average cooling rate of the sample as a function of the initial quenching temperature in liquid nitrogen. The figure has three domains:

domain I which remains inaccessible regardless of the value of thermal resistance;

domain II where the cooling rates are determined by the thermal resistance;

domain III which corresponds to a cooling rate slower than the one normally obtained through film boiling. This domain of cooling rate (or of heat flux density) may be obtained by means of cooling other than an evaporating liquid.

It is therefore interesting to discuss methods effective in domain II:

The upper curve Ac in figure 5 represents the fastest cooling rate, in the case where the thermal resistance of the coating reaches its critical value R_c. Each point in domain II represents one of two situations:

$R < R_c$. Cooling is assured by larvate boiling and further by nucleate boiling.

$R > R_c$. Nucleate boiling is slowed down by too high a value of the thermal resistance.

The curve αc represents the upper limit for the existence of larvate boiling. It corresponds to an ideal coating with zero heat capacity.

2.2 *Condition of superficial effusivity of the coating*

The condition of effusivity, introduced qualitatively in the model proposed to describe the mechanism of larvate boiling, imposes a small value of the superficial effusivity of the solid. The

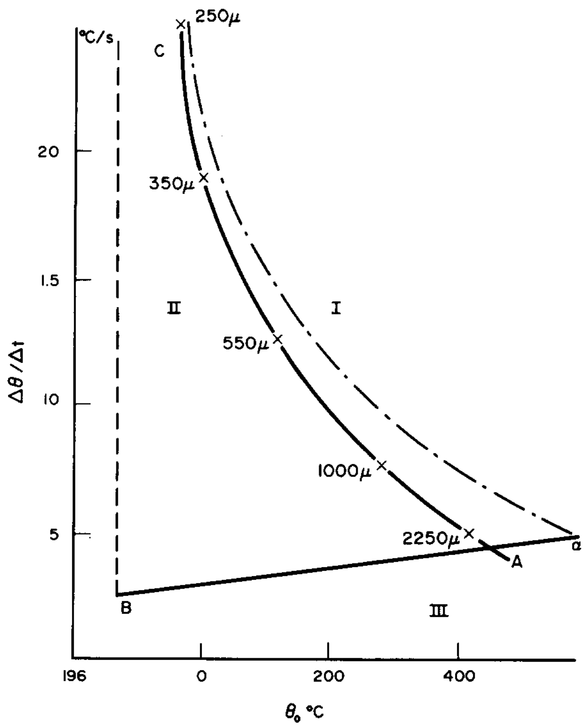

Figure 5. Quenching of a cylindrical nickel sample ($D = 10$ mm, $H = 10$ mm) in liquid nitrogen. Variations in average cooling rate as a function of initial quenching temperature for: AB—sample bare during film boiling. AC—sample coated with calcium sulfate of thickness e. αC—sample covered with ideal coating of zero heat capacity.

existence of this condition may be proved experimentally by varying the value of superficial effusivity of the sample by means of a very thin coating of silver (Moreaux *et al.* 1973), completely covering the zirconium oxide film ($E_{Ag} = 31574$, $E_{ZrO_2} = 2230$ W.m^{-2}.sec$^{1/2}$.(°C)$^{-1}$).

Case of boiling water. A nickel cylinder, with diameter $D = 16$ mm and length $H = 48$ mm, coated with a film of zirconium oxide 200 μm thick, quenched from 850°C in boiling water, gives the cooling curve 2 of figure 6. A layer of silver 50 μm thick of negligible thermal resistance is sufficient to bring the cooling curve back to the vicinity of that of a bare cylinder.

If the coated zirconium oxide sample gives rise to larvate boiling, it is enough to coat it with 50 μm of silver to observe a regime of film boiling very similar to that of a bare cylinder.

Similar results were obtained in the case where the thermal resistance consisted of an anhydrite [figure 7].

Case of liquid nitrogen. Identical experiments have been performed with liquid nitrogen. The curves of figure 8 demonstrate the same phenomena as in the case of water.

DISCUSSION

The condition of effusivity required for destabilization of film boiling, cannot be defined quantitatively. Nevertheless, in order to verify the effect of superficial effusivity of the solid on

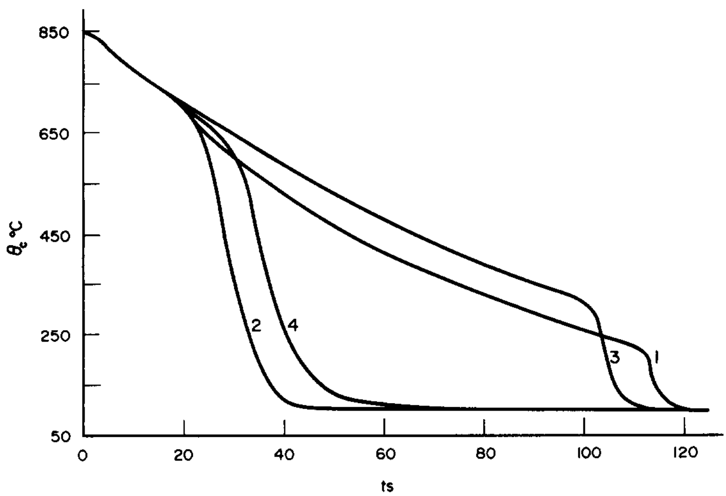

Figure 6. Cooling rates in the center of cylindrical nickel sample ($D = 16$ mm, $H = 48$ mm) quenched from 850°C in water at 100°C. (1) Bare. (2) Coated with 200 μm zirconium oxide. (3) Coated with 200 μm zirconium oxide + 50 μm silver. (4) Coated with 200 μm zirconium oxide + 50 μm silver + 10 μm sodium silicate.

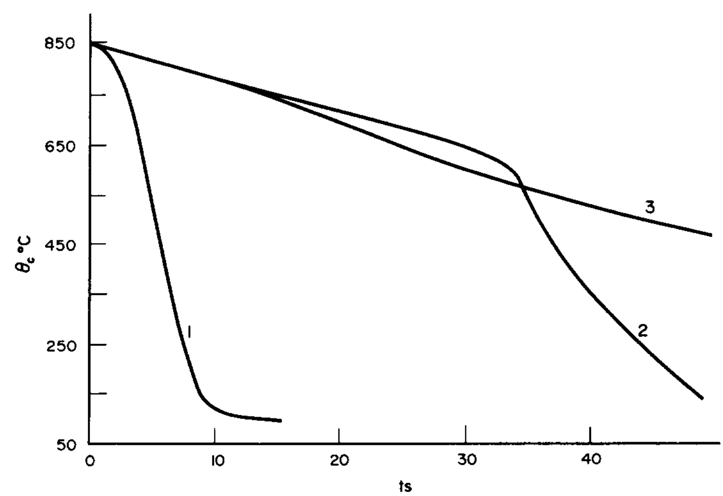

Figure 7. Cooling rates in the center of a cylindrical nickel sample ($D = 16$ mm, $H = 48$ mm) quenched from $\theta_0 = 850$°C in water at 100°C. (1) Coated with 400 μm anhydrite. (2) Coated with 400 μm anhydrite + 50 μm silver. (3) Bare.

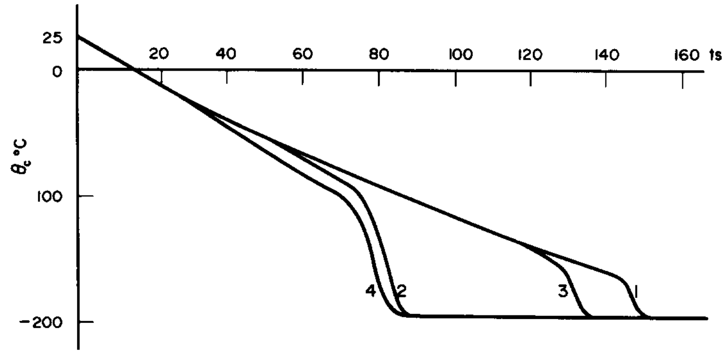

Figure 8. Cooling rates in the center of a cylindrical nickel sample (D = 16 mm, H = 48 mm) quenched from θ_0 = 25°C in liquid nitrogen. (1) Bare. (2) Coated with 200 μm zirconium oxide. (3) Coated with 200 μm zirconium oxide + 50 μm silver. (4) Coated with 200 μm zirconium oxide + 50 μm silver + 10 μm polystyrene.

larvate boiling, a very thin film (10 μm) of sodium silicate (thermal effusivity $s_i E$ = 2091 MKS) was applied to the sample already coated with zirconium oxide and silver. The sample, now covered with three coatings was quenched in boiling water, and regained the same cooling rate as in the case where it only had the 200 μm thick ZrO_2 coating (curve 4 of figure 6).

Note that in practice, the condition of effusivity is always satisfied since the coating material with low diffusivity, always has an equally small effusivity.

CONCLUSIONS

The presence of a thermal resistance at the interface between a high temperature solid and a volatile liquid acts to destabilize the very stable mode of vaporisation of film boiling.

Larvate boiling which replaces it, increases the heat transfer considerably. This mode of vaporisation is characterized by an alternating effect of wetting and non-wetting of the coating, in periods of the order of a hundredth of a second. Larvate boiling can only exist if the superficial effusivity of the surface is small.

The average heat flux passing from the solid to the liquid varies as a function of the thermal resistance of the cooling. On the other hand it varies as a function of the effusivity of the solid surface.

This allows, in practice, to control the heat transfer and, in the case of quenching, to obtain various cooling rates.

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Résumé—Dans le cas de la trempe, on décrit le mode de vaporisation qui s'établit quand on interpose entre la paroi de l'éprouvette métallique et le liquide de refroidissement, une résistance thermique constituée par un film d'une substance de faible conductivité thermique. Ce mode de vaporisation, l'ébullition larvée, se caractérise par une alternance mouillage-non-mouillage de la surface solide.

Deux conditions sont nécessaires à l'établissement de l'ébullition larvée: une condition de résistance thermique et une condition d'effusivité superficielle.

La substitution de l'ébullition larvée à la caléfaction permet d'accroître fortement la densité de flux de chaleur transmise entre une paroi solide à haute température et le liquide de refroidissement. Deux cas des liquides sont abordés: celui de l'eau et celui de l'azote liquide.

Auszug—Es wird die Form der Verdampfung beschrieben, die sich einstellt, wenn ein durch den Film eines Stoffes niedriger Waermeleitfaehigkeit gebildeter Waermewiderstand die Oberflaeche einer abgeschreckten Probe von der Kuehlfluessigkeit trennt. Diese Verdampfungsart, das teilweise Filmsieden, wird durch ein abwechselndes Benetzen und Abtrocknen der festen Oberflaeche gekennzeichnet. Zwei Bedingungen sind fuer teilweises Filmsieden erforderlich: Waermewiderstand, und Waermeabgabevermoegen der Oberflaeche. Durch teilweises anstelle von vollem Filmsieden kann der Waermestrom von einer festen Oberflaeche mit hoher Temperatur an die Kuehlfluessigkeit wesentlich gesteigert werden.

Резюме—Описан тип испарения, имеющий место при нахождении термического сопротивления, состоящего из пленки вещества с низкой теплопроводностью, между поверхностью закаливаемого вещества и охлаждающей жидкостью. Этот тип испарения, «личиночное» кипение, характеризуется перемежающимся смачиванием-несмачиванием твердой поверхности.

Для «личиночного» кипения необходимы два условия – термическое сопротивление и распространение по поверхности.

Замена пленочного кипения «личиночным» допускает сильное увеличение теплового потока между твердой поверхностью, находящейся при высокой температуре, и охлаждающей жидкостью.