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Hysteresis and contact angle effects in transition pool boiling of water

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Abstract—Transition boiling on a horizontal surface has been studied. Both transient heating and transient cooling were used. The heat flux into the boiling liquid is calculated from temperatures at two levels using an inverse heat conduction solution. The heat transfer is strongly affected by the value of the liquid contact angle, lower contact angles giving higher heat fluxes. Also there is pronounced hysteresis between heating and cooling with higher heat fluxes in heating. This hysteresis is observed at all contact angles.

INTRODUCTION

THERE has been a growing realization in recent years that transition boiling heat transfer is strongly affected by other parameters in addition to the known trend with wall temperature and the hydrodynamic limitations on the critical heat flux and the minimum film boiling heat flux. Although there is limited, generally qualitative, information in the literature, these ideas started to crystallize with the paper by Witte and Lienhard [1] which suggested the existence of a type of hysteresis in transition boiling, with higher heat transfer rates if the transition region is approached from the nucleate boiling side than if it is approached from the film boiling side. Also, in a comment on that paper, ref. [2] emphasized the importance of surface energy, with higher heat transfer rates being associated with better wetting, i.e. lower contact angles.

A number of early studies [3–7] associate improved heat transfer, particularly at the critical heat flux point, with improved wetting, but there appeared to be only two studies where data has been obtained over the entire transition region and where an attempt has been made to quantify the surface condition. Berenson [8] measured contact angles (0° and 10°) in his study of pool boiling of n-pentane on copper and found that the lower contact angle was associated with improved heat transfer. He also found that the value of the critical heat flux was unaffected by changes in the contact angle, in contradiction to other workers. Nishikawa *et al.* [9] made measurements with ethanol at contact angles of 0° and 50° and also found that the better wetting led to improved heat transfer but commented that there was evidence of gross contamination of the heat transfer surface.

More recently ref. [10] using a simple quenching technique with metal cylinders was able to cover a large range of contact angles in a reasonably continuous manner and thus confirm that transition boiling heat transfer increases with a decrease in the contact angle and that this effect includes both the critical heat flux and the minimum film boiling point. These results have been confirmed by Liaw and Dhir [11].

A number of studies have shown that the critical heat fluxes obtained in quenching tests are lower than steady-state values [12–14]. The same trend is observed when transient cooling (quenching) tests are compared with transient heating tests. Sakurai and Shiotsu [15] found higher heat fluxes at a given surface temperature in heating for wires and Liaw and Dhir [11] found the same for a vertical surface in pool boiling. Witte and Lienhard [1] hypothesized the existence of two transition boiling curves, one for cooling from film boiling and one for heating from nucleate boiling. They also suggested the possibility of sudden jumps from one curve to the other. Although there seems little evidence for jumps between two transition boiling curves a jump from steady-state transition boiling to an apparently novel type of steady two-phase heat transfer process was described in ref. [16].

The work to be described in this paper follows on from the work of ref. [10]. Although that appeared to be the first study of transition boiling to cover a reasonably wide and continuous range of contact angle the geometry of the heat transfer surface was not particularly well defined (a vertical cylinder). It was thought possible that the heat transfer conditions might vary slightly on different parts of the surface. This would have no effect on the observed trends with changing contact angle but might possibly explain the rather low peak heat fluxes that were recorded. Consequently it was decided to repeat the work with a horizontal, upward facing, surface. A transient technique was still used and it became possible to make measurements in both heating and cooling. At an early stage it became evident that there was a large hysteresis between the boiling curves obtained in heating and cooling. For these measurements sheathed thermocouples were used and the heat flux values calculated from a heat balance. Although it was not felt that either of these aspects of the technique could explain the hysteresis it was decided to repeat the

NOMENCLATURE

A_n defined by equations (3) and (4)
C defined by equation (2)
c specific heat
D defined by equation (2)
d thickness of control volume below boiling surface area (6 mm)
h_{lv} enthalpy change on evaporation
k thermal conductivity
q heat flux
q_s heat flux at boiling surface
q_{crit} critical heat flux
T_s temperature of boiling surface
TA_1, TA_2 average temperature at levels 1 and 2
T_{av} average temperature of a layer 3 mm deep below boiling surface
t time
z thickness between upper thermocouples and surface.

Greek symbols
α thermal diffusivity
δ separation of levels 1 and 2
θ contact angle
ρ density of metal
ρ_l density of liquid
ρ_v density of vapour
σ surface tension.

measurements using exposed junction thermocouples and to analyse the results using an inverse heat conduction routine.

POOL BOILING APPARATUS

The apparatus is shown in Fig. 1. The cylindrical test section made of commercially pure copper is heated from below by contact with a larger metal block containing four Watlow cartridge heaters (300 W/240 V). The pool of boiling water is confined to the top surface of the cylinder by means of a glass tube and a preformed PTFE seal. The diameter of the boiling heat transfer area at 26.6 mm is made close to that of the main part of the cylinder (32 mm) to encourage one-dimensional heat flow. The 1.5 mm thick flange at the top of the cylinder is needed to securely bolt on the glass tube but is not considered to significantly interfere with the heat flow (since it is well lagged).

Four thermocouple holes, 2 mm in diameter, were drilled at two levels, 3 and 20 mm, below the boiling surface. At each level one thermocouple is placed on the axis of the cylinder and one at a distance of 8 mm from the cylindrical surface. Type K thermocouples with 0.19 mm wires and bare junctions were used, soldered to the bottom of the holes with solder (Fry's metals number LS4, a lead/silver mixture). This solder is fully solid below 294°C and fully liquid above 305°C. The response time of the thermocouples in water is 0.01 s (manufacturer's data).

Prior to the boiling tests the thermocouples were calibrated against a further thermocouple by placing all five in a single hole in a metal block which was heated up and then allowed to cool slowly by natural convection in the room air. The fifth thermocouple was connected to a Comark 6110 digital thermocouple and the others to the data acquisition system. In this way an absolute error of around 1°C in all four temperature readings is likely, but the relative errors amongst the four thermocouples should be much less.

The four measurement thermocouples were each attached via a Flyde FE-254-GA amplifier and an A/D converter to a BBC microcomputer. To reduce

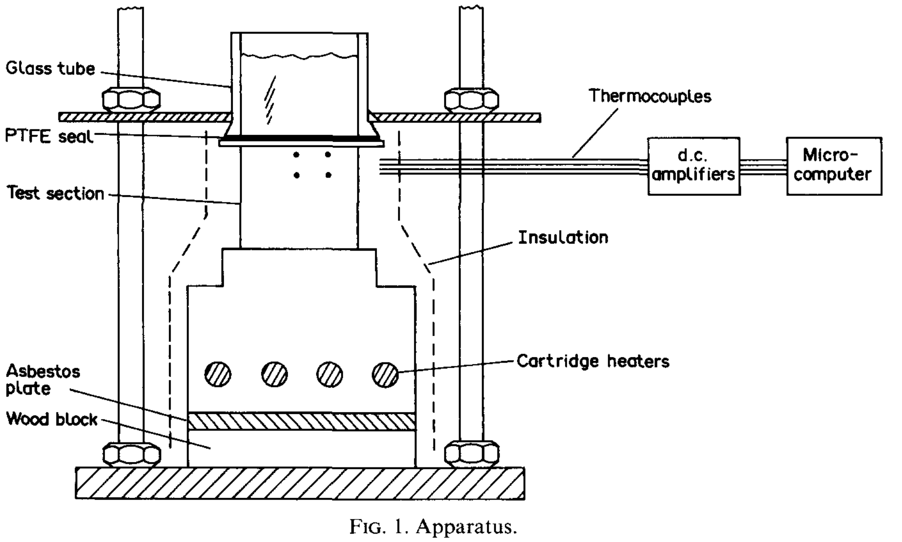

FIG. 1. Apparatus.

interference that was sometimes experienced the mains power supply to the equipment is taken through an R.S. plug-in mains filter.

Heating tests, with the cartridge heaters in the lower block on until the critical heat flux was exceeded, and cooling tests with the heater off, were conducted alternately. All four temperatures were read every 0.05 s and readings from an individual thermocouple averaged in groups of 12 (for most of the transition region including the critical heat flux) and groups of 110 (for low heat flux nucleate and transition boiling).

Only the average temperatures were printed out or used in calculating the heat flux, but even so the amount of data proved excessive. On average a heating run took 1 min and a cooling run 16 min, but the times decreased significantly for the later cooling runs (times from 300°C to the critical heat flux or vice versa).

Contact angles were measured at intervals at room temperature. The glass tube was removed and a drop of water placed on the heat transfer surface. The angle was measured using a microscope with a protractor eyepiece. It would be preferable to make the measurement at the temperatures of the boiling surface. It is doubtful if this could be done during boiling. Any apparent contact angles would be strongly influenced by dynamic effects. A true contact angle reading would require a separate pressure vessel, with windows capable of withstanding at least 20 bar.

DATA ANALYSIS USING KUDRYAVTSEV'S TWO-POINT METHOD

A literature survey revealed a number of analytical solutions to the one-dimensional inverse heat conduction problem but all the other methods required a known heat flux value at one position in the solid block. The Kudryavtsev two-point method [17] requires two known temperatures as a function of time. In our case these are TA_1, the average of the readings of the two thermocouples 3 mm below the surface, and TA_2 the average at 20 mm below the boiling surface. The method also assumes a uniform temperature at time $t = 0$, which is not true in our case, but after only a short time the results of the calculations become independent of the initial distribution.

The expression given by Kudryavtsev is (apart from a change of sign because we have heat flux towards the boiling liquid as positive)

$$q = \frac{k}{\delta}[TA_2(t) - TA_1(t)] + \frac{2k}{\delta}\sum_{n=1}^{n=\infty} e^{-(n\pi/\delta)^2\alpha t}\int_0^t e^{(n\pi/\delta)^2\alpha t} \times \left[(-1)^n \frac{d}{dt}TA_2(t) - \frac{d}{dt}TA_1(t)\right]dt \quad (1)$$

where q is the heat flux at position 1 and $\alpha = k/\rho c$ is the thermal diffusivity. In order to evaluate the expression degree six polynomials were fitted to the experimental data, i.e.

$$TA_1(t) = \sum_{i=0}^{6} C_{i+1}t^i$$

and (2)

$$TA_2(t) = \sum_{i=0}^{6} D_{i+1}t^i$$

and equation (1) evaluated by integrating by parts six times. After about $t = 1$ s certain transient terms become negligible and the heat flux at position 1 becomes

$$q = \frac{k}{\delta}[TA_2 - TA_1] + \frac{2k}{\delta}\sum_{n=1}^{\infty} A_n \quad (3)$$

where the A_n are given by

$$\begin{aligned}A_n = {} & \frac{1}{\left(\frac{n\pi}{\delta}\right)^2 \alpha}\{(-1)^n(6D_7t^5 + 5D_6t^4 + 4D_5t^3 \\ & + 3D_4t^2 + 2D_3t + D_2) - (6C_7t^5 + 5C_6t^4 + 4C_5t^3 \\ & + 3C_4t^2 + 2C_3t + C_2)\} - \frac{1}{\left(\frac{n\pi}{\delta}\right)^4 \alpha^2}\{(-1)^n(30D_7t^4 \\ & + 20D_6t^3 + 12D_5t^2 + 6D_4t + 2D_3) - (30C_7t^4 \\ & + 20C_6t^3 + 12C_5t^2 + 6C_4t + 2C_3)\} \\ & + \frac{1}{\left(\frac{n\pi}{\delta}\right)^6 \alpha^3}\{(-1)^n(120D_7t^3 + 60D_6t^2 \\ & + 24D_5t + 6D_4) - (120C_7t^3 + 60C_6t^2 + 24C_5t \\ & + 6C_4)\} - \frac{1}{\left(\frac{n\pi}{\delta}\right)^8 \alpha^4}\{(-1)^n(360D_7t^2 \\ & + 120D_6t + 24D_5) - (360C_7t^2 + 120C_6t + 24C_5)\} \\ & + \frac{1}{\left(\frac{n\pi}{\delta}\right)^{10} \alpha^5}\{(-1)^n(720D_7t + 120D_6) \\ & - (720C_7t + 120C_6)\} - \frac{1}{\left(\frac{n\pi}{\delta}\right)^{12} \alpha^6} \\ & \times \{(-1)^n(720D_7) - (720C_7)\}. \end{aligned} \quad (4)$$

We have given the result in full since, so far as we are aware, it is not available anywhere else. In ref. [17] the analysis stops at equation (1). The choice of degree six polynomials to fit the temperature data is a compromise between accuracy (less than degree six and the fit to the data was often poor) and computational convenience (equation (4) is already rather cumbersome). If a degree six polynomial is found to give an inadequate fit then the data can always be split into two or three regions and a different polynomial fitted to each. In this way it should be possible to use equation (4) over a wide range of practical situations.

The two-point method unfortunately only gives the heat flux at position 1, whereas what is needed is the heat flux at the boiling surface. However, we now have an accurate value of heat flux at a position only 3 mm below the surface and the extrapolation to the surface is small. The surface temperature is given by linear extrapolation as

$$T_s = TA_1 - z(TA_2 - TA_1)/0.017 \quad (5)$$

and the average temperature of the 3 mm surface layer is

$$T_{av} = (T_s + TA_1)/2. \quad (6)$$

The corrected surface heat flux q_s is given by

$$q_s = q - z\rho c \, \mathrm{d}T_{av}/\mathrm{d}t \quad (7)$$

where z is the thickness of material between position 1 and the boiling surface.

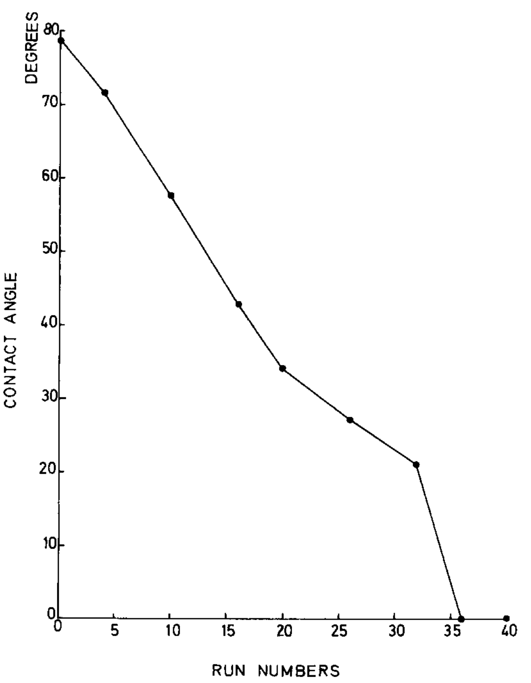

FIG. 2. Decrease of contact angle with time. The contact angle was zero after run 36.

RESULTS

A number of preliminary tests were conducted with a slightly different test section. The thermocouples were placed 3 and 9 mm below the boiling surface. This made it reasonable to calculate the heat flux into the boiling liquid from a simple heat balance on the top 6 mm of the metal block facilitating a check on the Kudryavtsev two-point calculation. The two methods of estimating the heat flux agreed extremely well except during rapid transients when a section of the boiling curve was traversed in less than 10 s. It is of course accepted that a simple lumped parameter approach, of the type used as a check in these preliminary tests, becomes inaccurate once the Biot number exceeds around 0.1. In ref. [14] an accurate inverse heat conduction solution diverged from the simple lumped parameter equation once the Biot number exceeded 0.45. In our experiments the Biot number (based on the 6 mm slab thickness) reached 0.7 at the critical heat flux.

However, with the two pairs of thermocouples only 6 mm apart it was felt that the measurement of temperature difference would be subject to error. For the main series of tests, described in this paper, the test section described earlier (Fig. 1) was used. All the main features of the results that follow also appeared in the preliminary tests.

The change of contact angle with run number is shown in Fig. 2 (all the results relate to a single surface). Boiling curves are shown in Figs. 3 and 4. Figure 5 shows the critical heat flux as a function of contact angle. In Fig. 6 is revealed the pronounced difference between heating and cooling.

Effect of contact angle

The change of contact angle with run number (or time) is shown in Fig. 2. The change in the boiling curve for successive runs was quite small so in Figs. 3 and 4 only three runs are shown. It is clear that the whole of the transition region is affected by the changing contact angle and that as the contact angle falls the heat transfer improves, confirming the results of ref. [10]. If attention is confined to the critical heat flux then more results can be displayed (Fig. 5). The steady rise of critical heat flux as the contact angle falls is clear.

The fine scale irregularities in the boiling curves probably result from experimental error in determining the temperatures. Some of the larger features, such as the dips at around 75 K superheat in runs 3 and 23 (Fig. 3) and the peak at around 95 K superheat in run 2 (Fig. 4), are considered to be real. On various occasions such peaks or dips were noted to be associated with increased noise, or the formation of a particularly large single vapour region that took a few seconds to clear the test section, or vapour bubble formation almost ceasing for a few seconds. Although in the majority of cases in the literature measurement of boiling curves has produced the expected trend, i.e. a monotonic decrease of heat flux as temperature rises in the transition region, other workers have reported a subsidiary heat flux peak at surface temperatures above the critical heat flux point, e.g. refs. [9, 18]. The dip in heat flux is possibly connected with the early stages of a change to a low heat flux mode where the surface becomes covered with a stable vapour film and evolution of vapour bubbles ceases. This has been

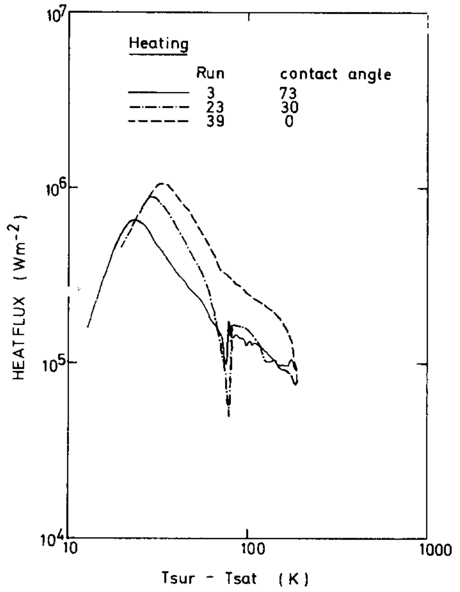

FIG. 3. Increase in heat flux at a given surface temperature as contact angle falls for heating.

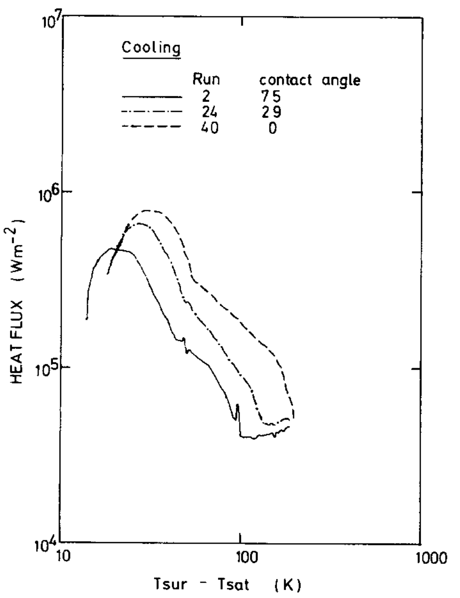

FIG. 4. Increase in heat flux at a given surface temperature as contact angle falls for cooling.

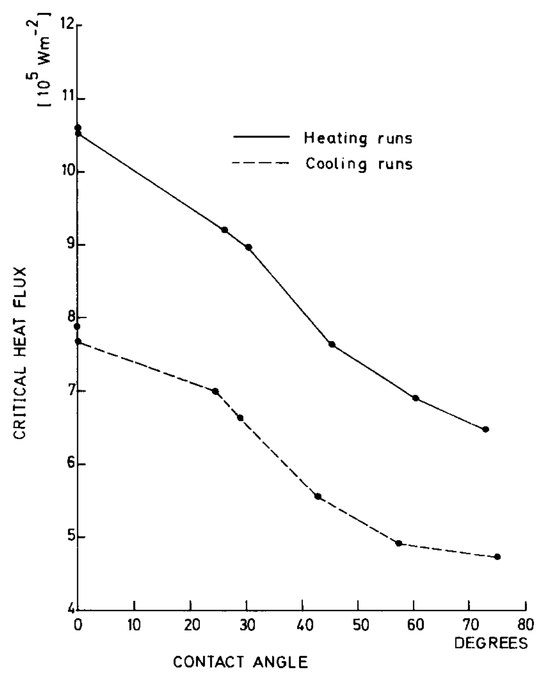

FIG. 5. Critical heat flux as a function of contact angle.

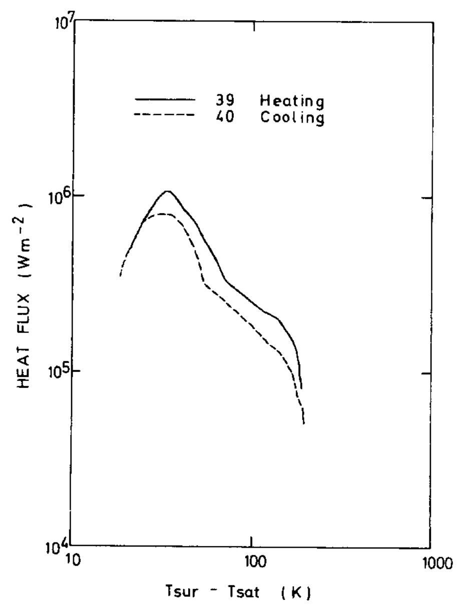

FIG. 6. Hysteresis between heating and cooling.

observed in steady-state boiling [16] and, once only, in quenching [19]. However, in the present measurements the low heat flux behaviour did not persist.

Hysteresis

Hysteresis between heating and cooling is implied by Figs. 3 and 4. To make this clearer a pair of successive heating and cooling runs is shown in Fig. 6. A large degree of hysteresis is present in all the results, not changing much with contact angle. For a given superheat the heat flux in heating is higher than that in cooling throughout the transition region including the critical heat flux. There is some indication that the curves are coming together in nucleate boiling.

DISCUSSION

In general we cannot expect the critical heat flux values to be predicted by theories of the hydrodynamic type since these theories give just a single value for water at atmospheric pressure, independent of surface energy or transient heating or cooling. However, if we assume that the hydrodynamic limi-

Table 1. Fall in critical heat flux associated with increase in contact angle

Reference	Low contact angle (deg)	High contact angle (deg)	Fall in critical heat flux (%) Heating	Cooling
Present work	0	75	39	40
[10]	0	73	—	31
[11]	38	107	30	65
[10]	58	102	—	66

tation on the counterflow of liquid and vapour does apply in the steady state when the wetting of the surface does not impose its own limitation, i.e. for a contact angle of zero, then a comparison can be attempted.

Zuber [20] and Kutateladze [21] both give equations as given below

$$q_{\mathrm{crit}} = K\rho_{\mathrm{v}}h_{\mathrm{lv}}\{\sigma g(\rho_{\mathrm{l}}-\rho_{\mathrm{v}})/\rho_{\mathrm{v}}^{2}\}^{1/4}. \quad (8)$$

Zuber gives $K = 0.13$ and Kutateladze gives $K = 0.168$. This means critical heat flux values for water of 1.1×10^6 and 1.4×10^6 W m^{-2}, respectively, in reasonable agreement with the experimental value for heating.

The improvement in transition boiling heat transfer with improved wetting is distinct. With the recently reported results of Liaw and Dhir [11] there are three sets of data available for water and it is of interest to compare all three. In ref. [11] there is no information for zero contact angle, but results are given for 38° and 107°. In Table 1 the comparison is attempted for the critical heat flux, both for a range of low, wetting, contact angles and for a range of higher contact angles extending into non-wetting conditions. The quantitative agreement is not particularly good but the trend is clear. The critical heat flux falls with increase in contact angle, over a wide range of angle, and regardless of the direction of the transient.

The hysteresis between heating and cooling is very marked in all pairs of runs. The concern mentioned in the introduction that the hysteresis might be an illusion resulting from the use of slow response thermocouples or an inaccurate method of calculating the heat flux has not been substantiated. At 0° contact angle the critical heat flux in cooling is 26% less than in heating. At 75° contact angle the reduction is 27%. In ref. [11] there are no results for a 0° contact angle with water but the reductions due to hysteresis at 38° and 107° are 18 and 60%, respectively. Partly by extrapolation of these results and partly using a measurement of virtually zero hysteresis in Freon-113 (which displayed 0° contact angle) it is argued in ref. [11] that there is no hysteresis at 0° contact angle. The results for water in this paper do not agree with this conclusion.

CONCLUSION

Heat transfer in transition boiling, including the critical heat flux, is strongly affected by both contact angle and the rate of heating or cooling. For a given surface temperature the heat flux throughout the transition region improves with better wetting, i.e. lower contact angle.

There is pronounced hysteresis between heating and cooling, heat fluxes being higher in heating. This was observed at all contact angles.

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EFFETS D'HYSTERESIS ET D'ANGLE DE CONTACT DANS L'EBULLITION D'EAU EN RESERVOIR

Résumé—On étudie l'ébullition de transition sur une surface horizontale. Le chauffage et le refroidissement variable sont tous deux utilisés. Le flux thermique dans le liquide bouillant est calculé à partir des températures à deux niveaux en utilisant la solution inverse de la conduction de chaleur. Le transfert thermique est fortement affecté par la valeur de l'angle de contact liquide, des angles plus faibles donnant des flux plus élevés. Il y a aussi une hysteresis prononcée entre le chauffage et le refroidissement avec des flux thermiques plus élevés dans le chauffage. Ce phénomène est observé pour tous les angles de contact.

DER EINFLUSS DER HYSTERESE UND DES RANDWINKELS BEIM BEHÄLTERSIEDEN VON WASSER IM ÜBERGANGSBEREICH

Zusammenfassung—An einer horizontalen Oberfläche wurde das Sieden im Übergangsbereich untersucht. Sowohl instationäre Beheizung als auch instationäre Kühlung wurden benutzt. Die an die siedende Flüssigkeit übertragene Wärmestromdichte wurde aus den Temperaturen an zwei Stellen durch Lösung des inversen Wärmeleitproblems berechnet. Der Wärmeübergang wird stark durch die Größe des Flüssigkeits-Randwinkels beeinflußt, kleinere Randwinkel ergeben höhere Wärmestromdichten. Ebenso existiert eine ausgeprägte Hysterese zwischen Heizen und Kühlen, mit höheren Wärmestromdichten beim Heizen. Diese Hysterese wurde bei allen Randwinkeln festgestellt.

ЭФФЕКТЫ ГИСТЕРЕЗИСА И КОНТАКТНОГО УГЛА ПРИ ПЕРЕХОДНОМ РЕЖИМЕ КИПЕНИЯ ВОДЫ В БОЛЬШОМ ОБЬЕМЕ

Аннотация—Исследовался переходный режим кипения на горизонтальной поверхности. Использовались нестационарные процессы нагревания и охлаждения. Тепловой поток в кипящую жидкость рассчитывался по температурам на двух уровнях с помощью решения обратной задачи теплопроводности. На теплообмен сильное влияние оказывает величина контактного угла жидкости; причем меньшим контактным углам соответствуют более высокие тепловые потоки. При нагреве в случае больших тепловых потоков ярко выражен гистерезис между нагревом и охлаждением. Он наблюдается при всех контактных углах.