BRIEF COMMUNICATION

CINEMICROPHOTOGRAPHIC STUDY OF BOILING OF WATER-IN-OIL EMULSIONS

YASUHIKO H. MORI, HIROSHI SANO and KAZUNARI KOMOTORI

Department of Mechanical Engineering, Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama 223, Japan

Abstract—Cinemicrophotography is applied to the boiling of emulsions of water dispersed in oils whose boiling points are higher than that of water. The layer of emulsion is made thin enough to make possible a microscopic observation by transmitted light through the layer. Two alternative processes of bubble formation are found: a periodical bubble formation at specified sites on the heated solid surface contacting the emulsion and a casual bubble formation some distance away from the surface. The basic mechanisms of those processes are discussed.

1. INTRODUCTION

The characteristics of boiling heat transfer to emulsions have been studied by a few groups of researchers. Tachibana & Enya (1972) and Satoh & Shoji (1973) carried out transient boiling experiments in which a heated piece of copper was dipped into a pool of emulsion composed of water and spindle oil. Recently Mori *et al.* (1978) studied the steady pool boiling heat transfer from an electrically heated nickel wire to various kinds of emulsions. Because of the opacity of emulsions, no observation of boiling behavior was made in those studies; and hence the mechanisms of boiling have not been revealed.

In general, an emulsion is composed of water and an oil immiscible with water. Some surfactant may be dissolved in either of them as an emulsifying agent. If the boiling points of water and the oil are quite different, the one with lower boiling point is exclusively subjected to the phase change. Therefore, it is presumed that there is a large difference in the boiling behavior between two cases: one is the case when a liquid with lower boiling point is continuous and the other the case when it is dispersed. The former case corresponds to oil-in-water emulsions and the latter to water-in-oil emulsions provided oils with higher boiling points are used as in the previous studies cited above. The basic mechanisms of nucleation and bubble growth in oil-in-water emulsions may not differ essentially from those in pure liquids, while those in water-in-oil emulsions seem to be significantly different. These matters will be revealed only by such microscopic observations that the performances of respective emulsion globules and the interactions between themselves and between them and the heating surface or vapor bubbles can be identified.

This work aims to observe microscopically the boiling behavior in water-in-oil emulsions and to shed some light on the matters described above. For this purpose, the layer of an emulsion in which boiling occurred was made so thin (1 mm or less) as to permit the light to transmit from behind to front and to make each water globule perceptible throughout the three kinds of experiments described in sections 2–4. Though such a peculiar condition may cause a minor difference in the boiling behavior from the usual pool boiling or flow boiling, it is unlikely to be a source of any essential trouble as far as the present objective described above is concerned.

In the first experiment described in section 2, a heating wire was immersed in an emulsion layer. Two alternative processes of bubble formation were distinguished: a periodical bubble formation at specified sites on the wire surface and a casual bubble formation away from the surface.

The former process was studied further in the experiment described in section 3. An emulsion was passed in the gap between parallel glass plates. One of the plates was heated so

that the periodical bubble formation was maintained at a specified site on its surface with artificial scratches.

The experiment described in section 4 aimed to examine the possibility of trapping of water globules into a bubble while growing. The peripheral region of a bubble growing two-dimensionally between parallel glass plates was observed.

The last section gives a summarized explanation, deduced from those experiments, of bubble formation processes and notes the possible heat transfer mechanism in relation to the bubble formation processes.

2. FREE CONVECTION BOILING ON WIRE SURFACE

Experimental

Figure 1 shows a schematic of the test section. The heating wire was made of nickel 0.2 mm in diameter and 15 mm long, stretched horizontally between two copper stays, and pressed on a glass strip fixed on the bottom of a glass pan. An emulsion was poured into the pan up to such a level that the wire was completely submerged. Since the width of the glass strip on which a thin layer of the emulsion was formed was restricted to 3 mm, the emulsion was continuously carried from the bulk in the pan to the wire surface by the free convection so that a rapid change in the constitution of the emulsion near the wire was prevented. Power was supplied to the heating wire from a regulated d.c. supply, and was determined from the resistance of the wire measured by use of a Wheatstone bridge and the voltage drop across the wire measured by use of a digital voltmeter. The temperature of the wire surface was known from the resistance of the wire. A thermocouple installed at a certain position in the pan yielded a representative bulk temperature which was controlled by two 200-W cartridge heaters beneath the pan. The bulk temperature was regulated to $40 \pm 1°C$ throughout this experiment, since higher temperatures were unfavourable for the stability of emulsions. The light from a 500-W ultra high voltage mercury-vapor lamp through a condenser was shed from below the glass strip. Directly above the heating wire was set vertically the body tube of a microscope (Nikon Type SIR, used at $30 \times$ magnification) connected to a 16-mm high-speed camera (Hycam Type 40-0001, operated at no more than 5000 frames/s). A nozzle was set close to the object lens of the microscope. The air jet from the nozzle protected the lens from the spattered oil droplets and the mist from the emulsion layer near the heating wire during the boiling operations. The thickness δ of emulsion layer on the glass strip was measured after each run by use of a probe attached to a micrometer head. It should by noted, however, that the actual thickness while some power was fed to the wire was less than the nominal thickness measured as described above, because the emulsion was drained outward from the position just above the wire due to a thermocapillary force exerted on the surface of the emulsion layer.
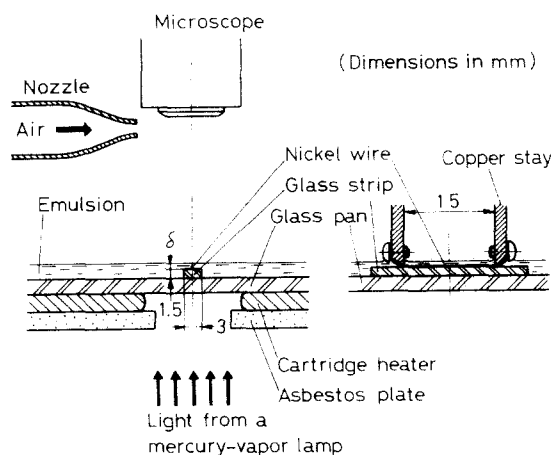

Figure 1. Schematic of test section.

Oils used in preparing emulsions were n-undecane of reagent grade and methylphenyl silicone oil (KF 54 fluid prepared by Shin-Etsu Kagaku Co.). Their boiling points are 196°C and above 400°C respectively under the atmospheric pressure (101.3 kPa). Span 80 (sorbitan monooleate) was addet to the oil phase as the emulsifying agent to 1 per cent of the expected volume of emulsion. Emulsions with adequate globule sizes for the microscopy were obtained by the intermittent shaking method (Gopal 1968). The ultrasonic method such as used in our previous study (Mori et al. 1978) was not used in the present experiment, because it provided so small globules that the superposition of a number of globules in the thickness of emulsion layer on the glass strip prevented the identification of each globule. The volume fraction of water was set at 10 per cent in water-in-KF 54 emulsion and 20 per cent in water-in-n-undecane emulsion for practical reasons. In emulsions with lower fractions of water, boiling hardly occurred at moderate heat fluxes; emulsions with higher fractions of water were unfavourable for the observation.

The details of this experiment and of those described in the later sections are presented elsewhere (Sano 1979).

Results and discussion

Figure 2 illustrates the boiling heat transfer characteristics in terms of the average heat flux on the wire surface, q, vs the excess temperature of the wire on the boiling point of water, ΔT_s, to demonstrate the conditions peculiar to this experiment. A large subcooling in the bulk and a vigorous circulation in the layer on the glass strip due to thermocapillarity gave high heat fluxes at low excess temperatures compared to those in our previous study (Mori et al. 1978). Cinemicrophotographs were taken exclusively at heat fluxes just above the boiling inception points, because at higher fluxes the high populations of bubbles near the heating wire made it hard to discern each process of bubble formation. In figure 2 are indicated by crosses the heat fluxes and the excess temperatures of the wire in the runs the sequences in which are exemplified below.

Examination of a number of cine films reveals that there are two different processes of bubble formation. Figure 3(a) shows a sequence, in one of the runs for water-in-KF 54 emulsion, in which those two processes are observed simultaneously. At frame 3, two bubbles start to grow at a certain site on the wire surface (indicated by the left-side arrow). Bubbles successively appear at the site as time passes. It is evident that some amount of water, though it is not perceptible, is attached to the site which has some favorable condition for nucleation. The mechanism of nucleation in this type of bubble formation seems to be the same as in the usual nucleate boiling of pure liquids. At frame 2, a bubble starts to grow in the emulsion a
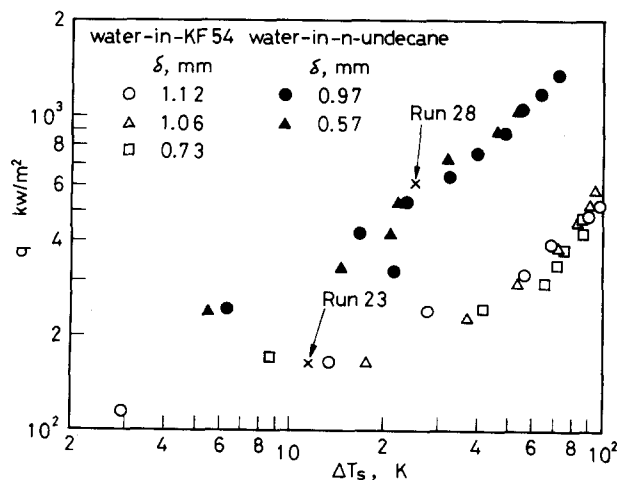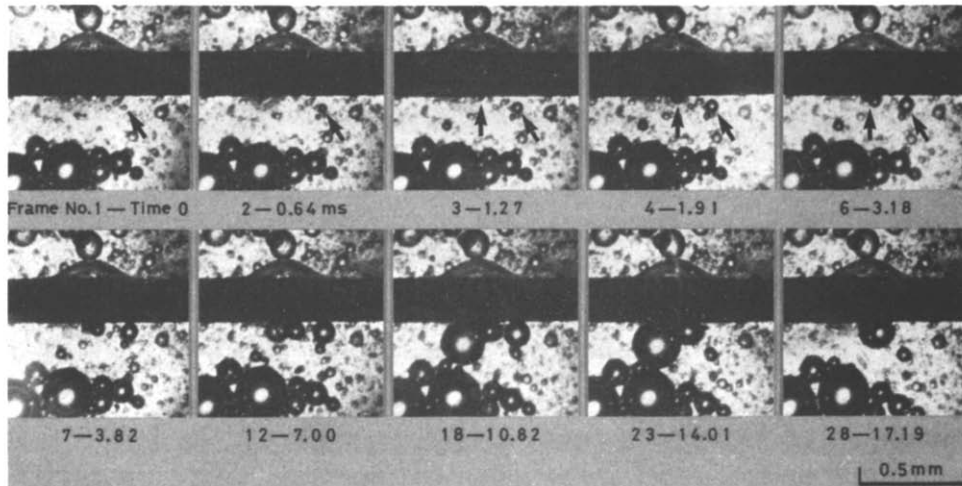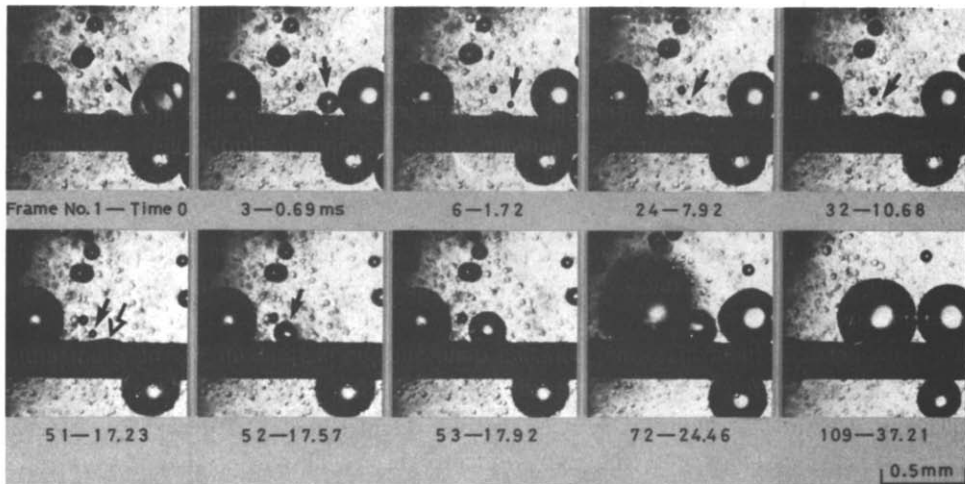

Figure 2. Heat transfer characteristics.

(a)

(b)

Figure 3. Sequences of boiling. (a) Water-in-KF 54 emulsion, Run 23, $q = 163 \, \text{kW/m}^2$, $\Delta T_s = 11.6 \, \text{K}$, $\delta = 0.93 \, \text{mm}$. (b) Water-in-$n$-undecane emulsion, Run 28, $q = 654 \, \text{kW/m}^2$, $\Delta T_s = 25.4 \, \text{K}$, $\delta = 0.82 \, \text{mm}$.

small distance away from the wire surface (indicated by an arrow), contacts with the wire surface during its growth (frame 7), and finally bursts into the air (frame 23). Apparently the nucleation in a water globule possibly superheated to some extent near the wire surface causes that bubble formation. Though this type of bubble formation occurs incidentally throughout this run, the former type (bubble formation on the surface) is apparently predominant.

Figure 3(b) shows a sequence in one of the runs for water-in-n-undecane emulsion. At frame 3 a bubble is released from the wire surface and then shrinks rapidly by condensation because of the large subcooling. It does not collapse completely, but becomes a tiny bubble probably because of a small amount of air contained in the bubble initially. It is assumed that the condensate envelopes or is attached to the bubble although this cannot be discerned on the photographs. As it approaches the wire surface again, it starts to grow and contacts the surface at frame 52. After contacting the surface, it grows to a larger size than before its release from the surface. It is assumed that the water attached to the wire surface (low conic lump indicated by the right-side arrow at frame 51) evaporates into the bubble after the bubble again contacts the wire surface. The numbers of bubble formation in the emulsion as shown in this figure and of periodical bubble formation at specified sites on the wire surface are 26 and 59 respectively

throughout the cine film which covers 1.2 mm in length of the wire and 2.0 s of the duration in this run.

Roughly speaking, the rate of bubble formation on the surface and that in the emulsion are comparable in water-in-KF 54 emulsion, but in water-in-n-undecane emulsion, the former is overwhelmingly predominant in most runs. This difference is primarily ascribed to the fact that KF 54, with viscosity two orders of magnitude greater than that of n-undecane, permits each tiny bubble, resulting from either the trapping of the air in preparing the emulsion or the bubble condensation as shown in figure 3(b), to drift around the wire for a longer time than does n-undecane, and thus yields a higher rate of bubble formation in the emulsion.

It should be noted that either of the two processes of bubble formation described above occurs not always but conditionally. The bubble formation on the surface requires the condition that the water separated from the emulsion comes into direct contact, displacing oil, with the surface at some specified sites which have some favorable condition for nucleation. Bubbles are formed at those sites periodically while those sites are wet with water. Apparently this process depends heavily on the the relative wettabilities of the surface with water and with oil. The bubble formation in the emulsion depends at least partly on the air contained in the emulsion in the form of tiny bubbles included in or attached to water globules or of dissolved air in water globules and the oil. The subcooling in the bulk also contributes to this process in some cases as seen in figure 3(b).

3. FLOW BOILING ON GLASS PLATE

Experimental

This experiment was designed for further examination of the periodical bubble formation at specified sites on the heated surface as seen in the experiment described in section 2. An emulsion flowed from a reservoir on to a heated surface to ensure observation of steady boiling for a much longer time than in the preceding experiment. It was desired that the heated surface be readily wet with water to yield bubble formation on the surface. For this reason and for convenience of observation, the surface of a flat glass plate was used as the heated surface.

Figure 4 shows a schematic of the test section. Two glass plates constituted a horizontal channel 23 mm wide and 1 mm high through which the water-in-KF 54 emulsion prepared as described in section 2 was passed and discharged into the atmosphere. The downward facing
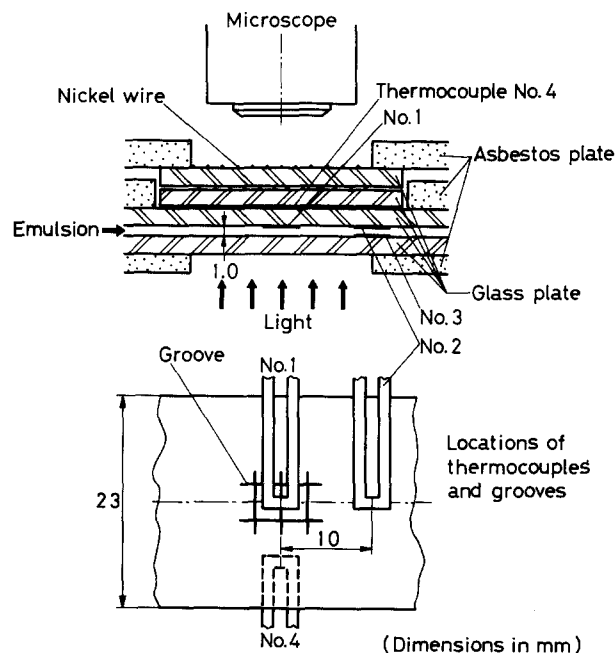

Figure 4. Schematic of test section.

surface of the upper plate served as the heated surface. By scratching with a diamond glass cutter, grooves each 0.18 mm wide, 0.02 mm deep at the maximum and 7–10 mm long, were provided at right angles to the surface. These grooves presented a number of potential nucleation sites. The surface was heated from behind by an electric heating wire wound round a glass plate so sparsely as to permit the observation of bubble formation on the surface from behind. The input to the heating wire was so regulated that bubbles were formed at only one site in the test section. For the purpose of rough estimation of surface and bulk temperatures, three film-type copper–constantan thermocouples (Nos. 1, 2, 3) 50 μm thick were stuck onto the inner surfaces of the channel, and one (No. 4) onto the reverse side of the upper plate. In each run, the temperatures T_1, T_2, T_3, from the former three were calibrated against the fourth, and the former three were then removed before the cine-photography. Still photography was also made by use of a 35-mm camera (Canon FTb) in two of the runs. By traversing the test channel in the reverse direction to emulsion flow, several pictures were taken to show the variation of bubble size with the distance x from the nucleation site in question, in a wider range of x than covered in the cine-photography. The stability of a nucleation site was examined by keeping an eye on it for 20–40 min in every run.

The experimental conditions in each run are given in table 1 together with some data dicussed later. The surface temperatures T_1, T_2, T_3 given there indicate that the bulk of emulsion in most of the test section was superheated to some extent above the boiling point of water contrary to the preceding experiment.

Results and discussion

The still photograph exemplified in figure 5 shows a train of bubbles flowing with the surrounding emulsion. The arrow in the photograph indicates the nucleation site generating the bubbles. Contrary to our expectation, we could not find on the photographs any blob of water covering a nucleation site. This may be ascribed to a very small contact angle of the blob surface (water–oil interface) to the glass surface.

The still photography shows the variation of bubble diameter with the distance x from the nucleation site as illustrated in figure 6(a). Figure 6(b) exemplifies, based on the cine-photography, the diameter-x histories of eight bubbles formed successively in a specified run. It should be kept in mind that each water globule 30 μm diameter (typical size of water globule in the water-in-KF 54 emulsion used in this study) would vaporize to a saturated-vapor bubble of 0.36 mm diameter and the thermal expansion of vapor due to the temperature rise from 100 to 200°C results in an increase of bubble diameter only by 9 per cent. Thus the possible reasons for such an increase of bubble diameter with x as shown in figures 6(a) and 6(b) are:

(i) Adjacent bubbles in a bubble train coalesce to each other.

(ii) Water globules around a bubble are trapped into the latter and evaporate while flowing.

(iii) Some amount of water is entrained with a bubble when the bubble is released from the blob of water covering the nucleation site, and evaporates into the bubble while flowing away from the site.

The cine-photography indicated that the coalescence of bubbles rarely occurred at least in the range covered. It gave us no information about the trapping of water globules into a flowing bubble. However, the experiment described in next section suggested that such trapping was not likely to occur in the short duration of each bubble flowing in the test section. Thus, the most probable reason is the last one. It is assumed that each bubble is enveloped by the entrained water. Such a type of "two-phase bubble" is permitted to be present transiently due to the limited rate of drainage of the enveloping water film (see Mercier *et al.* 1974, Mori 1978), though it may not be stable in the present system.

It was confirmed in every run that the bubble formation at a specified site continued steadily throughout the duration of observation from 20 to 40 min. The quantity of water consumed by the vapor generation in that duration was estimated tentatively by introducing a somewhat

Table 1. Summarized data on flow-boiling experiment

Run No.	Temperatures T_1, T_2, T_3 °C			Superficial velocity of emulsion mm/s	Bubble formation frequency Hz	Bubble diameter at $x = 1$ mm ** mm	Duration of observation † ks	Volume of saturated water vaporized in duration of observation †† mm^3
82	135	*	*	4.9	11.7	0.44	2.40	0.77
83	139	*	*	5.4	11.1	0.53	1.20	0.64
85	170	214	140	7.0	45.5	0.37	1.35	1.02
86	159	204	132	6.5	9.4	0.45	1.20	0.32
87	138	184	116	5.6	10.8	0.35	1.80	0.25

* Temperature fluctuated due to the bulk boiling.

** Sauter mean based on cine-photography.

† Bubble formation continued throughout this duration.

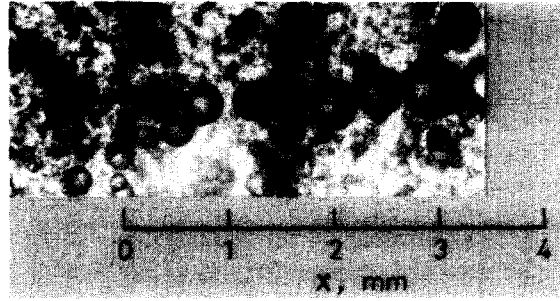†† Calculated by assuming the completion of vaporization and no superheating of vapor at $x \approx 1$ mm.

Figure 5. Train of bubbles formed at a specified site (Run 85).
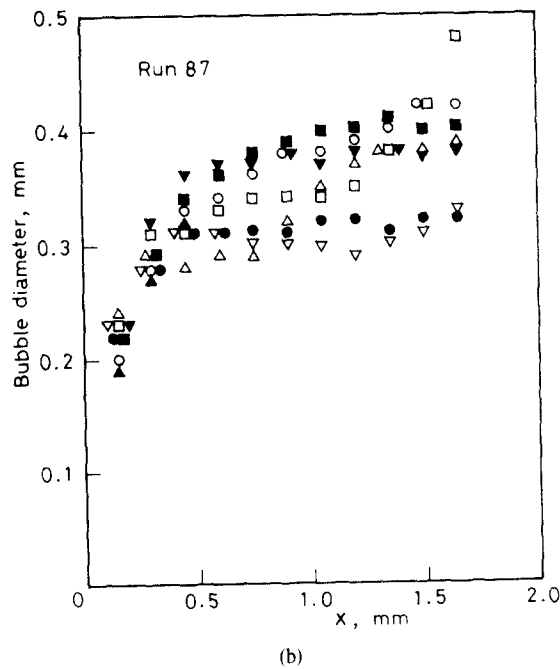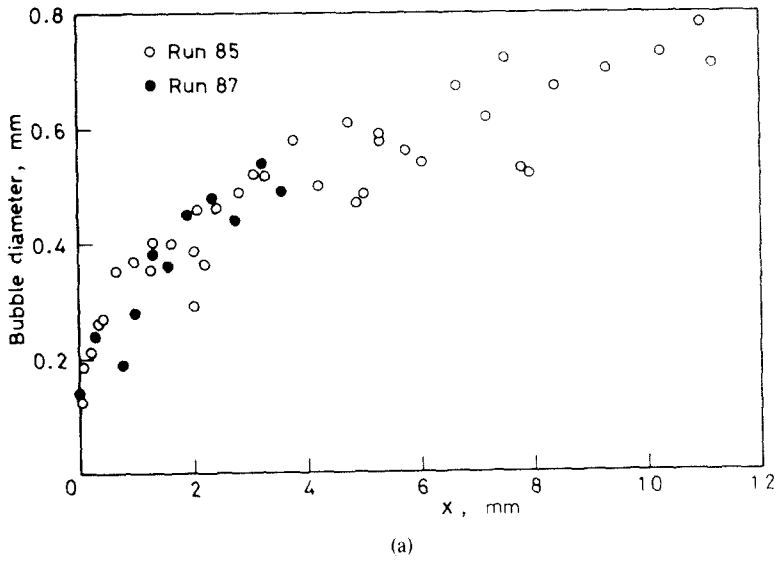

(a)

(b)

Figure 6. Variation of bubble diameter with distance from nucleation site. (a) Measurements based on still photography.
(b) Histories for respective bubbles based on cine-photography.

arbitrary assumption and is given in table 1. Here, each bubble was assumed, at $x = 1$ mm, to be filled with saturated vapor and not accompanied by any amount of unvaporized water, i.e. the vaporization was assumed to be just completed at $x = 1$ mm. Since some amount of water seems to be still present with bubble at $x = 1$ mm based on figures 6(a) and 6(b), this assumption would result in an underestimation of the quantity of water taken away from the nucleation site in that duration. Nevertheless the quantities of water calculated as stated above are 10^4–10^5 times as large as that of a water globule 30 μm diameter.

This fact can be explained by assuming a blob of water to cover the nucleation site, its volume exceeding that of a typical water globule in the bulk of emulsion at least by a factor of 10^4. Such a blob of water may result from the separation of water in the emulsion layer adjacent to the heated surface. It should be mentioned, in this connection, that the volume of each groove on the heated surface is about 0.02 mm^3 and so is much less than that of the water blob. The water blob must spread overflowing around the groove, even if the water fills up the groove extending its whole length displacing the emulsion.

However, this may not always be the case. A water blob smaller than the one estimated above can maintain the bubble formation long enough, if water is supplied continually at some rate to the water blob by, for example, the collision and coalescence of flowing water globules with the water blob to compensate fully or partly the loss of water by the bubble formation. For full compensation for the loss of water 10–50 globules have to coalesce with the water blob per second. We cannot evaluate for the present the possibility of such a coalescence.

Single bubbles were observed to appear at times in the bulk of emulsion throughout the test section, in a part of which the bulk was little superheated above the boiling point of water. This fact is reasonably ascribed to the presence of tiny air bubbles, included in or attached to water globules, which were entrapped in the emulsion during the process of its preparation.

3. BULK BOILING DUE TO DEPRESSURIZATION

Experimental

This supplementary experiment aimed at obtaining some knowledge about the interaction between a growing bubble and adjacent water globules in the emulsion. Bubbles were made to grow nearly two-dimensionally in a narrow gap between parallel glass plates so as to permit the observations of inside and outside regions abutting the bubble surface. To make bubbles form in a uniformly superheated emulsion at a desired instant, an emulsion pressurized and uniformly heated beforehand in a test cell was suddenly subjected to a depressurization to the atmospheric pressure while the high-speed camera was running.

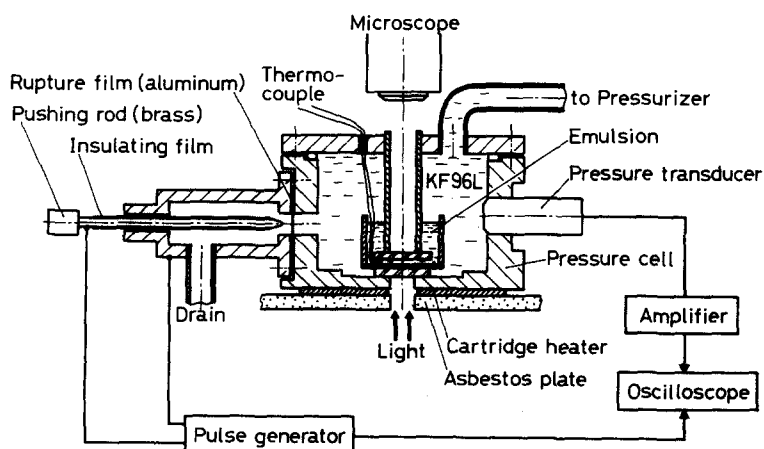Figure 7 shows a schematic of the apparatus. A cylindrical cell with a glass bottom plate was

Figure 7. Schematic of apparatus.

filled with the water-in-KF 54 emulsion and was set on the window at the bottom of the cylindrical pressure cell made of brass. A hollow vertical cylinder whose lower end was sealed with a glass disk was so dipped into the emulsion that an emulsion layer $\delta = 0.6$–1.2 mm thickness was formed between the glass disk and the bottom plate of the cell containing the emulsion. A copper-constantan thermocouple was installed in this emulsion layer. The residual volume in the pressure cell was filled with a dimethyl silicone oil (KF 96L fluid, Shin-Etsu Kagaku Co.) immiscible with KF 54. After the pressure in the cell was raised to about 1.5 atm by the head of mercury in a U-tube connected to the cell, the temperature was increased to a prescribed level T_1 (up to 125°C) by two 200-W cartridge heaters beneath the pressure cell. Then the pressure was decreased rapidly by breaking the rupture film attached to the side wall of the pressure cell with a pushing rod while the high-speed camera was running. The pressure decreased to the atmospheric pressure within 0.3 s in every run.

Results and discussion

It was observed that a number of bubbles appeared in the bulk of the emulsion layer in the gap between the glass plates as the pressure decreased. As a bubble grew by evaporation and coalescence with other bubbles, it occupied the whole height of the gap and spread nearly two dimensionally in the gap. Figure 8 exemplifies a sequence showing growing peripheries of such two-dimensional bubbles. Some irregular-shaped drops of water attached to the glass plates are seen through the bubbles. No water globules in the emulsion adjacent to the bubble surfaces are found to break through the surfaces into the bubbles, the same as in all of the other sequences not shown here. This finding suggests that the entrapping of water globules into a bubble rarely occurs in the boiling conditions as employed in the experiments described in this paper, though it may occur at extremely high superheatings of the emulsion.

5. SUMMARY AND GENERAL DISCUSSION

Cinemicrophotography has distinguished two different processes of bubble formation in the boiling of water-in-oil emulsions contacting a heated solid surface. They are a periodical bubble formation at specified sites on the heated surface and a casual bubble formation in the emulsion away from the surface.

The former is modelled as illustrated in figure 9(a). Some amount of water separated from the emulsion wets a site on the heated surface displacing the emulsion. If the site has a cavity (or cavities) favorable for nucleation, bubbles appear there one by one just the same as in the usual nucleate boiling of pure liquids. Each bubble grows in contact with the surface by the evaporation of the water covering there, and then breaks through the water–oil interface into the bulk of the emulsion entraining a certain volume of water. The bubble continues to grow in the bulk of emulsion by the evaporation of the entrained water provided the bulk is superheated above the boiling point of water. If it is subcooled, the bubble condenses in the bulk to a water droplet. This process apparently depends on the wettabilities of the surface with
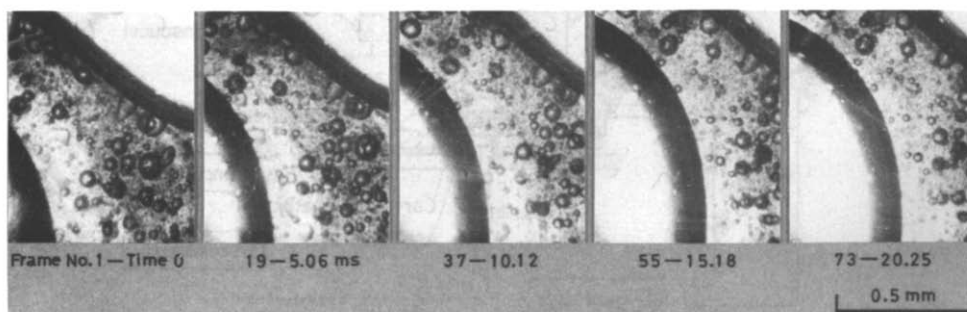

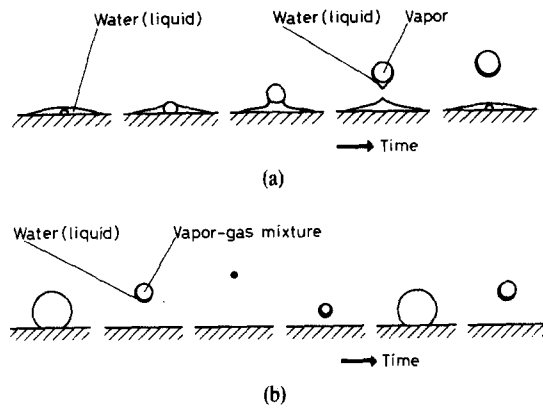Figure 8. Growing periphery of bubbles (Run 77, $T_1 = 112°C$, $\delta = 0.90$ mm).

Figure 9. Models of bubble formation processes. (a) Bubble formation on the surface when the bulk is superheated above the boiling point of water. (b) Bubble formation in the emulsion when the bulk is subcooled below the boiling point of water.

water and with oil. If the surface is wetted quite well with the oil, the bubble formation will be strongly suppressed.

The latter process is reasonably ascribed to the air contained in the emulsion. The intermittent shaking method used in this study as well as most of the other established methods of preparing emulsions are free from neither the dissolving of the air nor the entrapping of tiny air bubbles in the emulsions. Some of these air bubbles are presumed to be attached to or included in some of the water globules and result in a vaporization of those globules when the temperature is raised to the boiling point of water. The dissolved air in the water globules and in the surrounding oil may also affect the bubble formation in some cases. For example, figure 9(b) shows a schematic model of bubble formation in the emulsion, based on the observation in the first experiment stated in section 2. A bubble growing on the heated surface contains a small amount of air because of the diffusion, through the bubble surface, of the dissolved air in the water and the oil contacting the bubble. When the bubble is released from the surface into the bulk of emulsion subcooled below the boiling point of water, it condenses to a water droplet including a tiny air bubble. When it is carried up to the vicinity of the heated surface again by the convection around the surface, the bubble starts to grow by the evaporation of the water sheath.

In either of the two processes, the contribution of adjacent water globules to the bubble growth through their entry in the bubble seems to be negligible. This means that the bubble growth in the former process is caused exclusively by the separated water on the surface, while the latter process is considered to represent an individual vaporization of each water globule.

Above consideration on the bubble formation processes indicates the existence of the following modes of heat transfer.

(1) The sensible-heat transfer through solid–oil interface where the heated surface is wet with oil.

(2) The sensible-heat transfer through solid–water and water–oil interfaces in the region where the separated water contacts the heated surface displacing the emulsion.

(3) The heat transfer related to the periodical bubble formation in the above-mentioned region. The latent heat transferred by each bubble is primarily determined by the mass of vapour in the bubble when it breaks the water-oil interface into the bulk of emulsion. The unvaporized water entrained by the bubble evaporates after the bubble enters the bulk of emulsion, if the bulk is superheated above the boiling point of water, and thus contributes to the sensible-heat transfer (modes 1 and 2) by suppressing the temperature in the bulk. When the bulk is subcooled the only contribution to heat transfer, by translation of the entrained water, is sensible heat due to its temperature being higher than the bulk temperature.

(4) The heat transfer related to the casual bubble formation in the emulsion away from the surface. If a water globule stays in the thermal boundary layer adjacent to the heated surface for a finite period while vaporizing, some amount of latent heat is transferred by this process depending on the mass of vapor generated in that period. If not, this process only contributes to the sensible heat transfer (modes 1 and 2) through suppressing the temperature in the bulk.

The heat transfer rate due to each mode mentioned above has to be definitely evaluated for the quantitative interpretation of the heat transfer characteristics in boiling of water-in-oil emulsions. It should be noted also that the consideration described above may not always be valid at higher heat fluxes, since the present observations were made exclusively at low heat fluxes slightly above the boiling inception points. Our previous study (Mori *et al.* 1978) indicates that the temperature of the heated surface increases monotonically, sometimes exceeding the superheat limit of water, as the heat flux increases. The mechanism of boiling in such a range seems to be different from that described in this paper. These problems excluded from the scope of the present study have to be settled in the future.

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