

Research on

NUCLEATE BOILING

RUSSELL MESLER
University of Kansas
Lawrence, KS 66045

RESearch TAKES AN especially exciting turn when it requires us to revise our views of long held beliefs. Such is the case of recent research results concerning nucleate boiling. Although the picture is by no means complete, it appears possible that we may be changing several of our views concerning nucleate boiling.

Nucleate boiling is a familiar topic to any chemical engineer. All heat transfer texts devote sections to it in which the rudimentary facts are stated. All such treatments advise that nucleate boiling is a complicated subject and that it is not possible to go into a detailed discussion of the subject.

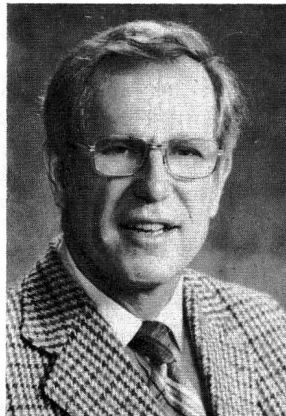
Nucleate boiling is classified as convective heat transfer. It is unusual convection on at least three counts. First, because of the latent heat, great quantities of heat can be exchanged with only a little change in temperature as long as liquid remains on the surface. Second, the density changes resulting from heat transfer are very large compared to usual convection. Third, it is only at a vapor-liquid interface that heat can be absorbed and vapor generated.

It is perhaps this third aspect that is most responsible for making nucleate boiling so inscrutable. Most of the vapor-liquid interface in boiling is provided by bubbles, but where do bubbles come from? Bubbles have short lives, escaping once they become large. A large source of nuclei is required.

It is important to recognize that liquids often resist the tendency to form bubbles. In the organic chemistry laboratory it is well known that liquids in a glass flask will superheat above their usual boiling point. Suddenly, when ebullition

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Russell Mesler is the Warren S. Bellows Professor of Chemical Engineering at the University of Kansas. His interest in nucleate boiling began in 1953 when he embarked on his doctoral research at the University of Michigan. He was a faculty member at the University of Michigan before coming to the University of Kansas in 1957.

does start, the boiling is so vigorous that the contents of the flask are likely to be expelled and wind up on the lab bench. The solution to the problem is the use of a boiling chip to reduce superheating and maintain steady ebullition.

The phenomenon of superheating is attributed to the action of surface tension. The pressure inside a small static spherical bubble is higher than outside by twice the surface tension over the radius, $2\sigma/r$. Such a static bubble would be in unstable equilibrium. If it were only slightly smaller, the increased pressure would tend to collapse it and if it were slightly larger, it would tend to grow and rise and thus escape.

The action of a solid on the stability of small bubbles is well exemplified by boiling chips. A boiling chip is able to retain gas or vapor on its surfaces and thus provides a vapor-liquid interface deep in the liquid able to generate vapor when the liquid becomes supersaturated. The vapor breaks away as bubbles rising to the surface but leaving behind some vapor from which the next bubble can grow.

Nucleate boiling is often characterized by the tendency for bubbles to rise repeatedly from certain points on the surface when boiling at low heat flux. These points are described as nucleation sites. They are thought to act as do boiling chips

by stabilizing a vapor-liquid interface on the surface.

Usually nucleation sites become active only after the surface temperature rises significantly above the boiling point. It is possible with very smooth surfaces to achieve even higher superheats before ebullition occurs, as in a glass flask. Once ebullition starts more nucleation sites become active, often with only a slight increase in surface temperature. It is only at low heat flux that the individual bubbles and nucleation sites can be effectively studied. At just a modest heat flux bubbles begin to merge and with just a bit more heat flux the commotion becomes so great that it is impossible to discern much detail at the center of activity. Views near the edge of a boiling surface may not be representative of the rest.

Because of the visual difficulty of studying bubbles at even modest heat fluxes most of the studies of bubbles have been made at low heat fluxes. The results of these studies have then been extrapolated to explain nucleate boiling at higher heat flux where applications are common.

At high heat fluxes more vapor is generated. In escaping the surface it pushes back the liquid and establishes a vapor region just above the surface. This leaves a film of liquid on the surface that becomes especially important in the transfer of heat [1, 2, 3, 4, 5].

Nucleate boiling is limited in its ability to transfer heat. Exceeding what is called the peak heat flux leads to the drying off of the surface. Without liquid the surface is unable to lose heat fast enough and the surface temperature rises unless the heat supply is reduced.

A surprising fact is that higher heat fluxes are possible if the depth of liquid on the surface is a few mm rather than much deeper [6]. Maintaining a film on the surface with a spray or with a jet permits even higher peak heat fluxes [4, 7, 8, 9].

PHASE TRANSITION PROCESSES

Boiling is not the only process involving a phase transition in which nucleation is important. Another is crystallization. In crystallization, crystal nuclei initiate crystal formation.

There are a number of similarities between crystal nucleation and vapor bubble nucleation. Both crystals and vapor bubbles will grow on surface imperfections such as scratches and pits. Crystallization also exhibits tribonucleation [10]. Crystallization is initiated by rubbing or dragging a stirring rod along the bottom of a beaker. Vapor

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nuclei can be produced in the same fashion.

There is another source for crystal nuclei for which no similar source of vapor bubbles has been reported. It is called secondary nucleation and is the production of nuclei from the break-up of larger crystals [11]. This source of nuclei is the most important one in industrial applications.

SEARCH FOR SECONDARY NUCLEATION

The reason secondary nucleation has never been seriously considered for nucleate boiling is probably because it was not easy to see how any of the abundant vapor could be returned to the liquid to serve as nuclei. Without a process for the return of vapor to the liquid it was impossible to see how secondary nucleation could occur in nucleate boiling.

The first clue to a process for secondary nucleation came in research on boiling in a thin liquid film [12, 13]. Nucleate boiling was studied on the outside of a vertical, steam-heated, copper tube. High speed motion pictures showed that when a bubble burst, bubble nuclei appeared where the bubble had just burst.

The next clue came in an experiment with superheated water in a glass tube [14]. An air bubble was formed in the water and was allowed to rise and burst at the surface. Soon after it burst, clusters of bubble nuclei appeared beneath the surface and these nuclei grew and coalesced to form a vapor bubble. When this bubble burst it too produced a cluster of nuclei beneath the surface. Viewing the high speed motion pictures of the event gave the impression that nuclei came from the top film of the bubble crashing upon the surface following the bursting of the bubble.

DROP-FORMED VORTEX RINGS

At this juncture a study of the tendency for small drops to entrain bubbles seemed appropriate. With just the simplest experiments using only an eye dropper and a beaker it was obvious that drops do entrain bubbles. With this encouragement high speed motion pictures were taken of drops falling several feet into water [15]. It was soon learned that entrainment occurred

not upon initial impact but later when the splash fell back upon the surface. This led to experiments with short falls where it was learned, by dyeing the drop, that when a dyed water drop strikes a water surface it usually produces a vortex ring. Furthermore, with optimized lighting it was seen that the vortex rings carry tiny bubbles on their axes as they plunge beneath the surface. See Figures 1 and 2.

Drop-formed vortex rings were news to us. Once one knows about them it's not difficult to find that they've been known a long time. They were described in an 1858 paper by W. B. Rogers [16] before he founded MIT. They are not mentioned in most texts on fluid mechanics, except for Sommerfield [17] and Batchelor [18]. A recent book *Bubbles, Drops and Particles* [19] includes no mention of them. Apparently few researchers have had occasion to learn of them.

We found that researchers at Los Alamos were not familiar with them. They developed a technique to solve the Navier-Stokes equations numerically with a computer [20, 21] and applied their method to the prediction of the behavior of a drop striking a pool of water. They could only solve for short falls. They compared their results to experiments with drops falling from greater heights. They found agreement and concluded that their method was sound, unaware that drops falling from short distances behave differently [22].

Now that drop-formed vortex rings have been recognized as providing a mechanism for secondary nucleation, it is pertinent to consider how important secondary nucleation might be. Vortex rings are capable of providing nuclei away from the interface. The tiny bubbles are carried within the rings wrapped in layers that come from near

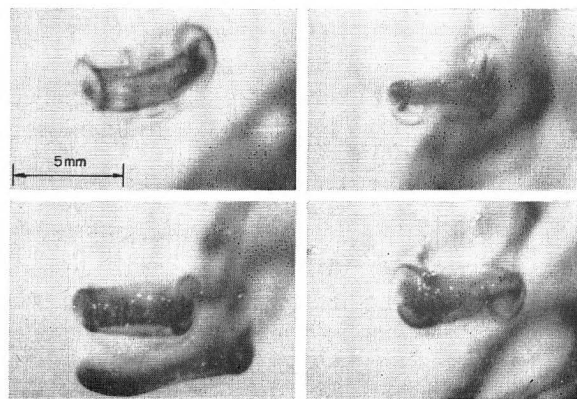


FIGURE 1. Examples of a drop-formed vortex ring entraining air bubbles.

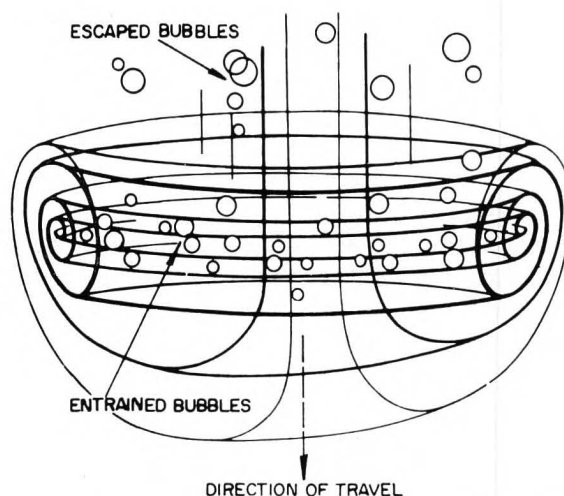


FIGURE 2. Sketch of a drop-formed vortex ring showing entrained and escaped bubbles.

the surface and the impinging drop that likely have temperatures close to saturation. The tiny bubbles are shielded from the local environment through which they are carried. If they escape to a hotter environment they can begin rapid growth. The fact that clusters of bubble nuclei are seen later beneath the surface when a bubble bursts on a superheated pool fits these circumstances.

When a bubble bursts on the surface, does its top film form drops that in turn form vortex rings? A bubble about 2 cm in diameter was blown from a detergent solution colored with food dye and placed on the surface of clear water. It was ruptured soon after placement and a picture was taken looking down on the surface. The picture showed tiny rings and bubbles where the meniscus had been [15]. Apparently the rings were vortex rings.

Another interesting aspect is the behavior of a vortex ring as it approaches a wall. It plows right in and spreads out, giving the impression that the tiny bubbles it might carry would be delivered very close to the wall. If the wall was a heater surface the tiny bubbles would seem to be delivered to a region of high temperature.

Although it goes unnoticed by current texts, it is possible to improve the performance of nucleate boiling by the simple means of boiling in thin liquid films. An early indication of this came from Japanese engineers who studied nucleate boiling on a horizontal surface with varying depths of liquid covering the surface [23, 24]. At depths below 5 mm, surface temperatures were lower and were lowest just before the surface dried off with the surface only partially covered with liquid.

They offered no explanation for the observation but did note that nucleation increased at lower liquid depths. Others have reported similar results [25, 26]. Boiling in thin liquid films offers an ideal opportunity for secondary nucleation to occur. When boiling thin liquid films, bubbles burst from the top of the film not far from the heater surface. Drops formed when a bubble bursts should be drawn into the film on the surface. Vortex rings formed by these drops would be well positioned to carry entrained bubbles to the surface. Near the surface the entrained bubbles would be exposed to the highest temperatures around. Whether secondary nucleation is indeed responsible for the better performance of boiling in thin liquid films cannot be said at present but it seems like a good bet. This would seem to be a likely candidate for further research.

One advantage of boiling from thin liquid films is that nucleate boiling can be achieved at lower surface temperatures and at lower heat fluxes. This is especially important in low heat flux applications where, judging only from boiling from submerged surfaces, nucleate boiling would not seem possible.

Nucleate boiling from thin films can be improved even more by the addition of traces of surfactants to water. This was reported by the Japanese and also more recently in desalination equipment [23, 27, 28]. Preliminary experiments in our laboratory show that surfactants increase nucleation, apparently from secondary nucleation.

There have been numerous studies of nucleate boiling on horizontal or vertical tubes or on vertical flat surfaces [28, 29, 30]. These studies have generally indicated better performance than nucleate boiling on submerged surfaces and the improvement has generally been attributed to the flow in the film. Not one of these studies has made any recognition that similar improvements occur when nucleate boiling occurs in thin liquid films on horizontal surfaces where there is no imposed flow.

Most texts on heat transfer describe flow boiling as being different from nucleate boiling on a submerged surface. In flow boiling, liquid is progressively converted to vapor so that a number of flow regimes exist along the flow. The predominant flow regime is annular flow with a liquid film on the wall. Early work hypothesized that in the annular flow region heat transfer was so good that it must be forced convection and that, consequently, nucleate boiling was suppressed [31].

The support for this hypothesis has recently been reanalyzed and found not to support the hypothesis [32]. Annular flow would seem to offer an ideal situation for secondary nucleation to augment nucleate boiling heat transfer.

In vigorous boiling on a submerged surface the view of the bubble activity is obscured. Extrapolating results on bubble nucleation at lower heat fluxes suggests that at higher heat fluxes more surface sites come into play. An alternate hypothesis is that at high heat fluxes secondary nucleation supplies the additional nuclei. The circumstances would seem to permit secondary nucleation in the same way as has been suggested for nucleate boiling in thin films. At high heat fluxes vapor does push the liquid away leaving the surface wet with a film of liquid. It would seem possible for drops to form from bubbles bursting from this film or perhaps from all the commotion above the surface. Drops striking the film would be well positioned to cause secondary nucleation. Current literature mentions only the former hypothesis, so the latter has not been ruled out. Who can say which hypothesis is more nearly correct? More research can help decide.

Secondary nucleation can also offer alternate hypotheses for two other characteristics of nucleate boiling. When heat flux is plotted against surface temperature excess over saturation, one obtains what is often called the boiling curve. The upper portion of the curve often moves in a parallel fashion to higher or lower temperature differences because of aging or other obscure reasons. It is usually said that changes in the nucleation characteristics of the surface are responsible for the shift. An alternate hypothesis is that changes in nucleation on the surface effects only the lower portion of the curve and that secondary nucleation ensues to move the rest of the curve.

The other characteristic is hysteresis in which surface temperatures are higher as the heat flux is increased but lower as it is decreased. On increasing heat flux it sometimes occurs that after bubbles begin to coalesce the surface temperature suddenly falls. The usual explanation for this is that new nucleation sites have acquired vapor and became active. The alternate hypothesis is that when bubbles begin to merge a new flow regime is established which favors secondary nucleation.

CONCLUSIONS

Secondary nucleation definitely occurs in

nucleate boiling. Indications are that drop-formed vortex rings are responsible for entraining tiny bubbles to serve as nuclei for secondary nucleation. How important secondary nucleation is in submerged boiling remains to be shown, but it seems likely to be responsible for the improved performance of nucleate boiling in thin liquid films with and without surfactants. Research opportunities abound in evaluating these new developments. □

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