F. B. Peterson*

The authors have presented data related to the subject of fluid nucleation, which could be relevant to a wide variety of engineering applications. In this regard, several questions concerning the manner in which the data was obtained and the conclusions derived from the data may be pertinent. First, in what way does the water sampling procedure influence the fluid history as it passes through the micro- orifice of the Coulter counter? In addition, what was the electrical conductivity of the water monitored and controlled to maintain a calibration of the Coulter counter?

In numerous experiments by others (e.g., [35-44]), it has been shown experimentally that stable hydrophobic particles can exist in water and that de novo particle production can occur with relative ease. Since the authors refer to their data as indicative of tap water "microbubble" spectra, it would be instructive to know how they determined that no particulate indicative of tap water "microbubble" spectra, it would be instructive to know how they determined that no particulate was present during the measurements.

The paper mentions that previous published work by the third author has not demonstrated a "most probable bubble size" when bubble sizes were directly measured in a water tunnel by a light scattering technique. In the other hand, the results presented in this paper do show a distinct peak in the "microbubble" spectra. Although the paper does not discuss this point further, it would be interesting to have the authors' comments on this difference in spectral characteristics. Could the reader reasonably conclude that the difference may be attributed to the fact that in a water tunnel the liquid is moving while in the present work the fluid was relatively stationary? As an alternative, could the difference be attributed to instrumentation and sampling techniques?

Since the authors' theoretical predictions of "suppression of Nucleation versus Water Temperature" give trends opposite to that observed in their experiments, the authors conclude that the unaccounted for "rectified diffusion" may have been of central importance. Since ultrasonics were used to generate the cavitation, it is possible that microstreaming due to the acoustic field may in fact have been of even greater importance than just "rectified diffusion" (see e.g., [45]). On the basis of the results presented in the paper, one real possibility is that the use of ultrasonic techniques to measure the superheat of water will give very misleading results. This may be particularly true if these same results were used to infer the nucleation susceptibility of water in a flowing system.

Additional References


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Numbers in brackets designate Additional References at end of discussion.
in my opinion, more relevant to divide superheat by same "characteristic temperature," taking into account mass and heat transfer between the two phases, such as \( \frac{\rho L}{D_p} \). The "dimensionless superheat" becomes in this case a Jakob number \( J \).

In the same way, the "suppression pressure" could usefully be related to the "characteristic pressure" \( \frac{\rho L \, dp}{C \, dP} \) giving so a \( B^* \) factor. It is quite analogous to \( B^* \) factor of Stepanoff.

D. Sette and F. Wanderlingh

The paper of Pyun, Hammit, and Keller [46] (hereafter referred to as PHK), concerning the distribution of microbubbles in liquids and their role on the onset of cavitation, contains very useful informations. In particular the direct experimental measurement of the population of microbubbles, both in normal tap water and in neutron irradiated water, allows a detailed discussion of the nucleation mechanism.

Generally speaking, the PHK's results, are in agreement with those already available, specially with the results obtained by our group. Before discussing in some details the questions involved in the nucleation process we wish to call the attention to some points which are relevant in their and our experiments (see, for comparison, references [2 and 4], and references quoted therein).

1. The threshold of cavitation is statistically related to the nuclei distribution.
2. Cavitation nuclei exist, as a rule, in normal tap water (microbubble persistence paradox).
3. The sizes of microbubbles created by neutron irradiation fall roughly in the same range of the pre-existing microbubbles.
4. The increase in the microbubbles concentration due to the neutron irradiation, although detectable, is always very small.

Finally, the PHK work has shown as a new result that the size distribution of microbubbles exhibits a maximum that shift, as temperature increases, toward larger sizes.

Some years ago we have tried to clarify the nucleation mechanism by studying from a thermodynamical point of view [48], the effect of high energy particles irradiation. The main points of our theory are as follows:

(i) While nucleation of microbubbles in the micron range, in a homogeneous water sample requires an energy of the order of hundreds MeV, in gassed water such an energy is dramatically lowered, and can even become negative, leading to an exoenergetic process.
(ii) There is a temperature-size threshold, that has to be reached in order to obtain microbubbles of theoretically unlimited size. (see Fig. 7 of reference [48]).
(iii) Such a threshold can be attained if a particle having an energy of, at least, 4 MeV, loses such an energy in a sufficiently short path length (order of ten microns). Such a particle can be an atomic nucleus struck by an energetic neutron. Its action is to bring a small volume of the liquid at a temperature suitable

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6. Numbers in brackets designate Additional References at end of discussion.
for the onset of a thermodynamic process of growth of microbubbles.

(iv) Once created, microbubbles of radius larger, say, than 1 micron, can statistically exist in the liquid, at least for a time interval during which some stabilizing process can take place (e.g., adhesion to dust particles).

In our opinion such a theory can satisfactorily explain the results of PHK experiments.

The circumstance that microbubbles created by neutron irradiation and pre-existing microbubbles have sizes that fall in the same range, and, moreover, the temperature dependence of the sizes that seems to be the same for the two kinds of microbubbles, strongly suggest a common origin. This is in agreement with our hypothesis, that also in tap water the existence of microbubbles can be largely ascribed to the action of high energy particles (e.g., cosmic rays).

One can easily show that neither hydrogen nor oxygen nuclei are in condition to fulfill the (iii) statement, if neutron sources like Ra-Be or Po-Be are used. Neutrons emitted by such sources have an energy spectrum that extend up to about 10 MeV, with a mean energy of the order of 4 ± 5 MeV. Due to its mass, an oxygen recoil nucleus can acquire, as an upper limit, an energy of 3 MeV. Hydrogen nuclei can acquire up to 10 MeV, but their specific energy loss is too low, so that the required energy cannot be dissipated in a region sufficiently small to make possible the nucleation process.

Atomic nuclei that can act as nucleating agents (in connection with 10 MeV neutrons) are those whose mass ranges from 6 to 12 [50].

A confirmation of such a circumstance has been given by detecting the microbubbles present in normal and irradiated (with 10 MeV neutrons) water, through a resonant acoustic absorption measurement [49]. Neutron irradiation can act only in connection with some impurity that furnish atomic nuclei of suitable mass, such as, e.g., CO₂ or N₂ dissolved in water. In such a case the rate of microbubble creation and their concentration will be determined by the impurity concentration rather than by the neutron flux. If the impurity content is artificially raised, as in reference [4], then a neutron source of very low intensity (10⁻⁴ Curie) is in condition to give a sizable effect.

An interesting point arising from PHK's measurements is the existence of a maximum in the microbubbles distribution. It seems that, at each temperature, there is a most probable size, around which the microbubbles distribute. Such a circumstance can be shown in reference [4].

The most probable size shifts as the temperature increases in a quite regular way toward larger sizes. In addition experimental results concerning both water and sodium become similar if thermodynamic reduced variables are used.

This behavior strongly supports our thermodynamical point of view. As anticipated before, as a result of our theoretical approach, there exist a threshold both in the size of microbubbles and in the local value of temperature (crossing of full line and dash-dot line in Fig. 7 of reference [48]). Above such a threshold further growth of a microbubble becomes an exoenergetic process, so that theoretically unlimited sizes could be reached. Actually, however, the growth is limited both by the local losses of energy, due to diffusion processes, and by the depletion of gas in the surrounding liquid.

Our calculations are concerned with a liquid (water) at ambient temperature (300°K), in which the amount of dissolved gases corresponds to the saturation value at such a temperature. As a consequence, at temperatures higher than 300°K (locally reached) the transition of a mixture of gas and water molecules from the solution status to the gaseous phase inside the growing microbubble, is thermodynamically allowed, because of the decrease in the Gibbs function. Moreover, the "evaporation" of gases is itself an exoenergetic process, and the corresponding decrease in the enthalpy can overcompensate the energy required both for the surface growing and the water evaporation.

Now, if the mean temperature of the liquid is raised, two circumstances take place: first, the same energy lost by a given recoil nucleus, brings a larger volume to a temperature sufficiently high for the starting of the process of growth. As a consequence the cooling of such a region, that takes place in the initial phase of the process, is decreased, so that a larger size of the microbubble can be attained (as in Fig. 7 of reference [3], dashed lines will be shifted to the right-hand side). Second, by increasing the mean temperature, the liquid tends to become oversaturated (if, as in the case of the PHK's experiments, the total gas content is left constant). The dash-dot line of Fig. 7 of reference [3] is then also shifted to the right hand side, and the threshold size increases.

The final result will be an increase in the size of the created microbubble.

It is to be noted that the proposed mechanism also explains the fact that the sizes of microbubbles are grouped around a most probable value. In our theory, in fact, the relevant condition is the loss of a suitable amount of energy in a sufficiently small volume. Also if an ionizing particle possesses a very high energy, the useful part of such an energy is that lost in the part of its path corresponding to the maximum of the specific energy loss (at least as far as atomic nuclei like C or N are concerned). As a consequence, there is a lower limit for the energy of the nucleating particle (of given mass), while the actual dimension of the created microbubble is essentially determined by the thermodynamic conditions of the sample.

As a final remark we want to consider the sizes of microbubbles. From the theoretical side, it has seemed reasonable to assume that the actual size should not be much larger than the threshold value, that is 1 ± 2 microns. Experimental results of resonant acoustic absorption measurements [49] indicate a size of about 2 microns for bubbles created by carbon recoil nuclei. The PHK's measurements, at ambient temperature, show the creation of microbubbles of about 3.5 microns.

It is to be noted that in our thermodynamic theory the numerical calculations are carried out by using some approximations, as, for example, the perfect gas law and the evaluation of the enthalphy of mixing. Also in the acoustic absorption measurements we find the relation between acoustic frequency and bubbles size by means of an approximate calculation.

In addition the time lag between neutron irradiation and measurements, as well as the duration of the irradiation, can be relevant parameters, in connection with the stabilizing mechanism.

Taking into account all these circumstances, we believe that the results obtained by PHK and by us on the sizes of bubbles agree fairly well.

Additional References


Author's Closure

The authors would like to express their deep appreciation for...
the comments and the interests by J. Bonnin, M. Plesset, F. Peterson, D. Sette, D. Taylor, and F. Wanderlingh.

The authors agree with all of the facts (particularly thermodynamic nucleation theory by Sette's group) pointed out by Professor J. Bonnin, D. Sette, and F. Wanderlingh.

Professor Plesset raises a question about the statistical significance of data associated with neutron irradiation. It is true that the statistical deviations are appreciable partly due to the statistical nature of the water sampling; however, the increases in the number of counts due to neutron irradiation especially for the smaller bubble size are significantly above this deviation (i.e., average 150 percent increase versus a maximum mean deviation of 17 percent).

The comments by Drs. Peterson and Taylor on test technique are an outgrowth of their long and continuing interest in, and work with this area. First, the authors calibrated the Coulter counter at room temperature (70°F). There is some change in the electrical conductivity of water at the higher temperature (160°F), but we find that the effect of temperature on the conductivity is negligible. Second, authors agree that some negligible particulate may exist during the measurements. To the author's knowledge there is not a measuring technique available to discriminate the particular matter and the bubble. Therefore, the system was pressurized up to 60 psig to find the effect of pressurization on the microbubble spectra. Since an appreciable change in the bubble size was found, we assumed that no particulate exists during the measurements. Third, the third author measured the microbubble spectra only at the room temperature under various conditions (e.g., filtered water, unfiltered water, etc.). Due to the limited resolution of his experimental equipment, he could not measure a microbubble with its radius less than 2–3 microns (refer to the microbubble spectrum without neutron irradiation in Fig. 4). Then he assumed that beyond this experimental limit the number of nuclei would increase rather than decrease. The first author assumed that it would decrease. Fourth, authors agree that this ultrasonic technique may not be the best method compared with other techniques available [12, 13, 25, e.g.]. But this method is simple and furthermore it provides a direct insight into the nucleation phenomena associated with cavitation-erosion.