Observation of second metastable state in superheated emulsion detector

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ABSTRACT

The superheated emulsion detector (SED), a well-known detector for neutrons, ions, electrons and photons, contains droplets of superheated liquid held in a viscoelastic gel. It is well known that these droplets are in a metastable state, decaying both spontaneously and when induced by energetic radiation. Here, it is shown that the SED comprises droplets of more than one metastable states nucleating to vapor bubbles at different frequencies. To the best knowledge of the authors the existence of two metastable states of the active liquid in SED has not been reported before.

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1. Introduction

A liquid is said to be in a superheated state when it maintains its liquid state at a temperature above its boiling point at a given pressure or at a pressure below its vapor pressure at a given temperature. This metastable state of the liquid may become unstable in presence of sharp edges or other heterogeneous nucleation sites, such as dust, air pockets, etc., at a temperature above its boiling point, leading to the phase transition to the next stable vapor state. However, one may maintain the liquid in steady superheated state by various techniques. One of them is the emulsification process [1], in which minute droplets of superheated liquid are dispersed homogenously in the absence of dust or air pockets in a viscoelastic gel as in the superheated emulsion detector (SED) or in a soft polymer matrix [2,3] as in a bubble detector (BD). This produces smooth interfaces and reduces the possibility of heterogeneous nucleation. SED and BD are a collection of minute drops of superheated liquid, each of which represents a microscopic bubble chamber [4]. These detectors have their place in almost all branches of radiation physics including space physics [3], high-energy physics [5,6], medical physics [7] and health physics [8], and have been extensively used in radiation detection [1,9–13], dosimetry [8,14–21] and spectrometry [11,22–25].

In superheated liquids there exist a large number of microbubbles that rapidly grow and collapse back due to surface tension. For the microbubbles to grow indefinitely, they need to reach a *critical radius* r_c [1]. The energy required to form such a microbubble of radius r_c is the threshold energy (W) for nucleation.

The nucleation of a superheated droplet, that is the sudden evaporation of a droplet to a vapor bubble, is accompanied with a sudden change in volume and pressure with the generation of an audible acoustic pulse. Droplet nucleation may be detected by using active detection method [12,26,27], based on the detection of acoustic pulse. In the present work the droplet nucleation is detected using active detection method.

When superheated droplets are exposed to energetic radiation, energy is deposited inside the liquid in the vicinity of microbubbles. The radiation comprises either energetic ions or energetic ions/electrons produced by secondary processes. These energetic charged particles pass through the superheated liquid depositing energy along their path in a very short duration in the form of heat spikes [17,28]. When this energy is deposited in the neighborhood of a microbubble, it may enable the microbubble to overcome the threshold energy barrier. When the deposited energy inside a microbubble exceeds the threshold energy (W) for nucleation of the liquid, it may cause nucleation and the process is known as *radiation-induced* or simply *induced* nucleation.

When exposed to energetic radiation, superheated droplets nucleate and vaporize to form vapor bubbles in a random, uncorrelated manner. Here the surviving number of droplets is a

Fig. 1. Counts observed by irradiating R12 with neutrons.

Fig. 2. Schematic diagram of the experimental setup, where (a) external diameter of the cylindrical neutron source is 4.0 cm (external length is 10.5 cm), (b) diameter of the SED vial is 2.5 cm, (c) height of the gel matrix is 2.5 cm, (d) distance between the axis of the neutron source and the axis of the vial were 16.0, 11.2 and 12.7 cm, respectively. The axis of the neutron source and that of the SED vial were perpendicular, with the SED vial placed corresponding to roughly the middle (along its length) of the neutron source.

Fig. 3. Counts observed by irradiating R12 with neutrons at 27 °C. The three successive irradiation intervals, at constant neutron fluence rate of 3.8×10^7 cm^{-2} s^{-1}, are separated by 70 and 60 min regions of no radiation.
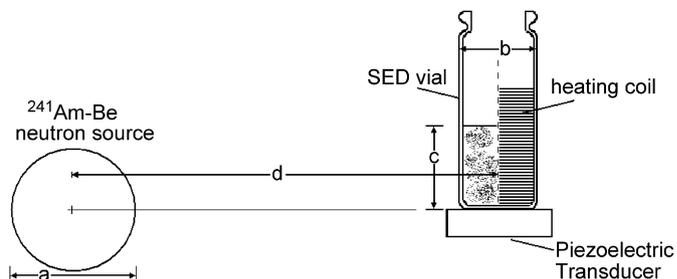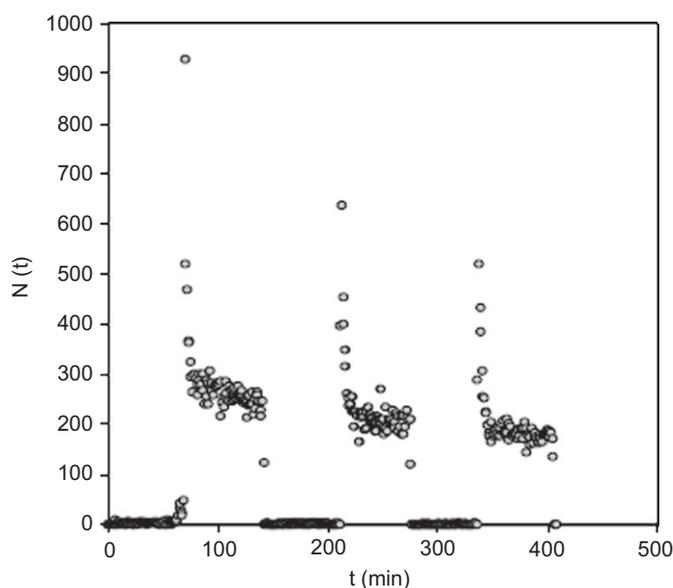
monotonically decreasing function of time. This decay in the number of superheated droplets is exponential in nature [12,29].

An experimental data for SED made of refrigerant R12 irradiated with neutrons from a ^{241}Am–Be neutron source is shown in Fig. 1. Here the data show a sharp curvature at the beginning of the irradiation. The count rate is observed to be much higher for initial few minutes compared to the rest of the data. The nature of the curve suggests the existence of two metastable states with different lifetimes, one with relatively short lifetime compared to the other. A possible explanation for this would be due to the nucleation of droplets of shorter lifetime (τ_2), after which only the long-lived (τ_1) droplets remain and decay. The decay of two states is shown by the dark lines, which implies that $\tau_2 \ll \tau_1$. For practical application of the detector the droplets should be long-lived. If the detector is irradiated once (over a period of time much longer than τ_2) and reused later, then the response should only be due to the long-lived droplets (since short-lived droplets have already been depleted during previous irradiation). We have exhibited a feature in contradiction to what was expected. This behavior is observed to be independent of the nature of the active liquid and irradiating radiations, implying that it is a physical property of the active liquid used in SED. We put forward in this paper its interpretation and explanation.

2. Experiment

The experiments were performed by irradiating R12 (CCl$_2$F$_2$, b.p. −29.8 °C) detector fabricated in our laboratory. A schematic diagram of the experimental setup is shown in Fig. 2. The detector was irradiated with energetic neutrons from a 3 Ci ^{241}Am–Be neutron source (manufactured by Bhabha Atomic Research Centre, India, cylindrical in shape, absolute source strength is 1.1×10^{11} n/s, average neutron energy is near about 4.3 MeV) at a known neutron flux. We have used active detection method [12] for the detection of nucleation by energetic neutrons. The acoustic signal from the exploding droplets are detected by a piezoelectric transducer and converted into an electrical signal. This electrical signal is then amplified, differentiated, discriminated, converted to a TTL pulse and finally acquired in an MCA operating in multi-channel scale (MCS) mode with a dwell time of 20 s.

The experiments were performed at a constant temperature and at a constant neutron flux. Initially the radiation was on and counts observed as a function of time were collected using MCA. The radiation was then turned off for a period of time (by removing the source) and then turned back on again with the same neutron flux. Such switching on and off the irradiation was repeated for two to four times where the "radiation off" period was varied. The response of the detector is shown in Fig. 3. Here we present the data for temperature 27 °C with a constant neutron fluence rate of 3.8×10^7 cm^{-2} s^{-1}. The data is shown in Fig. 3, where each of three irradiation regions shows initial high counts, as if the short-lived droplets have populated again.

To observe the behavior of counts received $N(t)$ and the initial spike-like feature at the beginning of the successive "radiation on" regions, several more experiments were done by varying the "radiation off" period. One such data, where the radiation was turned off four times, is shown in Fig. 4. Here the irradiation was done at a constant neutron fluence rate of 3.0×10^7 cm^{-2} s^{-1}. The durations when the radiation was on were 44 min each, while the durations when the radiation was off were 2, 45, 8 and 25 min, respectively.
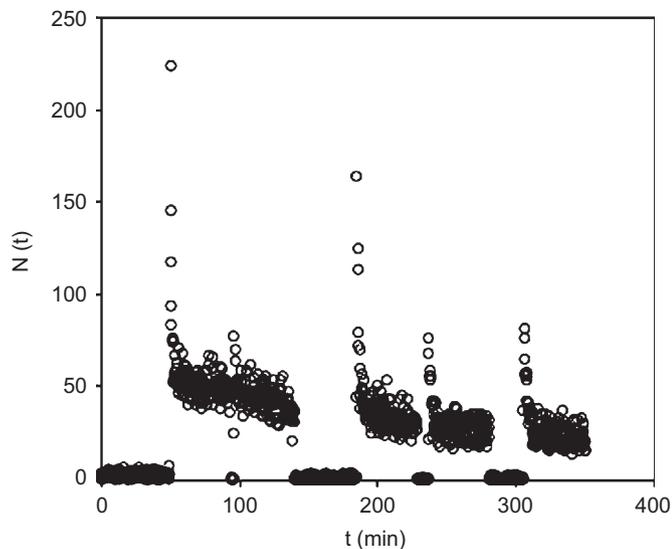
Fig. 4. Counts observed by irradiating R12 with neutrons at 30.5 °C. The five successive irradiation intervals, at constant neutron fluence rate of 3.0×10^7 $cm^{-2} s^{-1}$ are separated by 2, 45, 8 and 25 min regions of no radiation.

3. Observation

The experimental data as seen in Figs. 3 and 4 show three and five distinct regions where the radiation was successively on at constant flux. The general feature in all these regions of high counts is that they have an initial spike-like feature spanning about 5 min. Each of the regions where the radiation was on has an initial sharp curvature following which the decay in the count rate is gradual. There are some, but very few counts due to spontaneous nucleation in the radiation off regions.

One feature which stands out is the spike-like feature, where the count rate initially jumps to a value (say $N' = N(t+\tau)$), which is higher than the count rate at the end of the previous radiation on region (say $N(t)$), i.e. $N' > N$.

One also observes that the ratio $(N'-N)/N$ is not only positive but also increases with the duration (τ) when the radiation was turned off. A plot of this ratio as a function of off-duration τ is shown in Fig. 5.

In the case of polydisperse sample, having a distribution in droplet volume, the expression for surviving number of droplets at time t will follow a multi-exponential decay [12,30] as given by

$$S(t) = \sum S_j(t) = \sum S_{0j} e^{-b_j t} \tag{1}$$

where S_{j0} is the initial number of drops of size v_j and of life time b_j. If $f(v_j) = S_{j0}/S_0$ is the *initial* distribution [12,30] in the droplet volumes, then Eq. (1) can be written as

$$S(t) = S_0 \sum f(v_j) e^{-b_j t} \tag{2}$$

where $S_0 = \sum S_{j0}$ is the total number of droplets initially present in the detector.

$N(t)$, the observed counts or the number of drops nucleated in successive intervals of time, is given as

$$N(t) = S(t - \Delta t) - S(t) \tag{3}$$

Hence one gets

$$N(t) = S_0 \sum f(v_j)[e^{b_j \Delta t} - 1] e^{-b_j t} \tag{4}$$

From the expression above, and considering the non-negative nature of b_j and $f(v_j)$, one can show that $N(t)$ *never increases with time*, when the flux is held constant. Hence for a given flux one can write $N(t') \leqslant N(t)$, for $t' > t$. This, however, excludes the departures
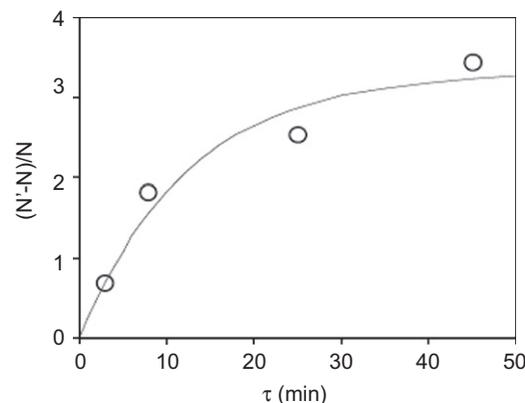
Fig. 5. The observed ratio $(N'-N)/N$ for R12 irradiated with neutrons at 30.5 °C. The solid line is the fit $(N'-N)/N = 3.3 \times (1 - e^{-\tau/12.5})$ to the data.

from monotonic decay due to counting fluctuations, the magnitude of which would be of the order of $\sqrt{N}$.

Thus it is clear from the above expressions that when an SED is exposed to a radiation field the number of observed counts, barring counting fluctuations, should decrease continuously. This should hold true irrespective of the magnitude of b_j. Thus we see that the observed feature that $N(t') \geqslant N(t)$ requires a special kind of explanation.

4. Discussion

To explain this contradiction between theoretical expectation and experimental observation we have investigated different possibilities. A probable explanation of this high rise or the additional counts is due to the inaccuracies in the placement of the source and is discussed here. If it is due to the placement of the source, then such an increase could only affect the counts during which the source was moving, which is small (a few seconds) compared to the dwell time per channel. Also, this might cause high counts at both the beginnings of the radiation on and off zones. However, such an increase in counts persisted for several channels at the beginning of radiation on zones and never occurred at the beginning of "radiation off" zones. The initial high counts were observed for all SED experiments, using neutrons, gamma rays and electrons. Hence such a simple explanation was discarded.

The facts that the initial channels after the radiation is turned on have a very high counts followed by a strong curvature in the count rate as a function of time indicate widely different lifetimes. The fact that when the radiation is turned off for a duration τ and turned back on (at the same flux, temperature and pressure) shows counts higher than before the radiation was turned off, indicating that there has to be more than one species of droplets of different lifetimes. The fact that the initial rise after each radiation off region increases to saturation (Fig. 5) indicates that these states are approaching equilibrium amongst themselves, thereby further reaffirming the existence of two metastable states. This is observed not only for R12 but also for several other liquids, indicating that this is an intrinsic characteristic of the active liquids used in SED. The fact that this behavior is observed when exposed to gamma rays or neutrons supports that this phenomenon is independent of the nature of irradiating radiation. We will, for simplicity, assume that there are two metastable states, the normal, which has the longer lifetime and the second metastable states having the smaller lifetime.

Other experiments show that at higher temperatures the transition from the normal to the second metastable state

becomes vanishingly small. This in effect isolates the two states and they decay independently, thereby depleting the second metastable state first, following which, the decay is simply that of the normal metastable state.

The nature of the second metastable state is still unknown to us. More work is under way to understand the nature of the second metastable state and the interstate transition kinetics.

5. Conclusion

It can, therefore, be concluded that the SED contains droplets of superheated refrigerants in two metastable states, one with a much shorter lifetime compared to the other. Though the exact nature of these states is still speculative, evidence of interstate transition is observed, indicating a continuous dynamical switching of states and a possible dynamic equilibrium between the states. The SED is a versatile neutron and ionizing particle detector and dosimeter. The existence of the two metastable states, in principle, suggests corrections to the quantities (such as dose, etc.) measured conventionally using SED.

Acknowledgments

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