SUPERHEATED DROP AS A NEUTRON SPECTROMETER

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Abstract

Superheated drops are known to vaporise when exposed to energetic nuclear radiation since the discovery of bubble chamber. The application of superheated drops in neutron research specially in neutron dosimetry is a subject of intense research for quite sometime. As the degree of superheat increases in a given liquid, less and less energetic neutrons are required to cause nucleation. This property of superheated liquid makes it possible to use it as a neutron spectrometer. Neutron detection efficiency of superheated drops made of R12 exposed to Am-Be neutron source have been measured over a wide range of temperature -17°C to 60°C and the results have been utilized to construct the energy spectrum of the neutron source. This paper demonstrates that a suitable neutron spectrometer may be constructed by using a single liquid and varying the temperature of the liquid suitably at a closer grid.

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1. INTRODUCTION

A fluid kept in the liquid state above its boiling temperature is called superheated. Application of superheated liquid to detect ionizing radiation is well known from the times of bubble chamber[1]. The resurgence of its use has been observed from the late seventies[2,3] and the investigations on the subjects in the last two decades turned into an almost maturing technology especially to detect neutrons and more recently to detect photons. The suitability of using superheated drops as a neutron dosimeter[4,5,6,7,8] has already been established. The superheated drops are now commercially available as neutron and photon dosimeter in the trade name superheated drop detector (SDD) and bubble detector (BD). In a superheated liquid minimum energy (threshold) required to nucleate decreases as the degree of superheat increases. The degree of superheat of a liquid could be defined simply by the difference of ambient temperature above the boiling point of the liquid. Therefore, liquids with lower boiling points possess higher degree of superheats at a given ambient temperature (above their boiling points) and as the ambient temperature increases the given liquid becomes more and more superheated. This property of the superheated liquid are being utilised to develop neutron dosimetry and neutron spectrometry[2,3,9,10]. There are two distinct types of methodologies used in developing neutron spectrometer. In one, a collection of superheated samples made of liquids with different boiling points (i.e. with different threshold neutron energies) are utilised[11], while in the other, two liquids are chosen and the temperature of the liquids are varied at four different temperatures to obtain eight sets of threshold energies[12,13,14] (equivalent to eight different samples with different boiling points). The temperature variation method is superior than using samples with different boiling points. By controlling the temperature of the sample one can, in principle, change the threshold neutron energy at any desired level (equivalent to using 'finer' windows to scan the spectrum), while in the other method one is limited by the availability of liquids with lower boiling points (equivalent to using 'coarser' windows). In the present work we measured the detection efficiency of a single sample made of R-12(Dichlorodifluoromethane : $CCl_2F_2$), which is known to be sensitive to neutrons of energies from thermal to tens of MeVs, by (almost) continuously changing temperatures over a wide range. The response of the sample to Am-Be neutrons have been measured at about thirty different temperatures in the range which is equivalent to using thirty different samples with thirty different boiling points (the experiment has been actually performed more than 50 different temperatures in the range -17°C to about 60°C but it has been observed that the sample started responding from about 0.5°C to Am-Be neutrons). In addition to the advantage of using single liquid, con-

\(^2\)Superheated Drop Detector is the registered trademark of Apfel Enterprises Inc, NewHaven, CT, USA

\(^3\)Bubble Detector is the registered trademark of Bubble Technology Industries Ltd.
trolling the temperature enables one to scan the energy spectrum by finer 'windows' thereby improving the inherent energy resolution of the spectrometer when compared with other such spectrometers using superheated liquid.

2. PRINCIPLE OF OPERATION

The superheated state of the liquid is a metastable state and the nucleation in this state can be initiated by the presence of heterogeneous nucleation sites such as air bubbles, solid impurites, gas pockets etc. or by radiation interactions. The nucleation in superheated state starts with the formation of a critical sized vapour embroy. The free energy required to form a spherical vapour bubble of radius \( r \) in a liquid is given by

\[
G = 4\pi r^2 \gamma(T) - \frac{4}{3} \pi r^3 (p_v - p_o)
\]  

where \( \gamma(T) \) is the liquid-vapour interfacial tension, \( p_v \) is vapour pressure of the superheated liquid and \( p_o \) is the ambient pressure. The difference \( p_v - p_o \) is called the degree of superheat of a given liquid. One can see from equation (1) that \( G \) is maximum at

\[
r = 2\gamma(T)/(p_v - p_o) = r_c
\]  

where \( r_c \) is called the critical radius. When a bubble grows to the size of the critical radius it becomes thermodynamically unstable and grows very fast till the entire liquid droplet vaporises.

The minimum amount of energy (\( W \)) needed to form a vapour bubble of critical size \( r_c \) as given by Gibbs[15] from reversible thermodynamics is

\[
W = 16\pi \gamma^3(T)/3(p_v - p_o)^2
\]  

where \( \gamma(T) = C (T_c - T - d) \) with \( T_c \) the critical temperature of the liquid and \( C \) and \( d \) are constants [16].

With increase in temperature, since the degree of superheat \( (p_v - p_o) \) increases and \( \gamma(T) \) decreases, the minimum energy \( (W) \) required for vapour bubble nucleation will be less. The variation of \( W \) with temperature for superheated drops of R-12 is shown in figure 1. Therefore \( W \), the threshold energy for nucleation depends on the type and the temperature of the liquid. When a neutron of energy \( E_n \) interacts with a nucleus of atomic weight \( A \), the maximum energy that can be transferred to the nucleus from the neutron is through the elastic head on collision and is given by,

\[
E_i = 4AE_n/(A + 1)^2
\]
After receiving the energy, the nucleus is scattered from its atom and moves through the liquid losing its energy through Coulombic interaction until it comes to rest. For a given neutron energy, different nuclei of the liquid will receive different amount of energy, depending on their atomic weight. The ion with the highest value of linear energy transfer (LET) or \((dE/dx)\) in the liquid, will play the major role in vapour nucleation[17]. The energy deposited along that part of the ion’s path (L) corresponding to about twice the critical radius contributes significantly to bubble formation[17,18]. For nucleation to occur this deposited must exceed \(W\) the minimum energy required for bubble formation.

Usually most of the energy is lost into heat and a very small fraction of the deposited energy is utilised in nucleation and \(W/E_c\) is called the thermodynamic efficiency (\(\eta_T\) of nucleation [17].

\[
W = kr_c dE/dx = 2\eta_T r_c dE/dx
\]

where \(k\) (constant) equals twice the thermodynamic efficiency (\(\eta\)). Hence, in the equation

\[
W/r_c = kdE/dx
\]

relates the threshold energy (corresponding to the \(dE/dx\)) for nucleation to the ambient temperature (corresponding to \(W/r_c\)). This enables us to convert the temperature scale of superheated drops to the (threshold) energy scale of incident neutrons. Therefore by varying the ambient temperature of the superheated drops, one can observe the variation of the SDD response at different neutron energies which has been used in neutron spectrometry.

3. EXPERIMENT

The experiment was performed with superheated drops of R12 at different temperatures by using the volumetric method, described by Das et al.[19]. The vial containing the sample was connected to a graduated horizontal glass tube with a small coloured water column as marker. The vapourization of a liquid drop displaces the water column by the distance corresponding to the volume of vapour formed. The details of the preparation of the sample is given elsewhere[20].

If neutrons of flux \(\psi\) are incident on superheated drops of volume \(V\), liquid of density\(\rho_L\) and molecular weight \(M\) the vaporization rate is given by

\[
\frac{dV}{dt} = V\psi \frac{N_A \rho_L}{M} \eta d \sum n_i \sigma_i
\]
where \( N_A \) = Avogadro Number

\[ d = \text{average droplet volume} \]

\[ n_i = \text{weight factor of the } i\text{th element in the molecule whose neutron nucleus elastic scattering cross section is } \sigma_i \]

\[ \eta = \text{efficiency of neutron detection}. \]

Due to nucleation by neutrons, the displacement of the water column along a horizontal glass tube was measured as a function of time. The procedure of calculating \( \eta \) from the measured displacement of water column has been explained in detail in one of our recent publications[19].

The temperature of the sample was controlled by an indigenously made temperature controller. For low temperature measurements the sample was placed in an alcohol bath sitting on the top of a cold finger dipped in liquid nitrogen. The upper part of the finger was wrapped with heating tape and by applying different voltages to the tape, different steady temperatures of the bath can be achieved. For measurements of higher temperatures the same setup was used without the liquid nitrogen. The fluctuation of temperature in these measurements was found to be within \( \pm 0.1^\circ C \).

In the experiment the ambient temperature of the sample was increased slowly from low to any desired higher value and the nucleation rate was measured at each temperature. The measurement was performed from in a temperature range of -17\(^\circ C\) to about 60\(^\circ C\) in a close grid necessary to obtain the energy spectrum of the source from temperature. The nucleation due to background radiation and due to other fluctuations has been subtracted. The liquid was observed to become unstable due to spontaneous nucleation at about 60\(^\circ C\).

4. EXPERIMENTAL RESULTS AND COMPUTATION OF THE NEUTRON ENERGY

The variation of neutron detection efficiency (\( \eta \)) for R12 with temperature in presence of neutrons from Am-Be neutron source is presented in figure 2. The solid line in figure 2 is the spline smoothing of the efficiency data at different temperatures. The uncertainties presented in the figure are the total experimental uncertainties of estimating \( \eta \). The derivative of efficiency, \( d\eta/dT \) against temperature is shown in figure 3. Now one has to estimate the equivalence of the energy of the detected neutrons with the temperature of the SDD sample. One way to do it is to expose the sample at different temperatures to different monoenergetic neutron sources and to note the threshold neutron energies for nucleation [12,14]. A novel approach has been used in this work.
As has been presented in Section 2, the different nuclei of the superheated liquid would receive different amount of energy and they must have different $dE/dx$. In case of R-12 containing C, Cl and F, $dE/dx$ of these ions with different neutron energy are presented in Figure 4. From Figure 4, it is clear that the $dE/dx$ values of C, Cl and F are comparable in the neutron energy of our interest and we take the average value of $dE/dx$ of all the ions using the equation below

$$(dE/dx)_{\text{average}} = \frac{\sum n_i \sigma_i (dE/dx)_i}{\sum n_i \sigma_i}$$

where $n_i$ is the number of ions of the i-th element in a R12 molecule, $\sigma_i$ is the neutron-nucleus elastic scattering cross section and $(dE/dx)_i$ is the LET of the i-th ion. The variation of average $dE/dx$ as a function of neutron energy is shown in figure 5.

From equations (3) and (5), we obtain

$$(dE/dx) = \frac{8\pi \gamma^2 (T)}{3k} (p_v - p_o)$$

From the equation above the $dE/dx$ has been plotted against temperature for different arbitrary values of $k$ of which only four such plots ($k=1, 0.1, 0.05, 0.0195$) are presented in figure 6 using the equivalence between the temperature of the sample and the incident neutron energy from figure 5. The variation of threshold neutron energy for nucleation with temperature of the sample for different $k$ has been studied of which four such variations are shown in figure 7. With the optimum value of $k$ the temperature axis of the figure 3 has been converted to neutron energy and the resulting spectrum was fitted with the peak neutron energy of the $^{241}$Am-Be neutron spectrum. The best fit is obtained for $k$ equals 0.0195. The analysis has been performed upto a maximum temperature of 42.5°C. The final neutron energy spectrum of $^{241}$Am-Be source obtained from our analysis is shown in figure 8.

If $L = 2r_c$ is taken as the distance in the ion’s path which contributes significantly in nucleation of superheated drops [17,18] in calculating the thermodynamic efficiency of nucleation, our experimental analysis produces the value close to 0.01. It may be noted here that Apfel et al.[17] obtained this value ranging from 0.03 to 0.05.
5. DISCUSSION

The result shows that $\eta$ increases with temperature. At low temperature, the threshold energy for nucleation ($W$) is high, which indicates that a larger energy is required to cause nucleation. According to equation 3 as temperature increases, $W$ decreases and more and more neutrons from the low energy range of the spectrum are taking part in nucleation. So $\eta$ increases with temperature. The sharp increase of $\eta$ near 25°C corresponds to detection of neutrons with energies ranging from highest available to those at the peak of the spectrum. At high temperature when all the neutrons of the spectrum contribute in nucleation, $\eta$ should be constant with temperature. But at about 45°C, $\eta$ increases again. We suspected that the sample becomes sensitive to gamma rays coming out of the Am-Be source. In a separate experiment, we indeed observed that R-12 becomes sensitive to gamma rays at about 45°C. Since, as mentioned before, the analysis has been performed up to a maximum temperature of 42.5°C, the contribution due to gamma rays is absent in the measured energy spectrum in this work.

Figure 3 shows that the $d\eta/dT$ vs. T graph resembles the neutron energy spectrum of $^{241}$Am-Be where the second peak corresponds to the gamma sensitivity of the sample. The ambient temperature of the superheated drops was converted to the energy of the neutrons following the method described in Section 4. So by using superheated drops at different temperatures, it is possible to obtain the neutron energy spectrum. This indicates important use of SDD in neutron spectrometry. The maximum uncertainty in neutron energy as could be found from figure 7 is within 5% in the entire region of our investigation. This method can be used to determine any other neutron energy outside the present range, only then one has to consider the ion with maximum $dE/dx$ and the rest of the analysis is same as this. The present study also helps to select the suitable material (liquid) for a given neutron energy spectrum.

It has been observed in this experiment that the nucleation rate of superheated drops rapidly changes for samples exposed to thermal shock compared to samples whose temperature was changed slowly. The liquid appeared to be more fragile when temperature was changed rapidly. Though this is not quite unexpected in the exact physics of this phenomenon, why the liquid becomes more fragile under heat shock, requires further investigation.
REFERENCES

1. D.A. Glaser, Phys. Rev. A 87, 665 (1952).
2. R.E. Apfel, US Patent 4,143,274 (1979).
3. R.E. Apfel, Nucl. Inst. Meth. 162, 603(1979).
4. R.E. Apfel and S.C. Roy, Nucl. Inst. Meth. 219, 582 (1984).
5. R.E. Apfel and Y.C. Lo, Health Phys. 56, 79 (1989).
6. R.E. Apfel, Rad. Prot. Dos. 44, 343 (1992).
7. S.C. Roy, R.E. Apfel and Y.C. Lo, Nucl. Inst. Meth. A255, 199 (1987).
8. H.Ing, Nuclear Tracks. 12, 49 (1986).
9. R.E. Apfel, Nucl. Inst. Meth. 179, 615 (1981).
10. K. Chakraborty, P. Roy, S.G. Vaijapurkar and S.C. Roy, Proc. of 7th National Conference on Particles and Tracks, Jodhpur pp 133 (1990).  
11. H. Ing, R. A. Noulty and T. D. Mclean, Rad. Meas.27, 1 (1995).
12. F. d’Errico, W. G. Alberts, G. Curzio, S. Guldbakke, H. Kluge, and M. Matzke, Rad. Proc. Dos. 61, 159 (1995).
13. F. d’Errico, R. E. Apfel, G. Curzio, E. Dietz, G. F.Gualdrini, S. Guldbakke, R. Nath, B. R. L. Siebert, Rad. Proc. Dos. 70, 1 (1997).
14. F. d’Errico, W. G. Alberts and M. Matzke, Rad. Proc.Dos. 70, 103 (1997).
15. J. W. Gibbs, Translations of the Connecticut Academy III, p.108 (1875).
16. F. H. Newman and V. H. L. Searle, The general properties of matter(fifth ed.), p.189 (1985).
17. R. E. Apfel, S. C. Roy and Y. C. Lo, Phys. Rev. A31, 3194 (1985).
18. M. J. Harper and M.E. Nelson, Radiat. Prot. Dosim. 47 , 535 (1990).
19. Mala Das,B. Roy, B. K. Chatterjee and S. C. Roy, Rad.Meas. 30, 35 (1999).
20. B. Roy, B. K. Chatterjee and S. C. Roy, Rad. Meas. 29, 173 (1998).
FIGURE CAPTIONS

Fig. 1: Variation of threshold energy (W) required for nucleation in R12 as a function of temperature (T).

Fig. 2: Observed variation of neutron detection efficiency (η) as a function of temperature (T) in R12.

Fig. 3: Variation of the derivative of neutron detection efficiency (dη/dT) as a function of temperature (T).

Fig. 4: Variation of stopping power (dE/dx) of different ions (C, Cl, F) in Freon-12 as a function of neutron energy.

Fig. 5: Variation of average stopping power (dEdx)_{\text{average}} over three different ions in Freon-12 as a function of neutron energy.

Fig. 6: Variation of stopping power (dE/dx) of ion in R12 as a function of temperature (T) of the sample, for different arbitrary values of k.

Fig. 7: Variation of neutron energy as a function of temperature (T) of the sample, for different k.

Fig. 8: The neutron energy spectrum of $^{241}\text{Am-Be}$ obtained from the experiment.