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Radiation Physics and Chemistry 61 (2001) 271–281

Radiation Physics and Chemistry

www.elsevier.com/locate/radphyschem

Superheated liquid and its place in radiation physics

S.C. Roy*

Department of Physics, Bose Institute, 93/1 Acharya Prafulla Chandra Road, Calcutta 700 009, India

Abstract

Superheated liquid drops suspended in a visco-elastic gel (known as a superheated drop detector) or in a more rigid polymer matrix (known as a bubble detector) are known to be a useful tool in radiation physics. Superheated liquids have been used as radiation detectors in health physics, medical physics, space physics, nuclear and high energy physics. In addition, the physics of nucleation is not fully understood and requires further investigation. The present paper discusses the special features of a superheated drop detector which has made its place in almost all branches of radiation physics within 20 years of its discovery.

Keywords: Superheated liquid; Nucleation; Neutron; Spectrometry; Dose; Detector

1. Introduction

Any liquid maintaining its liquid state at a temperature above its boiling temperature is called a superheated liquid. It is a metastable state of the liquid and can maintain this state if 'cared' for appropriately. By this, we mean that the container of the liquid should be perfectly smooth, there should be no air bubbles, gas pockets, or impurities and there should be no arbitrary fluctuations of temperature. However, it has been known from before the construction of the first bubble chamber (Glaser, 1952) that ionising radiations can initiate boiling of the superheated liquid. Drops of superheated liquid suspended either in a visco-elastic gel (known as the superheated drop detector, Apfel, 1979), or in a more rigid polymer matrix (known as the bubble detector, Ing and Birnboim, 1984) are found to be more versatile and useful compared to the bubble chamber due to its continuous sensitivity and small size. Within the 20 years since its discovery, the superheated drop detector has made its place in almost all branches of radiation physics, including, nuclear physics, health physics, medical physics, space physics and high energy physics. The application of these devices as neutron dosimeters has already been established (Apfel and Roy, 1984, 1985a; Apfel and Lo, 1989). Superheated liquids as neutron dosimeters also being presented by this author (Roy et al., 1987) in one of this series of conferences. The particular emphasis of this work is on the application of these devices in neutron spectrometry, in medical physics, in gamma ray detection. The paper has been organised to explain the basic mechanism of nucleation, the method of detecting nucleation, the method of measurement, application of superheated liquid on different areas of radiation physics and its potential applications in different areas of radiation physics.

2. Basic theory of nucleation

Using classical thermodynamics, one can calculate the minimum work (W) required in creating a spherical bubble of radius r, given by

$$W = 4\pi r^2\gamma(T) - \tfrac{4}{3}\pi r^3\Delta P. \tag{1}$$

Here $\gamma(T)$ is the surface tension of the liquid at temperature T, ΔP is the difference between the equilibrium vapour pressure (P_V) and the externally applied pressure (P_0) which is atmospheric pressure in our case.

This minimum work to create a bubble increases with the size of the bubble, reaching a maximum and then decreasing. The maximum constitutes an energy

*Corresponding author. Fax: +91-33-350-6790.
E-mail addresses: scroy@boseinst.ernet.in (S.C. Roy).

0969-806X/01/$ - see front matter
PII: S0969-806X(01)00250-X

barrier which must be overcome in order for nucleation to occur. The radius of the bubble corresponding to this maximum is called the critical radius (r_c) and is given by

$$r_c = 2\gamma(T)/\Delta P. \quad (2)$$

Substituting Eq. (2) into Eq. (1), the minimum reversible work needed to form the critical size bubble is given by

$$W = (16\pi/3)[\gamma^3(T)/(\Delta P)^2]. \quad (3)$$

This important equation implies that as the temperature increases, W decreases thus requiring less energy for vapour nucleation.

Although a complete theory of radiation-induced nucleation in superheated liquid is not available, Seitz's 'thermal spike' model (Seitz, 1958) is found to be satisfactory in explaining the basics of radiation induced nucleation. Seitz's theory suggests that when a heavy charged particle slows down in moving through a liquid, kinetic energy is transferred as thermal energy in extremely small regions (as temperature spikes). The intense heating induces localised boiling, creating trails of submicroscopic vapour seeds of different sizes. Only the vapour seed which reaches the critical size will grow into a macroscopic observable vapour bubble and the drop nucleates. Considering other dynamic factors such as viscosity, etc., the minimum energy E_m required for bubble formation is given by (Bell et al., 1974)

$$E_m = W + H + E_{wall} + F, \quad (4)$$

where H is the vaporisation energy, E_{wall} is the kinetic energy imparted to the liquid by the motion of the vapour wall and F is the energy imparted to the liquid during the growth of the bubble by the viscous forces. It has been found that the last two terms can be neglected. However, W is a more universal quantity in bubble formation and our major discussion will be based on that.

Radiation-induced nucleation is a dynamic process, involving fluid thermodynamics of the growth of the bubble. This was not included in the static equilibrium thermodynamic approach and therefore cannot provide a complete answer. The complete dynamical model for bubble nucleation in superheated liquid by ionising radiation has been proposed recently by Sun et al. (1992) using a numerical technique. The numerical treatment involves description of the behaviour of the liquid by the usual macroscopic fluid equations and assumed that the energy is deposited instantly and uniformly along an infinite line in the immediate vicinity of a heavy charged particle. This has been solved in selected cases by means of hybrid computational methods, but the required amount of computation prohibits its general use. Therefore models (Apfel et al., 1985b) and even semi-empirical approach (D'Errico, 1999) to understand the mechanism are in use for many practical purposes.

In the case of the neutron, the neutron deposits energy through the secondary ionizing particles produced during interaction with the nuclei of the liquid. When a neutron of energy E_n interacts with a nucleus of atomic weight A, the maximum kinetic energy that can be transferred to the nucleus from the neutron is through the elastic head-on collision and is given by

$$E_A = 4AE_n/(A+1)^2. \quad (5)$$

After receiving the energy, the nucleus is scattered off from the atom and moves through the liquid losing its energy through Coulombic interactions until it comes to rest. For a given neutron energy, different nuclei of the liquid will receive different amounts 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. The energy deposited by the charged particle within a distance L along the radiation particle track, must be larger than W required for bubble formation and is given by

$$W = L(dE/dx) = kr_c(dE/dx). \quad (6)$$

We have associated the length L with the critical bubble radius by a numerical constant k.

By considering that the energy deposited along that part of the ion's range corresponding to twice the critical radius contributes significantly to bubble formation, Apfel et al. (1985b) found that W corresponds to only 3–5% of the energy deposited by the ions in the critical diameter $2r_c(dE/dx) = E_c$, say. Interestingly, this has been found to be true when applied to the experimental data of Greenspan and Tschiegg (1982), who measured temperature dependence of the acoustic cavitation threshold for liquids exposed to a Pu–Be neutron source. Here the liquids were superheated by the negative pressure imposed by the acoustic waves. That the thermodynamic factor (defined as W/E_c) is more or less constant for different experimental situations suggests that physics of nucleation process is the same irrespective of the varying conditions of irradiation and nucleation. This thermodynamic factor has been utilised to calculate the threshold neutron energy for a given liquid and temperature (Apfel et al., 1985b). Recently, D'Errico (1999) observed, although empirically, that by introducing a non-dimensional quantity 'reduced superheat' defined as $(T - T_b)/(T_c - T_b)$, where T_b and T_c are the boiling temperature and the critical temperature of the liquid respectively, it is possible to predict the threshold neutron energy of a superheated liquid at a given temperature.

3. Detection of nucleation

There are two distinct ways by which one may detect nucleation: using active devices and passive devices. The Easiest passive device is that of counting vapour bubbles visually with the naked eye. In fact, there is one such instrument known as NeutrometerS (marketed by Apfel Enterprises Inc., CT, USA) in which bubbles are counted with the naked eye while in BDPND (marketed by Bubble Technology Industries Inc., Chalk River, Canada) counting of bubbles is done optically by digital scanner. These methods of counting bubbles are reasonable only when the incoming flux of radiation is low or when the nucleation rate is small.

Other passive devices essentially rely on the measurement of the volume of vapour formed due to nucleation. In NeutrometerHD (marketed by Apfel Enterprises Inc.), a 1-ml pipette with 0.1 ml resolution containing thick gel is attached to the vial containing superheated drops. As the drops nucleate, the vapour formed pushes the gel along the pipette. Once the graduation of the pipette is calibrated with dose, one may read the dose from the change of the marks of the gel. In the passive dosimeter developed by Das et al. (2000c), a glass vial containing superheated drops is connected to a horizontal glass tube containing a small coloured water column (used as a marker) which is placed along a graduated scale. Vapour formed due to drop nucleation then pushes the water marker along the glass tube. The displacement of the water column is directly related to the nucleated volume. One may increase the sensitivity of the apparatus, in principle, to a large extent by choosing a glass tube of smaller diameter. However, in the present set-up, glass tube of 1 m length has been used in order to keep the size of the instrument to a reasonable length. The present apparatus is capable of measuring neutron dose as small as 0.1 μSv. In addition to its superior sensitivity, this device, unlike others, is capable of taking real time measurement by placing the vial in the radiation area while placing the measuring apparatus in the control room. A schematic diagram of the apparatus is shown in Fig. 1.

The active device developed by Apfel and Roy (1983) senses the pressure change every time a drop nucleates by a piezoelectric transducer. When a drop vaporises, there is a 'burst' or 'explosion' due to the release of pressure from the vapour tension of the liquid to the atmospheric pressure. This pressure pulse, associated with each drop nucleation, picked up by the piezoelectric transducer coupled at the bottom of the vial containing superheated drops, is converted into electric signal. This electric signal is then sequentially amplified, fed to a rectifier detector, compared, digitised and counted by a counter. The schematic diagram of the arrangement is presented in Fig. 2. Since the output pulse of the transducer has a finite width which is a function of transducer damping (known as ringing), it is not convenient to feed the amplifier output to the comparator to compare with the voltage set above the noise level (one trigger will otherwise produce multiple counts). In order to avoid this problem, a rectifier detector is used which produces only the profile or the envelope of the amplified transducer pulse as shown in Fig. 2. This pulse is then fed to the comparator and the comparator output is used to produce the required TTL output, which is fed to the digital display unit for direct display

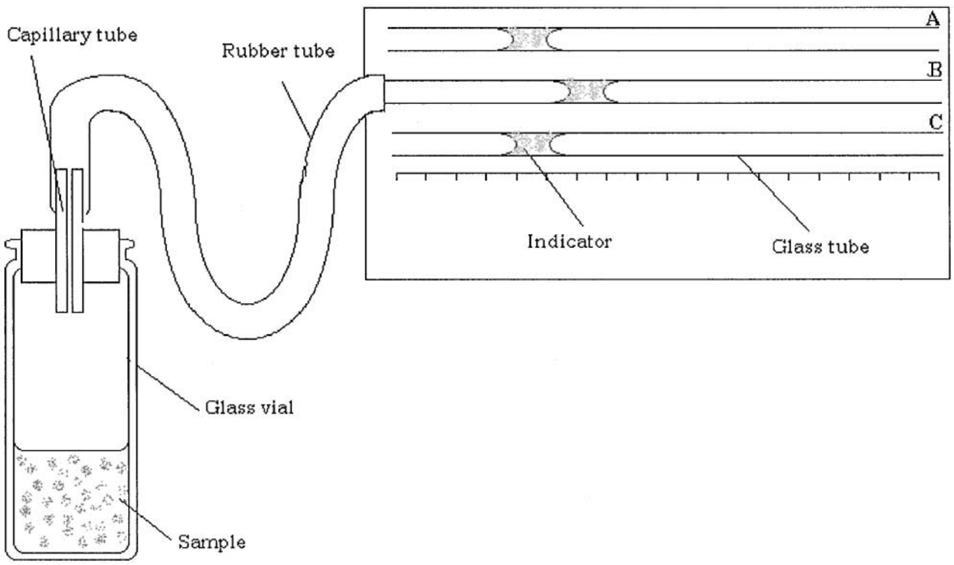

Fig. 1. Schematic diagram of the passive device used to detect radiation induced vapour nucleation.

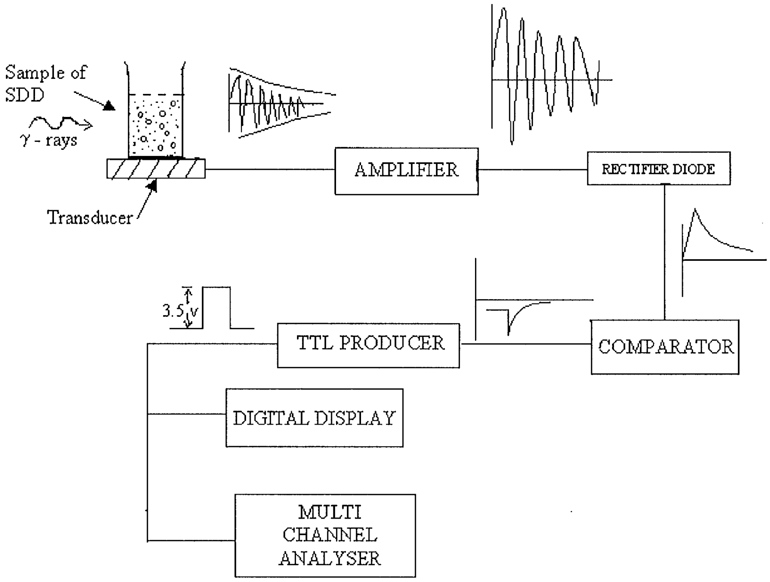

Fig. 2. Schematic diagram of the active device used to detect radiation induced vapour nucleation.

of counts. The entire electronics is built in our laboratory and is now available in a convenient portable size, operating with both 110 and 220 V.

Although the active device is more accurate than the passive device, the passive device is found to be more suitable in many practical applications. The passive device is easy to use, does not require any power source and has been used extensively in medical and health physics in view of the level of accuracy needed in such studies. Passive device could be used in intense radiation field without 'electronic saturation' problem associated with electronic measuring devices.

4. Basic principle of measurement

When a superheated liquid is exposed to energetic radiation, drops nucleate and the rate of nucleation depends on the flux of incoming particles, initial volume of the liquid (or the initial number of drops), and the efficiency of detection of the liquid at that temperature.

In passive devices, the rate of change of vapour volume formed due to vaporisation (also called as the rate of nucleation) is measured. Superheated drops are suspended in the gel without physically touching each other and they nucleate in a random manner independent of each other. Under such conditions it can be shown that the number of drops would decay exponentially with a time constant τ (lifetime). If a vial containing superheated drops is kept in a neutron field such that neutrons of flux ψ are incident on the drops of total volume V_0, liquid density ρ_L and molecular weight M, the initial vaporisation rate is given by

$$dV/dt = V_0\psi N_A\rho_L\eta d\sum n_i\sigma_i/M, \tag{7}$$

where N_A is the Avogadro number, d is the average droplet volume, n_i is the number of nuclei of the ith element of the molecule whose neutron-nucleus elastic cross-section is σ_i and η is the efficiency of neutron detection.

In the present air displacement system the change in position (h) of the water column along the glass tube, produced due to nucleation of the drops by neutrons, could be measured as a function of time. So, the rate of increase of vapour volume during nucleation should be equivalent to the rate of decrease of the volume of superheated liquid. Therefore, one can write

$$\begin{aligned}\rho_V A(dh/dt) &= -\rho_L(dV/dt)\\ &= -\rho_L V\psi N_A\rho_L\eta d\sum n_i\sigma_i/M,\end{aligned} \tag{8}$$

where A is the cross-sectional area of the horizontal tube, ρ_V is the density of vapour of the liquid and V is the total volume of the superheated drops existing at any time t.

Integrating and solving the equation above, one may obtain

$$(hA/m) = a[1 - \exp\{-b(t - t_0)\}], \tag{9}$$

where

$$a = \rho_L V_0/\rho_v m, \qquad b = (1/\tau) = \psi N_A\rho_L\eta\, d\sum n_i\sigma_i/M.$$

Here hA/m is the volume of accumulated vapour in time t per unit mass of the superheated sample containing superheated drops and gel. By fitting Eq. (9) for different values of h and t, values of a, b and t_0 are obtained. The slope of this curve at initial time t_0 can be obtained from Eq. (9) as

$$[\mathrm{d}/\mathrm{d}t(hA/m)] = ab. \quad (10)$$

This slope is nothing but the initial nucleation rate at time $t = t_0$, which can be determined as the product of the fitting parameters a and b. From b, one can find the value of η when all other quantities are known. For complete derivation and the method of measurement readers are referred to one of our previous articles (Roy et al., 1997).

In case of active device volumes, V_0 and V will be replaced by number of drops present initially (N_0) and at any time $t(N)$, respectively.

5. Limit of superheat

Since the threshold energy of detection depends on the degree of superheat, one may ask the relevant question as to how high the temperature of a liquid can be raised without boiling or what the maximum limit of superheat is. It could be easily understood that boiling temperature of a liquid is the lower limit of superheat since at a given pressure above this temperature the liquid becomes either a vapour or a superheated liquid. On the other hand, the critical temperature (T_c) of the liquid is the theoretical upper limit since above this temperature the liquid phase could no longer exist. However, critical temperature cannot be reached in practice although there is a maximum attainable temperature for a given liquid without boiling known as the 'limit of superheat of the liquid'. The knowledge of the limit of superheat (T_{sl}) of a liquid is also important in a number of industrial operations where a hot, non-volatile liquid comes in contact with a cold volatile liquid. If the temperature of the hot liquid reaches the limit of superheat of the cold liquid, explosive boiling would result posing a potential hazard to personnel and equipment in the vicinity (Reid, 1983).

The limit of superheat can be estimated either from the thermodynamic stability theory or from the analysis of the dynamics of formation of the critical sized vapour embryos (statistical mechanical theory). Using van der Waals equation of state, one may calculate the maximum limit of superheat as

$$T_{sl} = 27T_c/32. \quad (11)$$

An empirical relationship valid for most of the organic liquids is also available (Reid, 1983) as given by

$$T_{sl} = T_c[0.11\,(P/P_c) + 0.89], \quad (12)$$

where P_c is the critical pressure and P is the ambient pressure.

The statistical mechanical theory predicts the rate of formation of critical sized vapour embryos per unit volume J as given by

$$J = Nf\exp(-W/kT), \quad (13)$$

where N is the number density of molecules in the superheated liquid. A J value of 10^6 nucleation/m^3 s is often used to define the limit of superheat temperature.

Experimental values of T_{sl} reported so far are far below the predicted limit of superheat. One of the reasons is that observing 'pure' homogeneous nucleation experimentally without any chance of heterogeneous nucleation is difficult, if not impossible, to achieve. So the challenge is how close to the predicted limit of superheat one can reach experimentally. Recently, experiments have been performed (Das et al., 2000a, 2001) to measure T_{sl} using the active device coupled to a multichannel scaler while heating the homogeneously suspended superheated drops slowly above room temperature till nucleation occurs. The rate of nucleation against temperature for three different liquids (R-12, R-114 and R-22) is presented in Fig. 3.

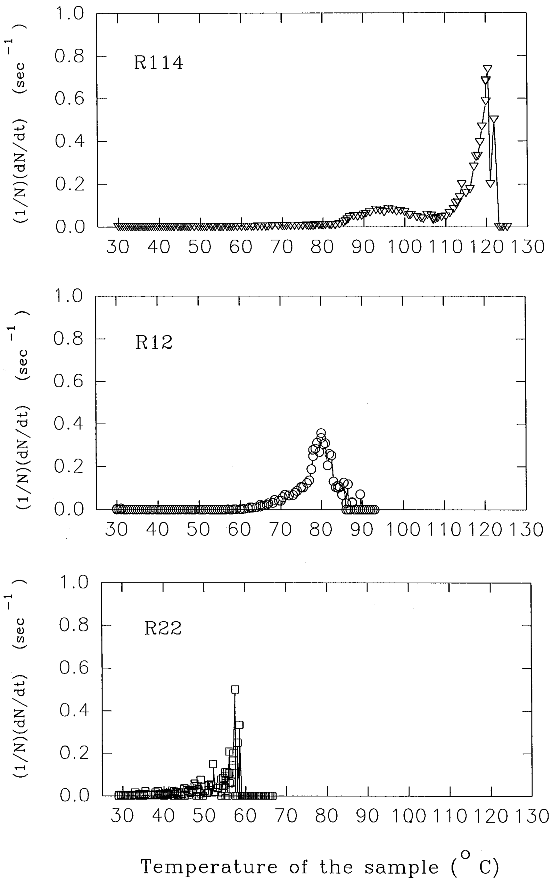

Fig. 3. Results showing the maximum limit of superheat for three different liquids.

Table 1
Comparison of experimental reduced limit of superheat [T_{sl} (K)/T_c (K)] of liquids with predicted limit

Liquid	T_c (K)	Predicted reduced limit of superheat			Experimental values	
		van der waals	Reid	Rate theory	Present	Others
R-12	384.5	0.844	0.89	0.899	0.92	0.89 [a]
R-114	418.7	0.844	0.89	0.94	0.94	0.896 [a]
R-22	369.0	0.844	0.89	0.89	0.89	0.886 [a] 0.89 [b]

[a] Reid (1983).
[b] Avedisian (1985).

Comparison of the results of this experiment with the theoretical limit and other experimental values has been presented in Table 1. As could be seen, the limit of superheat measured by the present method exceeded the theoretically predicted values indicating the inadequacy of the available theoretical calculation and warranting improved calculation. The present experimental values also superseded other measurements in reaching the temperature closest to the critical temperature. The experimental values prior to this measurement reveal that the reduced limit of superheat (defined as $T_{sl}(K)/T_c(K)$) of only 14 liquids out of 56 liquids studied by Avedisian (1985) hardly exceeded 90%.

6. Superheated liquid in neutron spectrometry

Neutron spectrometer utilises the basic property of the superheated liquid such that as the degree of superheat increases, less and less energetic neutrons are required to cause nucleation. 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 degrees of superheat at a given ambient temperature (above their boiling points) and as the ambient temperature increases the given liquid becomes more and more superheated. There exist, therefore, two distinct types of methodologies used in developing the neutron spectrometer. In one, a collection of superheated samples made of liquids with different boiling points (i.e. responses with different threshold energies) are utilised (Ing et al., 1995), while in the other, two liquids with different boiling points are chosen and the temperatures of the liquids are varied at four different temperatures to obtain eight sets of threshold energies, equivalent to eight different samples with different boiling points (D'Errico et al., 1995, 1997). Since these methods are limited by the availability of liquids with lower boiling points of choice and the temperature being varied to only four different values, the 'windows' set to scan the neutron energy spectrum is found to be 'coarse'.

However, by controlling the temperature of a superheated liquid one can, in principle, change the threshold neutron energy to any desired level (equivalent to using 'finer' windows to scan the spectrum). Recently, Das et al. (2000b) utilised the temperature variation method over a wide range of temperature in a single liquid (R-12) to demonstrate its applicability as a suitable neutron spectrometer. In this work, temperature of the sample made of R-12 (dichlorodifluromethane), which is known to be sensitive to neutrons of energies from thermal to tens of MeVs, has been varied almost continuously for a wide range of temperature. Also, the relationship between the temperature and threshold neutron energy has been established using a novel means. The response of the sample exposed to an Am–Be neutron source has been measured at about 30.different temperatures (equivalent to using 30 different samples with 30 different boiling temperatures). The temperature of the sample has been controlled by an indigenously built temperature controller. The temperature is found to be stable within $\pm 0.1°C$. The detection efficiency of the sample liquid at each temperature has been measured using the passive device (Das et al., 2000c) as described in Section 3. The variation of neutron detection efficiency (η) for R-12 with temperature in presence of neutrons from an Am–Be neutron source is presented in Fig. 4. The solid line in the figure is the spline smoothing of the efficiency data at different temperatures. The derivative of the efficiency ($d\eta/dT$) against temperature is shown in Fig. 5, and resembles the neutron energy spectrum of the neutron source. Now, one has to find the equivalence of the temperature of the sample with the neutron threshold energy of nucleation. One of the approaches used by D'Errico et al. (1995, 1997) is to expose the sample at different temperatures to different monoenergetic neutron sources and to note the threshold neutron energies for nucleation. However, this method requires the availability of calibrated monoenergetic neutron sources, which prohibits its general

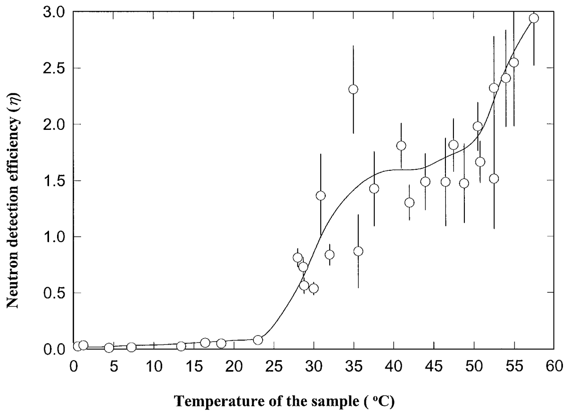

Fig. 4. Observed variation of neutron detection efficiency (η) of superheated drops of R-12 with temperature (T) of the sample.

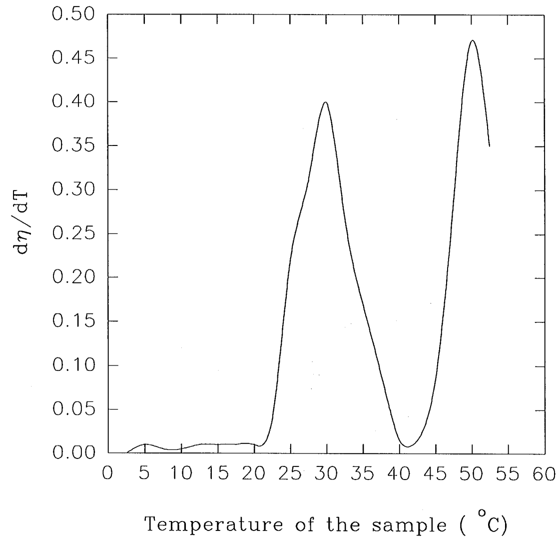

Fig. 5. Variation of ($d\eta/dT$) of R-12 drops with temperature.

application by many research establishments with limited research facilities. We have recently developed a new method of converting temperatures to threshold neutron energies (Das et al., 2000b) as described below.

As presented in Section 2, the different nuclei of the superheated liquid would receive different amounts of energy according to Eq. (5) and they must have different (dE/dx). In case of R-12 which contains C, Cl and F, (dE/dx) of all these ions corresponding to their energies have been determined and a weighted average stopping power for the liquid has been determined. The average (dE/dx) of the liquid with incident neutron energy could then be established. By using Eqs. (3) and (6), the following relationship may be derived:

$$dE/dx = (8\pi/3)\gamma^2(T)/k\Delta P. \quad (14)$$

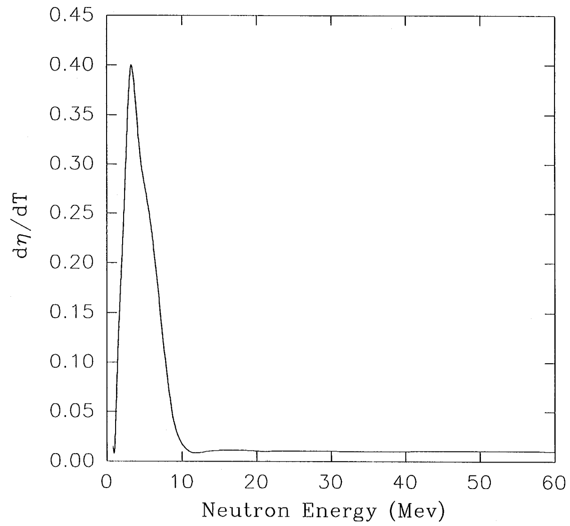

Fig. 6. The neutron energy spectrum of ^{241}Am–Be obtained from the experiment.

(dE/dx) could then be plotted against temperature for different arbitrary values of k. With the optimum value of k, the temperature axis of the figure 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 obtained for $k = 0.0195$. The final neutron energy spectrum of the source obtained from this analysis has been presented in Fig. 6. In another experiment, while studying the neutron energy spectrum of Am–Be using superheated drops of R-114 (Das et al., 2001), it has been found that the threshold neutron energy for different temperatures obtained by this method agrees well with the values obtained by D'Errico et al. (1995) using monoenergetic neutron source, thereby confirming the validity of the present method of determining the threshold neutron energy.

7. Superheated liquid in neutron dosimetry

It is difficult to achieve accurate neutron dosimetry due to the wide range of energy of neutrons (from thermal to tens of MeVs) to be covered and its varying quality factor. An ideal dosimeter should correspond to the ICRP equivalent curve over the entire range of neutron energy. It has been shown that superheated drop detector (SDD) corresponds quite well to the ICRP equivalent curve over the entire energy range when compared with the other commercially available dosimeters such as NTA film, TLD albedo dosimeter, fission track dosimeter or CR-39. Apfel and Lo (1989) reported that the measured response curve of SDD follows the trend of the ideal ICRP dose equivalent curve within

40% for neutrons of energies above 100 keV and within a factor of 10 below 100 keV. In addition, the following unique properties such as its photon insensitivity, passive operation, tissue equivalent composition, isotropic response, small size and low cost, make these detectors one of the most useful tools in neutron dosimetry. Interested readers are referred to some of the relevant literature on this subject (Apfel and Roy, 1984, 1985a; Apfel and Lo, 1989; D'Errico and Egger, 1994; D'Errico et al., 1996).

8. Application of superheated liquid in medical physics

The special properties that these superheated drop dosimeters possess make them an ideal choice for used in medical physics both in in-phantom and in-vivo neutron dosimetry. Recent application in the areas of radiation therapy using high energy medical accelerator produced some interesting and useful results (Nath et al., 1993; D'Errico et al., 1998b, c). High energy X-ray radiotherapy machines are known to generate neutrons by photo disintegration or electron-disintegration of atomic nuclei in various components of the accelerator such as the target, filter, collimator, etc., from various parts of the patient treatment room and also from the patient's body. These contamination neutrons expose the patient to an unwanted dose and also produce secondary radiation by neutron capture reactions occurring in the patient's body and accelerator superstructure. The minimum energy required to remove one neutron from a stable nucleus with $Z > 6$ lies between 6 and 16 MeV. Therefore an electron or photon with energy greater than this minimum energy interact and produce neutrons. Above this energy threshold, the neutron production cross-section increases with energy, reaches a maximum and decreases. Typically, the peak occurs at photon energies of 15–20 MeV in most light elements and at about 8 MeV in heavy elements. Therefore medical accelerators producing photons with energies above 6–8 MeV are potential sources of unwanted neutrons. In case of neutrons produced by electron disintegration, the cross-sections are about two orders of magnitude smaller than the corresponding photo disintegration processes. Therefore, in order to assess the potential risk to the radiotherapy patient due to these neutrons produced in a high energy medical accelerator, it is essential to measure the neutron dose equivalent in and around the radiotherapy beam and in patients. The accurate measurement of neutron dose was difficult to achieve before the availability of the superheated drop detector, due to the complex neutron energy spectrum produced due to room scattering and scattering from treatment head, etc., and the presence of large background of photon flux (three to four orders of magnitude larger than neutrons). Superheated drop detectors are photon insensitive and can produce dose equivalent measurements directly without the prior knowledge of the neutron energy spectrum and are, therefore, a natural choice in this situation. Neutron dose equivalent in and around the high energy photon and electron beam in medical accelerators of 25 MV X-ray and elctron beam with different field sizes and at different distances from the beam axis have been studied by Nath et al. (1993) using the superheated drop detector. It has been observed in case of 25 MV photon beam, that the neutron dose equivalent to the beam axis increases with the field size, while outside the beam axis it varies slightly with field size. The same is true for 25 MV electron beam except that the neutron dose equivalent is found to be 1/5th of that for 25 MV X-ray beam. The energy distribution of photo neutrons has also been measured in 18 MV X-ray beam using superheated drop detector by D'Errico et al. (1998c). The neutron energy spectrum was measured by using three superheated drop detectors made of R-12 (sensitive to neutrons above thermal to fast neutrons), R-114 (sensitive to neutrons above 1 MeV) and C-318 (sensitive to neutrons above 5.5 MeV). The irradiation was performed on a tissue equivalent liquid phantom. The measurements were performed at different depths and at different lateral distances from the beam axis. It has been found that within the primary beam, fast neutrons contribute significantly to the total dose equivalent, particularly neutrons above 1 MeV which deliver most of the total neutron dose. On the other hand, immediately outside the beam their contribution drops significantly and most of the dose comes from the scattered neutrons. The analysis of the spectral distribution of the photo neutron field has clear implications for the radiation protection of radiotherapy patients and also in the design of the shield for the protection of critical organs.

The first in-vivo measurement of neutron dose in patients has been performed recently using superheated drop detector by D'Errico et al. (1998b). An in-vivo vaginal probe was fabricated which contains the vial of superheated drops to measure neutron dose and a photon-sensitive diode to measure photon dose. This probe has been used to measure photon and neutron dose at the potential foetus when a woman patient is irradiated by an 18 MV X-ray beam. Two patients have been treated: one with upper chest irradiation and one with the pelvic irradiation. Therefore, in one case the potential fetus is within the field of the direct beam axis while in the other, the potential fetus is about 50 cm away from the beam axis. The measurement revealed an important and interesting result. The measured neutron dose equivalent per unit photon treatment dose (Gy) at 50 cm from the mantle field is four times lower than that measured in pelvic irradiation, whereas photon dose per unit treatment photon dose is about 200 times less. As concluded by the authors, the dose to the potential fetus clearly exceeded the limit regardless of the presence of neutrons, but it must be kept in mind that that the latter contributed a dose in the order of magnitude of the maximum admissible level. Therefore, under these circumstances a pregnant patient would require the application of a radiation shield suitably designed to attenuate both photons and neutrons. Recently, such a shield using alternate layers of lead and borated polyethylene has been designed by Roy and Sandison (2000) which reduces the photon dose by about 50% and the neutron dose by a factor of about 10 when tested in an 18 MV X-ray beam.

9. Application of superheated liquid in gamma ray detection

Although the use of superheated drops in neutron dosimetry and neutron spectrometry has already been established, the application of superheated drops in detecting photons is still in its infancy. The reason for a superheated liquid being sensitive to neutrons at a given temperature and yet insensitive to gamma rays, is the fact that gamma rays, being low LET radiation, could not contribute sufficient energy required to nucleate, while neutrons can do so through their production of secondary charged particles having high LET values. Therefore, most of the liquids suitable for neutron detection at room temperature could detect photons only at higher temperatures. Some liquids have been identified which are sensitive to gamma rays at room temperature (Apfel Enterprises Inc., D'Errico et al., 1998a, Ing et al., 1997, Roy et al., 1999). However, these detectors have not yet reached the stage where they can be used in quantitative measurements at any photon energy. Further research is continuing in order to make the superheated liquid a practical gamma detector.

It has been noted empirically by D'Errico et al. (1998a) that a liquid becomes photon sensitive at a temperature midway between its boiling point and critical temperature. Although the above finding is more or less correct, it is not true as has been observed recently by Roy et al. (2000). According to D'Errico et al., superheated drops of R-12 (boiling point $-29°C$ and critical temperature 111.5°C) will be photon sensitive at a temperature of about 41°C. In our recent experiments, Roy et al. (2001) have observed that superheated drops of R-12 are sensitive to different energetic photons at different temperatures.

10. Application of superheated liquid in high-energy physics

Superheated liquid has recently been considered as a suitable detector to search for cold dark matter due to its unique property of being sensitive to heavy particles, yet insensitive to low ionising radiations (Collar, 1996; Hamel et al., 1997). Current models explaining the evolution of the universe and the measured slight anisotropy of the cosmic background radiation predict an appreciable contribution of non-luminous, non-baryonic matter in the form of a mixture of relativistic, light particles and non-relativistic, massive particles (the so-called hot and cold dark matter). Initial accelerator experiments suggest that cold dark matter is made up of weakly interacting massive particles with masses ranging from 20 GeV/c^2 up to a few hundred GeV/c^2.

For all kinds of detector materials the recoil energies are expected to be smaller than 100 keV. Depending on more detailed assumptions on cross-sections, the expected event rates in the range 4–20 keV are between 0.01 and 100 events per kg per day. Therefore in order to ensure a reasonable count rate, a large target mass of more than 50–100 kg is needed especially if one wants to detect the slight annual variation in count rates of several percent due to the relative motion of the earth around the sun. The latter would be the decisive signature for the detection of dark matter candidates. Due to background limitations, however, current experimental sensitivities are still far too small (> factor of 100) in order to reach the small interaction rates predicted. The main reason for this is the fact that present detectors are sensitive to all kinds of ionising radiation. Therefore, an extremely low level of radioactive impurities in the detector material and its surroundings is necessary, as well as powerful active and passive background rejection techniques. In order to reduce the overall background sensitivity, a detector which is exclusively sensitive to the high ionisation density of recoiling nuclei with $A > 10$, but which is insensitive to radiation of much smaller specific ionisation like alpha, beta, gamma radiations is the ideal choice. It is understood that superheated liquid possesses much of these properties.

It has been shown by Collar (1996) that under realistic background considerations, an improvement in Cold Dark Matter sensitivity of several orders of magnitude is expected from a detector based on superheated liquid droplets. Such devices are totally insensitive to minimum ionising radiation but responsive to nuclear recoils of energies $\sim$few keV. They operate on the same principle as the bubble chamber, but offer unattended, continuous and safe operation at room temperature and atmospheric pressure.

11. Application of superheated liquid in space physics

The knowledge of terrestrial dosimetry, which essentially derives from the nature of interaction of radiation with matter, when extended to space radiation requires the characterisation of the radiation found in space. With this in mind, superheated liquid droplets dispersed in a more rigid elastic polymer, known as bubble detector, has been used in space radiation dosimetry by placing these detectors in satellites (Ing and Mortimer, 1994, 1996). Contrary to expectations, data revealed that a large number of neutrons (32% of the total) above 10 MeV are present. These high energy neutrons are believed to be due to high energy protons striking the aluminium shell of the satellite. The total neutron dose was found to be 0.75 mSv and 57% of the dose equivalent was delivered by neutrons above 10 MeV. The effects of protons and high energy transfer radiation on bubble detector were also examined to ascertain that the dose as obtained from bubble detector is from neutrons only.

By comparing the mechanism of energy deposition in superheated drops for nucleation with several models of biological damage, Ing et al. (1996) argued that the bubble detector may be used to mimic the biological damage explained by these models. This is not surprising when one considers that superheated drops are sensitive to energy deposition within a distance of tens of nanometres while the biological damage is also responsible for energy deposition in a nanometer scale. Therefore they suggest that without going into the details of characterising the space radiation, the superheated drops may be used as a direct and better indicator of true biological detriment from space radiation than dosimetric quantities currently derived from a variety of radiation detection devices.

12. Potential applications

12.1. Targeted drug delivery and enhanced imaging

Apfel Enterprises Inc., USA, is now exploring a new breakthrough in medical technology for both targeted drug delivery and enhanced diagnostic imaging. The three major challenges for efficiently treating localised disease are the delivery and distribution of drugs and the efficacy of diagnosis. If the delivery is not precise, the drug may injure crucial body systems, and if one cannot distribute the drugs to interstitial tissues the effectiveness is compromised. If one does not know whether any or how much drug gets to the site, through diagnostic imaging or other methodologies, then one is working in the dark in assessing the effectiveness of the delivery system. The new technology involves the use of dispersion of drug-laden superheated drops that can be infused into a specific site of the patient (say a tumour) and then triggered by ex-vivo radiation to release the drugs at the diseased site of the body. The therapeutic ratio and hence the potential to cure is enhanced by sparing the normal tissue of drug toxicity while enabling more drug to be delivered to the diseased tissue.

In the process of triggering, the liquid drops are transformed into bubbles with a volume of more than 200 times the drop volume. These bubbles are effective imaging contrast agents for ultrasound or MRI. Therefore the new system (temporarily labeled as ZapRx) combines an important therapeutic function-targeted drug delivery—with an important diagnostic function—enhanced imaging.

A special feature of this new system is that it is generic in that it can potentially carry a wide range of therapeutic agents (e.g. cytotoxic drugs, immunologic agents, gene therapies, anti-angiogenesis compounds). It is also unique because it addresses three of the most important aspects of curing local disease.

The company has applied for US and worldwide patent protection of this breakthrough technology. The company led by Prof. Robert E. Apfel who is the Higgins Professor at Yale University has teamed up with the Yale School of Medicine to perform and evaluate tests both with cell culture systems and with animals in order to assess the therapeutic and diagnostic potential.

12.2. Photon detector and photon dosimetry

Superheated liquid holds promise for use in gamma detection and in gamma dosimetry. Precise calibration of the detecting efficiency of superheated liquids to gamma detection is needed before its practical application as a gamma detector. Preliminary investigation on the use of superheated liquid as a suitable photon dosimeter has been tested by D'Errico et al. (1998a) and it shows great promise for the use of superheated liquid in three-dimensional photon dosimetry. The dependence of threshold energies on temperature as observed in one of our recent experiments suggests its use as a gamma ray spectrometer, similar to the neutron spectrometer.

12.3. Applications in proton and other heavy particles

Preliminary investigation on the application of superheated drops in proton detection and proton beam dosimetry has been done by D'Errico and Egger (1994). More investigations on proton beam dosimetry are needed and are in progress. Responses of superheated liquid drops taken in the form of bubble detectors when exposed with high-energy Kr, Ar, and N ions are also being studied by Ing et al. (1999).

13. Conclusion

We may, therefore, conclude that superheated drops have already been established as a useful tool in radiation physics within twenty years of its discovery and its potential applications in many other areas are envisaged. We are looking forward to seeing further progress.

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