Applied Radiation and Isotopes 77 (2013) 61–67

Contents lists available at SciVerse ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Study of gamma ray response of R404A superheated droplet detector using a *two-state* model

P.K. Mondal *[,1], B.K. Chatterjee

Department of Physics, Bose Institute, 93/1 A P C Road, Kolkata 700009, India

HIGHLIGHTS

- We report a room temperature gamma ray sensitive superheated droplet detector (SDD) with R404A as the active liquid.
- Gamma ray detection threshold temperature of the SDD is measured.
- Temperature variation of nucleation parameters of this SDD is studied using a two-state model.

ARTICLE INFO

Article history:
Received 5 February 2013
Received in revised form
22 February 2013
Accepted 25 February 2013
Available online 6 March 2013

Keywords:
Superheated liquid
Nucleation
Superheated droplet detector
Gamma ray detection threshold
Detection efficiency
Two-state model

ABSTRACT

The superheated droplet detector (SDD) is known to be gamma ray insensitive below a threshold temperature which made them excellent candidates for neutron detection in the presence of gamma rays. Above the threshold temperature, the gamma ray detection efficiency increases with increase in temperature. In this work the gamma ray threshold temperature has been studied for SDD using R404A as the active liquid and is compared to the theoretical prediction. The temperature variation of gamma ray detection efficiency and interstate transition kinetics has also been studied using a two-state model. The experiments are performed at the ambient pressure of 1 atm and in the temperature range of 17–32 °C using a 662 keV ^{137}Cs gamma ray source.

1. Introduction

It is well known that a liquid can be superheated to a temperature above its boiling point. The superheated state is a metastable state, where a minor perturbation like mechanical vibration, thermal fluctuation, energy deposition by energetic radiation etc., can trigger the formation of a stable vapor phase. Application of superheated liquid in radiation detection dates back to the time of the bubble chamber invented by Glaser (Glaser, 1952). The use of a superheated liquid in a more convenient form of radiation detector, known as the superheated drop detector, was reported by Apfel (Apfel, 1979). In superheated droplet detector (SDD), the whole liquid is dispersed in the form of minute droplets in a gel medium. SDDs are used as radiation detector in various areas, from health physics (Apfel and Lo, 1989) to high energy physics (Collar et al., 2000). Superheated liquid is also considered as a suitable detector to search for dark matter (Archambault et al., 2009; Felizardo et al., 2010; Behnke et al., 2011), since by choosing the operating temperature and pressure it can be made sensitive to high ionizing radiations yet virtually insensitive to the majority of backgrounds like gamma rays, beta rays, cosmic muons etc. PICASSO (Archambault et al., 2009), SIMPLE (Collar et al., 2000; Felizardo et al., 2010) and COUPP (Behnke et al., 2011) are the groups working on the dark matter detection using superheated liquids. For the preparation of SDD different low boiling point liquids are used, such that at their operating temperature they are in reasonably superheated state and can be used for detection of ionizing radiations. Recent studies also indicated that in superheated emulsion there exist two groups of droplets one having much shorter lifetime than the other (Sarkar et al., 2008; Mondal and Chatterjee, 2009). The decay of these droplets in SDD has been modeled earlier (Mondal and Chatterjee, 2009) using a *two-state* model which explains the ambiguity in the nucleation rate data. In this paper we have studied the characteristics of R404A (nearly azeotropic blend of $C_2H_3F_3$, $C_2H_2F_4$ and C_2HF_5; b.p. −46.5 °C) based SDD, which is gamma ray sensitive at room temperature. The gamma ray detection

* Corresponding author. Tel.: +913323031185; fax: +913323506790.
E-mail addresses: prasanna.mondal@saha.ac.in, prasanna_ind_82@yahoo.com (P.K. Mondal), barun_k_chatterjee@yahoo.com (B.K. Chatterjee).
[1] Present address: Astroparticle Physics and Cosmology Division, Saha Institute of Nuclear Physics, 1/AF Bidhannagar, Kolkata 700064, India.

0969-8043/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.apradiso.2013.02.021

threshold temperature of R404A is obtained using a 662 keV ^{137}Cs gamma ray source. The nucleation parameters, detection efficiency and interstate transition kinetics of the two metastable state is studied using the *two-state* model (Mondal and Chatterjee, 2009). To the best of our knowledge R404A based SDD has not been studied earlier.

2. Theory

In the superheated state of a liquid, the liquid is under tension and at a temperature higher than its boiling point at a given pressure. Here the liquid teams with a dynamic population of microbubbles (Roy et al., 1987), which pool the energy from the liquid into pockets where the tear grows (spherically) to a maximum size before imploding back and vanishing. A barrier (Roy et al., 1987) (in the radial coordinate) due to the interplay between the surface and volume energy governs the stability of the microbubbles. Occasionally a microbubble will have sufficient energy to overcome the barrier to cause spontaneous homogeneous nucleation. Nucleation may also be triggered by energetic radiation which deposits energy in the liquid and causes radiation induced nucleation. The frequency of spontaneous homogeneous nucleation is quite low compared to the induced nucleation, and this enables one to use SDD as a radiation detector.

During the bubble nucleation, after overcoming the barrier, the microbubble grows by the evaporation of the superheated liquid. This growth of microbubble and subsequent droplet vaporization is accompanied by the emission of an acoustic pulse (Apfel and Roy, 1983). Each individual nucleation is accompanied by the emission of an acoustic pulse and a change in volume. The acoustic pulse or the change in volume can be detected electronically (Apfel and Roy, 1983; Felizardo et al., 2008; Mondal and Chatterjee, 2008), which enables one to detect the nucleation.

Superheated droplets, when exposed to energetic radiation, are expected to decay monotonically. However, when the droplets are irradiated multiple times with a radiation-off period in between successive irradiations, it is observed that the nucleation rate at the beginning of the irradiation considerably increases compared to the rate at the end of the previous irradiation (Sarkar et al., 2008; Mondal and Chatterjee, 2009). This discrepancy in the nucleation rate data indicates that in SDD the droplets are in two metastable states. The droplets continuously move from one of these states to the other and these two states are in thermal equilibrium with each other. When the droplet is in the *normal metastable state* (**N**), it has much longer lifetime than when it is in the *second metastable state* (**S**). These two states were observed for various superheated liquids when irradiated with neutrons and gamma rays (Sarkar et al., 2008, 2010; Mondal, 2011).

At thermal equilibrium the droplets in the detector are distributed among the two metastable states. When irradiated the short lived droplets in *second metastable state* decay much faster than the others in *normal metastable state*, giving a sharp initial fall in the nucleation rate, after which only the long-lived droplets remain and decay (Sarkar et al., 2008; Mondal and Chatterjee, 2009). The short lived droplets repopulate from the *normal metastable state* during the radiation-off period resulting in an increase in the nucleation rate at later irradiations. It was observed that d, the transition rate from second metastable state to normal metastable state (**S**→**N**), is larger than the transition rate from normal metastable state to second metastable state (**N**→**S**) c (Mondal and Chatterjee, 2009).

During irradiation the superheated droplets decay due to induced and spontaneous nucleations and the nucleation frequency of the droplets in normal and second metastable states can respectively be expressed as (Mondal and Chatterjee, 2009),

$$b = b^{spont} + b^{induced} = k_o v + k_1 v\psi \tag{1}$$

and

$$a = a^{spont} + a^{induced} = q_o v + q_1 v\psi. \tag{2}$$

here v is the droplet volume, ψ is the radiation flux, k_o, q_o are the spontaneous nucleation rate per unit volume for the normal and second metastable states respectively and k_1, q_1 are the radiation induced nucleation frequency per unit volume per unit flux for the normal and second metastable states respectively. The first term on the right hand side of Eqs. (1) and (2) accounts for the spontaneous nucleation and the second term accounts for the radiation induced nucleation.

The nucleation parameters of the two metastable states k_o, q_o, k_1 and q_1, and the interstate transition rates c and d can be obtained by fitting the multi-exposure nucleation rate data (Mondal and Chatterjee, 2009). In this work it is done by using the *two-state* model and the temperature variations of the parameters k_o, q_o, k_1, q_1, c and d are obtained for R404A detector.

2.1. Detection efficiency

It is well known that for gamma rays the energy deposition takes place by the secondary electrons, having much larger ranges compared to that of the ions. The detection efficiency of the detector (η_{det}) is defined as the ratio of the number of nucleation events recorded to the number of energetic particles incident on the detector and can be expressed as,

$$\eta_{det} = \frac{\text{no. of counts received}}{\text{no. of particles incident on the detector}}. \tag{3}$$

In case of SDD the detection efficiency can be expressed as (Sarkar et al., 2004a, 2010),

$$\eta_{det} = \frac{-(dS/dt)_{t=0}}{\psi A}. \tag{4}$$

Here, dS/dt is the rate of nucleation of the superheated droplets, ψ is the radiation flux and A is the sagittal sectional area of the detector. Considering the presence of two metastable states Eq. (4) can also be written as,

$$\eta_{det} = \frac{-(dS/dt)^{N}_{t=0}}{\psi A} + \frac{-(dS/dt)^{S}_{t=0}}{\psi A}. \tag{5}$$

Here the first and second terms on the right hand side of Eq. (5) correspond to normal and second metastable states, respectively. Since the spontaneous nucleation rate is negligibly small, Eq. (5) can be expressed as,

$$\eta_{det} = \frac{k_1 S^{N}_{o} \overline{v}}{A} + \frac{q_1 S^{S}_{o} \overline{v}}{A} \tag{6}$$

where, $\overline{v}$ is the average volume of the droplet, $S^{N}_{o} = S_o c/(c+d)$ is the number of normal metastable drops initially present in the detector and $S^{S}_{o} = S_o d/(c+d)$ is the number of second metastable drops initially present in the detector, S_o being the total number of drops initially present in the detector. Hence the detection efficiency of the detector is

$$\eta_{det} = \frac{(k_1 c + q_1 d) S_o \overline{v}}{A(c+d)} = \frac{(k_1 c + q_1 d) V}{A(c+d)} \tag{7}$$

here $S_o \overline{v} = V$ is the total volume of the active liquid initially present in the detector. Thus the detection efficiency per unit volume of the active liquid is expressed as

$$\eta_D = \frac{(k_1 c + q_1 d)}{A(c+d)} \tag{8}$$