PRAMANA
— journal of
physics

Vol. 75, No. 4
October 2010
pp. 675–682

Nucleation efficiency of R134a as a sensitive liquid for superheated drop emulsion detector

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MS received 23 February 2010; revised 21 April 2010; accepted 21 May 2010

Abstract. Superheated emulsion detector is known to detect neutrons, γ-rays and other charged particles. The present work includes the study of nucleation efficiency of superheated drops of one of the CFC-free liquids, R134a ($C_2H_2F_4$), to fast neutrons, its response to γ-rays from ^{241}Am and ^{137}Cs and compare its nucleation efficiency with that of R12. The observation indicates that because of the presence of hydrogen, the nucleation efficiency is less in R134a than in R12 in the present neutron energy range of consideration. R134a is one of the most environment-friendly, commercially available liquid that is suitable for superheated drop detector, specially in neutron dosimetry and one needs to investigate it in detail.

Keywords. R134a; superheated emulsion; chlorafluorocarbon; nucleation efficiency; hydrogen; gamma.

PACS Nos 29.00; 29.90.+r

1. Introduction

Superheated emulsion detector, consisting of drops of superheated liquid, is known to detect neutrons, γ-rays and other charged particles [1–5]. The detector consists of drops of superheated liquid of low boiling point, suspended in aquasonic gel or polymer medium. The minimum energy required for nucleation to occur is called the critical energy. The critical energy decreases with increase in the operating temperature of the liquid. The critical energy obtained from the reversible thermodynamics is given by Gibbs [6] as

$$W = \frac{16\pi\gamma^3(T)}{3[P_{\mathrm{v}}(T) - P_0]^2}, \tag{1}$$

where $\gamma(T)$ is the surface tension, $P_{\mathrm{v}}(T)$ is the vapour pressure of the liquid at temperature T and P_0 is the atmospheric pressure. There are other expressions also for the determination of critical energy [7]. When radiation falls on the drops, if the energy deposition in the liquid exceeds the critical energy required for bubble formation, the drops form bubbles. The critical energy depends on the temperature and pressure of the liquid and on the type of the liquid.

The most popularly used sensitive liquid is dichlorodifluoromethane (R12, CCl_2F_2: b.p. -29.79°C). Other frequently used liquids for neutron detection are R114 ($C_2Cl_2F_4$), C318 (C_4F_8), R142b ($C_2H_3ClF_2$) etc. [3,8,9]. The response of R134a and R12 dosimeters were studied at 25°C as a function of neutron energies in the range of 85 keV–2.5 MeV from tandem proton accelerator using (p, n) reactions [10]. The response of the dosimeters as a function of temperature in 10–40°C range was also studied. It was observed that R134a (HFC) neutron dosimeter was approximately twice as sensitive as R12 dosimeter, although the HFC dosimeter did not give a consistently sensitive response at different temperature. Like the R12 dosimeter, the HFC material dosimeter was insensitive to γ radiation from ^{60}Co source up to 50 Gy (5000 rads) at 20°C. R134a is not sensitive to thermal neutrons because of the absence of chlorine atom.

The neutron dose response of R134a dosimeter and the γ sensitivity at 20°C were studied in details [10], but the study on the nucleation efficiency and determination of γ sensitive temperature remains unexplored. The lifetime of the superheated drops of R134a in the gel matrix has also not been studied earlier. In the present work we intend to study the nucleation efficiency of R134a and to compare it with that of R12. The study of R134a ($C_2H_2F_4$: b.p. -26.6°C) is important in the sense that the physical properties are similar to R12 but it is free of chlorine content and easily available. Therefore, it is more environment-friendly and its use in superheated emulsion detector will be acceptable in the present scenario. The investigation on γ-sensitive temperature gives us the temperature range over which the liquid can be used for neutron detection in γ background which is important for neutron spectrometry also. To choose R134a as an alternative medium for CFC-free liquid, detailed studies are needed.

Some other studies on the neutron detection sensitivity and detector response were given in [11–14]. The photon sensitization of various superheated liquids as a function of the reduced superheat and the reduced superheat limit common to all halocarbons were investigated [3]. The results showed that the photon sensitization arise above the mid-point between the boiling and the critical temperatures of the halocarbons.

The concept of thermodynamic efficiency (η) was introduced by Apfel *et al* [15]. Thermodynamic efficiency is the ratio of the critical energy of nucleation to the deposited energy in a path length of $2r_{\mathrm{c}}$. There is another efficiency term called the detection efficiency, which is the usual efficiency defined for any detector and that is the ratio of the number of neutrons detected to the number of neutrons falling on the detector. The other term is the nucleation efficiency which is a measure of the probability of nucleation occurring for each scattering event of the neutron.

In the present work, the nucleation efficiency of the superheated emulsion detector, R134a, irradiated by neutrons from an Am–Be source are presented and are

compared with that of R12. Comparison is made with R12, because R12 is the most studied liquid in neutron detection. The threshold temperature of nucleation in R134a to ^{241}Am and ^{137}Cs γ-rays has also been investigated. The long-term existence of superheated drops of R134a in aquasonic gel-type medium has been noted.

2. Experimental methods

The superheated emulsion detectors were fabricated at Bose Institute, India, using both the liquids, R12 and R134a, separately and the drops were suspended in a soft aquasonic gel matrix. The prepared superheated drops in gel were taken in several 15-ml glass vials. It was exposed to Am–Be (3Ci) neutrons at room temperature (average 32.5°C) and the number of drops nucleated was counted by an active drop counting device described earlier [16].

In a separate experiment, the superheated emulsion detector made with R134a drops was irradiated by ^{241}Am (450 mCi) and ^{137}Cs (32.5 mCi) γ-rays by varying the operating temperature at a slow rate (about 1°C per 10 min). Spontaneous nucleation with varying operating temperature was also observed. Temperature of the emulsion was varied by heating the detector vial with a heating tape. To measure the temperature of the emulsion accurately, a thermocouple thermometer probe was inserted in the emulsion at the centre of the glass vial. Variation in temperature within the emulsion was about ±0.5°C.

3. Results

The nucleation rate for such a detector can be expressed as originally described by Apfel [17] and later by Das *et al* [14],

$$\frac{\mathrm{d}N}{\mathrm{d}t} = -\frac{N(t)N_{\mathrm{A}}\rho_{\mathrm{l}}V_{\mathrm{avg}}\Psi(E_{\mathrm{n}})\eta\sum_i n_i\sigma_i(E_{\mathrm{n}})}{M} = -bN(t), \tag{2}$$

where V_{avg} is the average volume of the drops, N_{A} is the Avogadro number, ρ_{l} is the density of the liquid, M is the molecular weight, $\Psi(E_{\mathrm{n}})$ is the neutron flux and energy E_{n}, $\sum_i n_i\sigma_i(E_{\mathrm{n}})$ is neutron–nucleus elastic scattering cross-section of the ith element in the molecule having n number of atoms in the molecule, η is the nucleation efficiency. $\mathrm{d}N/\mathrm{d}t$ is the number of drops nucleated at time t, $N(t)$ is the existing number of drops at time t and

$$b = \frac{N_{\mathrm{A}}\rho_{\mathrm{l}}V_{\mathrm{avg}}\Psi(E_{\mathrm{n}})\eta\sum_i n_i\sigma_i(E_{\mathrm{n}})}{M}.$$

The nucleation efficiency described here is the probability of nucleation occurring for each neutron scattering event of the neutron. The details of evaluating neutron–nucleus elastic scattering cross-section, nucleation efficiency and count rate in the present case, have already been described elsewhere [16,18–20]. The experimentally observed variation of integral (accumulated) count as a function of exposure time for both R134a and R12, irradiated with Am–Be neutrons at an average room

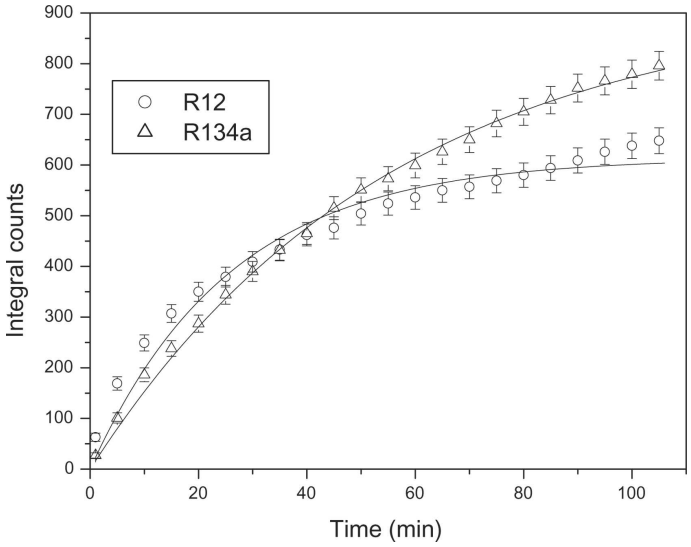

Figure 1. The response of superheated emulsion detector of R12 and R134a irradiated with Am–Be neutrons at room temperature. Solid curves are the fitted curves as explained in the text.

temperature of 32.5°C is shown in figure 1. The measurement was done with the dwell time of 1 min but in the figure the counts at 5 min interval are displayed as sample points for clarity. The solid curves are the exponential fitted curves to the experimental data. The nucleation efficiency of R134a with respect to R12 can be expressed as

$$\frac{\eta_{\mathrm{R134a}}}{\eta_{\mathrm{R12}}} = \frac{[V_{\mathrm{avg}}(\rho/M)\sum_i n_i\sigma_i(E_{\mathrm{n}})]_{\mathrm{R12}}\,[b]_{\mathrm{R134a}}}{[V_{\mathrm{avg}}(\rho/M)\sum_i n_i\sigma_i(E_{\mathrm{n}})]_{\mathrm{R134a}}\,[b]_{\mathrm{R12}}}. \tag{3}$$

The value of b is obtained by fitting the experimentally observed accumulated (integral) number of drops with time in the presence of neutrons (figure 1). The relative nucleation efficiency (η) of R134a with respect to R12 is estimated using expression (3). The value of η thus obtained is given in table 1. Some of the physical properties of both R12 and R134a are also tabulated in table 1.

The γ-ray sensitive temperature was investigated by observing the variation of count rate with increasing operating temperature for ^{241}Am and ^{137}Cs independently. The spontaneous nucleation rate (without any irradiation) as a function of increasing temperature of the detector was also observed. The experiments were performed twice independently, with source and without source, to observe the repeatability of the work. The response to ^{241}Am and ^{137}Cs γ-rays, spontaneous nucleation and the response (solid curve) for only ^{241}Am and ^{137}Cs by subtracting the spontaneous nucleation are shown in figures 2 and 3. The mean of the two experimental data for two different sets at a given temperature was considered while subtracting, though in figures 2 and 3 only one set of experimental data is shown for clarity. Interpolation was done through the experimental mean data points both for the source and spontaneous nucleation. Then the spontaneous nucleation

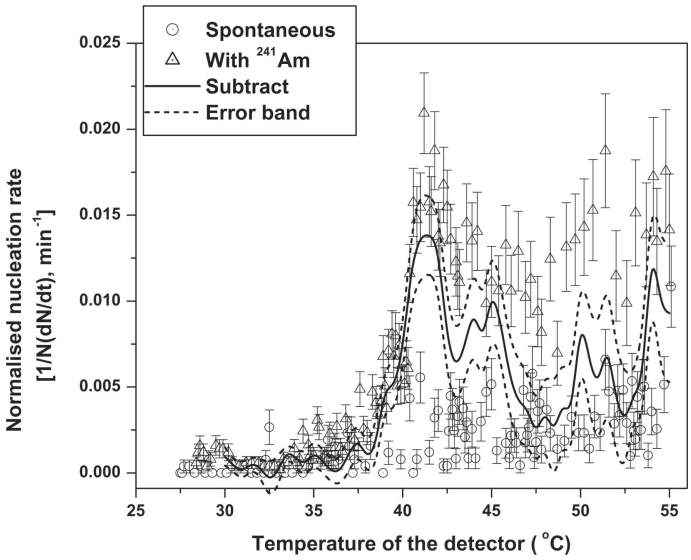

Figure 2. The response of R134a detector to ^{241}Am γ-rays and spontaneous nucleation with varying operating temperatures.

Table 1. Physical properties and results for R12 and R134a (temperature = 32.5°C).

Liquids	Critical temperature (°C)	Critical pressure (atm.)	Degree of superheat (atm.)	W (eV)	σ_{eff}[1] (barn)	Relative nucleation efficiency (η)
			$[P_{\text{v}}(T) - P_0]$ $P_0 = 1$ atm			
R134a	100.9	40.07	7.18	73.7	11.88	8.5 % ± 1.7 %
R12	112.0	40.61	6.85	98.7	10.02	

[1]Estimated for those recoil nuclei responsible for nucleation at this temperature, e.g. C, Cl, F for R12 and C, F for R134a.

data were subtracted from that of the data with source at each temperature. The corresponding response curve is shown as solid line with the error bands in short dash lines in figures 2 and 3.

4. Discussions

It is observed that the R134a detector is sensitive to ^{241}Am and ^{137}Cs γ-rays at about 40°C. Therefore, the observed response at 32.5°C is due to neutrons only.

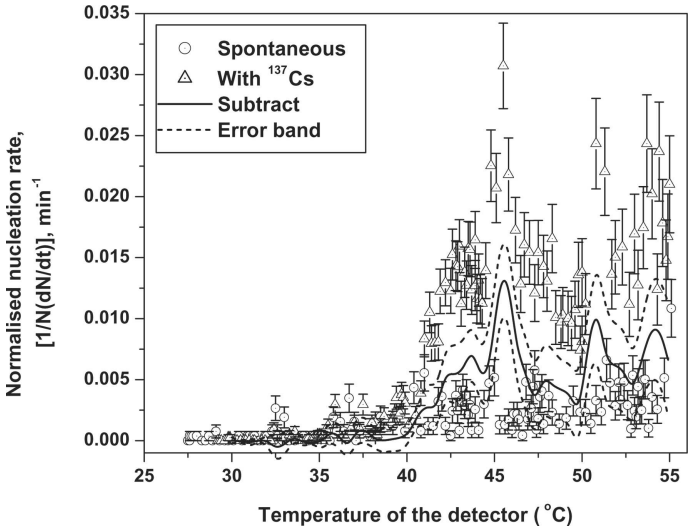

Figure 3. The response of R134a detector to ^{137}Cs γ-rays and spontaneous nucleation with varying operating temperatures.

It was already reported [21] that R12 became sensitive to ^{241}Am γ-rays at about 38.5°C and to ^{137}Cs at about 46°C.

Figure 1 shows that the integral count for R134a is higher than that for R12 as both the detectors contain almost the same number of drops (about 2000), which is expected as the critical energy, W is lower and the total neutron nucleus elastic scattering cross-section is higher in R134a than that in R12. But table 1 shows that the nucleation efficiency of R134a superheated emulsion is lower than that of R12. Lower nucleation efficiency indicates that the probability of nucleation occurring for each scattering event of incident neutron is less and larger fraction of the incident neutrons are lost and a comparatively smaller fraction is utilized in the nucleation process. The most possible reason for the lower value of nucleation efficiency of R134a is the presence of hydrogen. The energy transfer to hydrogen atom via neutron–nucleus elastic scattering is maximum among the constituent nuclei but because of the low LET (linear energy transfer) of hydrogen nuclei in the liquid, the nucleation is not possible at 32.5°C. The available energy responsible for the nucleation becomes less for R134a than that for R12. The critical LET at 32.5°C in R134a is about 44 MeV/mm and the maximum LET of C, F, H nuclei produced because of the elastic head-on collision with the incident neutron, is about 525, 389, 10.6 MeV/mm, respectively for a neutron energy of 3.5 MeV (average peak value of Am–Be neutrons). The critical LET, as was demonstrated earlier [18,22] is calculated using the expression given below:

$$\frac{W}{kr_{\mathrm{c}}}(T) = \frac{\mathrm{d}E}{\mathrm{d}x}(E_{\mathrm{n}}), \tag{4}$$

where r_{c} is the critical radius of the vapour bubble for nucleation and k is the nucleation parameter. It is clear that the LET of hydrogen recoil nuclei is not

sufficient for nucleation at this operating temperature. Another possible reason would be the absence of chlorine in R134a. As the LET of chlorine is the largest in the energy range of interest and among the constituent nuclei of these two liquids, the nucleation is more favourable in R12 than that in R134a at a given neutron energy. The effective incident neutron flux interacting with hydrogen nuclei in R134a is $((\Psi \times 2\sigma_{\mathrm{H}})/\sum_i n_i\sigma_i) = 25\%\,\Psi$, at the neutron energy of 3.5 MeV. This 25% of the incident neutrons are not taking part in the nucleation in R134a. The remaining, $((\Psi \times 2\sigma_{\mathrm{c}} + \Psi \times 4\sigma_{\mathrm{F}})/\sum_i n_i\sigma_i) = 75\%\Psi$ of the incident neutrons are significant in energy deposition and the subsequent steps of nucleation while for R12, there is no such loss.

For a given neutron energy, the nucleation rate is proportional to the neutron–nucleus elastic scattering cross-section, neutron flux, number of drops and nucleation efficiency. The response also depends on the critical energy at a given temperature, and lower the critical energy, higher is the response. Here the response is higher in R134a due to higher cross-section and lower W, and the lower value of nucleation efficiency is compensated by these two factors. But for the range of neutron energies where the cross-section is smaller than that of R12, the lower nucleation efficiency effect would be visible and then, the response of R134a would be lower than that of R12.

5. Conclusions

It is observed that the critical energy (W) of nucleation for R134a at different operating temperatures is lower than that of R12 though the photon sensitization temperature is higher ($\sim$40°C) and is almost similar to R12. This will help in investigating the extended range of neutrons in the relatively lower energy region. The insensitivity to thermal neutrons restricts its application as a thermal neutron dosimeter but it will help in fast neutron spectrometry in the presence of both thermal and fast neutrons. The present investigation, in addition to the earlier observation [10], implies that as the commercial use of chlorofluorocarbon is restricted to protect the environment, the easily available R134a would be a possible alternative for superheated emulsion detector. The hydrogen recoil nuclei produced by neutron elastic scattering are not likely to be responsible for bubble formation because of insignificant LET contribution. This phenomenon lowers the probability of nucleation occurring for each scattering event of the neutrons from R134a when compared with R12 and subsequently lowers the nucleation efficiency.

Acknowledgement

Mala Das is funded by the Department of Science and Technology (DST), Govt. of India, under Women Scientist Scheme-A programme (SR/WOS-A/PS-15/2006).

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