Radiation Protection Dosimetry (2005) Vol. 115, No. 1–4, pp. 398–402
doi:10.1093/rpd/nci124

NEUTRON SPECTROMETRY WITH LARGE VOLUME, HEAVY-LOADED SUPERHEATED DROPLET DETECTORS: A SIMPLE SPIN-OFF

A. R. Ramos[1,2,*], F. Giuliani[2], M. Felizardo[1,2], T. A. Girard[2], T. Morlat[2,3], J. G. Marques[1,2], C. Oliveira[1], D. Limagne[3], G. Waysand[3] and A. C. Fernandes[1,2]
[1]Instituto Tecnológico e Nuclear, EN 10, 2686-953 Sacavém, Portugal
[2]Centro de Física Nuclear da Universidade de Lisboa, Av. Prof. Gama Pinto 2, 1649-003 Lisbon, Portugal
[3]Groupe de Physique des Solides (UMR 75-88 CNRS), Universités Denis Diderot and Pierre et Marie Curie, 75251 Paris, France

SIMPLE is a superheated droplet detector (SDD) experiment designed to search for the evidence of spin-dependent weakly interacting neutralino dark matter (WIMPs). SDDs, a type of emulsion detector, consist of a uniform suspension of superheated liquid droplets in a compliant material such as a polymeric or aqueous gel. We report on the first neutron spectrometry experiments with SIMPLE SDDs, a spin-off of the neutron detector calibrations performed at the Portuguese Research Reactor. SIMPLE SDDs differ from most SDDs available commercially as they have a 10 times higher loading factor, containing 10^3 times more freon than their commercial counterparts and a 100 times larger volume. We have analysed the response of SIMPLE SDDs to two quasi-monochromatic neutron beams of energies 54 and 144 keV obtained with passive filters. Results show that the characteristic peaks in the fluence distribution of both filters could be determined and their energy position obtained using a simple thermodynamic relation.

INTRODUCTION

SIMPLE is a superheated droplet detector (SDD) experiment that is designed to search for the evidence of spin-dependent weakly interacting neutralino dark matters (WIMPs). The particular SDDs used are a type of superheated emulsion, consisting of a uniform dispersion of over-expanded halocarbon droplets (C_2ClF_5–R-115) suspended in a hydrogenated gel. Each droplet functions as a mini-bubble-chamber: charged particles liberated by radiation interactions nucleate the phase transition of the superheated liquid and generate detectable bubbles.

SIMPLE SDDs are large (1 litre) devices, containing 10^3 times freon than those available commercially. Application of previous neutron response studies (see Ref. (1,2) and references therein) is therefore impaired. Moreover, SDDs' neutron response has generally been explored at energies >100 keV, whereas the interest range for SIMPLE can be as low as 2 keV.

SDDs have been investigated for applications in neutron spectrometry for more than a decade[3–5]. Two distinct methodologies are used: (1) a collection of superheated samples made of liquids with different boiling points (i.e. with different threshold neutron energies) is utilised[4]; (2) two liquids are chosen and the temperatures of each liquid are varied to two or three different values to obtain sets of threshold energies[6]. The temperature variation method is superior, since one can, in principle, change the threshold neutron energy to any desired level[5]. We report on the first neutron spectrometry experiments with SIMPLE SDDs, a spin-off of the neutron detector calibrations performed at the Portuguese Research Reactor (RPI). Spectra were obtained using a variation of the second method, with continuously ramped temperature.

MATERIALS AND METHODS

Two neutron passive monochromator filters were set up in the thermal column of RPI. These were composite filters of Si + S and Si + Ti, designed for enhanced neutron transmission at 54 keV (Si + S) and 144 + 54 keV (Si + Ti). Specific details regarding filter structure, output spectra calculation and optimisation can be found in Ref. (7).

Two SIMPLE R-115 SDDs were irradiated. Fabrication procedures are described elsewhere[8,9]. Each SDD was placed in a temperature-controlling water-bath, which was shielded from external background neutrons. During measurement, the bath temperature was ramped from −2 to 15°C over a period of ~10–12 h at a constant rate of ~1.4°C h^{-1}, with the precise temperature range chosen according to the operating pressure as well as the neutron beam energy. The 15°C (16°C) upper limit at 1(2) atm was imposed in order to remain below the high dE/dx Auger cascades from environmental gamma interactions with the chlorine in the refrigerant, observed in underground measurements[8]. The bath

*Corresponding author: ariel@itn.mces.pt

temperature was monitored with three HOBO probes. Each detector was operated at either room pressure or that of 2 atm, monitored with a fourth HOBO probe. Results presented here correspond to experiments performed at 1 atm. Bubble nucleations were acoustically registered and stored in a Lecroy 9350 oscilloscope. Further details regarding the experimental set-up can be found in Ref. (10).

Measurements were performed at a reactor power of 1 MW. The thermal and epithermal flux were measured with uncovered and Cd-covered gold discs; the thermal neutron flux was 2.1×10^5 n cm^{-2} s^{-1} and the epithermal neutron flux (at 1 eV) was 5.3×10^2 n cm^{-2} s^{-1}. The gamma field measured was 19 mSv h^{-1}.

DETECTOR PHYSICS

Radiation-induced nucleation is explained in the framework of the Seitz 'thermal spike' model[11]. Seitz's theory suggests that when a heavy charged particle slows down while 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 sub-microscopic vapour seeds of different sizes. Only that vapour seed which reaches the critical size will grow into a macroscopic observable vapour bubble.

The energy of formation of a critical nucleus (E_c) can be obtained from a thermodynamic model[12,13]:

$$E_c = \frac{4\pi r_c^3}{3}\rho_v h_{fg} + 4\pi r_c^2\left(\gamma - \frac{d\gamma}{dT}T\right) + \frac{4\pi r_c^3}{3}P, \quad (1)$$

where $\gamma(T)$ is the surface tension of the liquid at temperature T; ρ_V is the vapour density, h_{fg} is the vaporisation enthalpy per unit mass, P is pressure and r_c is given by:

$$r_c = \frac{2\gamma(T)}{\Delta P}, \quad (2)$$

where ΔP is the difference between the equilibrium vapour pressure (P_V) and the superheated liquid pressure (P_1), the latter being equal to the externally applied pressure (P). Note that Equation 1 does not take into account any dissipative effects.

Neutrons deposit energy through the secondary ionising particles produced during their interaction with the nuclei of the liquid. The most important interaction to consider is nuclear scattering. When a neutron of energy E_n interacts with a nucleus of mass A, the maximum kinetic energy that can be transferred to the nucleus from the neutron is by means of an elastic head-on collision and is given by:

$$E_A = \frac{4A}{(A+1)^2}E_n. \quad (3)$$

After collision, the nucleus is scattered and moves through the liquid, losing its energy through Coulombic interactions. For a given neutron energy, different nuclei of the liquid will receive different amounts of energy, depending on their mass. For a bubble to nucleate, two conditions must be met: (1) the maximum energy of the recoiling ion (E_A) must be greater than E_c and (2) the energy-loss of the recoiling ion must be high enough to ensure that the energy necessary for the formation of a critical nucleus is deposited in a sufficiently short track-length $L = ar_c$. This last expression associates the length L with the critical bubble radius by a numerical constant a, which is given in Ref. (13):

$$a(T) = 4.3\left[\frac{\rho_v}{\rho_1}\right]^{1/3}, \quad (4)$$

where ρ_1 is the superheated liquid density. These two conditions define an energy threshold for the recoiling nucleus with the capability to originate a bubble, $E_{A,th}$, which in turn defines a neutron energy threshold ($E^A_{n,th}$) via Equation 3.

The response of a superheated drop detector to monoenergetic neutrons can be calculated as follows[14]:

$$R(E_n) = \Phi(E_n)V\sum_i N_i \sum_j \sigma_{ij}(E_n)S_{ij}(E_n). \quad (5)$$

$R(E_n)$ is the response of the SDD, e.g. the number of bubbles formed per unit time, E_n is the incident neutron energy, Φ is the neutron fluence rate given in n cm^{-2} s^{-1}, V is the total volume of the superheated drops and N_i is the atomic density of the ith atomic species of the superheated material, at cm^{-3}. σ_{ij} is the neutron cross section, in barn, of the jth interaction with the ith species and S_{ij} is the so-called 'superheat factor', which weighs the response by the amount by which the neutron energy is above the threshold energy. Considering scattering interactions, S_i is given in Ref. (14):

$$S_i(E_n) = 0, \quad \text{if } E_n \le E^i_{n,th};$$
$$S_i(E_n) = \frac{(E_n - E_{n,th})}{E_n}, \quad \text{if } E_n > E^i_{n,th}. \quad (6)$$

In the case of continuous neutron sources, the response is calculated by integrating Equation 5 with respect to neutron energy:

$$K(E) = \int_E^\infty R(E_n)\,dE_n = \int_E^\infty \Phi(E_n)V\sum_i N_i\sum_j \sigma_{ij}(E_n)S_{ij}(E_n)\,dE_n. \quad (7)$$

RESULTS AND DISCUSSION

Each measurement produced two time-ordered files (one from the bath temperatures and one of pulses from the Lecroy oscilloscope), which required merging and binning in order to obtain spectra. The temperatures from the three thermocouples were averaged to obtain the results presented. Binning was done with respect to time, with bin-size varied between 2 and 60 min. The spectrum is obtained by associating to each bin the average temperature during that bin. Binning time is set by a compromise between spectrum temperature resolution and the count rate statistic error. The detector's thermal inertia creates a difference between surface and inner detector temperature. A rigid shift of -1°C has been applied to the measured temperature to account for this effect, following a simple thermal diffusion simulation[10].

Our experimental results do not correspond to $K(E)$ directly (see Equation 7), but to $K(T(E))$, since we measure detector response as a function of temperature. Figure 1a shows the response results obtained for irradiations using the Si + S filter, binned in 10 min intervals. The squares correspond to the experimental $K(T)$ values. The line corresponds to a five-point adjacent averaging smoothing. Since SDDs are threshold devices with an integral response, deriving the temperature response $K(T)$ should allow the determination of peaks in fluence rate: a peak in fluence rate corresponds to a kink in the detector response. Figure 1b shows the derivative of the smoothed $K(T)$ values. Derivative results have been normalised to a maximum peak value. A single peak is observed at $T = 8.9$°C.

Figure 2a shows the response results obtained for irradiations using the Si + Ti filter, binned in 40 min intervals. The squares correspond to the experimental $K(T)$ values. The line corresponds to a two-point adjacent averaging smoothing. Figure 2b shows the derivative of the smoothed $K(T)$ values. Derivative results have been normalised to a maximum peak value. Two peaks are observed at $T = 4.2$ and 9.4°C. Peak positions were obtained

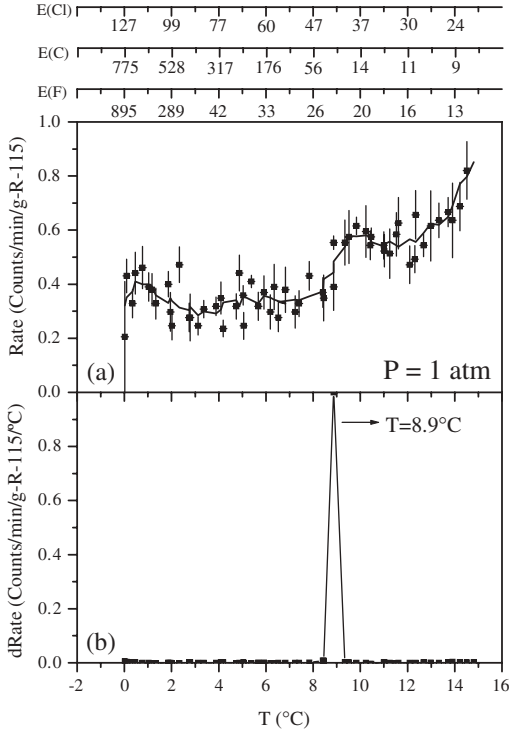

Figure 1. (a) Response results [$K(T)$] obtained for irradiations using the Si + S filter. The squares correspond to the experimental $K(T)$ values and the line corresponds to a five-point adjacent averaging smoothing. (b) Derivative of the smoothed $K(T)$ values.

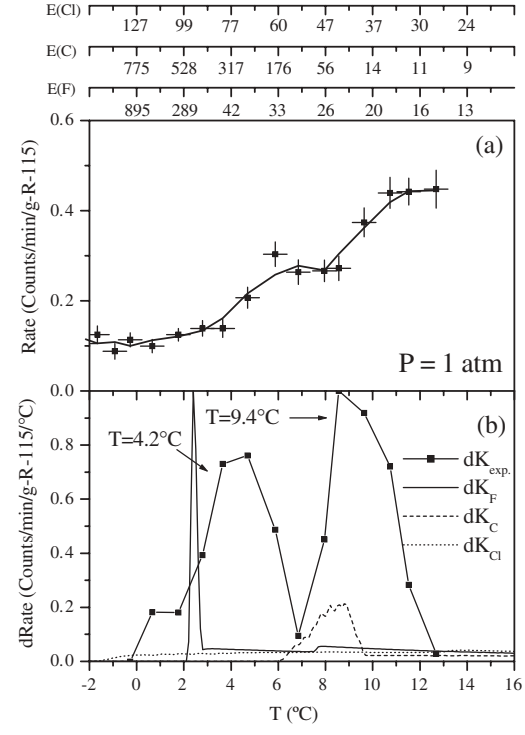

Figure 2. (a) Response results [$K(T)$] obtained for irradiations using the Si + Ti filter. The squares correspond to the experimental $K(T)$ values and the line corresponds to a two-point adjacent averaging smoothing. (b) Derivative of the smoothed $K(T)$ values (closed squares). The line graphs correspond to the derivative of the simulated partial responses for the Si + Ti filter.

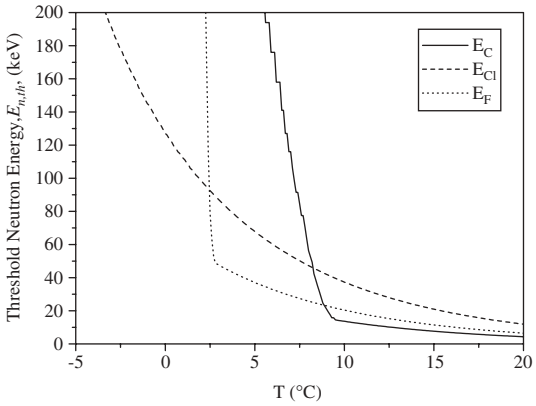

Figure 3. Neutron energy-threshold versus temperature, plotted for the three elements present in the halocarbon droplet: F, C and Cl.

using the following expression:

$$T_{\text{peak}} = \frac{\sum(T \times \mathrm{d}K/\mathrm{d}T)}{\sum \mathrm{d}K/\mathrm{d}T}. \quad (8)$$

The neutron energy-threshold–temperature correspondence was calculated[10] using Equations 1, 2 and 4. Figure 3 shows the correspondence between neutron energy threshold and temperature for the three elements present in the halocarbon droplet, C, Cl and F. It is clear that for R-115 in the temperature range chosen, the threshold neutron energies vary significantly, especially below 10°C. As the temperature increases the differences decrease and for high-enough temperatures the curves for the three elements coincide. The existence of three independent curves for C, Cl and F implies that the observed detector response K at any given temperature T is the sum of three non-zero components, corresponding to three integrals, each with a different lower limit of integration. Figure 2b includes the derivative of the simulated response for the Si + Ti filter for all three partial contributions. Derivative values have been normalised to a maximum peak value. An additional normalisation was introduced to account for the different intensity of each of the three components. For simulation details, see Ref. (10).

According to the energy–temperature relationship given in Figure 3, neutrons of 144 keV (Si + Ti filter) should elicit a response at $T = -0.9$°C for Cl, $T = 2.4$°C for F and $T = 6.5$°C for C. Neutrons of 54 keV (Si + Ti and Si + S) should elicit a response at $T = 6.8$°C for Cl, $T = 2.7$°C for F and $T = 8.0$°C for C. The single peak observed for the Si + S filter at 8.9°C and the peak at $T = 9.4$°C in the Si + Ti filter can therefore be attributed to C recoils originated by the 54 keV neutrons. As shown in Figure 2b, F also adds a contribution originated from 27 keV neutrons. This contribution is related to a distinct peak in the F scattering cross section[10]. The peak observed in Si + Ti filter at $T = 4.2$°C is the result of the convoluted response from C or F recoils originated by the 144 keV neutrons. Note that below ~10°C the neutron energy threshold varies abruptly with temperature, especially for C and F, thus limiting energy resolution in this region. Also, given the experimental procedure adopted, with a continuously ramped temperature, response results have relatively large temperature incertitude, resulting in less than precise derivative peak position.

The analysis of the derivative of the neutron detector response indicates that the features observed are mainly due to the scattered C and F ions. The threshold neutron energy–temperature relationship given in Figure 3 can be adopted to convert the temperature scale of Figures 1 and 2 into an energy scale (top x-axis in Figures 1 and 2). Figures 1b and 2b can thus be interpreted as fluence distribution spectra, recapturing the basic expected features of the monochromatic filters.

CONCLUSIONS

We demonstrate the use of SIMPLE SDD detectors for spectrometry. Results show that the characteristic peaks in the fluence distribution of the Si + Ti and Si + S filters could be determined, and their energy position obtained using a simple thermodynamic relation.

REFERENCES

1. d'Errico, F., Prokofiev, A., Sanniko, A. and Schuhmacher, H. *High energy neutron detection and spectrometry with superheated emulsions*. Nucl. Instrum. Methods Phys. Res. A **505**, 50–53 (2003).
2. d'Errico, F. *Radiation dosimetry and spectrometry with superheated emulsions*. Nucl. Instrum. Methods Phys. Res. B **184**, 229–254 (2001).
3. d'Errico, F., Alberts, W. G., Curzio, G., Guldbakke, S., Kluge, H. and Matzke, M. *Active neutron spectrometry with superheated drop (bubble) detectors*. Radiat. Prot. Dosim. **61**, 159–162 (1995).
4. Ing, H., Noulty, R. A. and Mclean, T. D. *Bubble detectors—a maturing technology*. Radiat. Meas. **27**, 1–11 (1997).
5. Das, Mala, Chatterjee, B. K., Roy, B. and Roy, S. C. *Superheated drop as a neutron spectrometer*. Nucl. Instrum. Methods Phys. Res. A **452**, 273–279 (2000).
6. Apfel, R. E. and d'Errico, F. *A neutron spectrometer based on temperature variations in superheated drop compositions*. Nucl. Instrum. Methods Phys. Res. A **476**, 298–303 (2002).
7. Oliveira, C., Giuliani, F., Girard, T. A., Marques, J. G., Salgado, J., Collar, J. I., Morlat, T., Limagne, D. and Waysand, G. *MCNP optimization of filtered neutron*

beams for calibration of the SIMPLE detector. Nucl. Instrum. Methods Phys. Res. B **213**, 172–176 (2004).
8. Collar, J. I., Puibasset, J., Girard, T. A., Limagne, D., Miley, H. S. and Waysand, G. *Prospects for SIMPLE 2000: a large mass, low background superheated droplet detector for WIMP searches*. New J. Phys. **2**, 14.1–14.14 (2000).
9. Collar, J. I., Puibasset, J., Girard, T. A., Limagne, D., Miley, H. S. and Waysand, G. *First dark matter limits from a large-mass, low-background superheated droplet detector*. Phys. Rev. Lett. **85**, 3083–3086 (2000).
10. Giuliani, F., Oliveira, C., Collar, J. I., Girard, T. A., Morlat, T., Limagne, D., Marques, J. G., Ramos, A. R. and Waysand, G. *Response of SIMPLE SDDs to monochromatic neutron irradiations*. Nucl. Instrum. Methods Phys. Res. A **526**, 348–358 (2004).
11. Seitz, F. *On the theory of bubble chamber*. Phys. Fluids **1**, 2–13 (1958).
12. Peyrou, Ch. *Bubble and Spark Chambers*. Vol. I, Shutt, R. P. Ed. (New York: Academic Press) (1967).
13. Harper, M. J. and Rich, J. C. *Radiation-induced nucleation in superheated liquid droplet neutron detectors*. Nucl. Instrum. Methods Phys. Res. A **336**, 220–225 (1993).
14. Lo, Y. C. and Apfel, R. E. *Prediction and experimental confirmation of the response function for neutron detection using superheated drops*. Phys. Rev. A **38**, 5260–5266 (1988).