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SUPERHEATED DROPLET DETECTORS: THEORETICAL MODEL AND TESTS TO NEUTRON FIELD*

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Abstract. Superheated droplet detectors are known to detect neutrons, gamma-rays or charged particles depending on the operating conditions. The sensitivity and response to neutron fluxes of commercially available detectors, in different experimental conditions, were investigated. This analysis represents the preliminary step in the use of this type of detectors for measurements of neutron background in Romanian low radioactivity underground laboratory.

Key words: superheated droplet detectors, model, neutron.

1. INTRODUCTION

The phase transition from a metastable state produced as a consequence of energy deposition by radiation has been exploited as a method for particle detection is the bubble chamber, where ionizing particles deposit enough local energy in a superheated liquid to produce vaporization along their track. The theory of the process was developed at a satisfactory level in the Seitz model. After three decades, Apfel extended this idea in the form of Superheated Droplet Detectors (SDD), [1] where small drops (with radius of the order of μm) of superheated liquid are uniformly dispersed in a gel or viscoelastic medium. Other recent results are discussed in references [2] and [3].

In a SDD, the gel matrix isolates the fragile metastable droplets from vibrations and convection currents. Hence, the lifetime of the superheated state is extended and thus new classes of applications are possible.

The neutron detectors used in this investigation are manufactured by Bubble Technology Industrie (BTI) as dosimeter devices. The sensitivity and response of these detectors to neutron fluxes in different conditions were investigated. In the future, this class of detectors will be used to characterize the neutron background in

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complex fields of radiation. This principle of detection is also one of the promising directions in searches for Dark Matter, in particular for WIMPs (Weakly Interacting Massive Particles).

2. DETECTION PRINCIPLE AND THEORETICAL MODEL

SDD use superheated liquid droplets as active medium, uniformly dispersed and suspended in a gel. The operation of the detector can be controlled by applying an adequate pressure which makes the boiling temperature of the liquid raise, preventing bubble formation. Under this external pressure, the detectors are insensitive to radiation. By removing the external pressure, the liquid becomes superheated and sensitive to incoming particles. These conditions are similar with those characterising classical bubble chamber detectors. This class of detectors are threshold detectors, bubble formation occurring through liquid-to-vapour phase transition, and being triggered by the energy deposited by incident particles and by the droplet dimension.

In principle, the energy deposition can be the result of nuclear or electronic recoils or the result of direct interactions of gamma or charged heavy particles. Thus, a trigger for particle /energy could be introduced.

In the case of the nuclear recoils produced by neutrons of low energy, the interaction is mainly through elastic scattering on nuclei. The recoil energy spectrum arises due to the familiar kinematics of elastic scattering. In the centre of momentum frame, the projectile scatters off a nucleus at angle ϑ, with $\cos\vartheta$ uniformly distributed between -1 and +1 for the isotropic scattering that occurs with zero-momentum transfer. If the nonrelativistic approximation is acceptable, thus, the recoil has the energy $E_R = E_i r \frac{(1-\cos\vartheta)}{2}$ (in the lab frame), where $r \equiv \frac{4\mu_A^2}{MM_A} = \frac{4MM_A}{(M+M_A)^2}$ is a dimensionless parameter related to the reduced mass μ_A. Note that $r \le 1$, with that $r = 1$ only if $M=M_A$. For this scattering the process is isotropic and the recoil energy is uniformly distributed between zero and $E_i r$. In the usual constructive models for detectors, the active liquids are compounds containing atoms of C, Cl, F and /or heavy elements, as Br for example. The factor r gives the maximum fraction of the energy of the incident neutron transmitted to the nucleus i, where r = 0.19 and 0.28 for ^{19}F and ^{12}C, respectively. When the de Broglie wavelength of the projectile, λ, is $\lambda < \lambda_n$ (r_n is the nuclear radius) the neutron interacts coherently with the entire nucleus. This coherence is manifested up to E_R < 100 keV.

Inelastic collisions only occur if the centre-of-mass kinetic energies of the neutrons are higher than the first excitation level of the nuclei (1.5 and 4.3 MeV for ^{19}F and ^{12}C, respectively). Absorption of neutrons by the ^{19}F nucleus followed by alpha particle emission requires neutron energy of 2.05 MeV.

The processes induced by thermal neutrons could also be the source of bubble nucleation, and the typical reactions are: ^{35}Cl(n, p)^{35}S, and ^{35}Cl(n, alpha)^{35}P. Other particles, as energetic muons, gamma rays, X rays and beta particles have a LET usually below the activation threshold of SSD's, which is typically $\leq \frac{200\text{keV}}{\mu\text{m}}$ at room temperature. Operating at temperatures lower than 30^0C, the detectors are insensitive to gamma rays of energy below 6 MeV [4].

For a phase transition to occur in a superheated liquid the prevailing theoretical model proposed by Seitz [5] predicts that a critical minimum amount of energy E_c has to be supplied within a local thermal spike and if the resulting proto-bubble reaches a critical radius R_c, it becomes thermodynamically unstable and grows rapidly. Thermodynamics predicts that the growth of the bubble passes through several stages of acceleration and deceleration, which also gives rise to a detectable pressure wave. The critical radius is:

$$R_c\left(T\right) = \frac{2\sigma}{\Delta p}.$$

In this equation the degree of superheat at the temperature T is $\Delta p = p_v - p_e$ which is the difference between the vapour pressure (including the gasses solved in liquid) and the external pressure applied to the detector. The quantity σ is the surface tension at the interface between liquid and vapour surface.

The expression for the critical energy which must be supplied is:

$$E_c\left(T\right) = -\frac{4\pi}{3} R_c^3 \Delta p + \frac{4\pi}{3} R_c^3 \rho_v h_{lv} + 4\pi R_c^2 \left(\sigma - T\frac{\mathrm{d}\sigma}{\mathrm{d}T}\right) + W.$$

Here ρ_v and h_{lv} are the density of the gas phase and the latent heat of evaporation. The first three terms of the equation are associated with reversible processes. The energy W includes all woks that go into irreversible processes (energy lost during the bubble growth by the action of viscous forces and the kinetic energy due to expanding bubble against the wall of the liquid, the acoustic energy lost generating the sound, the thermal energy lost during the time of expanding the bubble up to critical radius). In accord with [6], the irreversible processes represent only about 1% of the energy deposed.

In order to produce the phase transition, it is necessary that the energy deposited locally, *i.e.* the kinetic energy of the recoil, exceeds the critical energy

$$E_{deposed}\left(T\right)=\int_{0}^{L_c(T)}\frac{\mathrm{d}E}{\mathrm{d}x}\mathrm{d}x\geq E_c\left(T\right).$$

In practice the natural length scale of the process is given in terms of the critical radius, $L_c = bR_c$, with b empirically determined (a possible value is $L_c=\left(\rho_v/\rho_l\right)^{1/3}\times R_c$), in accord with Azuelos et co-workers [7].

The formation of bubbles is a probabilistic process which has a Bernoulli distribution. The rate of bubble production *R(T)* could be estimated considering the incident particle energy (neutrons), E_n, and their flux $\Phi\left(E_n\right)$ (neutrons/cm^3/s). Thus, if these particles are incident on N superheated drops of total volume V_l with liquid density ρ and molecular weight M, the total number of drop vaporization is small compared with N, the vaporization interaction rate – the response rate (as number bubbles/s) is given by the equation:

$$R\left(T\right)=\int_{E}\Phi\left(E_n\right)V_l\sum_{i}N^i\sum_{j}\sigma_j^i\left(E_n\right)F_j^i\left(T,E_n\right)\mathrm{d}E,$$

where T is the superheated drop liquid temperature. If the target used for detection, is a compound with more atomic species, thus N^i is the atomic density of the i^{-th} species in liquid, and $\sigma_j^i\left(E_n\right)$ is the microscopic cross-section for a *j*-type reaction in the i^{-th} species for neutrons with defined energy. $F_j^i\left(T,E_n\right)$ is the efficiency factor.

3. EXPERIMENTAL STUDIES AND RESULTS

The detectors used in this investigation are manufactured by BTI-Canada. Detectors are available in plastic tubes, with sensitivities ranging from 1.4 to 1.8 bubbles/μSv. All the detectors used in these tests are calibrated to an Am-Be source (about 1.13 x 10^7 neutrons/s, with an average energy of 4.15 MeV). The conversion factor for the source is 3.10×10^5 mrem/n$\cdot$cm^{-2}. In accord with the manufacturer prescriptions, for low doses, it is possible to count the number of bubbles by eye, but the number of bubbles should not exceed 350 before reading them.

The Pu-Be isotopic neutron source (with an activity of $\propto 10^7$ neutrons/s) was used for experimental tests. This source has a complex spectrum, with fast neutrons up to more than 11 MeV. In Fig. 1, the Pu-Be spectrum is presented. This spectrum was measured by Rubbino *et al.* [8] and the adapted figure was obtained using the data from the paper of Notarrigo and co-workers [9].

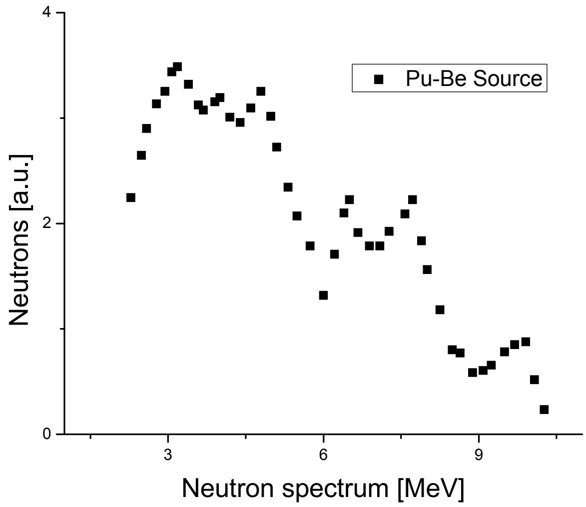

Fig. 1 – Neutron spectra of Pu-Be source.

The source is immersed in water, in a cylindrical stainless steel basin, with a radius of about 85 cm. As the water thickness is increased, fast neutrons are moderated shifting the spectra toward lower energies. For all experimental results reported in this paper the geometry presented in Fig. 2 was used:

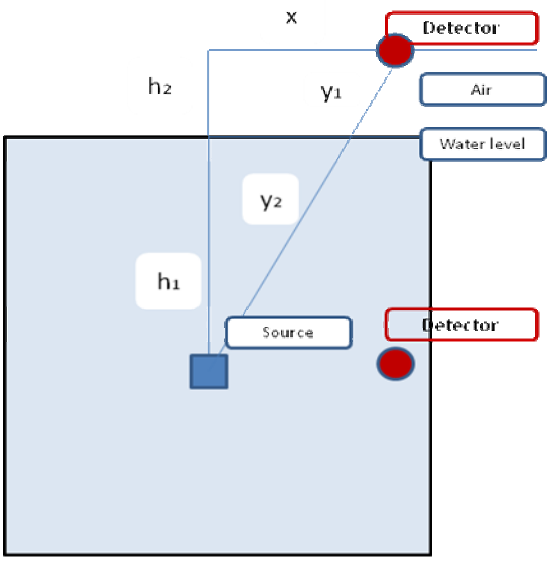

Fig. 2 – Experimental setup.

The average background in the laboratory was estimated after forty eight hours of measurements and a limit of $< 0.025\,\mu\text{Sv/h}$ were established. The neutron background of a value of $0.52 \pm 0.30\mu\text{Sv/h}$ was obtained in the chamber of the sources.

In the experimental geometry, the vertical axis is the symmetry axis, the source is in the centre, and measurements were performed in water and in air.

In air, an isotropy of the doses is obtained after the investigation of doses measured for two different radial directions. The results are presented in Table 1.

Table 1

Doses measured in air, radial, for two different directions

Radius [cm]	Debit of dose [μSv/h] measured for directions angular separated at 120°	
11±0.5	17.8±1.5	21.9±2.2
22±0.5	14.5±1.4	13.8±2.0
33±0.5	10.5±1.4	9.6±1.9

Monte Carlo simulations [10] predict that for a Pu-Be source, inserted in a water moderator, at a radius of 7 cm, low energy neutrons represent about 10% of total number of neutrons and the average energy is 2.04 MeV, and at 21.5 cm this fraction is increased at 30% for a 1.65 MeV average energy. Our measurements put in evidence the fact that the dose response is independent of energy in this energy range – the results are presented in the Table 2. For these detectors, the energy range of response is between about 200 keV, up to more 15 MeV.

Table 2

Experimental results (present work) of the debit of dose and MC simulated neutron spectra at the corresponding positions

Radius [cm]	Debit of dose [mSv/h]	Simulated data [10]		
		Radius [cm]	Low energy neutrons [%]	Average energy [MeV]
7	8.64±0.85	7	10.3	2.04
15	2.34±0.28	16.5	29.34	1,62
21	2.34±0.45	21.5	30.26	1.65

The linearity of the response was investigated studying the response of the detectors (as number of bubbles and thus as dose equivalent) as a function of time in a fixed geometry, detectors being exposed to fast neutrons from the Pu-Be source. A typical result is shown in Fig. 3. The errors are considered as statistical ones. In fact these errors are overestimated, especially at low numbers of bubbles.

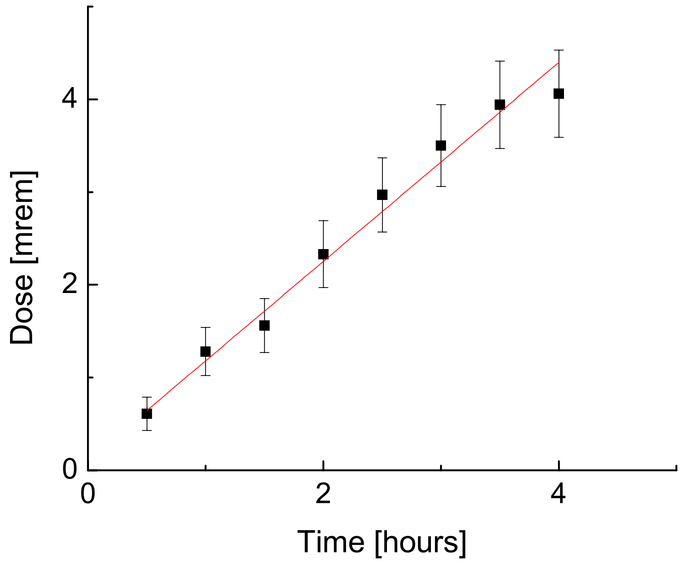

Fig. 3 – Integrated dose *versus* time of irradiation.

Starting from the mechanism of formation of bubbles, the sensitivity of the detector depends on the degree of superheat and hence on the difference between the vapour pressure and the external pressure applied to detector. As the temperature increases, the vapour pressure increases, and thus an increase in sensitivity is expected. In accord with the information from the manufacturer, the detectors are compensated in the range 20^0C to 37^0C. In fact, an aspect important associated with the temperature is the radius of the bubbles formatted. At lower temperatures the radius in larger and this variability in dimension represent an aspect that must be considered especially if the measurement of number of bubble is determined by eyes. In Fig. 4 the variability of the dimensions of bubbles is put in evidence as a function of temperature. Consequently, for detectors there exists an inherent thermal lag, *i.e.*, the detector medium inside the tube takes some time to reach the ambient temperature when moved from an area where the temperature is different. Also, measurements at constant temperature of the ambient medium, in the range of correct response, are recommended. As an observation, if the number of bubbles is counted by eye, the room temperature must be between (20–25)°C because this regime ensures a large size of the bubbles and thus an accurate counting especially in overlapping cases. Also the storage time of detectors produces variation of the dimensions of the bubbles as is an irreversible process – see for example images a) and d) from Fig. 4. The overlapping between bubbles in contact is a possible source of imprecision in the evaluation of the dose.

Fig. 4 – Effect of temperature and storage time.

4. MEASUREMENTS IN AN UNDERGROUND LOCATION

It is preferable that underground laboratories are located in rocks with a low content of natural radionuclides such as limestone, coal or salt in order to reduce the natural level of radiation. The Romanian underground low radiation laboratory was constructed in the former Unirea (Slanic-Prahova) salt mine at 208 m below surface. The natural temperature is about 13.6 ^{0}C and humidity around 65%.

Neutrons from radioactivity are originated in spontaneous fission (of ^{238}U mainly) and *(α,n)* reactions on low and intermediate Z isotopes ($Z < 30$). Generally, their energies are limited to about 10 MeV. Neutrons from cosmic-ray muons have spectra extended to GeV energies. Although the flux of muon - induced neutrons in the deep underground is far below that from radioactivity, they are more penetrating and can be responsible for a significant background component. Any high-*A* material, usually used as a shielding against gammas, is also a good target for neutron production by muons or their secondaries [11].

The muon flux data obtained in underground measurements, (depth from surface is –208 m, equivalent with 610 ± 7 mwe), estimated in accord with [12] is: 0.18 ± 0.01 m^{-2}s^{-1} [13, 14].

Epithermal neutron activation analysis has shown no detectable traces of potassium and thorium in salt and a concentration of uranium of about 0.92 +/– 0.3 ppm [15].

All these information and results suggest that superheated droplet detectors are adequate to measure low energy component of the neutron spectra. For high neutron energies, moderating materials are necessary for measurements. Also, for long time of measurements another important aspect in measurements is the time evolutions of the bubbles. An adequate and controlled temperature during the measurements is necessary.

These detectors could be used as veto systems to reject neutrons in direct WIMP detector systems [16].

5. SUMMARY

The superheated droplet detectors are devices with high sensitivity, able to identify very low doses of neutrons in complex fields of radiations. With integrated response and a good linearity of response for a large range of doses and energies of neutrons, they are adequate detectors for low background measurements.

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REFERENCES

1. R. E. Apfel, B.-T. Chu, J. Mengel, Applied Scientific Research, **38**,117–122 (1982).
2. I. Lazanu, S. Lazanu, JCAP07(2011)013.
3. I. Lazanu, S. Lazanu, Romanian Reports in Physics, **63**, 707–716, 2011.
4. Yuan-Chyuan Lo, Robert E. Apfel, Phys. Rev., **A 38**, 5260–5266, (1988).
5. F. Seitz, The Physics of Fluids, **1**, 2–13 (1958).
6. M. J. Harper, M. E. Nelson, Radiation Protection Dosimetry, **47**, 535–(1993).
7. G. Azuelos *et. al.*, Radiation Protection Dosimetry, **120**, 499–502 (2006).
8. A. Rubbino *et. al.*, Il Nuovo Cimento, **XLIV**, 178–181, (1966).
9. S. Notarrigo *et.al.*, Nucl. Phys., **A125**, 28–32 (1969).
10. H. R. Vega-Carrillo *et. al.*, Revista Mexicana de Fisica, **48**, 405 (2002).
11. V. A. Kudryavtsev, Journal of Physics: Conference Series, **120** (2008) 042028.
12. J. Formaggio, C Martoff, Ann. Rev. Nucl. Part. S, **54**, 361–412, 2004.
13. B. Mitrica *et.al.*, Astrophys. Space Sci. Trans., **7**, 163–166, 2011.
14. B. Mitrica *et. al.*, Romanian Reports in Physics, **62**, 750–757, 2010.
15. C. Cristache, *et. al.*, Radiochimica Acta, 333–337 (2009).
16. Ye Xu *et. al.*, JCAP06(2011)009.