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NEUTRON RESPONSE FUNCTIONS OF SUPERHEATED DROPLET DETECTORS

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The response functions of various C_2ClF_5 superheated droplet detectors fabricated by our team were calculated using the MCNPX-PoliMi and GEANT4 Monte Carlo radiation transport simulation codes. The simulation approach was validated by measurements using an Am-Be source.

INTRODUCTION

Superheated droplet detectors (SDD) developed at C^{2}TN have been employed to search for the evidence of dark matter (DM) in the context of the SIMPLE collaboration [1]. As the interaction of DM candidates with the superheated liquid is expected to mimic neutron-induced signals, the neutron background characterization at the underground experimental site is crucial for the analysis of a DM experiment. This task often relies on calculation tools and facility models, since the extremely reduced intensity of the neutron environment hinders most experimental approaches.

Calculations and measurements of the intrinsic signal of SIMPLE SDDs (containing ~14 g C_2ClF_5) have yielded a neutron-induced noise level of ~6×10^{-3} events per day, corresponding to a fast neutron fluence rate detection limit smaller than 10^{-7} cm^{-2} s^{-1} [2, 3]. The devices can be therefore operated as low-noise neutron detectors particularly fit for neutron measurements in massively shielded facilities. Their neutron response functions are herein investigated via neutron transport simulation as an initial step towards their application in various low neutron intensity frameworks.

MATERIALS AND METHODS

Detectors

Superheated liquids are employed as radiation detectors by identifying the vaporization of the liquid following energy absorption from radiation. A SDD is an emulsion of micrometric liquid droplets in a gel matrix that reduces the occurrence of spontaneous nucleations. This work is focused on 1-3 wt.% C_2ClF_5 SDDs; details on devices fabrication using food gels and on their acoustic instrumentation can be found in Ref. 1 and references therein. Detector volumes were (Fig.1): 1 L (employed in SIMPLE), 150 mL (standard test prototype at C^{2}TN) and 4 mL (with a modified fabrication protocol, for cell irradiation dosimetry).

Figure 1. The evaluated C_2ClF_5 devices. From left to right: 4 mL SDD and microphone; 150 mL SDD with a modified cap to embed the signal and pressure feedthroughs (note the microphone embed in a glycerin layer covering the translucent emulsion); 1 L SDD displaying gel fractures after exposure to high fast neutron fluence.

Radiation-induced nucleations are subject to a dual threshold condition regarding the energy deposited within the droplet and the deposition distance along the particle track, i.e., the radiation linear energy transfer (LET). These are found to depend on the liquid and on the operation thermodynamic conditions [4]. Stopping power tables of Ziegler/SRIM are used to extract the overall critical energy (E_c).

By operating at reduced superheat (low T and high p) the SDD can be rendered insensitive to minimum ionizing particles (mip). At terrestrial levels the SDD is sensitive to only nuclear recoils following neutron interactions and to alpha particles (α) that originate

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*Corresponding author: anafer@ctn.tecnico.ulisboa.pt from natural emitters. These can be further discriminated on the basis of the acoustic signal amplitudes, α's inducing larger amplitudes due to the production of various proto-bubbles [1,5]. The intrinsic insensitivity to mip's and the n/α discrimination are crucial to the low neutron-induced noise of the SDD.

Measurements

The dependance of E_c with the thermodynamic conditions yields the possibility to perform neutron spectrometry by changing the operation temperature (T) and/or pressure (p). In this work, the response of a 150 mL SDD (1.6 g liquid) to Am-Be neutrons was measured as a function of temperature (in the range 4-13 ºC at steps of 1 ºC) at 2 bar. The neutron source (external dimensions: dia. 17.4 mm x 19.3 mm thickness) with 0.09 mCi activity was placed at a distance of 1.5 m from the SDD. A low count rate was sought in order to avoid acoustical effects induced by the gas bubbles accumulating during the experiment.

The test SDD was placed in a small thermostatic water bath (yielding a layer of 2 cm water around the SDD) and covered with aluminum foil to improve thermal insulation. A dummy SDD (without liquid) was used to monitor the detector temperature T. The devices were left to stabilize for 1 hour following any temperature change. The test SDD is pressurized at 2 bar prior to each measurement in order to minimize small leaks potentially occurring. Each measurement ran for 60 min. Background measurements, with the neutron source removed, were also performed.

Simulations

The response of the three SDDs was calculated using general-purpose radiation transport Monte Carlo simulation codes. The MCNPX-PoliMi code was generally employed, with MNCPX (v.2.7.0) simulating the neutron transport and MCNPX-PoliMi (v.2.0) extracting the corresponding recoil distributions.

Material compositions and densities were extracted from Ref. 2. An homogeneous liquid-gel mixture with 10 wt.% liquid was considered. The liquid concentration is increased relative to actual in order to reduce the statistical uncertainty in the recoil distribution retrieval. The atomic concentration of hydrogen - the main gel ingredient, followed by oxygen - is diminished by only 2 at.% (from 58.4 at.% at 1 wt% liquid) yielding a negligible systematic error on the calculated detector signal.

The GEANT4 code (v.10.3–p.01) with the neutron high precision model from the QGSP_BERT_HP physics was employed to retrieve both neutron and recoil distributions - to be compared with the MCNPX-PoliMi results.

Neutron fluence rates (track length estimator) and recoil distributions in the detector volume were calculated as a function of the incident neutron energy. The response as a function of T at p=1 bar and 2 bar was initially extracted from the MCNPX-calculated reaction rates assuming maximum energy transfer from the neutrons to the nuclei. This simplification was later replaced by the usage of the –PoliMi or GEANT4 recoil distributions subject to the $E_c(T,p)$ energy cutoff.

For the calculation of response functions, monoenergetic, monodireccional plane neutron sources at 10 cm from the SDD axis were considered. The simulation of the Am-Be irradiation assumed a point isotropic source at 1.5 m from the SDD. The neutron energy spectrum was extracted from the ISO-8529-1:2001 standard; the data refers to an emission undisturbed by the source structure hence underestimates the low energy region of the spectrum. Neutron interactions within the experimental set-up were neglected except for the thermostatic SDD bath included in the model. The results were scaled per neutron incident on the detector surface and per unit mass of liquid when applicable.

RESULTS AND DISCUSSION

Neutron fluence

The energy distribution of the on-detector neutron fluence rates (Fig. 2) evidences the modification exerted by the SDD over the incident neutron spectrum, with a downscatter region that, for the larger volume devices (150 mL and 1 L), terminates in a thermal neutron component. The discrepancy between GEANT4- and MCNPX-calculated neutron fluence rates is smaller than 7% for all neutron energies and within 1-2% for neutron energies larger than 1 eV and for the total neutron fluence rate. This result is similar to that of other works using the high precision neutron physics models of GEANT4 [6].

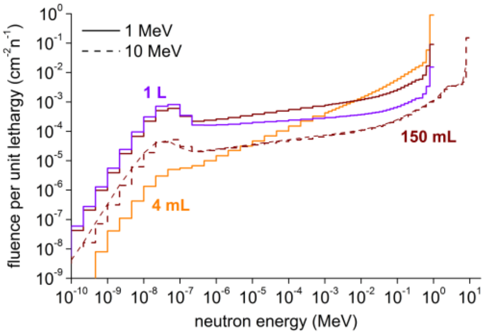

Figure 2. On-detector neutron fluences. The smooth line plots to a GEANT4 calculation (150 mL, 10 MeV).

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Event-producing reaction rates

Among the various reaction channels available the predominant event-producing reactions in C_2ClF_5 (for the neutron energy, T and p ranges considered) are elastic and inelastic scattering with the liquid atoms and the exoergic (n,p) and (n,α) reactions in ^{35}Cl [4]. Values of E_c for the reaction products are represented in Fig. 3, which evidences an increased E_c with increasing superheat. The case of ^{16}O was evaluated as a potential contributor from the gel to the detector signal (hydrogen does not reach the LET required for a nucleation). Most curves in Fig. 3 are characterized by a low-T region where E_c is defined by the LET threshold, followed by a sharp transition as the energy threshold becomes sufficiently low to set itself E_c.

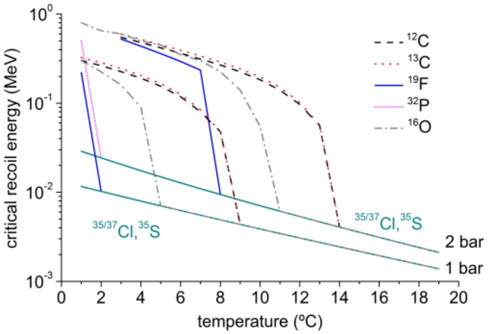

Figure 3. Critical recoil energy (E_c) of recoil atoms in C_2ClF_5 as a function of p and T.

The neutron energy transfer to the recoils, determined by the reaction kinematics equations, is the product of the maximum energy transferred and an angular factor. For elastic scattering, the minimum neutron energy required to induce a nucleation (E_{min}) is directly proportional to E_c and therefore varies with T, p for each atom of the liquid. In contrast, the energy of nuclei emerging from ^{35}Cl(n,p)^{35}S (17 keV) and ^{35}Cl(n,α)^{32}P (104 keV) is always sufficient to provoke an event at T≥2ºC. Finally, the various excited states induced by inelastic scattering were analyzed individually for T≥4 ºC. At p=1 bar all states induce events, except the first excited state of ^{19}F with a small threshold energy (115.84 keV) that always provokes a nucleation if T≥5 ºC. At 2 bar the same generally applies: all emerging states of $^{35/37}$Cl, $^{12/13}$C and ^{19}F contribute beyond 4-7 ºC while T-dependent thresholds apply for the first excited state of ^{19}F up to 11ºC

The energy distributions of reaction rates are plot in Fig. 4, the part above E_{min} corresponding to the fraction of elastic scattering reactions that yields events assuming maximum energy transfer. The sharp features found in the 10-100 keV range correspond to the resonances in the ^{19}F elastic scattering cross section – the largest contributor to the detector signal.

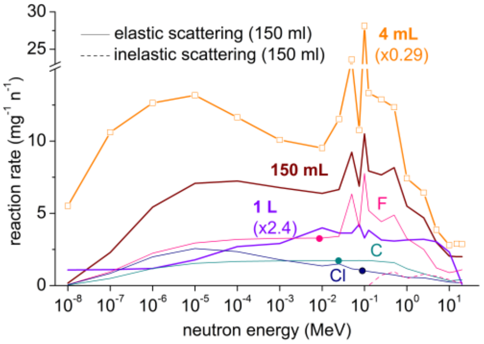

Figure 4. Reaction rate of neutron scattering reactions. The solid circles identify E_{min} for elastic scatterings at 9 ºC, 2 bar.

Recoil distributions

Figure 5 shows calculated energy distribution of recoils, and the E_c corresponding to elastic scattering at 9 ºC and 2 bar (similarly to Fig.4). The inclusion of the angular distribution of recoils is found to reduce the calculated event rate by 10-20% relative to estimates assuming maximum energy transfer. MCNPX-PoliMi and GEANT4 recoil distributions are in good agreement at energies higher than 1-10 keV.

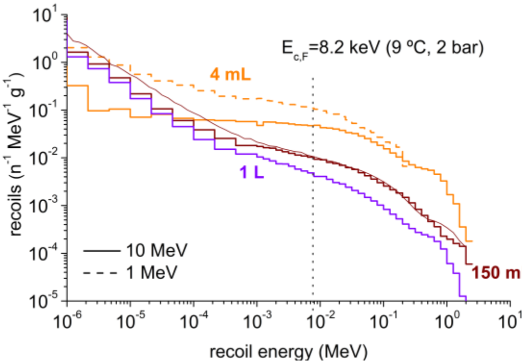

Figure 5. Recoil energy distributions (C+F+Cl). The smooth line plots a GEANT4 calculation (150 mL, 10 MeV).

Response functions

Detector responses as a function of energy (for fixed T and p) and as a function of T (for fixed energy and p) are shown in Fig. 6. The energy dependence corresponds to the convolution of the reaction rate above E_{min} with the recoil distribution (Figs. 4 and 5). The temperature dependence is similar to a threshold response curve, due to the features discussed in Fig. 3: the base level and the two kinks in the response display the contribution of the Cl, F and C recoils.

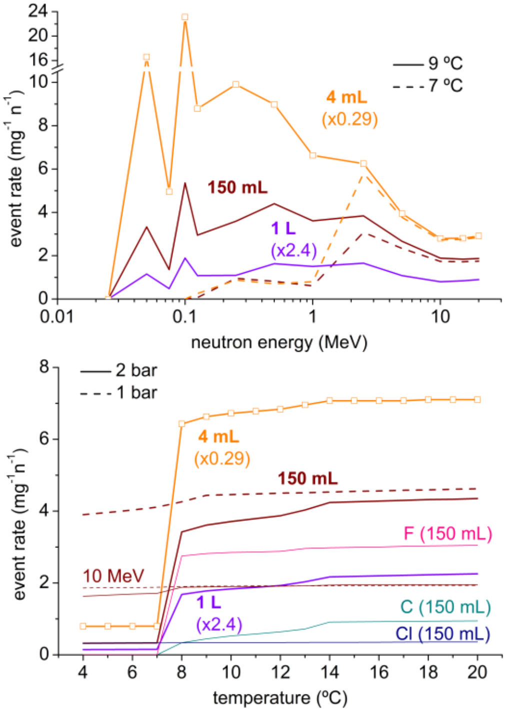

Figure 6. Detector response functions. Top: energy dependence (2 bar). Bottom: T dependence (1 MeV neutrons).

Comparison with measurements

The calculated and measured responses as a function of temperature of a 150 mL SDD at p=2 bar irradiated with Am-Be are shown in Fig. 7. The measurement uncertainties correspond to counting statistics. There is a fast rise in the detector signal as the temperature increases due to the threshold-like character of the response curve and the accounting of lower energy neutrons. The measurements follow generally the calculated distribution curve and the expected count rate. Various factors may justify the discrepancies observed: actual neutron energy, room scattering, counting statistics, disregarded contribution from the gel. Nevertheless the aim of validating the simulation approach has been accomplished.

CONCLUSIONS

The response functions of the various C_2ClF_5 SDDs fabricated by our team were calculated for the first time using Monte Carlo radiation transport simulation. The calculated response functions exhibit the characteristics found by other authors [4]. The response measured as a function of temperature for Am-Be neutrons follows the calculated shape and intensity. GEANT4 provided results in good agreement with those of MCNPX-PoliMi with respect to neutrons of all energies and to recoils beyond 1-10 keV. We look forward for a revised GEANT4 release that accounts properly for the transport of low energy recoils, allowing to model the energy deposition process of the liquid and gel atoms within the micrometric droplets. The improved simulation of the detector response is planned for the near future, followed by benchmarking measurements using monoenergetic neutrons.

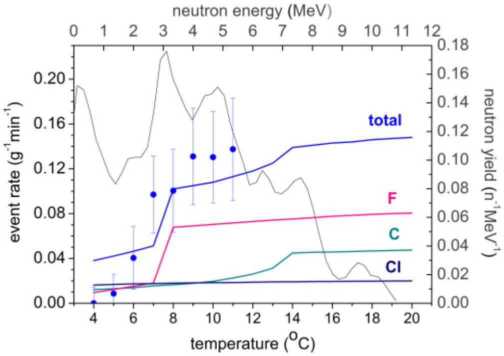

Figure 7. Measured (circles) and calculated (color lines) response as a function of T for Am-Be neutrons (gray line).

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