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# Advancements in Gd-based neutron detection: $\gamma$ - $\gamma$ coincidence approach

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**Abstract** — CEA LIST is working of the development of a high-sensitivity thermal neutron counter, based on the insertion of gadolinium in spherical plastic scintillators. As the radiation sensor comprises one or several volumes of scintillating polymers, it is, by nature, sensitive to both neutron and gamma radiations. This property allows the development of a versatile detector, provided that it is possible to algorithmically separate the signature of both types of radiations. The technologically building brick is, moreover, to be implemented in a transportable (< 15 kg) system, handled by a first intervention task force in an exposed area. The aim of this paper is to study, both through Monte Carlo simulation and experimentally, an algorithmic architecture of the measurement chain discriminating neutron and gamma events. The presentation is subdivided in three main sections. First, we detail a comparative study between the outputs of a numerical modelling tool, developed to simulate the total response of the sensor to varying radiation sources and the results of a series of experiments carried out in a controlled environment. Second, we describe the building and numerical pre-validation of a simulation code dedicated to the study of consolidated algorithmic architecture, namely  $\gamma$ - $\gamma$  quasi-coincidence on two adjacent plastic scintillators. Third, we comment on an experimental study of  $\gamma$ - $\gamma$  quasi-coincidence, Gd-based neutron detection and a comparison between the results and the estimated of the numerical study. We conclude with the presentation of first estimates of neutron sensitivity), gamma-background vulnerability, and  $n/\gamma$  discrimination ratio for the scaled and assessed, material and algorithmic architectures.

**Index Terms** — Neutron counter. Plastic scintillator. Gadolinium. Coincidence filtering.

## I. INTRODUCTION

IN THE FRAME OF THE INTER-MINISTERIAL R&D PROGRAM ON CHEMICAL, BIOLOGICAL, RADIOLOGICAL, NUCLEAR, AND HIGH YIELD EXPLOSIVE (CBRNE) COUNTERMEASURES, French Atomic Energy Commission (CEA LIST) is working of the development of a high-sensitivity thermal neutron counter, based on the insertion of gadolinium (Gd) in spherical plastic scintillators. The concept of the said detector was patented [1] and the first theoretical and experimental studies have shown the deployment potential of the concept, as well as the necessary incremental improvements for the definition of a consolidated technological solution [2]. In the original, modeled, assembled and tested prototype, neutron detection was based on the signature of gamma rays with energy above 5 MeV, emitted in the radiative cascade following the capture of a thermal neutron by a Gd nucleus. This signature is thus isolated *via* a mere pulse height discrimination (PHD). The estimated neutron sensitivity, of the order of 1 cps/(n.cm<sup>-2</sup>.s<sup>-1</sup>), was found compatible with Special Nuclear Material detection issues. The false count rate evolution, however, highlights a vulnerability to cosmic rays (muons) and high-energy gamma rays (fission), which may severely impact neutron detection limits for medium gamma dose rates (10-100  $\mu$ Sv.h<sup>-1</sup>).

As the radiation sensor comprises one or several volumes of scintillating polymers, it is, by nature, sensitive to both neutron and gamma radiations. This property allows the development of a versatile detector, provided that it is possible to algorithmically separate the signature of both types of radiations. Contrary to modified plastic scintillators with neutron/gamma ( $n/\gamma$ ) discriminating properties [3], such a detection ensemble allows the detection of slow neutron capture events that are not preceded by a proton recoil event. The combination of both sensing bricks should lead to wide-spectrum neutron detection, and ultimately, spectral information on the incident neutron radiation. The technological building brick is, moreover, to be implemented in a transportable (< 15 kg) system, handled by a first intervention task force on an exposed area. The aim of this paper is to study, both through Monte Carlo simulation and experimentally, an algorithmic architecture of the measurement chain discriminating neutron and gamma events.

## II. THERMAL NEUTRON CAPTURE BY GD NUCLEI AND STATE OF THE ART

In this section, we briefly recall the main characteristics of <sup>155</sup>Gd( $n,\gamma$ ) and <sup>157</sup>Gd( $n,\gamma$ ) neutron capture reactions. On the basis of the identification of the reaction products of interest, we establish a state of the art of the various material and algorithmic architectures dedicated to the quantification of this signature. Several architectural designs are then proposed, that are to be studied throughout the course of the program.

47 A. High-energy signature of  $^{155}\text{Gd}(n, \gamma)$  and  $^{157}\text{Gd}(n, \gamma)$  neutron capture reactions

48 Neutron capture by Gd-155 and Gd-157 results in the isotopic change of the nucleus, with reaction products freed as nuclear  
49 and atomic structures rearrange: gamma rays, internal conversion electrons, X rays and Auger electrons [4]. This de-excitation  
50 source term, amounting for an average  $8048.2 \pm 0.6$  keV Q-value in natural Gd, is largely dominated by a gamma-ray continuum  
51 characterized by a high multiplicity: about 99 % of the Q-value being carried by an average number of 3 to 5 simultaneously-  
52 emitted photons [5]. The emission spectrum shows a high-energy signature between 2 and 8 MeV, with several intense gamma  
53 rays (6.75 MeV; 7.86 MeV) dominating a continuum. This observation is at the root of the neutron detection strategy we  
54 followed, that consists in isolating the signature, inside the plastic sensors, of the high-energy gamma source term released after  
55  $\text{Gd}(n, \gamma)$  capture reaction. This signature indeed provides access to favorable  $n/\gamma$  signal-to-noise ratio (SNR) above a 2.6 MeV  
56 energy threshold (PHD). The said threshold corresponds to the highest-energy prominent gamma ray (Tl-208) emitted by  
57 radionuclides in the disintegration chains of U-238 and Th-232 of natural radioactivity [6]. Now the quantification of a  
58 significant fraction of this energetic electromagnetic source term requires a high-density or large-volume sensor, and governs the  
59 conception and optimization rules for dedicated detection schemes, whose state of the art is summarily described in the next  
60 paragraph.

61 B. Reference work on large-volume solid neutron detectors based on the gamma-ray signature of Gd captures

62 The previously detailed neutron detection strategy requires the interaction of photons with more than 3 MeV energy. Hence a  
63 scale-up of the radiation sensor must be operated according to a tradeoff between cost and detection efficiency. The latter  
64 consideration is the reason why liquid-or-plastic-scintillator-based technological solutions tend to be favored. As an illustration,  
65 attenuation coefficients for 3-MeV-energy photons [7] show that 50 % absorption yield is expected after 17.5 cm range in  
66 plastic.

67 As far as fieldable detectors are concerned, solid, plastic sensors are preferentially selected as mechanically robust, watertight,  
68 and non-fusible. De Vita *et al.* [8] thus introduced a neutron detection scheme based on  $\text{Gd}(n, \gamma)$  reactions in which  
69  $200 \times 6 \times 6$  cm<sup>3</sup> plastic scintillator (PS) blocks are wrapped in 50- $\mu\text{m}$  Mylar sheets covered with a 25- $\mu\text{m}$  Gd<sub>2</sub>O<sub>3</sub> layer. PS  
70 blocks act as a neutron moderator the fast, incident fission spectrum, and as a sensor for  $\text{Gd}(n, \gamma)$ -reaction gamma rays up to 8  
71 MeV. Neutron/gamma discrimination was based on hit multiplicity, *i.e.* on the number of events registered in several isolated  
72 bars following neutron or gamma-ray diffusion. GEANT4 simulations showed that, by enforcing a hit multiplicity equal to 3 for  
73  $n/\gamma$  discrimination, a rejection ratio of 145 was accessible while maintaining a theoretical neutron detection efficiency of 28 %.  
74 The concept was transposed to Radiation Portal Monitors (RPM), and Fanchini reported [9] a 100-% detection yield in presence  
75 of a moderated  $2 \cdot 10^5 \cdot \text{n} \cdot \text{s}^{-1}$  Cf-252 source a distance of 2.5 m, and a null false alarm rate in presence of a  $20 \cdot \mu\text{Sv} \cdot \text{h}^{-1}$  gamma-ray  
76 dose rate (Cs-137).

77 The development of large-volume PS with heterogeneous Gd-loading was also pursued in the fields of fundamental research  
78 and fixed facility instrumentation. Pawełczak *et al.* [10] described a prototype said “Neutron Sandwich Transmuter/Activation- $\gamma$   
79 Radiator” (NSTAR), whose purpose was to determine the neutron burst multiplicity at the output of D-D fusion neutron  
80 generator. NSTAR is formed by a stack of  $10 \times 20 \times 2$  cm<sup>3</sup> fast-response PS (BC-408, Saint-Gobain), interlined by 1-mm thick,  
81 Gd<sub>2</sub>O<sub>3</sub>-loaded polydimethylsiloxane foils (resulting heterogeneous mass load of 0.5 wt % (Gd). The module performs  $n/\gamma$   
82 discrimination on the basis of a temporal correlation between the signature of proton recoils, associated with the first diffusions  
83 of fast neutrons within the plastic, and the delayed signature of gamma rays released after the capture of the moderated neutron  
84 by a Gd-155 or Gd-157 nucleus. By setting a [7-97  $\mu\text{s}$ ] correlation gate, and rejecting signals with measured energies above  
85 5 MeV-electron equivalent (MeVee), the authors obtained  $n/\gamma$  SNR between  $10^2$  and  $10^3$ .

86 Another noticeable application field relates to the detection of electron antineutrinos *via* neutron emission, following the  
87 interaction of said antineutrinos proton nuclei in a hydrogenous target. In view of this indirect detection, Huroda *et al.* [11]  
88 described a transportable and reconfigurable detector said “Plastic AntiNeutrino Detector Array” (PANDA). The sensor is made  
89 of  $10 \times 10 \times 100$  cm<sup>3</sup> PS bars, wrapped in Gd-coated Mylar films (surface mass density of  $4.9 \text{ mg} \cdot \text{cm}^{-2}$ ), and  
90 concatenated to form a several hundred of kilogram target. The volume and granularity of the target allow neutron/background  
91 discrimination based on a combination of PHD, event multiplicity and temporal correlation approaches. In this algorithmic  
92 architectures, neutrons are detected on the grounds of:

- 93 - a temporal correlation between prompt and delayed ( $\text{Gd}(n, \gamma)$  capture) events lying within a [6-200  $\mu\text{s}$ ] gate;
- 94 - a measured delayed energy, integrated over all Gd-coated PS, between entre 3 and 8 MeVee;
- 95 - additional filters based on first maximal and second maximal energies laid down in separately considered modules.

96 A 360-kg prototype [12] said PANDA36 was assembled and set at sea level, at a distance of 36 m from the 3.4-GWth reactor  
97 of Ohi Power Station (Kansai Electric Power Co., Inc.). Electron antineutrino are indeed produced along the disintegration  
98 chains of U-235 and Pu-239, and thus may be used as a signal for surveilling and monitoring nuclear power plants. The expected  
99 antineutrino-induced count rate was confirmed experimentally by the authors, at the expense of a complexified discrimination  
100 scheme, based on the topology of coincidence events in view of muon rejection.

101 In order to address the issues of very-high-energy (20 MeV-1 GeV) neutron counting and dosimetry, Roecker *et al.* [13]  
102 conceived a 1-ton “Multiplicity and Recoil Spectrometer” (MARS) transportable by hoist. MARS is formed by two mixed PS/Gd  
103  $100 \times 75 \times 25$  cm<sup>3</sup> ensembles, each of them constituted of 12-cm-thick PS interlined with Gd-coated Mylar foils. The counting  
104 of 20-100 MeV neutrons is based on a capture-gated temporal correlation as described in references [9,10]. To detect higher-

energy neutrons, a  $101 \times 71 \times 20 \text{ cm}^3$  Pb plate is inserted between both PS/Gd ensembles, and acts a nuclear spallation target: very fast neutrons the induce lower-energy neutrons, in the order of 1-2 MeV, with multiplicity depending on incident neutron radiation. Secondary neutrons are subsequently diffused in plastic, and undergo Gd( $n, \gamma$ ) capture reaction within mean delay  $19 \pm 3 \text{ }\mu\text{s}$ . GEANT4 simulations led to an estimation of MARS neutron detection efficiency of  $12.8 \pm 0.1 \%$ .

### 109 C. Algorithmic filtering from neutron/background discrimination

110 Abovementioned reference work highlights three types of event filtering, dedicated to the discrimination of neutron-induced  
111 pulses from gamma-ray and muon background:

- 112 - by the measured energy (proportional to the amplitude or area of pulses), above the lower threshold lying between 3 and  
113 5 MeVee, and an upper threshold between 8 and 9 MeVee. This approach is based on the spectral extent of the  
114 continuum deprived of the extent of significant natural gamma background;
- 115 - by the multiplicity of gamma-ray events detected coincidentally. This approach relies on the multiplicity of gamma  
116 emission following reactions, forming a consolidated signature of neutron-induced events. However, this multiple  
117 signature requires a separation between both events;
- 118 - by the temporal correlation between two pulses within a window typically comprised between 10 and 200  $\mu\text{s}$ . This  
119 approach has roots in the fact that some slow neutron captures are preceded by fast neutron diffusion in the PS medium  
120 (recoil of a directly ionizing ion).The following of both events therefore constitutes a consolidated signature of the  
121 interaction history of one neutron within the detection ensemble. As previously, this method requires the separation of  
122 both events, and is only effective when a fast neutron-induced ion recoil is detectable.

123 The first PHD filter was systematically used in the algorithmic architecture of the detectors. Nonetheless, previous studies  
124 [2,8] show that an algorithmic architecture reduced to a sole PHD filter remains vulnerable to background radiations under an  
125 elevated gamma-ray dose rate, typically between 20 to 30  $\mu\text{Sv}\cdot\text{h}^{-1}$ . Thus it appears necessary to enrich the architecture with one  
126 or more filters to increase the SNR. The hereby detailed study concerns a filter based on gamma-ray-event temporal coincidence,  
127 with the purpose of identifying multiple gamma-ray emissions associated with Gd de-excitations. This study relies on both  
128 simulations and laboratory experiments.

## 129 III. STUDY OF A NUMERICAL MODEL FOR MATERIAL DESIGN

130 The conception and optimization of a material architecture for the detector was carried out by means of the MCNP6.1 code  
131 for particle transport [14]. The first step was to verify that valid modelling tools and schemes were available for the delivery of  
132 output data, representing real physical processes inside the sensor and useable for the computation of neutron sensitivity and  
133 detection limits in a mixed  $n/\gamma$  field. We based the validation study on the total response of the radiation sensor and a PHD  
134 detection algorithm to the radioactivity of a lead-shielded, neutron-emitting source (Cf-252). In Monte Carlo simulations, the  
135 output corresponds to an energy deposition yield labeled  $Y$  (expressed in deposition per disintegration,  $\text{d}\cdot\text{dis}^{-1}$ ) between a lower  
136 threshold varying between 2 and 4 MeVee and a 9 MeVee upper threshold. This expected response is then to be compared with  
137 the experimental count rate  $R$  (in count per disintegration,  $\text{c}\cdot\text{dis}^{-1}$ ) integrated between the same thresholds.

### 138 A. Description of the numerical model and simulated response to Cf-252

139 We modeled two  $10 \times 10 \times 10 \text{ cm}^3$  PS commercialized by Eljen Technology, and referenced EJ-200 [15]. As neutron diffusion  
140 strongly depends on molecular organization in dense and hydrogenous media, we used transport data associated with benzene  
141 nuclei at 300 °K. Both SP are positioned head-to-tail, and respective labeled S1 and S2. A Pb shield with same geometry and  
142 dimensions as each PS is placed in front of them, with the plane containing the barycenter of the shield being identical to the one  
143 containing the interface between S1 and S2. Thermal neutron converters are modeled as two natural, 100-% pure Gd. This model  
144 is representative of 250- $\mu\text{m}$  sheets commercialized by Goodfellow [16], and whose purity is certified as superior or equal to  
145 99 %.

146 Californium-252 has a typical spontaneous-fission-type neutron emission spectrum [17]. This neutron spectrum is modeled  
147 with MCNP6.1 by means of a Watt distribution, which associates to every energy  $E_n$  of a fission neutron a value of the emission  
148 density of probability  $f(E_n)$  as

$$149 \quad f(E_n) = C \cdot \exp\left(-\frac{E_n}{a}\right) \cdot \sinh(\sqrt{bE_n}) \quad (1)$$

150 with parameters  $(C; a; b) = (0.3016; 1.025 \text{ MeV}; 2.926 \text{ MeV}^{-1})$ . Spontaneous fissions account for  $\Gamma_f = 0,0309 \text{ f}\cdot\text{dis}^{-1}$   
151 (fission per disintegration) of Cf-252. A sealed Cf-252 sample is a mixed  $n/\gamma$  source, with mean number of emitted neutrons per  
152 fission  $\nu_n = 3.77 \text{ n}\cdot\text{f}^{-1}$ , and mean number of emitted gamma rays per fission  $\nu_\gamma = 7.98 \text{ }\gamma\cdot\text{f}^{-1}$  [18], as kept for this study. The  
153 gamma-ray spectrum accompanying spontaneous fissions is modeled as a piecewise density function [19]. The Cf-252 emitter is  
154 simulated under the form of an isotropic point source, in contact with the Pb shield and aligned with its center of mass.

155 Eight irradiation configurations were simulated, where the number of Gd sheets used (from 0 up to 2) and their disposition are  
156 varied in the setup. Configurations are indexed from 1 to 8, with description in Table I. These series have been defined to  
157 highlight the increase in total response ( $\text{d}\cdot\text{dis}^{-1}$ ) brought by the inversion of Gd. The front face of one PS is the one in contact  
158 with the lead shield.

with Pb, the back face as the one parallel to the front face and farther from the source. Fig. 1a) represents the model of the test bench (Moritz1.23 software was used for visualization [20]).

Neutron and gamma-ray emissions are simulated separately. The macroscopic estimate of the energy laid down by incident gamma rays in S1 and S2 is delivered by tally 8 of MCNP.1: it is labeled  $\tau_\gamma(E_\gamma)$ , and expressed in gamma-ray energy depositions per generated gamma ray and per energy bin. The PS response to the neutron emission of Cf-252 has two components:

- the energy laid down by gamma rays released after radiative capture of moderated neutrons. In series 2 to 8, where Gd converters are added to the sensor, this contribution is filled by the de-excitation source term provided by the MCPN6.1 code-backed, ENDF/B-VII.1 library [21]. The selection of this library was justified by a verification of the faithful estimation of the freed energy [22]. Moreover, in all configuration, S1 and S2 have intrinsic gamma-ray response to moderated neutrons, mainly through  $^1\text{H}(n,\gamma)$  and  $^{12}\text{C}(n,\gamma)$  capture reactions [23]. Prompt gamma rays from these captures are also generated with ENDF/B-VII.0. The sum of all  $(n,\gamma)$  contributions, provided by tally 8, is labeled  $\tau_{(n,\gamma)}(E_\gamma)$  and expressed in gamma-ray energy depositions per generated neutron and per energy bin;
- the energy laid down by recoil nuclei along the diffusion of incident neutrons. Given the  $1000\text{-cm}^3$  volume of both PS, and the neutron emission spectrum (mean energy around 2.3 MeV), its evaluation may be narrowed down to  $^1\text{H}(n,n)$  diffusions in plastic [24]. This contribution is delivered by tally 8 used with tally 6 and PHL option of MCNP6.1. It is labeled  $\tau_{(n,n)}(E_p)$  and expressed in proton energy depositions per generated neutron and per energy bin.

TABLE I. DESCRIPTION OF THE EIGHT SIMULATED CONFIGURATIONS OF IRRADIATION

Series index	Description
1	No Gd sheet
2	1 Gd sheet (250 $\mu\text{m}$ ) between S1 et S2
3	2 Gd sheets (500 $\mu\text{m}$ ) between S1 et S2
4	1 Gd sheet on the front face of S1
5	1 Gd sheet on the front face of S2
6	1 Gd sheet on the front face of S1 1 Gd sheet on the back face of S2
7	1 Gd sheet on the front face of S1 1 Gd sheet on the front face of S2
8	1 Gd sheet on the back face of S1 1 Gd sheet on the front face of S2

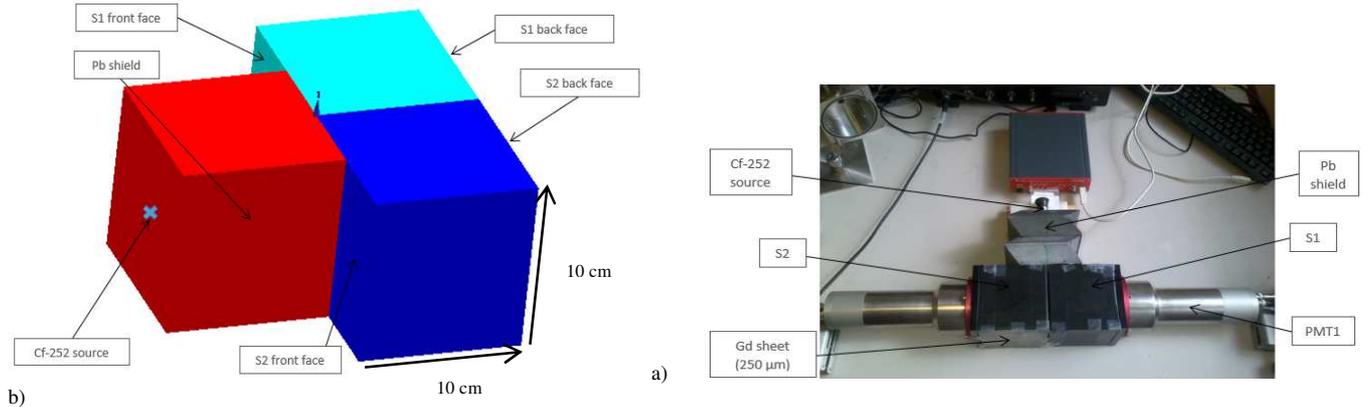


Fig. 1a) 3D-Representation of the bench model under MCNP6.1 (Moritz 1.23), and b) Experimental test bench (CEA LIST).

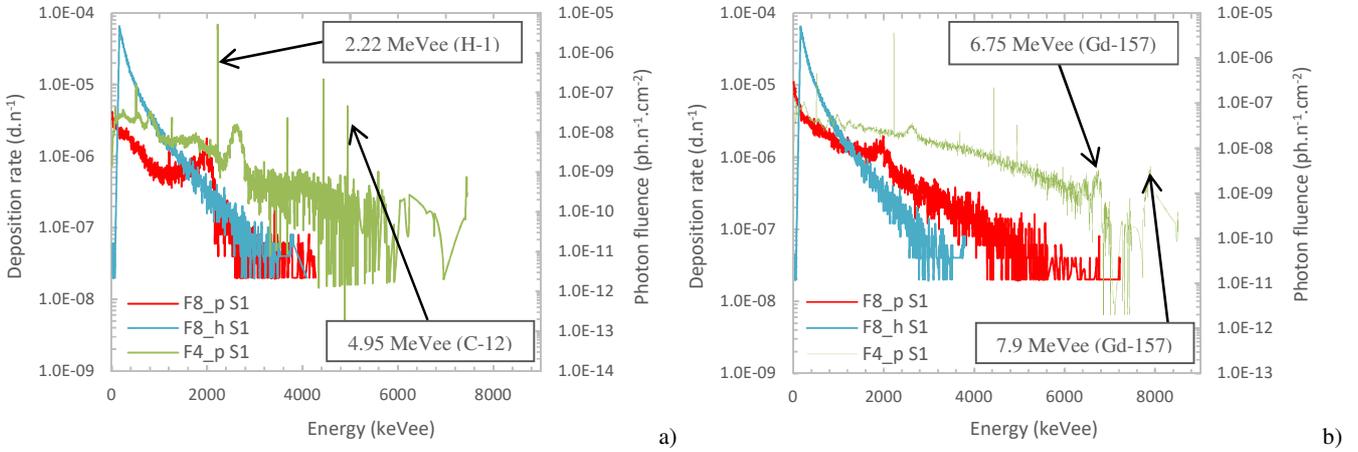
As we aimed at estimating the expected response at the output of a detection chain, it was necessary to convert every deposited energy  $E_\gamma$  or  $E_p$  (MeV) into the photoelectron response of the scintillation detectors, labeled  $E_{ee}$  and expressed in MeVee. This conversion depends on fluorescence quenching for recoil protons and secondary electrons, and is taken into account using the calibration by Pozzi *et al.* [25]. The expected spectral deposition rate per disintegration and per energy bin  $Y(E_{ee})$  is ultimately derived as

$$Y(E_{ee}) = Y_\gamma(E_{ee}) + Y_{(n,\gamma)}(E_{ee}) + Y_{(n,n)}(E_{ee}) = \tau_\gamma(E_{ee}) \cdot \nu_\gamma \cdot \Gamma_f + (\tau_{(n,\gamma)}(E_{ee}) + \tau_{(n,n)}(E_{ee})) \cdot \nu_n \cdot \Gamma_f \quad (2)$$

195 Figures 2a) and b) below present the estimated  $\tau_{(n,\gamma)}(E_{ee})$  (F8\_p) and  $\tau_{(n,n)}(E_{ee})$  (F8\_h) responses for S1 in configurations 1  
 196 (no Gd) and 3 (500  $\mu\text{m}$  of Gd). The comparison between both figures shows, according to Part II, a significant increase in the  
 197 high-energy ( $> 3$  MeVee) component of  $\tau_{(n,\gamma)}$  from series 1 to 3, attributable to Gd( $n,\gamma$ ) captures. This interpretation is  
 198 corroborated by the spectral evolution of photon fluence  $\psi_{(n,\gamma)}(E_{ee})$  in S1, expressed in photon per generated neutron and  
 199 centimeter square ( $\text{ph. n}^{-1} \cdot \text{cm}^{-2}$ ), and estimated by means of tally 4. Rays at 2.22 MeVee from  $^1\text{H}(n,\gamma)$  and 4.95 MeVee from  
 200  $^{12}\text{C}(n,\gamma)$  are clearly identified on both figures. Fluence  $\psi_{(n,\gamma)}(E_{ee})$  associated with series 3 additionally reveals a continuum up  
 201 to the Q-values of  $^{157}\text{Gd}(n,\gamma)$  (7.9 MeVee) and  $^{155}\text{Gd}(n,\gamma)$  (8.5 MeVee) reactions. Thus the input of Gd in the high-energy  
 202 response of the detector is qualitatively established.

203 In view of a quantitative review of simulation results, the full spectral response  $Y(E_{ee})$  for all eight configurations is  
 204 integrated between a varying lower bound  $s \in \{2; 2.5; 3; 4\}$  MeVee and an upper bound set equal to 9 MeVee. The integrated  
 205 response  $Y(s)$  is expressed in  $\text{d. dis}^{-1}$ . Figures 3a) and 3b) illustrate the evolution of  $Y$  as a function of series index from Table I,  
 206 for  $s = 2.5$  MeVee and  $s = 3$  MeVee in S1. All configurations in which a Gd converter is added to the sensor  
 207 have higher  $Y$  response than series 1, whatever the value of  $s$ . The highest expected increase is noted for series 2 and 3, where  
 208 Gd is inserted between S1 and S2, and is more drastic as  $s$  increases: from series 1 to series 3 in S1, for instance,  $Y$  is multiplied  
 209 by a factor 2.5 for  $s = 2$  MeVee, 4 for  $s = 2.5$  MeVee and  $s = 3$  MeVee, and 7 for  $s = 4$  MeVee.

210



211

212

213

214

Fig. 2. Macroscopic estimates from MCNP6.1 for a) series 1, and b) series 3.

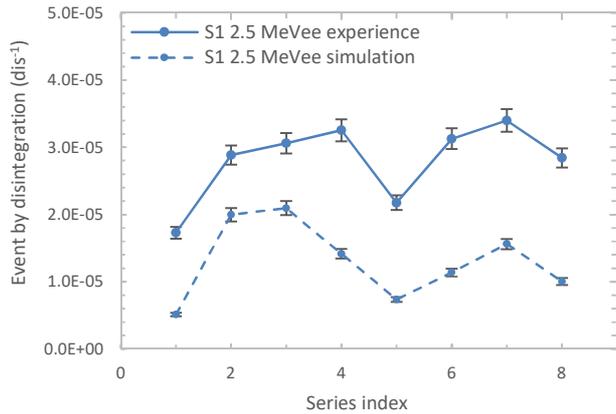
### 215 B. Test bench and experimental response to a shielded Cf-252 source

216 To investigate the modeling scheme presented in paragraph III.A, we reproduced experimentally the irradiation conditions  
 217 detailed above and illustrated in Fig. 1a). The test bench, depicted in Fig. 1b), has two EJ-200 and two photomultiplier tubes  
 218 (PMT) mounted by pairs and isolated from ambient light. PMT are polarized between -1500 V (channel S1) and -1535 V (S2).  
 219 The acquisition chain samples the signal with the frequency of 200 MS.s<sup>-1</sup> on both channels. A calibration coefficient equal to  
 220 76 keVee.channel<sup>-1</sup> was estimated with Co-60, so that the trigger set at channel 7 corresponds to a 531-keVee counting threshold.

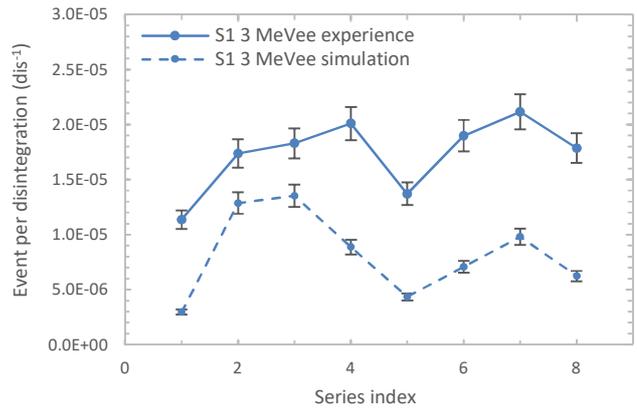
221 Net count rate spectra, labeled  $\lambda(E_{ee})$  and expressed in count per second and per energy bin, are acquired for experimental  
 222 configurations 1 to 8 from Table I. The high-energy background count rate from gamma-ray activity and cosmic muons,  
 223 integrated between a lower bound  $s$  and an upper bound of 9 MeVee, was found to range from  $\lambda(s = 4 \text{ MeVee}) = 3.11 \pm$   
 224  $0.06$  cps to  $\lambda(s = 2 \text{ MeVee}) = 5.80 \pm 0.08$  cps.

225 The mixed emitter was a sealed Cf-252 source of activity  $A = 580 \pm 87$  kBq. Count rate is divided by  $A$  to form a spectral  
 226 distribution  $R(E_{ee})$ , expressed in count per disintegration and per energy bin, which is homogenous to the estimated  $Y(E_{ee})$  from  
 227 paragraph III.A. The experimental spectral response  $R(E_{ee})$  is integrated between  $s \in \{2; 2.5; 3; 4\}$  MeVee and 9 MeVee. The  
 228 integrated response  $R(s)$  is expressed in count per disintegration of Cf-252 ( $\text{c. dis}^{-1}$ ). Figures 3a) and 3b) illustrate the evolution  
 229 of  $R$ , superimposed to  $Y$ , as a function of series index from Table I, for  $s = 2.5$  MeVee and  $s = 3$  MeVee in S1. Error bars  
 230 correspond to one standard deviation in Poisson counting statistics.

231



a)



b)

Fig. 3. Experimental  $R$  and simulated  $Y$  responses of S1 as a function of series index for a)  $s = 2.5$  MeVee, and b)  $s = 3$  MeVee.

### C. Comparative study and first conclusions

Comparison between  $Y$  and  $R$  shows similar trends as series indices are screened, for all threshold values  $s$  and both PS. We can noticeably verify that the lowest response  $R$  always corresponds to configuration 1 (no Gd), in agreement with simulations. The highest experimental increase is noted for series 2, 3, 6 and 7, and lies around a factor of 2 for all considered values of  $s$ . The relative increase of high-energy signal, obtained by insertion of Gd converters, is therefore in the order of our expectations, although by lesser values. This tends to indicate that the relative contribution of  $(n, \gamma)$  to the total response is lower in our experiments than in the idealized model of paragraph III.A.

We thus confirmed experimentally the relative evolution of  $Y$  as the Gd-insertion configuration varies, while highlighting discrepancies that need to be discussed. As the numerical code presented in paragraph III.A is to serve the conception of a PHD discrimination scheme in a composite PS/Gd sensor, we still have to investigate the absolute agreement of responses  $Y$  and  $R$ , at least as far as orders of magnitude are concerned. The most interesting series for the rest of the study are configurations 2 and 3, in which a 250 to 500- $\mu\text{m}$  layer is inserted between S1 and S2. Now, over S1 and S2, relative differences between  $R$  and  $Y$  lie between 47 and 49 % for  $s = 2$  MeVee, 45 and 46 % for  $s = 2.5$  MeVee, and around 35 % for  $s = 3$  MeVee. Relative differences over other series are found below 100 % for  $s = 2$  MeVee, and below 200 % for  $s = 2.5$  MeVee to  $s = 4$  MeVee.

Given the number and the variety of contributions to the high-energy response we listed in paragraph III.A, an experimental corroboration of the expected order of magnitude is a satisfactory result, and confirms the usability of the modelling scheme as a scaling tool for an optimized material design. The comparative study shows, however, that the measured total signal is systematically higher than the expected response, while relative trends appear consistent with numerical estimated. This may be imputed to several factors that non-mutually exclusive:

- simulations do not account for neutron and gamma-ray room return in the environment of the measurement bench, that only tends to increase the number of interaction events in the detection ensemble;
- while simulations describe as finely as possible the material bricks used in experiment, dimension (in the order of 1 mm) and position (in the order of 1 cm) uncertainties are admitted. The same statement is true as far the composition and structure (for PS) of interaction media, not to mention the 15-% one standard-deviation attached to Cf-252 source activity;
- fluorescence quenching afflicting the measured energy from proton recoils may be underestimated, as the phenomenon is scarcely described in literature ;
- limits exist regarding de-excitations models for Gd-156\* and Gd-158\* nuclei [26,27], as already mentioned in Section II.

The following sections will be devoted to the coupling of this model of PHD to the implantation of multiplicity-based temporal filtering.

## IV. BUILDING AND PRE-VALIDATION OF A NUMERICAL CODE FOR THE STUDY OF GAMMA-RAY COINCIDENCE FILTERING

Gamma-ray events in S1 and S2 may be due to background radiation and as well as neutron capture in the matrix and in Gd converters. However, due the emission multiplicity in Gd( $n, \gamma$ ) captures, and provided that the Gd-converter is inserted between adjacent PS, there is a probability that at least one photon will interact in S1 and one photon will interact in S2. In such a scenario, illustrated in Fig. 4a), neutron capture will be followed by two quasi-coincident energy depositions on a both channels. By designing a  $\gamma$ - $\gamma$  coincidence filtering algorithmic architecture, we may thus expect a drastic reduction of high-energy gamma vulnerability and an increase in  $n/\gamma$  discrimination ratio, at the expense, however, of neutron sensitivity. To quantify numerically the impact of such a filtering, we needed to the complete history of every primary and secondary particle inside the detection ensemble.



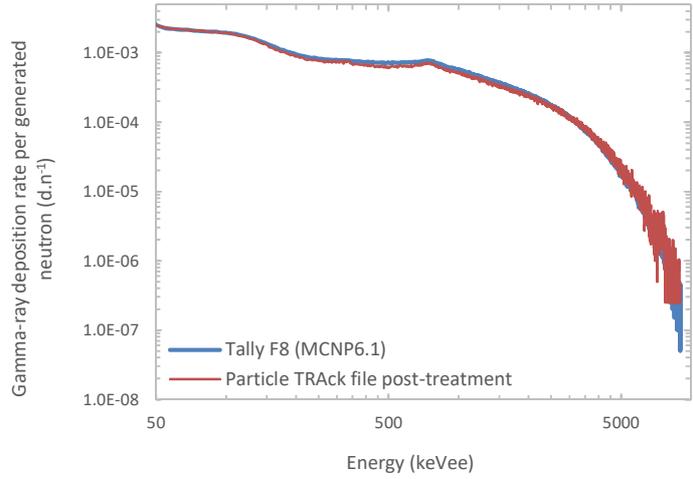
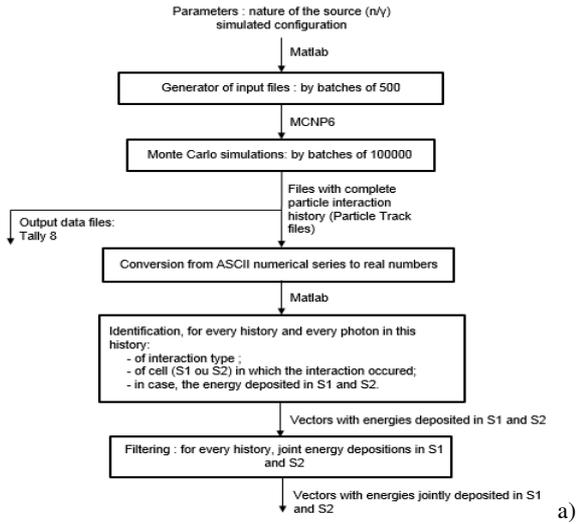


Fig. 5a) Flow chart of Particle Track file post-treatment, and b) Spectral rates  $\tau_{MCNP}(E_{ee})$  and  $\tau_{TRAC}(E_{ee})$  for Test 3.

The quantitative comparison of outputs is based on the relative difference  $\varepsilon_{\%}(s)$  between spectral rates integrated from a varying lower bound  $s \in \{50; 531; 2000\}$  keVee to an upper bound equal to 9 MeVee:

$$\varepsilon_{\%}(s) = 100 \% \cdot \frac{\tau_{PTRAC}(s) - \tau_{MCNP}(s)}{\tau_{MCNP}(s)} = 100 \% \cdot \frac{\int_s^{9 \text{ MeVee}} \tau_{PTRAC}(E_{ee}) dE_{ee} - \int_s^{9 \text{ MeVee}} \tau_{MCNP}(E_{ee}) dE_{ee}}{\int_s^{9 \text{ MeVee}} \tau_{MCNP}(E_{ee}) dE_{ee}} \quad (3)$$

Values of  $\varepsilon_{\%}(s)$  are presented in Table II, and range from 1 to 15 % in absolute. This confirms the accurate reconstruction of unfiltered gamma-ray deposition rates from gamma-ray and neutron sources by means of the numerical post-treatment code developed for this study. We therefore transposed the implementation of the said code to the simulation of expected filtered response  $Y_{\text{filt}}$ , in c. dis<sup>-1</sup>, to a Pb-shielded Cf-252 source.

TABLE II. RELATIVE DIFFERENCES BETWEEN INTEGRATED OUTPUTS  $\tau_{MCNP}(s)$  ET  $\tau_{TRAC}(s)$

Test label \ Integration threshold $s$	50 keVee	531 keVee	2 MeVee
<b>Test 1</b>	- 2.9 %	- 11.1 %	-
<b>Test 2</b>	- 2.2 %	- 2.4 %	14.7 %
<b>Test 3</b>	- 6.3 %	- 6.2 %	1.2 %
<b>Test 4</b>	- 6.2 %	- 9.0 %	- 1.3 %

### C. Estimates of signal evolution in a multiplicity-based gamma-ray event filtering

The entire algorithmic sequence illustrated in Fig. 5a) was implemented in Particle TRAcK file post-treatment. To highlight the benefit from Gd in multiplicity-based detection scheme, we computed the expected filtered spectral response  $Y_{\text{filt}}(E_{ee})$  in configurations 1 (no Gd) and 2 (250- $\mu\text{m}$  converter between S1 and S2) from Table I. As in paragraph III.A, responses gamma-ray and neutron emissions of Cf-252 are obtained separately, and averaged to one disintegration of the radionuclide using Eq. (2). The Pb shield used in experiments is modeled, and the position of the source is identical to the one considered in paragraph III.A.

The multiplicity-filter described in paragraph IV.A is an ideal  $\gamma$ - $\gamma$  coincidence filter. It follows that  $Y_{(n,n'),\text{filt}}(E_{ee}) = 0$  by construction. In simulated configuration, contribution from  $Y_{\gamma,\text{filt}}(E_{ee})$  are also negligible. The comparison between unfiltered  $Y(E_{ee})$  and filtered  $Y_{\text{filt}}(E_{ee})$  spectral responses thus essentially informs us on the reduction of post-filtering neutron signal  $Y_{n,\text{filt}}(E_{ee})$ . Integrated responses  $Y(s)$  et  $Y_{\text{filt}}(s)$ , between  $s = 531$  keVee and 9 MeVee, and expressed in energy deposition per disintegration of Cf-252 (d.dis<sup>-1</sup>), are presented in Table III below.

TABLE III. SIMULATED (IN D.DIS<sup>-1</sup>) AND EXPERIMENTAL (IN C.DIS<sup>-1</sup>) RESPONSES INTEGRATED FROM 531 KEVEE

Series index \ Response	$Y(531 \text{ keVee})$	$Y_{\text{filt}}(531 \text{ keVee})$	$R(531 \text{ keVee})$	$R_{\text{filt}}(531 \text{ keVee})$
<b>Series 1</b>	$(3.05 \pm 0.03) \cdot 10^{-4}$	$(5.13 \pm 0.77) \cdot 10^{-6}$	$(3.00 \pm 0.03) \cdot 10^{-4}$	$(1.7 \pm 0.2) \cdot 10^{-6}$

Series 2	$(4.20 \pm 0.04) \cdot 10^{-4}$	$(2.75 \pm 0.41) \cdot 10^{-5}$	$(3.57 \pm 0.03) \cdot 10^{-4}$	$(3.3 \pm 0.3) \cdot 10^{-5}$
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Two major conclusions may be drawn from the simulation-based estimates consigned in Table III:

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- Comparing  $Y_{\text{filt}}$  from series 1 to series 2, the adding of a central Gd layer leads to an increase of neutron response by a factor of 60. It thus appears necessary to use multiple-gamma-ray-emitter Gd nuclei after neutron capture to consolidate the detection scheme *via* a  $\gamma$ - $\gamma$  coincidence filtering method;

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- In presence of a 250- $\mu\text{m}$  Gd converter (series 2), the filtered neutron response  $Y_{\text{filt}}$  is found reduced by a factor of 15 when compared to the unfiltered response  $Y$ .

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We then estimated a neutron sensitivity reduction by approximately one order of magnitude in an ideal  $\gamma$ - $\gamma$  coincidence filtering. The benefit of such a discrimination scheme as far as gamma-ray vulnerability and detection limits will be studied experimentally in the following section.

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## V. EXPERIMENTAL STUDY OF N/ $\Gamma$ DISCRIMINATION BY QUASI-COINCIDENCE FILTERING

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An experimental study reproducing the irradiation configurations 1 and 2 described in Section IV was conducted in order to verify the one-order-of-magnitude neutron signal reduction and the impact on background-induced false counting *via* a multiplicity-based event discrimination. Given the sampling frequency of 200 MS.s<sup>-1</sup> of the pulse acquisition chain, the n/ $\gamma$  discrimination scheme was implemented as a quasi-coincidence-gated filter with a temporal window of 5 ns between channels 1 and 2. Pulse processing returns a filtered count rate  $\lambda_{\text{filt}}(s)$  (in c.s<sup>-1</sup>) integrated between the trigger threshold  $s = 531$  keVee on both channels and the upper threshold of 9 MeVee. The test bench is otherwise identical to the one described in paragraph III.B and depicted in Fig. 1b).

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### A. Experimental results and comparison with simulations

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We considered irradiation configurations 1 and 2 of Table I, with the same Pb-shielded Cf-252 source as used in paragraph III.B. For the sake of investigating of the merits of quasi-coincidence filtering, both filtered  $\lambda_{\text{filt}}(s)$  and unfiltered  $\lambda(s)$  count rates were averaged to one disintegration of radionuclide using the activity of the source. The obtained experimental responses  $R(s)$  (no filtering) and  $R_{\text{filt}}(s)$  (5-ns quasi-coincidence filtering), in c. dis<sup>-1</sup>, are presented in Table III supra to allow a quantitative comparison with simulation outputs.

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Responses  $Y$  and  $R$  stand in in good absolute agreement, with relative differences between experience and simulation about 1.7 % for series 1 and 18 % for series 2, which goes to the validation of the PHD modeling scheme argued in paragraph III.C. Filtered responses  $Y_{\text{filt}}$  and  $R_{\text{filt}}$  are also found compatible, with -17 % relative difference between experience and simulation for series 2, and same order of magnitude over series 1. Furthermore, both key conclusions from paragraph IV.C are experimentally corroborated. First, we verified the insertion of a Gd-converter has a drastic impact on detection with a quasi-coincidence filter,  $R_{\text{filt}}$  being multiplied by a factor of 20 from series 1 to series 2 (against 60 in simulations). Second, as far as series 2 is concerned, the ratio  $\frac{R}{R_{\text{filt}}} = 9$  confirms the one-order-of magnitude reduction of neutron response when implementing a quasi-coincidence filtering method (to be compared with  $\frac{Y}{Y_{\text{filt}}} = 15$  in simulations).

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The next point of the experimental study regards the impact of quasi-coincidence filtering on gamma- and cosmic-ray background reduction. We selected configuration 2 from Table I for this assessment. First acquisitions were carried out in the absence of radiation source, the dose rate at the detector level being then measured as equal to 75 nSv.h<sup>-1</sup> with a commercial Thermo Electron ESM FH 40G-L 10 radiameter. Integrated count rates between 531 keVee and 9 MeVee read as  $\lambda(531 \text{ keVee}) = 35,9 \pm 0,2 \text{ c. s}^{-1}$  without filter and  $\lambda_{\text{filt}}(531 \text{ keVee}) = 0$  with quasi-coincidence filter. Next acquisitions were performed in presence of an unshielded Na-22 source in front of the Gd/PS detection ensemble, inducing a 14- $\mu\text{Sv.h}^{-1}$ -dose rate at the detector level. Integrated unfiltered and filtered count rates then read as  $\lambda(531 \text{ keVee}) = 7989,5 \pm 40,0 \text{ c. s}^{-1}$  and  $\lambda_{\text{filt}}(531 \text{ keVee}) = 3 \pm 0,8 \text{ c. s}^{-1}$ . Therefore, cancellation of natural background and reduction of Na-22 background rate by a factor of 2700 were observed with the implementation of the filter, emphasizing the consolidation of the algorithmic architecture with regards to background radiations.

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### B. Estimates of figures of merits

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The general agreement between simulated and experimental responses from Table III allows us, in first approach, to discuss the values of  $R_{\text{filt}}$  in light of the conclusions drawn in the end of Section IV.

Observations from paragraph IV.B highlight that filtered response  $Y_{\text{filt}}$ , and therefore its consistent counterpart  $R_{\text{filt}}$ , may be identified to the detector response to the sole neutron component of Cf-252 emission. We may thus deduce from  $R_{\text{filt}}$  a first estimate of neutron sensitivity, labeled  $\widehat{S}_n(s)$ , of the multiplicity-based counting scheme (hereby the detection threshold  $s = 531$  keVee on channels 1 and 2). Neutron dose rate at the detector level is measured as  $\Delta_n = 10 \mu\text{Sv.h}^{-1}$  by means of a calibrated, commercial neutron probe reference LB 6411 (BERTHOLD FRANCE SAS). Using Cf-252 source activity  $A$ , the neutron sensitivity estimated in configuration 2 reads a

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$$\widehat{S}_n(531 \text{ keVee}) = \frac{R_{\text{filt}}(531 \text{ keVee}) \cdot A}{\Delta_n} \sim 7 \text{ c. nSv}^{-1} \quad (4)$$

402 The obtained estimate is quite high, in the 1-c.nSv<sup>-1</sup> order of magnitude of commercial, He-3 proportional counter  
 403 sensitivity [29].

404 We recall that this estimate of  $\widehat{S}_n$  is based on the assumption that gamma-ray contribution from Cf-252 to  $R_{\text{filt}}(531 \text{ keVee})$  is  
 405 negligible. Let alone the absolute agreement between  $Y_{\text{filt}}(531 \text{ keVee})$  and  $R_{\text{filt}}(531 \text{ keVee})$ , this assumption is backed by the  
 406 measurement of Cf-252-induced gamma-ray dose rate at the detector level in configuration 2, found equal to  $\Delta_\gamma = 2 \mu\text{Sv} \cdot \text{h}^{-1}$ .  
 407 Now, using the Na-22 measurement described at the end of paragraph V.A, it is possible to estimate the neutron detection  
 408 scheme vulnerability to a high-energy (1274.5 keV) gamma-ray background, labeled  $\widehat{V}_\gamma(s)$ , as  
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$$411 \quad \widehat{V}_\gamma(531 \text{ keVee}) = \frac{\lambda_{\text{filt},\gamma}(531 \text{ keVee})}{\Delta_\gamma} \sim 0.8 \text{ c.nSv}^{-1} \quad (5)$$

412 Using this estimate, we may derive an expected contribution from Cf-252 gamma-ray background to experimental response in  
 413 configuration 2:  $\widehat{R}_{\text{filt},\gamma}(531 \text{ keVee}) = 7 \cdot 10^{-7} \text{ c. dis}^{-1}$ , hence a relative contribution from false neutron counting  
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$$416 \quad \frac{\widehat{R}_{\text{filt},\gamma}(531 \text{ keVee})}{R_{\text{filt}}(531 \text{ keVee})} = \frac{7 \cdot 10^{-7}}{3,3 \cdot 10^{-5}} \sim 2 \% \quad (6)$$

417 which is indeed negligible. The main bias attached to this verification lies within the wide spectral extent of prompt gamma-  
 418 ray emission from Cf-252, notably above 1.5 MeV. Now penetrating power increases with gamma-ray energy, meaning that for  
 419 the same dose rate, a higher-energy source would be likely to induce a superior true quasi-coincident response, and therefore  
 420 higher gamma-ray vulnerability  $\widehat{V}_\gamma$ . This point is however to be tempered by the fact that mean energy of Cf-252 prompt gamma-  
 421 ray [30] is calculated as  $\overline{E}_\gamma = 875 \text{ keV}$ , and consequently far below to the energy of Na-22 prominent gamma-ray line. The  
 422 evolution of figure  $\widehat{V}_\gamma$ , for a given dose rate, a function of gamma-ray energy of emission, will be studied in future word, notably  
 423 as far as higher-energy emitters, such as Th-232, are involved.  
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425 Experimental quasi-coincidence filtered response over S1 and S2, above a PHD threshold set equal to 531 keVee, led to a  
 426 one-order-of-magnitude loss in neutron sensitivity, and a 1700-factor of gamma-ray background count rate when compared to  
 427 unfiltered response above the same PHD threshold. Estimates of neutron detection limit, labeled  $\widehat{DL}_n$  and expressed in neutron  
 428 per centimeter square and per second ( $\text{n} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}$ ), are conventionally [31] derived as a function of acquisition time  $t$  and  
 429 gamma-ray dose rate  $\Delta_\gamma$   
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$$432 \quad \widehat{DL}_{\text{filt},n}(\Delta_\gamma, t) = \frac{2,71 + 4,61 \sqrt{\lambda_{\text{filt},\gamma}(\Delta_\gamma) \cdot t}}{\widehat{S}_n \cdot t} \quad (7)$$

433 From Eq. (7), it follows that quasi-coincidence filtering leads a lowering of detection limit by a factor of 5 from the unfiltered  
 434 scheme in configuration 2, with implemented temporal gate and integration thresholds. From the prospective of  $n/\gamma$   
 435 discrimination ratio, estimated rates result in SNR around 150, which is close to the rejection ratio of 145 reported by De Vita *et*  
 436 *al.* [8], in the frame of another multiplicity-based discrimination architecture.

437 The estimates of key factors of merit attached to the assessed, material and algorithmic architectures are summarized in  
 438 Table IV.  
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TABLE IV. SETUP CHARACTERISTICS AND ESTIMATES OF KEY FIGURES OF MERIT

Material architecture	Algorithmic architecture	Neutron sensitivity estimate	Gamma-ray background vulnerability	$n/\gamma$ discrimination ratio estimate
Two PS EJ-200 10 × 10 × 10 cm <sup>3</sup> 250-μm Gd-nat between S1 et S2	Energy threshold: 531 keVee Quasi-coincidence filtering over S1 et S2 ( $\Delta t = 5 \text{ ns}$ )	7 c.nSv <sup>-1</sup> (Cf-252)	0.8 c.nSv <sup>-1</sup> (Na-22, 14 μSv.h <sup>-1</sup> )	150 (Cf-252/Na-22)

## 442 VI. CONCLUSION

443 A numerical model of the material bricks forming the detection ensemble (PS, Gd, shields), as well as of two  $n/\gamma$   
 444 discrimination algorithmic architectures, was developed by means of the MCNP6.1. The post-treatment of output data allowed  
 445 us to reconstruct the total spectral responses of both PS, as well as the responses associated with  $\gamma$ - $\gamma$  coincidence filtering of  
 446 multiple-gamma-ray events following Gd( $n, \gamma$ ) captures (Particle TRAcK files). A complete test bench, and an electronic chain

dedicated to pulse acquisition and treatment was used to calibrate our numerical models of detection schemes, to interpretate relative trends and absolute orders-of-magnitude of neutron responses.

While neutron sensitivity was estimated close to  $10 \text{ c.nSv}^{-1}$ , thus matching the gold standard of He-3 proportional counters, we derived, for our first setup and quasi-coincidence scheme, a gamma-ray vulnerability around  $1 \text{ c.nSv}^{-1}$ . It follows that the estimated  $n/\gamma$  discrimination ratio of 150 was found inferior, by one order-of-magnitude, to the  $10^3$ -figure of merit of He-3 counters. Furthermore, the latter ratio is consolidated up to  $1\text{-mSv.h}^{-1}$  whereas the experiments described in this paper were limited to a  $14\text{-}\mu\text{Sv.h}^{-1}$  gamma-ray dose rate, *i.e.* almost two orders of magnitude below. In order to consolidate further the discrimination scheme, we will study, both by simulation and experimentally, the sensitivity and vulnerability of the PS/Gd ensemble when conserving quasi-coincidence filtering (5-ns gate), but elevating the PHD threshold on channels 1 and 2 above 500 keVee. Moreover, the experimental validation of numerical models, both in the case of standard and expert use of MCNP6.1 output files, will allow us to extract rules for an optimized material design, closer to the ideal spherical geometry introduced in previous work

Eventually, alternative approaches such as a gamma-proton temporal correlation, within a 10 to 100- $\mu\text{s}$  gate, and their articulation with  $\gamma\text{-}\gamma$  coincidence, will be investigated in future work.

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