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1 Investigation of CLYC-6 for thermal neutron detection and CLYC-7 for fast neutron spectrometry

2

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5 Abstract

6 Highly enriched ⁶Li and ⁷Li Cs₂LiYCl₆ (CLYC) scintillators were investigated in view of their
7 application as thermal neutron counter (CLYC-6) and as fast neutron spectrometer (CLYC-7) in the
8 recently developed B-RAD radiation survey meter, an instrument designed to work in regions of
9 high magnetic fields, currently equipped with a probe for photon dose rate measurements and γ -
10 spectrometry. Both crystals were irradiated with a 2.5 MeV mono-energetic neutron beam from a
11 Deuterium-Deuterium (D-D) generator and characterised in terms of neutron/ γ -ray (n/ γ)
12 discrimination capability by the Pulse Shape Discrimination (PSD) method, energy resolution and
13 quenching factor. Both crystals exhibit an excellent n/ γ discrimination, quantified by a Figure of
14 Merit higher than 2. The measured thermal energy resolution of CLYC-6 is 6%. The quenching
15 factor for the ⁶Li(n,t) α thermal neutron reaction is 0.65 and for the ³⁵Cl(n,p)³⁵S reaction is 0.91. The
16 measured neutron detection efficiency per unit volume of CLYC-6 is 60% higher than that of a ³He
17 proportional counter. CLYC-7 was tested as a fast neutron spectrometer below 10 MeV, when
18 irradiated with continuum neutron spectra from Am-Be and ²⁵²Cf sources. A proton/ α -particle (p/ α)
19 discrimination was performed exploiting the PSD technique, without any unfolding procedure. The
20 resulting spectra match the corresponding ISO spectra better than the spectra without the p/ α
21 discrimination, especially in the range 3 MeV to 5 MeV. A more sensitive PSD algorithm might
22 help to improve particle discrimination at lower energies, while different reaction channels arise at
23 higher energies preventing a spectrometry analysis.

24 **Keywords**– CLYC-6, CLYC-7, fast neutron spectrometry, thermal neutron counter, Pulse Shape
25 Discrimination.

26 1. Introduction

27 Cs₂LiYCl₆:Ce (CLYC) is a promising inorganic scintillator from the elpasolite family. It is
28 characterised by high density (3.3 g cm⁻³), high Z_{eff} (54), light yield linearity down to low energies
29 (the light yield variation is 1.6% from 662 keV down to 14.4 keV) and good energy resolution (5%
30 at 662 keV) [1]. It can detect both γ -ray and neutron radiation with excellent neutron/ γ -ray (n/ γ)
31 discrimination capability by the Pulse Shape Discrimination (PSD) technique [2].

32 Two CLYC crystals were considered in this work, one enriched in ⁶Li and one enriched in ⁷Li. We
33 will refer to them as CLYC-6 and CLYC-7 respectively. The former is suitable for thermal neutron
34 detection via the ⁶Li(n,t) α reaction [3]. The latter is commonly used for fast neutron measurements
35 through the ³⁵Cl(n,p)³⁵S and ³⁵Cl(n, α)³²P reactions below \sim 8 MeV [4]. Between 8 MeV and
36 18 MeV, the cross section of the (n,p) and (n, α) reactions to the ground and excited states of ³⁵S and
37 ³²P, respectively, are comparable and the different reactions cannot be distinguished [4]. Above
38 \sim 20 MeV the cross sections of the (n,t) and (n,d) reactions become also comparable [5]. CLYC-7
39 was also recently investigated [6–8] as a fast neutron spectrometer without unfolding below about
40 10 MeV, since the energy deposited by the emitted proton and α -particle is linearly proportional to

41 the kinetic energy of the impinging neutron. A recent paper has evaluated the capabilities of CLYC-
42 6 and CLYC-7 for the detection and characterization of Special Nuclear Materials (SNM) [9].

43 In this work CLYC-6 and CLYC-7 crystals were characterised for application as a thermal neutron
44 counter and as a fast neutron spectrometer, respectively, in view of their possible integration in the
45 novel B-RAD radiation survey meter. The B-RAD is novel handheld instrument jointly developed
46 and patented by CERN and the Polytechnic of Milan [10,11] and now commercially available from
47 ELSE Nuclear [12], specifically designed to work in regions of high magnetic fields. It consists of a
48 LaBr₃ crystal coupled with a Silicon photomultiplier array for photon dose rate measurements and
49 γ -spectrometry. Additional probes are under development and CLYC has been selected as a
50 potential candidate for neutron detection.

51 In particular, the n/γ discrimination capability, the energy resolution and the quenching factor for
52 the thermal neutrons produced by the ${}^6\text{Li}(n,t)\alpha$ reaction in CLYC-6 were measured with a mono-
53 energetic neutron beam from a Deuterium-Deuterium (D-D) generator. For CLYC-7, the n/γ
54 discrimination and the quenching factor of the ${}^{35}\text{Cl}(n,p){}^{35}\text{S}$ reaction were measured with the same
55 neutron beam. The fast neutron spectrometric capability of CLYC-7 was also investigated with the
56 continuum neutron spectra from Am-Be and ${}^{252}\text{Cf}$ sources.

57 In [4], the response of CLYC-7 to monoenergetic neutrons showed promising results as a
58 spectrometer, since the energy peak coming from the ${}^{35}\text{Cl}(n,p){}^{35}\text{S}$ reaction was still detected up to
59 8 MeV. However, above 2.5 MeV a second peak due to the (n,α) reaction is present [4], and the
60 spectrum degenerates in a continuum above 5.5 MeV. Therefore, as already underlined in [13], fast
61 neutron spectrometry with CLYC-7 in presence of a continuum spectrum is quite difficult already
62 above 2.5 MeV. Figure 1 shows the cross sections of the main reactions occurring in CLYC-7
63 below 14 MeV. The plot shows that the energy threshold for the (n,α) reaction and for the (n,p)
64 reactions to the excited states of ${}^{35}\text{S}$ is around 2 MeV. Above 4 MeV the cross sections are not
65 negligible, *i.e.*, neutrons with the same energy undergo competing nuclear reactions and the
66 univocal correlation between the initial neutron energy and the energy deposited by the emitted
67 particle is lost.

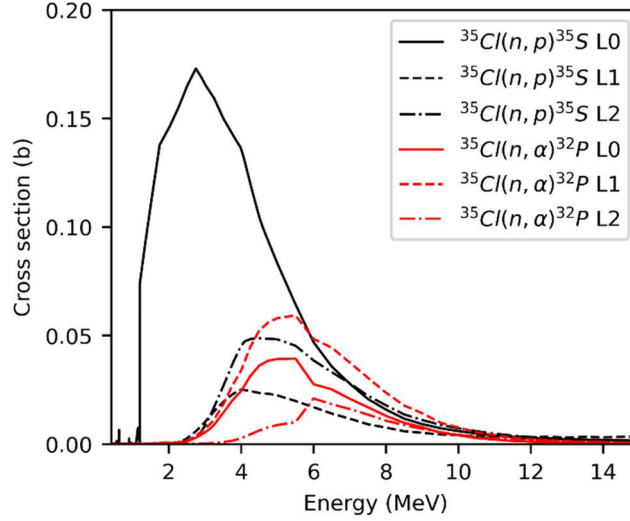
68 In this work we exploited the different Linear Energy Transfer (LET) of protons and α -particles to
69 perform proton/ α -particle (p/α) discrimination via the PSD method. A preliminary analysis was
70 made with Am-Be and ${}^{252}\text{Cf}$ sources demonstrating the feasibility of the analysis at least up to
71 5 MeV. However, due to the small difference of their LET, the proton and α -particle distributions in
72 the 2D PSD plot are not completely separated. This limitation can be overcome with a more
73 sophisticated PSD analysis and/or specifically designed electronics (e.g. shaper or analog PSD)
74 and/or a faster photodetector.

75 On the other side, CLYC-6 cannot be employed as a spectrometer because the acquired neutron
76 spectrum is usually dominated by a narrow peak due to the ${}^6\text{Li}(n,t)\alpha$ reaction from thermal neutrons.
77 However, its large cross section makes it suitable for thermal neutron detection and an appealing
78 alternative to the commonly used ${}^3\text{He}$ proportional counter, especially because of the diminishing
79 stockpile of ${}^3\text{He}$ gas [14].

80

81

82



83

84 *Figure 1. The (n,p) and (n,α) reaction cross sections on ^{35}Cl , where L0 stands for the ground state*
 85 *and L1 and L2 for the first and second excited states. Data are taken from the ENDF/B VIII.0*
 86 *library [15].*

87 2. Materials and methods

88 Both CLYC-6 and CLYC-7 crystals were grown by Radiation Monitoring Devices Inc. (RMD)
 89 [16]. The former (serial number 730-3D1, April 2020) is a parallelepiped with dimensions of
 90 $18 \times 18 \times 5 \text{ mm}^3$, enriched in ^6Li to $> 95\%$. The latter (serial number 039-2, September 2017) is a 1–
 91 inch right cylinder enriched in ^7Li to $> 99\%$. Both crystals were wrapped with a reflective inner
 92 coating and encapsulated in an aluminium casing. The energy resolution at 662 keV declared by the
 93 manufacturer is $< 5\%$ FWHM for both crystals.

94 Each crystal was coupled with a R6231-100 Superbialkali Hamamatsu Photomultiplier Tube (PMT)
 95 [17] operating at 1,100 V through optical grease, and placed inside a light-tight box. The output
 96 signal was directly sent to a 1 GHz, up to 2.5 Gs/s, 12 bit HDO6104 Teledyne Lecroy Oscilloscope,
 97 by a 50Ω termination. The sampling frequency in this work was set to 250 Ms/s. Each signal was
 98 saved and offline analysed with a Python code [18]. For the measurement of the neutron detection
 99 efficiency of CLYC-6 (section 3.4) a CAEN DT5720 digitizer (12 bit, 250 Ms/s) was used in place
 100 of the oscilloscope, because it allows faster signal processing and is provided with the DPP-PSD
 101 software for on-line PSD analysis (which was used instead of the Python analysis).

102 Before each measurement, the experimental set-up was energy calibrated with γ -ray sources of
 103 different energies (^{137}Cs and either ^{60}Co or ^{22}Na). The charge spectrum of each γ -ray source was
 104 acquired by calculating the charge of each signal as the pulse integral over the first $8 \mu\text{s}$. The charge
 105 to energy equation was calculated by linear fitting of the data points corresponding to each
 106 photoelectric peak.

107 In order to discriminate neutrons from γ -ray events, the PSD analysis was performed by means of
 108 the Charge Integration Method [19,20]. Two different time windows were selected, the first from 0
 109 s to $t = t_{\text{prompt}}$ and the second from t_{prompt} to $t = t_{\text{delay}}$, where t_{prompt} and t_{delay} are two selected times of
 110 the pulse tail. The integrals of the signals along the corresponding intervals were calculated and
 111 labelled as Q_{prompt} and Q_{delay} respectively. The PSD was calculated as the ratio $Q_{\text{delay}} / (Q_{\text{prompt}} +$
 112 $Q_{\text{delay}})$. Because of the different time profiles of the pulses generated by neutrons and γ -rays, the
 113 PSD histogram plot defines two Gaussian distributions ascribed to the two kinds of radiation. Each

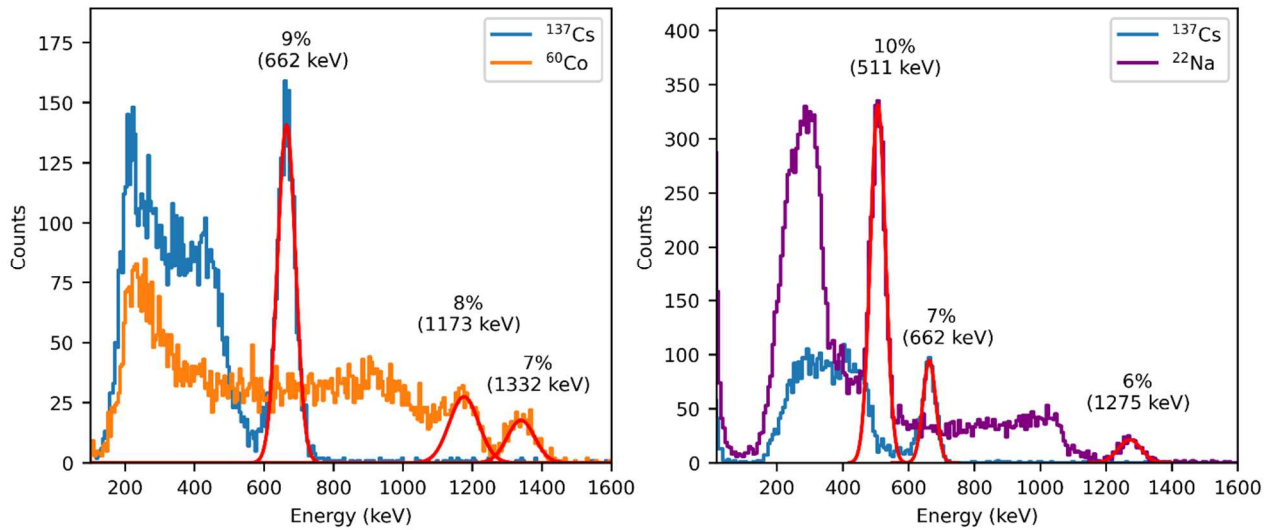
114 distribution was fitted using a Gaussian equation and a Figure-of-Merit (FOM) was calculated to
115 evaluate the goodness of the PSD. The FOM was defined as follows:

$$116 \quad \text{FOM} = \frac{x_n - x_\gamma}{FWHM_n + FWHM_\gamma} \quad (1)$$

117 where x_γ and x_n are the peak centroids of the photon and neutron Gaussian distributions and
118 $FWHM_\gamma$ and $FWHM_n$ the corresponding full widths at half maximum. The n/γ PSD was performed
119 by always setting the time windows t_{prompt} and t_{delay} to 70 ns and 2 μs respectively, unless otherwise
120 specified in the corresponding section. The values of t_{prompt} and t_{delay} were selected as the time
121 windows that optimise the FOM.

122 A Thermo Scientific MP 320 Deuterium-Deuterium (D-D) generator [21] emitting 2.5 MeV
123 neutrons was used for the measurements of the n/γ discrimination, energy resolution and quenching
124 factor. The neutron energy resolution of CLYC-7 was not calculated because of a sudden
125 degradation of the crystal, which only concerns the measurements with the D-D generator (section
126 4.1 and 4.2). All other measurements (with the Am-Be and ^{252}Cf sources) were performed before
127 the worsening of the crystal's performance. As an example, the energy calibrated spectra of ^{137}Cs
128 and ^{60}Co and the corresponding energy resolutions, acquired simultaneously with the measurements
129 described in section 4.4 are shown in figure 2 (left). For completeness, figure 2 (right) shows the
130 ^{137}Cs and ^{22}Na spectra and the corresponding energy resolutions obtained with CLYC-6
131 simultaneously with the measurements described in section 3.1. The energy resolution is worse than
132 the reference one (9% for CLYC-7 and 7% for CLYC-6 at 662 keV versus < 5% declared by the
133 company). The energy calibration applied in figure 2 was calculated by integrating the signals over
134 2 μs, selected as the integration time between 500 ns and 8 μs that optimises the energy resolution.
135 The energy calibration used for the calculation of the neutron spectra was calculated instead by
136 setting the same integration time for both the γ-ray and the neutron sources. The reason of the worse
137 energy resolution might be the digital electronics, an analog shaping electronics (*e.g.* preamplifier,
138 amplifier and multi-channel analyser as suggested by the company) might improve the energy
139 resolution by 2% [5]. In addition, we cannot exclude a degradation of the performance of the PMT,
140 and for CLYC-7 of the crystal itself and/or of his housing (CLYC is highly hygroscopic) since they
141 were purchased three years before these measurements and used for several past tests. The neutron
142 detection efficiency of CLYC-6 was measured with the thermal neutron field available at the
143 Calibration Laboratory (Cal Lab) of CERN Radiation Protection group [22]. The study of CLYC-7
144 as a fast neutron spectrometer was performed by irradiating the crystal with neutrons from both an
145 Am-Be source and a ^{252}Cf source.

146



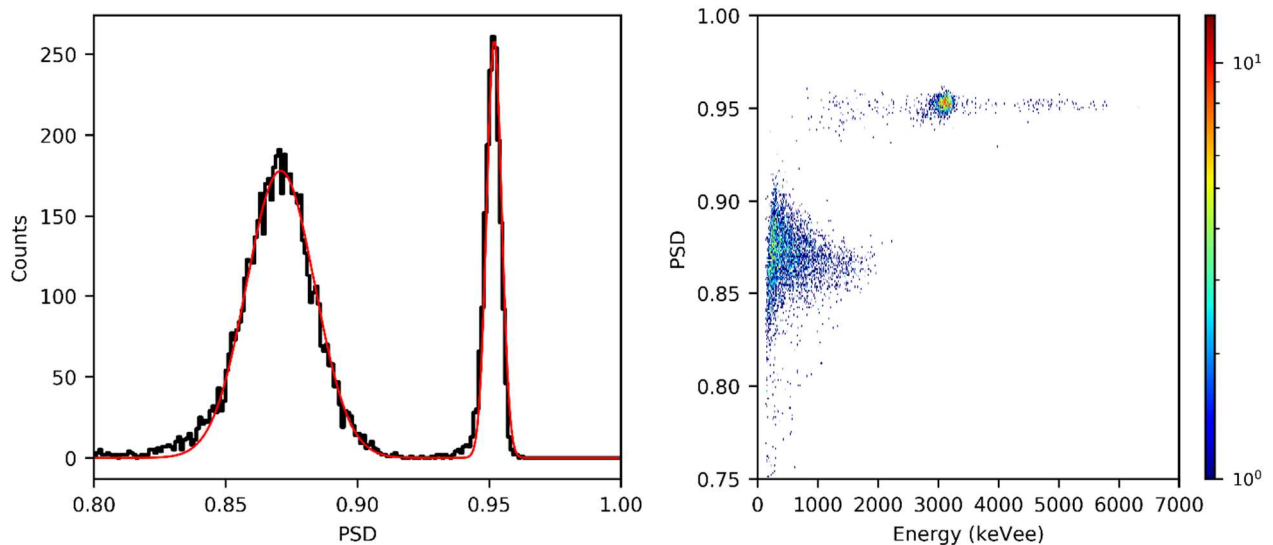
147

148 *Figure 2. Left: ^{137}Cs and ^{60}Co energy calibrated spectra acquired with CLYC-7 when irradiated*
 149 *with Am-Be source neutrons. Right: ^{137}Cs and ^{22}Na energy calibrated spectra acquired with CLYC-*
 150 *6 when irradiated with 2.5 MeV source neutron. The energy resolution of each photopeak is*
 151 *indicated.*

152 3. Measurements with CLYC-6

153 3.1. n/ γ discrimination capability

154 Figure 3 shows the 1D and 2D histogram plots of the PSD and PSD versus energy acquired with
 155 CLYC-6 and the D-D generator. The calculated FOM is 2.20 (for 100 keVee energy threshold),
 156 which confirms the excellent n/ γ discrimination capability of the crystal. The 2D PSD plot clearly
 157 shows the separation between the neutron events (on top of the plot) and the γ -ray events (on the
 158 bottom) with no overlap between the two distributions. The neutron distribution is dominated by a
 159 blob at around 3 MeVee corresponding to the $^6\text{Li}(n,t)\alpha$ reaction from thermal neutrons [7,13,23]. A
 160 single narrow energy peak is seen in the neutron spectrum (see figure 4) as discussed in the next
 161 section.



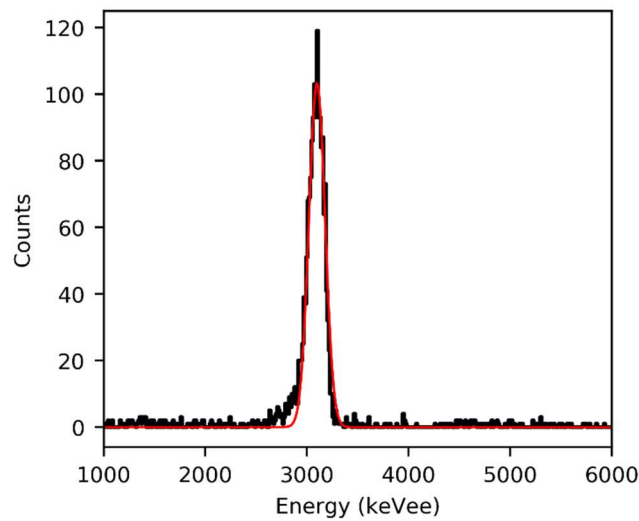
162

163 *Figure 3. Left: the 1D PSD histogram plot acquired with CLYC-6 irradiated with 2.5 MeV neutrons*
 164 *from a D-D generator. The red line is the Gaussian fit to the γ -ray (left peak) and neutron (right*
 165 *peak) distributions. Right: the 2D PSD plot for the same measurement.*

166 3.2. Neutron spectrum and energy resolution

167 From the PSD analysis the neutron events were selected and plotted in terms of counts versus
 168 energy, after the energy calibration. Figure 4 shows the resulting neutron spectrum acquired with
 169 the D-D generator. The main peak is attributed to the thermal neutron absorption on ${}^6\text{Li}$. Thermal
 170 neutrons are probably produced by the scattering of the 2.5 MeV source neutrons with the
 171 surroundings. The 2.5 MeV source neutrons can also directly interact with CLYC-6 via the ${}^6\text{Li}(n,t)\alpha$
 172 reaction. These events should appear on the right-end side of the thermal peak [7], however due to
 173 the difference between the thermal and fast neutron cross sections (940 b at 25 meV versus 0.17 b at
 174 2.5 MeV), the thermal peak is dominant. Neutrons of 2.5 MeV are also detected via the ${}^{35}\text{Cl}(n,p){}^{35}\text{S}$
 175 reaction (the cross section for 2.5 MeV neutrons is 0.19 b) as a single energy peak at 2.5 MeV + the
 176 Q-value of the reaction, multiplied by the proton quenching factor equal to 0.9 (see section 3.3 for
 177 the definition and section 4.2 for the calculation and references), i.e. at 2.8 MeV. The corresponding
 178 peak should appear at the left of the thermal neutron peak and can be the origin of the small bump
 179 visible just below the thermal peak [7,8]. The detected peak was fitted using a Gaussian fitting
 180 equation and the energy resolution calculated as the ratio between the corresponding FWHM and
 181 the mean value of the peak. The thermal neutron energy resolution is 6%.

182



183

184 *Figure 4. The neutron spectrum acquired with CLYC-6 irradiated with 2.5 MeV neutrons from a D-*
 185 *D generator. The red line is the Gaussian fit to the peak corresponding to the ${}^6\text{Li}(n,t)\alpha$ reaction.*

186

187 3.3. Quenching factor of the ${}^6\text{Li}(n,t)\alpha$ reaction

188 The quenching factor q is defined as:

189
$$q = \frac{E_{eq.}}{E_N + Q} \quad (2)$$

190 where $E_{eq.}$ is the energy in equivalent electron-volt (eVee) of the reaction, E_N the energy in
191 electron-volt of the impinging neutron and Q the Q-value of the reaction in electron-volt. In this
192 case, E_N is negligible (*i.e.*, thermal energy) and the Q-value is 4.78 MeV. $E_{eq.}$ was calculated as the
193 mean value of the Gaussian fit of the peak in the neutron spectrum (figure 4), *i.e.*, 3.1 MeV. The
194 resulting q is 0.65 in agreement with [24] which reports a value of 0.7 for the ${}^6\text{Li}(n,t)\alpha$ reaction.

195 3.4. Neutron detection efficiency

196 A tailored cut polyethylene cylinder was placed around the Am-Be source of Cal Lab. The cylinder
197 moderates the fast neutrons emitted by the source and produces a thermal neutron field that
198 irradiated the CLYC-6 crystal [22]. The crystal was placed at 86 cm from the source at the same
199 height above the floor, aligned with two orthogonal lasers. The neutron count rate was obtained
200 after the PSD analysis. For comparison, the neutron count rate at the same irradiation position was
201 measured with a Centronic SP9/152/Kr ${}^3\text{He}$ proportional counter (17 cm³ sensitive volume), filled
202 with a gas mixture of ${}^3\text{He}$ at 2.33 kPa (2.3 atm) and Kr at 1.22 kPa (1.2 atm). The counter was
203 supplied with +820 V and the output signal processed by an ORTEC 142IH preamplifier, an
204 ORTEC 570 amplifier (assembled in a portable NIM crate) and an Amptek Pocket Multi-Channel
205 Analyser 8000D. The neutron count rate was measured integrating the recoil spectrum acquired by
206 the multi-channel analyser setting a proper energy threshold to cut the electronic noise and γ -ray
207 events.

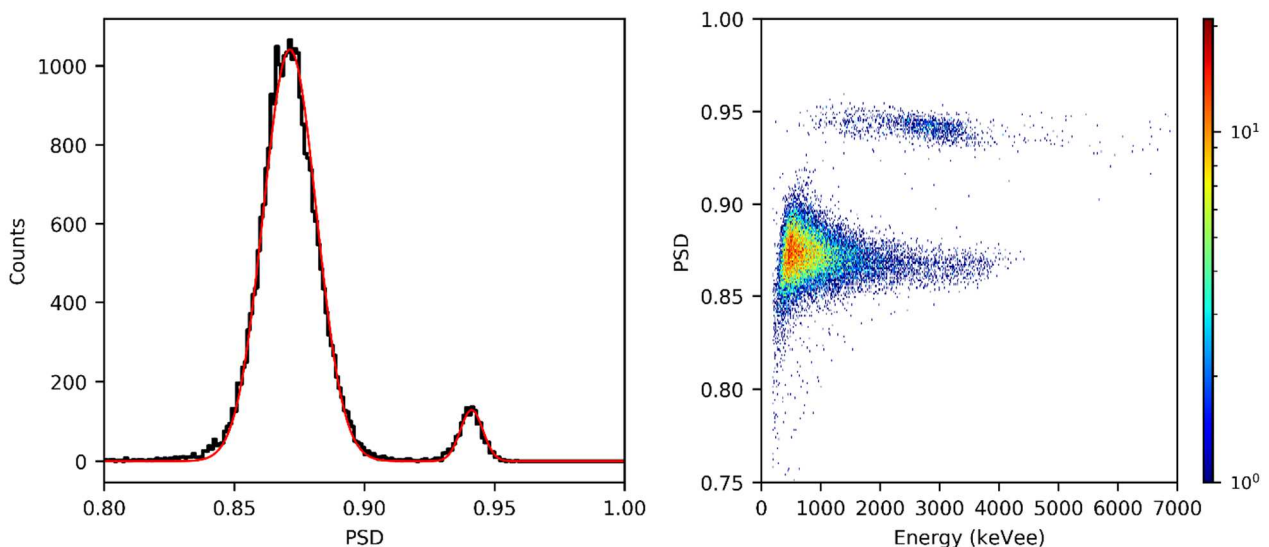
208 Taking into account the sensitive volume of each detector, the measurements confirmed that the
209 neutron detection efficiency per unit volume (cm³) of CLYC-6 is higher than that of the ${}^3\text{He}$ counter
210 [14], 50 counts per second (cps) per cm³ with CLYC-6 (1.62 cm³) versus 20 cps per cm³ with the
211 ${}^3\text{He}$ counter (17.15 cm³).

212 4. Measurements with CLYC-7

213 4.1. n/ γ discrimination capability

214 Figure 5 shows the 1D and 2D histogram plots of the PSD and PSD versus energy acquired with
215 CLYC-7 irradiated with the D-D generator. The Gaussian fits of the neutron and γ -ray distributions
216 are also plotted in figure 5. The corresponding FOM is 2.04, demonstrating excellent n/ γ
217 discrimination. The FOM for CLYC-7 is slightly lower than for CLYC-6 in agreement with [9].

218



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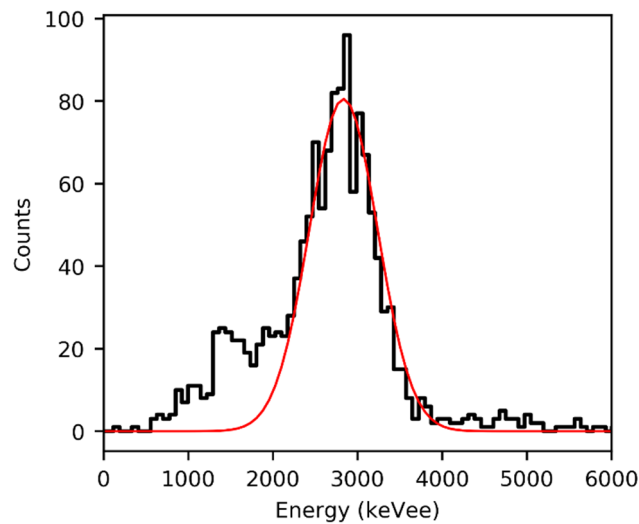
220 Figure 5. Left: the 1D PSD histogram plot acquired with CLYC-7 irradiated with 2.5 MeV neutrons
221 from a D-D generator. The red line is the Gaussian fit to the γ -ray (left peak) and neutron (right
222 peak) distributions. Right: the 2D PSD plot for the same measurement.

223

224 4.2. Quenching factor of the $^{35}\text{Cl}(n,p)^{35}\text{S}$ reaction

225 Figure 6 shows the neutron spectrum acquired with CLYC-7 irradiated with 2.5 MeV neutrons from
226 the D-D generator. The main peak at 2.83 MeVee is due to the (n,p) reaction on ^{35}Cl , the counts
227 detected at the left of the main peak (at around 1.1 MeVee and 1.7 MeVee) come from the (n, α)
228 reaction [13] (to the ground and to the first excited state of ^{32}P) and the (n,p) reaction to the excited
229 states of ^{35}S . Some counts in this energy range can also derive from scattered neutron background.
230 The quenching factor for the $^{35}\text{Cl}(n,p)$ reaction was calculated using equation (2), where $E_{eq.}$ is the
231 centroid of the Gaussian fit to the 2.83 MeVee peak shown in figure 6, $E_N = 2.5$ MeV and $Q =$
232 0.615 MeV. A value of 0.91 was found, in perfect agreement with literature data (from 0.85 to 0.92
233 in [13], 0.9 in [24]).

234



235

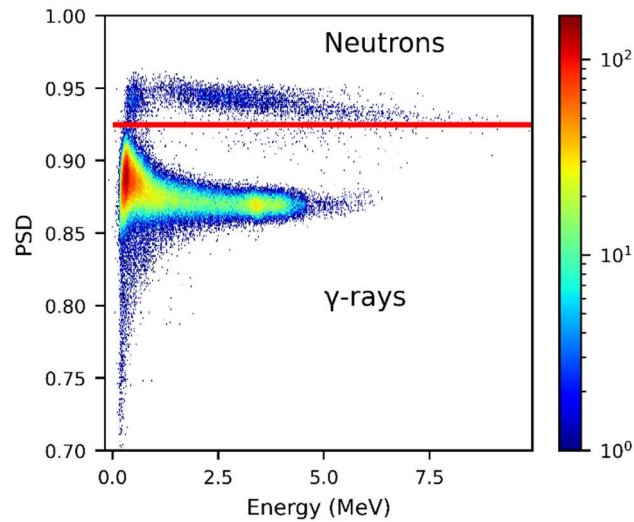
236 Figure 6. Neutron energy spectrum acquired with CLYC-7 irradiated with 2.5 MeV neutrons from a
237 D-D generator. The red line is the Gaussian fit to the peak at 2.83 MeVee corresponding to the
238 $^{35}\text{Cl}(n,p)^{35}\text{S}$ reaction.

239

240 4.3. The Am-Be neutron spectrum

241 CLYC-7 was irradiated with Am-Be source neutrons. The n/ γ PSD analysis was first performed as
242 in the previous study to discard the γ -ray events. Figure 7 shows the 2D histogram plot of the PSD
243 versus energy where the red line delimits the neutron and γ -ray regions in the PSD space. The
244 distance between x_γ and the red line is larger than $3\sigma_\gamma$, where x_γ and σ_γ are the centroid and the
245 standard deviation of the Gaussian equation fitting the γ -ray distribution. Few signals were detected
246 in between the neutron and γ -ray distributions. Because of the uncertainty associated to these
247 signals, they were discarded from the analysis. It is worth mentioning that these signals are 7% of
248 the total neutron signals and are spread over about 5 MeV. Their effect on the resulting neutron

249 spectrum is negligible (the neutron spectrum with these signals differs by less than 1% compared
250 with the spectrum without).

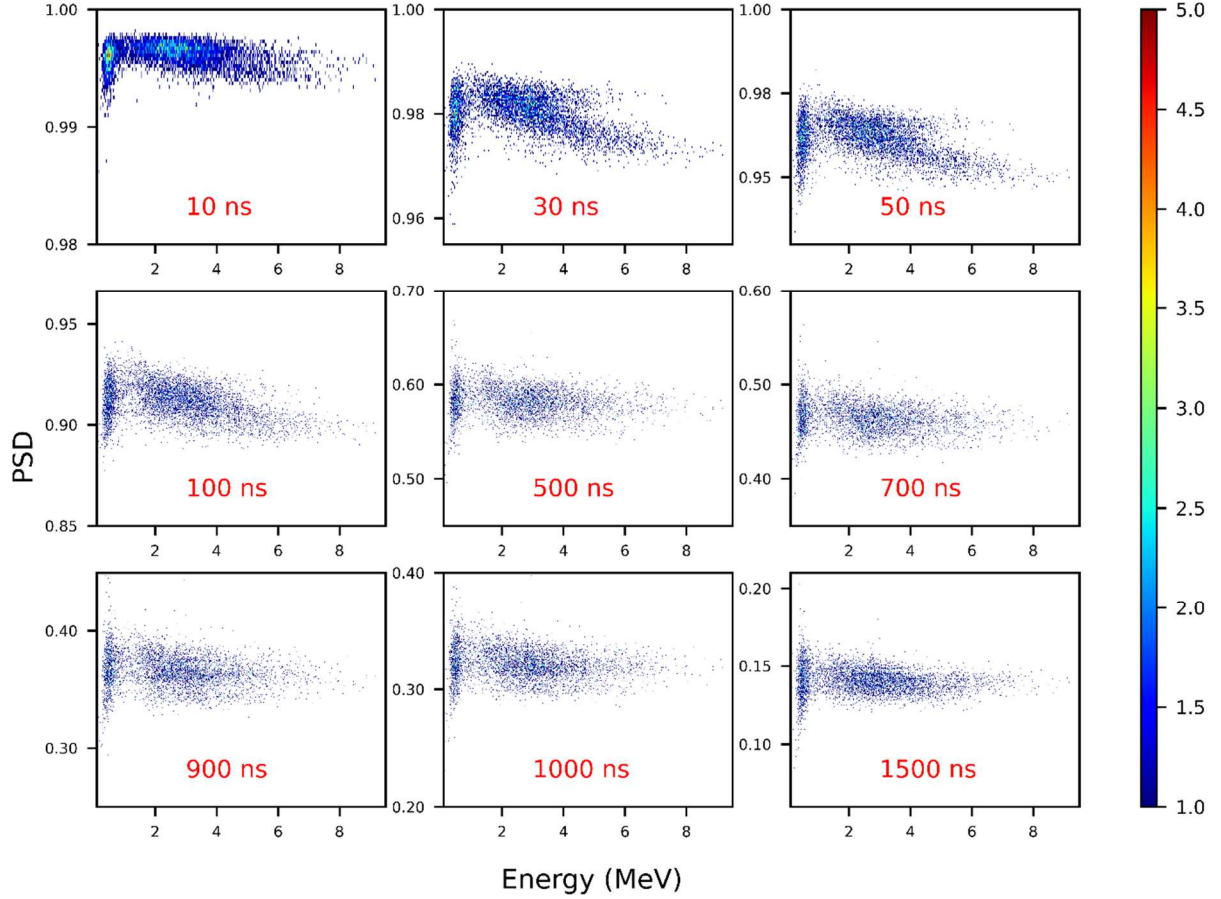


251

252 *Figure 7. 2D histogram plot of the PSD versus energy for CLYC-7 irradiated with Am-Be source*
253 *neutrons.*

254 The γ -ray events were discarded, and the PSD analysis was repeated on the selected neutron signals
255 by varying t_{prompt} from 10 ns to 1.5 μs while t_{delay} was kept constant at 2 μs . Figure 8 shows the 2D
256 histogram plots of the PSD versus energy for the different values of t_{prompt} , as indicated in each plot.
257 The width of the binning and the scale of the x-axis are the same for all plots, while the y scale
258 changes to better visualise the region of interest of each plot. For larger t_{prompt} values (bottom plots)
259 the neutron signals follow a unique distribution. By decreasing the value of t_{prompt} , the neutron
260 distribution starts to broaden and below 100 ns a second distribution arises that deviates from the
261 main distribution. This trend indicates that the signals differ in the first part of their time profile.
262 Variations on the first part of the signals become indeed negligible for larger values of t_{prompt} , when
263 t_{prompt} is almost equal to t_{delay} .

264 This second distribution appears at higher PSD values, which means that it is composed of slightly
265 slower signals (as defined in section 2). For this reason, we associated this distribution to α -particle
266 events: α -particles are higher LET particles (*i.e.*, slower signal) as compared to protons. The α -
267 particle distribution should appear starting from about 2 MeV. At this energy, its cross section is
268 negligible compared with the (n,p) cross section (less than 1%). However, due to the small
269 difference of their signal profiles, the two distributions overlap between 2 MeV and up to around
270 4 MeV. Only above this energy two distributions can be identified.



271

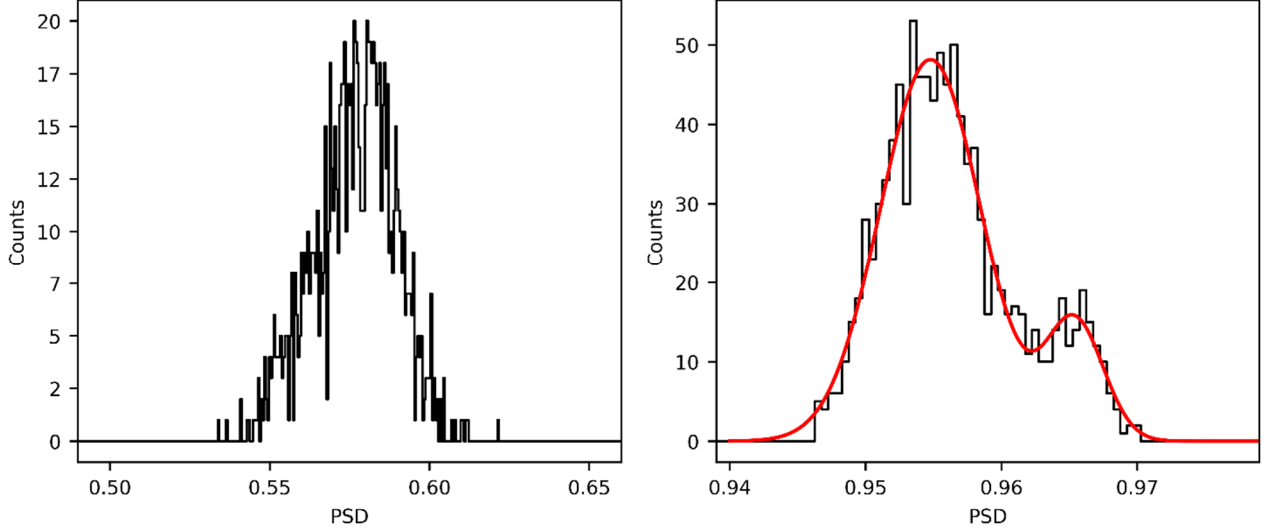
272 *Figure 8. 2D histogram plots of the PSD versus energy for CLYC-7 of the neutron signals from an*
 273 *Am-Be source at different t_{prompt} . The value of t_{prompt} is indicated in each plot, whilst t_{delay} is constant*
 274 *(2 μ s).*

275

276 For each value of t_{prompt} , the PSD values corresponding to the neutron signals were plotted into a
 277 histogram, selecting only the signal with $E > 4$ MeV. For t_{prompt} equal to 30 ns and 50 ns only, two
 278 Gaussian-like distributions appear which were fitted using a Gaussian equation, and the FOM
 279 calculated as follows:

$$280 \quad FOM = \frac{x_{\alpha} - x_p}{FWHM_{\alpha} + FWHM_p} \quad (3)$$

281 where x_{α} and x_p are the peak centroids of the α -particle and proton Gaussian distributions and
 282 $FWHM_{\alpha}$ and $FWHM_p$ the corresponding full widths at half maximum. The calculated FOM was
 283 0.68 and 0.75 for 30 ns and for 50 ns respectively. The latter was thus selected for the p/ α
 284 discrimination. Figure 9 compares the PSD histogram plot obtained for $t_{prompt} = 500$ ns and $t_{prompt} =$
 285 50 ns. The separation between the proton and the α -particle distributions is not very good (a FOM <
 286 1.25 implies that $x_{\alpha} - x_p < 3(\sigma_{\alpha} + \sigma_p)$), however two distributions have been detected, while it
 287 was not possible for other t_{prompt} values (such as for $t_{prompt} = 500$ ns, as shown in figure 9).



288

289 *Figure 9. The 1D histogram plot of the PSD for CLYC-7 for the neutron signals at $t_{prompt} = 500$ ns*
 290 *(on the left) and $t_{prompt} = 50$ ns (on the right). The right plot also shows the Gaussian fit to the*
 291 *selected proton and α particle distributions.*

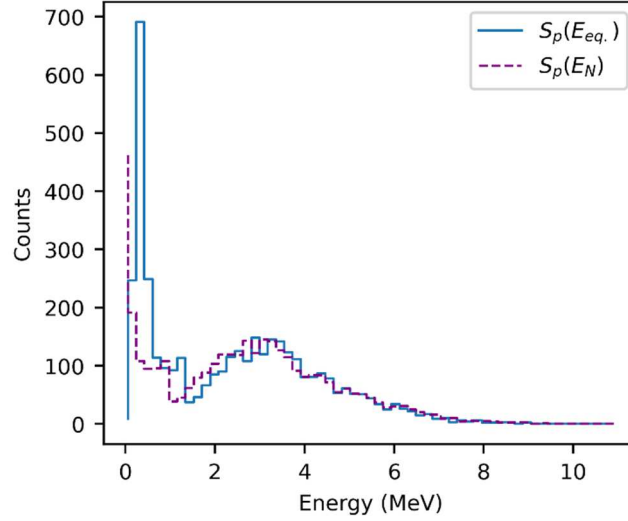
292 A PSD cut-off value equal to 0.9622 was defined, below which the pulses were classified as
 293 produced by protons and above by α -particles, excluding from the α -particles all signals detected as
 294 a blob below 1.5 MeV which spread over a wider PSD range (0.95-0.98). The latter are instead
 295 classified as protons since the (n, α) cross section is negligible below 1.5 MeV (0.10977 b the (n,p)
 296 cross section versus 0.0005 b the (n, α) cross section at 1.5 MeV [15]). According to this criterion,
 297 the pulses from protons and α -particles were collected separately and plotted into a histogram. The
 298 spectra so calculated correspond to the spectra of the energy deposited by protons and α -particles
 299 respectively, as a function of $E_{eq.}$. We will refer to them as $S_p(E_{eq.})$ and $S_\alpha(E_{eq.})$.

300 Inverting equation (2), the corresponding neutron spectra can be calculated as follows:

$$301 \quad E_N = (E_{eq.}/q) - Q \quad (3)$$

302 The neutron energy spectra, labelled as $S_p(E_N)$ and $S_\alpha(E_N)$, were calculated applying equation (3)
 303 to $S_p(E_{eq.})$ and $S_\alpha(E_{eq.})$, assuming q equal to 0.9 (see section 4.2) for protons and to 0.5 for α
 304 particles [24]. The Q_{value} is 615 keV for the (n,p) reaction and 938 keV for the (n, α) reaction.

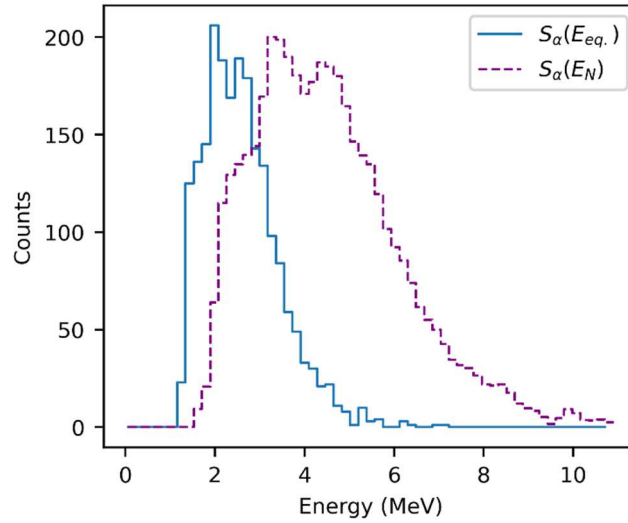
305 Figure 10 shows $S_p(E_{eq.})$ and $S_p(E_N)$, figure 11 $S_\alpha(E_{eq.})$ and $S_\alpha(E_N)$.



306

307 *Figure 10. The spectrum of the energy deposited by protons in CLYC-7 (continuous blue line) and*
 308 *the related spectrum of the incoming neutrons (dotted purple line), calculated taking into account*
 309 *the quenching factor (0.9) and the Q -value of the reaction (615 keV).*

310



311

312 *Figure 11. The spectrum of the energy deposited by α particles in CLYC-7 (continuous blue line)*
 313 *and the related spectrum of the incoming neutrons (dotted purple line), calculated taking into*
 314 *account the quenching factor (0.5) and the Q -value of the reaction (938 keV).*

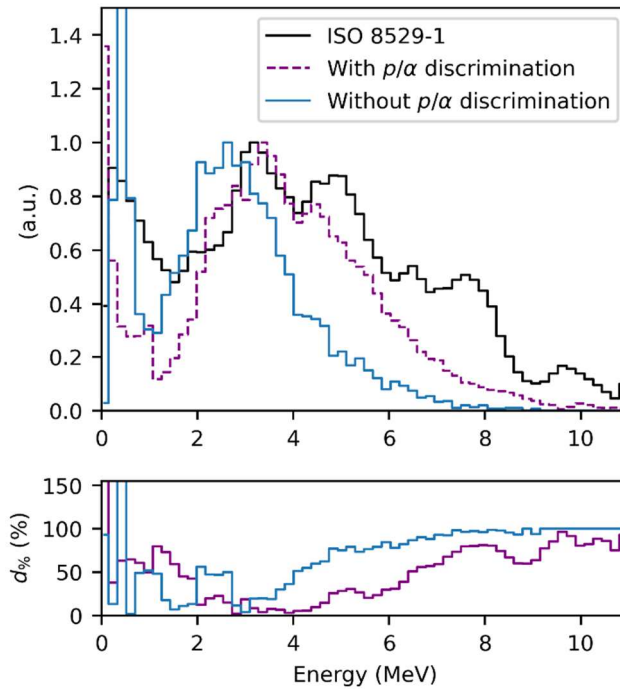
315 The total spectrum of the incoming neutrons was calculated as the sum of $S_p(E_N)$ and $S_\alpha(E_N)$ and is
 316 compared with the ISO spectrum [25] and with the spectrum calculated without the p/ α
 317 discrimination in figure 12. The three spectra have been normalised to their maximum (discarding
 318 the energy bins below 500 keV). Figure 12 also shows the percent difference ($d_\%$) between the Am-
 319 Be ISO spectrum and the spectrum with and without the p/ α discrimination. $d_\%$ is defined as:

$$320 \quad d_\% = \frac{|y_{ISO,i} - y_i|}{y_{ISO,i}} \times 100 \quad (4)$$

321 where $y_{ISO,i}$ is the intensity of the normalised Am-Be ISO spectrum in the i -th bin and y_i the
 322 normalised counts of the experimental spectrum for the same bin. The average $d_\%$ for $E > 500$ keV

323 (i.e. for fast neutron energies) is 46% with the p/α discrimination and 68% without the p/α
324 discrimination.

325

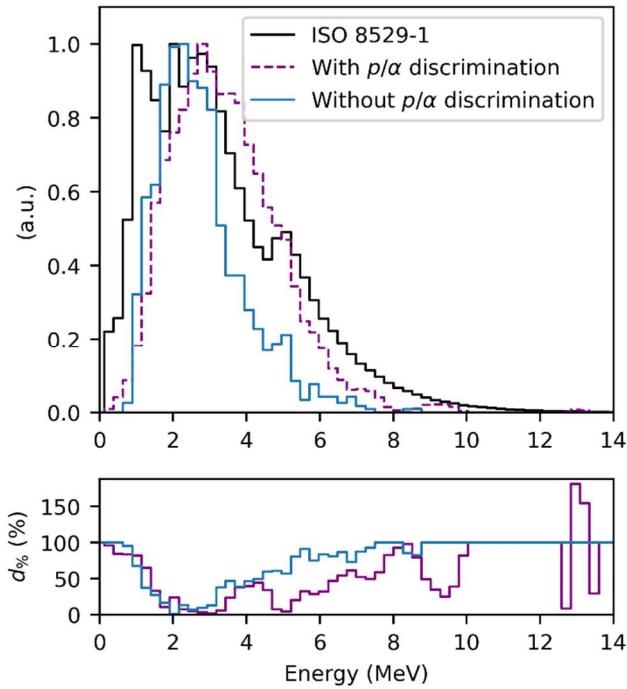


326

327 *Figure 12. Top: comparison between the Am-Be ISO spectrum (continuous black line) and the*
328 *neutron spectrum measured with CLYC-7, without discrimination of neutron signals (continuous*
329 *blue line) and with the p/α discrimination (dotted purple line). Bottom: percent variation between*
330 *the Am-Be ISO spectrum and the spectrum obtained with (continuous purple line) and without*
331 *(continuous blue line) p/α discrimination.*

332 4.4. The ^{252}Cf neutron spectrum

333 The procedure described in section 4.3 was applied to the data acquired irradiating CLYC-7 with
334 ^{252}Cf source neutrons. For this case only, t_{delay} were set to 1.5 μs while t_{prompt} was kept at 70 ns for
335 the n/γ discrimination and at 50 ns for the p/α discrimination. Figure 13 compares the ISO spectrum
336 [25] and the experimental spectrum before and after the analysis proposed in this work. The
337 resulting $d_{\%}$ parameters for $E > 500$ keV were 61% and 77% with and without the p/α
338 discrimination respectively.

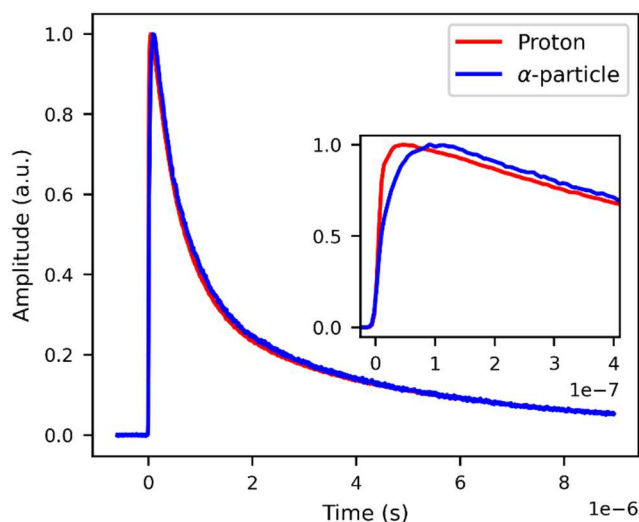


339

340 *Figure 13. Top: comparison between the ^{252}Cf ISO spectrum (continuous black line) and the*
 341 *neutron spectrum measured with CLYC-7, without discrimination of neutron signals (continuous*
 342 *blue line) and with the p/α discrimination (dotted purple line). Bottom: the percentage variation*
 343 *between the ^{252}Cf ISO spectrum and the spectrum obtained with (continuous purple line) and*
 344 *without (continuous blue line) the p/α discrimination.*

345 **4.5. Pulse shape analysis of the neutron signals**

346 To quantify the difference in terms of time profile between the proton and α -particle signals defined
 347 by the p/α discrimination proposed in this work, a pulse shape analysis was performed. The signals
 348 acquired with the CLYC-7 irradiated with the Am-Be source were considered and the so-called
 349 standard pulse of protons and α -particles was calculated as follows. All signals were aligned and the
 350 PSD was calculated as described in section 2 ($t_{\text{prompt}} = 70$ ns and $t_{\text{delay}} = 2$ μs). The γ -ray signals, *i.e.*
 351 the signals below the red line in figure 7, were discarded and the p/α discrimination was applied to
 352 the remaining signals as described in section 4.3 ($t_{\text{prompt}} = 50$ ns and $t_{\text{delay}} = 2$ μs). Only the events
 353 with $E > 4$ MeV were selected, where the recognised proton and α -particle distributions do not
 354 overlap (see figure 8, case $t_{\text{prompt}} = 50$ ns). The pulses recognised as protons were averaged and
 355 normalised to obtain the proton standard pulse. The same calculation was performed to obtain the α -
 356 particle standard pulse. Figure 14 shows the results.



357

358 *Figure 14. The proton and α -particle standard pulses obtained with the proposed p/ α*
 359 *discrimination, in CLYC-7 irradiated with Am-Be source neutrons*

360 **4.6. Discussion on the p/ α discrimination**

361 The p/ α discrimination via the PSD method is in principle possible. In fact, with the properly
 362 selected time windows, different kinds of particles are collected in different distributions in the PSD
 363 plot. Giaz *et al* [3] also detected two different distributions irradiating CLYC-7 with 14.1 MeV
 364 neutrons. The two distributions were also attributed to different LET particles, *i.e.*, protons and α
 365 particles. However, Giaz *et al.* did not exploit this difference to improve the CLYC spectroscopic
 366 capability. A similar analysis was instead proposed by Dolymphia *et al* [4] with a 7.23 MeV
 367 monoenergetic neutron beam. However, a continuous neutron spectrum, as used in this work,
 368 represents a more realistic scenario.

369 The pulse shape analysis demonstrates the correctness of the method since the signals ascribed to
 370 protons are faster than the signals ascribed to α -particles, *i.e.* the PSD is able to distinguish even
 371 small differences in the time profile if the proper time windows are selected. This was also
 372 demonstrated for higher neutron energies (up to 60.5 MeV) in [5]. For this case, the proton and α -
 373 particle's standard pulses differ the most in the first 100 ns of the signal, during their rise time. The
 374 rise time of the proton and α -particle standard pulses is 16 ns and 48 ns respectively. The tails of the
 375 two signals are slightly different up to around 4 μ s. However, the corresponding fall times are more
 376 comparable, 5.38 μ s for protons and 5.54 μ s for α -particles.

377 After the p/ α discrimination the measured Am-Be and ^{252}Cf spectra better match the corresponding
 378 ISO spectra. The percent difference between the experimental and the ISO spectra, averaged over
 379 the spectrum energies, improves for both cases by more than 15% when the p/ α discrimination is
 380 applied. Above 2 MeV the spectrum with p/ α discrimination is always closer to the corresponding
 381 ISO spectrum than the spectrum without discrimination (except at 13 MeV for two bins for the ^{252}Cf
 382 source). However, above 5 MeV the difference between ISO and experimental spectra increases in
 383 both cases, probably because of the drop of the (n,p) cross section at 5 MeV and the increasing
 384 importance of different reaction channels (see figure 1).

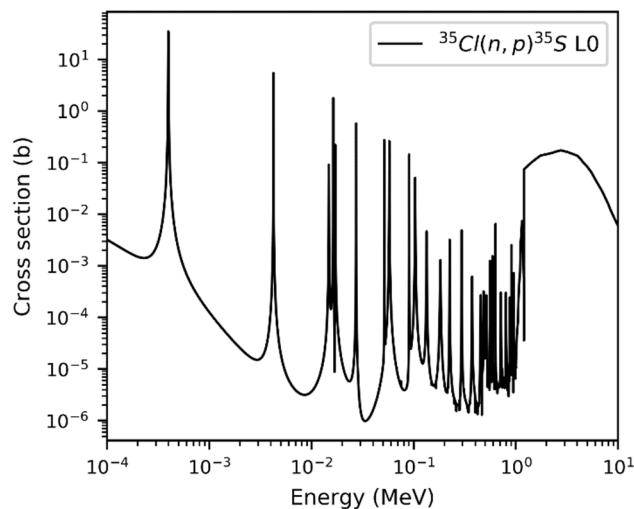
385 The non-match between the reference spectrum and the experimental one, in particular below 3-
 386 4 MeV, is due to the uncertainty associated with the p/ α discrimination since, in this region, many
 387 proton and α -particle pulses overlap. For Am-Be, a high peak at low energy is also visible, which
 388 can be due to either the scattered neutron component [3] or the resonance peaks of the (n,p) cross

389 section below 1 MeV [23] (figure 15). Because of the lack of thermal neutrons in the ^{252}Cf
390 spectrum, the same peak is not revealed.

391 The ^{252}Cf neutron spectrum has a more pronounced low energy component as compared to the Am-
392 Be spectrum and, as a consequence, the shape of the experimental spectrum without the p/ α
393 discrimination already matches quite well the peak of the ISO spectrum between 2 MeV and 3 MeV
394 [6]. However, the tail of the spectrum is not well reproduced, whilst the p/ α discrimination partially
395 restores the correct shape above 3 MeV.

396 Despite the p/ α discrimination performed in this study improves the performance of CLYC-7 as a
397 spectrometer, the method is still not rigorous since the overlapping region between the proton and
398 α -particle is dominant and the selection of the discrimination parameter is affected by high
399 uncertainty. A second limitation exists which cannot be overcome: the contribution to the neutron
400 spectrum of the (n,p) and (n, α) reactions leaving the ^{35}S and ^{32}P in their excited states. These
401 reactions contribute to the neutron spectrum also below 8 MeV, depositing per event less energy
402 compared with the corresponding reaction leaving the ^{35}S and ^{32}P to the ground states. However,
403 protons emitted by the (n,p) (ground state) reactions cannot be distinguished by protons emitted
404 after the (n,p) (excited state) reactions. The same is valid for α -particles. In general, the analysis
405 must be improved for example with the use of a faster photodetector, a specific electronic readout
406 or a more sensitive PSD analysis, and further investigations are needed before its application.
407 Nevertheless, the application of CLYC-7 as a portable fast-neutron spectrometer for the B-RAD
408 detector, is still attractive. A small, lightweight, single detector, instead of the commonly used
409 Bonner Sphere Spectrometer, represents a great advantage especially in some applications in
410 radiation protection where some uncertainty is acceptable.

411



412

413 *Figure 15. Zoom of the total cross section of the (n,p) and (n, α) reactions on ^{35}Cl , in the region of*
414 *interest. Below 1 MeV several resonance peaks are visible [15].*

415 5. Conclusions

416 CLYC-6 (enriched in ^6Li to > 95%) and a CLYC-7 (enriched in ^7Li to > 99%) crystals were studied
417 as thermal neutron detector and as fast neutron spectrometer, respectively. Both crystals were
418 irradiated with a 2.5 MeV monoenergetic neutron beam and their n/ γ discrimination capability
419 evaluated by the PSD method, both showing excellent performance (FOM > 2). The energy

420 resolution of CLYC-6 to 2.5 MeV neutrons is 6% and the measured quenching factor for the
421 ${}^6\text{Li}(n,t)\alpha$ reaction is 0.65. The quenching factor of the ${}^{35}\text{Cl}(n,p){}^{35}\text{S}$ reaction in CLYC-7 is 0.91.

422 As neutron counter, the thermal neutron detection efficiency of CLYC-6 was measured relative to
423 that of a ${}^3\text{He}$ proportional counter. For the thermal neutron field available at Cal Lab and for the
424 same irradiation position, CLYC-6 shows a 60% higher efficiency per unit volume.

425 The neutron spectrometry capability of CLYC-7 was investigated irradiating the crystal with the
426 continuum neutron spectra from Am-Be and ${}^{252}\text{Cf}$ sources. A p/α discrimination was carried out by
427 the PSD method, to distinguish between the protons and α -particles produced by the two main
428 reactions occurring in the crystal below 10 MeV. After p/α discrimination the experimental neutron
429 spectra better match the corresponding ISO spectra above 2 MeV. The best match was found
430 between 2 MeV and 4 MeV, where the difference with the ISO spectrum is below 30%. Below 2
431 MeV, the overlapping of proton and α -particle pulses is not negligible, whilst above 5 MeV
432 different reaction channels become comparable with the abovementioned ones.

433 The results here reported show that CLYC-7 is a suitable candidate for a probe for fast neutron
434 spectrometry for B-RAD below ~ 5 MeV and, simultaneously, as a stationary environmental
435 detector to monitor both the γ -ray (with the current probe [12]) and the neutron dose rates.
436 However, the proposed p/α discrimination method cannot yet be applied in an actual detector as
437 many events are still erroneously classified. CLYC-6 can instead be used for neutron count rate and
438 measurement of neutron ambient dose equivalent if embedded in a moderator (typically
439 polyethylene) to increase the thermal neutron component of the neutron field.

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