Response of LaBr₃(Ce) scintillators to 14 MeV fusion neutrons

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dedicated neutron capture measurements in present tokamak devices[8]

In this paper, we complete our previous investigation by presenting the response of LaBr₃(Ce) scintillators to 14 MeV neutron irradiation, which is most interesting for operations in deuterium-tritium plasmas. Measurements were performed at the Frascati neutron generator (FNG) with 2.5 MeV mono-energetic neutrons and at tokamak devices run in deuterium. A dedicated MCNP model was used to interpret the results.

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Similarly to our previous investigation, an MCNP model is used to aid in the interpretation of the results and, in particular, to identify the most relevant processes determining the measured response.

2. 14 MeV neutron measurements at FNG

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2. 14 MeV neutron measurements at FNG

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experiment, a 3′ × 3′ (diameter × height) LaBr₃(Ce) scintillator was placed at 90 degrees with respect to the incoming deuterium beam, at the same height of the target. The circular face of the scintillator was facing the source. The distance from the target (1.25 m) was chosen so to obtain a neutron flux of about 2.4 × 10⁷ n/(cm² s) on the detector front surface for a neutron yield of 5 × 10⁸ n/s generated by the machine. The latter was measured by the standard neutron counting diagnostic of FNG, which consists of an absolutely calibrated detector measuring alpha particles produced by deuterium–tritium reactions in the target. The impinging neutron energy spectrum at the detector position was centred at 14 MeV, with an estimated 1% (FWHM) energy spread.

The light emitted by the scintillator was collected by a photomultiplier tube and signals were fed directly into a 12 bit–250 Msample/s CAEN DT5720 digitizer. Each waveform above a selectable threshold was stored and processed off-line using a dedicated algorithm based on pulse fitting [7]. The pulse height spectrum was energy calibrated with laboratory ¹³⁷Cs and ⁶⁰⁰Co radioactive sources. It is worth to point out that the step to neutron data (up to 14 MeV) requires a quite large extrapolation compared to the available calibration points. An energy resolution of 3.2% was measured at the 0.662 MeV line of ¹³⁷Cs, which is close to the nominal value expected for LaBr₃ crystals (about 3%). In the following, the letter $E$ indicates the γ-ray (or electron equivalent) energy; a subscript is otherwise used to indicate the energy of a different particle.

Fig. 1 shows the measured energy spectrum during 14 MeV neutron irradiation. Data were integrated over a time of 120 s. Gamma-rays peaks at distinctive energies and more complex structures can be clearly observed in the figure, with most of the events in the region $E < 3$ MeV, where the detection efficiency to γ-rays is known to be higher [18]. There is however a clear tail of events extending to energies up to about $E = 14$ MeV. An average count rate of 8 × 10⁶ cps above the experimental $E = 0.35$ MeV threshold was observed during measurements. After about 1 h of 14 MeV neutron irradiation, the FNG beam was switched off, and a post-irradiation measurement was started. Fig. 1 shows the spectrum measured 10 min after irradiation, which can be compared to that obtained during irradiation, for the same experiment time. There is a clear difference in magnitude and shape between the two spectra. No events are found above $E = 4$ MeV in the post-irradiation spectrum. Many of the structures observed during irradiation have also disappeared and the shape of the spectrum is now dictated by the superposition of beta decays of different short lived unstable isotopes (see Section 5). An intense 511 keV annihilation peak still remains, together with a second peak at $E = 0.8$ MeV. The post irradiation spectrum can also be compared to that due to the crystal intrinsic radioactivity (Fig. 1, dotted line). The latter was measured at FNG right before irradiation and also shows a 662 keV peak due to a ¹³⁷Cs calibration source. Again, we find no similarity between the two spectra, both in terms of structure and magnitude, which indicates that the post-irradiation spectrum must be dominated by decays of unstable isotopes generated by 14 MeV neutron irradiation. The contribution of intrinsic radioactivity to the count rate before neutron irradiation was 700 cps, a value much lower than the (average) 2.5 × 10⁶ cps measured 10 min after neutron irradiation.

3. Monte Carlo simulation of the LaBr₃ response

As discussed in our previous paper on 2.5 MeV neutrons [16], nuclear interactions between 14 MeV neutrons and lanthanum and bromine isotopes dominate the measured response. ⁷⁹Br and ⁸¹Br are the Bromine isotopes in the crystal, with approximately the same abundance, while ¹³⁹La is the only stable isotope of Lanthanum. Few other isotopes are also present, but only in trace concentrations and were not considered in the analysis. Besides inelastic scattering, the main neutron interaction mechanism with these isotopes is the production of secondary particles (neutrons, protons, deuterons, alpha particles) which in turn deposit their energy into the crystal, resulting in recordable pulses. The cross-sections for the most relevant reactions are shown in Fig. 2. It is worth noticing here that there is a clear difference between the dominant neutron interaction channels with LaBr₃ at $E_n = 2.5$ MeV and $E_n = 14$ MeV: while at $E_n = 2.5$ MeV inelastic scattering dominates the response [16], secondary particle production is the most important process at $E_n > 10$ MeV and, particularly, (n,2n) reactions. The latter have a multiplicative effect on the detector response: secondary neutrons generated in the crystal can in turn undergo nuclear inelastic scattering, which generates excited nuclear states decaying by γ-ray emission, that adds up to the detector gamma-ray load due to inelastic scattering of the primary (14 MeV) neutrons. Among the reactions leading to charge particle production, proton generation due to n + ⁷⁹Br interactions is the most relevant.

A MCNP [19] model was implemented to interpret the experimental results, assuming a beam of mono-energetic 14 MeV neutrons directed onto a 3′ × 3′ LaBr₃ and tracking all the processes listed in Fig. 2. A summary of the results of this simulation is presented in Table 1, where numbers are normalized per 14 MeV neutron history. Of most interest is γ-ray production, which exceeds unity with 1.17 γ-rays produced per 14 MeV neutron. This result can be explained by the large additional contribution of secondary neutrons generated in the crystal (0.45), a fraction of which also undergoes inelastic scattering. Here we note that γ-rays born in the crystal volume have a significant detection probability (about 40%), given the large volume, high density and high Z of the scintillator, and are thus of relevance to understand the measured crystal response. Charged particle production is less important, mostly because of the smaller cross-sections of the associated processes, although, on the other hand, positive ions have a detection probability approaching 100%. For comparison, the simulation was also run for a 3′ × 6′ crystal, which is the size of the scintillator currently in use at JET [15]. As noted from Table 1, doubling the crystal volume determines a 55% increase in secondary neutron production, which is turn enhances gamma-ray creation by almost the same amount. Charged particle production remains small.

The contribution of secondary neutron/gamma-rays and other particles to the crystal response was also analysed in terms of spectral shapes, as shown in Fig. 3 for the 3′ × 3′ case. γ-rays dominate in the
region $E < 5$ MeV, while proton production contributes most to the spectral shape at $E > 5$ MeV, leading to a change of slope in this region. Alpha particle and deuterium generation introduces two additional structures centred around $E = 6$ and $E = 10$ MeV, respectively, which are however an order of magnitude lower than that due to protons and would be hard to distinguish in the overall detector response, without using pulse shape discrimination algorithms as investigated in recent studies [21]. By summing up all the different contributions, we can determine a 43% detection efficiency to 14 MeV neutrons above the experimental threshold of 0.35 MeV, based on the MCNP results. The efficiency is defined as the number of counts above threshold per impinging neutron.

In order to compare the simulated response with that measured, a separated calculation of the (external) neutron/gamma-ray background in the FNG hall at the detector position was performed, which was finally added to the (intrinsic) response described above. In fact, the radiation field impinging on the detector in the FNG hall is a complex mixture of neutron and $\gamma$-rays. Of these two, the neutron field has a dominant 14 MeV (direct) component, with an additional scattering contribution at the 50% level, the latter arising from direct neutrons that degrade their energy by interacting with materials in the experimental hall. Gamma-ray background originates from inelastic scattering of the direct neutrons. For the calculation of the scattered neutron and gamma-ray fields, we relied on an existing MCNP model of the FNG facility [22]. Its results were in turn used as input to separately determine the crystal response to such background for comparison with measurement.

Table 2 summarizes the output of the calculations, with numbers normalized per 14 MeV neutron history. Scattered neutrons are responsible of about 50% of the detector load due to the primary component. Gamma-ray background contributes to a further 30% fraction. In terms of energy spectrum (Fig. 4), the scattered neutron component has an exponential shape without clear structures, while gamma-rays are manifested as distinctive peaks and dominate the $E > 4.5$ MeV region. Fig. 4 also shows the response to direct 14 MeV neutrons (presented in Fig. 3) and which can be here compared to background. In order to obtain the $\gamma$-ray equivalent energy for the

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**Table 1**

<table>
<thead>
<tr>
<th></th>
<th>LaBr$_3$ 3$''$ x 6$''$</th>
<th>LaBr$_3$ 3$''$ x 3$''$</th>
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</thead>
<tbody>
<tr>
<td>Gamma creation</td>
<td>1.868</td>
<td>1.178</td>
</tr>
<tr>
<td>Neutron creation (n,2n)</td>
<td>6.982 $\times$ 10$^{-1}$</td>
<td>4.513 $\times$ 10$^{-1}$</td>
</tr>
<tr>
<td>Proton creation</td>
<td>1.53 $\times$ 10$^{-2}$</td>
<td>9.54 $\times$ 10$^{-3}$</td>
</tr>
<tr>
<td>Deuteron creation</td>
<td>1.13 $\times$ 10$^{-3}$</td>
<td>7.63 $\times$ 10$^{-4}$</td>
</tr>
<tr>
<td>Alpha creation</td>
<td>1.58 $\times$ 10$^{-3}$</td>
<td>1.30 $\times$ 10$^{-3}$</td>
</tr>
</tbody>
</table>

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**Table 2**

<table>
<thead>
<tr>
<th></th>
<th>Counts per 14 MeV neutron history</th>
</tr>
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<tr>
<td>Counts from direct 14 MeV neutrons</td>
<td>0.432</td>
</tr>
<tr>
<td>Counts from scattered neutrons at FNG</td>
<td>0.206</td>
</tr>
<tr>
<td>Counts from gamma background at FNG</td>
<td>0.138</td>
</tr>
<tr>
<td>Sum</td>
<td>0.776</td>
</tr>
</tbody>
</table>
The finite energy resolution of the detector was included in Fig. 4 by convolution of the simulated spectra with a Gaussian function of energy dependent width. This was obtained from the measured value of 3.2% (FWHM/E) at 662 keV and assumed to follow the Poisson law, i.e. to scale as $E^{-1/2}$.

4. Data analysis

The results of the MCNP simulations are compared to the measured spectrum in Fig. 5. To this extent, the three components of Fig. 4 (direct and scattered neutrons, gamma-ray background) were summed to obtain the expected measured spectrum during the FNG irradiation experiment. The comparison is limited to a range that well exceeds the accuracy needs for the use of LaBr$_3$ orders of magnitude below the most intense spectral structures, a range where the accuracy needs for the use of LaBr$_3$ in gamma-ray spectroscopy applications at ITER are concerned. In particular, when shown in logarithmic scale, the measured spectrum exhibits the change of slope predicted by MCNP in the region 5–10 MeV and due to the contribution of secondary charged particles (see Section 3). At a more detailed level, we find that most of the gamma-ray peaks observed in the measurement are also reproduced by the simulation, although, at times, there is a mismatch in terms of intensity. In few cases only, peaks predicted in the simulation do not appear in the measurement, and vice versa. We ascribe these minor differences to uncertainties in the cross-sections used by MCNP, especially since they are derived by statistical models, which may not accurately depict reality [26]. A second source of uncertainty arises from the FNG model, which may not specify the material composition of the experimental hall at a level of detailed necessary to explain all peaks found in the measurement. In spite of these minor discrepancies, we consider the level of agreement between simulation and measurement as satisfactory, given the capability of the simulation to match well the experimental count rate and spectral shape, which are of most interest to design filters for neutron background reduction on the scintillator at ITER [11]. In this context, we note in particular that the measured spectrum is rather structureless in the range $E = 3$ to 5 MeV, i.e. where gamma-ray lines from plasma reactions of principal interest in thermonuclear fusion applications occur.

5. Short lived activation induced by 14 MeV neutrons

Activation of the LaBr$_3$ crystal is of interest in view of ITER for two reasons. First, a significant activation level requires some special care when handling the crystal after an extended irradiation period. Second, the activation itself is a background source that interferes with the measured signal and is thus of relevance for signal-to-noise ratio considerations in view of measurements at ITER. In order to investigate the activation of the crystal induced by 14 MeV neutrons, post-irradiation measurements were carried out at FNG. The LaBr$_3$ crystal was first irradiated for about 1.5 h at a total neutron yield of $1.5 \times 10^{10}$ n/s, corresponding to a flux on the detector front surface of approximately $5.4 \times 10^5$ n/s/cm$^2$. A measurement of the residual count rate, as a function of time, due to the crystal activation was then started immediately after FNG had been switched off, with the results shown in Fig. 6. Data points were acquired every second, for 30 min.

Based on our previous analysis [16], short lived activation is expected to be due to $^{79}$Br and $^{80}$Br isotopes, which have a decay time of 540 s and 1500 s, respectively. $^{79}$Br and $^{80}$Br are produced by (n,2n) reactions on $^{79}$Br and $^{81}$Br. On the other hand, the (n,2n)
reaction on $^{139}$La produces $^{138}$La, which has a half-life of billions of years, and is therefore practically stable on the time scale of minutes.

The conclusions of our previous analysis are experimentally confirmed by a fit to the measurement of Fig. 6 using the following equation:

$$I = A \times \exp \left( -\frac{t}{\tau_1} \right) + B \times \exp \left( -\frac{t}{\tau_2} \right) + C$$

(1)

where $I$ indicates the intensity (count rate) and $t$ is the time. In Eq. (1) the time constants $\tau_1$ and $\tau_2$ were fixed and set to the decay times of $^{78}$Br (540 s) and $^{80}$Br (1500 s). The free fit parameters $A$, $B$, and $C$ are proportional to the initial number of $^{78}$Br and $^{80}$Br nuclei produced by 14 MeV irradiation. C is a fit parameter that takes into account long lived ($t \gg 30$ min) activation, such as that due to materials of the FNG hall that were also activated by 14 MeV neutrons. As seen from Fig. 6, the fit reproduces very well the measurements, confirming the role of $^{78}$Br and $^{80}$Br as main responsible of the crystal post-irradiation activity. The values obtained for the fit parameters $A$, $B$, and $C$ are summarized in Table 3.

### 6. Conclusions

The response of a $3^\times3^\times3^\times3$ LaBr₃ crystal to 14 MeV neutron irradiation was measured at FNG in a dedicated experiment. The results were interpreted by means of a MCNP model of 14 MeV neutron interactions with LaBr₃, including background contributions from scattered neutrons and gamma-rays generated in the FNG experimental hall. A good overall agreement between simulation and measurements was found. These measurements are novel with respect to the previously reported response function at 2.5 MeV, since the interaction physics of 14 MeV neutrons with the crystal is completely different. The simulations revealed that (n,2n) reactions and nuclear inelastic scattering on $^{79}$Br, $^{81}$Br and $^{138}$La isotopes are the dominant interaction processes of 14 MeV neutrons in the crystal. Reactions leading to the production of secondary charged particles are of relevance to explain events at deposited energies higher than 5 MeV. The overall efficiency of the $3^\times3^\times3$ detector to 14 MeV neutrons was found to be 43%, above an experimental threshold of 0.35 MeV. Measurements of the residual crystal activation after irradiation were also performed and were found to be dominated by decays of short lived $^{78}$Br and $^{80}$Br isotopes produced by (n,2n) reactions. The results presented in this paper are of relevance for the design of $\gamma$-ray detectors in burning plasma fusion experiments of the next generation, such as ITER, where capability to perform measurements in an intense 14 MeV neutron flux is required.

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### References


