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The light output and the detection efficiency of the liquid scintillator EJ-309

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HIGHLIGHTS
- New light output function for an EJ-309 detector and comparison with published data.
- Experimental neutron and gamma ray detection efficiencies for an EJ-309 detector.
- Comparison of measured efficiency with Monte Carlo calculations.
- Role of the light output function in neutron efficiency calculations.

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ABSTRACT
The light output response and the neutron and gamma-ray detection efficiency are determined for liquid scintillator EJ-309. The light output function is compared to those of previous studies. Experimental efficiency results are compared to predictions from GEANT4, MCNPX and PENELOPE Monte Carlo simulations. The differences associated with the use of different light output functions are discussed.

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1. Introduction

Organic liquid scintillators are commonly employed for fast-neutron detection thanks to their pulse shape discrimination (PSD) capability, to separate neutrons from the gamma-ray component of a radiation field (Knoll, 2010). The toxicity and the low flash point that characterize liquid scintillators limit their use in security applications. Moreover, the limits in detecting neutrons in a very intense gamma-ray field have been taken in the past as a weakness of those detectors that exhibit good gamma-ray detection efficiency (Kouzes et al., 2010). Recently, new liquid scintillation material, the EJ-309 type (Eljen Technology, Sweetwater Texas, USA), have become available. It is characterized by low toxicity and high flash point, compared to the more traditional ones, such as the well-known NE213. The EJ-309 scintillator has been employed in pure and applied research confirming a PSD capability well suited to perform neutron spectroscopy (Lavietes et al., 2010; Pozzi et al., 2009), while the gamma rejection power of EJ-309 has been the subject of recent studies (Stevanato et al., 2012; Swiderski et al., 2011). It is worth noting that it has been shown by Stevanato et al. (2012) that EJ-309 can be used to detect neutrons in a gamma-ray background with a dose rate of 300 μSv/h, much higher than the requirements given in Kouzes et al. (2010).

The selection of a scintillation material is generally driven by a detailed knowledge of the light output function, because it is essential to obtain the proton-recoil spectrum, hence the neutron spectrum. In this respect, the light output of EJ-309 detectors has been reported in recent studies (Enqvist et al., 2013; Takada et al., 2011), showing a light output dependence on the detector geometry (Enqvist et al., 2013).

Discrepancies between the published light output functions (Enqvist et al., 2013; Takada et al., 2011) for identical detector geometry motivated the present study. In this work, new light output function is developed and folded into Monte Carlo simulations in order to predict neutron efficiency values to compare directly to measured ones. The gamma-ray efficiency of the EJ-309 scintillator is also studied.
2. Experimental setup

In this work we studied a right-cylinder (51 mm diameter, 51 mm thick) cell, filled with EJ-309 liquid scintillator. The cell was coupled through an EJ-560 optical silicon rubber interface to a H8500 HAMAMATSU flat panel photomultiplier (PMT). The detector's technical characteristics and its performance are fully described in Cester et al. (2013). The detector assembly was placed at a distance of about 2 m from a $10^6$ fissions/s $^{252}$Cf source tagged by a 51 mm diameter, 51 mm thick plastic EJ-228 detector coupled to a PHOTONIS XP2020 photomultiplier. This fast plastic detector was positioned very close (approximately 10 mm) to the emission point to detect the burst of neutrons and gamma-rays emitted during each fission event. The front-end electronics used in this work was composed of CAEN VME modules: a V6533 Programmable HV Power Supply (6 Ch., 4 kV, 3 mA, 9 W), a Digitizer 4 Channel 10 bit 1 GS/s working in a coincidence mode, and a Bridge USB V1718 connected to a PC with a data acquisition software. The pulse height and the neutron-gamma pulse shape discrimination parameter (PSD) were obtained online by means of a digital pulse processing technique, and the time-of-flight was determined for each event offline employing a digital constant fraction discriminator technique. Details of the pulse processing procedure are given in Stevanato et al. (2012) and Cester et al. (2013).

A sample of time-of-flight spectrum measured with the aforementioned setup is illustrated in Fig. 1, showing the gamma-ray peak and the distribution of fission neutrons. The effect of performing the gamma-ray suppression by PSD is also shown, illustrating the complete disappearance of the gamma-ray peak and a significant reduction in the uncorrelated timing background. The overall timing resolution obtained with the $^{252}$Cf source is $\delta t = 0.850$ ns (FWHM), measured from the gamma-ray peak when the low energy threshold of the digital constant fraction discriminator is set at $E_{\text{th}} = 200$ keVee (i.e. in keV electron equivalent) for EJ-309.

Detector energy calibration, which is for the correct determination of the light output function, was performed using a $^{22}$Na gamma-ray source following the procedure detailed in Stevanato et al. (2011). Calibration spectra were recorded continuously during the measurements. The linearity of the setup energy response (scintillation detector and DAQ) has been tested in the energy window from 59.6 keV ($^{241}$Am) up to 4.4 MeV gamma-ray produced by an Am/Be source. As a test of the calibration procedure, Fig. 2 shows a comparison between the calibrated energy, obtained by using the $^{22}$Na transitions, and the nominal energy for the $^{241}$Am, $^{137}$Cs, $^{60}$Co (only the 1.17 MeV transition is reported) and 4.4 MeV from the Am/Be source. It is worth noting that nominal Compton edge values include the energy shift due to the finite resolution of the detection system (see Stevanato et al. (2011) for details). The full-energy peak is detected for the 59.6 keV transition of $^{241}$Am.

3. Monte Carlo simulations

Several Monte Carlo simulations were performed using GEANT4 (Agostinelli et al., 2003; Allison et al., 2006), MCNPX (version 2.7) (Pelowitz, 2011) and PENELOPE (Salvat et al., 2001) codes with the aim of comparing the measured neutron and gamma efficiencies to the simulated results, and verifying the role of the light output function in the energy range up to 7 MeV. Monte Carlo simulations offer an important tool in the design of detection system and it is therefore interesting to assess the capability of such calculation by benchmarking with experimental results.

GEANT4 offers access to particle tracking (position/time of the particle, kinetic energy, deposited energy, etc.), by means of its G4Track.hh which is used to determine the kinetic energy of the recoil protons. Each neutron history in MCNPX is followed by several charged particles (recoil protons, carbon ions, etc.) that may either fully deposit their energy or undertake partial energy

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Fig. 1. Sample of the time-of-flight spectrum measured in this work. The upper (lower) curve refers to the data without (with) gamma-ray rejection by PSD. The low energy threshold in the EJ-309 detector is $E_{\text{th}} = 200$ keVee (keV electron equivalent).

Fig. 2. Calibrated versus nominal energy. The crosses mark the $^{22}$Na Compton edges used to obtain the calibration, whereas the (-+) markers indicate the Compton edge positions of the other sources ($^{137}$Cs, $^{60}$Co (1.17 MeV) and Am/Be) and the full-energy peak position of the $^{241}$Am.

Fig. 3. Light output of our 51 mm $\times$ 51 mm EJ-309 detector compared with data from Takada et al. (2011) and Enqvist et al. (2013) relative to 127 mm $\times$ 127 mm cells.
deposition and escape from the sensitive volume of the scintillator. The light produced through the interaction of these charged particles was related to their kinetic energy via an appropriate light output formula. A total of $10^5$ neutron histories were simulated; this represents an excellent compromise having good statistics in the determination of neutron efficiency and keeping the data handling time manageable. In the PENELOPE code a mixed procedure is used for the simulation of electron and positron interactions (elastic scattering, inelastic scattering and bremsstrahlung emission), in which events with deflection angle and/or energy loss larger than pre-selected cutoffs are fully simulated and the other interactions are calculated from multiple scattering approach. Photon interactions (Rayleigh scattering, Compton scattering, photoelectric effect and electron-positron pair production) and positron annihilation are also fully reconstructed.

A sufficient number of particles ($10^5$ for MCNPX and $10^7$ for GEANT4 and PENELOPE) were tracked so that statistical uncertainty associated with the simulation reported in the next section is negligible, being lower than 1%.

4. Light output determination

A new light output function of EJ-309 was obtained using the method proposed by Kornilov et al. (2009). Analyzing the neutron fission spectrum of a $^{252}$Cf source, the pulse height distributions corresponding to different neutron energies were obtained by discriminating gamma-ray events by a proper windowing in the PSD scatter plot and then setting a 2-ns time windows on the measured time-of-flight. The uncertainty on the neutron energy reconstructed from the measured time-of-flight is discussed in detail by Kornilov et al. (2009) and depends on the uncertainty in flight path, on the neutron energy, and on the time resolution. Taking into account these effects, typical uncertainties in our study are approximately 27 keV at the neutron kinetic energy of 1.0 MeV and 300 keV at 8.2 MeV.

The first minimum in the derivative function is associated with multiple scattering events, whereas the second one at higher energy correlates with the maximum proton recoil distribution, corresponding to the average neutron kinetic energy in the time of flight window. The second minimum of the derivative function was fitted with a Gaussian distribution to determine its position.

The light output neutron energy correlation was derived in the energy range 0.54–8.2 MeV for neutrons. The light output of EJ-309 as a function of the proton recoil energy is presented in Fig. 3. Uncertainties associated with this procedure are discussed in details in Stevanato et al. (2011). The uncertainty in the light output associated with each time-of-flight bin reflects also the quality of the Gaussian fit of the second minimum, resulting in sizeable variations of the overall uncertainty, as evident in Fig. 3.

A fit to the experimental data is also reported according to the expression

$$L(E_p) = A E_p - B (1 - e^{-CE_p})$$

where the light $L(E_p)$ is given in electron equivalent units, $E_p$ is the proton energy and $A,B,C,D$ are fitted parameters.

The relevant parameters of the light output curves determined by Enqvist et al. (2013) and Takada et al. (2011) for EJ-309 are compared to our determination in Table 1. Moreover the Takada et al. (2011) light output curve and the one from Enqvist et al. (2013) relative to the same detector geometry are also reported in Fig. 3 together with our light function. The experimental data from Enqvist et al. (2013) extend to 6 MeV and extrapolation is given from 6 to 8 MeV in Fig. 3. It is worth noting that the other light output functions determined by Enqvist et al. (2013) and relative to smaller detectors, not reported in Fig. 3, are characterized by larger light outputs in the energy range 2–6 MeV, and therefore exhibit larger deviations with respect to our determination.

Light output functions from previous studies exhibit two important features: (i) a strong dependence on the size of the scintillation cells characterize the data on light yield from Enqvist et al. (2013) and Takada et al. (2011) for EJ-309; (ii) even for detectors of identical size (i.e. 127 mm × 127 mm), the two data sets show a rather large difference. It is found that our results are generally closer to the data from Takada et al. (2011), in particular for energies greater than 4 MeV, for which large discrepancies with Enqvist et al. (2013) are found. Moreover, at low proton energies up to about 3 MeV, some differences are still present between our data and both light outputs previously reported, as clearly shown in the inset of Fig. 3.

The effects of the difference in the light output, once the effects also the

<table>
<thead>
<tr>
<th>Energy threshold (MeV)</th>
<th>Measured efficiency (%)</th>
<th>Calculated efficiency GEANT4 (%)</th>
<th>Calculated efficiency MCNPX (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_{th} = 0.1$</td>
<td>33.0 ± 1.6</td>
<td>34.9</td>
<td>33.1</td>
</tr>
<tr>
<td>$E_{th} = 0.2$</td>
<td>22.0 ± 1.1</td>
<td>22.8</td>
<td>21.9</td>
</tr>
</tbody>
</table>

Table 2

Measured and predicted efficiency values for the $^{252}$Cf fission neutrons.
compared. It is clear that lower light outputs at a given neutron kinetic energy result in response functions, which are lower in energy.

5. Neutron efficiency

Neutron events from a $^{252}$Cf source, identified in the pulse shape discrimination 2-D plot, were sorted as a function of the time-of-flight in order to obtain the neutron yield as a function of energy $Y = Y(E)$. The relative neutron efficiency curve $\varepsilon = \varepsilon(E)$ was then obtained dividing the experimental distribution $Y = Y(E)$ by the spectrum of $^{252}$Cf from Cub et al. (1989). This procedure was repeated for different energy thresholds $E_{th} = 0.1$, 0.2, 0.3 and 0.4 MeVee (i.e. in MeV electron equivalent).

An absolute efficiency value was obtained from a measurement in which data were acquired by triggering on the fast plastic scintillator used to tag the $^{252}$Cf source. The number of counts registered by the plastic scintillator provided an estimate of the number of fission events. A suitable low energy threshold of $E_{th} = 0.2$ MeVee was used in the plastic in order to remove the signals resulting from isomeric states of the fission products. The neutron events in coincidence with the plastic were then used to

Fig. 5. Experimental neutron efficiency for the four different energy thresholds $E_{th} = 0.1$, 0.2, 0.3 and 0.4 MeVee (MeV electron equivalent) compared with predictions of Monte Carlo calculations.

Fig. 6. Comparison between experimental and simulated (MCNPX) neutron efficiency using different light output functions. The low energy threshold is 0.2 MeVee (MeV electron equivalent). For details see the text.

Fig. 7. Gamma-ray efficiency of the EJ-309 detector. The experimental points corresponding to the 662 keV gamma line of the $^{137}$Cs source are given.
extract the efficiency associated with the average neutron energy from the $^{252}$Cf source that is known to be 2.1 MeV. In determining the average efficiency, a neutron multiplicity value of $\nu = 3.76$ was used (Mannhart, 1986). As a quality check of this procedure it was verified that: (1) the count rate of the tag detector with a 0.2 MeV energy threshold is compatible with the expectation from the source activity and (2) the use of larger energy threshold values does not bias the resulting efficiency. The latter check demonstrates that the single spectrum of the tag detector is not contaminated by radiation sources different from fission. Absolute efficiencies for low energy thresholds $E_{\text{th}} = 0.1$ and $E_{\text{th}} = 0.2$ MeVee in the EJ-309 detector are reported in Table 2. The estimated overall uncertainty of the measured efficiency values is about 5%. Results from Monte Carlo simulations are also reported in Table 2. In this case the simulation considered a 51 mm diameter, 51 mm thick cylindrical cell made of aluminium with thickness of about 1.52 mm, filled with an organic material with a H:C ratio = 1.25 and density = 959 kg/m$^3$. Point-like monoenergetic neutron sources from 500 keV to 8.5 MeV collimated into a cone and located at 2 m from the face of the scintillator cell were simulated to derive the neutron efficiency as a function of neutron energy.

The measured efficiency values were used to normalize the energy-dependent efficiency data that are shown in Fig. 5 for energy thresholds $E_{\text{th}} = 0.1, 0.2, 0.3$ and 0.4 MeVee. GEANT4 and MCNPX simulations are also reported in Fig. 5, showing a rather good agreement with experimental data. Moreover it is worth noting that the differences between GEANT4 and MCNPX simulations are smaller than 7%, the larger differences corresponding to the maximum of the efficiency for all energy thresholds are considered. It is also interesting to note that similar results were obtained by Naem et al. (2013).

Simulations with monoenergetic neutrons were needed to construct the energy dependent efficiencies, whereas the predicted efficiency averaged over the typical fission energy spectrum was needed for comparison to the absolute efficiency data measured with the $^{252}$Cf source. Results from monoenergetic simulations were averaged to obtain an estimate corresponding to the fission spectrum.

In order to demonstrate the effect of the light output, a comparison of neutron efficiency data with MCNPX simulations for $E_{\text{th}} = 0.2$ MeVee is reported in Fig. 6. Small discrepancies in the light output curves (i.e. inset of Fig. 3) produce considerable differences in the predicted efficiency curves and, as expected, the differences are especially pronounced at energies near the threshold. This effect is clearly understood by looking at the inset of Fig. 3. The light output functions of Enqvist et al. (2013) and Takada et al. (2011) (both for 127 mm diameter, 127 mm thick detectors) predict a lower light output with respect to our evaluation for neutrons in the 1–3 MeV energy range. The relative weight of the threshold becomes ever larger with decreasing light output. In fact, as shown in Fig. 6, the effective neutron threshold curve is shifted towards larger energy values depending on the light output function used. This effect is extremely important in the case of systems used to detect fission neutrons since the 0.2 MeV energy threshold used in Fig. 6 will affect significantly the detection of neutrons with average energy of 2.1 MeV.

### 6. Gamma-ray efficiency

The gamma-ray efficiency curve of EJ-309 was obtained with the PENELOPE code (Salvat et al., 2001). Efficiency calculations with and without low-energy threshold with values of $E_{\text{th}} = 0, 100, 200, 300$ and 400 keV were performed. The results are reported in Fig. 7 and compared to data obtained with the 0.662 MeV photons of a $^{137}$Cs source. Excellent agreement between experimental data and simulation is found, for all threshold values larger than $E_{\text{th}} = 100$ keV, with PENELOPE. In Fig. 8 a comparison between the experimental $^{137}$Cs spectrum and the simulated response for the 662-keV line is shown. The simulated distribution reproduces well the experimental data for energies greater than $E_{\text{th}} = 100$ keV. However, the experimental spectrum at lower energies shows the strong photopeak associated with X-rays from $^{137}$Ba ($K_{\alpha}$ lines at about 32 keV and $K_{\beta}$ lines at about 36 keV) that are not taken into account in the simulations, thus giving rise to disagreement between the model and the experimental data.

### 7. Conclusions

A light output function for neutrons detected in the EJ-309 liquid scintillator was determined using a $^{252}$Cf source and the analysis proposed by Kornilov et al. (2009). This function shows some deviations from the data previously published by Enqvist et al. (2013) and Takada et al. (2011). When the different light output curves were folded in the Monte Carlo simulations for a 51 mm diameter, 51 mm thick detector, different response functions were generated for monoenergetic neutrons and, consequently, the predicted efficiencies do not agree, specially near-threshold energies. Good agreement between experimental neutron efficiencies and Monte Carlo simulations using GEANT4 and MCNPX was achieved when our light output function was used.

The gamma-ray efficiency predicted by the PENELOPE code was compared with the measured value at 662 keV. Excellent agreement was found for all threshold values greater than $E_{\text{th}} = 100$ keV. The dependence of the light output function on the size of the scintillator cell deserves future study.

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### References


