Nuclear Instruments and Methods in Physics Research A 784 (2015) 184–188

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Deuterated scintillators and their application to neutron spectroscopy

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ARTICLE INFO

Available online 5 November 2014

Keywords:
Deuterated scintillators
Neutron spectroscopy
Nuclear reactions

ABSTRACT

Deuterated scintillators have been used as a tool for neutron spectroscopy without Neutron Time-of-Flight (n-ToF) for more than 30 years. This article will provide a brief historical overview of the technique and current uses of deuterated scintillators in the UM-DSA and DESCANT arrays. Pulse-shape discrimination and spectrum unfolding with the maximum-likelihood expectation maximization algorithm will be discussed. Experimental unfolding and cross section results from measurements of (d,n), (^{3}He,n) and (α,n) reactions are shown.

1. Introduction

Neutron spectroscopy is an essential tool for the study of nuclei. The neutron provides a spectroscopic tool to probe nuclear interactions and structure essentially unperturbed by the Coulombic field. However, this introduces difficulties in the direct detection of neutrons and thus indirect techniques must be used. Organic scintillators have served as the primary choice for indirect detection of fast neutrons for physics applications mainly through recoil protons. They typically have high detection efficiency, fast response time (thus useful for timing applications such as neutron time-of-flight), n/γ discrimination, and are low cost. In addition to standard liquid organic scintillators such as NE-213/EJ-301, deuterated organic scintillators have shown enhanced properties compared to such hydrogen-based scintillators [1–3]. In particular, it has been shown that deuterated scintillators exhibit an advantage for spectrum unfolding [4] due to improvement in the condition of the response matrix. This improvement originates from a better defined response matrix due to the enhanced n+d elastic cross section at back angles. This yields a peak at the maximum recoil deuteron energy ($E_n = 8/9E_d$), the *recoil-peak* [2,3]. We must clarify that this nomenclature is solely based on the observation that the response appears to have, for lack of a better word, a 'peak' in the light response for incident mono-energetic neutrons. This feature is particularly useful as a quasi-quantitative identification of neutron energy groups in a spectrum prior to unfolding.

The history of such neutron spectroscopy without use of n-ToF measurement dates back more than 30 years. In a 1979 paper titled "Development of Organic Scintillators", Frank D. Brooks postulated the potential benefits of deuterated scintillators over conventional hydrogen-based scintillators for neutron spectrum measurements [5]. An early demonstration was made by P.M. Lister in his Ph.D. thesis, in which neutron spectrum unfolding was used with deuterated benzene (benzene-d_6, C_6D_6)-based liquid scintillator and deuterated anthracene (anthracene-d_{10}, $C_{14}D_{10}$) crystal scintillators [6], with results published the same year [7]. In 1988, Brooks again made a significant contribution in a publication introducing the deuterated-anthracene spectrometer (DAS) [8]. Shortly after, advantageous use of the recoil peak in deuterated liquid scintillators (NE230) was demonstrated in an experiment to search for neutrons from cold fusion [9,10]. This was a particularly novel approach to the measure of d+d fusion neutrons for which n-ToF was not possible (setup shown in Fig. 1).

Another experiment in that era involved the measurement of the energy spectrum of neutrons from decay of ^{120}Sb [11]. In this measurement, neutrons were detected in coincidence with charged particles, and the deuteron recoil peak served as the basis for determining the neutron energy spectrum. Later M. Ojaruega et al. [1] (deuterated benzene) showed that conventional pulse-shape discrimination (PSD) techniques, using discrete analog NIM and CAMAC modules, can effectively be used to study reactions such as (d,n) involving neutrons, again without the measurement of n-ToF. New arrays of deuterated scintillators are now in active use, such as the DESCANT [12] spherical array and the University of Michigan Deuterated Scintillator Array (UM-DSA) [13,14], for reaction studies with both stable and radioactive ion beams (RIBs).

In this article, we explore the use of neutron spectrum unfolding in nuclear reaction studies using the UM-DSA. Unfolding results from recent (d,n), (^{3}He,n), and (α,n) measurements are shown.

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http://dx.doi.org/10.1016/j.nima.2014.10.072
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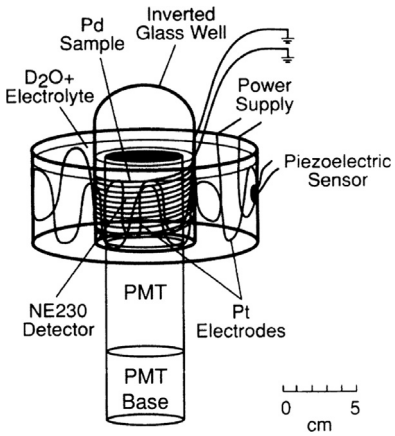

Fig. 1. Experimental setup for the detection of cold fusion neutrons (from [9]).

1.1. Description of the problem

As an indirect technique for neutron detection, organic scintillators rely on the elastic scattering of neutrons with atomic nuclei in the bulk scintillation material. The resulting recoil ion induces molecular excitation and subsequent photo emission which is detected and amplified by high-gain photo detector (PMT, APD, etc). Because each interaction results in a continuous probabilistic scattering distribution from $\theta \in (0,\pi)$, a mono-energetic neutron group results in a continuum in light-response spectra. Mathematically [13–18], this system can be described by a Fredholm integral equation of the first kind where the kernel function is the detector response function $\mathscr{R}(\ell,E)$ with incident neutron spectra $\varphi(E)$ and light response $N(\ell)$.

$$N(\ell) = \int \mathscr{R}(\ell,E)\varphi(E)dE \tag{1}$$

What we then obtain from a measurement is a superposition of individual light responses $N(\ell)$ for the incident spectra $\varphi(E)$. It is convenient to approximate Eq. (1) as a linearly-discretized matrix equation as follows,

$$\overline{S} = \overline{\overline{R}}_{\overline{x}} \tag{2}$$

where $\overline{\overline{R}}$ is known as the response matrix of the detector with incident neutron spectra $\overline{x}$ and light response $\overline{S}$. The extraction of the neutron spectrum $\overline{x}$ from the measured light-response spectrum $\overline{S}$ results in an ill-posed matrix inversion problem which must be solved. There have been many algorithms developed for solving these types of inverse problems with organic scintillators [13–18]. The resulting neutron energy spectrum obtained from spectrum unfolding is a *probabilistic distribution* of neutron counts per energy bin, thus one cannot directly correlate neutrons on an event-by-event basis without additional information. This is drastically different than n-ToF in which 'ideally' neutrons of specific energy (i.e. flight time) correlate to specific events. For example, if an experiment requires correlated observables, experimental designs using spectrum unfolding must permit projection of neutron spectra from another physical observable rather the contrary.

2. Spectrum unfolding using maximum-likelihood expectation-maximization

One particularly attractive spectrum-unfolding algorithm pertaining to experimental nuclear physics measurements is the maximum-likelihood expectation maximization (MLEM) algorithm [16]. The MLEM method starts with defining a likelihood function for the process. In the case of liquid scintillators, the likelihood function can be represented with a Poisson distribution. This representation is quite natural since it accounts for the Poisson nature of noise and photon counting statistics in the light-response spectra [16]. As derived in [16], the resulting iterative form of the MLEM algorithm take the form of Eq. 3.

$$x_j^{(k+1)} = x_j^{(k)} \sum_{i=1}^{I} R_{ij} \frac{S_i}{\sum_{l=1}^{J} R_{il} x_l^{(k)}}, \quad j = 1,\ldots,J \tag{3}$$

3. Experiments

An experimental campaign was established to evaluate the neutron spectral unfolding performance of deuterated scintillators for reaction studies as part of the UM-DSA developmental program. The campaign consisted of a series of (d,n), (^{3}He,n) and (α,n) reaction measurements on light nuclei at $E_d = 16$ MeV. Energy levels of the populated nuclei are fairly well-known and several cross-section measurements using n-ToF have been performed at similar energies. This provided an ideal set of spectra for evaluation which were then obtained at the University of Notre Dame's ISNAP laboratory. The ISNAP 10 MV FN tandem Van de Graaff accelerator was used to accelerate a deuteron beam to $E_d = 16.0$ MeV. Likewise, the accelerator was also used to accelerate ^{3}He and α beams with $E(^3He) = 16.0$ MeV and $E_\alpha = 7.5$ MeV for the (^{3}He,n) and (α,n) measurements, respectively. Beam currents were typically limited to 0.2–10.0 nA on target. To cleanly dump the beam, a 1.9 cm thick electrically-isolated graphite beam stop, which also served as Faraday cup for beam-charge integration, was fabricated. The Faraday cup was encased in a 60 cm × 60 cm × 60 cm paraffin-lined lead cave to reduce beam-induced background so as not to contaminate the neutron spectra. Borated-polyethylene pellets and plastic boron-loaded water jugs were used for additional shielding. A gold beam stop was used inside of the target chamber for the (^{3}He,n) measurement. This was done so that a 0 degree measurement could be performed to identify 0+ states. For more information regarding the experimental setup see [2].

A 5 cm × 5 cm diameter EJ-315 deuterated-benzene (C_6D_6)-based organic scintillator supplied by Eljen Technologies was used for the (d,n) measurements. A larger 12.7 cm × 12.7 cm diameter EJ-315 M enhanced-PSD deuterated-benzene-based liquid scintillator was later used for the (^{3}He,n) and (α,n) measurements. Both detectors consisted of an aluminum housing coated with TiO_2-based reflective coating (EJ-520) and a 6.3 mm Pyrex® glass window. An expansion gap 3% by volume of nitrogen gas is contained within the aluminum housing. The PMTs are magnetically shielded within a formed MuMetal® housing and operated horizontally.

4. Results and discussion

Pulse-shape discrimination was used to discriminate deuteron and electron recoils from incident neutrons and gamma rays, respectively. Each light-response spectrum $\overline{S}$ evaluated was constructed by binning the deuteron-gated spectra into a 10 keVee bin-width histograms. The incident neutron spectra were then extracted using a MLEM unfolding program written by the authors in C++ language. The program accepts an input response matrix $\overline{\overline{R}}$ (in our case from simulation using MCNP-PoliMi [19]), a light response spectrum to unfold $\overline{S}$, the maximum number of iterations to be used, the energy threshold cutoff, and if *a priori* information is to be supplied. The *a priori* information supplied is of the form of an energy range for which possible neutron groups may exist. The states populated in many reaction studies are often known, and thus this information can be used to constrain the possible solutions