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Deuterated-xylene (xylene-d_{10}; EJ301D): A new, improved deuterated liquid scintillator for neutron energy measurements without time-of-flight

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ABSTRACT

In conjunction with Eljen Technology, Inc. (Sweetwater,TX) we have designed, constructed, and evaluated a 3 in. $\times$ 3 in. deuterated-xylene organic liquid scintillator (C_8D_{10}; EJ301D) as a fast neutron detector. Similar to deuterated benzene (C_6D_6; NE230, BC537, and EJ315) this scintillator can provide good pulse-shape discrimination between neutrons and gamma rays, has good timing characteristics, and can provide a light spectrum with peaks corresponding to discrete neutron energy groups up to ca. 20 MeV. Unlike benzene-based detectors, deuterated xylene is less volatile, less toxic, is not known to be carcinogenic, has a higher flashpoint, and hence is much safer for many applications. In addition EJ301D can provide slightly more light output and better PSD than deuterated-benzene scintillators. We show that, as with deuterated-benzene scintillators, the light-response spectra can be unfolded to provide useable neutron energy spectra without need for time-of-flight (ToF). An array of these detectors arranged at many angles close to a reaction target can be much more effective ($\times$ 10 to $\times$ 100 or more) than an array of long-path ToF detectors which must utilize a narrowly-bunched and pulse-selected beam. As we demonstrate using a small Van de Graaff accelerator, measurements can thus be performed when a bunched and pulse-selected beam (as needed for time-of-flight) is not available.

1. Introduction

The detection of high-energy neutrons (i.e. a few MeV and beyond) as first done originally by James Chadwick [1], typically requires detection of a secondary, recoiling charged particle. The latter usually is a proton recoil from n+p elastic scattering although other neutron-induced reactions can occur in the detector medium. For example ^{3}He also may be used, especially if energy information of the neutron or fast timing is not needed [2,3]. In contrast to the latter, ^{1}H-based organic plastic or liquid scintillators can in principle provide information on the energy of the incident neutron either directly from the scintillation light or using a neutron time-of-flight (n-ToF) measurement given the fast nsec-level timing capability for such detectors [2,3]. In addition ^{1}H-based liquid organic scintillators such as NE213, BC501, EJ301, EJ309 and others as well as some new ^{1}H-based organic plastic scintillators such as EJ299-33 [4–6] can provide pulse-shape discrimination (PSD) to separate gamma rays from neutrons incident on the detector [2,3,7–9].

Good PSD is essential for optimal use of scintillators in certain nuclear-reaction measurements and especially for applications in nuclear non-proliferation where fission neutrons from 0.1 to several MeV must be detected and characterized in the presence of many gamma rays. Unfortunately as they are iso-spin states of the same basic nucleon, there is fore–back symmetry in n+p elastic scattering. This produces a rather featureless recoil-proton scintillation light spectrum (L) up to some maximum related to the maximum proton recoil energy and hence the energy of the incident neutron [2,3,9]. While it is possible in some cases to unfold the light spectrum to obtain the energy spectrum of the incident neutrons, this is often problematic. One then typically must use n-ToF for neutron energy measurements, often requiring a long flight path e.g. several meters for MeV-energy neutrons [2,3,9]. This becomes less feasible when low-intensity, secondary, short-lived radioactive ion beams are being studied as is now often the case at many nuclear-research accelerators e.g. at the University of

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Michigan (UM) -University of Notre Dame (UND) low-energy radioactive ion-beam facility *TwinSol* at UND [10]. At the *TwinSol* facility, we have used deuterated-benzene detectors (benzene-d_6; EJ315) for neutron measurements with both stable and short-lived, low-intensity secondary beams [11]. Likewise n-ToF experiments are more complex when the accelerator beam cannot be narrowly bunched in time and pulse selected. In such a situation, a secondary reaction particle [12] must be used to initiate the n-ToF signal, with often a significant sacrifice in overall detection efficiency.

Brooks, et al. pioneered the concept of using ^{2}H-based scintillators to remove the limitations of ^{1}H-based scintillators and in particular the limitations on unfolding the scintillation light spectra to extract the incident neutron energy spectra [13,14]. It was realized that in contrast to n+p scattering at MeV energies, the large asymmetry in the corresponding n+d elastic scattering for incident neutrons at these energies produces distinct peaks in the light spectrum. In addition to the creation of distinct peaks, these detectors provide somewhat better PSD, in principle at least, due to the larger density of ionization and hence larger pulse-height defect associated with recoiling deuterons in the scintillator relative to recoiling protons. These features *greatly* facilitate the identification of discrete neutron energy peaks and the subsequent spectral unfolding to obtain usable neutron energy spectra.

Brooks et al. originally demonstrated this effect using small deuterated-anthracene detectors [13] and later with deuterated benzene (benzene-d_6, C_6D_6), specifically NE230 [14]. Subsequently other groups including our group at UM, have exploited deuterated-benzene detectors (BC-537, NE-230), now marketed as EJ315, in a number of nuclear reaction studies using both stable and short-lived nuclear beams, as well as demonstrating applications of such detectors in nuclear security and nuclear non-proliferation [11,15–21]. However, benzene-based liquid scintillators have potential safety issues e.g. for field applications, and in particular the low flashpoint (Table 1) and its carcinogenic properties. Likewise as noted their PSD and light output are somewhat poorer than other organic scintillators such as xylene-based NE213 and EJ301. Better PSD for low-energy fission neutrons (E less than 1 MeV), and for neutron energies >10 MeV where n+d breakup can start to produce a significant number of recoil protons is important [11,18] . Breakup protons must be separated using PSD from the recoil deuterons in order to preserve the neutron peaks in the recoil-deuteron spectrum. As noted in the abstract, this generally limits the useful neutron energy region to <20 MeV. This limits the use of deuterated benzene and other deuterated detectors in some applications unless specific dopants are employed to enhance the PSD, such as in the special EJ315M detector we have previously evaluated [11,18].

Table 1
Comparison of xylene-d_{10} (EJ301D) and benzene-d_6 (EJ315,NE230,BC537)[a] scintillators.

Scintillator	EJ-301D[b]	EJ-315[c]
Chemical Formula	C_8D_{10}	C_6D_6
Light Output (% Anthracene)	78%	60%
L max	425 nm	425 nm
Blue Photons (per 1 MeVee)	12,000	9200
Ratio D to C	1.20[d]	0.99[d]
Short Decay Component	3.2 ns	3.5 ns
Index Refraction (n_D)	1.505	1.498
Flash Point	26 °C (79 °F)	−11 °C (12 °F)
Mean free path of fission neutrons[e]	4.94 cm	4.96 cm

[a] Adopted from Eljen Technology, Inc. data sheets.
[b] Xylene based (custom fabricated by Eljen Technology, Inc.).
[c] Benzene based.
[d] Includes carbon from the added ($<5\%$) fluor compound.
[e] Mean free path averaged over ^{252}Cf fission-neutron spectrum.

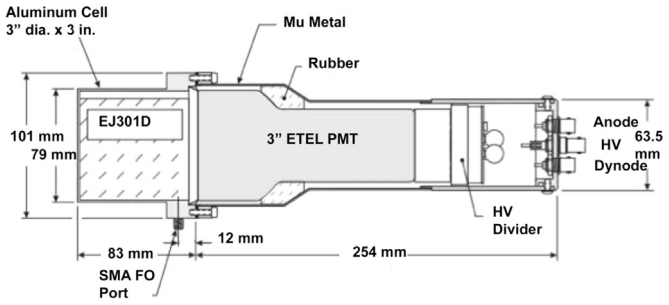

Fig. 1. Schematic of the Eljen 3 × 3 EJ301D (xylene-d_{10}) custom detector assembly.

Working with C. Hurlbut and colleagues at Eljen Technology, Inc. (Sweetwater TX) we have designed, constructed and evaluated a prototype 3 in. dia. × 3 in. (ca. 75 mm dia. × 75 mm) xylene-d_{10} liquid scintillator (denoted here as 3 × 3 EJ301D; Fig. 1). While presently about twice as expensive as a similar deuterated-benzene based scintillator, the improved performance and added safety may justify the increased cost in many applications. (The formulation of deuterated scintillators requires that all solvents and additives, where possible, also be highly-enriched deuterated compounds and this is an expensive process relative to producing normal ^{1}H-based scintillators). The custom EJ301D detector cell, PMT (a 3 inch dia., ETEL 9821KEB), and PMT base are similar to the other Eljen detectors (EJ315 in particular) we have designed and have in active use. The design includes an SMA fiber optic port at the cell-PMT interface (Fig. 1) for insertion of a stabilized LED light pulser signal.

In parallel with the evaluation of the EJ301D detector we also have for comparison data obtained for several xylene ^{1}H-based EJ301 detectors of similar design (including the PMT and base) as well as several EJ315 and EJ315H (^{1}H-based version of EJ315) detectors of similar size and design. This includes a 3 in. dia. × 2 in. EJ315 detector from Scionix, Inc. (which we will denote as 3 × 2 EJ315) that will be used here to compare with many of the 3 × 3 EJ301D detector measurements.

The characteristics of the EJ301D and EJ315 deuterated scintillator liquids are shown in Table 1. In addition to the different flashpoints, the xylene-based scintillator has slightly more light output and more ^{2}H content relative to carbon than a benzene-based scintillator which should yield a somewhat better PSD and intrinsic neutron detection efficiency for a given detector volume. However this can be impacted by the efficiency and intrinsic resolution of the light collection for a specific detector+PMT combination (see below).

2. PSD and neutron scintillation spectrum from ^{252}Cf fission neutron sources

Initial PSD and spectrum measurements were performed using various commercial ^{252}Cf fission sources that were previously available, or ones recently purchased specifically for scintillator evaluations having accurately-known activity. This included a very intense ^{252}Cf fission source (5.63 mCi). Owing to the gas-filled expansion volume inside the cell (about 3% of the volume), the Eljen detector is operated horizontally. This minimizes interference of the gas expansion volume (Fig. 1) with the light collection if the detector were operated vertically, e.g. face down with the gas volume at the PMT face [11].

The initial measurements of the PSD and ^{252}Cf fission-neutron spectra were obtained using the digital signal processing (DSP)