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C^7LYC: A NEW SCINTILLATOR FOR FAST NEUTRON SPECTROSCOPY*

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The scintillator Cs_2LiYCl_6 (CLYC) has emerged as a versatile detector for both gammas and neutrons, with excellent pulse shape discrimination. Originally developed as a thermal neutron counter, the discovery of its unexpected and unprecedented $\sim 10\%$ pulse height resolution for fast neutrons in the few MeV range has prompted studies to benchmark its use in low-energy nuclear science and applications. Since the typical long time-of-flight arms are not needed for achieving good energy resolution, geometrical efficiency can be enhanced by positioning the detectors much closer to the target. We have constructed a 16-element array of 1" $\times$ 1" ^{7}Li-enriched C^7LYC, to eliminate the dominant peak in the spectrum from thermal neutron capture on ^{6}Li, leaving the energy region above 3 MeV with a clean baseline for fast neutron spectroscopy. We have also procured the first ever 3" $\times$ 3" C^7LYC crystal. Test experiments under way with C^7LYC include elastic and inelastic neutron scattering cross sections at Los Alamos with a pulsed white neutron source, efficiency measurements using mono-energetic neutron beams up to a few MeV at UMass Lowell, and fission neutron measurements with GRETINA and CHICO detector arrays at Argonne. Beta-delayed neutron spectroscopy experiments have also been initiated at the CARIBU and NSCL facilities, to evaluate C^7LYC as a possible candidate for auxiliary scintillator arrays for stopped beam physics at the next generation rare isotope accelerators.

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1. Introduction

The detection and spectroscopy of neutrons are currently enjoying renewed interest in both fundamental and applied nuclear science. In nuclear structure and nuclear astrophysics, for example, there is a new excitement in

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being able to venture deeper into neutron-rich *terra incognita* in the nuclear chart through new facilities for rare isotope beams. Strength functions, neutron capture rates and r-process freeze-out scenarios are hot topics in the present-day nuclear astrophysics [1]. These studies of neutron-rich nuclei far from stability would involve more sophisticated techniques of beta-delayed neutron spectroscopy than has been possible earlier. In the applied nuclear sciences, beta-delayed neutrons have traditionally played a key role in reactor decay heat and control, while precise measurements of neutron scattering cross sections continue to be a priority for the nuclear energy and reactor design communities [2]. In addition, fission neutron multiplicities are still not measured/understood at a satisfactory level, and new neutron detection systems and arrays continue to be built to improve the experimental status [3]. Neutron detection is also being seriously considered for homeland security applications such as cargo inspection for special nuclear materials [4]. Finally, neutron detectors are being deployed in space explorations where cosmic ray interactions are used to infer the presence of water (or hydrogen) on our moon and planets [5, 6].

Neutrons, being uncharged, are more difficult to detect compared to charged particles or photons, since they do not interact with matter via electromagnetic interactions. Since neutron capture cross sections are low, typical neutron detection relies on the transfer of energy from the neutron to charged particles through scattering processes. These interactions, however, rarely capture the full energy of the incident neutron, and traditional "spectroscopy" of neutrons, especially of fast neutrons, relies on time-of-flight (TOF) techniques. These necessarily require a valid "start" signal for the reaction and the neutron detector to provide the "stop" signal. The energy resolution of this method depends on the distance of the detector from the source of neutrons, the farther the better. This leads to either a loss in geometrical efficiency or increased cost in covering the available solid angle around the neutron source.

New scintillators have recently emerged with capabilities that make them worth exploring as alternatives to the *status quo*. This work details the emergence of the dual neutron–gamma inorganic scintillator Cs_2LiYCl_6 (or CLYC) as an attractive candidate for fast neutron spectroscopy, and our recent efforts to benchmark its effectiveness in a variety of applications.

2. History

CLYC made its entry into the scintillator market roughly two decades ago as a versatile detector that can detect both gamma rays and neutrons [7]. The two quanta can be distinguished very effectively via digital signal processing since there is excellent separation between the two pulse shapes [8].

The gamma response and resolution of CLYC is better than sodium iodide, a scintillator that has for decades served as a benchmark and gamma-detection workhorse of the spectroscopy community. The neutron pulses have a slower rise and fall times compared to the gamma pulses, and the two can be very well discriminated by integrating the charge in two adjacent appropriately chosen time windows [8–10].

CLYC was originally developed as a thermal neutron detector, with the intrinsic neutron detection efficiency arising from the comparatively high cross section of the ^{6}Li$(n, \alpha)t$ reaction. Subsequent independent work by both our group [9] and others [12] led to the discovery of an unexpected $\sim 10\%$ pulse height resolution for fast neutrons in the few MeV range with a linear response via the ^{35}Cl$(n, p)^{35}$S reaction [11, 12]. This unprecedented intrinsic energy resolution introduces a new paradigm in fast neutron spectroscopy, as it obviates the need for long TOF arms, and allows placement of neutron detectors much closer to the source, thereby increasing geometrical efficiency significantly.

Following detailed characterization and optimization of the response of CLYC to fast neutrons over a range of energies at multiple accelerator facilities, we chose to focus on developing and benchmarking CLYC specifically for fast neutron spectroscopy in the few MeV range. This energy range encompasses much of the physics we are interested in both fundamental and applied science, from spectroscopy of beta-delayed and fission neutrons to reaction cross sections of elastic and inelastic neutron scattering. A dominant thermal neutron "peak" appears in CLYC spectra at a gamma-equivalent energy of 3.5 MeV, which arises from the Q-value of the ^{6}Li$(n, \alpha)t$ reaction tempered by a quenching factor for the light output of the alpha and triton. This peak appears in a prime energy region where we would like to keep a clean baseline for fast neutron spectroscopy. To focus exclusively on the spectroscopy fast neutrons, we chose to eliminate the significantly stronger thermal neutron response by modifying the lithium isotope content in CLYC, depleting the ^{6}Li and using $> 99.9\%$ ^{7}Li (C^{7}LYC) [11].

An additional advantage of CLYC is that while traditional neutron detectors do not have gamma-spectroscopy capabilities, the better-than-NaI gamma response of CLYC opens up the possibility of using the same detector for both gamma and neutron spectroscopy. We have constructed a Small C^{7}LYC Array for Neutron Spectroscopy (SCANS), a 16-element array of 1" $\times$ 1" ^{7}Li-enriched C^{7}LYC (the largest crystals that were being grown at the time of purchase) [11, 13], and have subsequently also procured the first ever 3" $\times$ 3" C^{7}LYC crystal (Fig. 1). Using these detectors, we have initiated studies to benchmark the use of C^{7}LYC in low-energy nuclear science and applications. A key goal is to evaluate how the comparatively low intrinsic efficiency of C^{7}LYC for fast neutrons ($< 1\%$) can be effectively offset by the gain in solid angle obtained by positioning the detectors much closer to the neutron source.

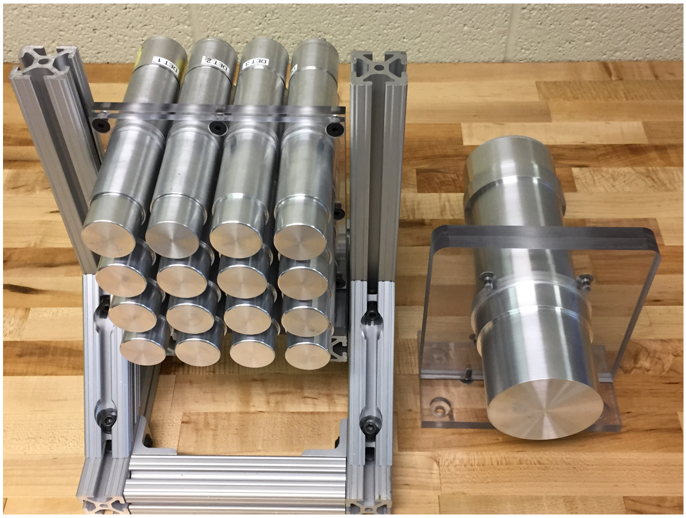

Fig. 1. The SCANS array (16 1" $\times$ 1") and the first 3" $\times$ 3" C^{7}LYC detectors.

3. Experiments

We report here on the status of three different avenues of exploration and benchmarking that we have embarked on to evaluate the utility of C^{7}LYC detectors in fast neutron spectroscopy. Some of the work described here has been accepted for publication in other peer-reviewed journals, and the reader is referred to those publications for further details.

3.1. Neutron scattering cross sections

Our first efforts at evaluating the application potential of C^{7}LYC have been to measure both elastic and inelastic neutron scattering cross sections from targets that are identified as high priority by the nuclear data community. We chose to start with the well-studied ^{56}Fe(n, n') reaction, and compare our results [13] to recent measurements of the same reaction with traditional tools using TOF techniques [14]. Our experiments were performed at the Weapons Neutron Research facility of the Los Alamos Neutron Science Center, where a white neutron beam with energies up to a few hundred MeV is generated by bombarding a tungsten spallation target with 800 MeV protons. The beam is pulsed with a < 1 ns time structure and a period of ~ 1.8 μs. The ^{56}Fe scattering target was situated ~ 20 m from the tungsten neutron source. The C^7LYC detectors of the SCANS array were placed in an angular range of 30° to 150° at $\sim$ 17 cm from the ^{56}Fe target. The TOF of the neutrons from the tungsten neutron source to the C^7LYC detectors (with appropriate minor corrections for the angle-dependent neutron scatters to the individual detectors) was used to define the incident neutron energy. The scattered neutron energy was obtained from the integrated charge in the C^7LYC detectors with a $\sim$ 10% resolution. Plotting the scattered neutron energy as a function of incident neutron energy clearly revealed a strong elastic scattering streak along a diagonal through the origin ($E_{\rm scattered} = E_{\rm incident}$), as well as a parallel streak ($E_{\rm scattered} < E_{\rm incident}$) for the inelastic scattering peak corresponding to the first excited state of ^{56}Fe at 847 keV [13]. Energy-dependent elastic and inelastic scattering yields were extracted from 100-keV wide bins of the incident neutron energy.

Extraction of the absolute energy-dependent elastic and inelastic scattering cross sections was not attempted in this first experiment since the efficiency of the detector as a function of neutron energy has not yet been firmly established, either through experiments or through simulations, the latter suffering from inadequate cross-section data for the $^{35}\mathrm{Cl}(n,p)^{35}\mathrm{S}$ reaction in current evaluated databases. Instead, the *relative* elastic and inelastic scattering yields as a function of angle were mapped out for incident neutron energies from below 1 MeV up to 5 MeV. The time interval between incident neutron pulses results in a lower cut-off in the neutron energy of $\sim$ 700 keVee (gamma- or electron-equivalent energy), below which the slowest neutrons of one beam pulse are overtaken by the fastest quanta from the succeeding pulse. The extracted angular distribution results were compared to recent high-resolution measurements in the same energy range [14], using a *single* arbitrary normalization constant for each energy bin for both elastic and inelastic data. Remarkable agreement is observed in the incident energy range of $\sim$ 2 to 3 MeV [13], where the intrinsic efficiency has a maximum and is, therefore, approximately flat [11]. This ensures that the intrinsic efficiency does not change significantly for the neutron energy difference between the elastic and inelastic channels. A more detailed comparison over the full energy range up to 5 MeV will have to await a better handle on the energy-dependent intrinsic efficiency measurements described in the next section.

3.2. Intrinsic efficiency measurements

Fast mono-energetic neutrons up to $\approx$ 3 MeV can be generated at the 5.5 MV Van de Graaff accelerator at UMass Lowell's Radiation Laboratory via the $^7\mathrm{Li}(p,n)^7\mathrm{Be}$ reaction, where protons are incident on a thin layer of ^{7}Li evaporated *in situ* under vacuum on a Ta foil [15]. For every neutron produced, a ^{7}Be nucleus is also produced. ^{7}Be decays back to ^{7}Li with a 53-day half-life via a $\sim$ 10% gamma-decay branch through a 479-keV transition. Thus, an assay of the Ta foil coated with the ^{7}Be residues after short irradiations of a few hours, using a germanium detector with well-calibrated efficiency, provides an excellent measure of the total neutrons generated during the experiment. Folding in the angular distribution of the neutron emission from the target as well as the geometrical efficiency of the C^{7}LYC detector placement provides the number of neutrons incident on the detector. Integrating the ^{35}Cl$(n,p)^{35}$S peak in the C^{7}LYC spectrum provides the number of neutrons detected. The ratio of these two quantities is a direct measure of the intrinsic neutron detection efficiency of the detector at a given incident neutron energy. Detailed measurements are currently in progress [13] for mono-energetic neutron energies from a few hundred keV to a few MeV. Preliminary results indicate that the intrinsic "spectroscopic" efficiency is $\sim$ 0.5% for a 1" $\times$ 1" crystal for 2 MeV neutrons, and that the efficiency increases approximately linearly with crystal depth.

3.3. Beta-delayed neutrons

Testing the capabilities of C^{7}LYC detectors for beta-delayed neutron spectroscopy is one of our high science priorities, as it is and will be a key measurement for neutron-rich nuclei far from stability. Our first attempt was at the CARIBU facility at Argonne [16] to study the beta-decay of ^{94}Rb, which has a $\sim$ 10% beta-delayed neutron branch. The set-up involved the SATURN tape-transport system and the X-Array consisting of five Ge-clover detectors [17]. The radioactive ^{94}Rb extracted from the $\sim$ 1 Ci ^{252}Cf fission source was implanted onto the tape and transported to the center of a near-4π plastic detector that is surrounded by the X-Array in a box geometry. We replaced one of the side clovers with our 16-element SCANS array of 1" $\times$ 1" C^{7}LYC detectors.

While the high-resolution gamma spectroscopy part of the experiment was successful, the neutron detection part was compromised partly due to the low yield of ^{94}Rb ions, and mostly due to the proximity of the $\sim$ 1 Ci ^{252}Cf source cask, which generated significant gamma and neutron background in spite of the available shielding. This shortcoming has now been addressed by moving the entire CARIBU low-energy decay station to a different vault area away from the ^{252}Cf source, where the measured gamma background is reduced by three orders of magnitude, with concomitant reduction in neutron background. The experiment is now planned to be repeated with our new 3" $\times$ 3" C^{7}LYC, which is almost twice as efficient overall as the entire SCANS array, and can also be placed much closer to the chamber than the SCANS array, with a significant increase in geometrical efficiency.

We have also recently conducted preliminary tests at NSCL using their beta-decay end station, piggy-backing on an experiment where an array of $LaBr_3$ detectors had been positioned with a HPGe array to measure lifetimes in neutron-rich nuclei around the $N = 20$ island of inversion via fragmentation of a ^{48}Ca beam. Beta-delayed neutron branches were present at or above $\sim 50\%$ in a number of reaction channels. Some C^7LYC detectors replaced an equivalent number of $LaBr_3$ detectors in the set-up for a small fraction of the experiment. Although this experiment was primarily to test the inclusion of C^7LYC detectors in the digital DAQ infrastructure at NSCL, an array of, say, four 3" $\times$ 3" C^7LYC detectors (a goal that is easily realizable with current technology) would be able to register a few thousand counts in individual reaction channels with the 80 pnA of ^{48}Ca beam that was used for this experiment. We plan to propose specific experiments with C^7LYC detectors at NSCL following analysis of this data set.

3.4. Fission neutrons

Another important application of C^7LYC is in the spectroscopy of fission neutrons, whose spectra peak in the $\sim$ 1–2 MeV range for fissile materials. Although fission was discovered relatively early in the development of nuclear physics and plays a dominant role in both nuclear energy as well as nuclear defense and deterrent sectors, the science of fission is still far from being satisfactorily understood. Measurements of neutron multiplicities and neutron spectra for both spontaneous and induced fission continue to be improved and repeated as new technologies in neutron detection emerge.

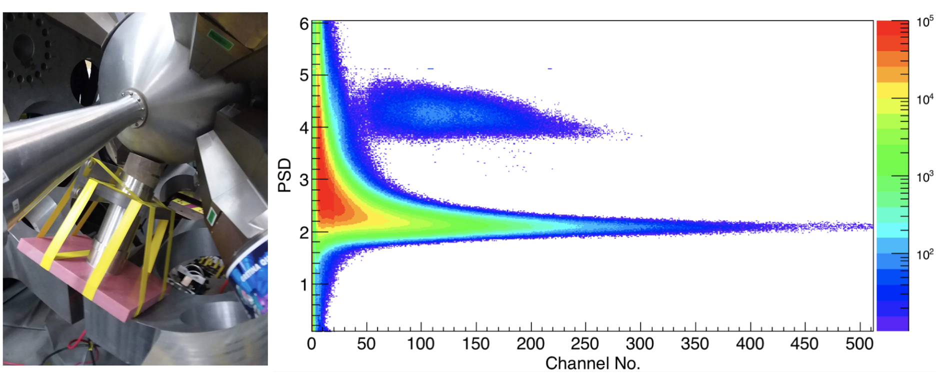

Fig. 2. (left) The 3" $\times$ 3" C^7LYC detector coupled with the GRETINA and CHICO arrays for fission neutron spectroscopy tests; (right) PSD plot showing the separation between neutrons (top) and gammas (bottom).

To this end, we have very recently acquired a first dataset using a $\sim$ 12 μCi ^{252}Cf fission source with our 3" $\times$ 3" C^{7}LYC detector, in conjunction with the GRETINA array of segmented HPGe tracking detectors, and the position-sensitive parallel-plate avalanche counter array CHICO for detecting the binary fission fragments. The 100 MHz digitizers of GRETINA were used for processing the C^{7}LYC pulses. The experimental set-up and the PSD plot for gammas and neutrons are shown in Fig. 2. This data is currently under analysis.

4. Machine learning n–γ discrimination

Pulse shape discrimination (PSD) between neutrons and gamma rays in a CLYC scintillator is a binary classification scheme that is a common goal of machine learning algorithms. Supervised learning algorithms such as artificial neural network analysis have already shown promise for PSD in liquid scintillators [18]. These require training data sets that can be generated using the usual charge comparison methods. On the other hand, cluster analysis algorithms, such as k-means++ [19], provide powerful machine learning techniques that are unsupervised and do not need to be "trained". We have utilized both of these machine learning approaches to compare their efficacy with charge comparison techniques. In this work, which has been reported recently [20], we find that both techniques are able to effectively discriminate between gamma and neutron pulse shapes in the energy range investigated. Machine learning can also be used to guide the choice of optimal parameters, *e.g.* gate widths, for charge comparison techniques [20].

5. Conclusions and future work

In summary, C^{7}LYC shows promise as a versatile dual neutron–gamma scintillator with excellent PSD and the unique distinction of $\sim$10% intrinsic spectroscopy-grade neutron energy resolution without TOF. Measurements of neutron scattering cross sections at Los Alamos have been very successful, and further measurements are being analyzed and/or proposed. Direct efficiency measurements of both 1" $\times$ 1" and 3" $\times$ 3" C^{7}LYC crystals are under way at the Van de Graaff accelerator at UMass Lowell. Detailed simulations are awaiting results of cross-section measurements of the ^{35}Cl$(n,p)^{35}$S reaction at Los Alamos. A first data set with a ^{252}Cf fission source has been obtained with the 3" $\times$ 3" C^{7}LYC coupled with the GRETINA and CHICO arrays. Following first attempts and preliminary measurements, beta-delayed neutron spectroscopy experiments are planned at both the CARIBU facility at ANL and the decay station at NSCL/MSU. These studies are aimed at assessing the viability of C^{7}LYC as a candidate for future compact scintillator arrays for stopped-beam physics at upcoming rare isotope facilities such as FRIB.

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