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Lithium indium diselenide: A new scintillator for neutron imaging

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ABSTRACT

Lithium indium diselenide, $^{6}LiInSe_2$ or LISe, is a newly developed neutron detection material that shows both semiconducting and scintillating properties. This paper reports on the performance of scintillating LISe crystals for its potential use as a converter screen for cold neutron imaging. The spatial resolution of LISe, determined using a 10% threshold of the Modulation Transfer Function (MTF), was found to not scale linearly with thickness. Crystals having a thickness of 450 μm or larger resulted in an average spatial resolution of 67 μm, and the thinner crystals exhibited an increase in spatial resolution down to the Nyquist frequency of the CCD. The highest measured spatial resolution of 198 μm thick LISe (27 μm) outperforms a commercial 50 μm thick ZnS(Cu):^{6}LiF scintillation screen by more than a factor of three. For the LISe dimensions considered in this study, it was found that the light yield of LISe did not scale with its thickness. However, absorption measurements indicate that the ^{6}Li concentration is uniform and the neutron absorption efficiency of LISe as a function of thickness follows general nuclear theory. This suggests that the differences in apparent brightness observed for the LISe samples investigated may be due to a combination of secondary charged particle escape, scintillation light transport in the bulk and across the LISe-air interface, and variations in the activation of the scintillation mechanism. Finally, it was found that the presence of ^{115}In and its long-lived ^{116}In activation product did not result in ghosting (memory of past neutron exposure), demonstrating potential of LISe for imaging transient systems.

1. Introduction

The interactions of cold neutrons with matter is a function of nuclear cross sections and crystalline structure [1], enabling neutrons to probe matter in a variety of ways. One such way is cold neutron imaging, where generated radiographs (or 3D images) reveal information about the interior or structural properties of imaged materials and devices [2]. Many modern thermal/cold neutron imaging systems utilize a scintillation screen doped with a neutron-sensitive isotope (e.g., ^{6}Li, ^{10}B, ^{nat}Gd) coupled to a light collection apparatus. The neutron interaction results in secondary charged particles that result in light production via scintillation. The range of secondary particles and thickness of the scintillator are critical factors in the spatial resolution of the imaging system. The range of secondary particles defines the intrinsic point spread function of the converter, and the thickness of the converter/scintillation screen results in a spread of the scintillation photons during internal transport to the surface. The result is overlapping point spread functions at the digitizer plane (e.g., CCD camera) generated from adjacent points, thereby blurring the image [3]. Therefore, the ideal neutron converter yields a narrow intrinsic point spread function (range of secondary charged particles), a large detection efficiency per unit path length, and generates a large number of light photons per neutron interaction. These characteristics limit both image blurring from thick scintillators and exposure time to gain the necessary signal-to-noise ratio (SNR) [4].

A general rule of thumb for scintillator-based converters assumes that the achievable spatial resolution is limited to the thickness of the scintillation screen, such as ZnS:^{6}LiF scintillation screens. Gadolinium-based scintillation screens are currently being explored for high resolution imaging because of its unparalleled neutron cross section and low energy of the conversion electrons. However, the gamma-rays produced result in image

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degradation and errors associated with false neutron counting [5]. Still, efforts are underway to increase the spatial resolution using Gadolinium-based scintillations screens by using micron-sized, ^{157}Gd enriched, scintillation screens [6]. Preliminary results have demonstrated a resolution of 7.6 μm [7] and recent results have yielded a resolution of 5.4 μm [8]. Another group has utilized infinity-corrected optics and an alternative Gadolinium-containing scintillation screen ($Gd_3Ga_5O_{12}$:Eu^{3+}), called GGG, and demonstrated a spatial resolution of 14.8 μm [9]. It should also be noted that GGG is optically transparent, and their experiment was conducted with a 1 mm thick scintillator. This result indicates that the resolution achieved was much higher than the thickness of scintillator, which is not possible on this scale using polycrystalline or epoxied scintillators, such as ZnS:^{6}LiF scintillation screens.

In an effort to identify additional converter screens that do not exhibit the dependence of spatial resolution on thickness while also maintaining all other desired attributes for neutron imaging, a new scintillating material, lithium indium diselenide (LISe), was investigated in this study [10]. Enriched in ^{6}Li to 95 at%, LISe provides the needed absorption efficiency and is optically transparent, demonstrating potential for overcoming limitations with existing neutron imaging converter materials. This paper reports on the efficacy of LISe for neutron imaging, considering light yield, neutron absorption efficiency, and spatial resolution as a function of scintillator thickness and surface finish.

2. Lithium indium diselenide

Lithium indium diselenide, or LISe, is a I-III-VI_2 chalcopyrite with neutron detection capability via the ^{6}Li(n,t)^{4}He reaction with a large Q-value of 4.78 MeV. The light yield of LISe has been determined to be 9,000 photons/MeV with a peak wavelength emission at 512 nm and a principle decay time constant of 31 ± 1 ns [11]. The mechanism of scintillation has yet to be identified, but appears to be reproducible. The 24 at% ^{6}Li concentration in LISe yields a mean free path for thermal neutrons of 920 μm, but the presence of ^{115}In reduces the ^{6}Li-based reactions to an effective absorption efficiency of 82% at thermal energy. The thermal neutron mean free path of LISe is smaller than any other lithium-containing solid-state scintillator known to the authors, and the high cold neutron absorption efficiency holds promise for its application to neutron imaging. However, the decay time constant of ^{116}In and the associated ionizing radiation emitted may limit the efficacy of LISe in dynamic neutron imaging studies, and is evaluated in this paper.

3. Experimental procedure

3.1. Neutron absorption efficiency

Experiments were conducted at the Oak Ridge National Laboratory (ORNL) High Flux Isotope Reactor (HFIR) CG-1D neutron imaging test station and beam line [12] and at the beam line for Neutron Optics and other Approaches (BOA) at Paul Scherrer Institut (PSI) [13]. Neutron absorption, related to detection efficiency, for each LISe sample was determined by evaluating the attenuation through LISe using a 50 μm ZnS(Cu):^{6}LiF scintillation screen. After open beam and dark field image corrections, the measured absorption, I_{abs}, is converted into absorption efficiency, ε, using Eq. (1), where I_o is the measured intensity at a given location from corrected open beam image, x is the thickness of the absorber/detector, and λ is the neutron mean free path.

$$\varepsilon = \frac{I_o - I_{abs}}{I_o} = 1 - e^{-x/\lambda} \quad (1)$$

3.2. LISe relative light yield (RLY)

The scintillation homogeneity and intensity for each sample was investigated by mounting the LISe samples onto a thin aluminum support plate and placing them in the position of the ZnS(Cu):^{6}LiF scintillation screen. At each beam line, the available optical transfer and digitation systems were utilized. At HFIR, two optical systems were used. The first utilizes lenses and a 45° mirror to focus onto a CCD camera (DW 936N-BV iKon-L ANDOR^{TM}). The second setup at BOA is similar to the first, but uses an ANDOR^{TM} Zyla 4.2 sCMOS. Both imaging systems can be used with 25 to 200 μm ZnS(Cu):^{6}LiF (2:1 ratio) convertor screens (a 50 μm thick scintillator was used for measurements reported in this study) deposited on an aluminum support plate with a peak emission at 530 nm from RC TRITEC. The relative light yield (RLY) measured by each sample was determined by dividing the acquired open beam images with each LISe sample by the response of the 50 μm ZnS(Cu):^{6}LiF converter. This serves to both remove the contribution of any variations of the neutron beam intensity and provide a relative measure of its light yield compared to a commonly used neutron imaging scintillator.

3.3. Spatial resolution

The spatial resolution of all LISe samples was determined using the knife-edge technique [14] or visual inspection with the PSI Siemens Star test pattern [15]. For the knife-edge technique, the sharp edge of a neutron absorbing mask in the presence of a uniform neutron field results in a predictable profile response called the edge spread function (ESF). The ESF is the convolution of the line spread function (LSF) and neutron intensity profile across the absorbing edge [14]. For step profiles, differentiation of the ESF results in the LSF. Assuming that the neutron imaging system is shift invariant, the width of the LSF profile describes the width of the system point spread function (PSF), which is a fundamental quantity used to describe the resolution of imaging systems.

The imaging system PSF is equal to the convolution of the various response functions from its components, which includes the beam divergence (geometric unsharpness), scattering degradation (knife-edge unsharpness), scintillator unsharpness, and the finite sampling frequency of the digitizer [14]. Further, the LISe scintillators tested are very thick (0.2–1 mm) when compared to typical ZnS(Cu):^{6}LiF screens, but also optically clear, which may lead to a defocusing contribution from the optical transfer between the scintillator and CCD. The gadolinium knife-edges used in these tests are thin-films, and owing to the current technological capabilities of lithographic printing, scattering degradation can be ignored. Therefore, the relevant contributing components of the imaging system are the scintillator unsharpness, beam divergence, and the optical transfer and digitization system (i.e., the lenses and CCD).

For an ideal cold neutron imaging beam line, the beam divergence is a function of the beam aperture diameter and aperture-object and object-scintillator distances. These parameters define the angular distribution of neutrons at the object plane and can be represented as a boxcar function in one dimension. The governing equation for the beam divergence, h_B, in one dimension is [16]

$$h_B(x) = \mathrm{rect}\left(\frac{x' - x_o'}{2 \cdot d \cdot Tan(\theta)}\right), \quad (2)$$

where x_o' is the location of the knife-edge in the object plane, x' is the position on the converter plane, d is the distance between the