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ZnO:Zn/^{6}LiF scintillator—A low afterglow alternative to ZnS:Ag/^{6}LiF for thermal neutron detection

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ABSTRACT

Current ZnS:Ag/^{6}LiF based scintillation detectors are often count rate limited by the long lifetime afterglow in the scintillator. Despite this drawback, new instruments at neutron scattering facilities, like ISIS in the UK, would still like to use ZnS:Ag/^{6}LiF detectors due to their low gamma sensitivity, high light output, simplicity of detector design and relatively inexpensive production. One particular advantage of ZnS:Ag/^{6}LiF detectors is their ability to provide strong pulse shape discrimination between neutrons and gammas. Despite the advantages of these detectors, it is becoming clear that new and upgraded instruments will be limited by the count rate capability of ZnS:Ag/^{6}LiF, so an alternative scintillator technology with equivalent simplicity is being sought. ZnO:Zn/^{6}LiF is investigated here as a low afterglow alternative to ZnS:Ag/^{6}LiF. Basic scintillation properties of ZnO:Zn are studied and are discussed. Pulse shape discrimination between neutrons and gammas is explored and taken advantage of through simple single photon counting methods. A further step toward a realistic detector for neutron scattering is also taken by fiber coupling the ZnO:Zn/^{6}LiF to a PMT. In an initial study of this fiber coupled configuration, ^{60}Co gamma sensitivity of $\sim 7 \times 10^{-6}$ is shown and improvements in count rate capability of at least a factor of 6 over ZnS:Ag/^{6}LiF based neutron detectors are demonstrated.

1. Introduction

Recent advances in neutron scattering facilities have led to a search for detectors that can cope with increasing neutron count rates [1–4]. At the present time, approximately half of the neutron instruments at the ISIS neutron and muon scattering facility in the United Kingdom [3] employ ZnS:Ag/^{6}LiF based neutron detectors [4] due to their low gamma sensitivity [5], simplicity of detector design and ability to adapt the design to many different neutron scattering applications [6,7].

Powder diffractometers on existing facilities typically operate within the count rate capability of the ZnS:Ag/^{6}LiF detectors [4]. In this case, the rate of neutrons incident on the detector is limited by a number of factors including the randomization of polycrystalline powders that spread the diffracted neutrons across large areas of the detectors in Debye Scherrer cones, large sample to detector distances, relatively low incident flux on the samples and collimation used to limit the field of view of the detectors to small Gauge volumes of the samples. Samples that tend away from pure powders (quasi-single crystals or single crystals) concentrate diffracted neutrons into more localized areas instead of cones. In this situation, the count rate capability of ZnS:Ag/^{6}LiF detectors is a limiting factor. Other neutron scattering techniques such as small angle neutron scattering (SANS) and reflectometry at existing facilities are currently restricted by count rate limitations in both conventional ^{3}He gas tubes and ZnS:Ag/^{6}LiF detectors. For small angle neutron scattering (SANS), neutrons are elastically scattered at shallow angles, thus resulting in neutrons being concentrated in small rings with the neutron intensity being dependent on the sample to detector distance [8]. In both SANS and neutron reflectometry, ZnS:Ag/^{6}LiF detectors are unable to cope with the direct beam.

New neutron scattering facilities and upgrades to existing facilities promise to deliver up to 100 times the flux available on current instruments [2]. Powder diffractometers at the European Spallation Source (ESS) are estimated to give between a factor of 2 and 20 increase in flux over the existing WISH diffractometer at ISIS [9]. Such increases in flux will challenge the rate capability of ZnS:Ag/^{6}LiF and large area ^{3}He gas detectors, thus requiring alternative detector solutions.

The primary decay of ZnS:Ag is on the order of 200 ns which is fast enough for almost any neutron scattering application. The problem is that ZnS:Ag/^{6}LiF based detectors are count rate limited by the long lifetime afterglow in the scintillator, which lasts up to several hundred microseconds. Opacity of traditional ZnS:Ag/^{6}LiF screens exacerbates

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the problem of afterglow. To achieve acceptable neutron detection efficiencies, discrimination thresholds are set to values corresponding to as low as 8 detected photo-electrons [5]. For bright events, afterglow occurring many microseconds following the primary decay will result in more than 8 photo-electrons being detected. This will cause such events to be counted multiple times and other events to be misplaced [10]. In ZnS:Ag/^{6}LiF detectors it is therefore a requirement of the signal processing to sufficiently suppress afterglow. Multi-count and position error suppression is typically accomplished by extending detector dead times leading to lower count rate capabilities and/or lower detector efficiencies. New facilities and upgrades to existing facilities will be limited by the count rate capability of the detectors, so an alternative scintillator technology with equivalent simplicity is being sought.

Most proposed alternative scintillators to ZnS:Ag have significant disadvantages for use in neutron scattering applications. For example, widely used lithiated glasses have low light yield and are too gamma sensitive for most applications [11]. Though pulse shape discrimination (PSD) in scintillators such as LiCaAlF:Ce reduce gamma sensitivity, light yield remains small and large area detectors are difficult and expensive because these scintillators are single crystals. Tokuyama Co. have recently developed a transparent rubber sheet with LiCaAlF:Eu which overcomes the single crystal problem and provides good gamma discrimination but at a severe cost to the neutron detection efficiency [12]. The elpasolite family of scintillators is limited by hygroscopicity, difficulties in manufacturing large areas [11] and the need to have an ADC for every readout channel. ADCs are required for the majority of current neutron sensitive scintillators with an exception being ZnS:Ag/^{6}LiF. Single photon counting is primarily used for ZnS:Ag signals thereby avoiding reliance on ADCs. Avoiding ADCs has several advantages including simplicity and lower cost of electronics and greater detector robustness. Variation in PMT gain causes detector instability leading to complex corrections in ADC based electronics. Gain corrections can be built into many electronic read-out boards but is challenging if needed over several thousand channels and will need readjustment over time. Single photon counting electronics alleviates these problems and is much more insensitive to gain variations. A scintillator detector capable of working with single photon counting is therefore preferred.

ZnO:Zn/^{6}LiF is investigated here as a low afterglow alternative to ZnS:Ag/^{6}LiF. ZnO based scintillators have previously been considered for thermal neutron detection with the majority of the work being focused on the fast (100s of ps) UV-violet component of the ZnO emission [13] and eliminating slower defect related emission [14]. The main disadvantage of this is that neutron–gamma discrimination has to be done by pulse height separation methods in a highly self-absorbing crystal. ZnO based ceramics, fabricated using a hot pressing technique, have been explored previously with some success showing low levels of afterglow as demonstrated by the absence of thermally stimulated luminescence at room temperature [15]. Creating transparent ceramics remains difficult [16] and ZnO:Zn alone is insensitive to neutrons.

Luminescence and scintillation properties of ZnO:Zn/^{6}LiF investigated here focus mainly on the brighter blue–green emission. Low afterglow of ZnO:Zn is shown in comparison to ZnS:Ag. For the first time, the application of ZnO:Zn/^{6}LiF based detectors for neutron scattering is assessed by fiber coupling the scintillator to a multi-anode photomultiplier tube (PMT). Neutron–gamma discrimination and an increase in neutron count rate capability is demonstrated using simple single photon counting algorithms, in keeping with the aforementioned requirements.

2. Experimental methods

2.1. Scintillator screens

ZnO:Zn/^{6}LiF screens were produced by Scintacor [17] to mimic their standard ZnS:Ag/^{6}LiF neutron detection screens. ZnO:Zn phosphor powder (3.5 μm grain size) produced by Phosphor Technologies was mixed with ^{6}Li enriched LiF in a ratio of 2:1 by weight together with a small amount of binder. The mixture is intended to uniformly distribute the ^{6}LiF throughout the scintillator. The scintillator screen is non-hygroscopic. ZnS:Ag/^{6}LiF samples with the same thickness and weight ratios were tested as a comparison. The ZnS:Ag grain size was slightly larger at 8 μm.

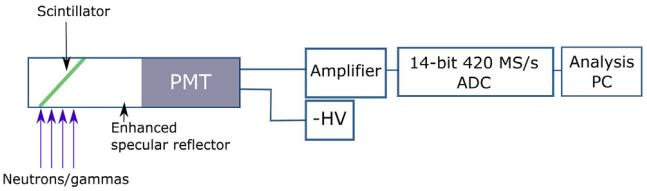

Fig. 1. Schematic of the experimental setup for determining basic scintillation properties — the scintillator is viewed by the PMT without additional light transport media such as optical fibers.

2.2. Luminescence

Photoluminescence (PL) spectra were obtained to determine possible light emission mechanisms. Steady state excitation–emission spectra were obtained using a Horiba Fluoromax-4 spectrofluorometer with a Xenon lamp as the excitation source. Alpha particle radioluminescence (RL) spectra were acquired by blocking the light source and installing a ^{226}Ra alpha source (5.11 MeV) 5 mm from the scintillator screens, which maintained a clear optical pathway to the emission monochromators. X-ray induced RL was measured using 60 kV peak X-rays and a ThorLabs CCS-100 spectrometer. All data was corrected for monochromator and detector efficiencies.

2.3. Scintillation characteristics

Neutron and gamma signals from the scintillator screens were acquired in several different configurations. In order to investigate basic scintillation properties of the ZnO:Zn/^{6}LiF, screens were directly viewed by a low noise, 38 mm diameter PMT with a green-extended photocathode (9902 from ET Enterprises Ltd.). Scintillator samples were inserted in an aluminum tube at an angle of 45 degrees to the axis of the tube. This tube was lined with an enhanced specular reflector to aid in light collection. The PMT was located approximately 50 mm away from the scintillator and shielded with lead to avoid direct interactions of gammas with the PMT. A diagram of the experimental configuration is shown in Fig. 1.

2.4. Clear fiber coupled detector

To assess the scintillator in a more realistic configuration for neutron scattering applications, a position sensitive detector with 1 m long transparent optical fiber readout (shown in Fig. 2) was constructed. The fiber block was designed to be able to swap scintillators for direct comparisons. Each detector pixel was 3 mm × 12 mm, had 1 mm diameter fibers aligned vertically and was optically isolated from its neighboring pixels. Scintillator to fiber distance was set at 3 mm to allow scintillation light to spread across multiple fibers. Every other fiber in each detector pixel was coupled to a separate pixel on a Hamamatsu H8711-MOD 16-channel PMT with a green enhanced photocathode. Two pixels on the PMT photocathode were used for each detector pixel. This arrangement allowed scintillation events to be discriminated from random PMT noise by using coincidences between the pairs of PMT pixels. As a consequence of fiber coincidence, light collection per PMT is automatically reduced by a factor of two. This detector geometry further reduces the overall light collection by limiting the solid angle of collection due to the numerical aperture of the fiber.