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Original Article

# Study on the neutron imaging detector with high spatial resolution at China spallation neutron source



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# ABSTRACT

Gadolinium oxysulfide (GOS) is regarded as a novel scintillator for the realization of ultra-high spatial resolution in neutron imaging. Monte Carlo simulations of GOS scintillator show that the capability of its spatial resolution is towards the micron level. Through the time-of-flight method, the light output of a GOS scintillator was measured to be 217 photons per captured neutron, ~100 times lower than that of a ZnS/LiF:Ag scintillator by neutron beam tests. The measured spatial resolution is ~36  $\mu$ m (28 line pairs/mm) at the modulation transfer function (MTF) of 10%, mainly limited by the low experimental collimation ratio of the beamline. The weak light output of the GOS scintillator requires an enormous increase in the neutron flux to reduce the exposure time for practical applications.

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### 1. Introduction

Neutron imaging is a powerful technique for non-destructive testing with the advantages of sensitivity for light elements, high penetrability for heavy metals, an ability to distinguish isotopes and magnetic analysis. However, the spatial resolution of neutron imaging was limited to tens of micrometers for a long time, while X-ray imaging has already reached micron-level spatial resolution [1] [–] [3]. A spatial resolution of better than 10  $\mu$ m is urgently demanded by the neutron imaging user community for the future development of this technology in a wide variety of domains [4]. The spatial resolution of neutron imaging is mainly affected by the collimation of the neutron beam, the resolution of optical coupling and the spot size of the scintillator. The spot size is a key property

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for the scintillator and it can be defined as the Full Width at Half Maximum (FWHM) of the intensity distribution of the light spot on the scintillator surface. The spot size of a traditional ZnS/LiF scintillator is usually larger than 30  $\mu$ m, which is the critical limitation for improving the spatial resolution. Gadolinium oxysulfide (GOS) scintillators can reach higher spatial resolution due to the significantly smaller spot size of several microns. The Neutron Microscope Project initiated by the Paul Scherrer Institute (CH-5232 Villigen PSI, Switzerland) reached a spatial resolution of 5  $\mu$ m by using an isotopically-enriched gadolinium oxysulfide (<sup>157</sup>GOS) scintillator [3] [[,5,6] ][][].

The China Spallation Neutron Source (CSNS) [7], a pulsed neutron source (25 Hz) with a power of 100 kW, has been in public operation since August 2018. An energy-resolved neutron imaging instrument is currently under construction at this facility and will be used for new materials, renewable energy and high-end manufacturing. A <10  $\mu$ m resolution is a novel requirement to explore fuel cells and lithium-ion batteries. A neutron imaging detector based on a GOS scintillator is an ideal method to satisfy this demand. In this study, a Monte Carlo simulation with the Geant4 toolkit demonstrates that a GOS scintillator could reach

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Fig. 1. Range projection of conversion electrons.



Fig. 2. Neutron capture efficiency versus thickness of GOS scintillator.

ultra-high resolution, as well as high neutron capture efficiency. The light output of a GOS scintillator excited by a single neutron is a key characteristic related to the imaging contrast and exposure time. However, it is difficult to measure because of the small light yield and poor ability to distinguish neutrons from gamma background. An experiment is carried out using a PMT to measure the weak scintillation light and the gamma background is suppressed significantly with the time-of-flight (TOF) method. A prototype of a neutron imaging detector with the GOS scintillator is developed and the performance evaluated through the imaging of a Siemens-Star test object and samples.



Fig. 4. Charge spectrum induced by a single neutron in the <sup>nat</sup>GOS scintillator.

#### 2. GOS scintillator

The neutron is detected through the scintillation light excited by conversion electrons induced from the nuclear reaction of  $^{155}Gd$  (n,  $\gamma$ )  $^{156}Gd$  and  $^{157}Gd$  (n,  $\gamma$ )  $^{158}Gd$ . The range distribution of conversion electrons and neutron capture efficiency were obtained by a Monte Carlo simulation with the Geant4 toolkit. The light output of a single neutron in the GOS scintillator was measured.

## 2.1. Monte Carlo simulation

The neutron capture efficiency and the spatial resolution of the detector are affected by the thickness of the GOS scintillator. In order to obtain the micron-level resolution, the thickness of the scintillator should be < 10  $\mu$ m. However, the intrinsic spatial resolution is limited by the spot size. To evaluate the spot size, tracks of conversion electrons in a 10  $\mu$ m thick GOS scintillator were simulated and the projection of the range on the surface of scintillator is shown in Fig. 1. The FWHM of the distribution is less than 2  $\mu$ m, which reveals that the spatial resolution could be toward the micron level. For the ultra-thin scintillator, the capture efficiency needs to be considered.

For the neutron imaging detector, the capture efficiency is expected to be as high as possible. In natural gadolinium, the abundances of <sup>155</sup>Gd and <sup>157</sup>Gd are 14.7% and 15.6%, respectively. The dashed line with triangle markers in Fig. 2 shows the capture efficiency of thermal neutrons with different thicknesses for the natural gadolinium oxysulfide (<sup>nat</sup>GOS) scintillator. For a thickness of 2  $\mu$ m, the efficiency is only ~20%, which can be increased by using an enriched isotope. The cross section of <sup>157</sup>Gd is much larger than that of <sup>155</sup>Gd. The efficiency of the <sup>157</sup>GOS scintillator, shown by the



Fig. 3. Schematic diagram of light output measurement (a) and experimental setup (b).



Fig. 5. Experimental setup (a) and neutron imaging detector prototype (b).



**Fig. 6.** MTF curves of the imaging of a Siemens-Star test object tested by neutrons (solid line) and LED light (dashed line), respectively.

solid line with star markers in Fig. 2, can be improved to 68% at the ultra-thin thickness of 2  $\mu$ m. This is triple that of the <sup>nat</sup>GOS scintillator. The <sup>157</sup>GOS scintillator is more suitable for <10  $\mu$ m spatial resolution detectors. In particular, for ultra-thin thicknesses, the advantages of the <sup>157</sup>GOS scintillator will be more significant.

#### 2.2. Light output of GOS scintillator

GOS is commonly coupled with a CCD for imaging readout, thus the light output of the scintillator is critical. Considering the high price of <sup>157</sup>GOS and nearly identical physical chemistry properties between <sup>157</sup>GOS and <sup>nat</sup>GOS, excluding the neutron capture efficiency, the <sup>nat</sup>GOS scintillator (10  $\mu$ m thickness, RC Tritec AG, Paul Scherrer Institute) is employed to measure its light output.

The experimental setup is shown in Fig. 3. A monochromator was placed on the neutron beam with a tilt angle of 45° to reduce the  $\gamma$  background and provide the 4.8 Å monochromatic neutrons. The scintillation photons were collected by the PMT (XP2020, Hamamatsu) in a dark chamber. An oscilloscope (Keysight DSO-X 3024A) was used as the data acquisition system to measure the charge spectrum from the PMT. The TOF method was used to further reduce the influence of the  $\gamma$  background.

The high voltage of the PMT was set at -1700 V and the single photoelectron peak of the PMT is  $\sim 139$  fC ( $Q_{spe}$ ) measured using an LED. The charge spectrum induced by a single neutron in the <sup>nat</sup>GOS scintillator was obtained through the integration of the pulse signal output from the PMT, and the result is shown in Fig. 4. The background and neutron signals were fitted by Gauss and Landau functions, respectively, with peaks of 262 fC ( $Q_0$ ) and 2076 fC ( $Q_1$ ).

The quantum efficiency ( $\eta$ ) of the XP2020 PMT is ~6% at the emission maximum wavelength of 544 nm. As a result, the light output of the <sup>nat</sup>GOS scintillator is 217 ± 2 photons per captured neutron (ph/n), calculated through ( $Q_1$ - $Q_0$ )/( $Q_{spe} \times \eta$ ), with only the statistical error considered in the estimation. Compared to the conventional ZnS/LiF:Ag scintillator, the light output of GOS is ~100 times lower. As a result, it is necessary to increase the neutron beam flux for neutron imaging with a higher spatial resolution.

## 3. Prototype of neutron imaging detector

The prototype of the neutron imaging detector using the <sup>nat</sup>GOS scintillator has been developed and several tests were performed to evaluate its characteristics at CSNS.

# 3.1. Experimental setup

A schematic diagram of the experimental setup is shown in Fig. 5(a). The detector prototype is assembled inside a dark chamber, as shown in Fig. 5(b). The neutrons penetrating through the sample are absorbed by the scintillator and converted into the green light, which is deflected with by  $90^{\circ}$  using a mirror and then focused by the lens system. Through loss-free light transmission



Fig. 7. Imaging of Siemens-Star test object tested at SANS.



Fig. 8. Imaging photographs of the small bearing (a) and mini motor (b).

with the appropriate magnification, the image is acquired by a CCD camera (Princeton Instrument, SOPHIA 2048B) with high quantum efficiency and spatial resolution.

detection efficiency and then the imaging time will be further reduced by half.

## 3.2. Performance of prototype

In order to evaluate the intrinsic spatial resolution, the prototype without a scintillator was firstly tested in the LED light field using a Siemens-Star test object with 128 line pairs at an optical magnification of 2.7. The light intensity variation of the line pairs at the same circle position of the Siemens-Star image were described by the sine wave. The MTF was calculated through the ratio of the amplitude to the bias level. As shown in Fig. 6 (dashed line), The spatial frequency is 62 lp/mm at an MTF of 10%, corresponding to the optical spatial resolution of 16  $\mu$ m, which can be improved by using a higher resolution sCMOS camera and larger optical magnification of the lens.

The prototype was then tested at the Small-Angle Neutron Spectrometer (SANS) of CSNS, with a collimation ratio (L/D) more than 200:1 and a neutron flux of ~3 × 10<sup>6</sup> n/cm<sup>2</sup> · s. The imaging of the test object is shown in Fig. 7. The red circle plotted in the figure corresponds to a 30  $\mu$ m spatial resolution. The star object inside the circle can be well distinguished. As shown in Fig. 6, the spatial resolution is 36  $\mu$ m at a MTF of 10% (solid line), much less than the intrinsic optical spatial resolution due to the poorer collimation ratio (L/D) of the neutron beam.

In addition to the spatial resolution measurement, a small bearing and a mini motor were tested to validate the feasibility of the imaging system by using the GOS scintillator at SANS. In Fig. 8(a), the image of the bearing clearly shows the balls with the diameter of 1 mm and retainer inside, and the lubricating oil with a black color can be seen around the inner ring. There are several blacker areas among the balls, which indicate the concentration of the lubricating oil. From the imaging of Fig. 8(b), the inner structure of the mini motor is clearly exhibited. The black rectangle is the rotor with enamel wire, which is assembled through a rotatable shaft. The results show that the imaging system can directly detect the weak light of the GOS scintillator. Both of the two images were acquired in one frame with a single exposure time of 10 min, which cannot be accepted in the imaging experiments with the CT method. In order to decrease the imaging time to less than 10 s, the neutron beam flux should be increased to at least the level of  $10^8$  n/ cm<sup>2</sup>.s. Moreover, the isotopically-enriched <sup>157</sup>GOS will double the

#### 4. Conclusion and outlook

GOS scintillators are a valuable solution to realizing the ultrahigh spatial resolution of neutron imaging. The simulation of a GOS scintillator shows that the ability of the spatial resolution tends towards the micron level and high efficiency by using the isotopically-enriched <sup>157</sup>GOS. According to the measurement with PMT, the light output of GOS is 217 ph/n, ~100 times lower than that of the ZnS/LiF:Ag scintillator. In order to verify the feasibility of the imaging system, a detector prototype was developed and tested at SANS. The experimentally determined resolution (36 µm) is compatible with the resolution obtained with ZnS/LiF. The results are about an order of magnitude larger than the simulated intrinsic resolution of GOS, which are limited through the beam divergence.

Due to the weak light output of the GOS scintillator, the critical requirement is to increase the neutron flux to obtain the acceptable exposure time. The Energy-Resolved Neutron imaging Instrument (ERNI) at CSNS is now being constructed, which will provide the much better neutron beam for the imaging experiments. Future work will be devoted to the study on the fabrication of GOS scintillator to improve the light output by using transparent ceramics.

#### **Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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