Method Article

Light Yield Enhancement of 157-Gadolinium Oxysulfide Scintillator Screens for the High-Resolution Neutron Imaging

Jan Crha\textsuperscript{a,b,1}, Joan Vila-Comamala\textsuperscript{c}, Eberhard Lehmann\textsuperscript{a}, Christian David\textsuperscript{d}, Pavel Trtik\textsuperscript{a,}\textsuperscript{*}

\textsuperscript{a} Laboratory for Neutron Scattering and Imaging, Paul Scherrer Institut, Villigen PSI, CH, 5232, Switzerland
\textsuperscript{b} Faculty of Civil Engineering, Czech Technical University, 166 29, Prague, Czech Republic
\textsuperscript{c} Institute for Biomedical Engineering, University of ETH Zürich, 8092, Zürich, Switzerland
\textsuperscript{d} Laboratory for Micro- and Nanotechnology, Paul Scherrer Institut, Villigen PSI, CH, 5232, Switzerland

A B S T R A C T

This paper reports on light yield enhancement of terbium-doped gadolinium oxysulfide based scintillator screens achieved by coating their substrates with thin layers of a high density and high atomic number material. For this purpose, iridium was chosen and layers of various thicknesses were applied by atomic layer deposition (ALD). We assessed newly developed scintillator screens for neutron absorption, light yield and spatial resolution and compared them to previously used non-iridium-coated scintillator screens. The addition of the iridium layer resulted in 65 \% light yield enhancement in comparison to uncoated scintillator screens while the spatial resolution and absorption power remained unchanged.

Highlights

- 65 \% light yield enhancement of the scintillator light output with preservation of the spatial resolution
- Use of atomic layer deposition for nanoengineering of the neutron sensitive scintillator screens

© 2018 Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

A R T I C L E  I N F O
Method name: Neutron imaging detection
Keywords: atomic layer deposition, gadolinium oxysulfide, iridium, neutron imaging, neutron microscope, scintillator screen
Article history: Received 24 June 2018; Accepted 11 December 2018; Available online 17 December 2018

* Corresponding author.
E-mail address: pavel.trtik@psi.ch (P. Trtik).
\textsuperscript{1} scientific guest at Neutron Imaging and Applied Materials Group, PSI

https://doi.org/10.1016/j.mex.2018.12.005
2215-0161/© 2018 Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).
Method details

Neutron imaging is a non-destructive testing (NDT) method which in many cases provides complementary information to more common X-ray imaging. While X-ray sources can yield relatively high photon flux, especially in synchrotron facilities, the neutron sources lack behind in terms of intensity. From this fact stems the necessity of thorough optimization of the neutron-sensitive scintillator screens, in particular for the high-resolution applications.

Nowadays, most widely used detection systems for high-resolution neutron imaging comprise a scintillator screen optically coupled to CCD or CMOS camera [1]. Standard scintillator screens consist of a compound (e.g. Gd$_2$O$_2$S: Tb$^{3+}$ - terbium-doped gadolinium oxysulfide) or of a mixture of compounds (e.g. $^6$LiF + ZnS:Ag) spread over a substrate with no further modifications. Work presented here is a part of the ‘Neutron Microscope project’ [2–4] performed at the Paul Scherrer Institut (PSI) and is focused on the spatial resolution enhancement through material and geometrical adjustments of the scintillator screen.

During this project light optics with fivefold magnification tailored for main emission peak of Tb$^{3+}$ in gadolinium oxysulfide host was developed. A substantial decrease of thickness of gadolinium scintillating powder layer was obtained by utilizing enriched $^{157}$Gd isotope (about 88 % of $^{157}$Gd) for $^{157}$Gd$_2$O$_2$S: Tb$^{3+}$ powder production. Details of scintillator screen production using enriched gadolinium can be found in [5].

The main source of scintillation light in terbium-doped gadolinium oxysulfide phosphor upon neutron irradiation are internal conversion electrons (ICE) and Auger electrons originating from (n, $\gamma$) reaction on $^{157}$Gd. Since scintillator screens using $^{157}$Gd$_2$O$_2$S: Tb$^{3+}$ require only a thin layer ($\sim$ 3 μm for cold neutron beam) to almost fully absorb incident cold neutron beam, it can easily occur that ICEs’ energy is not fully deposited within the scintillating powder thus travelling outside the scintillating layer. Therefore, the objective is to utilize (n, $\gamma$) reaction products more efficiently.

Methodology

As already mentioned some of the ICEs do not fully deposit their energy in the scintillating powder layer. This is caused by the fact that $\gamma$-rays from (n, $\gamma$) reaction are emitted isotropically implying that ICE emission is also an isotropic process. Considering small dimensions of the phosphor powder layer and ICEs’ initial momentum it is obvious that certain portion of ICEs can escape the phosphor. Therefore, iridium, which combines high mass density and high atomic number, was selected as a suitable candidate material for maximizing the ICEs’ backscatter. To determine the appropriate thickness of iridium coating, the Monte Carlo code for electron transport CASINO [6] was utilized to calculate the electron backscattering coefficients (the ratio of the number of electrons leaving the sample to the number of electrons entering the sample).

Internal conversion electrons from (n, $\gamma$) reaction on $^{157}$Gd possess a distinct energy spectrum [7]. The most important lines are those at 29 keV and 71 keV (the 29 keV line being $\sim$6.4 × more intense than the 71 keV line). It is worth noting that the electron spectrum emitted due to $^{158}$Gd de-excitation also includes K-Auger electrons of 35 keV and ICE electrons of 78 keV, both with relatively high yield. Although, when electrons of similar energies (29 keV + 35 keV and 71 keV + 78 keV) are grouped

---

**Specifications Table**

| Subject area | Physics and Astronomy |
|-------------|-----------------------|
| More specific subject area | Neutron-sensitive scintillator screen |
| Method name | Neutron imaging detection |
| Name and reference of original method | - |
| Resource availability | - |
together, their relative yield ratio remains practically the same, i.e. $\sim$6. Backscattering coefficient were, therefore, calculated for 29 keV and 71 keV ICEs.

The results of simulated 29 keV and 71 keV electrons striking the surface of iridium layer on a silicon substrate from two different angles, namely orthogonal to the surface (0°) and at 45° are concisely presented in Fig. 1. It can be seen that the curves of 29 keV ICEs saturate at about 300 nm thickness of the iridium layer. Therefore, increasing the thickness further would not have any impact on the amount of backscattered electrons. The results of calculations with 71 keV electrons follow the same trend as those with 29 keV electrons. However, the state of saturation is reached for a thicker layer (the backscattering coefficient for 1500 nm of iridium and an orthogonal 71 keV electron beam is 0.46, which corresponds to the saturation state) that is similar to the size of $^{157}$Gd$_2$O$_2$S:Tb$^{3+}$ particles used in the scintillator screen. Considering the relative ratio of the 29 keV and 71 keV ICEs and the thickness of the iridium layer necessary for effective backscattering of 71 keV ICEs, and the slow growth rate of the atomic layer deposition (ALD) coating, the final thickness of iridium layer was chosen on the basis of 29 keV electron backscattering simulation.

In Fig. 2, the trajectories of simulated electron transport in a layer of iridium on a silicon substrate are shown. The two-layer geometry including an infinitely thick silicon substrate and an iridium layer of a variable thickness was used for the simulation. The electron beam of 10 nm width impinged iridium surface first. It is clearly shown that increasing the iridium layer thickness significantly lowers the interaction volume of incident electrons in the material and causes more electrons to backscatter. In Fig. 3, the cross-section of the actual iridium-ALD-coated scintillator screen is shown on the scanning electron microscope (SEM) micrograph. The layer has been built up in two separate 100 nm thick deposition runs.

**Fabrication and experimental characterization of the iridium coated scintillator screens**

The original 11 mm x 11 mm silicon substrates cut from a wafer were coated with an iridium thickness of 200 nm and 460 nm by atomic layer deposition (ALD) using iridium acetylacetonate,
Ir(acac)₃ and oxygen, O₂, as precursor gases at 350 °C. The iridium layer was nucleated using a plasma-enhanced ALD recipe to obtain a finer iridium grain size, but the bulk of the iridium thickness was grown by thermal ALD process [8]. A PICOSUN™ R-200 Advanced ALD system was used for the deposition. The resulting substrates and uncoated control substrates were then deposited with ¹⁵⁷Gd₂O₂S:Tb³⁺ powder, as described in [5].

Neutron imaging experiments were carried out at Paul Scherrer Institut using the cold neutron imaging facility ICON [9]. The mean neutron wavelength 3.1 Å of the ICON’s cold spectrum in combination with ¹⁵⁷Gd₂O₂S:Tb³⁺ phosphor allows for the utilization of very thin scintillator powder layers. Test samples comprise square scintillator screens of 11 mm x 11 mm in dimension consisting of silicon substrates (250 μm thick), iridium layers (200 nm or 460 nm thick) and ¹⁵⁷Gd₂O₂S:Tb³⁺ powder.

Fig. 2. CASINO simulation results of electron transport in a layer of iridium on a silicon substrate. The numbers in the left bottom corner of each image denote the iridium layer thickness. The incident electron beam energy is 29 keV. Red trajectories denote backscattered electrons, blue trajectories denote electrons that do not backscatter.

Fig. 3. SEM micrograph of a silicon substrate coated with iridium by atomic layer deposition (ALD).
phosphor powder of a mean particle size of 2 μm. The thickness of phosphor layer is approximately 2.5 μm. Two samples per each given thickness of phosphor layer were tested. The newly developed iridium-coated scintillators were compared to uncoated scintillator screens to assess the influence of iridium layer on scintillator performance in neutron absorption power, light output and spatial resolution. For the neutron absorption measurements the ‘Sample’ position was used, for light output measurements and spatial resolution measurements the ‘Scintillator’ position was used (see Fig. 4).

The neutron absorption and light output measurements were carried out using the standard high-resolution setup available at the ICON facility with 20 μm thick natGd2O2S:Tb3+ scintillator screen coupled to the camera with a nominal pixel size of 13.5 μm [9]. Four camera pixels were binned resulting in a nominal image pixel size of 27 μm. Spatial resolution measurements were carried out using the Neutron Microscope setup equipped with sCMOS camera (ORCA-Flash4.0 V2) with image pixel size of 1.3 μm. All acquired neutron radiographs were corrected for open beam and dark current.

**Results**

During the measurements, the newly developed 200 nm and 460 nm iridium-coated scintillator screens were tested and compared to the uncoated predecessor. Neutron absorption measurements yielded 0.395, 0.415 and 0.41 for scintillator screen with 460 nm of iridium coating, scintillator screen with 200 nm of iridium coating and the uncoated scintillator screen, respectively. The results of individual samples of each iridium coating thickness are shown in Table 1 and Fig. 5b. The very similar results of neutron absorption measurements were expected as the amount of scintillating powder in each scintillator screen which is responsible for neutron absorption was approximately the same. Light output measurements resulted in 977, 930 and 583 counts for scintillator screen with 460 nm of iridium coating, scintillator screen with 200 nm of iridium coating and uncoated scintillator screen, respectively. The difference in light output results for individual sample can be seen in Table 2. Fig. 5a also confirms the data from the Table 2 where iridium-coated scintillator screens appear clearly brighter. Therefore, the influence of iridium coating is apparent, as it yields approximately 65% higher light output in comparison to uncoated substrates.

The scintillator screens were tested for spatial resolution using a test sample consisting of a gadolinium Siemens star. Gadolinium Siemens star [10] is a standardized test object for the spatial resolution assessment. The resulting neutron radiograph is shown in Fig. 5c). Applying the Fourier
Ring Correlation (FRC) method [11,12] for spatial resolution assessment yielded a resolution of 5.2 μm (compared to 4.9 μm uncoated). A visual inspection of the gadolinium Siemens star reveals spatial resolution of 4.5 μm (see Fig. 5d).

**Discussion**

The main sources of scintillation light in $^{157}$Gd$_2$O$_2$S:Tb$^{3+}$ are ICE and Auger electrons origination from (n, γ) reaction on $^{157}$Gd. However, only 29 keV ICE and 35 keV Auger electrons have a continuous slowing down approximation (CSDA) range in the order of single micrometers (computed using [13] and compared with [7]), i.e. similar to phosphor particle size. The next energy of ICEs is 71 keV. Yet, these conversion electrons’ CSDA range is in the order of tens of micrometers meaning that full energy deposition in the scintillation powder is much less probable. As seen in the results, the increase of the iridium layer thickness does not seem to lead to significant increase in the light output despite the clearly higher backscattering coefficient of the thicker layer. To explain this apparent inconsistency

### Table 1

The mean values and the standard deviations of the neutron absorption. Scintillators 1, 2 - 460 nm iridium layer; 3, 4 - 200 nm iridium layer; 5, 6 - uncoated scintillator screens.

| Scint. number | Neutron absorption (-) |
|---------------|------------------------|
| 1             | 0.38 ± 0.02            |
| 2             | 0.41 ± 0.02            |
| 3             | 0.41 ± 0.02            |
| 4             | 0.42 ± 0.02            |
| 5             | 0.41 ± 0.02            |
| 6             | 0.41 ± 0.02            |

Fig. 5. Neutron radiographs of (a) light output measurements, (b) neutron absorption measurements, (c) spatial resolution measurements using standard sample of gadolinium Siemens star and (d) zoom-in of center part of Siemens star image presented in 5c. Scintillators 1, 2: 460 nm iridium layer; 3, 4: 200 nm iridium layer; 5, 6: uncoated screens. The width of the central spokes is equal to 4.5 μm.
Table 2
The mean values and the standard deviations of the light output. Scintillators: 1. 2 - 460 nm iridium layer; 3, 4 - 200 nm iridium layer; 5, 6 - uncoated scintillator screens.

| Scint. number | Light output (counts) |
|---------------|-----------------------|
| 1             | 1035 ± 39             |
| 2             | 919 ± 32              |
| 3             | 944 ± 31              |
| 4             | 916 ± 32              |
| 5             | 595 ± 27              |
| 6             | 571 ± 27              |

between CASINO calculations of electron backscattering coefficients and the measured results, further simulations were carried out. For this purpose the Monte Carlo simulation code GEANT4 [14] was employed and the influence of iridium layer on the energy deposition in Gd₂O₂S:Tb³⁺ was assessed. GEANT4 simulation consisted of simple geometrical arrangement of adjacent boxes filled with respective materials (silicon for the substrate, iridium for the backscattering layer and gadolinium oxysulfide for the scintillating material). For each energy of ICEs (based on [7]) and for each representative position along the axis of the scintillator, 10⁵ ICEs were simulated to be emitted isotropically. Energy deposition, which is proportional to the amount of generated light, in the volume containing scintillating material for a given energy of an ICE was recorded. Assuming the Beer-Lambert law and based on the knowledge of the conversion electron yields upon neutron irradiation, the energy deposition was calculated.

The results of the additional simulations showed that the thicker layer of iridium coating, i.e. 460 nm, used for the measurements yield only about 10 % increase in energy deposition in the phosphor (in comparison to uncoated scintillator screen). The 200 nm layer increases the energy deposition only by about 8.5 %. The result of this simulation therefore indicates that the difference of 1.5 % between the 200 nm and 460 nm thick iridium layers is too small to be observed in the measured results obtained with the limited sample set.

Considering the combination of results presented in this paper and the energy deposition simulations, major influence of the light output enhancement is to be associated with the increase in the backscattering (reflection) of the scintillation light due to the higher reflectivity of iridium compared to silicon. The minor part is therefore caused by the backscatter of the ICEs.

Conclusion

We demonstrated the influence of iridium coating of the scintillator screens on light output. The iridium coating on silicon substrate resulted in a 65 % increase in light output while the neutron absorption power and spatial resolution have been preserved. Since the results of this investigation suggest that the major part of the light output enhancement is due to the better light reflection from the iridium surface, the additional layer of a material of even better reflectivity will be considered. Iridium layer should be, however, preserved not only for its electron backscattering capabilities but also for its ability to significantly reduce ICEs interaction volume. This reduction of the interaction volume causes the backscattered electrons to reappear on the surface closer to the position of its entrance and thus the probability of energy deposition in the proximity of their emission from (n, γ) reaction is increased (prevents the spatial resolution degradation).

In the future, further light output enhancement of the scintillator screens is foreseen as it is planned to coat nanostructured phosphor surfaces in order to facilitate light escape from the powder particles. On the side of spatial resolution, scintillator powder should be placed into microchannels in order to contain conversion reaction product and light as close as possible to the location of its origin, which is an approach already successfully applied in the field of x-ray imaging [15].
References

[1] E. Lehmann, P. Trittik, D. Ridikas, Status and perspectives of neutron imaging facilities, Phys. Procedia 88 (88) (2017) 140–147.
[2] P. Trittik, et al., Improving the Spatial Resolution of Neutron Imaging at Paul Scherrer Institut – The Neutron Microscope Project, Phys. Procedia 69 (2015) 169–176.
[3] P. Trittik, E.H. Lehmann, Progress in High-resolution Neutron Imaging at the Paul Scherrer Institut - The Neutron Microscope Project. J. Phys. Conf. Ser. 746 (Sep. (1)) (2016) 012004.
[4] P. Trittik, Neutron microtomography of voids in gold, MethodsX 4 (2017) 492–497.
[5] P. Trittik, E.H. Lehmann, Isotopically-enriched gadolinium-157 oxysulfide scintillator screens for the high-resolution neutron imaging, Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. 788 (2015) 67–70.
[6] D. Drouin, A.E. Couture, D. Joly, X. Tastet, V. Aimez, R. Gauvin, “CASINO V2.42—A Fast and Easy-to-use Modeling Tool for Scanning Electron Microscopy and Microanalysis Users, Scanning 29 (2007) 92–101.
[7] D.A. Abdushukurov, et al., Modeling the registration efficiency of thermal neutrons by gadolinium foils, (2010).
[8] J. Vila-Comamala, L. Romano, V. Guzenko, M. Kagias, M. Stampanoni, K. Jefimovs, Towards sub-micrometer high aspect ratio X-ray gratings by atomic layer deposition of iridium, Microelectron. Eng. (2018).
[9] A.P. Kaestner, et al., The ICON beamline A facility for cold neutron imaging at SINQ, Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. 659 (1) (2011) 387–393.
[10] C. Grünzweig, G. Frei, E. Lehmann, G. Kühne, C. David, Highly absorbing gadolinium test device to characterize the performance of neutron imaging detector systems, Rev. Sci. Instrum. 78 (5) (2007).
[11] M. Van Heel, M. Schatz, Fourier shell correlation threshold criteria, J. Struct. Biol. 151 (3) (2005) 250–262.
[12] J. Vila-Comamala, et al., Characterization of high-resolution diffractive X-ray optics by ptychographic coherent diffractive imaging, Opt. Express 19 (22) (2011) 21333.
[13] M.J. Berger, ESTAR, PSTAR and ASTAR: Computer Programs for Calculating Stopping Powers and Ranges for Electrons, Protons and Helium Ions (1995).
[14] S. Agostinelli, et al., GEANT4 - A simulation toolkit, Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. 506 (3) (2003) 250–303.
[15] U.L. Olsen, et al., Structured scintillators for X-ray imaging with micrometre resolution, Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. 607 (1) (2009) 141–144.