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Scintillator screen development for fast neutron radiography and tomography and its application at the beamline of the 10 MW BNC research reactor

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HIGHLIGHTS

- Inexpensive, simple ZnS-based fast neutron imaging screens have been developed.
- The screen performance has been compared with that of commercial counterparts at a 10-MW research reactor beamline and 70% efficiency of the best commercial one can be achievable.
- Radiographic and tomographic imaging has been performed using the different screens.
- High quality fast neutron imaging capabilities have been demonstrated on dense and robust objects.

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ABSTRACT

Simple and inexpensive ZnS-based fast neutron imaging screens have been developed and their performance has been tested and compared to a commercially available one using the RAD beamline of the 10 MW research reactor of the Budapest Neutron Centre (BNC), Hungary. ZnS(Ag) and ZnS(Cu) powders have been mixed with optical epoxy, deaerated and casted into sheet form using an aluminum frame. The ZnS concentration and the screen thickness have been optimised using sample screen pieces. The in-house screens have been tested in camera-based neutron imaging detectors in a reactor beamline and compared with a commercially available polypropylene/ZnS(Cu) fast neutron imaging screen and with a BC400 plastic scintillator slab screen. It has been found that the in-house screen produces only about 60 % of light intensity of the commercial polypropylene/ZnS screen, which is mainly due to the lower hydrogen density of the optical epoxy compared to polypropylene by the same amount. The BC400 performs inferior compared to any ZnS-based scintillator tested here. Fast neutron tomography has been performed with both the commercial and the in-house screens on the reactor beamline. A spatial resolution of around 1.6 mm has been achieved. Typically 10–15 min exposures were needed to obtain good quality radiographic images, whereas several hours of acquisition were needed to obtain the full tomographic set images. High quality imaging results have been obtained on large (150 mm in diameter) and dense objects (hydraulic couplings) proving the feasibility and utility of fast neutron imaging for such samples.

1. Introduction

Fast neutron imaging is a promising nondestructive technique for testing dense and voluminous objects of practically any material composition. Only fast neutrons can provide images free of artifacts and with reasonable contrast for dense and voluminous samples containing mixed low-Z/high-Z materials, where photon-based techniques or even thermal neutron imaging would fail. This is mainly due to the fact that in the fast energy range (at few MeV) the macroscopic neutron interaction cross sections are generally of the same order regardless of energy and material, allowing high penetrating power regardless of sample composition. This makes fast neutron imaging a promising inspection modality for many practical problems.

Several applications of fast neutron imaging have been investigated in past works. One of them is homeland security to detect heavily shielded explosives (Mor et al., 2015) or contraband (Buffler, 2004). It

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is also foreseen for nuclear stockpile stewardship (Hall et al., 2007). Fast neutrons imaging is also used as a highly penetrating tool in cultural heritage investigations complementing X-ray techniques (Osterloh et al., 2015) or for industrial applications like looking into glued wooden boards (Osterloh et al., 2011). Furthermore it is also of interest as non-intrusive, non-contact method for investigating industrial processes in heavy enclosures under potentially harsh conditions such as two-phase flows in nuclear fuel bundles (Takenaka et al., 1999; Zboray et al., 2014, 2015; Adams et al., 2016).

One of the main limiting factors for the applications of fast neutron imaging is the detection technology, offering relatively poor spatial resolution and low detection efficiency due to the high penetrating power compared to X-ray or thermal neutron imaging. This results usually in long exposure times to enable reasonable image quality. Therefore, the development of efficient imaging screens for fast neutrons is of high importance. In a recent paper, Makowska et al. (2017) gives a good overview of fast neutron imaging detector and scintillator developments while focusing on quantifying the performance of Polypropylene(PP)/ZnS type commercially available scintillator screens produced by RC Tritec AG (Tritec, 2017).

In a previous study (Zboray et al., 2017), we have demonstrated the feasibility of fast neutron radiography and tomography on the RAD beamline of the 10 MW research reactor of the Budapest Neutron Centre (BNC), Hungary, using a BC400 plastic scintillator slab screen in combination with an inexpensive detector setup. The beamline is normally used for thermal neutron imaging (Kis et al., 2015), however it also provides a significant fast flux that can be utilized for fast neutron imaging. We have used again this beamline in the present study to improve the imaging detector efficiency (mainly that of the scintillator screen) and to test different ZnS-based scintillator screens. We have indicated in our previous paper, and we prove it experimentally below, that due to its much higher light yield, a ZnS-based scintillator might give higher detection efficiency compared to the organic BC400 scintillator in a camera-based imaging detector setup with a low light collection efficiency. The scintillator screens we examined include inexpensive, in-house made ones and a commercial one from RC Tritec AG similar to the ones used by Makowska et al. (2017). We have produced inexpensive ZnS(Ag) and ZnS(Cu) based fast neutron imaging scintillator screens using relatively simple laboratory infrastructure and means with the idea to see if such screens can deliver acceptable performance that could be somewhat comparable to that of commercial ones. Commercial screens are produced typically using a vulcanizer which requires specialized equipment and is a sensitive and challenging process to master. Furthermore we also wanted to examine the performance in terms of screen thickness and phosphor content over a wider range to see if there is still some potential for optimization of those parameters. For example, for applications in a fission spectrum reactor beamline, containing neutrons up to 10 MeV, increasing the screen thickness might be beneficial as the first sections of the screen facing the beam can act as proton radiator and contribute to a higher light output. The next section focuses on a description of the experimental setup consisting of the fast neutron beam, the imaging detector system and the different scintillator screen types. Then we discuss the study on the optimal ZnS concentration and screen thickness using a special sample holder and small-sized sample screens. After that, radiographic and tomographic images obtained using the different scintillator screens are shown to illustrate the system performance in terms of efficiency and resolution.

2. The experimental setup

2.1. The neutron beamline at BNC

Imaging was performed at the RAD beamline (radial channel number 2) of the 10 MW research reactor of the BNC (Kis et al., 2015) shown in Fig. 1. The beamline is routinely utilized for thermal neutron imaging utilizing a thermal flux of around $4 \times 10^7 \text{cm}^{-2}\text{s}^{-1}$. The beam features further a significant gamma background of 8.5 Gy/h and a significant fast neutron contribution of $2.7 \times 10^7 \text{cm}^{-2}\text{s}^{-1}$ (E > 2.5 MeV), both figures expressing non-attenuated values (BNC, 2018). These values have been carefully evaluated by the instrument crew. The fast flux specifically has been measured by the standard procedure activating a Cd-clad Ni foil utilizing the Ni-58(n,p)Co-58 threshold reaction (~ 2.5 MeV) as described in ASTM (2002). Just as in our previous fast neutron imaging study, we have used a 10 mm MirroBor (MirroBor, 2012), a borated rubber mat, filter enabling the suppression of practically the entire thermal neutron component of the beam to avoid unnecessary sample activation. This time we used only a 200 mm thick lead filter in the beam against the gammas, as this should be sufficient against the direct in-beam gamma contribution because most of the relatively modest gamma background in the experimental hatch is originating from prompt gammas due to activation of surrounding structures by scattered neutrons (Zboray et al., 2017). The lead filter decreases the fast neutron flux to around $3.3 \times 10^5 \text{cm}^{-2}\text{s}^{-1}$, which has been utilized for the experiments below. The primary aperture of the beamline (situated at the boundary of the biological shield and the core reflector) with a diameter of 28 mm results in a calculated L/D of 177 at the position of the detector placed at 4960 mm from the primary aperture, enabling a quasi-parallel beam imaging geometry.

2.2. The imaging detector and scintillator screens

An ANDOR Neo 5.5 (2560 × 2160 pix) sCMOS camera, being the part of the standard thermal neutron imaging detector setup at the beamline, has been used for the experiments (Kis et al., 2015). The optics we used on the camera was a 50 mm, f # 1.2 objective with a total field of view (FOV) of the detector of about $270 \times 230 \text{ mm}^2$ at an effective pixel size of about 106 μm. For the given optics simple geometrical consideration show that the light collection is relatively poor just due to the solid angle under which the screen is seen by the objective amounting to a factor of about 1.41×10^{-4}. This figure is about the same order of magnitude for any other typical camera-based imaging detector setup. Therefore if one uses a scintillator with relatively low light yield in such setup, neutron collision events can go undetected. This we have pointed out in our earlier work in connection with using an 8 mm thick BC400 organic slab scintillator screen from St. Gobain (St. Gobain, 2011). ZnS-based inorganic scintillators provide about ten times higher light yield compared to BC400. Due to their transparency, much thicker organic scintillator screens could be applied compared to the relatively nontransparent ZnS-based scintillator screens, however a very large screen thickness has an adverse effect on the spatial resolution. Furthermore plastic scintillators, as BC400, have a higher gamma sensitivity, while thinner ZnS-based screen are less sensitive to gammas. Therefore to improve the overall detector performance, we started to experiment with ZnS-based scintillator screens. Our main motivation was to try to develop simple, inexpensive screens that could be produced by basic laboratory means and deliver comparable performances as commercial screens. As a reference, we compare their performance to that of the aforementioned BC400 screen and of a 3.8 mm thick, commercially available polypropylene(PP)/ZnS(Cu) screen with 30 %, weight percent, ZnS(Cu) from RC Tritec AG.

We have produced fast neutron imaging scintillator screens by mixing ZnS(Ag) and ZnS(Cu) powder with optical epoxy. The optical epoxy, EJ500 from Eljen (EljenTechnology, 2016a) has a hydrogen density of $4.68 \times 10^{22}/\text{cm}^3$, about 40 % less than that of PP $7.7 \times 10^{22}/\text{cm}^3$. Its optical transmission is close to 100 % above 400 nm. The ZnS(Ag) powder, EJ600 also from Eljen (EljenTechnology, 2016b), had a typical particle size of 8 μm. ZnS(Cu) has been obtained from Phosphor Technology (PhosporTechnology, 2016) with a median particle size of 4 μm. Note that the above particle sizes might strongly underestimate the actual grains sizes, as at these particle sizes powders tend to agglomerate to larger grains up to typically around 50 μm unless

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