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New type of neutron image scintillator based on

\( H_3^{10}\text{BO}_3/ZnS(\text{Ag}) \)

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Abstract

In this contribution we present a system of converter \((H_3^{10}\text{BO}_3)\) and phosphor \((ZnS:Ag)\) which were fabricated by spraying method. The particle sizes and the mass ratio of the neutron converter and phosphor materials were optimized by simulation and experimental methods, respectively. Furthermore, the spatial resolution of scintillators at thicknesses of 0.40nm and 0.35mm were 1.11 lp/mm (MTF=0.1) and 1.33 lp/mm (MTF=0.1), respectively. The results indicated that \(H_3^{10}\text{BO}_3/ZnS(\text{Ag})\) scintillator have potential application in thermal neutron radiography.

Keywords: neutron scintillator, \(H_3^{10}\text{BO}_3/ZnS(\text{Ag})\), neutron radiography;

1. Introduction

Neutron scintillators have found widespread application in the detection of thermal neutrons especially in neutron radiography [1, 2]. Commercially available neutron scintillators [3~5] consist of the neutron converter \(^6\text{LiF}\) and the phosphor \(ZnS:Ag/Cu\) dispersed in an organic binder which is supported by a aluminium foil. Although composite neutron scintillators based on \(ZnS:Ag\) fluorescent material have been in use for many decades, there are still some points pertaining to be optimized.

In order to improve the detection efficiency and the spatial resolution, some new kind of phosphors containing the neutron absorbing isotopes \(\text{Gd, }^{10}\text{B or }^6\text{Li}\) such as \(\text{Gd}_2\text{Si}_2\text{O}_7:Ce\) [6], \(^6\text{Li}_{\text{dep}}\text{Gd(BO}_3\text{)}_3:Ce\),

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$^6$Li$_6$natGd(BO$_3$)$_3$:Ce, Cs$_2$$^6$LiYBr$_6$:Ce, Cs$_2$$^6$LiYCl$_6$:Ce [7], Y$^{10}$BO$_3$:Ce [8], and organic polymers containing $^{10}$B (BP) [9] were investigated. T. Kojima et al. [10] developed ZnS:Ag/$^{10}$B$_2$O$_3$ scintillators by substituting $^6$LiF by $^{10}$B$_2$O$_3$, which exhibited the detection efficiency of 20–40% for thermal neutrons. In order to optimize ZnS:Ag/$^{10}$B$_2$O$_3$ scintillators, in this paper, we developed the $^3$H$_3$10BO$_3$/ZnS(Ag) scintillators.

2. Experimental

The neutron cross-section of $^{10}$B (3830 barn) is about four times higher than that of $^6$Li (910 barn) for thermal neutrons [11]. Hence, one expects that replacing the $^6$LiF with the compounds containing $^{10}$B will improve the detection efficiency. However, it should be noted that using the $^{10}$B$_2$O$_3$ compounds as a neutron converter decreases the light yield due to degradation in the transparency of the $^{10}$B$_2$O$_3$ compounds. To solved this problem, we used the $^3$H$_3$BO$_3$ (B$_2$O$_3$.3H$_2$O) which has much better transparency.

Single crystal $^3$H$_3$10BO$_3$ ($^{10}$B abundance >92.4%) and ZnS(Ag) powders were mixed in 25 wt% binder (epoxy) homogeneously. Then, they were poured into a spray pot and sprayed on the aluminium foil (Ø300mm×0.5mm). The luminous intensity of the scintillating light was measured by a multi-channel analyzer after the amplification of the light by a photomultiplier and a spectroscopy amplifier when the samples were irradiated with the $^{241}$Am–Be neutron source. Some samples with sizes of 100mm×100mm×0.35mm and 0.40mm (mass ratio of the $^3$H$_3$10BO$_3$/ZnS:Ag was 1:6) were characterized by using the neutron radiography facility with an accelerator driven D-Be neutron source at Peking University of China [12].

3. Result and discussion

Since the neutron image is formed by a number of successive processes, the results of each of them can affect the final detector efficiency. Here we consider these parameters and processes in the order they appear in the image formation process.

3.1. Crystals size of the neutron converter and phosphor

The neutron capture reaction gives rise to a secondary ionizing radiation that carries away the energy released in the reaction. However, only part of this energy can be deposited into the phosphor crystals. The rest is deposited into neutron converter crystals and the organic binder (if any) and thus is lost for our purpose. The reactions between neutron and boron ($^{10}$B) are [7]:

$$^10\text{B} + n \rightarrow (7\%)^7\text{Li}(1.0\text{MeV}) + ^4\text{He}(1.78\text{MeV})$$

(1)

$$^10\text{B} + n \rightarrow (93\%)^7\text{Li} + ^4\text{He} \rightarrow ^7\text{Li}(0.83\text{MeV}) + ^4\text{He}(1.47\text{MeV}) + \gamma(0.48\text{MeV})$$

(2)

The neutron capture by the $^{10}$B nucleus is accompanied by the emission of two type of charged particles: alpha particle ($^4$He) with energy of 1.78 MeV(7%)/1.47 MeV (93%) and lithium ($^7$Li) with energy of 1.0 MeV(7%)/0.83 MeV (93%). As the 7% part contribution is minor and the energy of the same particles is similar, only the 93% part was considered. Simulated transfer ranges of these particles in neutron converter and phosphor crystals by the software of Stopping and Range of Ions in Materials (SRIM) [13, 14] are presented in Fig.1.

It is can be see that the average maximum transmission paths of $^7$Li particles in $^3$H$_3$10BO$_3$ are 9.06μm and 5.09μm, respectively. The number of photons produced per single neutron capture event is
proportional to the fraction of the energy deposited into phosphor crystals. This suggests that the transfer ranges of alpha and lithium particles in the neutron converter material must exceed the crystal sizes of the converter. From Fig.1 it follows that the crystals of the H$_3^{10}$BO$_3$ converter have to be at most several micrometers in size. In order to decrease the energy wastage during the charged particles passing through the converter material, the H$_3^{10}$BO$_3$ size should be as small as possible in theory, but when the size was decreased to be nanometre scale, the clumping phenomenon will appeared [15].

As a result, the particle size of H$_3^{10}$BO$_3$ was chose to be 1~3μm in radius. The simulation results show that more than 80% of the nuclear reaction energy is then deposited into H$_3^{10}$BO$_3$ particles. As for ZnS:Ag, the optimal size is determined by the requirement that the charged particle must deposit the remaining energy into phosphor crystals as much as possible. The results were shown in Fig.2.

From Fig. 2a and 2b it is obvious that remaining energy of the charged particles could be deposited into the ZnS:Ag crystals within the 2.5μm radius completely, after they have passed through the H$_3^{10}$BO$_3$
(2μm). Moreover, a mixture with high homogeneity is obtainable if the particles sizes of the converter and phosphor are similar.

So far, the particles sizes of H$_3^{10}$BO$_3$ and ZnS:Ag were determined to be 2μm and 2.5μm respectively. The results of TEMs (Fig.3) show that the particle’s sizes of the raw materials were 1–2.5μm (H$_3^{10}$BO$_3$) and 2.5–4μm (ZnS:Ag), which fulfils the experimental requirements.

3.2. Optimal mass ratio between neutron converter and phosphor

In order to get the maximum amount of photons generated by a given neutron flux, the mass ratio between the converter and the phosphor had been optimized by experiments as indicated above. The result is presented in Fig.4.

The thicknesses of the scintillators with different mass ratios were chosen to be 400μm coincidentally. There is an increase in the neutron counting efficiency as the ratio changes from 4:1 to 6:1, but for higher ZnS(Ag) fractions it results in diminishing returns. The optimal mass ratio of the ZnS(Ag)/H$_3^{10}$BO$_3$ is approximate 6.5:1 depending on the spline connected curve.
3.3. Spatial resolution

In order to evaluate the spatial resolution, the scintillators were partially covered with a cadmium tile and exposed to the thermal neutrons at the neutron radiography facility with accelerator driven D-Be neutron source at Peking University of China. From the image of the cadmium edge the line spread function and the modulation transfer function were calculated. The modulation transfer function obtained in this way is shown in Fig. 5.

![MTF graph](image)

Fig.5 The MTFs of ZnS(Ag)/H$_3^{10}$BO$_3$ scintillators with 0.35mm and 0.40mm thickness respectively

The spatial resolutions are 1.11 lp/mm (MTF=0.1) and 1.33 lp/mm (MTF=0.1), with the thicknesses of 0.40mm and 0.35mm, respectively (Fig.5). It decreases with increasing thickness.

3.4. Neutron radiography of beam purity indicator and sensitivity indicator

Finally, the radiographic quality of the H$_3^{10}$BO$_3$/ZnS(Ag) scintillators was evaluated by inspection of the beam purity indicator (BPI) and beam sensitivity indicator (BSI) [16] at the neutron radiography facility described above. The obtained images were presented in Fig.6.

![Radiography images](image)

Fig.6 NR of beam sensitivity indicator (left) and purity indicator (right)

It is apparent with visual inspection of the beam sensitivity indicator (BPI) radiograph in Figure 6 (right) that the Pb disks are not noticeable darker or lighter than the surrounding polytetrafluoroethylene
(PTFE), therefore, the gamma and pair production content can be deemed to be low. One cadmium wire appeared to be less sharp than the other, thus, the L/D ratio is not sufficient. From the beam purity indicator (BSI) radiograph in Figure 6 (left), it could inspect different thicknesses of acrylic resin and the gaps of 1mm and 0.5mm formed by aluminium shims.

4. Conclusion

The ZnS(Ag)/ $H_3^{10}BO_3$ scintillators were fabricated successfully by a spraying method. In order to improve the performance of the scintillators, the particle sizes of the neutron converter and phosphor materials were optimized to be 2µm and 2.5µm in radius, respectively, based on the results of simulation studies. The mass ratio between the ZnS(Ag) and $H_3^{10}BO_3$ were calculated to be approximate 6.5:1 by the experimental curve. The spatial resolution of the scintillators with the thickness of 0.40mm and 0.35mm are 1.11 lp/mm (MTF=0.1) and 1.33 lp/mm (MTF=0.1) respectively, which are close to the commercially available ZnS:Ag/6LiF scintillators. This indicates that $H_3^{10}BO_3$/ZnS(Ag) scintillator have a potential for application for thermal neutron radiography.

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