The study of zinc sulphide scintillator for fast neutron radiography

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Abstract

Fast neutron radiography is a promising application for accelerators. The potential effectiveness of this technique depends on the development of suitable imaging detectors for fast neutrons. Zinc sulphide based scintillators have the largest light output per event in the family of imaging scintillators used so far in fast neutron radiography. This paper investigated different aspects of this scintillator in order to determine the factors which might affect the light output. A mathematical model was established to estimate effectiveness of this scintillator. Zinc sulphide screens were prepared with ZnS particles of different concentration in polypropylene matrix. A 14MeV fast neutron source was used in the experiments. The light output was detected using a CCD camera or a film coupled to the scintillator screen. The results showed that the optimum scintillators is 3-mm in thickness with the weight ratio of 1:1 to 2:1 for ZnS and polypropylene.

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Key word: zinc sulphide scintillator, neutron radiography, analog calculation, experiment

1. Introduction

Fast neutron radiography is a potentially powerful method for non-destructive inspection and testing due to the extremely high penetration depth of fast neutrons in comparison with other radiation in most materials of industrial interest [1]. Particularly, it has great advantage for detecting voids, cracks or other defects in low-Z materials (e.g. plastics, ceramics, etc.) shielded by thick, high-Z parts [2-4]. This
property of the fast neutrons which makes it unique and valuable also makes it difficult to be detected. Detection efficiency is thus the key factor, particularly with the limited flux usually produced by accelerator based neutron sources.

Most significant methods for the detection of fast neutrons depend on the elastic scattering of protons and the subsequent conversion of the energy of the recoil protons into a number of electrons\(^5\). In turn these are often converted into light in a scintillator material. This light subsequently produces an electrical signal by a CCD, or an amorphous silicon or other semiconductor screens. The cross-section for the scattering of protons is very much smaller than that for the absorption reactions \(^6\)Li(n, \(\alpha\)) or \(^{10}\)B(n, \(\alpha\)) used for detection in thermal neutron radiography, so the detection of neutrons is an intractable problem in fast neutron radiography.

The zinc sulphide scintillator has a highest quantum efficiency in all imaging scintillators used in fast neutron radiography [5]. These screens consist of silver or copper activated zinc sulphide particles suspended in a rich hydrogenous material. The ratio of the two components can vary with the maximum thickness of the scintillator, and hence its efficiency is limited because ZnS is essentially opaque to itself-emitted light. Some of the light is however transmitted through or between the grains and passes through the plastic component to the detection system. Therefore plastic acts a source of recoiled protons as well as a light guide for the scintillations in this type of detector.

2. Theoretical model

As pointed out in the introduction, the performance of the zinc sulphide scintillator for fast neutrons is based on the recoiled protons generated in hydrogenous material and consequent deposition of proton energy in luminophore. The energy lost in the luminophore grains is transferred into visible light with given conversion efficiency. This process is shown in Fig. 1.

![Fig. 1 Neutron detection in the zinc sulphide scintillator](image)

At any point in the scintillator, the neutrons which react with the scintillator can be calculated from the following equation:

\[
I(z) = \frac{dn}{dz} = u_t n_0 e^{-u z}.
\]
Where \( n_0 \) is the total incidence neutron flux, \( \mu_1 \) is the linear coefficient of neutron attenuation. We assume that the number of photons emitted in the screen per neutron is \( k \), \( N_{zns} \) is the atomic density of zinc sulphide powder, \( \delta_{zns} \) is the cross-section of zinc sulphide interacting with a proton. So the photons density can be given:

\[
Q(z) = N_{zns} \delta_{zns} k u_1 n_0 e^{-\mu_1 z} = N_{zns} \delta_{zns} k' e^{-\mu_1 z}
\]  
(2)

Where \( k'=k\mu_1 n_0 \) in the function, it will be constant if the composition of scintillator is certain. We can get the photon energy at this point is:

\[
E(z) = N_{zns} h v k' e^{-\mu_1 z}
\]  
(3)

Where \( h \) is planck constant, \( v \) is frequency of photon. It can be predigested as below:

\[
E(z) = N_{zns} K e^{-\mu_1 z}
\]  
(4)

here \( K=h v k' \). We can assume that the light intensity of \((x,y,z)\) point is \( E_0 \), so the output plane light which contributed by the point is:

\[
E = E_0 \frac{1}{4\pi L^2}
\]  
(5)

\[L = \sqrt{(d-z)^2+(x-x_0)^2+(y-y_0)^2} \]

\( L \) is the light transport distance. On the other hand, the light will be absorbed by scintillator because the ZnS neutron radiography scintillator is opaque. It is assumed that the linear coefficient of light attenuation is \( \mu_2 \), so the output plane light for the contribution of the light point at \((x_0,y_0,d)\) is:

\[
q(x_0, y_0) = \frac{N_{zns} K}{4\pi [(d-z)^2+(y-y_0)^2+(x-x_0)^2]} e^{-\{u_1 z+u_2 [(d-z)^2+(y-y_0)^2+(x-x_0)^2]^{1/2}\}}
\]  
(6)

There are a lot of light points in scintillator, so the total output plane light intensity is:

\[
Q(x_0, y_0) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{0}^{d} \frac{N_{zns} K}{4\pi [(d-z)^2+(y-y_0)^2+(x-x_0)^2]} e^{-\{u_1 z+u_2 [(d-z)^2+(y-y_0)^2+(x-x_0)^2]^{1/2}\}} dxdydz
\]  
(7)

In fact, the output plane is not the \( z=d \) one, it is a plane which have a very short distance between the \( z=d \) plane. Here it is assumed the distance is \( \varepsilon =0.01 \text{mm} \). So we can get:

\[
Q(x_0, y_0) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{0}^{d} \frac{N_{zns} K}{4\pi [(d-\varepsilon-z)^2+(y-y_0)^2+(x-x_0)^2]} e^{-\{u_1 z+u_2 [(d-\varepsilon-z)^2+(y-y_0)^2+(x-x_0)^2]^{1/2}\}} dxdydz
\]  
(8)

3. Calculation results

3.1. Thickness and composition of scintillator vs. intensity of export light

Here \( \mu_1 \) and \( \mu_2 \) is determined by the composition of zinc sulphide scintillator, \( \mu_1=\rho w N_A \delta \mathcal{A} \), here \( \rho \) is density of scintillator, The zinc sulphide scintillator detect neutron by the \((n,p)\) reaction, so \( w \) is the number of hydrogen atoms per polypropylene molecule. The \((n,p)\) cross section of hydrogen(6.89*10^{-1} barn for 14MeV neutron) is much large than carbon(1.06e^{-4} bar for 14MeV neutron), otherwise, the
atomic density of zinc($1.93 \times 10^{-1}$ barn for 14MeV neutron) and sulfur($2.54 \times 10^{-1}$ barn for 14MeV neutron) are too small than polypropylene, so the cross-section of carbon, zinc and sulfur can overlook here. $N_A$ is Avogadro’s number, $\delta$ is 14MeV neutron microscopic cross-section for the H(n,p) reaction, A is the atomic weight of the polypropylene molecule. Polypropylene was adopted as organic hydrogenous material in the paper, the linear coefficient of neutron attenuation $\mu_1$ for the ZnS:Ag+polypropylene can be calculated by the above equation. The linear coefficient of light attenuation $\mu_2$ for the screen ZnS:Ag+polypropylene was taken in Yoshii (1994) $^5$, $\mu_2$ is between $0.3 \text{mm}^{-1}$ and $6.2 \text{mm}^{-1}$ when the ZnS relative content is between 10% to 89%. Now we can calculate light intensity at any point on the surface of scintillator by the equation (3). The calculated curves for thickness and the volume portion of the ZnS vs. relative light intensity for ZnS:Ag+polypropylene scintillator are given in Fig. 2.

![Fig.2 Thickness and the volume portion of scintillator vs. export light intensity](image)

When the thickness of the scintillator $d$ equals 2.2mm, the export light intensity reaches a maximum value. The Fig.2 shows that the export light increases with the thickness of scintillator. On the other hand, the scintillator is opaque to visible light, if the thickness of scintillator exceeds certain value, the export light intensity will decrease. The results show that the thickness of scintillator has an optimal value, if the composition of scintillator is ascertained.

3.2. **Point spread function of scintillator vs. thickness of scintillator**
It is worth while to notice that the resolution can be limited by secondary scattering neutron and light in the scintillator. Fig.3 and Fig.4 show the PSF plotted against a distance from the line neutron source for various scintillators. One can see that the FWHH (full-width at half height) of the PSF increases with the thickness of scintillator. The thickness of scintillator will influence the radiography quality, although the influence may be negligible for a thickness of less than 5 mm. Because the spatial resolution of fast neutron radiography is usually about 1 mm.

3.3. Experiment study

Some zinc sulphide scintillators of polypropylene resin and ZnS powder mixture were developed. The processing technique of zinc sulphide scintillators is showed in Fig.5. A vulcanizer and a set mould is needed. The prototype of zinc sulphide scintillators is showed in Fig.6.
Several pieces of fast neutron scintillator by five types of ZnS fluorescent (ZnS:Ag-Al, ZnS:Cu-Al, p22-g, p22-b, p31) powder were prepared. The decay time of these ZnS fluorescent powder is less than 1ms, graininess is less than 9μm. The ‘ZnS:Ag-Al’ and ‘p22-b’ ZnS fluorescent powder are silver activated, the ‘p22-g’, ‘p31’, ‘ZnS:Cu-Al’ are copper activated. The mixture composition and thickness of each are shown in Table 1.

### Table 1 The mixture composition and thickness of each scintillators

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Composition</th>
<th>Thickness[mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>ZP1</td>
<td>ZnS(wt)/PP(wt)=1:1</td>
<td>1,2,3,4</td>
</tr>
<tr>
<td>ZP2</td>
<td>ZnS(wt)/PP(wt)=2:1</td>
<td>1,2,3,4</td>
</tr>
<tr>
<td>ZP3</td>
<td>ZnS(wt)/PP(wt)=1:2</td>
<td>1,2,3,4</td>
</tr>
</tbody>
</table>

A accelerator was used in the experiment. They were exposed to a neutron flux generated by the T(D,n)He reaction, the neutron energy is 14MeV. The neutron source have an effective yield of $10^{11}$n/sec, along the beam axis and an effective focal spot size of $\leq 6$ mm (FWHM). The scintillators were coupled to a CCD camera with a battery of lens (shown in Fig.7). The specification for the CCD-detectors is shown in Table 2.

### Table 2 Main parameters of CCD chip

<table>
<thead>
<tr>
<th>parameter</th>
<th>unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Format</td>
<td>1024×1024 pixels</td>
</tr>
<tr>
<td>Pixel size</td>
<td>24×24 μm</td>
</tr>
<tr>
<td>Imaging area</td>
<td>24.6×24.6 mm</td>
</tr>
<tr>
<td>Fill factor</td>
<td>100 %</td>
</tr>
<tr>
<td>Dark current</td>
<td>10 pA/cm²</td>
</tr>
<tr>
<td>Sensitivity</td>
<td>2.4 μV/e</td>
</tr>
</tbody>
</table>
3.3.1 The sensitivity of different zinc sulphide scintillators

In the experiment, the distance of neutron source and zinc sulphide scintillators is 1.5m, CCD imaging system was used here, the exposure time is about 15min. The experiment results are shown in Fig.8 and Fig.9. The results indicate the optimal thickness of scintillator is about 3mm, and the best material composition is proportionately about 2:1 for ZnS(wt) and polypropylene(wt). The result approximately corresponds to the above calculation result. Finally, we find the P22-b fluorescent powder has the highest efficiency.
3.3.2 Fast neutron radiography by zinc sulphide scintillator

For assess the imaging quality of zinc sulphide scintillators, the under mentioned experiment was proceeded. Some 5cm thick steel sample were made for the experiment(shown in Fig.10). Fig.11(right) is the image made by on Kodak AA400 film that we have taken based on 3mm thick ZnS scintillator. The 1mm hole in the sample can be distinguished clearly, the distance between the neutron source and zinc sulphide scintillators is 2m , the collimation ratio is 200, the exposure time is about 40min. Fig.11(left) is the image made by on CCD that we have taken based on 3mm thick ZnS scintillator. The 1mm slit in the sample can be distinguished clearly, the distance between the neutron source and zinc sulphide scintillators is 1m , the collimation ratio is 100 the exposure time is about 15 min. The sample was appressed to the zinc sulphide scintillator in all experiment.
3.3.3 The contrast of different fast neutron converters

Plastic scintillator (for instance BC408) and optical fiber matrix can also be used for fast neutron radiography, whose irradiance efficiency is higher than zinc sulphide scintillator. However, the thickness of plastic scintillator and optical fiber matrix are usually several centimeters, which could influence the resolution badly. A 4cm thick BC408, a 5cm thick optical fiber matrix, a 3mm thick zinc sulphide scintillator and a 4cm thick steel block sample were prepared for the experiment to study the performance of different converters of fast neutron radiography.

The luminescent efficiency of the 3mm thick zinc sulphide scintillator (made by p22-b zinc sulphide powder, zinc sulphide weight: polypropylene weight=2:1) and BC408 (4cm thickness) were tested in the experiment (show in Table 3). CCD imaging system was used here, the distance of neutron source and zinc sulphide scintillators is 1m, the sample was appressed to the detector (zinc sulphide scintillator, BC408 and fiber matrix), the collimation ratio is 100, the exposure time is about 15 min. Fig.12 and Fig.13 show that the spatial resolution of zinc sulphide scintillator is the best one in all fast neutron converters, and the boundary of the image of zinc sulphide scintillator is the sharpest.
BC408 scintillator

Fig. 12 Imaging results of steel sample boundary with fast neutron

Fig. 13 Imaging results of steel sample-air interface with different converters

4. Conclusion

In this paper, a mathematical model is developed for analyzing performance of zinc sulphide scintillator on the basis of the fast neutron radiography theory. At the same time, the study result shows the model is approximately correct. The following observations are notable:

a. With the thickness of zinc sulphide scintillator increasing, the efficiency of scintillator is improved, but the light output is reduced because the scintillator is opaque to itself emitted light, and the best thickness of zinc sulphide scintillator is between 2mm and 3mm.
b. The FWHM of the PSF broadens with the thickness of zinc sulphide scintillator increasing, but the influence on image resolution can be neglected if the thickness below 5mm.

c. The composition of zinc sulphide scintillator influence its efficiency, the optimal ratio for ZnS(wt) and polypropylene(wt) is 1.5:1.

d. The p22-b has the highest efficiency in the five zinc sulphides.

e. The image quality of zinc sulphide scintillator is better than plastic scintillator and optical fiber matrix, but the imaging efficiency, which is affected by the thickness, zinc sulphide scintillator is lower than that of plastic scintillator and optical fiber matrix. So the zinc sulphide scintillator is suitable for the fast neutron imaging system which has the highest neutron flux and require highest image resolution.

5. acknowledgement

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Reference


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