EPJ Web of Conferences **225**, 07009 (2020) https://doi.org/10.1051/epjconf/202022507009
ANIMMA 2019

Development of a Coated-Micro-Particle Neutron Detector Based on LiF/ZnS Scintillator

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Abstract—**^{6}LiF:ZnS(Ag) micro-particle neutron detectors are a promising technology to further improve neutron detection capabilities for a variety of applications. Specifically, we have been investigating ^{6}LiF micro-particles coated with ZnS(Ag) to increase the neutron detection efficiency, light production, and light collection efficiency when compared to the existing powder-based technology (EJ-426 from Eljen Technology). Extensive radiation and light transport simulations with single micro-particles have been performed to find the optimal ^{6}LiF diameter and ZnS(Ag) coating thickness. Full-scale multi-particle simulations also have been performed to determine the optimal pitch (particle-to-particle distance) and detector thickness. Randomizations of ^{6}LiF radius, ZnS(Ag) coating thickness, position of particles, as well as shape of particles and partial coating have been performed to account for possible manufacturing imperfections. EJ-426 sheets have been modeled for reference purposes by defining spherical grains of ^{6}LiF and ZnS(Ag) and compared against experiments. The simulation results show that the coated micro-particles should dramatically increase the neutron detection efficiency, light production, and light collection efficiency when compared to the existing EJ-426 technology.**

Keywords*—*Micro-particles, ^{6}LiF:ZnS(Ag), Neutron-detection optimization, Geant4

I. INTRODUCTION

Neutron detectors based on ^{6}LiF:ZnS(Ag) are used for various applications including neutron imaging, high efficiency cold neutron detection [1,2] and neutron multiplicity counting [3]. Current existing technology employs mixing of ^{6}LiF and ZnS(Ag) powders to create the active volume for neutron detection. Although this technique works well, it has some serious shortcomings such as clustering of the same type of grains and non-uniform grain size, which reduce both charged particle energy deposition in ZnS(Ag) and light propagation through the detection medium. In order to overcome these problems and increase the neutron detection efficiency, it was proposed to coat ^{6}LiF particles with ZnS(Ag) and distribute these coated micro-particles throughout the volume to create the active detection medium.

Optimization of ^{6}LiF radius, ZnS(Ag) coating thickness, pitch (particle-to-particle distance), and detector thickness is required to find the ideal geometry for neutron capture, charged particle energy deposition (light production) and light propagation. Geant4 and MCNP6 have been used to optimize the ^{6}LiF radius and Geant4 has been used to determine the optimal ZnS(Ag) coating thickness, micro-particle pitch, and detector thickness since it has a capability of simulating light photons, as well as neutrons and charged particles.

After determining the optimal dimensions, randomizations of particle position, particle size, particle shape, and partial coating have been performed within a specified tolerance to account for the possible future manufacturing imperfections.

Four different EJ-426 sheets have been modeled to compare the performance of coated micro-particle detectors to existing technology.

II. OPTIMIZATION

Extensive simulations were performed to determine the various optimal parameters of the geometry and individual optimization steps are described in the following sections.

An illustration of the proposed geometry is shown in Fig. 1.

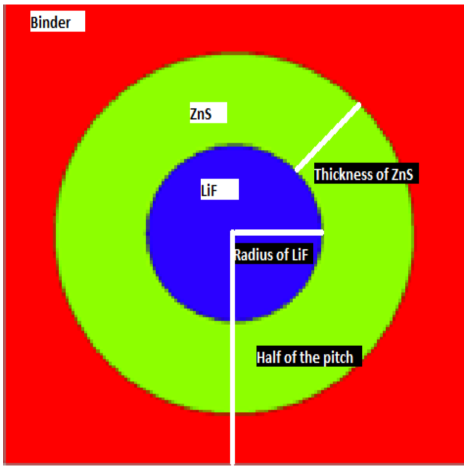

Fig. 1. Illustration of a single coated micro-particle (created with MCNP6).

A. Optimization of LiF radius

First, the radius of ^{6}LiF particles was optimized. ^{6}LiF radius should be chosen large enough to capture as many neutrons as possible, but small enough so that the alpha and triton particles upon neutron capture can escape the neutron converter volume without substantial energy loss.

Coating thickness was set constant at a relatively large value (90 μm) and ^{6}LiF radius was varied. Pencil beam of thermal neutrons were incident on the single particle and the energy deposition in scintillator medium per primary neutron was tallied in both Geant4 and MCNP6. The results are shown in Fig. 2.

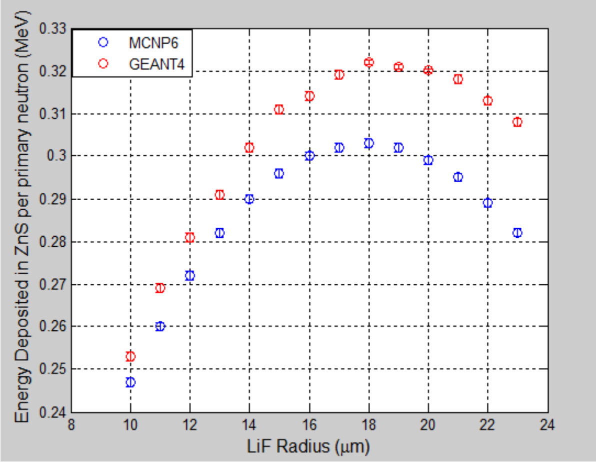

Fig. 2. Energy deposited in ZnS per primary thermal neutron as a function of LiF radius simulated in MCNP6 and Geant4.

Although there is a relatively small disagreement between the codes regarding how much energy is deposited in ZnS(Ag), both codes have agreed upon the optimal radius for ^{6}LiF, which was found to be 18μm.

B. Optimization of ZnS coating thickness

After determining the optimal ^{6}LiF radius, the ZnS(Ag) coating thickness was optimized. Although ZnS(Ag) is a bright scintillator (50000 photons/MeV) [4], its relative opacity to its own luminescence limits its practical size when it is used as the scintillating medium for radiation detection. Therefore, the optimization is required to find the ideal thickness. The coating thickness should be large enough to capture most of the energy from the charged particles, but small enough so that the light produced in ZnS(Ag) can escape without significant internal attenuation.

^{6}LiF radius was set constant at its optimal value of 18μm and ZnS(Ag) thickness was varied. The number of light photons escaping a single micro-particle per primary thermal neutron was tallied in Geant4. The results are shown in Fig. 3.

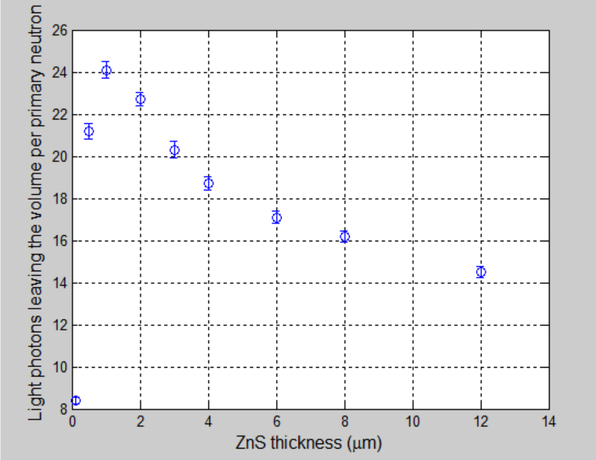

Fig. 3. Number of light photons leaving the micro-particle as a function of coating thickness in Geant4.

Optimal ZnS(Ag) thickness was found to be 1μm. Although approximately 85% of the charged particle energy is not deposited in ZnS(Ag) at this thickness, the produced light photons escape more efficiently. Besides the small absorption length of the ZnS(Ag), another problem is the higher refractive index of

ZnS(Ag) (2.49) compared to the refractive index of ^{6}LiF (1.39). In addition, the refractive index of the organic binder simulated is 1.51. This refractive index mismatch causes substantial internal reflection, which results in less efficient light collection.

C. *Optimization of pitch and detector thickness*

Optimization of micro-particle pitch and detector thickness requires the simulation of a large number of micro-particles. Pitch is the particle-to-particle distance and it is measured from the centers of the particles. The pitch needs to be small enough to maximize the concentration of ^{6}Li, but large enough to allow for efficient light propagation without significant attenuation. Therefore, the binder material needs to have favorable optical properties; e.g., it has to be transparent to light.

In simulations, micro-particles were placed at a constant pitch inside the detector medium and circular thermal neutron beam was made incident on the detector surface. Keeping the detector cross-sectional area constant, pitch and detector thickness were varied to find the optimal geometry. The active detector volume was placed between two polyester sheets and the light photons were collected on the photocathode surface with the same cross-sectional area as that of the scintillator.

An illustration of the simulation geometry is shown in Fig. 4.

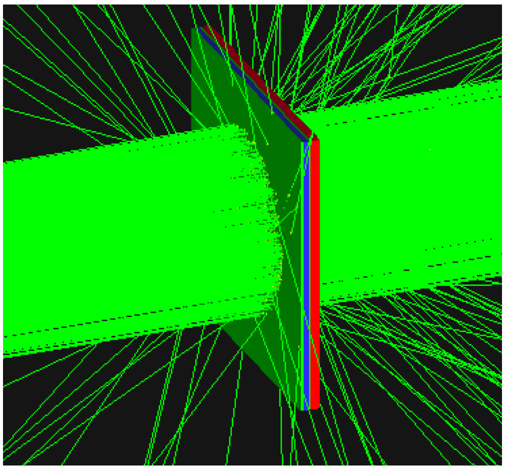

Fig. 4. Illustration of full-scale simulation geometry. Green, blue, and red surfaces represent polyester, active detector volume, and photocathode surface, respectively.

The ideal detector thickness and the micro-particle pitch are correlated since the specification of a certain pitch size dictates the optimal detector thickness. If the pitch is large, the detector thickness can also be relatively large since the areal density of ZnS(Ag) in the detector would be less than in the case of a smaller pitch. The results are shown in Fig.s 5 and 6.

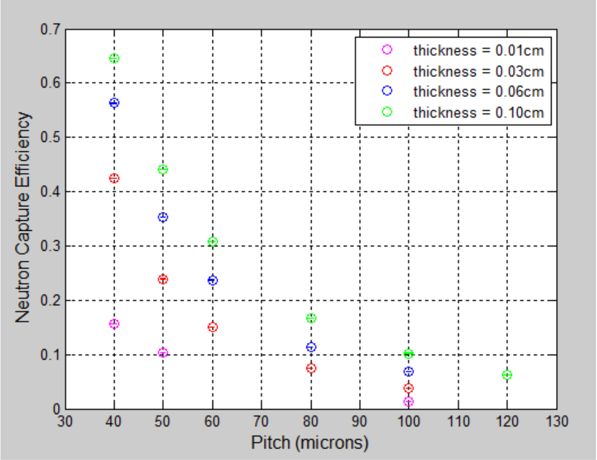

Fig. 5. Neutron capture efficiency as a function of pitch for different detector thicknesses.

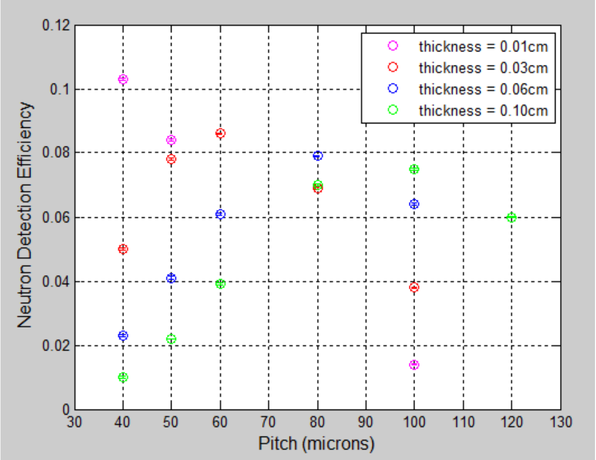

Fig. 6. Neutron detection efficiency as a function of pitch for different detector thicknesses with a light photon threshold of 20.

Neutron capture efficiency decreases with increased pitch and with smaller detector thickness, because of smaller amount of ^{6}Li in the active detection volume. Neutrons are attenuated exponentially in the micro-particle geometry according to

$$I = I_0 \exp(-\Sigma t) \qquad [Eq. 1]$$

where I, Σ, and t are the neutron intensity, the total macroscopic cross section, and the effective detector thickness, respectively. In Eq.1, t is not the overall detector thickness but rather the thickness that is occupied by ^{6}LiF, the neutron capturing material. The attenuation in ZnS(Ag) and binder can be neglected for small thicknesses.

Neutron detection efficiency is defined as the number of neutrons that are captured and subsequently result in the detection of 20 light photons on the photocathode surface. Detection of 20 photons include quantum efficiency of the photocathode surface so it is the number of photoelectrons generated upon a neutron capture event. This number of detected photons is somewhat arbitrary; however, it is well above the single photoelectron noise signal level. It was observed that decreasing this threshold increases the neutron detection but doesn't affect the overall trends.

It is shown in Fig. 6 that there is no single optimal pitch size that applies to each detector thickness. Instead, it is a function of detector thickness and it decreases as the detector thickness increases since the detector thickness compensates for the sparsely placed micro-particles. However, when the thickness is made smaller and smaller, the overall neutron detection efficiency increases and peaks at 0.1mm detector thickness, suggesting that photon propagation properties dominates over the neutron capture probability.

III. RANDOMIZATION

The aforementioned optimization of the micro-particle parameters was performed assuming an ideal geometry with perfectly spherical particles, symmetric placement of these particles throughout the detector volume and perfect coverage of ^{6}LiF with ZnS(Ag) without any bare surfaces.

It is possible to have coated micro-particles with non-ideal dimensions, different shapes and partial coverage in the manufacturing process. Moreover, these microparticles may not be aligned perfectly to have constant pitch between them. It is expected that the deviation from optimal dimensions and shape would result in worsened charged particle energy deposition and light propagation through the detection medium.

Therefore, randomization of these parameters was performed in Geant4 within certain tolerance levels. An illustration of randomization steps is shown in Fig. 7.

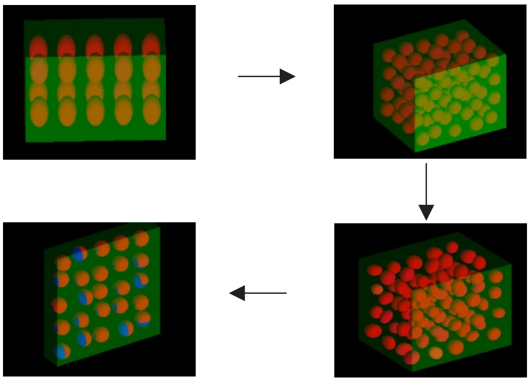

Fig. 7. An illustration of several randomization steps: a) Ideal geometry with symmetrically placed particles and fixed LiF and ZnS size, b) randomization of position of micro-particles, c) randomization of shape, rotation, and position of micro-particles, and d) randomization of coating coverage.

Since there are thousands of micro-particles in a given detector volume, due to the complexity of simulation models it is not feasible to randomize all of them individually. Instead, a conventional unit cell was defined with multiple micro-particles in it and only those particles were randomized. After that randomization step, the conventional unit cell was repeatedly placed throughout the detector medium. The effect of the number of randomized micro-particles is shown in Fig. 8.

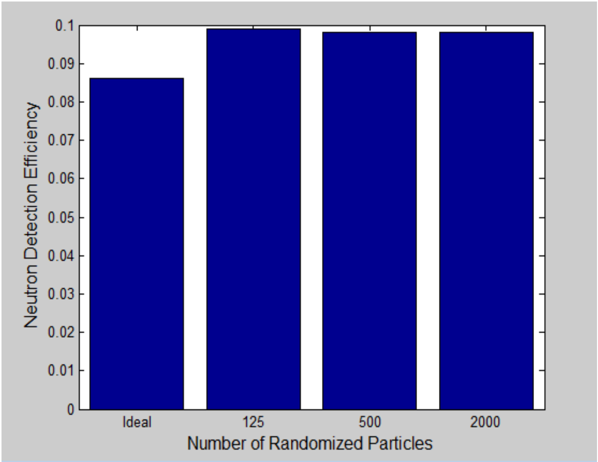

Fig. 8. Neutron detection efficiency as a function of number of particles in a conventional unit cell.

As shown in Fig. 8, the neutron detection efficiency quickly saturates at 125 randomized micro-particles. It is also observed that the neutron detection efficiency is somewhat increased when the geometry deviates from ideal. The main reason is that there are empty spaces in the ideal geometry where a neutron has no chance of undergoing a capture reaction with ^{6}Li. Since circular thermal neutron beam is directly incident on the detector some of the source neutrons travel through these empty spaces. However, when the particles are randomly placed, some of these empty spaces are filled with ^{6}LiF, increasing the neutron capture probability.

After the randomization of the position of particles, the ^{6}LiF radius, and the ZnS(Ag) coating thickness was carried out. The overall particle size was kept constant and only ^{6}LiF and ZnS(Ag) dimensions were randomized. Again, the particles were randomly placed inside the conventional unit cell. The results are shown in Fig. 9.

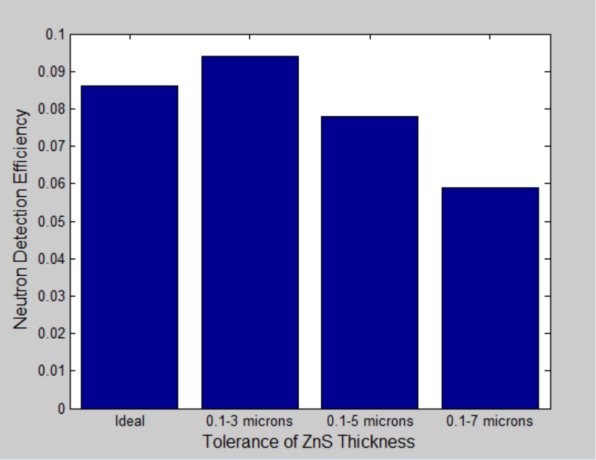

Fig. 9. Neutron detection efficiency as a function of different tolerance intervals of ZnS thickness.

Although the deviation from the optimal ZnS(Ag) thickness result in deterioration of the neutron detection efficiency, a random placement of micro-particles surpasses the effect of non-ideal ZnS(Ag) thickness when the tolerance is between 0.1µm-3µm. However, when the tolerance is increased, neutron detection efficiency clearly suffers.

In addition, the randomization of shape and coating coverage was performed, and their effects are shown in Fig. 10 and 11.

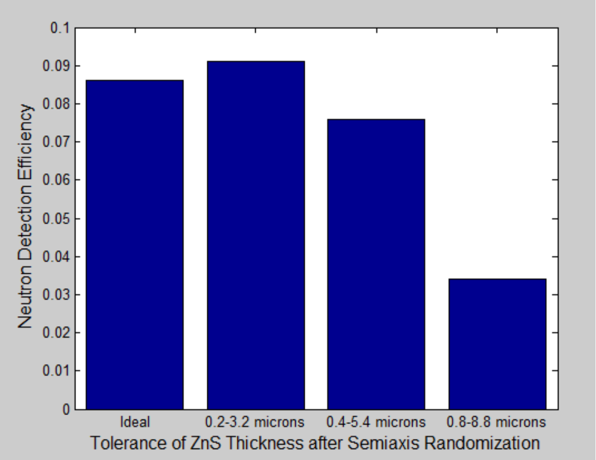

Fig. 10. Neutron detection efficiency as a function of different tolerance intervals of ZnS thickness. The difference is that the shapes are not spherical but ellipsoid.

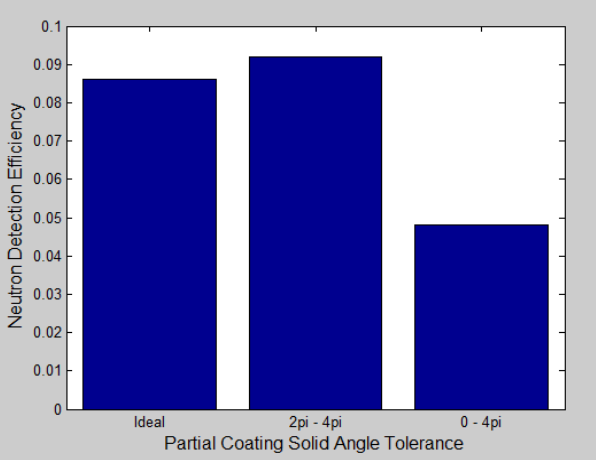

Fig. 11. Neutron detection efficiency as a function of different tolerance intervals of solid angle for partial coating.

Again, there is a small increase in the neutron detection efficiency compared to the ideal case when the tolerance interval is narrow. When the tolerance intervals become wider, the particles become more distorted and the detection efficiency suffers significantly.

When the partial coating tolerance is between 2π-4π steradians, the neutron detection efficiency increases compared to the ideal case. However, the position randomization is always implicit so the partial coating itself actually decreases the neutron detection efficiency. The effect is much pronounced when the tolerance interval is narrow. When the tolerance interval is made 0-4π, the detection efficiency decreases rapidly.

IV. EJ-426 SIMULATIONS

In addition to the micro-particle simulations, EJ-426 sheets were simulated in Geant4. ^{6}LiF and ZnS(Ag) grains were modeled as spheres and placed in a unit cell respecting the LiF:ZnS mass ratio and LiF concentrations of these sheets. A unit cell represents the building block of the sheets which comprise multiple unit cells.

An illustration of a unit cell is given in Fig. 11.

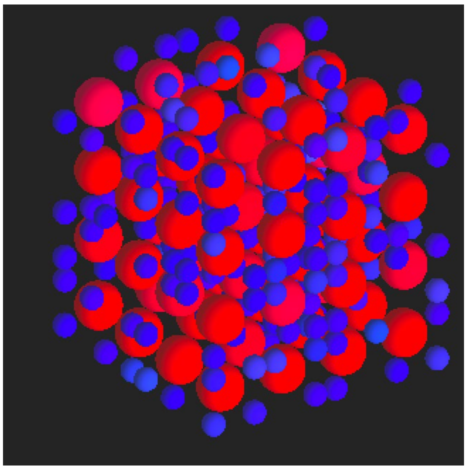

Fig. 12. An illustration of a unit cell in an EJ-426 sheet.

Again, a circular thermal neutron beam was incident on the detector sheet as shown in Fig. 4. Four different EJ-426 sheets with different LiF:ZnS mass ratios and thicknesses were tested and the results are given in TABLE I.

Detector Thickness (mm)	LiF:ZnS Mass Ratio	Neutron Detection Efficiency (%)
0.32	1:3	1.9
0.32	1:2	2.6
0.60	1:3	1.5
0.60	1:2	1.6

TABLE I. Neutron detection efficiencies for different EJ-426 sheets.

With the same cross sectional detector area and light photon threshold, it is observed that the best performing EJ-426 sheet is the 0.32mm thick one with the LiF:ZnS mass ratio of 1:2. As seen from Fig. 6, most of the coated micro-particle detectors have higher neutron detection efficiencies than 2.6%. While 0.1mm thick micro-particle detector performs almost 4 times better, the 0.32mm thick micro-particle detector performs 3 times better than EJ-426.

V. CONCLUSION

Coated micro-particles were optimized and the ideal ^{6}LiF radius, ZnS(Ag) coating thickness, pitch, and detector thickness were found by performing extensive simulations of neutrons, charged particles, and light photons in Geant4. EJ-426 sheets were also modeled and simulated in Geant4. The neutron detection efficiencies of coated-micro-particle detectors for thermal neutrons were found higher than the best performing EJ-426 sheet, by a factor of 2-4. Non-idealities were also tested in coated micro-particle geometries and it was observed that the proposed technology under imperfect conditions still performs better than the existing technology.

ACKNOWLEDGEMENT

The authors would like to thank the Penn State's College of Engineering and the Applied Research Laboratory for supporting this research.

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