# Neutron Shielding Gd<sub>2</sub>O<sub>3</sub> – Polymer Coating Development for Dry Cask Storage System

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## 1. Introduction

Due to the saturation of spent nuclear fuel wet storage facilities, the importance of developing dry storage casks that ensure both economic efficiency and safety has emerged [1]. Also, the potential increase in nuclear fuel enrichment will require higher neutron shielding capability for dry cask storage systems.

Previous studies have focused on developing neutron absorbers within the basket used to maintain the subcriticality of spent nuclear fuel. For instance, the cold spray additive manufacturing method was used to coat B<sub>4</sub>C/Al composite onto an aluminum blanket surface [2]. Another study proposed that the design of epoxy contains Gd<sub>2</sub>O<sub>3</sub> [3]. These studies emphasize the potential to enhance the neutron shielding performance of the dry cask storage system through the design of neutron absorbing coatings.

It is desired to develop efficient neutron shielding materials for the conventional Transportation and Storage Cask to accommodate higher enriched spent fuel assemblies. This issue can be addressed by adding neutron shielding materials to the existing dry cask storage system through coating technologies. The intended application of these coatings is for the outer surface of canisters or the inner surface of casks. The neutron shield coatings can be applied to a complex geometry and has the advantage of enhancing performance while maintaining the current design.

# 2. Overview of Dry Cask Storage System (DCSS)

The Dry Cask Storage System (DCSS) consists of three major parts: Basket, Canister, and Cask.

The Basket consists of a bundle of fuel tubes that hold spent nuclear fuel rods. The specifications of the Basket, i.e., the number of fuel tubes, vary depending on its design. To maintain the subcriticality of spent nuclear fuel, a neutron absorber is inserted between adjacent fuel tubes. The commonly used neutron absorber is a  $B_4C/Al$  composite.

The Canister has a metal shell structure that encloses the Basket, and it is typically made of 304L stainless steel. The Canister provides structural support to the Basket, isolates the nuclear fuel rods from the external environment, and partially shields radiation.

The Cask can be classified into three types depending on its purpose: storage cask, transport cask, and dualpurpose cask for both storage and transport. In the past, separate casks were used for transport and storage. Storage casks usually use concrete or metal as a neutron shield. Transport casks use polymers or hydrogencontaining materials as neutron shielding to reduce weight and enhance mobility. Recently, the use of dualpurpose casks for both storage and transport has been increasing due to economic and safety reasons. This trend is driven by the delay in the construction of final disposal facilities, leading to the expansion of interim storage facilities. The dual-purpose casks shield neutrons using polymers, hydrogen-containing materials, and boron, similar to transport casks.

In this background, this study aims to develop neutron absorbing  $Gd_2O_3$ -polymer coating for conventional DCSS to enhance the neutron shielding capability. The developed coatings have been analyzed experimentally and the neutron shielding performance will be simulated using the Monte Carlo method.

# 3. Modeling

The following represents the process of simulating a simplified dry cask storage system with the neutron shielding coatings using the Monte Carlo N-Particle (MCNP) simulation.

# 3.1 Geometry of Dry Cask Storage System

In this study, neutron shielding was simulated by applying the specifications of the KORAD-21, which is designed as dual-purpose cask with cylindrical shape. As shown in Table 1, the inner diameter of the KORAD-21 cask is 1,696 mm, and the internal height is 4,905 mm [4], while the outer diameter is 2,126 mm, and the external height is 5,285 mm [5]. The canister has an outer diameter of 1,686 mm and a height of 4,267 mm. The cask is made of carbon steel [4], while the canister is made of stainless steel [5].

Table 1. KORAD-21 Design

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	Cask (mm)	Canister (mm)	
Outer Diameter	2,126	1,686	
Outer Height	5,285	4,267	
Inner Diameter	1,696	-	
Inner Height	4,905	-	
Material	Carbon Steel	Stainless Steel	

3.2 Neutron Source and Neutron Flux Modelling

Since a goal of the simulation is evaluating the neutron shielding capability of  $Gd_2O_3$ -polymer coating material, the following conditions were assumed to construct the model. Fig. 1 describes thermal neutrons located inside the canister, emitting through the cylindrical surface and bottom and top lids. The coating material is applied to the inner surface of the cask surrounding the canister. The ratio of neutron flux between the incident and emitted coating surfaces will be determined using MCNP calculation. Using this ratio, the attenuation of neutron flux is calculated, and the macroscopic neutron cross-section is obtained.



Fig .1. The simulated cylindrical structure

### 4. Experimental

Gd<sub>2</sub>O<sub>3</sub>-polymer coating was applied to a Zn-plated steel substrate, followed by surface characterization. Polyester was used as the epoxy binder mixing with gadolinia powder, and the coating thickness was about  $50 \pm 10 \ \mu\text{m}$ . The coating was prepared by an external company using the electrostatic coating method. The electrostatic coating is a process where electrically charged coating particles are attracted to a surface with an opposite charge.

Surface analysis was conducted for Zn-plated steel coated with  $Gd_2O_3$ -polymer composites where concentration of  $Gd_2O_3$  powder were varied such as 50wt.%, 70wt.%, and 90wt.%. The coating was prepared using a mixture of gadolinia powder and polyester binder based on the mass fraction.

The coating phase, morphology, and thickness were examined using X-Ray Diffraction (XRD) and Optical Microscopy (OM), respectively. To measure the coating thickness, the sample was hot-mounted vertically, polished, and then its cross-section was observed using OM.

### 5. Results and Discussions

#### 5.1 MCNP Simulation

The calculation for macroscopic cross section was conducted as a function of the concentrations: 50wt.%, 70wt.%, and 90wt.% Gd<sub>2</sub>O<sub>3</sub>, which were tested in the experiment (see Fig.2). Additionally, since the coating

stability was not great at 70wt.% Gd<sub>2</sub>O<sub>3</sub> in the experiment, another simulation (see Fig. 4) was performed at higher concentrations than 70wt.%.



Fig. 2. Macroscopic total neutron cross-section depending on Gd<sub>2</sub>O<sub>3</sub> concentration

As the concentration increases, the neutron shielding efficiency improves due to the increase in macroscopic neutron cross-section. At concentrations above 85wt.%, the macroscopic neutron cross-section exceeds 1200 cm<sup>-1</sup>, which is approximately 70 times higher than the neutron cross-section of the conventional B<sub>4</sub>C/Al composite neutron absorber 16.9 cm<sup>-1</sup> as absorption cross-section[7]. This means that the same shielding performance can be achieved using a coating that is 70 times thinner than the conventional neutron absorber.

#### 5.2 Experimental Analysis of Gd<sub>2</sub>O<sub>3</sub>–Polymer Coating

In the low-magnification images, it was observed that as the concentration of gadolinia decreased, more peeloff of the composite coating occurred. The volume fraction was calculated using high-magnification images (see Table 2). There was a significant difference between the mass fraction that the manufacturer provided and the one measured. The coating thickness was determined through OM (see Table 2), and it was found that the coating thickness slightly increased as the concentration decreased. The coating thickness was measured to be in the range of 50  $\pm$  10  $\mu$ m. The tendency of peel-off and coating thickness at low concentrations are attributed to coating porosity. It is speculated that a higher content of Gd<sub>2</sub>O<sub>3</sub> powder reduces the porosity in the Gd<sub>2</sub>O<sub>3</sub>-polymer coating. The porosity decreased due to the surface tension effect of the Gd<sub>2</sub>O<sub>3</sub> powder particles in the binder composite.

 
 Table 2. Coating Characteristics Depending on Concentration

Weight fraction (%)	90	70	50	
Thickness (µm)	44.8	52.6	57.6	
Volume fraction (%)	9.0	7.8	1.7	

For 90wt.% coating, where delamination was hardly observed, the gadolinia peak was predominantly detected, and it can be concluded that the coating remains robust. The observed gadolinia XRD peak corresponds to the FCC phase. At 70wt.% which is delamination started to be observed, and at 50wt.% which is severe delamination occurred, the XRD results showed that the Zn peak from the substrate was dominant. To ensure neutron shielding capability and coating integrity, a certain level of concentration of gadolinia is required.

## 6. Conclusions and Further Works

In this study, the neutron shielding  $Gd_2O_3$ -polymer composite coatings were developed and their efficacy was evaluated using MCNP simulations as a function of  $Gd_2O_3$  concentration. In the thermal neutron region, the macroscopic neutron cross-section value increased as the  $Gd_2O_3$ -concentration increased. While fabrication of the  $Gd_2O_3$ -polymer coatings, it is speculated that a more number of  $Gd_2O_3$  powder in the polymer binder reduced the coating's porosity due to its surface tension effect leading to a more structurally stabilized coating layer. As the  $Gd_2O_3$  concentration increases, the  $Gd_2O_3$ polymer coating is presumed to exhibit higher neutron shielding capability and improved coating integrity.

Future studies will focus on increase in the  $Gd_2O_3$  concentration to optimize coating integrity and neutron shielding performance. Additionally, neutron irradiation experiments would be conducted to verify the neutron shielding capability which observed in the MCNP simulation.

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