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An evaluation of the design and performance for a new neutron absorber based on an artificial rare-earth compound

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ABSTRACT

In this study, a neutron absorber based on an artificial rare earth compound, which is a radioactive waste generated from pyro-process, is proposed for use in spent fuel storages. To secure the stable control of criticality with physical and chemical durability, a neutron absorber was designed and fabricated using borosilicate glass and a rare earth compound. The performance of the developed neutron absorber was evaluated in terms of the: (1) criticality controllability with various artificial rare earth compositions, (2) stability after neutron irradiation generated from the spent fuel, (3) radioactivity of the neutron absorber, and (4) physical and chemical properties. Our results show that the neutron absorber can successfully control the criticality regardless of the artificial rare earth composition. Also, we demonstrate that the neutron absorber can be utilized without any additional radiation shielding of the spent fuel storages for a long period of time (more than 100 years). In addition, analysis shows that the absorber has sufficient physical and chemical strength for use in spent fuel storage. We expect that this study will help to minimize the number of radioactive waste storage sites as well as reduce the disposal costs.

1. Introduction

The artificial rare earth compounds used in this study are from radioactive waste generated while pyro-processing spent nuclear fuels (Lee et al., 2011). It has been noted that the nuclides in artificial rare earth compounds have high radioactivity (Kim, 2012) because they are produced by the fission reaction of fissionable materials. Therefore, the disposal costs of these materials are substantial and securing an appropriate disposal site is difficult. In a previous study, a method to reuse artificial rare earth compounds as neutron absorbers in spent fuel storages was first performed (Kim, 2012). Conventionally, for dense spent fuel storage in a limited disposal site, expensive neutron absorbers have been used as the criticality control material in spent fuel storage. On the other hand, the method reusing artificial rare earth compounds to control the criticality has advantages on reducing the number of radioactive waste disposal sites and has additional economic benefits. However, in spite of the advantages, reusing radioactive wastes as criticality control materials has led to some technical problems. These include the uncertainty of the nuclide compositions of artificial rare earth compounds, which are dependent on the nuclear fuel and burn-up conditions, and whether or not the physical and chemical durability are suitable for use in spent fuel storage. Therefore, to use artificial rare earth compounds in spent fuel storage, the following considerations must be verified: (1) the neutron absorbing performance with various artificial rare earth compositions under burn-up conditions, (2) the sustainability of the neutron absorbing performance caused by the decay of unstable nuclides in the compound, (3) the stability of the criticality controllability with neutron irradiation generated from the spent fuels, (4) the chemical stability and uniformity of the nuclides in a neutron absorber, (5) the physical strength to withstand external shock, (6) the thermal stability to withstand increased temperature.

In this study, a new neutron absorber, designed by mixing artificial rare earth compounds and borosilicate glass materials, is proposed to secure stable criticality control as well as physical and chemical durability. Using the design of the proposed neutron absorber, criticality controllability was evaluated in a conceptual design for spent fuel storage under various conditions. In addition, after manufacturing trial neutron absorbers, the chemical and physical properties were estimated.

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2. Design of new neutron absorber

2.1. References for the criticality safety and the neutron absorber

The basic design requirements for neutron absorbers were investigated by examining the regulation guideline (ANSI/ANS-8.1, 1998b; ANSI/ANS-8.17, 2004; ANSI/ANS-8.24, 2007) and ASTM documents (ASTM:C1671-07, 2007; ASTM:C750-03, 2010). Among various design requirements, the main criticality safety requirements are given as follows:

- According to the evaluation criteria of critical analysis for spent fuel storage and transport cask, the effective multiplication factor should be less than a 0.95 within 95% confidence interval including any bias and uncertainty for normal, abnormal, and accidental conditions.

The verification and certification for neutron absorbers based on boron oxide are as follows:

- Generally, neutron absorbers that have been loaded into fuel storage are inaccessible and difficult to monitor. Therefore, it is necessary to verify that the performance of the neutron absorbers can be maintained for an extended period of time.
- The main issues for verification of neutron absorbers are whether the absorbers are durable, possess satisfactory physical properties, and have a homogeneous distribution of the neutron absorbing nuclide. Therefore, it should be confirmed that the physical properties of the neutron absorber do not change when it is subjected to external influences. This confirmation is made through various tests such as visual inspection, mechanical inspection, and the neutron attenuation test.

2.2. Design of the neutron absorber and spent fuel storage

To meet the criticality safety design, all uncertainties, which are about the code verification, manufacturing uncertainty, and the other accident scenarios, should be considered and evaluated. With the $k_{eff} < 0.95$ criterion referred in Section 2.1, the goal of the multiplication factors in this study was set to $k_{eff} < 0.91$, empirically. To design a neutron absorber with chemical and physical durability and stable control of criticality, using the borosilicate glass materials mixed with artificial rare earth compounds was proposed. The optimum composition of the neutron absorber was defined through trial fabrication (Choi et al., 2014). The neutron absorber density and the mass ratio of materials constituting the absorber are as follows:

- Density of neutron absorber: 3.8–4.1 g/cm^3 (depending on the nuclide composition of RE_2O_3).
- Shape: cylindrical.
- Radius: 0.55 cm.
- Accumulated height: fuel active height of each assembly.
- Material composition.
- RE_2O_3 = 50 wt%.
- SiO_2 = 25 wt%.
- B_2O_3 = 8.33 wt%.
- Al_2O_3 = 16.67 wt%.

RE_2O_3 is an artificial rare earth tri-oxide that it can be directly extracted from one of the pyro-chemical processes during the pyro-processing of spent nuclear fuels. To obtain a representative of the artificial rare earth compound, a burn-up calculation was performed under the following conditions:

- Initial fuel enrichment (to extract the artificial rare earth compound): 5 wt%.
- Total burn-up (to extract the artificial rare earth compound): 45 GWD/MTU.
- Property of initial fuel: without gadolinium burnable poisons (Group 1).
- Cooling time: 10 years (the cooling time until the fabrication of the neutron absorber is predicted more than 10 years).

PLUS7 (an improved model of CE 16 × 16) is the main assembly types currently used in Korea. Thus, ORIGEN-S code (Bowman and Leal, 2009) was employed for the burn-up calculation using the CE 16 × 16 library (a previous model of PLUS7 with minor design changes). The mass ratios in the neutron absorber calculated are shown in Table 1. Group 1 indicates the composition of the artificial rare earth compound without gadolinium burnable poison while Group 2 has the burnable poisons. Fig. 1 shows the trial products of the neutron absorbers based on the Groups 1 and 2 conditions.

In this study, a cylindrical neutron absorber with a radius of 0.55 cm was designed to be able to load the neutron absorber into the guide tubes of each fuel assembly type. The cylindrical shape of the absorber was determined to consider two aspects which are the manufacturing strength and high criticality control ability of the absorber. Also, the radius of the absorber was decided to insert it into both WH 17 × 17 and PLUS7 guide tubes as well as considering manufacturing process cost. When designing the spent fuel storage for WH 17 × 17 (Wagner et al., 2001) and PLUS7 (Sohn and Kim, 2011), the size and position of the guide tube in each fuel assembly type is not identical. For this reason, the arrangement of the neutron absorber was designed differently according to the form of the fuel assembly. The preliminary designs of WH 17 × 17 and PLUS7 spent fuel storages made in this study are shown in Fig. 2. The fuels in WH 17 × 17 are located in a 17 × 17 lattice structure; each fuel is cylindrical with 0.41 cm radius and 10.412 g/cm^3 effective density. Twenty-five guide tubes with an inner diameter of 0.56134 cm are positioned in the fuel assembly as shown in Fig. 2(a). The fuels and guide tubes were shrouded in a cladding of ZIR-4. The pitch between the fuels is 1.25984 cm. More information on the WH 17 × 17 assembly is shown in Table 2. The fuels in the PLUS7 assembly are located in a 16 × 16 lattice structure; each fuel is cylindrical with a 0.41 cm radius and an effective density of 10.313 g/cm^3. Five guide tubes with an inner diameter of 1.1430 cm are positioned in the fuel assembly as shown in Fig. 2(b). More information on the PLUS7 assembly is shown in Table 3. For the criticality control, in this study, the

Table 1
Isotopes and mass ratios included in the neutron absorber for group 1.

Zaid number	Mass ratio	Zaid number	Mass ratio	Zaid number	Mass ratio
B-10	0.004745366	Ce-144	0.01025268	Sm-147	0.00232727
B-11	0.021133872	Pr-141	0.04407384	Sm-148	0.00657481
O-16	0.341965771	Pr-142	0.00000282	Sm-149	0.00005495
Al-27	0.088208408	Pr-143	0.00054749	Sm-150	0.01352001
Si-28	0.107778807	Nd-142	0.00163719	Sm-151	0.0004944
Si-29	0.005457303	Nd-143	0.02209677	Sm-152	0.00543124
Si-30	0.003622621	Nd-144	0.04965084	Sm-153	0.00004906
Y-89	0.01349185	Nd-145	0.02444869	Sm-154	0.0022301
Y-90	0.00000451	Nd-146	0.03142699	Eu-153	0.00614456
Y-91	0.00091401	Nd-147	0.00022287	Eu-154	0.00127595
La-139	0.05029868	Nd-148	0.01597756	Eu-155	0.00031976
La-140	0.00008922	Nd-150	0.00880913	Eu-156	0.00028948
Ce-140	0.05246751	Pm-147	0.00466863	Gd-154	0.00014041
Ce-141	0.00152523	Pm-148	0.00003291	Gd-156	0.00803455
Ce-142	0.04556667	Pm-149	0.00004716	Gd-157	0.00000573
Ce-143	0.00005728	Pm-151	0.00000912	Gd-158	0.0018759