Nuclear Engineering and Technology 50 (2018) 788–793

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Technical Note

Performance evaluation of METAMIC neutron absorber in spent fuel storage rack

Kiyoung Kim [a,*], Sunghwan Chung [a], Junhee Hong [b]

[a] Central Research Institute, Korea Hydro & Nuclear Power Co., Ltd., 70, Yuseong-daero 1312 beon-gil, Yuseong-gu, Daejeon 34101, Republic of Korea
[b] Chungnam National University, 99 Daehak-ro, Yuseong-Gu, Daejeon, 34134, Republic of Korea

ARTICLE INFO

Article history:
Received 8 September 2017
Received in revised form
28 November 2017
Accepted 28 January 2018
Available online 15 February 2018

Keywords:
Blister
Criticality
METAMIC
Neutron Absorber
Neutron Attenuation Test
Scanning Electron Microscope

ABSTRACT

High-density spent fuel (SF) storage racks have been installed to increase SF pool capacity. In these SF racks, neutron absorber materials were placed between fuel assemblies allowing the storage of fuel assemblies in close proximity to one another. The purpose of the neutron absorber materials is to preclude neutronic coupling between adjacent fuel assemblies and to maintain the fuel in a subcritical storage condition. METAMIC neutron absorber has been used in high-density storage racks. But, neutron absorber materials can be subject to severe conditions including long-term exposure to gamma radiation and neutron radiation. Recently, some of them have experienced degradation, such as white spots on the surface. Under these conditions, the material must continue to serve its intended function of absorbing neutrons. For the first time in Korea, this article uses a neutron attenuation test to examine the performance of METAMIC surveillance coupons. Also, scanning electron microscope analysis was carried out to verify the white spots that were detected on the surface of METAMIC. In the neutron attenuation test, there was no significant sign of boron loss in most of the METAMIC coupons, but the coupon with white spots had relatively less B-10 content than the others. In the scanning electron microscope analysis, corrosion material was detected in all METAMIC coupons. Especially, it was confirmed that the coupon with white spots contains much more corrosion material than the others.

1. Introduction

Korea Hydro & Nuclear Power Co., Ltd (KHNP) is the nation's largest electric power company, generating approximately 31.5% of the total electric power produced in South Korea. It operates 25 nuclear power and hydroelectric plants. KHNP recently performed a test to verify the performance of neutron absorber materials in spent fuel (SF) pool racks. Neutron absorber materials were placed between fuel assemblies to allow the storage of fuel assemblies in close proximity to one another. The purpose of neutron absorber materials is to preclude neutronic coupling between adjacent fuel assemblies and to maintain the fuel in a subcritical storage condition. Various neutron absorber materials have been used in SF pools to increase the storage capacity of SF. METAMIC is one of the commonly used neutron poison materials and boron carbide–aluminum composite materials for reactivity control in SF storage racks. According to KHNP procedures, coupon surveillance tests, such as visual inspection dimensional measurement and measurements of dry weight and density except for B-10 area density, have been performed about 10 times during the past 50 years. Recently, degradation, such as white spots, was detected on the surface of METAMIC surveillance coupons. Under these conditions, the material must continue to serve its intended function of absorbing neutrons. This article examines the performance of METAMIC surveillance coupons through neutron attenuation test, conducted for the first time in Korea. Also, scanning electron microscope (SEM) analysis was performed to verify the white spots that were detected on the surface.

2. Test method

2.1. METAMIC surveillance coupon

METAMIC is a material that has been used for criticality control in SF storage racks as well as storage and transportation casks for nuclear fuel. In these applications, it is typically used in plate form with approximate dimensions of 5–8 inches width, 12 feet length,

* Corresponding author.
E-mail address: kiyoungkim@khnp.co.kr (K. Kim).

https://doi.org/10.1016/j.net.2018.01.017

and up to 0.125 inches thickness. A plate of METAMIC is generally sealed within the rack structure and cannot readily be subjected to in-service inspection. So, when new fuel storage racks are installed, a series of surveillance coupons of the neutron absorber material are placed in the racks. The coupons are housed in a specially designed assembly and placed in a storage cell surrounded by recently discharged SF assemblies. Periodically, one or two coupons are removed from the assembly and sent to a laboratory for testing and inspection [1–3].

METAMIC is an aluminum–boron carbide composite produced via a powder metallurgy process. The feed materials for METAMIC are boron carbide and AA 6061 powder. The boron carbide (B_4C) particulate is fine and classified using the Federation of European Products of Abrasive (FEPA) method, which assures a tighter distribution of particle sizes. The AA 6061 powder particle size is also carefully controlled to obtain a homogeneous finished product. METAMIC generally consists of boron carbide loading over the range of 15–40 vol. % [4].

Five METAMIC surveillance coupons were harvested from a KHNP unit to verify the performance of the neutron absorber materials. A lot of white spots were detected on the surface of the two coupons. So, neutron attenuation test and SEM analysis were performed. SEM analysis is generally used to verify the ingredients of the white spots. Fig. 1 shows METAMIC surveillance coupons that were removed from the SF pool. For reference, all of them were set in 2008 [5].

2.2. Neutron attenuation test

The neutron attenuation test is designed to identify the B-10 area density in the neutron absorber. The neutron attenuation test facility used in this research generates about 60 neutrons, less than 0.6 eV per second, and irradiates neutrons on the surface of a neutron absorber for 20 min per coupon. Neutron attenuation tests were performed for all of the METAMIC surveillance coupons.

2.2.1. Test facility

Fig. 2 shows the neutron attenuation test facility. It consists of a graphite layer, an SP^9-He^3 detector, and a neutron source. The graphite layer moderates fast neutrons and generates thermal and epithermal neutrons; the SP^9-He^3 detector measures thermal and epithermal neutrons. The neutron source is Am^{241}-Be (α,n). Neutrons that are generated in the test facility are thermal and epithermal neutrons. So, neutrons are measured before the Cd filter to exclude epithermal neutrons over 0.6 eV from total neutrons. Fig. 3 shows the neutron spectrum with a Maxwell–Boltzmann distribution that is irradiated on the surface of coupons.

2.2.2. Test procedure

The neutron attenuation test is generally performed at five locations on the surveillance coupon, as shown in Fig. 4. With this approach, a neutron beam is transmitted through a METAMIC surveillance coupon. The measured B-10 area density is compared to the preirradiation B-10 area density to verify the performance of the neutron absorber. The procedure of the neutron attenuation test is shown below.

(a) The SP^9-He^3 detector is set within the thermal and epithermal neutron field and shielded by the Cd cover (thickness: 0.0236 inch). Then, the detector measures the count rate, which is defined as Re. Re is the count rate of neutrons over 0.6 eV.
(b) The count rate measured after removing the Cd cover is defined as Rt. Rt is the total count rate.
(c) The difference between Rt and Re is the count rate of neutrons less than 0.6 eV, defined as Rth. Rth = Rt−Re
(d) The surveillance coupon is set in the test facility, and the count rate (St) is measured without a Cd cover.
(e) The count rate (Se) is measured with Cd cover in place. Se is defined as the count rate of neutrons over 0.6 eV penetrating the surveillance coupon.
(f) The difference of St and Se is the count rate of neutrons less than 0.6 eV penetrating the surveillance coupon. It is defined as Sth. (Sth = St−Se)
(g) The transmission rate of a surveillance coupon is defined as P. (P = Sth/Rth = (St−Se)/(Rt−Re). About an average of 60 neutrons per second were irradiated for 20 min. As can be seen in Table 1, the uncertainty of the transmission rate is ±0.0035%. Uncertainty is defined as the root mean square error of the transmission rate for all of the tests.

Fig. 1. METAMIC surveillance coupons. (A) Coupons without white spots. (B) Coupons with white spots.

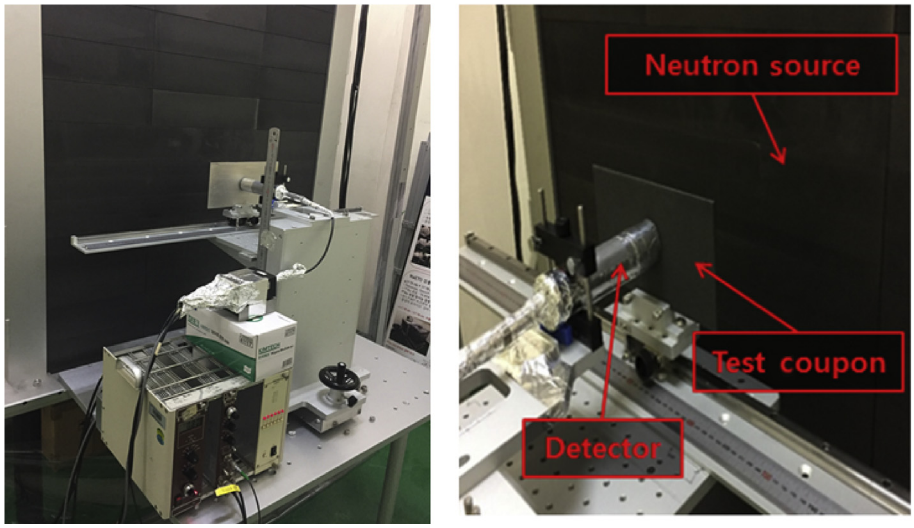

Fig. 2. Neutron attenuation test facility.

(h) Convert the transmission rate into a B-10 area density by MCNPX, Ver. 2.7.0 code [6].

2.3. SEM analysis

A SEM is a type of electron microscope that produces images of a sample by scanning the surface with a focused beam of electrons. The electrons interact with atoms in the sample, producing various signals that contain information about the surface topography of the sample and composition. In this study, SEM equipment was used to analyze the composition of the white spots on the surface of the METAMIC surveillance coupons. Fig. 5 shows the SEM equipment used to analyze the composition of white spots.

3. Test results

3.1. Performance of METAMIC surveillance coupon

The objective of this study is to verify the performance of METAMIC surveillance coupons. The neutron attenuation test was performed to measure B-10 area density at five locations on all of the surveillance coupons. With this approach, a neutron beam less than 0.6 eV is transmitted through the METAMIC surveillance coupons. Measured B-10 area density (postirradiation) is compared to the initial condition (preirradiation). The preirradiation condition means the manufactured condition.

Table 1 gives the measured B-10 area density and preirradiation condition of the METAMIC surveillance coupons. As a result, coupons without white spots, such as HU3C10, HU3C11, and HU4C10, as well as the HU4C11 coupon with low-level white spots showed no significant sign of boron loss. Boron levels were even higher than in the preirradiation condition. However, the HU3C13 coupon with high-level white spots has less B-10 area density than the preirradiation condition. About 1.2 % boron loss occurred in the HU3C13 coupon. For reference, the limit of boron loss in the KHNP procedure is 5% [7].

3.2. Composition of white spots on the surface of METAMIC surveillance coupon

SEM analysis was performed to verify the composition of the white spots on the surface of METAMIC surveillance coupon. SEM scans a sample with a focused electron beam and delivers images with information about the samples' topography and composition. Fig. 6 shows surface micrographs of the METAMIC surveillance

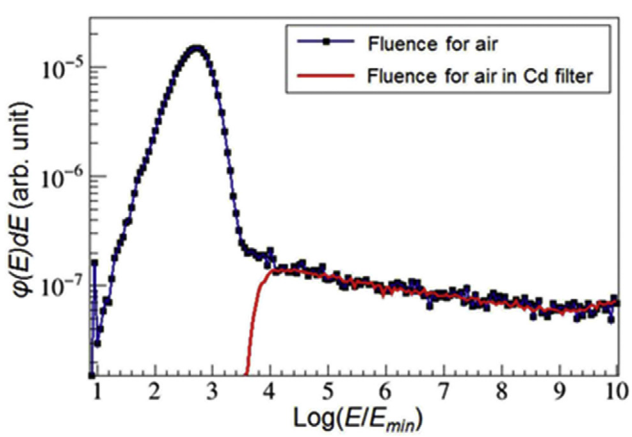

Fig. 3. Neutron spectrum.

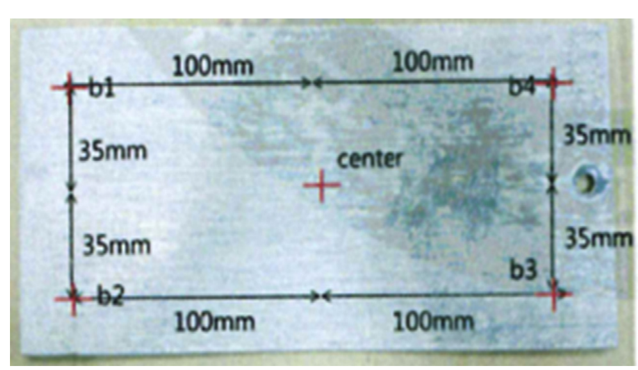

Fig. 4. Test location (center, b1, b2, b3, b4).

Table 1
B-10 area density of METAMIC surveillance coupons.

Coupons	Location	P (%) ± error (%)	B-10 (g/cm^2)	Preirradiation (g/cm^2)	Relative difference
			Postirradiation		
	center	0.28 ± 0.0034	0.03708	0.03393	+2.3%
	b1	0.42 ± 0.0035	0.03388		
	b2	0.37 ± 0.0034	0.03490		
	b3	0.41 ± 0.0035	0.03408		
	b4	0.40 ± 0.0035	0.03429		
	average	0.38 ± 0.0035	0.03470 ± 0.00006		
	center	0.33 ± 0.0034	0.03572	0.03332	+2.3%
	b1	0.40 ± 0.0035	0.03429		
	b2	0.45 ± 0.0034	0.03338		
	b3	0.43 ± 0.0035	0.03368		
	b4	0.43 ± 0.0036	0.03368		
	average	0.41 ± 0.00348	0.03408 ± 0.00006		
	center	0.45 ± 0.0034	0.03353	0.03300	-1.2%
	b1	0.50 ± 0.0036	0.03295		
	b2	0.52 ± 0.0035	0.03258		
	b3	0.54 ± 0.0035	0.03222		
	b4	0.52 ± 0.0035	0.03175		
	average	0.51 ± 0.0035	0.03260 ± 0.00006		
	center	0.31 ± 0.0034	0.03623	0.03297	+5.9%
	b1	0.48 ± 0.0036	0.03295		
	b2	0.33 ± 0.0034	0.03572		
	b3	0.37 ± 0.0036	0.03490		
	b4	0.34 ± 0.0035	0.03551		
	average	0.37 ± 0.00352	0.03490 ± 0.00007		
	center	0.31 ± 0.0034	0.03623	0.03145	+8.4%
	b1	0.43 ± 0.0035	0.03368		
	b2	0.46 ± 0.0034	0.03324		
	b3	0.43 ± 0.0035	0.03368		
	b4	0.42 ± 0.0036	0.03388		
	average	0.41 ± 0.00348	0.03408 ± 0.00006		

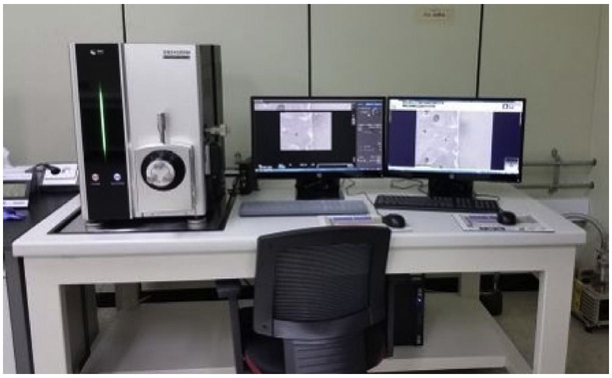

Fig. 5. Scanning electron microscope equipment.

coupons. Sample coupons HU3C13 and HU4C11 include white spots. Fig. 7 shows the surface composition of the METAMIC surveillance coupons. Basically, the METAMIC coupons are composed of boron, carbon, and aluminum. From the results of SEM analysis, corrosion material containing oxygen is found to be included on the surface of all of the METAMIC surveillance coupons. Especially, the HU3C13 coupon, with high levels of white spots, includes much higher oxygen content than the other coupons. Thus, it can be concluded that corrosion is progressing in the METAMIC neutron absorber.

4. Conclusion

The neutron absorber METAMIC is the main material for criticality control in SF pools. KHNP has harvested five METAMIC surveillance coupons to verify the performance of METAMIC from the KHNP SF pool. Three coupons were clear, but the others contained white spots on their surfaces. Thus, for the first time in Korea, a neutron attenuation test and SEM analysis were performed.

B-10 area density was measured through the neutron attenuation test to verify the performance of the METAMIC surveillance coupons. As the result of the neutron attenuation test, it was confirmed that the coupons without white spots, e.g. HU3C10, HU3C11, and HU4C10, as well as the HU4C11 coupon, with low level white spots, experienced no significant sign of boron loss. Boron levels were even higher than preirradiation condition. However, the HU3C13 coupon, with high-level white spots, exhibited a reduced B-10 area density (−1.2%: relative difference) compared to the preirradiation condition. The results are shown in Table 1. Relative difference is defined as the rate of postirradiation over preirradiation.

SEM analysis was performed to verify the composition of the white spots on the surface of the METAMIC surveillance coupons. Basically, METAMIC material is composed of boron, carbon, and aluminum. As for the results of the SEM analysis, corrosion material containing oxygen was conformed on the surfaces of all of the METAMIC surveillance coupons. Thus, the HU3C13 coupon, with high levels of white spots, contained much higher oxygen content than the others. Thus, corrosion is progressing in the METAMIC neutron absorber.

The degradation of METAMIC coupons was confirmed as evidence showing boron loss or surface corrosion, even though the B-10 area density of the coupons was less than the -5% limit. Therefore, surveillance coupons should be monitored every cycle because the surface corrosion can accelerate under wet conditions (e.g. in the SF pool). According to KHNP procedures, coupon surveillance tests have been carried out about 10 times over the past 50 years. KHNP has a plan to revise the KHNP procedure for inspection

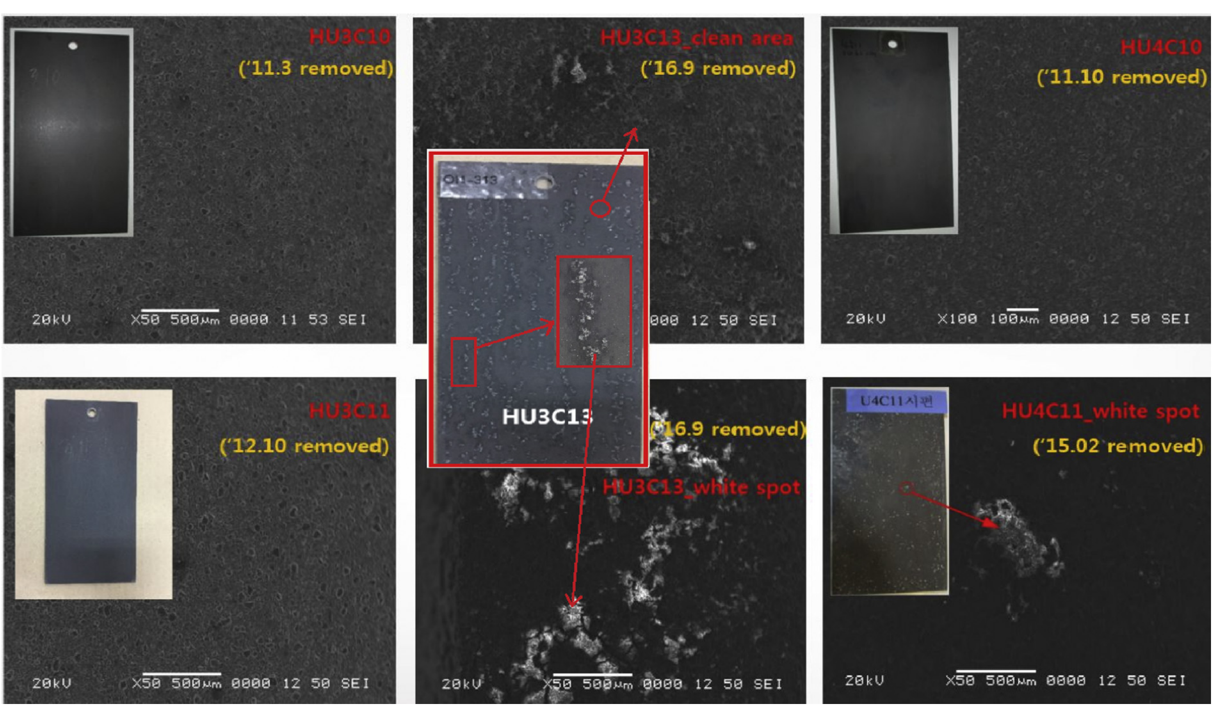

Fig. 6. Surface micrographs of METAMIC coupons.

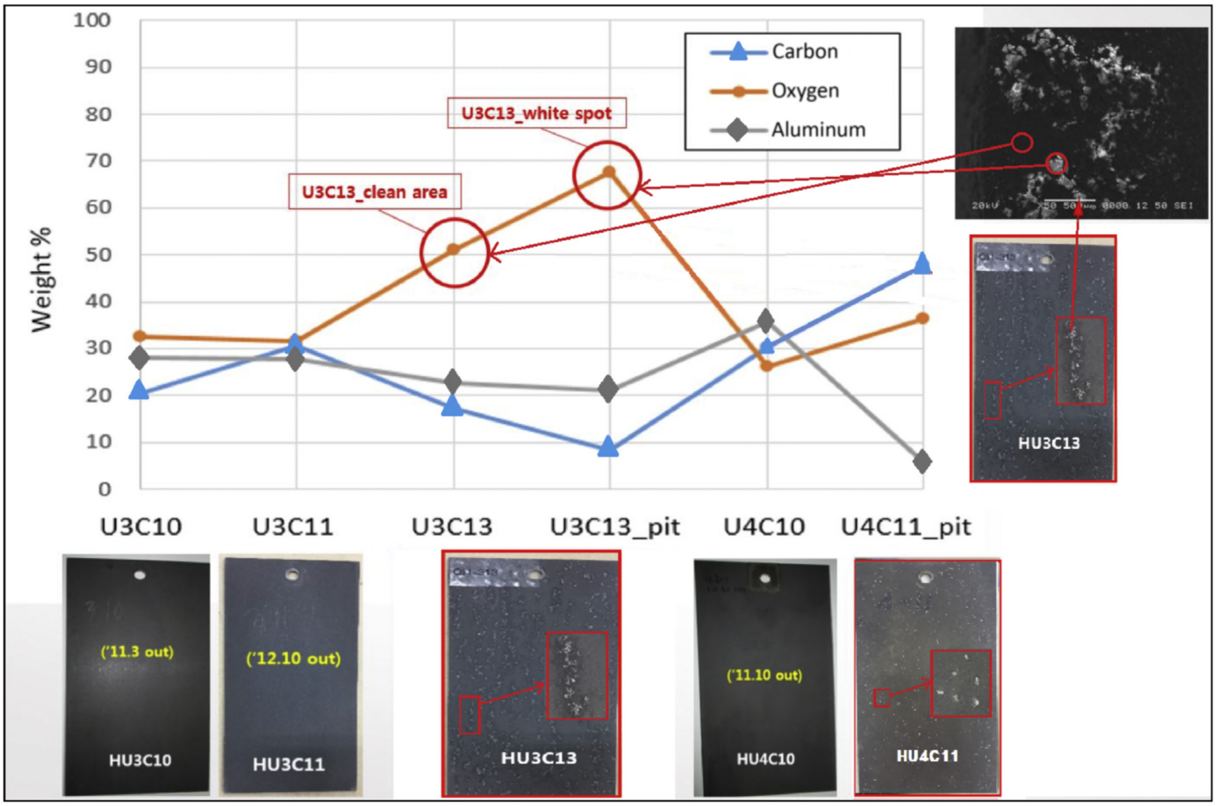

Fig. 7. Surface composition of METAMIC coupons.

period, including the neutron attenuation test. Further, KHNP is proceeding with a project to verify the root cause of white spots, to prevent further corrosion.

Conflicts of interest

None.

Acknowledgments

This work was supported by the research and development program of the Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Ministry of Trade, Industry and Energy (MOTIE; 20141710201 66B).

References

[1] W. Emma, Degradation of Neutron-Absorbing Materials in Spent Fuel Pool, Nuclear Regulatory Commission, USA, 2009. NRC Information Notice 2009–2026.

[2] A. Machiels, Overview of BORAL Performance Based Upon Surveillance Coupon Measurements, EPRI, USA, 2010, 1021052.

[3] H. Akkurt, K. Cummings, Overview of Neutron Absorber Materials Used in Spent Fuel Pools, International Conference on Nuclear Criticality Safety, 2015.

[4] A. Machiels, R. Lambert, Handbook of Neutron Absorber Materials for Spent Nuclear Fuel Transportation and Storage Applications, EPRI, USA, 2009, 1019110.

[5] Ulchin Units 3 and 4 Spent Fuel Pool Region 1 and Region 2 Criticality Evaluation, HOLTEC, USA, 2007. HI-2073685.

[6] Gregg W. McKinney, MCNPX User's Manual Version 2.7.0, LANL, USA, 2011. LA-CP-1100438.

[7] KHNP Operation Procedure, Fuel-1012 Inspection of Surveillance Coupon in Spent Fuel Storage Rack. Rev04, KHNP, KOREA, 2017.