Experimental emulation of neutron absorber degradation under spent nuclear fuel dry storage environment

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1. Introduction

Aluminum-boron carbide (Al- B_4C) metal matrix composite (MMC) is one of the most widely used neutron absorber in various spent nuclear fuel storage systems to assure the criticality safety. Long-term safety and performance of the neutron absorber has become a critical issue in nuclear industry, since the longer-term interim/temporary storage of SNF has become inevitable due to the delay in final disposal of SNF [1]. In the case of Korea, the final repository facility is planned to be secured until 2060 and until then, the interim SNF storage, utilizing dry cask, is also planned to be constructed to secure the SNF storage capacity [2].

The premature degradation of Al-B₄C neutron absorber (i.e., pit corrosion, unexpectedly fast B-10 depletion, and bubble accumulation) was recently discovered from the surveillance coupon used in domestic spent nuclear fuel pool (SPF) for ~8 years [3]. The limitation of current absorber qualification tests has been suggested based on the findings and very few studies has been conducted to properly evaluate the safety and performance of Al-B₄C absorbers [4-6]; however, only for the absorber used in wet storage environments.

In this study, the degradation of $Al-B_4C$ neutron absorber was emulated by conducting high-temperature gas corrosion test on irradiated absorber sample.

2. Methods and Results

Commercial Al-B₄C MMC type neutron absorber, MAXUS[®], was provided by Nikkeikin Aluminium Core Technology Co., Ltd and utilized to evaluate the safety and performance for the SNF dry storage application.

The improved test methodology was utilized in this study, composed of the series of ion-beam irradiation and high-temperature humid gas corrosion test to investigate the possible synergistic degradation of the absorber.

2.1 Radiation damage emulation

In-situ high-temperature ion irradiation was conducted by utilizing 120-keV ion beam accelerator in Korea multi-purpose accelerator complex (KOMAC) to emulate radiation damage accumulated mainly via $10^{B}(n, \alpha)^{7}$ Li reaction. Various radiation damage levels

(0.01, 0.1, and 1 dpa) were emulated at 400 °C, corresponding to the regulatory limit temperature of SNF cladding for conservative evaluation.

The significant amount of helium bubbles was discovered even from the lowest tested dose (0.01 dpa) and accumulation of bubbles was observed at the interfacial boundaries (Fig.1) [7].



Fig. 1. Intragranular helium bubbles within the Al alloy matrix of the absorber observed in 0.01 dpa (left) and helium bubble coalesced at grain boundary observed in 1 dpa case (right) [7].

2.2 Emulation of corrosion behavior under dry storage environments

The high-temperature humid gas corrosion test was conducted on the ion-irradiated absorber specimens for 500 hours at 400 °C by utilizing gas corrosion loop (Fig. 2) consist of mass flow controller (MFC) and test section (furnace). The humidity condition was chosen by assuming internal pressure of dry canister at the off-normal condition (~7 atm) is solely exerted by water vapor. The humidity of the system was maintained by controlling the flow of argon gas and water, and monitored by flowmeter attached on the MFCs (Fig. 3).



Fig. 2. Schematic illustration of high-temperature humid gas corroion loop.



Fig. 3. Flow of water and agron gas during the corrosion test measured with the software, FlowPlot.

2.3 Post characterization

The microstructure of the absorber specimens after the series of ion irradiation and high-temperature corrosion test was analyzed by using transmission electron microscopy (HR-TEM, JEM-2100F, JEOL) and TEM specimens were prepared by utilizing lift-out technique of focused ion beam (FIB, Helios 450HP, FEI). The absorber samples were intermittently discharged from the test section for micro-scale characterization after 100, 300, and 500 hours.

The formation of the corrosion layer was confirmed by energy dispersive spectroscopy (EDS) mapping analysis, which showed non-uniform thickness of oxygen-rich layer at the surface of the absorber (Fig 4). The corrosion layer seemed to preferentially grow along the interface between Al alloy matrix and secondary phase within the matrix; which may indicate possible intergranular corrosion behavior of the Al-B₄C neutron absorber under the dry storage environments.



Fig. 4. STEM-EDS mapping of the absorber after the hightemperature corrosion test.

Further analysis utilizing line-EDS confirmed that the corrosion layer was formed up to the depth of ~500 nm

after the 300 hours of corrosion test on irradiated absorber sample (Fig. 5)



Fig. 5. Line-EDS analysis on the absorber after the hightemperature corrosion test.

The number of intragranular helium bubbles was seemed to be decreased after the 500 hours of hightemperature irradiation and the average size of intergranular helium bubbles was increased (Fig. 6); compared to that observed right after the in-situ high temperature ion irradiation.



Fig. 6. Microstructure of the irradiated absorber sample after the 500 hours of high-temperature corrosion test.

3. Conclusions

The degradation of Al-B₄C MMC neutron absorber under the dry storage environments was experimentally emulated. The series of ion irradiation and hightemperature corrosion test was conducted on the absorber specimens to properly evaluate the material degradation induced by high energy radiation and high temperature condition of SNF dry storage.

Microstructural evolution of the Al-B₄C MMC induced by ion irradiation and high-temperature corrosion test was notable, especially at the interfacial boundaries. The corrosion induced by water vapor progressed along the interface of secondary phases up to ~500 nm. The average size of intergranular helium bubbles was seemed to be increased and tend to agglomerate at the interfacial boundaries after the high-temperature corrosion test.

The longer-term corrosion test will be conducted on the absorber specimens to fully understand the synergistic effect of irradiation and corrosion on the degradation of the absorber for the extended usage of neutron absorber in SNF dry storage.

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