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A Preliminary Study on Time Limited Aging Analysis for Radiation Embrittlement of Components in Dry Storage System

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1. Introduction

In dry storage systems (DSS), Metal alloys consist of most part of the dry storage systems are neutron irradiated. High energy neutron (>1 MeV) is the dominant source of metal alloys' embrittlement [1]. Due to the radiation embrittlement, ability of a material to resist fracture is degraded (i.e., loss of fracture toughness and ductility) [2].

The primary mechanism of radiation embrittlement is the hardening as a consequence of irradiation. High energy neutron generates high energy recoil atoms by scattering and reactions as a result it produces lattice defects. The primary defects are in the form of single and small clusters of vacancies and self-interstitials. Diffusion of primary defects occur and defect clusters, dislocation loop etc. are formed. These defects suppress the movement of the dislocation, so that the yield strength of the material increases (i.e., hardening) [1].

US NRC estimated the accumulated high energy neutron fluence conservatively (i.e., 70 GWD/MTU average burnup, 4.0 wt% initial enrichment, 40 Fuel assemblies, 100 years of storage) during the extended dry storage period (~100 years) was about 10^{16} #/cm^2 [2]. Usually, threshold of high energy neutron fluence that induced degradation of intended function was about 10^{18}-10^{22} #/cm^2 in most of the materials that consist of DSS components [2].

Especially, polymer-based neutron shielding material, the threshold for radiation embrittlement has been found to be 10^4 Gy for polyethylene and significantly lower for other polymers. Radiation can reduce ductility and fracture toughness altering the polymer structure by molecular-scission and cross linking. It was reported that this dose can be reached in 10-100 years for specific SNF, so that it was a credible aging mechanism for polymer materials [2]. But there were no specified data evaluating the radiation susceptible SSCs in DSS.

The purpose of this study is identifying any SSCs that can be susceptible for radiation embrittlement and providing fundamental data for Time Limited Aging Analysis (TLAA) relevant to managing the aging (i.e., radiation embrittlement) of components in DSS. In this study, we evaluated the safety margin between threshold of radiation dose/neutron fluence (i.e., high energy neutron fluence, absorbed dose) and calculated (using Monte Carlo simulation code) irradiated radiation doses /neutron fluence in each SSCs.

2. Methods and Results

For estimating radiation fields in DSS, MCNP 6.2 computational code and ORIGEN-ARP module was used (see Fig. 1). Firstly, source term was calculated using ORIGEN-ARP module. Source term can be categorized into neutron sources (the main neutron producing mechanisms are spontaneous fission and (α,n) reaction) and gamma sources (^{60}Co activity of the steel structural material, decay of fission products, (n,γ) reactions in the material consisting DSS and environment). Secondly, for generating emitted radiations from the source, the source term data (i.e., energy spectrum of gamma/neutron) were provided into MCNP 6.2 code. Thirdly, for transporting neutron and gamma, the cross section library for gamma/neutron and geometrical/dimensions and material composition data of all SSCs (including helium gas etc.) consisting of dry storage cask and environment were entered. For scoring entering radiation into the components, F4 (for neutron fluence)/F6 (for deposited energy; absorbed dose) tallies were used. Finally, for evaluating radiation susceptibility, gap analysis was conducted.

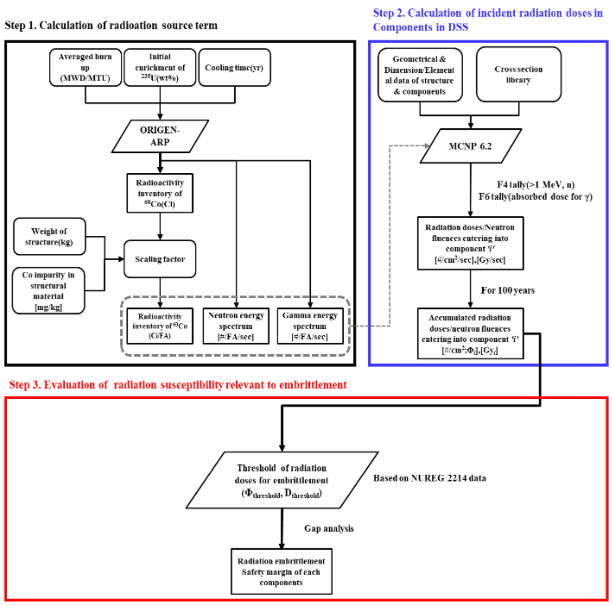

Fig. 1. Framework for evaluating radiation susceptibility of components in dry storage system (DSS) due to radiation embrittlement ('i' indicates type of component)

2.1 Source term calculation

Since late 2010 in Korea, the initial enrichment of nuclear fuel used for Pressurized Water Reactor (PWR) increased to about 4.5 wt%, so that the maximum burnup increased to 55 GWD/MTU and expected to increase [3].

The characteristics relevant to spent nuclear fuel was assumed considering conservative irradiated neutron fluences and gamma absorbed doses [4]. Cooling time was set to 20 years for satisfying maximum allowable heat load according to the reference metal cask (see Table I) [4]. Calculation results for neutron and gamma production rate per Fuel Assembly (FA) are shown in Fig. 1 and Fig. 2.

Table I: Assumptions for characteristic data of spent nuclear fuel in this study [3, 4]

Input parameters	Description
Initial enrichment	4.5 wt%
Averaged burnup	55 GWD/MTU
Weight of U	461.5 kg
Fuel type	WH 17×17(RFA)
ARP cross-section library	W 17×17
Active fuel region length	381 cm
Width of FA	21.4 cm
Density of active fuel region	3.77 g/cm^3
Cooling time after discharge	20 years
Irradiation schedule	1090 days

In case of spent fuel assembly hardware, the structural materials are composed of various alloys of stainless steel, Inconel etc. There are some impurities such as Cobalt (Co), Niobium (Nb), Nickel (Ni), and become neutron activated. Due to the low Nb and Ni impurities, their radioactivity inventory generated by neutron activation was negligible, so that in this study we only consider ^{60}Co ($T_{1/2}$=5.27 yr) activation products. Eq. 1 is used to calculate ^{60}Co radioactivity in FA hardware region [5].

$$A_{f\text{-hardware}} = W_{i\text{-hardware}} \times F \times A_{f\text{-active fuel region}} \quad (1)$$

$A_{f\text{-hardware}}$=Activity of activated radionuclide ‘f’ in hardware [Bq]
$W_{i\text{-hardware}}$=Weight of nuclide *‘i’* in hardware [g]
F=Scaling factor of hardware [-]
$A_{f\text{-active fuel region}}$=Activity of activated radionuclide *‘f’* in active fuel region in case 1 g of nuclide *‘i’* was existed in fresh nuclear fuel [Bq]

Table II: Assumptions for characteristic relevant to hardware components of fuel assembly in this study [6, 7]

Component	Scaling factor	Material	Composition of Co impurity	Weight [kg]
TOP spacer grid	0.2	Inconel-625	10,000	0.8
Bottom spacer grid	0.3	Inconel-625	10,000	1.4
Top end fitting	0.1	Stainless steel 304	800	12.9
Bottom end fitting	0.2	Stainless steel 304	800	7.3
Cladding +Middle support grid	1.0	Ziracaloy -4	20	125.1

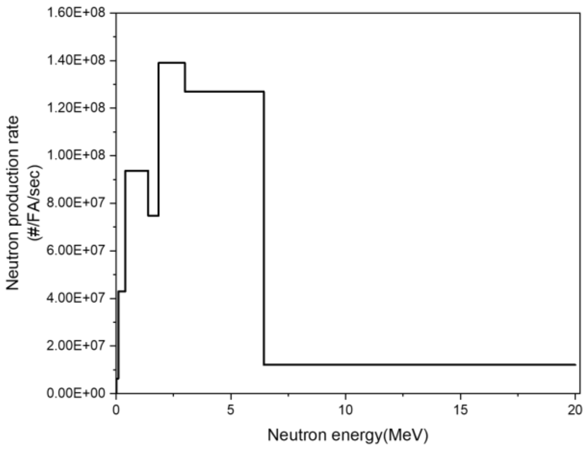

Fig. 1. Calculation result of neutron energy spectrum for spent nuclear fuel in this study.

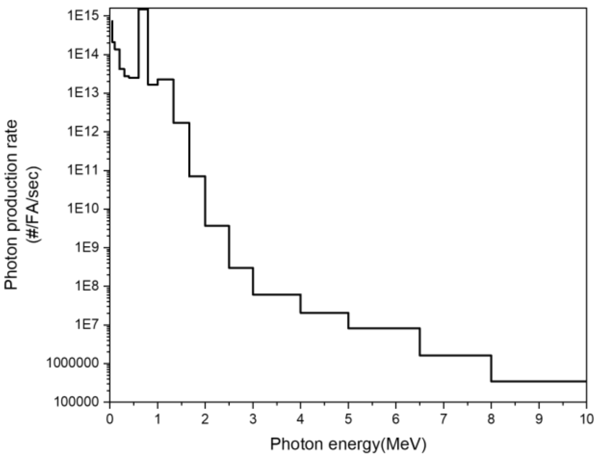

Fig. 2. Calculation result of photon energy spectrum for spent nuclear fuel in this study.

Table III: Calculation results of ^{60}Co residual radioactivity in hardware region

Component	Radioactivity [Bq]
TOP spacer grid	7.44E+11
Bottom spacer grid	3.21E+12
Top end fitting	2.53E+12
Bottom end fitting	5.43E+11

Cladding+Middle support grid	1.16E+12

2.2 Reference metal cask

There were lots of types for dry storage casks such as horizontal/vertical concrete overpacked storage cask and metal storage cask etc. In this study, our reference cask was assumed to be a metal cask which can storage 21 FAs. The established model for the reference cask was shown in Fig. 3.

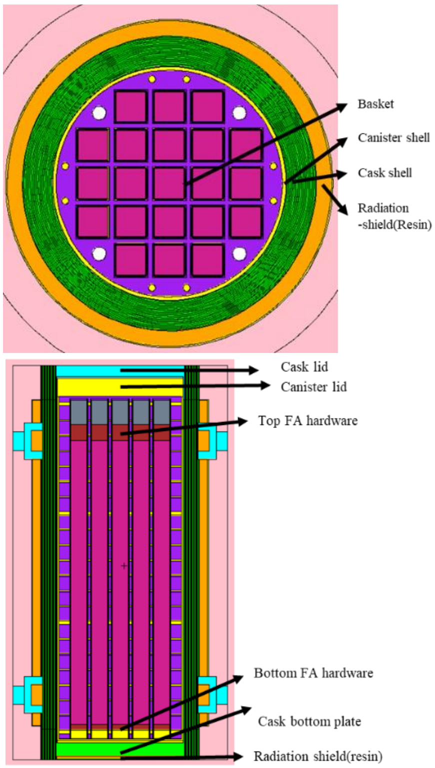

Fig. 3. Result of MCNP modeling for reference metal cask.

2.3 Susceptibility of radiation embrittlement of components

Gap analysis was performed considering extended long term operation (~100 years) of DSS, and results (see step 3 in Fig. 1.) were shown in Table III. For conservatism, in case of calculating accumulated neutron fluence and absorbed dose, the radioactive decay of source term was not considered.

According to the conservative calculation results, it turns out that all of the components in reference DSS will have no aging effect. Polymeric material (e.g. epoxy resin) mostly used for neutron shielding was the most susceptible to radiation embrittlement (see Table III). According to this analysis, the absorbed dose due to gamma rays entering the neutron shield accounted for approximately 67 % (Gamma rays emitted due to decay of ^{60}Co account for 24 % of the total absorbed dose) of the total absorbed dose. It was estimated that in case considering radioactive decay of fission products and ^{60}Co, absorbed dose will be drastically decreased.

Table III: Result of gap assessment in this study ($\Phi_{accumulate,i}$ and $D_{accumulate,i}$ indicates accumulated neutron fluence and absorbed dose)

Component	Material	$\Phi_{accumulate,i}/\Phi_{threshold}$ or $D_{accumulate,i}/D_{threshold}$
Basket	SA-240 Type 304	2.70E-06
BORAL cover	Al alloy	2.94E-08
Canister shell	SA-240 Type 304	9.06E-07
Top FA hardware	Inconel SA-240 Type 304	2.19E-07
Bottom FA hardware	Inconel SA-240 Type 304	5.99E-07
Neutron shield	Resin	4.52E-02
Cladding	Zr-4	3.31E-08
Cask bottom Plate	SA-350 LF.3	9.79E-07
Canister lid	SA-240 Type 304	2.62E-08
Cask lid	SA-182 GR.F6NM	2.82E-09
Cask shell	SA-350 LF.3	3.27E-06

3. Conclusions

In this study, we preliminary identified components which were susceptible to radiation embrittlement using ORIGEN-ARP and MCNP 6.2. All of the components in reference DSS will have no degradations caused by radiation embrittement. Especially, components consisting of steel alloys have large safety margin, and neutron shield consist of organic resin is the most susceptible to radiation embrittlement. When, performing time limited aging analysis (TLAA), components consist of that material should be analyzed carefully, considering thickness of surrounding components such as neutron shield cover sheet, gaps or cracks etc. It is expected that the results of this study would be useful in the aging management review/TLAA process of dry storage systems.

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