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Application of Niobium to Determine Fast Neutron Exposure for Reactor Pressure Vessel

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1. Introduction

Ex-Vessel Neutron Dosimetry (EVND) is widely used in the nuclear industry to monitor the neutron exposure of the Reactor Pressure Vessel (RPV). And all PWR in Korea has EVND system. EVND employs advanced sensor sets that are recommended by, and designed to, the latest ASTM neutron dosimetry standards. [1-7] Especially, ASTM E 844 [4] describes 23 dosimeter elements for fast (E>1.0 MeV) neutron exposure which damages RPV integrity.

Traditionally, vanadium-encapsulated ^{238}U and ^{237}Np fission monitors are used in EVND sensor sets for fast neutron. These fission monitors fulfilled two roles. First, by counting the ^{137}Cs fission product (30.07 year half-life), the fission monitors serve as a long term fluence integrator. Second, ^{237}Np is useful for measuring neutrons with energies from approximately 0.68 to 5.61 MeV, [8] which is important for confirming the fast neutron response spectrum. However, these fission monitors would be constrained and limited on use because ^{238}U is fissionable material and ^{237}Np does not exist in nature.

Niobium comprises one stable isotope (^{93}Nb) in nature and when ^{93}Nb get a neutron response, niobium undergo following reaction, ^{93}Nb (n, n') ^{93m}Nb. The neutron response energy range is 0.95 – 5.79 MeV. [9] And the daughter nuclide, ^{93m}Nb has proper half-life (16.13 year) to monitor fast neutron and long term.

The objective of this paper is to introduce neutron dosimeter measurement including ^{93}Nb and neutron transport calculation to determine fast neutron exposure of the RPV. Also uncertainty analysis and possibility caused by using ^{93}Nb dosimeter will be discussed in a view of the global situation.

2. Methods and Results

This section is composed of simple introduction of EVND, niobium measurement, and best-estimated flux & uncertainty evaluation results with or without niobium.

2.1 Ex-Vessel Neutron Dosimetry (EVND)

EVND system is a surveillance program coupled with In-vessel surveillance capsule to monitor RPV integrity against irradiation embrittlement phenomena. Fig. 1 has shown example of In/Ex-Vessel capsules.

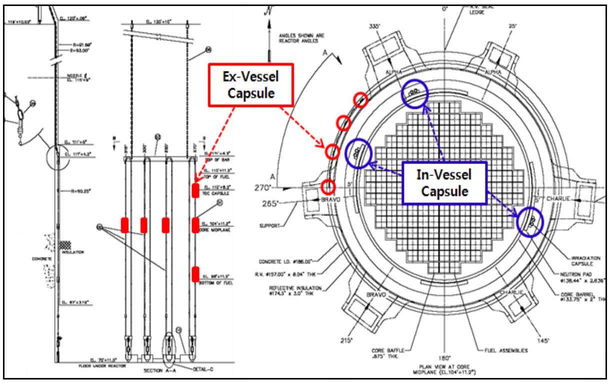

Fig. 1. In/Ex-Vessel capsule location for surveillance program of Westinghouse 3-loop plant

EVND system is installed in the cavity region between the reactor vessel reflective insulation and the primary concrete shield. The sensor sets consist of various neutron dosimeters to monitor fast neutron. Table 1 lists the neutron reactions that are of interest.

Table 1. Neutron Dosimetry Reactions of Interest for EVND

Material	Reaction of Interest$^{(a)}$	Neutron Energy Response	Product Half-Life
Copper	^{63}Cu(n, α)^{60}Co	4.53-11.0 MeV	5.272 y
Titanium	^{46}Ti(n, p)^{46}Sc	3.70-9.43 MeV	83.79 d
Iron	^{54}Fe(n, p)^{54}Mn	2.27-7.54 MeV	312.1 d
Nickel	^{58}Ni(n, p)^{58}Co	1.98-7.51 MeV	70.82 d
^{238}U$^{(b, c)}$	^{238}U(n, f)^{137}Cs	1.44-6.69 MeV	30.07 y
Niobium	^{93}Nb(n, n')^{93m}Nb	0.95-5.79 MeV	16.13 y
^{237}Np$^{(b, c)}$	^{237}Np(n, f)^{137}Cs	0.68-5.61 MeV	30.07 y
Cobalt	^{59}Co(n, γ)^{60}Co	Thermal	5.272 y

Notes:

a) Energies between which 90% of activity is produced (^{235}U fission spectrum) [4]
b) For the fission monitors, ^{95}Zr (64.02 d) and ^{103}Ru (39.26 d) activities are also reported
c) Vanadium-encapsulated ^{238}U and ^{237}Np fission monitors are subject to availability

The EVND program can monitor important azimuthal and axial exposure gradients over the entire beltline region of reactor vessel (unavailable with In-vessel capsule) and minimize the uncertainty in reactor vessel exposure projections using a combination of measurements and analytical predictions.

2.2 Measurement of niobium with Liquid Scintillation Counter (LSC)

When high purity niobium is irradiated in a neutron field, the activation reaction of niobium is ^{93}Nb (n, n') ^{93m}Nb. This activation reaction is useful for monitoring neutrons with energies above approximately 0.5 MeV and for irradiation times up to about 30 years. [9] The metastable state, ^{93m}Nb decays to the ground state by the virtual emission of 30 keV gamma rays that are all internally converted giving rise to the actual emission of orbital electrons followed by X rays.

If liquid scintillation counting of the irradiated niobium is being used to determine the ^{93m}Nb activity, the niobium must be dissolved using nitric acid and hydrochloric acid. It is measured for 30 minutes and the specific activity is obtained by applying the efficiency with the quenching correction curve by the standard solution.

2.3 Results

Evaluations of neutron sensor sets contained in the Ex-vessel dosimetry capsules withdrawn to date from several PWRs in Korea were completed using current state-of-the-art least-squares methodology. Fig. 2 shows fast neutron flux spectrum in log scale per neutron energies with niobium and with fission monitor.

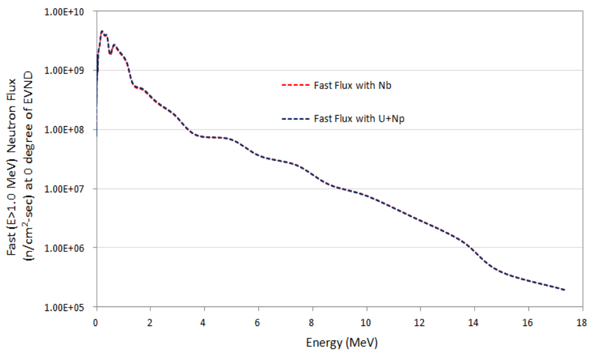

Fig. 2. Fast neutron flux spectrum with Nb and with U+Np.

They are good agreement in the entire fast neutron energy range (E>1.0 MeV). Maximum relative difference is 5% at the low energy region which is from 0.01 MeV to 1.0 MeV. This means determining fast neutron flux with niobium monitor instead of fission monitor is appropriate.

Also, Regulatory Guide 1.190 [10] for calculational and dosimetry methods for determining pressure vessel neutron fluence describes that the neutron transport calculation results typically agree with the measurements to within 30% for cavity dosimetry. Fig. 3 is showing the results of measurement to calculation ratio for niobium which is measured at the different cavity location of azimuthal angle. And the results show that the ratios are within 30% criteria.

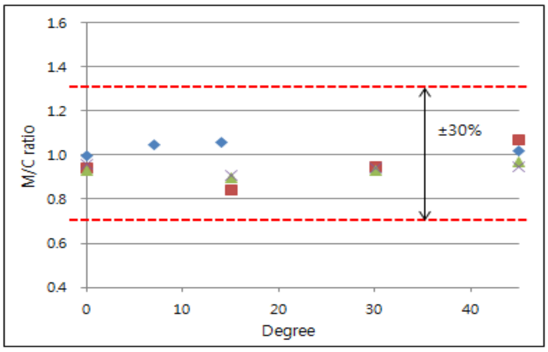

Fig. 3. The ratio of measurement over calculation for niobium.

The final results include bias and uncertainty in order to predict neutron exposure rate at the end of life or at future. Table 2 shows such bias factor and related uncertainty by using fission monitor, niobium, and both for 4 nuclear plants in Korea.

Table 2. The summary of Flux Bias and Uncertainty with fast neutron monitors

		U + Np	U+Np+Nb	Nb
Unit 1	Flux Bias	1.09	1.07	1.06
	Flux Unc.	6 %	5 %	5 %
Unit 2	Flux Bias	1.05	1.01	0.96
	Flux Unc.	6 %	5 %	6 %
Unit 3	Flux Bias	0.94	0.93	0.90
	Flux Unc.	6 %	5 %	5 %
Unit 4	Flux Bias	0.99	0.97	0.94
	Flux Unc.	6 %	5 %	6 %

The lowest uncertainty can be seen surely when all of the fast neutron monitors are used. Also a niobium monitor could cover the two fission monitors from an uncertainty aspect because the uncertainties obtained from niobium are 5-6% while the uncertainties obtained from fission monitors are 6%. These results suggest that niobium is a good fast neutron monitor as an alternative to fission monitors which would be constrained in the future.

3. Conclusions

Ex-vessel neutron dosimetry (EVND) system is employed for all PLWR in Korea to determine fast neutron exposure to reactor pressure vessel (RPV). The fast neutron is defined as energy above 1.0 mega electric volt in irradiation embrittlement evaluation field. In order to provide both the fast neutron energy response and the long term integration, two fission monitors are used traditionally. However these fission monitor has several disadvantages which are availability, correction of fission yield, impurity, and so on. Niobium is in nature and has proper energy response range for fast neutron and half life of daughter nuclide. Through evaluation results with niobium for neutron flux spectrum, measurement to calculation ratio, final bias and uncertainty, niobium could be an alternative to fission monitors.

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