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Neutron Dosimetry for the GE Hitachi 16-10393 Irradiation in ATR

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Summary

PNNL project 74242 involves the analysis of neutron fluence monitors and melt wires irradiated in the Advanced Test Reactor (ATR) at Idaho National Laboratory in accordance with MPO 00236287 and Statement of Work (SOW) No. 17370, Rev. 0, *PNNL Analysis of NSUF Flux and Melt Wire Capsules*. This report is for the GE Hitachi 16-10393 irradiation which was conducted in positions B11 of the ATR. Three other irradiations included in the SOW will be reported separately. The neutron fluence monitors were prepared by PNNL and loaded into the GE Hitachi assemblies prior to irradiation. Following irradiation, the capsules were returned to PNNL for analysis. The neutron dosimetry capsules were opened, the flux wires were removed for gamma analysis. The measured activities were used to determine the activation rates for various activation products. Following suitable corrections, the measured activation rates were used to adjust calculated neutron spectra at each fluence monitor location. The adjusted neutron spectra were then used to determine displacement per atom (dpa) and gas production for irradiated materials.

Irradiation History

The GE Hitachi irradiation occurred in ATR cycles 162A, 162B1, 162B-2, 164A, and 164B starting on October 6, 2017 and ending January 17, 2019 with a total exposure of 208.5 FPD (full power days) at 24 MW for a total of 5004.6 MWD (Megawatt days).

Preparation of Neutron Fluence Monitors

The preparation of the neutron fluence monitors is documented in the report PNNL-70233, *Preparation of Fluence capsules for the GE-Hitachi Drop-In Experiment for Idaho National Laboratory,* MPO#00181356, SOW-13604, Rev. 0 sent to Doug Stacey on March 13, 2017. Small high-purity wires of Fe, Ti, Nb, and 0.116 % Co-Al alloy were encapsulated in vanadium capsules measuring 0.05" OD by about 0.42" long. The vanadium capsules have identification codes stamped on the bottom and each wire and the final sealed capsules were accurately weighed. The vanadium capsules were electron beam welded in a vacuum and helium leak tested. The capsules and weights are listed in Table 1. Weights were measured on a calibrated balance, with daily performance checks.

Table 1. GE Hitachi Neutron Fluence Monitors (Weight in mg)

Capsule ID	Fe	Ti	Nb	0.116% Co-Al	Final Capsule Weight
3H	2.189	2.467	2.482	1.071	49.730
UR	2.402	2.371	2.584	1.205	52.119
2Y	2.389	2.054	2.243	1.083	52.044

The neutron fluence monitors were placed into the GE Hitachi assemblies as documented in drawings provided by INL. The position and elevation of each capsule relative to the midplane of the ATR are listed in Table 2. Unfortunately, when capsule 3H was examined, we found that the capsule was broken and did not contain any of the fluence monitor wires.

Table 2. Location of the Neutron Fluence Monitors in the GEH Assemblies

Capsule ID	Height, in.	Position	KGT#
3H	0	Center	3337
UR	10.46	Top	3340
2Y	-10.46	Bottom	3342

Post-Irradiation Analyses

Following irradiation, the neutron fluence monitors were shipped to PNNL for analysis. Each monitor was cleaned prior to visual examination under a microscope to confirm the capsule identification. The entire capsules were initially gamma counted and then opened in a fume hood to remove the individual wires for final gamma counting. Gamma counting was performed according to procedure RPG-CMC-450 Rev. 3, Gamma Energy Analyses (GEA) and Low-Energy Photon Spectrometry (LEPS). Nuclear decay data were adopted from the NuDat 2.8 database at the National Nuclear Data Center at Brookhaven National Laboratory. Analyses were performed using the Genie2000 software from Mirion. The gamma detectors were calibrated using NIST-traceable standards obtained from Eckert and Zeigler. The performance of the gamma detectors is checked daily on use using control standards to confirm the energy and efficiency calibrations and the energy resolution.

Niobium wires were dissolved in a combination of nitric and hydrofluoric acid. A small aliquot was then deposited on filter paper and the x-rays emitted by Nb-93m were detected using LEPS detectors. The very thin mount eliminates concerns about x-ray absorption, fluorescence, and backscatter effects. The x-ray mounts were verified by gamma counting the Nb-94 activity on each mount to the activity detected in the original wire. Table 3 lists the gamma and x-ray activities measured in the samples. The neutron activation products that we were able to measure are due to three thermal neutron reactions and four fast neutron threshold reactions. The thermal neutron reactions are ^{58}Fe(n,g)^{59}Fe, ^{59}Co(n,g)^{60}Co, and ^{93}Nb(n,g)^{94}Nb and the threshold reactions are ^{54}Fe(n,p)^{54}Mn, ^{46}Ti(n,p)^{46}Sc, and ^{93}Nb(n,n')^{93m}Nb.

Table 3. Measured Activities, Bq/mg
(decay corrected to EOI at Jan. 17, 2019; heights relative to midplane)

Monitor/ Position	**Height, in.**	**^{54}Fe(n,p)^{54}Mn**		**^{46}Ti(n,p)^{46}Sc**		**^{93}Nb(n,n')^{93m}Nb**	
		x10^{5}	±%	x10^{5}	±%	x10^{5}	±%
3H	0	-	-	-	-	-	
UR	10.46	3.24	2	1.36	2	9.27	4
2Y	-10.46	3.62	2	a	2	9.51	4
		^{58}Fe(n,g)^{59}Fe		**^{59}Co(n,g)^{60}Co**		**^{93}Nb(n,g)^{94}Nb**	
		x10^{6}	±%	x10^{6}	±%	x10^{4}	±%
3H	0	-		-	-	-	-
UR	10.46	2.14	16	3.71	2	2.66	2
2Y	-10.46	2.30	3	3.85	2	2.97	2

a The 2Y Ti wire was found to be broken and the ^{46}Sc activity could not be measured.

Table 4. Saturated Activation Rates (atom/atom-sec)
(Uncertainties estimated at ±2%)

Monitor/ Position	**Height, in.**	**^{54}Fe(n,p)^{54}Mn**		**^{46}Ti(n,p)^{46}Sc**		**^{93}Nb(n,n')^{93m}Nb**	
		x10^{-12}	±%	x10^{-13}	±%	x10^{-12}	±%
3H	0	-		-		-	
UR	10.46	1.88	2	3.03	2	6.02	4
2Y	-10.46	2.10	2	-	-	6.18	4
		^{58}Fe(n,g)^{59}Fe		**^{59}Co(n,g)^{60}Co**		**^{93}Nb(n,g)^{94}Nb**	
		x10^{-10}	±%	x10^{-9}	±%	x10^{-10}	±%
3H	0	-		-		-	
UR	10.46	1.48	16	4.77	2	2.23	2
2Y	-10.46	1.60	3	4.96	2	2.50	2

The saturated reaction rates for the neutron activation reactions listed in Table 4 were calculated from the measured activities in Table 3 by correcting for the decay over the irradiation history, atomic weight, isotopic abundance, neutron burnup, and gamma absorption in each wire. The saturated reaction rate is equal to the product of the average neutron flux times the spectral-averaged neutron activation cross section for each reaction. The decay during irradiation correction was determined by calculating the growth and decay of each activation product over the entire irradiation history using the BCF computer code. The irradiation history was provided by staff at Idaho National Laboratory (INL). Gamma self-absorption corrections in the wires averaged around 1% and was calculated from the total photon absorption cross sections given in the NIST XCOM database (https://physics.nist.gov/PhysRefData/Xcom/html/xcom1.html). Neutron burnup refers to the depletion of target or product atoms due to neutron absorption. Corrections were applied in an iterative method using the measured reaction rates as the first approximation and iterating until the process converges. The largest correction was around 1.5%. Neutron self-absorption corrections were estimated to be less than 1% due to the small size of the neutron flux wires and relatively low thermal neutron cross sections. In the case of the Co-Al alloy, the Co fraction is only 0.00116 so neutron absorption is negligible in such a dilute alloy.

Neutron Spectral Adjustment

The STAY'SL PNNL [1] computer code was used to adjust the neutron energy spectrum at each location using the calculated reaction rates and uncertainties as input. The starting neutron spectra were provided by Jill Mitchell (INL) using the Monte Carlo Neutral Particle (MCNP) neutron transport code. STAY'SL PNNL performs a least-squares adjustment to determine the most likely neutron spectrum at each position considering the uncertainties and covariances of all of the input data (activation data, neutron cross sections, and neutron flux spectra). The neutron activation cross sections and covariances were taken from the International Reactor Dosimetry File, IRDF V1.05 [2].

The adjusted neutron fluences from STAY'SL are listed in Table 5. The thermal fluence includes all neutrons < 0.5 eV, the epithermal energy range is from 0.5 eV to 0.11 MeV, and the fast neutron fluences are listed and plotted for thresholds of 0.11 MeV and 1 MeV. Neutron spectral adjustments are shown in Figure 1.

Table 5. Adjusted neutron fluences for the GEH experiment, n/cm^2

Monitor/ Height, in.	**Total**		**Thermal* <0.5 eV**		**Epithermal 0.5 eV to 0.11 MeV**		**Fast >0.11 MeV**		**Fast > 1 MeV**	
	x10^{21}	±%	x10^{20}	±%	x10^{21}	±%	x10^{21}	±%	x10^{20}	±%
3H 0										
UR 10.46	7.39	14	3.21	27	2.90	35	1.28	8	5.00	5
2Y -10.46	7.96	12	3.57	13	3.01	27	1.38	8	5.33	5

*Thermal fluence was calculated as the sum of all neutrons < 0.5 eV

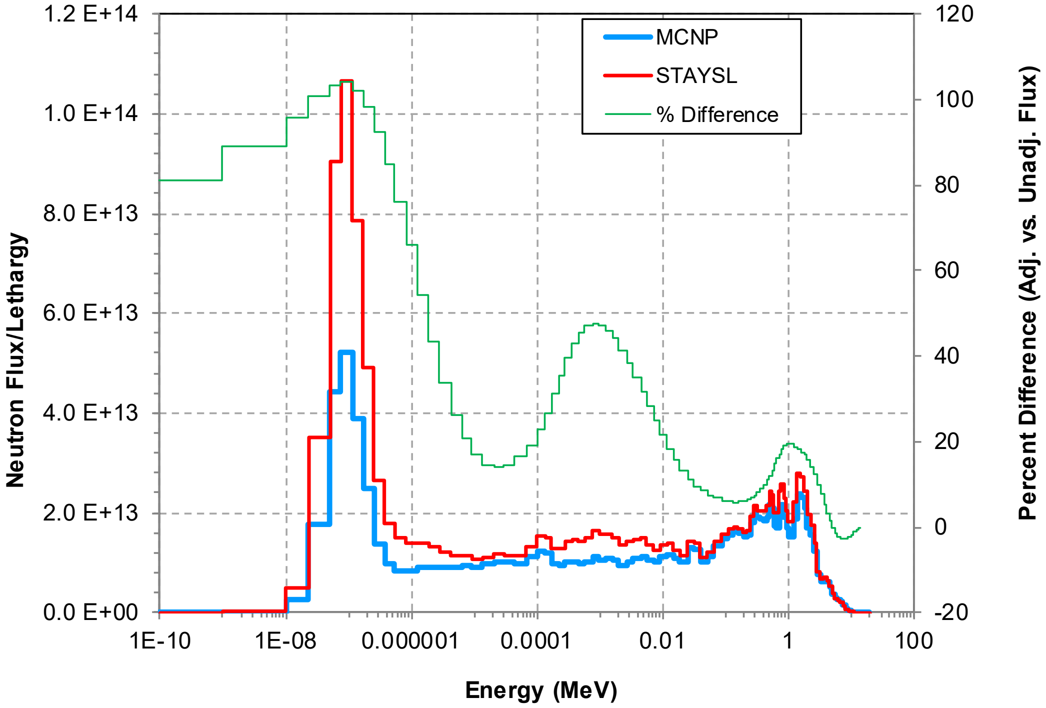

Figure 1 – Adjusted neutron flux spectrum for GEH compared to the MCNP calculation for monitor UR at 10.46 in.

Radiation Damage Calculations

The adjusted neutron spectra were used to calculate radiation damage parameters using the SPECTER computer code [3]. Displacement per atom (dpa) values are listed for several important elements, Inconel 718, and Type 316 stainless steel in Table 6. The contribution to the stainless steel and Inconel dpa values from the ^{59}Ni reaction [4] are included in the calculations.

Table 6. Calculated DPA Values for the GEH Experiment

Monitor/ Height, in.	Fe	Al	Inconel718+	316SS*
3H 0	-	-	-	-
UR 10.46	0.79	1.55	0.94	0.84
2Y -10.46	0.85	1.67	1.03	0.92

*Type 316 stainless steel – Fe (0.67) Cr (0.18) Ni (0.13) Mn (0.02)
+Inconel 718 – Ni(53.5)Fe(17)Cr(20)Nb(5)Mo(3)Ti(1)Al(0.5)

References

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[2] A. Trkov, P.J. Griffin, S.P. Simakov, L.R. Greenwood, et al, IRDFF-II: A New Neutron Metrology Library, Nuclear Data Sheets, 163, pp 1-108, 2020.

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