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Neutron Dosimetry for the University of Central Florida (UCF3) Irradiation in ATR

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Idaho National Laboratory

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Neutron Dosimetry for the University of Central Florida (UCF)-3 Irradiation in ATR

Summary

PNNL project 74242 involves the analysis of neutron fluence monitors irradiated in the Advanced Test Reactor (ATR) at Idaho National Laboratory in accordance with MPO 00236287 and Statement of Work No. 17370, Rev. 0, *PNNL Analysis of NSUF Flux and Melt Wire Capsules*. This report is for the University of Central Florida (UCF)-3 third stage experiment which was conducted in position B8 of the ATR. Three other irradiations included in the scope of work are reported separately. The neutron fluence monitors were prepared by PNNL and loaded into the UCF-3 assemblies at INL 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, and 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 8 fluence monitor locations. The adjusted neutron spectra were then used to determine displacement per atom (dpa) and gas production for irradiated materials.

Irradiation History

The UCF-3 irradiation occurred in ATR cycles 160B-1, 160B-2, 162A, 162B-1, 162B-2, and 164-A starting on December 20, 2016 and ending August 17, 2018 with a total exposure of 206.3 EFPD (effective full power days) for a total of 4126.3 MWD (Megawatt days) at an average power of 20 MW.

Preparation of Neutron Fluence Monitors

The preparation of the neutron fluence monitors is documented in the report PNNL-67962, *Preparation of Fluence capsules for Idaho National Laboratory,* MPO#00158722, SOW-12043, Rev. 0 sent to Clint Baker on August 27, 2015. Small high-purity wires of Fe, Ti, and Nb were encapsulated in vanadium capsules measuring 0.05" OD by about 0.34" long. Separate vanadium capsules measuring 0.050" OD by about 0.034" long and containing only 0.116% Co-Al alloy wires were also prepared. 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, helium leak tested, and subjected to additional tests and inspections as documented in our report. The capsules, wires, and weights are listed in Table 1. Weights were measured on a calibrated balance, with daily performance checks.

The neutron fluence monitors were placed into the UCF-3 assemblies as documented in drawings provided by INL [1]. The position and elevation of each capsule relative to the midplane of the ATR are listed in Table 2.

Table 1. UCF-3 Neutron Fluence Monitors with ID Codes and weights

Capsule ID	Fe (mg)	Ti (mg)	Nb (mg)	Capsule (mg)
9R	2.695	2.123	2.804	44.965
5C	2.832	1.791	2.564	48.120
5V	2.730	1.912	2.778	47.343
8H	3.063	1.948	2.744	48.058
9A	2.338	1.895	2.531	45.709
L1	2.421	1.632	2.253	45.540
71	2.324	1.715	2.556	47.337
7V	2.028	1.515	2.310	44.462
Capsule ID	**Co-Al (mg)**	**Capsule (mg)**		
8Z	1.954	42.617		
9X	2.460	42.716		
2F	1.935	41.333		
1D	2.163	39.961		
21	1.989	41.107		
7Z	2.141	39.083		
8V	2.221	42.565		
8L	2.184	41.425		

Table 2. Location of the Neutron Fluence Monitors in the UCF-3 Assemblies

Fe-Ti-Nb Capsule ID	0.116% Co-Al Capsule ID	UCF-3 Positions	Elevation, in
9A	21	UCF-34	-16.94
71	8V	UCF-35	-14.69
9R	8Z	UCF-15	-10.18
5V	2F	UCF-17	-5.67
8H	1D	UCF-19	1.10
5C	9X	UCF-21	5.61
7V	8L	UCF-38	10.12
L1	7Z	UCF-39	12.37

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.

Table 3. Measured Activities, Bq/mg
(decay corrected to EOI at Aug. 17, 2018)

Height, in	$^{54}Fe(n,p)^{54}Mn$		$^{46}Ti(n,p)^{46}Sc$		$^{93}Nb(n,n')^{93m}Nb$	
	x10^6	±%	x10^5	±%	x10^6	±%
-16.94	0.955	2	3.60	2	2.23	4
-14.69	1.06	2	3.94	2	2.47	4
-10.18	1.17	2	4.61	2	2.83	4
-5.67	1.22	2	5.14	2	2.84	4
1.10	1.29	2	5.06	2	3.09	4
5.61	1.24	2	4.91	2	2.94	4
10.12	1.11	2	4.24	2	2.52	4
12.37	1.05	2	4.02	2	2.50	4
Height, in.	**$^{58}Fe(n,g)^{59}Fe$**		**$^{59}Co(n,g)^{60}Co$**		**$^{93}Nb(n,g)^{94}Nb$**	
	x10^6	±%	x10^6	±%	x10^4	±%
-16.94	2.79	13	4.84	2	3.32	2
-14.69	3.62	5	5.30	2	3.79	2
-10.18	4.03	19	6.02	2	4.32	2
-5.67	3.89	4	5.36	2	4.06	2
1.10	3.68	5	5.24	2	4.11	2
5.61	4.05	4	6.10	2	4.19	2
10.12	2.82	22	4.56	2	3.61	2
12.37	3.41	11	5.02	2	3.64	2

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 4. Saturated Activation Rates (atom/atom-sec)

Height, in	**$^{54}Fe(n,p)^{54}Mn$**		**$^{46}Ti(n,p)^{46}Sc$**		**$^{93}Nb(n,n')^{93m}Nb$**	
	$x10^{-12}$	±%	$x10^{-13}$	±%	$x10^{-11}$	±%
-16.94	5.85	2	7.79	2	1.47	4
-14.69	6.51	2	8.54	2	1.63	4
-10.18	7.19	2	9.99	2	1.87	4
-5.67	7.49	2	11.1	2	1.88	4
1.10	7.92	2	11.0	2	2.04	4
5.61	7.63	2	10.6	2	1.94	4
10.12	6.79	2	9.17	2	1.66	4
12.37	6.44	2	8.71	2	1.65	4
Height, in.	**$^{58}Fe(n,g)^{59}Fe$**		**$^{59}Co(n,g)^{60}Co$**		**$^{93}Nb(n,g)^{94}Nb$**	
	$x10^{-10}$	±%	$x10^{-9}$	±%	$x10^{-10}$	±%
-16.94	1.69	13	6.47	2	2.83	2
-14.69	2.19	5	7.12	2	3.25	2
-10.18	2.45	19	8.17	2	3.73	2
-5.67	2.36	4	7.21	2	3.50	2
1.10	2.23	5	7.04	2	3.54	2
5.61	2.46	4	8.29	2	3.61	2
10.12	1.71	22	6.07	2	3.09	2
12.37	2.07	11	6.72	2	3.12	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 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 [2] 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 spectrum was calculated by J. Parry (INL) using the Monte Carlo Neutral Particle (MCNP) neutron transport code for prior irradiations for UCF [3]. 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 [4].

The adjusted neutron fluences from STAY'SL are listed in Table 5 and are shown in Figures 1 and 2. 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. A typical neutron spectral adjustment is shown in Figure 3. The thermal and epithermal flues are lower than calculated although the fast neutron fluences above 0.1 MeV agree quite well with the MCNP calculations. It would of course be preferable to perform an MCNP calculation that takes the experimental assembly into account. However, the STAY'SL adjustment resulted in good agreement for the three neutron activation reactions that are sensitive to neutron fluxes in the thermal and epithermal energy region such that the integral neutron fluence values should accurately represent the neutron spectra at the locations of the flux monitors.

Table 5. Adjusted Neutron Fluences for the UCF-3 Experiment (x10^{21})

	Thermal*		Epithermal		Fast		Fast	
	< 0.5 eV		0.5 eV to 0.11 MeV		> 0.1 MeV		> 1 MeV	
Height, in.	**n/cm²**	**±%**	**n/cm²**	**±%**	**n/cm²**	**±%**	**n/cm²**	**±%**
-16.94	3.09	5	3.98	12	3.04	7	1.34	4
-14.69	3.59	5	4.32	11	3.38	6	1.49	4
-10.18	4.12	5	4.91	11	3.84	6	1.68	4
-5.67	3.64	5	4.88	11	3.98	7	1.70	4
1.10	3.42	5	5.22	10	4.23	6	1.84	4
5.61	4.12	5	4.97	11	4.02	6	1.76	4
10.12	2.90	5	4.58	11	3.50	6	1.53	4
12.37	3.35	5	4.28	11	3.39	6	1.47	4

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

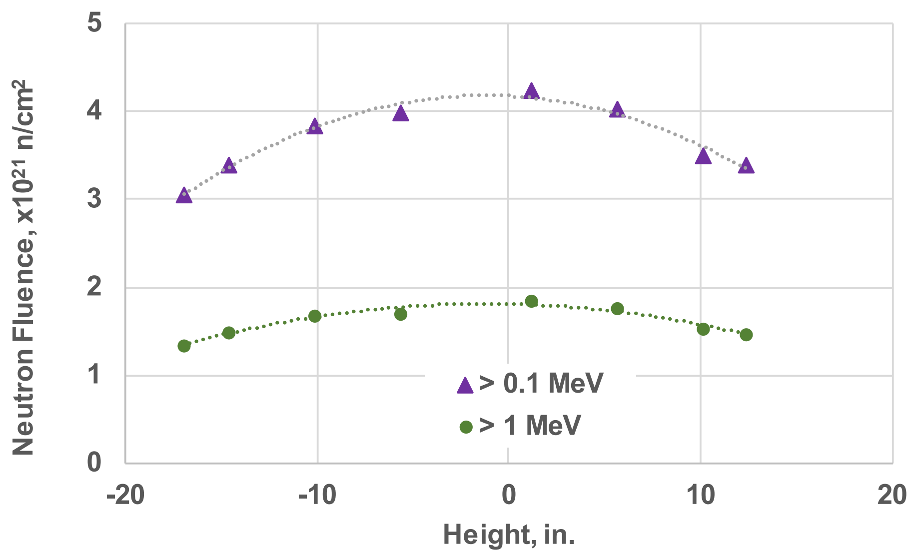

Figure 1. Fast neutron fluence values

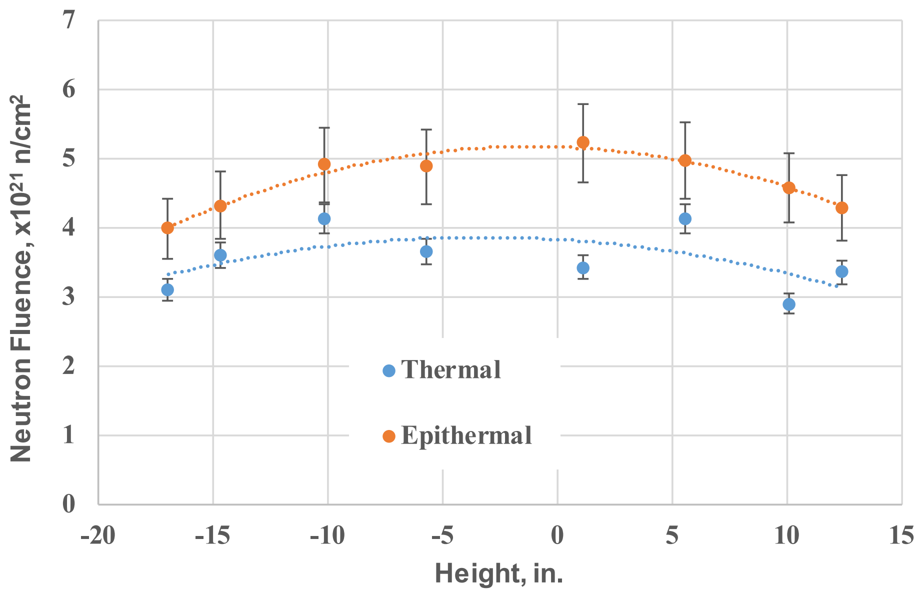

Figure 2 – Thermal and epithermal neutron fluences

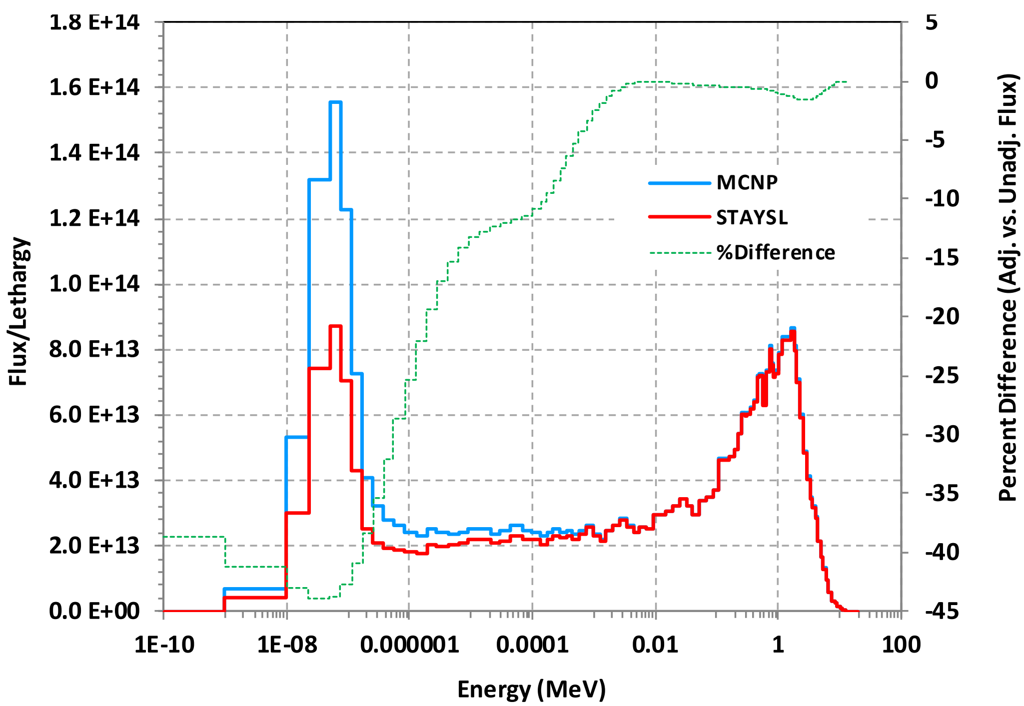

Figure 3. Adjusted neutron flux spectrum for UCF-3 at 1.1 cm compared to the MCNP calculation.

Radiation Damage Calculations

The adjusted neutron spectra were used to calculate radiation damage parameters using the SPECTER computer code [5]. Displacement per atom (dpa) for several important elements are listed in Table 6 and plotted in Figure 4.

Table 6. Calculated DPA Values for the UCF-3 Experiment

Height, in.	Al	Cr	Fe	Zr	Mo
-16.94	3.75	2.19	1.94	2.13	1.47
-14.69	4.17	2.44	2.16	2.36	1.64
-10.18	4.72	2.76	2.44	2.68	1.86
-5.67	4.87	2.83	2.51	2.76	1.92
1.10	5.19	3.01	2.67	2.95	2.05
5.61	4.94	2.88	2.55	2.80	1.94
10.12	4.30	2.51	2.22	2.44	1.70
12.37	4.16	2.42	2.15	2.36	1.64

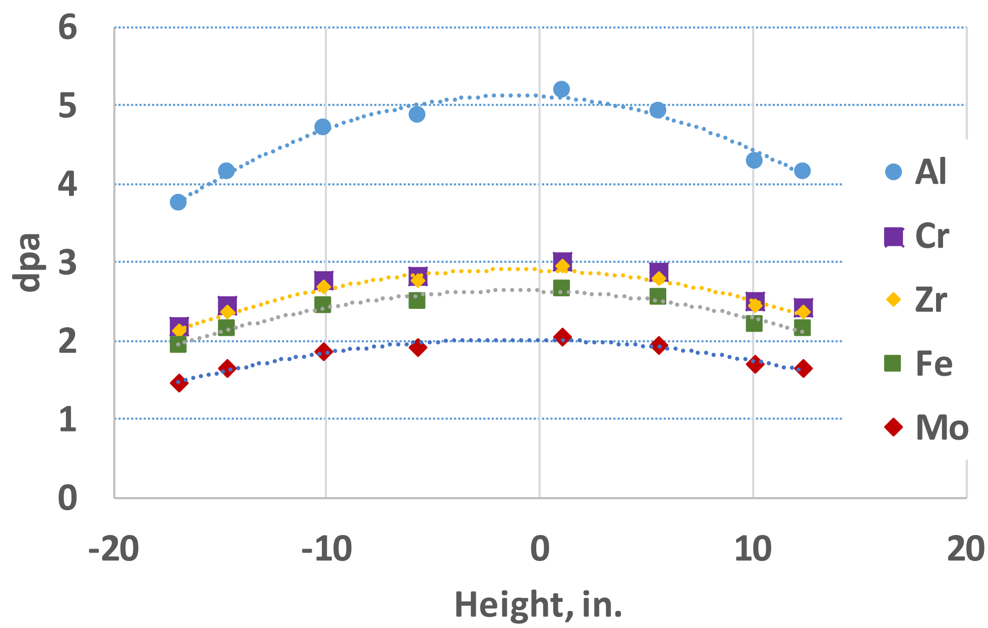

Figure 4 – dpa values vs. height in the UCF-3 assembly

The measured activities listed in Table 3 and saturated reaction rates listed in Table 4 show a decrease in the three thermal neutron reactions $^{58}Fe(n,g)^{59}Fe$, $^{59}Co(n,g)^{60}Co$, and $^{93}Nb(n,g)^{94}Nb$ near midplane, as shown in Figure 2. All three of the thermal neutron reactions show the same decrease near midplane. No such decrease is observed in the fast neutron reactions $^{54}Fe(n,p)54Mn$, $^{46}Ti(n,p)^{46}Sc$, and $^{93}Nb(n,n')^{93m}Nb$, all of which show that the highest reaction rates occur near midplane, as would be expected, as shown in Figure 1. The reason for the decrease of the thermal neutron flux near midplane is not known but is likely due to a thermal flux depression caused by the materials in the UCF3 assembly. This decrease in the thermal neutron flux does not have any significant impact on the dpa values shown in Table 6 since the radiation damage is predominantly caused by fast neutrons.

References

[1] *University of Central Florida Static Capsule Third Stage Experiment (UCF-3) Data Package*, DP-112, Rev. 1, 11/10/2016, INL Project Number 31226 with provided drawings.

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