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Neutron Dosimetry for the SAM-2 Irradiation in ATR

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Lawrence R Greenwood
Digitally signed by Lawrence R Greenwood
Date: 2023.02.20 16:14:44 -08'00'

Larry R. Greenwood, Author

Dana Byram
Digitally signed by Dana Byram
Date: 2023.02.21 08:48:13 -08'00'

Technical Reviewer

Veronika Vazquez
Digitally signed by Veronika Vazquez
Date: 2023.04.11 15:08:41 -07'00'

Quality Engineer

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NATIONAL LABORATORY

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L. R. Greenwood (Pacific Northwest National Laboratory)

Summary

PNNL project 79550 provides for the analysis of neutron fluence monitors irradiated in the Advanced Test Reactor (ATR) at Idaho National Laboratory in accordance with MPO 00269673 and Statement of Work No. 19704, Rev. 0, *PNNL Analysis of NSUF Flux Capsules*. This report is for the SAM-2 irradiation which was conducted in position B8 of the ATR. Other experiments included in the statement of work for this project will be reported separately. The neutron fluence monitors were prepared by PNNL and sent to INL for loading into the SAM-2 assembly prior to irradiation. The SAM-2 experiment has 8 capsules labelled A through H designed for different exposures in successive irradiation cycles. This report is for the first three capsules, A, B and C, co-irradiated for 1 cycle. The remaining capsules and fluence monitors will be removed and analyzed after additional irradiation.

Following irradiation, the fluence monitors from capsules A, B, and C were returned to PNNL for analysis. The neutron dosimetry capsules were opened, the flux wires were removed for gamma or x-ray 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 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 SAM-2 experiment was irradiated in position B8 of the ATR for cycle 169A from February 19, 2021, to April 23, 2021 for a total irradiation of 63 EFPD (effective full power days) at an average power of 20.7 MW for a total exposure of 1304.1 MWD. Following irradiation, the fluence monitors were removed from the SAM-2 assembly and returned to PNNL for analysis.

Preparation of Neutron Fluence Monitors

The preparation of the neutron fluence monitors is documented in the report PNNL-77150, *Fluence capsules for the SAM-2 Experiment,* MPO#00239221, SOW-17494, Rev. 0 sent to Douglas Stacey on September 22, 2020 [1]. 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 0.39" 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. Weights were measured on a calibrated balance, with daily performance checks. A total of 8 fluence monitors were delivered and this report is for the analysis of 3 capsules as listed in Table 1.

Table 1. SAM-2 Neutron Fluence Monitors (Weight in mg)

Capsule ID	Fe	Ti	Nb	0.116% Co-Al	Final Capsule Weight
1F	2.516	1.195	2.165	0.695	49.619
97	2.613	1.264	1.863	0.830	48.352
9J	2.610	1.191	2.281	0.863	48.197

The neutron fluence monitors were placed into the SAM-2 assembly 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. A fourth capsule A5 will be sent to PNNL for analysis later.

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

Capsule ID	KGT#	SAM-2	Elevation, in
1F (A)	4486	B8-A NNW	+39
97 (B)	4505	B8-B NNW	+31
9J (C)	4524	B8-C NNW	+23

Post-Irradiation Analyses

Following irradiation, the neutron fluence monitors were shipped to PNNL for analysis. Each monitor was cleaned prior to weighing the capsule to confirm the 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 each gamma detector is checked daily on use with control standards to confirm the energy, efficiency, resolution of three gamma peaks from Am-241, Cs-137, and Co-60.

The niobium wires were then dissolved in a combination of nitric and hydrofluoric acid. A small aliquot was deposited on filter paper and the x-rays emitted by Nb-93m were detected using low energy Ge (LEGe) detectors. The very thin mount of the activities on filter paper 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 and comparing the activities to the activities detected in the original wires. 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/g
(Decay corrected to EOI April 23, 2021; heights relative to midplane)

Monitor/ Position	**Height, in.**	**$^{54}Fe(n,p)^{54}Mn$**		**$^{46}Ti(n,p)^{46}Sc$**		**$^{93}Nb(n,n')^{93m}Nb$**	
		bq/g	±%	bq/g	±%	bq/g	±%
1F	+39	5.21e+5	2	5.37e+5	2	5.11e+5	5
97	+31	8.31e+6	2	6.53e+6	2	1.14e+7	5
9J	+23	1.58e+8	2	1.20e+8	2	2.56e+8	5
		$^{93}Nb(n,g)^{94}Nb$		**$^{59}Co(n,g)^{60}Co$**		**$^{58}Fe(n,g)^{59}Fe$**	
		bq/g	±%	bq/g	±%	bq/g	±%
1F	+39	1.46e+4	2	3.30e+6	2	6.16e+6	9
97	+31	1.79e+5	2	3.21e+7	2	7.77e+7	6
9J	+23	3.44e+6	2	5.04e+8	2	1.09e+9	3

Table 4. Saturated Activation Rates (atom/atom-sec)
(Uncertainties estimated at ±2%; heights relative to midplane)

Monitor/ Position	**Height, in.**	**$^{54}Fe(n,p)^{54}Mn$**		**$^{46}Ti(n,p)^{46}Sc$**		**$^{93}Nb(n,n')^{93m}Nb$**	
		at/at-s	±%	at/at-s	±%	at/at-s	±%
1F	+39	6.47e-15	2	1.29e-15	2	1.07e-14	5
97	+31	1.03e-13	2	1.57e-14	2	2.39e-13	5
9J	+23	1.97e-12	2	2.89e-13	2	5.35e-12	5
		$^{93}Nb(n,g)^{94}Nb$		**$^{59}Co(n,g)^{60}Co$**		**$^{58}Fe(n,g)^{59}Fe$**	
		at/at-s	±%	at/at-s	±%	at/at-s	±%
1F	+39	3.93e-13	2	1.24e-11	2	3.30e-13	9
97	+31	4.82e-12	2	1.21e-10	2	4.16e-12	6
9J	+23	9.29e-11	2	1.91e-09	2	5.84e-11	3

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 2%. 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 spectra were provided by Jill Mitchell (INL) using the Monte Carlo Neutral Particle (MCNP) neutron transport code [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 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 II [4]. The adjusted neutron fluences from STAY'SL are listed in Table 5. The neutron spectral adjustments are shown in Figures 1, 2, and 3.

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

Monitor/Height, in.	1F +39" A		97 +31" B		9J +23" C	
		±%		±%		±%
Total	**3.41E+18**	**5**	**4.18E+19**	**5**	**1.01E+21**	**5**
Thermal <0.5 eV	**1.99E+18**	**5**	**1.91E+19**	**4**	**2.86E+20**	**5**
Epi 0.5 eV to 0.11 MeV	**8.21E+17**	**20**	**1.20E+19**	**16**	**4.04E+20**	**10**
Epi 0.5 eV to 0.18 MeV	**8.56E+17**	**20**	**1.27E+19**	**16**	**4.34E+20**	**10**
Fast > 0.11 MeV	**5.98E+17**	**6**	**1.07E+19**	**6**	**3.15E+20**	**6**
Fast > 0.18 MeV	**5.63E+17**	**6**	**1.00E+19**	**6**	**2.86E+20**	**6**
Fast > 1.0 MeV	**3.32E+17**	**4**	**6.00E+18**	**5**	**1.39E+20**	**5**

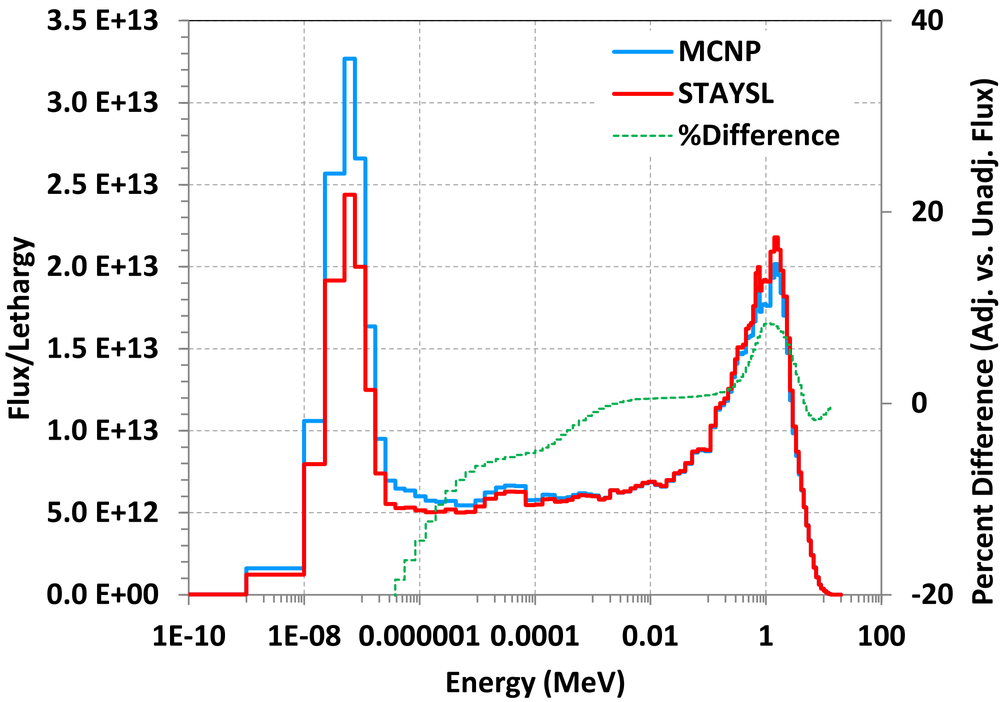

Figure 1 – Adjusted neutron flux spectrum compared to the MCNP calculation for SAM2 monitor 9J at +23" in position B8

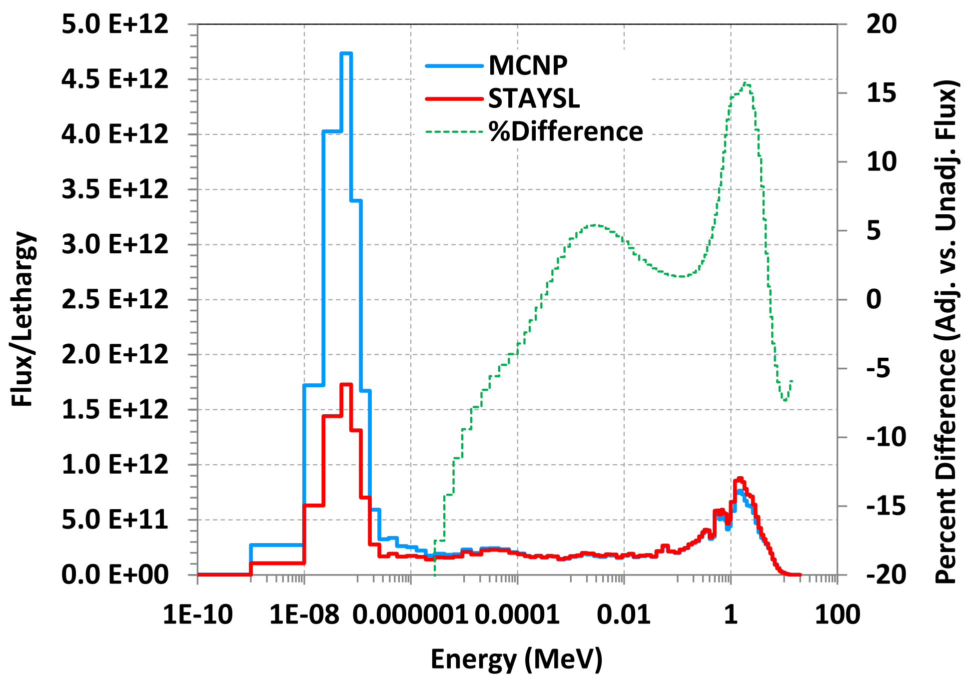

Figure 2. Adjusted neutron flux spectrum compared to the MCNP calculation for monitor 97 at +31" in B8.

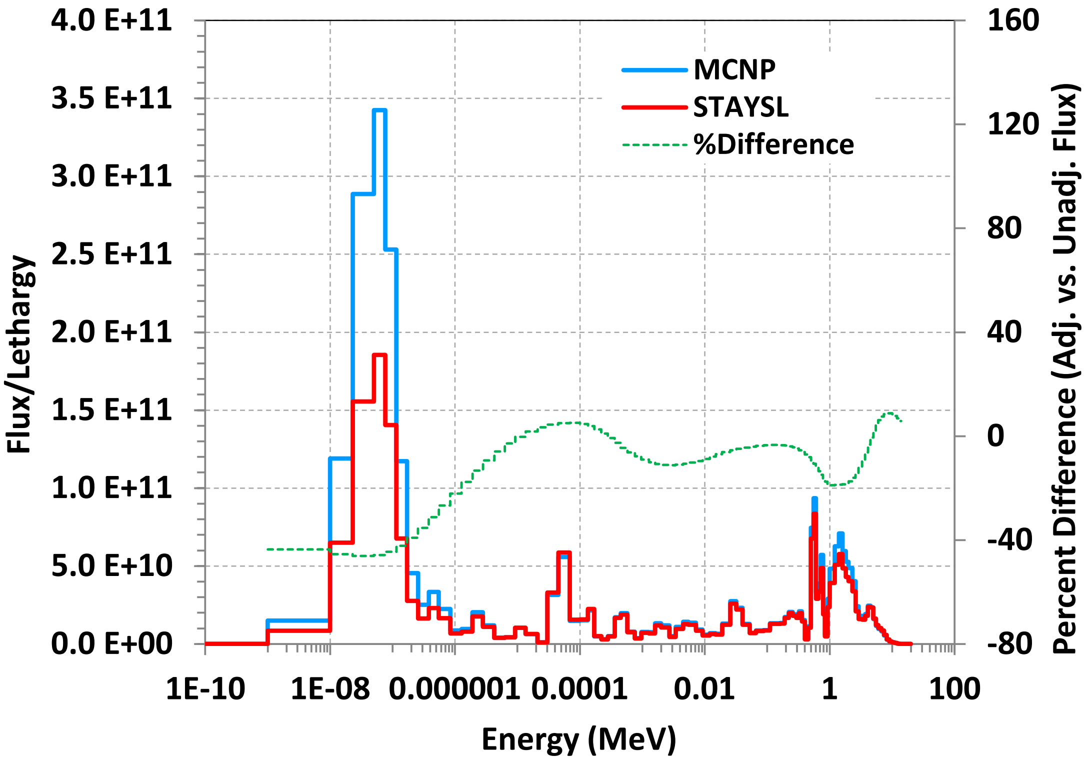

Figure 3. Adjusted neutron flux spectrum compared to the MCNP calculation for monitor 1F at +39" in position B8.

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 and Type 316 stainless steel are listed in Table 6. The small contribution to the stainless steel dpa values from the Ni-59 reaction [6] were included for the SAM2 irradiation.

Table 6. Calculated DPA Values for the Sam2 Experiment

Monitor/ Height, in.	SiC[a]	Fe	Al	Zr	316SS*
1F +39 A	6.83e-4	4.60e-4	8.03e-4	4.70e-4	4.78e-4
97 +31 B	1.22e-2	7.83e-3	1.43e-2	8.36e-3	8.34e-3
9J +23 C	3.45e-1	1.97e-1	3.91e-1	2.22e-1	2.10e-1

[a]SiC ref. 7

*Type 316 stainless steel – Fe (0.67) Cr (0.18) Ni (0.13) Mn (0.02).
The contribution from the ^{59}Ni reaction (6) was negligible.

References

[1] PNNL-77150, *Fluence capsules for the SAM-2 Experiment,* MPO#00239221, SOW-17494, Rev. 0 sent to Douglas Stacey on September 22, 2020

[2] L. R. Greenwood and C. D. Johnson, *Least-Squares Neuron Spectral Adjustment with STAYSL PNNL*, International Symposium on Reactor Dosimetry 15, EPJ Web of Conferences Vol 106, 586-594, Aix-en-Provence, France, May 2015, ISBN:978-1-5108-1940-5.

[3] Jill Mitchell, ECAR-5567, ATR As-Run Physics Evaluation for the SAM-2 Experiment, November 20, 2019.

[4] 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.

[5] L. R. Greenwood and R. K. Smither, *SPECTER: Neutron Damage Calculations for Materials Irradiations*, ANL/FPP-TM-197, January 1985.

[6] L. R. Greenwood, *A New Calculation of Thermal Neutron Damage and Helium Production in Nickel*, Journal of Nuclear Materials 115 (1983) 137-142.