INL/RPT-24-79215-Revision-0

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Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

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Lawrence R Greenwood Digitally signed by Lawrence R Greenwood
Date: 2021.02.17 16:29:06 -08'00'

Larry R. Greenwood, Author

Truc Trang-Le Digitally signed by Truc Trang-Le
Date: 2021.02.17 16:33:13 -08'00'

Technical Reviewer

Samuel A. Adams Digitally signed by Samuel A. Adams
Date: 2021.02.17 16:47:43 -08'00'

Quality Engineer

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Neutron Dosimetry for the Colorado School of Mines (CSM 16-10584) 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 Colorado School of Mines (CSM 16-10584) experiment which was conducted in position B5 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 CSM 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 12 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 CSM assembly consisted of 5 sections referred to as A, B, C, D, and E. All CSM sections were irradiated in ATR cycle 164A from June 12, 2018 to August 17, 2018, for 54 days EFPD (effective full power days) at 22.6 MW for a total of 1220.7 MWD (Megawatt days). Capsules A and E, located at the highest and lowest elevations in the assembly, were then removed and capsules B, C, and D were further irradiated in ATR cycle 164B from September 19, 2018 to January 17, 2019 for a total exposure of 117.6 EFPD at 22.6 MW for a total exposure of 2658.6 MWD.

Preparation of Neutron Fluence and Melt Wire Monitors

The preparation of the neutron fluence monitors is documented in the report PNNL-70745, *CSM-10584 Monitor Procurement for Idaho National Laboratory,* MPO#00184726, SOW-13744, Rev. 2 sent to Craig Tyler on August 31, 2017. Small high-purity wires of Fe and Ti, were encapsulated in vanadium capsules measuring 0.05” OD by about 0.39” long. The neutron fluence monitors are listed in Table 1 including the weights of the wires and final sealed capsules. Weights were measured on a calibrated balance, with daily performance checks. Melt wires were encapsulated in separate vanadium capsules containing Bi, Pb, and 95Zn-5Al alloy wires, as listed in Table 2. All 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 neutron fluence monitors and melt wire monitors were placed into the CSM 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 3.

Table 1. CSM Neutron Fluence Monitors with ID Codes and weights

Capsule ID (Stamped on bottom)	Fe Weight (mg)	Ti Weight (mg)	Final Capsule Weight (mg)
E5	3.611	2.493	51.281
1Z	3.954	2.757	51.626
1U	3.999	2.587	49.090
I2	3.886	2.259	46.789
8H	3.763	2.620	47.582
B1	4.144	2.714	51.808
1Y	4.017	2.376	51.086
E2	3.967	2.584	51.260
7Y	4.175	2.365	50.980
5L	3.538	2.335	50.571
9Y	4.234	2.317	50.018
87	4.560	2.242	51.969

Table 2. Summary of Melt Wire Monitors with ID, Melting Point, and Average Length

Bi 271.58 ^{0}C 0.403”		Pb 328.51 ^{0}C 0.323”		95Zn-5Al 383.73 ^{0}C 0.450”
9V		BA		5I
B9		7B		9E
Y5		D2		97
J1		IR		8Z
87		A9		5U
2Z		80		8B
50		J2		1H
01		EI		B2
7D		IX		I7
1J		7I		9B
5Z		2V		8A
Y8		98		7B

Table 3. Location of the Neutron Fluence and Melt Wire Monitors in the CSM Assembly

Height, in	Capsule CSM-	Fluence Monitor	Bi	Pb	Zn
-24.3	E1	1Z	B9	7B	9E
-22.9	E2	E5	9V	BA	5I
-21.4	D1	I2	J1	IR	8Z
-19.9	D2	1U	Y5	D2	97
-2.0	C3	1Y	50	J2	1H
-0.6	C4	8H	87	A9	5U
0.7	C1	B1	2Z	80	8B
2.0	C2	E2	01	E1	B2
19.8	B1	5L	1J	71	9B
21.3	B2	7Y	7D	1X	17
22.8	A1	87	Y8	98	7B
24.2	A2	9Y	5Z	2V	8A

Post-Irradiation Analyses of Neutron Fluence Monitors

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 4 lists the gamma activities measured in the samples. The neutron activation products that we were able to measure are due to one thermal neutron reaction and two fast neutron threshold reactions. The thermal neutron reaction is ^{58}Fe(n,g)^{59}Fe and the threshold reactions are ^{54}Fe(n,p)^{54}Mn and ^{46}Ti(n,p)^{46}Sc. The fluence monitor 8H located in CSM-C4 was stuck in a metal block. Upon our receipt examination under a microscope, the capsule was found to be broken and none of the neutron fluence wires could be located.

Table 4. Measured Activities, Bq/mg
(AE monitors 1Z, E5, 87 and 9Ydecay in shaded rows corrected to Aug. 17, 2018)
(Remaining BCD monitors decay corrected to Jan. 17, 2019)

Height, in	Monitor	^{54}Fe(n,p)^{54}Mn		^{46}Ti(n,p)^{46}Sc		^{58}Fe(n,g)^{59}Fe	
		x10^{5}	±%	x10^{5}	±%	x10^{6}	±%
-24.3	1Z	1.27	2%	0.864	2%	1.51	5%
-22.9	E5	2.04	2%	1.42	2%	1.54	7%
-21.4	I2	4.47	2%	2.12	2%	2.18	4%
-19.9	1U	5.35	2%	2.61	3%	2.78	6%
-2.0	1Y	11.8	2%	5.64	2%	5.66	7%
0.7	B1	11.7	2%	5.77	2%	6.00	6%
2.0	E2	12.0	2%	5.65	2%	5.76	4%
19.8	5L	4.59	2%	2.35	2%	1.74	4%
21.3	7Y	3.51	2%	1.75	2%	1.28	4%
22.8	87	1.34	2%	0.995	5%	0.985	11%
24.2	9Y	0.996	2%	0.735	4%	0.759	20%

Table 5. Saturated Activation Rates (atom/atom-sec)

Height, in	Monitor	^{54}Fe(n,p)^{54}Mn		^{46}Ti(n,p)^{46}Sc		^{58}Fe(n,g)^{59}Fe	
		x10^{-12}	±%	x10^{-12}	±%	x10^{-10}	±%
-24.3	1Z	1.84	2%	0.242	2%	0.938	5%
-22.9	E5	2.96	2%	0.397	2%	0.956	7%
-21.4	I2	3.59	2%	0.483	2%	1.44	4%
-19.9	1U	4.31	2%	0.595	3%	1.84	6%
-2.0	1Y	9.58	2%	1.29	2%	3.76	7%
0.7	B1	9.50	2%	1.32	2%	3.99	6%
2.0	E2	9.74	2%	1.29	2%	3.83	4%
19.8	5L	3.69	2%	0.535	2%	1.15	4%
21.3	7Y	2.82	2%	0.398	2%	0.842	4%
22.8	87	1.94	2%	0.278	5%	0.611	11%
24.2	9Y	1.43	2%	0.199	4%	0.447	20%

The saturated reaction rates for the neutron activation reactions listed in Table 5 were calculated from the measured activities in Table 4 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 were 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.

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 calculated 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 [3].

The adjusted neutron fluences from STAY'SL are listed in Table 6 and are shown in Figure 1. 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. The neutron spectral adjustment for CSM monitor E2 at 2.0" above midplane is shown in Figure 2. The STAY'SL spectral adjustment resulted in good agreement for all of the neutron activation reactions such that the integral neutron fluence values should accurately represent the neutron spectra at the locations of the flux monitor packages.

Table 6. Adjusted Neutron Fluences for the CSM Experiment (x10^{21} n/cm^2)

AE monitors (shaded rows) irradiated for 1 cycle; BCD monitors irradiated for 2 cycles

Monitor	Total		Thermal* < 0.5 eV		Epithermal 0.5 eV to 0.11 MeV		Fast > 0.1 MeV		Fast > 1 MeV	
Ht, in.	n/cm^2	±%	n/cm^2	±%	n/cm^2	±%	n/cm^2	±%	n/cm^2	±%
-24.3 1Z	1.01	16	0.550	16	0.271	43	0.190	7	0.091	7
-22.9 E5	1.26	18	0.550	15	0.401	48	0.307	7	0.149	6
-21.4 I2	4.05	12	1.81	15	1.33	29	0.910	7	0.408	7
-19.9 1U	5.02	16	2.33	17	1.60	40	1.09	7	0.488	7
-2.0 1Y	10.42	15	4.81	19	3.31	35	2.30	7	1.04	7
0.7 B1	10.83	16	5.14	18	3.38	42	2.30	7	1.03	7
2.0 E2	10.80	19	4.93	19	3.46	51	2.41	7	1.08	7
19.8 5L	3.64	19	1.46	19	1.28	50	0.895	7	0.401	7
21.3 7Y	2.70	16	1.07	16	0.952	40	0.672	7	0.304	7
22.8 87	0.863	18	0.361	16	0.290	50	0.213	7	0.097	6
24.2 9Y	0.632	18	0.264	16	0.212	50	0.157	7	0.072	6

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

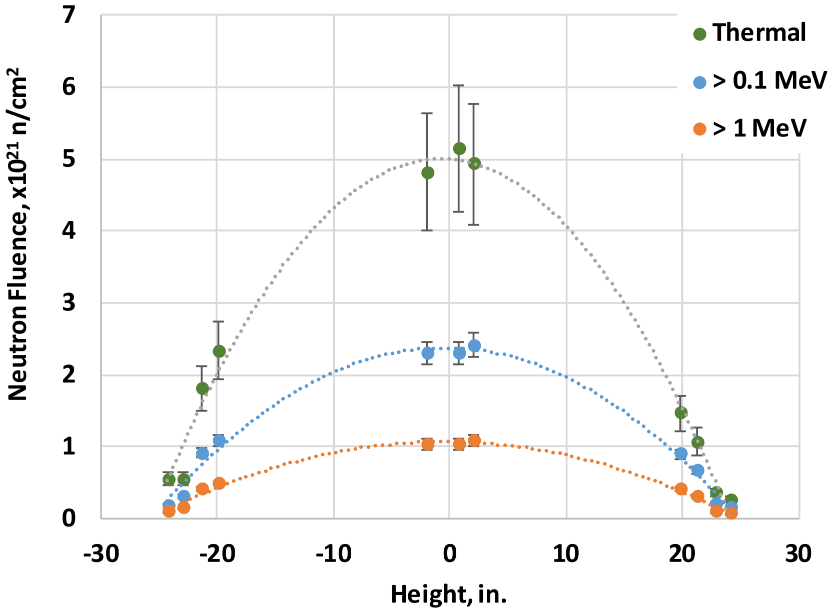

Figure 1. Neutron fluence values. The two top and bottom monitors were irradiated for only 1 cycle.

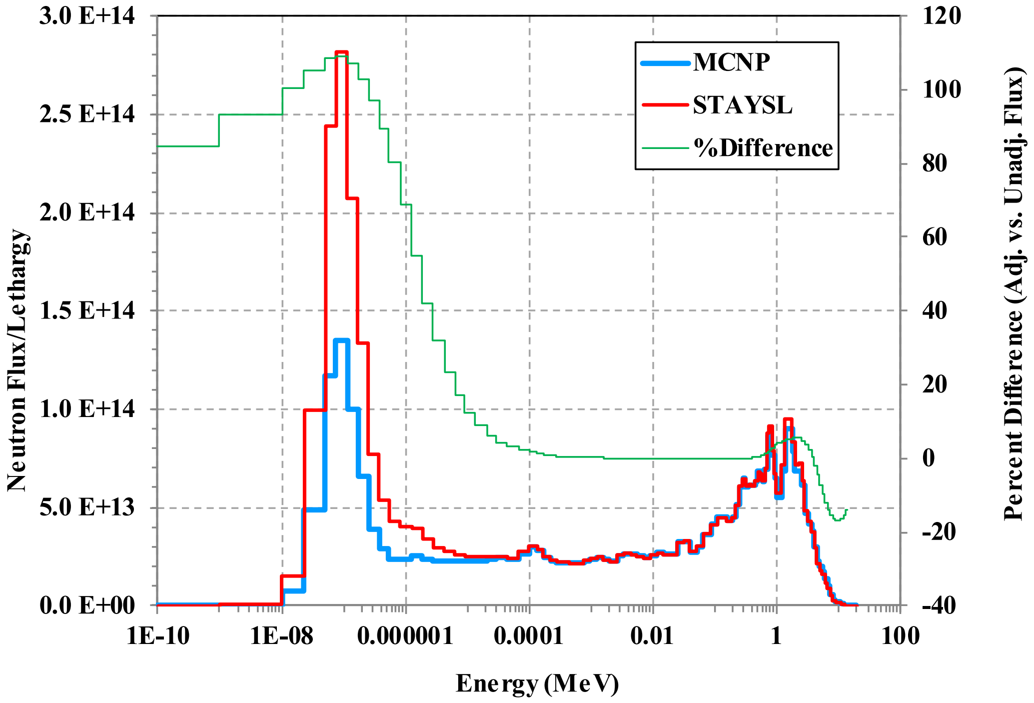

Figure 2. Comparison of the calculated MCNP neutron flux spectrum (blue) with the STAYSL adjustment (red) for CSM monitor E2 at 2.0” above midplane. The green line shows the percent difference for the spectral adjustment which is mainly to the thermal flux.

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Radiation Damage Calculations

The adjusted neutron spectra were used to calculate radiation damage parameters using the SPECTER computer code [4]. Displacement per atom (dpa) for several important elements and Type 316 stainless steel are listed in Table 7 and plotted in Figure 3. The small contributions to the stainless steel and Inconel dpa values from the Ni-59 reaction [4] are included in the calculations.

Table 7. Calculated DPA Values for the CSM Experiment

AE monitors (shaded rows) irradiated for 1 cycle; BCD monitors irradiated for 2 cycles

Height, in.	Fe	Al	Inconel718+	316SS*
-24.3	0.13	0.24	0.15	0.14
-22.9	0.21	0.39	0.23	0.22
-21.4	0.60	1.14	0.67	0.63
-19.9	0.72	1.36	0.82	0.75
-2.0	1.55	2.90	1.81	1.61
0.7	1.55	2.91	1.84	1.62
2.0	1.61	3.04	1.88	1.68
19.8	0.60	1.13	0.66	0.62
21.3	0.45	0.85	0.49	0.47
22.8	0.14	0.27	0.15	0.15
24.2	0.11	0.20	0.11	0.11

*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)

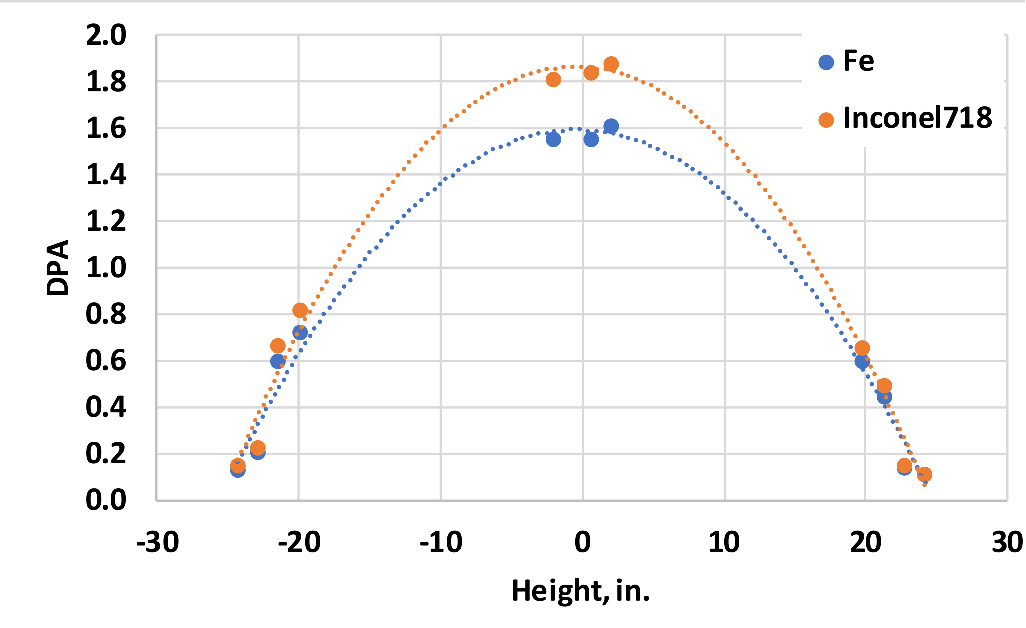

Figure 3 – Fe dpa values vs. height in the CSM assembly. The two monitors at the top and bottom were irradiated for only 1 cycle whereas the rest of the monitors were irradiated for two cycles in ATR.

References

[1] *CSM-10584 Loading and Marking Arrangement*, PLN-5306, Rev. 1, 02/04/2019, with provided drawings.

[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] 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.

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