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Changing the World's Energy Future

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Idaho National Laboratory

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Summary

The USU (Utah State University) Advanced Test Reactor National Scientific User Facility (ATR NSUF) experiments to test the performance of hafnium-aluminum composites were conducted in the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL) during cycles 149A through 151A from April 14, 2011 to February 13, 2012. Four sets of two neutron fluence monitor capsules were irradiated in four different combinations of reactor cycles. The assemblies were irradiated in the B2 position of the ATR. Neutron fluence monitors were located at 8 positions in the assemblies. These monitors were previously fabricated at Pacific Northwest National Laboratory (PNNL). After irradiation, the monitors were sent to PNNL for analysis. The neutron fluence monitor wires were removed from the vanadium capsules and analyzed by gamma and x-ray spectrometry to determine the activities of the activation products. The measured activation rates were used to adjust the neutron spectra at each location in the assemblies to determine the best fit to the neutron fluence spectra. Radiation damage (dpa) calculations were performed at all locations using the adjusted neutron spectra. The report was revised after receipt of a corrected irradiation history for Cycle 149A. A plot of the fast neutron flux as a function of height in the B2 position was also added to the report.

Preparation of Neutron Fluence Monitors

The neutron fluence monitors were previously prepared by PNNL and delivered to INL in July 2010 as per requirements specified in MPO# 00100896 and Statement of Work (SOW) for INL Project 59407 (SOW-8903, Rev. 0), dated 4/26/2010 . Small high-purity Fe, Ti, Nb, and 0.116% Co-Al wires were encapsulated in 8 vanadium capsules measuring 0.05" O.D. by about 0.34" long. The vanadium capsules had identification codes stamped on the bottom and each wire was accurately weighed. The vanadium tubes were electron beam welded in a vacuum and helium leak tested. The wire segments were typically 0.04" long and had a diameter of about 0.02". The monitor identification codes and wire weights are listed in Table 1. The complete documentation for the fabrication of the fluence monitors was provided to INL in a report dated July 2010[1].

The client ID numbers indicate the location of the fluence monitors in the four irradiation assemblies referred to as A, B, C, and D. The suffix -1 and -2 further indicates whether the capsules were irradiated in Al (-1) or Al_3Hf (-2) sections of each irradiation assembly.

[1] L.R. Greenwood, Preparation of Fluence Monitors For Idaho National Laboratory MPO# 00100896/SO -8903, Rev. 0. Pacific Northwest National Laboratory, July 2010.

Table 1. Neutron Fluence Monitors and Wire Weights

Client ID	PNNL Capsule ID	Fe Weight (mg)	Nb Weight (mg)	Ti Weight (mg)	Co-Al Weight (mg)	Capsule Weight (mg)
A-1	8R	1.217	1.333	1.150	1.094	48.286
B-1	1U	1.427	1.211	1.326	1.355	49.007
C-1	AU	1.449	1.487	1.047	1.058	49.378
D-1	X8	1.266	1.421	1.166	1.221	47.755
A-2	7H	1.326	1.098	1.066	1.210	51.184
B-2	7O	1.305	1.294	1.110	1.273	49.243
C-2	1L	1.518	1.344	1.109	1.153	43.181
D-2	2D	1.245	1.513	1.321	1.125	44.810

Irradiation History

The experiment was conducted in the small B position of ATR, B2 as per Figure 1. The total duration of the in-pile experiment was four ATR cycles (149A, 149B, 150B and 151A), commencing in April 2011 and lasting through February 2012. The experiment was removed from the reactor for a high power cycle that occurred during cycle 150A. The experiment assembly housed three capsules per cycle, stacked vertically in the reactor core. Four identical capsules, designated A through D, were assembled. Capsule E was a neutronically equivalent dummy capsule, prepared by INL. Three capsule assemblies were loaded in the reactor in a given cycle as indicated in Table 2. An identical set of specimens were contained in each capsule pair.

Table 2. Capsule irradiation schedule.

Position	Cycle 149A	Cycle 149B	Cycle 150B	Cycle 151A
Top	B	B	E	E
Middle	A	A	A	A
Bottom	C	D	D	D

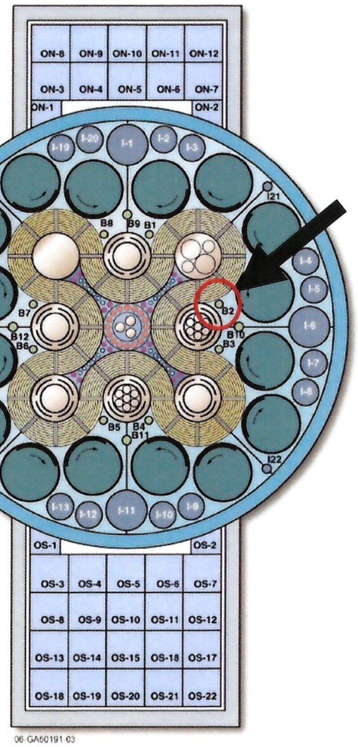

Figure 1. Diagram of irradiation positions in the ATR with B2 position highlighted.

Each capsule was laser etched with the capsule and assembly designator. This marking was placed at the top end of the capsule to indicate orientation in the reactor. The top (Assembly -1) of each capsule pair is designated USUA-1, USUB-1, USUC-1 or USUD-1; whereas, the bottom (Assembly -2) capsules are designated USUA-2, USUB-2, USUC-2 or USUD-2. Each capsule pair experienced a different irradiation history as shown in Table 2. For the cycle indicated (see Table 3), the color green indicates that the capsule pair was in the reactor, whereas the color red indicates that the capsule pair was removed. The EFPD (effective full power days) values were calculated as the average of the fluxes in the NE, C, and SE positions, which are thought to best represent the flux in the B2 position. The values are normalized to a full power flux of 21.35 MW, which was the average flux for cycle 149A. These values are not used in any subsequent analyses in this report and are only meant to indicate the relative exposure times of the four different assemblies.

Table 3. Capsule irradiation history. Green indicates that the monitor pair was irradiated in that reactor cycle whereas red indicates that they were not present. All capsules were removed during cycle 150A (not shown).

Capsule	149A	149B	150B	151A	EOI cycle*	EFPD**
A					151A	186.3
B					149B	91.9
C					149A	37.5
D					151A	148.8

*EOI = End of irradiation **EFPD = Effective full power days

Post-Irradiation Analyses

Following irradiation, the vanadium tubes were retrieved from the assemblies at INL and sent to PNNL for analysis. All of the vanadium tubes were opened using tube cutting pliers and the wires were removed. Each wire was then placed in a small plastic vial for gamma counting using procedure, RPG-CMC-450, Rev. 2, *Gamma Energy Analysis (GEA) and Low-Energy Photon Spectrometry (LEPS)*. Each fluence monitor wire was assigned a unique Radiochemical Processing Laboratory (RPL) # and an Analytical Service Request# 9377 for tracking purposes.

Each flux monitor was counted on a gamma detector calibrated with standards traceable to the National Institute of Standards and Technology (NIST). Gamma detector control counts are performed daily when the gamma spectrometers are in use and background counts are measured at least weekly. Nuclear decay data were taken from the National Nuclear Data Center at Brookhaven National Laboratory in 2011.

After gamma counting, the Nb wires were dissolved in an acid mixture of HNO_3 and HF and aliquots were mounted on very thin filter paper for x-ray counting using low energy photon spectrometers, which are thin Ge detectors optimized to have high-efficiency and high resolution for x-rays. The analysis procedure used was RPG-CMC-450, Rev. 2, *Gamma Energy Analysis (GEA) and Low-Energy Photon Spectrometry (LEPS).* The initial gamma count of the Nb wires detected the activity of ^{94}Nb and this isotope was used to verify the preparation of the x-ray mounts.

All of the gamma and x-ray counting results are listed in Table 4. Neutron activation reactions and products measured include ^{54}Fe(n,p)^{54}Mn; ^{93}Nb(n,g)^{94}Nb and ^{93}Nb(n,n')^{93m}Nb (from the x-ray counting); ^{59}Co(n,g)^{60}Co; and ^{46}Ti(n,p)^{46}Sc. This provides three reactions sensitive to fast neutrons with different energy thresholds and two reactions sensitive to thermal/epithermal neutrons.

The saturated reaction rates for the neutron activation reactions were calculated for the measured data 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, which is a module in our STAYSL PNNL code [1]. BCF calculates the decay for each time segment of constant flux using the equation A_i (1 – exp(- LT_i)), where A_i is the relative flux for time period T_i and L is the decay constant of each activation product given by ln(2)/halflife. The irradiation history was provided by staff at INL. Table 5 lists the correction factors that were used to determine the saturated reaction rates. Gamma self-absorption in the wires was calculated using the STAYSL PNNL software and the corrections averaged around 1%. 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 for neutron burnup was around 7% as shown in Table 5 for ^{60}Co. The minimum value is for the shortest irradiation time for the C samples and the maximum values correspond to the longest irradiation time for the A samples. Neutron self-absorption corrections are calculated and applied to the neutron activation cross sections prior to the spectral adjustment described later. The saturated reaction rates are listed in Table 6. The activation rates are normalized to an average power level of 21.35 MW using the EFPD values in Table 3.

Table 4. Measured Activities in Bq/mg for ASR 9377
(Decay corrected to End of Irradiation (EOI) dates as listed; absolute 1-sigma uncertainty 2%)

Sample ID	EOI (24 hr)	Sc-46	Mn-54	Co-60	Nb-94	Nb-93m
A1 8R	2/13/2012 9:59	5.30E+5	1.31E+6	6.28E+6	3.96E+4	2.35E+6
B1 1U	8/1/2011 6:59	4.17E+5	7.28E+5	3.02E+6	1.92E+4	1.08E+6
C1 AU	5/26/2011 12:59	8.50E+4	1.18E+5	4.71E+5	2.88E+3	1.65E+5
D1 X8	2/13/2012 9:59	1.70E+5	3.33E+5	1.67E+6	1.04E+4	6.54E+5
A2 7H	2/13/2012 9:59	4.91E+5	1.28E+6	5.52E+6	3.78E+4	2.21E+6
B2 7O	8/1/2011 6:59	1.32E+5	2.07E+5	5.45E+5	4.22E+3	2.99E+5
C2 1L	5/26/2011 12:59	2.51E+5	3.43E+5	1.08E+6	7.01E+3	4.96E+5
D2 2D	2/13/2012 9:59	4.68E+5	9.51E+5	4.28E+6	2.79E+4	1.81E+6

Table 5. Saturated Activity Correction Factors

Reaction	Atomic Weight	Isotopic Abundance	Gamma Abs.	Neutron Burnup Correction	
				Min	Max
$^{93}Nb(n,g)^{94}Nb$	92.906	1	0.987	0.997	0.956
$^{93}Nb(n,n')^{93m}Nb$	92.906	1	1	0.999	0.994
$^{59}Co(n,g)^{60}Co$*	58.933	0.00116	1.000	0.998	0.926
$^{54}Fe(n,p)^{54}Mn$	55.845	0.05845	0.989	0.999	0.981
$^{46}Ti(n,p)^{46}Sc$	47.867	0.0825	0.994	0.999	0.990

*Alloy of Co-Al wire NIST Standard Reference Material 953

Table 6. Saturated Reaction Rates in atom/atom-second (1-sigma uncertainties are 2%)

Sample ID	$^{54}Fe(n,p)^{54}Mn$	$^{46}Ti(n,p)^{46}Sc$	$^{93}Nb(n,n')^{93m}Nb$	$^{59}Co(n,g)^{60}Co$	$^{93}Nb(n,g)^{94}Nb$
A1 8R	7.12 E-12	8.76 E-13	1.69 E-11	9.01 E-09	3.72 E-10
B1 1U	6.46 E-12	8.03 E-13	1.56 E-11	8.12 E-09	3.57 E-10
C1 AU	2.40 E-12	3.21 E-13	5.79 E-12	2.98 E-09	1.29 E-10
D1 X8	2.09E-12	2.93E-13	5.86E-12	2.80E-09	1.18E-10
A2 7H	6.93 E-12	8.10 E-13	1.59 E-11	7.84 E-09	3.54 E-10
B2 7O	1.83 E-12	2.53 E-13	4.30 E-12	1.43 E-09	7.72 E-11
C2 1L	6.98 E-12	9.50 E-13	1.74 E-11	6.88 E-09	3.15 E-10
D2 2D	6.01E-12	8.11E-13	1.63E-11	7.40E-09	3.24E-10

Neutron Spectral Adjustment

The STAYSL PNNL [1] computer code was used to adjust the neutron energy spectrum at each location using the calculated reaction rates and uncertainties as input. STAYSL PNNL performs a least-squares adjustment to determine the most likely neutron spectrum at each capsule taking into account 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, IRDF2002 [2].

Calculations with STAYSL PNNL indicate good agreement with the initial fast neutron spectra for the Al and Al_3Hf assemblies provided by J. Parry at INL, although some adjustment was required for the thermal/epithermal neutrons. The adjusted neutron fluences from STAYSL PNNL are listed in Table 7. The thermal fluence is reported including all neutrons < 0.55 eV. The epithermal neutron fluence is listed with energy ranges of 0.55 eV to 0.11 MeV. The fast neutron fluences are listed for thresholds of 0.11 MeV and 1 MeV. The uncertainties take into account uncertainties in the saturated activation rates and nuclear activation cross sections. Typical neutron spectral adjustments are shown in Figures 2a and 2b for capsule C2 1L. The -2 positions showed the largest adjustments for the thermal neutron fluence.

Table 7. Adjusted Neutron Fluences (x 10^{21} n/cm^2)

		Thermal*		**Epithermal**		**Fast**		**Fast**	
	Height	**< 0.55 eV**		**0.55 eV to 0.11 MeV**		**> 0.11 MeV**		**> 1 MeV**	
Monitor	**Inches**	**n/cm^2**	**±%**	**n/cm^2**	**±%**	**n/cm^2**	**±%**	**n/cm^2**	**±%**
A1 8R	-6.6	4.21	3	4.28	7	3.22	5	1.41	3
B1 1U	9.4	1.87	3	1.95	7	1.46	5	0.638	3
C1 AU	-22.6	0.280	3	0.292	7	0.220	5	0.0959	3
D1 X8	-22.6	1.03	3	1.14	7	0.886	5	0.377	4
A2 7H	6.6	3.74	4	3.89	9	3.06	5	1.33	3
B2 7O	22.6	0.315	4	0.489	9	0.404	5	0.174	3
C2 1L	-9.4	0.644	4	0.805	9	0.668	5	0.286	3
D2 2D	-9.4	2.81	4	3.00	9	2.47	5	1.05	4

*Thermal fluence is defined as the sum of all neutrons < 0.55 eV.

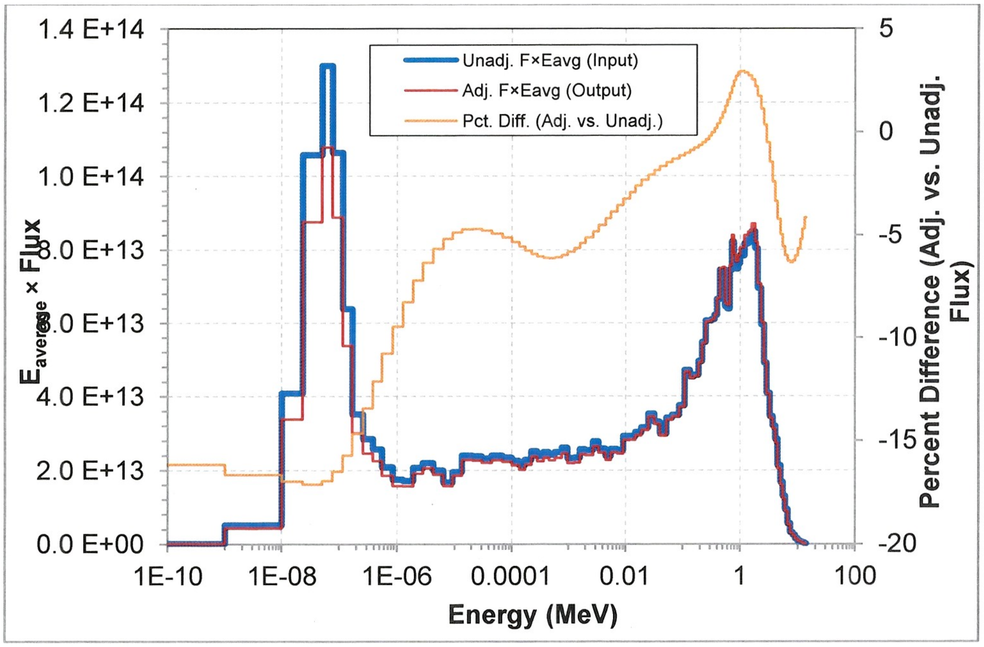

Figure 2a. Example STAYSL PNNL adjustment for position C2 1L. The blue curve is the starting neutron spectrum and the red curve shows the adjusted spectrum. The orange curve shows the percent difference using the y-axis on the right side.

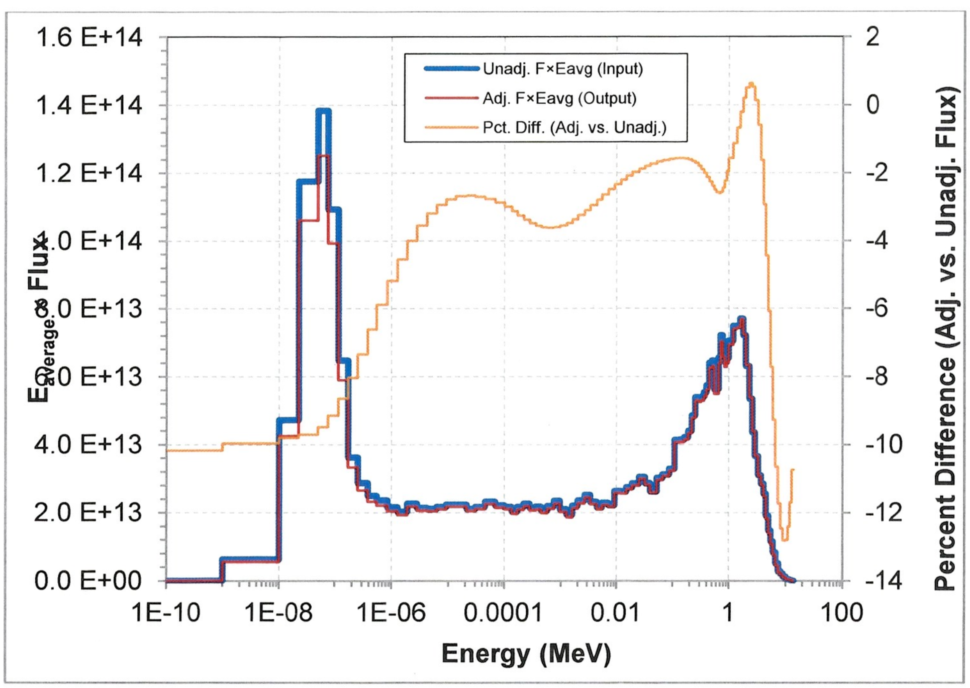

Figure 2b. Example STAYSL PNNL adjustment for position A1 8R. The blue curve is the starting neutron spectrum and the red curve shows the adjusted spectrum. The orange curve shows the percent difference using the y-axis on the right side.

Table 7 lists the vertical heights relative to midplane for the eight different experimental capsules. The average neutron fluxes can be compared for all of the positions by normalizing them to a common average power level of 21.35 MW resulting in the EFPD values listed in Table 3. It is important to note that this normalization is arbitrary and has no impact on the neutron fluence values listed in Table 7. The flux values can also be quoted at any other power level by simple linear renormalization. Figure 3 shows the flux values > 1 MeV as a function of vertical height above midplane for all of the experimental capsules. The measured flux values agree within experimental uncertainties with the expected cosine curve (IN-1260) [4] and MCNP calculations by J. Parry [5].

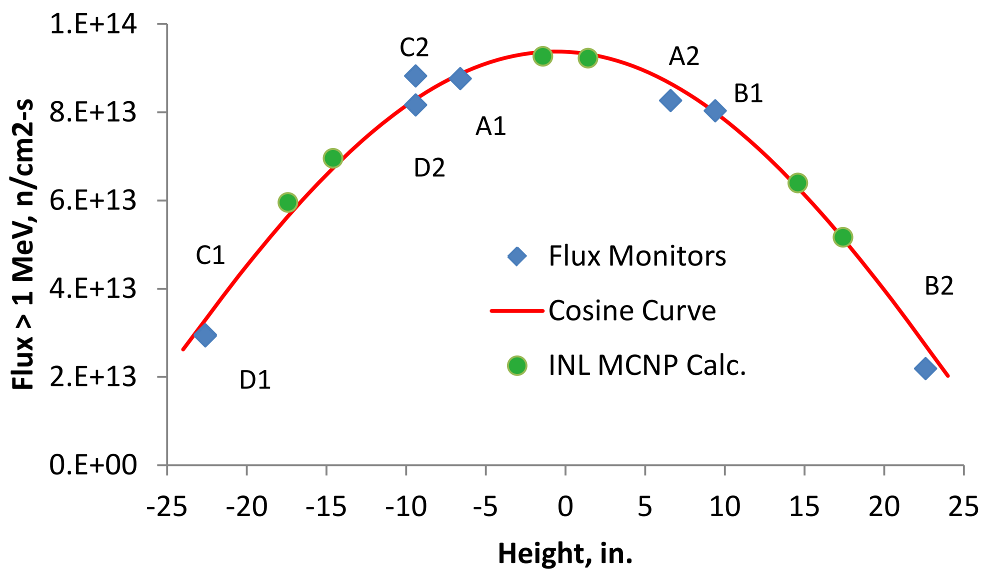

Figure 3. Neutron flux > 1 MeV normalized to an average power of 21.35 MW. Measured values (blue diamonds) are compared to the expected cosine curve (IN-1260) and MCNP calculation by INL (green dots).

Radiation Damage Calculations

The adjusted neutron spectra were used to calculate radiation damage parameters using the SPECTER computer code [3] and the results are listed in Table 8. The dpa (displacements per atom) values are defined as the average number of times an atom has been displaced from a lattice site and are calculated from the total damage energy assuming a typical binding energy of 40 eV for pure Fe and Ni. The damage energy is determined from the nuclear stopping energy of all primary atomic recoils that are created by the neutron exposure, including all possible nuclear reactions. The dpa unit represents the fundamental neutron-induced damage in materials. However, most of this damage is not permanent due to recombination of the vacancies and interstitial atoms. The dpa concept has proven to be a very useful concept for the characterization of irradiation experiments and correlation of neutron-induced radiation effects in very different neutron spectra such as thermal and fast reactors, 14-MeV sources, and accelerator based neutron sources.

Table 8. Calculated dpa values

Monitor	Fe	Ni
A1 8R	2.06	2.67
B1 1U	0.93	1.11
C1 AU	0.14	0.16
D1 X8	0.55	0.63
A2 7H	1.97	2.47
B2 7O	0.26	0.28
C2 1L	0.42	0.47
D2 2D	1.54	1.87

References

[1] L. R. Greenwood and C. D. Johnson, User Guide for the STAYSL PNNL Suite of Software Tools, PNNL Report 22253, January 2013.

[2] International Reactor Dosimetry File 2002 (IRDF-2002), IAEA Technical Reports Series No. 452, International Atomic Energy Agency, Vienna, Austria, November 2006.

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

[4] IN-1260, Reactor Physics Results from Low-Power Measurements in the Advanced Test Reactor, Idaho Nuclear Corporation, 1969.

[5] J. Parry, Idaho National Laboratory, personal communication, 2013.