INL/RPT-24-79219-Revision-0

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October 2014

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http://www.inl.gov

Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

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October 2014

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PNNL Project Number: 64525

INL SOW-11013, Rev. 1
MPO 00140214, with Amendment 01

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Neutron Fluence Measurements for the UCF HSIS Experiment in ATR

Summary

The University of Central Florida (UCF) experiments were irradiated in the Hydraulic Shuttle Irradiation System (HSIS) of the Advanced Test Reactor (ATR) at Idaho National Laboratory (INL) to study the effects of low neutron fluence on metallic fuels. Four samples were irradiated for 20 days from 2/20/2014 to 3/12/2014 and four additional samples were irradiated for 2 days from 3/17/2014 to 3/19/2014. All eight of the samples included neutron fluence capsules that were previously prepared at PNNL. After irradiation, the neutron fluence monitors were returned to Pacific Northwest National Laboratory (PNNL) for analysis to determine the neutron fluence that was received by each sample.

Preparation of Neutron Fluence Monitors

Ten neutron fluence monitors were previously prepared by PNNL and delivered to INL in December 2013 as per requirements specified in MPO# 00140214 and Statement of Work SOW-11013, Rev. 0). Small high-purity Fe, Ti, Nb, and 1% Co-V wires were encapsulated in ten 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. Whereas PNNL prepared ten neutron fluence monitors, two were not irradiated in these experiments. The complete documentation for the fabrication of the fluence monitors was provided to INL in the report, PNNL-64525, *Preparation of Fluence Monitors for Idaho National Laboratory MPO# 00140214/SOW-11013, Rev. 0* dated December 2013.

Table 1. Neutron Fluence Monitors and Wire Weights

Reactor Position	PNNL Capsule ID	Fe Weight (mg)	Ti Weight (mg)	Nb Weight (mg)	1%Co-V Weight (mg)	Capsule Weight (mg)
UCF-1	8A	2.277	1.560	2.106	1.355	43.809
UCF-3	8U	2.131	1.398	2.075	1.213	46.593
UCF-5	A9	1.924	1.643	2.176	1.505	46.686
UCF-7	8Y	2.052	1.559	2.191	1.229	47.887
UCF-8	1J	2.202	1.580	2.212	1.374	49.963
UCF-10	1V	2.071	1.578	2.204	1.415	47.390
UCF-12	2Y	2.052	1.633	1.925	1.558	46.022
UCF-14	A2	1.948	1.638	2.158	1.331	46.908

Post-Irradiation Analyses

Following irradiation, the neutron fluence monitors 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# of 9612 for tracking purposes.

Each fluence monitor wire 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 as a tracer to check for any loss of Nb during the preparation of the x-ray mounts.

All of the gamma and x-ray counting results are listed in Table 2. Neutron activation reactions and products measured include ^{54}Fe(n,p)^{54}Mn; ^{58}Fe(n,g)^{59}Fe; ^{93}Nb(n,g)^{94}Nb (and subsequent activation to ^{95}Nb) and ^{93}Nb(n,n')^{93m}Nb (detected by x-ray counting); ^{59}Co(n,g)^{60}Co; and ^{46}Ti(n,p)^{46}Sc. These results provide three neutron activation reactions sensitive to fast neutrons with different energy thresholds and three reactions sensitive to thermal/epithermal neutrons.

The saturated reaction rates for the neutron activation reactions were calculated for the measured data listed in Table 2 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 as 20 days for capsules UCF8, UCF10, UCF12, and UCF14 and 2 days for capsules UCF1, UCF3, UCF5, and UCF7. 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 during the irradiation period. Corrections were applied in an iterative method using the measured reaction rates as the first approximation and iterating until the process converges. Due to the very brief time of these irradiations, these corrections were less than 0.2% in all cases. 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 3.

Table 2. Measured Activities in Bq/mg
(Decay corrected to End of Irradiation (EOI) dates as listed)

Reactor Position	EOI	^{46}Sc	^{54}Mn	^{59}Fe	^{60}Co	^{94}Nb	^{93m}Nb
20-day Irradiation		±2%	±2%	±3%	±2%	±2%	±4%
5-UCF08 1J	3/12/2014	1.49E+5	1.91E+5	1.92E+6	5.45E+6	4.46E+3	2.57E+5
7-UCF10 1V	3/12/2014	1.42E+5	2.01E+5	1.55E+6	4.54E+6	4.01E+3	2.52E+5
9-UCF12 2Y	3/12/2014	1.55E+5	2.05E+5	1.51E+6	4.37E+6	3.92E+3	2.80E+5
11-UCF14 A2	3/12/2014	1.38E+5	1.77E+5	1.84E+6	5.19E+6	4.23E+3	2.58E+5
2-day Irradiation		±2%	±2%	±3%	±2%	±2%	±4%
5-UCF01 8A	3/19/2014	1.48E+4	1.76E+4	2.15E+5	4.94E+5	4.28E+2	2.38E+4
7-UCF03 8U	3/19/2014	1.64E+4	2.04E+4	1.83E+5	4.11E+5	3.99E+2	2.83E+4
9-UCF05 A9	3/19/2014	1.46E+4	1.86E+4	1.67E+5	4.01E+5	3.92E+2	2.55E+4
11-UCF07 8Y	3/19/2014	1.56E+4	1.81E+4	2.09E+5	4.81E+5	4.21E+2	2.61E+4

Table 3. Saturated Reaction Rates in atom/atom-second
(1-sigma uncertainties are ±2% except for ^{59}Fe at ±3% and ^{93m}Nb at ±4%)

Sample ID	$^{54}Fe(n,p)^{54}Mn$	$^{46}Ti(n,p)^{46}Sc$	$^{93}Nb(n,n')^{93m}Nb$	$^{59}Co(n,g)^{60}Co$	$^{93}Nb(n,g)^{94}Nb$	$^{58}Fe(n,g)^{59}Fe$
UCF08 1J	7.07 E-12	9.48 E-13	1.69 E-11	7.48 E-09	3.75 E-10	2.38 E-10
UCF10 1V	7.43 E-12	9.04 E-13	1.66 E-11	6.23 E-09	3.37 E-10	1.92 E-10
UCF12 2Y	7.58 E-12	9.86 E-13	1.84 E-11	5.99 E-09	3.29 E-10	1.87 E-10
UCF14 A2	6.55 E-12	8.78 E-13	1.69 E-11	7.13 E-09	3.55 E-10	2.28 E-10
UCF01 8A	6.37 E-12	8.74 E-13	1.56 E-11	6.72 E-09	3.58 E-10	2.33 E-10
UCF03 8U	7.39 E-12	9.69 E-13	1.86 E-11	5.59 E-09	3.34 E-10	1.98 E-10
UCF05 A9	6.73 E-12	8.63 E-13	1.67 E-11	5.45 E-09	3.28 E-10	1.81 E-10
UCF07 8Y	6.55 E-12	9.22 E-13	1.71 E-11	6.54 E-09	3.52 E-10	2.26 E-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]. The starting neutron spectra were taken from reference 4.

The adjusted neutron fluences from STAYSL PNNL are listed in Table 4. The thermal fluence is reported including all neutrons < 0.55 eV. The epithermal neutron fluence is listed with an energy range 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.

Table 4. Adjusted Neutron Fluences (x 10^{20} n/cm^2)

	Total		Thermal*		Epithermal		Fast		Fast	
			< 0.55 eV		0.55 eV to 0.11 MeV		> 0.11 MeV		> 1 MeV	
Position	**n/cm²**	**±%**	**n/cm²**	**±%**	**n/cm²**	**±%**	**n/cm²**	**±%**	**n/cm²**	**±%**
UCF08 1J	11.7	3	3.67	3	4.54	7	3.45	5	1.50	4
UCF10 1V	10.8	3	2.96	3	4.43	7	3.42	5	1.52	4
UCF12 2Y	11.0	3	2.81	3	4.56	7	3.65	5	1.62	4
UCF14 A2	11.2	3	3.50	3	4.34	7	3.36	5	1.46	4
UCF01 8A	1.06	3	0.330	3	0.414	7	0.315	5	0.137	4
UCF03 8U	1.09	4	0.258	3	0.462	7	0.370	5	0.162	4
UCF05 A9	1.00	3	0.257	3	0.414	7	0.330	5	0.146	4
UCF07 8Y	1.10	3	0.316	3	0.442	7	0.343	5	0.147	4

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

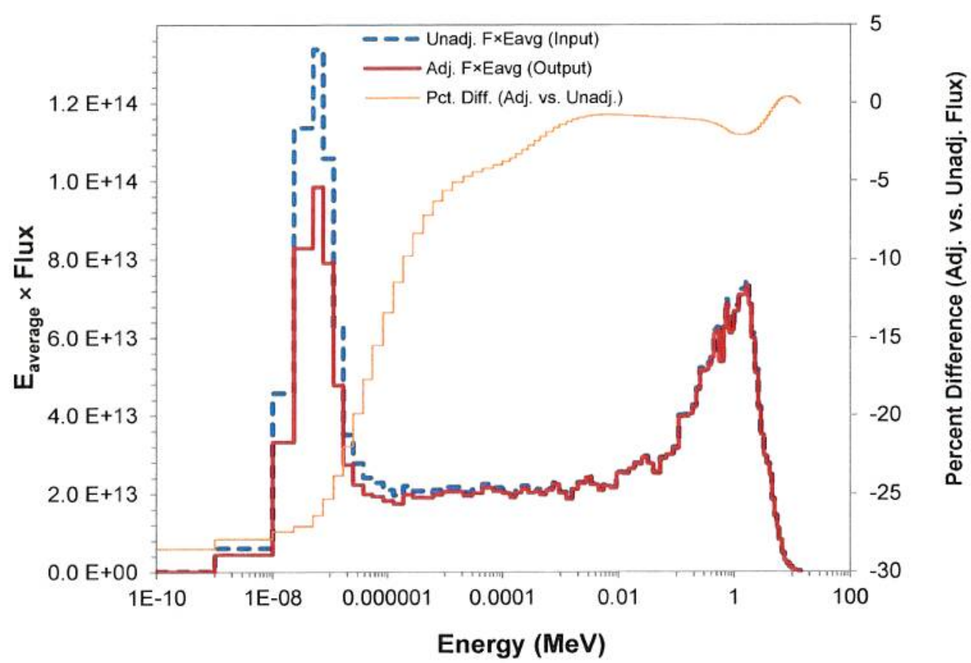

Figure 1. Example of the STAYSL PNNL adjustment for position UCF8. The blue curve is the starting neutron spectrum and the red curve shows the adjusted spectrum. The orange curve shows the percent difference using the y-axis on the right side.

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 5. 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 5. Calculated dpa Values

Monitor	Fe	Ni
UCF08 1J	0.22	0.24
UCF10 1V	0.22	0.24
UCF12 2Y	0.23	0.25
UCF14 A2	0.21	0.23
UCF01 8A	0.020	0.022
UCF03 8U	0.023	0.026
UCF05 A9	0.021	0.023
UCF07 8Y	0.022	0.024

References

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