NEUTRON DOSIMETRY AND RADIATION DAMAGE CALCULATIONS FOR HFBR -

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OBJECTIVE

To provide neutron dosimetry and radiation damage analyses for fusion materials irradiations.

SUMMARY

Neutron dosimetry measurements have been conducted for various positions of the High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory (BNL) in order to measure the neutron flux and energy spectra. Neutron dosimetry results and radiation damage calculations are presented for positions V10, V14, and V15.

PROGRESS AND STATUS

Introduction

The HFBR has an enriched fuel core and a heavy water reflector, thus offering a wide range of neutron spectra for irradiation experiments. Researchers from Oak Ridge National Laboratory (ORNL) recently utilized this spectral capability to investigate effects of neutron spectrum on radiation damage in steels. In support of this effort, comprehensive measurements of neutron flux and energy spectra were conducted, as documented in this report.

Irradiation History

The first dosimetry measurements in HFBR were conducted in 1976 in position V15 in support of experiments planned by C. L. Snead (BNL). More recent dosimetry measurements were started in 1989 with two irradiations in positions V10 and V15. Additional planned measurements were delayed by the shutdown of HFBR in the 1990-1991 timeframe. Measurements were resumed in 1994 in V10 and V14. Recently, measurements were conducted in 1996 with several irradiations in positions V10, V14, and V15. Planned additional measurements have been delayed by the present shutdown of the reactor. The irradiation histories for all of these irradiations are summarized in Table 1 below.

The first (V15-S3) and last (V10-5) irradiations included complete spectral sets of dosimeters with both bare and cadmium-covered monitors. The spectral sets consisted of small wires of Fe, 0.1% Co-Al, Al, Ni, Ti, 0.1% Au-Al, 2.2% Lu-Al, Nb, 80% Mn-Cu, and encapsulated oxides of ^{237}Np and ^{238}U. All of the other capsules contained a reduced set of monitors including Fe, 0.1% Co-Al, Al, Ni, and Ti. The irradiations in 1994, denoted as ORNL, were

*Pacific Northwest National Laboratory is operated for the U.S. Department of Energy by Battelle Memorial Institute under Contract DE-ACO6-76RLO 1830.

conducted by C.A. Baldwin at Oak Ridge National Laboratory. Following gamma counting at ORNL, the results were sent to PNNL for analysis. There was one additional irradiation in HFBR denoted as V15-1 that included a full spectral set of monitors and a cadmium cover. The capsules were doubly encapsulated in quartz resulting in very high heating that melted the cadmium; however, most of the monitors were recovered for analysis. The dual irradiations denoted as V10-2 and V10-10 were designed to compare bare and cadmium covered reaction rates; however, due to a miscommunication the two capsules were identical without any cadmium cover.

Table 1. Summary of HFBR dosimetry measurements

Position-Dosimeter	Power (MW)	Height,in.@	Start Date/Time	Stop Date/Time	EFPH*
V15-S3	40	-0.85	4/30/76	4/30/76	#
V15-7	60	0	3/07/89 13:53	3/07/89 17:53	4
V10-3	60	-7.1	3/03/89 13:50	3/07/89 13:53	96
V10-ORNL	30	-7.1	10/21/94 9:55	10/21/94 17:55	8
V14-ORNL	30	-6.1	10/12/94 10:47	10/12/94 18:47	8
V10-2	30	-7.1	4/29/96 14:03	4/30/96 14:03	24
V10-10	30	-7.1	4/29/96 14:03	4/30/96 14:03	24
V10-1	30	-7.1	5/08/96 10:14	5/08/96 15:19	5:08
V14-4	30	-6.1	5/20/96 15:12	5/21/96 15:20	24:13
V10-5	30	-7.1	7/31/96 10:00	7/31/96 17:17	6:17
V15-1	30	0	8/26/96 10:42	8/26/96 15:07	4:25

@Most irradiations were at the bottom of each thimble except for the V15 runs, as noted.
*EFPH = effective full power hours at stated power.
#Details of the 1976 irradiation could not be retrieved from the available records.

Gamma Counting and Data Reduction

L. R. Greenwood analyzed the irradiations in 1976 and 1989 at Argonne National Laboratory. Chuck Baldwin at ORNL conducted the two irradiations in 1994 and the data were analyzed at PNNL. All of the other irradiations were analyzed by the present authors at Pacific Northwest National Laboratory. In all cases, individual monitors retrieved from the dosimetry capsules were gamma-counted using high-resolution Ge spectrometers. The measured activities were then converted to saturated activation rates by correcting for the decay during and after irradiation, gamma self shielding, atomic weight, and fission yield, as appropriate. The resultant activation rates are listed in Tables 2-3 in units of product atom/target atom-second. The values have an estimated accuracy of 2-3%, except as noted. The largest sources of uncertainty are due to the counting statistics and detector calibration. Results measured in 1976 are not listed since these values may contain some small differences in the nuclear decay data and neutron activation cross sections that need to be evaluated.

The cadmium cover and neutron self-shielding corrections are not included for the values listed in Tables 2-3. Since these effects depend on the neutron flux spectrum, we corrected the energy-dependent neutron activation cross sections so that spectral adjustments, described later, automatically include the proper corrections for these two effects.

Several problems were encountered in the analysis of the activation data for position V10. All of these problems are due to the very thermalized neutron spectrum, which makes it possible for thermal neutron or photon effects to compete with fast neutron reactions. In particular, the $^{27}Al(n,a)^{24}Na$ and $^{46}Ti(n,p)^{46}Sc$ rates are deemed to be unreliable due to competition from (n,γ) reactions with Na and Sc, respectively. The fission rates for ^{237}Np and ^{238}U are also deemed to be unreliable due to photofission effects which were calculated using photon flux calculations provided by Eugene Hu (BNL).

The data for position V10 listed in Table 2 also show an unexplained decrease (about 15%) in the activation rates measured with the full spectral set (V10-5). For the thermal neutron reactions, this decrease might be explained by a flux depression effect caused by the cadmium cover. However, similar effects are seen with the threshold reactions, which are not sensitive to thermal flux depression effects. Another possible cause for the difference may be the reactor power history, which involved a 1 hour unplanned reactor shutdown for this particular irradiation although reactor power history corrections attempted to correct for this effect. Finally, such effects may be caused by differences in the reactor fuel cycle. However, measurements in V10 were deliberately designed to sample different parts of the fuel cycle and the good agreement between the other four experiments would suggest that such effects are quite small. The decreased reaction rates for V10-5 are thus not understood at this time.

Neutron Flux and Spectral Adjustments

The activation rates listed in Tables 2-3 are integral quantities equal to the energy integral of the neutron activation cross section times the neutron flux spectrum. Since the neutron activation cross sections are relatively well known, the set of integral equations represented by the data can be solved by a least-squares technique to determine the neutron flux spectrum that provides the best fit to the data. This spectral adjustment was performed with the STAY'SL computer code [1] which takes into account all known uncertainties.

Eugene Hu (BNL) provided starting neutron flux spectra calculated at a reactor power level of 40 MW. The neutron spectral adjustment results are presented in Table 4 and shown in Figure 1. As can be seen, in general the measured neutron flux values are about 20-40% lower than the calculations. This difference is seen with both the thermal and fast neutron fluxes, which have the lowest uncertainty in the measurements. In the case of V10-5 and V15-S3, full spectral sets were analyzed resulting in reasonably low uncertainties for all neutron energy ranges. However, for the other measurements, the dosimetry reactions used do not result in much sensitivity to neutron energies between 0.5 eV and about 1 MeV; hence, fluxes in this range have a larger uncertainty, as stated in Table 4. Hopefully, additional measurements using full spectral sets will be conducted when HFBR resumes operation.

For position V15, the flux values determined from run V15-7 at 60 MW in 1989 are in good agreement with the more recent results from run V15-1 at 30 MW in 1996. The flux values measured for this position in 1976 agree within 15% except in the epithermal flux region, as discussed above. The current flux values are believed to be more reliable since they are based on more recent neutronics calculations that better define the flux spectrum.

Radiation Damage Calculations

The adjusted neutron spectra were used to calculate displacements per atom (dpa) values for various elements and compounds using the SPECTER computer code [2]. The results are listed in Table 5. Values are quoted for the specific irradiations that were conducted. Total and fast (> 1 MeV) neutron fluences are also listed and the ratios (dpa/E+22 n/cm^2) are given so that damage rates can be calculated for any length of irradiation. The calculations for SiC were performed using the SPECOMP computer code to calculate the dpa cross sections. Displacement threshold values of 20 eV were assumed for both the Si and C atoms.

Table 2. Activation rates (atom/atom-s) for position V10 of HFBR

Reaction	Run 1	Run 2	Run 5	Run10	ORNL	Run 3	Comments
	30MW	30MW	30MW	30MW	30MW	30MW*	
^{54}Fe(n,p)^{54}Mn			7.37E-15				
+Cd			7.16E-15				
^{58}Fe(n,γ)^{59}Fe	1.80E-10	1.76E-10	1.54E-10	1.75E-10	1.59E-10	1.63E-10	
+Cd			5.06E-12				
^{59}Co(n,γ)^{60}Co	5.53E-09	5.33E-09	4.59E-09	5.31E-09	4.89E-9	5.06E-09	
+Cd			1.54E-11				
^{27}Al(n,α)^{24}Na	5.57E-16	4.19E-16		6.88E-16		4.13E-16	23Na(n,γ)?
^{58}Ni(n,p)^{58}Co	1.01E-14	9.23E-15	9.17E-15	8.96E-15	7.72E-15	7.18E-15	
^{46}Ti(n,p)^{46}Sc	1.65E-15	1.64E-15	1.29E-15	1.69E-15		1.62E-15	45Sc(n,γ)?
^{47}Ti(n,p)^{47}Sc	1.92E-15	2.27E-15		2.20E-15		1.59E-15	
^{48}Ti(n,p)^{48}Sc	5.66E-17	6.09E-17		6.32E-17		4.99E-17	
^{197}Au(n,γ)^{198}Au			1.26E-08				
+Cd			3.54E-10				
^{176}Lu(n,γ)^{177}Lu			4.73E-07				
+Cd			2.68E-09				
^{93}Nb(n,γ)^{94}Nb			1.48E-10				
^{55}Mn(n,2n)^{54}Mn			7.05E-17				
^{237}Np(n,γ)^{238}Np +Cd			4.52E-10				
^{238}U(n,γ)^{239}Np +Cd			1.93E-11				
^{238}U(n,fission) +Cd			1.29E-13				Photofission
^{237}Np(n,fission) +Cd			5.32E-13				Photofission

*Rates for run V10-3 were renormalized from 60 MW to 30 MW.

Table 3. Activation rates (atom/atom-s) for positions V14 and V15 in HFBR

Position	V14	V14	V15	V15
Reaction	Run 4	ORNL	Run 1	Run 7
	30 MW	30 MW	30 MW	30 MW*
$^{54}Fe(n,p)^{54}Mn$	2.92E-12	3.19E-12	1.12E-11	1.06E-11
$^{58}Fe(n,\gamma)^{59}Fe$	4.98E-10	4.94E-10	1.78E-10	1.80E-10
$^{59}Co(n,\gamma)^{60}Co$	1.69E-08	1.67E-08	8.06E-9	8.50E-09
$^{93}Nb(n,\gamma)^{94}Nb$			8.36E-10	
$^{27}Al(n,\alpha)^{24}Na$	4.56E-14			8.75E-14
$^{58}Ni(n,p)^{58}Co$	3.06E-12	3.54E-12	1.46E-11	1.38E-11
$^{46}Ti(n,p)^{46}Sc$	4.55E-13			1.50E-12
$^{47}Ti(n,p)^{47}Sc$	7.23E-13			2.51E-12
$^{48}Ti(n,p)^{48}Sc$	1.35E-14			3.78E-14
^{238}U(n,fission)+Cd			4.82E-11	
^{237}Np(n,fission+Cd			3.06E-10	

*Rates for run V15-7 were renormalized from 60 MW to 30 MW.

Table 4. Comparison of calculated and adjusted neutron fluxes (n/cm^2-s)

V10 (-7.1")

E,MeV	Exp.(PNNL)		Calc.(BNL)	E/C/Norm*
Power	30	+/–%	40	
Th, <0.5 eV	1.57E+14	6	3.48E+14	0.60
0.5 eV-0.1 MeV	2.01E+12	16	1.92E+12	1.40
>0.11 MeV	2.84E+11	22	2.05E+11	1.85
>1.0 MeV	1.03E+11	23	3.91E+10	3.51
Total	1.59E+14	6	3.50E+14	0.61

V14 (-6.1")

E,MeV	Exp.(PNNL)		Calc.(BNL)	E/C/Norm*
Power	30	+/–%	40	
Th, <0.5 eV	4.96E+14	10	8.45E+14	0.78
0.5 eV-0.1 MeV	4.99E+14	20	6.80E+14	0.98
>0.11 MeV	5.99E+13	10	1.24E+14	0.64
>1.0 MeV	2.38E+13	11	5.05E+13	0.63
Total	1.06E+15	8	1.65E+15	0.86

V15 (Midplane) (1996)

E,MeV	Exp.(PNNL)		Calc.(BNL)	E/C/Norm*
Power	30	+/–%	40	
Th, <0.5 eV	1.23E+14	8	2.36E+14	0.69
0.5 eV-0.1 MeV	1.03E+15	15	1.82E+15	0.76
>0.11 MeV	2.64E+14	10	4.89E+14	0.72
>1.0 MeV	1.11E+14	11	1.83E+14	0.81
Total	1.42E+15	8	2.54E+15	0.75

V15 (Midplane) (1989)

E,MeV	Exp.(PNNL)		Calc.(BNL)	E/C/Norm*
Power	60	+/–%	40	
Th, <0.5 eV	2.40E+14	8	2.36E+14	0.68
0.5 eV-0.1 MeV	2.19E+15	15	1.82E+15	0.80
>0.11 MeV	4.80E+14	10	4.89E+14	0.85
>1.0 MeV	1.87E+14	11	1.83E+14	0.68
Total	2.92E+15	8	2.54E+15	0.77

V15 (-0.85") (1976)

E,MeV	Exp.(PNNL)		Calc.(BNL)	E/C/Norm*
Power	40	+/–%	40	
Th, <0.5 eV	1.55E+14	11	2.36E+14	0.66
0.5 eV-0.1 MeV	9.41E+14	12	1.82E+15	0.52
>0.11 MeV	5.84E+14	12	4.89E+14	1.19
>1.0 MeV	1.43E+14	10	1.83E+14	0.78
Total	1.68E+15	6	2.54E+15	0.66

*Ratio of experimental (PNNL) to calculated (BNL) flux normalized to the same reactor power.

Table 5. Radiation damage calculations for HFBR (per day)

Material	dpa	dpa/ 10^{22} n/cm^2
V14 Fluence >1 MeV = 2.05×10^{18} n/cm^2		
Fe	4.00E-3	19.5
Al	6.86E-3	33.5
Cu	5.60E-3	27.3
V	6.39E-3	31.2
SiC	7.82E-3	38.1
V15 Fluence >1 MeV = 9.62×10^{18} n/cm^2		
Fe	1.57E-2	16.3
Al	2.95E-2	30.7
Cu	2.10E-2	21.8
V	2.29E-2	23.8
SiC	3.36E-2	34.9
V10 Total Fluence = 13.8×10^{18} n/cm^2		
Fe	1.23E-4	0.089
Al	5.43E-5	0.039
Cu	2.18E-4	0.159
V	2.64E-4	0.192
SiC	4.41E-5	0.032

FUTURE WORK

At the present time, the HFBR reactor is not operational pending the resolution of regulatory concerns with the State of New York. Further work is planned to study additional positions in the reactor when operations are resumed.

ACKNOWLEDGEMENTS

This work was supported by Contract 19292A with K. Farrell of Oak Ridge National Laboratory, Oak Ridge, TN 37831. The research was sponsored by the Division of Materials Science, US Department of Energy under Contract DE-AC05-96OR22464 with Lockheed Martin Energy Research Corp. The authors would like to acknowledge the extensive support and cooperation of N.E. Holden and J.R. O'Conor of BNL.

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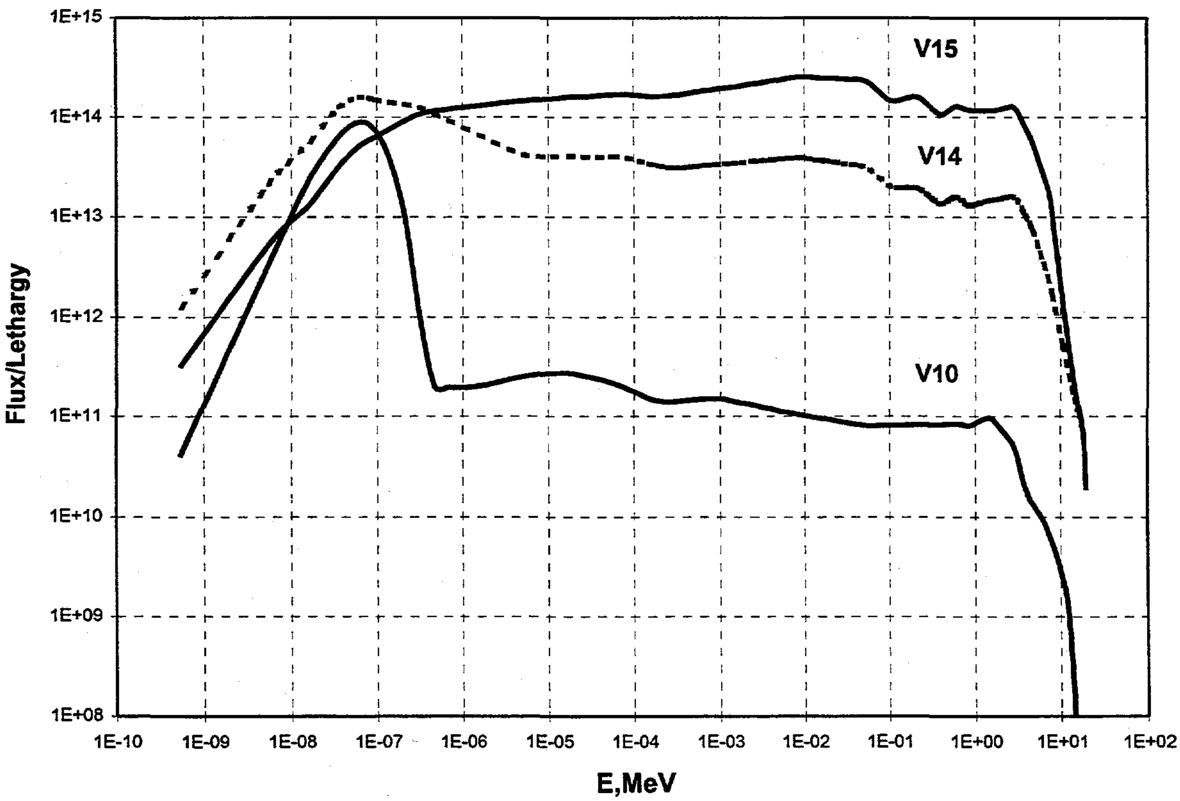

Figure 1. Adjusted neutron spectra for HFBR irradiation positions