

NEUTRON DOSIMETRY, DAMAGE CALCULATIONS, AND HELIUM MEASUREMENTS FOR THE HFIR-MFE-60J-1 AND MFE-330J-1 SPECTRAL TAILORING EXPERIMENTS - L. R. Greenwood (Pacific Northwest Laboratory<sup>a</sup>), C. A. Baldwin (Oak Ridge National Laboratory), B. M. Oliver (Rockwell International)

## OBJECTIVE

To provide dosimetry and damage analysis for fusion materials irradiation experiments.

## SUMMARY

Neutron fluence measurements and radiation damage calculations are reported for the joint U.S.-Japanese MFE-60J-1 and MFE-330J-1 experiments in the hafnium-lined removable beryllium (RB\*) position of the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL). These experiments were continuations of the ORR-6J and 7J irradiations performed in the Oak Ridge Research Reactor (ORR). The combination of irradiations was designed to tailor the neutron spectrum in order to achieve fusion reactor helium/dpa levels in stainless steel. These experiments produced maximum helium (appm)/dpa (displacement per atom) levels of 10.2 at 18.5 dpa for the ORR-6J and HFIR-MFE-60J-1 combination and 11.8 at 19.0 dpa for the ORR-7J and HFIR-MFE-330J-1 combination. A helium measurement in one JPCA sample was in good agreement with helium calculations.

## PROGRESS AND STATUS

The design of these spectral tailoring experiments has been published,<sup>1,2</sup> as were neutron dosimetry and damage calculations for the ORR-6J and -7J irradiations.<sup>3,4</sup> The ORR irradiations were conducted from June 28, 1983, to March 26, 1987, for a total exposure of 475 effective full power days (EFPD) at 30 MW. The 6J capsules were irradiated in position C7 at 60°C and 200°C. The 7J capsules were in position C3 at 330°C to 400°C. Materials effects specimens were then repackaged for insertion in HFIR. The materials from the 60°C irradiation in ORR-6J were subsequently irradiated in HFIR-MFE-60J-1 and those from ORR-7J were irradiated in HFIR-MFE-330J-1.<sup>1</sup> The 60J-1 and 330J-1 irradiations were conducted from July 20, 1990, to November 14, 1992, for a total of 522.7 EFPD at 85 MW. Both experiments were positioned in hafnium-lined removable beryllium (RB\*) positions of HFIR.

The goal of these extended irradiations was to achieve fusion reactor levels of helium(appm)/dpa in stainless steel of about 10/1. Helium production is enhanced by the two-step nickel reactions  $^{58}\text{Ni}(n,\gamma)^{59}\text{Ni}(n,\alpha)^{56}\text{Fe}$ . The ORR portion of the irradiation produced about 1.5 at%  $^{59}\text{Ni}$  in nickel. Further irradiation required a thermal neutron shield (hafnium) to prevent excessive helium accumulation.<sup>1</sup>

Dosimetry for the HFIR-60J-1 and 330J-1 experiments consisted of 24 aluminum capsules, four at each of six vertical levels, about 0.05 in OD by 0.25 in. long. Each capsule contained small wire segments of Fe, Ni, Nb, Ti, 0.1% Co-Al alloy, and 80.2% Mn-Cu alloy. After irradiation, eight capsules (four from each experiment) were opened in hot cells and gamma counted at ORNL.

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The gamma-counting results were forwarded to Pacific Northwest Laboratory (PNL) for analysis. The measured activities were converted to activation rates as listed in Table 1 by correcting for burnup, gamma self-absorption, decay during irradiation, isotopic abundance and atomic weight. Burnup corrections are based on an iterative procedure for the thermal/epithermal monitor reactions. The resultant estimates of the thermal/epithermal neutron fluences were then used to calculate burnup corrections for the threshold fast neutron monitor reactions. Burnup corrections averaged 10-15% for the thermal/epithermal reactions and only a few percent for the threshold reaction rates. The activation rates listed in Table 1 are normalized to full reactor power of 85 MW and have a net absolute uncertainty of about 5%.

The two irradiations were nearly identical, the largest difference being for the  $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$  reaction where the midplane 60J results are about 8.5% higher than the 330J results. The absence of this difference in the  $^{93}\text{Nb}(n,\gamma)^{94}\text{Nb}$  reaction suggests that the difference may be due to epithermal neutrons near the Co resonance rather than to remnant thermal neutrons.

The two runs were averaged to determine the best fit to the activation rates. The values in Table 1 were fit to a polynomial function of form  $f(x) = f(0) [ 1 + a x^2 ]$ , where  $x$  is the vertical height from reactor centerline in cm. All of the data are reasonably well fit by the average polynomial (coefficient  $a = -1.276 \times 10^{-3}$ ). Midplane activation rates were then used in the STAY'SL computer code to adjust the neutron flux spectrum determined in spectral measurements at the RB\* position in HFIR.<sup>5</sup> STAY'SL performs a generalized least-squares adjustment of all measured and calculated values including the measured activities, calculated spectra, and neutron cross sections.<sup>6</sup> Neutron cross sections and their uncertainties were generally taken from ENDF/B-V. The resultant neutron fluence values are listed in Table 2, and the neutron energy spectrum is shown in Figure 1. The activation rates and the derived neutron spectra and fluences are in excellent agreement with previous measurements in the RB\* position of HFIR.<sup>5</sup>

Neutron damage calculations with the SPECTER computer code<sup>7</sup> were performed for the midplane position. Midplane dpa and helium (appm) values are also listed in Table 2. The fluence and damage values at other experimental positions can be calculated by the gradient equation given above. Damage parameters for other elements or compounds have been calculated and are readily available on request.

Helium production in nickel and nickel alloys requires a more complicated non-linear calculation.<sup>8</sup> Values for the ORR-6J and -7J irradiations were published.<sup>3,4</sup> A further calculation was required to determine the net production of  $^{59}\text{Ni}$ . The result predicts maximum  $^{59}\text{Ni}$  content in pure nickel of 1.33 at% and 1.52 at% for ORR-6J and -7J, respectively. These values were then assumed as the starting conditions for the HFIR-MFE-60J-1 and -330J-1 irradiations. The growth of  $^{59}\text{Ni}$  and helium production in pure nickel for the combined irradiations is illustrated in Figure 2 which illustrates the maximum helium production in both irradiations. The growth of helium in ORR rises sharply with the production of  $^{59}\text{Ni}$ ; however, helium grows at a much slower rate in HFIR because of the hafnium thermal neutron shield. Helium production in stainless steel is detailed for the MFE-60J-1 and -330J-1 irradiations in Table 3. Table 4 lists the maximum net helium production in stainless steel for the ORR and HFIR irradiation sequences. Because the growth of helium in nickel is non-linear, detailed helium calculations must be performed for each material specimen, depending on the relative location in each reactor assembly.

The helium content in one JPCA sample was measured at Rockwell International. JPCA has the following nominal composition in wt%: C, 0.06; Si, 0.50; Mn, 1.77; P, 0.027; S, 0.005; Ni, 15.6; Cr, 14.22; Mo, 2.28; Ti, 0.24; B, 0.0031; N, 0.0039; Co, 0.002; and Fe, balance. The JPCA sample, identified as ONO5, was irradiated in TEM holder J2 in Level 1 of the 330°C section of ORR-7J at about 0.62" below the reactor centerline and then at 2.6" above the reactor centerline in the HFIR-RB\*-330J-1 assembly.

Before helium measurement, the JPCA sample was acid etched to remove surface material that might affect helium recoil into or out of the sample during irradiation. Two pieces were cut from the sample

Table 1. Activation Rates (at/at-s) - MFE-60J/330J

Pos.	Ht,cm	$^{54}\text{Fe}(n,p)^{54}\text{Mn}$ (E-11)	$^{46}\text{Ti}(n,p)^{46}\text{Sc}$ (E-12)	$^{55}\text{Mn}(n,2n)^{54}\text{Mn}$ (E-14)	$^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ (E-9)	$^{93}\text{Nb}(n,\gamma)^{94}\text{Nb}$ (E-10)
60-1	15.08	0.83	1.18	2.70	3.44	3.06
60-5	9.05	1.04	1.49	3.39	3.85	3.84
60-9	3.02	1.11	1.59	3.60	4.26	4.25
330-11	3.02	a	1.72	3.82	3.94	4.33
330-14	-3.02	1.31	1.74	4.01	4.02	4.37
60-16	-3.02	1.23	1.65	3.82	4.38	4.29
330-18	-9.05	1.23	1.57	3.65	3.58	3.92
330-22	-15.08	a	1.24	2.95	a	3.09

<sup>a</sup> Wire not recovered in hot cell.

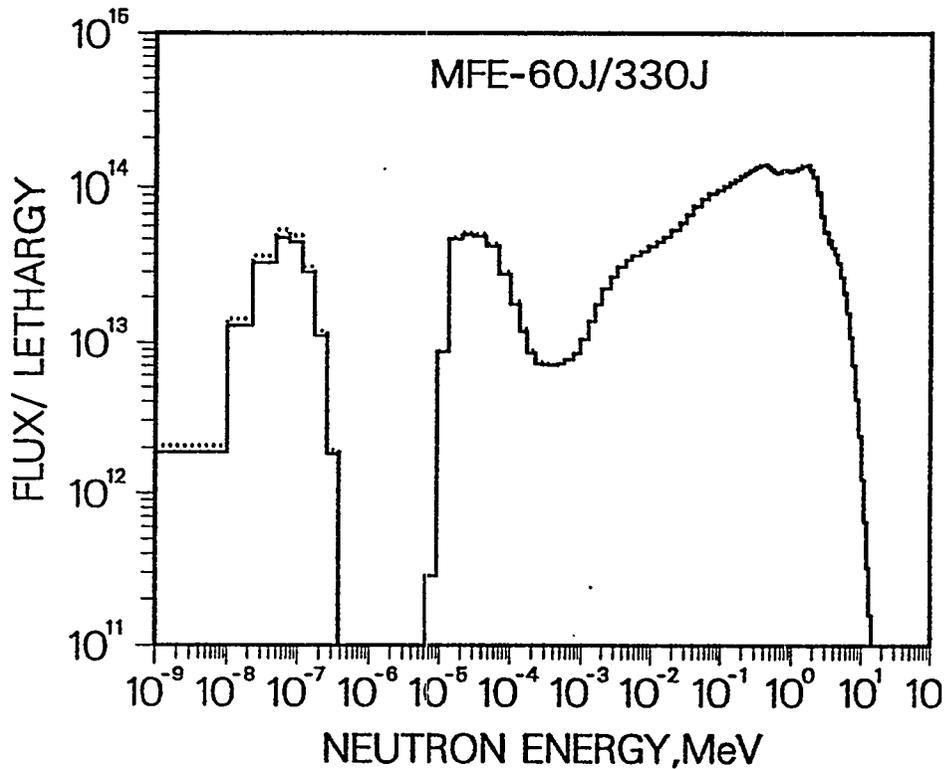


Fig. 1. The STAY'SL adjusted neutron flux spectrum is shown for the hafnium-lined RB\* position in HFIR. The structure at low energies results from the thermal neutron absorption and epithermal neutron resonances in hafnium.

Table 2. Midplane Fluence and Damage Values for MFE-60J/330J Only

Neutron Fluence, n/cm <sup>2</sup>	Element	dpa	He, appm
Total	C	14.1	13.9
Thermal (<.5 eV)	Al	22.5	5.4
0.5 eV - 0.1 MeV	V	14.6	0.18
> 0.1 MeV	Cr	12.4	1.3
> 1 MeV	Fe	11.1	2.4
	Ni Fast	12.3	34.2
	59-Ni	<u>0.5</u>	<u>276.0</u>
	Total	12.8	310.2
	Cu	14.7	2.0

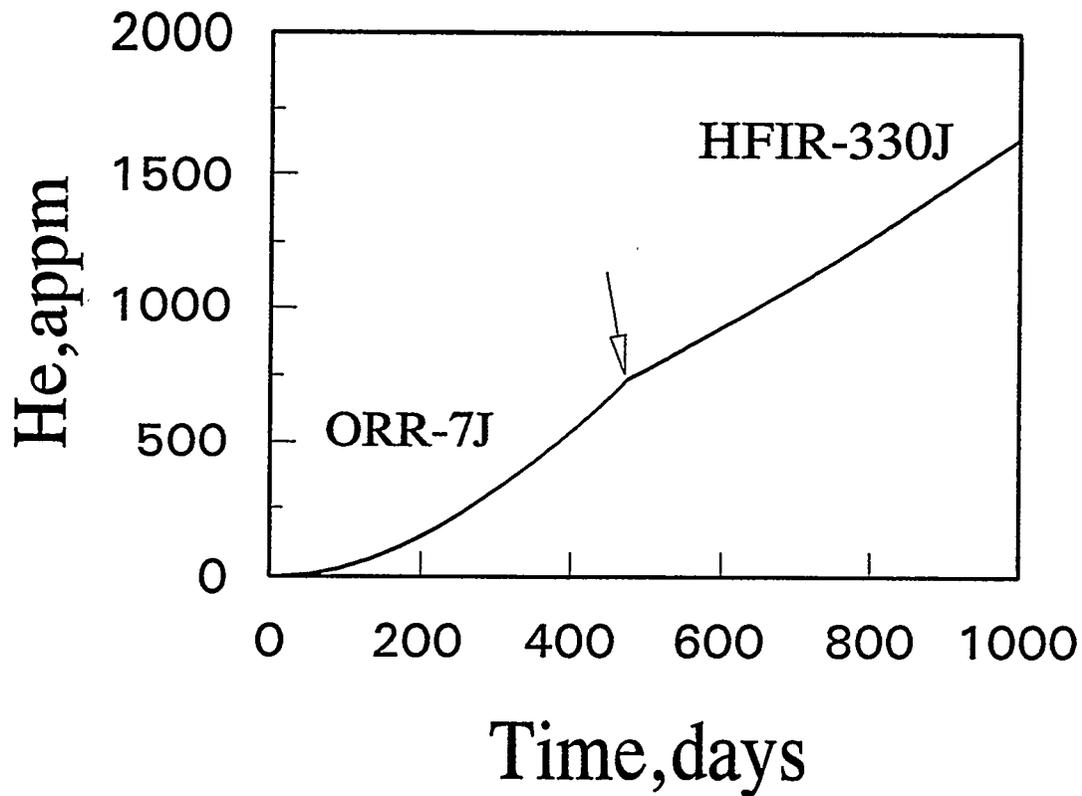


Fig. 2. Helium production in nickel is shown for the ORR-7J and HFIR-330J-1 irradiation sequence as a function of full power days. The arrow indicates the start of the HFIR irradiation.

Table 3. DPA and He Values for 316 SS in MFE-60J/330J only  
(Includes  $^{59}\text{Ni}$  effect)

<u>Ht.cm</u>	<u>dpa</u>	<u>He.appm</u>
0	11.5	42.0
3	11.4	41.3
6	11.0	38.9
9	10.4	35.0
12	9.5	29.9
15	8.4	23.8
18	7.0	17.2

316SS = Fe(0.645), Ni(0.13), Cr(0.18), Mn(0.019), Mo(0.026) wt%

Table 4. Maximum DPA and Helium Production in ORR/HFIR Irradiations  
(Calculations for 316 Stainless Steel)

	<u>dpa</u>	<u>He.appm</u>		<u>dpa</u>	<u>He.appm</u>
ORR-6J	6.9	75.3	ORR-7J	7.4	102.0
HFIR-60J-1	11.6	112.5	HFIR-330J-1	11.6	122.5
Total	18.5	187.8	Total	19.0	224.5

for duplicate analysis. The helium content in each piece was determined by isotope-dilution gas mass spectrometry after vaporization in a resistance-heated graphite crucible in one of the mass spectrometer system's high-temperature vacuum furnaces.<sup>9</sup> The absolute amount of helium released was measured relative to a known quantity of added <sup>3</sup>He spike. The helium spikes were obtained by expanding and partitioning a known quantity of gas through a succession of calibrated volumes.<sup>10</sup> The mass spectrometer was calibrated for mass sensitivity during each series of runs by analyzing known mixtures of <sup>3</sup>He and <sup>4</sup>He.

The average measured helium content was  $250 \pm 1$  appm. To calculate the helium content, it is necessary to consider both irradiations in ORR and HFIR, as illustrated in Table 4 and Figure 2 for 316 stainless steel. A computer program was written to follow the growth of helium through both irradiations for any given combination of sample positions. This net helium calculation yields a value of 286 appm, about 14% higher than the measurement. One uncertainty in the calculation is the boron content of the JPCA specimen. Given the nominal content of 31 wppm (160 appm) and assuming total burnout of the <sup>10</sup>B, then this boron impurity adds about 32 appm helium. Previous measurements by Rockwell International of other stainless steels suggests a much lower boron content, about 1-3 appm, which would provide excellent agreement between the measurement and calculation of helium. Additional helium or boron measurements are needed to resolve this question. Nevertheless, the agreement between measurements and calculations provides some confidence that we can predict helium levels in other materials irradiated in these spectral tailoring experiments.

#### FUTURE WORK

Detailed helium and damage calculations are being performed for each material specimen package in the joint irradiation sequences. Additional experiments still in progress in HFIR include MFE-200J-1 and MFE-400-J1 as well as JP9-16 and JP20-22. Dosimeters from the JP-10, 11, 13, and 16 irradiations are awaiting analysis at PNL. Dosimeters are being fabricated at PNL for the JP-24-25 irradiations.

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