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Prediction of Stainless Steel Activation in Experimental Breeder Reactor 2 (EBR-II) Reflector and Blanket Subassemblies

by

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DISCLAIMER

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INTRODUCTION

Stainless steel structural components in nuclear reactors become radioactive wastes when no longer useful. Prior to disposal, certain physical attributes must be analyzed. These attributes include structural integrity, chemical stability, and the radioactive material content among others. The focus of this work is the estimation of the radioactive material content of stainless steel wastes from a research reactor operated by Argonne National Laboratory.

Experimental Breeder Reactor Number II (EBR-II) is a 62.5 mega-watt thermal (MWt) metal-fueled liquid metal cooled fast neutron spectrum nuclear reactor. EBR-II was operated by Argonne National Laboratory from August 1964 until decommissioning began in September 1994.

Regulatory basis

The next few paragraphs contain a brief discussion of the regulations covering commercially generated radioactive wastes in the United States. The waste classification scheme will be discussed. Following that, a discussion of the impact of these regulations on the United States Department of Energy (DOE).

Title 10 Code of Federal Regulations (CFR) Part 61 Section 55 (10 CFR 61.55) includes two lists of nuclides. ^{14}Carbon, ^{59}nickel, ^{94}niobium, and ^{99}technetium are on the first list. Irradiated stainless steel will have quantities of these four nuclides. The only nuclide on the second list that should be present in irradiated stainless steel is ^{63}nickel.

Determining the waste classification of a specific container of waste is a three step process. The first step is to divide the actual concentration by the concentration limit for each nuclide (these products are termed "fractions" in the regulation and "Activity Ratios" in the "Results" section of this thesis). The "Results" section of this thesis reports the results of this type of calculation. Step two, the fractions are summed for the nuclides in each table. Step three, compare the sum of the fractions to unity. If either sum of the fractions is greater than unity or any one fraction is greater than unity, the waste is GTCC-LLW.

The disposal of irradiated metals that are classified GTCC-LLW has become a major issue for the commercial nuclear power utilities and DOE. Several nuclear power plants have been permanently shut down and decommissioned. Some of the core internals from these reactors have been determined to be GTCC-LLW. DOE was given the responsibility for disposal of GTCC-LLW by the Low-Level Radioactive Waste Policy Amendments Act of 1985 Public Law 99-240. Several DOE owned reactors are also being

decommissioned. This places DOE at both ends of the GTCC-LLW problem: waste generator and waste disposal facility owner.

Storage space for DOE owned GTCC-LLW is being filled rapidly, a disposal facility is required. DOE does not currently have a disposal facility permitted to accept GTCC-LLW or SPAR-LLW. Because the Environmental Impact Statement for the high-level waste deep geologic disposal facility at Yucca Mountain will not address GTCC-LLW, the facility will not accept wastes of this category. The Waste Isolation Pilot Plant (WIPP) located near Carlsbad, New Mexico also rejects GTCC-LLW because these wastes do not meet the waste acceptance criteria.

METHODS AND PROCEDURES

The question to be addressed in this thesis is whether or not EBR-II reflector and blanket hardware exceeds the Class C limits as defined by 10 CFR 61.55. Regarding EBR-II irradiated materials, calculational tools used in the past did not address this question directly. The focus of this thesis is to develop a calculational procedure to assist with the estimation of the activation product content of irradiated EBR-II stainless steel hardware. Chemical analysis of selected samples will be accomplished to prove the procedure accurate.

Direct measurement of the β emitters requires radiochemical analysis (chemical dissolution followed by measurement) due to the β particle self-shielding nature of metal plates. For instance, thin metal foils are used on Geiger-Mueller tubes to shield the tube from β particles. The nuclides of waste disposal concern are primarily β emitters (^{94}niobium is also a γ emitter). Therefore, comparisons between the concentration of an easily measured γ emitter (i.e. ^{60}cobalt) and the β emitting nuclides of interest are made, called 'scaling factors'. The correlation between these concentrations is dependent on the irradiation history, neutron spectrum, and the cooling time. In most light water reactors, the neutron spectrum is a slowly varying function of position. The neutron spectrum varies greatly with position in EBR-II. This complicates the calculation of 'scaling factors' greatly.

The procedure developed for this thesis involves four major steps. Step one, determine the bounding values for the chemical composition of stainless steels used in the fabrication of EBR-II reflector and blanket hardware. This step was accomplished by Carole Trybus [Ref. 1] prior to the start of work on this thesis. The mean and standard deviation for the chemical compositions determined from archival samples along with the chemical compositions input to ORIGEN for SS304 and SS316 are tabulated in Table 1.

Table 1: Chemical Analysis Results of SS304 and SS316 Samples (all compositions are in weight percent)

Element	SS304 μ^*	SS304 σ^{**}	SS304 ORIGEN input	SS316 μ^*	SS316 σ^{**}	SS316 ORIGEN input
Mn	1.41	0.27	2.15	1.70	0.12	1.94
Cr	18.78	0.39	19.56	17.23	0.70	18.63
Co	0.17	0.11	0.39	0.17	0.08	0.33
C	0.07	0.01	0.09	0.06	0.01	0.08
Mo	0.37	0.16	0.69	2.40	0.25	2.90
N	0.047	0.011	0.069	0.047	0.013	0.063
Ni	9.23	0.68	10.59	11.75	0.77	13.29
Cu	0.25	0.15	0.45	0.26	0.18	0.62
Nb	0.012	0.008	0.028	0.005	0.006	0.017
Fe***			65.98			62.13

* average value ** standard deviation

*** The remainder of each sample was iron. The iron composition input to ORIGEN was set such that the total equals 100%.

Step two, calculate an accurate neutron flux using a standard transport theory neutronics code, TWODANT [Ref. 2]. Several models using different reactor composition information, different geometric information, and different scattering orders were modeled to determine the uncertainty in the flux calculation. The reactor composition and geometry information for a recent reactor run was chosen as the model to use in the final analysis.

Step three, collapse the neutron cross sections from an up-to-date multi-energy group library to a single group. Cross sections for the nuclides in stainless steel were generated from ENDF/B Version VI tapes using the NJOY code [Ref. 3]. Weighting fluxes from several locations in the EBR-II blanket were used so that the uncertainty in

the cross sections could be determined. Table 2 provides a comparison between the ORIGEN standard fast reactor cross sections and the high and low values calculated using the COLLAPSE code.

Table 2: ORIGEN cross sections in barns

Nuclide	ORIGEN $(^0n_1,\gamma)$	COLLAPSE lower value	COLLAPSE upper value
^{58}Ni	0.00721	0.015	0.025
^{59}Ni	0.0204	0.02	1.2
^{62}Ni	0.00489	0.015	0.035
^{93}Nb	0.108	0.08	0.28
^{94}Nb	0.846	0.1	0.8
^{98}Mo	0.0432	0.04	0.16
^{99}Tc	0.204	0.2	1.3

Step four, calculate the expected nuclide concentrations using a standard isotope generation and depletion code, Oak Ridge Isotope Generation (ORIGEN) code [Ref. 4]. The CURIES code was used to edit the ORIGEN output and create a tabulated listing of the "Activity Ratios". These "Activity Ratios" are reported in the "Results" section of this thesis.

RESULTS

Detailed calculations were performed using irradiation histories for five stainless steel subassemblies. Thirty-seven axial zones were defined for each subassembly. The results for these subassemblies using both SS304 and SS316 are summarized below. Both SS304 and SS316 calculations were performed because the fabrication instructions for stainless steel reflector subassemblies specify that either type of steel may be used. Each type of steel was analyzed using the unmodified ORIGEN cross sections and the

NJOY cross sections. All of the subassemblies are stainless steel reflectors irradiated in the reflector region or the blanket region.

The "Activity Ratio" (activation product concentration divided by the Class C limit) was calculated for each nuclide in each of the subassemblies. The peak "Activity Ratios" have been tabulated in Table 3 for several subassembly irradiation histories. The irradiation histories are also listed. The suffix "-O" denotes "Activity Ratios" calculated using the cross sections from the ORIGEN library. The suffix "-C" denotes "Activity Ratios" calculated using the cross sections produced using the COLLAPSE code.

Table 3: ORIGEN calculated Peak Activity Ratios

subassembly	irradiation History	^{59}Ni SS304 SS316	^{63}Ni SS304 SS316	^{94}Nb SS304 SS316	^{99}Tc SS304 SS316
U1603-O	349000 MWd row 14	0.07 0.08	0.07 0.08	9.0 7.0	0.1 0.5
U1603-C		0.22 0.28	0.33 0.43	15 9.0	0.3 1.1
U8972-O	134000 MWd row 13 91000 MWd row 9	0.18 0.22	0.19 0.21	25 15	0.4 1.5
U8972-C		0.55 0.72	1.05 1.3	63 38	1.3 5.8
U9006-O	187000 MWd row 10 3344 MWd row 14	0.25 0.30	0.25 0.30	34 22	0.5 2.2
U9006-C		0.8 1.0	1.5 1.8	86 52	1.8 7.8
U9007-O	187000 MWd row 10	0.25 0.30	0.25 0.30	33 21	0.5 2.2
U9007-C		0.75 0.99	1.4 1.8	85 51	1.8 7.6
U9807-O	60000 MWd row 8 107000 MWd row 16	0.15 0.18	0.15 0.18	21 13	0.2 1.3
U9807-C		0.44 0.55	0.75 0.95	45 26	1.0 4.5

Error Analysis

Many avenues exist for errors to intrude into this analysis. The cross sections used in the transport calculations could be in error. The composition files could contain inaccuracies. The transport code input could contain errors directing an incorrect calculation. The 28 group activation cross sections could be incorrect. The FORTRAN codes used to produce the one-group cross sections could be in error. The input file directing the ORIGEN calculations could be in error. The chemical compositions of the stainless steels may not represent the steels used in EBR-II hardware.

The cross sections used in the 28 group transport calculations are used for reactor physics calculations. The 28 group transport calculation proceeds to completion with no unusual output. The resulting k-effective is very close to 1.00. This leads to the conclusion that this calculation is correct. Running the same model with a higher order scattering cross section library produces a similar value for k-effective while providing a much harder neutron spectrum (higher percentage of fast neutron flux) in the blanket. At best, the neutron flux calculation can produce results within 10% of actual irradiation conditions.

The extreme lower portion of the reactor, including the grid plate was not modeled, nor was the extreme upper portion of the reactor. This facet of the model will introduce some inaccuracies into the calculation. Far from the top and bottom edge, the results should not be effected. A TWODANT model that included an additional twenty cm of material added to the top and bottom of the Run 168 model was analyzed to explore this problem. The results from this model follow the results from the Run 168 model except within 15 cm of the top and bottom of the smaller model. The differences in the results from the two models away from the upper and lower boundaries were less than the uncertainty in the chemical composition.

The activation cross sections produced using the NJOY code are dependent on the input weighting function. In order to determine the effect this weighting function has on the cross sections, an additional cross section set was produced using the Row 14 reactor midplane flux as a weighting function. Below 1 KeV, the weighting

functions are greatly divergent. Above this energy the weighting functions are similar.

The effects of the two weighting functions on the cross sections were analyzed by running the ORIGEN calculation of SS304 in subassembly U1603. The activity ratios were quite similar. The use of a neutron flux data set from a P3 transport calculations offers lower activity ratios by a factor of 2. The one-group cross sections are largely unaffected by the weighting function used, providing that it is from the region of interest. Clearly, a weighting function from Row 1 would not be appropriate for use in the blanket. Further, these figures point out a need for continued analysis of the blanket. Higher order scattering appears to significantly affect the flux calculation in the blanket.

The largest errors are in the chemical compositions input to ORIGEN. The standard deviation, for SS304, of the niobium concentration is 66% of the mean value and for SS316, it is 100%. The chemical compositions vary by as much as a factor of 25 from the smallest to the mean plus two standard deviation value used in the ORIGEN calculation. The largest Activity Ratio calculated is near 80, so the subassembly will still be GTCC-LLW even if the starting chemical composition is near the smallest value. Smaller values for the starting chemical composition will allow more of the subassembly to be segregated into different waste streams for disposal. Higher values will diminish the benefit of waste stream segregation.

CONCLUSIONS

The results presented in the previous section point to a significant problem. EBR-II irradiated stainless steel hardware exceeds the 10 CFR 61.55 Class C limits for near surface waste disposal. The amount by which the hardware exceeds the limit is perhaps not as great as shown in Table 3 due to the conservatism built into the calculations. However, even if the starting concentration of niobium is reduced to the mean value determined by the chemical analysis, the maximum value of the ratio of the calculated concentration to the allowed will be reduced by a factor of 3. But, this value would still far exceed the Class C limit.

The collapsed cross sections produced for this thesis are very different than those in the ORIGEN library. Clearly the cross sections used in a calculation of this type heavily influence the results. The neutron spectrum in EBR-II changes dramatically from the inner core to the blanket. Cross sections collapsed using an inner core neutron spectrum will not be valid in the blanket.

What effect does the calculation of reactor power have on this thesis? The uncertainty in the thermal power calculations for EBR-II is estimated to be 5% and perhaps it is as great as 10%. Assuming that the flux is 10% lower over the entire operating lifetime of EBR-II, the ratio calculated in this thesis will still exceed 1.00. The stainless steel hardware will still exceed the limit.

FUTURE WORK

The current implementation of the cross section collapsing code does not account for neutron flux differences between depleted uranium subassemblies and steel reflector subassemblies. A detailed three-dimensional model could be constructed to perform this calculation. Two-dimensional diffusion or transport calculations in the axial plane could also be accomplished. Preliminary studies indicate that the neutron flux is somewhat softer in reflector subassemblies than in depleted uranium subassemblies. The magnitude of this flux difference should be investigated.

The Analytic Laboratory will conduct chemical analysis on several hex duct specimens cut from a row 10 reflector subassembly in the near future (FY 1997). This analysis will prove the validity of the calculational procedure presented in this thesis. Chemical analysis of other specimens may be warranted by the results of the first set of chemical analysis. The decommissioning of EBR-II will provide a large number of stainless steel specimens for analysis that would not have been available if the reactor were still in operation.

1 C. L. Trybus, personal communication, April 25, 1995.

2 R. E. Alcouffe, R. S. Baker, F. W. Brinkley, D. R. Marr, R. D. O'Dell, and W. F. Walters, 'DANTSYS: A Diffusion Accelerated Neutral Particle Transport Code System', LA-12969-M, Los Alamos National Laboratory, Los Alamos, New Mexico, June 1995. Distributed as RSIC Computer Code Collection CCC-547 by the Radiation Shielding Information Center at Oak Ridge National Laboratory, Oak Ridge, Tennessee.

3 R. E. MacFarlane, D. W. Muir, and R. M. Boicourt, 'The NJOY Nuclear Data Process System', LA-9303-M (ENDF-324), Los Alamos National Laboratory, Los Alamos, new Mexico, May 1982.

4 M. J. Bell, 'ORIGEN - The ORNL Isotope Generation and Depletion Code', ORNL-4628, Oak Ridge National Laboratory, Oak Ridge, Tennessee, May 1973.