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Reevaluation of HFIR Source Term

W. E. Thomas

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MARTIN MARIETTA ENERGY SYSTEMS, INC.
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DEPARTMENT OF ENERGY**

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National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
NTIS price codes—Printed Copy: A02 Microfiche A01

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Operations Division

REEVALUATION OF HFIR SOURCE TERM

Supplement 2

W. E. Thomas

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MASTER

ORNL-6324
(Supplement No. 2 to ORNL-3573)
Dist: Category UC-80

Operations Division

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W. E. Thomas

Manuscript Completed - August 1986
Date Published - November 1986

NOTICE: This document contains information of a preliminary nature. It is subject to revision or correction and therefore does not represent a final report.

Prepared for the
Office of Basic Energy Sciences

Prepared by the
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37831
operated by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-84OR21400

CONTENTS

	<u>Page</u>
Abstract	1
Introduction	1
Fuel Assembly Loadings	1
Cycle Time and Computational Time Steps	1
Nuclide Microscopic Cross Sections	2
Results	3
Iodine Activities and Dose	4
Noble Gas Activities and Dose	5
Other Fission Products	8
Conclusions	11
References	12
Distribution	13

LIST OF TABLES

<u>Table</u>	<u>Page</u>
1 HFIR initial fuel assembly loadings	2
2 One-group microscopic cross sections (barns).	3
3 Activities used for iodine dose calculation (curies).	5
4 Approximate change in thyroid dose.	5
5 Activities used to compute external dose from noble gasses (curies)	6
6 Approximate change in external dose from direct-emission nuclides.	7
7 Activities used to compute internal dose due to fission products other than iodine (curies)	8
8 Boneseekers, approximate change in dose	9
9 Other activities used to compute internal dose due to fission products other than iodine (curies)	10
10 Others, approximate change in dose.	10

REEVALUATION OF THE HFIR SOURCE TERM

W. E. Thomas

ABSTRACT

The HFIR source term has been reevaluated to assess the impact of the increase in core lifetime from 15 to 24 days. Calculations were made to determine the nuclide activities of the iodines, noble gasses, and other fission products. The results show that there is no significant change in off-site dose due to the increased fuel cycle for the release scenario postulated in ORNL-3573.

INTRODUCTION

The contents of this document are a supplement to the original reactor safety analysis reported in ORNL-3573⁽¹⁾. The postulated fission product release scenarios in that document were based on nuclide activity from fuel assemblies irradiated for the designed 15-day operation at full power. Results from those analyses showed that the airborne dose to the thyroid from the iodine isotopes and the whole-body dose from the noble gasses were within existing federal limits.

The average HFIR core lifetime is on the order of 21 full-power days, thus, a reevaluation of the nuclide activity was undertaken. This report presents the details of the calculations and the results. For simplicity, all subsequent references to reactor operating time (in days) are defined as full-power days of operation.

FUEL ASSEMBLY LOADINGS

Typical HFIR fuel assembly loadings are shown in Table 1 and are based on data from the Babcock and Wilcox manufacturing inspection reports together with the necessary supplemental data from fuel assay reports.⁽²⁾ Two typical fuel assemblies were chosen to illustrate the range of values expected.

CYCLE TIME AND CALCULATIONAL TIME STEPS

A review of the records indicates that HFIR has achieved a maximum cycle time of 24 days. As a basis for the study, an optimum 11-time-step ORIGEN-2 calculation was chosen with the nuclide activities evaluated at the end of 1, 2, 5, 7, 10, 13, 15, 17, 19, 21, and 24 days. The ORIGEN-2 calculation expands the flux in a Taylors series through the second-order terms and integrates the expression over a time step to obtain an average neutron flux.⁽³⁾ Even with this degree of sophistication, some variation in nuclide activity will be observed if very coarse time steps are used. Eleven time steps were considered sufficient to minimize the associated flux error.

Table 1. HFIR initial fuel assembly loadings

	Fuel assembly identification number	
	293	298
A. From inspection reports:		
Grams of ^{235}U	9427.658	9466.356
^{235}U Enrichment	0.93168	0.93167
$\text{U}/\text{U}_3\text{O}_8$ (wt. fraction)	0.84603	0.84601
Inner element only:		
Grams of ^{10}B	2.7166	2.7143
^{10}B Isotopic (wt. fraction)	0.1852	0.1852
$\text{B}/\text{B}_4\text{C}$ (wt. fraction)	0.7787	0.7787
B. Breakdown for ORIGEN-2 calculations (g):		
Oxygen	1841.566	1849.429
Carbon	4.1687	4.165
$^{234}\text{U}^*$	101.625	102.058
^{235}U	9427.658	9466.356
$^{236}\text{U}^*$	42.172	42.351
$^{238}\text{U}^*$	547.532	549.866
^{10}B	2.7166	2.7143
^{11}B	11.9519	11.9417

*Distribution based on fuel assay reports.

NUCLIDE MICROSCOPIC CROSS SECTIONS

AMPX⁽⁴⁾ weighted cross sections, generated for a HFIR fuel assembly, were used for the actinides and ^{135}Xe . ORIGEN-2 (PWR) cross sections were used for the remaining fission products. HFIR "cell-weighted" cross sections for the actinides are considerably larger than the PWR cross sections.

To test the applicability of a mixed library of cross sections, we compared the fission-product activities with an identical ORIGEN-2 case executed with an all-PWR library. The ORIGEN-2 group neutron flux was about a factor of four lower with the mixed library. Only two fission products, ^{135}Xe and ^{105}Rh , which have very high cross sections, showed a significant difference in the 15-day activities. Since HFIR cross sections are used for ^{135}Xe , only the ^{105}Rh activity can have a significant

error. PWR fuel rods are more "self-shielded" than HFIR fuel plates. This means that the ^{105}Rh capture reaction rate will be low, which will cause the activity to be high and, hence, in the conservative direction. Activities for almost all of the remaining 35 fission products of interest differed by <0.1%. The mixed library of cross sections is, therefore, considered adequate for HFIR safety analysis work. HFIR cross sections for ^{135}Xe and the actinides are shown in Table 2.

Table 2. One-group microscopic cross sections (barns)*

Nuclide	σ_f	$\sigma(n, \gamma)$	$\sigma(n, 2N)$	$\sigma(n, 3N)**$
^{234}U	0.294	42.28	1.67E-4	1.345E-5
^{235}U	189.64	35.43	1.77E-3	1.16E-6
^{236}U	0.234	12.66	8.8E-4	2.21E-5
^{238}U	0.037	10.683	1.77E-3	4.597E-5
^{135}Xe	--	8.44E+5	--	--

*Collapsed from XSDRNPM(4) 4-Gp cross sections for the fuel element region of the HFIR reactor.

**ORIGEN-2 PWRU cross sections.

RESULTS

Nuclide activities at the end of a 15-day operation are given in various tables of Reference 1.

Two activity ratios, called Ratio 1 and Ratio 2, will be used here to compare results and indicate the magnitude of activity change due to extended operation (beyond 15 days) of HFIR. Ratio 1 is the ORIGEN-2 activities divided by the corresponding activities from ORNL-3573 after 15 days of operation. Hence, Ratio 1 is a measure of calculational agreement. Ratio 2 is the ORIGEN-2 activity after 24 days divided by ORIGEN-2 activity after 15 days. Therefore, this ratio is a consistent measure of nuclide activity change due to longer burnup.

Total activities are also given in each table. "Net change" is defined as the ORIGEN-2 total activity at the end of 24 days divided by total activity for the nuclide group reported in ORNL-3573 at the end of 15 days of operation. This parameter is a measure of the overall change in group activity.

IODINE ACTIVITIES AND DOSE

Activities for the five isotopes of iodine used in the dose calculation were reported in Table 1.A.2 of ORNL-3573.(1) Those activities, reported in units of curies/kW have been converted to curies and listed in Table 3. Notice that the total activity (for the group) at 15 days of operation differed by only 0.3%. The net change for the group of five isotopes shows only a 2% increase due to extended HFIR operation.

The greatest change in nuclide activity for a single nuclide is associated with ^{131}I . The ORIGEN-2 activity is about 7% larger than those reported in ORNL-3573 at 15 days, and the ratio of ORIGEN-2 activities (Ratio 2) indicates that extended operation might increase the ^{131}I activity by about 20%.

Parameters which affect the thyroid dose are: (1) downwind distance to the recipient, (2) activity release rate from the stack, (3) windspeed, (4) meteorological conditions, (5) stack height, (6) breathing rate, (7) nuclide decay constant, (8) time, (9) dose conversion factor, and (10) specific activity. With all other conditions equal, the thyroid dose is directly proportional to the product of specific activity, dose conversion factor, and a time-dependent constant summed over appropriate nuclides. The constant accounts for air dilution and nuclide decay following an accident. Therefore, if one assumes an infinite exposure and a rapid release rate and neglects the nuclide decay during travel to a receptor, then the thyroid dose becomes proportional only to the sum of the product of nuclide specific activity and dose conversion factor.(5) Using this method, the net change in thyroid dose due to extended operation of the HFIR is illustrated in Table 4. Again, the most significant single-nuclide change is associated with ^{131}I . The approximately 13% dose increase for the group of five iodine isotopes is well within the accuracy of any off-site estimates; therefore, operating HFIR for 24 days (rather than 15 days) is not expected to significantly increase the thyroid dose during accident conditions nor cause the thyroid dose to exceed 10CFR100 limits(6) for the accident conditions postulated in Reference 1.

Table 3. Activities used for iodine dose calculation (curies)

Nuclide	15-day operation			24-day operation	
	ORNL-3573*	ORIGEN-2	Ratio 1	ORIGEN-2	Ratio 2
¹³¹ I	1.62E+6	1.729E+6	1.07	2.095E+6	1.21
¹³² I	3.711E+6	3.513E+6	0.95	3.627E+6	1.03
¹³³ I	5.483E+6	5.563E+6	1.01	5.541E+6	0.996
¹³⁴ I	6.411E+6	6.280E+6	0.98	6.204E+6	0.99
¹³⁵ I	4.977E+6	5.202E+6	1.05	5.128E+6	0.99
Total	2.2202E+7	2.2287E+6	1.003	2.2595E+7	1.01

24-day net change (relative to ORNL-3573) = 1.02

*Table 1.A.2 of Reference 1.

Table 4. Approximate change in thyroid dose

Nuclide	15-day operation			24-day operation		
	$\mu\text{Ci} \times 10^{-12}$ *	rem/ μCi	rem $\times 10^{-12}$	$\mu\text{Ci} \times 10^{-12}$ **	rem/ μCi	rem $\times 10^{-12}$
¹³¹ I	1.62	x 1.484 =	2.404	2.095	x 1.484 =	3.109
¹³² I	3.711	x 0.054 =	0.200	3.627	x 0.054 =	0.196
¹³³ I	5.483	x 0.399 =	2.188	5.541	x 0.399 =	2.211
¹³⁴ I	6.411	x 0.025 =	0.160	6.204	x 0.025 =	0.155
¹³⁵ I	4.977	x 0.123 =	<u>0.612</u>	5.128	x 0.123 =	<u>0.631</u>
Totals			5.564			6.302

Dose, Net change 6.302/5.564 = 1.13

*From Table 1.A.2 of Reference 1.

**Calculated using ORIGEN-2.

NOBLE GAS ACTIVITIES AND DOSE

A comparison of ORIGEN-2-generated activities with those reported in ORNL-3573 for the 10 noble gas nuclides of interest is given in Table 5. Activities of the three daughter nuclides are also compared in Table 5.

We note first that there is almost no change in total activity for the group of 10 direct-emission (parent) nuclides nor the three daughter nuclides due to 24-day operation of the HFIR reactor. An inspection of the values for Ratio 1 shows the largest disagreements at 15 days are associated with the metastable nuclides. The cause for this is not known. It is not due to the nuclide decay constant, nor does it appear to be caused by a difference in capture cross sections. However, it may be associated with a difference in the branching ratio data.

There are two reasons for the 15% reduction in ^{135}Xe activity at the higher burnup. First, equilibrium occurs very early in the cycle because of its high cross section; and second, the progressive fuel depletion with associated time-dependent neutron flux increase will affect the nuclide loss reaction rate.

The expected change in external gamma dose may be estimated by comparing the sum of the products of nuclide activity and average gamma energy per transition as shown in Table 6. The dominant contributor is ^{88}Kr because of its energetic gamma spectrum.

The data in Tables 5 and 6 support the conclusion that there is essentially no change in noble gas activity or off-site dose caused by 24-day operation of the HFIR reactor.

Table 5. Activities used to compute external dose from noble gasses (curies)

Nuclide	15-day operation			24-day operation	
	ORNL-3573*	ORIGEN-2	Ratio 1	ORIGEN-2	Ratio 2
Direct emission (curies):					
^{84}Kr	4.05E+5	4.431E+5	1.09	4.376E+5	0.99
^{86}Kr	1.265E+6	1.048E+6	0.83	1.035E+6	0.99
^{87}Kr	2.277E+6	2.120E+6	0.93	2.093E+6	0.99
^{88}Kr	3.121E+6	2.997E+6	0.96	2.960E+6	0.99
^{89}Kr	3.88E+6	3.801E+6	0.98	3.754E+6	0.99
^{133}Xe	1.32E+5	1.622E+5	1.23	1.646E+5	1.01
^{133}Xe	4.718E+6	4.654E+6	0.99	5.292E+6	1.14
^{135}Xe	1.518E+6	9.414E+5	0.62	9.284E+5	0.99
^{135}Xe	1.6E+5	1.598E+5**	0.999	1.366E+5**	0.85
^{138}Xe	4.639E+6	5.145E+6	1.11	5.082E+6	0.99
Totals	2.2115E+7	2.1472E+7	0.97	2.18832E+7	1.02

24-day net change (relative to ORNL-3573) = 0.99

Table 5 (continued)

Daughters (curies):					
⁸⁸ Rb	3.121E+6	2.997E+6	0.96	2.960E+6	0.99
⁸⁹ Rb	3.88E+6	3.801E+6	0.98	3.754E+6	0.99
¹³⁸ Cs	4.639E+6	5.145E+6	1.11	5.082E+6	0.99
Totals	1.164E+7	1.1943E+7	1.026	1.1796E+7	0.99
24-day net change (relative to ORNL-3573) = 1.013					

*Table 1.A.4 of Reference 1.

**Using HFIR cross-sections.

Table 6. Approximate change in external γ dose from direct-emission nuclides

Nuclide	15-day operation			24-day operation		
	$\mu\text{Ci} \times 10^{-12}$	$\bar{E}\gamma$ MeV/dis*	$\mu\text{Ci}(\text{MeV/dis}) \times 10^{-12}$	$\mu\text{Ci} \times 10^{-12}$	$\bar{E}\gamma$ MeV/dis	$\mu\text{Ci}(\text{MeV/dis}) \times 10^{-12}$
^{83m} Kr	0.405	0.415	0.168	0.437	0.415	0.181
^{85m} Kr	1.265	0.181	0.229	1.035	0.181	0.187
⁸⁷ Kr	2.277	0.56	1.275	2.09	0.56	1.170
⁸⁸ Kr	3.121	2.07	6.460	2.96	2.07	6.127
⁸⁹ Kr	3.88	0	0	3.754	0	0
^{133m} Xe	0.132	0.237	0.031	0.165	0.237	0.039
¹³³ Xe	4.718	0.081	0.382	5.292	0.081	0.429
^{135m} Xe	1.518	0.528	0.801	0.928	0.528	0.490
¹³⁵ Xe	0.16	0.268	0.043	0.137	0.268	0.037
¹³⁸ Xe	4.639	0	0	5.082	0	0
Totals			9.389			8.660

Dose change $\sim 8.66/9.389 = 0.92$

*From Table 1.A.4 of Reference 1.

OTHER FISSION PRODUCTS

There are two other categories of fission products labeled "bone-seekers" and "others," listed in Table 1.A.3 of ORNL-3573(1). The 11 "bone-seekers" and eight "other" nuclides were used to calculate an other-than-iodine internal dose in Reference 1.

ORIGEN-2-generated activities for the "bone-seekers" are compared with those from Table 1.A.3 in Table 7 of this report. With the exception of ^{89}Sr , the 15-day ORIGEN-2 activities are all lower than those in ORNL-3573.

Comparing ORIGEN-2 activities at 24 days with the ORIGEN-2 activities at 15 days (Ratio 2) indicates a significant (~28%) increase in the total group activity due to the longer cycle time. However, if the total activity for the group of nuclides at 24 days (ORIGEN calculation) is compared with the total group activity reported in ORNL-3573, then only a modest (<10%) increase is noted.

Using the sum of the product of activity and dose conversion factor as an index of the change in dose (shown in Table 8), about a 40% increase is expected for this group of fission products.

Table 7. Activities used to compute internal dose due to fission products other than iodine (curies)

Nuclide	15-day operation			24-day operation	
	ORNL-3573*	ORIGEN-2	Ratio 1	ORIGEN-2	Ratio 2
^{89}Sr	6.94E+5	7.448E+5	1.07	1.124E+6	1.51
^{90}Sr	5.1E+3	4.681E+3	0.92	7.487E+3	1.60
^{91}Y	8.16E+5	7.770E+5	0.95	1.199E+6	1.54
^{95}Zr	8.21E+5	8.042E+5	0.98	1.228E+6	1.53
^{97}Zr	5.23E+6	4.877E+6	0.93	4.847E+6	0.99
^{95}Nb	1.387E+6	1.106E+5	0.08	2.591E+5	2.34
^{140}Ba	3.095E+6	2.890E+6	0.93	3.779E+6	1.31
^{141}Ce	1.404E+6	1.327E+6	0.95	1.952E+6	1.47
^{143}Pr	2.779E+6	2.379E+6	0.86	3.307E+6	1.39
^{144}Ce	1.78E+5*	1.628E+5	0.91	2.576E+5	1.58
^{147}Nd	1.321E+6	1.152E+6	0.87	1.463E+6	1.27
Totals	1.773E+7	1.523E+7	0.86	1.942E+7	1.28

24-day net change (relative to ORNL-3573) = 1.095

*Table 1.A.3 of Reference 1.

Table 8. Boneseekers, approximate change in dose

Nuclide	15-day operation			24-day operation		
	$\mu\text{Ci} \times 10^{-12}$	rem/ μCi	rem $\times 10^{-12}$	$\mu\text{Ci} \times 10^{-12}$	rem/ μCi	rem $\times 10^{-12}$
⁸⁹ Sr	0.694	0.416	0.289	1.124	0.416	0.468
⁹⁰ Sr	0.0051	44.3	0.225	0.0075	44.3	0.332
⁹¹ Y	0.816	0.337	0.275	1.199	0.337	0.404
⁹⁵ Zr	0.821	0.062	0.051	1.228	0.062	0.076
⁹⁷ Zr	5.23	0.004	0.021	4.847	0.004	0.019
⁹⁵ Nb	1.387	0.012	0.017	0.259	0.012	0.003
¹⁴⁰ Ba	3.095	0.090	0.279	3.779	0.090	0.340
¹⁴¹ Ce	1.404	0.019	0.027	1.952	0.019	0.037
¹⁴³ Pr	2.779	0.023	0.064	3.307	0.023	0.076
¹⁴⁴ Ce	0.178	1.200	0.214	0.258	1.200	0.310
¹⁴⁷ Nd	1.321	0.189	0.250	1.463	0.189	0.277
Totals			1.712			2.342
Change in dose = 2.342/1.712 = 1.37						

The last category of nuclides, the "others," are compared in Table 9. ORIGEN-2 activities at 15 days were much lower than those reported in ORNL-3573. The principal discrepancies were associated with the Te isotopes. Note also that the ORIGEN-2 activities at 24 days for the group of nuclides are actually less than those reported in ORNL-3573 at 15-day operation (net change less than unity).

With the above trend in activity, the dose is also expected to be lower as shown in Table 10. Again, it is emphasized that the "dose" given in Tables 4, 6, 8, and 10 is not a true dose but simply a means of estimating expected change in dose at 24 days of operation relative to those reported in ORNL-3573.

Table 9. Other Activities used to compute internal dose due to fission products other than iodine (curies)

Nuclide	15-day operation			24-day operation	
	ORNL-3573*	ORIGEN-2	Ratio 1	ORIGEN-2	Ratio 2
¹⁰³ Ru	5.49E+5	6.125E+5	1.12	9.089E+5	1.48
¹⁰⁵ Rh	7.59E+5	6.709E+5	0.88	6.506E+5	0.97
¹⁰⁶ Ru	9.0E+3	9.738E+3	1.08	1.547E+4	1.59
¹²⁷ Te	2.3E+4	1.12E+3	0.05	2.06E+3	1.84
¹²⁹ Te	2.27E+5	2.237E+4	0.10	3.301E+4	1.47
¹³¹ Te	2.246E+6	3.033E+5	0.14	3.026E+5	0.998
¹³² Te	3.565E+6	3.429E+6	0.96	3.553E+6	1.04
¹³⁷ Cs	5E+3	4.855E+3	0.97	7.766E+3	1.60
Totals	7.383E+6	5.0538E+6	0.68	5.4734E+6	1.08

24-day net change (relative to ORNL-3573) = 0.74

*Table 1.A.3 of Reference 1.

Table 10. Others, approximate change in dose

Nuclide	15-day operation			24-day operation		
	$\mu\text{Ci} \times 10^{-12}$	rem/ μCi	rem $\times 10^{-12}$	$\mu\text{Ci} \times 10^{-12}$	rem/ μCi	rem $\times 10^{-12}$
¹⁰³ Ru	0.549	0.007	0.0038	0.9089	0.007	0.0064
¹⁰⁵ Rh	0.759	0.0014	0.0011	0.6506	0.0014	0.0009
¹⁰⁶ Ru	0.009	0.065	0.0006	0.01547	0.065	0.00101
¹²⁷ Te	0.023	0.018	0.0004	0.00206	0.018	0.00004
¹²⁹ Te	0.227	0.046	0.0104	0.03301	0.046	0.0015
¹³¹ Te	2.246	0.0077	0.0173	0.3026	0.0077	0.0023
¹³² Te	3.565	0.0015	0.0053	3.553	0.0015	0.0053
¹³⁷ Cs	0.005	0.0086	0.00004	0.007766	0.0086	0.00007
Totals			0.03894			0.01752

Change in dose = $0.01752/0.03894 = 0.45$

CONCLUSIONS

Operating the HFIR at 24 full-power days as opposed to 15 full-power days has the following effect on the groups of fission products of interest to a safety analysis:

1. Iodine activities for the group of five isotopes remain essentially unchanged.
2. Since the reported off-site thyroid dose was significantly lower than the 10CFR100 limit, a small change in iodine dose will not cause a problem. Also, the estimated change in dose lies well within the accuracy associated with atmospheric dispersion model calculations.
3. Noble gas activities and activities for the daughter nuclides also remain essentially unchanged. Thus, the dose for this nuclide group is not expected to increase.
4. Activities for the solid nuclides (bonesearchers) do increase with increased fuel burnup. However, compared with the activities reported in ORNL-3573, the increase should be less than ~10% for the group. Deposition of these solid nuclides will be largely confined to the federal reservation and will have a negligible effect on the off-site areas.
5. The ORIGEN-2 activities at 24 days of operation for the group of eight "other" solid nuclides are lower than that reported in ORNL-3573. The principal difference is associated with the Te isotopes. This would indicate that there could be a dose reduction for these nuclides.

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