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WANL-TME-650
JANUARY 1964

RELEASE OF FISSION PRODUCTS FROM WANL CAPSULE FP-4

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February 4, 1964

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NASA Lewis Research Center
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Cleveland 35, Ohio

Subject: WANL-TME-650, "Release of Fission Products from WANL Capsule
FP-4", dated January 20, 1964

Dear Mr. Schroeder:

Transmitted herewith are three (3) copies of the subject report.

This report is transmitted for your information.

Respectfully,

H. F. Faught
Program Manager
NERVA Nuclear Subsystem

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19

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RELEASE OF FISSION PRODUCTS FROM WANL CAPSULE FP-4

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TABLE OF CONTENTS

<u>Section</u>	<u>Title</u>	<u>Page</u>
1.0	Introduction	1
2.0	Experimental	1
2.1	Material	1
2.2	Irradiations	2
2.3	Radiochemical Analysis	2
2.4	Post Irradiation Heat Anneal	3
2.5	Calculation of Release Fractions	3
3.0	Results and Discussion	5
3.1	Release during Steady State Irradiation	5
3.2	Release during Post Irradiation Heat Anneal	6
3.3	Release during Transient irradiation in TREAT	8
4.0	Conclusions	14
	Bibliography	16

LIST OF TABLES

<u>Table No.</u>	<u>Title</u>	<u>Page</u>
1	Fractional Release of Fission Nuclides during irradiation in GETR	7
2	Fractional Release of Fission Isotopes During Post Irradiation Heat Anneal	9
3	Arrangement of Elements to Boiling Point and Diffusion Rate	10
4	Change in Initial Cross Activity in NERVA Fuel as a Result of Transient	11
5	Fractional Release of Fission Nuclides during transient irradiation in TREAT	13

1.0 INTRODUCTION

This report describes the release of fission products from type II, beaded NERVA fuel irradiated in capsule WANL FP-4 in conjunction with the fission product diffusion program. The experiments with the fuel samples in this capsule were performed with three main objectives in view:

1. A determination of the fractional release of fission products from the fuel during irradiation at elevated temperatures
2. A determination of the fractional release of fission products during post irradiation heat anneal experiments.
3. A determination of fractional release of fission products during transient irradiations in the TREAT reactor.

The data obtained from post irradiation heat anneals is necessary for determining basic diffusion parameters which can be utilized to formulate a mathematical model which will be incorporated into a modified fission product inventory code. Fractional releases of the various fission products during irradiation will serve to corroborate release from the operating reactor predicted by the fission product inventory code. Release data obtained during transient irradiations in TREAT will be utilized in predicting the release of fission products during NERVA reactor excursions.

2.0 EXPERIMENTAL

2.1 MATERIAL

The material used in this study consisted of 12, cylindrical samples of Type II, beaded NERVA fuel cut from a NERVA B-4 fuel element. As such, they consisted of 20 Wt% UC_2 in the form of 100 μ diameter beads, encased in 25 μ

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thick pyrocarbon coat and dispersed throughout a graphite lattice. The U^{235} enrichment of this fuel was 93%. Sample dimensions at the time of GETR irradiation were 0.25" long by 0.25" in diameter with a 0.095" hole located axially within the cylinder. Prior to the post irradiation heat anneal, the samples were cut in half to yield 24 samples with dimensions 0.25" in diameter by 0.125" in length.

2.2 IRRADIATIONS

Capsule WANL-FP-4 was irradiated on July 2, 1963. Cobalt flux monitor wires located at the top and bottom of the capsule indicated flux levels of 0.995×10^{14} and 1.14×10^{14} n/cm²-sec in these respective positions during the 15 minute irradiation. A temperature of 2200°C, as determined by a Tungsten 26% Rhenium Tungsten thermocouple, was achieved during irradiation. In addition to the GETR irradiation six of the "half" samples were subjected, in pairs, to transient irradiation energies of 177, 451, and 806 megawatt seconds in the TREAT reactor on September 3, 1963. These energies pertain to the TREAT reactor; equivalent NERVA fuel energies would be greater perhaps as much as a factor of twelve. Temperatures achieved during the respective irradiations were 1250, 2200 and $\approx 3000^\circ\text{C}$. Each irradiation was accomplished in approximately 1 second.

2.3 RADIOCHEMICAL ANALYSIS

In order to determine the initial concentration of various isotopes in the twelve samples irradiated in GETR each sample was cut in half, and the half sample dissolved and analyzed for Sr^{89} , Ba^{140} , Ce^{144} , Y^{91} , Zr^{95} and Cs^{137} . After TREAT irradiations, analyses were made on the test samples for these nuclides, as well as for Mo^{99} . In addition, gross γ activity measurements

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were made on all 24 samples prior to dissolution of the 12 control samples and a further heating or TREAT irradiation of the test samples.

2.4 POST IRRADIATION HEAT ANNEAL

Four of the test samples which had been irradiated in GETR were heated in an induction vacuum furnace described previously⁽¹⁾ at a temperature of 2000°C at time intervals varying from 15 to 60 minutes. All runs were made in a vacuum of 10^{-4} mmHg.

2.5 CALCULATION OF RELEASE FRACTIONS

In order to determine the fractional release of a nuclide during the various tests it is necessary to know the initial concentration of the nuclide, C_0 , and the concentration of the nuclide after the test, C_f with the appropriate decay correction back to the time of the test. The fraction of the nuclide retained is C_f/C_0 , and the fraction released is given by the expression $f = 1 - C_f/C_0$. Determination of C_f is straightforward by radiochemical analysis after sample dissolution. The determination of C_0 is somewhat more complex and values of C_0 are determined by different methods according to the particular test performed.

Designating the initial concentration of isotope, i , C_G , from the GETR irradiation, a value of C_G for the post irradiation heat anneal may be obtained by analysis of a control sample irradiated concurrently with the test samples. If the concentration of isotope i in the control sample is C_G^i , the gross activity of the control sample as A_c and the gross activity of the test sample as A_T , C_G is found from the simple relationship

$$C_G = C_G^i A_T / A_c$$

The determination of the amount of a nuclide, i , released during

steady state irradiation in GETR requires a knowledge of the concentration of that isotope formed by the irradiations, C_s , and the concentration of that isotope after the irradiation is complete, C_f . The value of C_s is the result of two processes; the direct fission yield, and the decay of the precursors of that isotope. A value of C_s for any isotope is easily obtained by the product of the number of fission and the chain yield of that isotope. As before, the fraction released during irradiations is $1 - C_f/C_s$ with C_s replacing C_o in the expression defined above. It is apparent that the release fraction for a nuclide determined by this method describes release through a mechanism which involves not only the nuclide of interest, but also the precursors to that nuclide. An example of this may be seen by considering the decay chain for Zr^{95} .

From this chain yield it may be seen that the direct fission yield of Zr^{95} is only 0.019% whereas the total amount of Zr^{95} and Y^{99} neither of which diffuse readily at the sample temperature (2200°C) during capsule irradiation. This method was used to determine the loss of fission products from the twelve control samples during the GETR irradiation.

The determination of C_o for the samples irradiated in TREAT can be made by combining the two previously described methods. In this case $C_o + C_G + C_T$. Where C_T is the original concentration of isotope i formed by the TREAT irradiation. Values of C_T were obtained by Mo^{99} analysis of the test samples after the TREAT irradiation. It should be noted that in order

to establish C_T values by this method, precaution must be taken to insure that all Mo^{99} from the initial GETR does not contribute to the final analysis of this nuclide. The Mo^{99} contribution from GETR was made negligible by allowing a 23 half life decay time between irradiations. The method of determining the fractional release of a nuclide is perhaps more easily followed by use of the following example. Number of atoms of Sr^{89} atoms from GETR control sample on September 2 = $2.05 \times 10^{14} = C'_G$
Gross γ Activity of control sample = 3.568×10^5 CPM = A_c
Gross γ Activity of test sample = 3.715×10^5 CPM = A_T
Hence number of atoms of Sr^{89} in test sample from GETR irradiation = C_G
 $= C'_G \cdot A_T / A_c \cdot C'_G = 2.05 \times 10^{14} \times \frac{3.715 \times 10^5}{3.568 \times 10^5} = 2.134 \times 10^{14}$
Number of Mo^{99} atoms on September 2 from TREAT irradiation in Test Sample = 4.02×10^{13}

Fission Yield of $\text{Mo}^{99} = 6.06$

Fission Yield of $\text{Sr}^{89} = 4.79$

Number of Sr^{89} atoms on September 2 from TREAT irradiation in test sample
 $4.02 \times 10^{13} \times \frac{4.79}{6.06} = 3.177 \times 10^{13} = C_T$. The total number of Sr^{89} atoms formed

by both irradiations, $C_o = C_G + C_T = (2.134 + 0.318) \times 10^{14} = 2.452 \times 10^{14}$.

Number of Sr^{89} atoms found in test sample by radiochemical analysis on Sept. 2,
 $C = 9.930 \times 10^{13}$. Fraction Sr^{89} retained = $C / C_o = \frac{9.930 \times 10^{13}}{2.452 \times 10^{14}} = 0.405$ and
fraction of Sr^{89} released by TREAT transient irradiation = $1 - 0.405 = 0.595$.

3.0 RESULTS AND DISCUSSION

3.1 RELEASE DURING STEADY STATE IRRADIATION

Values of the fractional release of various nuclides during the GETR

irradiation are given in Table I normalized to Mo^{99} . The concentration of Mo^{99} was found to be essentially constant from sample to sample validating the premise of no diffusional loss of Mo^{99} or its precursors during irradiation. All other nuclides show substantial losses from the samples located in the center of the capsule. These losses are not surprising in the case of Sr^{89} , Ba^{140} and Cs^{137} which are rapid diffusers, however the losses of Ce^{144} , Y^{91} and Zr^{95} probably occurred via their precursors, since these nuclides do not readily diffuse at the irradiation temperature of 2200°C . It is interesting to note that release fractions greater than 1, (negative release fractions) are obtained on the end samples for the nuclides Sr^{89} , Ce^{144} , and Y^{91} . This apparent gain in activity suggests that material diffusing from the central samples redeposits on the end samples. This phenomenon would certainly occur in a static system where diffusing material would transfer from hot to cool zones. The design of the irradiation capsule used for this test conforms to the above description and it is probably a redeposition mechanism rather than a flux gradient which is responsible for the change of activity from sample to sample.

3.2 RELEASE DURING POST IRRADIATION HEAT ANNEAL

The fractional release of various nuclides during post irradiation heat anneals at 2000°C in vacuum are given in Table 2. Here it may be seen that essentially all of the Sr^{89} , Ba^{140} and Cs^{137} are released in 30 minutes. No release of Zr^{95} was obtained under these conditions, while Y^{91} and Ce^{144} were somewhat depleted. These data permit arrangement of the various nuclides into four groups, according to their release fraction at 2000°C for 30 minutes.

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- 7 -

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TABLE 1

FRACTIONAL RELEASE OF FISSION NUCLIDES DURING IRRADIATION IN GETR

Sample Position	Ba ¹⁴⁰	Sr ⁸⁹	Cs ¹³⁷	Ce ¹⁴⁴	Y ⁹¹	Zr ⁹⁵	Mo ⁹⁹
1	0.056	-0.276	0.076	-0.715	-1.08	0.076	0.00
2	0.791	0.468	0.326	0.112	0.787	0.421	0.00
3	0.360	0.733	0.672	0.589	0.935	0.550	0.00
4	0.906	0.675	0.527	0.602	0.890	0.276	0.00
5	0.952	0.758	0.707	0.729	0.976	0.579	0.00
6	0.951	0.741	0.703	0.846	0.975	0.512	0.00
7	0.939	0.708	0.639	0.835	0.968	0.511	0.00
8	0.905	0.640	0.484	0.801	0.948	0.459	0.00
9	0.828	0.480	0.260	0.807	0.874	0.314	0.00
10	0.082	-0.167	0.072	0.050	0.783	0.282	0.00
11	0.062	-0.280	0.122	-0.520	0.764	0.160	0.00
12	0.062	-0.216	0.127	-0.350	-1.200	0.102	0.00

<u>Group</u>	<u>% Released</u>	<u>Nuclide</u>
Group 1	> 90%	Sr ⁸⁹ Ba ¹⁴⁰
Group 2	< 90% > 70%	Cs ¹³⁷
Group 3	< 70% > 10%	Y ⁹¹ C ¹⁴¹
Group 4	< 10%	Zr ⁹⁵ Mo ⁹⁹

The relative release rate of these nuclides are in accord with their elemental boiling points. If we assign a range of boiling points of each of the groups it is possible to include elements which represents the majority of the activity after irradiation, but which were not analysed in this study. Such an arrangement is given in Table 3. The elements listed here are comprised of 144 isotopes and represent 96% of the radioactive species present after a 10 minute NERVA operation. This arrangement is preliminary and justification by determinations of additional nuclides to those presently studied is necessary before release rates can be assigned to all nuclides. The value of such an arrangement however, is the reduction of radiochemistry required to assign diffusion constants to all nuclides over a range of temperature, time or other parameters.

3.3 RELEASE DURING TRANSIENT IRRADIATION IN TREAT

The change in gross γ activity as a result of the TREAT irradiations are given in Table 4. The gross γ activities of the test samples measured before the irradiation were corrected to time of irradiation by a decay factor determined from a control sample. Only for the 806 megawatt second irradiation was there any reduction of the initial gross γ inventory. If the change of gross activity is plotted as a function of irradiation energy, as in Figure I,

TABLE 2

FRACTIONAL RELEASE OF FISSION ISOTOPES DURING POST IRRADIATION HEAT ANNEAL

Temperature (Deg. C.)	Time (Min)	Sr ⁸⁹	Ba ¹⁴⁰	Y ⁹¹	Zr ⁹⁵	Cs ¹³⁷	Ce ¹⁴⁴
2000	15	0.189	0.275	0.140	0.00	0.795	0.312
2000	30	0.999	0.999	0.163	0.00	0.828	0.320
2000	45	0.999	0.999	0.218	0.00	0.823	0.334
2000	60	0.999	0.999	0.146	0.00	0.806	0.390

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TABLE 3
ARRANGEMENT OF ELEMENTS ACCORDING TO BOILING POINT AND DIFFUSION RATE

GROUP	Boiling Point Range (Deg. C.)	% Release
1. Rb, Sr, Kr, Ba, Br, Xe, I, Se, As	-153 to 1200	90%
2. Sb, Ta, Cs	1200 to 1400	90%
3. Y, La, Ce, Sn, Ru, Rh	1400 to 2900	70% 10%
4. Nb, Zr, Mo	2900 to 3700	10%

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TABLE 4

CHANGE IN INITIAL GROSS ACTIVITY IN NERVA FUEL AS A RESULT OF TRANSIENT

IRRADIATION IN TREAT

Sample Number	Energy Transient (Megawatt seconds)	Number of Fissions (1)	Weight Loss (%)	Change in Gross Activity (%)	Average Change in Gross Activity (%)
1	177	1.57×10^{13}	0.0	+5.35	+4.18
2	177	1.55×10^{13}	0.0	+3.01	
3	451	4.13×10^{13}	0.0	+11.69	+11.69
4	451	4.07×10^{13}	0.0	+11.69	
5	806	6.34×10^{13}	12.8	-6.35	-6.40
6	806	6.56×10^{13}	10.8	-6.45	

(1) Determined by a Mo^{99} analysis

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FIGURE I

CHANGE IN GROSS ACTIVITY, OF NERVA SAMPLES IRRADIATED IN TREAT

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TABLE 5

FRACTIONAL RELEASE OF FISSION NUCLIDES DURING TRANSIENT RADIATION IN TREAT

TREAT Transient Irradiation energy megawatt seconds	Temperature (Deg. C)	Ba ¹⁴⁰	Sr ⁸⁹	Cs ¹³⁷	Zr ⁹⁵	Y ⁹¹	Ce ¹⁴⁴
177	1250	0.084	0.005	0.0175	0.00	0.00	0.00
451	2200	0.164	0.109	0.0180	0.086	0.00	0.00
806	~3000	0.558	0.595	0.619	0.086	0.00	0.187

an activity increase of 21% at 806 MWS would be predicted by linear extrapolation. Some justification for this linear extrapolation is given by the values obtained for Mo⁹⁹ concentration which has been normalized to the gross γ activity and included in Figure I.

Release fractions for the various nuclides comprising the initial inventory from the GETR irradiation are given in Table 5. Small losses of Ba¹⁴⁰, Sr⁸⁹ and Cs¹³⁷ occurred during the 177 and 451 MW's irradiations, with no loss of Zr⁹⁵, Y⁹¹ and Ce¹⁴⁴. During the 806 MW's irradiation there was release of all nuclides for which an analysis was made with the exception of Y⁹¹. The losses for Sr⁸⁹, Ba¹⁴⁰ and Cs¹³⁷ are substantial amounting to approximately 50% for each of the three nuclides. The 50% losses obtained for these nuclides are not adequately reflected in the 6% gross γ activity loss, for the obvious reason that the activity represents only a small portion of the total activity present. Calculation of the fission product inventory after 62 days decay,⁽²⁾ (the time lapse between GETR and TREAT irradiations) shows the percentage contribution to the total activity by the elements Ba, Sr, and Cs, is 13.7%. A 50% reduction of these elements would constitute only a 6.9% reduction in gross activity which is in qualitative agreement with the 6.4% value obtained for gross γ activity.

4.0 CONCLUSIONS:

1. Substantial release of Ba¹⁴⁰, Sr⁸⁹, Cs¹³⁷, Ce¹⁴⁴, Y⁹¹ and Zr⁹⁵ occur during irradiation of NERVA fuel in GETR at 2200°C.
2. The release of these nuclides is the net result of diffusion of both the nuclides and their precursors.
3. Greater than 90% release of Sr⁸⁹, Ba¹⁴⁰ and Cs¹³⁷ is obtained from NERVA

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fuel heated to 2000°C in vacuum for 30 minutes.

4. No release of Zr^{95} and Mo^{99} occurs during post irradiation heating 2000°C over a 60 minute interval.
5. The relative release order of the nuclides studied are Ba^{140} , Sr^{89} , Cs^{137} , Ce^{144} , Y^{91} , Zr^{95} and Mo^{99} . This corresponds to the same order obtained when these elements are arranged according to their boiling points.
6. Transient irradiations at 177 and 451 megawatt second TREAT energies, increase the inventory in pre-irradiated NERVA fuel.
7. A transient irradiation of 800 megawatt seconds the original inventory by less than 10%.
8. The majority of the loss of inventory is due to the decrease of Ba, Cs, Sr and Ce isotopes
9. There is negligible loss of Y, Zr, and Mo during transient of 800 megawatt seconds or less.

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