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September 1965

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Westinghouse Astronuclear Laboratory

INTERIM REPORT ON THE EXPERIMENTAL
INVESTIGATION OF
FISSION PRODUCT DIFFUSION
(Title Unclassified)

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Westinghouse Astronuclear Laboratory

September 1965

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1.0 INTRODUCTION AND SUMMARY

This Interim Report covers the work performed on the Fission Product Diffusion Program between April 1, 1965, and September 1, 1965. The prime objective of the Fission Product Diffusion Program is to provide experimental data on the diffusion of fission product nuclides in NERVA fuel as a function of time and temperature, geometry, and fuel material. The experimentally determined diffusion constants are used in analytical codes to predict the magnitude of fission product release from the NERVA reactor under accidental and normal operating conditions.

Since the diffusion constants of the fission products may vary with the nature of the fuel material from which they diffuse, studies have been made on "Duplex" fuel some of which was used in the NRX-A3 reactor.

In order to test the validity of releases predicted by the analytical codes, during accident conditions, experiments have been run using a clustered assembly of fuel elements intended to represent the NERVA reactor during post operational loss of coolant conditions.

The final experimental effort described in this report concerns post operational radiochemical analysis of samples from the NRX-A3 reactor. This analysis has been made to provide fission product release data for comparison with code predictions for normal full power reactor operation.

2.0 POST IRRADIATION THERMAL ANNEAL EXPERIMENTS ON DUPLEX FUEL

2.1 EXPERIMENTAL

2.1.1 Sample Preparation

The samples used in this experiment were $\frac{1}{4}$ inch sections of a regular nineteen hole, hexagonal fuel element. The duplex fuel is a dispersion of 100 μ diameter UC₂ beads coated with two concentric 12.5 μ thick layers of laminar pyrolytic graphite. After sectioning, the samples were leached to remove exposed UC₂, then irradiated at the Westinghouse Reactor Evaluation Center, in a thermal neutron flux of $\approx 1 \times 10^{10}$ n/cm² for $\frac{1}{2}$ hour. After irradiation, the samples were returned to WANL for thermal anneal studies.

2.1.2 Apparatus

The irradiated samples were counted on a low geometry gamma counter, then heated in an induction heated vacuum furnace at a pressure of 4×10^{-4} torr. Sample temperatures which varied over the range 2250°C to 2550°C were measured with a dual wave length, recording optical pyrometer.

2.1.3 Calculation of Gross Fission Product Release Fractions

The fractional release of gross activity was determined by counting samples in a constant geometry before and after thermal treatment. The only correction required for this counting data was a decay correction for

the time lapse between the two counting times. This decay correction was made empirically by counting an unheated control sample and using a single relationship:

$$A_c = A_1 C_2 / C_1 \quad (1)$$

where: A_c was the corrected activity in the test sample at time, t_2 .

A_1 was the initial test sample activity at time, t_1 .

C_1 was the control sample activity at time, t_1 .

C_2 was the control sample activity at time, t_2 .

The fraction of the gross activity, f , retained in the test sample during the thermal anneal, was given by:

$$f = A_t / A_c \quad (2)$$

where: A_t was the measured activity of test sample at time, t_2 .

The fraction of the gross gamma activity released was then simply $1-f$.

2.2 RESULTS

The fractional release of fission products determined by gross gamma activity measurements for samples of duplex fuel heated for five minutes at various temperatures is given in Figure 1. Included in the figure are gross fission product releases from Type II fuel heated under identical
(1)
conditions. The fractional gross fission product release from duplex fuel

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FIGURE I

FRACTIONAL RELEASE OF GROSS GAMMA FISSION PRODUCT INVENTORY
FROM TYPE II AND DUPLEX NERVA FUELS HEATED 5 MINUTES IN VACUUM

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appears to increase linearly with temperature over the temperature range studied. This release data for duplex fuel appears identical with the release data from the Type II fuel.

2.3 DISCUSSION

(2)

It has been reported that duplex fuels exhibit superior UC_2 migration resistance over the Type II fuel which is currently in use in the NERVA reactor. Type II fuel consists of 100μ diameter UC_2 beads coated with a single 25μ thick layer of laminar pyrolytic graphite which was deposited on the bead in one operation. The duplex fuel used in these experiments has two 12.5μ thick layers of pyrolytic graphite deposited in two separate operations with a cool down between operations. This process results in a similar 25μ thickness of pyrolytic graphite surrounding the fuel bead but exposes the UC_2 to lower temperatures during the second coating operation. The duplex fuel should exhibit less total migration during reactor operations because less UC_2 migration takes place during the fabrication of this material.

The difference in fabrication techniques between this duplex fuel and Type II fuel result in essentially identical physical systems with respect to fission product release, namely a 25μ thickness of laminar pyrolytic graphite through which fission products must diffuse. It is to be expected then that the extent of fission product release from these two materials

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should be similar as shown in Figure 1.

It should be noted that the designation of "duplex" to describe the various types of multiple pyrolytic graphite coatings on UC_2 beads is a broad definition. It is outside the realm of this report to describe the various duplex fuels which are under consideration for use as NERVA fuel. Since some of the double laminar fuel described in this report was used in NRX-A3, it was necessary to determine fission product diffusion characteristics from this particular fuel. When other duplex fuels are actually incorporated in NERVA fuel elements for full power reactor tests, determination of their fission product release characteristics will be required.

3.0 POST IRRADIATION THERMAL ANNEAL EXPERIMENTS ON UNIT CELLS

3.1 INTRODUCTION

Previous fission product diffusion studies on degraded samples of NERVA fuel provided data on the fission product release rates from the fuel which could be applied during a loss of coolant accident following normal reactor operation. The question remains whether these release rates apply to escape of fission products from the reactor proper as well as the fuel material.

In order to simulate the reactor components and geometry from a fission product release standpoint, experiments have now been made on

an assembly containing fuel elements, non-fueled graphite, support plates, or orifices, and a tie rod. The geometry of this assembly was chosen to represent a unit cell of the NERVA core and thus provide identical diffusion path lengths for fission product release.

Three unit cell experiments were made. The first of these was a feasibility study with non-irradiated fuel to test the heating rates of the vacuum furnace employed, and to acquaint personnel with the complete experimental operation. The second experiment was an attempt to heat an irradiated unit cell to 2300°C. This experiment was not successful due to failure of the unit cell components early in the course of the heating run. The third experiment was a second attempt to heat a unit cell to 2300°C. A description and the results of this third experiment constitutes this section of the report.

3.1.1 Sample Preparation

The unit cell is a cluster of 4" long segments of NERVA fuel held together with a modified NERVA tie rod assembly and contained in a hollow graphite cylinder, 3" in diameter. A list of the unit cell components is given in Table I. The unit cell construction is shown schematically in Figure 2 and pictorially in Figure 3. The fueled portions of the unit cell were irradiated, together with 4, 1" long, hexagonal control samples, at the Westinghouse Reactor Evaluation Center for a total of 0.75Kwh. After irradiation, the fueled components were returned to WANL, allowed to decay for

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TABLE I
COMPONENTS OF UNIT CELL

ITEM NUMBER	COMPONENT DESCRIPTION	NUMBER OF LIKE COMPONENTS	COMPONENTS DIMENSIONS AND CONFIGURATION	MATERIAL
1	Fueled Elements	6	Regular hexagonal, side 0.437", length 4". 19, 0.097 diameter cooling holes.	100 μ diameter UC ₂ beads with 25 μ diameter coating of laminar pyrolytic graphite, in graphite matrix NbC lining in coolant holes.
2	Fueled Elements	6	$\frac{1}{2}$ of a regular hexagon, 4" long with 11, 0.097" diameter cooling holes	"
3	Fueled Elements	6	$\frac{1}{4}$ of a regular hexagon, 4" long with 5, 0.097" diameter cooling holes	"
4	Unfueled Element	1	Regular hexagon, side 0.437", length 4". 1 axial hole, 12, 0.097" coolant holes	Graphite, with NbC lining in coolant holes.
5	Bottom Support Plate	1	Disc, 2 3/4" diameter, 1/8" thick provided with 222, 0.097" diameter cooling holes	Graphite
6	Top Support Plate	1	Disc, 2 3/4" diameter, $\frac{1}{4}$ thick provided with 222, 0.097" cooling holes	Silver, Indium, Cadmium
7	Tie Rod Assembly	1	Length 9.5", diameter 0.111" plus assorted hardware	Inconel
8	Graphite Can	1	Cylindrical tube, closed at one end, inside diameter, 2 3/4", outside diameter, 3.03 length 11 $\frac{1}{2}$ "	Graphite

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FIGURE 2
CROSS-SECTIONAL VIEW OF UNIT CELL

FIGURE 3
EXPLODED VIEW OF UNIT CELL ASSEMBLY

five days, then assembled with the remaining unirradiated components into the cluster assembly.

The cluster was inserted into the graphite cylinder, which in turn was placed in a quartz cylinder, 17" long by $5\frac{1}{2}$ " in diameter, closed at one end. The $2\frac{1}{2}$ " annulus between the graphite and quartz cylinders was packed with powdered graphite which served as thermal insulation during the subsequent heating operation.

3.1.2 Apparatus and Procedure

The unit cell assembly in the quartz container was heated in a 1000 cycle, 100 kilowatt radio frequency--vacuum furnace at the Westinghouse Research Laboratory. The sample temperature was monitored with a dual wave-length recording optical pyrometer, through a quartz view port in the vacuum furnace. To prevent clouding of the quartz sight glass due to carbon deposition during the course of the run, a mylar film strip, interposed between the sight glass and the heated sample, was continuously passed across the sight glass by a motor driven reel assembly.

The unit cell was heated to 2300°C and maintained at this temperature for 15 minutes. During the run a dynamic vacuum of 1×10^{-4} mm was achieved with a 4" diffusion pump system.

During the heating process, movies were taken to provide a continuous visual record of the unit cell component disposition.

After the thermal anneal, the unit cell was returned to WANL and disassembled in a shielded glove box. Photographs were taken of the various components during the disassembly process. Individual fuel elements as well as the central unfueled element and a representative sample of the outer graphite can were monitored for gross gamma activity in a high pressure ionization chamber. Selected samples were then ignited in O_2 and dissolved for radiochemical analyses.

3.1.3 Sample Dissolution and Radiochemical Analysis

Five representative samples from the unit cell were chosen for radiochemical analysis. These were:

1. A 4" length of a full hexagonal fuel. (Item 1, Table I)
2. A 4" length of a $\frac{1}{2}$ hexagonal fuel element. (Item 2, Table I)
3. A 4" length of a $\frac{1}{4}$ hexagonal fuel element. (Item 3, Table I)
4. A 4" length of unfueled element. (Item 4, Table I)
5. A 1" x 11 $\frac{1}{2}$ " strip from the graphite can plus a 1" arc wedge from the bottom of the graphite can. (Item 8, Table I).

In addition to the above samples, radiochemical analyses were made on the four irradiated control samples. Samples with NbC lining were leached with $HF-HNO_3$ to remove the lining. Then the samples were ignited in a quartz tube in an atmosphere of flowing oxygen. The residual ash in the ignition tube was

dissolved in 6N-HNO₃ and diluted to a known volume. Radiochemical analyses were then performed in duplicate with aliquots from the known volume of solution to determine the concentration of the following nuclides: Sr⁸⁹, Ba¹⁴⁰, La¹⁴⁰, Ce¹⁴¹-Ce¹⁴⁴, Zr⁹⁵, and I¹³¹.

3.1.4 Calculation of Release Fractions

The calculation of fractional retention of the individual fission product nuclides during the post irradiation thermal anneal experiment required knowledge of the initial concentration of the nuclide of interest before the thermal treatment and the concentration of the same nuclide after the thermal treatment. Determination of the concentration of any nuclide after thermal treatment was made by radiochemical analyses of the heated test sample. The initial concentrations of the various nuclides in the test samples before heating was determined by radiochemical analysis of four unheated control samples together with gross gamma measurements of the test and control samples before the heat treatment. The initial concentration of the nuclide in the test sample, C₁, is given by:

$$C_1 = C_2 G_1/G_2 \quad (3)$$

where: C₂ is the concentration of the nuclide in the control sample.

G₁ is the gross gamma activity in the test sample.

G₂ is the gross gamma activity in the control sample.

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In this experiment, four control samples were used, hence, the values of C_2 and G_2 used in the calculations were average values of these four.

With the initial concentrations of various nuclides, C_1 , determined as outlined, and the final concentrations of these isotopes, C_f , the fractional retention of the various isotopes is given by:

$$C_f/C_1$$

and the fractional release as:

$$1 - C_f/C_1$$

3.2 RESULTS

3.2.1 Visual Examination

Complete melting of the metal tie rod assembly, orifices, and top support plate occurred during the thermal anneal. On a weight basis, approximately 60% of the metallic components escaped the graphite can by vaporization. The majority of the metal which remained, collected in the bottom of the graphite can. The redeposited metal served to bond the graphite can to the inner fuel element bundle. After removal of the redeposited metal the fuel element segments were easily separated and showed no evidence of graphite to graphite self welding.

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Some of the vaporized metal condensed in cooling channels of the fuel elements, resulting in 40% complete blockage and 24% partial blockage of the channels. The remaining 36% of the channels were completely free of redeposited metals.

Figures 4 through 7 are a series of photographs taken during disassembly of the unit cell. The first step in the disassembly was removal of those portions of the graphite can which extended beyond the ends of the fuel element bundle. Figure 4 is a side view of the fuel element bundle with the portion of the graphite can adjacent to the bundle still intact. A top view of the bottom section of the graphite can containing the bulk of the redeposited metal is shown at the extreme right in Figure 4.

Figure 5 is a top view of the graphite fuel bundle at the same stage of disassembly as Figure 4. Condensed metal can be seen in some of the channels of the graphite support plate.

A side view of the fuel bundle with the graphite can completely removed is shown in Figure 6. At this stage of the disassembly some of the peripheral fuel elements were found to be unbonded to the inner fuel elements. The graphite support plate, however, is affixed to the fuel element bundle by redeposited metal in the cooling channels.

The individual components of the fuel element bundle are shown in Figure 7, after complete disassembly. In order to separate the

FIGURE 4
SIDE VIEW OF UNIT CELL ASSEMBLY AFTER THERMAL ANNEAL

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FIGURE 5

TOP VIEW OF UNIT CELL ASSEMBLY AFTER THERMAL ANNEAL

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FIGURE 6

UNIT CELL FUEL BUNDLE WITH OUTER GRAPHITE CAN REMOVED

FIGURE 7

UNIT CELL COMPONENTS DISASSEMBLED AFTER THERMAL ANNEAL

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individual elements it was necessary to break away portions of the graphite support plate. After removal of this plate, the fuel elements were easily separated with no evidence of self welding.

3.2.2 Radiochemical Analysis

The release fractions of the various nuclides as well as the gross gamma inventory release from the regular hexagonal elements, $\frac{1}{2}$ hexagonal elements, and $\frac{1}{4}$ hexagonal segments are given in Table II. Included in Table II are the values for the fractional retention of the nuclides, by the center unfueled element and the outer graphite can. Also included in Table II are the effective release fractions from the unit cell. These numbers were calculated by subtracting the total fraction of the nuclide inventory which was retained by the unfueled portions of the unit cell, from the average fractional release from the fuel.

In general, it appears that the fractional release of fission products was greatest from the $\frac{1}{4}$ hexagonal fuel segments and least from the regular hexagonal fuel segments.

Except for Ba^{140} , the retention of the various fission product nuclides by the unfueled portions of the unit cell was small with respect to the release fraction of the nuclides.

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TABLE II

FRACTIONAL RELEASE OF FISSION PRODUCTS FROM UNIT CELL

NUCLIDE	FRACTIONAL RELEASE FROM REGULAR HEXAGONAL FUEL ELEMENTS	FRACTIONAL RELEASE FROM $\frac{1}{2}$ HEXAGONAL FUEL ELEMENTS	FRACTIONAL RELEASE FROM $\frac{1}{4}$ HEXAGONAL FUEL ELEMENTS	AVERAGE FRACTIONAL RELEASE FROM FUEL	FRACTION RETAINED BY UNFUELED CENTER ELEMENT	FRACTION RETAINED BY OUTER GRAPHITE CAN	EFFECTIVE RELEASE FROM UNIT CELL
Sr ⁸⁹	0.743	0.858	0.878	0.826	0.034	0.004	0.788
Zr ⁹⁵	0.043	0.063	0.061	0.052	0.007	0.002	0.052
I ¹³¹	0.669	0.602	0.624	0.632	0.00	0.00	0.632
La ¹⁴⁰	0.301	0.460	0.457	0.406	0.011	0.001	0.394
Ba ¹⁴⁰	0.585	0.738	0.734	0.686	0.153	0.013	0.520
Ce ¹⁴¹⁻¹⁴⁴	0.026	0.316	0.201	0.258	0.036	0.060	0.162
Gross γ Inventory	0.314	0.378	0.399	0.364	—	—	0.364

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3.2.3 Comparison of Radiochemical Results with FIPDIF Prediction

A prediction of the release of various nuclides during the unit cell experiment was made using the FIPDIF code. The diffusion constants for the nuclides in this code were based on thermal anneal experiments using $\frac{1}{4}$ " by $\frac{1}{4}$ " hollow cylindrical samples of Type II and Type III fuel. The assignment of diffusion constants for diffusion of various elements from these fuels has been discussed in previous reports, however, for the sake of clarity, these diffusion constants and a description of the two fuel types are summarized in Tables III and IV.

The effective release of various fission products from the unit is compared with predicted releases for these nuclides in Table V. The predicted gross fission product release is higher than the experimentally determined gross fission product release. This is consistent with the high release fractions of various nuclides, predicted by the code. The predicted release for Ce¹⁴¹⁻¹⁴⁴ is almost 60% greater and the release for Ba¹⁴⁰ is 45% greater than was found by radiochemical determinations.

3.3 DISCUSSION

The fuel used for the assembly of the unit cell consisted of Type II NERVA fuel with NbC lining in the coolant channels. There are thus two paths for diffusional release of fission products from this fuel, the first

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TABLE III

⁺
DIFFUSION CHARACTERISTICS OF FISSION PRODUCT ELEMENTS IN TYPE II, NERVA FUEL

Group Number	1 - G **	G *	D ₀ (Sec ⁻¹)	E/R (Kcals/mole)	Elements
1	0.85		3.977×10^2	33.2	Sr, Sn, Ag, Cd, Zn, In
2		0.15	8.70	15.5	SrO, Sn-O, Ag-O, Cd-O, Zn-O, In-O
3	0.85		1.83×10^{-3}	6.00	Ba
4		0.15	9.17×10^{-3}	2.50	Ba-O
5	1.00		7.97	26.0	Ce, Y, La, Kr, Xe, Sb, Pr
6	1.00		5.383×10^{10}	87.2	I, Cs, Br, Te, Rb
7	1.00		0	0	Mo, Zr, Ru, Nb, Tc

* G = fraction of concentration which diffuses at fast rate

** 1-G = fraction of concentration which diffuses at slow rate

+ Type II NERVA fuel is a dispersion of 100u diameter beads of UC₂ in graphite. The UC₂ beads are coated with a 25 micron thickness of laminar pyrolytic graphite.

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TABLE IV

DIFFUSION CHARACTERISTICS OF ELEMENTS IN TYPE III⁺ NERVA FUEL

Element	% Abundance at Zero Time	Representative Nuclide	Diffusion Group	D ₀ (sec ⁻¹)	E/R (Kcals/mole)
Nb	10.18	Nb ⁹⁵	11	** 0	0
Sr	8.05	Sr ⁸⁹	2	1.63	11.4
Rb	7.71	* -	11	0	0
Y	6.81	Y ⁹¹	10	0.379	17.0
Xe	5.98	Xe ¹³³	2	1.63	11.4
Cs	5.86	Cs ¹³⁷	4	0.477	10.5
Kr	5.73	Kr ⁸⁷	2	1.63	11.4
Zr	5.62	Zr ⁹⁵	11	0	0
Br	5.36	* -	11	0	0
Ba	4.72	Ba ¹⁴⁰	3	1800	26.2
Sb	4.70	Sb ¹²⁷	9	0.779	17.0
I	4.70	I ¹³¹	6	2.67	18.2
La	3.70	La ¹⁴⁰	7	21.6	25.0
Ce	3.50	Ce ¹⁴¹⁻¹⁴⁴	9	0.779	17.0
Tc	2.97	* -	11	0	0
Te	2.97	Te ¹²	8	0.0805	13.1
Mo	2.83	Mo ⁹⁹	11	0	0
Se	2.37	* -	11	0	0
Pr	1.65	* -	11	0	0
Sn	1.45	Sn ¹²⁵	5	18.4	18.2
Ru	.10	Ru ¹⁰³	11	0	0
Ag	0.03	Ag ¹¹¹	1	1	20.0

TOTAL — 96.99

+ Type III NERVA fuel is prepared by thermal degradation of "as received" (Type II) fuel. The Thermal treatment at 2500°C for 30 minutes causes migration of UC₂ resulting in a dispersion of small UC₂ crystallites throughout the graphite lattice.

* Diffusion constants for nuclides of these elements have not yet been measured.

** Values of zero indicates no diffusion of these elements.

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TABLE V

COMPARISON OF EXPERIMENTAL FISSION PRODUCT RELEASE FRACTIONS
WITH FIPDIF PREDICTION

	RADIOCHEMICAL RESULTS	FIPDIF PREDICTION
Sr ⁸⁹	0.788	0.999
Zr ⁹⁵	0.052	0.001
I ¹³¹	0.632	0.870
La ¹⁴⁰	0.394	0.704
Ba ¹⁴⁰	0.520	0.999
Ce ¹⁴¹⁻¹⁴⁴	0.162	0.630
Gross Gamma	0.364	0.441

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through the graphite matrix and NbC coating to the cooling channels and the second through the graphite matrix to an unclad face of the hexagonal fuel element.

Those fission products which migrate to an unclad fuel face would be expected to continue into an adjacent face of an unfueled element. Radiochemical analyses of the unfueled elements, however, show almost a negligible portion present of the fission products which are released from the fuel. This lack of fission products in the unfueled graphite suggests that the majority of the fission products are released through NbC coating into the cooling channels, and out of the unit cell.

If the primary fission product release mechanism is via the NbC lining, the differences between the predicted and measured release fractions may be due to input data for the FIPDIF prediction which were based on unclad Type III fuel samples. Diffusion data pertaining to NbC coated, Type III fuel will be incorporated into the code as they become available.

An alternate reason for the low release fractions may be plugging of the coolant channels by redeposited metal vapor. Approximately 40% of the cooling channels were plugged by redeposited metal vapor at the termination of the unit cell thermal anneal. If the diffusion path for fission product release is solely via these channels, plugging of some of these channels should inhibit fission product release. It is perhaps coincidental that the predicted

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gross release is 40% higher than the measured gross release.

It is expected that at higher temperatures and longer times, evaporation of the metallic components in the unit cell will be complete, and no plugging of cooling channels will occur. Future unit cell experiments should resolve the effect, if any, of cooling hole blockage by redeposited metal.

4.0 POST OPERATION EXAMINATION OF NRX-A3 SAMPLES USING RADIOCHEMICAL TECHNIQUES

4.1 SAMPLES USED IN STUDIES

A post operation radiochemical analysis was performed on selected samples obtained from the NRX-A3 reactor after it had been operated at full power for approximately $3\frac{1}{2}$ minutes, shutdown for 29.3 days then restarted and operated at full power for an additional 13 minutes. These samples consisted of segments of both fueled and unfueled elements. The purpose of this investigation was to determine the extent of fission product release as a result of full power testing. Since, the samples were available for analysis after completion of both runs, only a determination of the total release of fission products from the complete series of tests can be made. No attempt has been made to determine the fission product release during the separate full power runs.

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4.2 SAMPLING AND EXPERIMENTAL PROCEDURE

Samples were taken from 3 fueled elements, 000G, 1D2E, and 6J9W which represent a radius vector in the NRX-A3 core. Additional samples were taken from the unfueled elements 000A and 6J9Q which were respectively adjacent to the fuel elements 000G and 6J9W in the core.

Four sections were cut from each element between 28 and 52 inches from the end of the core. Three sections were also cut from the opposite ends of the elements to be used as control samples. These sections near the inlet end of the core experienced temperatures no greater than 900°C, and hence, suffered no loss of fission products via diffusion.

A quarter inch section was then taken from the center of each of the sections. The sample locations are given in Table VI. This sampling procedure resulted in 21 fuel sample slices and 8 slices from unfueled elements. In the prior radiochemical examination of NRX-A2, four $\frac{1}{4}$ inch slices were taken for analyses. The reproducibility of radiochemical analyses between these slices indicated that one slice would serve as a representative sample from a five inch section, hence, for the NRX-A3 examination only one $\frac{1}{4}$ inch fuel slice was taken.

The $\frac{1}{4}$ inch slices were placed in separate plastic containers and shipped to WANL in an 890 pound cask which provided radiation shielding. At WANL the samples were transferred to a shielded glove box, then removed individually from the box for subsequent analyses.

The samples were weighed, then monitored for γ activity in a high pressure ionization chamber. Data on the number of fissions per sample as measured by the ion chamber are given in Table VI.

The samples were dissolved using the following process:

1. HF-HNO₃ leach to remove NbC lining,
2. Combustion in flowing O₂ followed by dissolution of residue in HNO₃, and
3. Trapping of the volatiles from the combustion process in NaOH.

The three solutions from the above dissolution process were combined and diluted to a standard volume for subsequent radiochemical analysis.

4.3 NRX-A3 RADIOCHEMICAL RESULTS

The fuel samples from the NRX-A3 reactor were analyzed for Mo⁹⁹, Y⁹¹, Zr⁹⁵, Sr⁸⁹, I¹³¹, Cs¹³⁷, and Ba¹⁴⁰, to determine the mass chain releases represented by these nuclides. The release fractions for these nuclides were determined using a control sample technique which assumes that:

$$\left(\frac{X_1}{I_1} = \frac{X_2}{I_2} \right) \dots \dots \dots (4)$$

where: X_1 is the concentration of the diffusing nuclide of interest in the control sample.

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TABLE VI

IONIZATION CHAMBER READINGS ON NRX-A3 FUEL SAMPLES

RADIOCHEMISTRY SAMPLE NUMBER	EXTRUSION NUMBER	REACTOR LOCATION	RADIUS (cm from center of core)	AXIAL LOCATION (Inches from inlet.)	SAMPLE WEIGHT (Grams)	ION CHAMBER APPARENT NUMBER OF FISSIONS PER SAMPLE
1	99-16043	000G.	1.91	5.00 - 5.25	3.0096	1.451×10^{17}
2				15.25 - 15.50	2.9686	2.224
3				25.5 - 25.75	2.9488	2.518
4				31.0 - 31.25	3.1148	2.588
5				36.25 - 36.50	2.9850	2.092
6				41.50 - 41.75	2.9836	1.621
7				47.75 - 48.00	2.9682	0.942
15	99-15659	1D2E	21.74	5.00 - 5.25	2.8570	1.268
16				15.25 - 15.50	2.8770	2.164
17				25.50 - 25.75	2.9745	2.401
18				31.0 - 31.25	2.9113	2.149
19				36.25 - 36.50	2.8312	1.831
20				41.50 - 41.75	2.8767	1.409
21				47.75 - 48.00	2.9174	0.769
29	69-15030	6J9W	41.99	5.00 - 5.25	2.8088	1.037
30				15.25 - 15.50	2.8755	1.831
31				25.50 - 25.75	2.8855	1.988
32				31.0 - 31.25	2.9058	1.859
33				36.25 - 36.50	2.9254	1.590
34				41.50 - 41.75	2.9450	1.235
35				47.75 - 48.00	2.8905	0.711

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X_2 is the concentration of the diffusing nuclide of interest in the test sample if no diffusion had occurred during reactor operation.

I_1 is the concentration of a non-diffusing nuclide in the control sample.

I_2 is the concentration of the non-diffusing nuclide in the test sample.

The expected concentration of a particular nuclide, X_2 , was established in this manner. The actual concentration of this nuclide, X_3 , was found by radiochemical analysis. The fractional retention is then given by X_3/X_2 and the fractional release by $1 - X_3/X_2$.

For these determinations, three samples from the cold end of each element were taken as controls. The indicator nuclide used was Cs^{137} which has negligible release from the fuel under normal operating conditions. It was originally intended to use Mo^{99} as an indicator nuclide, since components of the mass 99 chain do not diffuse even during extreme temperatures. Unfortunately, at the time of analyses, the Mo^{99} had practically all decayed, thus the Mo^{99} concentrations based on low count rates were unreliable.

The maximum fractional releases for the various mass chains are given in Table VII. The release fractions are given in terms of the total activity of the particular nuclide in the entire core.

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TABLE VII
RELEASE OF FISSION PRODUCTS FROM NRX-A3 FUEL
AND RETENTION BY NON-FUELED ELEMENTS

MASS CHAIN NUMBER	END MEMBER NUCLIDE	FRACTIONAL RELEASE FROM FUEL A	FRACTION RETAINED IN UNFUELED ELEMENTS B	B/A x 100
89	Sr	0.013	0.00064	4.9
91	Y	0.026	0.00060	2.3
95	Zr	0.031	0.00017	0.5
115	Cd-m	0.305	0.00016	0.05
131	I	< 0.018	0.00004	0.2
137	Cs	0.000	0.000	-
140	Ba	0.019	0.00032	1.7
Gross Gamma Inventory		0.009	0.00027	3.0

4.3.1 Unfueled Samples

The unfueled samples were analyzed for the same nuclides as the fueled samples. A comparison with the data for release from fueled samples was then made to determine the extent of diffusion into unfueled graphite. These results are also given in Table VII.

4.4 COMPARISON OF FIPDIF FISSION PRODUCT RELEASE PREDICTION WITH RADIOCHEMICAL ANALYSIS

4.4.1 Diffusion Constants Used in FIPDIF

The diffusion constants used as input for the FIPDIF code prediction of fission product release from NRX-A3, were based on experimental data using NbC coated samples. These samples were hollow cylinders, $\frac{1}{4}$ " long by $\frac{1}{4}$ " diameter, machined from Type II NERVA fuel. After fabrication, the samples were leached in HNO_3 , then coated with NbC by a vapor deposition process. The fractional release data for various nuclides from this material (3) has been reported previously. The release fraction as a function of time for the various data were fitted to a first order rate equation of the form:

$$C = C_0 e^{-Dt} \dots \dots \dots (5)$$

where: C is the concentration of a particular nuclide at time, t.

C_0 is the initial concentration at $t = 0$.

D is the diffusion rate constant.

Equation (5) can be rearranged into the logarithmic form:

$$\ln C/C_0 = -Dt \quad \dots \quad (6)$$

where C/C_0 is now the fractional retention of the nuclide of interest.

A plot of $\ln C/C_0$ versus t gives the diffusion constant, D , as the slope. Such a plot is given in Figure 8 for Sn^{125} .

The diffusion constants for the various nuclides were found to obey the Arrhenius equation as a function of temperature. This equation is:

$$D = D_0 e^{E/RT} \quad \dots \quad (7)$$

where: D_0 is an experimental constant with dimensions of reciprocal time.

E is the activation energy in calories per mole.

R is the gas constant.

T is the temperature in degrees Kelvin.

A typical Arrhenius relationship for Sn^{125} is given in Figure 9.

A summary of the diffusion constants and activation energies used as input for the FIPDIF code is given in Table VIII.

4.4.2 Power and Temperature Input for FIPDIF

For this analysis of the predicted fission product

RETENTION OF Sn^{125} IN NbC COATED TYPE II FUEL

FIGURE 8

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ARRHENIUS RELATIONSHIP FOR Sn^{125} IN NbC

COATED TYPE II FUEL

FIGURE 9

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TABLE VIII

DIFFUSION CONSTANTS FOR FISSION PRODUCT RELEASE
FROM NbC COATED TYPE II FUEL

Diffusion Group	D_0 (Sec^{-1})	E/R (K cals/mol)	Nuclide
1 B	8.04	13.1	Ag ¹¹¹
2 B	0.754	17.9	Sr ⁸⁹
3 B	16.25	27.9	Ba ¹⁴⁰
4 B	1387	37.7	Sn ¹²⁵
5 B	4.02×10^6	55.7	Cd ^{115m}
6 B	72.8	33.6	Ce ¹⁴¹ , Y ⁹¹ , La ¹⁴⁰
7 B	5.38×10^{10}	87.2	I ¹³¹ , Cs ¹³⁷ , Sb ¹²⁵ , Xe ¹³³ , Kr ⁸⁵
8 B	0	0	Mo ⁹⁹ , Zr ⁹⁵ , Ru ¹⁰³ , Nb ⁹⁵

release during the NRX-A3 test, the core was divided into six sections. The first section at the exhaust end of the core was 8.5 inches thick. The next four sections nearest the exhaust end of the core, were equivalent sections five inches thick; the remaining 23.5 inches at the cold inlet end of the core was treated as a unit from which no diffusion occurred.

Table IX lists the time, temperature, and power inputs used for the FIPDIF prediction for the NRX-A3 full power runs EP IV and EP V. These data were provided by the WANL, Reactor Analysis section, and were based on the latest analysis of the NRX-A3 test data.

Since radiochemical analyses provide a measure of the total release from the NERVA fuel during all operational history, it was necessary to include the various low power run input data in the code calculation. Although the fission products during these low power runs would not be released at time of formation, they would be available for release during subsequent high power-high temperature runs. The data for these low power runs have been omitted from Table IX for brevity, since there was a total of 21 separate power holds.

4.4.3 Results of FIPDIF Prediction Compared with Radiochemical Analysis

The predicted release of selected mass chains together with the release of the gross gamma inventory are given in Table X. Included

TABLE IX

INPUT DATA FOR THE FIPDIF NRX-A3 FISSION PRODUCT RELEASE PREDICTION

EP IV START TIME 1.24×10^5 seconds

SECTION NUMBER	AXIAL DISTANCE (INCHES)	FRACTION OF INTEGRATED POWER IN SECTION	FIRST RAMP 250 MEGAWATTS FOR 56 SECONDS TEMPERATURE °K*	SECOND RAMP 511 MEGAWATTS FOR 128 SECONDS TEMPERATURE °K	THIRD RAMP 894 MEGAWATTS FOR 36 SECONDS TEMPERATURE °K	FOURTH RAMP 1113 MEGAWATTS FOR 203 SECONDS TEMPERATURE °K
1	0 - 23.5	0.4381	273	273	920	1060
2	23.5 - 28.5	0.1307	"	1140	1545	1785
3	28.5 - 33.5	0.1264	"	1340	1740	1990
4	33.5 - 38.5	0.1126	"	1480	1880	2170
5	38.5 - 43.5	0.0920	"	1585	1975	2300
6	43.5 - 52	0.1002	"	1640	2030	2360

EP V START TIME 2.66×10^6 seconds

SECTION NUMBER	AXIAL DISTANCE (INCHES)	FRACTION OF INTEGRATED POWER IN SECTION	FIRST RAMP 250 MEGAWATTS FOR 56 SECONDS TEMPERATURE °K	SECOND RAMP 506 MEGAWATTS FOR 48 SECONDS TEMPERATURE °K	THIRD RAMP 874 MEGAWATTS FOR 32 SECONDS TEMPERATURE °K	FOURTH RAMP 1080 MEGAWATTS FOR 788 SECONDS TEMPERATURE °K
1	0 - 23.5	0.4381	273	273	860	1070
2	23.5 - 28.5	0.1307	"	1150	1560	1790
3	28.5 - 33.5	0.1264	"	1330	1750	2020
4	33.5 - 38.5	0.1126	"	1480	1915	2185
5	38.5 - 43.5	0.0920	"	1590	2040	2300
6	43.5 - 52	0.1002	"	1610	2100	2360

* All temperatures are average temperatures taken at the center of the section.

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TABLE X

COMPARISON OF PREDICTED FISSION PRODUCT RELEASE
FROM NRX-A3 WITH RADIOCHEMICAL ANALYSES

<u>MASS CHAIN NO.</u>	<u>END MEMBER NUCLIDE</u>	<u>PREDICTED FRACTION RELEASE FROM WHOLE CORE</u>	<u>FRACTION RELEASE DETERMINED BY RADIOCHEMISTRY</u>
89	Sr	0.015	0.013
91	Y	0.022	0.026
95	Zr	0.028	0.031
115	Cd	0.390	0.30
131	I	0.008	0.018
137	Cs	0.000	0.000
140	Ba	0.010	0.019
Gross Gamma Inventory		0.016	0.009

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in the table are the releases of these mass chains determined by radiochemical analyses of end member nuclides of the various chains.

There appears to be good agreement between the predicted and measured fractional releases for all mass chains except 131. The measured iodine-131 release is substantially higher than the predicted release. There was considerable scatter in the iodine radiochemical analyses, however, and for this reason the radiochemical result is suspect.

4.5 DISCUSSION

The agreement between predicted and measured releases of the fission product mass chains with the exception of the 131 chain, indicates that the diffusion constants for the nuclides which are members of these chains are correct. The disagreement between predicted and measured releases for the 131 chain is probably due to uncertainties in radiochemical analyses of iodine, rather than incorrect assignment of diffusion constants to chain members. There is, however, disagreement between the predicted release of the gross gamma inventory and the release determined by radiochemical means. The higher predicted release suggests the use of incorrect diffusion constants of nuclides in mass chains other than those studied. In order to refine the FIPDIF code more comprehensive radiochemical analyses will be required.

For the prediction of 1.6% release of fission products from NRX-A3, diffusion constants of various nuclides in NbC coated samples were used.

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A prediction of 3.87% release of fission products from NRX-A3 was made using diffusion constants based on Type II fuel without NbC coating. Obviously the NbC coated Type II diffusion constants are closer to the measured 1% release of fission products.

There is, of course, some uncertainty in the release of the fission products determined by post mortem radiochemical analyses of the NERVA fuel; since the fission product losses are small, the measured releases are based on differences in large numbers. For the NRX-A2 reactor test however, a measured release of 1.02% was obtained, while the measured release for the NRX-A3 reactor was 0.90%. Since both reactors were run at essentially identical steady state conditions of power and temperature, similar fission product releases would be expected. On this basis then the 12% difference in the radiochemical measurement of the fission product releases from the two reactors represents fair precision.

Some substantiation for the 1% release of fission products from
(4)
a NERVA type reactor during full power operation has been obtained from LASL.

During full power operation of the Phoebus-1A reactor, samples of fission products in effluent cloud indicated a 1.0% release of the gross fission product inventory.

The predicted 39% release of $\text{Cd}^{115\text{m}}$ is somewhat higher than the 30% measured release. This discrepancy between the two releases for

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NRX-A3 is much less than the discrepancy between a previous prediction of 1.45% release of $\text{Cd}^{115\text{m}}$ as opposed to a measured release of 18.7%, in the NRX-A2 reactor. The low predicted release for $\text{Cd}^{115\text{m}}$ in the NRX-A2 was due to lack of correct diffusion constant for the cadmium precursor, Ag^{115} . Analysis of Ag release from NbC coated, Type II fuel has now provided a proper diffusion constant for this nuclide as evidenced by the agreement between the predicted and measured release fraction for NRX-A3.

As may be seen in Table VII, the concentration of the gross fission product inventory deposited in the non-fueled graphite elements was approximately 3% of the gross fission products released from the fuel. This is higher than the retention of fission products in the non-fueled elements of NRX-A2. The 3% retention is, however, still negligible with respect to the total fission products lost from the fuel. It is therefore believed that fission product release from NERVA fuel element samples represent release from the reactor.

5.0 CONCLUSIONS

As a result of the work reported here the following conclusions can be made:

1. The release of fission products from double laminar coated UC_2 beads, ("Duplex fuel") in a graphite matrix is identical with the release of fission products from Type II, fuel. Both bead coatings had the same thickness.

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2. During a thermal anneal at 2300°C for 15 minutes, complete melting of the metallic components in a unit cell occurs, resulting in complete blockage of 40% of the cooling channels in fuel and non-fueled elements.
3. No graphite to graphite self welding occurs during a thermal anneal at 2300°C for 15 minutes in vacuum.
4. During a simulated loss of coolant accident where a temperature of 2300°C is maintained for 15 minutes, 36% the gross gamma fission product inventory is lost from a unit cell.
5. The loss of fission products from a unit cell predicted by the FIPDIF code is 40% higher than the measured release.
6. Retention of fission products by unfueled portions in the unit cell is negligible except for Ba¹⁴⁰. Approximately 24% of the barium released by the fuel remains in the non-fueled-graphite portions.
7. Approximately 1% of the gross fission product inventory was released during the combined full power runs of NRX-A3.
8. Approximately 3% of the fission products released from the fueled elements in NRX-A3 was retained by adjacent non-fueled elements.
9. The FIPDIF code prediction of fission product release during NRX-A3 full power runs was at least 60% higher than the actual release. The use of diffusion constants which do not consider the

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inhibitory effects of NbC resulted in a prediction of fission product release which was approximately 400% higher than the measured release.

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