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March 1966

Westinghouse Astronuclear Laboratory

PROGRESS REPORT ON THE EXPERIMENTAL INVESTIGATION OF FISSION PRODUCT DIFFUSION

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MAR 31 1966

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Investigation of Fission Product Diffusion, dated March 1966 (2)

Dear Mr. Dooling:

Transmitted herewith for your information are ten (10) copies
of the subject report.

Respectfully,

H. F. FAUGHT

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SECTION 1

INTRODUCTION

This report covers the work performed on the Fission Product Diffusion Program between October 1, 1965, and March 1, 1966. This program is an experimental study to determine the fission product release rate of NERVA fuel.

The objectives of the program during this report period were threefold:

- 1) To determine the effect of NbC coating on the release rate of fission products from NERVA fuel at times and temperatures corresponding to normal reactor operation.
- 2) To determine the effect of NbC coating on the release rate of fission products from NERVA fuel at times and temperatures corresponding to reactor accident conditions.
- 3) To determine the effects of sample size and metallic reactor components on the release rate of fission products from NERVA fuel at times and temperatures corresponding to reactor accident conditions.

The greater part of the work performed to satisfy the first of these objectives has been reported previously.¹ During this report period new work to satisfy this objective consisted of additional post irradiation thermal anneals on NbC coated "as received" fuel.

The second objective is satisfied by the work reported here as fission product release studies from NbC coated "as received" fuel samples which were heated at temperatures of 2300 and 2400°C.

The third objective is satisfied by the post irradiation thermal anneal studies on unit cells, which are clustered assemblies of segments of NERVA fuel elements.¹

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SECTION 2

PROTECTIVE COATING STUDIES

2.1 EXPERIMENTAL METHOD

Sample Preparation

The samples used in these studies consisted of 21 one quarter inch slices cut from a 19 hole, hexagonal NERVA fuel element, designated Type II fuel. The samples were leached in 6 N HCL to remove any exposed UC_2 , then completely coated with a 2 mil thickness of NbC via a vapor deposition process. These samples are now referred to a Type II NbC fuel.

Two batches of samples were irradiated at the Westinghouse Reactor Evaluation Center. During the first irradiation, FP A-33, an average of 2.13×10^{13} fissions per sample was achieved for 12 samples. The second irradiation, FP A-35, achieved an average of 2.56×10^{13} fissions per sample in the remaining nine samples. After irradiation, the samples were returned to WANL for post-irradiation thermal anneals.

Apparatus and Procedure

The gross gamma activity of each sample was measured on a single channel gamma pulse height analyzer. In addition, the number of fissions in each sample was determined in a high pressure ionization chamber, as previously described². The gross gamma activity and the number of fissions per sample are given in table 1.

The samples from irradiation capsule FP A-33 were heated inductively in a vacuum bell jar at a pressure of 1×10^{-4} , over the temperature range of 1800 to 2200°C for times from 300 to 900 seconds. The samples from capsule FP A-35 were heated in the same apparatus at temperatures of 2300 and 2400°C. The samples heated above 2200°C are designated Type III NbC fuel. The time-temperature anneal conditions for each sample is given in table 1. Several samples from each capsule were not heated; these were controls used to determine the initial concentration of the various nuclides.

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TABLE 1

ACTIVITY LEVELS AND THERMAL ANNEAL CONDITIONS OF NbC COATED NERVA FUEL SAMPLES

Irradiation Capsule	Sample Number	Gross Gamma (Counts/min)	Fissions per Sample $\times 10^{13}$	Thermal Anneal Temperature (Deg. C)	Thermal Anneal Time (Seconds)
FP A-33	1	59,342	2.37	Control	Control
	2	54,004	2.17	1800	300
	3	52,474	2.10	1800	600
	4	50,872	2.03	1800	900
	5	44,347	1.79	2000	300
	6	50,919	2.05	2000	600
	7	46,680	1.88	2000	900
	8	47,804	1.94	2200	300
	9	46,998	1.88	2200	600
	10	51,085	2.01	2200	900
	11	61,045	2.47	Control	Control
FP A-35	12	67,536	2.20	Control	Control
	13	62,909	2.07	2300	300
	14	50,741	1.68	2300	600
	15	60,687	1.98	2300	900
	16	53,847	1.86	2300	1800
	17	52,936	1.84	Control	
	18	52,766	1.83	2400	300
	19	54,005	1.88	2400	180
	20	54,372	1.87	2400	600
	21	54,433	1.90	2400	900
	22	56,749	1.97	2400	1800
	23	67,311	2.35	Control	Control

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After the thermal anneals, all samples were gross gamma counted and etched with HF-HNO₃ to remove the NbC coating. The samples were then ignited in O₂; this was followed by dissolution of the residue in HNO₃. The resulting solutions were analyzed radiochemically to determine the amount of fission product released.

Radiochemical Analyses and Calculation of Release Fractions

Duplicate analyses were made on each sample for the following nuclides: Sr⁸⁹, Y⁹¹, Zr⁹⁵, Mo⁹⁹, Ag¹¹¹, Sb¹²⁷, I¹³¹, Ba¹⁴⁰, La¹⁴⁰, and Ce¹⁴¹.

The calculation of the fractional release of each nuclide was determined by comparison of the heated test samples with the control sample as shown in the following example:

- 1) Average gross gamma activity of control samples 12, 17, and 23 = $(67,536 + 52,936 + 56,749)/3 = 59,074$ cpm.
- 2) Gross activity of test sample 13 = 62,909 cpm.
- 3) Average Ba¹⁴⁰ activity in control samples = 49,835 cpm.
- 4) Initial concentration of Ba¹⁴⁰ in test 13 sample
$$= \frac{\text{Gross activity, test sample}}{\text{Gross activity, control sample}} \times \text{Ba}^{140} \text{ activity, control sample}$$
$$= \frac{62,909}{59,074} \times 49,835 = 53,070 \text{ cpm}$$
- 5) Final concentration of Ba¹⁴⁰ in test 13 sample = 40,652 cpm
- 6) Fraction of Ba¹⁴⁰ activity remaining in test sample 13 after thermal anneal
$$= \frac{\text{Final concentration Ba}^{140}}{\text{Initial concentration Ba}^{140}}$$
$$= \frac{40,652}{53,070} = 0.766$$
- 7) The fraction release of Ba¹⁴⁰ from test sample 13 is then
$$1 - 0.766 = 0.234$$

2.2 RESULTS OF PROTECTIVE COATING STUDIES

The fractional release of various fission products from Type II NbC fuel heated at temperatures below 2300°C are given in table 2. These data were obtained to complete the

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TABLE 2

RELEASE FRACTIONS OF FISSION PRODUCTS FROM NbC TYPE II FUEL

Temp. Deg. C	Time (Min)	La ¹⁴⁰	La ^{140*}	Ag ¹¹¹	Ag ^{111*}	Ce ¹⁴¹	Ce ^{141*}	Y ⁹¹	Y ^{91*}	Ba ¹⁴⁰	Ba ^{140*}	Sr ⁸⁹	Sr ^{89*}
1800	5	0.01	0.01	0.98	0.99	0	0.02	0	0	0.01	0	0.05	0.09
	10	0.01	0.03	0.83	0.97	0	0	0	0	0.06	0.01	0.14	0.12
	15	0.01	0.02	0.91	0.98	0	0	0	0	0.02	0.02	0.08	0.12
2000	5	0.02	0.03	0.98	0.97	0	0	0.2	0	0.06	0.04	0.15	0.14
	10	0.05	0	0.99	0.95	0.02	0	0.04	0.02	0.10	0.05	0.20	0.14
	15	0	0.02	0.99	0.97	0.02	0.04	0.01	0	0.10	0.09	0.23	0.19
2200	5	0	0.02	0.99	0.91	0.02	0.03	0.03	0.12	0.18	0.13	0.36	0.25
	10	0.09	0.09	0.99	0.91	0.09	0.10	0.09	0.12	0.32	0.34	0.43	0.38
	15	0.06	0.07	0.99	0.98	0.06	0.10	0.06	0.09	0.39	0.39	0.55	0.46

* The data taken from WANL-TME-1294 are included for comparison with present data.

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fission product release studies on NbC coated "as received" fuel during normal reactor operation. The bulk of these studies has been reported previously¹ however, it is necessary to include in table 2 some of the previously reported data here for the sake of comparison. It had been expected that the new data would clarify the functional relationship of the release fractions with time, unfortunately there is still considerable scatter in the present data.

In general, it was found that the fractional release as a function of time could be approximated by an exponential function. In figure 1, it can be seen that the logarithm fraction of Ba¹⁴⁰ retained is linear with time. Included in figure 1 are both the present data and previously obtained data. The maximum deviation between the two sets of Ba¹⁴⁰ data is approximately 3 percent.

The fractional release from NbC coated samples of Type III NbC fuel heated at 2300 and 2400°C at various times are given in table 3. For most nuclides there was an inhibiting effect on the release from these coated samples as compared with uncoated samples heated under similar conditions. A comparison between the release of various fission from uncoated and coated fuel is given in figure 2. The reduction in release varied from 4.5 percent for Sr⁸⁹ to 60.2 percent for La¹⁴⁰, for samples heated at 2400°C for 15 minutes. Te¹²⁹ was an exception to the general trend; for this nuclide greater release was obtained with coated rather than uncoated samples. This may be due to exchange problems in radiochemical analyses; there is no reason for NbC to enhance tellurium release.

In order to obtain rate constants which describe the fractional release as a function of time, the release data for each nuclide was empirically fitted to an exponential function:

$$f = e^{-Dt} \quad (1)$$

where

- f = Fraction of the nuclide concentration retained
- D = Diffusion rate constant in seconds⁻¹
- t = Time in seconds

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Figure 1. Retention of Ba^{140} in NbC Coated Type II Fuel

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TABLE 3
RELEASE FRACTIONS OF FISSION PRODUCTS FROM NbC TYPE III FUEL

Temperature Deg. C	Time (Min)	La ¹⁴⁰	Mo ⁹⁹	Sb ¹²⁷	I ¹³¹	Ba ¹⁴⁰	Sr ⁸⁹	Te ^{129, 132}
2300	5	0.02	0	0	0.21	0.23	0.47	0.17
	10	0.01	0	0	0.03	0.14	0.37	0.05
	15	0.11	0.02	0.32	0.07	0.49	0.65	0.08
	30	0.28	0.09	0.63	0.23	0.81	0.83	0.38
2400	3	0.09	0.23	0.76	0.30	0.53	0.80	0.49
	5	0.09	0.06	0.52	0.23	0.47	0.67	0.25
	10	0.13	0.03	0.39	0.65	0.78	0.95	0.43
	15	0.14	0.19	0.80	0.67	0.80	0.96	0.62
	30	0.70	0.21	0.89	0.95	1.00	1.00	0.88

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Figure 2. Comparison of Release of Fission Products from Types III and NbC Type III Fuels, at 2400°C, 15 Minutes

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Typical plots showing this exponential relationship are shown for Ba¹⁴⁰, Sr⁸⁹, Y⁹¹, and I¹³¹, in figures 3, 4, 5, and 6, respectively.

Since experiments were run at only two temperatures, determination of the functional relationship of the diffusion constants with temperature was not possible. However, in all previous studies on NERVA fuel the diffusion constants obeyed the Arrhenius equation:

$$D = D_o e^{-\frac{E}{RT}} \quad (2)$$

with

- D_o = Constant
- E = Activation energy in K cals per mole
- R = Gas constant
- T = Temperature in degrees K

The present data have been fitted to this relationship to yield values of D_o and E/R for the nuclides listed in table 4.

These values can be used to represent the release data in the range 2300 to 2400°C, however, extrapolation outside this range is not warranted. Confirmation of the Arrhenius equation will require release studies as a function of time at additional temperatures.

2.3 DISCUSSION OF DIFFUSION RESULTS

NERVA fuel consists of a homogeneous dispersion of 100 micron diameter UC₂ beads coated with a 25 micron thickness of pyrolytic graphite in a graphite matrix. The fuel elements, in the form of a hexagon, 0.43 inch across a face, contain axial cooling channels which are lined with a vapor deposited coat of NbC. If the fuel element is maintained at a temperature of 2200°C for 15 minutes, the UC₂ migrates and degrades the pyrolytic coating of the fuel bead.

In order to establish the mechanism of fission product loss from these complex media during NERVA reactor operation, fission product diffusion studies must be made on the four systems shown in figure 7.

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Figure 3. Retention of Ba^{140} in NbC Coated Type III Fuel

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Figure 4. Retention of Sr^{89} in NbC Coated Type III Fuel

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Figure 5. Retention of Y^{91} in NbC Coated Type III Fuel

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Figure 6. Retention of I^{131} in NbC Coated Type III Fuel

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TABLE 4
DIFFUSION CONSTANTS FOR TYPE III NbC FUEL

Nuclide	E/R (Deg.)	D_0 (Sec ⁻¹)
Sr ⁸⁹	95.5×10^3	1.35×10^{13}
Sb	95.6×10^3	9.27×10^{12}
Ba	74.0×10^3	2.29×10^9
La	63.7×10^3	7.02×10^6
I ¹³¹	174.2×10^3	7.91×10^{21}
Te	107.3×10^3	2.61×10^{14}

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Figure 7. Fission Product Diffusion Paths in UC_2 Fuel

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Post irradiation thermal anneal studies have been made on the following samples, corresponding to the four systems:

- 1) Type II fuel which is the "as received" fuel with unlined cooling channels.
- 2) Type II, NbC fuel which is Type II fuel completely coated with NbC.
- 3) Type III fuel which is the "as received" fuel thermally degraded at 2500°C prior to irradiation.

4) Type III NbC which is Type II NbC which has been subjected to thermal anneals at temperatures above 2200°C, the thermal degradation temperature.

The results of studies on Type II, Type II NbC, and Type III fuel have been reported previously^{1,2}. The results of these studies are summarized by the following statements:

- 1) The diffusion of fission product from Type II fuel can be represented empirically by simple exponential rate equations.
- 2) NbC coatings on Type II fuel inhibits the release rate of fission products. The release of fission products could be represented by simple exponential rate equations.
- 3) The release of fission products from Type III fuel is accelerated by degradation of the pyrolytic graphite coating of the fuel bead.

During the current study it was found that NbC again exhibited an inhibiting effect on the release of fission products. There are at least three possible explanations for this inhibiting effect.

The first possibility is the formation of compounds of fission products with NbC. This reaction may be written for the general case as:

where

M is a metallic fission product.

In this instance the resulting products are carbides of the various fission products plus Nb metal. Investigation of the literature³ discloses the following carbide compounds:

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SrC_2 , BaC_2 , LaC_2 , CeC_2 , ZrC , and MoC . Of the compounds listed, only ZrC and MoC are stable at the temperatures at which the diffusion experiments were made.

The possibility of zirconium reacting with NbC can be determined by consideration of the free energies of formation.

$$\Delta F = F_{\text{ZrC}}^{\circ} - \Delta F_{\text{NbC}}^{\circ}$$

where

ΔF = the free energy of the $\text{Zr} + \text{NbC}$ reaction

$\Delta F_{\text{ZrC}}^{\circ}$ = free energy of formation of ZrC

$\Delta F_{\text{NbC}}^{\circ}$ = free energy of formation of NbC

$$\Delta F_{\text{ZrC}}^{\circ} = -40,000 - 7.7T^4 \quad (3)$$

where

T = temperature in degrees K

$\Delta F_{\text{ZrC}}^{\circ} = -55.40 \text{ K cal/mole}$

$$\Delta F_{\text{NbC}}^{\circ} = -33,980 + 2.45T - 9.27 \times 10^{-3}T + 0.98 \times 10^{-5}T^{-1} \quad (4)$$

$$\Delta F_{\text{NbC}}^{\circ} \text{ at } 2000^{\circ}\text{C} = 32,788 \text{ cal/mole}$$

ΔF for the reaction $\text{Zr} + \text{NbC} \longrightarrow \text{ZrC} + \text{Nb}$

$$= -55.40 + 32.79 + -22.6\Delta \text{K cal/mole}$$

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The large negative value of the free energy indicates favorable conditions for the Zr + NbC reaction to proceed.

A similar calculation for the reaction:

$2 \text{ Mo} + \text{NbC} \longrightarrow \text{Mo}_2\text{C} + \text{Nb}$ yields a free energy of + 32 K cal. This positive value of free energy indicates that this reaction is unfavorable.

In summary, it appears that except for Zr, the possibility of compound formation of fission product with NbC as the cause for the inhibition of diffusion is not likely.

The second possibility* is that NbC interatomic distances are smaller than graphite. In this instance, we are making the assumption that diffusion proceeds through both graphite and NbC via crystalline interstices.

The graphite crystal structure consists of layer planes of hexagonally arranged atoms with tight bindings in the planes and loose binding between the planes.⁵ The interplanar distances as shown in figure 8 is $6.7\text{\AA}/2 = 3.35$ Angstroms.

NbC is a face centered cubic lattice with four atoms associated with each unit cell. For NbC with a density of 7.788 gms/cc and a molecular weight of 104.91, 1 mole occupies $104.91/7.82 = 13.41 \text{ cm}^3$. There are 6.02×10^{23} atoms of niobium in this volume, hence, the total number of unit cells contained is $6.02 \times 10^{23}/4 = 1.51 \times 10^{23}$. The volume of each cell is $13.41/1.51 \times 10^{23} = 8.88 \times 10^{-23} \text{ cm}^3$, thus, the edge of the unit cell is $(8.88 \times 10^{-23})^{1/3} = 4.34 \text{ \AA}$. The distance then between alternate Nb and C atoms is $4.34/2 = 2.17 \text{ \AA}$. Thus the distance between adjacent atoms is less in the NbC lattice than in the graphite lattice.

A relationship between the diffusion constants of diffusing species in various crystal lattices has been derived by Langmuir and Dushman in Barrer's text on diffusion.⁶ This relationship is given by:

$$D = \frac{E}{N_o h} d^2 e^{-\frac{E}{RT}} \quad (5)$$

* For the inhibition of fission product diffusion by NbC.

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where

D	=	is the diffusion constant (cm^2/sec)
E	=	is the activation energy for diffusion in (cals/mole)
N_o	=	is Avogadro's number. (atoms per mole)
h	=	is Planck's constant (erg second)
d	=	is the crystalline interlayer distance ($\text{\AA}^o$)
R	=	is the gas constant (cals/degree-mole)
T	=	is the temperature (degrees K)

For Ba^{140} , the activation energy in Type III fuel was found experimentally¹ to be 36 K cals per mole. Substitution of this value in Equation 5 with temperature equal to 2000°C results in a value of the diffusion constant D, of $8.5 \times 10^{-4} \text{ cm}^2/\text{sec}$ for the graphite lattice and a D of $1.30 \times 10^{-4} \text{ cm}^2/\text{sec}$ for the NbC lattice. Apparently the difference in the lattice spacings between graphite and NbC can reduce the diffusion constant for Ba^{140} by a factor of 6.5. This difference agrees well with the difference in the experimental diffusion contents for Ba^{140} , which are 5.8×10^{-4} in Type II fuel and 6.40×10^{-5} in Type II NbC fuel.

The third possibility* arises if it is assumed that diffusion occurs via pores as a result of differences in void volumes of graphite and NbC. The NbC coating deposited by vapor phase reaction has a density approaching 99 percent of theoretical. The density of the reactor graphite, however, is only 75 percent of the theoretical density. On the basis of the diffusion occurring in pores, one would anticipate much faster diffusion of fission products occurring in the graphite. It is difficult to determine to what extent the diffusion in the graphite occurs by interstitial diffusion and how much occurs via pore diffusion. It is evident, however, that a NbC coating on the surface of a graphite sample can inhibit fission product release regardless of the diffusion process.

Having determined that the NbC coating can retard the release of fission products, the question which now arises is which of the systems, Type II or Type II NbC, is applicable to the NERVA reactor. Two fission product release predictions using the FIPDIF¹ code have

* For the inhibition of fission product diffusion by NbC.

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$$a_o = 2.46 \text{ \AA}$$

$$b_o = 4.28 \text{ \AA}$$

$$c_o = 6.71 \text{ \AA}$$

Figure 8. Graphite Lattice Schematic

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TABLE 5

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

Mass Chain Number	End Member Nuclide	Radiochemical Analysis % Release	Predicted Release With Type II Diffusion Constants	Predicted Release With Type II-NbC Diffusion Constants
89	Sr	1.30	3.92	1.61
91	Y	2.60	5.20	2.80
95	Zr	3.10	2.45	0.66
131	I	1.80	2.52	0.80
137	Cs	0.00	0.12	0.00
140	Ba	1.9	8.64	0.87

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been made. One prediction used diffusion constants which apply to Type II fuel and the other prediction used Type II NbC diffusion constants. In table 5 the two predictions are compared with actual radiochemical measurements of release¹ of several nuclides for the NRX-A3 reactor run. For all nuclides other than Zr^{95} there was better comparison with experimental results using Type II NbC diffusion constants. This would lead us to believe that the primary diffusion path for release of fission products from the NERVA reactor is through the NbC lining into the cooling channels.

The basic conclusion to be drawn from these studies is that diffusion constants based on Type II NbC fuel better represents the release of fission products from NERVA.

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SECTION 3

POST IRRADIATION THERMAL ANNEAL EXPERIMENTS ON UNIT CELLS

Earlier studies on degraded samples of NERVA fuel² provided fission product release rate data which was applicable to a loss of coolant accident following normal reactor operation. The question remains whether this data is applicable to the 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 been made on individual unit cell assemblies, each containing irradiated fuel elements, non-fueled graphite, support plates, orifices, and a tie rod. The geometry of an assembly was chosen to represent a unit cell of the NERVA core and thus provide identical diffusion path lengths for fission product release.

Five unit cell experiments have been completed. The first of these was a feasibility study with non-irradiated fuel to test the heating rates of the vacuum furnace to be used, and to acquaint personnel with the complete experimental operation. The second experiment, in which an irradiated unit cell was to be heated to 2300°C, was not successful due to failure of the unit cell components early in the heating run. The third experiment, unit cell 2, in which an irradiated unit cell was heated to 2300°C, has been reported in WANL-TME-1294, dated September 1965.¹

Two additional unit cell experiments, unit cell 3 and unit cell 4, have been conducted and subsequently analyzed. The descriptions and results of these recent experiments constitute this section of the report.

3.1 EXPERIMENTAL METHOD

Unit Cell Description and Sample Preparation

The unit cell is a cluster of 4 inch long segments of NERVA fuel held together with a modified NERVA tie rod assembly and contained in a hollow graphite cylinder, 3 inches in diameter. The unit cell construction is illustrated schematically in figure 9 and pictorially

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Figure 9. Cross-Sectional View of Unit Cell

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in figure 10. A list of the unit cell components is given in table 6.

The fueled components of the unit cell were irradiated, together with four 1 inch long, full hexagonal, fueled control samples, at the Westinghouse Reactor Evaluation Center for a total of 8.0×14 fissions. Following irradiation, the fueled components were returned to WANL, allowed to decay for five days, then assembled with the remaining unirradiated components into the cluster assembly. The cluster assembly was then inserted into the graphite cylinder and subjected to thermal annealing. Control samples were likewise irradiated but were not subjected to subsequent heating.

Thermal Annealing - Apparatus and Procedure

The annealing operations were performed in a 3000 cycle, 50 kilowatt radio frequency vacuum furnace. The temperature was monitored with a optical pyrometer through a herculite viewport in the furnace wall. During the annealing runs, a dynamic vacuum of 2×10^{-4} mm of Hg. was maintained.

During the individual runs the furnace was brought up to the annealing temperature with the unit cell assembly inside the furnace but not in the elevated temperature zone. When the annealing temperature was reached the assembly was lowered into the "hot" zone. The furnace with the unit cell assembly in the "hot" zone was kept at the annealing temperature for the required length of time. In unit cell 3, the annealing temperature was 2300°C with a 30 minute dwell-time at this temperature. In the case of unit cell 4, the annealing temperature was 2450°C and a 30 minute exposure at this temperature.

This heating procedure differed from the conditions under which unit cell 2 was heated. Unit cell 2 was heated from ambient to 2300°C in situ, requiring approximately 30 minutes to achieve the 2300°C temperature. Unit cells 3 and 4 by insertion into a "hot zone" achieved the thermal anneal temperature in about 1-1/2 minutes. The experimental time-temperature profiles for unit cells 2 and 3 are given in figure 11.

Following the thermal annealing runs, each unit cell was returned to the Physical Sciences Laboratory and disassembled in a shielded glove box. For each experiment, individual

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Figure 10. Exploded View of Unit Cell Assembly

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TABLE 6
COMPONENTS OF UNIT CELL

Item Number	Component Description	Number of Like Components	Component Dimensions and Configuration	Material
1	Fueled Element	6	Regular hexagonal, side 0.437 inch, length 4 inches - eighteen 0.097 inch 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 Element	6	1/2 of a regular hexagon, 4 inches long with eleven 0.097 inch 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.
3	Fueled Element	6	1/4 of a regular hexagon, 4 inches long with five 0.097 inch 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.
4	Unfueled Element	1	Regular hexagon, side 0.437 inch length 4 inches with 1 axial hole.	Graphite, with pyrolytic graphite sleeve.
5	Bottom Support	1	Disc, 2 3/4 inches diameter, 1/8 inches thick provided with 222 cooling holes, 0.97 inch in diameter	Graphite
6	Top Support Plate	1	Disc, 2 3/4 inch diameter, 1/4 inch thick provided with 222 cooling holes 0.97 inch in diameter with orifice jets inserted.	Silver, Indium, Cadmium, Stainless Steel
7	Tie Rod Assembly	1	Length 9.5 inches, diameter 0.111 inch plus assorted hardware	Inconel
8	Graphite Can	1	Cylindrical tube, closed at one end, inside diameter, 2 3/4 inch, outside diameter, 3.03 length 11 1/2 inches	Graphite

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Figure 11. Heating Profiles for Unit Cells 2 and 3

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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 individually ashed, using an oxygen combustion technique, dissolved and analyzed radiochemically.

Sample Selection and Dissolution

Representative samples from each unit cell were selected for radiochemical analysis.

In the unit cell three experiment samples were chosen as follows:

- 1) Two-full hexagonal fuel elements, 4 inches long. (Item 1, table 6)
- 2) Two-1/2 hexagonal fuel elements, 4 inches long. (Item 2, table 6)
- 3) Two-1/4 hexagonal fuel elements, 4 inches long. (Item 3, table 6)
- 4) One-full hexagonal unfueled element, 4 inches long. (Item 4, table 6)
- 5) A 10 percent portion of the graphite can. (Item 8, table 6)
- 6) Two-full hexagonal, fueled control samples, 1 inch long

In unit cell 4 the samples were chosen as follows:

- 1) Three-full hexagonal fuel elements, 4 inches long. (Item 1, table 6)
- 2) Three-1/2 hexagonal fuel elements, 4 inches long. (Item 2, table 6)
- 3) Three-1/4 hexagonal fuel elements, 4 inches long. (Item 3, table 6)
- 4) One-full hexagonal unfueled element, 4 inches long. (Item 4, table 6)
- 5) A 10 percent portion of the graphite can. (Item 8, table 6)
- 6) Three-full hexagonal fueled control samples, 1 inch long

The individual samples noted above were treated in the following manner. Selected samples with NbC lining were first leached with HF-HNO_3 to remove the lining. The samples were then combusted in a quartz tube in an atmosphere of flowing oxygen. The residual ash in the ignition tube was dissolved in 6N-HNO_3 and diluted to a known volume in each case.

Radiochemical analyses were then performed in duplicate with aliquots from the known volume of solution to determine the concentration of the following nuclides: Sr^{89} , Y^{91} , Zr^{95} , I^{131} , Ba^{140} , and $\text{Ce}^{141-144}$ (in the unit cell 3 experiment). In addition to the above noted

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nuclides, concentrations of $\text{Te}^{129\text{m}}$ and Mo^{99} in unit cell 4, were determined. The determination of the various fractional releases was based on concentrations present in the unheated control samples as discussed in WANL-TME-1294, Calculation of Release Fractions, Section 3.14.

3.2 RESULTS OF UNIT CELL EXPERIMENTS

Visual Examination

Complete melting of the metal tie rod assembly, orifices and support plate occurred during the thermal annealings in both unit cell 3 and 4. Although vaporization of metallic components did occur, there was little deposition on individual fuel element sections and less coolant channel blockage than was present in unit cell 2.

Radiochemical Analysis and Diffusion Results

The release fractions of the various nuclides as well as the gross gamma inventory release from the regular hexagonal elements, 1/2 hexagonal elements, and 1/4 hexagonal segments in unit cell 3 and unit cell 4 are given in tables 7 and 8 respectively. Included are the values for the fractional retention of the nuclides, by the center unfueled element and the outer graphite can. Also included are the effective release fractions from the respective unit cells. These values were calculated by subtracting the total fraction of the nuclide inventory which was retained by the unfueled portions of the unit cells, from the average fractional release from the fuel in each case.

Certain values previously reported for unit cell 2 were found to be in error because they had not been weighted. The release values for the individual nuclides in the columns headed "Average Fractional Release from Fuel" and "Effective Release from Unit Cell" have been changed to reflect these corrections. An arithmetic error in the cerium results was also corrected. The unit cell 2 data, as corrected, is included in table 9 for comparison.

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

TABLE 7

FRACTIONAL RELEASE OF FISSION PRODUCTS FROM UNIT CELL 3
(THERMAL ANNEAL-30 MINUTES AT 2300°C)

Nuclide	Fractional Release from Regular Hexagonal Fuel Elements	Fractional Release from 1/2 Hexagonal Fuel Elements	Fractional Release from 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	0.89	0.94	0.94	0.92	0	0.09	0.83
Zr ⁹⁵	0	0	0	0	0	0	0
I ¹³¹	0.22	0.31	0.36	0.28	0	0	0.28
Ba ¹⁴⁰	0.57	0.73	0.82	0.67	0.02	0.24	0.41
Ce ¹⁴¹⁻¹⁴⁴	0.05	0.15	0.22	0.11	0.01	0.05	0.05
Y ⁹¹	0.02	0.03	0.11	0.04	0.01	0.02	0.02
Gross γ Inventory	0.25	0.32	0.36	0.29	0.01	0.08	0.21

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TABLE 8

FRACTIONAL RELEASE OF FISSION PRODUCTS FROM UNIT CELL 4
(THERMAL ANNEAL-30 MINUTES AT 2450°C)

Nuclide	Fractional Release from Regular Hexagonal Fuel Elements	Fractional Release from 1/2 Hexagonal Fuel Elements	Fractional Release from 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}	0.94	0.97	0.98	0.96	0.01	0	0.95
Zr^{95}	0.01	0.02	0.07	0.03	0	0	0.03
I^{131}	0.30	0.35	0.33	0.32	0	0	0.32
Ba^{140}	0.48	0.77	0.83	0.65	0.02	0.12	0.51
$\text{Ce}^{141-144}$	0	0.11	0.21	0.08	0.01	0.06	0.01
Y^{91}	0.06	0.11	0.18	0.10	0	0.01	0.09
Mo^{99}	0.02	0.04	0.06	0.03	0	0	0.03
$\text{Te}^{129\text{m}}$	0.23	0.24	0.37	0.26	0	0.01	0.25
Gross γ Inventory	0.21	0.30	0.34	0.26	0.01	0.07	0.19

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TABLE 9

FRACTIONAL RELEASE OF FISSION PRODUCTS FROM UNIT CELL 2
(THERMAL ANNEAL-15 MINUTES AT 2300°C)

Nuclide	Fractional Release from Regular Hexagonal Fuel Elements	Fractional Release from 1/2 Hexagonal Fuel Elements	Fractional Release from 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}	0.74	0.86	0.88	0.81	0.03	0	0.77
Zr^{95}	0.04	0.06	0.06	0.05	0.01	0	0
I^{131}	0.67	0.60	0.62	0.61	0	0	0.61
La^{140}	0.30	0.46	0.46	0.35	0.01	0	0.34
Ba^{140}	0.59	0.74	0.73	0.66	0.15	0.01	0.50
$\text{Ce}^{141-144}$	0.03	0.32	0.20	0.16	0.04	0.06	0.06
Gross γ Inventory	0.31	0.38	0.40	0.36	--	--	0.36

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Except for Ba¹⁴⁰, in unit cell 2, 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 from the fuel.

3.3 DISCUSSION OF EXPERIMENTAL RESULTS

In order to check the validity of the diffusion constants used for the prediction of fission product release during normal operation of NERVA type reactors comparison can be made of measured fission product release from reactor fuel with predictions based on Type II or Type II NbC diffusion constants. Such a comparison was made earlier in this report where it was found that a prediction based on Type II NbC diffusion constants gave a better agreement with measured releases of fission products during normal operation.

An analogous procedure for comparison of predicted releases with measured releases from a reactor subjected to loss of coolant conditions would require post mortem examination of fuel from a reactor that had experienced such a loss of coolant. Since this fuel is not available, a compromise has been made utilizing small reactor models called unit cells.

Three experiments have been made in which two unit cells were heated to a maximum of 2300°C while a third unit cell was heated to a maximum temperature of 2450°C.

The relative magnitudes of fission product release from the unit cells heated to 2300°C appears incongruous since greater release was obtained from unit cell heated to 2300°C from the unit cell heated 30 minutes. This incongruity can be resolved if the experimental heating profiles shown in figure 11 are considered. Unit cell 2 required approximately 28 minutes to reach thermal equilibrium at 2300°C where it held at this temperature for 30 minutes. Since diffusion is a time-temperature dependent phenomena, fission product release will have occurred from unit cell 2 during the long period required to reach thermal equilibrium.

In a previous report¹ a comparison was made between the measured effective release of fission products and a predicted release based on Type III diffusion constants. At that time it was found that the predicted release of various nuclides was greater than the measured release. Two possible reasons for this difference were offered, one being blockage of cooling channels

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by redeposited metal vapor and the other the use of Type III diffusion constants that did not account for the effect of NbC coating on fission product release.

In the two unit cell experiments, 3 and 4, completed in the report period, the coolant channels were found to be free of redeposited metal, and again a discrepancy occurs between Type III prediction and measured fission product release. Thus it seems that the coolant channel blockage is not the cause of the discrepancy between the measured and predicted releases.

As has been pointed out, the diffusion constants and activation energies given in Table 4 for Type III NbC fuel are preliminary. Nevertheless, it is instructive to attempt a comparison between predicted releases based on these constants and the measured releases for the unit cell. In table 10, the measured release of Sr^{89} and Ba^{140} are compared with predictions based on Type III and Type III NbC diffusion constants. It appears that the Type III NbC data provides better agreement with the measured releases. This finding tends to reinforce our hypothesis that the release of fission products from the NERVA reactor proceeds by diffusion through the NbC coating in the fuel element cooling channels.

Although there are some indications that Type III NbC diffusion constants are adequate for describing the effective release of Sr^{89} from unit cells, it does not necessarily follow that these same diffusion constants also apply to other nuclides.

Of particular concern is the apparent effect of the sample size on the release of other fission products. In table 8, it may be seen that the release of Zr^{95} , Ba^{140} , $\text{Ce}^{141-144}$, Y^{91} , Mo^{99} and Te^{129} is greatest from the 1/4 hexagonal elements and least from the regular elements. These results raise questions concerning which fuel geometry is described by the Type III NbC data, and which fuel geometry best describes release from the reactor.

Since the reactor core consists of regular fuel elements and contains none of the 1/2 and 1/4 hexagonal elements, it is logical to assume that the release of fission products from the regular fuel elements best describes release from the reactor.

In view of these considerations the best method of obtaining diffusion constants which describe release of fission products from the NERVA reactor during loss of coolant conditions is by further unit cell experiments. Unit cell experiments as presently conducted suffer the

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TABLE 10
COMPARISON OF MEASURED FISSION PRODUCT RELEASES FROM UNIT CELLS
WITH PREDICTED RELEASES

Unit Cell Number and Experimental Conditions	% Sr ⁸⁹ Release Radiochemical Determination	% Sr ⁸⁹ Release Predicted with Type III, Diffusion Constants	% Sr ⁸⁹ Release Predicted with Type III-NbC Diffusion Constants	% Ba ¹⁴⁰ Release Radiochemical Determination	% Ba ¹⁴⁰ Release Predicted with Type III, Diffusion Constants	% Ba ¹⁴⁰ Release Predicted with Type III, NbC, Diffusion Constants
2 Heated 2300°C for 15 minutes 26 minutes to achieve temp. equilibrium	77	100	61	50	100	47.3
3 Heated to 2300°C for 30 minutes. 1.5 minutes to achieve temp. equilibrium	83	100	85	41	100	72.2
4 Heated to 2450°C for 30 minutes. 1.5 minutes to achieve temp. equilibrium	95	100	97	50	100	83

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disadvantage of time consuming radiochemical analyses. In order to eliminate some of this disadvantage it is suggested that in future experiments radiochemical analyses be restricted to regular fuel elements. Although $1/2$ and $1/4$ fuel segments are necessary to provide diffusional geometry simulation to the reactor core, fission product measurements from these partial elements are not required.

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SECTION 4

CONCLUSIONS

As a result of the work reported here, the following conclusions can be drawn.

- 1) NbC coating inhibits the release of fission products from Type III NERVA fuel. At 2400°C, the reduction in release varied from 4.5 percent for Sr^{89} to 60.2 percent for La^{140} .
- 2) Diffusion constants derived from studies on NbC coated Type II fuel samples predict a 1.6 percent gross release of fission products from NERVA during normal operation. This predicted release is in better agreement with the 1.0 percent measured release than the 3.2 percent release predicted by uncoated Type II diffusion constants.
- 3) Diffusion constants derived from Type III fuel predict a 100 percent release of Sr^{89} and Ba^{140} which is higher than the measured release of 83 percent for Sr^{89} and 41 percent for Ba^{140} for a unit cell heated 2300°C for 30 minutes.
- 4) Diffusion constants derived from NbC covered Type III fuel samples also give a predicted value of release which is greater than the measured release values.
- 5) Modified unit cell experiments should be performed in future studies to obtain diffusion constants that describe the release of fission products from NERVA during loss of coolant conditions.

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SECTION 5

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