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PART II: PRELIMINARY INVESTIGATION OF THE FISSION PRODUCT
RETENTION OF METAL AND CERMET COMPACTS

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PART II: PRELIMINARY INVESTIGATION OF THE FISSION PRODUCT
RETENTION ABILITY OF METAL AND CERMET COMPACTS

Abstract

Between 360° C and 560° C, the compacts composed of .010 in. of uranium-aluminum alloy clad in .020 in. of aluminum lost approximately .01% to 2.0% of the volatile fission products. Between 900° C and 1100° C, the elements composed of .010 in. of stainless steel-UO₂ clad in .010 in. of stainless steel allowed approximately .01% to .03% of volatile fission products to escape. A dark surface crust or film (resembling rust) was observed on the stainless steel specimens. The Al₂O₃-Cr element with .020 in. cladding allowed approximately .04% to 1.5% of the volatile fission products to escape between 1000° C and 1100° C. The .025 in. cladding of MgO-Ni allowed approximately 2.4% to 12% of the volatile fission products to escape between 1000° C and 1100° C.

047-002

Introduction

The diffusion of fission products through solids is of great importance in the design and construction of nuclear power reactors. A study of the diffusion of fission products through simulated fuel elements has been in progress at ORNL since 1953^{1,2}. This study was continued at North Carolina State College during the academic year of 1954-55 and the results are contained herein.

Experimental Procedure

The experimental procedure is described in detail in a previous report². In brief, the specimens were irradiated in the X-10 Graphite Reactor for a period of 7 days in a flux of $5 \times 10^{11} \text{ n/cm}^2/\text{sec}$. and shipped by air express to North Carolina State College. Following irradiation, the specimens were enclosed in alumina-silicate wool and sealed in a quartz glass tube with a condenser projection. After heating for 24 hours at selected temperatures, the condenser projections were cooled for 4 hours in liquid air, sealed off, and analyzed with the aid of a gamma ray spectrometer. The alumina-silicate "wool" used to enclose the specimens was also examined for diffused fission products. Specimens were washed before and after heating and the washes were examined for radioactive fission products. The fission product retention factor was determined by comparing activities from gamma ray spectrometer peaks with the UO_2 powder standard, Specimen G-33-3. Figure 1 shows the activity of the standard as determined by different methods.

¹Metallurgy Division Semi-Annual Progress Report for period ending October 10, 1954, ORNL-1875, 1955.

²W. E. Moody, A. J. Taylor, J. R. Johnson, ORNL-1778, 1955.

A standard quantity consisting of UO_2 powder in a glass ring in a quartz glass container provided with a friable stem was irradiated in the X-10 pile for 7 days, (specimen G-224-12). This container was then sealed into a standard process tube with an aluminum-silicate wool filter between the condensing stem of the process tube and the container. The friable stem on the inner container was then broken allowing the fission products to escape. They were condensed in the normal manner without any heating period.

Physical descriptions of the test specimens are given in Table I.

Discussion of Results

It is estimated that the specimens reached a temperature of $100^\circ\text{--}200^\circ\text{C}$ during irradiation and that some of the gaseous fission products may have been lost during this time. In the case of the unheated specimen, G-224-12, Table III, about 22% of the Xe^{133} and 0.62% of the I^{133} calculated to have been generated was collected in the liquid air trap. 62% of the Xe^{133} was collected from the standard specimen, G-33-3, processed normally. This, together with the 22% accounted for as loss during irradiation, is 84% of the Xe^{133} calculated to have been generated. This relatively high accountability value would indicate the adequacy of the experimental procedure. Fission product losses during irradiation of the cermet and metal specimens have been assumed to be negligible as the diffusion rate of isotopes such as Xe and I at the low irradiation temperatures have been calculated to be very low.

The "A" series were specimens of $\text{Cr-Al}_2\text{O}_3$ which had been fabricated so as to eliminate the tungsten impurity which was present in specimens reported previously. Tests on these specimens indicated that there was a factor of about 10^2 improvement in fission product retention at 1000°C over retention in previously tested specimens.

The "H" series specimens, which were an aluminum clad uranium-aluminum alloy, were too large for the available quartz glass tubes and it was necessary to roll the specimens into a coil. The rolling probably caused breaks in the aluminum cladding and resulted in greater diffusion of fission products. However, the relatively low temperatures used would account for the low diffusion rates. Some La^{140} was found on the specimens after heating.

The results for the MgO-Ni specimens, D-33-14, D-33-15 and D-33-16, are probably in error as the gamma spectrometer was not functioning properly at the time of measurement.

The amount of activity found in all washes before and after heating was relatively small and in many cases could not be analyzed.

Other data of Table II are self explanatory. Table IV gives detailed results of the diffusion from each specimen.

Acknowledgment

The assistance of the Ceramic Laboratory, Metallurgy Division, ORNL, in particular J. M. Warde and J. R. Johnson, is greatly appreciated.

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TABLE I

PROPERTIES OF COMPACTS

Compact Specimen No.	Composition Cladding	Cladding Thickness (in.)	Core Composition	Amount Uranium	Core Thickness (in.)	Core Size (in.)	Specimen Size (in.)
A series	Al ₂ O ₃ -28% Cr -72%	.020	UO ₂ + Al ₂ O ₃ + Cr	3.3 mg UO ₂	.010	.35 D.	.050 x .450 D.
D Series	MgO -80% Ni -20%	.025	UO ₂ + MgO+ Ni	3.3 mg. UO ₂	.025	.35 D.	.075 x .450 D.
H Series	Al	.020	U+Al Alloy	16.49 mg. U metal .4% enrichment	.010	.4 x 2.0	.050 x .675 x 2.325
SS Series	Stainless Steel No. 304	.010	UO ₂ + Stainless Steel Chips+ B ₄ C .375 gm. Core wt.	74 gm. UO ₂ .713 mg. B ₄ C	.010	.25 x .875 Oval shape	.030 x .5 x 1.0

TABLE II

DIFFUSION RESULTS FOR COMPACTS

Compact Specimen No.	Temp. of Heating	Type of Activity Detected as Products of Diffusion	Uranium Compound in Compact	Ratio of Diffused Activity to Activity of G-33-3 Standard* (Fission Product) (Retention Factor)	Remarks
A-33-11	1000° C	Xe	UO ₂	3.7×10^{-4}	
A-33-12	1100° C	Xe, I, Te	"	2.5×10^{-3}	
A-33-13	1100° C	Te, I	"	1.5×10^{-2}	
A-33-14	1100° C	Xe, I, Te	"	1.1×10^{-2}	
D-33-11) D-33-12) D-33-13)	Used to set up and check equipment.				Specimens D-33-11, D-33-12 and D-33-13 probably in error as gamma ray spectrometer was slightly out of calibration.
D-33-14	900° C	Xe, I, Te	UO ₂	2.5×10^{-2}	
D-33-15	900° C	"	"	1.4×10^{-2}	Identification of Np in surface wash of some specimens.
D-33-16	900° C	"	"	6.6×10^{-3}	
D-33-17	1100° C	"	"	1.3×10^{-1}	
D-33-18	1100° C	"	"	5.9×10^{-2}	
D-33-19	1100° C	"	"	9.6×10^{-2}	
D-33-20	1000° C	"	"	3.6×10^{-2}	
D-33-21	1000° C	"	"	6.1×10^{-3}	
D-33-22	1000° C	"	"	5.1×10^{-3}	
G-1300-11	25° C		UO ₂		Too radioactive to make satisfactory measurements.
G-224-11	25° C	Xe, I, Te, Mo	UO ₂	Standard	
H-1	560° C	Xe, I, Te	U-Al Alloy	1.52×10^{-2}	La ¹⁴⁰ found in surface wash after heating in H series.
H-2	460° C	Xe	" "	1.1×10^{-4}	
H-3	460° C	Xe	" "	8.8×10^{-5}	
H-4	360° C	Xe	" "	8.5×10^{-5}	
H-5	360° C	Xe	" "	1.1×10^{-4}	
SS-740-1	900° C	Xe, I, Mo	UO ₂	7.6×10^{-5}	Some activity at 1 mev gamma that could not be identified. Dark surface crust formed on 900° C specimen.
SS-740-2	1000° C	Xe, I, Te	"	2.4×10^{-5}	
SS-740-3	1100° C	I, Te	"	2.8×10^{-4}	

* Calculated from activities under gamma ray spectrometer peaks. 647 007

TABLE III

TABULATED RESULTS FOR TUBE FROM STANDARD COMPACT

(Not Heated)

Compact Specimen No.	Isotope	Theoretical Amount Fission Products Present at Seal off of Tube Time	Measured Fission Products in Tube at Seal off Time	% Fission Products Collected
G-224-12	Xe ¹³³	17.61×10^{11}	3.37×10^{11}	22.2%
	I ¹³³	5.64×10^{11}	22.5×10^8	0.624%

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TABLE IV

ISOTOPE DIFFUSION RESULTS

Compact Specimen No.	Heating Time Hr.	Elapsed Time from Flux to Furnace Removal Hr.	Elapsed time from Furnace Removal to Measurement Hr.	Fission Products Present at removal from Furnace T_2	Calculated Fission Products Diffused Through Specimen T_2 at Removal from Furnace	Fission Products Present at time of Gamma Spectrometer Analysis of Tube T_3
A-33-11	24	52-1/2	23-3/4	I133-1.35 x 10 ¹⁰ Xe133-2.36x 10 ¹¹ Te132-1.30x 10 ¹¹ I132 -4.20x 10 ⁹ I131 -1.83x 10 ¹⁰	Xe133-2.83 x 10 ⁸	Xe133-2.46 x 10 ⁸
A-33-12	24	76-1/2	23-1/4	I133-6.26 x 10 ⁹ Xe133-2.17x 10 ¹¹ Te132-1.05x 10 ¹¹ I132 -3.38x 10 ⁹ I131 -1.67x 10 ¹⁰	Xe133-1.07 x 10 ⁸ Te132-1.04 x 10 ⁸ I131 -1.52 x 10 ⁹ I132 -1.73 x 10 ⁹ I133 -6.43 x 10 ⁶	Xe133-9.6 x 10 ⁷ Te132-8.43 x 10 ⁷ I131 -1.40 x 10 ⁹ I132 -4.85 x 10 ⁶ I133 -3.05 x 10 ⁶
A-33-13	19	29-1/2	51	I133 -2.78x 10 ¹⁰ Xe133-2.53x 10 ¹¹ Te132-1.60x 10 ¹¹ I132 -3.11x 10 ⁹ I131 -1.98x 10 ¹⁰	Te132-4.32 x 10 ⁹ I132 - Small	Te132-2.72 x 10 ⁹ I132 -1.31 x 10 ⁷
A-33-14	19	29-1/2	6-1/2	I133 -2.78x 10 ¹⁰ Xe133-2.53x 10 ¹¹ Te132-1.60x 10 ¹¹ I132 -3.11x 10 ⁹ I131 -1.98x 10 ¹⁰	Te132-1.96 x 10 ⁹ I132 -Small Mo99 -4.71 x 10 ⁷ I133 -1.36 x 10 ⁸	Te132-1.85 x 10 ⁹ I132 -4.36 x 10 ⁷ Mo99 -4.41 x 10 ⁷ I133 -1.10 x 10 ⁸
D-33-14	24	35	44-1/2	Xe133-2.51x 10 ¹¹ I133 -2.36x 10 ¹⁰ Te132-1.53x 10 ¹¹ I132 -5.10x 10 ⁹ I131 -1.95x 10 ¹⁰	Xe133-8.55 x 10 ⁹ Te132-2.69 x 10 ⁹ I131 -5.99 x 10 ⁹ I133 -7.62 x 10 ⁸ I132 -Small	Xe133-7.12 x 10 ⁹ Te132-1.50 x 10 ⁹ I131 -5.1 x 10 ⁹ I133 -1.84 x 10 ⁸ I132 -2.07 x 10 ⁷

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TABLE IV (CONT'D)

Compact Specimen No.	Heating Time Hr.	Elapsed Time from Flux to Removal Hr.	Elapsed time from Furnace Removal to Measurement Hr.	Fission Products Present at removal from Furnace T_2	Calculated Fission Products Diffused Through Specimen at Removal from Furnace T_2^1	Fission Products Present at time of Gamma Spectrometer Analysis of Tube T_3
D-33-15	24	35	45	Xe133-2.51 x 10 ¹¹ I133 -2.36 x 10 ¹⁰ Te132-1.53 x 10 ¹¹ I132 -5.10 x 10 ⁹ I131 -1.95 x 10 ¹⁰	Xe133-6.37 x 10 ⁹ I133 -1.93 x 10 ⁸ Te132-5.42 x 10 ⁸ I131 -3.13 x 10 ⁹	Xe133-5.05 x 10 ⁹ I133 -4.6 x 10 ⁷ Te132-3.57 x 10 ⁸ I132 -1.62 x 10 ⁷ I131 -2.67 x 10 ⁹
D-33-16	24	35	45-1/2	I133 -2.36 x 10 ¹⁰ Xe133-2.51 x 10 ¹¹ Te132-1.53 x 10 ¹¹ I132 -5.10 x 10 ⁹ I131 -1.95 x 10 ¹⁰	Xe133-5.02 x 10 ⁹ Te132-6.84 x 10 ⁷ I131 -1.24 x 10 ⁸	Xe133-3.86 x 10 ⁹ Te132-4.51 x 10 ⁷ I131 -1.05 x 10 ⁸
D-33-17	18	30	6-1/4	Xe133-2.53 x 10 ¹¹ I133 -2.74 x 10 ¹⁰ Te132-1.60 x 10 ¹¹ I132 -5.04 x 10 ⁹ I131 -1.98 x 10 ¹⁰	Xe133-4.47 x 10 ⁹ I133 -3.52 x 10 ⁹ I131 -9.02 x 10 ⁹	Xe133-4.9 x 10 ⁹ I130 -2.85 x 10 ⁹ I131 -8.79 x 10 ⁹ (?)
D-33-18	18	30	6-3/4	I133 -2.74 x 10 ¹⁰ Xe133-2.53 x 10 ¹¹ Te132-1.60 x 10 ¹¹ I132 -5.04 x 10 ⁹ I131 -1.98 x 10 ¹⁰	Xe133-3.97 x 10 ⁸ I133 -2.14 x 10 ⁹ I131 -6.50 x 10 ⁹	Xe133-.774 x 10 ⁹ I133 -1.71 x 10 ⁹ I131 -6.32 x 10 ⁹ Mo ⁹⁹ (?)
D-33-19	18	30	7-1/4	I133 -2.74 x 10 ¹⁰ Xe133-2.53 x 10 ¹¹ Te132-2.1 x 10 ¹¹ I132 -5.63 x 10 ⁹ I131 -2.21 x 10 ¹⁰	Xe133-5.54 x 10 ⁹ I133 -4.23 x 10 ⁹ I131 -16.1 x 10 ⁹	Xe133-6.09 x 10 ⁹ I133 -3.34 x 10 ⁹ I131 -15.7 x 10 ⁹ Mo ⁹⁹ (?)
D-33-20	24	52-1/2	6	Xe133-2.36 x 10 ¹¹ I133 -1.35 x 10 ¹⁰ Te132-1.30 x 10 ¹¹ I132 -4.20 x 10 ⁹ I131 -1.83 x 10 ¹⁰	Xe133-6.37 x 10 ⁹ I133 -9.70 x 10 ⁸ I131 -7.05 x 10 ⁹	Xe133-6.27 x 10 ⁹ I133 -7.95 x 10 ⁸ I131 -6.9 x 10 ⁹ Te132 (?)

TABLE IV (CONT'D)

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Compact Specimen No.	Heating Time Hr.	Elapsed Time from Flux to Furnace Removal Hr.	Elapsed time from Furnace Removal to Measurement Hr.	Fission Products Present at removal from Furnace T_2	Calculated Fission Products Diffused Through Specimen at Removal from Furnace T_{21}	Fission Products Present at time of Gamma Spectrometer Analysis of Tube T_3
D-33-21	24	52-1/2	6-3/4	$I^{133} -1.35 \times 10^{10}$ $Xe^{133} -2.36 \times 10^{11}$ $Te^{132} -1.30 \times 10^{11}$ $I^{132} -4.20 \times 10^9$ $I^{131} -1.83 \times 10^{10}$	$Xe^{133} -2.95 \times 10^8$ $I^{133} -3.22 \times 10^8$ $I^{131} -2.24 \times 10^9$	$Xe^{133} -3.39 \times 10^8$ $I^{133} -2.57 \times 10^8$ $I^{131} -2.18 \times 10^9$ (?)
D-33-22	24	52-1/2	21-3/4	$I^{133} -1.35 \times 10^{10}$ $Xe^{133} -2.36 \times 10^{11}$ $Te^{132} -1.30 \times 10^{11}$ $I^{132} -4.20 \times 10^9$ $I^{131} -1.83 \times 10^{10}$	$Xe^{133} -2.72 \times 10^8$ $I^{133} -2.70 \times 10^8$ $I^{131} -2.61 \times 10^9$	$Xe^{133} -3.64 \times 10^8$ $I^{133} -1.35 \times 10^8$ $I^{131} -2.4 \times 10^9$ (?)
G-224-12	0	14	38	$Xe^{133} -17.61 \times 10^{11}$ $I^{133} -3.61 \times 10^{11}$ (Seal off time)	$Xe^{133} -3.73 \times 10^{11}$ $I^{133} -22.5 \times 10^8$ (Seal off time)	$Xe^{133} -3.03 \times 10^{11}$ $I^{133} -6.7 \times 10^8$ (Measurement Time)
Escape						
H-1	18	30	252-1/2	$I^{133} -9.08 \times 10^{10}$ $Xe^{133} -8.39 \times 10^{11}$ $Te^{132} -5.30 \times 10^{11}$ $I^{132} -1.67 \times 10^{10}$ $I^{131} -6.56 \times 10^{10}$	$Xe^{133} -Small$ $I^{133} -2.04 \times 10^{10}$ $Te^{133} -5.74 \times 10^8$ $I^{132} -Small$ $I^{131} -2.12 \times 10^9$	$Xe^{133} -4.29 \times 10^9$ $I^{133} -6.75 \times 10^6$ $Te^{132} -5.91 \times 10^7$ $I^{132} -3.19 \times 10^6$ $I^{131} -8.76 \times 10^8$
H-2	24	54	5-1/2	$I^{133} -4.27 \times 10^{10}$ $Xe^{133} -7.82 \times 10^{11}$ $Te^{132} -4.25 \times 10^{11}$ $I^{132} -1.38 \times 10^{10}$ $I^{131} -6.03 \times 10^{10}$	$Xe^{133} -2.50 \times 10^8$	$Xe^{133} -2.40 \times 10^8$
H-3	24	54	6	$I^{133} -4.27 \times 10^{10}$ $Xe^{133} -7.82 \times 10^{11}$ $Te^{132} -4.25 \times 10^{11}$ $I^{132} -1.38 \times 10^{10}$ $I^{131} -6.03 \times 10^{10}$	$Xe^{133} -1.99 \times 10^8$	$Xe^{133} -1.91 \times 10^8$

*4 hour cooling period only.

TABLE IV (CONT'D)

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Compact Specimen No.	Heating Time Hr.	Elapsed Time from Flux to Furnace Removal Hr.	Elapsed time from Furnace Removal to Measurement Hr.	Fission Products Present at removal from Furnace T_2	Calculated Fission Products Diffused Through Specimen at Removal from Furnace T_2^1	Fission Products Present at time of Gamma Spectrometer Analysis of Tube T_3
H-4	24	78	4	$I^{133} - 1.98 \times 10^{10}$ $Xe^{133} - 7.16 \times 10^{11}$ $Te^{132} - 3.46 \times 10^{11}$ $I^{132} - 1.11 \times 10^{10}$ $I^{131} - 5.53 \times 10^{10}$	$Xe^{133} - 1.73 \times 10^8$	$Xe^{133} - 1.67 \times 10^8$
H-5	24	78	4-1/2	$I^{133} - 1.98 \times 10^{10}$ $Xe^{133} - 7.16 \times 10^{11}$ $Te^{132} - 3.46 \times 10^{11}$ $I^{132} - 1.11 \times 10^{10}$ $I^{131} - 5.53 \times 10^{10}$	$Xe^{133} - 2.12 \times 10^8$	$Xe^{133} - 2.04 \times 10^8$
SS-740-1	24	35	44	$I^{133} - 5.06 \times 10^{11}$ $Xe^{133} - 5.63 \times 10^{12}$ $Te^{132} - 3.43 \times 10^{12}$ $I^{132} - 1.14 \times 10^{11}$ $I^{131} - 4.37 \times 10^{11}$	$Xe^{133} - 5.42 \times 10^7$ $I^{131} - 1.62 \times 10^9$ $Mo^{99} - 4.25 \times 10^7$	$Xe^{133} - 4.22 \times 10^7$ $I^{131} - 1.39 \times 10^9$ $Mo^{99} - 2.87 \times 10^7$
SS-740-2	24	52-1/2	22	$I^{133} - 3.02 \times 10^{11}$ $Xe^{133} - 5.29 \times 10^{12}$ $Te^{132} - 2.92 \times 10^{12}$ $I^{132} - 9.42 \times 10^{10}$ $I^{131} - 4.10 \times 10^{11}$	$Xe^{133} - 2.18 \times 10^8$ $Te^{132} - 1.95 \times 10^7$ $I^{132} - 1.92 \times 10^8$ $I^{133} - 1.98 \times 10^7$ $I^{131} - 1.91 \times 10^7$	$Xe^{133} - 2.01 \times 10^8$ $Te^{132} - 1.60 \times 10^7$ $I^{132} - 8.62 \times 10^5$ $I^{133} - 9.85 \times 10^6$ $I^{131} - 1.76 \times 10^7$
SS-740-3	19	29-1/2	72-1/2	$I^{133} - 6.24 \times 10^{11}$ $Xe^{133} - 5.67 \times 10^{12}$ $Te^{132} - 3.59 \times 10^{12}$ $I^{132} - 6.96 \times 10^{10}$ $I^{131} - 4.45 \times 10^{11}$	$Te^{132} - 1.61 \times 10^9$ $I^{132} - \text{Small}$	$Te^{132} - 8.4 \times 10^8$ $I^{132} - 1.96 \times 10^7$

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