FEASIBILITY STUDIES
FOR DECONTAMINATION AND DENSIFICATION
OF CHOP-LEACH CLADDING RESIDUES

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

JULY 1974

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by

B. Griggs

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ABSTRACT

The properties of the chop-leach cladding hull waste stream is reviewed with respect to aqueous, molten salt, and molten metal decontamination. The removal of transuranic contamination and densification of the waste stream by melting appear feasible but technically difficult to produce an acceptable product with only a small increase in the mass of the total waste stream. Research is indicated to develop a process utilizing a gaseous film alteration, aqueous film removal and etching, and flux extraction of the molten metal.

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FEASIBILITY STUDIES FOR
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B. Griggs

INTRODUCTION

The treatment of light water reactor (LWR) fuel elements to recover the residual uranium and plutonium after exposure in a power reactor requires the removal of the corrosion resistant cladding. The currently typical fuel assembly for a boiling water reactor (BWR) consists of forty-nine 0.563-inch diameter Zircaloy tubes, 160 inches long, containing 144 inches of uranium dioxide pellets and a 16-inch spring. The typical pressurized water reactor (PWR) fuel assembly consists of two hundred four 0.422-inch diameter Zircaloy tubes, 148 inches long, containing 144 inches of uranium dioxide pellets and a 4-inch spring. The assemblies are held together by stainless steel end fittings and seven Zircaloy or nine Inconel* spacers for the BWR and PWR respectively. In the chop-leach process these fuel assemblies are sheared into 1- to 2-inch lengths, and the uranium dioxide dissolved in nitric acid. The fuel hulls are then washed in dilute nitric acid and stored. Any metal fines generated by sawing or shearing are specially handled. The main advantage of this chop-leach process has been the economies resulting from the avoidance of corrosive head end solvents. The chop-leach process assumes inexpensive disposal of cladding residue (hulls).

The current fuel assemblies have evolved from an all stainless steel design. Further reductions in the current design percentage of stainless steel and Inconel may occur in the future. In order to have a base to work from, fuels designed for Browns Ferry-1 and Diablo Canyon-1 were chosen as typical of BWR and PWR fuel, respectively. Fuel hulls now in storage will have a higher percentage of stainless steel. Fuel hulls of the future may

* Inconel - Trade Mark of the International Nickel Company

have an even higher percentage of Zircaloy. Fuel hulls from one dissolver run may have a percentage of stainless steel that varies from one extreme to the other.

The LWR fuel hulls are characterized as slightly deformed 1- to 2-inch pieces of Zircaloy and stainless steel tubing together with sheet metal spacers and massive stainless steel end fittings. All metal will be contaminated with transuranic elements to some degree. The fuel hulls have been estimated^(1,2) to contain not more than 0.1% of the input fuel. The high surface area part of the hulls ($\sim 4.4 \text{ cm}^2/\text{g}$) will have been in contact with the high concentration solution of the dissolver in addition to exposure to the fuel on one side throughout its irradiation. The high level of fuel contamination on the hulls together with the reactive nature of thin Zircaloy makes storage and shipment of this material somewhat hazardous. Even though Zircaloy as massive as the fuel hulls will not spontaneously ignite at room temperature, they will readily burn if heated in air and the intense heat generated will make the burning hulls difficult to contain. The surface fuel contamination will classify the hulls as transuranic⁽³⁾ waste requiring special handling.

The objectives of this research is to (1) reduce volume and reactivity of the cladding hulls to a minimum and (2) remove the objectionable transuranic contamination in a minimum volume to a level consistent with waste disposal practice.⁽³⁾ The method of densification under study is melting and casting the zirconium-stainless steel-Inconel hulls into minimum volume, corrosion resistant ingots. The methods of decontamination under study are: (1) The removal of transuranic contamination by aqueous decontaminants, (2) the removal of transuranics by molten salt decontamination, and (3) the removal of transuranic contamination from the molten alloys before casting.

This report will review pertinent literature to establish the desired processes and techniques for laboratory testing and will point out areas where insufficient information exists to adequately plan processes. The necessary characterization of the waste stream will be treated first,

followed by the three methods of decontamination and finally densification. The treatment processes will be considered for technical feasibility and waste volume. Economics will not be treated but the fact that any process will have to be conducted remotely in a shielded facility will be considered.

WASTE CHARACTERISTICS

The waste stream hulls generated from reprocessing fuels from the rapidly expanding nuclear power industry are not yet available in quantity, but will be in the near future. Excepting laboratory, pilot plant and some reprocessing of early power reactor fuels at Nuclear Fuel Services (NFS), the characterization of the waste stream must be based on projections and assumptions about design of commercial reactor fuel and head end processing in the separation plants. The "typical" fuel design chosen was for Browns Ferry-1⁽⁴⁾ and Diablo Canyon-1,⁽⁵⁾ BWR and PWR respectively. The separations plant head end considered "typical" was a continuous or batch leach process in nitric acid as described for NFS⁽⁶⁾ and Barnwell⁽⁷⁾ or Midwest Fuel Recovery Plant⁽⁸⁾ (MFRP).

Hull Generation

The generation of the hull waste stream starts with the removal of the end hardware and fuel rod spacers during or before shearing. If the end hardware is sawed off before shearing, the metal fines generated during sawing are handled separately.⁽⁹⁾ The removed end hardware and spacers are combined with the fuel hulls after fuel leaching and stored as a solid waste. The bundle of prepared fuel rods are fed into a shear with one or two hold-down clamps and sheared into 1- or 2-inch length pieces which fall into a continuous dissolver or batch dissolver basket. The Zircaloy clad and Inconel springs or spacers, relatively unaffected by the subsequent 6 to 12 hours' treatment in intermediate strength nitric acid, emerge from the dissolver as fuel clad hulls. After cursory examination to insure that most (99.9%) of the fuel has been removed from the clad, the leached metal is combined with the previously removed hardware and stored as solid waste. The fines generated in the shearing process are handled separately. The hulls are either stored under water⁽⁹⁾ or buried in barrels.^(7,10)

Physical Characteristics

The physical characteristics of the hulls are nonhomogenous varying from massive, lightly irradiated, stainless steel end fittings to light guage, highly irradiated Zircaloy and Inconel. The surface to mass ratio for the Zircaloy hulls is $3.7 \text{ cm}^2/\text{g}$ and $4.9 \text{ cm}^2/\text{g}$ from the BWR and PWR fuel respectively. The surface-to-mass ratio for the stainless steel is about $0.8 \text{ cm}^2/\text{g}$ while the Inconel is about $5.7 \text{ cm}^2/\text{g}$. Irradiation reduces the ductility of Zircaloy⁽¹¹⁾ causing highly irradiated material to have less than 1% elongation in tensile tests. The fuel cladding will therefore not smear much in shearing but break in a nearly brittle manner.⁽¹²⁾ The bulk density of the hulls is about 1.1 kg/l .

The mass of scrap metal produced in processing a metric ton of uranium was estimated at 334 kg for a BWR, and 320 kg for a PWR, from the fuel specification in Table I. This averages to $0.3 \text{ m}^3/\text{MTU}$. Estimation of fuel reprocessing loads for the future⁽¹³⁾ show 250 metric tons uranium per year by 1974, 2400 MTU/y by 1980, and 7650 MTU/y by 1990. These estimates correspond to 75 m^3 , 720 m^3 , and 2295 m^3 or 817 MT, 785 MT, and 2500 MT of hulls for the years 1974, 1980, and 1990 respectively. If one assumes a 10 MTU/day plant for process estimation, the hull treatment facility would need a minimum capacity of 3270 kg or 3 m^3 of hulls per day.

Chemical Characteristics

The elemental composition of the hulls will vary depending on the reactor type and fuel design. Using the fuel element designs considered typical of BWRs and PWRs for this report (see Table I), the estimated masses of various metals and elements from one metric ton of uranium are as follows:

	<u>BWR</u>	<u>PWR</u>
Zircaloy	263 kg/MTU 79%	227 kg/MTU 71%
Stainless Steel	44 kg/MTU 13%	66 kg/MTU 21%
Inconel	27 kg/MTU 8%	27 kg/MTU 8%
Total Metal	334 kg/MTU	320 kg/MTU

TABLE I. Fuel Design Specifications

	PWR (Diablo-Canyon-1) ⁽⁵⁾	BWR (Browns Ferry-1) ⁽⁴⁾
Assembly Arrangement	15 x 15 Rods	7 x 7 Rods
Number of Rods per Assembly	204	49
Spacer Material	Inconel 718	Zircaloy-4
Number of Spacers per Assembly	9	7
Assembly Dimensions	8.426" x 8.426"	5.278" x 5.278"
Number of Control Rod Guides	20	--
Material of Rod Guides	304 Stainless Steel	--
End Fitting Material	304 Stainless Steel	304 Stainless Steel
Uranium Oxide per Assembly	1141 lbs	485 lbs
Rod Diameter, OD	0.422"	0.563"
Rod Diameter, ID	0.373	0.499
Rod Length	148"	160"
Fuel Column Length	144"	144"
External Spring Material	--	Inconel-X
Cladding Material	Zircaloy	Zircaloy-2
Assemblies/MTU	2.19	5.15

The Zircaloy is generally alloy 2 in BWRs and alloy 4 in PWRs. The stainless steel is usually Type 304. The Inconels are Type X, 718, and 600. Future fuel designs may replace some of the stainless steel and Inconel with Zircaloy.

The chemical characteristics of the cladding are fairly well known from the many fuel examinations that have been made and from Zircaloy technology in general. The Zircaloy clad will be coated on both inside and outside with a layer of zirconium oxide which is relatively inert to most aqueous solutions. The inside oxide layer is derived from fuel outgassing during operation and is relatively thin, about 1 to 10 microns.⁽¹⁴⁾ The oxide layer on the outside of the Zircaloy will run between 10 and 100 microns⁽¹⁴⁾ and is formed by reaction with the reactor coolant. Shearing of the Zircaloy tubes will tend to fracture the protective oxide layers near the shear cut as well as exposing a fresh surface in the cross section. Zircaloy and zirconium oxide are relatively inert to the dissolver solution and will emerge from the dissolver unchanged. Certain alloys of zirconium with beryllium,⁽¹⁵⁾ uranium,⁽¹⁶⁾ and presumably other metals will undergo a selective corrosion in the dissolver solution which dissolves one phase of the alloy and leaves a finely divided zirconium which is pyrophoric. Fires⁽¹⁵⁾ have occurred in hulls which contained a small percentage of selectively attacked alloy that initiated burning in the more massive hulls.

Zirconium alloys will burn in air if heated sufficiently. The temperature and reaction rate at ignition is almost completely dependent on the particle size.^(17,18) Particles as massive as the cladding hulls are not likely to burn unless heated above 750°C.⁽¹⁸⁾ The surface oxide on most of the surface from previous corrosion will impede the reaction rate even at 750°C. Nevertheless, if a basket of hulls was ignited by a collection of zirconium fines, there is a fair probability they would continue to burn in a smoldering fashion unless the heat was rapidly dissipated.

Radioactive Contamination

The radioactivity associated with the hulls comes from several sources and is located on the surface, in the surface oxide and uniformly through the metal. The uniformly distributed activity comes from activation of the

main alloying elements, from alloy impurities, and from diffusion of fission produced tritium out of the fuel. The calculation of the expected activity from these sources (except ^3T) by the computer code ORIGEN⁽¹⁹⁾ is shown in Tables II and III based on an input of 254 kg/MTU of Zircaloy-2 with typical impurities placed into a fluence representative of a PWR for a burnup of 33,000 megawatt days per metric ton of uranium (MWD/MTU). One of the impurities in Zircaloy is uranium which is allowed by ASTM-B-353-71⁽²⁰⁾ to the extent of 3.5 ppm. The input for this impurity at 1 ppm is shown in Table IV. Two average thermal fluence levels and two impurity levels for uranium, namely $5.76 \cdot 10^{13}$ and $2.88 \cdot 10^{13}$ n/cm²/sec and 1 and 0.1 ppm, were examined to evaluate the effects of concentration and the higher fluence levels in the cladding than in the average of the fuel. In this case the thermal fluence was increased by a factor of two while the resonance and fast fluence were kept the same in both runs in order to approximate the standard 33,000 MWD/MTU burnup in the standard exposure time of 1100 days. The effect of increased fluence with 1 ppm uranium is shown in Table V. The effect of lowering the uranium concentration is essentially linear with concentration. The fission products from the impurity uranium are shown in Table VI. The activation products for 1 ppm thorium contamination are shown in Table VII.

The uniformly distributed activity from the metal alloy and its impurities is very much less than the activities associated with the residual fuel associated with the cladding. NFS experience^(1,2) with dissolving fuel from early power reactor fuel indicates that $\leq 0.1\%$ of the input fuel is associated with the cladding after treatment in the dissolver. Table VIII shows the major transuranic activities to be expected in 0.1% of a typical metric ton of fuel as calculated by the ORIGEN code.⁽²¹⁾ These values divided by the expected mass of hulls per metric ton of uranium will give an estimate of the anticipated activities associated with the cladding which are shown in the table for the more significant transuranic elements. The location of this fuel contamination is more a matter of speculation. It has been speculated⁽²²⁾ that some of the fuel had diffused into the cladding. Considering that the maximum cladding temperature is probably less than 450°C,

TABLE II. Activation Products from Zircaloy-2 Plus Impurities Basis:
Irradiation at 2.88×10^{13} n/cm²/sec avg. for 1100 days

ELEMENT CONCENTRATIONS, GRAMS BASIS = MT OF U CHARGED TO REACTOR												
	CHARGE	DISCHARGE	1.MO	3.MO	6.MO	12.MO	36.MO	60.MO	120.MO	1200.MO	12000.MO	120000.MO
H	3.15E+01	3.15E+01	3.15E+01	3.15E+01	3.15E+01	3.15E+01	3.15E+01	3.15E+01	3.15E+01	3.15E+01	3.15E+01	3.15E+01
HE	0.	2.00E-02	2.00E-02	2.00E-02	2.00E-02	2.00E-02	2.00E-02	2.00E-02	2.00E-02	2.00E-02	2.00E-02	2.00E-02
LI	0.	9.69E-03	9.69E-03	9.69E-03	9.69E-03	9.69E-03	9.69E-03	9.69E-03	9.69E-03	9.69E-03	9.69E-03	9.69E-03
BE	0.	6.69E-05	6.69E-05	6.69E-05	6.69E-05	6.69E-05	6.69E-05	6.69E-05	6.69E-05	6.69E-05	6.69E-05	6.69E-05
B	7.56E-02	6.36E-02	6.36E-02	6.36E-02	6.36E-02	6.36E-02	6.36E-02	6.36E-02	6.36E-02	6.36E-02	6.36E-02	6.36E-02
C	3.42E+01	3.43E+01	3.43E+01	3.43E+01	3.43E+01	3.43E+01	3.43E+01	3.43E+01	3.43E+01	3.43E+01	3.43E+01	3.43E+01
N	1.02E+01	1.02E+01	1.02E+01	1.02E+01	1.02E+01	1.02E+01	1.02E+01	1.02E+01	1.02E+01	1.02E+01	1.02E+01	1.02E+01
O	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02
F	0.	2.13E-07	2.13E-07	2.13E-07	2.13E-07	2.13E-07	2.13E-07	2.13E-07	2.13E-07	2.13E-07	2.13E-07	2.13E-07
NE	0.	2.18E-09	2.18E-09	2.18E-09	2.18E-09	2.18E-09	2.18E-09	2.18E-09	2.18E-09	2.18E-09	2.18E-09	2.18E-09
NA	0.	2.71E-10	1.69E-10	1.69E-10	1.69E-10	1.69E-10	1.69E-10	1.69E-10	1.69E-10	1.69E-10	1.69E-10	1.69E-10
MG	0.	1.02E-04	1.02E-04	1.02E-04	1.02E-04	1.02E-04	1.02E-04	1.02E-04	1.02E-04	1.02E-04	1.02E-04	1.02E-04
AL	8.88E+00	8.88E+00	8.88E+00	8.88E+00	8.88E+00	8.88E+00	8.88E+00	8.88E+00	8.88E+00	8.88E+00	8.88E+00	8.88E+00
SI	1.53E+01	1.53E+01	1.53E+01	1.53E+01	1.53E+01	1.53E+01	1.53E+01	1.53E+01	1.53E+01	1.53E+01	1.53E+01	1.53E+01
P	0.	9.89E-05	9.89E-05	9.89E-05	9.89E-05	9.89E-05	9.89E-05	9.89E-05	9.89E-05	9.89E-05	9.89E-05	9.89E-05
S	0.	2.03E-08	2.12E-08	2.12E-08	2.12E-08	2.12E-08	2.12E-08	2.12E-08	2.12E-08	2.12E-08	2.12E-08	2.12E-08
CL	0.	1.17E-14	1.17E-14	1.17E-14	1.17E-14	1.17E-14	1.17E-14	1.17E-14	1.17E-14	1.17E-14	1.17E-14	1.17E-14
AR	0.	3.48E-11	3.48E-11	3.48E-11	3.48E-11	3.48E-11	3.48E-11	3.48E-11	3.48E-11	3.48E-11	3.48E-11	3.48E-11
K	0.	1.18E-12	1.14E-12	1.14E-12	1.14E-12	1.14E-12	1.14E-12	1.14E-12	1.14E-12	1.14E-12	1.14E-12	1.14E-12
CA	0.	1.60E-05	1.60E-05	1.60E-05	1.60E-05	1.60E-05	1.60E-05	1.60E-05	1.60E-05	1.60E-05	1.60E-05	1.60E-05
SC	0.	4.74E-06	3.51E-06	2.21E-06	1.17E-06	4.53E-07	2.59E-07	2.59E-07	2.59E-07	2.59E-07	2.59E-07	2.59E-07
TJ	6.33E+00	6.33E+00	6.33E+00	6.33E+00	6.33E+00	6.33E+00	6.33E+00	6.33E+00	6.33E+00	6.33E+00	6.33E+00	6.33E+00
V	0.	2.98E-01	3.05E-01	3.10E-01	3.11E-01	3.11E-01	3.11E-01	3.11E-01	3.11E-01	3.11E-01	3.11E-01	3.11E-01
CR	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02	2.54E+02
MN	6.38E+00	6.19E+00	6.19E+00	6.19E+00	6.20E+00	6.20E+00	6.22E+00	6.24E+00	6.25E+00	6.26E+00	6.26E+00	6.26E+00
FE	3.43E+02	3.44E+02	3.44E+02	3.44E+02	3.44E+02	3.44E+02	3.44E+02	3.44E+02	3.44E+02	3.43E+02	3.43E+02	3.43E+02
CO	2.54E+00	2.51E+00	2.51E+00	2.50E+00	2.49E+00	2.47E+00	2.47E+00	2.38E+00	2.31E+00	2.24E+00	2.25E+00	2.30E+00
NI	1.40E+02	1.40E+02	1.40E+02	1.40E+02	1.40E+02	1.40E+02	1.40E+02	1.40E+02	1.40E+02	1.40E+02	1.40E+02	1.40E+02
CU	6.36E+00	6.29E+00	6.29E+00	6.29E+00	6.29E+00	6.30E+00	6.30E+00	6.30E+00	6.30E+00	6.37E+00	6.43E+00	6.43E+00
ZN	0.	3.40E-02	3.40E-02	3.40E-02	3.40E-02	3.40E-02	3.40E-02	3.40E-02	3.40E-02	3.40E-02	3.40E-02	3.40E-02
SH	0.	2.22E-01	2.21E-01	2.21E-01	2.21E-01	2.20E-01	2.20E-01	2.20E-01	2.20E-01	2.20E-01	2.20E-01	2.20E-01
Y	0.	6.61E-03	2.84E-03	1.38E-03	4.71E-04	5.47E-05	1.15E-08	1.48E-09	1.31E-09	1.42E-10	3.23E-20	0.
ZR	2.49E+05	2.49E+05	2.49E+05	2.49E+05	2.49E+05	2.49E+05	2.49E+05	2.49E+05	2.49E+05	2.49E+05	2.49E+05	2.49E+05
NR	0.	7.34E-01	6.85E-01	4.66E-01	2.07E-01	3.23E-02	9.74E-05	1.22E-04	2.16E-04	1.92E-03	1.90E-02	1.89E-01
MO	0.	1.20E+01	1.24E+01	1.31E+01	1.37E+01	1.41E+01	1.41E+01	1.41E+01	1.41E+01	1.41E+01	1.41E+01	1.41E+01
TC	0.	3.24E-10	3.31E-10	3.31E-10	3.31E-10	3.31E-10	3.31E-10	3.31E-10	3.31E-10	3.31E-10	3.30E-10	3.21E-10
CD	9.39E-03	1.63E-07	1.60E-07	1.60E-07	1.60E-07	1.60E-07	1.60E-07	1.60E-07	1.60E-07	1.60E-07	1.60E-07	1.60E-07
SN	3.65E+03	3.65E+03	3.65E+03	3.65E+03	3.65E+03	3.65E+03	3.65E+03	3.65E+03	3.65E+03	3.65E+03	3.65E+03	3.65E+03
SB	0.	1.25E+00	1.25E+00	1.25E+00	1.27E+00	1.24E+00	1.23E+00	1.22E+00	1.21E+00	1.21E+00	1.22E+00	1.22E+00
TE	0.	1.84E-02	1.93E-02	2.12E-02	2.37E-02	2.82E-02	4.13E-02	4.92E-02	5.76E-02	6.08E-02	6.08E-02	6.08E-02
TA	0.	2.03E-04	2.09E-04	2.18E-04	2.27E-04	2.40E-04	2.48E-04	2.48E-04	2.48E-04	2.48E-04	2.48E-04	2.48E-04
W	1.27E+01	1.12E+01	1.12E+01	1.12E+01	1.12E+01	1.12E+01	1.12E+01	1.12E+01	1.12E+01	1.12E+01	1.12E+01	1.12E+01
TOTALS	2.54E+05	2.54E+05	2.54E+05	2.54E+05	2.54E+05	2.54E+05	2.54E+05	2.54E+05	2.54E+05	2.54E+05	2.54E+05	2.54E+05

TABLE III. Activation Products from Zircaloy-2 Plus Impurities
Basis: Irradiation at 2.88×10^{13} n/cm²/sec avg. for
1100 days

NUCLIDE RADIOACTIVITY, CURIES
BASIS = MT OF U CHARGED TO REACTOR

	CHARGE	DISCHARGE	1.MO	3.MO	6.MO	12.MO	36.MO	60.MO	120.MO	1200.MO	12000.MO	120000.MO
H 3	0.	5.37E-03	5.35E-03	5.30E-03	5.22E-03	5.08E-03	4.54E-03	4.05E-03	3.06E-03	1.92E-05	0.	0.
BE 10	0.	6.50E-08	6.50E-08	6.50E-08	6.50E-08	6.50E-08	6.50E-08	6.50E-08	6.50E-08	6.50E-08	6.50E-08	6.48E-08
C 14	0.	2.08E-01	2.08E-01	2.08E-01	2.08E-01	2.08E-01	2.08E-01	2.08E-01	2.07E-01	2.05E-01	1.84E-01	6.19E-02
NA 24	0.	2.34E-01	5.11E-16	0.	0.	0.	0.	0.	0.	0.	0.	0.
P 32	0.	2.60E-04	5.95E-05	3.11E-06	3.72E-08	5.32E-12	2.22E-27	9.28E-43	0.	0.	0.	0.
AR 39	0.	1.48E-17	1.47E-17	1.47E-17	1.47E-17	1.47E-17	1.46E-17	1.46E-17	1.44E-17	1.14E-17	1.12E-18	9.49E-29
K 43	0.	1.41E-07	2.15E-17	4.97E-37	0.	0.	0.	0.	0.	0.	0.	0.
CA 45	0.	1.09E-03	9.62E-04	7.45E-04	5.07E-04	2.36E-04	1.09E-05	5.09E-07	2.37E-10	0.	0.	0.
CA 47	0.	6.78E-06	6.44E-08	5.80E-12	4.95E-18	3.61E-30	0.	0.	0.	0.	0.	0.
SC 46	0.	1.44E-01	1.12E-01	4.76E-02	3.18E-02	7.03E-03	1.68E-05	4.02E-08	1.13E-14	0.	0.	0.
SC 47	0.	2.32E-01	4.94E-04	2.26E-09	4.20E-17	1.49E-29	0.	0.	0.	0.	0.	0.
SC 46	0.	2.22E-02	2.18E-07	2.11E-17	2.01E-32	0.	0.	0.	0.	0.	0.	0.
CR 51	0.	1.20E+03	5.53E+02	1.23E+02	1.27E+01	1.33E-01	1.64E-09	2.01E-17	3.36E-37	0.	0.	0.
MN 54	0.	2.16E+01	2.02E+01	1.75E+01	1.42E+01	9.37E+00	1.76E+00	3.31E-01	5.08E-03	0.	0.	0.
FE 55	0.	1.73E+02	1.69E+02	1.62E+02	1.51E+02	1.32E+02	7.77E+01	4.56E+01	1.20E+01	4.55E-10	0.	0.
FE 59	0.	1.35E+01	8.43E+00	3.30E+00	8.04E-01	4.85E-02	6.29E-07	8.16E-12	4.94E-24	0.	0.	0.
CO 58	0.	1.33E+02	9.86E+01	5.45E+01	2.24E+01	3.80E+00	3.13E+03	2.58E-06	5.01E-14	0.	0.	0.
CO 60	0.	3.01E+02	2.98E+02	2.91E+02	2.84E+02	2.64E+02	2.03E+02	1.56E+02	8.06E+01	5.69E-04	0.	0.
NI 59	0.	5.47E-02	5.47E-02	5.47E-02	5.47E-02	5.47E-02	5.47E-02	5.47E-02	5.47E-02	5.47E-02	5.42E-02	5.02E-02
NI 63	0.	8.60E+00	8.59E+00	8.58E+00	8.50E+00	8.53E+00	8.40E+00	8.28E+00	7.97E+00	4.05E+00	4.59E-03	0.
CU 64	0.	1.71E+02	1.13E-15	0.	0.	0.	0.	0.	0.	0.	0.	0.
ZN 65	0.	6.86E-02	6.30E-02	5.30E-02	4.09E-02	2.44E-02	3.09E-03	3.91E-04	2.23E-06	0.	0.	0.
SR 89	0.	3.77E+01	2.51E+01	1.12E+01	2.30E+00	2.90E-01	1.71E-05	1.01E-09	2.69E-20	0.	0.	0.
SR 90	0.	9.09E-04	9.07E-04	9.03E-04	8.98E-04	8.87E-04	8.44E-04	8.03E-04	7.10E-04	7.71E-05	1.76E-14	0.
Y 90	0.	1.39E+03	5.09E-01	9.04E-04	8.98E-04	8.87E-04	8.44E-04	8.04E-04	7.10E-04	7.71E-05	1.76E-14	0.
Y 91	0.	9.90E+01	6.92E+01	3.38E+01	1.15E+01	1.34E+00	2.43E-04	4.42E-08	1.97E-17	0.	0.	0.
ZR 93	0.	1.05E-01	1.05E-01	1.05E-01	1.05E-01	1.05E-01	1.05E-01	1.05E-01	1.05E-01	1.05E-01	1.05E-01	1.05E-01
ZR 95	0.	2.99E+04	2.16E+04	1.13E+04	4.26E+03	6.08E+02	2.51E-01	1.04E-04	3.62E-13	0.	0.	0.
NB 93M	0.	7.29E-03	7.70E-03	8.52E-03	9.74E-03	1.21E-02	2.11E-02	2.93E-02	4.63E-02	1.04E-01	1.05E-01	1.05E-01
NB 94	0.	1.30E-13	1.30E-13	1.30E-13	1.30E-13	1.30E-13	1.30E-13	1.30E-13	1.30E-13	1.30E-13	1.30E-13	1.30E-13
NB 95	0.	2.89E+04	2.69E+04	1.83E+04	8.12E+03	1.27E+03	5.34E-01	2.21E-04	7.68E-13	0.	0.	0.
NR 96	0.	1.34E+01	3.69E-09	2.78E-28	0.	0.	0.	0.	0.	0.	0.	0.
MO 99	0.	3.67E-06	1.92E-09	5.22E-16	7.43E-26	1.50E-45	0.	0.	0.	0.	0.	0.
TC 99M	0.	3.20E-06	1.83E-09	4.99E-16	7.10E-26	1.44E-45	0.	0.	0.	0.	0.	0.
TC 99	0.	5.56E-12	5.70E-12	5.70E-12	5.70E-12	5.70E-12	5.70E-12	5.70E-12	5.70E-12	5.70E-12	5.68E-12	5.52E-12
CD115	0.	1.68E-03	1.30E-07	7.81E-16	3.64E-28	0.	0.	0.	0.	0.	0.	0.
SN117M	0.	1.16E+04	2.57E+03	1.26E+02	1.37E+00	1.62E-04	3.17E-20	6.20E-36	0.	0.	0.	0.
SN119M	0.	2.27E+01	2.09E+01	1.76E+01	1.37E+01	8.25E+00	1.09E+00	1.44E-01	9.08E-04	0.	0.	0.
SN121M	0.	3.36E-01	3.36E-01	3.35E-01	3.35E-01	3.33E-01	3.27E-01	3.21E-01	3.07E-01	1.35E-01	3.68E-05	0.
SN121	0.	2.06E+03	1.48E-05	7.56E-22	0.	0.	0.	0.	0.	0.	0.	0.
SN123	0.	4.35E-01	3.68E-01	2.62E-01	1.58E-01	5.74E-02	9.99E-04	1.74E-05	6.95E-10	0.	0.	0.
SN125	0.	3.60E+00	3.82E-01	4.28E-03	5.10E-06	7.22E-12	0.	0.	0.	0.	0.	0.
SB124	0.	4.65E+00	3.27E+00	1.62E+00	5.64E-01	6.84E-02	1.48E-05	3.19E-09	2.19E-18	0.	0.	0.
SB125	0.	4.47E+01	4.38E+01	4.19E+01	3.93E+01	3.46E+01	2.07E+01	1.24E+01	3.43E+00	3.16E-10	0.	0.
SB126	0.	2.18E-01	4.03E-02	1.38E-03	8.70E-06	3.48E-10	8.86E-28	2.26E-45	0.	0.	0.	0.
TE125M	0.	1.62E+01	1.65E+01	1.66E+01	1.60E+01	1.43E+01	8.58E+00	5.13E+00	1.42E+00	1.31E-10	0.	0.
TA182	0.	9.80E-02	8.15E-02	5.65E-02	3.26E-02	1.08E-02	1.33E-04	1.62E-06	2.69E-11	0.	0.	0.
W181	0.	3.13E-01	2.70E-01	1.99E-01	1.27E-01	5.14E-02	1.38E-03	3.71E-05	4.38E-09	0.	0.	0.
W185	0.	4.59E+01	3.46E+01	1.97E+01	8.48E+00	1.57E+00	1.83E-03	2.14E-06	1.00E-13	0.	0.	0.
W187	0.	1.22E+03	7.66E-07	3.03E-25	0.	0.	0.	0.	0.	0.	0.	0.
SUBTOT	0.	7.73E+04	5.25E+04	3.05E+04	1.30E+04	2.35E+03	3.23E+02	2.28E+02	1.06E+02	4.65E+00	4.53E-01	3.21E-01
TOTAL	0.	7.90E+04	5.25E+04	3.05E+04	1.30E+04	2.35E+03	3.23E+02	2.28E+02	1.06E+02	4.65E+00	4.53E-01	3.21E-01

TABLE IV. Activation Products from 1 ppm Uranium Impurity in Zircaloy-2
Basis: Irradiation at $2.88 \cdot 10^{13}$ n/cm²/sec avg. for 1100 days

NUCLIDE RADIOACTIVITY, CURIES BASIS = MT OF U CHARGED TO REACTOR												
	CHARGE	DISCHARGE	1.MO	3.MO	6.MO	12.MO	36.MO	60.MO	120.MO	1200.MO	12000.MO	120000.MO
TL207	0.	4.76E-14	5.08E-14	5.75E-14	6.75E-14	9.24E-14	1.70E-13	2.46E-13	4.26E-13	2.67E-12	2.21E-11	2.58E-10
TL208	0.	3.56E-11	3.83E-11	4.63E-11	5.40E-11	8.37E-11	1.97E-10	2.97E-10	4.38E-10	2.23E-10	3.88E-14	5.17E-15
TL209	0.	2.70E-16	1.53E-16	5.34E-17	4.60E-17	4.91E-17	6.02E-17	7.23E-17	1.07E-16	1.98E-15	4.19E-13	6.72E-11
PR209	0.	1.23E-14	6.98E-15	2.43E-15	2.09E-15	2.23E-15	2.74E-15	3.29E-15	4.85E-15	8.99E-14	1.90E-11	3.06E-09
PR210	0.	3.19E-17	3.42E-17	3.90E-17	4.71E-17	6.59E-17	1.89E-16	4.14E-16	1.65E-15	6.64E-13	2.21E-10	9.56E-09
PR211	0.	4.77E-14	5.10E-14	5.77E-14	6.77E-14	9.27E-14	1.71E-13	2.47E-13	4.28E-13	2.68E-12	2.21E-11	2.59E-10
PR212	0.	9.89E-11	1.06E-10	1.29E-10	1.61E-10	2.33E-10	5.46E-10	8.25E-10	1.22E-09	6.20E-10	1.08E-13	1.44E-14
PR214	0.	8.57E-16	8.84E-16	9.62E-16	1.08E-15	1.33E-15	2.75E-15	4.79E-15	1.28E-14	1.21E-12	2.21E-10	9.56E-09
BI210	0.	3.19E-17	3.26E-17	3.91E-17	4.71E-17	6.59E-17	1.89E-16	4.14E-16	1.65E-15	6.64E-13	2.21E-10	9.56E-09
BI211	0.	4.77E-14	5.10E-14	5.77E-14	6.77E-14	9.27E-14	1.71E-13	2.47E-13	4.28E-13	2.68E-12	2.21E-11	2.59E-10
BI212	0.	9.89E-11	1.06E-10	1.29E-10	1.61E-10	2.33E-10	5.46E-10	8.25E-10	1.22E-09	6.20E-10	1.08E-13	1.44E-14
BI213	0.	1.23E-14	6.98E-15	2.43E-15	2.09E-15	2.23E-15	2.74E-15	3.29E-15	4.85E-15	8.99E-14	1.90E-11	3.06E-09
BI214	0.	8.57E-16	8.84E-16	9.62E-16	1.08E-15	1.33E-15	2.75E-15	4.79E-15	1.28E-14	1.21E-12	2.21E-10	9.56E-09
PO210	0.	2.03E-17	2.21E-17	2.57E-17	3.20E-17	4.68E-17	1.49E-16	3.42E-16	1.65E-15	6.64E-13	2.21E-10	9.56E-09
PO211	0.	1.43E-16	1.53E-16	1.73E-16	2.03E-16	2.78E-16	5.13E-16	7.41E-16	1.28E-15	8.04E-15	6.64E-14	7.78E-13
PO212	0.	6.33E-11	6.80E-11	8.23E-11	1.03E-10	1.49E-10	3.50E-10	5.28E-10	7.79E-10	3.97E-10	6.90E-14	9.19E-15
PO213	0.	1.20E-14	6.82E-15	2.37E-15	2.05E-15	2.18E-15	2.68E-15	3.21E-15	4.75E-15	8.80E-14	1.86E-11	2.99E-09
PO214	0.	8.57E-16	8.84E-16	9.62E-16	1.08E-15	1.33E-15	2.75E-15	4.79E-15	1.28E-14	1.21E-12	2.21E-10	9.56E-09
PO215	0.	4.77E-14	5.10E-14	5.77E-14	6.77E-14	9.27E-14	1.71E-13	2.47E-13	4.28E-13	2.68E-12	2.21E-11	2.59E-10
PO216	0.	9.89E-11	1.06E-10	1.29E-10	1.61E-10	2.33E-10	5.46E-10	8.25E-10	1.22E-09	6.20E-10	1.08E-13	1.44E-14
PO218	0.	8.57E-16	8.84E-16	9.62E-16	1.08E-15	1.33E-15	2.75E-15	4.79E-15	1.28E-14	1.21E-12	2.21E-10	9.56E-09
AT217	0.	1.23E-14	6.98E-15	2.43E-15	2.09E-15	2.23E-15	2.74E-15	3.29E-15	4.85E-15	8.99E-14	1.90E-11	3.06E-09
RN219	0.	4.77E-14	5.10E-14	5.77E-14	6.77E-14	9.27E-14	1.71E-13	2.47E-13	4.28E-13	2.68E-12	2.21E-11	2.59E-10
RN220	0.	9.89E-11	1.06E-10	1.29E-10	1.61E-10	2.33E-10	5.46E-10	8.25E-10	1.22E-09	6.20E-10	1.08E-13	1.44E-14
RN222	0.	8.57E-16	8.84E-16	9.62E-16	1.08E-15	1.33E-15	2.75E-15	4.79E-15	1.28E-14	1.21E-12	2.21E-10	9.56E-09
FR221	0.	1.23E-14	6.98E-15	2.43E-15	2.09E-15	2.23E-15	2.74E-15	3.29E-15	4.85E-15	8.99E-14	1.90E-11	3.06E-09
FR223	0.	7.31E-16	7.78E-16	8.72E-16	1.01E-15	1.29E-15	2.39E-15	3.45E-15	5.98E-15	3.75E-14	3.10E-13	3.63E-12
RA223	0.	4.77E-14	5.10E-14	5.77E-14	6.77E-14	9.27E-14	1.71E-13	2.47E-13	4.28E-13	2.68E-12	2.21E-11	2.59E-10
RA224	0.	9.89E-11	1.06E-10	1.29E-10	1.61E-10	2.33E-10	5.46E-10	8.25E-10	1.22E-09	6.20E-10	1.08E-13	1.44E-14
RA225	0.	1.21E-14	4.44E-15	2.18E-15	2.10E-15	2.23E-15	2.74E-15	3.29E-15	4.85E-15	8.99E-14	1.90E-11	3.06E-09
RA226	0.	8.57E-16	8.91E-16	9.62E-16	1.08E-15	1.33E-15	2.75E-15	4.79E-15	1.28E-14	1.21E-12	2.21E-10	9.56E-09
RA228	0.	1.31E-19	1.42E-19	1.65E-19	2.02E-19	2.93E-19	7.97E-19	1.50E-18	3.89E-18	8.02E-17	8.67E-16	1.44E-14
AC225	0.	1.23E-14	6.98E-15	2.43E-15	2.09E-15	2.23E-15	2.74E-15	3.29E-15	4.85E-15	8.99E-14	1.90E-11	3.06E-09
AC227	0.	5.22E-14	5.56E-14	6.23E-14	7.24E-14	9.23E-14	1.71E-13	2.47E-13	4.27E-13	2.68E-12	2.21E-11	2.59E-10
AC228	0.	1.31E-19	1.42E-19	1.65E-19	2.02E-19	2.93E-19	7.97E-19	1.50E-18	3.89E-18	8.02E-17	8.67E-16	1.44E-14
TH227	0.	4.87E-14	5.20E-14	5.86E-14	6.85E-14	9.13E-14	1.68E-13	2.44E-13	4.22E-13	2.64E-12	2.18E-11	2.56E-10
TH228	0.	9.85E-11	1.08E-10	1.28E-10	1.60E-10	2.32E-10	5.46E-10	8.23E-10	1.22E-09	6.20E-10	1.08E-13	1.44E-14
TH229	0.	1.99E-15	2.01E-15	2.05E-15	2.11E-15	2.23E-15	2.74E-15	3.29E-15	4.85E-15	8.99E-14	1.90E-11	3.06E-09
TH230	0.	9.29E-13	9.58E-13	1.02E-12	1.10E-12	1.28E-12	1.99E-12	2.74E-12	4.72E-12	6.41E-11	1.23E-09	1.23E-08
TH231	0.	4.21E-08	9.47E-10	9.47E-10	9.47E-10	9.47E-10	9.47E-10	9.47E-10	9.47E-10	9.55E-10	1.03E-09	1.68E-09
TH232	0.	1.41E-18	1.47E-18	1.60E-18	1.80E-18	2.19E-18	3.74E-18	5.30E-18	9.20E-18	8.02E-17	8.67E-16	1.44E-14
TH234	0.	8.18E-08	8.18E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08
PA231	0.	1.31E-12	1.31E-12	1.32E-12	1.32E-12	1.33E-12	1.37E-12	1.41E-12	1.51E-12	3.34E-12	2.21E-11	2.59E-10
PA232	0.	1.91E-08	2.18E-15	2.85E-29	4.26E-50	0.	0.	0.	0.	0.	0.	0.
PA233	0.	2.46E-08	2.53E-08	2.59E-08	2.60E-08	2.61E-08	2.61E-08	2.63E-08	2.67E-08	4.93E-08	1.81E-07	2.21E-07
PA234M	0.	8.23E-08	8.18E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08
PA234	0.	5.60E-10	8.18E-11	8.17E-11	8.17E-11	8.17E-11	8.17E-11	8.17E-11	8.17E-11	8.17E-11	8.17E-11	8.17E-11
U232	0.	4.04E-10	4.27E-10	4.71E-10	5.32E-10	6.44E-10	9.69E-10	1.16E-09	1.33E-09	6.04E-10	1.04E-13	0.
U233	0.	2.43E-12	2.44E-12	2.46E-12	2.49E-12	2.54E-12	2.78E-12	3.02E-12	3.58E-12	1.79E-11	5.19E-10	8.86E-09
U234	8.69E-08	3.97E-08	3.98E-08	3.99E-08	4.01E-08	4.05E-08	4.21E-08	4.38E-08	4.78E-08	9.87E-08	1.50E-07	1.48E-07
U235	3.88E-09	9.47E-10	9.47E-10	9.47E-10	9.47E-10	9.47E-10	9.47E-10	9.47E-10	9.47E-10	9.55E-10	1.03E-09	1.68E-09
U236	0.	1.58E-08	1.58E-08	1.58E-08	1.58E-08	1.58E-08	1.59E-08	1.59E-08	1.59E-08	1.62E-08	1.93E-08	3.87E-08

TABLE IV. (contd.)

		NUCLIDE RADIOACTIVITY, CURIES											
		BASIS = MT OF U CHARGED TO REACTOR											
	CHARGE	DISCHARGE	1. MO	3. MO	6. MO	12. MO	36. MO	60. MO	120. MO	1200. MO	12000. MO	120000. MO	
U237	0.	6.15E-02	2.70E-03	5.88E-06	6.75E-07	6.59E-07	6.00E-07	5.47E-07	4.33E-07	6.39E-09	2.18E-12	1.03E-12	
U238	8.38E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	8.17E-08	
NP236	0.	2.14E-07	2.20E-17	2.24E-37	0.	0.	0.	0.	0.	0.	0.	0.	
NP237	0.	2.55E-08	2.60F-08	2.61E-08	2.61E-08	2.61E-08	2.61E-08	2.63E-08	2.67E-08	4.93E-08	1.81E-07	2.21E-07	
NP238	0.	3.07E-02	1.33E-06	2.49E-15	2.03E-28	0.	0.	0.	0.	0.	0.	0.	
NP239	0.	4.86E+00	6.22E-04	4.84E-06	4.84E-06	4.84E-06	4.84E-06	4.83E-06	4.83E-06	4.79E-06	4.42E-06	1.95E-06	
PU236	0.	2.84E-08	2.80E-08	2.84E-08	2.84E-08	2.84E-08	2.84E-08	2.84E-08	2.84E-08	2.84E-08	2.84E-08	2.84E-08	
PU238	0.	2.56E-04	2.63E-04	2.72E-04	2.81E-04	2.91E-04	2.95E-04	2.91E-04	2.80E-04	1.40E-04	1.47E-07	7.62E-26	
PU239	0.	8.47E-05	8.60E-05	8.60E-05	8.60E-05	8.60E-05	8.60E-05	8.60E-05	8.60E-05	8.58E-05	8.37E-05	6.55E-05	
PU240	0.	1.24F-04	1.25E-04	1.25E-04	1.25E-04	1.25E-04	1.25E-04	1.25E-04	1.25E-04	1.25E-04	1.14E-04	4.53E-05	
PU241	0.	2.76E-02	2.75F-02	2.73E-02	2.70E-02	2.63E-02	2.45E-02	2.10E-02	1.73E-02	2.55E-04	8.73E-04	4.11E-08	
PU242	0.	3.61E-07	3.61E-07	3.61E-07	3.61E-07	3.61E-07	3.61E-07	3.61E-07	3.61E-07	3.61E-07	3.61E-07	3.55E-07	
PU243	0.	9.54E-02	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	
AM241	0.	2.26E-05	2.63E-05	3.36E-05	4.44E-05	6.57E-05	1.46E-04	2.19E-04	3.72E-04	8.43E-04	2.02E-04	4.12E-08	
AM242	0.	2.46E-06	2.46E-06	2.46E-06	2.46E-06	2.46E-06	2.46E-06	2.46E-06	2.46E-06	1.56E-06	2.57E-08	3.84E-26	
AM243	0.	4.83E-06	4.84E-06	4.84E-06	4.84E-06	4.84E-06	4.84E-06	4.83E-06	4.83E-06	4.79E-06	4.42E-06	1.95E-06	
AM245	0.	1.57E-14	1.47E-14	1.28E-14	1.05E-14	7.00E-15	1.39E-15	2.78E-16	4.93E-18	0.	0.	0.	
CM242	0.	8.80E-03	7.78E-03	6.01E-03	4.07E-03	1.88E-03	8.58E-05	5.73E-06	1.93E-06	1.28E-06	2.11E-08	3.16E-26	
CM243	0.	9.59E-07	9.57E-07	9.54E-07	9.48E-07	9.38E-07	8.98E-07	8.60E-07	7.72E-07	1.10E-07	3.75E-16	0.	
CM244	0.	6.62E-04	6.60E-04	6.55E-04	6.49E-04	6.37E-04	5.95E-04	5.46E-04	4.51E-04	1.44E-05	1.76E-20	2.23E-20	
CM245	0.	9.48E-08	9.48E-08	9.48E-08	9.48E-08	9.48E-08	9.48E-08	9.48E-08	9.48E-08	9.48E-08	9.48E-08	9.48E-08	
CM246	0.	1.88E-08	1.88E-08	1.88E-08	1.88E-08	1.88E-08	1.88E-08	1.88E-08	1.88E-08	1.88E-08	1.88E-08	1.88E-08	
CM247	0.	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	7.08E-14	
CM248	0.	2.25E-13	2.25E-13	2.25E-13	2.25E-13	2.25E-13	2.25E-13	2.25E-13	2.25E-13	2.25E-13	2.25E-13	2.21E-13	
CM249	0.	7.43F-09	1.14E-15	1.07E-16	3.06E-18	2.50E-21	1.12E-33	5.03E-46	0.	0.	0.	0.	
CM250	0.	2.63E-20	2.63E-20	2.63E-20	2.63E-20	2.63E-20	2.63E-20	2.63E-20	2.63E-20	2.62E-20	2.53E-20	1.77E-20	
BM249	0.	1.04E-09	9.77E-10	8.54E-10	6.98E-10	4.66E-10	9.30E-11	1.85E-11	3.29E-13	0.	0.	0.	
BM250	0.	1.85E-09	2.63E-20	2.63E-20	2.63E-20	2.63E-20	2.63E-20	2.63E-20	2.63E-20	2.62E-20	2.53E-20	1.77E-20	
CF249	0.	3.24E-13	4.94E-13	7.93E-13	1.17E-12	1.74E-12	2.64E-12	2.81E-12	2.83E-12	2.37E-12	4.03E-13	8.08E-21	
CF250	0.	1.08E-11	1.08E-11	1.07E-11	1.06E-11	1.03E-11	9.27E-12	8.34E-12	6.40E-12	5.43E-14	2.53E-20	1.77E-20	
CF251	0.	8.01E-14	8.01E-14	8.01E-14	8.01E-14	8.01E-14	7.98E-14	7.98E-14	7.98E-14	7.42E-14	3.71E-14	3.62E-17	
CF252	0.	1.46E-11	1.43E-11	1.37E-11	1.28E-11	1.13E-11	6.67E-12	3.95E-12	1.07E-12	0.	0.	0.	
CF253	0.	1.20E-12	3.67E-13	3.44E-14	9.43E-16	8.05E-19	3.61E-31	1.62E-43	0.	0.	0.	0.	
CF254	0.	3.15E-16	2.22E-16	1.10E-16	3.86E-17	4.83E-18	1.12E-21	2.60E-25	2.12E-34	0.	0.	0.	
ES253	0.	8.60E-13	7.17E-13	1.67E-13	1.22E-14	3.56E-17	1.01E-27	2.51E-38	0.	0.	0.	0.	
SUBTOT	1.75E-07	5.10E+00	3.98E-02	3.45E-02	3.22E-02	2.94E-02	2.53E-02	2.31E-02	1.86E-02	1.48E-03	4.10E-04	1.16E-04	
TOTAL	1.75E-07	1.00E+01	3.98E-02	3.45E-02	3.22E-02	2.94E-02	2.53E-02	2.31E-02	1.86E-02	1.48E-03	4.10E-04	1.16E-04	

TABLE V. Activation Products from 1 ppm Uranium Impurity in Zircaloy-2
Basis: Irradiation at 5.76×10^{13} n/cm²/sec avg. for 1100 days

REFERENCE PKW	3.3 PERCENT UNENRICHED U	PROPERTIES AFTER	IRRADIATION	1. MO	3. MO	5. MC	12. MO	36. MO	60. MO	12. MC	1200. MO	12000. MO	120000. MO
POWER=	0.00 MW	BURNUP=	0.00 MW, FLUX= 5.76×10^{13} n/cm ² -SEC	NUCLIDE RADIOACTIVITY, CURIES									
				BASIS = MT OF U CHARGED TO REACTOR									
CHARGE	DISCHARGE												
TL217	4.25E-14	4.53E-14	5.19E-14	5.93E-14	8.41E-14	1.45E-13	2.06E-13	3.47E-13	3.47E-13	3.47E-13	1.59E-12	8.59E-12	1.13E-10
TL208	3.71E-11	4.03E-11	4.84E-11	6.08E-11	8.77E-11	8.77E-11	3.27E-10	4.15E-10	4.15E-10	4.15E-10	2.30E-09	4.00E-09	5.61E-08
TL209	3.05E-16	3.05E-16	3.05E-16	3.05E-16	3.05E-16	3.05E-16	3.05E-16	3.05E-16	3.05E-16	3.05E-16	1.79E-15	3.80E-13	5.35E-11
PB209	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	3.14E-14	1.73E-11	2.78E-09
PB210	3.27E-17	3.27E-17	3.27E-17	3.27E-17	3.27E-17	3.27E-17	3.27E-17	3.27E-17	3.27E-17	3.27E-17	6.31E-15	2.35E-10	1.02E-08
PB211	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	1.56E-12	8.59E-12	1.14E-10
PB212	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	6.38E-10	1.11E-13	1.56E-14
PB213	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	1.16E-12	2.35E-10	1.02E-08
PB214	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	6.31E-13	2.35E-10	1.02E-08
BI211	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	1.56E-12	8.59E-12	1.14E-10
BI212	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	6.38E-10	1.11E-13	1.56E-14
BI213	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	1.16E-12	2.35E-10	1.02E-08
BI214	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	3.38E-17	6.31E-13	2.35E-10	1.02E-08
PO210	1.93E-17	2.11E-17	2.47E-17	3.06E-17	4.44E-17	1.38E-16	3.12E-16	1.04E-15	1.04E-15	1.04E-15	6.31E-13	2.35E-10	1.02E-08
PO211	1.29E-15	1.36E-16	1.53E-16	1.78E-16	2.41E-16	4.35E-16	6.20E-16	1.04E-15	1.04E-15	1.04E-15	6.31E-13	2.35E-10	1.02E-08
PO212	2.44E-14	1.35E-14	4.17E-15	3.40E-15	1.24E-15	2.49E-15	4.28E-15	1.14E-14	1.14E-14	1.14E-14	1.16E-12	2.35E-10	1.02E-08
PO213	8.32E-16	1.35E-16	9.33E-16	3.48E-16	1.24E-15	2.49E-15	4.28E-15	1.14E-14	1.14E-14	1.14E-14	1.16E-12	2.35E-10	1.02E-08
PO214	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	1.56E-12	8.59E-12	1.14E-10
PO215	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	6.38E-10	1.11E-13	1.56E-14
PO216	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	1.16E-12	2.35E-10	1.02E-08
AT217	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
RA219	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	1.56E-12	8.59E-12	1.14E-10
RA220	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	6.38E-10	1.11E-13	1.56E-14
RA221	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	1.16E-12	2.35E-10	1.02E-08
FR221	2.53E-14	1.38E-14	4.26E-15	3.48E-15	1.24E-15	2.49E-15	4.28E-15	1.14E-14	1.14E-14	1.14E-14	1.16E-12	2.35E-10	1.02E-08
FR222	6.93E-16	7.03E-16	1.12E-15	2.03E-15	2.03E-15	2.03E-15	2.03E-15	2.03E-15	2.03E-15	2.03E-15	2.14E-14	1.73E-11	2.70E-09
RA223	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	4.54E-14	1.56E-12	8.59E-12	1.14E-10
RA224	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	1.11E-10	6.38E-10	1.11E-13	1.56E-14
RA225	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	8.08E-15	1.16E-12	2.35E-10	1.02E-08
RA226	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
AC225	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
AC227	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
AC228	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
TH227	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
TH228	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
TH229	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
TH230	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
TH231	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
TH232	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
TH233	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
TH234	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
PA231	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
PA232	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
PA233	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
PA234	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	1.38E-14	4.08E-10	7.10E-14	9.97E-15
U232	4.28E-10	4.32E-10	4.96E-10	5.59E-10	6.72E-10	1.34E-09	1.92E-09	2.51E-09	2.51E-09	2.51E-09	6.21E-10	1.07E-13	7.94E-09
U233	1.31E-12	1.32E-12	1.37E-12	1.37E-12	1.43E-12	1.43E-12	1.92E-12	2.51E-12	2.51E-12	2.51E-12	4.66E-10	1.60E-07	1.58E-07
U234	3.39E-08	3.39E-08	3.42E-08	3.42E-08	3.47E-08	3.66E-08	3.85E-08	3.85E-08	3.85E-08	3.85E-08	1.02E-07	1.60E-07	1.58E-07
U235	3.88E-09	3.88E-09	3.88E-09	3.88E-09	3.88E-09	3.88E-09	3.88E-09	3.88E-09	3.88E-09	3.88E-09	3.35E-10	3.81E-10	8.08E-10
U236	1.69E-08	1.69E-08	1.69E-08	1.69E-08	1.69E-08	1.69E-08	1.69E-08	1.69E-08	1.69E-08	1.69E-08	1.73E-08	2.07E-08	4.28E-08

TABLE V. (contd.)

[illegible]

TABLE VI. Fission Products to be Expected from 1 ppm of Uranium Impurity in Zircaloy Fuel Cladding

Basis: Exposure at 2.88×10^{13} n/cm² avg. for 1100 days

ELEMENTAL ACTIVITY, CURIES												
BASIS = MT OF U CHARGED TO REACTOR												
	CHARGE	DISCHARGE	1.MO	3.MO	6.MO	12.MO	36.MO	60.MO	120.MO	1200.MO	12000.MO	120000.MO
H	J.	1.25E-04	1.25E-04	1.24E-04	1.22E-04	1.18E-04	1.06E-04	9.45E-05	7.13E-05	4.47E-07	0.	0.
ZN	J.	5.33E-06	3.38E-11	3.26E-20	2.11E-34	0.	0.	0.	0.	0.	0.	0.
GA	J.	2.22E-04	1.35E-10	4.68E-20	3.04E-34	0.	0.	0.	0.	0.	0.	0.
GE	J.	1.77E-03	6.02E-24	0.	0.	0.	0.	0.	0.	0.	0.	0.
AS	J.	7.77E-03	1.13E-09	4.74E-21	4.24E-39	0.	0.	0.	0.	0.	0.	0.
SE	J.	6.57E-02	5.03E-08	5.03E-08	5.03E-08	5.03E-08	5.03E-08	5.03E-08	5.03E-08	5.03E-08	4.98E-08	4.52E-08
BR	J.	3.06E-01	1.93E-10	6.69E-23	1.37E-41	0.	0.	0.	0.	0.	0.	0.
KR	J.	5.59E-01	1.19E-03	1.18E-03	1.16E-03	1.12E-03	9.85E-04	8.66E-04	6.29E-04	1.95E-06	0.	0.
RB	J.	9.34E-01	1.64E-05	1.72E-06	5.83E-08	6.89E-11	2.01E-12	2.00E-12	2.00E-12	2.00E-12	2.00E-12	2.00E-12
SR	J.	9.78E-01	7.57E-02	3.80E-02	1.68E-02	8.54E-03	7.38E-03	7.03E-03	6.21E-03	6.74E-04	1.54E-13	0.
Y	J.	1.56E+00	1.36E-01	5.57E-02	2.41E-02	9.65E-03	7.38E-03	7.03E-03	6.21E-03	6.74E-04	1.54E-13	0.
ZR	J.	7.88E-01	1.77E-01	9.24E-02	3.49E-02	4.98E-03	2.29E-06	2.28E-07	2.27E-07	2.27E-07	2.27E-07	2.26E-07
NB	J.	1.97E+00	2.29E-01	1.55E-01	6.86E-02	1.67E-02	4.55E-06	6.46E-08	9.95E-08	2.26E-07	2.27E-07	2.26E-07
MO	J.	1.63E+00	1.56E-04	4.25E-11	6.04E-21	1.22E-40	0.	0.	0.	0.	0.	0.
TC	J.	1.82E+00	1.51E-04	2.08E-06	2.08E-06	2.08E-06	2.08E-06	2.08E-06	2.08E-06	2.08E-06	2.07E-06	2.01E-06
RU	J.	8.83E-01	2.82E-01	1.66E-01	1.06E-01	6.78E-02	1.69E-02	4.26E-03	1.35E-04	0.	0.	0.
RH	J.	1.36E+00	2.82E-01	1.66E-01	1.06E-01	6.78E-02	1.69E-02	4.26E-03	1.35E-04	0.	0.	0.
PD	J.	1.24E-01	2.73E-08	2.73E-08	2.73E-08	2.73E-08	2.73E-08	2.73E-08	2.73E-08	2.73E-08	2.73E-08	2.73E-08
AG	J.	1.49E-01	1.59E-03	8.38E-04	5.51E-04	3.95E-04	5.33E-05	7.21E-06	4.84E-08	0.	0.	0.
CD	J.	1.10E-02	8.14E-05	3.26E-05	9.19E-06	2.72E-06	2.14E-06	1.34E-06	1.51E-06	1.75E-08	7.77E-28	0.
IN	J.	2.08E-02	6.18E-07	1.99E-07	5.60E-04	4.46E-09	1.78E-13	7.11E-18	7.19E-29	0.	0.	0.
SN	J.	6.76E-01	2.35E-03	1.21E-03	7.28E-04	2.65E-04	4.88E-06	2.18E-07	1.16E-07	1.16E-07	1.15E-07	1.08E-07
SB	J.	1.18E+00	2.07E-03	1.89E-03	1.75E-03	1.53E-03	9.17E-04	5.49E-04	1.52E-04	2.30E-07	2.29E-07	2.15E-07
TE	J.	1.45E+00	1.78E-02	7.85E-03	3.43E-03	1.35E-03	3.87E-04	2.28E-04	6.30E-05	5.81E-15	0.	0.
I	J.	2.27E+00	1.37E-02	7.35E-05	3.34E-08	6.30E-09	6.30E-09	6.30E-09	6.30E-09	6.30E-09	6.30E-09	6.30E-09
XE	J.	1.57E+00	7.53E-03	2.20E-05	9.71E-09	2.14E-12	4.93E-31	1.14E-49	0.	0.	0.	0.
CS	J.	1.31E+00	4.92E-02	4.54E-02	4.28E-02	3.84E-02	2.64E-02	2.00E-02	1.37E-02	1.57E-03	4.13E-08	4.12E-08
BA	J.	1.24E+00	6.75E-02	1.67E-02	1.46E-02	1.45E-02	1.38E-02	1.32E-02	1.17E-02	1.47E-03	1.36E-12	0.
LA	J.	1.21E+00	6.37E-02	2.25E-03	1.60E-05	8.10E-10	5.33E-27	3.51E-44	0.	0.	0.	0.
CE	J.	1.12E+00	2.85E-01	1.68E-01	1.11E-01	6.76E-02	1.14E-02	1.91E-03	2.21E-05	0.	0.	0.
PR	J.	9.29E-01	2.06E-01	1.34E-01	1.05E-01	6.75E-02	1.14E-02	1.91E-03	2.21E-05	0.	0.	0.
NC	J.	2.16E-01	1.64E-02	3.66E-04	1.22E-06	1.36E-11	2.09E-31	3.22E-51	0.	0.	0.	0.
PM	J.	2.63E-01	2.25E-02	1.86E-02	1.63E-02	1.40E-02	8.22E-03	4.84E-03	1.29E-03	5.87E-14	0.	0.
SM	J.	1.09E-01	2.21E-04	2.19E-04	2.19E-04	2.18E-04	2.14E-04	2.11E-04	2.03E-04	9.89E-05	7.60E-08	0.
EU	J.	5.04E-02	1.26E-02	2.98E-03	2.26E-03	2.03E-03	1.45E-03	1.13E-03	7.81E-04	1.52E-05	1.77E-22	0.
GD	J.	1.23E-03	4.55E-06	3.82E-06	2.94E-06	1.74E-06	2.15E-07	2.65E-08	1.42E-10	0.	0.	0.
TB	J.	6.17E-04	2.47E-04	1.33E-04	5.53E-05	9.55E-06	8.51E-09	7.58E-12	1.79E-19	0.	0.	0.
DY	J.	1.13E-04	5.69E-09	2.28E-14	1.83E-22	1.18E-38	0.	0.	0.	0.	0.	0.
HQ	J.	3.15E-05	8.68E-09	1.78E-10	1.78E-10	1.78E-10	1.78E-10	1.78E-10	1.77E-10	1.68E-10	1.00E-10	5.53E-13
TOTALS	J.	2.68E+01	1.92E+00	1.38E+00	6.56E-01	3.78E-01	1.24E-01	6.76E-02	4.13E-02	4.51E-03	3.07E-06	2.90E-06

TABLE VII. Activation Products to be Expected from 1 ppm of Thorium Impurity in Zircaloy Fuel Cladding
Basis: Exposure at $2.88 \cdot 10^{13}$ n/cm²/sec avg. for 1100 days

		NUCLIDE RADIOACTIVITY, CURIES											
		BASIS = MT OF U CHARGED TO REACTOR											
	CHARGE	DISCHARGE	1.MO	3.MO	6.MO	12.MO	36.MO	60.MO	120.MO	1200.MO	12000.MO	120000.MO	
TL207	0.	3.29E-08	3.46E-08	3.81E-08	4.33E-08	5.61E-08	9.53E-08	1.32E-07	2.15E-07	6.61E-07	6.74E-07	5.56E-07	
TL208	0.	1.15E-05	1.21E-05	1.39E-05	1.62E-05	2.01E-05	3.00E-05	3.44E-05	3.65E-05	1.57E-05	1.18E-08	9.10E-09	
TL209	0.	4.18E-10	3.85E-10	3.84E-10	4.32E-10	5.54E-10	4.64E-10	1.37E-09	2.40E-09	2.07E-08	1.95E-07	1.29E-06	
PR209	0.	1.90E-08	1.75E-08	1.74E-08	1.96E-08	2.52E-08	4.38E-08	6.24E-08	1.09E-07	9.42E-07	8.88E-06	5.87E-05	
PR210	0.	8.99E-14	9.71E-14	1.12E-13	1.34E-13	1.98E-13	5.60E-13	1.11E-12	3.31E-12	2.12E-10	1.67E-08	6.09E-07	
PR211	0.	3.30E-08	3.47E-08	3.82E-08	4.34E-08	5.63E-08	9.56E-08	1.33E-07	2.15E-07	6.62E-07	6.76E-07	5.58E-07	
PR212	0.	3.18E-05	3.36E-05	3.87E-05	4.50E-05	5.59E-05	8.32E-05	9.55E-05	1.01E-04	4.36E-05	3.28E-08	2.53E-08	
PR214	0.	2.65E-12	2.76E-12	3.04E-12	3.44E-12	4.23E-12	7.51E-12	1.09E-11	2.01E-11	3.32E-10	1.67E-08	6.09E-07	
BI210	0.	9.00E-14	9.54E-14	1.13E-13	1.34E-13	1.98E-13	5.61E-13	1.11E-12	3.31E-12	2.12E-10	1.67E-08	6.09E-07	
BI211	0.	3.30E-08	3.47E-08	3.82E-08	4.34E-08	5.63E-08	9.56E-08	1.33E-07	2.15E-07	6.62E-07	6.76E-07	5.58E-07	
BI212	0.	3.18E-05	3.36E-05	3.87E-05	4.50E-05	5.59E-05	8.32E-05	9.55E-05	1.01E-04	4.36E-05	3.28E-08	2.53E-08	
BI213	0.	1.90E-08	1.75E-08	1.74E-08	1.96E-08	2.52E-08	4.38E-08	6.24E-08	1.09E-07	9.42E-07	8.88E-06	5.87E-05	
BI214	0.	2.65E-12	2.76E-12	3.04E-12	3.44E-12	4.23E-12	7.51E-12	1.09E-11	2.01E-11	3.32E-10	1.67E-08	6.09E-07	
PO210	0.	5.58E-14	6.10E-14	7.21E-14	9.12E-14	1.38E-13	4.46E-13	9.43E-13	3.31E-12	2.12E-10	1.67E-08	6.09E-07	
PO211	0.	9.91E-11	1.04E-10	1.15E-10	1.30E-10	1.69E-10	2.87E-10	3.98E-10	6.46E-10	1.99E-09	2.03E-09	1.67E-09	
PO212	0.	2.04E-05	2.15E-05	2.48E-05	2.84E-05	3.57E-05	5.33E-05	6.11E-05	6.49E-05	2.79E-05	2.10E-08	1.62E-08	
PO213	0.	1.86E-08	1.71E-08	1.71E-08	1.92E-08	2.46E-08	4.28E-08	6.10E-08	1.07E-07	9.22E-07	8.68E-06	5.74E-05	
PO214	0.	2.65E-12	2.76E-12	3.04E-12	3.44E-12	4.23E-12	7.51E-12	1.09E-11	2.01E-11	3.32E-10	1.67E-08	6.09E-07	
PO215	0.	3.30E-08	3.47E-08	3.82E-08	4.34E-08	5.63E-08	9.56E-08	1.33E-07	2.15E-07	6.62E-07	6.76E-07	5.58E-07	
PO216	0.	3.18E-05	3.36E-05	3.87E-05	4.50E-05	5.59E-05	8.32E-05	9.55E-05	1.01E-04	4.36E-05	3.28E-08	2.53E-08	
PO218	0.	2.65E-12	2.76E-12	3.04E-12	3.44E-12	4.23E-12	7.51E-12	1.09E-11	2.01E-11	3.32E-10	1.67E-08	6.09E-07	
AT217	0.	1.90E-08	1.75E-08	1.74E-08	1.96E-08	2.52E-08	4.38E-08	6.24E-08	1.09E-07	9.42E-07	8.88E-06	5.87E-05	
RN219	0.	3.30E-08	3.47E-08	3.82E-08	4.34E-08	5.63E-08	9.56E-08	1.33E-07	2.15E-07	6.62E-07	6.76E-07	5.58E-07	
RN220	0.	3.18E-05	3.36E-05	3.87E-05	4.50E-05	5.59E-05	8.32E-05	9.55E-05	1.01E-04	4.36E-05	3.28E-08	2.53E-08	
RN222	0.	2.65E-12	2.76E-12	3.04E-12	3.44E-12	4.23E-12	7.51E-12	1.09E-11	2.01E-11	3.32E-10	1.67E-08	6.09E-07	
FR221	0.	1.90E-08	1.75E-08	1.74E-08	1.96E-08	2.52E-08	4.38E-08	6.24E-08	1.09E-07	9.42E-07	8.88E-06	5.87E-05	
FR223	0.	4.96E-10	5.20E-10	5.69E-10	6.42E-10	7.85E-10	1.34E-09	1.85E-09	3.01E-09	9.27E-09	9.46E-09	7.81E-09	
RA223	0.	3.30E-08	3.47E-08	3.82E-08	4.34E-08	5.63E-08	9.56E-08	1.33E-07	2.15E-07	6.62E-07	6.76E-07	5.58E-07	
RA224	0.	3.18E-05	3.36E-05	3.87E-05	4.50E-05	5.59E-05	8.32E-05	9.55E-05	1.01E-04	4.36E-05	3.28E-08	2.53E-08	
RA225	0.	1.86E-08	1.70E-08	1.77E-08	2.00E-08	2.52E-08	4.38E-08	6.24E-08	1.09E-07	9.42E-07	8.88E-06	5.87E-05	
RA226	0.	2.65E-12	2.78E-12	3.04E-12	3.44E-12	4.23E-12	7.51E-12	1.09E-11	2.01E-11	3.32E-10	1.67E-08	6.09E-07	
RA228	0.	7.09E-09	7.24E-09	7.55E-09	8.00E-09	8.87E-09	1.19E-08	1.44E-08	1.88E-08	2.53E-08	2.53E-08	2.53E-08	
AC225	0.	1.90E-08	1.75E-08	1.74E-08	1.96E-08	2.52E-08	4.38E-08	6.24E-08	1.09E-07	9.42E-07	8.88E-06	5.87E-05	
AC227	0.	3.54E-08	3.72E-08	4.06E-08	4.54E-08	5.61E-08	9.55E-08	1.32E-07	2.15E-07	6.62E-07	6.76E-07	5.58E-07	
AC228	0.	7.09E-09	7.24E-09	7.55E-09	8.00E-09	8.87E-09	1.19E-08	1.44E-08	1.88E-08	2.53E-08	2.53E-08	2.53E-08	
TH227	0.	3.34E-08	3.52E-08	3.86E-08	4.37E-08	5.54E-08	9.43E-08	1.31E-07	2.12E-07	6.53E-07	6.66E-07	5.50E-07	
TH228	0.	3.17E-05	3.40E-05	3.86E-05	4.44E-05	5.57E-05	8.31E-05	9.53E-05	1.01E-04	4.36E-05	3.28E-08	2.53E-08	
TH229	0.	1.59E-08	1.67E-08	1.82E-08	2.05E-08	2.52E-08	4.38E-08	6.24E-08	1.09E-07	9.42E-07	8.88E-06	5.87E-05	
TH230	0.	3.62E-09	3.63E-09	3.64E-09	3.66E-09	3.70E-09	3.87E-09	4.03E-09	4.44E-09	1.18E-08	8.54E-08	7.81E-07	
TH231	0.	1.77E-02	8.35E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	
TH232	2.77E-08	2.53E-08	2.53E-08	2.53E-08	2.53E-08	2.53E-08	2.53E-08	2.53E-08	2.53E-08	2.53E-08	2.53E-08	2.53E-08	
TH234	0.	1.27E-03	5.30E-04	9.21E-05	6.66E-06	3.48E-08	2.61E-17	2.83E-22	5.52E-22	5.38E-21	5.36E-20	5.32E-19	
PA231	0.	6.88E-07	6.90E-07	6.90E-07	6.90E-07	6.90E-07	6.90E-07	6.90E-07	6.90E-07	6.88E-07	6.75E-07	5.58E-07	
PA232	0.	1.01E-02	1.16E-09	1.51E-23	2.26E-44	0.	0.	0.	0.	0.	0.	0.	
PA233	0.	1.76E+01	8.15E+00	1.75E+00	1.73E+01	1.71E+03	1.05E+09	1.03E+09	1.03E+09	1.03E+09	1.03E+09	1.03E+09	
PA234M	0.	3.59E-01	5.30E-04	9.21E-05	6.66E-06	3.48E-08	2.61E-17	2.83E-22	5.52E-22	5.38E-21	5.36E-20	5.32E-19	
PA234	0.	3.42E-01	5.30E-07	9.21E-08	6.66E-09	3.48E-11	2.64E-20	2.83E-25	5.52E-25	5.38E-24	5.36E-23	5.32E-22	
U232	0.	1.11E-04	1.11E-04	1.11E-04	1.10E-04	1.10E-04	1.08E-04	1.06E-04	1.01E-04	4.24E-05	7.31E-09	0.	
U233	0.	8.99E-05	9.43E-05	9.73E-05	9.84E-05	9.81E-05	9.81E-05	9.81E-05	9.81E-05	9.80E-05	9.77E-05	9.40E-05	
U234	0.	9.50E-06	9.50E-06	9.50E-06	9.50E-06	9.50E-06	9.50E-06	9.50E-06	9.50E-06	9.50E-06	9.47E-06	9.24E-06	
U235	0.	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	7.90E-10	
U236	0.	2.13E-09	2.13E-09	2.13E-09	2.13E-09	2.13E-09	2.13E-09	2.13E-09	2.13E-09	2.13E-09	2.13E-09	2.13E-09	

TABLE VII. (contd.)

NUCLIDE RADIOACTIVITY, CURIES
BASIS = MT OF U CHARGED TO REACTOR

	CHARGE	DISCHARGE	1.MO	3.MO	6.MO	12.MO	36.MO	60.MO	120.MO	1200.MO	12000.MO	120000.MO
U237	0.	5.97E-03	2.62E-04	5.05E-07	4.46E-11	1.84E-12	1.67E-12	1.52E-12	1.21E-12	1.78E-14	2.54E-19	1.19E-19
NP236	0.	8.37E-09	8.44E-14	6.59E-39	0.	0.	0.	0.	0.	0.	0.	0.
NP237	0.	9.80E-10	1.03E-09	1.03E-09	1.03E-09	1.03E-09	1.03E-09	1.03E-09	1.03E-09	1.03E-09	1.03E-09	1.03E-09
NP238	0.	1.18E-03	5.10E-08	9.57E-17	7.77E-30	0.	0.	0.	0.	0.	0.	0.
NP239	0.	2.02E-12	2.02E-12	2.02E-12	2.02E-12	2.02E-12	2.02E-12	2.02E-12	2.02E-12	2.01E-12	1.85E-12	8.18E-13
PU236	0.	5.24E-10	5.14E-10	4.97E-10	4.48E-10	4.14E-10	2.55E-10	1.57E-10	4.64E-11	1.44E-20	0.	0.
PU238	0.	4.02E-06	4.09E-06	4.04E-06	4.08E-06	4.06E-06	4.00E-06	3.94E-06	3.78E-06	1.88E-06	1.70E-09	6.54E-32
PU239	0.	1.54E-09	1.54E-09	1.54E-09	1.54E-09	1.54E-09	1.54E-09	1.54E-09	1.54E-09	1.54E-09	1.50E-09	1.16E-09
PU240	0.	7.60E-10	7.60E-10	7.60E-10	7.60E-10	7.60E-10	7.60E-10	7.60E-10	7.59E-10	7.52E-10	6.86E-10	2.73E-10
PU241	0.	7.70E-08	7.67E-08	7.61E-08	7.52E-08	7.35E-08	6.69E-08	6.09E-08	4.82E-08	7.12E-10	1.02E-14	4.78E-15
PU242	0.	3.49E-13	3.49E-13	3.49E-13	3.49E-13	3.49E-13	3.49E-13	3.49E-13	3.49E-13	3.49E-13	3.49E-13	3.43E-13
PU243	0.	9.23E-08	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.53E-21
AM241	0.	2.77E-11	3.80E-11	5.83E-11	8.86E-11	1.48E-10	3.72E-10	5.75E-10	1.00E-09	2.32E-09	5.55E-10	5.09E-15
AM242M	0.	2.11E-12	2.11E-12	2.11E-12	2.11E-12	2.10E-12	2.08E-12	2.06E-12	2.02E-12	1.34E-12	2.21E-14	3.30E-32
AM242	0.	2.04E-08	2.11E-12	2.11E-12	2.11E-12	2.10E-12	2.08E-12	2.06E-12	2.02E-12	1.34E-12	2.21E-14	3.30E-32
AM243	0.	2.02E-12	2.02E-12	2.02E-12	2.02E-12	2.02E-12	2.02E-12	2.02E-12	2.02E-12	2.01E-12	1.85E-12	8.18E-13
CM242	0.	5.29E-09	4.71E-09	3.63E-09	2.47E-09	1.13E-09	5.24E-11	3.96E-12	1.66E-12	1.10E-12	1.81E-14	2.71E-32
CM243	0.	2.94E-13	2.94E-13	2.93E-13	2.91E-13	2.88E-13	2.76E-13	2.64E-13	2.37E-13	3.37E-14	1.15E-22	0.
CM244	0.	1.25E-10	1.25E-10	1.24E-10	1.23E-10	1.21E-10	1.12E-10	1.03E-10	8.55E-11	2.72E-12	2.96E-27	4.70E-28
CM245	0.	1.10E-14	1.10E-14	1.10E-14	1.10E-14	1.10E-14	1.10E-14	1.10E-14	1.10E-14	1.09E-14	1.01E-14	4.77E-15
CM246	0.	1.12E-15	1.12E-15	1.12E-15	1.12E-15	1.12E-15	1.12E-15	1.12E-15	1.12E-15	1.11E-15	9.69E-16	2.58E-16
CM247	0.	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.54E-21	2.53E-21
CM248	0.	4.76E-21	4.76E-21	4.76E-21	4.76E-21	4.76E-21	4.76E-21	4.76E-21	4.76E-21	4.76E-21	4.76E-21	4.67E-21
BK249	0.	1.57E-17	1.47E-17	1.28E-17	1.05E-17	7.01E-18	1.43E-18	2.57E-19	2.65E-20	0.	0.	0.
CF249	0.	3.38E-21	5.96E-21	1.04E-20	1.61E-20	2.46E-20	3.81E-20	4.08E-20	4.10E-20	3.44E-20	5.78E-21	1.16E-28
CF250	0.	1.27E-19	1.28E-19	1.28E-19	1.28E-19	1.25E-19	1.12E-19	9.97E-20	7.68E-20	1.33E-21	6.79E-32	6.79E-32
CF252	0.	6.89E-20	6.89E-20	6.89E-20	6.89E-20	6.89E-20	3.28E-20	1.56E-20	-4.84E-21	-2.78E-31	0.	0.
SUBTOT	2.77E-08	1.83E+01	8.15E+00	1.75E+00	1.74E-01	2.32E-03	8.04E-04	8.88E-04	9.26E-04	4.70E-04	1.85E-04	5.84E-04
TOTAL	2.77E-08	3.66E+01	8.15E+00	1.75E+00	1.74E-01	2.32E-03	8.04E-04	8.88E-04	9.26E-04	4.70E-04	1.85E-04	5.84E-04

TABLE VIII. Concentration of Important Transuranic Elements on Fuel Cladding Hulls from a Residual 0.1% UO₂ Fuel as Calculated by ORIGEN Code for One Year Cooling⁽²¹⁾

<u>Element</u>	<u>Grams of Element in 0.1% tonne of Exposed Fuel</u>	<u>Weight ppm on 330 kg(1 MTU) of Cladding Hulls</u>	<u>nCi/g of Hulls</u>
U	955	2893	11
Np	0.48	1.5	54
Pu	10	30	9×10^5
Am	0.14	0.4	538
Cm	0.03	0.1	1.97×10^4

the fuel is a stable oxide, and the Zircaloy surface has a zirconium dioxide surface layer a few microns thick,⁽¹⁴⁾ the likelihood of substantial diffusion of transuranic elements appears remote. The energetic radiation environment will increase the diffusion rates but even fast reactor fuel cladding has not shown large gradients of plutonium^(23,24) in the fuel cladding exposed in more energetic radiation environments at higher temperatures. The thin oxide layer that develops on the interior of the Zircaloy cladding during exposure is being formed in a heavily fuel contaminated environment and undoubtedly incorporates fuel contamination in pores, cracks and other imperfections, if not in the ZrO₂ lattice itself. Frequent observations^(25,26) of fuel oxide appearing to be bonded to the cladding could be explained as an intergrowth of the fuel oxide and the zirconium oxide. During early runs at NFS on Dresden⁽²⁷⁾ fuel an assay of the residual fuel in the hulls was made by treating the hulls for 8 hours in concentrated nitric acid. The corrosion rate of Zircaloy in concentrated nitric acid is so low⁽²⁸⁾ that the test would probably only leach the surface contamination in the hulls but in many cases significant amounts of fuel were leached from the hulls indicating that a significant portion of the fuel contamination was on the surface or could be leached from the oxide.

The shearing of the bundle of fuel rods requires the bundle be held by clamps, moved along contaminated tables, handled by contaminated manipulators, and most importantly contacted by the shear blade. All of these contacts are potential contamination points that will embed small fuel particles into the surface. The shear blades will be the most highly contaminated and exert the highest pressures which will embed particles in the surface and on the sheared surface. The extent that these particles will be subsequently removed in the dissolver will be dependent on whether they are smeared over with metal which will not be dissolved. The amount of smearing of irradiated Zircaloy should be minimal because its ductility has been substantially reduced by radiation^(11,12) damage. Nevertheless, some particles of fuel will undoubtedly be embedded and smeared over on the sheared surface. Each 10 micron particle so trapped will add about 1 nCi of transuranic contamination to the piece.

The external surface of fuel element cladding in a reactor develops a corrosion product layer of zirconium dioxide about 20 microns thick, and a deposited layer of corrosion product oxides from other parts of the system which vary between 3 and 30 microns.⁽¹⁴⁾ Ideally, there should be no transuranic contamination of either of these oxide layers, however, if ruptures have occurred in the reactor, the deposited layer can be significantly contaminated with transuranic elements. The treating of this material in the dissolver solution (dilute HNO_3) will undoubtedly remove some of the complex iron, chrome, nickel and copper oxides from the surface but will not remove all of them. The residual oxide will come to equilibrium with the highly contaminated dissolver solution and probably leave the process with more transuranic contamination than it started with unless it was severely contaminated to start with. No data on the absorbed transuranics from a dissolver treatment were located and the process of partial dissolution and absorption is too complex to attempt an estimate of the final surface concentration.

In summary, the characterization of a typical fuel cladding hull is a 1-inch piece of thin walled one-half-inch Zircaloy tubing covered with

scratched and fracture zirconium dioxide. On the average, it will have about 25 mg of fuel left attached to the inside surface or pressed and smeared in the cut ends. Most of the transuranic activity will be associated with the surface oxide or in a thin surface layer of metal, however, a small amount of transuranics activity will be uniformly distributed in the metal from the uranium and thorium impurities. This uniformly distributed activity will be primarily Pu^{241} which may not have to be removed to treat the waste as nontransuranic.⁽³⁾

Stainless steel end fittings and Inconel springs and spacers represent another portion of the hull inventory. The transuranic contamination of these alloys will only arise by activation of uranium and thorium impurities and from a contaminated reactor coolant crud deposition. If these alloys are contaminated by uranium or thorium to the same extent as the Zircaloy the transuranic contamination produced by irradiation would be roughly equivalent to the Zircaloy. The internal springs will be contaminated with fuel and operate at a higher temperature which may result in some diffusion of transuranics into the metal where it is in contact with fuel particles.

The induced radioactivity in the stainless steel and Inconel exclusive of transuranic elements is substantially higher than the Zircaloy. One computer estimate⁽²⁹⁾ of activation of the Inconel and stainless steel during irradiation in a PWR shows the predominant activities to be Co^{60} (82%) and Fe^{55} (17%) after 3 years' cooling and Ni^{63} (78%) and Co^{60} (21%) after 30 years' cooling with 0.28 and 0.03 Ci/g respectively. The Co^{60} is the isotope which will control the shielding requirements because of its 1.33 MeV gamma radiation.

WASTE TREATMENT PROCESSES

Process Requirements

The previously described waste stream is a low density, reactive, transuranic contaminated, radioactive material. The desired product is high density, unreactive, nontransuranic, radioactive material. The initial guidelines for transuranic contamination were taken from the AEC Radioactive Waste Management Manual⁽³⁾ which are quoted in part below.

"23. Transuranium-Contaminated Solid Waste--Those contaminated with certain alpha-emitting radionuclides of long half-life and high specific radiotoxicity to greater than 10 nanocuries/gram (10 microcuries/kilogram), subject to the following conditions and understandings:

- a. The radionuclides included are U^{233} (with its daughter products), plutonium, and transplutonium nuclides except Pu^{238} and Pu^{241} . (Note that Pu^{238} and Pu^{241} waste should be handled as transuranium-contaminated waste when so indicated by Pu^{239} impurities or when required by local burial criteria.)
- b. The value of 10 nCi/g is derived from the upper range of concentrations of Ra^{226} in the earth and is subject to modification based on long-term studies of nuclide migration in soil.
- c. The activity density may be averaged over the contents of individual shipping containers, such as 55-gallon drums, including materials added for shielding or sorption of liquids. Late discovery (for example, on recalculation of data) that an individual container is above this level will not be considered as necessitating its retrieval provided there is reasonable assurance that the average of the container and the balance of the associated containers is below the level.
- d. For typical Pu^{239} waste at this activity density, it is recognized that indirect measurements or estimates and administrative controls must be used instead of direct external measurements. An example of such administrative controls is the establishment of specific in-plant working areas from which typical wastes have been established by suitable studies as being either above or below the control value."

Reuse of Metal

The initial treatment plan objectives were to separate the cladding hull waste stream into two waste streams: (1) a low volume transuranic contaminated waste, and (2) a dense, nonleachable, nontransuranic, non-reactive, zirconium-stainless steel-Inconel alloy waste which could be

disposed of by burial. An interest in reuse of the metallic waste stream subsequently developed and a logical treatment of the various alternatives was examined as shown in Figure 1. The major changes from the original plan that reuse would introduce are briefly--separation of the various alloys, avoidance of chemical contamination, higher melting temperatures, and radioactive refabrication procedures. Inasmuch as reuse implies storage, some relaxation of the final transuranic contamination might be permissible. Relaxation of final transuranic contamination of material for reuse from the 10 nCi/g level has not yet been quantified.

The main waste stream will have residual radioactivity even though it will have most of the transuranic contamination removed. The disposal of these radioactive zirconium alloy ingots will require that the residual radioactivity is relatively nonleachable by groundwaters. However, an absolute leach rate which would be universally acceptable has not yet been determined because of the variability of groundwater chemistry and movement. Presumably, the natural corrosion resistance of stainless steels and zirconium would be sufficient to adequately control the release of the dilute radioactivity in the ingots. The small transuranic waste stream can be incorporated into or treated like high-level waste.

The reuse of cladding and structural alloys will require storage of the material during which the material should produce no threat of dispersion because of manmade or natural disasters. The natural corrosion resistance and insolubility of corrosion products for massive Zircaloy, stainless steel, and Inconel should be sufficiently low to preclude any objectionable dispersal of corrosion products in typical environments.

The density of a waste stream should be maximized consistent with heat dissipation and economics of transport, storage or disposal. The high thermal conductivity of a metal together with the low heat generation rate of the cladding hulls will not cause any problems for densified fuel hulls, but may produce some heating of large piles of uncooled, low density hulls. The high surface area of the original waste stream must be reduced to control

FIGURE 1. Treatment Logic for Decontamination and Densification of Fuel Hulls

Decontamination and Densification of Fuel Hulls

1. The use of the chop-leach head-end process has been based on the anticipated low cost of separate treatment and burial of cladding hulls and the economy of using stainless steel dissolvers and conventional reprocessing schemes. If the cost of hull disposal is substantially increased or the cost of tritium removal from waste streams requires the alternative head-end processes such as liquid metal clad dissolution, gas phase clad oxidation, or even aqueous dissolving of the clad, the fuel hull waste will be combined with other waste.

2. It is assumed there is currently no alternative to storage for waste streams heavily contaminated with transuranic elements.

3. The disposal of a chemically reactive, high volume, transuranic-contaminated waste streams is assumed to be currently unacceptable.

4. The mixture of stainless steel, Inconel and Zircaloy will have to be segregated into three waste streams for processing. This separation can probably be done magnetically.

5. No Reuse - the decontamination of the cladding hulls is for the purpose of removing transuranic contamination to such a degree (<10 nCi/g) that the resulting product can be more easily stored. The resulting transuranic and non-transuranic waste streams must be minimized to produce only a small increase in total mass and a large reduction in total volume and chemical reactivity. The transuranic contaminated waste stream should be a very small fraction of the total. The process considered most likely to accomplish this result at present is a gas phase oxide metathesis followed by an

aqueous bath to do a final decontamination by a transuranics and during melting contamination to be purified standpoint of waste volume. The higher is performed for the separate presumably a desire to keep of the total fluorine, and

It may restrict

6. Reuse - is a cold wall waste stream. Decontaminates detrimental such restriction the waste still. The waste alloys or to produce an be handled directly processed by different desirable. Similar that all of waste to cast the may reduce that can be directly from those melting and alloying waste. The consumable arc

The decontamination of Zircaloy will be a case because of its ductility and corrosion the oxide from free alloys which melting. The waste by their surface have transuranic contamination is any event might decontamination similarly may large (0.3%) amounts aqueous processing during melt must eventually a very high D.F. to reuse, thus high a decontamination intervention achieved to allow amount of fuel material as non-contaminated at less which will ding hulls. intensive subsequent

7. The estimation of cladding hull surface to make in 19, the oxygen This large surface in the melt must makes storage use. A technique rial sensitivity of complete carefully controlled conium, is not employed. The steels and Inconels the low density by suitable oxygen water or cast keep the temperature

21. The storage of densified material will present few problems. The storage of the decontaminated hulls presents a problem in corrosion and inflammability which will require cool storage and oxide removal before melting.

22. Storage of these ingots should not present major problems unless the guidelines change or the decontamination cannot be achieved to meet the current standards. The storage of hulls will require cool, high volume storage and may contain more than 10 nCi/g of transuranic contamination.

23. & 24. The refabrication of these alloys will have to be done in a radioactive facility even if the transuranic content of these alloys has been reduced below 10 nCi/g. The attached tables of ORIGEN computer runs for a Zircaloy-2 with normal expected impurities show several long half life materials such as ^{63}Ni and ^{93}Zr which though they present little dose rate problem will require control of atmosphere, etc., to prevent ingestion or inhalation. It would thus be desirable to do as little refabrication work as can be done to acquire a satisfactory product. For example, if directly extrudable billets were made in 18 of the proper alloy, the tubing could be extruded with a minimum of handling. The actual fabrication of fuel will require radioactive facilities in any event and the only changes will be in the handling of the final fuel element.

25. The waste streams from fuel manufacture will be slightly radioactive from the cladding. The insolubility of zirconium dioxide in reactor coolant should not add any new radioactivity to the reactor system.

leaching and pyrophoricity. From an economical point of view, the highest density will require a minimum mass of shielding for transport. The required product is thus a minimum surface-to-mass ratio of metal consistent with a minimum shielding requirement for transport.

In summary, the required product is a high density, low surface-to-volume ratio metal ingot or bar with less than 10 nCi/g of transuranic contamination (may be higher for reuse) and good corrosion resistance to natural environments (reactor environments if intended for reuse). The transuranic waste should be a minimum volume of material compatible with waste storage treatments.

DECONTAMINATION

General

The decontamination of a waste stream is accomplished by selectively removing one or more types of contaminants which will allow some desired use or disposal of the major portion of the stream. In the case of a solid waste stream, such as the cladding hulls, the location, chemical form and amount of the contamination to be removed must be known to design an optimum decontamination process. As previously discussed (page 6), the transuranic contamination of the cladding hulls is mostly as oxides on surfaces or pressed into the surface during handling. Substantial amounts of contamination are probably included in the zirconium oxide corrosion product layer and a still smaller amount uniformly distributed in the zirconium metal from activation of original uranium and thorium impurities in the metal.

If all of the contamination is to be removed in one step, it will have to occur during melting because of the Pu^{241} and Cm^{244} daughters of impurity uranium in the zirconium. If Pu^{241} and Cm^{244} can be considered nontransuranic⁽³⁾ it is probable that the hulls can be satisfactorily decontaminated in a molten salt bath that will remove the oxide layer. If the contamination is completely on the surface, the hulls might be satisfactorily decontaminated by an aqueous process. The high (10^5) total decontamination factor that will

be necessary to go from 0.4% fuel contamination of the cladding hulls (0.1% of input uranium) to less than 10 nCi/g will be difficult to assure in the melting process step, even if it were attainable. A two-step decontamination process removing most of the activity prior to melting appears more desirable. If oxide removal is required either because it contains too much transuranic contamination or to avoid oxygen contamination of the Zircaloy during melting, the molten salt or a modified aqueous process will probably be required.

In order to minimize the mass of the waste stream the decontaminant must be either reclaimable or easily reduced in mass to a small fraction of its original mass. The low volatility of the fused salts, either from melt flux or from a lower temperature wash, makes a reduction in mass very difficult. Therefore, the fused salts must be purified and reused or the volume of the waste stream will be substantially increased. The aqueous processes can frequently be selected to achieve nearly complete incineration of the decontaminant or to be amenable to a side stream cleanup. Decontaminant process selection must consider the ability to minimize the mass of contaminated waste.

The basis of selection of premelting decontamination processes for laboratory testing are the following:

1. The reagents must effectively remove UO_2 and PuO_2 .
2. The reagents must be potentially reusable with treatment or easily decontaminated for disposal.
3. The reagents should corrode Zircaloy metal uniformly at a slow rate.
4. The reagents should dissolve or remove zirconium oxide surface layers.
5. The cost of reagent usage should be reasonable.
6. The removed zirconium, uranium, and transuranic contamination should be easily concentrated to a small volume.

7. Process should be easily conducted remotely with reliable equipment.
8. Process should not create any potential chemical reaction hazards.

Aqueous Decontamination

The technology of aqueous decontamination processes has been extensively developed for reactor and equipment decontamination.^(30,31) Most processes can be carried out in open vessels at temperatures up to the boiling point. The aqueous waste can frequently be concentrated, treated and stored with existing plant waste streams. The chemical technology for treatment has been studied and the economics can be optimized. The main disadvantage to aqueous decontamination is that zirconium dioxide corrosion product layers, which probably incorporate transuranic contamination, cannot be effectively dissolved without severe attack on the substrate metal. If the zirconium is to be reused, the oxygen level must be reduced before melting by removing the corrosion product layer.

Aqueous decontaminants can be formulated to have the desired corrosivity for the process application. In the case of cladding hulls that may have small particles of fuel smeared into the cut surface, corrosion of the zirconium metal is desirable to remove the thin, smeared metal and exposed the fuel for dissolution. If one assumes a smeared metal thickness of one mil or less and a processing time of 30 minutes, a metallic corrosion rate of about 2 mils/hour appears to be optimum. The actual surface area corroded by the solvent will be a small fraction of the total surface area because most of the surface is covered by an oxide film which will only be lightly corroded. The removal of the decontaminant from the treated metal is usually accomplished by water rinsing.

The corrosion of zirconium alloys in aqueous solutions is generally very low because the zirconium oxide corrosion films act as a barrier. Corrosion rates in the mil per hour range are only seen in hot concentrated

sulfuric and phosphoric acids or dilute acid fluoride solutions.^(32,33) Concentrated sulfuric acid can be reduced in volume and reused by volatilizing the SO_3 at high temperature and readsorption in water while the concentrated phosphoric acid would be difficult to clean up and nearly irreducible in volume. Dilute fluoride solutions with organic or nitric acids can be easily concentrated by distillation and the distillate reused. Even though volatilization of some of the fission products such as iodine, tritium, and ruthenium will take place during distillation,⁽³⁴⁾ the recovered water and acids should be reusable for the decontamination process. If nonvolatile organic acids are used, they can be calcined to carbon dioxide and water and fresh makeup acid added to the distillate. The extent that any of these potential decontaminants must be cleaned up in order to decontaminate with the desired efficiency is unknown.

The main purpose of the decontaminant is to dissolve the adherent fuel particles which are on or in the surface. These particles are currently high temperature fired uranium dioxide but may include plutonium oxide in the future. The dissolution of uranium oxide is usually accomplished with hot nitric acid or a buffered hydrogen peroxide mixture. Small additions of hydrofluoric acid have been used⁽³⁵⁾ to promote the solution of plutonium oxide-uranium oxide mixtures, thorium oxide and zirconium-uranium oxide ceramics.⁽³⁶⁾ Mixtures of hydrogen peroxide-gluconic acid and ammonium oxalate were developed and used⁽³⁷⁾ for dissolving uranium oxide-plutonium oxide fuel rupture debris from the PRTR rupture loop and gas system. This solvent proved to be reasonably effective in dissolving the fuel debris but was not corrosive to Zircaloy. A somewhat similar mixture containing oxalic acid-hydrogen peroxide-hydrogen fluoride has been reported⁽³⁸⁾ to be mildly corrosive to Zircaloy and zirconium oxide. Presumably this solution would effectively dissolve all fuel oxides as well as remove very thin zirconium oxide coatings and dissolve zirconium metal. The concentrated acids, sulfuric and phosphoric, also dissolve ceramic plutonium and uranium oxides.⁽³⁹⁾ It is not known whether they can be compounded to dissolve or remove zirconium oxides fast enough to avoid excess corrosion of the metal. Other

acids such as HCl have been used⁽³⁶⁾ to dissolve uranium and zirconium oxide under heat and pressure but this treatment would attack the metal severely and be difficult to perform.

None of the aqueous processes are known to meet all of the process selection guidelines previously discussed (page 25). None of the aqueous reagents is known to dissolve the ZrO_2 formed in service without serious attack on the metal. The decontamination of the reagents for reuse is not certain except for the distillation process. The concentrated reagents like phosphoric or sulfuric acid must be cleaned up sufficiently by a suitable side stream technique even though the exact nature of such a side stream cleanup has not been determined.

The most serious disadvantage with aqueous processes is their inability to remove the corrosion product oxide layers from Zircaloy without dissolving substantial amounts of the underlying metal. If the Zircaloy is heated for melting with the oxide layer on the hulls it will contaminate the resulting ingot with one to two percent oxygen which will adversely affect its corrosion resistance for normal applications. The oxide layer will also cause problems during melting by preventing intimate contact between the various alloying materials (Inconel and stainless steel) in the melt and may require excessive temperatures to achieve a fluid melt. The effect of oxygen on the room temperature corrosion of zirconium-stainless steel alloy was not found. The deduction that the internal oxide layer of the fuel cladding probably contains substantial transuranic contamination within the oxide layer, if correct, throws an increased decontamination load on the melting process. If the melting process decontamination develops large decontamination factors, this increased transuranic contamination could be tolerated. However, large DF's in the melting process are not expected which casts a shadow on processes that will not remove the ZrO_2 layer.

The aqueous decontamination of stainless steel and Inconel hardware can, in general, be accomplished. Reagents such as nitric-hydrofluoric acid, nitric acid-ceric ion, oxalic-citric-peroxide-fluoride, and sulfuric

acid-hydrogen peroxide have been effective in removing transuranic contamination from stainless steels.^(31,38,40) Most of the stainless steel and Inconel scrap has not been heavily contaminated with transuranic elements because it was not exposed to the fuel. The oxide films formed on this material by the reactor coolant can be very difficult to dissolve and would probably withstand the nitric acid treatment in the dissolver, if it was fed into the dissolver with the sheared fuel. This surface oxide layer would adsorb transuranic elements from the dissolver solution and would have to be removed in a subsequent decontamination step. The internal fuel element springs are exposed to fuel particles during operation and may have some transuranic element diffusion into the metal where they contact the top insulator pellet and reach a temperature significantly higher than the cladding. Where the insulator pellet is not uranium dioxide, the chances for transuranic element diffusion is greatly decreased. To the extent the contamination is diffused into the metal it cannot be removed by surface decontamination without a substantial corrosion of the metal.

If fuel dissolution residues are to be separated before decontamination, (i.e., Inconel, stainless steel, and Zircaloy), the decontamination process can be suited to the particular metal and contaminant to be removed. If material is not separated, the decontamination process must be an optimization for all materials and contamination characteristics. The most likely optimization will occur with the acid fluoride solutions such as nitric-hydrofluoric acid and ammonium oxalate-citric acid-hydrogen peroxide-hydrofluoric acid. Available fluoride in these solutions will have to be kept quite low in order to avoid severe attack on the zirconium metal before the fuel oxides have been dissolved and the stainless steels defilmed. The addition of a fluoride complexing agent such as aluminum ion will control the available free fluoride ion and at the same time allow enough fluoride to be available to avoid a rapidly changing bath composition. This type of formulation was reported⁽³⁸⁾ to produce several orders of magnitude reduction in the corrosion of Zircaloy when an equimolar aluminum ion addition was made. The corrosivity of these solutions can also be regulated by pH adjustment or other fluoride complexing agents. Thus, the most likely aqueous decontaminating agent for all three metals is an acid fluoride solution complexed or pH buffered to control the corrosivity toward Zircaloy.

Such a solution will be an excellent solvent for residual fuel and corrosion product oxides except zirconium and be easily reduced to a minimum volume for disposal.

Molten Salt Decontamination

Molten salts have been periodically examined as process fluids in the nuclear industry and found to have some very desirable characteristics. The use of fused $\text{NaF-ZrF}_4\text{-UF}_4$ in the Aircraft Reactor Experiment⁽⁴¹⁾ and $\text{LiF-BeF}_2\text{-UF}_4$ in Molten Salt Reactor Experiment⁽⁴²⁾ have generated extensive research^(43,44,45) on the properties of these particular system and engineering equipment to utilize them. More closely related to decontamination are the dissolution of fuel in a fused LiF-NaF-ZrF_4 for the fluoride volatility process⁽⁴⁶⁾ and the initial study⁽⁴⁷⁾ of fused salt decontamination of stainless steel from separations facilities. Fused salts have also been studied for electrodeposition media⁽⁴⁸⁻⁵²⁾ and reaction media⁽⁵³⁾ whenever the low temperature or hydrogen or oxygen of aqueous media interferes with the desired process. In the case of the zirconium dioxide-zirconium system, any aqueous medium that rapidly dissolves zirconium dioxide will even more rapidly attack zirconium metal because the metal is only stable as long as an oxide layer separates the metal from the aqueous medium.

In general, fused salt systems can be contained, pumped, electrolysed, purified, etc., in a manner similar to aqueous systems, however, fused salt systems are not exposed to air or water except in a few special cases where the salts are oxidants and nonhydrolyzable. If a salt system is to remove the protective layer from a metal, the salts will have to be thermodynamically more stable than the metal being stripped or the salt will be reduced and the metal oxidized. The common cations more stable than zirconium are the alkali metals most of the alkaline earths, yttrium, cerium, scandium, uranium and most of the actinides. The anion must not be reducible by the zirconium, which leaves the halides. Even though zirconium will resist attack by the hydroxides, it is only because of the formation of an oxide layer.

Zirconium and its alloys dissolve their own oxide layers when heated to form a zirconium-oxygen alloy. If the oxide layer is contaminated by transuranic elements, they will probably be absorbed into the metal also

and thus defeat the purpose of the decontamination. Zirconium starts dissolving its own oxide at an appreciable rate about 500°C and the rate increases rapidly with temperature. Thus, for a mildly corrosive salt system treatment of about 30 minutes, the upper temperature limit is probably about 600°C. This may be adjusted somewhat by increasing the corrosivity of the salt bath.

The most pertinent previous work was the studies of fuel dissolution in the Fluoride Volatility Process.⁽⁵⁴⁾ The LiF-NaF-ZrF₄ system flushed with hydrogen fluoride will dissolve fuel oxides, zirconium oxide and zirconium metal. The maximum uranium oxide dissolved⁽⁵⁵⁾ with no HF at 1150°C was 0.1 weight percent for pure salt mixtures, and 2.3 weight percent for salt with 45 weight percent UF₄. Early corrosion data for Zircaloy-2 in ZrF₄-KF-NaF at 675°C was as high as 25 mils/hr⁽⁴⁶⁾ when flushed with excess HF to convert the zirconium to zirconium fluorides. Later corrosion data⁽⁵⁶⁾ for many runs at 650°C showed dissolution rates between 1 and 2 mils/hr and reduced this rate to about 1/2 mil/hr at 500°C.⁽⁵⁷⁾ The corrosion rate of Zircaloy without HF flushing would be less but exact rates would depend on salt impurities including the zirconium and uranium oxides dissolved. The tolerance of the salt bath to an oxide solute such as zirconium oxide was not found but it obviously must be converted to fluoride by HF treatment or otherwise removed from the bath. Conceivably, the zirconium oxide might be removed from the bath by electrolysis together with transuranics similar to a plating operation.^(48,51)

The action of the fused fluoride baths on stainless steels and Inconel is to remove the oxide coating and lightly corrode the metal.⁽⁵⁷⁾ The effect of corroding Zircaloy in electrical contact with the easily reduced iron and nickel alloys is to reduce their rate of corrosion when HF is being flushed through the bath.⁽⁴⁶⁾ Presumably the corrosion rate without an HF flush will be similarly reduced. The cathodic member of the couple (stainless steel and Inconel) will probably tend to plate out easily reduced impurities and fission products on their surface such as ruthenium, nickel, etc., but the transuranic elements will probably not be plated out because of their strong electropositive character.

The oxidizing salt baths will either be too corrosive to zirconium or will not remove the oxide coating, however, they have been effectively used for decontaminating stainless steels.⁽⁴⁷⁾ The decontamination achieved for cerium was usually better than 10^3 which would probably apply to transuranic elements as well. The problems inherent in many oxidizing salt baths are the potential for violent chemical reactions, especially with zirconium, and the volatilization of oxides such as ruthenium. The advantages of the oxidizing salts is their lack of sensitivity to air and moisture.

The hull waste stream has an average density of 1.1 kg/l and a maximum attainable density of 6.8 kg/l. The difference in these densities determines the minimum volume of salt that must be used for dipping the hulls. This large volume of salt is essentially irreducible in mass and thus must be reused many times if the volume and mass of the waste streams are to be minimized. The fused salt decontamination⁽⁴⁷⁾ of relatively dense stainless steel scrap produced a volume of waste salt about three times the volume of the original steel, on a used once basis. Obviously, the use of a fused salt decontaminant is predicated on cleanup and reuse of the salt. The Molten Salt Breeder reactor concept has promoted research on side stream purification of a fused $\text{LiF-BeF}_2\text{-UF}_4\text{-ThF}_4$ salt by reductive extraction into liquid metals.⁽⁵⁸⁾ Plating research has explored electrolytic reduction of metals from fused fluoride baths.⁽⁴⁸⁻⁵¹⁾ These methods can probably be adapted for gross transuranic element removal in the fused salts but there is currently no information on how clean the salt bath must be to avoid serious redeposition of transuranic elements on the zirconium. Another major factor in waste stream volume will be the drag out of fused salts on the Zircaloy as it is removed from the bath. The fused salts are fairly viscous (5 to 8 centipoises at 600°C)⁽⁴¹⁾ and the drag out of salt on the large surface area Zircaloy will be substantial. These salts can be removed from the surface easily in aqueous solutions, however, the recovery of the salt for reuse from the water phase will be difficult and expensive. If the salt covering on the hulls is transferred to the melting operation,

some of it will be volatilized (mostly ZrF_4) and some will form a slag on the melt. Any easily reducible elements in the slag will probably alloy with the metal.

Examining the fused salt decontamination methods in the frame of the basic requirements (page 25) shows they will meet many of the requirements but not all of them. The salt systems will dissolve fuel oxides and corrosion product oxides, can be adjusted to corrode Zircaloy at a proper rate, can be a chemically stable system, and the transuranics can probably be removed in a small volume from a side stream. The equipment can be made reliable but will not be as simple as aqueous equipment because of the exclusion of the atmosphere and moisture. The cost of reagent usage and potential waste stream volume from drag out of salts on the high surface area hulls appears to be a problem area. If the salts can be satisfactorily recovered for reuse from the melting operation, the waste stream size and salt replacement cost might be reduced to a reasonable level.

Decontamination of Molten Metal

The necessity of decontaminating the molten metal rests on four currently uncertain bases: (1) The amount of uranium and thorium contamination of the original zirconium, stainless steel, and Inconel alloys; (2) the necessity for removing Pu^{241} and Cm^{244} contamination; (3) the amount of transuranic element diffusion from the fuel into the cladding alloy during service; and (4) the effectiveness of surface decontamination prior to melting. If all of the conditions were favorable, no melt decontamination would be necessary and standard zirconium melting practice could be used. If all of the conditions were unfavorable, decontamination factors during melting as high as 10^5 might be necessary to reduce the product to less than 10 nCi/g of all transuranic elements. Such a high DF in the melting process will be difficult to achieve and difficult to assure by analyses. Thus, the amount of decontamination necessary during the melting process should be held to a practical minimum.

If premelting surface decontamination is complete, uranium and thorium contamination of the original metal are 1 and 2 ppm, respectively, no diffusion of fuel transuranic into the clad metal occurs beyond that removed in premelting decontamination, and the Pu^{241} and Cm^{244} must be removed; a melting decontamination factor of about 10 will be necessary (see Tables IV and VI). This DF is a more reasonable level and probably can be achieved by fluxing and/or absorption on the melt container.

The physical properties of the hull waste stream of importance to melt decontamination are shown in Table IX. The Inconel, stainless steel and Zircaloy must be treated separately if the material is to be reused (see Figure 1) at a later date. In addition, no undesirable (for later reactor use) chemical contamination of the metals must occur in processing. As can be seen in Table IX, the separate treatment of the alloys will raise the treatment temperature by 400 and 700°C for the nickel alloys and Zircaloy, respectively. This increase in temperature for the Zircaloy together with controlling desired contamination severely restricts the melting methods, crucible materials and potential fluxing agents which might be useful for decontamination. The melting or fluxing of stainless steels and Inconel are not as difficult to achieve.^(47,59,60,61)

Zircaloy melting practice for nuclear components is usually by a vacuum, consumable arc process utilizing a water cooled copper crucible which freezes the adjacent layer of Zircaloy.^(62,63,64) In affect, the Zircaloy is melted in a Zircaloy crucible in contact with nothing else but a noble gas. Some volatile impurities may be partially removed in the process but none of the transuranic elements are very volatile. Tritium would be nearly quantitatively removed. Thus, decontamination by this process would be small. Early zirconium melting was frequently done^(64,65) in graphite crucibles with induction heating, but the zirconium picked up enough carbon from the crucible to affect its corrosion resistance in various acids.⁽³²⁾ Some laboratory scale melts of zirconium⁽⁶⁵⁾ and the chemically similar titanium⁽⁶⁷⁾ have been made by melting under calcium fluoride and lanthanum fluoride slags by electric arc and induction heating. Both of these methods

TABLE IX. Properties of Zircaloy, Stainless Steel and Inconel Alloys Used in Fuel Elements

ASTM-B-353 Gr RA-1 (20) (Zr-2)		ASTM-B-353 Gr RA-2 (20) (Zr-4)		304 Stainless Steel (70)		Inconel 718 (71)		Inconel 600 (71)	
<u>Chemical</u>									
Sn - 1.2 to 1.7 %		Sn - 1.2 to 1.7%		Cr - 18 to 20%		Ni - 50 to 55%		Ni - 72% Min.	
Fe - 0.07 to 0.20%		Fe - 0.18 to 0.24%		Ni - 8 to 10%		Cr - 17 to 21%		Cr - 14 to 17%	
Cr - 0.05 to 0.15%		Cr - 0.07 to 0.13%		Mn - 2%		Mo - 2.8 to 3.3%		Fe - 6 to 10%	
Ni - 0.03 to 0.08%		Ni - 0.007% Max.		Si - 1%		Nb+Ta 4.7 to 5.5%		Mn - 1% Max.	
Zr - Bal		Zr - Bal		Fe - Bal		Co - 1% Max		Si - 0.5% Max.	
						Ti - 0.65 to 1.15%		Cu - 0.5% Max.	
						Al - 0.2 to 0.8%		S - 0.015% Max.	
						Si - 0.35%		C - 0.15% Max.	
						Mn - 0.35%			
						Cu - 0.3%			
						Fe - Bal			

Density

6.55 g/cc	6.55 g/cc	8.0 g/cc	8.21 g/cc	8.43 g/cc
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Melting Range

1800 to 1850°C	1800 to 1850°C	1400 to 1450°C	1260 to 1335°C	1354 to 1413°C
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involved the use of a water cooled copper melting crucible which introduced no contamination because a layer of calcium fluoride was frozen on the crucible wall. Titanium has been melted⁽⁶⁸⁾ in a partially reduced yttrium oxide crucible by induction heating. The crucible was kept cooler than the melt which was held to less than 100°C superheat. Some oxygen and yttrium pickup occurred but was low enough to make the technique of interest, especially as a potential solid adsorbent for melt decontamination. The reaction with zirconium may be much more than with titanium and in fact was stated to "react rapidly"⁽⁷²⁾ with molten zirconium. Thorium oxide, beryllium oxide, and zirconium oxide react with molten zirconium less than most oxides⁽⁶⁴⁾ and may prove useful for solid state adsorption.

The melting of stainless steels and Inconel, under flux, is more easily accomplished and can be conducted in refractory oxide crucibles, vacuum arc melted, induction melted, resistance heated or gas fired. Previous work^(61,59) on removing uranium from stainless steel, copper, and aluminum indicates that uranium in nickel and stainless steel was reduced by $3 \cdot 10^3$ to less than 1 ppm by melting in an arc furnace. In this case the uranium concentrated in the slag. The uranium in the aluminum was only reduced a factor of 2 during melting. Thus, melting of stainless steel and Inconel under a simple flux should purify the melt of transuranic elements which are, with the exception of neptunium, all more electropositive than uranium. The use of a fluoride flux would make all the transuranic elements more stable than uranium. The decontamination of stainless steel melts employing oxide type fluxes was investigated⁽⁴⁷⁾ as a method of disposal. The system used in the investigation did not stir the melt to equilibrate it with the flux but was still capable of producing DF's of 10^2 to 10^3 for cerium-praseodymium which should be reasonably representative of plutonium. However, the removal of transuranic element to a fractional ppb is still uncertain (see Table X).

The melting of a Zircaloy-10 to 30% stainless steel alloy was reported⁽⁷¹⁾ to be possible at 1150°C with reasonably fluid melts. The melting was accomplished in a graphite crucible in a resistance furnace

under high vacuum. The interaction of the melt and crucible was not severe, but carbon concentration in the ingot was not determined. If graphite can serve as a crucible material the use of fluoride type fluxes will be facilitated and the decontamination of the melt will be higher than in the cases where no flux is used. No decontamination data for stainless steel-Zircaloy melts were located, and thus an accurate decontamination factor cannot be calculated. The zirconium content of the melt is the controlling factor on removing transuranic elements by selective oxidation. Zirconium is an active element with a free energy of oxidation similar to uranium. The selective oxidation of the transuranic impurities and subsequent extraction into a salt flux will be more difficult than with the stainless steels. The flux must be more stable than zirconium to avoid reaction with the melt but at the same time must contain an oxidant that will react with the transuranic elements. The amount of transuranic elements expected in normally contaminated Zircaloy-2 (see Table X) is less than 1 ppb and thus will probably not require a specific oxidant beyond the normal impurities in the flux and atmosphere of the melt furnace. The similarity of the free energies of the zirconium and some of the transuranic elements (see Table XI) will mean that the zirconium will compete actively for the oxidant and establish equilibria which only slightly favors some of the transuranic elements. The transuranic element(s) present from original uranium contamination of the alloys is primarily plutonium which will be extracted more efficiently than neptunium. This marginal extraction expected of transuranic element, together with the probability that elements such as uranium and thorium may not be extracted at all, underscore the importance of minimizing the transuranic contamination input to the melting operation by efficient premelting decontamination.

The substantial effort⁽⁷³⁻⁷⁶⁾ to develop pyrochemical fuel reprocessing schemes for irradiated uranium alloys produced data which should be somewhat relevant to the decontamination of Zircaloy-stainless steel melts because of the similarity of free energies for uranium and zirconium and the similar processing temperature. In general the extraction of plutonium from

TABLE X. Concentration of Several Transuranic Elements
Derived from Activation of 1 ppm Uranium or
Thorium Impurity in Zircaloy as Calculated by
ORIGEN Code for Five Years' Cooling⁽¹⁹⁾

Element	1 ppm U Contamination*		Element	1 ppm Th Contamination*	
	conc ppb	nCi/g		conc ppb	nCi/g
U	972	0.003	Th	913	0.4
Np	0.1	0.02	U	48	0.8
Pu	9	88	Np	0.006	$4 \cdot 10^{-6}$
Am	0.3	0.9	Pu	0.001	0.02
Cm	0.03	2.2			
Others			Others		
Total	<0.4	<0.1	Total	<0.04	2.3

*Assumed 253 kg Zircaloy/MTU

TABLE XI. Free Energy of Zirconium and Important
Transuranic Element Fluorides

(kg calories per gram atom of fluoride)⁽⁷⁸⁾

Compound	1400°K	1800°K	2100°K
Zr F ₃	92	86	84
Zr F ₄	90	88	87
U F ₃	87	79	74
U F ₄	87	83	82
Np F ₃	94	88	84
Pu F ₃	99	92	90
Am F ₃	102	96	93

irradiated uranium by $\text{CaF}_2\text{-LiF-UF}_4$ fluxing at 1300°C was in the range of 95 to 99%.⁽⁷⁹⁾ Similar chloride based systems employing $\text{CaCl}_2\text{-UCl}_3$ at 1150 and 1200°C extracted between 49 and 89% of the plutonium in the uranium metal.⁽⁸⁰⁾ Other more electropositive elements such as cerium, rare earths, strontium, and cesium were more than 90% transferred to the fused salt flux.^(74,75,79) No data for extremely low levels of plutonium were found. Zirconium only slightly extracted into the fused salt layer.

The bases for selecting potential melt decontamination process for further study are substantially affected by requirements to reuse the metal at a later time. If reuse is desired, the bases of selection in decreasing order of importance are:

1. The extent of chemical contamination during melting.
2. The extent of removal of transuranic impurities.
3. The ease of remote operation and reliability.
4. The minimizing of waste stream mass.

If reuse is not indicated, the bases of selection in order of decreasing importance are:

1. The extent of transuranic element removal.
2. The minimizing of waste stream mass.
3. The ease and reliability for remote operation.
4. The extent of chemical contamination of the melt which will affect its low temperature corrosion resistance.

Reviewing the process information available and the bases for material to be reused, it is doubtful that any melting practice other than vacuum arc with a cold wall crucible will produce an acceptable Zircaloy ingot. Even this process will require extreme care to avoid oxygen contamination. The calcium fluoride slag^(66,67) should be tested but the current indication of fluoride pickup appears undesirable. The melt decontamination of stainless steel by a flux⁽⁴⁷⁾ appears to be about equal to the uranium decontamination achieved in straight arc melting.^(59,61) If this same relationship is

established for plutonium, the use of a separate flux will be superfluous. Thus, for the reuse of material the process choice appears to be a nonconsumable arc melt in a cold wall crucible with alloying adjustment as needed. The feed to the system would have to be very clean and provision made to condense the volatilized impurities so they could be periodically removed.

In the case of nonreuse of metals where decontamination becomes most important process, the metals can be blended to produce an alloy of lower melting point. The reduced temperature of the melt and the relaxed requirements on chemical contamination will probably allow the use of a graphite crucible and a flux during melting. Induction heating effectively stirs the melt which will enhance the decontamination by promoting flux-metal equilibrium not inhibited by long diffusion paths. The flux will have to be recovered for reuse, probably by electrolysis and unusable crucibles reduced to a minimum waste by combustion. Crucible ashes and electrolysis products become the transuranic waste stream from melting.

In summary, none of the melt decontamination processes are known to reduce transuranic contamination into the fractional ppb range. The vacuum arc process for Zircaloy will probably produce very little decontamination. The fluxed induction melted Zircaloy-stainless steel alloy may produce decontamination factors of about 10. The arc melted stainless steels may produce DF's of 100 to 1000. However, none of the processes may reduce the level of transuranic contamination to the very low level desired.

Total Process Considerations

A review of the three potential modes of decontamination shows that each one has a potential problem. The aqueous process will not remove the corrosion formed oxides from the metal without severe metal attack. The fused salt processes will have problems with "drag out" which will increase waste stream size. The molten metal processes will probably not attain large decontamination factors. One potential solution to these interacting problems would be a gaseous treatment of the hulls that would make the corrosion product oxide subject to removal in an aqueous solution. Hydrogen

fluoride in fused fluoride salt mixtures at 600°C is known to dissolve zirconium dioxide twice as fast at the metal.⁽⁵⁴⁾ Hydrogen fluoride-oxygen mixture reacts with Zircaloy cladding and completely oxidizes it at 550°C.⁽⁸¹⁾ Thus it would appear that hydrogen fluoride gas at about 500°C would interact with the zirconium dioxide and render the oxide liable to attack by available aqueous decontaminants probably by forming $ZrOF_2$ in the film. Other gaseous fluorides such as CF_4 might be used. In such a process the fluoride film would partially stay in place and hopefully inhibit the gas reaction with the metal sufficiently to avoid rapid metal reaction and hydrogen production from the metal-hydrogen fluoride reaction. Extensive hydrogen production would be undesirable because the metal would react with it at this temperature. The gaseous fluoride would be diluted with inert gas to give a concentration where temperature control could be maintained. The inert gas would be recirculated through a bed or trap to remove water and HF.

The fuel cladding without a protective oxide layer can be decontaminated to bright metal and slightly etched in a nitric-hydrofluoric acid solution or an oxalic-hydrofluoric acid-hydrogen peroxide solution. The bright metal can then be melted after rinsing and thorough drying. The metal melting process would depend on the need for further decontamination and the intentions on reuse.

The waste from this process would consist of the flaked off zirconium fluoride from the gaseous process, the evaporated aqueous decontaminant, any flux from the melting process that could not be reclaimed and reused and broken or unusable crucibles and molds from the melting process. If graphite molds and crucibles are employed, they can be reduced to a low volume ash. If cold wall crucibles are used, they will be quite durable.

The tritium content of Zircaloy hulls may run 50 ppb (0.19 ml/kg STP)^(11,82) while total hydrogen will be 50 to 150 ppm (560 to 1640 ml/kg STP). The hydrogen and tritium will be mostly removed during melting. However, some of the tritium may exchange with hydrogen containing decontaminants or gases during the premelting decontamination.

The tritium and hydrogen from the melting operation can be readsorbed in a previously cast zirconium alloy billet which is nonuniformly heated (700 to 300°C). The nonuniform heating is necessary to avoid the trace oxygen and nitrogen in the system from fouling the hydrogen absorption and at the same time try to achieve the lower equilibrium hydrogen pressure over zirconium at the lower temperature. The tritium released to the aqueous decontaminants will be largely recycled by reuse of the distillate from evaporation. Any tritium released to a gas phase pretreatment will be concentrated in a small volume of water which can be added to the other aqueous distillate or otherwise treated.

In summary, the above treatment process offers the potential to treat cladding hulls with more or less standard equipment and with low total waste volume. It depends on several uncertain factors such as a gas treatment to make the oxide removable by aqueous etchants, the character of the hull waste stream, and melt decontamination factor. However, problem areas with the other methods of decontamination appear to be even more difficult to resolve. Only experimentation with actual hull samples will resolve some of the uncertainties.

DENSIFICATION

The densification of the waste stream includes the hull metal, the transuranic contamination, the decontaminants, and the scrap equipment. The metal portion of the waste will be increased in density from about 1.1 kg/l to about 6.8 kg/l which represents the maximum densification obtainable by any means. The reduction of the transuranic contaminated waste stream volume by distillation, combustion, or electrolytic purification is necessary to avoid creating much more waste volume than the beginning material.

The aqueous decontaminants can be compounded to be completely volatile and thus recoverable by distillation. Small amounts of volatile activities such as tritium, ruthenium, etc., will be transferred with the distillate⁽³⁴⁾

and tend to increase in concentration in the distillate with time. The residues left in the evaporator will be fluorides and oxides of zirconium, iron, chromium, and nickel together with the transuranic contamination and removed fission products. Assuming complete removal of the crud, corrosion product oxide and average corrosion of 0.5 mil of metal, this waste will approximate 25 kg/MTU or about 8% of the input hull stream.

If nonvolatile decontaminants are used they will have to be purified by a side stream system and reused. A cleanup system for a concentrated acid was not investigated. The use of a nonvolatile concentrated decontaminant would be undesirable from a waste minimization standpoint if it has to be replaced frequently.

The use of molten salt decontaminants requires that they be cleaned up and reused to avoid increasing the waste volume. The cleanup of uranium, thorium, plutonium, and rare earths from fused fluoride melts has been extensively investigated for the molten salt reactor and the molten salt breeder reactor. The process evolved^(45,58) generally included volatilization of uranium as a hexafluoride and extraction of other elements into liquid bismuth-lithium metal reductants. The precipitation of uranium and protoactinium as oxides from the fused fluoride salt was also studied. Electrolytic plating from fused fluorides^(49,50,51) is a simpler but less selective method of purification. The cathode deposits are frequently granular⁽⁵¹⁾ and would have to be filtered from the system unless a liquid metal cathode is used. A liquid metal cathode could then be circulated to a subsequent vessel to remove the plated transuranic elements. The oxides of the transuranic elements are soluble enough that oxide precipitation would not yield a high purity salt though the process may be satisfactory if the salt phase can stand a substantial transuranic element contamination and still perform satisfactorily.

A second source of waste associated with the fused salt decontamination is the separation of the fuel hulls from the salt after submerging them in the somewhat viscous medium. The high surface area hulls will have heavy

surface contamination of salt which will generate a waste stream difficult to recover if removed in an aqueous wash. If the salt is carried into the melting process it will generally be too volatile and too contaminated with transuranic elements to use as a flux. If the molten salt is volatilized from the hulls before melting, it will leave its transuranic contamination on the hulls because the transuranics are nonvolatile.

Decontamination during melting is potentially the product of four processes (i.e., solid adsorption, flux solution, volatilization, or oxide slagging). Not all of these will be operating significantly in any one melt system. Solid adsorption has usually been studied as the adsorption from the melt by the crucible wall. A crucible which is slightly reactive will transform some of the melt impurities to oxides or carbides and leave them attached to the crucible wall, if they are more active than the melt.⁽⁷⁷⁾ This process may also create some unattached oxides which separate by gravity from the melt and can be removed. If the crucible or another solid introduced in the melt can be recovered for reuse by simple scraping, heating, or treatment by volatile solvents (such as HNO_3), the waste generated is minimal. If the crucible cannot be reused, the waste stream will be large or small depending on whether it is graphite which can be burned or oxide which is irreducible. None of the transuranic metals or oxides are very volatile and thus will not be volatilized from the melt itself, but because of their active nature they may be partially reacted in an arc and concentrate to an extent on furnace side walls or a condensor deliberately added for that purpose. The decontamination of zirconium by this process will not be large because it is also an active metal very close to the the chemical activities of the transuranics. The amount of waste produced by this process will be minimal. The use of a flux will suppress volatilization but serves as an extractant and stabilizer for oxidized impurities in the melt. The equilibrium between the flux and the melt must favor the flux by a large margin because there will only be a fractional ppb of transuranics to be removed from the melt. When these transuranic impurities build up in the flux it will no longer extract the elements

with the desired distribution ratio. For example, if the flux is one-tenth the volume of the melt and the distribution favors the flux by a factor of 10, then about 50% would be extracted the first use. The second use with the same starting conditions would only remove 25%. If the flux is only used once, a waste flux stream at least half the volume of the metal ingots will be required. If distribution coefficients are not very favorable, two or three times that much may be necessary. The cleaning and reuse of a flux is possible by separate process such as electrolysis but the purity required to extract a ppb level impurity from a melt will be difficult to maintain unless very favorable distribution coefficients are found.

In summary, the minimization of the total waste stream, the cost of reagents and the ease of operation all favor a dilute aqueous decontamination which removes all contamination not alloyed with the metal followed by melting in graphite crucibles or cold wall crucibles with a low decontamination factor necessary in the melt. When actual metal contamination levels and melt-flux distribution ratios are determined, this view may be changed. The largest uncertainty in the proposed process is finding an easy to use, safe gaseous method for making the zirconium oxide film subject to removal by dilute aqueous reagents.

DEACTIVATION

The large surface area Zircaloy hull waste stream presents a substantial area for corrosion ($100 \text{ m}^2/\text{MTU}$). The waste stream, with the exception of sawing fines, is not subject to spontaneous ignition if kept cool. The extremely low corrosion rate of Zircaloy in natural environments should not produce much spontaneous heating but intense radiation may produce some if relatively adiabatic conditions exist. The surface contamination of the hulls from the fuel and the dissolver solution will probably be easily leached in natural waters. The contamination in the Zircaloy will be very tightly bound because of the insolubility of the zirconium dioxide corrosion product layer in natural waters. The fact that zirconium dioxide sands are

found naturally after surviving the dissolution of their parent rock formations is testimony to their insolubility in fresh water streams.⁽⁸³⁾ The retention of nearly all of the oxide corrosion product during reactor exposure of the metal is further indication of its insoluble characteristic.

The conversion of the large surface area contaminated hull waste stream to a nontransuranic low surface area casting will reduce the surface area to about $1.3 \text{ m}^2/\text{MTU}$, improve the heat transfer so internal heating will be insignificant, and reduce the corrosion availability of the internal contamination linearly with the reduction of surface area. The corrosion resistance of Zircaloy-stainless steel-Inconel ingots is not known and will have to be determined. The corrosion resistance⁽⁶⁴⁾ of Zircaloy is reduced for some engineering applications if it is melted in graphite crucibles which add a significant amount of carbon to the melt. The combined effect of graphite crucibles at a much lower temperature in the presence of stainless steel alloy will also have to be determined as well as the effect of any flux used which may contaminate the melt chemically. Somewhat higher uniform corrosion rates could be tolerated but the incidence of localized corrosion, such as intergranular corrosion, dendritic attack or destructive hydriding, could not be. Corrosion testing must necessarily be done under environments typical of storage or disposal which have not yet been defined.

CONCLUSIONS

Examination of decontamination and densification methods applicable to the chop-leach hull waste stream has indicated processes which may be effective. Some deficiency has been found with each aqueous, molten salt, or melting decontamination process. A potential process based on reasonable speculation has been described which requires experimental verification in the following areas:

1. Analyses of typical fuel element hulls to determine amount, location, chemical form and kind of transuranic contamination.
2. Test of gaseous fluoride process to make corrosion product covered metal subject to dilute aqueous etching without seriously attacking the metal.
3. Determination of transuranic element distribution coefficients between applicable fluxes and Zircaloy-stainless steel melts.
4. Determine corrosion resistance of Zircaloy-stainless steel-Inconel alloys of typical compositions melted in graphite and cold wall crucibles with and without flux.
5. When a process can be reasonably defined, estimate the cost of the process on an industrial scale.

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