DR-423

K-1734

MASTER

NUCLEAR SAFETY STUDIES FOR
LOW ENRICHMENT FLUORIDE VOLATILITY
FUEL REPROCESSING PLANTS

AUTHORS:

K. E. Habiger
D. L. Breton

UNION CARBIDE CORPORATION
NUCLEAR DIVISION
OAK RIDGE GASEOUS DIFFUSION PLANT

operated for the ATOMIC ENERGY COMMISSION under U. S. GOVERNMENT Contract W-7405 eng 26

**UNION
CARBIDE**

OAK RIDGE GASEOUS DIFFUSION PLANT
P. O. Box P
Oak Ridge, Tennessee 37830

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Printed in the United States of America. Available from Clearinghouse for Federal
Scientific and Technical Information, National Bureau of Standards,
U.S. Department of Commerce, Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

— LEGAL NOTICE —

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

- A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Date of Issue: September 4, 1968

Report Number: K-1734

Subject Category: Criticality
Studies

NUCLEAR SAFETY STUDIES
FOR LOW ENRICHMENT FLUORIDE VOLATILITY FUEL REPROCESSING PLANTS

K. E. Habiger
D. L. Breton

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

UNION CARBIDE CORPORATION
NUCLEAR DIVISION
Oak Ridge Gaseous Diffusion Plant
Oak Ridge, Tennessee

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

A B S T R A C T

In cooperation with the national laboratories, the Oak Ridge Gaseous Diffusion Plant has participated in conceptual and hardware development studies related to the design of fluoride volatility fuel reprocessing plants for low enrichment nuclear reactor fuels. As a part of this effort, criticality calculations have been made to establish reactivities of various assemblies of uranium and plutonium with other volatility process materials. These results were then used as bases for estimating the geometry or fissile mass limitations for the conceptual plant processing equipment. The DTF-II and ANISN one-dimensional S_n multigroup transport codes were used in the S_4 option for the calculations. For uranium, calculations were made for cases involving homogeneous mixtures of uranium dioxide and alumina, uranium dioxide and water, uranium hexafluoride and hydrogen fluoride, and disodiumoctofluorouranate and sodium bifluoride. The maximum uranium-235 assay considered was 5%. Included in the plutonium studies were calculations for plutonium dioxide, plutonium hexafluoride, and plutonium tetrafluoride-sodium fluoride complex; calculations were also made for homogeneous mixtures of plutonium dioxide and water, plutonium dioxide and alumina, plutonium hexafluoride and hydrogen fluoride, and plutonium tetrafluoride-sodium fluoride complex and sodium bifluoride. Various geometries, including cylinders, spheres, and slabs were considered. In the plutonium studies, all plutonium was assumed to be plutonium-239 except in one calculation series where a plutonium isotopic distribution similar to that expected in spent light water reactor fuel was considered. It was concluded that, based on the assumptions made, it should be possible to design most of the equipment in a volatility plant to be geometrically safe for uranium and plutonium, provided water is excluded from the process vessels. Mass control for plutonium might be necessary in large vessels, however, if the possibility of plutonium segregation is allowed.

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION	17
SUMMARY	18
CALCULATION PROCEDURES	18
CHECKS WITH CRITICAL EXPERIMENTAL DATA	20
Uranium Cross Sections	20
Plutonium Cross Sections	20
URANIUM CRITICALITY STUDIES	23
URANIUM DIOXIDE-ALUMINA MIXTURES	23
URANIUM DIOXIDE-WATER MIXTURES	25
URANIUM HEXAFLUORIDE-HYDROGEN FLUORIDE MIXTURES	25
DISODIUM OCTOFLUOROURANATE AND SODIUM BIFLUORIDE MIXTURES	27
PLUTONIUM CRITICALITY STUDIES	30
POWER REACTOR PLUTONIUM	33
PLUTONIUM DIOXIDE CALCULATIONS	36
Plutonium Dioxide-Water Mixtures	36
Plutonium Dioxide Slabs	36
Plutonium Dioxide Cylinders	36
Plutonium Dioxide Spheres and Spherical Shells	40
Plutonium Dioxide-Alumina Mixtures	42
Plutonium Dioxide-Alumina Cylinders	42
Plutonium Dioxide-Alumina Spheres	42
PLUTONIUM HEXAFLUORIDE CALCULATIONS	45
Plutonium Hexafluoride Cylinders	45
Plutonium Hexafluoride Spheres	45

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

TABLE OF CONTENTS
(Continued)

	<u>Page</u>
Plutonium Hexafluoride-Hydrogen Fluoride Mixtures	45
Plutonium Hexafluoride-Hydrogen Fluoride Cylinders	45
Plutonium Hexafluoride-Hydrogen Fluoride Spheres	49
Plutonium Hexafluoride with Reinforced Phenolic Foam Reflectors	49
PLUTONIUM TETRAFLUORIDE-SODIUM FLUORIDE COMPLEX AND SODIUM BIFLUORIDE SYSTEMS	54
Cylinder Calculations	55
Slab Calculations	55
CONCLUSIONS	55
ACKNOWLEDGEMENT	57
BIBLIOGRAPHY	58
APPENDIX	61

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

LIST OF TABLES

<u>Table</u>		<u>Page</u>
I	Summary of Selected Criticality Calculations for Uranium and Plutonium Systems	19
II	Computed Multiplication Factors and Critical Experiment Data for Unreflected Assemblies Containing Plutonium . .	21
III	Computed Multiplication Factors and Critical Experiment Data for Plutonium Nitrate Solutions	22
IV	Plutonium Isotope Compositions in Irradiated Fuel	34
V	Critical Thicknesses for Infinite Slabs Containing Plutonium Dioxide	38
VI	Critical Masses for Plutonium Dioxide Spheres and Shells	40
VII	Critical Radii for Infinite Cylinders Containing Homogeneous Plutonium Hexafluoride-Hydrogen Fluoride Mixtures	48
VIII	Critical Spheres Masses for Homogeneous Plutonium Hexafluoride-Hydrogen Fluoride Mixtures	49
IX	Composition of Reinforced Phenolic Foam Insulation	50
X	Effective Neutron Multiplication Factors for Infinite Cylinders of Plutonium Hexafluoride	53
XI	Effective Neutron Multiplication Factors for Plutonium Hexafluoride Contained in Annular Cylinders	54
A-I	Critical Radii for Infinite Cylinders Containing Homogeneous Uranium Dioxide-Water Mixtures	61
A-II	Radii Corresponding to k_{eff} 's of 0.9 and 1.0 for Infinite Cylinders Containing Uranium Hexafluoride and Hydrogen Fluoride	62
A-III	Radii for Infinite Cylinders Containing $UF_6 \cdot 2NaF \cdot HF \cdot NaF$ Mixtures for k_{eff} 's of 0.9 and 1.0	62
A-IV	Critical Radii for Plutonium Dioxide Spheres	63
A-V	Critical Masses for Plutonium Dioxide Spheres	63

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

LIST OF TABLES
(Continued)

<u>Table</u>		<u>Page</u>
A-VI	Critical Radii for Infinite Cylinders of Plutonium Dioxide	64
A-VII	Critical Radii for Infinite Cylinders Containing Homogeneous Plutonium Dioxide-Alumina Mixtures	64
A-VIII	Critical Masses of Plutonium Dioxide-Alumina Spheres	65
A-IX	Critical Radii for Infinite Cylinders of Plutonium Hexafluoride	65
A-X	Critical Masses of Plutonium Hexafluoride Spheres	66
A-XI	Critical Radii for Infinite Cylinders of Plutonium Tetrafluoride-Sodium Fluoride and Sodium Bifluoride	66

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
1	Infinite Neutron Multiplication Factors for Mixtures of Uranium Dioxide and Alumina	24
2	Critical Radii for Infinite Cylinders of Uranium Dioxide and Water Mixtures	26
3	Radii for Infinite Cylinders of Uranium Hexafluoride and Hydrogen Fluoride Mixtures	28
4	Infinite Neutron Multiplication Factors for Mixtures of $\text{UF}_6 \cdot 2\text{NaF}$ and $\text{HF} \cdot \text{NaF}$	29
5	Radii for Infinite Cylinders of $\text{UF}_6 \cdot 2\text{NaF}$ and $\text{HF} \cdot \text{NaF}$ Mixtures	31
6	Critical Mass of PuO_2 Spheres for Varying Thicknesses of Water and Alumina Reflectors	32
7	Calculated Critical Mass of Spheres Containing PuO_2 Powder	35
8	Critical Mass for Spheres of Plutonium Dioxide and Water Mixtures	37
9	Critical Radii for Infinite Cylinders of Plutonium Dioxide	39
10	Plutonium Mass in Plutonium Dioxide Spheres Versus Effective Neutron Multiplication Factors	41
11	Critical Radii for Infinite Cylinders of Plutonium Dioxide and Alumina Mixtures	43
12	Critical Masses for Spheres of Plutonium Dioxide and Alumina Mixtures	44
13	Critical Radii for Infinite Cylinders of Plutonium Hexafluoride	46
14	Critical Masses of Plutonium Hexafluoride Spheres	47
15	Critical Radii for Infinite Cylinders of Plutonium Hexafluoride Versus Phenolic Foam Reflector Thickness	51

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

LIST OF FIGURES
(Continued)

<u>Figure</u>		<u>Page</u>
16	Effective Neutron Multiplication Factors for Infinite Cylinders of Plutonium Hexafluoride	52
17	Critical Radii for Infinite Cylinders of $\text{PuF}_6 \cdot \text{NaF}$ and $\text{HF} \cdot \text{NaF}$ Mixtures	56

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

NUCLEAR SAFETY STUDIES
FOR LOW ENRICHMENT FLUORIDE VOLATILITY FUEL REPROCESSING PLANTS

For several years, the United States Atomic Energy Commission has sponsored a number of programs related to the development of fluoride volatility processes as potential alternatives to established aqueous schemes for recovering values from irradiated nuclear fuel. In the dry gas routes, fuels are volatilized as the hexafluorides, thereby achieving decontamination from nonvolatile fission product compounds. Further purification by selective sorption, distillation, or other techniques is then used to separate the uranium hexafluoride and plutonium hexafluoride from volatile fission products.

One of the principal advantages of the fluoride volatility route is that only gas-solids contacting operations are employed in the primary processing steps and aqueous solutions, with their good neutron moderating capabilities, are almost entirely avoided. Characteristically, aqueous plant design considerations include use of soluble neutron absorbers, concentration control, and stringent equipment size restrictions with concomitant multiline staging of vessels in order to avoid nuclear criticality situations. For the most part, however, such considerations do not apply to the volatility process. Of course, certain size and mass limitations do exist in a fluoride volatility plant, and fixed neutron absorbers might be necessary in some instances; however, there generally appears to be much more latitude in certain aspects of choice of equipment and/or batch sizes than for aqueous plant design.

Since fiscal year 1964, the Gaseous Diffusion Development Division of the Oak Ridge Gaseous Diffusion Plant has participated with the national laboratories in conceptual and hardware development studies leading to the design of a commercial fluoride volatility fuel reprocessing plant. Included in these studies were computer calculations made to establish reactivity of various assemblies of uranium, plutonium, and other volatility process materials. These results were used as bases for estimating the geometry or fissile mass limitations involved in the conceptual volatility plant studies. Thus, the conceptual reprocessing plant designs could reasonably be assumed to be satisfactory from a nuclear safety viewpoint.

Although a wealth of information on uranium and plutonium criticality does exist in the form of calculations and experiments; much of it is concerned with high uranium-235 enrichments, with aqueous or metal systems, or with other systems not directly applicable to the uranium and plutonium oxide and fluoride compounds that will be encountered in a volatility plant. Thus, additional nuclear safety studies were considered essential.

This report is a summary of the criticality calculations performed for a fluoride volatility fuel reprocessing plant handling spent fuel from a light water reactor. Earlier, similar information for high enrichment fuel reprocessing schemes had been developed⁽²⁴⁾. Although slanted toward

the volatility process, the results of this report may be applicable to other situations. It should be recognized, however, that, although the calculations should be valid, the conclusions as to mass limitations, size restrictions, etc, cannot be utilized in general without corroboration of the assumptions involved.

SUMMARY

The DTF-II and ANISN one-dimensional S_n transport codes were used to perform criticality calculations for the uranium and plutonium compounds expected in a fluoride volatility plant. The codes were used in the S_4 option with 16-group cross sections and were shown, after some modification of uranium-238 cross-section data, to be sufficiently accurate for conceptual design studies by comparing code estimates with the results of critical experiments.

A summary of the results of criticality calculations performed for various uranium and plutonium systems is shown in table I. Since, in most cases, a range of densities or concentrations was studied, the data in table I are not complete, and the text should be consulted for other results and trends. In addition to the cases presented in the table, effective neutron multiplication factors were calculated versus infinite cylinder radii or sphere masses for systems of uranium hexafluoride and hydrogen fluoride, for plutonium dioxide, and for plutonium hexafluoride. Also not shown in the table are results for plutonium dioxide slab and spherical shell calculations and comparisons of critical plutonium dioxide sphere masses for various plutonium isotopic distributions typical of fuel discharged from light water thermal reactors.

It was concluded that, in the absence of water or other good neutron moderating materials, there would be no nuclear safety problems due to uranium in the main fluid-bed reactor of a fluoride volatility plant. Mass limits for plutonium would be necessary in this vessel and its associated filters, however, if the possibility of plutonium segregation is allowed. With the possible requirement of neutron absorbing materials in certain cases, it should be feasible to design the remaining equipment in the plant to be geometrically safe for both uranium and plutonium.

CALCULATION PROCEDURES

The DTF-II⁽⁴⁾ and ANISN⁽⁵⁾ one-dimensional S_n multigroup transport codes were employed in all of the criticality calculations described in this report. These codes were used in the S_4 mode and, in almost all cases, with 16-group cross-section data. Most of the calculations were made with the ANISN code, a DTF-II successor, although DTF-II was used for some of the initial uranium system studies. The two codes are basically the same, except that ANISN is written in FORTRAN IV and has several more options, including general anisotropic scattering, while DTF-II is written

TABLE I

SUMMARY OF SELECTED CRITICALITY CALCULATIONS FOR URANIUM AND PLUTONIUM SYSTEMS

Fissile Material*	Density, g/cc	Diluent Material Compound	Density, g/cc	Reflector		Minimum Critical Conditions**			
						Infinite Cylinders		Spheres	
				Compound	Thickness, cm	Fissile Compound, vol %	Critical Radius, cm	Fissile Compound, vol %	Critical Mass, kg Pu
UO ₂	10.9	Al ₂ O ₃	2.5			All k _∞ 's less than 1.0			
UO ₂	10.9	H ₂ O	1.0	H ₂ O	30	20	12.8		
UF ₆	5.0	HF	1.0	H ₂ O	30	30	25.6		
UF ₆ ·2NaF	4.16	HF·NaF	2.16	H ₂ O	30	35	39.2		
PuO ₂	3.50			Al ₂ O ₃	30	100+			
PuO ₂	3.50			Al ₂ O ₃	30	100	8.0	100	43.9
PuO ₂	3.63	H ₂ O	1.0	Al ₂ O ₃	30			57.26	11.4
PuO ₂	3.63	Al ₂ O ₃	2.5	Al ₂ O ₃	30	20	21.1	20	113.7
PuF ₆	6.0			H ₂ O	30	100	6.9	100	25.8
PuF ₆	6.0	HF	1.749	H ₂ O	30	0.5	12.7	0.5	0.55
PuF ₆	6.0			Phenolic Foam	100	100	10.5		
PuF ₄ ·NaF	6.47	HF·NaF	2.16	H ₂ O	30	100	6.7		
PuF ₄ ·NaF	6.47			H ₂ O	30	100s			

* Uranium-235 isotopic composition = 5%. All plutonium is plutonium-239.

† Thickness of Infinite Slab = 3.22 cm.

§ Thickness of Infinite Slab = 3.98 cm.

** In most cases, the plutonium systems did not exhibit minima between the end points of the independent variables studied.

in FORTRAN II and allows only a first order Legendre expansion of the scattering cross sections. Other differences between the codes are listed in reference (5); for the types of computations made in this study, it was felt that these differences did not significantly affect the consistency of the results.

CHECKS WITH CRITICAL EXPERIMENTAL DATA

Prior to making the fluoride volatility system calculations, it was decided to test the accuracy of the cross sections used with the computer codes. This was done by comparing the results of computer calculations for both uranium and plutonium systems with the results of critical experiments.

Uranium Cross Sections

ORNL and PCTR^(13,15,16) experimental values of the infinite multiplication factor (k_{∞}) for undermoderated uranium were employed in the uranium cross-section checks, and it was found that the 16-group Hansen and Roach⁽⁷⁾ cross sections used in the codes tended to predict low values of k_{∞} . To remedy this, the uranium-238 absorption cross sections in groups 8 through 12 were adjusted by applying reduction factors until the computed and experimentally determined k_{∞} values were in agreement. Groups 8 through 12 included the uranium-238 resonance region. The reduction factors finally chosen are given in appendix I of reference (17).

Plutonium Cross Sections

The plutonium cross sections, obtained from Hansen and Roach⁽⁷⁾ and from the GAM-II code⁽¹⁰⁾, were first checked using both the Jezebel plutonium metal data of Los Alamos⁽⁹⁾ and the plutonium dioxide-polystyrene data of the Hanford Laboratories⁽²²⁾. The experimental data and calculated results for these assemblies are shown in table II. Each assembly was unreflected. In both cases, actual critical dimensions were input to the code, and effective neutron multiplication factors were calculated. As shown in table II, the percentages of error in the calculated effective multiplication factors were 0.04% for the Jezebel assembly and 3.0% for the plutonium dioxide-polystyrene assembly.

TABLE II

COMPUTED MULTIPLICATION FACTORS AND CRITICAL EXPERIMENT DATA
FOR UNREFLECTED ASSEMBLIES CONTAINING PLUTONIUM

	<u>Jezebel Assembly(9)</u>	<u>Plutonium Dioxide- Polystyrene Assembly(22)</u>
Plutonium Density, g/cc	15.66	1.12
Plutonium-240*, %	4.5	2.2
Hydrogen-to-Plutonium Ratio	0	15
Critical Mass, kg	16.28 ± 0.05	-
Critical Radius, cm	-	18.58 ± 0.22
Reflector	none	none
Experimental k_{eff}	1.0	1.0
Calculated k_{eff}	1.0004	1.030
Percent Error in Calculated k_{eff}	0.04	+ 3.0

* The remainder of the plutonium was plutonium-239.

Comparisons were also made with the results of Hanford Laboratory plutonium nitrate critical experiments(19,20,21). The experimental data and ANISN results for five plutonium nitrate spheres reflected with water are shown in table III. Again, critical dimensions were used as input to the code, and effective neutron multiplication factors were computed. Except for one case, the error in the calculated effective multiplication factor was less than 1%. It was concluded, therefore, that the accuracies of the calculation method and of the plutonium cross sections were satisfactory for both moderated and relatively poorly moderated situations.

It should be pointed out that several of the early calculations performed for plutonium systems, described in a later section, were made using the uranium-235 fission spectrum in the code instead of the plutonium spectrum. Several check calculations were made to determine the error introduced because of this, and the differences in results were found to be less than 0.5% in each case. Furthermore, the error was in the conservative direction; i.e., estimated critical dimensions were slightly

TABLE III

COMPUTED MULTIPLICATION FACTORS AND CRITICAL EXPERIMENT DATA FOR PLUTONIUM NITRATE SOLUTIONS

Plutonium Concentration, g/l	Acid Molarity	Water, g/l	Total Nitrate, g/l	Specific Gravity	H/Pu	Critical Volume, liters	Calculated k_{eff}	Percent Error in Calculated, k_{eff}
140*	2.2	882	284	1.307	171	12.97	0.994	- 0.6
44.5†	5.49	825	386	1.261	522	23.24	0.991	- 0.9
96*	1.67	919	203	1.219	258	12.99	0.996	- 0.4
53†	5.95	790	424	1.277	423	21.51	0.981	- 1.9
34†	2.06	930	163	1.124	741	22.52	0.991	- 0.9

* Contained 4.6% plutonium-240; reflected with 0.049-inch stainless steel plus water. The remaining plutonium was plutonium-239.

† Contained 4.6% plutonium-240; reflected with 0.044-inch stainless steel plus water. The remaining plutonium was plutonium-239.

smaller when using the uranium spectrum. Thus, it was decided that the use of the uranium fission spectrum for the plutonium calculations was acceptable for this study.

URANIUM CRITICALITY STUDIES

Criticality calculations were performed for uranium dioxide and uranium hexafluoride systems, since these compounds are two of the most prevalent forms of uranium present in the fluoride volatility process. A third uranium compound, U_3O_8 , is also present in large quantities. Criticality calculations for this compound were not made, however, since the uranium density in U_3O_8 is lower than in uranium dioxide; thus, the results of the calculations for uranium dioxide should be conservative when applied to vessels which contain U_3O_8 . It was assumed that the uranium-235 assay was 5%, which is higher than even the initial enrichment of present-day light water power reactor fuels, thereby adding a further degree of conservatism to the results as applied to a volatility plant. Additional conservative assumptions, such as that voids in powder or pellet beds were completely filled with moderator or fissile material and that the beds were fully water reflected, were also usually made.

The uranium system criticality studies included calculations for mixtures of uranium dioxide and alumina, uranium dioxide and water, uranium hexafluoride and hydrogen fluoride, and disodiumoctofluorouranate ($UF_6 \cdot 2NaF$) and sodium bifluoride ($HF \cdot NaF$). The calculations primarily centered around spherical or cylindrical geometries, since, provided the basic assumptions are reasonable, these cases should bracket the actual physical situations expected. Most of the results are presented in graphical form to show trends, while tabulations of some of the computed results are given in the appendix for more precise interpretation.

URANIUM DIOXIDE-ALUMINA MIXTURES

Infinite neutron multiplication factors were calculated for various homogeneous mixtures of uranium dioxide and alumina. The uranium dioxide density was assumed to be 10.9 g/cc, which is near its theoretical density(6), while the alumina density was assumed to be 2.5 g/cc, which is somewhat greater than its typical bulk density(8).

As shown in figure 1, the infinite neutron multiplication factors are less than 1.0 for all uranium dioxide volume fractions, suggesting that mixtures of slightly enriched uranium dioxide and alumina cannot maintain a nuclear chain reaction in the absence of good neutron moderating materials and, therefore, are inherently safe for homogeneous geometries. Heterogeneous geometries of uranium dioxide and alumina, with a water-filled lattice, are considered unlikely in view of volatility plant design precautions.

DWG. NO. G-68-207

Figure 1

INFINITE NEUTRON MULTIPLICATION FACTORS FOR
MIXTURES OF URANIUM DIOXIDE AND ALUMINA

URANIUM DIOXIDE-WATER MIXTURES

Unlike the previous case for uranium dioxide and alumina, certain homogeneous mixtures of uranium dioxide and water can maintain a nuclear chain reaction for the uranium assay considered in this study. The critical radii for infinite cylinders of uranium dioxide and water mixtures were calculated assuming a 30 cm water reflector. Again, the uranium dioxide density was assumed to be 10.9 g/cc, while the water density was assumed to be 1.0 g/cc. The results are plotted in figure 2 and are presented in table A-I. (Tables numbered using the prefix "A" appear in the appendix.)

As shown in figure 2, the critical radius reaches a minimum of approximately 12.8 cm at about 20 volume percent uranium dioxide. In view of these small radii for uranium dioxide-water mixtures, it is essential that extreme caution be employed to exclude water from the fluid-bed reactor system. A 12.8 cm limitation on the cylinder radius for safe geometry equipment, corresponding to approximately a 10-inch-diameter limit, would be overly restrictive for the main reactor of a volatility plant where several whole fuel assemblies, each from 5 to 9 inches square, would be charged simultaneously. In addition, the large quantities of uranium dioxide that must be charged to the reactor, if throughput requirements are to be met economically, preclude uranium mass control as a method of guaranteeing nuclear safety. In the absence of good moderators, however, no nuclear safety problems due to uranium appear to exist for the main reactor and its associated filters, since, as noted previously, mixtures of uranium dioxide and alumina are inherently safe for the uranium assay considered.

URANIUM HEXAFLUORIDE-HYDROGEN FLUORIDE MIXTURES

Calculations were made for mixtures of uranium hexafluoride and hydrogen fluoride. The latter compound is an impurity which may be present with uranium hexafluoride in such equipment items as cold traps, purification stills, and hold tanks. Hydrogen fluoride is a good neutron moderator and, if present in substantial quantities, can significantly reduce the critical radii of uranium hexafluoride cylinders or spheres. Even though only small amounts of hydrogen fluoride are anticipated in the volatility plant, calculations were made for the full range of concentrations to establish minima in the critical dimensions.

The radii for infinite cylinders containing uranium hexafluoride and hydrogen fluoride were calculated for effective neutron multiplication factors of 0.9 and 1.0. Uranium hexafluoride and hydrogen fluoride densities of 5.0 and 1.0 g/cc, respectively, were used, and a 30 cm water reflector was assumed. Since it is improbable that this relatively high uranium hexafluoride density would be attained in any of the volatility process equipment, the calculations should be somewhat conservative. The effective neutron multiplication factor of 0.9 was chosen arbitrarily and does not necessarily represent a recommended design criteria. In general, such relatively high values of k_{eff} were chosen for this study since, as noted by Clark⁽²⁾, k_{eff} 's ranging from 0.90 to

DWG. NO. G-68-208

Figure 2
CRITICAL RADII FOR INFINITE CYLINDERS OF
URANIUM DIOXIDE AND WATER MIXTURES

0.95 may be acceptable for design purposes when the fissile material is located in a shielded area or when calculations are known to be conservative. Volatility plant process cells will be heavily shielded with concrete, and as mentioned above, the calculations themselves are also felt to be conservative.

As shown in figure 3 and table A-II, the minimum infinite cylinder radii for both k_{eff} values occur at a uranium hexafluoride concentration of about 30 volume percent; the radii are about 20.1 and 25.6 cm for k_{eff} 's of 0.9 and 1.0, respectively. Although it is extremely doubtful that the hydrogen fluoride concentration would ever be as high as 70% during processing, the above minimum dimensions may be applied for design purposes in the absence of experimental data showing more realistic concentration ranges.

DISODIUM OCTOFLUOROURANATE AND SODIUM BIFLUORIDE MIXTURES

Sodium fluoride sorption traps have been considered for several applications in the fluoride volatility process. For example, with the proper choice of operating conditions, certain fission products can be selectively removed from uranium hexafluoride streams, while allowing essentially all of the uranium to pass through the trap. Under other conditions, sodium fluoride can be employed to trap uranium hexafluoride as a sodium fluoride complex; the sorbed uranium can then be desorbed and transferred to product collection equipment. Criticality calculations have been completed for sodium fluoride-uranium hexafluoride systems in which hydrogen fluoride was also assumed to be present, again as an impurity. The calculations were made assuming the formation of sodium bifluoride ($HF \cdot NaF$) and disodium octafluorouranate ($UF_6 \cdot 2NaF$) complexes in the system. While other complexes of these components are possible, their formation is considered to be unlikely at the anticipated operating conditions of volatility plant trapping systems. As before, although the amount of hydrogen fluoride present should be small under normal circumstances, computations were extended to cover the full range of concentrations.

Two types of calculations were made for various mixtures of the above complexes. These were (a) calculation of neutron multiplication factors for infinite media with uranium-235 enrichments of both 2 and 5%, and (b) calculation of radii of infinite cylinders, reflected with 30 cm of water, corresponding to effective neutron multiplication factors of 0.9 and 1.0. The latter calculations were made only for a uranium-235 enrichment of 5%. Mixtures of the complexes were assumed to be homogeneous with no voids. The uranium density in the pure uranium hexafluoride complex was taken as 2.25 g/cc, corresponding to a disodium octafluorouranate density of 4.13 g/cc⁽¹²⁾, while the hydrogen density in the hydrogen fluoride complex was assumed to be 0.035 g/cc, based on a sodium bifluoride density of 2.16 g/cc⁽¹⁾.

Neutron multiplication factors for infinite media are shown in figure 4. Since the maximum calculated k_{∞} for the system with 2% enriched uranium

DWG. NO. G-68-209

Figure 3
RADII FOR INFINITE CYLINDERS OF
URANIUM HEXAFLUORIDE AND HYDROGEN FLUORIDE MIXTURES

Figure 4
INFINITE NEUTRON MULTIPLICATION FACTORS FOR
MIXTURES OF $\text{UF}_6 \cdot 2\text{NaF}$ AND $\text{HF} \cdot \text{NaF}$

is less than 1.0, a critical homogeneous assembly does not appear to be possible for the given system at that enrichment. The maximum k_{∞} for the system with 5% enriched uranium is approximately 1.24 and occurs at about 0.4 volume fraction disodiumoctofluorouranate.

Radii of infinite cylinders, corresponding to effective neutron multiplication factors (k_{eff}) of 0.9 and 1.0 for 5% uranium assays, are shown in figure 5 and are tabulated in table A-III. The minimum critical radius ($k_{\text{eff}} = 1.0$) is approximately 39.2 cm at a disodiumoctofluorouranate volume fraction of about 0.35. This radius corresponds to a vessel diameter of about 2.57 feet. The minimum radius for $k_{\text{eff}} = 0.9$ is about 29.4 cm at a uranium hexafluoride-sodium fluoride complex volume fraction of 0.4. This radius corresponds to a vessel diameter of about 1.9 feet.

PLUTONIUM CRITICALITY STUDIES

Criticality calculations were also made for plutonium dioxide and plutonium hexafluoride systems. As in the uranium studies, several conservative assumptions were usually applied to each calculation. Among these were that fissile material was reflected with 30 cm of either alumina or water, and that all plutonium was present as plutonium-239 at densities higher than those which normally would be expected.

In the criticality studies for high enrichment uranium systems, it was found that, for high assay uranium, alumina is actually a better neutron reflector than water for large reflector thicknesses. It was found in the present work that the same also applies to plutonium systems. (The critical masses for spheres of plutonium dioxide reflected with varying thicknesses of water and alumina are compared in figure 6.) A water reflector was chosen for the low enrichment uranium calculations because the large volumes of uranium present in the main fluid bed preclude the possibility of thick alumina reflection. This is not necessarily true for plutonium systems, however. Since the plutonium mass is relatively small, it may be possible (although seemingly improbable) for the plutonium to segregate in the center of a processing vessel and be reflected by a substantial quantity of alumina. Therefore, to be conservative, a 30 cm reflector of alumina was assumed for all situations where alumina was present. For cases where no alumina was present, a 30 cm water reflector was used, as before.

Included in the plutonium criticality studies were calculations for plutonium dioxide and plutonium hexafluoride cylinders and spheres, and for mixtures of plutonium dioxide and water, plutonium dioxide and alumina, plutonium hexafluoride and hydrogen fluoride, and plutonium tetrafluoride-sodium fluoride complex and sodium bifluoride. The results of some plutonium dioxide slab calculations are also included, and additionally, calculations for plutonium hexafluoride systems reflected with reinforced phenolic foam insulation are discussed. The results of each of these calculation sets are presented in the subsequent sections,

Figure 5

RADI FOR INFINITE CYLINDERS OF
 $UF_6 \cdot 2NaF$ AND HF · NaF MIXTURES

DWG. NO. G-68-212

Figure 6
CRITICAL MASS OF PuO_2 SPHERES FOR
VARYING THICKNESSES OF WATER AND ALUMINA REFLECTORS

following a brief discussion as to why the assumption that the plutonium is present as plutonium-239 was made.

POWER REACTOR PLUTONIUM

Plutonium-239 is produced in nuclear power reactors as a result of absorption of neutrons by uranium-238 in the fuel. The major portion of the other plutonium isotopes, such as plutonium-240, -241, and -242, are then formed from plutonium-239 by the capture of additional neutrons. Plutonium-239 and -241 are fissile nuclides, while plutonium-240 and -242 are essentially nonfissionable.

Criticality calculations were made for plutonium dioxide systems in which the plutonium isotopic composition corresponded approximately to that for fuel burned to 10,000, 20,000, and 30,000 Mwd/tonne. The three irradiation levels were chosen to cover the expected range at which current and near-future power reactors may operate. The plutonium isotopic compositions for these irradiation levels are listed in table IV and are based on calculations by Dawson, et al⁽³⁾, assuming that 30% of the neutron flux is in the resonance region. The 30% resonance flux value is typical of currently designed pressurized light water reactors.

Since the percentage of total fissile plutonium decreases as burnup increases, the mass required for a critical assembly should increase correspondingly. Calculations were made to confirm this and to provide quantitative estimates of the increase. In addition, calculations were made for 100% plutonium-239 for comparison purposes.

The critical masses of plutonium in plutonium dioxide spheres were calculated for the three irradiation levels above, as well as for the case in which all plutonium is the 239 isotope. A 30 cm reflector of alumina at a density of 2.5 g/cc was assumed. The results are plotted in figure 7, and the sphere radii corresponding to these masses are listed in tables A-IV and A-V. As shown in figure 7, the critical masses for plutonium-239 are about 10 to 20% lower than for the irradiated cases, indicating that it is conservative, as expected, to assume that all plutonium is plutonium-239. The critical mass for plutonium-239 is about 33 kg at a plutonium dioxide density of 4.5 g/cc, while the critical mass at the same density for material burned to the 10,000 to 30,000 Mwd/tonne level ranged from 37 to 39 kg, respectively. For this reason, it was decided to perform subsequent plutonium calculations assuming pure plutonium-239.

The limiting plutonium density that might be achieved in the fluoride volatility plant is not definitely known; however, the density of uranium in bulk uranium salt powders encountered in salvage operations usually does not exceed 3.2 grams of uranium per cubic centimeter unless the material has been pressed and/or sintered⁽²⁵⁾. The 3.2 g/cc value is similarly considered to be a good assumption for plutonium, since it is estimated that the crystal densities of the oxide salts of uranium and plutonium are roughly the same. Since operational data were not

TABLE IV

PLUTONIUM ISOTOPE COMPOSITIONS IN IRRADIATED FUEL

<u>Plutonium Isotope</u>	<u>Isotopic Composition at Indicated Irradiation Levels, atom percent*</u>		
	<u>10,000 Mwd/tonne of Uranium</u>	<u>20,000 Mwd/tonne of Uranium</u>	<u>30,000 Mwd/tonne of Uranium</u>
239	75	65	59
240	15	18	19
241	9	15	17
242	1	2	5
Total Fissile	84	80	76

* Plutonium isotopic distributions were taken from 30% resonance flux values reported by Dawson, F. G., et al, "Plutonium Buildup and Depletion", Nuclearonics, 23, No. 8, p. 101 (1965).

Figure 7
CALCULATED CRITICAL MASS OF SPHERES
CONTAINING PuO_2 POWDER

available, the above calculations were made for a plutonium dioxide density range of 1.0 to 4.5 g/cc, which corresponds to a plutonium density range of 0.88 to 4 g/cc. Bulk and tap densities for plutonium dioxide, prepared in several different ways, are presented in reference (14); the densities reported generally fall into the above range.

PLUTONIUM DIOXIDE CALCULATIONS

Plutonium Dioxide-Water Mixtures

The effects of small amounts of water on the critical mass of plutonium dioxide spheres were calculated assuming a plutonium density of 3.2 g/cc, a water density of 1.0 g/cc, and a 30 cm reflector of alumina at a density of 2.5 g/cc. For these calculations, it was assumed that the plutonium density did not change, i.e., that the total volume was constant. As shown in figure 8, the critical mass of plutonium decreases rapidly with increasing water concentrations. In fact, for a water concentration of 8% on a weight basis (31.6 volume percent), the critical mass is decreased by over 50%. This calculation serves to support the already-mentioned conclusion that water or other good neutron moderating material must be prevented from entering the main reactor vessel used in the fluoride volatility process.

Plutonium Dioxide Slabs

Critical thicknesses for infinite slabs of plutonium dioxide were calculated assuming a 30 cm reflector of alumina. The calculations were made for plutonium dioxide densities ranging from 1.0 to 4.5 g/cc. The critical slab thicknesses, shown in table V, were found to decrease from 11.26 to 2.50 cm for increasing plutonium dioxide densities. As expected, the product of slab thickness and plutonium dioxide density remained essentially constant. Since slabs of these small thicknesses would probably be unrealistic in a volatility plant handling present-day fuels, no further slab calculations were made.

Plutonium Dioxide Cylinders

Critical radii for infinite cylinders of plutonium dioxide have been calculated assuming a 30 cm reflector of alumina. The calculations were made for plutonium dioxide densities ranging from 1.0 to 4.5 g/cc. Plotted in figure 9, the results are also detailed in table A-VI.

In figure 9, the critical radius is seen to decrease rapidly with increasing plutonium dioxide density, especially at the low end of the density scale. The critical radius decreased from about 20 cm at a density of 1 g/cc to about 6.5 cm at a density of 4.5 g/cc; these radii correspond to vessel diameters of about 16 inches and 5 inches, respectively. It is interesting to note that vessel diameters as large as 26 inches are desirable for the main fluid-bed reactor in a nominal 1 tonne per day plant. Thus, based on the hypothesized conditions under which these calculations were performed, the primary reactor would not

DWG. NO. G-68-213

Figure 8
CRITICAL MASS FOR SPHERES OF
PLUTONIUM DIOXIDE AND WATER MIXTURES

TABLE V

CRITICAL THICKNESSES FOR INFINITE SLABS CONTAINING PLUTONIUM DIOXIDE

Plutonium Dioxide Density, g/cc	Critical Thickness*, cm
1.0	11.26
1.5	7.56
2.0	5.62
2.5	4.50
3.0	3.74
3.5	3.22
4.0	2.82
4.5	2.50

* A 30 cm alumina reflector at 2.5 g/cc was assumed. All plutonium was assumed to be plutonium-239.

DWG. NO. G-68-214

Figure 9
CRITICAL RADII FOR INFINITE
CYLINDERS OF PLUTONIUM DIOXIDE

be geometrically safe for plutonium, and plutonium mass control would be necessary to ensure nuclear safety in this vessel.

Plutonium Dioxide Spheres and Spherical Shells

Calculations were made to obtain critical plutonium masses for spherical shells of plutonium dioxide, both where the central region was void and where the central region contained alumina. A 30 cm outer alumina reflector was assumed for both cases. The purpose of these calculations was to show that the solid plutonium-239 dioxide sphere critical masses were greater than the masses for the hollow spheres and, thus, that the assumption of solid spheres should represent a conservative limit.

The results of the calculations are shown in table VI for a solid plutonium dioxide sphere and for spherical shells where the inside shell radius varied from 2 to 10 cm. The plutonium density in plutonium dioxide was assumed to be 3.2 g/cc, while the alumina density was assumed to be 2.5 g/cc. As can be seen from the table, the critical masses for the void and alumina center cases were about equal, and were larger than for the solid plutonium dioxide sphere.

TABLE VI

CRITICAL MASSES FOR PLUTONIUM DIOXIDE SPHERES AND SHELLS

Inside Shell Radius, cm	Outside Shell Critical Radius, cm		Critical Mass*, kg plutonium	
	Void Center	Alumina Center	Void Center	Alumina Center
Solid Sphere	14.68	14.68	42.40	42.40
2	14.75	14.74	42.88	42.85
5	15.18	15.18	45.26	45.19
10	17.31	17.26	56.12	55.52

* A 30 cm alumina reflector at a density of 2.5 g/cc was assumed; the plutonium density in plutonium dioxide was 3.2 g/cc; all plutonium was plutonium-239.

Calculations were also made to estimate plutonium masses corresponding to effective neutron multiplication factors ranging from 0.8 to 1.0 for solid spheres reflected with 30 cm of alumina at 2.5 g/cc. The plutonium density was taken as 3.2 g/cc. As shown in figure 10, the plutonium mass ranges from about 22 to 42 kg for effective neutron multiplication factors ranging from 0.80 to 1.0. Thus, the allowable plutonium mass in the main reactor of a volatility plant will be highly dependent on

DWG. NO. G-68-215

Figure 10

PLUTONIUM MASS IN PLUTONIUM DIOXIDE SPHERES
VERSUS EFFECTIVE NEUTRON MULTIPLICATION FACTORS

the particular value of k_{eff} chosen as a reasonable design guide for nuclear safety, as well as on the assumed conditions of density, reflection, etc, already mentioned. Again, it should be pointed out that it was assumed all plutonium was present as plutonium-239, which, as shown previously, is a conservative assumption.

Plutonium Dioxide-Alumina Mixtures

The plutonium calculations which were discussed in the preceding sections considered spherical or cylindrical geometries of relatively concentrated plutonium reflected by 30 cm of alumina. This implies the possibility that plutonium dioxide may segregate to form these geometries in the process. A more realistic situation in volatility processing might well involve plutonium dioxide mixed, for the most part, homogeneously with alumina; this mixture could then be reflected by thick layers of alumina.

Criticality calculations were performed for various homogeneous plutonium dioxide-alumina mixtures for a plutonium density of 3.2 g/cc and an alumina density of 2.5 g/cc. A 30 cm alumina reflector was again used.

Plutonium Dioxide-Alumina Cylinders. Critical radii for infinite cylinders of plutonium dioxide-alumina mixtures were calculated first; the results are presented in figure 11 and in table A-VII. The critical radius increases rapidly with decreasing plutonium dioxide concentration, ranging from about 8 to 30 cm for plutonium dioxide concentrations between 100 and 10%, respectively. It is interesting to note that, at 10 volume percent plutonium dioxide, the weight ratio of alumina to plutonium is about 7.0. In the main fluid-bed reactor of a fluoride volatility plant handling LWR fuels, the actual alumina-to-plutonium weight ratio is expected to be much greater than this; i.e., on the order of 200. Thus, if the plutonium dioxide mixes homogeneously with alumina, the critical radius of infinite cylinders of these mixtures will be greater than 30 cm, and the vessel, depending on its radius, may be judged to be geometrically safe. Of course, if the possibility of partial or complete plutonium dioxide segregation is allowed, the results of the earlier calculations for pure plutonium dioxide must be considered.

Plutonium Dioxide-Alumina Spheres. The critical masses for spheres containing homogeneous mixtures of plutonium dioxide and alumina, reflected with 30 cm of alumina, were also estimated. The densities were assumed to be the same as those used in the cylinder calculations. As can be seen from figure 12 and table A-VIII, the critical mass of plutonium increases rapidly with decreasing concentrations of plutonium dioxide, ranging from about 42 to 114 kg for plutonium dioxide concentrations ranging from 100 to 20%. Thus, considerable relaxation of the plutonium mass limit may also be possible if it can be shown experimentally that plutonium dioxide will mix homogeneously with alumina during processing.

Figure 11
CRITICAL RADII FOR INFINITE CYLINDERS OF
PLUTONIUM DIOXIDE AND ALUMINA MIXTURES

Figure 12
CRITICAL MASSES FOR SPHERES OF
PLUTONIUM DIOXIDE AND ALUMINA MIXTURES

PLUTONIUM HEXAFLUORIDE CALCULATIONS

After the plutonium fluorination step in the volatility process, plutonium hexafluoride will be the major compound of plutonium present. Accordingly, criticality calculations have been made for this compound in various geometries and at various densities. Emphasis was placed on calculations pertaining to areas outside of the main fluid-bed reactor. For this reason, reflection by water rather than by alumina was assumed.

Plutonium Hexafluoride Cylinders

Critical radii for infinite cylinders of plutonium hexafluoride at various densities were calculated first, using a 30 cm water reflector. The results are plotted in figure 13 and are tabulated in table A-IX.

The critical radius decreases rapidly with increasing plutonium hexafluoride density; at a density of 1 g/cc, the radius is about 33 cm, while at a density of 6 g/cc, the radius is near 7 cm. Thus, as was the case for plutonium dioxide, the critical radius of a process vessel will be highly dependent on the particular plutonium hexafluoride density achieved. While no data on the density of plutonium hexafluoride are available, it is reasonable to expect a density analogous to that of uranium hexafluoride; the theoretical density of solid uranium hexafluoride is 4.68 g/cc at a temperature of 21°C⁽⁶⁾.

Plutonium Hexafluoride Spheres

Critical masses of plutonium hexafluoride spheres were calculated for various densities and for a 30 cm water reflector. The results are plotted in figure 14 and are listed in table A-X. The critical mass decreases quite rapidly with increasing plutonium hexafluoride densities, varying from about 413 kg plutonium at 1 g/cc to 26 kg plutonium at 6 g/cc.

Plutonium Hexafluoride-Hydrogen Fluoride Mixtures

Calculations were also made for homogeneous mixtures of plutonium hexafluoride and hydrogen fluoride. As discussed in the uranium criticality sections, hydrogen fluoride may be present in the fluorine as an impurity, although its concentration should be relatively low.

Plutonium Hexafluoride-Hydrogen Fluoride Cylinders. Critical radii for infinitely long cylinders containing plutonium hexafluoride-hydrogen fluoride mixtures were calculated for hydrogen-to-plutonium-239 ratios varying from 0 to about 1,000. It was assumed that the cylinders were reflected with 30 cm of water and that the plutonium hexafluoride and hydrogen fluoride densities were 6.0 and 1.749 g/cc, respectively. These rather high densities were chosen for conservative coverage of operating conditions. In at least one situation where cold traps might be used, the above mixtures may be present at very low temperatures; i.e., down to minus 180°C. Therefore, the plutonium hexafluoride density was taken as being comparable to that of uranium hexafluoride; the solid density

Figure 13
CRITICAL RADII FOR INFINITE CYLINDERS
OF PLUTONIUM HEXAFLUORIDE

Figure 14
CRITICAL MASSES OF
PLUTONIUM HEXAFLUORIDE SPHERES

of uranium hexafluoride, in turn, was estimated by linearly extrapolating values given at two higher temperatures in the Reactor Handbook⁽²⁶⁾. The hydrogen fluoride density used was the value reported by Ryss⁽²³⁾ at minus 191°C. The results of the calculations are shown in table VII.

TABLE VII

CRITICAL RADII FOR INFINITE CYLINDERS
CONTAINING HOMOGENEOUS PLUTONIUM HEXAFLUORIDE-HYDROGEN FLUORIDE MIXTURES

Plutonium Hexafluoride, volume percent	H/Pu Ratio	Critical Radius*, cm
0.50	1,024	12.69
1.00	509	10.06
1.25	406	9.56
1.50	338	9.24
2.00	252	8.86
2.50	201	8.62
3.00	166	8.48
5.00	98	8.15
10.00	46	7.72
100.00	0	6.87

* Reflected with 30 cm of water. The plutonium hexafluoride and hydrogen fluoride densities were assumed to be 6.0 and 1.749 g/cc, respectively. All plutonium was assumed to be plutonium-239.

The minimum critical radius of an infinite cylinder of plutonium hexafluoride and hydrogen fluoride occurs at a plutonium hexafluoride concentration of 100%. The radius at this concentration is 6.87 cm, which corresponds to a cylinder diameter of about 5.4 inches.

It should be pointed out that if a nuclear chain reaction did occur in a low-temperature vessel, an immediate result would be an increase in temperature due to the heat evolved in the reaction. Since fissile and moderator material densities would be expected to decrease with increasing temperature, it is likely that the temperature coefficient of reactivity would be negative. Thus, the chain reaction would tend to

be self-suppressing. Of course, considerable vessel damage might occur before the reaction was brought under control.

Plutonium Hexafluoride-Hydrogen Fluoride Spheres. Critical sphere masses for homogeneous mixtures of plutonium hexafluoride and hydrogen fluoride were also calculated, assuming the same densities and reflection as in the cylinder calculations. The results are shown in table VIII; as can be seen, the critical mass of plutonium reaches a minimum of about 0.55 kg plutonium at a plutonium hexafluoride concentration of about 0.5 volume percent.

TABLE VIII
CRITICAL SPHERE MASSES
FOR HOMOGENEOUS PLUTONIUM HEXAFLUORIDE-HYDROGEN FLUORIDE MIXTURES

Plutonium Hexafluoride, volume percent	H/Pu-239	Critical Radius, cm	Critical Mass, kg Pu-239*
0.2	2,567	40.04	2.19
0.5	1,024	18.58	0.55
1	509	15.32	0.61
2	252	13.81	0.90
3	166	13.39	1.23
5	98	13.02	1.88
10	46	12.84	3.60
100	0	11.49	25.81

* The spheres were reflected with 30 cm of water. The plutonium hexafluoride and hydrogen fluoride densities were 6.0 and 1.749 g/cc, respectively. All plutonium was assumed to be plutonium-239.

Plutonium Hexafluoride with Reinforced Phenolic Foam Reflectors

Several of the volatility process vessels in which plutonium hexafluoride is handled will require insulation. This insulation will act as a neutron reflector and will reduce the critical radius to a certain extent, depending on the nuclear properties of the insulation. This effect can be minimized by choosing a lagging material that is a good neutron absorber.

One insulation material, a reinforced phenolic foam, adapted by the ORGDP for use with uranium hexafluoride containers⁽¹¹⁾, was evaluated for possible use on vessels in which plutonium hexafluoride will be present. The approximate composition of this material is shown in table IX⁽¹⁸⁾.

TABLE IX
COMPOSITION OF REINFORCED PHENOLIC FOAM INSULATION

Element	Weight Percent
Carbon	57.02
Hydrogen	4.80
Boron	3.08
Chlorine	3.92
Oxygen, Silicon, Fluorine	31.18

Critical radii of infinitely long cylinders of plutonium hexafluoride at a density of 6 g/cc and reflected with varying thicknesses of reinforced phenolic foam were calculated assuming a phenolic foam density of 0.112 g/cc. The results are plotted in figure 15. The calculated decrease in critical radius for increasing reflector thickness was quite small, which indicates that the insulating material has good neutron absorption properties. The critical radius varied from 11.50 to 10.48 cm for reflector thicknesses between 0 and 100 cm. At a thickness of 10 cm, which is probably near the insulation thickness that will be required in the volatility process, the critical radius was about 11.12 cm.

Effective neutron multiplication factors for various radii of infinite cylinders reflected with 10 cm of the foam insulation have also been calculated, again assuming that the densities of plutonium hexafluoride and the foam were 6.0 and 0.112 g/cc, respectively. As can be seen from the results of these calculations, plotted in figure 16, the relationship between the effective multiplication factors and the cylinder radii is approximately linear for the range of k_{eff} 's considered.

Effective neutron multiplication factors were next calculated for infinitely long cylinders with dimensions corresponding to the actual sizes of standard schedule-40 pipe. Three k_{eff} calculations were made for each cylinder. These were for (a) bare cylinders, (b) cylinders reflected with 100 cm of reinforced phenolic foam at a density of 0.096 g/cc, and (c) cylinders reflected with the pipe wall thickness of metal (nickel) plus 10 cm reinforced phenolic foam at a density of

Figure 15

CRITICAL RADII FOR INFINITE CYLINDERS OF PLUTONIUM
HEXAFLUORIDE VERSUS PHENOLIC FOAM REFLECTOR THICKNESS

DWG. NO. G-68-221

Figure 16

EFFECTIVE NEUTRON MULTIPLICATION FACTORS FOR
INFINITE CYLINDERS OF PLUTONIUM HEXAFLUORIDE

0.096 g/cc plus 30 cm of water. In these calculations, the plutonium hexafluoride density was assumed to be 5 g/cc; the results are given in table X.

TABLE X
EFFECTIVE NEUTRON MULTIPLICATION FACTORS
FOR INFINITE CYLINDERS OF PLUTONIUM HEXAFLUORIDE

Nominal Pipe Diameter, inch	Inside Diameter, inch	Wall Thickness, inch	Effective Multiplication Factor*		
			Bare	Reflected†	Reflected§
4	4.026	0.237	0.350	0.396	0.434
6	6.065	0.280	0.545	0.612	0.664
8	7.981	0.322	0.730	0.811	0.871
10	10.020	0.365	0.922	-	-

* Plutonium hexafluoride density = 5.0 g/cc.

† Reflected with 100 cm reinforced phenolic foam at 0.096 g/cc.

§ Reflector consisted of the wall thickness of metal (assumed to be nickel) plus 10 cm reinforced phenolic foam at 0.096 g/cc plus 30 cm water.

As shown in table X, for nominal pipe diameters up to 8 inches, the cylinders were subcritical even for the reflected cases. The bare 10-inch cylinder was also subcritical; calculations were not made for the reflected 10-inch cylinders, but it is likely that the k_{eff} 's for these cases would have been greater than 1.0.

Effective neutron multiplication factors were then calculated for plutonium hexafluoride contained in two infinitely long concentric nickel cylinders. Again, all dimensions were taken as those corresponding to standard schedule-40 pipe. Plutonium hexafluoride was assumed to be in both the center cylinder and the annular region. The outer cylinder was reflected with the wall thickness of metal plus 10 cm of reinforced phenolic foam, while the inner cylinder was reflected only by its wall. The densities of plutonium hexafluoride and reinforced phenolic foam were assumed to be 5.0 and 0.096 g/cc, respectively. The results of these calculations are shown in table XI.

TABLE XI

EFFECTIVE NEUTRON MULTIPLICATION FACTORS
FOR PLUTONIUM HEXAFLUORIDE CONTAINED IN ANNULAR CYLINDERS

Nominal Cylinder Diameter†, inches		k_{eff}^*
Inner Cylinder	Outer Cylinder	
4	8	0.764
6	10	0.968
8	12	1.148

* Outer cylinder reflected by 10 cm reinforced phenolic foam at a density of 0.096 g/cc. The plutonium hexafluoride density was 5.0 g/cc.

† Dimensions were taken for standard schedule-40 pipe. The pipe wall thicknesses were taken into account in the calculations.

PLUTONIUM TETRAFLUORIDE-SODIUM FLUORIDE COMPLEX
AND SODIUM BIFLUORIDE SYSTEMS

Although a definite scheme for recovering plutonium in the volatility process has not been established, there is a possibility that sorption of plutonium hexafluoride on sodium fluoride will be used. Accordingly, criticality calculations were made to evaluate the nuclear safety aspects of this system.

It was assumed for this work that plutonium hexafluoride complexes with sodium fluoride to form plutonium tetrafluoride-sodium fluoride ($\text{PuF}_4 \cdot \text{NaF}$). Other complexes may be formed with higher ratios of sodium fluoride to plutonium tetrafluoride, with the particular ratio being temperature sensitive. The former compound was chosen, however, because it should be the most conservative case from a criticality standpoint, i.e., complexes with higher sodium fluoride-to-plutonium tetrafluoride ratios would be less reactive and, thus, have larger critical dimensions. For these calculations, it was assumed that the plutonium density in the complex was 4.33 g/cc. This number was computed by assuming that the plutonium density was the same as in the plutonium trifluoride-sodium fluoride complex. The theoretical density of the latter compound is 6.13 g/cc (1). As with the uranium complexes, it was assumed that the impurity, hydrogen fluoride, was present as sodium bifluoride at 2.16 g/cc.

Cylinder Calculations

The critical radii for infinite cylinders of various plutonium tetrafluoride-sodium fluoride complex and sodium bifluoride mixtures were computed assuming a 30 cm water reflector. As shown in figure 17 and in table A-XI, the minimum critical radius occurs at a plutonium tetrafluoride-sodium fluoride concentration of 100% and is 6.67 cm.

Slab Calculations

Since a cylinder radius of 6.67 cm might be too restrictive considering throughput requirements, the critical thickness of an infinite slab of plutonium tetrafluoride-sodium fluoride complex was computed to evaluate the possibility of using annular cylinder configurations for the plutonium sorbent traps. The critical thickness of the annulus would approach that of the slab, at least for large cylinder radii. It was assumed for this calculation that the slab was reflected with 30 cm of water and that the plutonium tetrafluoride-sodium fluoride density was the same as before. Because these calculations were preliminary, no hydrogen fluoride-sodium fluoride complex was considered.

The calculations showed that the critical thickness of the infinite slab was 3.98 cm, corresponding to 1.57 inches. It was concluded, therefore, that, under the assumed conditions, neutron absorbing materials would probably be necessary to ensure nuclear safety for those plutonium tetrafluoride-sodium fluoride annular cylinder configurations which would be practical from a process throughput standpoint. No further calculations were made because it was felt that they were not justified until a more definite plutonium purification scheme is established.

CONCLUSIONS

Criticality calculations have shown that, in the absence of water or other good neutron moderating materials, homogeneous mixtures of uranium dioxide (5% enriched) and alumina cannot maintain a neutron chain reaction. Thus, it was concluded that there will be no nuclear safety problems due to uranium in the main fluid-bed reactor portion of a volatility plant, since process conditions and controls should be such that elimination of water is ensured. No other good neutron moderating materials will be present. The same conclusion applies to other areas of the plant, such as in the main reactor filter section, where uranium will again be present in the absence of good neutron moderators.

If the possibility of plutonium dioxide segregation in the main fluid-bed reactor is allowed, mass control for plutonium will be necessary to ensure nuclear safety. The particular limiting mass chosen will depend on several factors, particularly administrative decisions on the proper density and degree of segregation that should be considered. If it is

DWG. NO. G-68-222

Figure 17

CRITICAL RADII FOR INFINITE CYLINDERS OF
 $\text{PuF}_4 \cdot \text{NaF}$ AND $\text{HF} \cdot \text{NaF}$ MIXTURES

not considered possible for plutonium dioxide to segregate in the main reactor, mass control for this compound may not be necessary, depending on the particular vessel diameters and densities assumed.

These criticality calculations have shown that it should be possible to design the remaining equipment in a volatility plant to be geometrically safe for uranium and plutonium, even though hydrogen fluoride, a good neutron moderator, may be present. It may be necessary, however, to employ neutron absorbers in certain instances, but their use in aqueous systems is a well-established practice and should present no particular design problems in a volatility plant.

In several instances, criticality calculations were made to point out the relationship between critical dimensions and the effective neutron multiplication factor, k_{eff} . These calculations were made to give the designer a feel for the degree of safety built into designs where vessel sizes or fissile material masses are chosen to be smaller than critical. Of course, the k_{eff} 's in this study were selected arbitrarily and do not necessarily represent recommended design criteria.

In conclusion, the nuclear safety advantages of the volatility process might be very substantial with respect to batch and equipment sizes, providing excessive segregation of plutonium can be ruled out as it has been in some aqueous processes for fissile materials. Furthermore, in choosing nuclear safety guides for a volatility plant, the fact that all of the equipment handling fissile material will be remotely operated in cells heavily shielded with concrete should be kept in mind. Thus, because of this contingency, it might be possible to apply less restrictive safety factors to volatility plant equipment design than would be possible in other instances.

ACKNOWLEDGEMENT

The authors wish to express their appreciation to Dr. J. R. Knight of Union Carbide Corporation, Nuclear Division, Computer Technology Center who helped develop the local versions of the computer codes used for the calculations and provided considerable assistance in program utilization. The assistance of Drs. A. D. Callihan and D. W. Magnuson of the Oak Ridge National Laboratory and Messrs. J. R. Merriman, J. H. Pashley, and R. B. Schappel of the ORGDP Process Systems Development Department in reviewing the report is also gratefully acknowledged.

BIBLIOGRAPHY

1. Brunton, G. D., Insley, H., McVay, T. N., and Thoma, R. E., Crystallographic data for Some Metal Fluorides, Chlorides, and Oxides, Union Carbide Corporation, Nuclear Division, Oak Ridge National Laboratory, February, 1965 (ORNL-3761).
2. Clark, H. K., Handbook of Nuclear Safety, E. I. du Pont de Nemours and Company, Inc., Savannah River Laboratory, January, 1961 (DP-532).
3. Dawson, F. G., et al, "Plutonium Buildup and Depletion", Nucleonics, 23, No. 8 (1965).
4. Engle, W. W., Jr., Boling, M. A., and Colston, B. W., DTF-II, A One-Dimensional Multigroup Neutron Transport Program, North American Aviation, March 25, 1966 (NAA-SR-10951).
5. Engle, W. W., Jr., A Users Manual for ANISN, A One-Dimensional Discrete Ordinates Transport Code with Anisotropic Scattering, Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, March 30, 1967 (K-1693).
6. Handbook of Chemistry and Physics, 45th Edition, The Chemical Rubber Company, Cleveland, Ohio (1964).
7. Hanson, G. E., and Roach, W. H., Six and Sixteen Group Cross Sections for Fast and Intermediate Critical Assemblies, Los Alamos Scientific Laboratory, 1961 (LAMS-2543).
8. Hydrated, Calcined, and Tabular Aluminas, Alumina Company of America, March 15, 1956 (Product Data Section GA2A).
9. Jarvis, G. A. Linenberger, G. A., Orndoff, J. D., and Paxton, H. C., "Two Plutonium-Metal Critical Assemblies", Nucl. Sci. Eng., 8, 525-531 (1960).
10. Joanou, J. D., and Dudek, J. S., GAM-II, A B₂ Code for the Calculation of Fast Neutron Spectra and Associated Multigroup Constants, General Atomic, 1963 (GA-4265).
11. Mallett, A. J., and Wheatley, S. J., Fissile Material Container and Packaging Development and Testing Program, Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, April 1, 1966 (K-1661).
12. McNeese, L. E., An Experimental Study of Sorption of Uranium Hexafluoride by Sodium Fluoride Pellets and a Mathematical Analysis of Diffusion with Simultaneous Reaction, Union Carbide Corporation, Nuclear Division, Oak Ridge National Laboratory, November 29, 1963 (ORNL-3494).

13. Mihalcz, J. T., and Neeley, V. I., "~~The~~ Infinite Neutron Multiplication Constant of Homogeneous Hydrogen-Moderated 2.0 Weight Percent Uranium-235 Enriched Uranium", Nucl. Sci. Eng., 13:6-11 (1962).
14. Moseley, J. D., and Wing, R. O., Properties of Plutonium Dioxide, Dow Chemical Company, Rocky Flats Division, August 24, 1965 (RFP-503).
15. Neeley, V. I., and Handler, H. E., Measurement of Multiplication Constant for Slightly Enriched Homogeneous Uranium Trioxide-Water Mixtures and Minimum Enrichment for Criticality, General Electric, Hanford Atomic Products Operation, August 21, 1961 (HW-70310)..
16. Neeley, V. I., Berberet, J. A., and Masterson, R. H., K_∞ of Three Weight Percent U-235 Enriched Uranium Trioxide and UO₂(NO₃)₂ Hydrogenous Systems, General Electric, Hanford Atomic Products Operation, September, 1961 (HW-66882).
17. Newlon, C. E., and Mallett, A. J., Hydrogen Moderation - A Primary Nuclear Safety Control for Handling and Transporting Low-Enrichment Uranium Hexafluoride, Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, May 31, 1966 (K-1663).
18. Newlon, C. E., Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, Personal Communication.
19. Physics Research Quarterly Report, July, August, September, 1962, General Electric, Hanford Atomic Products Operation, October 15, 1962 (HW-75228).
20. Physics Research Quarterly Report, October, November, December, 1962, General Electric, Hanford Atomic Products Operation, January 15, 1963 (HW-76128).
21. Physics Research Quarterly Report, January, February, March, 1963, General Electric, Hanford Atomic Products Operation, April 15, 1963 (HW-77311).
22. Richey, C. R., White, J. D., Clayton, E. D., and Lloyd, R. C., "Criticality of Homogeneous Plutonium Oxide-Plastic Compacts at H:Pu = 15", Nucl. Sci. Eng., 23, 150-158 (1965).
23. Ryss, I. G., The Chemistry of Fluorine and Its Inorganic Compounds, A U. S. Atomic Energy Commission translation from a publication of the State Publishing House for Scientific, Technical, and Chemical Literature, Moscow (1956) (AEC-tr-3927, pt. 1).
24. Smiley, S. H., Brater, D. C., and Pashley, J. H., ORGP Fuel Reprocessing Studies, Summary Progress Report, Fiscal Year 1964 through Fiscal Year 1965, Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, October 29, 1965 (K-1649).

25. Smiley, S. H., Pashley, J. H., and Schappel, R. B., ORGDP Fuel Reprocessing Studies Summary Progress Report, January through June, 1966, Union Carbide Corporation, Nuclear Division, Oak Ridge Gaseous Diffusion Plant, January 18, 1967 (K-1691).
26. Stoller, S. M., and Richards, R. B., eds, Reactor Handbook, Volume II, Fuel Reprocessing, Interscience Publishers, Inc., New York (1961).

APPENDIX

The following tables present computed results for many of the cases discussed in the text.

TABLE A-I

CRITICAL RADII FOR INFINITE CYLINDERS
CONTAINING HOMOGENEOUS URANIUM DIOXIDE-WATER MIXTURES

<u>Uranium Dioxide, volume percent</u>	<u>H/U-235 Ratio</u>	<u>Critical Radius*, cm</u>
5	1,045	25.3
10	495	14.6
20	220	12.8
30	128	13.1
40	82	14.2
60	37	19.3

* A 30 cm water reflector was assumed. The uranium dioxide and water densities were assumed to be 10.9 and 1.0 g/cc, respectively. The uranium-235 assay was 5%.

TABLE A-II

RADII CORRESPONDING TO k_{eff} 's OF 0.9 AND 1.0
FOR INFINITE CYLINDERS
CONTAINING URANIUM HEXAFLUORIDE AND HYDROGEN FLUORIDE

Uranium Hexafluoride, volume percent	H/U-235 Ratio	Cylinder Radius*, cm	
		$k_{eff} = 0.9$	$k_{eff} = 1.0$
10	633	24.8	31.4
20	281	20.6	26.1
30	164	20.1	25.6
40	105	21.2	27.2
50	70	23.4	30.5

* A 30 cm water reflector was assumed, and the densities of uranium hexafluoride and hydrogen fluoride were assumed to be 5.0 and 1.0 g/cc, respectively. The uranium-235 assay was 5%.

TABLE A-III

RADII FOR INFINITE CYLINDERS
CONTAINING $UF_6 \cdot 2NaF$ - $HF \cdot NaF$ MIXTURES
FOR k_{eff} 's OF 0.9 AND 1.0

$UF_6 \cdot 2NaF$, volume fraction	H/U-235 Ratio	Cylinder Radius*, cm	
		$k_{eff} = 0.9$	$k_{eff} = 1.0$
0.20	294	34.2	ca 50
0.30	172	29.9	40.9
0.35	138	29.8	39.2
0.40	110	29.4	39.9
0.50	73.5	31.9	42.6

* A 30 cm water reflector was assumed, and the densities of the $UF_6 \cdot 2NaF$ and $HF \cdot NaF$ complexes were assumed to be 4.13 and 2.16 g/cc, respectively. The uranium-235 assay was 5%.

TABLE A-IV

CRITICAL RADII FOR PLUTONIUM DIOXIDE SPHERES

Plutonium Dioxide Powder Density, g/cc	Critical Radii for the Plutonium Isotope Composition at Indicated Irradiation Levels *, cm		
	10,000 Mwd/tonne	20,000 Mwd/tonne	30,000 Mwd/tonne
4.5	13.1	13.1	13.3
4.0	14.2	14.3	14.5
3.5	15.7	15.7	15.9
3.0	17.5	17.6	17.8
2.5	19.9	20.0	20.2
2.0	23.3	23.4	23.7
1.5	28.5	28.6	29.0
1.0	38.0	38.1	38.8

* A 30 cm alumina reflector at 2.5 g/cc was assumed.

TABLE A-V

CRITICAL MASSES FOR PLUTONIUM DIOXIDE SPHERES

Plutonium Dioxide Density, g/cc	Critical Radius*, cm	Critical Mass of Plutonium, kg
4.5	12.57	33.02
4.0	13.67	37.75
3.5	15.03	43.90
3.0	16.75	52.08
2.5	19.03	63.65
2.0	22.24	81.27
1.5	27.11	110.41
1.0	36.09	173.65

* A 30 cm alumina reflector at 2.5 g/cc was assumed. All plutonium was assumed to be plutonium-239.

TABLE A-VI
CRITICAL RADII FOR INFINITE CYLINDERS
OF PLUTONIUM DIOXIDE

Plutonium Dioxide Density, g/cc	Critical Radius*, cm
4.5	6.69
4.0	7.29
3.5	8.04
3.0	9.00
2.5	10.29
2.0	12.12
1.5	14.98
1.0	20.29

* A 30 cm reflector of alumina at a density of 2.5 g/cc was assumed.
All plutonium was assumed to be plutonium-239.

TABLE A-VII
CRITICAL RADII FOR INFINITE CYLINDERS
CONTAINING HOMOGENEOUS PLUTONIUM DIOXIDE-ALUMINA MIXTURES

Plutonium Dioxide, volume percent	Weight Ratio, alumina/plutonium	Critical Radius*, cm
10	7.0	30.08
20	3.1	21.10
40	1.2	14.20
60	0.5	10.93
80	0.2	9.11
100	0.0	7.85

* Reflected with 30 cm of alumina. The plutonium and alumina densities were assumed to be 3.2 and 2.5 g/cc, respectively. All plutonium was assumed to be plutonium-239.

TABLE A-VIII
CRITICAL MASSES OF PLUTONIUM DIOXIDE-ALUMINA SPHERES

Plutonium Dioxide, volume percent	Weight Ratio, alumina/plutonium	Critical Radius, cm	Critical Mass of Plutonium*, kg
20	3.1	34.87	113.66
40	1.2	24.26	76.55
60	0.5	19.49	59.54
80	0.2	16.61	49.14
100	0.0	14.68	42.40

* Reflected with 30 cm of alumina. The plutonium and alumina densities were assumed to be 3.2 and 2.5 g/cc, respectively. All plutonium was assumed to be plutonium-239.

TABLE A-IX
CRITICAL RADII FOR INFINITE CYLINDERS OF PLUTONIUM HEXAFLUORIDE

Plutonium Hexafluoride Density, g/cc	Critical Radius*, cm
1	32.71
2	17.79
3	12.54
4	9.81
5	8.12
6	6.87

* Reflected with 30 cm of water. All plutonium was assumed to be plutonium-239.

TABLE A-X
CRITICAL MASSES OF PLUTONIUM HEXAFLUORIDE SPHERES

Plutonium Hexafluoride Density, g/cc	Sphere Radius, cm	Critical Mass of Plutonium*, kg
1	52.58	412.26
2	28.86	136.34
3	20.51	73.41
4	16.12	47.52
5	13.37	33.89
6	11.49	25.81

* Reflected with 30 cm of water. All plutonium was assumed to be plutonium-239.

TABLE A-XI
CRITICAL RADII FOR INFINITE CYLINDERS
OF PLUTONIUM TETRAFLUORIDE-SODIUM FLUORIDE AND SODIUM BIFLUORIDE

PuF ₄ ·NaF, volume percent	Critical Radius*, cm
100	6.67
80	7.40
60	8.27
40	9.37
20	11.46

* A 30 cm water reflector was assumed, and the densities of plutonium tetrafluoride-sodium fluoride and sodium bifluoride were assumed to be 6.47 and 2.16 g/cc, respectively. All plutonium was plutonium-239.