ANL/CSE-13/28

Front-End Processes for Conversion of Current HEU-Based Alkaline Processes to LEU Foil Targets: Volumes and Compositions of All Waste, Product, and Off-Gas Streams from Both Front-End Options

Chemical Science and Engineering Division

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Front-End Processes for Conversion of Current HEU-Based Alkaline Processes to LEU Foil Targets: Volumes and Compositions of All Waste, Product, and Off-Gas Streams from Both Front-End Options

by
James L. Jerden Jr., Dominique C. Stepinski, Artem Gelis, and George F. Vandegrift
Chemical Science and Engineering Division, Argonne National Laboratory

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CONTENTS

1 INTRODUCTION 1

2 COMPOSITION OF IRRADIATED LEU-FOIL 4

2.1 Actinides 4
2.2 Fission Products 5
2.3 Noble Fission Gases 16
2.4 Decay Heat 19

3 NITRIC-ACID-DISSOLUTION FRONT-END OPTION 22

3.1 Dissolver System 22
3.2 Molybdenum Recovery Using Titania Columns 24
3.3 Waste Streams Generated 25
3.4 Waste Stream 1: Activated Aluminum Cladding Waste Stream 26
3.5 Waste Stream 2: Spent Acid Gas And Nitrogen Oxide Off-Gas Trap 28
3.6 Waste Stream 3: Spent Iodine Trap Material And Fission Gas Stream 29
3.7 Waste Stream of Titania Column 33

4 ELECTROCHEMICAL LEU DISSOLUTION IN BICARBONATE 36

5 ANNULAR LEU-FOIL TARGET QUALIFICATION EFFORT 42

6 REFERENCES 44

FIGURES

1-1 Schematic of the Argonne Annular LEU-Foil Target 1

2-1 Mass Percent of Fission Products after 12 hr of Cooling for 200 g of LEU after 200-hr Irradiation 9

2-2 Mass Percent of Fission Products after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation 9

2-3 Percent of Fission Products after 12 hr of Cooling for 200 g of LEU after 200-hr Irradiation 15

2-4 Percent of Fission Products after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation 15

FIGURES (Cont.)

2-5 Mole Percent of Noble Fission Gases after 12 hr of Cooling for 200 g of LEU after 200-hour Irradiation 17

2-6 Mole Percent of Noble Fission Gases after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation 17

2-7 Percent of Noble Fission Gases after 12 hr of Cooling for 200 g of LEU after 200-hr Irradiation 18

2-8 Percent of Noble Fission Gases after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation 18

2-9 Cumulative Decay Heat for Fission Products and Actinides for 200 g of LEU after 200-hr Irradiation 21

3-1 Conceptual Rendering of the Nitric-Acid Dissolver System 23

3-2 Conceptual Rendering of the Nitric-Acid Dissolver System Showing the Steps Involved in Starting the Dissolution and Gas Flows during Operation 24

3-3 Processes and Waste Streams for the Nitric-Acid-Based Front-End Process 25

3-4 Mole Percent of Fission Gases after 12 hr of Cooling for 200 g of LEU after 200-hr Irradiation 31

3-5 Mole Percent of Fission Gases after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation 31

3-6 Percent of Fission Gases after 12 hr of Cooling for 200 g of LEU after 200-hr Irradiation 32

3-7 Percent of Fission Gases after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation 32

4-1 Product and Waste Streams for the Electrochemical Front-End Process 36

TABLES

2-1 Yields of Actinides in Grams per 200 g of LEU; Yields after Five Cooling Times Are Shown 4

2-2 Yields of Actinides in Ci per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown 5

2-3 Yields of Selected Fission Products in Grams per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown 6

2-4 Yields of Selected Alkali Metals, Alkaline Earths, Iodine and Lanthanide Fission Products per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown 7

2-5 Total Element Yields of Fission Products per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown 8

2-6 Yield of Selected Fission Products per 200 g of Irradiated LEU 10

2-7 Yields of Selected Alkali Metals, Alkaline Earths, Iodine, and Lanthanide Fission Products per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown 12

2-8 Total Element Yields of Fission Products per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown 14

2-9 Fission Gas Yields in Moles per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown..... 16

2-10 Fission Gas Yields in Ci per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown 19

2-11 Yield of Thermal Watts for Selected Actinide Isotopes per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown..... 20

2-12 Thermal Watts Based on Element Yields of Fission Products per 200 g of Irradiated LEU 20

3-1 Preliminary Design of Chromatography Column for Separation of Mo from Target Solution..... 25

3-2 Waste Stream Chemical Composition for the Nitric-Acid Front-End Process for 200 g of Irradiated LEU Foil 26

TABLES (Cont.)

3-3 Waste-Stream Isotopic Composition for Cladding Waste from Nine Irradiated Targets Containing 200 g of LEU, in Grams 27

3-4 Waste-Stream Isotopic Composition for Cladding Waste from Nine Irradiated Targets Containing 200 g of LEU, in Curies 27

3-5 Thermodynamic Modeling Results Simulating the Off-Gas Composition for the Dissolution of Uranium Metal after the Dissolver Has Cooled to a Temperature of 25°C 28

3-6 Iodine Yields in Moles per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown 30

3-7 Iodine Yields in Curies per 200 g of Irradiated LEU; Yields after Five Cooling Times Are Shown 30

3-8 Partitioning of Elements from 200 g Irradiated Target between Product and Waste Streams Following a 200-hr Irradiation and 12-hr Cooling Period before Processing 34

4-1 Waste/Product Stream Chemical Composition 37

4-2 Partitioning of Elements from 200-g Irradiated Target between Product and Waste Streams Following a 200-hr Irradiation and 12-hr Cooling Period before Processing 38

4-3 Selected Fission Products in Waste Stream 1 39

4-4 Selected Fission Products in Waste Stream 2 40

4-5 Nuclide Composition of Waste Stream 3 41

4-6 Nuclide Composition of Waste Stream 4 41

FRONT-END PROCESSES FOR CONVERSION OF CURRENT HEU-BASED ALKALINE PROCESSES TO LEU-FOIL TARGETS: VOLUMES AND COMPOSITIONS OF ALL WASTE, PRODUCT, AND OFF-GAS STREAMS FROM BOTH FRONT-END OPTIONS

1 INTRODUCTION

The Global Threat Reduction Initiative (GTRI) Conversion Program develops technology necessary to enable the conversion of civilian facilities using high enriched uranium (HEU) to low enriched uranium (LEU) fuels and targets. The main technology components of the program are as follows:

- The development of advanced LEU fuels,
- Design and safety analysis for research reactor conversion, and
- Development of targets and processes for the non-HEU production of the medical isotope molybdenum-99 (^{99}Mo).

The conversion of conventional HEU dispersion targets to LEU for the production of ^{99}Mo requires approximately five times the uranium in a target to maintain the ^{99}Mo yield per target; therefore, a much denser form of uranium in the target is necessary. To meet this requirement, Argonne National Laboratory (Argonne) has developed the annular LEU-foil target, where a uranium metal foil (~125–150 µm thick) is inserted between two aluminum tubes. Figure 1-1 is a schematic of this target.

Because of the high density of uranium metal, the annular LEU-foil target can contain much greater quantities of uranium compared to aluminide, silicide, or oxide targets, and therefore can produce equivalent yields in the same irradiation position. An example of how this can be accomplished is to look at the possible conversion of the current Institute of Radio Elements (IRE) HEU target (Fallais et al. 1978). The IRE target is in the form of an HEU-aluminide dispersion plate that has been curved and then welded into an annular target. The current target contains 3.7 g of ^{235}U, with 93% enriched uranium. An LEU-foil target with a

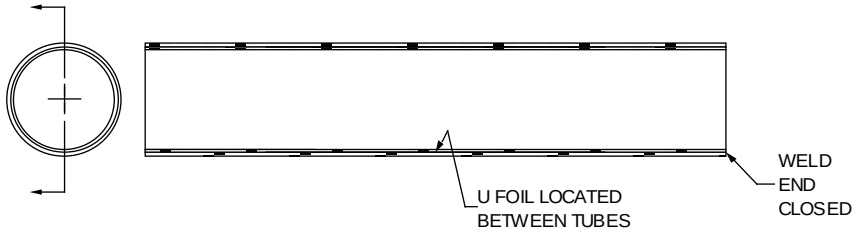

FIGURE 1-1 Schematic of the Argonne Annular LEU-Foil Target

standard 135-µm-thick foil of the same dimensions, with 19.9% enriched uranium, would contain 4.1 g of ^{235}U. If the LEU foil were made as thick as the current fuel meat (510 µm), the target would contain 16.6 g of ^{235}U.[1]

However, the main concern in using the LEU-foil target is that this target cannot be used in the current processes for recovery and purification of molybdenum, which were developed for the HEU-aluminide targets. In the current alkaline-based processes used by IRE, Covidien, and NTP Radioisotopes SOC Ltd. (NTP), the entire target is digested in a solution of either sodium hydroxide or sodium hydroxide/sodium nitrate. In the digestion process, uranium and the alkaline-insoluble fission and activation products precipitate as hydrous metal oxides; Mo is solubilized and stays in solution with most of the Na and Al, as well as a few other alkaline-soluble fission products (e.g., I, Cs, Se, Te). The digestion process is the first purification step, and the bulk of the radioactive impurities report to the solid precipitate. The Mo contained in the filtrate is then put through a series of purification steps to achieve the purity requirements for medical application of its daughter, 99mTc.

Under GTRI, Argonne is developing two front-end options for the current processes to allow the use of LEU-foil targets. In both processes, the goal is to produce a product after the front end that will be compatible with current purification operations and that will, with the same number of targets irradiated, provide the same or higher yield of 99Mo at the end of processing. The goal of both front-end processes is to deliver a product solution with (1) the same or higher Mo purity as the current target-digestion filtrate, (2) an equal or lower volume than the current filtrate solution, and (3) equal or better compatibility with current purification process.

In the first front-end option, the LEU foil (contained in a thin [10–15 µm] Ni fission-recoil barrier[2)] is removed from the annular target and then dissolved in nitric acid. In the dissolution, uranium, nickel, and all fission and activation products are dissolved; the nitric-acid concentration after dissolution will be ~1 M. Following dissolution, the solution is fed to a column containing a TiO2 sorbent, which is highly specific for Mo. After washing the column with 0.1-M nitric acid and then water, Mo is stripped using 0.1-M sodium hydroxide. This product solution will be compatible with current purification processes.

In the second front-end option, the LEU-foil will be contained in a 40-µm Al foil (see footnote 2). After being removed from the aluminum target, the Al fission-recoil barrier is dissolved from the LEU foil using an alkaline solution. The mass of the Al fission recoil barrier is approximately 2 g for a 24-g LEU foil and will contain about 2% of the Mo and other fission products (due to fission recoil into the barrier).[3] After the fission-recoil barrier is removed from

---

[1] However, a target containing such a high loading of ^{235}U in the geometry of the current IRE target would certainly be unusable in the reactors currently used by IRE for irradiation. The power density in the target would be well above the ability of the reactors' cooling system to irradiate the target safely; geometry of the target will not limit the uranium loading.

[2] This fission-recoil barrier is wrapped around the LEU foil to assure that the foil does not bind to the inner and outer Al tubes and can be removed from the target after irradiation. The inner and outer tubes will then be disposed of as solid low-level waste.

[3] Whether this Mo is worth recovering will be an economic decision. If it is decided to recover this Mo, it can be combined with the filtrate from the U/carbonate precipitation.

the LEU foil, the LEU foil will be electrolytically dissolved into a sodium bicarbonate solution. Uranyl carbonate is soluble to approximately 80 g-U/L. Other spent fuel components have limited solubility in sodium bicarbonate, and after uranium dissolution the solution will likely contain a fine precipitate of other components as hydroxides. At the end of dissolution, the slurry will be moved into a second stirred vessel where the addition of CaO will precipitate the carbonate, uranium, and alkaline-insoluble fission and activation products. The pH of this solution will be set by Ca(OH)2 solubility at 12.5. The filtrate from the precipitation step will be compatible with current alkaline-based processes.

In the following three sections, the expected irradiated-foil composition and the two front-end processes will be described in detail, and masses/volumes and compositions of the product and waste streams will be identified. The last section will discuss the annular-LEU-foil-target qualification experiments that will be performed by IRN in Pitesti Romania.

2 COMPOSITION OF IRRADIATED LEU-FOIL

The composition (actinides and fission products) of the LEU foils that will be processed in the two front-end processes described below was calculated by Charlie Allen, University of Missouri, using ORIGEN2, Version 2.2. The assumptions used for these calculations were as follows:

- Irradiation of uranium foil enriched to 19.75% ^{235}U.
- Power = 1.9×10^{-3} MW; burnup = 1.59×10^{-2} MW-days; flux = 2.1×10^{14} N/cm^2 s.
- Burnup is for 200 hours; foil composition is given for cooling times of 12, 24, 36, and 48 hours.

The results from the ORIGEN calculations were used to determine the actinide and fission product content (in grams and curies) for 200 g of irradiated LEU. This mass of LEU was chosen so that the compositions calculated would reflect the waste streams produced in the operation of a production-scale ^{99}Mo generation processes.

Selected results from these calculations are presented in Tables 2-1 through 2-12 and Figures 2-1 through 2-5. Descriptions of the chemical, elemental, and radioisotope compositions of the specific waste streams discussed in Sections 3 and 4 are derived from these ORIGEN calculation results.

2.1 ACTINIDES

The actinide content of the irradiated foils is shown in Tables 2-1 and 2-2. In terms of mass, the primary actinides are the uranium isotopes, with minor amounts of plutonium and neptunium (other actinides are all less than 0.001 g). In terms of curie content, ^{239}Np and ^{237}U dominate (other actinides are less than 0.001 Ci).

TABLE 2-1 Yields of Actinides in Grams per 200 g of LEU (200-hr irradiation at a flux of 2.1×10^{14} neutron/ cm^2 s); Yields after Five Cooling Times Are Shown

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
U-238	160.4	160.44	160.44	160.44	160.44
U-236	0.56	0.56	0.56	0.56	0.56
U-235	35.64	35.64	35.64	35.64	35.64
Pu-239	0.04	0.04	0.05	0.05	0.05
Np-239	0.02	0.02	0.02	0.02	0.01
Total	196.70	196.71	196.71	196.71	196.71

TABLE 2-2 Yields of Actinides in Ci per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1×10^{14} N/cm^2 s); Yields after Five Cooling Times Are Shown.

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Np-239	5.7E+03	4.9E+03	4.3E+03	3.7E+03	3.2E+03
U-237	1.4E+01	1.3E+01	1.2E+01	1.2E+01	1.1E+01
Np-238	5.4E-02	4.6E-02	3.9E-02	3.3E-02	2.8E-02
Np-240m	1.1E+01	3.3E-02	1.8E-02	1.0E-02	5.6E-03
U-240	5.9E-02	3.2E-02	1.8E-02	1.0E-02	5.5E-03
Pu-239	2.4E-03	2.6E-03	2.8E-03	3.0E-03	3.1E-03
U-235	7.7E-05	7.7E-05	7.7E-05	7.7E-05	7.7E-05
U-238	5.4E-05	5.4E-05	5.4E-05	5.4E-05	5.4E-05
Total	5.7E+03	4.9E+03	4.3E+03	3.7E+03	3.2E+03

2.2 FISSION PRODUCTS

The mass (in grams) of fission products produced by irradiation of 200 g of LEU are shown in Tables 2-3, 2-4, and 2-5 (only isotopes with more than 0.001-g yields are shown). Isotopes of zirconium, cerium, molybdenum, neodymium, barium, ruthenium, cesium, strontium, lanthanum, and praseodymium are most abundant. The total mass of fission products produced is approximately 2.7 g per 200 g of irradiated LEU (Table 2-5). Figures 2-1 and 2-3 show the mass percent of the major fission product isotopes for 12-hr and 48-hr cooling times.

Some of the shortest lived radionuclides (i.e., those not present after 12 hr of cooling) were omitted from the radionuclide tables in this section.

The yields of fission products in terms of curie content are shown in Tables 2-6, 2-7, and 2-8. The most abundant isotopes (based on activity) are molybdenum, cerium, technetium, tellurium, barium, lanthanum, praseodymium, niobium, strontium, rhodium, promethium, and ruthenium (Table 2-8). The total curie content for the fission products produced by irradiating 200 g of LEU is 1.6×10^5 Ci after 12 hr of cooling and 8.0×10^4 after 48 hr of cooling (Table 2-8). Figures 2-3 and 2-4 show the percent fission product yield in terms based on activity (curie content) for cooling periods of 12 and 48 hr, respectively.

TABLE 2-3 Yields of Selected[a] Fission Products in Grams per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1×10^{14} N/cm^2 s); Yields after Five Cooling Times Are Shown

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Se-82	3.9E-03	3.9E-03	3.9E-03	3.9E-03	3.9E-03
Se-80	1.4E-03	1.4E-03	1.4E-03	1.4E-03	1.4E-03
Br-81	2.4E-03	2.4E-03	2.4E-03	2.4E-03	2.4E-03
Y-91	6.7E-02	7.0E-02	7.0E-02	7.0E-02	7.0E-02
Y-89	3.4E-03	3.8E-03	4.2E-03	4.5E-03	4.9E-03
Zr-97	9.3E-03	5.7E-03	3.5E-03	2.1E-03	1.3E-03
Zr-96	8.5E-02	8.5E-02	8.5E-02	8.5E-02	8.5E-02
Zr-95	8.2E-02	8.2E-02	8.1E-02	8.1E-02	8.0E-02
Zr-94	8.3E-02	8.3E-02	8.3E-02	8.3E-02	8.3E-02
Zr-93	7.9E-02	8.2E-02	8.4E-02	8.4E-02	8.5E-02
Zr-92	7.5E-02	7.8E-02	7.8E-02	7.8E-02	7.8E-02
Zr-91	3.4E-03	3.8E-03	4.2E-03	4.7E-03	5.1E-03
Nb-95	3.6E-03	4.0E-03	4.4E-03	4.8E-03	5.2E-03
Mo-99	3.5E-02	3.1E-02	2.7E-02	2.4E-02	2.1E-02
Mo-98	8.0E-02	8.0E-02	8.0E-02	8.0E-02	8.0E-02
Mo-97	7.0E-02	7.4E-02	7.7E-02	7.8E-02	7.9E-02
Mo-100	8.9E-02	8.9E-02	8.9E-02	8.9E-02	8.9E-02
Tc-99m	2.8E-03	2.6E-03	2.3E-03	2.1E-03	1.8E-03
Tc-99	4.9E-02	5.3E-02	5.7E-02	6.1E-02	6.4E-02
Ru-106	5.8E-03	5.8E-03	5.8E-03	5.8E-03	5.8E-03
Ru-104	2.7E-02	2.7E-02	2.7E-02	2.7E-02	2.7E-02
Ru-103	4.2E-02	4.2E-02	4.2E-02	4.1E-02	4.1E-02
Ru-102	6.0E-02	6.0E-02	6.0E-02	6.0E-02	6.0E-02
Ru-101	7.2E-02	7.2E-02	7.2E-02	7.2E-02	7.2E-02
Rh-105	2.2E-03	2.1E-03	1.7E-03	1.4E-03	1.1E-03
Rh-103	3.2E-03	3.6E-03	3.9E-03	4.3E-03	4.7E-03
Pd-108	1.1E-03	1.1E-03	1.1E-03	1.1E-03	1.1E-03
Pd-107	2.5E-03	2.5E-03	2.5E-03	2.5E-03	2.5E-03
Pd-106	4.8E-03	4.9E-03	4.9E-03	4.9E-03	4.9E-03
Pd-105	7.7E-03	8.2E-03	8.6E-03	9.0E-03	9.3E-03
Sn-126	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03
Te-132	3.6E-02	3.3E-02	2.9E-02	2.6E-02	2.4E-02
Te-130	2.6E-02	2.6E-02	2.6E-02	2.6E-02	2.6E-02
Te-129M	1.5E-03	1.6E-03	1.5E-03	1.5E-03	1.5E-03
Te-128	6.3E-03	6.3E-03	6.3E-03	6.3E-03	6.3E-03

[a] Selected based on their yield being >0.001 g/200g-LEU.

TABLE 2-4 Yields of Selected[a] Alkali Metals, Alkaline Earths, Iodine and Lanthanide Fission Products (grams) per 200 g of Irradiated LEU (200-hr Irradiation at a Flux of 2.1×10^{14} N/cm^2 s); Yields after Five Cooling Times Are Shown

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Rb-87	3.1E-02	3.2E-02	3.2E-02	3.2E-02	3.2E-02
Rb-85	1.2E-02	1.2E-02	1.2E-02	1.2E-02	1.2E-02
Sr-90	7.3E-02	7.3E-02	7.3E-02	7.3E-02	7.3E-02
Sr-89	5.7E-02	5.7E-02	5.6E-02	5.6E-02	5.6E-02
Sr-88	4.5E-02	4.5E-02	4.6E-02	4.6E-02	4.6E-02
I-133	1.8E-02	1.3E-02	8.4E-03	5.7E-03	3.8E-03
I-131	3.6E-02	3.5E-02	3.4E-02	3.3E-02	3.1E-02
I-129	1.0E-02	1.0E-02	1.0E-02	1.0E-02	1.0E-02
I-127	9.5E-04	1.0E-03	1.1E-03	1.2E-03	1.3E-03
Cs-137	1.2E-01	1.2E-01	1.2E-01	1.2E-01	1.2E-01
Cs-135	4.4E-03	6.2E-03	8.1E-03	9.2E-03	9.8E-03
Cs-133	3.8E-02	4.3E-02	4.7E-02	5.2E-02	5.6E-02
Ba-140	9.9E-02	9.6E-02	9.4E-02	9.1E-02	8.9E-02
Ba-138	1.3E-01	1.3E-01	1.3E-01	1.3E-01	1.3E-01
La-140	9.9E-03	1.0E-02	1.1E-02	1.1E-02	1.1E-02
La-139	1.3E-01	1.3E-01	1.3E-01	1.3E-01	1.3E-01
Ce-144	1.1E-01	1.1E-01	1.1E-01	1.1E-01	1.1E-01
Ce-143	2.7E-02	2.1E-02	1.7E-02	1.3E-02	1.0E-02
Ce-142	1.2E-01	1.2E-01	1.2E-01	1.2E-01	1.2E-01
Ce-141	1.0E-01	1.1E-01	1.0E-01	1.0E-01	1.0E-01
Ce-140	1.5E-02	1.7E-02	1.9E-02	2.1E-02	2.4E-02
Pr-143	7.7E-02	8.1E-02	8.4E-02	8.6E-02	8.6E-02
Pr-141	9.6E-03	1.1E-02	1.2E-02	1.3E-02	1.4E-02
Nd-150	1.4E-02	1.4E-02	1.4E-02	1.4E-02	1.4E-02
Nd-148	3.5E-02	3.5E-02	3.5E-02	3.5E-02	3.5E-02
Nd-147	3.6E-02	3.5E-02	3.4E-02	3.3E-02	3.2E-02
Nd-146	6.1E-02	6.2E-02	6.2E-02	6.2E-02	6.2E-02
Nd-145	7.7E-02	7.9E-02	8.0E-02	8.0E-02	8.0E-02
Nd-144	1.8E-03	2.0E-03	2.1E-03	2.2E-03	2.4E-03
Nd-143	1.4E-02	1.6E-02	1.9E-02	2.1E-02	2.3E-02
Pd-149	7.4E-03	6.5E-03	5.6E-03	4.8E-03	4.1E-03
Pd-147	1.0E-02	1.2E-02	1.3E-02	1.4E-02	1.5E-02
Sm-154	1.6E-03	1.6E-03	1.6E-03	1.6E-03	1.6E-03
Sm-152	9.8E-03	9.8E-03	9.8E-03	9.8E-03	9.8E-03
Sm-151	3.2E-03	3.6E-03	3.9E-03	4.2E-03	4.4E-03
Sm-150	1.2E-02	1.2E-02	1.2E-02	1.2E-02	1.2E-02
Sm-149	3.0E-03	4.1E-03	5.0E-03	5.8E-03	6.5E-03
Eu-153	2.4E-03	2.6E-03	2.8E-03	2.9E-03	3.0E-03

[a] Selected based on their yield being >0.001 g/200g-LEU.

TABLE 2-5 Total Element Yields of Fission Products (grams) per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1×10^{14} N/cm^2 s); Yields after Five Cooling Times Are Shown

Element[a]	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Zr	4.2E-01	4.2E-01	4.2E-01	4.2E-01	4.2E-01
Ce	3.7E-01	3.7E-01	3.7E-01	3.7E-01	3.6E-01
Mo	2.7E-01	2.7E-01	2.7E-01	2.7E-01	2.7E-01
Nd	2.4E-01	2.4E-01	2.5E-01	2.5E-01	2.5E-01
Ba	2.3E-01	2.3E-01	2.3E-01	2.2E-01	2.2E-01
Ru	2.1E-01	2.1E-01	2.1E-01	2.1E-01	2.1E-01
Cs	1.6E-01	1.7E-01	1.7E-01	1.8E-01	1.8E-01
Sr	1.8E-01	1.8E-01	1.8E-01	1.8E-01	1.7E-01
La	1.4E-01	1.4E-01	1.4E-01	1.4E-01	1.4E-01
Pr	9.0E-02	9.3E-02	9.6E-02	9.9E-02	1.0E-01
Y	7.9E-02	7.7E-02	7.6E-02	7.6E-02	7.5E-02
Tc	5.2E-02	5.6E-02	5.9E-02	6.3E-02	6.5E-02
Te	7.3E-02	6.8E-02	6.5E-02	6.2E-02	5.9E-02
I	7.3E-02	6.2E-02	5.5E-02	5.1E-02	4.8E-02
Rb	4.3E-02	4.4E-02	4.4E-02	4.4E-02	4.4E-02
Sm	3.1E-02	3.2E-02	3.4E-02	3.4E-02	3.5E-02
Pm	2.0E-02	1.9E-02	1.9E-02	1.9E-02	1.9E-02
Pd	1.7E-02	1.7E-02	1.7E-02	1.8E-02	1.8E-02
Se	6.1E-03	6.1E-03	6.1E-03	6.1E-03	6.1E-03
Rh	5.5E-03	5.7E-03	5.7E-03	5.7E-03	5.8E-03
Nb	4.3E-03	4.4E-03	4.7E-03	5.0E-03	5.3E-03
Eu	3.4E-03	3.6E-03	3.8E-03	3.9E-03	4.0E-03
Sn	2.8E-03	2.7E-03	2.7E-03	2.6E-03	2.6E-03
Total	2.7	2.7	2.7	2.7	2.7

[a] Selected based on their yield being >0.001 g/200g-LEU.

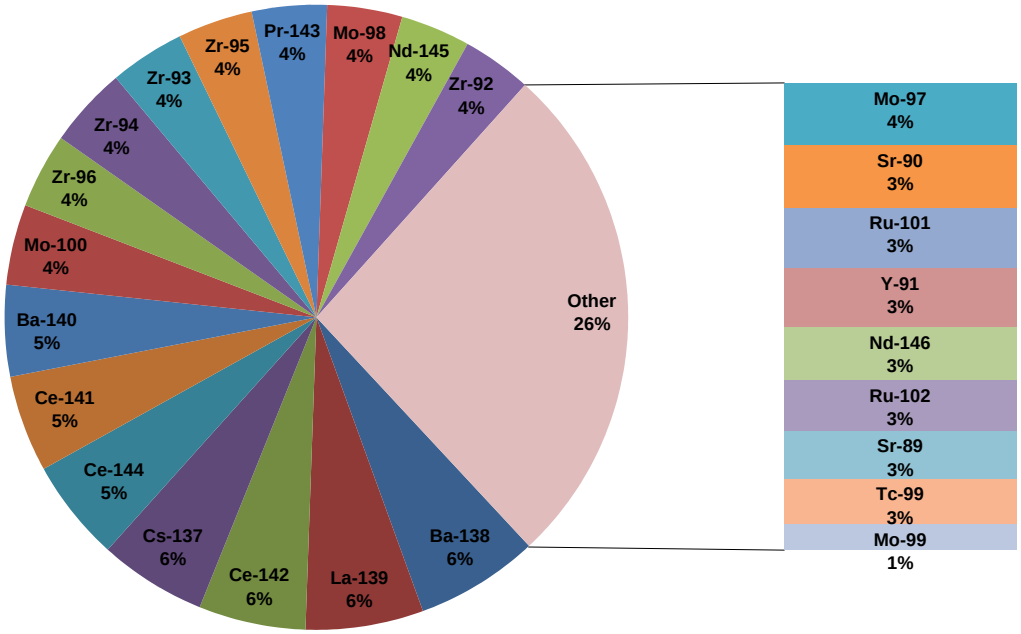

FIGURE 2-1 Mass Percent of Fission Products after 12 hr of Cooling for 200 g of LEU after 200-hr Irradiation

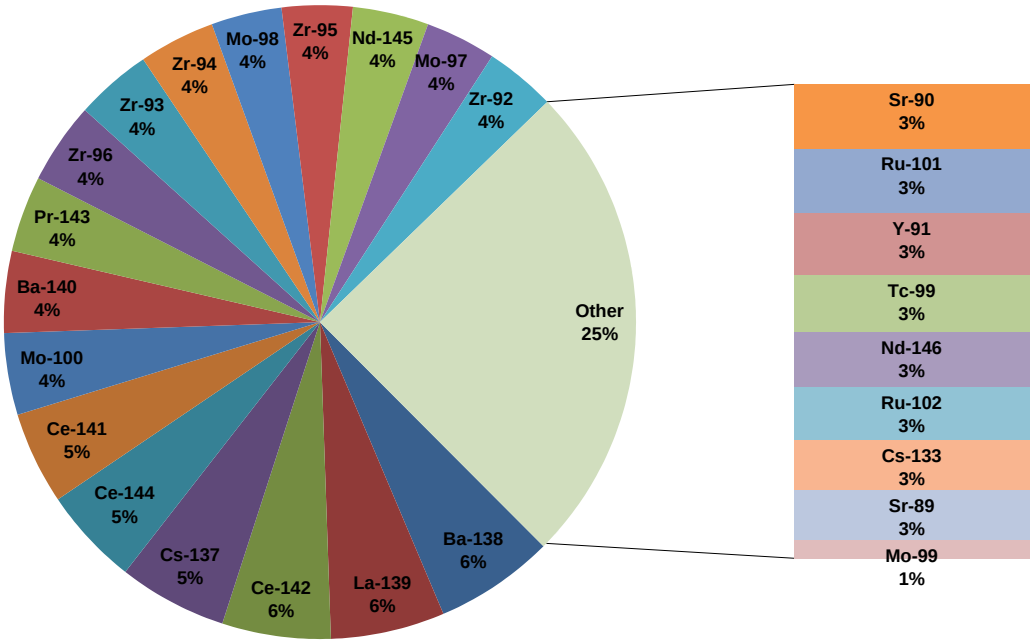

FIGURE 2-2 Mass Percent of Fission Products after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation

TABLE 2-6 Yield of Selected[a] Fission Products (Ci) per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1 × 1014 N/cm^2 s)

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
As-77	2.4E+01	2.1E+01	1.7E+01	1.4E+01	1.2E+01
Ge-77	8.9E+00	4.3E+00	2.0E+00	9.8E-01	4.7E-01
Br-82	5.9E-01	4.6E-01	3.7E-01	2.9E-01	2.3E-01
Y-90	6.7E+00	7.1E+00	7.4E+00	7.8E+00	8.0E+00
Y-93	2.0E+04	8.7E+03	3.8E+03	1.7E+03	7.4E+02
Y-92	1.8E+04	4.7E+03	5.8E+02	6.1E+01	6.2E+00
Y-91M	1.0E+04	4.7E+03	2.0E+03	8.2E+02	3.4E+02
Y-91	1.6E+03	1.7E+03	1.7E+03	1.7E+03	1.7E+03
Zr-97	1.8E+04	1.1E+04	6.7E+03	4.1E+03	2.5E+03
Zr-95	1.8E+03	1.8E+03	1.7E+03	1.7E+03	1.7E+03
Nb-97M	1.7E+04	1.0E+04	6.3E+03	3.9E+03	2.4E+03
Nb-97	1.8E+04	1.1E+04	6.7E+03	4.1E+03	2.5E+03
Nb-96	2.5E+00	1.8E+00	1.2E+00	8.6E-01	6.0E-01
Nb-95M	6.7E+00	7.2E+00	7.7E+00	8.1E+00	8.5E+00
Nb-95	1.4E+02	1.6E+02	1.7E+02	1.9E+02	2.0E+02
Mo-99	1.7E+04	1.5E+04	1.3E+04	1.1E+04	1.0E+04
Tc-99M	1.5E+04	1.4E+04	1.2E+04	1.1E+04	9.7E+03
Ru-106	1.9E+01	1.9E+01	1.9E+01	1.9E+01	1.9E+01
Ru-105	3.1E+03	4.9E+02	7.6E+01	1.2E+01	1.8E+00
Ru-103	1.4E+03	1.4E+03	1.3E+03	1.3E+03	1.3E+03
Rh-106	8.4E+02	1.9E+01	1.9E+01	1.9E+01	1.9E+01
Rh-105M	8.7E+02	1.4E+02	2.1E+01	3.3E+00	5.0E-01
Rh-105	1.9E+03	1.8E+03	1.5E+03	1.2E+03	9.1E+02
Rh-103M	1.2E+03	1.2E+03	1.2E+03	1.2E+03	1.2E+03
Pd-112	3.9E+01	2.6E+01	1.7E+01	1.1E+01	7.5E+00
Pd-109	9.7E+01	5.2E+01	2.8E+01	1.5E+01	8.2E+00
Ag-112	3.9E+01	3.0E+01	2.0E+01	1.3E+01	8.8E+00
Ag-111	3.4E+01	3.2E+01	3.1E+01	3.0E+01	2.8E+01
Ag-109M	9.6E+01	5.2E+01	2.8E+01	1.5E+01	8.2E+00
Cd-115	2.7E+01	2.3E+01	2.0E+01	1.7E+01	1.5E+01
In-115M	2.7E+01	2.5E+01	2.2E+01	1.9E+01	1.6E+01
Sn-125	1.9E+01	1.9E+01	1.8E+01	1.7E+01	1.7E+01
Sn-123	6.1E-01	6.1E-01	6.1E-01	6.1E-01	6.0E-01
Sn-121	3.9E+01	2.9E+01	2.1E+01	1.6E+01	1.1E+01
Sb-129	1.9E+03	2.9E+02	4.2E+01	6.1E+00	8.9E-01
Sb-128	3.4E+01	1.3E+01	5.3E+00	2.1E+00	8.4E-01
Sb-127	3.2E+02	3.0E+02	2.7E+02	2.5E+02	2.3E+02
Sb-126	1.1E+00	1.0E+00	9.9E-01	9.7E-01	9.4E-01
Sb-125	3.6E-01	3.6E-01	3.7E-01	3.7E-01	3.8E-01
Te-132	1.1E+04	9.9E+03	8.9E+03	8.0E+03	7.2E+03
Te-131M	1.1E+03	8.5E+02	6.4E+02	4.9E+02	3.7E+02
Te-131	7.7E+03	1.9E+02	1.4E+02	1.1E+02	8.3E+01

TABLE 2-6 (Cont.)

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Te-129M	4.6E+01	4.7E+01	4.7E+01	4.6E+01	4.6E+01
Te-129	1.7E+03	3.7E+02	8.0E+01	3.7E+01	3.1E+01
Te-127	2.7E+02	2.7E+02	2.5E+02	2.4E+02	2.2E+02

[a] Selected based on their yield being >0.1 Ci/200g-LEU.

TABLE 2-7 Yields of Selected[a] Alkali Metals, Alkaline Earths, Iodine, and Lanthanide Fission Products (Ci) per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1×10^{14} N/cm^2 s); Yields after Five Cooling Times Are Shown

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Rb-88	1.1E+04	6.6E+02	3.5E+01	1.9E+00	1.0E-01
Sr-91	1.8E+04	7.4E+03	3.1E+03	1.3E+03	5.4E+02
Sr-90	1.0E+01	1.0E+01	1.0E+01	1.0E+01	1.0E+01
Sr-89	1.7E+03	1.6E+03	1.6E+03	1.6E+03	1.6E+03
I-135	1.9E+04	5.5E+03	1.6E+03	4.4E+02	1.3E+02
I-134	2.3E+04	7.3E+00	5.9E-04	4.5E-08	3.4E-12
I-133	2.1E+04	1.4E+04	9.6E+03	6.4E+03	4.3E+03
I-132	1.1E+04	1.0E+04	9.2E+03	8.3E+03	7.4E+03
I-131	4.5E+03	4.4E+03	4.2E+03	4.0E+03	3.9E+03
I-130	7.1E+00	3.6E+00	1.9E+00	9.5E-01	4.8E-01
I-129	1.8E-06	1.8E-06	1.8E-06	1.8E-06	1.8E-06
I-128	2.0E-01	4.2E-10	8.8E-19	1.9E-27	0.0E+00
Cs-137	1.0E+01	1.0E+01	1.0E+01	1.0E+01	1.0E+01
Cs-136	6.3E+00	6.1E+00	5.9E+00	5.8E+00	5.6E+00
Ba-140	7.2E+03	7.0E+03	6.8E+03	6.6E+03	6.5E+03
Ba-137M	1.0E+01	9.7E+00	9.7E+00	9.7E+00	9.7E+00
Ba-136M	1.0E+00	1.0E+00	9.8E-01	9.5E-01	9.3E-01
Ba-140	7.2E+03	7.0E+03	6.8E+03	6.6E+03	6.5E+03
Ba-137M	1.0E+01	9.7E+00	9.7E+00	9.7E+00	9.7E+00
Ba-136M	1.0E+00	1.0E+00	9.8E-01	9.5E-01	9.3E-01
La-141	1.8E+04	2.3E+03	2.8E+02	3.4E+01	4.1E+00
La-140	5.5E+03	5.8E+03	6.0E+03	6.1E+03	6.2E+03
Ce-144	3.5E+02	3.5E+02	3.5E+02	3.5E+02	3.5E+02
Ce-143	1.8E+04	1.4E+04	1.1E+04	8.6E+03	6.7E+03
Ce-141	3.0E+03	3.0E+03	3.0E+03	3.0E+03	2.9E+03
Pr-145	1.2E+04	3.0E+03	7.4E+02	1.8E+02	4.6E+01
Pr-144M	4.3E+00	4.2E+00	4.2E+00	4.2E+00	4.2E+00
Pr-144	5.1E+02	3.5E+02	3.5E+02	3.5E+02	3.5E+02
Pr-143	5.2E+03	5.5E+03	5.7E+03	5.8E+03	5.8E+03
Pr-142	2.7E+00	1.7E+00	1.1E+00	7.3E-01	4.7E-01
Nd-147	2.9E+03	2.8E+03	2.7E+03	2.7E+03	2.6E+03
Pm-151	1.3E+03	9.5E+02	7.1E+02	5.3E+02	3.9E+02
Pm-149	2.9E+03	2.6E+03	2.2E+03	1.9E+03	1.6E+03
Pm-148M	5.0E-01	5.0E-01	4.9E-01	4.9E-01	4.8E-01
Pm-148	6.4E+00	6.0E+00	5.6E+00	5.3E+00	4.9E+00
Pm-147	9.6E+00	1.1E+01	1.2E+01	1.3E+01	1.4E+01
Sm-156	4.1E+01	1.7E+01	7.0E+00	2.9E+00	1.2E+00
Sm-153	5.1E+02	4.3E+02	3.6E+02	3.0E+02	2.5E+02
Sm-151	8.3E-02	9.5E-02	1.0E-01	1.1E-01	1.1E-01
Eu-157	2.0E+01	1.2E+01	6.7E+00	3.9E+00	2.2E+00
Eu-156	2.2E+01	2.2E+01	2.2E+01	2.1E+01	2.1E+01

TABLE 2-7 (Cont.)

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Eu-155	2.5E-01	2.5E-01	2.5E-01	2.5E-01	2.5E-01
Gd-159	3.2E+00	2.1E+00	1.4E+00	8.6E-01	5.5E-01
Tb-161	1.6E-01	1.5E-01	1.5E-01	1.4E-01	1.3E-01

[a] Selected based on their yield being >0.1 Ci/200g-LEU.

TABLE 2-8 Total Element Yields of Fission Products (Ci) per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1×10^{14} N/cm^2 s); Yields after Five Cooling Times Are Shown

Element[a]	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
I	1.1E+05	3.4E+04	2.5E+04	1.9E+04	1.6E+04
Mo	6.4E+04	1.5E+04	1.3E+04	1.1E+04	1.0E+04
Ce	5.7E+04	1.8E+04	1.4E+04	1.2E+04	9.9E+03
Tc	6.3E+04	1.4E+04	1.2E+04	1.1E+04	9.7E+03
Te	8.0E+04	1.2E+04	1.0E+04	8.9E+03	8.0E+03
Ba	1.0E+05	7.1E+03	6.8E+03	6.7E+03	6.5E+03
La	9.8E+04	8.2E+03	6.3E+03	6.2E+03	6.2E+03
Pr	4.5E+04	8.8E+03	6.8E+03	6.3E+03	6.2E+03
Nb	1.3E+05	2.1E+04	1.3E+04	8.1E+03	5.1E+03
Zr	8.8E+04	1.3E+04	8.4E+03	5.8E+03	4.2E+03
Y	1.4E+05	2.0E+04	8.1E+03	4.3E+03	2.8E+03
Nd	9.0E+03	2.9E+03	2.8E+03	2.7E+03	2.6E+03
Sr	1.1E+05	1.0E+04	4.8E+03	2.9E+03	2.2E+03
Rh	6.3E+03	3.2E+03	2.7E+03	2.4E+03	2.1E+03
Pm	6.1E+03	3.6E+03	2.9E+03	2.4E+03	2.0E+03
Ru	5.5E+03	1.9E+03	1.4E+03	1.4E+03	1.3E+03
Sm	6.8E+02	4.4E+02	3.6E+02	3.0E+02	2.5E+02
Sb	3.2E+04	6.0E+02	3.2E+02	2.6E+02	2.3E+02
Ag	5.0E+02	1.2E+02	8.1E+01	5.8E+01	4.5E+01
Sn	1.1E+04	5.3E+01	4.0E+01	3.3E+01	2.9E+01
Eu	5.6E+01	3.4E+01	2.9E+01	2.5E+01	2.3E+01
Cs	8.6E+04	1.7E+01	1.6E+01	1.6E+01	1.6E+01
In	1.3E+03	3.1E+01	2.2E+01	1.9E+01	1.6E+01
Pd	4.2E+02	7.8E+01	4.5E+01	2.7E+01	1.6E+01
Cd	3.5E+02	2.6E+01	2.0E+01	1.7E+01	1.5E+01
As	5.5E+03	2.2E+01	1.7E+01	1.4E+01	1.2E+01
Gd	3.7E+00	2.1E+00	1.4E+00	8.6E-01	5.5E-01
Ge	1.7E+03	4.5E+00	2.0E+00	9.8E-01	4.7E-01
Br	4.0E+04	5.4E+01	2.0E+00	3.4E-01	2.3E-01
Rb	9.5E+04	6.6E+02	3.5E+01	2.0E+00	1.9E-01
Tb	3.1E-01	1.5E-01	1.5E-01	1.4E-01	1.3E-01
Ga	2.4E+02	2.4E+02	2.4E+02	2.4E+02	2.4E+02
Total	1.4E+06	1.9E+05	1.4E+05	1.1E+05	9.5E+04

[a] Selected based on their yield being >0.1 Ci/200g-LEU.

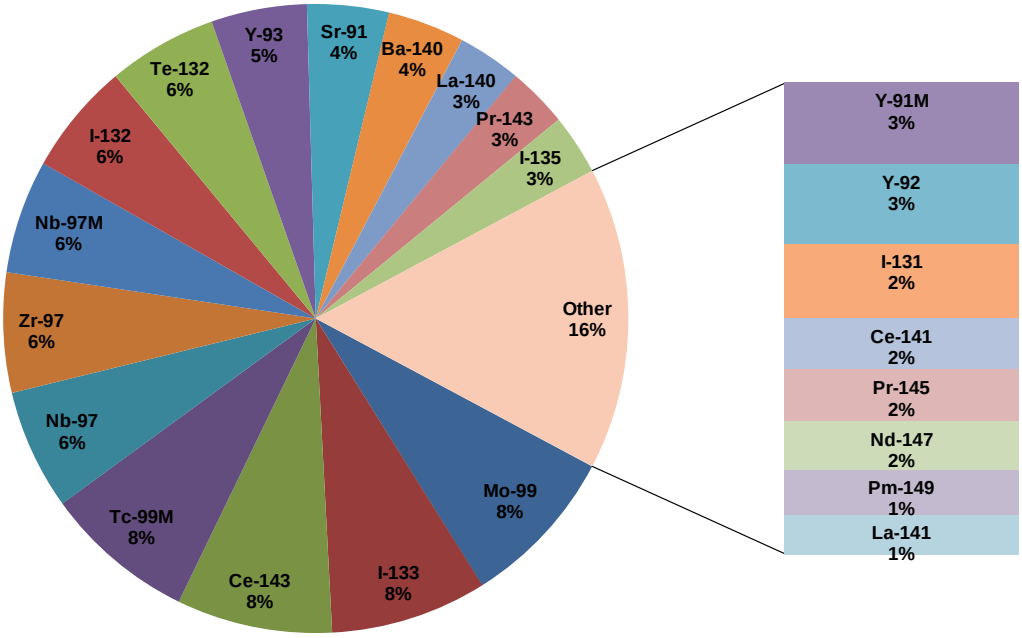

FIGURE 2-3 Percent of Fission Products (based on curie content) after 12 hr of Cooling for 200 g of LEU after 200-hr Irradiation

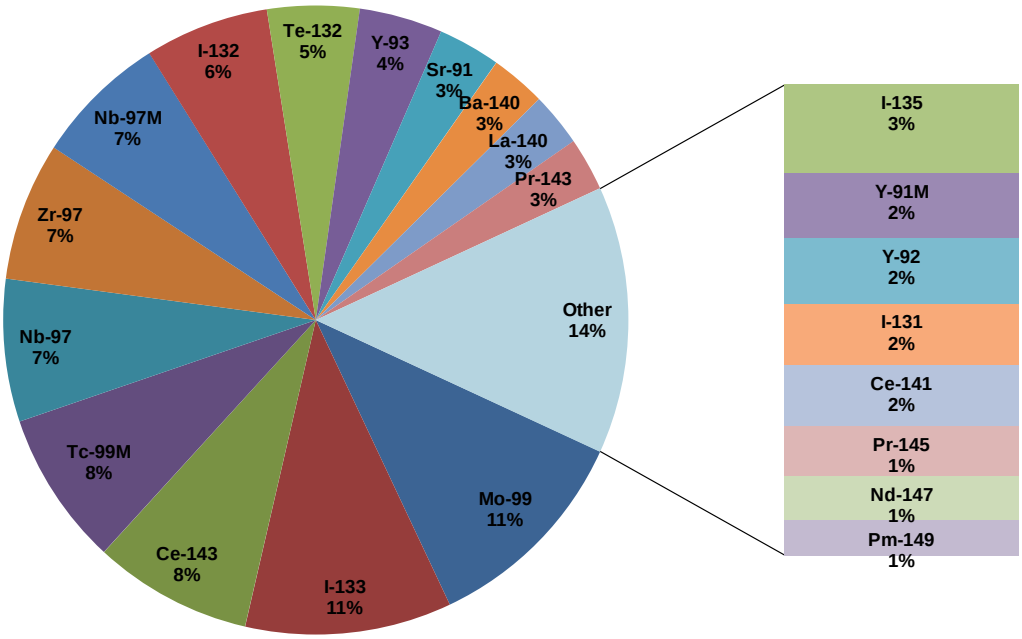

FIGURE 2-4 Percent of Fission Products (based on curie content) after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation

2.3 NOBLE FISSION GASES

Noble fission gas yields are shown in Tables 2-8 and 2-9 in terms of moles and curie content (respectively). These tables show that xenon isotopes are the most important relatively long-lived fission gases. It should be noted that, even though iodine is not listed with the noble fission gases, its speciation will depend on the thermochemical conditions during processing. For example, in the acid-based front-end process, the relatively volatile I_2 species is expected to be the main form of iodine; in the more basic solutions of the electrochemical front-end process, however, much of the iodine is expected to stay in solution as I^-. Figures 2-5 and 2-6 show the mole percent of the major fission gas isotopes for 12-hr and 48-hr cooling times. Figures 2-7 and 2-8 show the percent fission gas yield in terms based on activity (curie content) for cooling periods of 12 and 48 hr, respectively.

It should also be noted that, for the nitric-acid-based front-end process, the fission gases will make up a small fraction (less than 1%) of the overall off-gas stream due to the large volumes of nitrogen oxides produced. The amount and speciation nitrogen oxide gases produced in the nitric acid front-end process is discussed below.

TABLE 2-9 Fission Gas Yields in Moles per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1 × 14 N/cm^2 s); Yields after Five Cooling Times Are Shown

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
H-3	1.5E-06	1.5E-06	1.5E-06	1.5E-06	1.5E-06
Kr-88	1.7E+00	9.2E-02	4.9E-03	2.6E-04	1.4E-05
Kr-87	1.1E+00	1.7E-03	2.4E-06	3.4E-09	5.0E-12
Kr-85M	1.1E-01	1.8E-02	2.7E-03	4.3E-04	6.7E-05
Kr-85	2.2E-05	2.2E-05	2.2E-05	2.2E-05	2.2E-05
Kr-83M	4.7E-03	4.8E-04	1.8E-05	6.1E-07	1.9E-08
Kr-81	9.8E-19	9.8E-19	9.8E-19	9.8E-19	9.8E-19
Total Kr	3.0E+00	1.1E-01	7.7E-03	7.1E-04	1.0E-04
Xe-138	1.5E+00	7.5E-16	3.8E-31	0.0E+00	0.0E+00
Xe-135M	8.0E-02	2.0E-02	5.8E-03	1.6E-03	4.6E-04
Xe-135	1.8E-02	1.6E-01	1.0E-01	5.4E-02	2.5E-02
Xe-133M	5.7E-03	5.6E-03	5.3E-03	4.8E-03	4.4E-03
Xe-133	1.0E-01	1.0E-01	1.0E-01	1.0E-01	9.8E-02
Xe-131M	8.4E-05	9.2E-05	9.9E-05	1.1E-04	1.1E-04
Xe-129M	1.0E-10	1.0E-10	9.6E-11	9.2E-11	8.8E-11
Total Xe	1.7E+00	2.9E-01	2.2E-01	1.6E-01	1.3E-01
Total	4.7	0.4	0.23	0.16	0.13

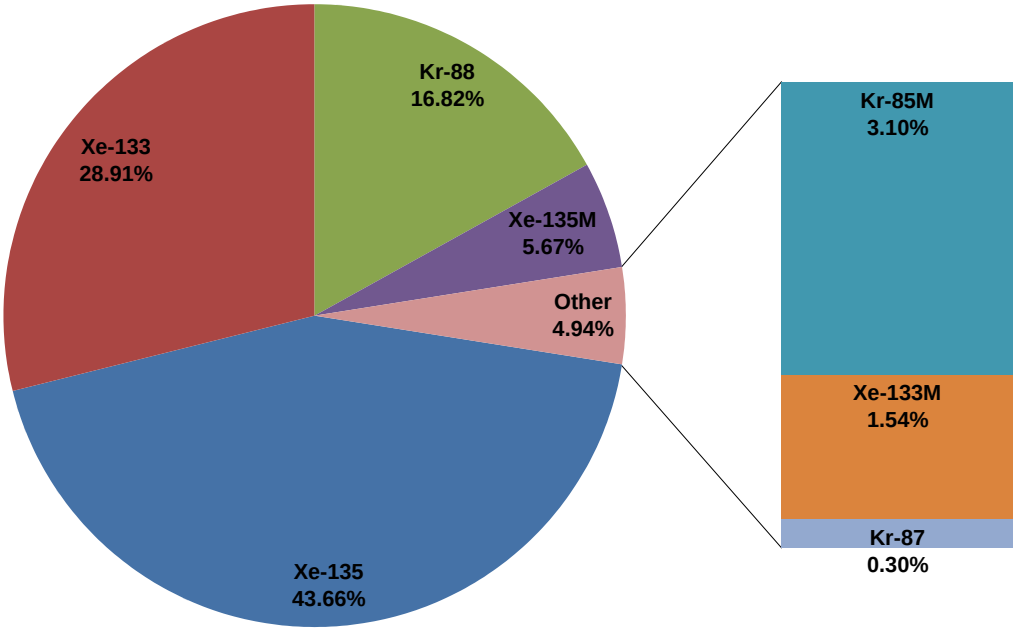

FIGURE 2-5 Mole Percent of Noble Fission Gases after 12 hr of Cooling for 200 g of LEU after 200-hour Irradiation

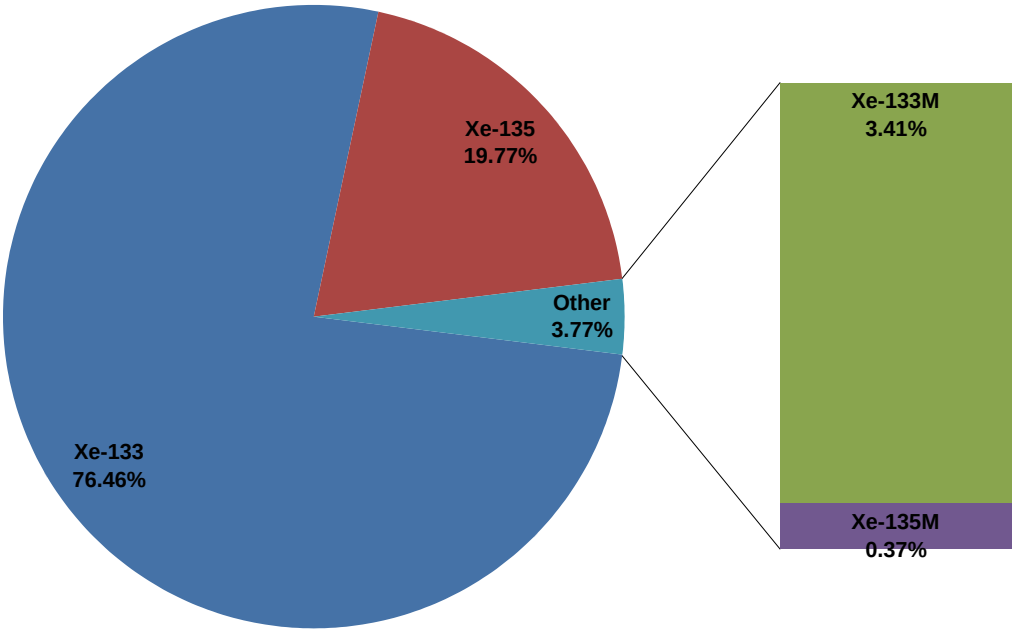

FIGURE 2-6 Mole Percent of Noble Fission Gases after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation

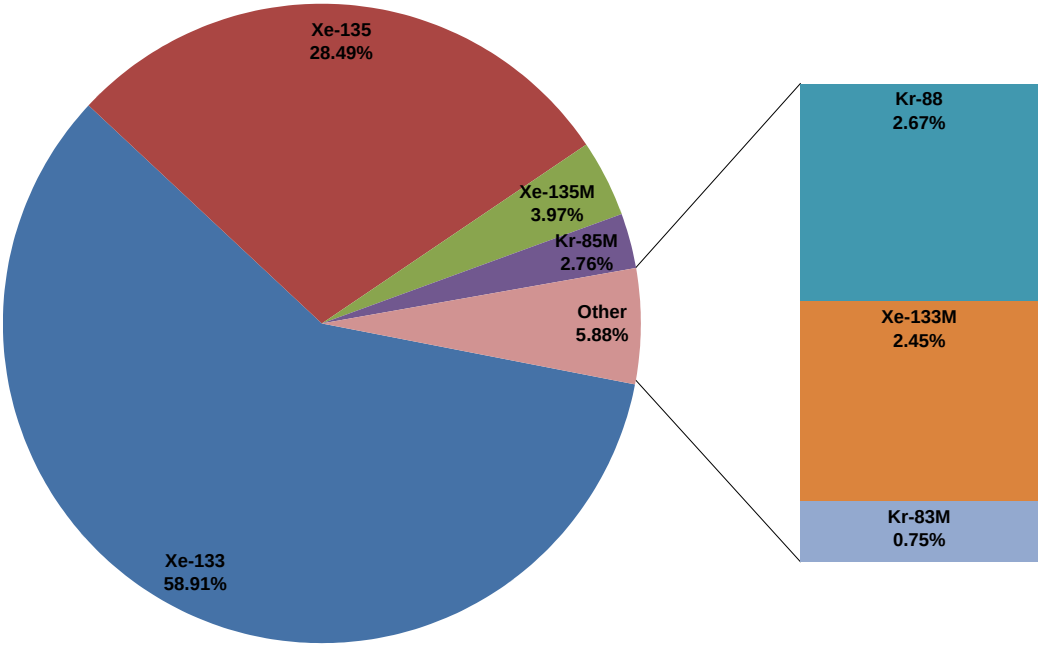

FIGURE 2-7 Percent of Noble Fission Gases (based on curie content) after 12 hr of Cooling for 200 g of LEU after 200-hr Irradiation

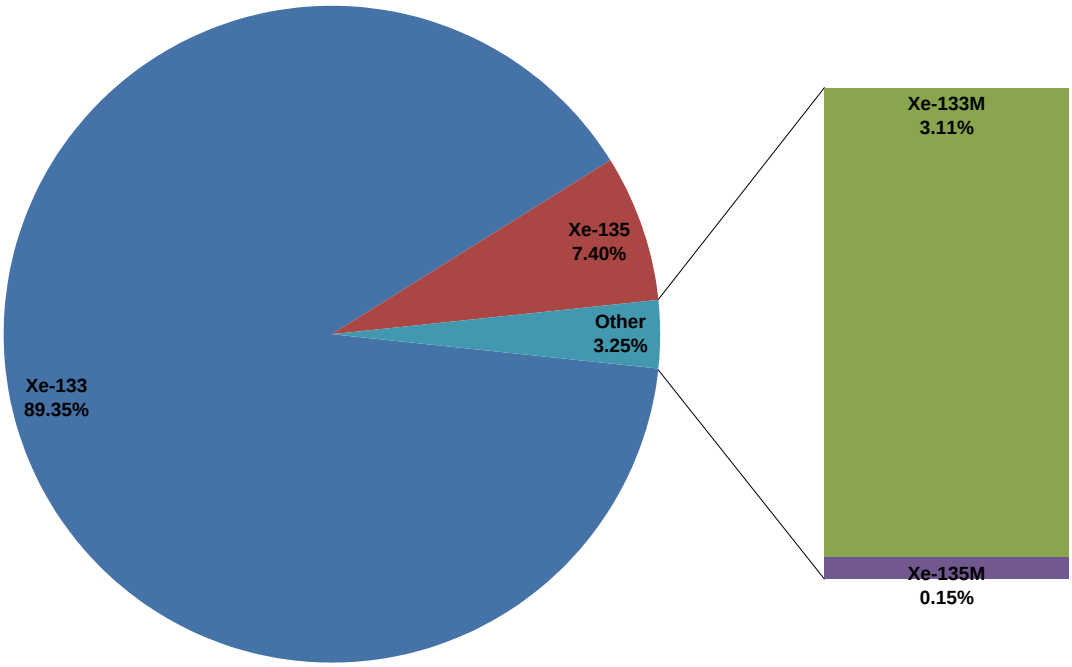

FIGURE 2-8 Percent of Noble Fission Gases (based on curie content) after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation

TABLE 2-10 Fission Gas Yields in Ci per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1 × 14 N/cm^2 s); Yields after Five Cooling Times Are Shown

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
H-3	4.5E-02	4.5E-02	4.5E-02	4.5E-02	4.5E-02
Kr-88	1.1E+04	5.9E+02	3.1E+01	1.7E+00	9.0E-02
Kr-87	7.8E+03	1.1E+01	1.6E-02	2.4E-05	3.4E-08
Kr-85M	3.9E+03	6.1E+02	9.5E+01	1.5E+01	2.3E+00
Kr-85	1.2E+00	1.3E+00	1.3E+00	1.3E+00	1.3E+00
Kr-83M	1.6E+03	1.7E+02	6.4E+00	2.1E-01	6.6E-03
Kr-81	6.5E-13	6.5E-13	6.5E-13	6.5E-13	6.5E-13
Total Kr	2.4E+04	1.4E+03	1.3E+02	1.8E+01	3.7E+00
Xe-138	1.9E+04	9.6E-12	4.9E-27	0.0E+00	0.0E+00
Xe-135M	3.4E+03	8.8E+02	2.5E+02	7.1E+01	2.0E+01
Xe-135	7.3E+02	6.3E+03	4.2E+03	2.2E+03	1.0E+03
Xe-133M	5.5E+02	5.4E+02	5.1E+02	4.7E+02	4.2E+02
Xe-133	1.3E+04	1.3E+04	1.3E+04	1.3E+04	1.2E+04
Xe-131M	1.1E+01	1.3E+01	1.4E+01	1.4E+01	1.5E+01
Xe-129M	9.6E-06	9.2E-06	8.8E-06	8.4E-06	8.1E-06
Total Xe	3.6E+04	2.1E+04	1.8E+04	1.5E+04	1.4E+04
Total	6.0E+05	2.4E+04	1.9E+04	1.5E+04	1.4E+04

2.4 DECAY HEAT

The thermal wattage produced by decay heat of the actinides and fission products was also calculated and is shown in Tables 2-11 and 2-12 and summarized in Figure 2-9. The total thermal output for 200 g of irradiated LEU is around 1000 W for a 12-hr cooling time and around 500 W for a 48-hr cooling time. These thermal values have implications for the front-end processes because they must be designed to account for elevated temperatures that may be caused by decay heat.

TABLE 2-11 Yield of Thermal Watts for Selected[a] Actinide Isotopes per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1 × 10^{14} neutron/cm^2 s); Yields after Five Cooling Times Are Shown

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Np-239	1.4E+01	1.2E+01	1.0E+01	8.9E+00	7.7E+00
U-237	2.6E-02	2.5E-02	2.4E-02	2.2E-02	2.1E-02
Np-238	2.6E-04	2.2E-04	1.9E-04	1.6E-04	1.3E-04
Pu-239	7.4E-05	8.1E-05	8.6E-05	9.1E-05	9.5E-05
Np-240M	6.3E-02	1.9E-04	1.1E-04	5.8E-05	3.2E-05
Np-240	5.2E-02	2.4E-05	1.1E-08	5.1E-12	2.4E-15
U-239	1.7E+01	1.0E-08	6.4E-18	3.9E-27	0.0E+00
Total	31	12	10	8.9	7.7

[a] Selected based on 1.0 × 10^{-6} W/200g-LEU at 0 hr cooling.

TABLE 2-12 Thermal Watts (decay heat) Based on Element Yields of Fission Products per 200 g of Irradiated LEU (listed in order of decreasing wattage)

Element	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
I	1.8E+03	3.3E+02	2.4E+02	1.9E+02	1.6E+02
La	1.5E+03	1.1E+02	1.0E+02	1.0E+02	1.0E+02
Ce	3.7E+02	6.4E+01	5.1E+01	4.1E+01	3.3E+01
Mo	4.8E+02	4.7E+01	4.1E+01	3.7E+01	3.2E+01
Nb	1.7E+03	1.2E+02	7.3E+01	4.5E+01	2.8E+01
Zr	8.8E+02	6.6E+01	4.4E+01	3.0E+01	2.2E+01
Te	8.3E+02	3.1E+01	2.6E+01	2.2E+01	1.9E+01
Ba	9.9E+02	2.0E+01	1.9E+01	1.9E+01	1.8E+01
Xe	9.4E+02	3.8E+01	2.9E+01	2.2E+01	1.7E+01
Pr	4.3E+02	2.5E+01	1.6E+01	1.4E+01	1.4E+01
Y	2.2E+03	1.3E+02	4.7E+01	2.2E+01	1.3E+01
Sr	1.5E+03	7.3E+01	3.1E+01	1.6E+01	9.9E+00
Tc	4.8E+02	1.2E+01	1.0E+01	9.2E+00	8.1E+00
Nd	4.6E+01	7.0E+00	6.6E+00	6.4E+00	6.2E+00
Pm	3.1E+01	9.5E+00	7.6E+00	6.2E+00	5.1E+00
Ru	3.5E+01	8.0E+00	5.0E+00	4.5E+00	4.4E+00
Rh	3.0E+01	3.2E+00	2.5E+00	2.0E+00	1.7E+00
Sb	6.5E+02	5.2E+00	2.2E+00	1.6E+00	1.4E+00
Sm	1.9E+00	8.8E-01	7.2E-01	5.9E-01	4.9E-01
Eu	4.9E-01	2.8E-01	2.5E-01	2.4E-01	2.2E-01
Ag	4.8E+00	5.2E-01	3.4E-01	2.4E-01	1.8E-01
Sn	1.5E+02	2.3E-01	1.5E-01	1.4E-01	1.3E-01

TABLE 2-12 (Cont.)

Element	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Cs	1.7E+03	9.5E-02	9.3E-02	9.1E-02	8.9E-02
Cd	4.2E+00	9.0E-02	6.6E-02	5.6E-02	4.8E-02
In	3.3E+01	7.3E-02	4.5E-02	3.7E-02	3.2E-02
Pd	3.0E+00	1.6E-01	9.1E-02	5.1E-02	2.9E-02
As	1.3E+02	4.8E-02	2.5E-02	2.0E-02	1.6E-02
Kr	1.1E+03	9.8E+00	6.7E-01	6.1E-02	8.8E-03
Ge	2.8E+01	4.4E-02	2.1E-02	1.0E-02	4.8E-03
Br	8.6E+02	1.1E-01	9.2E-03	4.9E-03	3.8E-03
Rb	2.1E+03	1.0E+01	5.6E-01	3.0E-02	2.0E-03
Gd	1.3E-02	6.9E-03	4.4E-03	2.8E-03	1.8E-03
Total	2.10E+04	1.13E+03	7.57E+02	5.94E+02	5.00E+02

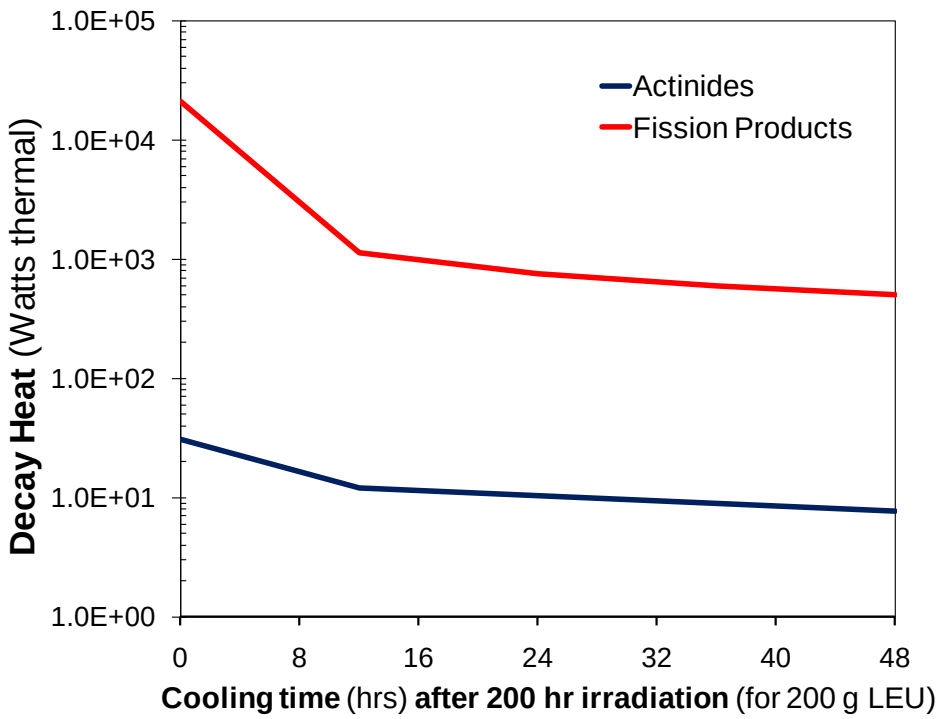

FIGURE 2-9 Cumulative Decay Heat for Fission Products and Actinides for 200 g of LEU after 200-hr Irradiation

3 NITRIC-ACID-DISSOLUTION FRONT-END OPTION

3.1 DISSOLVER SYSTEM

The nitric-acid process involves dissolving the LEU foil and nickel fission-recoil barrier (once Al cladding is removed) in a non-pressurized, steel dissolver at elevated temperature. The dissolver is designed to operate at pressures less than 2 atm (absolute) and at temperatures less than 115°C. The dissolver system was designed to process up to 250 g of irradiated LEU foil. The conceptual design of the dissolver is shown in Figure 3-1.

The dissolver system consists of a 304 stainless-steel vessel (approximately 2-L volume) connected to an approximately 65-L (30 cm × 90 cm) off-gas reservoir. The dissolver vessel is open to the off-gas reservoir during the dissolution process. The volume of the reservoir was chosen to provide passive containment of all gas reaction products at a pressure less than 2 atm (absolute). The reservoir is designed to contain reaction-product gases, at low pressure, during both normal and off-normal (loss of cooling during reaction) conditions. In order to keep the temperature of the gas within the reservoir to below 100°C during a potential loss of cooling, it is wrapped with an aluminum heat sink (Al-fin rings).

The dissolution process is started by first lowering the uranium foil (contained within a steel mesh basket into the dissolver vessel and then sealing the vessel with a metal cap. Pre-heated acid (~100°C) is then added to the vessel using an acid feed system that is designed to avoid pressurization of the acid bottle in the event that the dissolution reaction begins instantaneously when the acid addition step is started.

The dissolver vessel is cooled by forced air blown from the base of the unit. The temperature of the dissolver solution is monitored by a thermocouple. The dissolver vessel is insulated so that the top of the vessel is cooled continuously during the reaction. Heat loss from the top of the vessel is optimized by the presence of aluminum cooling fins. This design causes the water vapor to condense along the walls at the top of the vessel during the dissolution reaction (as acid is boiling). This process is shown schematically in Figure 3-2.

In addition to the heat produced during the exothermic uranium-metal-dissolution reaction, the dissolver cooling system was designed to dissipate up to 1500 W of continuous thermal power resulting from the decay heat of the irradiated LEU foils. The thermal output for all actinides and fission products present in the irradiated foil was calculated as part of the ORIGEN runs discussed above (Section 2; see Tables 2-11 and 2-12 and Figure 2-9).

Following dissolution of the LEU foils, any iodine remaining in the product will be volatilized using a standard chemical technique (e.g., adding hydrogen peroxide at elevated temperature: see Cathers and Shipman 1975). The product solution is then drained from the base of the vessel and run through a titania column for molybdenum recovery. The product solution from the dissolver contains uranium, nickel, and non-volatile fission products in a 1-M nitric acid solution.

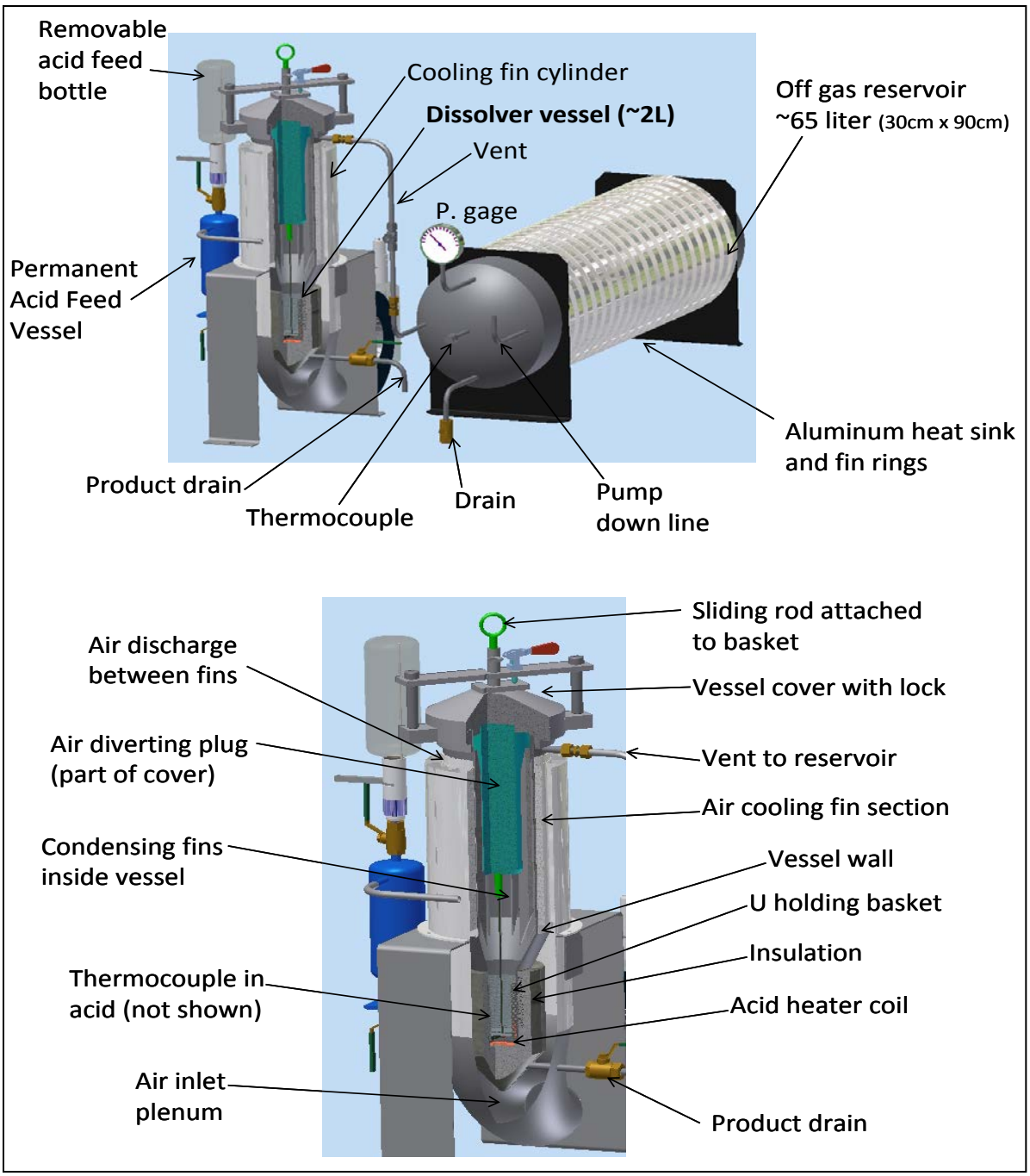

FIGURE 3-1 Conceptual Rendering of the Nitric-Acid Dissolver System

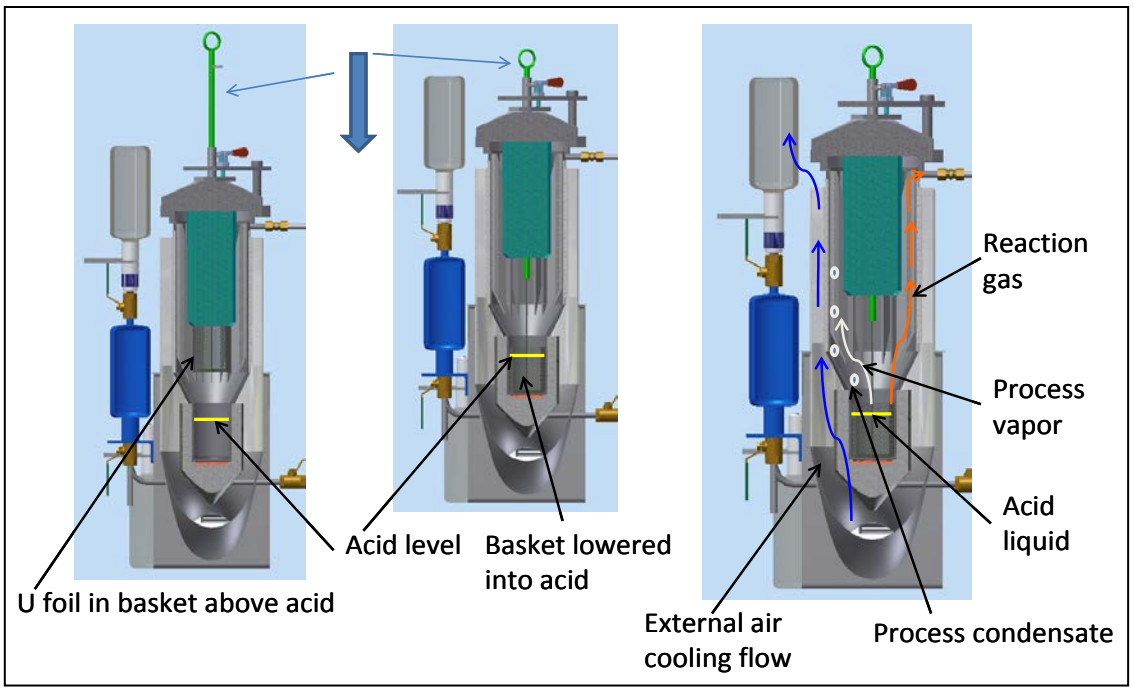

FIGURE 3-2 Conceptual Rendering of the Nitric-Acid Dissolver System Showing the Steps Involved in Starting the Dissolution and Gas Flows during Operation

3.2 MOLYBDENUM RECOVERY USING TITANIA COLUMNS

For recovery of Mo from 200 g of LEU, preliminary column designs are shown in Table 3-1. A method of non-constant pattern mass transfer zone (NCP-MTZ) analysis was used to calculate the preliminary process-scale column length and inner diameter for a titania S80 sorbent (ZirChrom Separations, Inc., Anoka, Minnesota) at different velocities using VERSE (VErsatile Reaction and SEparation) simulation software. Batch and pulse and frontal column tests were conducted for the sorbent to estimate the sorbent parameters and effective isotherm and mass transfer parameters in UNH solution. The effective Mo isotherm was estimated in 450 gU-/L uranyl-nitrate 1-M HNO_3 solution, using batch tests. Capacity, q_{Mo}, obtained from batch and breakthrough experiments indicates that q_{Mo} at 7 mM Mo calculated from column breakthrough curves is 0.31 mmol/g versus q_{Mo} 0.39 mmol/g predicted from batch studies. The results are not unexpected under the non-equilibrium conditions of the Mo-recovery column. The nonlinear isotherm parameter was modified with the prediction of breakthrough experiments. Mo intraparticle diffusivity, D_p, 4.4×10^{-5} cm^2/min, was estimated from the Mo breakthrough curve data in 145 g/L UNH solution at a pH of 1. Other mass transfer parameters, axial dispersion, E_b, mass transfer coefficient, k_f, and Mo Brownian diffusivity, D_∞, were estimated from the literature, and VERSE was used to calculate MTZ. Breakthrough curves were run at linear velocities of 5 and 9 cm/min using a solution containing 450 gU/L, 7 mM Mo, and 1-M HNO_3 to confirm model predictions. Loaded columns were stripped with 30 bed volumes of 0.1-M NaOH solution at 3 cm/min and 100 ± 5% Mo recovery was achieved.

TABLE 3-1 Preliminary Design of Chromatography Column for Separation of Mo from Target Solution

	Loading Time ~ 15 min								
Sorbent	ID (cm)	$u_{superficial}$ (cm/min)	L_{min} (cm)	ΔP (atm)	CV_{min} (mL)	$W_{sorbent}$ (g)	Flow Rate (cm^3/min)	Time (min)	Cost[a] $
S80	3	5	4	0.1	28	37	35.3	14.1	77
	2.5	9	6	0.2	29	38	44.2	11.3	80
	2	15	8	0.4	25	33	47.1	10.6	68
	1.5	21	10	0.8	18	23	37.1	13.5	48

[a] Sachtopore-NP 80µm, 60 Å; $2.09/g (Zirchrom, Anoka, Minnesota).

The flow sheet for the nitric-acid-dissolution front-end process is shown in Figure 3-3. This flowsheet identifies the waste streams (indicated in red) generated during processing.

3.3 WASTE STREAMS GENERATED

As shown in Figure 3-3 and Table 3-2, the primary waste streams produced by the nitric-acid-dissolution front-end process are (1) activated aluminum from target cladding; (2) spent NaOH gel/solution used for neutralization of acid gas and NO_x off-gas from dissolver; (3) spent copper-based iodine trap; (4) fission gases: I, Xe, and Kr; (5) eluent and wash solutions from the titania column containing uranium, nickel, and fission products; and (6) spent titania from the

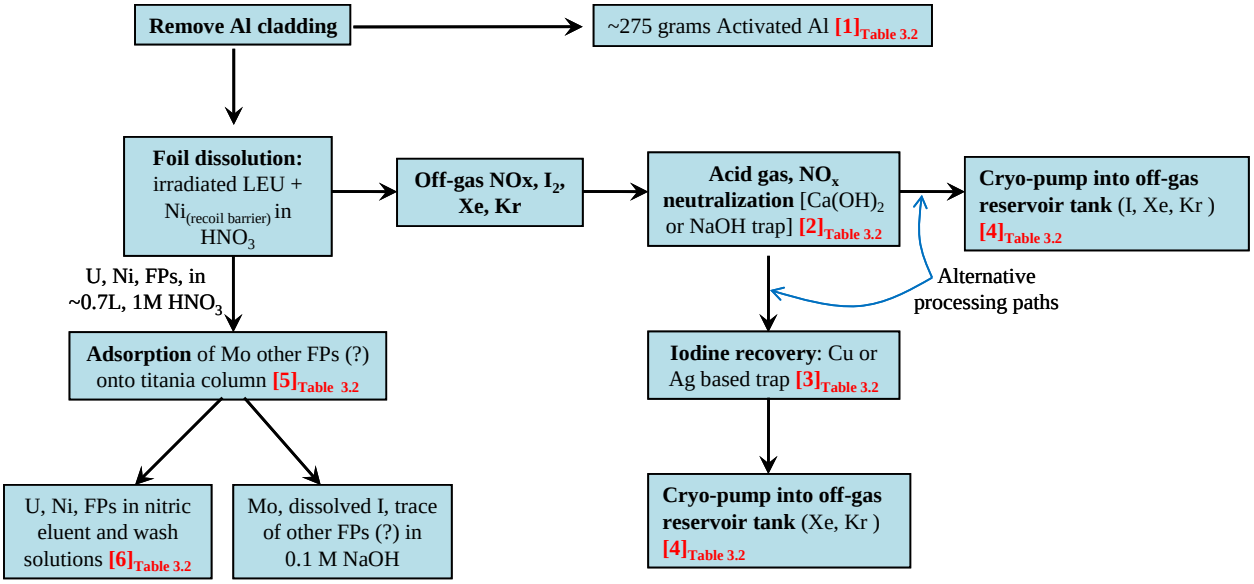

FIGURE 3-3 Processes and Waste Streams for the Nitric-Acid-Based Front-End Process

TABLE 3-2 Waste Stream Chemical Composition for the Nitric-Acid Front-End Process for 200 g of Irradiated LEU Foil

Process Step	Stream Number	Stream Description	Chemical Composition
Decladding	1	Al tubing	275 g Al with activation products (Table 3-3)
Neutralization NO_x with NaOH gel/solution	2	$NaNO_2$ and $NaNO_3$ solids produced by reaction of NO_2(g) with NaOH	70 g $NaNO_2$ and 80 g $NaNO_3$ solids + excess NaOH
Trap and recovery of Iodine isotopes on Cu_{metal}	3	CuI solid produced by reaction of I_2 and HI with Cu_{metal}	1–10 g of Cu_{metal} potentially contaminated with trace I
Unprocessed fission gases	4	Gaseous stream	I_2 and/or all Xe and Kr isotopes (Tables 2-9 and 2-10 and Figures 2-5, 2-6, 2-7, and 2-8)
Spent titania from molybdenum separation column	5	TiO_2 solids	~30 g TiO_2
Eluent and wash solutions from titania column containing U + FP + Ni	6	Nitric acid and water wash solutions containing U + FP + Ni	400 mL eluent (Table 3-4), 300 mL of 0.1-M HNO_3 column wash solution and 150 mL of water, for a total of 850 mL

molybdenum separation column. The product stream is the strip solution from the column; it will be a 0.1-M NaOH solution containing the Mo and small amounts of other fission products. The physical characteristics and the chemical composition of each of these waste and product streams are discussed in detail in the following sections.

3.4 WASTE STREAM 1: ACTIVATED ALUMINUM CLADDING WASTE STREAM

For this exercise, we assumed that the 200 g of LEU would be contained in nine targets of the same dimensions of the current IRE dispersion target. Therefore, each target would contain 22.2 g of LEU and the cladding would be Al 3003, the same cladding Argonne has used to fabricate targets for Indonesia and Argentina. Each target would have 30.5 g of Al-3003 cladding that would be removed before foil dissolution. In addition, the target would contain a 40-µ Al (electrolytic dissolution) or 15-µ Ni (nitric-acid dissolution) fission-recoil barrier. The target is disassembled by cutting off both ends of the target, then making a longitudinal cut. After making the cuts, the outer tube is pulled away from the inner tube, and the LEU foil with the fission-recoil barrier attached is removed from the target and further processed. The cladding is disposed of as low-level waste. The mass and Ci composition of this waste is shown in Tables 3-3 and 3-4.

TABLE 3-3 Waste-Stream Isotopic Composition for Cladding Waste from Nine Irradiated Targets Containing 200 g of LEU, in Grams

Isotope	Mass (g)	Fraction (%)
Al-27	266.4	96.89
Mn-55	4.1	1.50
Fe-56	1.8	0.65
Si-28	1.5	0.56
Cu-63	0.38	0.14
Cu-65	0.17	0.06
Zn-64	0.13	0.05
Fe-54	0.11	0.04
Zn-66	0.08	0.03
Si-29	0.08	0.03
Zn-68	0.05	0.02
Si-30	0.05	0.02
Fe-57	0.04	0.02
Total	275	100

TABLE 3-4 Waste-Stream Isotopic Composition for Cladding Waste from Nine Irradiated Targets Containing 200 g of LEU, in Curies

		Activity (Ci)					Fraction of Total Activity at 12 hr
Isotope	Half-life (s)	EOB	12 hr	24 hr	36 hr	48 hr	(%)
Mn-56	9.3E+03	6.8E+02	2.7E+01	1.1E+00	4.3E-02	1.7E-03	71.4
Cu-64	4.6E+04	1.8E+01	9.5E+00	4.9E+00	2.6E+00	1.3E+00	24.9
Na-24	5.4E+04	2.2E+00	1.3E+00	7.4E-01	4.2E-01	2.4E-01	3.4
Zn-65	2.1E+07	2.5E-02	2.5E-02	2.5E-02	2.5E-02	2.5E-02	0.065
Zn-69	3.4E+03	5.3E-01	2.2E-02	1.2E-02	6.6E-03	3.6E-03	0.059
Zn-69m	5.0E+04	3.8E-02	2.1E-02	1.1E-02	6.2E-03	3.4E-03	0.054
Fe-55	8.6E+07	1.8E-02	1.8E-02	1.8E-02	1.8E-02	1.8E-02	0.048
Fe-59	3.8E+06	9.0E-03	9.0E-03	8.9E-03	8.8E-03	8.8E-03	0.024
Si-31	9.4E+03	1.3E-01	5.3E-03	2.2E-04	9.2E-06	3.9E-07	0.014
Total		7.0E+02	3.8E+01	6.8E+00	3.1E+00	1.6E+00	100

3.5 WASTE STREAM 2: SPENT ACID GAS AND NITROGEN OXIDE OFF-GAS TRAP

The dissolution of the irradiated LEU foil in nitric acid can be represented by the following general reaction:

$$U_{metal} + 4HNO_3 \rightarrow UO_2(NO_3)_2(aq) + 2H_2O + 2NO(g) \quad (1)$$

Therefore, for every mole of uranium dissolved, 4 moles of HNO_3 are consumed and 2 moles of NO(g) are produced. The exact speciation of the nitrogen oxide gases produced will depend on chemical conditions during dissolution. For example, in the presence of oxygen N(II) is converted to an N(IV) species:

$$2NO(g) + O_2 \leftrightarrow 2NO_2(g) \quad (2)$$

Using the kinetic rate law presented by Chilton (1968), one finds that the rate of reaction (2) is on the order of milliseconds to seconds, even at relatively low O_2 partial pressures (0.1–0.001 atm). When water vapor and oxygen are present, NO_2 can be converted to both nitrous and nitric acid vapors [HNO_2(g) and HNO_3(g)]. The relative amounts of the important nitrogen oxide gases produced by the dissolution of different amounts of LEU were calculated using the thermodynamic code OLI-ESP (Table 3-5).

TABLE 3-5 Thermodynamic Modeling Results Simulating the Off-Gas Composition for the Dissolution of Uranium Metal after the Dissolver Has Cooled to a Temperature of 25°C (Calculations were done in the absence of oxygen. Under oxygenated conditions, NO_2 rather than NO will be the dominant species due to the relatively fast kinetics of Reaction [2].)

U metal (g)	H_2O (moles)	NO (moles)	NO_2 (moles)	HNO_3 (moles)	HNO_2 (moles)	Total (moles)
5	1.2E-03	0.0404	3.5E-05	3E-08	2.2E-04	0.0418
10	2.3E-03	0.0816	1.4E-04	2E-07	6.0E-04	0.0847
20	3.8E-03	0.1632	9.8E-04	2E-06	2.0E-03	0.17
25	4.7E-03	0.2041	1.2E-03	2E-06	2.5E-03	0.2125
50	9.4E-03	0.4083	2.4E-03	5E-06	5.0E-03	0.4252
75	0.0141	0.6124	3.7E-03	7E-06	7.6E-03	0.6377
100	0.0188	0.8164	4.9E-03	9E-06	0.0101	0.8502
125	0.0235	1.0205	6.1E-03	1E-05	0.0126	1.0628
150	0.0283	1.2248	7.3E-03	1E-05	0.0151	1.2755
175	0.033	1.4288	8.5E-03	2E-05	0.0176	1.488
200	0.0377	1.6329	9.8E-03	2E-05	0.0201	1.7005
225	0.0424	1.8372	0.011	2E-05	0.0227	1.9132
250	0.0471	2.0412	0.0122	2E-05	0.0252	2.1257

It should be noted that the results shown in Table 3-2 do not account for the presence of oxygen. Under oxygenated conditions, NO_2 rather than NO will be the dominant species due to the relatively fast kinetics of reaction (2); however, this does not affect the NO_x off-gas totals.

As indicated in Figure 3-6, the nitrogen oxide gases are neutralized using a hydroxide NO_x trap or scrubber (e.g., NaOH). The conversion can be described by the following general reactions:

$$NO_2(g) + NaOH + 0.25O_2(aq) \leftrightarrow NaNO_3 + 0.5H_2O \quad (3)$$

$$NO_2(g) + NaOH \leftrightarrow NaNO_2 + 0.5H_2O + 0.25O_2(aq) \quad (4)$$

which combine to produce

$$NO_2(g) + NaOH \leftrightarrow 0.5NaNO_2 + 0.5NaNO_3 + 0.5H_2O \quad (5)$$

Assuming that 2 moles of nitrogen oxide gas need to be neutralized (this corresponds to just over 200 g of U_{metal} dissolved; Table 3-3), 2 moles of NaOH will be needed. This corresponds to approximately 80 g of NaOH. Therefore, for every 200 g of LEU dissolved, a waste stream of approximately 70 g $NaNO_2$ + 80 g $NaNO_3$ will be produced (waste stream 2 in Table 3-1).

The fission product noble gases (Xe, Kr) are not anticipated to not be retained within the hydroxide NO_x scrubber; however, trace amounts of iodine species may be present within the spent hydroxide trap.

3.6 WASTE STREAM 3: SPENT IODINE TRAP MATERIAL AND FISSION GAS STREAM

The iodine fission gas produced by LEU dissolution can be selectively removed from the other fission gases using a copper-based trap or scrubber (Figure 3-3). The following is a generalized reaction for how such an iodine trap would work:

$$Cu + 0.5I_2 \leftrightarrow CuI \quad (6)$$

This simplistic reaction can be used to estimate the mass of copper that would be required to trap the amount of iodine produced during the LEU foil dissolution process and thus provide information on the mass of the copper waste stream that would be generated by this process. The maximum amount of $I_2(g)$ that will be processed in the iodine trap is around 4.7×10^{-4} moles for a 12-hr cooling period and 3.6×10^{-4} moles for a 48-hr cooling period (Table 3-6). Therefore, the amount of copper that will be reacted in the iodine trap is less than 1 g. Because this is a surface reaction, more than a stoichiometric amount of Cu will be required in the trap. As a conservative estimate, 10 g of copper metal is assumed for the iodine trap for each 200 g of irradiated LEU (waste stream 3, Table 3-2). The iodine can be recovered from the copper trap by reacting the

CuI product with H_2 gas at elevated temperatures. It can also be removed by anion exchange using an aqueous solution.

The mass and curie content amounts of the noble fission gases plus iodine are shown in Tables 3-6 and 3-7, as well in Figures 3-4, 3-5, 3-6, and 3-7. After 48 hours of cooling, the key isotopes are Xe-133, Xe-135, Kr-88, and Kr-85. These isotopes dominate waste stream 4 (Table 3-2). It is envisioned that the noble-gas fission products will be stored in gas cylinders until their activities decrease beyond regulatory thresholds.

TABLE 3-6 Iodine Yields in Moles per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1×10^{14} N/cm^2 s); Yields after Five Cooling Times Are Shown

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
I-135	4.1E-05	1.2E-05	3.3E-06	9.3E-07	2.6E-07
I-134	6.5E-06	2.0E-09	1.6E-13	1.2E-17	9.5E-22
I-133	1.4E-04	9.5E-05	6.3E-05	4.3E-05	2.9E-05
I-132	8.1E-06	7.5E-06	6.7E-06	6.1E-06	5.4E-06
I-131	2.8E-04	2.7E-04	2.6E-04	2.5E-04	2.4E-04
I-130	2.8E-08	1.4E-08	7.3E-09	3.7E-09	1.9E-09
I-129	7.8E-05	8.0E-05	8.1E-05	8.1E-05	8.1E-05
I-128	2.6E-11	5.6E-20	1.2E-28	2.5E-37	0.0E+00
I-127	7.4E-06	8.2E-06	8.9E-06	9.5E-06	1.0E-05
Total	5.5E-04	4.7E-04	4.2E-04	3.9E-04	3.6E-04

TABLE 3-7 Iodine Yields in Curies per 200 g of Irradiated LEU (200-hr irradiation at a flux of 2.1×10^{14} N/cm^2 s); Yields after Five Cooling Times Are Shown

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
I-135	1.9E+04	5.5E+03	1.6E+03	4.4E+02	1.3E+02
I-134	2.3E+04	7.3E+00	5.9E-04	4.5E-08	3.4E-12
I-133	2.1E+04	1.4E+04	9.6E+03	6.4E+03	4.3E+03
I-132	1.1E+04	1.0E+04	9.2E+03	8.3E+03	7.4E+03
I-131	4.5E+03	4.4E+03	4.2E+03	4.0E+03	3.9E+03
I-130	7.1E+00	3.6E+00	1.9E+00	9.5E-01	4.8E-01
I-129	1.8E-06	1.8E-06	1.8E-06	1.8E-06	1.8E-06
I-128	2.0E-01	4.2E-10	8.8E-19	1.9E-27	0.0E+00
Total	7.9E+04	3.4E+04	2.4E+04	1.9E+04	1.6E+04

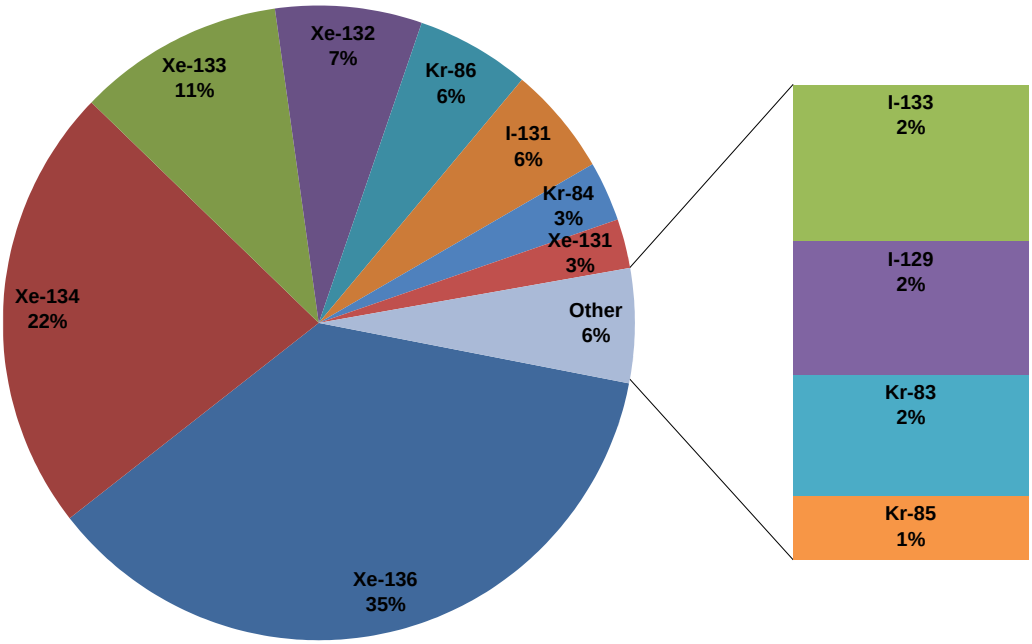

FIGURE 3-4 Mole Percent of Fission Gases after 12 hr of Cooling for 200 g of LEU after 200-hr Irradiation

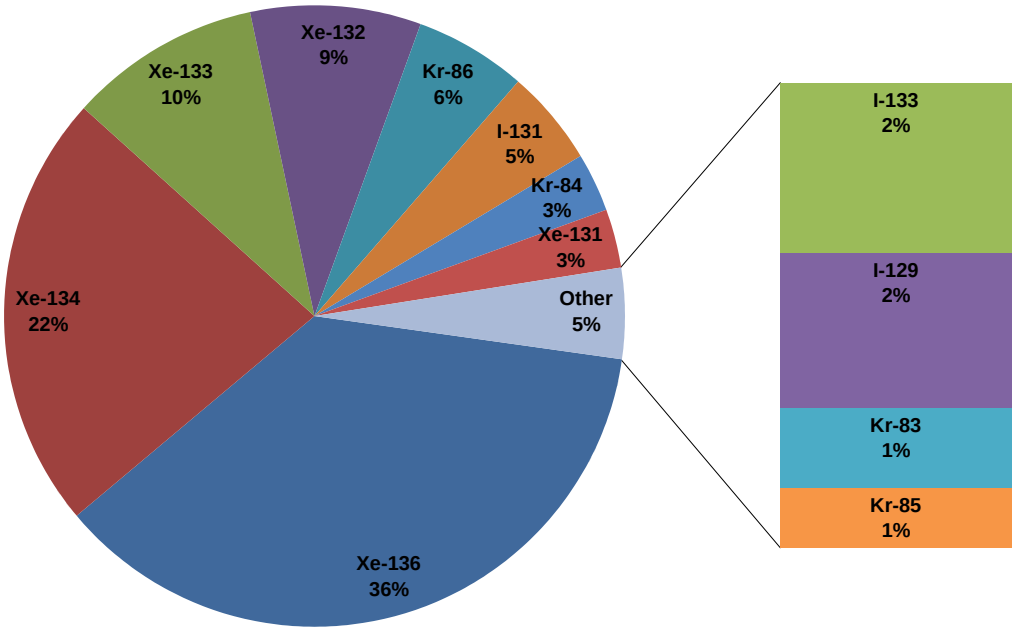

FIGURE 3-5 Mole Percent of Fission Gases after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation

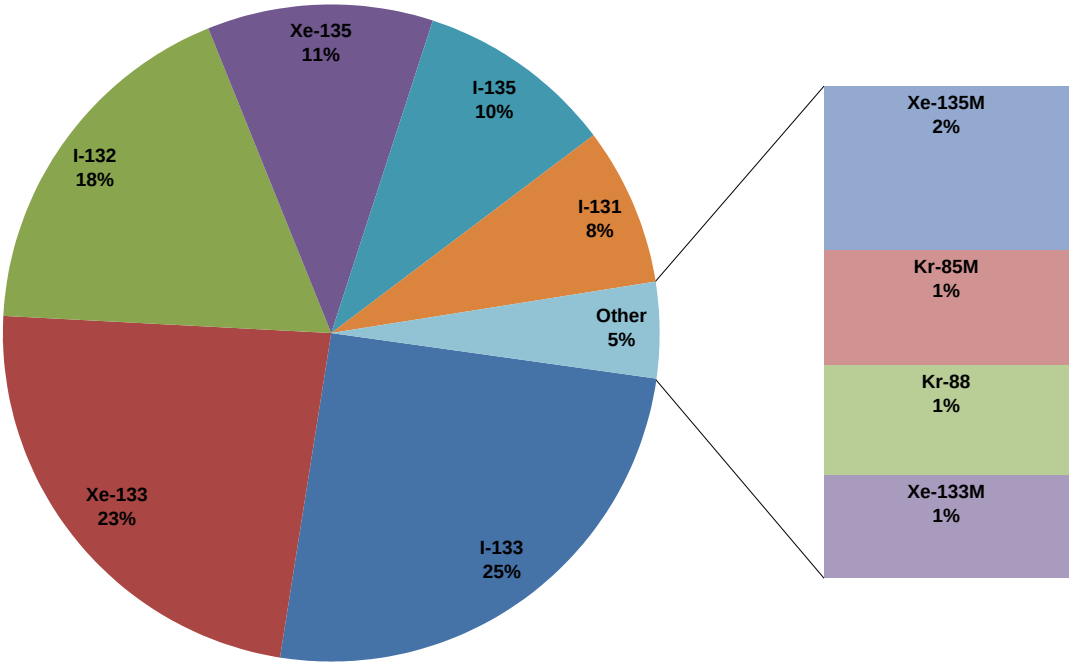

FIGURE 3-6 Percent of Fission Gases (based on curie content) after 12 hr of Cooling for 200 g of LEU after 200-hr Irradiation

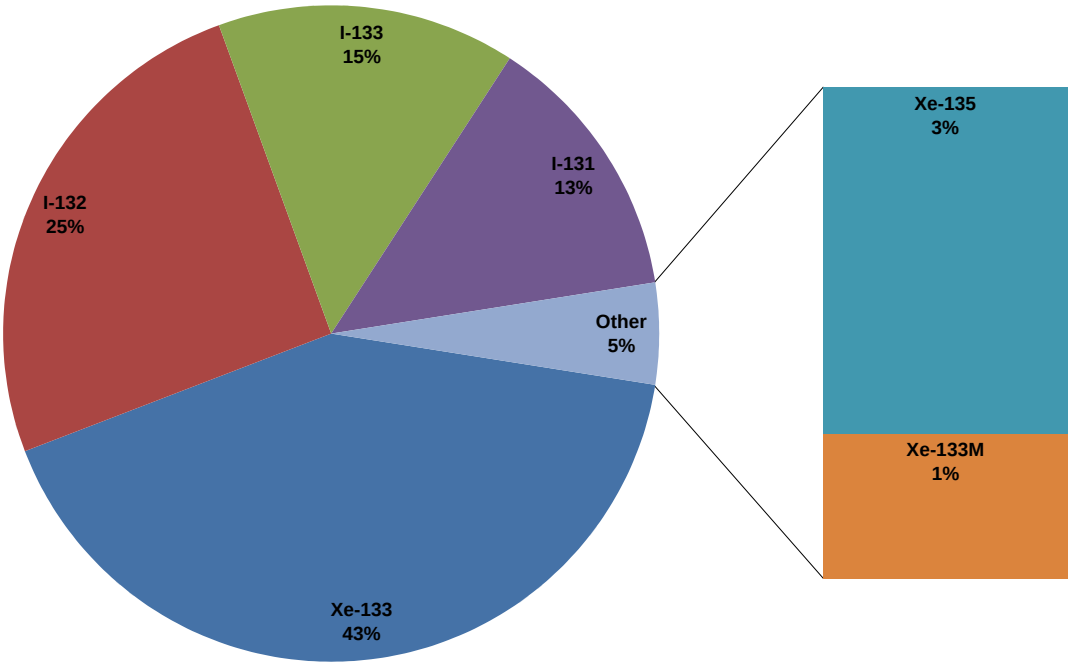

FIGURE 3-7 Percent of Fission Gases (based on curie content) after 48 hr of Cooling for 200 g of LEU after 200-hr Irradiation

3.7 WASTE STREAM OF TITANIA COLUMN

The partitioning of fission products and actinides among the sorbent waste stream (stream 5), eluent and wash waste streams (stream 6), and the Mo-product stream was predicted for elements that are either ≥0.01% of Mo activity or ≥1% of Mo mass (Table 3-2). The sorbent waste stream is predicted to contain approximately 30 g of TiO_2, as well as Zr as it forms colloids that are suspended on the column. Waste stream 6 is expected to contain 400 mL of the Mo-depleted feed, 300 mL of 0.1-M HNO_3 column wash solution, and 150 mL of water wash, for a total of 850 mL. Waste stream 6 is expected to contain all of the actinides, lanthanides, and alkali and alkaline earth metal ions. It will also contain approximately 8 g of Ni from the fission-recoil barrier dissolved with the LEU foil (assuming 0.04 g Ni per gram of LEU). The Mo-product stream is expected to contain 900 mL of 0.1-M NaOH and a fraction of the fission products known to have chemistries somewhat similar to Mo, specifically elements known to be present as oxoions or anions in acidic solution. Therefore, the Mo product stream is expected to contain a small fraction of As, Se, Nb, Tc, Ru, Rh, Sn, Sb, and Te, as well as the iodine not volatilized in the dissolution; the fraction in the Mo product is assumed to be 1%. Partitioning of halides and oxoions has not yet been measured for the expected feed composition. In 0.1-M HNO_3, their K_d values tend to be quite a bit lower than that of Mo; therefore, it is likely that a significant fraction of As, Se, Nb, Tc, Ru, Rh, Sn, Sb, Te, Br, and the remaining I will be found in waste stream 6 rather than the product stream. On the other hand, if these oxoions strongly compete with Mo for adsorption, column size may need to be increased by up to 100% to ensure complete capture of Mo. Table 3-8 summarizes the partitioning of irradiated foil components in the various product and waste streams.

TABLE 3-8 Partitioning of Elements from 200 g Irradiated Target between Product and Waste Streams Following a 200-hr Irradiation and 12-hr Cooling Period before Processing

	Feed Composition		Sorbent Waste Stream (5) Table 3-2		Eluent and 0.1 M HNO_3 Wash Waste Stream (6) Table 3-2		Product Stream, 0.1 M NaOH	
Fission Products	Ci/200 g Target	g/200 g Target	Ci/200 g Target	g/200 g Target	Ci/200 g Target	g/200 g Target	Ci/200 g Target	g/200 g Target
Ge	4.48E+00	4.45E-05			4.43E+00	4.40E-05	4.48E-02	4.45E-07
As	2.20E+01	3.17E-05			2.18E+01	3.14E-05	2.20E-01	3.17E-07
Se	6.08E-02	6.05E-03			6.02E-02	5.99E-03	6.08E-04	6.05E-05
Br	5.37E+01	2.40E-03			5.32E+01	2.37E-03	5.37E-01	2.40E-05
Rb	6.58E+02	4.37E-02			6.58E+02	4.37E-02		
Sr	9.95E+03	1.78E-01			9.95E+03	1.78E-01		
Y	1.98E+04	7.65E-02			1.98E+04	7.65E-02		
Zr	1.26E+04	4.19E-01	1.26E+04	4.19E-01				
Nb	2.14E+04	4.44E-03			2.12E+04	4.39E-03	2.14E+02	4.44E-05
Mo	1.47E+04	2.74E-01					1.47E+04	2.74E-01
Tc	1.37E+04	5.59E-02			1.36E+04	5.53E-02	1.37E+02	5.59E-04
Ru	1.87E+03	2.07E-01			1.85E+03	2.05E-01	1.87E+01	2.07E-03
Rh	3.17E+03	5.72E-03			3.13E+03	5.66E-03	3.17E+01	5.72E-05
Pd	7.84E+01	1.70E-02			7.84E+01	1.70E-02	7.84E-01	1.70E-04
Ag	1.22E+02	6.54E-04			1.22E+02	6.54E-04	1.22E+00	6.54E-06
Cd	2.55E+01	9.04E-04			2.55E+01	9.04E-04	2.55E-01	9.04E-06
In	3.07E+01	1.06E-04			3.07E+01	1.06E-04	3.07E-01	1.06E-06
Sn	5.31E+01	2.66E-03			5.31E+01	2.66E-03	5.31E-01	2.66E-05
Sb	5.96E+02	1.92E-03			5.96E+02	1.92E-03	5.96E+00	1.92E-05
Te	1.16E+04	6.83E-02			1.16E+04	6.83E-02	1.16E+02	6.83E-04
I	3.43E+04	6.16E-02			3.43E+04	6.16E-02	3.43E+02	6.16E-04
Cs	1.66E+01	1.67E-01			1.66E+01	1.67E-01		
Ba	7.07E+03	2.29E-01			7.07E+03	2.29E-01		
La	8.21E+03	1.38E-01			8.21E+03	1.38E-01		
Ce	1.75E+04	3.72E-01			1.75E+04	3.72E-01		
Pr	8.82E+03	9.29E-02			8.82E+03	9.29E-02		
Nd	2.86E+03	2.43E-01			2.86E+03	2.43E-01		

TABLE 3-8 (Cont.)

Fission Products	Feed Composition Ci/200 g Target	Feed Composition g/200 g Target	Sorbent Waste Stream (5) Table 3-2 Ci/200 g Target	Sorbent Waste Stream (5) Table 3-2 g/200 g Target	Eluent and 0.1 M HNO_3 Wash Waste Stream (6) Table 3-2 Ci/200 g Target	Eluent and 0.1 M HNO_3 Wash Waste Stream (6) Table 3-2 g/200 g Target	Product Stream, 0.1 M NaOH Ci/200 g Target	Product Stream, 0.1 M NaOH g/200 g Target
Pm	3.57E+03	1.94E-02			3.57E+03	1.94E-02		
Sm	4.44E+02	3.24E-02			4.44E+02	3.24E-02		
Eu	3.37E+01	3.61E-03			3.37E+01	3.61E-03		
Gd	2.11E+00	2.98E-04			2.11E+00	2.98E-04		
Total	2.15E+05	3.29E+00	1.26E+04	4.19E-01	1.66E+05	2.03E+00	1.55E+04	2.79E-01
Actinides								
Np	4.92E+03	2.13E-02			4.92E+03	2.13E-02		
U	1.32E+01	1.97E+02			1.32E+01	1.97E+02		
Pu	3.73E-03	4.28E-02			3.73E-03	4.28E-02		
Total	4.94E+03	1.97E+02	0	0	4.94E+03	1.97E+02		
Total Fission Products and Actinides	2.20E+05	2.00E+02	1.26E+04	4.19E-01	1.71E+05	1.99E+02	1.55E+04	2.79E-01

4 ELECTROCHEMICAL LEU DISSOLUTION IN BICARBONATE

The schematic of the electrochemical-dissolution process is presented in Figure 4-1. The process is designed for the irradiation of nine LEU targets with a total uranium weight of 200 g. The amount of Al is estimated to be 30.5 g per target, 275 g total. De-cladded Al tubes contain activated ^{28}Al ($T_{1/2}$ = 2.25 min) and some activated impurities, which depend on the Al alloy used for the target fabrication. The Al alloy's composition is reported in Tables 3-3 and 3-4.

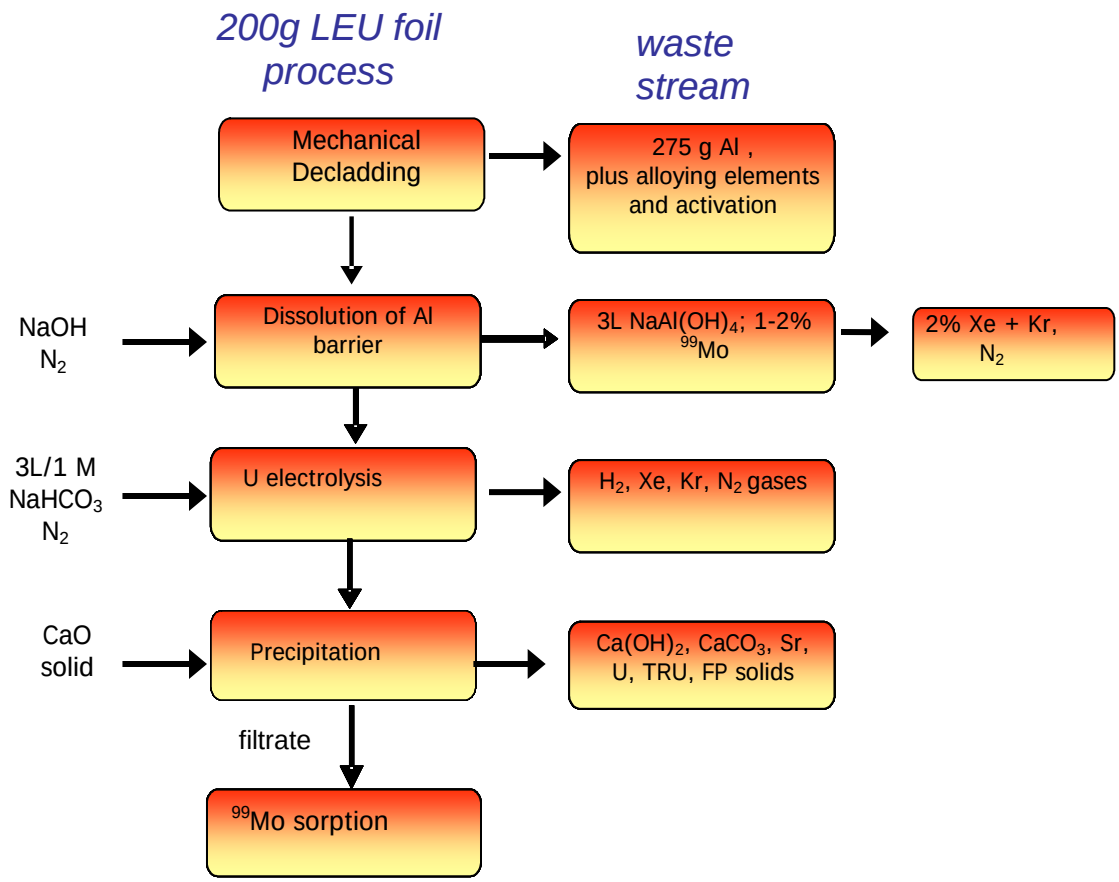

FIGURE 4-1 Product and Waste Streams for the Electrochemical Front-End Process

After mechanical decladding, the Al fission-recoil barrier is dissolved in 3 L of 0.5-M NaOH. Dissolution of the fission-recoil barrier will release approximately 2% of the fission products formed during irradiation; it is likely to contain a fine precipitate of alkaline-insoluble elements. Nitrogen is used as a sweep gas. After draining the spent NaOH solution from the dissolver, 3 L of 1-M $NaHCO_3$ is added, and the U foil is anodically dissolved. Nitrogen is used again to remove xenon and krypton radioactive gases, as well as hydrogen forming on the cathode.

The solution is then drained from the dissolver into a precipitation vessel. By adding CaO to the solution, all the uranium, transuranics, and most of the fission products are precipitated in the vessel. The amount of CaO needed for a complete precipitation is about 200 g, making the weight of the cake around 460 g. The pH of the filtrate is around 12.5, and it is set by $Ca(OH)_2$, soluble of about 1 mM. We assume that Rh, Ru, and most of the transition metals and rare earths completely precipitate at this step. The filtrate contains Mo, I, Cs, Sn, Sb, Te, Tl, and some other base-soluble fission products. The filtrate can be either acidified (a low volume of HNO_3 will be required), or it can be fed into an anion exchange column (e.g., AG-1) to recover ^{99}Mo. The chemical compositions and quantitative parameters of each stream are given in Table 1-1; the radionuclide compositions are given in Tables 4-2 through 4-6.

TABLE 4-1 Waste/Product Stream Chemical Composition

Process Step	Waste/Product Stream No.	Waste Stream Description	Chemical Composition
Decladding	1[a]	Al tubing	275 g of Al and alloying elements
Dissolution of Al barrier	2	Alkaline solution	3L of 0.5-M NaOH, 0.25-M $NaAl(OH)_4$, and 2% of all fission products
Dissolution of Al barrier	3	Gaseous stream	N_2, 2% of total Xe and Kr
Uranium electrolysis	4	Gaseous stream	N_2, H_2, 98% of total Xe and Kr
U precipitation/filtration	5	Solid carbonates	~460 g $Ca(OH)_2$, $CaCO_3$, and hydrated oxides of U, TRU, FP
Filtration	6	Mo product	~3 L of 1-mM $Ca(OH)_2$, pH 12.5, Mo, I, Sn, Sb, Te, Tl

[a] For a description of waste stream 1 see Tables 3-3 and 3-4.

TABLE 4-2 Partitioning of Elements from 200-g Irradiated Target between Product and Waste Streams Following a 200-hr Irradiation and 12-hr Cooling Period before Processing

	Alkaline Solution, Stream (2) Table 4-1		Solid Caronates Stream (5) Table 4-1		Product Stream (6) Table 4-1	
Fission Products	Ci/200 g Target	g/200 g Target	Ci/200 g Target	g/200 g Target	Ci/200 g Target	g/200 g Target
Ge	9.0E-02	8.9E-07	4.4E+00	4.4E-05		
As	4.4E-01	6.3E-07			2.2E+01	3.1E-05
Se	1.2E-03	1.2E-04			6.0E-02	5.9E-03
Br	1.1E+00	4.8E-05			5.3E+01	2.3E-03
Rb	1.3E+01	8.7E-04	6.4E+02	4.3E-02		
Sr	2.0E+02	3.6E-03	9.8E+03	1.7E-01		
Y	4.0E+02	1.5E-03	1.9E+04	7.5E-02		
Zr	2.5E+02	8.4E-03	1.2E+04	4.1E-01		
Nb	4.3E+02	8.9E-05	2.1E+04	4.3E-03		
Mo	2.9E+02	5.5E-03			1.4E+04	2.7E-01
Tc	2.7E+02	1.1E-03			1.3E+04	5.5E-02
Ru	3.7E+01	4.1E-03			1.8E+03	2.0E-01
Rh	6.3E+01	1.1E-04			3.1E+03	5.6E-03
Pd	1.6E+00	3.4E-04			7.7E+01	1.7E-02
Ag	2.4E+00	1.3E-05			1.2E+02	6.4E-04
Cd	5.1E-01	1.8E-05	2.5E+01	8.9E-04		
In	6.1E-01	2.1E-06	3.0E+01	1.0E-04		
Sn	1.1E+00	5.3E-05	5.2E+01	2.6E-03		
Sb	1.2E+01	3.8E-05			5.8E+02	1.9E-03
Te	2.3E+02	1.4E-03			1.1E+04	6.7E-02
I	6.9E+02	1.2E-03			3.4E+04	6.0E-02
Cs	3.3E-01	3.3E-03	1.6E+01	1.6E-01		
Ba	1.4E+02	4.6E-03	6.9E+03	2.2E-01		
La	1.6E+02	2.8E-03	8.0E+03	1.3E-01		
Ce	3.5E+02	7.4E-03	1.7E+04	3.6E-01		
Pr	1.8E+02	1.9E-03	8.6E+03	9.1E-02		
Nd	5.7E+01	4.9E-03	2.8E+03	2.4E-01		
Pm	7.1E+01	3.9E-04	3.5E+03	1.9E-02		
Sm	8.9E+00	6.5E-04	4.3E+02	3.2E-02		
Eu	6.7E-01	7.2E-05	3.3E+01	3.5E-03		
Gd	4.2E-02	6.0E-06	2.1E+00	2.9E-04		
Total	3.9E+03	5.4E-02	1.1E+05	2.0E+00	7.9E+04	6.9E-01
Actinides						
U			1.3E+01	2.0E+02		
Np			3.7E-03	4.3E-02		
Pu			4.9E+03	2.1E-02		
Total			4.9E+03	2.0E+02		
Total Fission Products and Actinides	7.7E+03	1.1E-01	2.3E+05	4.0E+02	1.6E+05	1.4E+00

TABLE 4-3 Selected[a] Fission Products (curies) in Waste Stream 1 (Table 4-1)

Isotope	0 hr	12 hr	24 hr	36 hr	48 hr
As-77	4.8E-01	4.2E-01	3.4E-01	2.8E-01	2.4E-01
Ge-77	1.8E-01	8.6E-02	4.0E-02	2.0E-02	9.4E-03
Br-82	1.2E-02	9.2E-03	7.4E-03	5.8E-03	4.6E-03
Y-90	1.3E-01	1.4E-01	1.5E-01	1.6E-01	1.6E-01
Y-93	4.0E+02	1.7E+02	7.6E+01	3.4E+01	1.5E+01
Y-92	3.6E+02	9.4E+01	1.2E+01	1.2E+00	1.2E-01
Y-91M	2.0E+02	9.4E+01	4.0E+01	1.6E+01	6.8E+00
Y-91	3.2E+01	3.4E+01	3.4E+01	3.4E+01	3.4E+01
Zr-97	3.6E+02	2.2E+02	1.3E+02	8.2E+01	5.0E+01
Zr-95	3.6E+01	3.6E+01	3.4E+01	3.4E+01	3.4E+01
Nb-97M	3.4E+02	2.0E+02	1.3E+02	7.8E+01	4.8E+01
Nb-97	3.6E+02	2.2E+02	1.3E+02	8.2E+01	5.0E+01
Nb-96	5.0E-02	3.6E-02	2.4E-02	1.7E-02	1.2E-02
Nb-95M	1.3E-01	1.4E-01	1.5E-01	1.6E-01	1.7E-01
Nb-95	2.8E+00	3.2E+00	3.4E+00	3.8E+00	4.0E+00
Mo-99	3.4E+02	3.0E+02	2.6E+02	2.2E+02	2.0E+02
Tc-99M	3.0E+02	2.8E+02	2.4E+02	2.2E+02	1.9E+02
Ru-106	3.8E-01	3.8E-01	3.8E-01	3.8E-01	3.8E-01
Ru-105	6.2E+01	9.8E+00	1.5E+00	2.4E-01	3.6E-02
Ru-103	2.8E+01	2.8E+01	2.6E+01	2.6E+01	2.6E+01
Rh-106	1.7E+01	3.8E-01	3.8E-01	3.8E-01	3.8E-01
Rh-105M	1.7E+01	2.8E+00	4.2E-01	6.6E-02	1.0E-02
Rh-105	3.8E+01	3.6E+01	3.0E+01	2.4E+01	1.8E+01
Rh-103M	2.4E+01	2.4E+01	2.4E+01	2.4E+01	2.4E+01
Pd-112	7.8E-01	5.2E-01	3.4E-01	2.2E-01	1.5E-01
Pd-109	1.9E+00	1.0E+00	5.6E-01	3.0E-01	1.6E-01
Ag-112	7.8E-01	6.0E-01	4.0E-01	2.6E-01	1.8E-01
Ag-111	6.8E-01	6.4E-01	6.2E-01	6.0E-01	5.6E-01
Ag-109M	1.9E+00	1.0E+00	5.6E-01	3.0E-01	1.6E-01
Cd-115	5.4E-01	4.6E-01	4.0E-01	3.4E-01	3.0E-01
In-115M	5.4E-01	5.0E-01	4.4E-01	3.8E-01	3.2E-01
Sn-125	3.8E-01	3.8E-01	3.6E-01	3.4E-01	3.4E-01
Sn-123	1.2E-02	1.2E-02	1.2E-02	1.2E-02	1.2E-02
Sn-121	7.8E-01	5.8E-01	4.2E-01	3.2E-01	2.2E-01
Sb-129	3.8E+01	5.8E+00	8.4E-01	1.2E-01	1.8E-02
Sb-128	6.8E-01	2.6E-01	1.1E-01	4.2E-02	1.7E-02
Sb-127	6.4E+00	6.0E+00	5.4E+00	5.0E+00	4.6E+00
Sb-126	2.2E-02	2.0E-02	2.0E-02	1.9E-02	1.9E-02
Sb-125	7.2E-03	7.2E-03	7.4E-03	7.4E-03	7.6E-03
Te-132	2.2E+02	2.0E+02	1.8E+02	1.6E+02	1.4E+02
Te-131M	2.2E+01	1.7E+01	1.3E+01	9.8E+00	7.4E+00
Te-131	1.5E+02	3.8E+00	2.8E+00	2.2E+00	1.7E+00
Te-129M	9.2E-01	9.4E-01	9.4E-01	9.2E-01	9.2E-01
Te-129	3.4E+01	7.4E+00	1.6E+00	7.4E-01	6.2E-01
Te-127	5.4E+00	5.4E+00	5.0E+00	4.8E+00	4.4E+00

[a] Selected based on their yield being >0.001 Ci/200g-LEU.

TABLE 4-4 Selected[a] Fission Products (curies) in Waste Stream 2 (Table 4-1)

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Rb-88	2.2E+02	1.3E+01	7.0E-01	3.8E-02	2.0E-03
Sr-91	3.6E+02	1.5E+02	6.2E+01	2.6E+01	1.1E+01
Sr-90	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01
Sr-89	3.4E+01	3.2E+01	3.2E+01	3.2E+01	3.2E+01
I-135	3.8E+02	1.1E+02	3.2E+01	8.8E+00	2.6E+00
I-134	4.6E+02	1.5E-01	1.2E-05	9.0E-10	6.8E-14
I-133	4.2E+02	2.8E+02	1.9E+02	1.3E+02	8.6E+01
I-132	2.2E+02	2.0E+02	1.8E+02	1.7E+02	1.5E+02
I-131	9.0E+01	8.8E+01	8.4E+01	8.0E+01	7.8E+01
I-130	1.4E-01	7.2E-02	3.8E-02	1.9E-02	9.6E-03
I-129	3.6E-08	3.6E-08	3.6E-08	3.6E-08	3.6E-08
I-128	4.0E-03	8.4E-12	1.8E-20	3.8E-29	0.0E+00
Cs-137	2.0E-01	2.0E-01	2.0E-01	2.0E-01	2.0E-01
Cs-136	1.3E-01	1.2E-01	1.2E-01	1.2E-01	1.1E-01
Ba-140	1.4E+02	1.4E+02	1.4E+02	1.3E+02	1.3E+02
Ba-137M	2.0E-01	1.9E-01	1.9E-01	1.9E-01	1.9E-01
Ba-136M	2.0E-02	2.0E-02	2.0E-02	1.9E-02	1.9E-02
Ba-140	1.4E+02	1.4E+02	1.4E+02	1.3E+02	1.3E+02
Ba-137M	2.0E-01	1.9E-01	1.9E-01	1.9E-01	1.9E-01
Ba-136M	2.0E-02	2.0E-02	2.0E-02	1.9E-02	1.9E-02
La-141	3.6E+02	4.6E+01	5.6E+00	6.8E-01	8.2E-02
La-140	1.1E+02	1.2E+02	1.2E+02	1.2E+02	1.2E+02
Ce-144	7.0E+00	7.0E+00	7.0E+00	7.0E+00	7.0E+00
Ce-143	3.6E+02	2.8E+02	2.2E+02	1.7E+02	1.3E+02
Ce-141	6.0E+01	6.0E+01	6.0E+01	6.0E+01	5.8E+01
Pr-145	2.4E+02	6.0E+01	1.5E+01	3.6E+00	9.2E-01
Pr-144M	8.6E-02	8.4E-02	8.4E-02	8.4E-02	8.4E-02
Pr-144	1.0E+01	7.0E+00	7.0E+00	7.0E+00	7.0E+00
Pr-143	1.0E+02	1.1E+02	1.1E+02	1.2E+02	1.2E+02
Pr-142	5.4E-02	3.4E-02	2.2E-02	1.5E-02	9.4E-03
Nd-147	5.8E+01	5.6E+01	5.4E+01	5.4E+01	5.2E+01
Pm-151	2.6E+01	1.9E+01	1.4E+01	1.1E+01	7.8E+00
Pm-149	5.8E+01	5.2E+01	4.4E+01	3.8E+01	3.2E+01
Pm-148M	1.0E-02	1.0E-02	9.8E-03	9.8E-03	9.6E-03
Pm-148	1.3E-01	1.2E-01	1.1E-01	1.1E-01	9.8E-02
Pm-147	1.9E-01	2.2E-01	2.4E-01	2.6E-01	2.8E-01
Sm-156	8.2E-01	3.4E-01	1.4E-01	5.8E-02	2.4E-02
Sm-153	1.0E+01	8.6E+00	7.2E+00	6.0E+00	5.0E+00
Sm-151	1.7E-03	1.9E-03	2.0E-03	2.2E-03	2.2E-03
Eu-157	4.0E-01	2.4E-01	1.3E-01	7.8E-02	4.4E-02
Eu-156	4.4E-01	4.4E-01	4.4E-01	4.2E-01	4.2E-01
Eu-155	5.0E-03	5.0E-03	5.0E-03	5.0E-03	5.0E-03
Gd-159	6.4E-02	4.2E-02	2.8E-02	1.7E-02	1.1E-02
Tb-161	3.2E-03	3.0E-03	3.0E-03	2.8E-03	2.6E-03

[a] Selected based on their yield being >0.001 Ci/200g-LEU.

TABLE 4-5 Nuclide Composition (curies) of Waste Stream 3 (Table 4-1)

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Kr-88	2.2E+02	1.2E+01	6.2E-01	3.4E-02	1.8E-03
Kr-87	1.6E+02	2.2E-01	3.2E-04	4.8E-07	6.8E-10
Kr-85M	7.8E+01	1.2E+01	1.9E+00	3.0E-01	4.6E-02
Kr-85	2.4E-02	2.6E-02	2.6E-02	2.6E-02	2.6E-02
Kr-83M	3.2E+01	3.4E+00	1.3E-01	4.2E-03	1.3E-04
Kr-81	1.3E-14	1.3E-14	1.3E-14	1.3E-14	1.3E-14
Total Kr	4.8E+02	2.8E+01	2.6E+00	3.6E-01	7.4E-02
Xe-138	3.8E+02	1.9E-13	9.8E-29	0.0E+00	0.0E+00
Xe-135M	6.8E+01	1.8E+01	5.0E+00	1.4E+00	4.0E-01
Xe-135	1.5E+01	1.3E+02	8.4E+01	4.4E+01	2.0E+01
Xe-133M	1.1E+01	1.1E+01	1.0E+01	9.4E+00	8.4E+00
Xe-133	2.6E+02	2.6E+02	2.6E+02	2.6E+02	2.4E+02
Xe-131M	2.2E-01	2.6E-01	2.8E-01	2.8E-01	3.0E-01
Xe-129M	1.9E-07	1.8E-07	1.8E-07	1.7E-07	1.6E-07
Total Xe	7.2E+02	4.2E+02	3.6E+02	3.0E+02	2.8E+02
Total	1.2E+04	4.8E+02	3.8E+02	3.0E+02	2.8E+02

TABLE 4-6 Nuclide Composition (curies) of Waste Stream 4 (Table 4-1)

Isotope	0 (hr)	12 (hr)	24 (hr)	36 (hr)	48 (hr)
Kr-88	1.1E+04	5.8E+02	3.0E+01	1.7E+00	8.8E-02
Kr-87	7.6E+03	1.1E+01	1.6E-02	2.4E-05	3.3E-08
Kr-85M	3.8E+03	6.0E+02	9.3E+01	1.5E+01	2.3E+00
Kr-85	1.2E+00	1.3E+00	1.3E+00	1.3E+00	1.3E+00
Kr-83M	1.6E+03	1.7E+02	6.3E+00	2.1E-01	6.5E-03
Kr-81	6.4E-13	6.4E-13	6.4E-13	6.4E-13	6.4E-13
Total Kr	2.4E+04	1.4E+03	1.3E+02	1.8E+01	3.6E+00
Xe-138	1.9E+04	9.4E-12	4.8E-27	0.0E+00	0.0E+00
Xe-135M	3.3E+03	8.6E+02	2.5E+02	7.0E+01	2.0E+01
Xe-135	7.2E+02	6.2E+03	4.1E+03	2.2E+03	9.8E+02
Xe-133M	5.4E+02	5.3E+02	5.0E+02	4.6E+02	4.1E+02
Xe-133	1.3E+04	1.3E+04	1.3E+04	1.3E+04	1.2E+04
Xe-131M	1.1E+01	1.3E+01	1.4E+01	1.4E+01	1.5E+01
Xe-129M	9.4E-06	9.0E-06	8.6E-06	8.2E-06	7.9E-06
Total Xe	3.5E+04	2.1E+04	1.8E+04	1.5E+04	1.4E+04
Total	5.9E+05	2.4E+04	1.9E+04	1.5E+04	1.4E+04

5 ANNULAR LEU-FOIL TARGET QUALIFICATION EFFORT

Approximately 50 Argonne-designed annular LEU-foil targets have been irradiated and processed in Indonesia (Badan Tenaga Atom Nasional—BATAN), Australia (Australian Nuclear Science and Technoology Organization—ANSTO), Argentina (Comisión Nacional de Energia Atómica, Argentina—CNEA), and the United States (University of Missouri Research Reactor—MURR) with no target failure. However, no systematic qualification study has been performed to qualify the use of these targets. The goal of the target-qualification effort to be performed by ICN Romania is to develop a sufficient database of irradiation and post-irradiation examination (PIE) of the annular LEU-foil target to qualify its use in reactors and ^{99}Mo production facilities around the world. The PIE data will be used to support the development and qualification of a LEU-foil target that can be used by any current or future ^{99}Mo producer who desires to evaluate the LEU-foil target option. The effectiveness of nickel-foil, electroplated-nickel, aluminum-foil, and electroplated-zinc fission-recoil barrier materials applied to LEU-foils will be evaluated by testing.[4] The use of rolled uranium foil (either cross-rolled to the specified thickness or rolled followed by a beta-quench step) and use of oil roll-cast by the Korean Atomic Energy Research Institute (KAERI) will also be tested,[5] leading to a test matrix of 12 target configurations. The tests will be performed under irradiation conditions that represent the maximum irradiation parameters (i.e., thermal neutron flux and irradiation time) of dispersion type targets that are now being irradiated to produce ^{99}Mo. The objectives of the experimental plan:

- Determine the change in dimensions of the LEU-foil caused by U-swelling and anisotropic growth and evaluate the integrity of a fission-recoil barrier material's bond to the U-foil during irradiation.

- Determine any change of irradiated-foil flexibility with time after its removal from the target. The purpose of this objective is to provide assurance that the foil does not become brittle and break before it is placed in the dissolver.

- Determine the amount of contamination on the surface of the fission-recoil barrier after the foil's removal from the target. The purpose of this objective is to address the issue of potentially contaminating the hot cell during target disassembly, as discussed at the recent International Atomic Energy Agency (AEA) Consultancy Meeting.

- Measure the amount (i.e., moles) of fission gas (Kr, Xe, and I) that escapes during target disassembly.

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[4] All these fission-recoil barriers were used successfully in earlier irradiations.

[5] All previous irradiations were performed using beta-quenched foils. In addition, one target was irradiated at MURR using a KAERI foil. Beta quenching was performed on rolled foils to produce a random, small-grain structure in the foils, thus eliminating concerns about anisotropic grain growth during irradiation. This process is time consuming and, therefore, will increase the cost of the annular LEU-foil target. Cross-rolling is being examined as an alternative process.

This five-stage program is scheduled to be completed in 13 months. The start-up has been delayed due to the United States Congress's long delay in passing a Fiscal Year 2011 (FY-11) budget. In the first stage, ICN and the U.S. team will develop a detailed test plan, design procedures, and equipment required for the PIE, and complete all planning, procedural, and safety documentation necessary to fabricate the targets at the Y-12 National Security Complex. In addition, they will irradiate, transport, provide PIE of the irradiated targets, as well as disposing of radioactive waste generated from these experiments at Pitesti.

In the second stage, ICN will test all procedures and equipment in the test plan to assure that (1) methods are applicable to performance in the ICN PIE hot cell and (2) the accuracy and precision of the methods developed and the equipment used for the measurements are sufficient to meet the requirements for qualifying the target. It is certain that during this "cold-testing" stage, means to improve the procedures and equipment will be developed. These modifications will be the basis for a final test plan for "hot testing."

In the third stage, targets will be irradiated at lower flux and/or shorter times to (1) assure safe irradiation in the reactor and (2) verify all operations and procedures can be performed safely and as planned. All target configurations and procedures planned for high-level "hot testing" will be part of this low-level-testing task. Experiments in this task will be performed in either duplicate or triplicate. During the performance of this task, modifications to the detailed test plan may also be required. At the end of this task, a revised detailed test plan will be completed.

In the fourth stage, targets will be irradiated at high flux for the time required to subject the targets to the most severe conditions anticipated for their use by current Mo-99 producers. All target configurations will be irradiated and subjected to PIE unless stage 3 reveals the unsuitability of some configurations for further testing. Experiments will be performed in triplicate.

In the fifth stage, a report will be written that can be used by reactors to develop a safety basis for the irradiation of annular LEU-foil targets. This report will be a compilation of all data and results collected during this program, in addition to a summary and conclusion section.

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Chemical Science and Engineering Division
Argonne National Laboratory
9700 South Cass Avenue, Bldg. 205
Argonne, IL 60439-4837

www.anl.gov

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