



# ORAU TEAM Dose Reconstruction Project for NIOSH

DOE Review Release 01/10/2013

Document Title:  <b>Exotic Radionuclides at the Savannah River Site</b>	Document Number:      ORAUT-RPRT-0054 Revision:                    01 Effective Date:            01/03/2013 Type of Document:        Report Supersedes:                Revision 00
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New     
  Total Rewrite     
  Revision     
  Page Change

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**PUBLICATION RECORD**

<b>EFFECTIVE DATE</b>	<b>REVISION NUMBER</b>	<b>DESCRIPTION</b>
01/20/2012	00	New document initiated in response to questions raised by SC&A that not all radionuclides have been identified and considered in the analysis of personnel exposures at the Savannah River Site. Incorporates formal internal and NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by Edward D. Scalsky.
01/03/2013	01	Revision initiated to add new information pertaining to <sup>147</sup> Pm in Attachment B, <sup>249</sup> Bk and <sup>254</sup> Fm in Attachment C, and <sup>171</sup> Tm in Attachment D. Responds to formal NIOSH review comments; For information added a sentence at the end of the first paragraph in Section 5.0; deleted the last sentence in the plutonium paragraph; added information at the end of the uranium paragraph related to the types of DU analysis used between 1986 through 1994 and for DU and EU after 1994; replaced the two paragraphs discussion in the neptunium section with a new paragraph on the separation of neptunium from the actinides and uranium and on the analysis of neptunium; added additional references to the information in the section on <sup>90</sup> Sr; clarified the references for the whole body counting information and added a discussion on the counting geometry and the energy regions of interest and added the references into the reference section; revised the title of Table A-1 to reflect the doses were due to inhalation from handling the $1.8 \times 10^{-6}$ g <sup>99</sup> Tc; added the words "estimates" and "handling" for clarification that estimated inhalation doses are a result of handling the <sup>99</sup> Tc. There were no review comments incorporated as a result of formal internal review. Training required: As determined by the Objective Manager. Initiated by Donald E. Bihl and Edward D. Scalsky.

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**ACRONYMS AND ABBREVIATIONS**

AHIB	alpha-hydroxyisobutyrate
AIMS	Automated Inventory Management System
ANL	Argonne National Laboratory
BNL	Brookhaven National Laboratory
Ci	curies
cm	centimeter
cpm	counts per minute
d	day
DNA	Delayed Neutron Analysis
DU	depleted uranium
EU	enriched uranium
g	gram
HDEHP	di,2-ethylhexylophosphoric acid
HFDP	High Flux Demonstration Program
hr	hour
in.	inch
IPHS	Isotopic Power and Heat Sources
kCi	kilocurie
keV	kiloelectron-volt
kg	kilogram
LRL	Lawrence Radiation Laboratory
mCi	millicurie
MCi	megacurie
mg	milligram
mm	millimeter
MPLB	maximum permissible lung burden
mrem	millirem
n	neutron
nCi	nanocurie
ng	nanogram
NIOSH	National Institute for Occupational Safety and Health
NMMSS	Nuclear Material Management and Safeguards System
NTA	nuclear track emulsion, type A
ORAU	Oak Ridge Associated Universities
ORNL	Oak Ridge National Laboratory
pCi	picocurie
ppm	parts per million

RC	Radiation Control
s	second
SC&A	Sanford Cohen & Associates
SRDB Ref ID	Site Research Database Reference Identification (number
SRL	Savannah River Laboratory
SRP	Savannah River Plant
SRS	Savannah River Site
TIOA	tri-isooctylamine
TRAMEX	transuranic metal extraction
UCB	University of California, Berkeley
UCL	University of California, Livermore
W(t)	watt-thermal
WTD	Works Technical Department
yr	year
μCi	microcurie
μg	microgram

## 1.0 EXECUTIVE SUMMARY

The first reactor (R-Reactor) at the Savannah River Plant (SRP) – now known as the Savannah River Site (SRS) – started operation on December 28, 1953 and shut down in June 1964. The K-, P-, and L-Reactors all started in 1954 and operated until 1988; the L-Reactor was shut down in 1968 and restarted in 1985. The C-Reactor operated from 1955 through 1985.

During the years the SRS reactors were operating, there were many nuclides that were irradiated. Originally, the irradiations were in support of the nuclear weapons program. However, when military requirements slowed down, other missions were sought. Some missions included the production of transplutonium elements; development of isotopic power and heat power sources; and development of sources for food sterilization, medical uses, radiography, and irradiating samples for national laboratories, universities, and other countries in support of their research.

In light of the many radionuclides that resulted from these irradiations, Sanford Cohen & Associates (SC&A) questioned if all radionuclides were identified and considered in the analysis of personnel exposures (SC&A 2010). According to the SC&A report, the main issues relate to the production, irradiation in reactors, and processing (if any) of target materials for the production of special radionuclides to clarify the dose reconstruction issues associated with them. SC&A classifies these special radionuclides as the “exotic” radionuclides that were produced in the C- and K-Reactors during the high flux years of operation (i.e., 1965 to 1970). This report attempts to identify the radionuclides, the bioassay methods that were used to evaluate the potential exposures to personnel handling and processing the radionuclides and, if possible, the approximate quantities that were available at SRS.

## 2.0 INTRODUCTION

In addition to the materials that were irradiated in the SRS reactors for the weapons program, a great deal of material was irradiated for use in commercial, industrial, and medical applications. This report discusses the nuclides that were irradiated and processed for various periods in the SRS facilities. The bioassay methods that were used to evaluate the potential exposure to these radionuclides are also discussed.

Section 3.0 describes production of exotic radionuclides, and Section 4.0 discusses Savannah River Laboratory (SRL) operations. Section 5.0 discusses bioassay practices, and Section 6.0 summarizes the findings of this report. Attachments A through E provide details on production of  $^{99}\text{Tc}$ ,  $^{147}\text{Pm}$ ,  $^{249}\text{Bk}$  and  $^{254}\text{Fm}$ , and  $^{242\text{m}}\text{Am}$ , respectively.

## 3.0 RADIONUCLIDE PRODUCTION

The term “exotic radionuclides” was used first in the SC&A report (SC&A 2010). SC&A stated its concerns that “exotic radionuclides” that are not explicitly addressed as matrix issues for which NIOSH is developing coworker models. Radionuclides excluded from analysis include  $^{232}\text{Th}$ ,  $^{241}\text{Am}$ ,  $^{244}\text{Cm}$ ,  $^{252}\text{Cf}$ ,  $^{237}\text{Np}$ , fission and activation products,  $^{210}\text{Po}$ , recycled uranium, natural uranium,  $^{239/240}\text{Pu}$ , and  $^{241}\text{Pu}$ . SC&A included among the “exotic radionuclides” those materials that were irradiated for offsite customers during the High Flux Demonstration Program (HFDP). However, for purposes of this report any radionuclide with some exceptions (e.g., tritium, thorium) that were produced as a result of irradiation in the SRS reactors is considered to be an exotic radionuclide, some of which are only briefly mentioned.

The SC&A report (SC&A 2010) identified the following radionuclides of concern:

- Alpha emitters:
  - Americium-243
  - Protactinium-231
  - Plutonium-238
  - Plutonium-242
  - Uranium-233
  
- Beta emitters:
  - Cesium-137
  - Europium-153
  - Iodine-131
  - Iridium-192
  - Promethium-147
  - Technetium-99
  - Thulium-170

In the early 1950s, five heavy-water reactors (R, P, L, K, and C) were constructed at SRS. All five reactors were operating by 1955. Reactor shutdown began in 1964 with R-Reactor. C-Reactor was shut down in 1987; L-Reactor was shut down in 1968, restarted in 1985, and shut down again in 1988. K- and P-Reactors operated with few interruptions until 1988. A restart of K-Reactor began in 1991 but was not completed (Till et al. 2001). None of the reactors are currently operational.

Originally, the primary function of the SRS reactors was the production of plutonium and tritium for nuclear weapons. However, even in the early 1950s, the reactors were producing radioisotopes for nonweapons uses, which continued in the 1960s with the reduction of the military mission at SRS (Reed et al. 2002). Depleted uranium (DU),  $^{237}\text{Np}$ , and lithium targets were irradiated to produce  $^{239}\text{Pu}$ ,  $^{238}\text{Pu}$ , and tritium, respectively. The targets were irradiated and transferred to the Tritium Facilities, H-Area Separations ( $^{238}\text{Pu}$ ), and F-Area Separations ( $^{239}\text{Pu}$ ) for processing. Spent reactor fuel was sent to H-Area Separations for processing. The reactors were also used to irradiate targets that produced transplutonic elements such as curium and californium.

Between 1955 and 1967 large quantities of special radionuclides were produced in the SRS reactors for various campaigns. For example, a series of campaigns produced 66 MCi of  $^{60}\text{Co}$  for Oak Ridge National Laboratory (ORNL), the U.S. Bureau of Mines, the U.S. Army Food Process Development Irradiator, Japan Atomic Energy Research Institution, Brookhaven National Laboratory (BNL), and Marine Products Development. SRS also produced 630 kg of  $^{233}\text{U}$  from irradiation of thorium for the thorium breeder reactor program. In addition, 275 kg of  $^{238}\text{Pu}$  from irradiation of  $^{237}\text{Np}$ , 600 g of  $^{210}\text{Po}$ , 1,700 Ci of  $^{152}\text{Eu}$ , and 370 MCi of  $^{170}\text{Tm}$  were produced. Other radionuclides produced during this period and in subsequent years included more  $^{210}\text{Po}$ ,  $^{121}\text{Sn}$ ,  $^{139}\text{Ce}$ ,  $^{166}\text{Ho}$ , and  $^{41}\text{Ca}$  for BNL, the University of Illinois, and Denmark (Bebbington 1990).

From 1965 through 1970, C- and K-Reactors were modified to produce a very high flux of neutrons for production of unusual (e.g.,  $^{170}\text{Tm}$ ,  $^{171}\text{Tm}$ ,  $^{46}\text{Sc}$ ) radionuclide sources. The HFDP began in C-Reactor in February 1965 and lasted 1 year. The HFDP in K-Reactor began in August 1969 to produce  $^{252}\text{Cf}$ . A peak flux of  $7 \times 10^{15}$  n/cm<sup>2</sup>-s was achieved, and 2.1 g of  $^{252}\text{Cf}$  were produced before termination of the production campaign in November 1970. A second Californium Production Campaign was planned but never executed because the market for  $^{252}\text{Cf}$  never materialized as expected (Boswell 2000). The target material in the C- and K-Reactors consisted primarily of  $^{242}\text{Pu}$ . In addition, three 1-in.-diameter thimbles contained  $^{243}\text{Am}$ ,  $^{244}\text{Cm}$ , and 150 target materials of 66 elements for nine

universities and laboratories (Boswell 2000). Table 3-1 lists the target materials that were used to supply irradiated samples to Argonne National Laboratory (ANL), ORNL, the Lawrence Radiation Laboratory (LRL) facilities at University of California, Berkeley (UCB) and the University of California, Livermore (UCL) (Smith et al. 1965). There were additional targets; for example, one tritium-producing core (Mark VI) was designed with a removable inner lithium target so fuel could be irradiated to a very high exposure. The high exposure depleted 70% of the  $^{235}\text{U}$  and thus concentrated  $^{236}\text{U}$ , which was formed from every sixth neutron capture in  $^{235}\text{U}$ . Irradiation of the  $^{236}\text{U}$  formed  $^{237}\text{Np}$ , which was the target material for the production of plutonium (Boswell 2000). In addition, the irradiation of the target material  $^{242}\text{Pu}$ , after a succession of neutron captures and beta decays, successively produced other target materials –  $^{242}\text{Pu}$ ,  $^{243}\text{Pu}$ ,  $^{243}\text{Am}$ ,  $^{244}\text{Am}$ ,  $^{244}\text{Cm}$ ,  $^{245}\text{Cm}$ ,  $^{246}\text{Cm}$ ,  $^{247}\text{Cm}$ ,  $^{248}\text{Cm}$ ,  $^{249}\text{Cm}$ ,  $^{249}\text{Bk}$ ,  $^{250}\text{Bk}$ ,  $^{250}\text{Cf}$ ,  $^{251}\text{Cf}$ , and  $^{252}\text{Cf}$  (Reed et al. 2002). There were also special irradiations during both the HFDP and the Curium I campaign to produce test quantities of  $^{170}\text{Tm}$ ,  $^{171}\text{Tm}$ ,  $^{46}\text{Sc}$ , and  $^{204}\text{Tl}$ .

Table 3-1. Examples of general samples irradiated during the HFDP.<sup>a</sup>

Sample	Laboratory	Sample	Laboratory	Sample	Laboratory
Si-30	ANL	Yb-168	ORNL	Os	UCL
S-33	ORNL	Tm-168	UCB, UCL	W	UCL
Ca-40	ORNL, ANL	Er-170	UCB	Re	UCL
Ca-48	ANL	Yb-176	ANL, UCB, UCL	Be	ANL
Fe-54	ORNL	Lu-176	UCB	Be <sub>3</sub> N <sub>2</sub>	ORNL
Fe-58	ORNL, ANL, UCB	Hf-177	UCB	Xe	UCL
Ni-58	ORNL, ANL	Hf-179	ANL, UCL, UCB	TiO <sub>2</sub>	ANL
Ni-62	ORNL	Hf-180	ANL, UCL, UCB		
Nb-93	ORNL	Ta-180	UCB		
Nb-96	ANL	Ta-181	UCB		
Ag-107	UCB	W-186	ANL, UCL, UCB	<b>Additional isotopes<sup>b</sup></b>	
Ag-108	UCB	Re-187	UCL, UCB	Zn-67	ANL
Sn-118	ORNL, UCB, ANL	Os-180	UCB	Te-124	ANL
Sn-124	ANL	Os-182	ANL, UCL, UCB	Lu-175	UCL
Ba-130	UCB	Bi-209	UCB	Be-9	ANL
Sm-144	ORNL, UCB	Am-241	ANL, UCL		
Nd-146	ORNL	Cm-244	ANL, UCL, UCB	Sn-120	ANL
Nd-150	ANL	Bk-249	ANL		
Sm-150	UCB, UCL	Cf-252	ANL, UCL, UCB	Cd-160	UCL
Sm-154	ORNL	Es-254	UCB	Am-243	UCL
Dy-156	UCB, UCL	LiCl	UCB		
Dy-158	ANL	MgO+Cr-50	UCL		
Tb-158	UCB, UCL	KPO <sub>3</sub>	UCL		
Gd-160	ANL, UCL, UCB	Ar	UCL		
Er-162	UCB, UCL	RbCl	ORNL		
Dy-164	ANL, UCB	Hf	ANL		
Ho-165	UCB, UCL	HFO <sub>2</sub>	UCL		

a. Source: Smith et al. 1965.

b. Additional isotopes from Table 3-2.

Some development work was done mostly with cold Tm<sub>2</sub>O<sub>3</sub> (DuPont 1974, p. 12), and a little work was done with  $^{170}\text{Tm}$ ,  $^{171}\text{Tm}$ ,  $^{46}\text{Sc}$ , and  $^{204}\text{Tl}$  (DuPont 1974, pp. 7–8). Scandium-46 is a gamma emitter. Thallium-204 is a beta-only emitter. Thulium-170 and -171 have low-yield, low-energy gammas that might show up in chest counting if someone was looking closely, but are mostly beta emitters. Thulium oxide was characterized as target material for irradiation to form  $^{170}\text{Tm}$ , with potential applications for short-term missions in space (128-day half-life). As with cobalt, the irradiated product is useful without chemical processing. Conditions were established for fabricating wafers, and the properties of Tm<sub>2</sub>O<sub>3</sub> were studied to prepare and irradiate it to yield a radioactive product suitable for use in heat sources. There was some activity that involved breaching of the source or chemistry on

the  $^{170}\text{Tm}$  and  $^{171}\text{Tm}$ . These were small research and development experiments before actual large-scale production of the sources. There was some breakage of  $^{170}\text{Tm}$  oxide wafers in the early experiments. Metal targets did not have that problem. There was no evidence of ongoing, large-scale breaching or chemistry on the  $^{170}\text{Tm}$  or  $^{171}\text{Tm}$  sources. Thulium-170 was mentioned as a possible heat source early in the program; it emits a gamma ray with about 3% yield. Thulium-171 only emits a beta in sufficient abundance for bioassay.

SC&A mentioned  $^{147}\text{Pm}$  and  $^{192}\text{Ir}$  as two radionuclides of concern (SC&A 2010). The National Aeronautics and Space Administration's interest in aged  $^{147}\text{Pm}$  as a source of isotopic heat for space missions led to a study of the quantity and quality of  $^{147}\text{Pm}$  that could be produced annually at SRS. The recovery of  $^{147}\text{Pm}$  from current high-activity waste was also studied. The major source of  $^{147}\text{Pm}$  was fission product waste; the quantity available after aging to allow for decay of associated  $^{148}\text{Pm}$  would have been about 1,700 W(t) per reactor-year of operation. Product quality from current waste, as represented by a low ratio of  $^{148}\text{Pm}$  to  $^{147}\text{Pm}$ , would presumably have been the same as that produced at the Hanford Site (about  $5 \times 10^{-7}$ ). Promethium-148 emits gamma rays that could require extra shielding. Irradiation of naturally occurring neodymium would produce only 50% as much  $^{147}\text{Pm}$  (DuPont 1974). No further mention of  $^{147}\text{Pm}$  has been found in this historical reference. DuPont (1974) mentions that iridium was experimented with but indicates that this experimentation was with several inactive iridium capsules. The report makes no mention of  $^{192}\text{Ir}$ .

Table 3-1 shows a list of materials that were irradiated for the U.S. Atomic Energy Commission laboratories only (i.e., ANL, ORNL, UCB, and UCL). Some of the materials were irradiated for the duration of the HFDP (i.e., from February to November 1965). Others were irradiated for single fuel cycles of about 1 week (Smith et al. 1965). Table 3-1 does not include isotopes that were irradiated for BNL, the University of Illinois, or Denmark, such as  $^{121}\text{Sn}$ ,  $^{138}\text{Ce}$ , and  $^{139}\text{Ce}$ , or those irradiated for all offsite customers.

Table 3-2 lists some of the specific target elements and compounds that were irradiated for other customers during the indicated months and years. Many of the samples were repeated in subsequent months; for example, March and May 1966 and March 1967 had many repeat samples. There were five shipments during January 1966. The  $^{257}\text{Fm}$  to LRL and the 2  $\mu\text{g}$  of  $^{252}\text{Cf}$  to ANL are listed in the table. The other shipments, not listed, included 380 mg of iridium to ORNL, 200 mg of calcium to Berkeley, and 1  $\mu\text{g}$  of  $^{253}\text{Es}$  to ANL.

The procedure for handling these samples is described by Gray (2006) as follows:

*All samples containing the material to be irradiated for off-site customers were in welded closed aluminum cans. They came to SRP in that condition. They were placed in a basket and the basket lowered into the spent fuel storage basin where the deep water provided shielding for handling radioactive components removed from the reactor. The research samples were placed in small perforated tubes about 1 inch in diameter; maybe five or so samples per a 12" tube. The tubes were then placed in a larger (about 1 3/8ths inch diameter) perforated tube about 21 feet long, called a thimble. The reactor had many of these thimbles for general use with safety rods, instruments, etc. Three of them near the center of the reactor were dedicated to my [Gray's] use for research samples. When it came time for either insertion or removal (or both) of a sample, the thimble was removed by remote machine from the reactor and placed in the basin. It was moved remotely under water to a location where I could have it turned upside down and the tubes unloaded onto a well-lit work table about 12 feet down. I sorted through the tubes and found the one from which a sample would be unloaded (or a new one loaded); all under water. The process was reversed to put the whole thing back together and return the loaded thimble to the reactor for the next*

Table 3-2. Samples irradiated during the HFDP for selected months.<sup>a</sup>

August 1965 <sup>b</sup>		January 1966 <sup>c</sup>		March 1966 <sup>d</sup>		May 1966 <sup>e</sup>		March 1967 <sup>f</sup>	
Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents
ORNL	280 mg natural iridium	SRP-SRL	1.4 g Co-59	LRL	11 mg Ag-107	LRL	11 mg Ag-107	LRL	11 mg Ag-107
SRP-SRL	1.4 g Co-59	ANL	40 mg Cm-244	LRL	1.52 g LiCl	LRL	1.52 g LiCl	LRL	1.52 g LiCl
ANL	40 mg Cm-244	ANL	2 µg Cf-252	LRL	217 mg MgO and Cr-50	LRL	217 mg MgO and Cr-50	LRL	217 mg MgO and Cr-50
ANL	2 µg Cf-252	LRL	240 mg Cm-244, 20 mg Cm-246	LRL	10 mg Fe-58	LRL	10 mg Fe-58	LRL	10 mg Fe-58
LRL	400 mg Cm-244	ANL	77.2 mg Be-9, 0.6 mg Ca-48, 2 mg S-30	LRL	2 mg Dy-156	LRL	2 mg Dy-156	LRL	2 mg Dy-156
LRL	450 mg Cm-244; 40 mg Cm-246	ANL	3.95 mg Fe-58, 1.4 mg Sn-124, 5.26 mg Hf-180	LRL	9 mg Hf-179	LRL	9 mg Hf-179	LRL	9 mg Hf-179
LRL	60 mg Cm-244; 25 mg Cm-246	LRL	1.9 mg Cf-252	LRL	10.3 mg Hf-180	LRL	10.3 mg Hf-180	LRL	10.3 mg Hf-180
ANL	240 mg Cm-244; 20 mg Cm-246	LRL	231 mg Gd-160	LRL	58 mg Sm-150	LRL	58 mg Sm-150	LRL	58 mg Sm-150
ANL	77.2 mg Be-9; 0.6 mg Ca-48; 2 mg Si-30		48 g Bi-209	GT	200 mg 27% Pb-204 metal, 1 mg PbO (Pb-204 enriched to 99.7%)	GT	200 mg 27% Pb-204 metal, 1 mg PbO (Pb-204 enriched to 99.7%)	GT	200 mg 27% Pb-204 metal, 1 mg PbO (Pb-204 enriched to 99.7%)
LRL	3.95 mg Fe-58; 1.4 mg Sn-124; 5.26 mg Hf-180	LRL	48 g Bi-209	BNL	0.5 g C (diamond in graphite)	LRL	1.5 mg Pu-240, 0.1 mg Pu-244	LRL	1.5 mg Pu-240, 0.1 mg Pu-244
ANL	1.9 mg Cf-252	LRL	99 mg Yb-176	ANL	40 mg Cm-244	LRL	48 g Bi-209 (2.5 in.)	LRL	48 g Bi-209
LRL	0.8 g Hf-180	UI	8.3 mg Ba(NO <sub>3</sub> ) <sub>2</sub> , 12.5 mg CeO <sub>2</sub> , 600 mg Sn-118	LRL	1.5 mg Pu-240, 0.1 mg Pu-244	LRL	48 g Bi-209 (2.54 in.)	LRL	48 g Bi-209
LRL	231 mg Cd-160	LRL	1 g Am-243	LRL	48 g Bi-209	LRL	10.8 mg Hf-177	LRL	10.8 mg Hf-177
LRL	48 g Bi-209	LRL	1 g Am-243	LRL ANL	240 mg Cm-244; 20 mg Cm-245	LRL	10 mg Os-192	LRL	10 mg Os-192

August 1965 <sup>b</sup>		January 1966 <sup>c</sup>		March 1966 <sup>d</sup>		May 1966 <sup>e</sup>		March 1967 <sup>f</sup>	
Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents
LRL	48 g Bi-209	LRL	5.7 g Lu-175	LRL	48 g Bi-209	LRL	10.9 mg Er-162	LRL	10.9 mg Er-162
LRL	99 mg Yb-176	ORNL	216 mg Sm <sub>2</sub> O <sub>3</sub> (enriched to 94.5% Sm-144) and 7.828 mg type 302 SS monitor	LRL	10.8 mg Hf-177	LRL	9.6 mg W-186	LRL	9.6 mg W-186
<b>SRP</b>	13.47 mg Al	ORNL	700 mg gold foil and 7.61 mg type 302 SS monitor	LRL	10 mg Os-192	LRL	9.74 mg Gd-160	LRL	9.74 mg Gd-160
UI	8.3 mg Ba(NO <sub>3</sub> ) <sub>2</sub> ; 12.5 mg CeO <sub>2</sub> ; 600 mg Sn-118	ORNL	418 mg La <sub>2</sub> O <sub>3</sub> and 7.718 mg type 302 SS monitor	LRL	10.9 mg Er-162	LRL	9.74 mg Re-187	LRL	9.74 mg Re-187
ORNL	280 mg natural iridium	ANL	1 g Sn-120	LRL	9.6 mg W-186	LRL	7.65 mg Lu-176	LRL	24 mg Sm-144
LRL	1 g Am-243	LRL	40 mg Ba-130 as BaCO <sub>3</sub>	LRL	9.74 mg Gd-160	LRL	24 mg Sm-144	LRL	50 mg Sn-118
LRL	1 g Am-243	LRL	40 mg Ba-130 as BaCO <sub>3</sub>	LRL	9.74 mg Re-187	LRL	9.6 mg Ag-109	GT	77 g lead
LRL	1 g Am-243	LRL	40 mg Ba-130 as BaCO <sub>3</sub>	LRL	24 mg Sm-144	LRL	50 mg Sn-118	<b>SRL</b>	1 g Er <sub>2</sub> O <sub>3</sub> enriched in Er-170
LRL	1 mg Hg-203 (as nitrate)	LRL	1.5 mg Pu-240, 0.1 mg Pu-244	LRL	9.6 mg Ag-109	ORNL	100 mg Te-128 (99.16% enriched)	ANL	60 mg Te-126 enriched (98.69%)
<b>SRL</b>	90 mg Nd <sub>2</sub> O <sub>3</sub> (enriched Nd-146)	BNL	0.5 g C (diamond in graphite)	LRL	50 mg Sn-118	GT	77 g Pb	Livermore	460 mg Cm-244, 90 mg Cm-246, 9 mg Cm-248
		BNL	0.5 g C (diamond in graphite)	ORNL	100 mg Te-128 (99.16% enriched)			NC	500 mg SnO <sub>2</sub> enriched 99% Sn-120
		BNL	0.5 g C (diamond in graphite)	ORNL	100 mg Te-128 (99.16% enriched)			Livermore	300 µg Cf-252
		ANL	Ca-40, Ca-42, Ni-58, Ni-62,	GT	77 g lead			ANL	Er-162, Yb-176, Mo-92

August 1965 <sup>b</sup>		January 1966 <sup>c</sup>		March 1966 <sup>d</sup>		May 1966 <sup>e</sup>		March 1967 <sup>f</sup>	
Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents	Sponsor	Slug contents
		ANL	Zn-67, Te-124, Pb-204 <sup>d,g</sup>					ANL	Fe-54, Yb-168, Cr-50, Os-192
		ANL	Se, Yb-176 <sup>g</sup>					ANL	Nd-150, Ti-50, W-186, Yb-176
		ANL	Nd-150, Os-190 <sup>g</sup>					ANL	Nb, Ce-136, Cf-249, Np-236
		ANL	U-236, Pa, Ni <sup>g</sup>					ANL	90 mg Cm-244, 30 mg Cm-246
		ANL	Np-236, Cf-249, Th-229 <sup>g</sup>					LRL	5 mg Ln-176 (70% enriched)
		LRL	200 mg calcium					ORNL	103.2 mg WO <sub>3</sub> (97.2% enriched W- 186)
		LRL	0.7 mg platinum metal						
		LRL	1 mg Hg-202 in quartz capsule						
		LRL	Fm-257 rabbit sample in Al capsule (10 µg)						
		ORNL	8.5 mg type 302 SS monitor						
		ORNL	150 µg <sup>236</sup> U <sub>2</sub> O <sub>3</sub> ; 8.5mg type 302 SS monitors						

a. GT = Georgia Tech; SS = stainless steel; UI = University of Illinois.

b. DuPont (WTD Aug 1965). (For citations of this form, see the Monthly and Quarterly Reports section at the end of the References list.)

c. DuPont (WTD Jan 1966).

d. DuPont (WTD Mar 1966).

e. DuPont (WTD May 1965).

f. DuPont (WTD Mar 1967).

g. The sample mass and chemical form of one or more nuclides were unreadable in the referenced document.

*cycle. Of the 15-18 hours for shut down, I had about 3-5 hours in the middle of it to do my work.*

*A removed research sample, without any opening of the welded-closed aluminum cans, was placed directly in a shielded shipping cask. The cask lid was put in place and sealed (or locked) down for transport. As the cask was removed from the basin it was washed down. Then it was placed on a truck for transport, usually to the Atlanta airport, for a flight to the outfit that sent it to us originally for irradiation. Many samples, because of short half lives of the desired product, were returned in well less than 24 hours. No samples were opened at SRP [Savannah River Plant].*

As discussed by Gray (2006), all samples that were to be irradiated for external customers were in welded-closed aluminum cans. They came to SRS in that condition. After irradiation they were prepared under water for shipment, so the potential for intake was minimal. Similarly, because the sources were inspected and loaded into casks under water, the potential for external exposure was reduced. As noted above, Table 3-2 lists specific samples that were irradiated for the sponsors listed in the odd numbered columns for the indicated month and year. As can be seen in the table, the samples were generally small for research purposes at the sponsor's facilities. However, there are five samples in the 5-month period listed in the table that were transferred to SRS-SRL. These samples would have been handled using SRS procedures. Based on the limited number of radionuclides transferred, it would appear that such irradiations were not routine.

In addition to the radionuclides and compounds listed in Tables 3-1 and 3-2, a request was made of the Isotopes Development Center at SRS to look at the possibility of using irradiation facilities as fully as possible for the production of unusual or quite large amounts of isotopes for the potential benefit to research and commerce. In response to that request, Rupp (1969) discussed some of the characteristics of the radionuclides in Table 3-3, the target materials or compounds that would be used to produce some of them, and difficulties that might be encountered. He noted that some of these radionuclides were already being produced. As noted above in this section, these radionuclides have been identified by SC&A (2010) as being of concern for dose reconstruction. The entire list discussed in Rupp (1969) is included in Table 3-3. No documentation has been found indicating that program was ever pursued.

Table 3-3. Radionuclides discussed for possible irradiation (Rupp 1969).

Plutonium-238	Thulium-170 and 171	Calcium-41	Barium-133	Samarium-154
Americium-242m	Potassium-40	Iron-55	Lanthanum-137	Samarium-155
Cesium-134	Argon-38 (stable)	Nickel-59	Terbium-157	Europium 155
Europium-152	Chlorine-36	Cadmium-109	Holmium-166m	
Europium-154	Argon-39	Cadmium-113m	Samarium-151	

In addition to the information above, a review of monthly reports was conducted to identify the radionuclides that were being irradiated or processed at SRS from 1954 through 1971. Table 3-4 lists the results of this review (Taulbee 2011). Some of these isotopes were part of the production process and many were shipped to off-site customers.

The cells containing an X indicate that the isotope was present several times during the year. Some isotopes were present in every month of the year, others only during several months. Those identified by an L or M indicate they were present on a limited basis during 1 or 2 months during the year. The letters represent the potential for exposure to that radionuclide [i.e., either low (L) or medium (M) during that period]. This represents the highest estimated exposure potential for those radionuclides. Because the radionuclides indicated by an X appeared many times during the year, an indication of their exposure potential could not be identified in the table. Most were in the low to medium range but others, such as <sup>241</sup>Am, <sup>252</sup>Cf, <sup>244</sup>Cm, and <sup>237</sup>Np, were often listed as a high exposure potential. The

Table 3-4. Isotopes identified in SRS documents from 1954 through 1971.

Isotope	1954	1955	1956	1957	1958	1959	1960	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970	1971
Am-241					X					X		X				X	X	X
Am-242m <sup>a</sup>															# <sup>b</sup>	M <sup>c</sup>		
Am-243						X	X	X		X	X	X	X	X	X	X		
Al <sup>a</sup>												L <sup>d</sup>						
Ba-133 <sup>a</sup>										L								
Bi-209 <sup>a</sup>													L					
Bk-249												X	X	X	X	X	X	X
Bk-250 <sup>a</sup>																L	M	
Ce-144 <sup>a</sup>						X				X	X			X	X			
Cf-252						X					X	X	X	X	X	X	X	X
Cm-244					X	X	X	X		X	X	X	X			X	X	X
Cm-248 <sup>f</sup>														f				
Co-59												X	X					
Co-60		X								X	X				X	X	X	X
Cs-137 <sup>a</sup>										X	X	X						
Er-170											X	X		X				
Es-253														X	X	X	X	
Es-254												X	X	X	X			
Es-255														L				
Eu-152 <sup>a</sup>														L	L			
Eu-154 <sup>a</sup>											L							L
Fm-254 <sup>a</sup>																	M	
Flux wire														L				
Ir-192 <sup>a</sup>													L					
Nd-148 <sup>a</sup>												L						
Np-237 <sup>a</sup>		X	X			X	X	X	X	X	X	X	X	X	X	X	X	X
Np-238 <sup>a</sup>																		M
Np-239									L									
Pa-231 <sup>a</sup>													M					
Pa-233 <sup>e</sup>											e							
Pm-147 <sup>a</sup>						X	X					X	X	X	X		X	
Po-210 <sup>a</sup>									X		X	X	X	X	X	X		
Pu-238 <sup>a</sup>									X		X	X	X	X	X	X	X	
Pu-239 <sup>a</sup>												X						
Pu-240 <sup>a</sup>			X				X	X	X			X	X	#	#			
Pu-241 <sup>a</sup>												X						
Pu-242 <sup>a</sup>										X		X	X	X	X	X	X	X
Ru-106 <sup>a,e</sup>														e				

Isotope	1954	1955	1956	1957	1858	1959	1960	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970	1971
Sb-124 <sup>a</sup>																		L
Sb-125 <sup>a</sup>																		L
Sc-46 <sup>a</sup>										L		L						
Si (quartz) <sup>a</sup>													L					
Ta <sup>a</sup>													L					
Tc-99 <sup>a</sup>												L						
Tl-204 <sup>a</sup>												L	L					
Tm-170 <sup>a</sup>										X		X	X	X	X			
Tm-171 <sup>a</sup>											X	X	X	X	X			
U-233 <sup>a</sup>		X							X	X		X	X	X	X			X
Y-91 <sup>a</sup>						L												
Zr-95 <sup>e</sup>													e					
Zr-95-Nb-95 <sup>e</sup>														e				
Thorium <sup>a</sup>	X	X			X	X	X	X	X	X	X	X	X	X	X			

a. These isotopes are not included in Table 3-2.

b. # Identified in calculations or plans.

c. M Represents an estimated moderate potential of exposure.

d. L Represents an estimated low potential of exposure.

e. Not identified in the review of the monthly reports but was identified by the SRL bioassay program (DuPont 1984a).

f. During separation and purification of transcurium nuclides, 1.7  $\mu\text{g}$  <sup>248</sup>Cm was obtained from a filter from which 14  $\mu\text{g}$  <sup>252</sup>Cf had previously been obtained.

# symbol indicates the isotopes were identified either through existing SRS calculations or in plans. There are four radionuclides added to the table that were not identified in the monthly reports but were identified in *History of the Savannah River Laboratory, Volume IV – General* (DuPont 1984a) and were observed in the bioassay results. These are  $^{106}\text{Ru}$ ,  $^{233}\text{Pa}$ ,  $^{95}\text{Zr}$ , and Zr/Nb-95 and are included for completeness.

#### 4.0 SAVANNAH RIVER LABORATORY OPERATIONS

The Savannah River Laboratory (SRL) (Building 773-A) and related organizations began organizing in July 1953. Over the next couple of years the Engineering Services that were provided in support of research and development personnel included operation of all building auxiliaries and waste disposal facilities, a glass shop, instrument pool, and a supply room. Close liaison was established with Works Engineering Department personnel to coordinate all maintenance, machine shop, and project work for the SRL. As radioisotopes were brought into 773-A to start operations in radiochemical laboratories, the Radiation Control (RC) Group assisted in establishing necessary procedures and handling techniques.

Operations formally accepted occupancy of the cave wing, 773-A, Part II, on December 7, 1953, and all construction work was completed in February 1954. Experiments with radioactive materials began in the caves in July 1954. In October 1954 the first plutonium produced at the SRP was isolated in the caves.

A review of the operations in the SRL was conducted because that was where the products of reactor operations were evaluated. While the SRL did not evaluate the radionuclides that were shipped to offsite customers, they did handle, evaluate, experiment with, and store "production" radionuclides such as  $^{244}\text{Cm}$ ,  $^{252}\text{Cf}$ ,  $^{60}\text{Co}$ , etc. Reactor incidents are not covered in this report because they would primarily involve reactor functionality, such as fuel failures, valve failures, etc., rather than exposure to the handlers of the transfers of highly radioactive radioisotopes, which was, of necessity, performed under water due to the high exposure potential. There were a considerable number of potentially serious incidents in the SRL, as discussed below. Several of the incidents have been investigated and have an associated Special Hazards Investigation Report (DuPont 1962–1974), while the nature of many of the incidents has not been identified. However, there was an extensive radiation control program in terms of identifying potential problems and steps taken to evaluate any internal or external exposure as a result of these incidents.

Radiation Control was established in Technical Division facilities so all the survey, monitoring, and "containment" services usually assigned to Health Physics could be provided in the Technical Division organization, together with a wholly integrated approach and direct responsibility (DuPont 1984a). The containment service function required close liaison work with technical personnel in the design and construction of the special equipment necessary to contain the radioactive materials completely during experimental work. This function was performed by the Apparatus Development Group, a subgroup of the RC Group (DuPont 1984a). As radioisotopes were brought into Building 773-A to start operations in radiochemical laboratories, the RC Group assisted in establishing necessary procedures and handling techniques. Major effort was directed to extensive training of laboratory personnel and personnel of assigned groups. The fundamentals of radiation and contamination control and aspects of personal protection were stressed. By December 1955, procedures basic to the program of personnel and area radiation protection were essentially complete (DuPont 1984a).

Much of the work in the SRL was carried out in the High-Level Caves, which were designed to perform chemical and metallurgical operations on highly radioactive materials. As indicated above, operations formally accepted occupancy of the cave wing, 773-A, Part II, on December 7, 1953. In October 1954 the first plutonium produced at the SRP was isolated in the caves to provide basic data

to substantiate and refine calculations of reactor performance. Operations in the caves continued at the maximum obtainable rate from that date.

The following information is a synopsis of the internal exposure history of the SRL from 1953 through December 1972, which encompasses the HFDP operational period. The information for these years is taken from *History of the Savannah River Laboratory, Volume IV – General* (DuPont 1984a).

From July 1953 to the early part of 1960 evaluation of internal doses was based on *in vitro* analysis. In 1960 the SRS whole body counter was completed and provided another means of assessing internal doses. From 1953 through 1972 there were thousands of urine samples and whole body counts but relatively few documented uptakes in the SRL that resulted in significant doses. There were no significant biological uptakes from 1953 through 1959 and none identified in 1963 and 1968. Table 4-1 shows the number of positive bioassay results that have been documented in DuPont (1984a). Not all positive bioassay results were confirmed to be significant uptakes.

Table 4-1. Positive bioassay results.

Year	Number of positive results	Radionuclide	% Maximum permissible body burden
1960	5	Not identified	<1
1961	3	Not identified	<1
1962	51	Not identified	<1
1964	1	Pa-233	<1.5
	1	Ce-144	<1.5
		Pu	<1.5
	1	Not identified	Not identified
1965	1	Am-Cm	30 <sup>a</sup>
	1	Not identified	<5
1966	1	Zr-95	3.2
		Ru-106	1.2
	4	Pa-231	<1
1967	1	Ru-106	4.2
		Ce-144	1.4
		Zr-Nb-95	0.36
1969	11	Cm-244	Not identified
1970	31	Cm-244	Not identified
	1	Cm-244	SHI No. 273 <sup>b</sup>
1971	21	Not identified	Not identified
	1 of the 21	Cm-244	SHI No. 283 <sup>c</sup>
1972	4	Not identified	Not identified
	2	Cf-252	Not identified

a. No Special Hazards Investigation Report was found for this uptake.

b. Lower (1970).

c. Lower (1971).

Two Special Hazards Investigation Reports (SHI) have been identified for this period. Report No. 273 reported that a general service operator received an uptake of <sup>244</sup>Cm when he released airborne activity to the decontamination room environment while removing solid radioactive waste from the Decontamination Chamber in Building 773-A, Decontamination Facility, Room C-003 (Lower 1970). Special Hazards Investigation Report No. 283 reported an Analytical Chemistry Division Chemist assimilated some curium while removing samples from storage in Building 773-A, Room B-15 (Lower 1971).

## 5.0 **BIOASSAY PROGRAMS**

Workers at SRS were monitored for internal deposition of radionuclides from the beginning of operations. The methods for evaluation of potential intakes included analysis of the amount of radioactivity excreted in the urine or feces (*in vitro* bioassay) and the measurement of the photon radiation emitted from the body by chest counting or whole-body counting (*in vivo* bioassay). The bioassay descriptions in this section are brief. More detailed descriptions can be found in the most recent version of the Internal Dosimetry Technical Basis Document (ORAU 2005).

In the beginning of operations at SRS, the only method available to evaluate the potential internal exposure of personnel was *in vitro* bioassay, primarily analysis of urine samples. This method was typically a gross beta activity analysis although a gross gamma analysis was included from at least 1959 (Boni undated). Over the years, the bioassay analyses methods changed as new techniques were developed.

### 5.1 **IN VITRO BIOASSAY**

*In vitro* bioassay methods changed considerably over time as new techniques and analyses were developed and refined. Some of the methods used for analysis of some of the radionuclides are briefly discussed below.

Before completion of a whole-body counting facility in 1960, only *in vitro* bioassay, primarily urinalysis, was performed. *In vitro* urine bioassay for nuclear byproducts performed during this period was typically a gross beta activity analysis and/or gross gamma analysis. Since that time, *in vitro* bioassay analysis has become more radionuclide specific, and can be divided into the following categories:

- Plutonium urinalysis
- Uranium urinalysis
- Trivalent actinide urinalysis
- Neptunium urinalysis
- Fission product urinalysis
- Fecal bioassay

#### **Plutonium Urinalysis**

The first procedure for plutonium urinalysis at SRS was implemented in 1954 and used until 1959. Changes in the processes to prepare the solution for counting were made, but the method for counting the alphas tracks on the nuclear track emulsion, Type A (NTA) remained the same until 1966. In 1964 solid-state surface barrier detectors (gross alpha counting) replaced the NTA detectors. In the early 1980s experimentation began with a new technique that used alpha spectroscopy, which allowed the detection of the individual radionuclides in the sample (Taylor 2000).

#### **Uranium Urinalysis**

Analysis of uranium by measurement of emitted alpha radiation was referred to as an analysis for "enriched uranium." The analysis of uranium based on its chemical properties was referred to as a "depleted uranium" analysis. Enriched uranium (EU) was determined starting in the mid-1950s. Analyses for DU were performed with the ORNL fluorophotometric method from 1954 to 1982. This method involved using a spectroscope to measure total uranium. The delayed neutron analysis (DNA) method was adopted for both EU and DU analyses around 1982 (Taylor 2000). Kinetic phosphorimetry analysis was also used for DU during 1986 through 1994. Alpha spectrometry and gross alpha counting have been used for both DU and EU since 1994.

### Trivalent Actinide Urinalysis

The trivalent actinides consist of americium, curium, and californium. There was increased production of these materials at SRS in the early 1970s. Analysis was accomplished by extracting the trivalent actinides from the plutonium tri-isooctylamine (TIOA) raffinate with di,2ethylhexylophosphoric acid (HDEHP), followed by gross alpha counting. The 1970 method also captured thorium, berkelium, einsteinium, and fermium (Butler and Hall 1970). In 1990, ion exchange resin was used to separate trivalent actinides from other actinides. The sample was direct-mounted (placed on a planchet with rimmed edges and heated to evaporation), and was "gross-alpha" counted by using a wide window on the alpha spectrometer. Since 1994, extraction chromatography resin has been used to separate trivalent actinides from other actinides. With the implementation of improved software and use of a  $^{243}\text{Am}$  tracer, chemical recovery and counting efficiency could be determined for each sample (Taylor 2000).

### Neptunium Urinalysis

From 1993-1996, extraction chromatography resin was used to separate neptunium from fission products and other actinides followed by electrodeposition and gross alpha counting (Taylor 2000). In 1996, analysis returned to SRS using calcium phosphate precipitation followed by capture on ion exchange resin from which plutonium and neptunium were extracted separately followed by alpha counting (WSRC 2001 pp. 183-184). Since 2001, TEVA and TRU resins are used to separate neptunium from actinides and uranium, and alpha spectrometry has been used (WSRC 2001, pp. 58, 175, 183-184).

### Fission Product Urinalysis

Fission products measurements in urine date to at least January 1957, although the method used at that time has not been discovered. Since 1959, select fission products in urine were analyzed by beta counting on a Geiger-Mueller or proportional counter. The beta count was primarily used to determine the presence of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$ , and  $^{90}\text{Y}$ , which emit no gamma radiation. Because high-energy betas are associated with  $^{89}\text{Sr}$ , a reliable measurement of its activity was made even though an average of 1 g of precipitate was mounted on the planchet. Yttrium-90, which emits a strong beta also yields a reliable count, and affords a satisfactory estimate of its parent isobar,  $^{90}\text{Sr}$ , since some  $^{90}\text{Sr}$  betas are not counted due to absorption by the solid. The low-energy beta of most of the induced activities along with the beta of zirconium-niobium and the rare earths also contributed to the total gross beta count. No interference from  $^{40}\text{K}$  was observed. The chemistry on the beta sample resulted in quantitative recovery of beta-emitting isotopes of iron, cobalt, strontium, niobium, zirconium, ruthenium, and cerium. Gross gamma counting was performed using sodium iodide detectors. The gamma count was primarily used to determine the presence of induced activity. A gamma count was used because of the low counting efficiency of the weak beta energies associated with the induced activities. The gamma from zirconium-niobium and rare earths also contributed to the total gross gamma count. Either the beta or the gamma counting statistics are good enough to accurately estimate 1% of the maximum permissible amount in the total body of any of the following radionuclides (i.e.,  $^{90}\text{Sr}/\text{Y}$ ,  $^{89}\text{Sr}$ ,  $^{95}\text{Zr}/\text{Nb}$ ,  $^{95}\text{Nb}$ ,  $^{144}\text{Ce}/\text{Pr}$ ,  $^{60}\text{Co}$ ,  $^{59}\text{Fe}$ ,  $^{51}\text{Cr}$ ,  $^{65}\text{Zn}$ ) with the exception of zirconium-niobium. The mixed beta-gamma isotope method provided a simple evaluation of the sample regarding the need for an extensive specific analysis, thus eliminating unnecessary and time consuming work (Boni undated).

SRS logbooks show that radionuclide-specific concentrations and associated uncertainties were quantified for some radionuclides for some of the gamma-counted samples starting in December 1965 (DuPont 1965–1968). The reported radionuclides were:

- Chromium-51
- Manganese-54
- Cobalt-60
- Zinc-65

- Zirconium/Niobium-95
- Ruthenium-106
- Iodine-131
- Cesium-137
- Barium/Lanthanum-140
- Cerium-144.

### Strontium Urine Bioassay

In the late 1950s and early 1960s,  $^{89/90}\text{Sr}$  was determined by beta counting on automatic and Geiger-Mueller counters along with zirconium, rare earths, and niobium. From 1969 to 1997, strontium was analyzed by a liquid ion exchange method that extracted  $^{90}\text{Y}$ , which was counted on a beta proportional counter (Taylor et al 1995, p. 79). In 1998, the strontium method was modified by using an ion exchange column to extract  $^{90}\text{Sr}$ , which was also counted on a proportional counter (Taylor 2000, p. 5). Since 2001,  $^{90}\text{Sr}$  has been separated as part of the use of TRU and TEVA resins for plutonium, uranium, actinides, and strontium (WSRC 2001, pp. 58, 168, 169, 172, 173).

Table 5-1 lists the isotopes that were listed as being present, their radiations, bioassay method for detection, half-life, and the number of times they were identified in the monthly reports from 1954 to 1971. An X indicates they were listed numerous times.

Table 5-1. Selected isotopes bioassay methods.

Radionuclide	Times identified	Half-life <sup>a</sup>	Principal radiation	Bioassay method used or available for use	Bioassay period of availability
Am-243	X	7.4E3 yr	Alpha, gamma	Trivalent actinide urinalysis	From mid 1960s
				Whole-body counting of Np-239	From 1960
				Chest counting	From 1966 <sup>b</sup>
Bk-249	X	320 d	Beta	None discovered <sup>c</sup>	
Co-60	X	5.26 yr	Beta, gamma	Whole-body counting	From 1960
				Gamma urinalysis IA	From 1960
Cs-137	X	30.2 yr	Beta, gamma	Whole-body counting	From 1960
Es-253	X	20.5 d	Alpha	Trivalent actinide urinalysis <sup>d</sup>	From mid 1960s
				Whole-body counting of Bk-250 progeny	From 1960
Fm-254	X	3.24 h	Alpha	None discovered <sup>e</sup>	
Pm-147	X	2.63 yr	Beta	None discovered <sup>f</sup>	
Pu-242	X	2.9E5 yr	Alpha	Pu urinalysis <sup>g</sup>	From 1954
Tm-170 <sup>h</sup>	X	129 d	Beta, gamma	Chest counting <sup>i</sup>	From 1966
Tm-171 <sup>h</sup>	X	1.92 yr	Beta	None discovered <sup>l</sup>	
U-233	X	1.6E5 yr	Alpha	EU urinalysis	From 1959
Bk-250	2	3.22 hr	Beta, gamma	Whole-body counting <sup>k</sup>	From 1960
Ce-144	2	284 d	Beta, gamma	Fission product urinalysis FP	From 1950s
				Whole-body counting	From 1960
Eu-152	2	12.7 y	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Eu-154	2	8.8 yr	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Tl-204	2	3.8 yr	Beta	Fission product urinalysis FP	From 1950s
Am242m <sup>l</sup>	1	152 yr	Beta (Am-242)	Trivalent actinide urinalysis	From mid 1960s

Radionuclide	Times identified	Half-life <sup>a</sup>	Principal radiation	Bioassay method used or available for use	Bioassay period of availability
Ba-133 <sup>n</sup>	1	10.5 yr	Gamma	Gamma urinalysis	From 1960
Cm-248	1	3.4E5 yr	Alpha	Trivalent actinide urinalysis	From mid 1960s
Es-255	1	38 d	Alpha, beta (including progeny)	Trivalent actinide urinalysis <sup>d</sup>	From mid 1960s
Np-239	1	2.35 d	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Pa-231	1	3.2E4 yr	Alpha, gamma (7%)	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Sb-124	1	60.2 yr	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole body counting	From 1960
Sb-125	1	2.73 yr	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Sc-46	1	83.8 d	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Tc-99	1	2.1E5 yr	Beta	None <sup>m</sup>	
Y-91	1	58.8 d	Beta	Fission product urinalysis FP	From 1950s
Ir-192	Shipment only	74.3 d	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
Cd-113m <sup>n</sup>	0 <sup>n</sup>	13.6 yr	Beta	Fission product urinalysis FP	From 1950s
Cs-134 <sup>n</sup>	0	2.06 yr	Beta, gamma	Fission product urinalysis FP	From 1950s
				Whole-body counting	From 1960
Eu-155 <sup>n</sup>	0	5.0 yr	Beta, gamma	Chest counting	From 1966
				(also whole-body counting of associated Eu-154)	From 1960
Ho-166m <sup>n</sup>	0	1.2E3yr	Beta, gamma	Gamma urinalysis IA	From 1960
				Whole-body counting	From 1960
I-131	0	8.06 d	Beta, gamma	Whole-body counting	From 1960
				Thyroid counting	
K-40 <sup>n</sup>	0	1.3E9 yr	Beta, gamma	Whole body counting <sup>o</sup>	From 1960
Np-238	1	2.12 d	Beta, gamma	Whole-body counting if soon after intake	From 1960

- a. Half-lives are taken from Lederer, Hollander, and Perlman (1967) and the *Radiological Health Handbook* (PHS 1970).
- b. The 40-cm arc whole-body detector had a lung calibration for the 98-keV gamma from Nd-147, which also would have detected the 99- 111-keV X-rays from Np-239.
- c. See Attachment C.
- d. The trivalent actinide method from 1970 forward captured Am, Cm ,Cf, Th, Bk, Es, Fm (Butler and Hall 1970). Alpha-emitting isotopes would have been counted.
- e. The half-life of Fm-254 is so short that bioassay would not have been feasible. However, the cooling of the target before handling in the laboratory resulted in little or no leftover <sup>254</sup>Fm. See Attachment C.
- f. See Attachment B.
- g. Pu-242 was used as an internal tracer starting in about 1981, but would have been detected in Pu urinalysis prior to that.
- h. Isotopes that were discussed by Rupp (1969) in response to the request to use the SRS irradiation facilities for commercial and industrial development.
- i. Because Tm is a bone seeker, the 84-kev gamma from the ribs would have been detectable in chest counts, despite the low yield (3.3%).
- j. See Attachment D.
- k. The half-life for Bk-250 is so short that the whole body count would have had to occur shortly after intake; however, the DAC (1E-7 μCi/cm<sup>3</sup>) is 50 times larger than for Sr-90 so a significant intake would have been readily detectable in the workplace.
- l. See Attachment E.
- m. See Attachment A.
- n. Not identified in the referenced monthly reports but were available along with bioassay methods for their detection.
- o. An occupational intake would have been detectable only if the count was significantly larger than natural background.

## Fecal Bioassay

Chemical bioassay of urine and fecal samples has been carried out at SRS since the 1950s to determine an individual's uptake of uranium and actinide elements such as americium, plutonium, and californium. Over the years, several techniques have been employed for these types of analyses. Methods ranging from autoradiography to high-resolution solid-state alpha spectroscopy have been used at one time or another.

Many different methods of sample preconcentration, treatment, and preparation have been evaluated and used over the years. Each technique had some particular advantage at the time it was being used, either in terms of analysis time, the number of samples that could be processed simultaneously, the physical form of the final material to be counted, compatibility with other analyses that could be carried out sequentially or in parallel with the main analysis, compliance with more stringent detection levels, etc. (WSRC 1990). In contrast to a urine bioassay, a fecal bioassay can (and usually does) contain material that was never in the systemic body (i.e., insoluble material cleared from the lungs and gastrointestinal tract). Fecal bioassay is thus complementary to urine bioassay. In fact, at SRS, fecal bioassay is always used in conjunction with a urine bioassay and never alone (WSRC 2001).

The primary method for fecal bioassay in the early and mid-1980s was gamma spectrometry with a phoswich detector. Some of the samples that were analyzed by this method were subsequently dried and sent to offsite laboratories for analysis. Since 1994, fecal samples have been analyzed by a modified urinalysis procedure (Taylor et al. 1995; Taylor 2000).

## 5.2 IN VIVO BIOASSAY

### History of Whole-Body Counting at SRS (Taylor 2000)

*In vitro* bioassay was the only method available for assessing occupational intakes of radionuclides in defense facilities like SRS before the mid-1950s. Before the development of large NaI detectors, *in vivo* bioassay was performed with ionization chambers or Geiger-Mueller tubes that were rather insensitive and not practical for assessing occupational intakes of radionuclides, with the possible exception of  $^{131}\text{I}$ . In 1957, a whole body counting facility similar to that at ANL using NaI detectors was proposed for SRS. Before the completion of the facility in 1960, only *in vitro* bioassay, primarily urinalysis, was performed at SRS. *In vitro* urine bioassay for fission and activation products during this period was typically a gross beta or gross gamma analysis.

The original SRS Whole Body Counter in Building 735-A was completed in 1960 and ceased operations in September 1995. The original geometry was a single 8- by 4-in. NaI detector with the person sitting in a chair. This was referred to as the 40-cm arc geometry. The energy spectrum was divided into 15 regions ranging from 60 -100 keV ( $^{147}\text{Nd}$ ) to 1760 -1990 keV (unnamed) (Taylor 2000, pp. 5-6, Taylor et al 1995 pp.60-63, Watts et al 1964, Watts 1961-67). Sometime during the early 1970s, the original chair geometry was replaced with a recumbent (bed) geometry. The single NaI detector was replaced with five 4- by 4-in. cylindrical NaI detectors under the bed. The NaI detectors were in an arc under the bed to provide constant efficiency for all detectors (Taylor 2000). Starting in about 1979, the gamma spectrum was reorganized into 10 regions of interest from 104 keV to 1,534 keV (as shown on the In Vivo Count Results form). In the mid-1980s, whole body counting using a stand-up geometry and large (4" x 4" x 16" or 5" x 3" x 16") NaI detectors and much shorter counting times (2-4 min.) was implemented (Taylor 2000, p. 7; Taylor et al 1995, pp. 74-76).

### History of Chest Counting at SRS (Taylor 2000)

The whole-body counter, which was useful for monitoring workers for intakes of gamma-emitting fission and activation products, was of no use for detecting or quantifying intakes of many actinides such as plutonium and americium. The 40-cm arc whole body counter was calibrated for  $^{147}\text{Nd}$  in the lung as well as in the whole body so low-energy gamma emitters (around 100 keV) could be quantified, although the sensitivity was poor. Efforts began at SRS in the mid-1960s to develop low-

energy photon detectors that would permit detection of actinides in the chest. Most efforts involved either thin NaI detectors or xenon-filled proportional counters. The xenon proportional counters, with their high resolution, showed great promise but were eventually abandoned because of their high background and low efficiency. Development of the thin NaI detector continued, and in 1966 a 1-mm-thick by 5-in.-diameter detector with a 0.001-in. window was first used to count workers involved in an incident. Thin NaI detectors were used with varying degrees of success from 1966 until 1971, when they were replaced with phoswich detectors. The results of the thin NaI counts were verified to be officially recorded in the personnel files of the workers counted during this period.

In the late 1960s, the phoswich detectors were developed for low-energy photon and X-ray measurement. Phoswich detectors consist of a “sandwich” of phosphorescent materials (NaI and CsI), hence, the name “phoswich.” This detector took advantage of its thin construction and inherent anti-coincidence counting capabilities to greatly reduce the background in the low-energy region of the spectrum. In 1972, a dual phoswich system composed of two 5-in.-diameter detectors was placed in service. Although improvements were made in the analysis of data and calibration over the years, this basic system was used for chest counting for nearly 15 years until the adoption of planar germanium detectors in the late 1980s.

In the late 1970s, small coaxial germanium detectors were installed in the steel room. These detectors were pointed at the sides of the chest in an effort to detect the 186-keV photon from the  $^{235}\text{U}$  in EU, which was not readily detected by the phoswich or NaI detector. These detectors were also used to identify high-energy photon emitters, taking advantage of the superior resolution of the germanium detectors. In the mid-1980s, thick phoswich detectors specifically designed for EU were purchased. This turnkey system was used for only a few years before the germanium chest counter replaced it. In the early 1980s, solid-state planar germanium detectors were introduced at several facilities in the complex for chest counting. Routine use of a six-detector germanium chest counter began at SRS in about 1987 or 1988. This was a turnkey system purchased from a commercial vendor.

The utility of routine chest counting for actinides declined throughout the 1980s and early 1990s. This decline can be related to fundamental changes in the occupational internal dose limits. During the 1970s, the occupational limit for plutonium was expressed in terms of a quantity of plutonium that could be present in the chest at any given time – the maximum permissible lung burden (MPLB). The MPLB for plutonium was 16 nCi, which at the time was considered to be detectable with the dual phoswich system. Thus, compliance with the occupational dose limits could be demonstrated with a simple chest count. In 1980, the limits changed to annual effective dose equivalent, which caused an increase in the complexity of calculations but did not cause insurmountable problems because it is closely related to the MPLB. The routine chest count was still a valuable tool for demonstrating compliance. In 1992, the limits were changed to committed effective dose equivalent. The effect of this was to reduce drastically the amount of plutonium in the chest that corresponded to the occupational limit. The reduction was so great that chest counters could no longer detect plutonium at compliance levels, which rendered routine chest counts useless for determining compliance. However, chest counters still play an important role for special chest counts and for routine chest counts if a tracer such as  $^{241}\text{Am}$  might be present.

Germanium detectors have become the detectors of choice for measurement of actinide radioisotopes in the lungs. They are less efficient but much more specific (i.e., better resolution) than NaI or phoswich detectors. The SRS germanium detector system became operational in December 1989, replacing the phoswich detector system.

## 6.0 SUMMARY

Based on the various isotopes that were irradiated in the SRS reactors, it is clear that more than 100 nuclides have been identified as having been irradiated for the SRL and many offsite customers. As indicated in the tables, aside from the “production” radioisotopes, most materials that were irradiated were relatively small quantities, which were, according to Gray (2006), sealed in aluminum cans on arrival, handled under water, and shipped as soon as possible on removal from the reactor. During the early years, urine bioassay (either gross alpha or gross beta analyses) was the only method of evaluating intakes of radioactive materials. SRS built a whole-body counter in 1960 for detecting fission and activation products. Because the whole-body counter had limited use for the detection of actinides, efforts began in the mid-1960s to develop low-energy photon detectors that would permit detection of actinides in the chest. Improvements in whole-body counters and chest counters provided the means of detecting lower amounts of activities in the body. The RC Group provided training and retraining of all personnel and maintained records of internal and external exposures from the beginning of operations including exposures related to incidents.

SRL did handle high activities of actinides, such as isotopes of americium, curium, and californium, for which bioassay was established. A program involving  $^{210}\text{Po}$  was conducted by a small group of personnel for whom bioassay was conducted (Mahathy 2011). Whole-body counting was used to monitor intakes of most of the exotic irradiated targets. The SRL did early development work involving chemistry on  $^{171}\text{Tm}$  and broken wafers ( $^{170}\text{Tm}$ ), which was one-time, small-scale work. Other than that, no evidence was found that the SRL worked with exotic radionuclide sources in a manner that breached the integrity of the source. Although this was small-scale work, a method for bioassay for the  $^{170}\text{Tm}$  was available as indicated in Table 5-1. No evidence was found for bioassay for  $^{171}\text{Tm}$ ,  $^{249}\text{Bk}$ ,  $^{254}\text{Fm}$ ,  $^{147}\text{Pm}$ , and  $^{99}\text{Tc}$ . These five radionuclides are discussed in Attachments A through E. There were exotic radionuclides created at SRS for which no bioassay was performed (mostly beta-only emitters), there was no evidence found that indicated that bioassay was needed.

This report addresses  $^{249}\text{Bk}$  during the HFDP (1965 to 1970) (see Attachment C). There was significant production of  $^{249}\text{Bk}$  in the reactors in the 1970s, but nothing was found in the records that indicated the targets were processed in a way that purified the  $^{249}\text{Bk}$  or that pure  $^{249}\text{Bk}$  was handled in quantities that could have produced significant potential for intakes.

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**ATTACHMENT A  
TECHNETIUM-99**

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In the latter half of 1964 and into 1965 work was performed in the SRL on extraction of  $^{99}\text{Tc}$  from high-level waste (DuPont 1984b, pp. 94, 118; DuPont SRL Sep 1964,<sup>1</sup> p. 44). The feed material would have had plenty of fission products, and monitoring by fission product urinalyses and whole body counting would have been appropriate. In June 1964, 1.1 g of  $^{99}\text{Tc}$  was isolated and work toward extracting another 1.8 g was conducted in September (DuPont SRL Sep 1964, p. 44). The purity of the product was not mentioned. No mention of technetium extraction was made for subsequent years in the historical summary document (DuPont 1984b), and no mention was made in the series of quarterly reports of the Isotopic Power and Heat Sources (IPHS) Project starting in July 1966 (for instance, DuPont IPHS Jul–Sep 1966, DuPont IPHS Jan–Mar 1967, and others). [Note: the October to December 1966 report has not been captured.] In addition, no mention of work with  $^{99}\text{Tc}$  was made in any of the SRL RC Group monthly reports in 1966 (whereas there is frequent mention of work specifically with curium, thorium,  $^{238}\text{Pu}$ ,  $^{237}\text{Np}$  and occasional mention of work with thulium and polonium in 1966 or other years).

The work with  $^{99}\text{Tc}$  extraction appears to be limited from about midyear 1964 to about midyear 1965, when it appears work on this radionuclide was abandoned.

Because the specific activity of  $^{99}\text{Tc}$  is small (0.017 Ci/g), the potential for consequential intakes was smaller than for the other heat source radionuclides and was very small in terms of potential for inducing cancer. Table A-1 shows estimates of potential chronic intakes and committed organ doses from handling 1.8 g of  $^{99}\text{Tc}$  using the common Brodsky's "magic" number ( $10^{-6}$ ) to relate activity in process to potential for intakes (Brodsky 1980).

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<sup>1</sup> For citations of this form, see the Monthly and Quarterly Reports section at the end of the reference list.

**ATTACHMENT A  
TECHNETIUM-99**

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Table A-1. Potential inhalation committed organ doses (mrem) from handling 1.8 g <sup>99</sup>Tc.<sup>a</sup>

Target organs	Organ committed doses	
	Chronic intake (365 d), 83.7 pCi/d	
	Type F inhalation	Type M inhalation
Adrenals	3.86E-03	3.47E-03
Urinary bladder	1.60E-02	1.44E-02
Brain	3.86E-03	3.47E-03
Breast	3.86E-03	3.47E-03
Gall bladder	3.86E-03	3.47E-03
Heart wall	3.86E-03	3.47E-03
Kidneys	3.86E-03	3.47E-03
Liver	5.14E-03	4.63E-03
Muscle	3.86E-03	3.47E-03
Ovaries	3.86E-03	3.47E-03
Pancreas	3.86E-03	3.47E-03
Testes	3.86E-03	3.47E-03
Thyroid	1.01E-01	9.07E-02
Red bone marrow	3.86E-03	3.47E-03
Bone surface	3.86E-03	3.47E-03
Stomach	2.07E-01	1.89E-01
Small intestine	5.18E-03	6.24E-03
Upper large intestine	5.59E-02	5.99E-02
Lower large intestine	1.56E-01	1.69E-01
Skin	3.86E-03	3.47E-03
Spleen	3.86E-03	3.47E-03
Thymus	3.86E-03	3.47E-03
Uterus	3.86E-03	3.47E-03
Extrathoracic airways	1.92E-01	9.33E-01
Lung	6.10E-03	2.69E+00
Colon	9.94E-02	1.07E-01
Esophagus	3.86E-03	3.47E-03
Gonads	3.86E-03	3.47E-03

a. Maximum doses highlighted.

## ATTACHMENT B PROMETHIUM-147

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Promethium-147 was among the unusual nuclides that were produced in research quantities and evaluated for a variety of uses from 1965 to 1970. Promethium-147 has a half-life of 2.6 years and is a beta emitter with an average energy of 62 keV. The low energy allows for ease of handling and does not require significant shielding, which, in turn, reduces the weight required of other radioisotopes for similar applications.

As early as 1958 there was an interest in using the fission product  $^{147}\text{Pm}$  as a fuel for Systems for Nuclear Auxiliary Power (SNAP) batteries. As a result, a process for the recovery of  $^{147}\text{Pm}$  was started. At that time a method was also developed for the radiochemical determination of  $^{147}\text{Pm}$ , along with  $^{144}\text{Ce}$  and  $^{91}\text{Y}$ , which were the major contaminants of promethium (DuPont 1984b, p. 23). As can be seen in Table 3-4  $^{147}\text{Pm}$  was identified several times in 1959 and 1960. However, it was not until 1965 that studies began on the use of various radionuclides as heat source materials, including  $^{147}\text{Pm}$ .

Because the National Aeronautics and Space Administration (NASA) had an interest in aged  $^{147}\text{Pm}$  as a source of isotopic heat for space missions, a study of the quantity and quality of  $^{147}\text{Pm}$  that could be produced annually at SRP was undertaken. The major source of  $^{147}\text{Pm}$  for its recovery would be fission product waste. The quantity available after aging to allow for decay of associated  $^{148}\text{Pm}$  would be about 1,700 W(t) per reactor-year of operation. Product quality from current waste in 1965, as represented by a low ratio of  $^{148}\text{Pm}/^{147}\text{Pm}$ , would presumably be the same as that produced at Hanford (about  $5 \times 10^{-7}$ ). Promethium-148 emits gamma rays that could require extra shielding, while  $^{147}\text{Pm}$  is a beta emitter as indicated above. In addition, a scoping study was made to determine the quality of  $^{147}\text{Pm}$  in fission product sludge from SRP operation. The measured isotopic ratio of  $^{148}\text{Pm}$  to  $^{147}\text{Pm}$  was found to be similar to the isotope ratio in Hanford fission products (0.4 ppm) (DuPont 1974, p. 7).

A process to recover  $^{147}\text{Pm}$  from plant high-activity waste was also developed in the laboratory in 1965 for use as a power source. During the extraction process, the  $^{147}\text{Pm}$  had to be separated from  $^{144}\text{Ce}$  since that is a major source of heat and its removal would allow easier storage of the  $^{147}\text{Pm}$ . After separation the  $^{147}\text{Pm}$  was then stored until most of the  $^{148}\text{Pm}$  impurity decayed and was then purified by ion exchange (DuPont 1984b, p.119). Starting in February 1965, an estimate was made of the annual production at SRP (in the fission product fraction) of  $^{147}\text{Pm}$ , and the recovery of  $^{147}\text{Pm}$  from current high-activity waste was studied (DuPont SRL Feb 1965, pp. 37–40; DuPont SRL Aug 1965, pp.43, 45). The February study was conceptual; work at the tracer level started in August. The early work had considerable  $^{144}\text{Ce}$ , so intake would have been monitorable by whole body counting (DuPont SRL Dec 1965, pp. 29–32). The isotopic purity of aged  $^{147}\text{Pm}$  (from the SRP waste tanks) was also measured (DuPont 1974, pp.5–6; DuPont SRL Mar 1965, p. 24). Laboratory-scale testing and improvements to the chemical separation procedure continued in 1966, still with issues with  $^{144}\text{Ce}$  in the  $^{147}\text{Pm}$  fraction (DuPont SRL Feb 1966, p. 20; DuPont SRL Sep 1966, pp. 23–24). A plant test to demonstrate the initial recovery steps was begun in November 1966 and demonstrated the recovery of a  $^{147}\text{Pm}$ -La mixture from plant waste (DuPont 1984b, p. 142; DuPont SRL Nov 1966, p. 33). Work continued in the laboratory on a method to remove the cerium (DuPont SRL Nov 1966, p. 34). The first test with essentially pure  $^{147}\text{Pm}$  occurred in June 1967 involving about 500 Ci (DuPont SRL Jun 1967, pp. 26–27). By late 1967 another test of the process to recover  $^{147}\text{Pm}$  from high-activity waste was successful in producing about 500 g from a lanthanide mixture. The first steps of the process were conducted in F Area to produce the Pm-La mixture, which underwent multiple extractions (back-extractions) to remove 80% to 90% of the cerium. The resulting material was to be stored 2 years for decay of  $^{148}\text{Pm}$ . Laboratory tests then demonstrated that pressurized ion exchange columns could be used for final purification of  $^{147}\text{Pm}$ . The initial separation steps also captured  $^{238}\text{Pu}$

**ATTACHMENT B  
PROMETHIUM-147**

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in a separate fraction (DuPont SRL Sep 1967, pp. 32–33; DuPont SRL Dec 1967, p. 37; DuPont 1984b, p. 163). Although the  $^{148}\text{Pm}$  contaminant in the Pm-La mixture is a strong gamma emitter and was of concern for the desired use of the  $^{147}\text{Pm}$ , the activity ratio was about 0.002  $^{148}\text{Pm}/^{147}\text{Pm}$ , so whole body counting would not have been a sensitive bioassay for this mixture; however, even with 80% to 90% removal, there was still considerable  $^{144}\text{Ce}$  (a little more than 1,700 Ci/g  $^{147}\text{Pm}$  or about 2/1 activity ratio  $^{144}\text{Ce}/^{147}\text{Pm}$ ) (DuPont SRL Apr 1968, p. 17). Whole body counting would have been an adequate bioassay for the Pm-La mixture in the F Area.

For another application, suitable isotopic purities were calculated for  $^{238}\text{Pu}$ ,  $^{171}\text{Tm}$ , and  $^{147}\text{Pm}$  as power sources for pumps to assist or replace the heart. A heart pump would be about 5 in. in diameter and installed in the abdomen. It would include a steam boiler and heat exchanger with blood as coolant (DuPont IPHS Oct–Dec 1967, p. 45). Plutonium-238 appeared to be the most promising power source for such pumps because it would be available in adequate supply and could be aged to remove  $^{236}\text{Pu}$  to sustain long cycles of operation with reasonable weight of shielding. Thulium-171 would require potentially the least shielding, but was expected to be more expensive than  $^{238}\text{Pu}$  in the necessary, highly purified form and was also handicapped by much shorter half-life (1.9 vs. 87 years). Promethium-147, readily available as a fission product, had similar disadvantages of being difficult and more expensive to produce the necessary isotopic purification required for minimum shielding, and it also has a shorter half-life (2.6 years). In 1968,  $^{147}\text{Pm}$  was also considered as a heat source for thermoelectric and thermionic power generation. Studies of compounds of promethium were conducted to provide information on the optimum fuel form selection (DuPont 1984b, p.182). A January to March 1968 plant test of the process in H Area recovered about 2 kg of  $^{147}\text{Pm}$  as a lanthanide mixture. In laboratory tests in April 1968 pressurized ion exchange columns were used to isolate approximately 1 g of  $^{147}\text{Pm}$  from a portion of the 500-g lanthanide mixture that had been created the previous December (DuPont 1984b, p. 182; DuPont SRL Apr 1968, pp. 15–17). The feed for these tests had 1,700 Ci of  $^{144}\text{Ce}$  per g  $^{147}\text{Pm}$ , but the final product had less than 1 mCi  $^{144}\text{Ce}$  per g  $^{147}\text{Pm}$  and similarly small amounts of  $^{106}\text{Ru}$ ,  $^{95}\text{Zr}/\text{Nb}$ , and  $^{154}\text{Eu}$  (DuPont SRL Apr 1968, p. 17). No further work with pure  $^{147}\text{Pm}$  was found in the SRL monthly reports for the remainder of 1968 or for 1969 to 1971.

While there was considerable development work with  $^{147}\text{Pm}$ , there is no record that indicates it was ultimately applied to the above-mentioned applications. The Pm-La separations of 500 g and 2 kg in different operations in the F and H Areas represented multicuries of  $^{147}\text{Pm}$  activity, but there was sufficient  $^{144}\text{Ce}$  contamination for adequate bioassay monitoring by whole body counting. The smaller amounts of pure  $^{147}\text{Pm}$  that were used for methods testing in June 1967 (about 500 Ci) and April 1968 (about 1,000 Ci) were too pure to use whole body counting for bioassay. These tests were conducted in the SRL.

In 1969 work on the development of  $^{147}\text{Pm}$  as a heat source was concluded and no further work was planned (DuPont 1984b, pp. 193, 200–201). In 1971 about 460 g  $^{147}\text{Pm}$  and 100 g  $^{144}\text{Ce}$ , stored for several years, were discarded to make room for  $^{244}\text{Cm}$  (DuPont 1971, p. 8).

## ATTACHMENT C BERKELIUM-249 AND FERMIUM-254

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Berkelium-249 and  $^{254}\text{Fm}$  were two of many radionuclides created as part of the Transplutonium Project, evolving out of the work to create  $^{252}\text{Cf}$  and to produce 3 kg of  $^{244}\text{Cm}$  by 1967 (DuPont SRL Sep 1964, p. 25). Berkelium-249 resulted from decay of  $^{249}\text{Cm}$ , which was produced from multiple (n, $\gamma$ ) reactions starting from  $^{244}\text{Cm}$ . Berkelium-249 emits a weak beta particle (average energy 33 keV) and no photons. Its progeny is  $^{249}\text{Cf}$ , which is an alpha emitter that would have been counted in the trivalent actinide bioassay procedure, but the long half-life (351 years) precludes ingrowth of sufficient activity to be useful for bioassay monitoring.

Fermium-254 resulted from multiple (n,  $\gamma$ ) reactions starting from  $^{252}\text{Cf}$  with the final two steps being an (n, $\gamma$ ) reaction of the  $^{253}\text{Es}$  and beta decay of  $^{254}\text{Es}$  (DuPont 1971, p. 38). Fermium-254 (half-life 3.2 hours) was an intermediate step in the pathway to produce  $^{257}\text{Fm}$  (half-life 84 days). It is an alpha emitter, but the short half-life precludes direct bioassay monitoring. Its progeny  $^{250}\text{Cf}$  is also an alpha emitter, but the difference in half-lives (3.2 hours versus 13.1 years) means the  $^{250}\text{Cf}$  activity would be insignificant. The transplutonium historical document states that "only  $^{257}\text{Fm}$  can be recovered in significant amounts because, even though it is at the end of the reaction chain, its 84-day half-life is much longer than those of the other fermium isotopes" (DuPont 1971, p. 39).

### Berkelium-249

The SRL monthly report for September 1964, in a section on the use of the transuranic metal extraction (TRAMEX) process for isolation of californium, stated, "Continuing analytical development study will include determination of Bk and Es, in addition to Cf, in the dissolver solution from the second Transplutonium campaign" (DuPont SRL Sep 1964, p. 34). There was no mention of berkelium in the October and November monthly reports for 1964 (DuPont SRL Oct 1964; DuPont SRL Nov 1964). "A series of laboratory tests was made in support of the second Transplutonium separations campaign, which was started in the H Area in October." These tests focused on the separations chemistry of plutonium, americium, and curium (DuPont SRL Nov 1964, p.38). In January 1965 a 0.03- $\mu\text{g}$  sample of  $^{252}\text{Cf}$  was extracted from curium and extraction of a very small amount of berkelium and einsteinium from the californium sample was begun (DuPont SRL Jan 1965, pp. 7, 14-18). This work continued through March; the activity of the extracted  $^{249}\text{Bk}$  was mentioned as about 2,600 cpm. The counters were not calibrated for  $^{249}\text{Bk}$  so the exact activity was not given, but it was probably 1 to 10 nCi (DuPont SRL Mar 1965, pp. 14-16). No mention is made in subsequent SRL monthly reports until March 1966, although the January 1966 monthly report states that a transplutonium campaign was started in the reactor in February 1965 and was due to be removed in February 1966 (DuPont SRL Jan 1966, p. 5). The historical summary document of the Separation Technology Section indicated that in 1965 analyses were developed for  $^{249}\text{Bk}$  and  $^{253}\text{Es}$  as part of the Transplutonium Project (DuPont 1984b, pp. 111-112). The March 1966 SRL monthly report shows that some  $^{249}\text{Bk}$  from LRL had been combined with a small amount produced at SRS (presumably from the 0.03- $\mu\text{g}$   $^{252}\text{Cf}$  sample) to produce a thin target for cross-section studies. The target was placed in the reactor in February 1966. It contained 485 ng (790  $\mu\text{Ci}$ ) (DuPont SRL Mar 1966, pp. 14-16). In February 1967, mass spectrometry was performed on a sample from an irradiated  $^{238}\text{Pu}$  target (P-5) to determine the yields of isotopes of californium, einsteinium, and berkelium. The entire P-5 element had 88  $\mu\text{g}$  of  $^{249}\text{Bk}$  but, of course, the analyzed sample had much less. The sample also had considerable alpha activity from the isotopes of californium and einsteinium (DuPont SRL Feb 1967, pp. 21-22). In May 1967 a similar analysis was performed on two samples from the P-6 element showing similar isotopic abundances of californium, einsteinium, and berkelium (DuPont SRL May 1967, p. 18). Another cross-section study was performed in August 1967 using two  $^{249}\text{Bk}$  targets containing 175  $\mu\text{g}$  (285 mCi) and 105  $\mu\text{g}$  (171 mCi) of  $^{249}\text{Bk}$ , respectively. Chemistry was involved in the target preparation (DuPont SRL Aug 1967, pp. 39-42).

**ATTACHMENT C**  
**BERKELIUM-249 AND FERMIUM-254**

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The chapter in the 50-year symposium document that discusses creation of curium and californium states, "In 1968-69, a high-pressure elution development cation exchange chromatographic process was developed. ... Milligram quantities of  $^{252}\text{Cf}$ , submilligram quantities of  $^{249}\text{Bk}$ , and subnanogram amounts of  $^{253}\text{Es}$  and  $^{254}\text{Fm}$  were separated from ...  $^{244}\text{Cm}$  and  $^{243}\text{Am}$  ..." (Harbour et al. 2000, p. 172). There is no mention of work with berkelium, einsteinium, or fermium in the SRL monthly reports from September 1967 through February 1968. In March 1968 the yield of berkelium in the curium campaign of 1967 was determined by analyzing a small sample from the stored solution. "The analytical method was also used to trace berkelium in the TrameX process." These were tests in preparation for extraction of larger amounts of berkelium from the TRAMEX process (DuPont SRL Mar 1968, pp. 9–10). Additional tests (scanning coulometry and mass spectrometry) were run on berkelium a year later, but information was not available on the amounts handled in these tests (DuPont SRL Mar 1969, p. 5; DuPont SRL Apr 1969, pp. 6–8.) Use of the pressurized cation exchange chromatographic process first shows in the October and December 1969 monthly reports. However,  $^{250}\text{Bk}$  was used as the tracer in these tests. The  $^{250}\text{Bk}$  was made from irradiation of a  $^{249}\text{Bk}$  target. Improvements in counting  $^{249}\text{Bk}$  using liquid scintillation counters were also being made at this time (DuPont SRL Oct 1969, p. 5; DuPont SRL Dec 1969, pp. 7–8).

Improvements in the chemical separation techniques were being tested in April 1970 "for isolating  $^{249}\text{Bk}$ ,  $^{254}\text{Es}$ , and  $^{257}\text{Fm}$  produced with the  $^{252}\text{Cf}$  in the present high flux campaign ...." That same monthly report also mentions that the  $^{250}\text{Bk}$  tracer (mentioned earlier) was created from irradiation of a 10- $\mu\text{g}$  (16 mCi)  $^{249}\text{Bk}$  target (DuPont SRL Apr 1970, pp. 18–19; DuPont 1971, pp. 37–40). The chemical separation techniques were apparently intended to be used in the Multipurpose Processing Facility, under construction in 1970 in the F Area hot canyon (DuPont 1984b, pp. 209–210). In this period SRS was also irradiating targets that were shipped to ORNL for separation of the berkelium, fermium, and einsteinium (DuPont 1971, pp. 39–40), and then, ironically, ordering the berkelium back from ORNL (DuPont SRL Jul 1970, p. 6; DuPont SRL Aug 1970, p. 6). The shipment from ORNL involved 250  $\mu\text{g}$  (407 mCi) of  $^{249}\text{Bk}$  (DuPont SRL Aug 1970, p. 6). More separations were performed in September in what were still referred to as tests. "Batch 9 containing 1.25 mg of  $^{252}\text{Cf}$  from the  $^{244}\text{Cm}$  campaign was purified by pressurized ion exchange chromatography with alpha-hydroxyisobutyrate (AHIB). This test confirms feasibility of separating Bk, Es, and Fm in the present  $^{252}\text{Cf}$  campaign." The amount of  $^{249}\text{Bk}$  separated was not given, but it would have been much less than the 1.25 mg of californium (DuPont SRL Sep 1970, pp. 6–7).

The Separation Technology Section history indicates that work on isolating berkelium, einsteinium, and fermium continued into 1971 (DuPont 1984b, p. 225). The April SRL monthly report confirms two additional tests on separation of  $^{249}\text{Bk}$  from  $^{252}\text{Cf}$  and  $^{154}\text{Eu}$  (apparently a stubborn contaminant) referred to as "small-scale" tests versus the "large-scale" tests in September 1970 (DuPont SRL Apr 1971, pp. 7–8). The Transplutonium Project history document, drafted in February 1971, indicates the "Californium I campaign is expected to yield 51 milligrams of  $^{249}\text{Bk}$ , 6 micrograms of  $^{254}\text{Es}$ , and 10 picograms of  $^{257}\text{Fm}$  ( DuPont 1971, p. 40). Note that no mention was made of  $^{254}\text{Fm}$ . There are conflicting reports on the time of the Californium I campaign. The February 1971 document refers to it in the future, and other documents show californium work underway in early 1971 (DuPont WTD Feb 1971, pp. 5–6, 8, 13). However, the reactor history states that the Californium I campaign occurred from August 1969 to November 1970 (DuPont 1984c, p. 158). The difference might be the time the targets were in the reactor versus the time the targets were being analyzed after a cooling period. The later article on this work (from about 2000) refers to submilligram quantities of  $^{249}\text{Bk}$  rather than 10s of milligrams (Harbour et al. 2000, p. 172), so it appears that the predicted amounts in the Californium I campaign were either not created or not recovered in the amounts expected. A presentation on the history of berkelium and californium through the end of 1974 stated that the

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material from the Californium I campaign was sent to ORNL for separation and purification of the berkelium in the Transplutonium Processing Plant (Crandall 1975, p. 11).

There is no indication that the elements higher than californium were actually isolated in the Multipurpose Processing Facility when it was operating. SRL monthly reports in 1972 and 1973 indicate that production was focused on  $^{238}\text{Pu}$  heat sources and  $^{252}\text{Cf}$  neutron sources. The 1975 historical presentation on berkelium and californium stated that "Slide 15 illustrates one of the latest flow sheets, that for the completed, but as yet unused, Multipurpose Processing Facility (MPPF) at Savannah River" (Crandall 1975, p. 11). Presumably "unused" referred to berkelium rather than californium. The 1982 risk analysis report for the facility indicated that the plant was capable of extraction of curies of berkelium from the plant feed, but it did not state that that had actually occurred (Stoddard 1982).

A waste characterization report in 1994 indicated that  $^{249}\text{Bk}$  was still being produced in the reactors in the 1970s. Specifically, Figure 4 of that document shows  $2.59 \times 10^5$ ,  $1.04 \times 10^4$ ,  $1.16 \times 10^4$ , and  $95 \mu\text{g}$  of  $^{249}\text{Bk}$  in the "receipt/production" column for fiscal years 1976, 1977, 1978, and 1980, respectively (Gibbs 1994, pp. 8, 19). The series of annual reports showing customers and shipments of various isotopes made in DOE laboratories for 1972 through 1980 showed that all  $^{249}\text{Bk}$  was made at ORNL and that SRS only received berkelium in 1972 (Simmons 1973, p. 14; Simmons and Mandell ca. 1975, p. 12; Simmons and Gano ca. 1976, p. 13; Simmons 1977, p. 13; Simmons 1978, p. 15; Burlison and Laidler 1979, p. 15; Burlison 1980, p. 11; Burlison 1981, p. 12). Therefore, the amounts in the Gibbs document must have been produced and used internally at SRS, which is consistent with the statement in the Gibbs document that the  $^{249}\text{Bk}$  was "tracked in the Accountability records and obtained from the reactor production codes." This is verified by the SRS Automated Inventory Management System (AIMS) monthly reports and the Nuclear Materials Management and Safeguards System (NMMSS) (DOE 2012a,b) files. For instance, the October 1975 AIMS report listed 65 "pieces" (presumably targets) in K-Reactor amounting to about  $2.95 \times 10^5 \mu\text{g}$ , which is nearly twice the inventory in the Gibbs document for the end of June 1975 (DuPont 1975–1979, pp. 2–5). The difference, according to the monthly NMMSS reports, was production of  $8.5 \times 10^4$ ,  $1.8 \times 10^4$ , and  $1.7 \times 10^4 \mu\text{g}$  in July, August, and September 1975, respectively. This production rate continued through December 1975 and occurred again at about the same rate during March through October 1976 (DOE 2012a). The NMMSS report lists this production as an SRS-to-SRS transaction of "in fuel element and target fabrication process," consistent with irradiation of targets in one of the reactors. The AIMS report for October 1976 listed the same 65 pieces in K-Reactor with an inventory increase to  $4.08 \times 10^5 \mu\text{g}$  and 24 pieces in P-Reactor with a small inventory of  $277 \mu\text{g}$ , which is consistent with the inventory in the Gibbs document for the same time (DuPont 1975–1979, pp. 22–24). The October 1977 AIMS report shows the 65 pieces still in K-Reactor but with much less activity, about  $3.10 \times 10^5 \mu\text{g}$ , which agrees well with the Gibbs report for calendar year 1977. This amount implies that there was some continued production of the  $^{249}\text{Bk}$  in the reactor for some of the year, followed by radioactive decay. So the targets were still at K-Reactor but not being irradiated at that time. There were also 23 pieces in P-Reactor amounting to about  $2,170 \mu\text{g}$ , implying that the targets in P-Reactor had continued to be irradiated during the year (DuPont 1975–1979, pp. 35–37). The NMMSS reports show continued production through January 1978, although the amounts are much smaller, generally in the hundreds of micrograms per month (as opposed to tens of thousands of micrograms per month in the 1975 to 1976 period). This production was listed as "in reactor, product" (DOE 2012a), and is consistent with the smaller targets in P-Reactor. In February and March 1978 there was a production of only  $3 \mu\text{g}$  each month, also listed as "in reactor, product," then there was no more production of this type. The last production occurred in November 1979, consisting of  $95 \mu\text{g}$  listed as "aluminum process, irradiated," which is consistent with the Gibbs report (DOE 2012b).

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None of the reports are completely clear as to what happened to the targets after irradiation (production). There were no shipments off the site from 1974 through 1992. All of the transactions are listed as from and to 703-42A with the same shipper and receiver code except for two shipments in 1979 listed as removals to "underground waste storage – liquid" (DOE 2012a). A little of the material is listed as being removed due to "fission and transmutation," which would have been occurring while the targets were still in the reactors. Most of the inventory is just shown as decaying away with only 7 µg left by the end of 1992 (DOE 2012b). The decay bookkeeping category does not preclude having the targets chemically manipulated and the <sup>249</sup>Bk purified. In the transaction reports, there is mention of production of a small amount of oxides (totaling 15 µg) in 1974 and a few listings of production of "flats, tubes, and extruded metal" in amounts varying from <1 to 44 µg in late 1975 through June 1976 (DOE 2012a). This might indicate some manipulation of the targets, although without chemical purification <sup>249</sup>Bk would not have been the predominant radionuclide. The category "aluminum process, irradiated" first shows in December 1978 with a small inventory of 95 µg that grew to 1,378 µg by March 1979. The inventory in this category was then reduced consistent only with radiological decay through 1989 when it decayed below 1 µg (DOE 2012b). So the dates of the first entries of "aluminum process, irradiated" align somewhat with the dates of transfer of some material to underground waste storage. This might indicate that some of the targets were chemically manipulated in late 1978 and 1979, producing some waste.

The NMMSS report covering 1993 to present (DOE 2012c) shows the continuation of the decay of the 7 µg of material shown in the 1974 through 1992 report, down to 4 µg by August 1993. Then in October 1993 the inventory jumps to 58 µg listed as burial material. This inventory stays on the books undecayed through June 2006. The transaction report for July 2006 shows all 58 µg being removed without any description or movement so presumably this was to show that the buried material had decayed away (DOE 2012d). Throughout this period there is no movement of material off the site or between material accountability areas.

From an intake perspective, it is important to note concerning general contamination in gloveboxes, caves, accessible surfaces, and air contamination, that the overwhelming constituents would have been <sup>244</sup>Cm, <sup>252</sup>Cf, and <sup>243</sup>Am, which were being handled in large amounts. For instance, disposal of <sup>244</sup>Cm contaminated waste to the burial grounds in 1969 totaled 12,185 Ci (DuPont RC Dec 1969, p. 8). The RC Group monthly reports show that handling of <sup>252</sup>Cf in the hundreds of curies range in the SRL was common during this period. Possible exposure to pure submilligram quantities of <sup>249</sup>Bk (1.6 Ci/mg) or the microgram/picogram quantities of einsteinium and fermium would have occurred only to those handling and packaging the final products of the chemical process, which would have been handled with extreme care because of the rarity of the products. For instance, there is no mention of contamination by berkelium, einsteinium, or fermium in the April 1970 RC Group report (DuPont RC Apr 1970) (when the highest activity tests were conducted), or in any of that series of reports in 1969 or 1971.

Possible exposure to pure <sup>249</sup>Bk appears to have occurred at nanocurie levels in 1965, sub millicurie levels in 1966, and tens to hundreds of millicurie levels in 1967 and intermittently thereafter through 1971. There was continued production of <sup>249</sup>Bk through irradiation of targets in the reactors from 1974 to 1979. No conclusive evidence has been found that <sup>249</sup>Bk was extracted from those targets, but the transfer of 58 µg to underground storage, apparently as a liquid, seems to indicate some chemical processing. Most of the about  $1 \times 10^5$  µg of material made in the 1970s seems to have just decayed away with some being transferred to waste storage. No facilities other than 703-42A are mentioned in the transaction reports. General contamination in the SRL workspace would have been dominated by other radionuclides, principally <sup>244</sup>Cm, <sup>252</sup>Cf and other isotopes of californium, <sup>243</sup>Am, and <sup>154</sup>Eu; the

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risk of intake from the small amounts of pure  $^{249}\text{Bk}$  seem low. It seems unlikely that there was exposure to purified  $^{249}\text{Bk}$  after 1972; the 1972 date is based on receipt of a shipment from ORNL in 1972.

#### Fermium-254

Both  $^{254}\text{Fm}$  and  $^{257}\text{Fm}$  are alpha emitters so, if present in the body, would have been excreted in urine and in theory would have been measured by the trivalent actinide urinalysis. However, because of its 3.2-hour half-life, essentially none of the excreted  $^{254}\text{Fm}$  would have been counted considering the time gap between excretion and counting of the sample. However, the same half-life consideration applies to time involved in separating and extracting the fermium from the californium and einsteinium. For instance, the 1971 history indicates that a 15-month cooling period between the time the targets were removed from the reactor until the start of chemical processing was normal (DuPont 1971, p. 40). If hardly any cooling time was employed, then no  $^{254}\text{Fm}$  would be left in the target (the parent  $^{254}\text{Es}$  also has a half-life in hours). Although the article from 2000 states "subnanogram amounts of ...  $^{254}\text{Fm}$  were separated" (Harbour et al. 2000, p. 172), that article does not mention  $^{257}\text{Fm}$ , which was the actual goal of the irradiation of einsteinium. It is possible that this was a typographic error and the sentence should have said  $^{257}\text{Fm}$ . The purpose of creating the higher order elements was for research on their physics and chemical natures, which would not have been possible on a very small amount of something with a 3.2-hour half-life.

Tracer quantities of  $^{254}\text{Fm}$  were created twice in 1970 (April and September) for the special purpose of testing the procedure for separating fermium and einsteinium from californium. The activity of  $^{254}\text{Fm}$  was about 1  $\mu\text{Ci}$  in the April test and about 20  $\mu\text{Ci}$  in the September test (DuPont SRL Apr 1970, pp. 21–23; DuPont SRL Sep 1970, pp. 6–7; DuPont SRL Oct 1970, pp. 9–10). These activities of such a short half-life radionuclide material are not of dosimetric concern. It is concluded that no significant exposure occurred to  $^{254}\text{Fm}$  as a pure radionuclide.

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THULIUM-171**

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Interest in  $^{171}\text{Tm}$  (and  $^{170}\text{Tm}$ ) as potential heat sources started in 1964. In the September monthly report of the SRL, under New Products – Special Test Irradiations, “quantities of  $^{46}\text{Sc}$ ,  $^{170}\text{Tm}$ ,  $^{171}\text{Tm}$ , and  $^{204}\text{Tl}$  are being made in the current  $^{244}\text{Cm}$ -producing charge” (DuPont SRL Sep 1964, p. 43), a test that was authorized in July (Allen 1964). The October and November monthly reports do not mention any actual handling of the material (DuPont SRL Oct 1964; DuPont SRL Nov 1964). An historical summary document also mentions the above irradiation in 1964 and states that an ion exchange process for separating the thulium from the erbium target had been developed and would be used on the irradiated material (DuPont 1984a, p. 93).

The first chemistry to extract  $^{170}\text{Tm}$  and  $^{171}\text{Tm}$  from the irradiated erbium targets occurred in 1965 using the aforementioned technique. This was at the laboratory development phase (DuPont 1984a, p. 118; DuPont SRL Feb 1965, pp. 43–44; DuPont SRL Apr 1965, p. 41). During this period the extracted material consisted of a combination of  $^{170}\text{Tm}$  and  $^{171}\text{Tm}$  because the target material was natural erbium. Because  $^{170}\text{Tm}$  is a strong beta emitter, intakes of this mixed isotopic material would have been detected by both air sampling and the fission product bioassay. Experimentation with enriching erbium to enhance the abundance of  $^{170}\text{Er}$ , thereby producing more  $^{171}\text{Tm}$ , began in the second half of 1965 (DuPont SRL Jun 1965, pp. 14–22; DuPont SRL Jul 1965, p. 38; DuPont SRL Sep 1965, p. 56; DuPont SRL Nov 1965, p. 31). Much of that work was on unirradiated erbium. Experimentation and improvement of this technique continued in 1966 (DuPont 1984a, p. 141). Experiments in January resulted in separation of 2.7 mg (2.9 Ci) of  $^{171}\text{Tm}$  (DuPont SRL Jan 1966, pp. 21–22). This material was used as a tracer in subsequent tests toward improving separation techniques (DuPont SRL Apr 1966, pp. 12–15). The RC Group monthly report for May 1966 indicates that a study on the radiation characteristics from and shielding design needed for  $^{170,171}\text{Tm}$  sources was underway (DuPont RC May 1966, p. 8). In June a different approach involving irradiation of  $^{169}\text{Tm}$  to produce  $^{171}\text{Tm}$  was started, which was estimated to produce about 500,000 Ci of  $^{170+171}\text{Tm}$  in each of two targets. In addition, irradiation of two 1-g samples of erbium oxide enriched in  $^{170}\text{Er}$  began (DuPont SRL Jun 1966, pp. 9–15). The irradiated  $^{169}\text{Tm}$  targets were later shown to contain mostly  $^{170}\text{Tm}$  (DuPont SRL Dec 1966, p. 9). The RC Group December 1966 monthly report indicated that proposed operations for about 100 Ci of  $^{171}\text{Tm}$  in gloveboxes B-126 to B-130 were approved and that the associated beta and low-energy gamma activity was to be easily shielded. The report added that the aluminum cladding was to be removed in the High Level Caves (DuPont RC Dec 1967, p.10).

Analyses of the irradiated enriched erbium oxide samples were conducted in May and June of 1967; these showed that nearly pure  $^{171}\text{Tm}$  was produced (DuPont SRL May 1967, p. 9; DuPont SRL Jun 1967, p. 8; DuPont IPHS Apr–Jun 1967, p. 21; DuPont 1984a, p. 163). Two hundred Ci of  $^{171}\text{Tm}$  had been produced by this method at that time. In the following quarter about 60 Ci of this material was purified from the residual erbium using cation exchange chromatography (DuPont IPHS Jul–Sep 1967, p. 22; DuPont SRL Jul 1967 p.14). No work with  $^{171}\text{Tm}$  was mentioned in subsequent SRL monthly reports until May 1968; it was also not mentioned in the fourth quarter (calendar year) 1967 IPHS report. The latter report indicated that  $^{238}\text{Pu}$  would be a better heat source than either  $^{171}\text{Tm}$  or  $^{147}\text{Pm}$  because of their short half-lives, difficulty in separation and purification, and cost (DuPont IPHS Oct–Dec 1967, p. 45). The first quarter 1968 IPHS report only discusses work with  $^{244}\text{Cm}$  and  $^{170}\text{Tm}$ ; external radiation measurements were made on a 3-kCi  $^{170}\text{Tm}$  source that also contained 1.7 kCi of  $^{171}\text{Tm}$ , but no chemistry was done (DuPont IPHS Jan–Mar 1968, p. 29; Sanders et al. 1969). In May 1968 another separation of  $^{171}\text{Tm}$  from enriched erbium was made with improved chemistry methods (30 mg  $^{171}\text{Tm}$ , 57 Ci) (DuPont IPHS Apr–Jun 1968, pp. 45–46; DuPont SRL May 1968, pp. 12–13). Both of these references also stated that this concluded the work with  $^{171}\text{Tm}$ . That the work was “deferred” was reiterated in a July 1969 letter from E. L. Albenesius, Research Manager in the SRL (Albenesius 1969). No further work with potential for intakes of pure  $^{171}\text{Tm}$  was found.

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**THULIUM-171**  
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No mention of the work with  $^{171}\text{Tm}$  was found in the series of monthly reports by the RC Group of the SRL in 1968 or 1969, which merely indicates that there was no significant loss of control.

Bebbington, in his history of DuPont years at Savannah River, concluded "among the unusual radionuclides produced in research quantities and evaluated as heat sources and for a number of other specialized uses were: curium-244, promethium-147, thulium-170 and -171, scandium-46, thallium-204, and polonium-210. ... None of these nuclides, though, found significant applications" (Bebbington 1990, p. 200).

The period of work with potential for intakes of essentially pure  $^{171}\text{Tm}$  appears to be limited to 1966 through 1968. The work took place in the SRL.

## ATTACHMENT E AMERICIUM-242M

Americium-242m is a beta emitter with a half-life of 152 years. It has an alpha decay branch, but the yield is only 0.5%. Because it does not emit significant alpha radiation, it would not be counted in the trivalent actinide bioassay method. The principal decay branch of  $^{242m}\text{Am}$  (99.5% yield) beta-decays to  $^{242}\text{Am}$ , which beta-decays to  $^{242}\text{Cm}$  with an 82.7% yield and 16-hour half-life. So this latter transition is nearly instantaneous, and equilibrium between  $^{242m}\text{Am}$  and  $^{242}\text{Am}$  is rapidly established. Curium-242 is an alpha emitter with a 163-day half-life that would have been counted by the trivalent actinide bioassay analysis. However, it takes quite a few years for equilibrium to be established between  $^{242m}\text{Am}$  and  $^{242}\text{Cm}$ , and equilibrium is never 100% because of the branching ratios.

The question is whether the buildup of  $^{242}\text{Cm}$  is rapid enough and sufficient such that the trivalent actinide bioassay would have been an appropriate bioassay for  $^{242m}\text{Am}$  exposure. Because americium and curium behave the same in the body, body biokinetics do not change the ratio of  $^{242}\text{Cm}$  and  $^{242m}\text{Am}$  as a function of time. Assuming workers were chronically exposed to initially pure  $^{242m}\text{Am}$ , the rate at which  $^{242}\text{Cm}$  activity would build up in the body is shown in Table E-1, assuming a unit activity intake per day of  $^{242m}\text{Am}$ .

Table E-1. Activity of  $^{242}\text{Cm}$  per unit daily intake of  $^{242m}\text{Am}$ .

Time after start of chronic intake	Cm-242 activity
1 d	0.0018
5 d	0.0087
10 d	0.017
50 d	0.082
90 d	0.140
180 d	0.249
1 yr	0.407
1.5 yr	0.504
2 yr	0.571
3 yr	0.648

The derivations and a plot of the buildup as a function of time are shown in La Bone (2011). It is unlikely that the  $^{242m}\text{Am}$  would have been pure in the workplace environment; that is, airborne or surface contamination would have been a mix of  $^{242m}\text{Am}$  and  $^{242}\text{Cm}$ , so the actual ratios would have been larger than those in Table E-1, up to an equilibrium of 0.83. The activity values in Table E-1 show that an annual or biennial trivalent actinide bioassay would have had sufficient  $^{242}\text{Cm}$  to monitor for chronic intakes of  $^{242m}\text{Am}$ .