LA-UR-12-22883
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Title: Expanding OSRP's Mission in Recovering New Isotopes for WIPP Disposal

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Intended for: INMM Conference, 2012-07-15/2012-07-19 (Orlando, Florida, United States)

Abstract

This presentation to the INMM Conference July 15, 2012, is a discussion on expanding the OSRP's mission by including Cm-244 and Cf-252 sealed sources for disposal as transuranic material to Waste Isolation Pilot Plant.

Expanding OSRP's Mission in Recovering New Isotopes for WIPP Disposal

INMM Conference July 2012

by Ioana Witkowski, Julia Whitworth, Alex Feldman, Mike Pearson

Offsite Source Recovery Project (OSRP)

- **OSRP's Mission – Eliminate excess, unwanted, abandoned, or orphan radioactive sealed sources that pose a potential risk to national security, public health, and safety**
- **OSRP is part of the National Nuclear Security Administration (NNSA) Office of Global Threat Reduction**
- **OSRP addresses radiological threat reduction for NNSA by aggressively removing radioactive materials (in the form of sealed sources) from the public sector that could pose a terrorist threat if acquired**

Project History

- **Project started in 1999 at LANL; since then, OSRP has recovered:**
  - **25,000 sources located in US at more than 1000 sites**
  - **2000 sources internationally**
- **OSRP started to recover actinides in 1999**
- **Recovery of beta-gamma sources started in 2004**
- **WIPP disposition of ^{239}Pu sources started in 2003 after DOE/NNSA Defense Determination approval. Similar Defense Determinations were obtained in 2006 for ^{238}Pu and ^{241}Am**
- **First NTS disposal of ^{60}Co occurred in 2008**
- **OSRP's source recovery mission expanded by including additional radionuclides, such as ^{237}Np, ^{244}Cm, ^{252}Cf**

Who Uses Radioactive Sealed Sources?

50+ Years of Isotope Distribution in the US

- **Oil and Gas Service Companies**
- **Colleges and Universities**
- **Manufacturing**
- **Medical Facilities**
- **Military Installations**
- **Construction Industry**
- **DOE and Government Sites**

Isotopes Managed

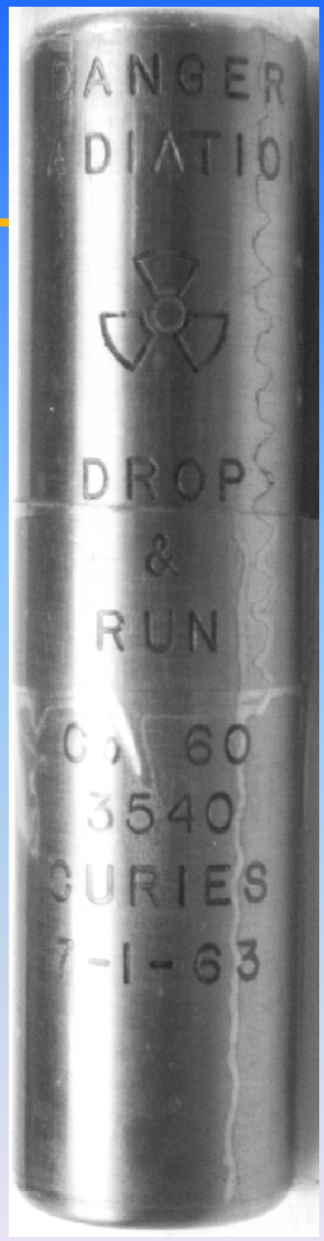
A Gammator containing ^{137}Cs recovered from a school district in Texas

Nuclides Originally Managed
^{241}Am
^{239}Pu
^{238}Pu
^{252}Cf

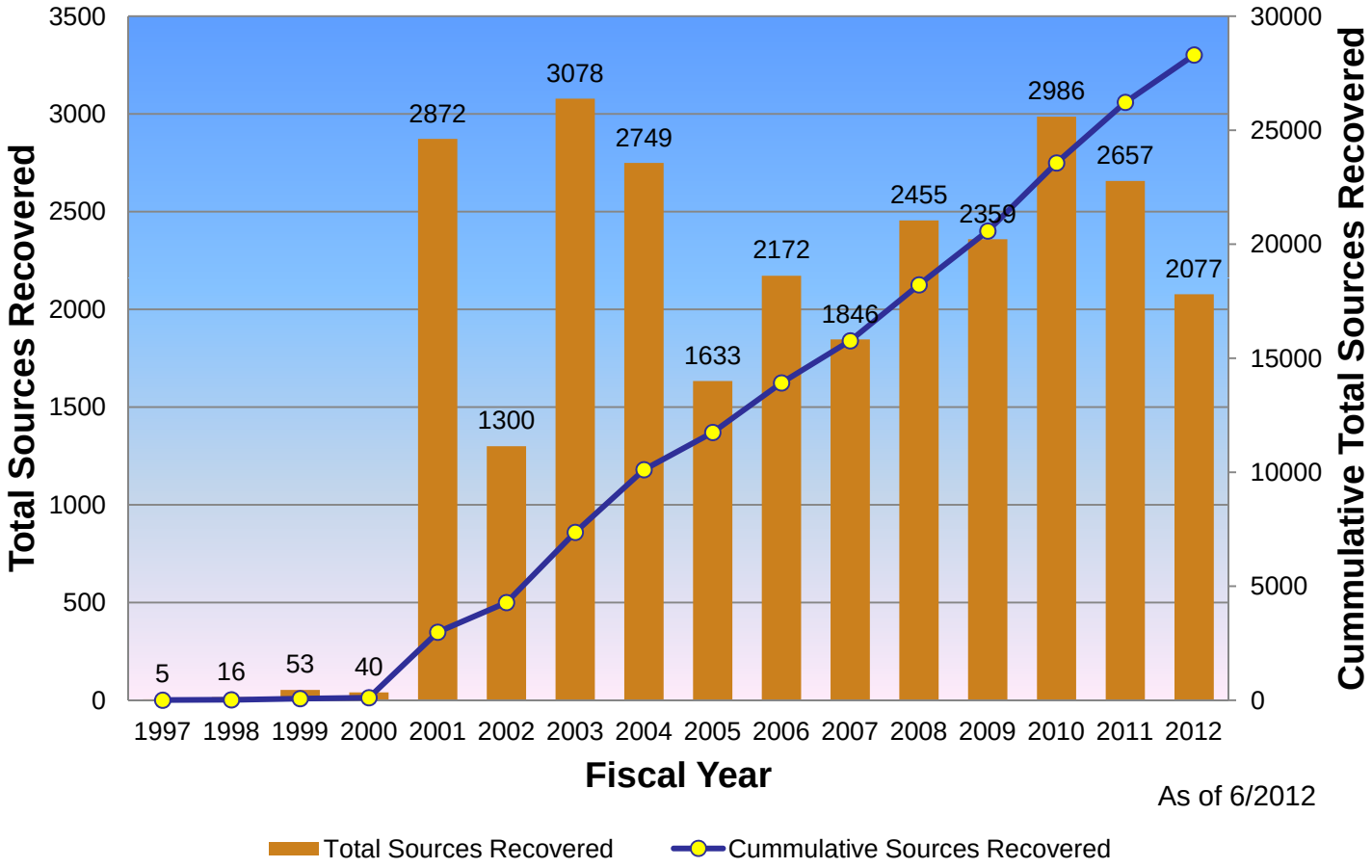
Additional Nuclides Currently Managed
^{244}Cm
^{226}Ra
^{90}Sr
^{60}Co
^{137}Cs
^{192}Ir

All nuclides currently found in IAEA sealed-sources-of-concern list

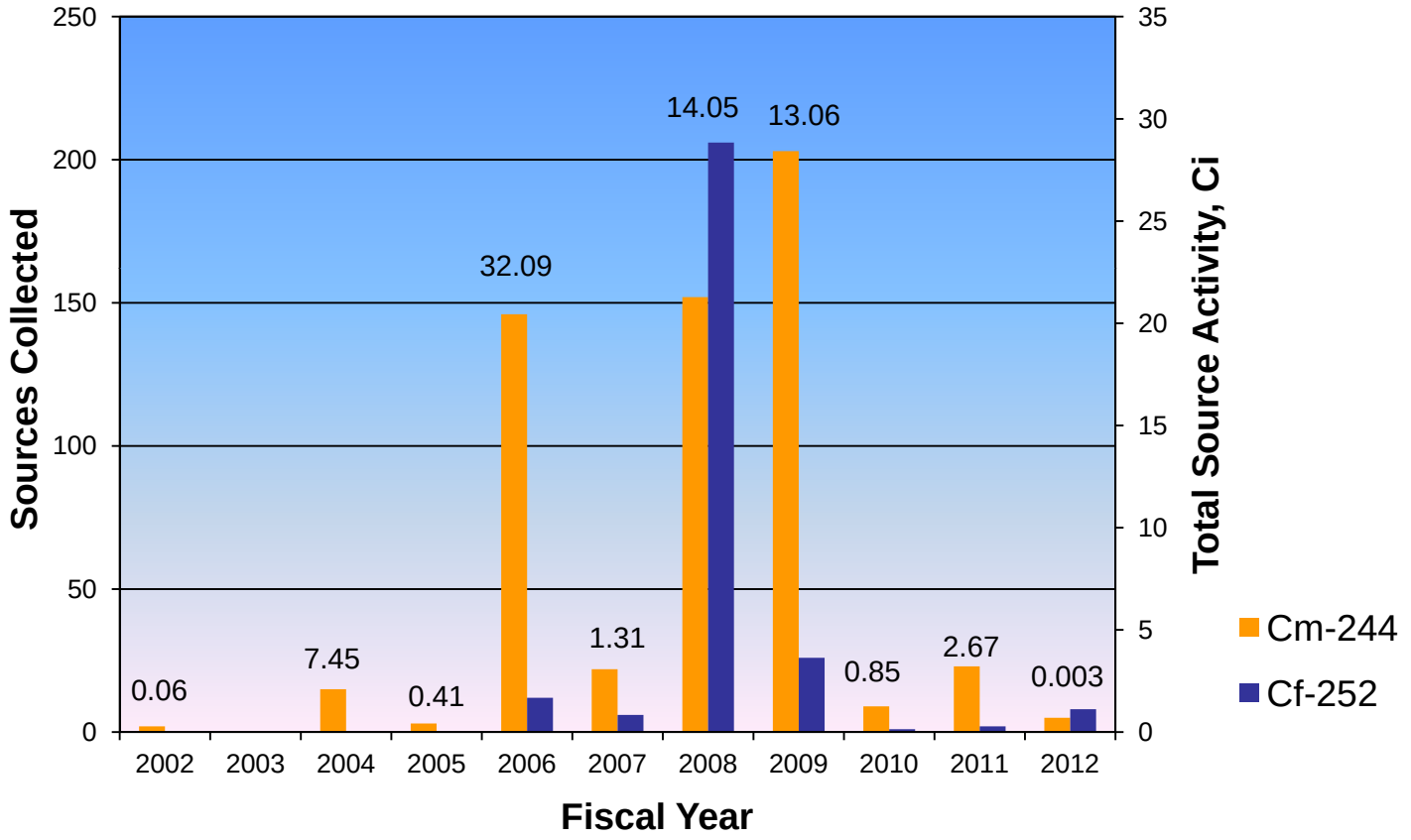
Identification: Source Self-Identification



Total Sources Recovered by OSRP per Fiscal Year

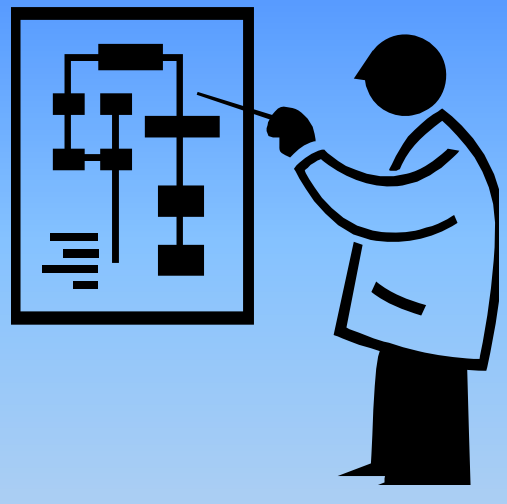


^{244}Cm and ^{252}Cf Recoveries per Fiscal Year



OSRP's Operation Processing Steps

1. Licensees Register Sources
2. Sources Entered into OSRP Database
3. OSRP Organizes Source Recovery or Self-Ships
4. Sources Consolidated in Interim Storage
5. Sources Shipped to Long-Term Storage
6. Characterization of Sealed Sources for WIPP
7. Final Disposal Approval by CCP
8. Transportation for Disposal to WIPP



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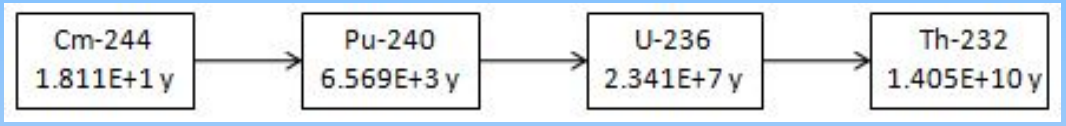
^{244}Cm in Sealed Source Production

- Sources used for γ and x-ray source applications
- ^{244}Cm has $t_{1/2}$= 18.11y < 20y for WIPP TRU isotopes
- Decay chain includes ^{240}Pu with $t_{1/2}$= 6.56E+3y -WIPP eligible



- ORNL producer of ^{244}Cm in US
- ^{244}Cm sealed sources manufactured by Amersham, IPL
- Batches provided to manufacturers were periodically purified to remove ingrowth of radionuclides, such as ^{240}Pu

Batch Data for ^{244}Cm and Associated Isotopic Fractions Used in Sealed Source Productions

Isotope	% Mass Fraction	Half-Life (Y)
Cm-244	86%-88%	18.11
Cm-245*	2%	8.50E+03
Cm-246*	10%-11%	4.75E+03
Cm-247*	0.2%	1.56E+07
Cm-248*	0.1%	3.39E+05

* TRU eligible radionuclides

- All of the “impurity” Cm isotopes listed meet the definition of TRU and contribute to the TRU content of ^{244}Cm source material

Typical Design of ^{244}Cm Sealed Sources

Construction of a sealed source with single encapsulation and tungsten backing.

Sources

Curium-244

γ and Primary X-ray Sources

Curium-244 incorporated in a ceramic enamel, sealed in a welded monel capsule with brazed beryllium window; the active component is recessed into a tungsten backing.

Nominal activity* GBq	mCi	A mm	B mm	Typical photon output in photons/s per steradian 17keV Pu LX-rays	Product code
0.37	10	10.8	7	0.8×10^6	CLC10990
1.11	30	10.8	7	2.4×10^6	CLC11564
3.7	100	10.8	7	7.8×10^6	CLC11562
7.4	200	10.8	7	15.0×10^6	CLC11377

* Tolerance ±10%

Nominal activity* GBq	mCi	A mm	B mm	Typical photon output in photons/s per steradian 17keV Pu LX-rays	Product code
0.37	10	8	4	0.8×10^6	CLC11932
1.11	30	8	4	2.4×10^6	CLC11284
3.7	100	8	4	7.8×10^6	CLC11933

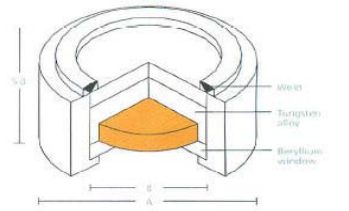
* Tolerance ±10%

Recommended working life: 10 years

Quality control
Wipe Test I
Immersion Test II
Bubble Test III

Neutron emission
All Curium-244 sources emit ~3.6 x 10^3n/s per GBq due to spontaneous fission and (α,n) reactions with the low atomic number elements (e.g. Si, Al, O) in the active material.

VZ-3069



Capsule dimensions and safety performance testing

Overall diam. 'A'mm	Active diam. 'B'mm	Window thickness 'C'mm	Safety performance testing ANSI/ISO classification	IAEA special form
8.0	4	1	C64343	YES
10.8	7	1	C64344	YES

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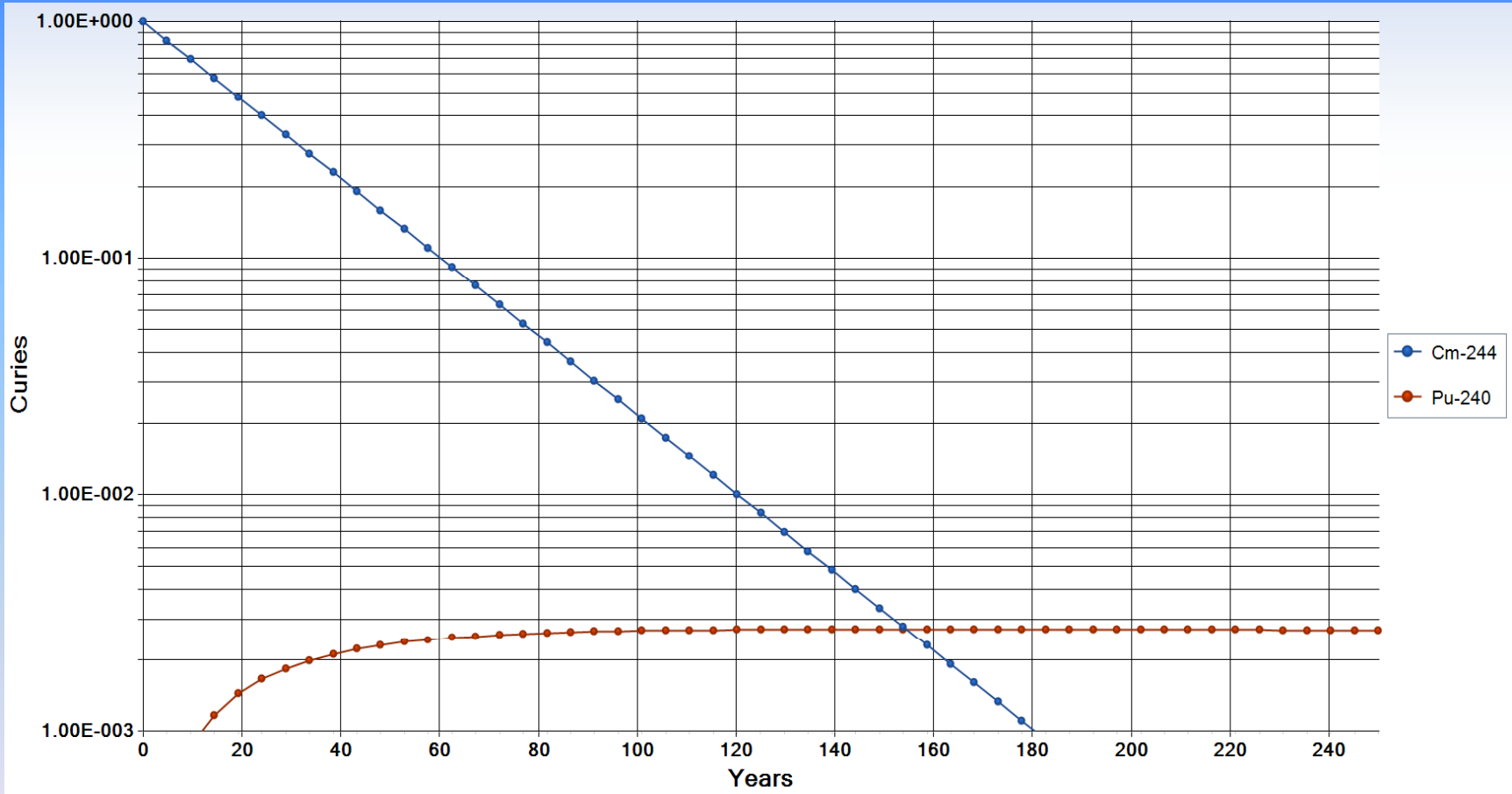
Decay Results for ^{244}Cm/ Ingrowth of ^{240}Pu with Time



Decay Results for 1 Ci ^{244}Cm Source after 10 years

Nuclide	Half-Life, Y	Specific Activity, Ci/g	Isotope Final Activity, Ci	Isotope Final Activity, nCi	TRU Isotope Activity, nCi/g capsule*	TRU Isotope Activity, nCi/g source**
Cm-244	1.81E+01	80.97	6.82E-01	6.82E+08	-	-
Cm-245	8.50E+03	0.172	4.87E-05	4.87E+04	6.77	9.75E+02
Cm-246	4.75E+03	0.307	4.70E-04	4.70E+05	65.3	9.41E+03
Pu-240	6.57E+03	0.227	8.76E-04	8.76E+05	121.7	1.75E+04
Am-241	4.32E+02	3.74	1.60E-07	1.60E+02	0.023	0.32E+01
Majority TRU Content					**193.8** nCi/g capsule	**2.79E+04** nCi/g source

Considerations:

- Minimum age of ^{244}Cm sources collected by OSRP is 10 years
- Decay calculations take into account an average distribution of ^{244}Cm isotopic data from ORNL
- TRU nCi/g calculations were done for the largest special form capsule *7200 g and **50 g per source
- The four main radionuclides contributors were considered

Discussion on ^{244}Cm Eligibility for WIPP Disposal

- The minimum decay interval of 10y for 1 Ci ^{244}Cm source produced ingrowth concentrations of ^{240}Pu that exceeded the 100-nCi/g requirement for WIPP disposal.

- Even with the heaviest capsule considered in the calculations for the TRU content and considering all Cm "impurity" isotopes, the TRU content is 193.8 nCi/g /Ci ^{244}Cm.

- If weight of capsule is excluded, TRU content increases to 28,000 nCi/g/Ci ^{244}Cm
  - lower quantities of material can be packaged
  - shorter decay period could be used

^{252}Cf in Sealed Source Production

- Sources used for neutron source applications
- ^{252}Cf has $t_{1/2}$= 2.65y < 20y for WIPP TRU isotopes
- Source material contains ^{249}Cf and ^{251}Cf impurities-WIPP eligible
- ORNL High Flux Isotope Reactor is producer of ^{252}Cf in US
- ^{252}Cf sealed sources are manufactured by Amersham, IPL, Frontier, and DOE
- Used in oil well logging and reactor start-up operations
- Decays by α emission primarily and spontaneous fission
- Batches provided to manufacturers were periodically purified to remove ingrowth of undesired radionuclides

Californium Source Material Isotopic Distribution

- Typical batches of ^{252}Cf provided to manufacturers

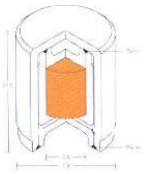
Nuclide	Half-Life, Yrs	Atom % Batch # CXCF-708 (6/9/03)	Atom % Batch # CAMP68 (7/18/91)	Atom % Batch # CXCF-579 (9/6/01)	Atom % Batch # COMP69 (6/30/96)	Average Atom %	Average Mass Fraction %	Total TRU Content, nCi/Ci Cf-252
Cf-249*	351 y	3.41	5.76	6.70	6.63	5.63	5.57	6.34E+05
Cf-250	13.20 y	8.70	9.22	9.63	12.66	10.05	9.98	
Cf-251*	898 y	2.60	2.85	2.97	4.06	3.12	3.11	
Cf-252	2.65 y	85.27	81.99	80.63	76.65	81.14	81.26	
Cf-253	17.81 d	0.004	<0.165	0.03	Not reported	0.05	0.05	
Cf-254	61.9 d	0.010	<0.018	0.04	0.00003	0.02	0.02	

- *TRU eligible radionuclides – α emitters and $t_{1/2}$ > 20 y
- At time of manufacture, the material meets the definition of WIPP transuranic

Typical Design of ^{252}Cf Sealed Sources

Sources

X.1



Sources up to 500µg can be manufactured in the X1 capsule design. Outer capsules are available in stainless steel or zircalloy:

Safety performance testing

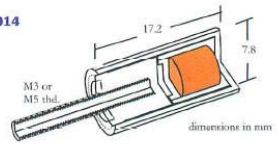
ANSI/ISO classification	IAEA special form	US-Model number
C66544	YES	CVN.CY2

Nominal content Cf-252	Nominal activity*		Emission n/s	Product code with –capsule- X1
1ng	20KBq	0.5µCi	2.3×10^3	CVN-X1-001ng-S
100ng	2MBq	54µCi	0.23×10^6	CVN-X1-100ng-S
500ng	10MBq	268µCi	1.15×10^6	CVN-X1-500ng-S
1.0µg	20MBq	536µCi	2.3×10^6	CVN-X1-001ug-S
2.0µg	40MBq	1.07mCi	4.6×10^6	CVN-X1-002ug-S
5µg	100MBq	2.7mCi	1.15×10^7	CVN-X1-005ug-S
10µg	200MBq	5.4mCi	2.3×10^7	CVN-X1-010ug-S
20µg	400MBq	10.7mCi	4.6×10^7	CVN-X1-020ug-S
50µg	1GBq	27mCi	1.15×10^8	CVN-X1-050ug-S
100µg	2GBq	54mCi	2.3×10^8	CVN-X1-100ug-S
200µg	4GBq	107mCi	4.6×10^8	CVN-X1-200ug-S
400µg	8GBq	214mCi	9.2×10^8	CVN-X1-400ug-S

* Tolerance -10%, +20% S indicates material S = stainless M = MP35N Z = Zircalloy

* Custom activities can be supplied within 12 weeks.

X.2014

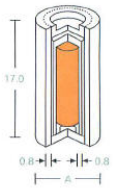


The X.2014 is an extended X.1 capsule with a female M3 threaded lid. An M3 (X.2014) or M5 (X2014/1) handling rod can be provided. Activities are available up to 8GBq (400µg).

Safety performance testing

Capsule	ANSI/ISO classification	IAEA special form	US-Model number
X2014	C66544	YES	CVN.CY12
X2014/1	C66544	YES	CVN.CY12

X.33, 35



Safety performance testing

Capsule	diam. 'A'mm	ANSI/ISO classification	IAEA special form
X.33	7.8	C66545	YES
X.35	9.5	C64545	YES

Nominal content Cf-252	Nominal activity*		Emission n/s	Capsule	Product code
500µg	10GBq	268mCi	1.15×10^9	X.33	CVN330
1mg	20GBq	536mCi	2.3×10^9	X.33	CVN331
2mg	40GBq	1.07Ci	4.6×10^9	X.35	CVN352
3mg	60GBq	1.61Ci	6.9×10^9	X.35	CVN353

* Tolerance -10%, +20%

* Custom activities can be supplied within 12 weeks.

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Sources

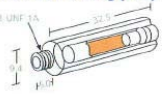
Californium-252

Spontaneous
Fission Neutron Sources

Savannah River Capsules

This design is the original Cf-252 capsule design created by the US Department of Energy at their South Carolina facility on the Savannah River. It is popular in many applications.

Savannah River Long (SRL)



Sources up to 2mg can be manufactured in the Savannah River Long capsule design (or the old X.224). Outer capsules are available in stainless steel or zircalloy:

SRL - stainless steel containing X1 or X33 inners
SRL - zircalloy containing X1 or X33 inners
X.224 (same dimensions as the SRL) - stainless steel

Safety performance testing

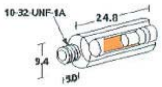
Capsule	ANSI/ISO classification	IAEA special form	US-Model number
SRL/X.1 inners	C66545	YES	CVN.CY7
SRL/X.33 inners	C64444	YES	CVN.CY8
X.224	C64545	YES	CVN.CY6

Nominal content Cf-252	Nominal activity*		Emission n/s	Product code with –capsule- SRL
1ng	20KBq	0.5µCi	2.3×10^3	CVN-SRL-001ng-S
100ng	2MBq	54µCi	0.23×10^6	CVN-SRL-100ng-S
500ng	10MBq	268µCi	1.15×10^6	CVN-SRL-500ng-S
1µg	20MBq	536µCi	2.3×10^6	CVN-SRL-001ug-S
5ug	100MBq	2.7mCi	1.15×10^7	CVN-SRL-005ug-S
10µg	200MBq	5.4mCi	2.3×10^7	CVN-SRL-010ug-S
20µg	400MBq	10.7mCi	4.6×10^7	CVN-SRL-020ug-S
50µg	1GBq	27mCi	1.15×10^8	CVN-SRL-050ug-S
100µg	2GBq	54mCi	2.3×10^8	CVN-SRL-100ug-S
200µg	4GBq	107mCi	4.6×10^8	CVN-SRL-200ug-S
500µg	10GBq	270mCi	1.15×10^9	CVN-SRL-500ug-S
1mg	20GBq	540mCi	2.3×10^9	CVN-SRL-001mg-S
2mg	40GBq	1.08Ci	4.6×10^9	CVN-SRL-002mg-S

* Tolerance -10%, +20% S indicates material S = stainless Z = Zircalloy

* Custom activities can be supplied within 12 weeks.

Savannah River Short (SRS)



Sources up to 1mg can be manufactured in the Savannah River Short capsule design. Outer capsules are available in stainless steel or zircalloy:

SRS - stainless steel containing X1 or X33 inners
SRS - zircalloy containing X1 or X33 inners

Safety performance testing

Capsule	ANSI/ISO classification	IAEA special form	US-Model number
SRS/X.1 inners	C64541	YES	CVN.CY14
SRS/X.33 inners	C64441	YES	CVN.CY15

Nominal content Cf-252	Nominal activity*		Emission n/s	Product code with –capsule- SRS
1ng	20KBq	0.5µCi	2.3×10^3	CVN-SRS-001ng-S
100ng	2MBq	54µCi	0.23×10^6	CVN-SRS-100ng-S
500ng	10MBq	268µCi	1.15×10^6	CVN-SRS-500ng-S
1.0µg	20MBq	536µCi	2.3×10^6	CVN-SRS-001ug-S
2.0µg	40MBq	1.07mCi	4.6×10^6	CVN-SRS-002ug-S
5µg	100MBq	2.7mCi	1.15×10^7	CVN-SRS-005ug-S
10µg	200MBq	5.4mCi	2.3×10^7	CVN-SRS-010ug-S
20µg	400MBq	10.7mCi	4.6×10^7	CVN-SRS-020ug-S
50µg	1GBq	27mCi	1.15×10^8	CVN-SRS-050ug-S
100µg	2GBq	54mCi	2.3×10^8	CVN-SRS-100ug-S
200µg	4GBq	107mCi	4.6×10^8	CVN-SRS-200ug-S
400µg	8GBq	214mCi	9.2×10^8	CVN-SRS-400ug-S
1mg	20GBq	540mCi	2.3×10^9	CVN-SRS-001mg-S

* Tolerance -10%, +20% S indicates material S = stainless Z = Zircalloy

* Custom activities can be supplied within 12 weeks.

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Decay Results for 1 Ci ^{252}Cf with Time

Considerations

- Decay of ^{252}Cf produces ingrowth of WIPP qualifying curium daughters
- The first eight transuranic radionuclides contributors were considered
- TRU nCi/g calculations were done for a model III special form capsule 3500 g
- Minimum age of ^{252}Cf sources collected by OSRP is 10 years

TRU Nuclide	Half-Life, yrs	TRU Nuclide Concentration, nCi		
		5 yrs nCi	10 yrs nCi	20 yrs nCi
Cf-249	351	5.20E+05	5.15E+05	5.05E+05
Cf-251	898	1.13E+05	1.12E+05	1.11E+05
Cm-246	4760	1.61E+04	2.84E+04	4.51E+04
Cm-248	3.39E+05	5.51E+03	7.00E+03	7.50E+03
Cm-245	8500	2.13E+02	4.24E+02	8.39E+02
Cm-250	9700	2.45E-01	2.45E-01	2.45E-01
Am-241	433	6.42E-02	4.85E-01	3.45E+00
Pu-242	3.75E+05	7.73E-02	2.85E-01	9.75E-01
Total TRU Content		6.55E+05	6.63E+05	6.70E+05
nCi/3500 g*		1.87E+02 nCi/g capsule	1.89E+02 nCi/g capsule	1.91E+02 nCi/g capsule

Discussion on ^{252}Cf Eligibility for WIPP Disposal

- The TRU content in ^{252}Cf material at 20y was 191 nCi/g, exceeding 100 nCi/g, even when the weight of the special form capsule is considered
- Evaluated from the time of production through 20y the material will increase their concentrations of transuranic nuclides
- If weight of capsule is excluded, TRU content increases to 6.7E+5 nCi/g/Ci ^{252}Cf
  - lower quantities of material can be packaged
  - shorter decay period could be used

Conclusions

•Although ^{244}Cm sealed sources are not TRU at the time of manufacture, they rapidly decay, producing ^{240}Pu in quantities that exceed the minimum requirement of 100 nCi/g

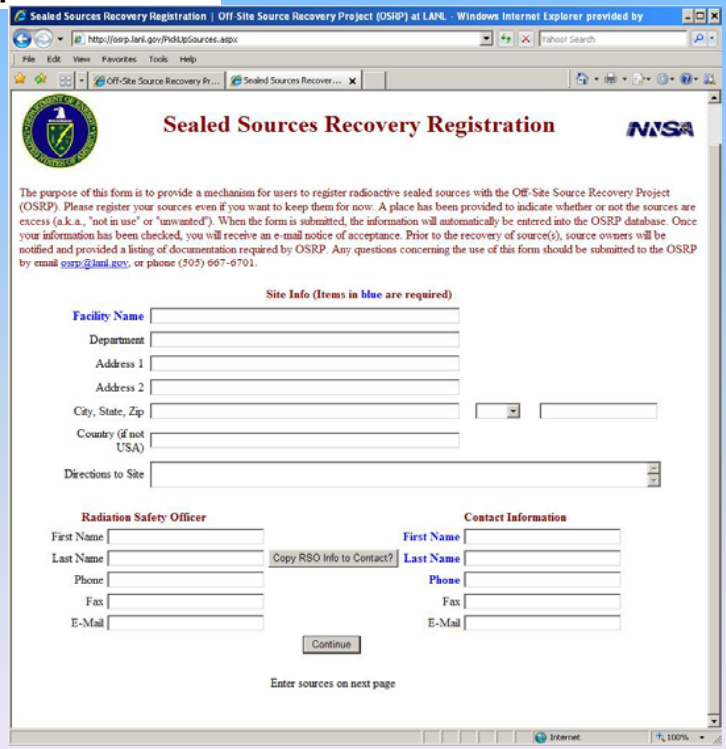
• ^{252}Cf source material contains significant amount of TRU impurities, increasing with time the TRU content for WIPP disposal

• Factors influencing the TRU concentration for each container:
  - isotopic distribution of the production batch
  - initial source activity
  - weight of sources and special form capsules
  - dates of source manufacture
  - contribution of other curium and californium isotopes and TRU ingrowth daughters

DO YOU HAVE UNWANTED SOURCES?

Register them at osrp.lanl.gov

- Register unwanted transuranic and selected beta, gamma sources with OSRP for recovery consideration.
- OSRP source recovery operations are generally prioritized on the basis of activity and level of security.
- Where numerous sources of lower activity are present at a single location, consideration is given to the total activity from a security perspective.



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