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Title: Selection of Russian Plutonium-Beryllium Sources for Inclusion in the Nuclear Materials Information Program Archive

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Introduction

Throughout the 1960s and 1970s, the former Soviet Union produced and exported PuBe neutron sources to various Eastern European countries. The Russian sources consist of an intermetallic compound of plutonium and beryllium encapsulated in an inner welded, sealed capsule and consisting of a body and one or more covers. The amount of plutonium in the sources ranges from 0.002 g up to 15 g. A portion of the sources was originally exported to East Germany. A portion of these sources were acquired by Los Alamos National Laboratory (LANL) in the late 1990s for destruction in the Offsite Source Recovery Program. When the OSRP was canceled, the remaining 88 PuBe neutron sources were packaged and stored in a 55-gal drum at TA-55. This storage configuration is no longer acceptable for PuBe sources, and the sources must either be repackaged or disposed of. Repackaging would place the sources into Hagan container, and depending on the dose rates, some sources may be packaged individually increasing the footprint and cost of storage. In addition, each source will be subject to leak-checking every six months. Leaks have already been detected in some of the sources, and due to the age of these sources, it is likely that additional leaks may be detected over time, which will increase the overall complexity of handling and storage. Therefore, it was decided that the sources would be disposed of at the Waste Isolation Pilot Plant (WIPP) due to the cost and labor associated with continued storage at TA-55. However, the plutonium in the sources is of Russian origin and needs to be preserved for research purposes. Therefore, it is important that a representative sample of the sources retained and archived for future studies. This report describes the criteria used to obtain a representative sample of the sources.

Background

The information for the Russian PuBe neutron sources at LANL comes from three sources. Limited information was obtained from the from the distribution company, Amersham Buchler, GmbH and includes the mass of plutonium, the neutron emission rates, and the dimensions for some sources. Additional data was obtained from the NDA campaign of the 88 sources and the destructive analysis of six of the sources conducted by LANL in between 2005 and 2007. The following information was obtained as part of the NDA campaign:

- Visual observation and inspection;
- Dose rate measurement at 30 cm (gamma and neutron);
- Isotopic analysis by gamma spectrometry;
- Prompt gamma-ray analysis for low-Z impurities;
- Total neutron count; and
- X-ray radiography.

FRAM v. 4.2 software was used to determine the isotopic composition of each PuBe source. The isotopic analysis results indicated that the sources can be divided into three groups based on the isotopic composition of the plutonium. The variation within each group is shown on the isotopic bar graph in Figure 1. The SNM value, or the weight of plutonium rounded to the nearest gram, is shown below the identification number for each source.

The sources in Group 1 have an average of 4.84% ^{240}Pu and 95.08% ^{239}Pu. As shown in Table 1, this isotopic composition is comparable to US weapons grade material but more enriched in ^{239}Pu. Group 2 (mean ^{240}Pu: 19.08, mean ^{239}Pu: 77.77) and Group 3 (mean ^{240}Pu: 19.06, mean ^{239}Pu: 68.39) have higher concentrations of ^{240}Pu and are similar to US fuel and power grade material. The major difference between groups 2 and 3 is in the concentration of ^{238}Pu and ^{242}Pu.

Table 1. Nominal isotopic compositions of US Pu

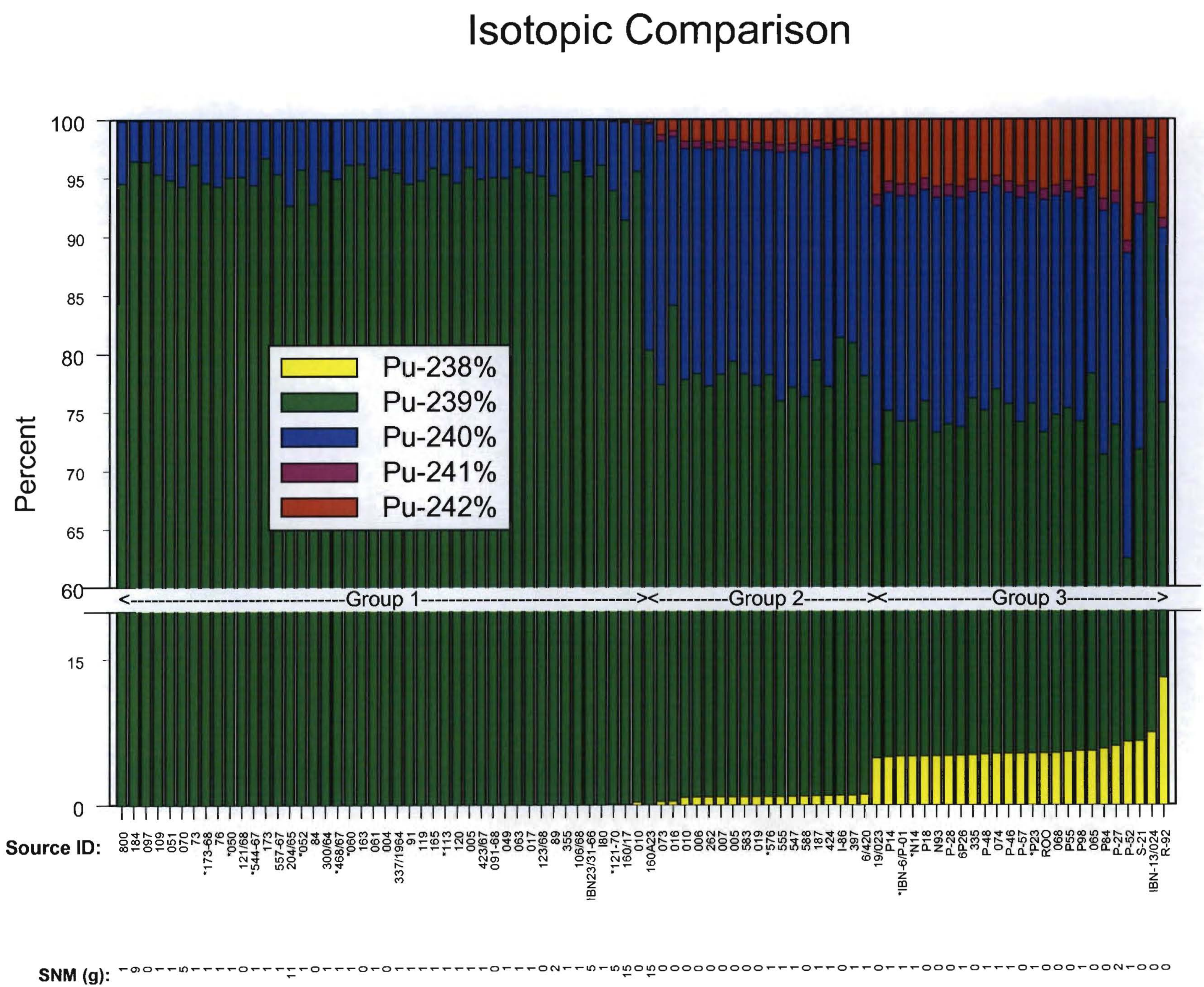
Grade	^{238}Pu %	^{239}Pu %	^{240}Pu %	^{241}Pu %	^{242}Pu %
Weapon Grade	0.05	93.50	6.00	0.40	0.05
Fuel Grade	0.1	86.1	12.0	1.6	0.2
Power Grade	1.0	63.0	22.0	12.0	3.0

Based on NDA results, representative sources from each isotopic group were selected for destructive analysis. The destructive analysis was performed on a total of six sources: four from Group 1 and one each from Groups 2 and 3. The analyses included the following:

- Gamma spectrometry;
- Pu & U Isotopic Analysis by Thermal Ionization Mass Spectrometry (TIMS);
- Pu & U Assay by Isotope Dilution Mass Spectrometry;
- Am concentration by radiochemistry;
- Trace Elemental Analysis by Plasma Spectrometry; and
- XRF analysis of the outer casing.

These data along with the NDA data are available in the "Russian plutonium beryllium neutron source database", LA-CP-09-00147.

Figure 1. Isotopic comparison of 88 Russian PuBe sources



* Indicates sources no longer available.

Selection Criteria

Several considerations were made in selecting sources for the NMIP archive. The primary consideration is the preservation of the Russian plutonium for research purposes. As indicated by the isotopic comparison in Figure 1, three subpopulations exist within the collection, and a representative sample must be chosen for each group. A second consideration is the preservation of as much plutonium as possible in the fewest number of sources. Most of the sources contain only a few hundred milligrams to 1 gram of plutonium. Disposing of these smaller sources will reduce the overall cost of repackaging, storage and handling. Another consideration is the preservation of unique samples within the collection. This would add additional sources to the archive and would provide bounding data for the archive sample.

As of March 2009, 77 sources remain in the collection at TA-55. Eleven of the original 88 sources were destroyed during DA. Of the remaining 77, three sources have developed leaks and have been excluded. In addition, one source has been excluded because it contains only 2 mg of plutonium, and the gamma-ray isotopic results from NDA are known to have considerable error. This brings the total number of sources eligible for selection to 73. The excluded sources are listed in Table 2.

Table 2. Sources excluded

*050	*052	*060	*113
*121-70	*173-68	*355	*468/67
*544-67	*576	*IBN-6/P-01	*N14
*P23	†163	†187	†424
‡IBN-13/024			
*Source destroyed; †Source found to be leaking; ‡Source is too small			

A set of parameters for selecting sources were identified in the complete data set available in the "Russian plutonium beryllium neutron source database". These parameters are as follows:

- Isotopic group designation;
- SNM value (mass of plutonium in the source);
- Isotopic composition; and
- Condition of source (i.e. intact or leaking).

Using these parameters the source selection was completed as described below.

1. One representative source was selected for every 20 sources within each isotopic group. The representative sources should be the largest source (by mass of plutonium) provided that it is representative of the average isotopic composition of the group and intact.
2. Up to two unique samples were selected that have isotopic compositions most different from that of the average for the particular group.

The sources were chosen for archival are listed in Table 3. The isotopic compositions of the sources selected for archival are listed in Table 4, and a bar chart comparing these data is shown in Figure 2.

Table 3. Sources chosed for NMIP archive

IsoGroup	SourceID	SNM(g)	Reason for Selection
	184	9	Representative IsoGroup 1
	204/65	11	Outlier: Highest Pu-240 in IsoGroup 1
	IBN23/31-66	5	Representative IsoGroup 1
1	160/17	15	Outlier: Highest Pu-238, Pu-241, and Pu-242 in IsoGroup 1
	016	0	Outlier: Lowest Pu-240 in IsoGroup 2
2	547	1	Representative IsoGroup 2
	P-48	1	Representative IsoGroup 3
	P-52	1	Outlier: High Pu-240, High Pu-242
3	R-92	0	Outlier: Highest Pu-238 in collection

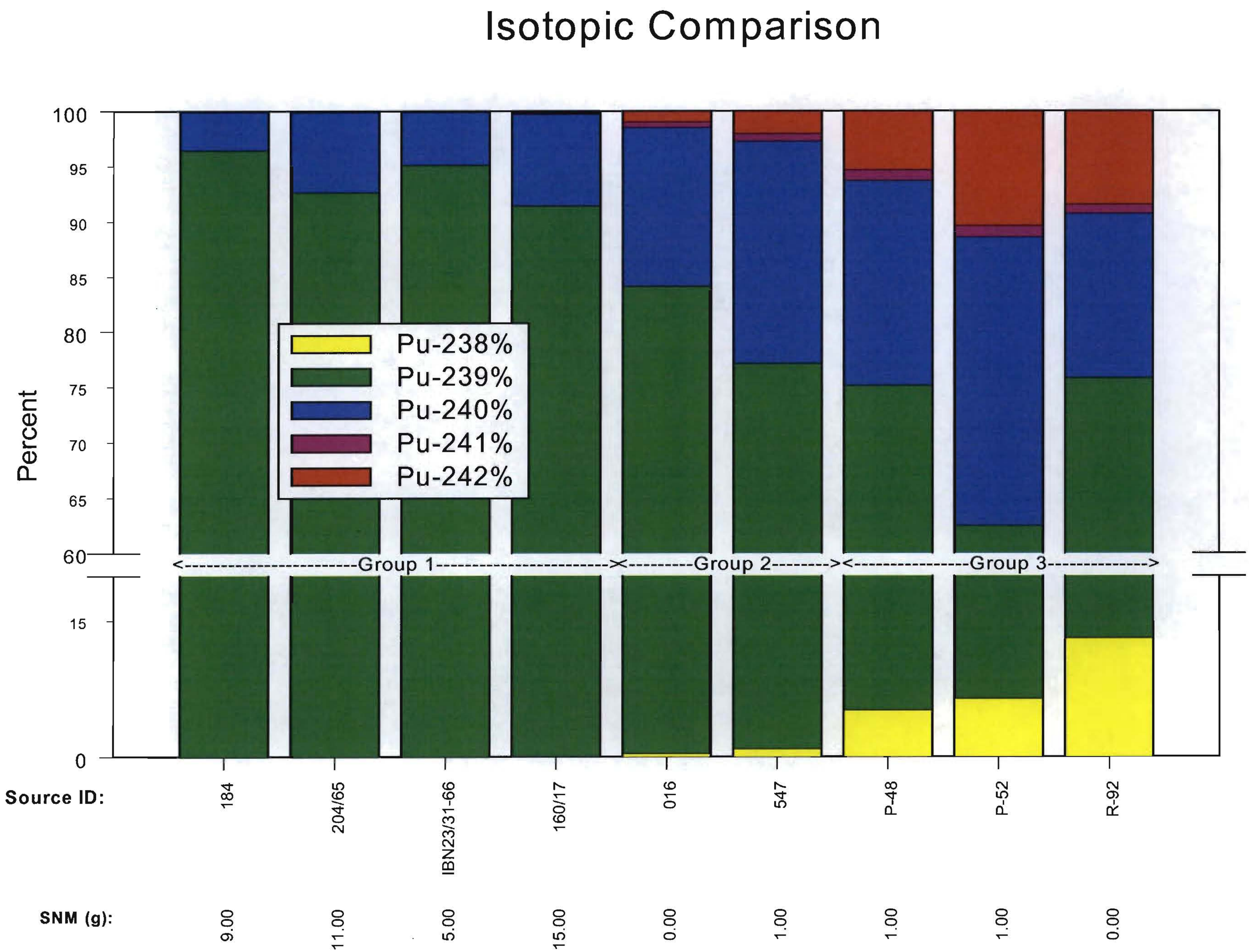
Table 4. NDA data for selected sources

IsoGroup	SourceID	Pu-238 (%)	Pu-239 (%)	Pu-240 (%)	Pu-241 (%)	Pu-242 (%)
1	184	0.0002	96.4502	3.5091	0.0371	0.0034
1	204/65	0.0035	92.6527	7.2855	0.034	0.0244
1	IBN23/31-66	0.0082	95.1041	4.8248	0.0364	0.0266
1	160/17	0.0607	91.3559	8.3599	0.0818	0.1416
2	16	0.3977	83.7508	14.3761	0.5402	0.9352
2	547	0.8888	76.2445	20.1517	0.6716	2.0434
3	P-48	5.1308	70.0012	18.5628	0.9745	5.3307
3	P-52	6.3997	56.0974	26.1022	1.0264	10.3743
3	R-92	13.1073	62.7046	14.9058	0.8386	8.4437

Conclusion

Nine Russian PuBe neutron sources have been selected out of a collection of 77 sources for inclusion in the NMIP archive. Selection criteria were developed so that the largest sources that are representative of the collection are included. One representative source was chosen for every 20 sources in the collection, and effort was made to preserve sources unique to the collection. In total, four representative sources and five unique sources were selected for the archive. The archive samples contain 40 grams of plutonium with an isotopic composition similar to that of weapon grade material and three grams of plutonium with an isotopic composition similar to that of reactor grade plutonium.

Figure 2. Isotopic comparison of Russian PuBe sources selected for NMIP archive



Appendix

Slides for NMIP Meeting

Sample Selection for Archive

LA-UR-09-01394

Selection of Russian PuBe Sources for NMIP Archive

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NNSA

Scope

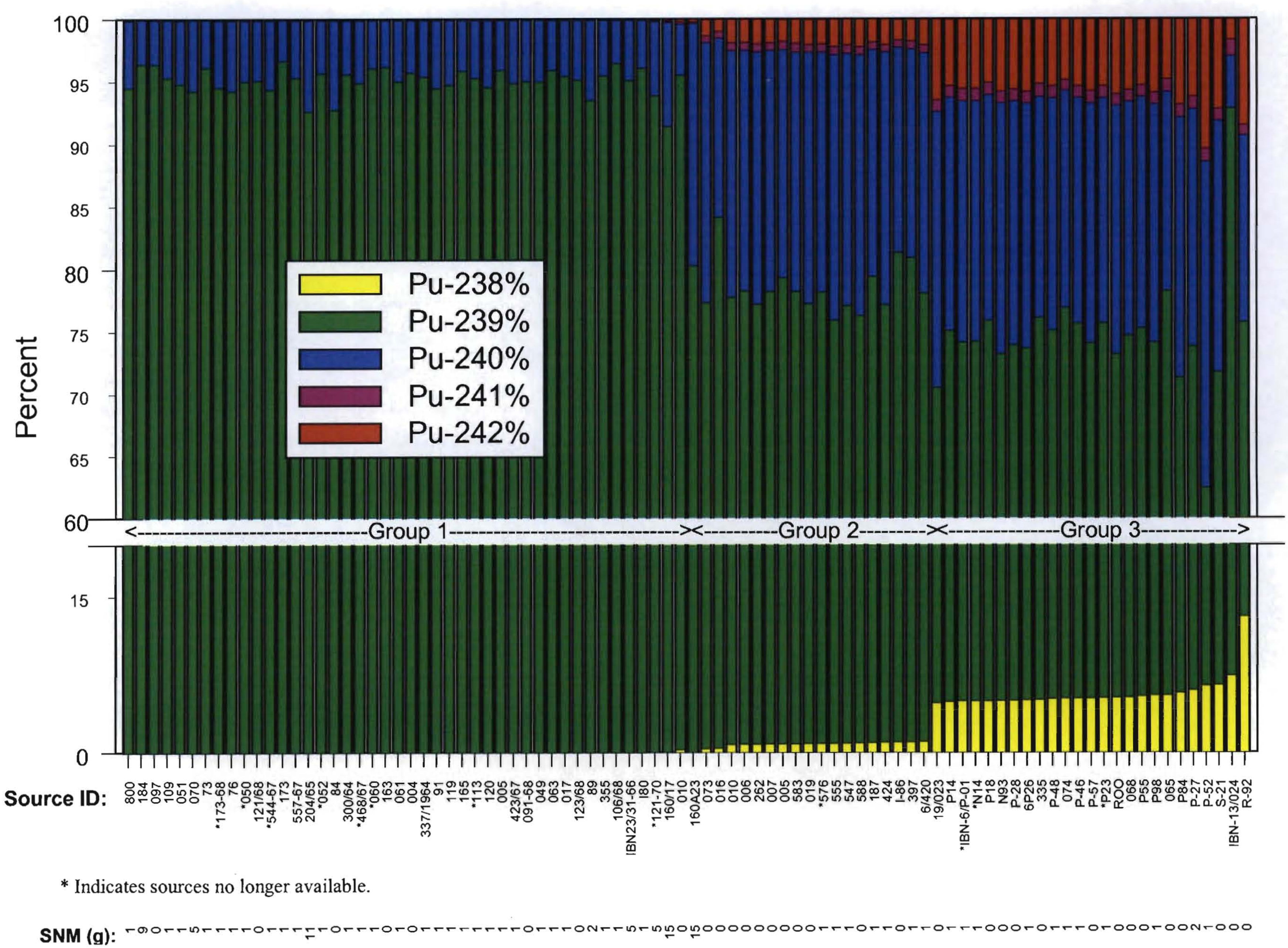
- 88 Russian PuBe neutron sources received at LANL as part of off-site source recovery program in late 1990s.
- Collection of sources is currently stored at TA-55
- Storage of entire collection is costly and labor intensive
  - 6-month leak checking required
  - Repackaging of each source is required
- Select a representative sample for inclusion into NMIP archive
- Disposition remaining sources to WIPP

Background Information

- **<u>NDA (88 sources)</u>**
  - Visual observation
  - Dose rate measurements;
  - Gamma spectrometry
  - Prompt gamma-ray analysis to detect if low-Z impurities
  - Neutron Count; and
  - Radiography
- **<u>Result:</u>** Based on isotopic analysis, 3 distinct groups exist within collection.

- **<u>DA (6 sources)</u>**
  - Gamma spectrometry;
  - Pu & U Isotopic Analysis by Thermal Ionization Mass Spectrometry (TIMS);
  - Pu & U Assay by Isotope Dilution Mass Spectrometry;
  - Am concentration by radiochemistry; and
  - Trace Elemental Analysis by Plasma Spectrometry
  - XRF analysis of casing.

Isotopic Comparison



* Indicates sources no longer available.

Selection Considerations

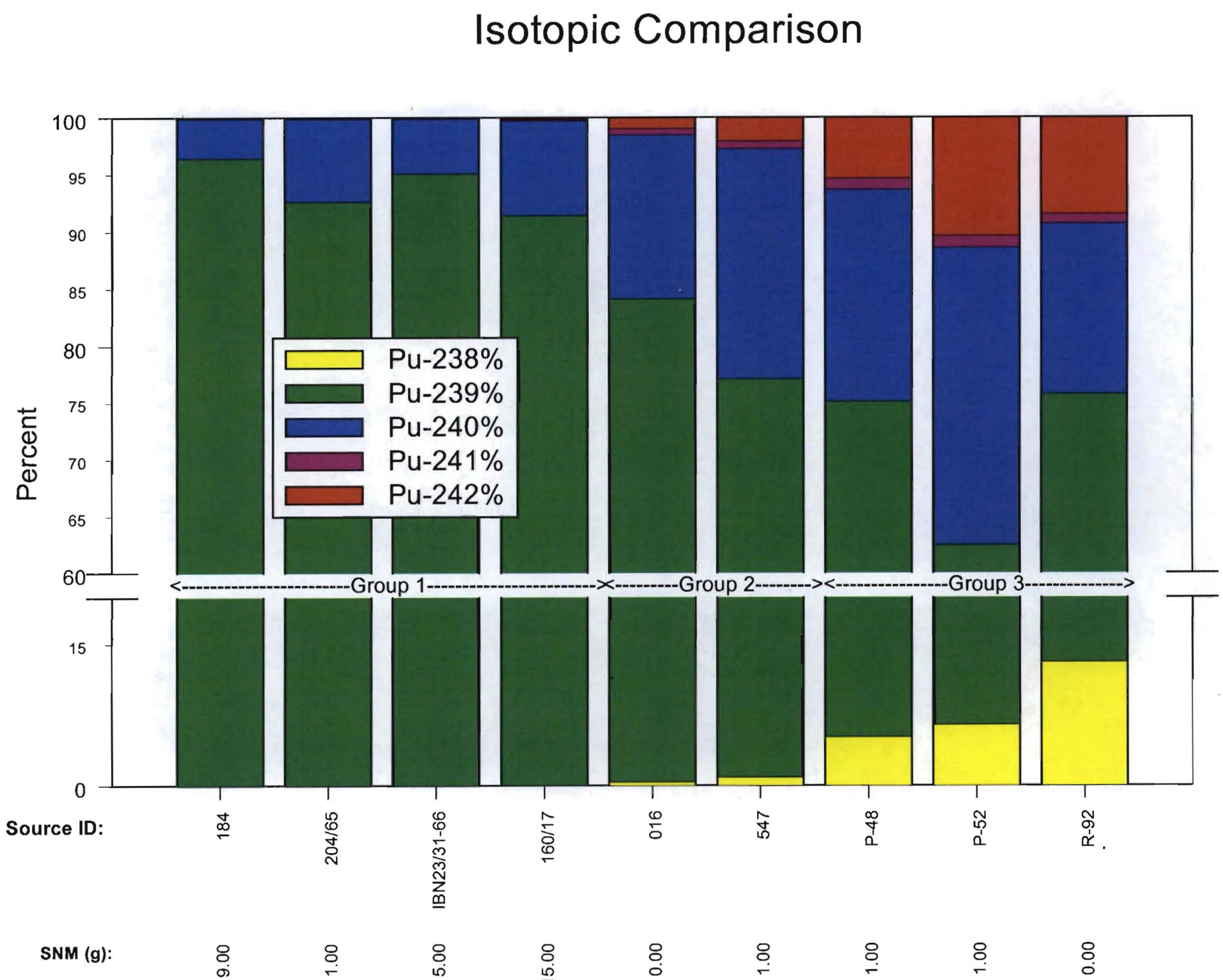
- Competing factors
  - Need to preserve Russian plutonium samples
    - Development of signatures
    - Future analyses
  - Reduce storage cost and labor
- Selection of representative sample
  - Preserve most plutonium with fewest number of samples.
  - Select sources from each isotopic group.
  - Preserve “representative” samples.
  - Preserve unique samples.

Selection Criteria

- **Criteria:**
  - Isotopic group designation
  - Mass of plutonium in the source
  - Isotopic composition
  - Condition of source (i.e. intact or leaking)
- **Selection Procedure:**
  - Selected one representative source for every 20 sources within each isotopic group. The representative sources should be the largest source provided that it is representative of the average isotopic composition of the group.
  - Selected up to two unique samples from each group, which are most different from the average isotopic composition of the group.

Sources Selected

IsoGroup	SourceID	SNM(g)	Reason for Selection
1	184	9	Representative IsoGroup 1
	204/65	11	Outlier: Highest Pu-240 in IsoGroup 1
	IBN23/31-66	5	Representative IsoGroup 1
	160/17	15	Outlier: Highest Pu-238, Pu-241, and Pu-242 in IsoGroup 1
2	016	0	Outlier: Lowest Pu-240 in IsoGroup 2
	547	1	Representative IsoGroup 2
3	P-48	1	Representative IsoGroup 3
	P-52	1	Outlier: High Pu-240, High Pu-242
	R-92	0	Outlier: Highest Pu-238 in collection



Isotopic Comparison
Percent
100
95
90
85
80
75
70
65
60
15
0
Pu-238%
Pu-239%
Pu-240%
Pu-241%
Pu-242%
<----Group 1----><----Group 2----><----Group 3---->
Source ID:
184
204/65
IBN23/31-66
160/17
016
547
P-48
P-52
R-92
SNM (g):
9.00
11.00
5.00
15.00
0.00
1.00
1.00
1.00
0.00

Conclusion

- Sources selected for NMIP archive: 9
  - Representative sources: 4
  - Unique sources 5
- Russian weapon grade Pu: 40 g
- Russian reactor grade Pu: 3 g

Sources Detail

IsoGroup	SourceID	Pu-238 (%)	Pu-239 (%)	Pu-240 (%)	Pu-241 (%)	Pu-242 (%)
1	184	0.0002	96.4502	3.5091	0.0371	0.0034
1	204/65	0.0035	92.6527	7.2855	0.034	0.0244
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