Recovery of Spent High Intensity Neutron Sources in Atalante Facility

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Abstract

The Atalante facility is required by law to recover both neutron and gamma sources with activity levels exceeding 300 mCi. Most of the neutron sources consist of mixtures of alpha-emitters (^{238}Pu, ^{239}Pu, ^{241}Am or ^{244}Cm) and beryllium. Several processes now under consideration are based on routine chemical separation techniques (selective precipitation, extraction chromatography, ion exchange). The treatment produces an actinide oxide (which is used later for R&D studies) and solid beryllium nitrate, which is considered as a waste and transferred to a surface interim storage site if the overall activity of the package after 300 years is less than 50 MBq (ANDRA specifications). The Material Analysis and Metrology Laboratory of Atalante validate the residual alpha activity in the waste. The techniques used include alpha spectrometry and L-line X-ray fluorescence for alpha emitters, and plasma torch measurements (ICP-AES and ICP-MS) for beryllium analysis. Specific equipments for transport (B type cask), storage and treatment (hot shielded cells) are used for this activity.

Keywords: *Neutron sources, Actinides, Beryllium, Analysis, Atalante, Waste.*

Context

In 1990, French government decided that suppliers had to recover spent radioactive sources at the end of their use or later ten years after. In this frame, the Atalante facility is in charge of the recovery of both gamma and neutron French sources which activity level is above 300 mCi (11 GBq). This task comprises two major parts. A management aspect consisting to:

- examine sources recovery requests,
- identify sources to be recovered from inventories,
- organize transports,
- manage small interim storage facilities.

A technical and scientific aspect concerning mainly:

- the development of specific devices to implement in hot cells,
- R&D studies to carry out actinides separation from beryllium.

The fabrication of sealed sources ended on 1992 at the Fontenay aux Roses center and the activity was transferred to Atalante in Marcoule where specific equipments have been implemented to keep on. This paper presents an overview of these equipments and gives the status on the R&D in progress concerning the recovery of spent high intensity neutron sources.

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Neutron sources to be recorded

Most of the neutron sources comprise a mixture of beryllium (metal or oxide) and an alpha emitter such as ^{241}Am, ^{244}Cm, ^{238}Pu or ^{239}Pu. Neutrons are produced by reaction between alpha particles and beryllium as follows:

$$^{9}_{4}Be + ^{4}_{2}He \rightarrow ^{1}_{0}n + ^{12}_{6}C + 5{,}71\ \text{MeV}$$

Table I shows some neutron sources characteristics.

Table I Neutron sources characteristics

Source Type	Half life (year)	Alpha emission energy (MeV)	Neutron Emission (10^6 n/s/Ci)	Neutron Emission (10^6 n/s/g)
^{241}AmBe	458	5.4	2,2	7,7
^{239}PuBe	24400	5.1	1,5	0,09
^{244}CmBe	18.1	5.8	2,6	200

PuBe sources are often used as standard for neutron counters because of their stability. AmBe sources are used in numerous fields: petroleum industry (well logging), soil science (density and moisture content determinations), and activation analysis…

The inventory of the neutron sources manufactured in Fontenay aux Roses and then in Marcoule has been done (cf. Table II). It allows identifying disused sources to be recovered which are stored in or out of Atalante.

Table II Neutrons sources inventory

Source Type	**Actinide mass stored in Atalante (g)**	**Actinide mass stored out of Atalante (g)**	**TOTALS (g)**
^{241}AmBe	80 (207 sources)	590 (919 sources)	670
^{239}PuBe	432 (4 sources)	713 (26 sources)	1145
^{244}CmBe	0.37 (10 sources)	3 g (30 sources)	17

^{241}AmBe and ^{239}PuBe represent significant masses that could be used for other applications (R&D studies, etc).

Main equipments used for the neutron sources recovery

A specific cask (photo n°1) has been developed by the CEA for high activity sources transports. The pending renewal of approval is scheduled for 2005. This shipping container allows to transport a large variety of actinides as shown in table III.

Table III CTB cask contents

Content	Isotopes	Mass (g)
Np	237	250
	239	0,001
Pu	238	30
	239 - 240 - 241- 242	250
U	Natural	1000
	233 - 234 - 235	250
	232	2
Am	241 - 243	30
Cm	242	0,002
	244	6
	248	0,001
Th	228	0,01
	229	250
Tc	99	100
Cf (special form)	252	5,00E-05
Alpha emetters and light elements (Be, B, F, C ou Li) mixtures in special form	^{241}Am	14,5
	^{244}Cm	0,617
	^{238}Pu	2,92
	^{239}Pu	250

In Atalante, the RESO room is devoted to the low activity radioactive sources storage. In the case of actinides beryllium sources, it cannot be used because of the intense neutron emission. So, sources are directly introduced in the C10 hot cell (cf. photo n°2) which biological shield is suitable with high neutron emissions.

Photo n°2 : C10 Hot cell – General view

Photo n°1 : CTB Cask

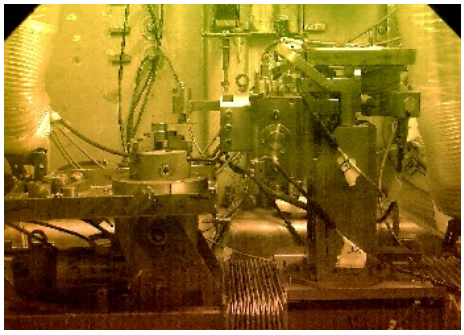

Photo n°3 : Sources opening machine

After their introduction, the sources are opened with a specific device (photo n°3). Since 1997, about 160 ^{241}AmBe sources have been opened, leading to the recovery of about 600 g of material (Am + Be). R&D studies are now in progress at the L6 laboratory to perform Am/Be separation. Specific analytical developments are also needed, in particular to characterize the waste containing the beryllium. These tasks are carry out in laboratories L19 (ICP measurements) and L27 (radiometric determinations). The R&D results will be used to perform the chemical operations in hot cells.

Studies on the reprocessing of the AMO_2/Be sources

Studies have been undertaken to reduce the neutron fluence of the mixture (Am/Be) and to recover the material with the following objectives:

- separate the americium from the beryllium and stabilize the americium as AmO_2;
- process the beryllium and effluent streams to obtain a Be chemical form and an Am concentration suitable for removal from the Atalante building.

Different steps have been studied:

- to dissolve the AmO_2/Be mixture,
- to perform the Am/Be separation,
- to obtain pure americium,
- to produce Be nitrate at an activity level suitable for a waste surface site disposal.

The obtained results allow proposing a diagram of the reprocessing that will have to be validated with genuine

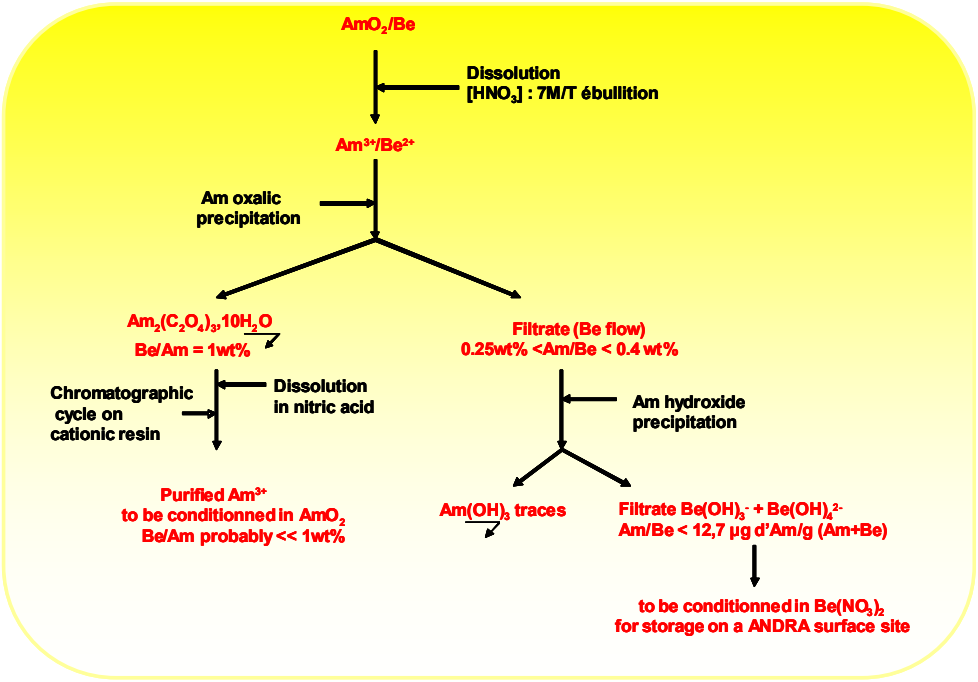

Fig. 1 AmO_2/Be source reprocessing flowsheet

The synthesis of beryllium nitrate and the liquid waste processing must be studied too.

Analytical studies

In collaboration with ANDRA (the French National Radioactive Waste Management Agency), the beryllium arising from source processing is placed in interim storage at a surface site. The resulting waste package must only contain beryllium nitrate, and its activity after 300 years must not exceed 50 MBq.

In Atalante, the waste drums destined for an ANDRA site are measured by passive neutron counting in addition to gamma spectrometry measurements. These techniques are unreliable in the case of waste arising from the destruction of neutron sources; it was therefore decided that the waste activity would be determined prior to conditioning. Specific protocols have been developed for this purpose.

Determination of the Be concentration in the Be nitrate solution

After the beryllium nitrate conversion in hot cell, a sample is then dissolved in nitric acid and transferred to Analysis Laboratory. The quantity of beryllium in the dissolution solution is determined by Inductively Coupled Plasma – Atomic Emission Spectrometry (ICP-AES) and by (Inductively Coupled Plasma – Quadrupolar Mass Spectrometry (ICP-QMS). In both techniques, a plasma is used to excite the atoms initially present in the liquid sample. ICP-AES detects the photon radiation emitted during the atom relaxation processes and the excited ions (emission spectrometry), while in ICP-QMS the ions formed in the plasma are collected and counted (mass spectrometry). ICP-AES is capable of measuring each element by its particular wavelengths, while ICP-QMS discriminates among the elements according to the ion mass/charge ratio.

The determination was performed with the equipment in laboratory L19. Beryllium analysis by ICP-AES and ICP-MS. ICP analyses are carried out on a highly diluted sample. An internal measurement standard is added to the ICP measurement sample to correct the matrix effects. The nature of the standard depends on the element being assayed and the technique used. Holmium (Ho) was used for ICP-AES measurements, and indium (^{115}In) for ICP-QMS. In these conditions, both techniques are very sensitive and the quantification limits are respectively equal to 0.2mg.L^{-1} and 0.01mg.L^{-1} for ICP/AES and ICP/MS.

Determination of the alpha activity in the residual beryllium nitrate

The alpha activity was determined for a surrogate solution containing beryllium (50 g/L) and plutonium 239 (11 mg/L) using a semiconductor detection system. The dilution factor and the quantity deposited for source preparation allowed usable spectra to be recorded in a reasonable time with an activity of about 5 Bq.
The plutonium concentration determined from the alpha activity was consistent with the value obtained by X-ray fluorescence. The system is capable of determining trace quantities of uranium and plutonium in aqueous or organic solutions arising from spent nuclear fuel reprocessing.

Comments

The results obtained show that the selected techniques and operating procedures correctly characterize beryllium waste. No interference was observed for analysis of beryllium by ICP-MS or by ICP-AES. For activity measurement, we demonstrated that sample dilution followed by semiconductor alpha spectrometry measurements yielded usable count rates and dead time values, avoiding to perform difficult chemical separation steps.