

CHARACTERISATION OF THE CONTENT OF A RADIOACTIVE WASTE DRUM: EXAMPLE OF TECHNIQUES USED IN FRANCE FOR PLUTONIUM MEASUREMENT

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1. INTRODUCTION

French ministerial directive of June 1988 gives the technical conditions for control and accountancy of nuclear materials contained in solid waste. As far as the nuclear materials follow-up is concerned, this text states that any nuclear material (plutonium or high enriched uranium) declared as waste should be characterized. For other materials, best estimates are allowed. Reversibility possibility (from waste to usable material) was not considered.

It is the operator's responsibility to declare as «waste» a nuclear material whose use is no longer economically profitable, generally due to low concentration of those materials. It is worth pointing out that the French regulation considers nuclear materials all along the fuel cycle (excluding ores) up to waste disposal with the adapted regime for waste mentioned above. That means that, for instance, uranium ores with a uranium concentration level of 1% currently met is not taken into account. That means also that nuclear materials in similar concentration could be legible to be treated as waste but possibly also to be exempted from French domestic safeguards. The experience gained from the French safeguards inspections shows a variety of «waste» from scraped materials to contaminated items, especially in facilities under dismantling. Setting concentration levels could help to reduce those discrepancies and either to define a safeguards termination for those materials or to harmonize what could be labeled as waste in different facilities.

Since precise quantification of nuclear materials in waste poses some problems, operators are often tempted to overestimate the nuclear material content of waste drums following basic criticality safety rules. This procedure is highly prejudicial for control and accountancy purposes since it results in biased nuclear materials balance and the obvious consequence that it is impossible to conclude on a possible loss or diversion of nuclear materials by the only analysis of the facility material balance. This is due to the fact that quantities that are shipped out of the facility are effectively smaller than those reported. This trend observed in the operators' behaviour reaches some limit for economical reasons since each waste drum should not exceed an activity limit (due to repository specifications in case of disposal), which corresponds to a nuclear materials mass limit.

The Nuclear Defense Expertise Division (IRSN) of the Institut de Radioprotection et de Sûreté Nucléaire (IRSN) as the technical support of the French Competent Authority for NMPC&A has shown the need to improve the traceability of nuclear materials in wastes and to perform more precise **physical measurements** despite low concentration in wastes of elements under French domestic safeguards. IRSN has developed, for inspections purposes, original and complementary methods for the measurement of plutonium and uranium in waste drums. This paper describes the principle of these devices. It also addresses the possibility to use them for the characterization of other radioactive material. In a second part, it addresses a

summary of the different kind of waste and measurement systems used in the French nuclear industry.

2. PLUTONIUM MEASUREMENTS IN WASTE DRUMS

The objective of the measurement is the determination of plutonium mass in 100-liter or 200-liter cylindrical waste drum, with a typical density in the range of 0.1 and 0.4 g/cm³.

The IRSN has developed original and complementary methods for the measurement of plutonium in waste drums. Those devices, further described in the text, are used to control periodically, on a random basis, the plutonium quantities declared by the operators. These measurements that are done by inspectors from the IRSN enable the French Authority to assess the measurement methods of the operator; it may also contribute to the request of improvement of the performance of the operators' instrumentation.

Measuring plutonium in waste drums is not a simple task. Several factors contribute to increase the uncertainties. Nuclear materials in waste drums may be in relatively low quantities. The localization of the plutonium in the drum as well as the matrix composition are unknown. Analysis methods may also be disrupted by the presence of "polluting" elements that emit neutrons or photons (curium, neptunium, americium etc.). The detection limit of the measurement devices as well as the uncertainties on the announced result are then raised.

2.1. MEASUREMENT'S ORGANIZATION

IRSN's inspectors are used to organizing on-site inspections on duration of a week, measures beginning on Monday and ending on Thursday. The material is **transported** by truck the week preceding the inspection in order to settle and test the equipment on the previous Friday. This day allows also doing the formalities for access. The last day of the inspection is foreseen to make some additional measures, to pack the material and to make a summary meeting of the inspection.

For each drum, several measurements are performed.

- Gross weight,
- Filling height of the drum, (using a gamma scanning with a transmission source)
 - Both information gives an indication of the density of the drum
- Passive neutron measurement using an instrument called FUNE
- Gamma spectrometric measurement using an instrument called PLUM (and COMPOS for isotopic composition)
- Isotopic composition, using MGA code.

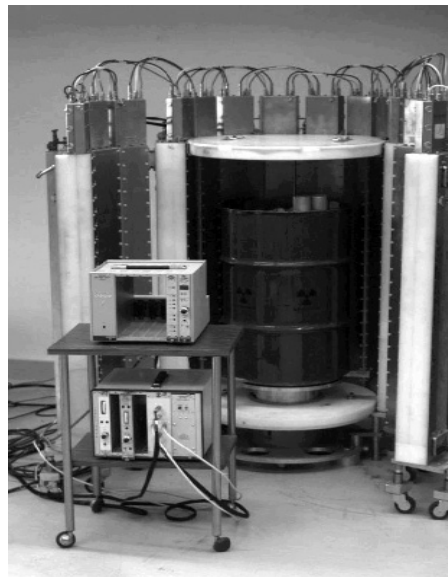
A typical duration for FUNE and PLUM counting and analysis is 30 min. This means an average duration of a drum's expertise in 45 min and an average of 30 drums' expertise for the total inspection week.

Regarding detection limits for both PLUM and FUNE, a typical value is on the range of a few dozen of milligram of plutonium. These detection limits depend on many factors like radioactive background conditions, matrix: every inspection is a special case.

2.2. QUANTIFICATION OF THE PLUTONIUM IN WASTE DRUM: FUNE [1]

The device FUNE (picture 1) developed by the IRSN is a passive neutron coincidence counting device with 28 helium 3 tubes. The whole system is designed to be easily **dismantled and transported** for on-site measurements.

Calibration curves have been established to relate the neutron coincidence counting to the equivalent mass of plutonium 240, according to the absorption of the matrix of the drum. For that purpose, a correction is done by means of a transmission measurement using an external californium 252 source.



Picture 1: FUNE

Concerning the determination of the matrix density, the content of a drum (metal, wood, concrete, vinyl, etc.) is never directly accessible to the inspectors. On the other hand, a simple weighing leads to the evaluation of the density. However this value does not indicate the real neutron absorption. Indeed, neutron absorption depends on the nature of the matrix and not only on the density. To achieve this study, the IRSN made several matrixes, of vinyl type, with density in the range of 0.1 and 0.4 g/cm³. A transmission measurement with a californium 252 source allows the evaluation of the neutron absorption of each matrix. With this method, the neutron absorption correction factor is measured experimentally, independently of the nature of the matrix. That allows to establish equation $\rho_i = f(T)$. T is the total counting rate measured during the transmission measurement. Three measurements are necessary to proceed to the quantification of the plutonium in a waste drum:

- Neutron coincidence counting for the drum,
- Transmission measurement with the californium 252 source. For this measurement, only half of the detectors of the chamber are used.
- Measurement of the drum alone, in the previous configuration.

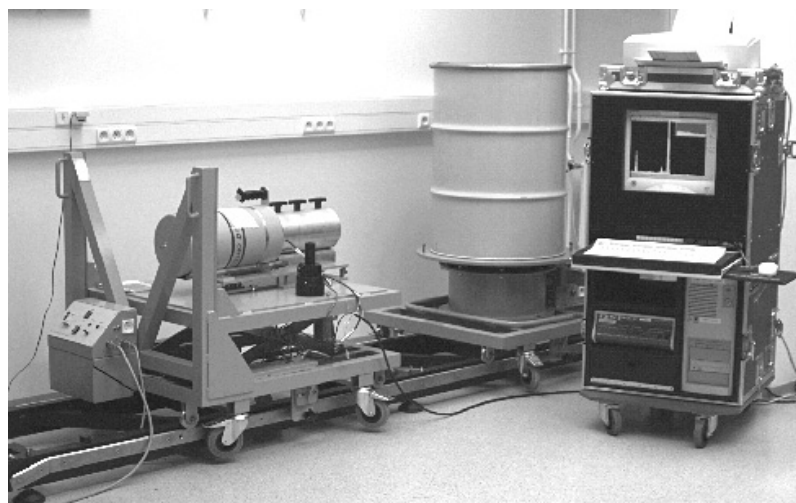
2.3. QUANTIFICATION OF THE PLUTONIUM IN WASTE DRUM: PLUM [2]

The instrumentation (Picture 2 and 3) was realized to meet the needs of inspectors for whom speed and flexibility of installation on site, as well as the geometrical reproducibility of the measurement conditions, are important. This measurement device is constituted with a frame on which come to position two mobile wagons; the first supports the detector germanium; the second is constituted with a rotating plateau on which is set the drum to measure. The instrumentation consists of a multichannel analyzer and of a high efficiency germanium detector. Dedicated software has been developed by the IRSN, linked to commercialized module, to acquire spectra and to perform the analyses. The whole devices are transported in flight cases and are operational in thirty minutes on site.

The principle of the measurement is based on a method, said of "infinite energy extrapolation". It requires the use of a high efficiency germanium gamma detector, and analyzes the main gamma rays emitted by the plutonium between 100 and 500 keV [2].



Picture 2 : Installation of PLUM



Picture 3: PLUM

It is generally difficult to estimate within a waste drum the corrections for self-absorption and attenuation by the usual equations that suppose to know the geometry of the sample. Methods used in the device PLUM are based on the evaluation of these corrections from countings observed on several gamma peaks of different energies in the region from 100 to 500 keV.

2.4. ISOTOPIC COMPOSITION MEASUREMENT

Both PLUM and FUNE need the isotopic composition value to get quantitative results on the plutonium mass. FUNE uses data obtained from MGA code. PLUM uses data obtained from COMPOS code. The main idea here is to have independent method to get isotopic data. Subsequently, PLUM and FUNE use independent methods to measure the mass of plutonium.

2.5. COMPLEMENTARITY OF FUNE AND PLUM

FUNE, as well as PLUM are devoted to the measurement of plutonium mass in waste drums. The interest of these two systems is to be complementary. They are based on two methods: measurement of the spontaneous neutron emission of even isotopes of the plutonium (plutonium 238, 240 and 242) for FUNE, measurement of the photon emission of odd isotopes of the plutonium (plutonium 239 and 241) for PLUM. It is necessary however to note that the two methods require the knowledge of the isotopic composition of the plutonium.

The interest of those two systems used together, is due to the fact that the plutonium waste drum could be "polluted" with the emission of neutrons from other material than the plutonium or could contain plutonium with a lot of self-absorption or metallic screen. The joint use of the two systems allows to enhance the measurement capability and to explain some discrepancies between values announced by the operator and those measured by the IRSN.

3. UTILISATION OF PLUM FOR MEASUREMENT OF OTHER RADIOACTIVE MATERIAL

PLUM is dedicated to the calculation of quantities of plutonium in standardized drums (100-liter or 200-liter drums, cylindrical geometry) with a density range between 0.1 and 0.4. The analysis has been automated with a user-friendly software interface.

However, this gamma spectrometric measurement system can be set to analyze any radioactive container (with the same kind of mass, volume and density than stated above) with qualitative (identification of radioelement) and quantitative results (calculation of activities).

For now on, the dedicated and automatic software for that purposes is not available. In order to measure radioactive material other than plutonium, an expert on gamma spectrometric measurements would have to do the setting of the instrumentation and the spectrum analysis. Nevertheless, small effort would be required to adapt the existing tool for the characterization of a specific radioactive material.

4. WASTE DRUM MEASUREMENT DONE BY THE OPERATORS

In order to improve the management of nuclear materials contained in waste, the IRSN/IRSN wanted to accurately know what technical facilities the operator was using to qualify and to quantify the nuclear material contained in the waste. In this respect, the operators completed a questionnaire sent to them by the IRSN/IRSN on this subject.

The production of waste is simply made up of their waste activities. Each installation performs a sort according to its own criteria. The criteria used for this categorization are the nature of the waste (solid or liquid), the measuring devices, the type of packaging. Therefore, earth and demolition rubbish is found, (plaster, breeze blocks, metals, dismantled machinery), metallic waste (small part tooling, shells and end pieces, electrical cables, fuse bases), technological waste from installation operations (plastics, cloth, papers, glass and metals), liquid waste (liquid effluent, silts and used solvents), as well as resins and incineration ashes.

Quantification of the mass of nuclear materials requires varied methods and facilities given the diversity of the waste. However, it is possible to mention that:

- Quantification of the plutonium and uranium in the solid technological waste calls, in general, for the use of gamma spectrometry and passive neutron counting (notably for the plutonium). These measuring facilities whether used separately or not, have been used to quantify the nuclear material contents in 87% of items containing materials put out for waste annually.
- Quantification of plutonium and uranium in the solid metallic waste (solid parts) requires the use of active neutron interrogation. In fact, if the metal objects are relatively voluminous they reduce the emission of the gamma photons.
- Quantification of the plutonium and uranium in liquid waste often requires optical emission spectrometry for the uranium and spectrometry α for the plutonium.
- For large volume containers, the dose flows are measured, the measurement results of which are not related to the mass of nuclear material. Weight measurements are also used to quantify the material contained in the bags.

The detection limits for the measuring facilities are very varied according to the devices, their use, the physical chemistry form of the material (solid, liquid, metallic oxide...) and the packaging. It is possible to summarize.

- The detection limits of plutonium (mass of materials per container) are much lower than those for uranium for all the measurements common to the above mentioned materials. The detection limit for plutonium is less to the gram with an average estimated value of 100mg/container. The detection limit for uranium is in the order of a hundred grams per container. However, the values may vary from 0.2 g/container to 500 g/container for uranium.
- The detection limit for plutonium quantified by gamma spectrometry whether linked or not to passive neutron counting is in the order of 100 mg/container of solid waste, even 67mg. In the case where the detection limit is very low and equal to 0.7 mg/container, it seems that the estimate of this value results from measurements and calculations even of spectra type. As a result, the validity of such detection limits depends on the reproduction of the manufacture of the waste packaging. In fact, these containers must contain, for example, uranium with a

constant mixture or plutonium of the same isotopic composition. Mixtures could lead to falsifying the value of the detection limit.

- Active neutron interrogation leads to a much higher detection limit than the previous limit for plutonium equal to 0.5 to 20 g/container.
- As indicated above, the detection limit for nuclear materials contained in liquid waste is of the same order of size as that in solid waste 100mg/container of plutonium and 200g/container of uranium.

Table 1. Nuclear material and detection limits depending on the means of measurement

Means of Measurement	Nuclear Material	Detection Limit
Gamma spectrometry	depleted uranium	4 10 ⁻³ Bq/g of ²³⁸ U 2 10 ⁻² Bq/g of ²³⁵ U
	uranium	10 ⁻² g to 0.2g of ²³⁵ U or 7 Bq/g of U
Gamma spectrometry and passive neutron counting	plutonium	0.7 mg/container to 140 mg/container average value 100 mg/container
Active neutron interrogation	uranium and plutonium	80 to 500 g/container of U 0.5 to 20 g/container of Pu
Passive neutron counting	uranium and plutonium	100 mg/container of Pu 1g of U or of Pu (gantry)
Optical emission spectrometry	uranium	161 g/container to 361 g/container average value 200 g/container
Alpha spectrometry	plutonium	0.27 mg/container to 798 mg/container average value 100mg/container
Measurement of dose flow and irradiation	uranium and plutonium	5 µGy/h or 0.5 rad/h
Weight	uranium	0.2 g/bag to 1 g/bag average value 0.5 g/bag
Analysis by liquid scintillation	tritium	10 ⁻⁴ g/base

The number of containers whose nuclear material is lower than the detection limit represents 22% of the annual number of containers of waste (approximately 10000). The mass of nuclear material put out to waste whose quantification by container is less than the detection limit is low, being 5% of the annual mass of uranium put out to waste and 1% of the annual mass of plutonium.

In relation to the type of packaging of materials put out as waste, they are numerous; it shall be noted that the 118 litre container is very often used. They are then often crushed and placed inside a 220 litre container into which concrete is injected. The solid waste is, in general, compacted into 220 litre containers sealed by concrete or into much larger volume containers (several m³). As a result, this waste is, and will no doubt over a number of years be

difficult to recycle.

5. CONCLUSIONS

IRSN has a long expertise in measuring plutonium waste drums. For instance, for the last five years, IRSN's inspectors, using on-site measurement devices, have controlled most French facility producing plutonium waste drums. This represents about 250 measurements on waste drums. From this feedback, we think that there is not a unique method that can solve every kind of measurement on waste. The use of passive neutron and gamma spectrometry techniques, which are complementary, is a powerful methodology to assess most cases.

The overview of measurements techniques used by French operators shows the diversity of waste and measurement systems. From this feedback, it appears that there is not a unique method that can solve every kind of measurement on waste.

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