DETECTION AND CHARACTERIZATION OF VERY LOW LEVELTRANSURANIC ELEMENTS IN DRUMMED WASTEBEFORE STORAGE

Nicole Bardy, D. Chardon
P. Giesberger, JY. Benard
Commissariat a' l'Energie Atomique
France

ABSTRACT

To be able to sort drummed waste before storage, their contents must first be characterized and quantified. A measuring assembly designed to detect low levels of contamination in 100 and 220 liters (25-55 gallons) drummed waste, has been used for the last 15 months at the CEA facilities at Bruyères-le-Chatel. The measurement takes place in a low-level background lead cell. The radioelements are detected using gamma spectroscopy.

The drum motions, data acquisition, data reduction and results generation are controlled by a central computing unit. The system has proven capable to detect a few milligrams of Plutonium in a 220-liter (55-gallon) drum holding waste of 0.2 g/cc (12.5 lb/ft^3) density.

INTRODUCTION

Drummed waste has to be characterized for transuranics contents in order to determine the destination of the drums (storage or recycling).

Several methods are used for this purpose at the S.P.R. (Service de Protection contre les Rayonnements), at the C.E.A. Centre in Bruyères-le-Chatel, in France.

Passive operational methods are based on gamma spectroscopy and the measurement of neutrons from spontaneous fissions. An active method using 14 MeV pulsed neutrons under development is nearly complete.

This paper presents the results obtained from a low-level activity gamma spectroscopy measurement system.

This system allows the detection of a few milligrams of plutonium present in a 220-liter (55-gallon) drum (density 0.2 [12.5 lb/cuft]) for an acquisition time of 1000 seconds.

This performance is obtained for waste containing exclusively transuranics.

This system is set to accept 220-liter (55-gallon) drums weighing 50 to 70 kgs (110-155 lbs) and for 100-liter (25-gallon) drums weighing between 20 to 30 kgs (44-66 lbs).

It can also be used to measure 20-liter (5-gallon) capacity bagged waste.

DESCRIPTION OF THE EQUIPMENT

The system is located near the drummed waste storage area, a relatively high background area. Therefore, shielding is necessary to be able to detect low levels of contamination.

Measuring Cell

The cell is manufactured using very low background level lead (0,3 to 1 Bq.g-^{1}), 10 cm (4 inches) thick (manufactured by LEMER, France).

It consists of the following:

- the shielded cell receiving the drum during the measurement: internal dimensions 170 x 82 x 90 cm (67 x 32 x 36 inches)
- a motor driven door
- a lead shield for the detector
- an electro-mechanical system which allows rotation of the drum and positions it in front of the detector.

The Electronic System

The detector is a high purity Germanium diode with a relative efficiency of 21 percent (HVPS, preamplifier, associated amplifier), connected to an encoder, and a GOUPIL 3 computer equipped with an analyzer (CANBERRA).

Measurement Sequence

The drum is introduced manually into the unit. A program automatically controls the measurement sequence.

- Drum Movement: During the measurement, the drum rotates and thus minimizes the effects of the non-homogeneity of the contamination.
- Drum is measured at three levels; it is therefore examined totally, and the position of the contamination could be determined in case of non-homogenities.
- The detector collimator has been designed so as to minimize the effects of the cross-talk between the three areas.

- Acquisition and Analysis of the Spectrum: In the present system, the program adds the three obtained spectra and analyzes the total result.
- Recording the Results: The results are printed in a standard format which presents:

the data concerning the drum: number, origin, weight, type (100 or 220 liters)

the level of alpha activity at the time of the determination, the radioelements detected and their project activity in 300 years with their descendants (mainly AM 241), for the purpose of long-term storage.

SYSTEM PERFORMANCE

Effect of the Low-Level Background Lead Shielding

Efficient shielding is provided by 10 cm of low activity lead. This shielding reduces the three main sources of background in the environment (Ref. 1, Fig. 1 and 2):

- Natural levels of TH 232, U 238 and K40 from the concrete building: the table in Fig. 2 shows the peaks of the background level, with the door open and closed. The effect of the lead shield is readily noticed (a factor of about eight on the peak areas).

The K 40 radiation is attenuated by a factor of 16 due to the lead shield. The activity measured inside the unit is probably partially due to the molecular sieve connected to the detector (dewar and sensor rod), and can be minimized by careful designs of the sieve location.

For a 220-liter drum monitored for 60,000 seconds, the detected activity K 40 is about 500 Bq.

- Co-60 and Cs-137 are present in the steel.

The steel used inside the unit has been chosen for its low level of activity.

For a 220-liter drum and 60,000 seconds acquisition time, the reading is:

Cs-137: < 10 Bq

Co-60: 25 Bq

With the door open (Fig. 2) there is a high level of background activity due to Co-60. This is certainly due to the metallic structures of the building (door, drum transfer system, etc.), made of standard commercial steel.

- Radioelements in the present drums in the storage areas: with the door open, uranium peaks are

detected. The lead protection greatly reduces their contribution to the background level.

Finally, the Compton background level is reduced by a factor of 140 around 120 KeV and 40 towards 600 KeV.

Detection Range

Critical Level: This is calculated from the Compton background level, following the recommendations of the Group of Work Normalisation No 5 of the CEA. The threshold area is calculated according to the formula:

$$S_D = 4{,}4\,(RB)^{1/2}$$

- S_D is calculated, for a given time of acquisition, for a probability level of 97.5 percent (alpha risk = 2.5%).
- R is the resolution in KeV of the detected peak
- B is the Compton background level in C.KeV.

Therefore, for Pu 239, the critical level is 8 mg in 1000 seconds, calculated at an energy of 375 keV.

Fig. 3 shows a spectrum obtained in three times 1000 seconds (measured in 3 parts), on a 220-liter drum, in which 15 mg of Pu 239 had been distributed.

On the other hand, using a 0.4 cm iron screen, several grams of plutonium can be measured.

Measurement Precision

There are three sources of errors:

- The error in the efficiency curve measured in the center of a standard matrix drum: ± 3%.
- The statistic error on the peaks of interest: this depends upon the measurement time.
- The error in the positioning of the source: this is the most important one.

The measurement method has been designed, considering that the activity is distributed evenly in the drum, which in our case is the most probable hypothesis.

In the case of a single source placed in the drum, the error could be big.

To estimate it, we have carried out measurements while locating a known quantity of Pu, in different positions within a drum (of 100 and 220 liters).

The results obtained are given in Fig. 4.

It can be noted that the error in the Pu amount is not more than ± 25% except in the case of a source placed in the center of the drum; the over estimate could be ~ 40% in this case.

This problem can be solved when the source has been located. This is possible if the spectrum obtained on every

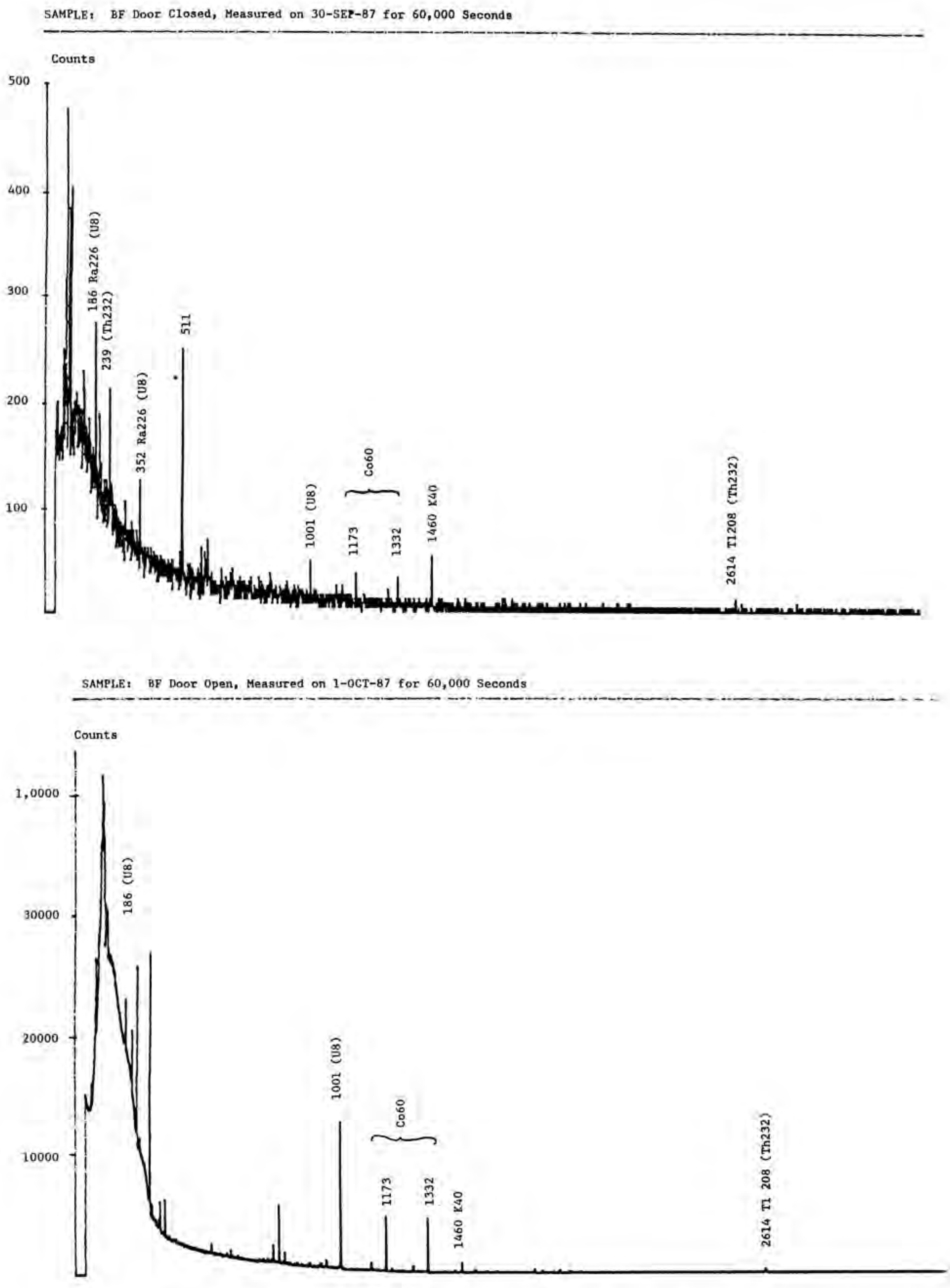

Fig. 1. Sample: BF Door Closed, Measured on 30-SEP-87 for 60,000 Seconds.

Element Assigned To (keV)	Door Open	Door Closed	Remarks
U-Ra (186)	6601	318	Uranium Storage
Th 232 (239)	1750	223	
U-Ra (352)	823	134	
U-Ra (609)	616	131	
Cs 137 (662)	200	45 (Background)	
U-Ra (1001)	32163	76	Uranium Storage
Co 60 (1332)	12721	81	Untreated Steel Structures
K 40 (1460)	2350	148	
Th 232 (2614)	1124	62	

Fig. 2. Comparison of the Background Noice Door Open and Closed in 60,000 Seconds.

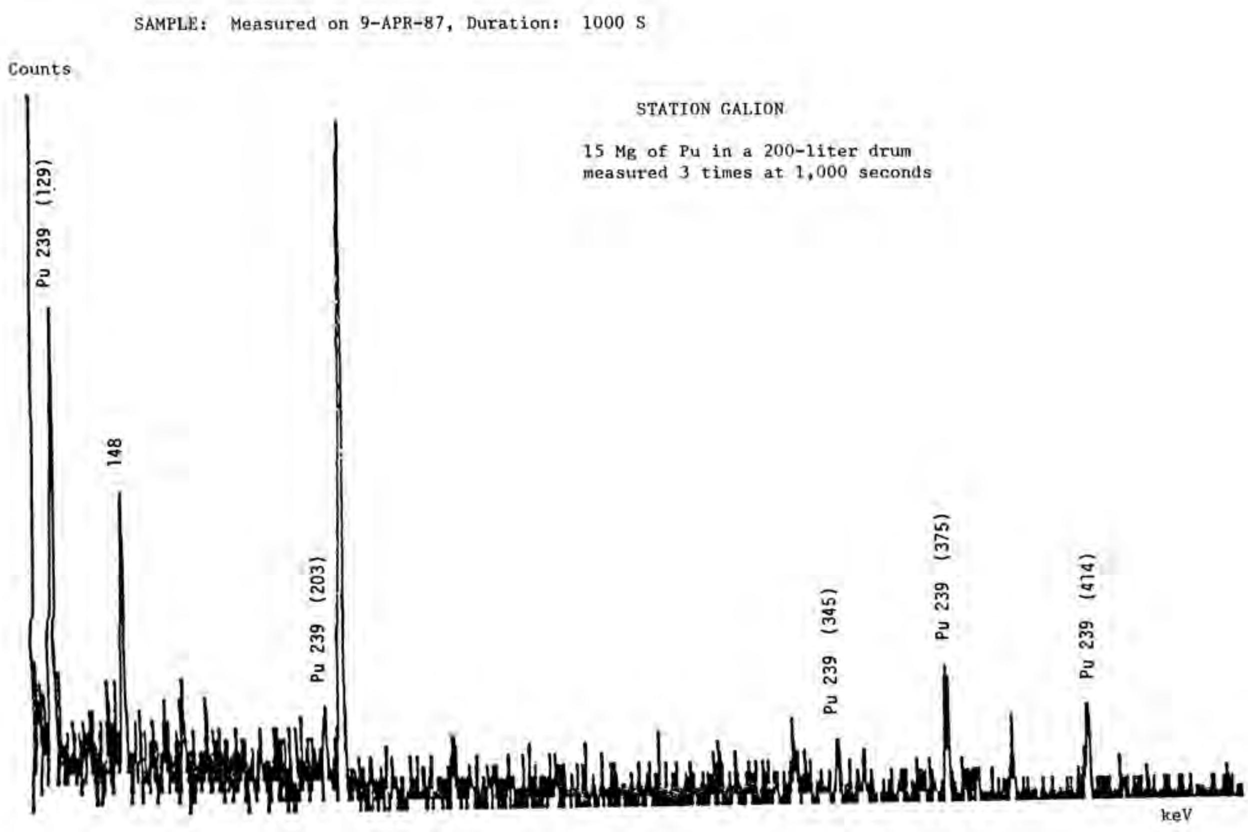

Fig. 3. Sample: Measured on 9-APR-87, Duration: 1000 S.

220 Liter Drum

Actual mass: 188 mg of Pu 239		Actual mass: 403 mg of Pu 239	
219 (+ 16%)	187 (- 0.5%)	463 (+ 15%)	413 (+ 2%)
265 (+ 41%)	218 (+ 16%)	568 (+ 40%)	393 (- 3%)
217 (+ 16%)	189 (- 0.8%)	370 (- 8%)	447 (+ 11%)
Axis of Drum	Edge of Drum	Axis of Drum	Edge of Drum

100 Liter Drums

Actual mass: 1,190 g of Pu 239
1,393 (+ 17%)
1,579 (+ 33%)
897 (- 25%)
Axis of Drum

Fig. 4. Measuring in a Low-Level Background Station Case of a Localized Source.

TABLE I

Actual Compared to Measured Values of Pu-239 in Standard Matrix Drums.

Actual Value (mg of Pu-239)	Measured Value (mg of Pu-239)	Difference
5.1	7.6	+ 49% (measurement near detection threshold)
20.7	20.8	+ 1%
49.5	55.4	+ 12%
99.8	90.2	- 10%
188.3	177.4	- 6%
357.0	429.0	+ 20%
1224.0	1137.0	- 8%

part of the drum is analyzed. A further development of the program has been foreseen for this purpose.

It must be borne in mind, however, that this is not a frequent event and that an even distribution is the most probable case in our drums.

Results obtained from drums with a known activity level:

Pu active sources were placed in standard matrix drums, and the results were compared with the actual quantities.

The combination of the theoretical errors is made quadratically, and is $\pm$ 20% TO $\pm$30%

The results obtained, as compared with the actual values, are given in the Table I here below:

CONCLUSIONS - FUTURE DEVELOPMENTS

The described system have been operational for several months and provides satisfactory results for the types of drums measured.

It must be pointed out that this equipment can also be used to measure 20-liter bags of waste.

The main developments foreseen are:

- Locating the source in the drum in case of uneven distribution. The precision of the measurement can thus be improved.
- Full automatization of the measuring unit: the drums are identified in the storage area, brought into the measurement cell, and returned to the storage area without human intervention.

BIBLIOGRAPHY

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