

## Measurement of the Plutonium Hold-Up in Glove Box before Dismantling - 15111

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### ABSTRACT

The French Bruyeres-Le-Châtel CEA (Commissariat à l'Énergie Atomique et aux Énergies Alternatives – French Alternatives Energies and Atomic Energy Commission) research center is going through a period of cleaning and dismantling of these old treatment facilities on fissile materials and others associated nuclides. Some very large glove boxes contained some machines like melting furnace which can't be moved and are dismantled in situ. We need to evaluate the quantity of fissile materials to prevent criticality risk, to bring the evidence to the safety authority, to manage for the best the radioprotection of the workers and to manage waste production. A melting furnace in a glove box was dismantled. Measurements by gamma spectrometry with an HPGe detector (High Purity Germanium) and passive neutron counting (coincidence counting with shift register) are done in situ (furnace in the glove box). In order to refine the measurements, some data acquisitions by sector of glove box were carried out. In view of the geometries to characterize, some modellings have been created, in order to correct the different attenuations and to determine the plutonium masses content in the glove box, including the use of infinite energy technique. For the measurement by passive neutron counting (coincidence counting with shift register), we have used two slabs in high density polythene including nine He3 counters in each slab. This measurement technique is complementary to gamma spectrometry, and allows quantizing the fissile material which would be included in interstice of high Z material. The 100-L and 200-L drums produced during the dismantling were also measured. Based on simplified assumptions to analyze in situ measurements, plutonium weight could be evaluated.

This approach of measurement in amount, during and after dismantling, has allowed to quantize the fissile material mass present in the glove box and didn't produce waste package which wouldn't respected the technical specifications of storage outlets. The results show good agreement between in situ and waste measurements.

### INTRODUCTION

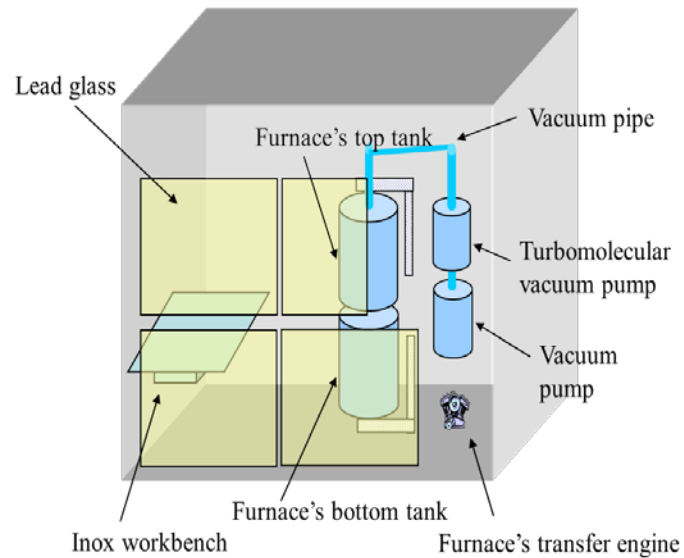
The French Bruyeres-Le-Chatel research center is going through a period of cleaning and dismantling. Estimation of fissile materials quantities in various process equipment is necessary to comply with criticality, safeguards, radioprotection and waste management, during the different phases. Some specific equipment can't be moved and must be dismantled in situ. Both neutron counting and gamma spectrometry are used for the determination of plutonium quantities in the presence of different radionuclides [1]. For attenuation correction, methods, from simple assumptions (straight line attenuation) to complex models through the infinite energy attenuation [2], can be used. HPGe spectrometry provides the isotopic composition of plutonium or other fissile materials. Neutron counting offers an additional method to the gamma spectrometry, less sensitive to high-Z materials. Used with specific electronics, multiplicity or others parameters are achievable.

### SET OF PROBLEMS

The installation wants to dismantle a melting furnace contained in a glove box (Figure 1 and Figure 2)

which contains fissile materials. The achievable quantity through the in/out weight (with uncertainties) gives a value greater than 350 g (considering only the Plutonium), incompatible with the criticality risk and safety.

One solution is to achieve in-situ measurement by spectrometry gamma and passive neutron counting, to reduce this value. We also provide a measurement system during the dismantling, to manage wastes.



**Figure 1 : Simplified scheme of the furnace in the glove box**



**Figure 2 : Furnace inside the glove box**

## HOLD-UP EVALUATION

### First Run of Measurements: Global Measurement

We first measure the entire box by gamma spectrometry with the following equipment:

- EGPC 15 HPGe detector (Intertechnique),
- DSPEC Plus spectrometer (Ortec),
- Visu software (Automatismes et Mesures),
- Cadmium shield (0,5 mm) in front of the detector,
- Lead shield (5 mm) around the detector.

This measurement (Figure 3) provides the identification of the major radionuclides (U-235, Pu-239, Pu-241, Am-241, Am-243) and the isotopic composition.

However, due to the complex geometry (furnace, pumps, engines...), we aren't able to interpret the results in term of mass or activity.

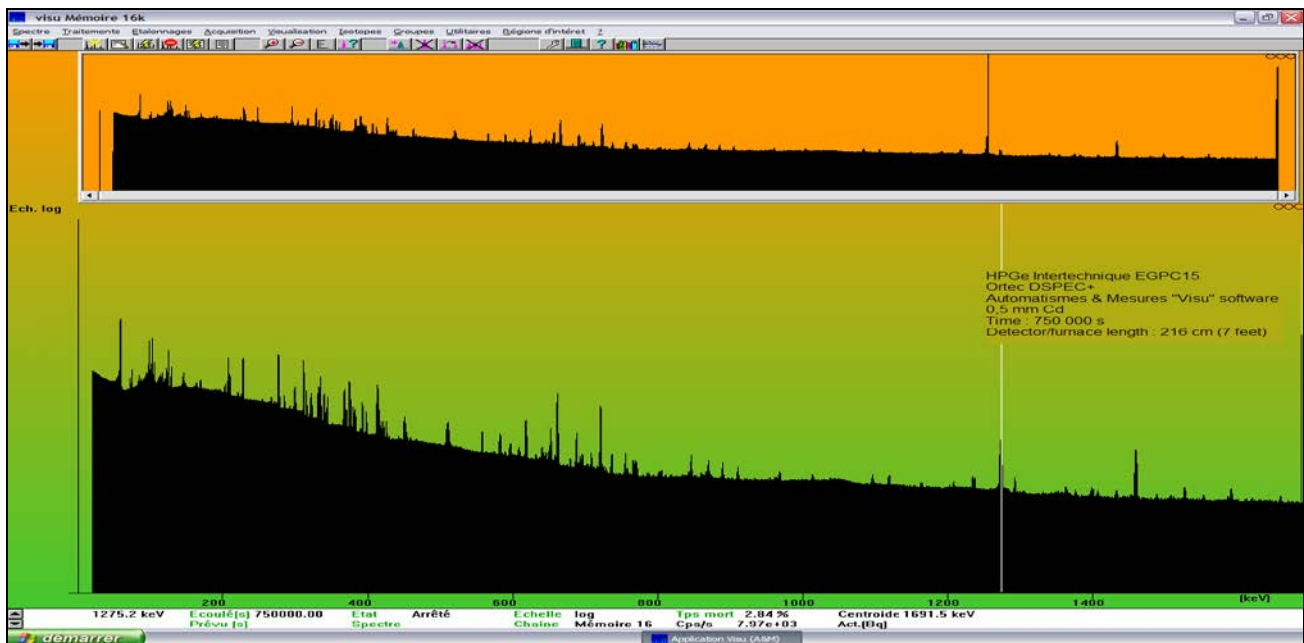


Figure 3 : Spectrum of the furnace

### Second Run of Measurements: "Quadrant" Measurement

In order to simplify the geometry, we divide the furnace and pumps in four quadrants. These quadrants are focused on the furnace and the pumps. This division gives cylindrical elementary volumes.

We use the same equipment as used in the first phase but the detector is now closer in order to view only the chosen quadrant (Figure 4).

The hypotheses are:

- all the fissile materials are into the middle of each volume,

- attenuation factor is only due to the layer thickness.



Figure 4 : HPGe detector in front of the glove box

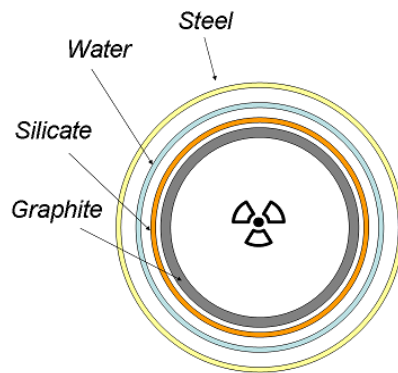


Figure 5 : Example of the simplified scheme of the bottom tank

These hypotheses are confirmed by a dose rate cartography which does not show hot spot of radiation, just a higher dose rate near the furnace and the pumps. Moreover, these assumptions are reasonably over estimated:

- due to the process, the fusion is in center of the furnace, so the major part of fissile materials is here,
- due to the cylindrical symmetry of the furnace, each point (front and behind) contribute equally and could be reduced to a middle point.

We obtain a total Plutonium weight equal to 48 g. The other radionuclides are minor to present a criticality risk.

In order to estimate the confidence in our results, we put all the fissile materials at the opposite side of the detector. Shields contribute twice to the attenuation correction factor. Taking this assumption in consideration, the total weight of plutonium is 98 g.

Quadrant		Pu weight
1	Bottom tank	≈ 16 g
2	Top tank	≈ 18 g
3	Vacuum pump	≈ 7 g
4	Turbo molecular pump	≈ 7 g
	Total	48 g ± 50 g

**Table 1**

So the final result, which avoids the criticality risk, is: 48 g ± 50 g.

Nota: using the infinite energy attenuation method, we obtain a similar weight. This confirms our relative simplified approach.

### **Neutron Measurements**

We use two neutron slabs which make up of an array of 3He tubes inside a slab of polyethylene moderator and cadmium shield (to reduce the neutron flux from the back face). The electronic units are: Antech shift register (AMSR150), with Novelec amplifier/discriminator (SADN) and summation modules (SADP).

The distribution of the R (real) value for hundred measurements shows a large dispersion (Figure 7). This is due to the neutrons originate from ( $\alpha$ , n) reactions from Plutonium and Americium-241, in oxide form, in low-atomic-number materials.

The results (from neutron measurement) give us a final plutonium weight less than 100 g ± 100 g. This value is coherent with the gamma value.



Figure 6 : Neutron slab in front of the glove box

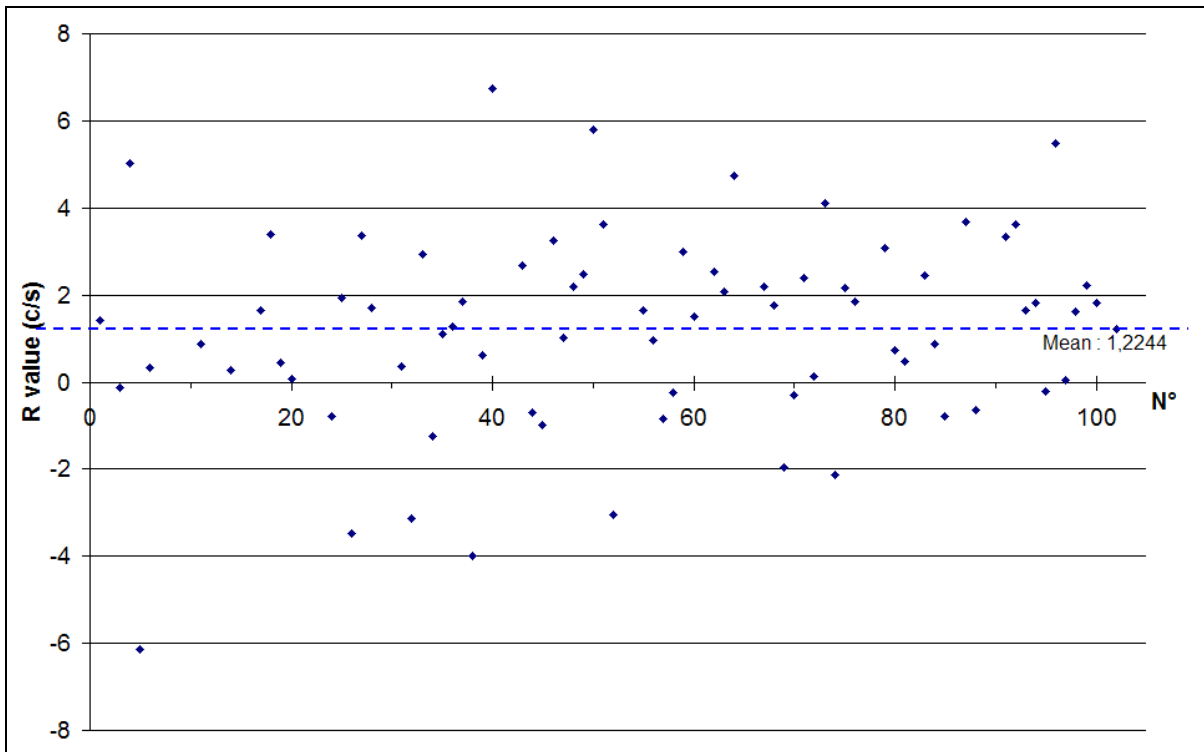


Figure 7 : Distribution of the R (real) value for hundred neutron total counting

## WASTE MANAGEMENT

We develop a fast characterization system (Figure 8) which could receive 20-L packets or 100/200 L drums during the dismantling of the glove box. It provides the operator a fast way (typically 10-15 minutes) to decide whether his waste could be evacuated, compared with the specifications of radioactive

waste collection centers and storage.

Due to the specificity of the radionuclides (most transuranics, many peaks mostly below 400 keV), the chosen detector is an HPGe detector.

The configuration is:

- HPGe LOAX detector (Ortec),
- DSPEC Plus spectrometer (Ortec),
- Visu software + embedded software (Automatismes et Mesures),
- 0,5 mm Cd shield,
- Lead collimator.



**Figure 8 : SYDECAR – Systeme DE CARacterisation Rapide (Fast Characterization System)**

During the dismantling, we follow the evolution of the cumulative weight of Plutonium (Figure 9). After the destruction of the furnace and the pumps, the weight of plutonium is equal to  $55 \text{ g} \pm 30\%$ . This value is coherent with the in-situ value.

The additional mass of plutonium obtained at the final stage of the dismantling, come from old wastes which are reprocessed during the work.

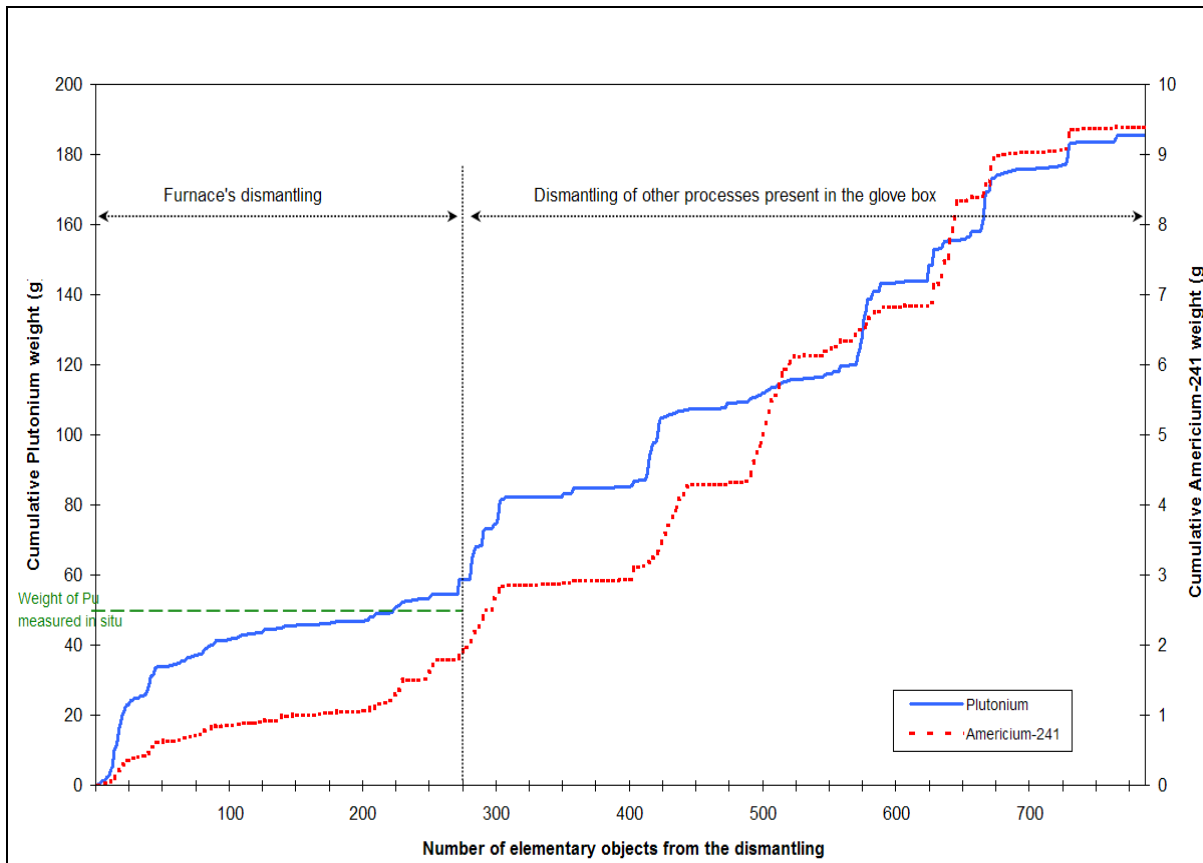


Figure 9 : Evolution of the cumulative weight of plutonium and americium during the dismantling

## CONCLUSION

By gamma spectrometry, and with simple hypotheses, it's possible to give a reasonable value of the fissile materials contained in this furnace. This is possible due to the relative simple geometry of the furnace.

With passive neutrons counters, the measure is difficult to interpret, due to the presence of  $(\alpha, n)$  reactions. But the results confirm those get by gamma spectrometry.

The value from the waste measurements and from in-situ measurements agree.

## REFERENCES

- [1] D. Reilly, N. Ensslin, H. Smith Jr, S. Kreiner, "Passive Nondestructive Assay of Nuclear Materials", NUREG/CR-5550, LA-UR—90-732, March 1991
- [2] J. Morel, D. Chauvenet, M. Etcheverry, J. Monier, T. Arnal, "Mesure des quantites de plutonium par spectrometrie  $\square$ ", ESARDA Symposium, Liege, 1985.