

Characterization of Low Level and Intermediate Nuclear Waste: improvements in calorimetric measurement combined radiological modelling.

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ABSTRACT

Since 40 years, calorimetry allows to determine precisely amounts of nuclear material within a container. Combined with gamma spectrometry, calorimetric measurements give plutonium contents with a low level of uncertainty for each isotope. Calorimetry is used in a lot of nuclear facilities all around the world for plutonium or tritium inventories. It is one of the best solutions to estimate the overall quantity of nuclear material on a wide range of mass, from a few milligrams up to kilograms of radionuclides.

In the case of drums containing nuclear waste, characterization was very challenging due to complex matrix, volume and low level activities. The measurement time was a real problem with large volume container and organic matrix with high air content.

New designs of SETARAM calorimeters allow to measure nuclear material contents in very large container (up to 385 liters) and/or with a very low detection limit (10 μ Watt). These new concepts are based on single measurement cell and vacuum chamber for low level uncertainties. The acquisition system and electronic temperature regulation are highly improved to increase the detection limit, accuracy and precision of the measurement. These modern technological developments within the calorimetry field give the promise of faster measurement times and precise measurements.

These devices can be used in waste management of nuclear facilities with a better accuracy and lower uncertainties than others techniques. It is a non-destructive and non-intrusive method. There is no matrix effect which can bias the results. Waste can also be in solid, liquid or gas phase. The new calorimeters are able to give amounts of plutonium, tritium or others alpha or beta emitter radionuclides and allow determining the classification of waste (low level or intermediate).

A measurement approach combining very large volume calorimeter, gamma spectrometry and radiological modelling will be presented as an integrated automatic waste characterization system. This system with a calculation step is taking into account the gamma emitters contribution on the total thermal power measured within the calorimeter. The aim of the system is to give a precise quantification of the radionuclides in the waste container with a precise value of the total uncertainties.

INTRODUCTION

The calorimetric signal results from the measurement of the heat flow between the measurement cell containing the radioactive sample and the thermal block. This thermal heat flow generates a signal (μV) based on time which is recorded by a data acquisition card. When the heat flow signal is stabilized, this one is converted to a thermal activity (W) by means of the linear calibration curve.

The applications of calorimetry in nuclear industry consist in the measurement of heat generation from radionuclides. Calorimetry is used as a nondestructive assay (NDA) technique for determining the power output of heat-producing nuclear materials. The heat is generated by the decay of radioactive isotopes within the item. Hence, the knowledge of the isotopy easily allows mass of each isotopes calculation:

$$M_i = R_i \times \frac{W}{\sum_{j=1}^N R_j \times P_j} \quad \text{Equation 1}$$

Where M_i is the mass of the i^{th} isotope, R_j isotope fraction, P_j specific power [1,2] and W the thermal power measured with calorimeter. $\sum_{j=1}^N R_j \times P_j$ is the effective specific power of the mix (P_{eff}).

The most successful radiometric calorimeter designs fit the general category of differential calorimeter for heat flow measurement (described in Figure 1).

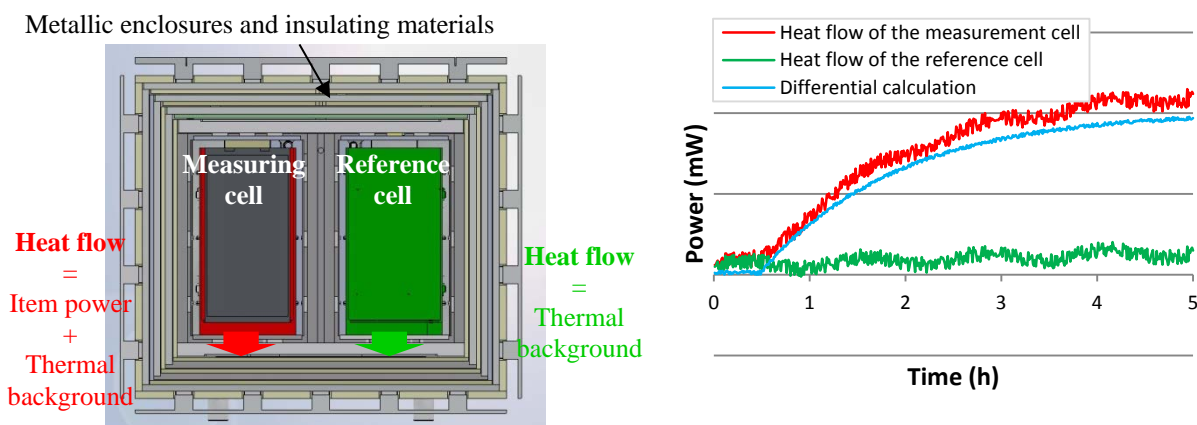


Figure 1. Schematic cross-section of the calorimeter with a thermal block insulated by multilayer insulation and several metallic plates. The calorimeter is surrounded by an external temperature control layer.

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In the Figure 1, a differential calorimeter with a measuring cell for a container loaded with an active product to be analyzed and a reference cell for a container that is either empty or loaded with an inactive product. Principle is based on measuring the heat flow rate between the block maintained at a constant temperature and the containers containing the sample and the reference. The differential calorimeter design is more precise because residual fluctuations caused by environmental changes (external temperature control variation) are cancelled out in the net signal formed between the measurement and reference.

LARGE VOLUME CALORIMETRIC ASSAY

Lot of calorimeters are designed for the study and characterisation of materials in the fields of organic, pharmaceutical products, food and biological products, mineral and ceramic products, metal and metallurgical products. In these cases measured volumes are less than 1 L [3].

Large volume calorimeters were developed for nuclear material control and accountability with volume from 1 L to 90 L [4]. Recently, the new differential calorimeter, LVC1380 (Figure 2) [5,6], was developed by KEP Technologies for nuclear waste measurement (dimensioned for waste drums having volumes up to 250 L) with a power range between 3 mW and 3.5 W (i.e. tritium accountability range of 10 mg to 10 g).



Figure 2. View of the LVC 1380.

The stabilization of the heat flow signal and so the duration of the experiment depends on many parameters such as the thermal power range, the weight of the sample or the thermal environmental conditions. In practice, it is necessary to wait for the heat flow signal to stabilize with a standard deviation in the same range as the baseline. The duration of the experiment of the calorimeter varies 3 to 7 days and the measurement duration is about 10 hours.

The calibration of the calorimeter is carried out by mean of a Joule effect device and allows the conversion of the heat flow voltage signal (μV) to thermal power (mW). This Joule effect consists in an electrical resistor placed in a canister having the same geometry as the radioactive drums. The heat flow is generated in the range 0-3.5 W to get the same measurement conditions than the waste drum.

Tests lasted many months in severe atmospheric conditions (temperature from 10 to 30 °C and hygrometry from 20 to 90 %) and with different types of containers. Finally, the accuracy of the measurement with the LVC1380 calorimeter is shown on TABLE 1. It has been estimated to be 2 % rel. for power up to 1000 mW and 15 % for low power near 10 mW.

TABLE 1. Accuracy of measurement.

Power Range (mW)	H-3 Range	Accuracy (% rel.)
10 mW	0.03 g H-3	15 %
500 mW	1.54 g H-3	2.5 %
1000 mW	3.08 g H-3	2 %
3000 mW	9.26 g H-3	2 %

In spite of the severe conditions, the measurement quality of this technique has been retained and the accuracy is considered as sufficient. The other advantages of the calorimetry for radioactive waste measurement are various: no sample preparation, no destructive test, no dependence on sample geometry, no matrix effect, absolute method for accountability.

ISOTOPY MEASUREMENT

In the case of calorimetric assay, isotopy measurement (e.g. with gamma spectrometry) has two goals:

- Measuring the concentration of each isotope to calculate the effective specific power (P_{eff} described in Equation 1).
- Combined with modeling codes, it allows calculation of heating induced by high energies and/or high activities gamma rays emitters, such as Cs-137 or Co-60.

Gamma rays spectrometry

The measurement of the isotopic distribution of a plutonium and uranium sample is a well-developed technique. To calculate the amount of nuclear materials, the global system needs to well-known the isotopic fractions and the specific power (mg/W) for each isotope. A planar HPGe detector is used in most applications. With a front surface area of 200 mm² and a thickness of 10 mm, this type of detector gives a good compromise between resolution and efficiency. The multi peak analysis of all gamma emitted in the 120-400 keV energy range allows the determination of the isotopic ratios with MGA [7] or PC/FRAM [8] software with accuracy better than 1 %.

Each isotope has different specific power (see TABLE 2) and different contribution on the output power. Hence, we applied the Morris method (described in the Figure 3) [9] to classify the importance of isotopes and understand how each isotope impacts the thermal power of an Am-Pu sample.

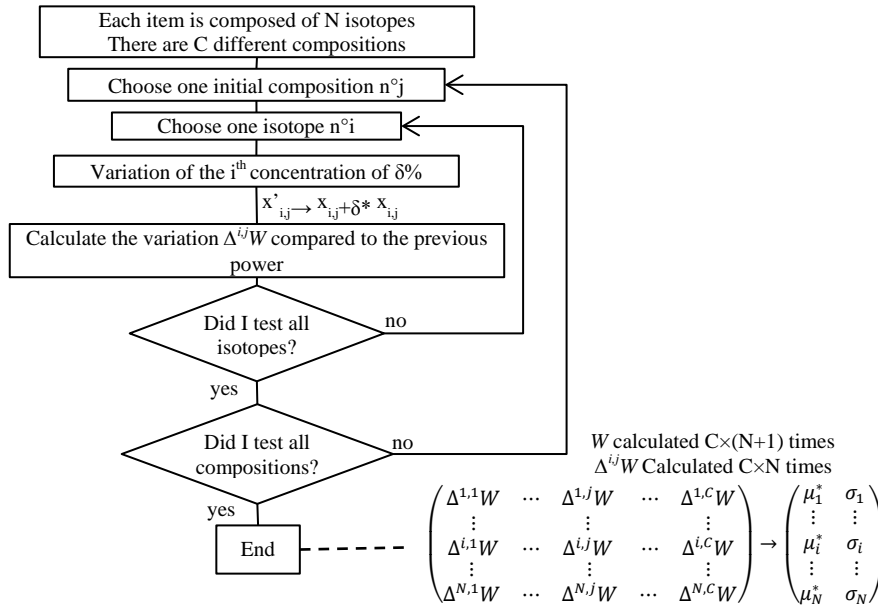


Figure 3. Morris method.

At the end of the process we calculate for each isotope:

- $\mu_i^* = E(|df_i|)$: Sensitivity. High value shows the output signal is very impacted by this parameter.
- $\sigma_i = \sigma(df_i)$: Interactions and/or no-linear effects. High value shows high importance in the signal due to interaction between parameters or no-linear effects.

In the Figure 4, we applied the Morris method, using 28 different isotopic compositions of mix plutonium-ameridium composed of six isotopes (mainly found in ref.[10]) and made varied each concentration in range $\delta = [+5\%; +20\%]$.

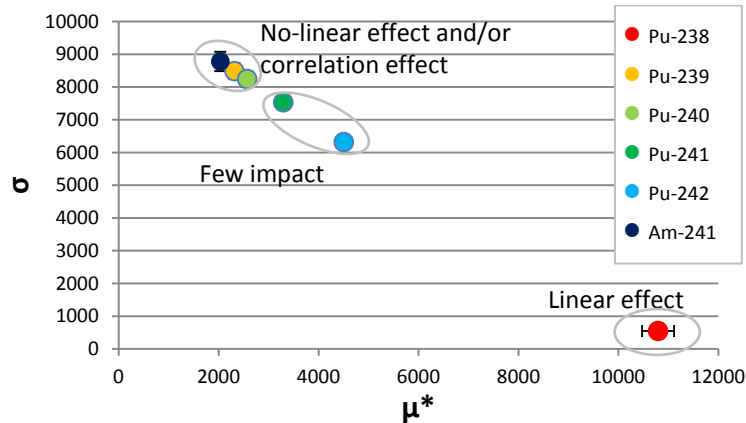


Figure 4. Thermal power sensitivity study.

Three groups appear in the Figure 4 :

- Am-241, Pu-239 and Pu-240 represent a large part of the mixing. So, a small variation induced large concentrations variations and so thermal power changing.

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- Pu-241 and Pu-242 are a small fraction of the AmPu mixing and have small specific power. These isotopes do not impact the thermal power
- The mixing AmPu contains little Pu-238, but this specific power is very high. Variation of the Pu-238 quantity induces proportional variation of the thermal power but do not impact the concentration of others isotopes.

Figure 4 shows that during the isotopic assay, we must be careful with Am-241 and Pu-238 which have an important impact on the effective specific power calculation.

Heating sources

Alpha and beta particles heating

In the case of beta emitter, the total reaction energy is shared between the beta particle, neutrino, the excitation energy and recoil energy of the daughter. The characteristic distance of 2 MeV electrons is a few millimeters in the matrix and all of the kinetic energy of the beta particle is absorbed by the sample. One 200 keV or 2 MeV electron in polyethylene produces respectively ~ 0.15 keV and ~ 11 keV X-ray due to bremsstrahlung radiation. The neutrino is not absorbed, so its energy is lost. At last, the thermal power measured by a calorimeter from beta emitters is the product of the activity and the average beta-particle energy, plus the de-excitation gamma-ray energy deposit taking into account the decay scheme. The gamma-ray contribution can be calculated with Monte Carlo code, e.g. MCNPX [11].

In the case of alpha emitter, the heat output produced by the decay of alpha emitter, in case of complete stopping of particles is distributed as follows for the Pu-239 [12]:

- Alpha particle: 98.17%
- Recoil: 1.67%
- Conversion electrons: 0.1%
- Auger electrons: 0.035%
- X-rays: 0.01%
- Gamma-rays: 0.001%

The range of 5-6 MeV alpha particles is around $5 \mu\text{m}$ in common materials. Thus, all of the energy released by charged particles during the alpha decay will remain within the item as heat. Low-energy gamma-rays and X-rays can be assumed to be totally absorbed in the matrix and the specific power of alpha emitter is directly deduced from the Q-value. The gamma-rays and X-rays have very low energy and can be assumed totally absorb by the plutonium itself or by the matrix.

Because the heat-measurement result is independent of material and matrix, it can be used for the inspection of any material form or matrix. The specific powers and the associated uncertainties are listed in TABLE 2 for all plutonium isotopes and Am-241.

TABLE 2. Specific powers, uncertainties and maximum energy loss due to escape of photons and neutrons [1Error! Bookmark not defined.,13].

Isotopes	specific power (mW/g)	Relative Standard Deviation (%)	Max. Energy Loss (%)
Pu-238	567.57	0.05	3.1E-2
Pu-239	1.9288	0.02	1.3E-3
Pu-240	7.0824	0.03	5.5E-4
Pu-241	3.412	0.06	2.5E-2
Pu-242	0.1159	0.22	3.2E-2
Am-241	114.2	0.37	5.1E-1

In the case of the alpha emitter study, the assumption “all particles are totally absorbed in the matrix” is pretty good; neutrons and photons represent less than 0.05% of the total release energy (exception for the Am-241).

Gamma heating

In “classic” nuclear waste, heat generation in matrix is assumed to be only due to absorption of alpha and/or beta particles and the heating induced by gamma rays energy deposits (gamma-heating) is negligible. But more and more nuclear wastes appear to have very high beta emitters activity and/or high energy gamma-rays emitters.

For example, irradiated graphite wastes are contaminated with pure beta emitter such as C-14, but also contain a significant amount of Co-60 due to activated steel-corrosion contamination. In this case we must measure dozen μW but the gamma-heating can reach a hundred μW . The spent fuel assay is another example where a significant amount of Cs-137 can induce a few watts heating.

In these specific cases, the energy deposit in matrix, over-pack and calorimeter due to gamma rays must be estimated using modelling codes, such as MCNPX code [11] and the MCNPX’s F6 tally which gives the energy deposit in medium.

Three points must be studied in case of no-negligible gamma-heating:

- Is the calorimeter’s working disturbed by the gamma-heating?
- How much the gamma-heating of the inspected item will bias the calorimetric assay?
- How to correct the bias due to gamma-heating?

In the frame of irradiated graphite characterization with calorimetry [14], we have studied the impact of the gamma-heating on the calorimeter working and the calorimetric assay. In worst case, the gamma-heating of the regulation blocs reached several tens of μW . The heating of these parts is not measured during the assay and so does not disturb the calorimeter performance. In addition, this heating is negligible compare to the power of bloc’s regulated thermal barriers (several watts) and the power injected by temperature sensor (around 1 mW is injected due to the sensor self-heating).

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We also studied the gamma-heating according to source localization inside the graphite waste (see Figure 5).

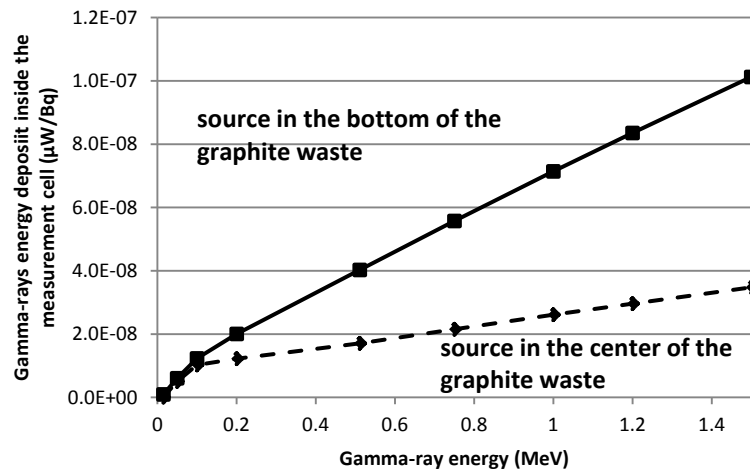


Figure 5. Gamma-heating induced by punctual source inside the measurement cell of a calorimeter.

The gamma-heating inside the graphite waste and the measurement chamber greatly depends on the source location:

- In graphite waste, if the source is not surrounding by graphite or is closed to the outside, gamma rays can easily escape and not induce heating.
- The heating of the measurement cell is very dependent of source position because of the cell is no symmetric. If the source in close to the lateral wall gamma rays mainly go through the thin copper wall. If the source is close to the ground, gamma rays are absorbed by the thick back part of the cell.
- We want measure a power range between several tens μW and several hundred μW to quantify C-14 inside graphite. But beta emitters' activity such as Co-60 could reach $1\text{E}9$ Bq per item [15], so the gamma-heating can reach $\sim 100\mu\text{W}$.

The knowledge of the beta emitters isotopy, thanks to gamma ray spectrometry, allows correction of the gamma-heating. But it is important to design the measurement cell to decrease the dependence of the gamma-heating vs. source localization.

CONCLUSIONS

Calorimetry has been used for many years in nuclear industry for accountability of nuclear materials and the many advantages are well known:

- The heat measurement is independent of material and matrix type.
- No physical standards are required.
- The heat from the entire item is measured.
- The response is independent of the source location.

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- Calorimetry is very precise.

The heating induced by gamma rays is generally not an issue and so calorimetry is considered nearly bias free. But in some specific cases the measured power and the gamma-heating have similar value. Hence, we must calculate the gamma-heating for a correct interpretation of the calorimetric assay.

With the development of the very large calorimeter LVC1380, calorimetry was shown to be efficient technique for the management of nuclear wastes within 100 L or 200 L containers. The accuracy of the measurement is 2% in the range of 1 W (3 g H-3), the limit of detection currently is 9 mW (28 mg H-3) and further improvements can be done to reduce to 3 mW (9 mg H-3) by improving the stabilization of the heat flow signal or reducing the duration of measurement.

REFERENCES

[1] D. S. Bracken, and Clifford R. Rudy, "Principles and Applications of Calorimetric Assay", PANDA 2007 Addendum.

[2] D. S. Bracken, R. S. Biddle, L.A. Carrillo, P. A. Hypes, C. R. Rudy, C. M. Schneider, M. K. Smith, "Application Guide to Safeguards Calorimetry", LA-13867-M, January (2002).

[3] http://www.setaram.com/product_categories/calorimetry/

[4] G. Jossens, C. Mathonat, G. Etherington, Large volume and sensitive calorimeter for nuclear applications: latest developments for greater accuracy and shorter measurement times. INMM 2011, July 17-21, 2010, Desert Springs, USA

[5] F. Bacheleta et al., "Large Nuclear Calorimeter for Non Destructive Assay on 55 gallons Tritiated Waste Drums", 55th annual meeting INMM – July 20-24, 2014 – Atlanta (USA).

[6] G. Jossens, C. Mathonat, F. Bachelet, "Nuclear waste calorimeter for very large drums with 385 liters sample volume", INMM 2013

[7] R. Gunnink, "MGA : A Gamma-Ray Spectrum Analysis Code For Determining Plutonium Isotopic Abunbances", LLNL report UCRL-LR-103220, April 1990.

[8] Thomas E. Sampson, Thomas A. Kelley and Duc T. Vo, "Application Guide to Gamma-Ray Isotopic Analysis Using the FRAM software", Report LA-14018, Los Alamos National Laboratory, September 2003.

[9] https://en.wikipedia.org/wiki/Morris_method

[10] D. Reilly, N. Enselin, H. Smith, Jr. and S. Kreiner, "Passive Nondestructive Assay Manual – PANDA", <http://www.lanl.gov/orgs/n/n1/panda/> (1991).

WM2016 Conference, March 6-10, 2016, Phoenix, Arizona, USA.

[11] <https://mcnpx.lanl.gov/>

[12] G. Bortels, "What information can alpha spectrometry provide to calorimetric assay of mixed alpha-particle emitters?" Proceedings of the international workshop on calorimetry, Ispra, Italy March 23-27 1992.

[13] S. Guardini, "Performance values for non-destructive assay (NDA) techniques applied to safeguards: The 2002 evaluation by the Esarda NDA working group", EC-JRC-IPSC, Ispra, Italy, 2002.

[14] P. Le Tourneur et al., "New concepts and instruments for ^{14}C and ^{36}Cl measurements in i-graphite", PREDEC 2016, February 16-18, Lyon, France.

[15] Inventaire national des matières et déchets radioactifs 2009, Catalogue descriptif des familles, ANDRA.