Proceedings of Global 2015
September 20-24, 2015 - Paris (France)
Paper 5190

Mobile laboratories: an innovative and efficient solution for radiological characterization of sites under or after decommissioning

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Abstract – *During the operation and the decommissioning of a nuclear site the operator must assure the protection of the workers and the environment. It must furthermore identify and classify the various wastes, while optimizing the costs. At all stages of the decommissioning radiological measurements are performed to determine the initial situation, to monitor the demolition and clean-up, and to verify the final situation.*

Radiochemical analysis is crucial for the radiological evaluation process to optimize the clean-up operations and to the respect limits defined with the authorities. Even though these types of analysis are omnipresent in activities such as the exploitation, the monitoring, and the cleaning up of nuclear plants, some nuclear sites do not have their own radiochemical analysis laboratory. Mobile facilities can overcome this lack when nuclear facilities are dismantled, when contaminated sites are cleaned-up, or in a post-accident situation.

The current operations for the characterization of radiological soils of CEA nuclear facilities, lead to a large increase of radiochemical analysis. To manage this high throughput of samples in a timely manner, the CEA has developed a new mobile laboratory for the clean-up of its soils, called SMaRT (Shelter for Monitoring and nucleAR chemisTry). This laboratory is dedicated to the preparation and the radiochemical analysis (alpha, beta, and gamma) of potentially contaminated samples. In this framework, CEA and Eichrom laboratories has signed a partnership agreement to extend the analytical capacities and bring on site optimized and validated methods for different problematic.

Gamma-emitting radionuclides can usually be measured in situ as little or no sample preparation is required. Alpha and beta-emitting radionuclides are a different matter. Analytical chemistry laboratory facilities are required. Mobile and transportable laboratories equipped with the necessary tools can provide all that is needed. The main advantage of a mobile laboratory is its portability; the shelter can be placed in the vicinity of nuclear facilities under decommissioning, or of contaminated sites with infrastructures unsuitable for the reception and treatment of radioactive samples. Radiological analysis can then be performed without the disadvantages of radioactive material transport. This paper describes how this solution allows a fast response and control of costs, with a high analytical capacity.

I. INTRODUCTION

The analysis of radionuclides in waste produced during the decommissioning operations or in soils located in the perimeter of nuclear facilities is an essential step of the global process of radiological characterization. In the case of the management of soils potentially contaminated by radioactive substances, several studies has showed that by investing money to get an accurate characterization of the initial state, before remediation works, enables to save money later especially by optimizing the radioactive waste volumes. In Fontenay-aux-Roses, the first remediation works began in 1955 after the dismantling of the plutonium facility. During these works, a large feedback made evaluate the practices. The evolution on the global cost of rehabilitation is illustrated on Fig. 1 below.

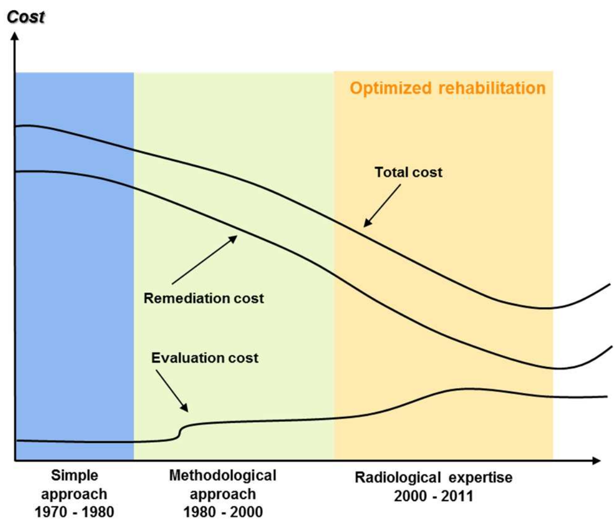

Fig. 1. Evolution on rehabilitation costs

Three different periods can be highlighted:

1970-1980: with the simple approach, remediation works were managed in function of direct radiation measurement. There was no control of the works duration and associated costs. In this situation, it was quite hard to finalize the rehabilitations.

1980-2000: In this period, a methodology was progressively established, clean-up methods were improved, and some soils evaluation was performed before remediation works. The consequent better characterization of the pollution made the cost lower.

2000-2015: Since the 2000s, several software dedicated to data treatment allows getting accurate maps of contamination and associated impact. Modern tools for in situ measurements allow performing a radiological expertise and so optimizing the rehabilitation costs while controlling the project deadline.

In terms of investments to get the initial state, the CEA has developed a mobile unit, composed of several tools to estimate the radioactive contamination in soils, such as the LAMAS [1] and VEgAS [2] vehicles, the Kartotrak software [3] that converts radiological data into maps, and SMART mobile laboratories [4].

The current operations for the characterization of radiological soils of CEA nuclear facilities, lead to a large increase of radiochemical analysis. To manage this high throughput of samples in a timely manner, the CEA has established a partnership with Eichrom laboratories. This partnership provides a solution to CEA's medium and long term decommissioning works, by bringing on site experimented staff, operating with validated methods, in modern laboratories in an auto sufficient way.

II. BENEFITS OF MOBILES LABORATORIES

II.A. Technical description

The laboratory is installed in a container called «shelter», transportable via road and airline [4]. It can be moved using a lifting crane via slings through integrated rings or using a fork lift via incorporated holes. Furniture and equipment is fastened to stabilize them during transport. The lab is split in different areas. In the first zone, samples are received in a dedicated hatch equipped with retention trays. A locker is also available for staff personal belongings. The second zone contains a wet laboratory bench and two ducted fume hoods provided by Ermaflux®, for the chemical preparation of the samples. In the first hood, solid samples are heated, dissolved in concentrated nitric acid, and diluted for specific radionuclides extractions. The main extraction techniques used are liquid/liquid extraction for plutonium and strontium, and resin exchange chromatography for other actinides. The laboratory equipment (pumps, centrifuge, and balances) is fixed on the workbench.

All chemical preparations are performed in the two fume hoods, especially designed for the shelter. Each one extracts the hazardous vapors from the work area with a flow rate of 850m3/h. The air is filtered by a F7 dust filter, a CAMACTIF active carbon chemical filter, and finally by a high efficiency H14 filter before being rejected in the environment.

An air-handling unit conditions the air and creates a -10Pa depression in the lab. Three split-type air conditioner units assure a stable temperature in the shelter. After the chemical preparation, the samples are transferred into containers adapted to the geometric requirements of the measurement and finally moved to the third zone, which contains the α, β, and γ spectrometers, described in the next section.

Waste created during the sample preparation is stored in specific containers in the last zone, the technical room. The containers used are in agreement with the French National Radioactive Waste Management Agency (ANDRA) specifications. This room also contains the electric supply system, the fire alarm system and the low oxygen detection system. The general design of the shelter is given in figure 2.

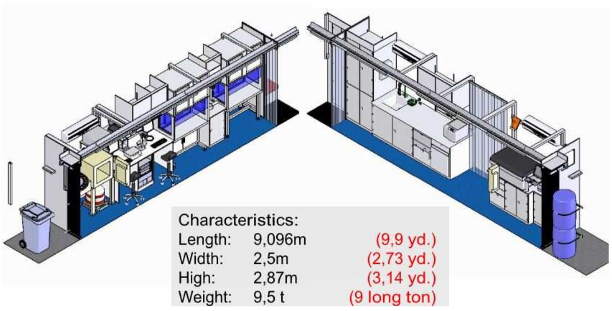

Fig.2: General design of the shelter

Liquid wastes are stored in suitable containers behind the fume hoods. The sink, the dishwasher, and the security shower are connected to a double tank, to control the quality of waste water before discharge.

II.B. instrumentation and equipment

Gamma spectrometer

The detector used for gamma spectrometry analysis is a coaxial type HPGe detector (Canberra® GC2518). Its characteristics (a relative efficiency of 25 % and an energy resolution of 1.8keV at 1.33MeV and 0.850keV at 122keV), are suitable for the measurement of low and medium activities of current gamma tracers. The detector head is set inside a shielded measurement cell covered with low-level lead (50Bq/kg). It is connected with an automatic sample loader (24/7 measurement) and with the analyzer software Interwinner 7 provided by Itech® Instruments (Fig. 3).

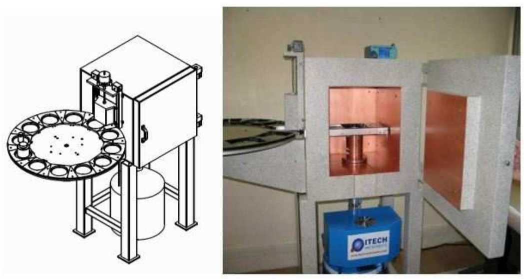

Fig. 3: Sample loader and shielded measurement cell

This heavy equipment (2500kg) is firmly fixed to the shelter, by the creation of fastening arms, as shown in figure 4.

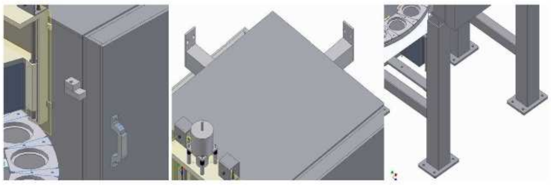

Fig. 4: Fastening arms, for holding the equipment during transports

Space is saved by the compact shape of the digital analyzer compared to analog electronic units. The software used for spectrum acquisition and to control the sample loader is installed on special computers with a reinforced hard disk (Fig. 5). All the data collected from the spectrometers is centralized inside a database developed by the team via a network.

Fig. 5: Digital analyzer ORION and computers with reinforced hard disk

Alpha spectrometers with semi-conductors

The alpha-emitting radionuclides are mainly encountered in the nuclear industry, at the front end (uranium, plutonium) and at the back end of the fuel cycle (plutonium 238, 239, 240 and 242, americium 241 and 243, neptunium 237 and curium 242 and 244).

The process of radiochemical separation and electrodeposition used in the SMαRT allows the realization of sub-samples optimized for measurement by alpha spectrometry. Two complementary methods have been selected for the measurement of alpha emitters: the alpha spectrometer with semi-conductor detectors and the liquid scintillation counter PERALS®.

An eight semi-conductors ensemble (Fig. 6) provided by AMETEK® and ITECH® (for SMART 1 and 2 respectively) can, with an adapted counting time and a specific chemical preparation, identify, quantify and discriminate the expected alpha-emitters radionuclides (resolution (full width at half maximum) around 20keV), and can reach very low detection limits.

Fig. 6: Alpha ensemble with semi-conductors

This technique involves a long chemical preparation step which may be an unsuitable in some cases such as post-accident situations. Liquid scintillation is more suitable in post-accident situations as it does not require a long chemical preparation.

Alpha liquid scintillation PERALS®

The PERALS® (Photon-Electron Rejecting Alpha Liquid Scintillation) allows a faster measurement of some alpha-emitting radionuclides, such as plutonium or uranium. The principle of the sample preparation is based on a liquid-liquid extraction in which the isotope of interest is isolated with a scintillating and complexing agent which is the matrix condition appropriated to the measurement. The principle of detection of alpha particles is based on the liquid scintillation technique, associated with an α/β pulse discriminator (PSD), which allows the rejection of 99 % of β pulses (Fig. 7).

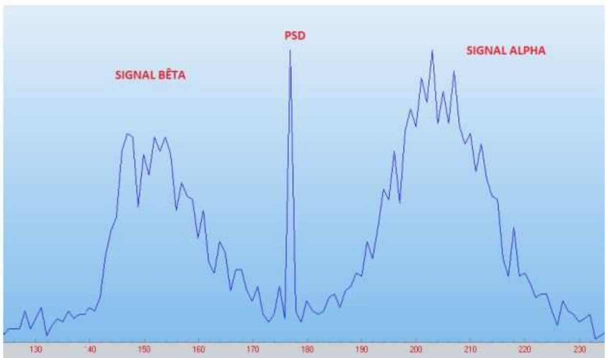

Fig. 7: Alpha-beta discriminator (PSD) of PERALS®

The two detectors 8100AB, provided by (ORDELA®) are inserted in an electronic NIM rack (Fig. 8). Pulses from the detector are analyzed by a double-card encoder.

Fig. 8: Detector and analyzer for PERALS®

The detector characteristics (efficiency 99.7%, background < 0.001cpm, resolution of 200keV at 4.78MeV), allows quick analysis (24h) of solutions containing uranium or plutonium.

Beta liquid scintillation

The SMαRT is equipped with a liquid scintillation counter connected to a sample loader (24/7 measurement), which allows the continuous counting of beta emitters such as ^{3}H, ^{14}C, ^{90}Sr and ^{241}Pu. Strontium 90 for example, is extracted from solid samples by acid dissolution, and isolated by coprecipitation and liquid-liquid extraction. Its daughter after decay, yttrium 90, is measured by liquid scintillation. The B2910TR model provided by Perkin Elmer® has a loading capacity of 408 samples. Beta spectra are analyzed with the Spectraworks® software.

II.C. Characteristics, advantages and benefits of mobile laboratory

The SMART shelters are built on shelf. Its built time (from 6months to 1 year) depends on the analysis design requested by the project to work on. If gamma spectrometry, alpha spectrometry or liquid scintillation analysis are required, the built will be short, as the new shelter will be sister shipped from SMART 1 and 2. This concept make SMART a “plug and analyze” device. Compared to a built of a new lab dedicated to the project, the SMART will be immediately available. This operational immediacy is a real benefit for project managers.

SMARTs are transportable via road and airline, as described in paragraph 2.1. It can be placed wherever the best place on the site to be inspected is. Closed to the drilling or the effluent treatment plant, closed to another laboratory, inside or outside the security barrier, the choice belongs to the project manager. This advantage to be directly on site, dedicated to the site will drop global costs.

As SMART is consecrated to one customer and one site, the samples to analyze will be from and just for the connected site; there will be no reassignment of urgency with samples from another contract.

This real time measurement is a main advantage to the project manager: he can orientate drillings and make quicker decisions.

This consecration to one customer and real time measurement involve quicker delays, less interrupted work and less spent money.

One main characteristic of SMART is to be on site, it means no samples will be released out of the site. Benefits are drop of transport costs, and less constraints for all involved departments (radioprotection, measurement, delivery).

The SMART solution is economically viable. Compared to construction of a new laboratory for a polluted site with no facilities, this is an affordable solution. A SMART is less expansive and has less constraints compared to an existing laboratory that would need a reassignment.

As described in paragraph 2.2, SMαRTs are designed with state of art technologies, with performant methods and devices. Compared to a stationary laboratory, analytical performances are equivalent, and showed in table I.

TABLE I

Routine Minimum Detectable Activity

Device	Radioelement	MDA (Bq/kg dry)
Gamma spectrometry	^{137}Cs	5
Alpha spectrometry	$^{239+240}Pu$	1
Liquid scintillation	^{90}Sr	30

Table II presents initial capabilities of analyzes per week for 2 radiochemists in a shelter. Capabilities of radiochemistry have been increased with the partnership with Eichrom laboratories (see part III).

TABLE II

Initial capabilities per week for 2 radiochemists

Device	Radioelement	Number per week
Gamma spectrometry (1h)	^{137}Cs	150
Alpha spectrometry	$^{239+240}Pu$	8
Liquid scintillation	^{90}Sr	8
Liquid scintillation	filters $\alpha+\beta$	500

III. BENEFITS OF MOBILE/FIX LAB COMBINATION

In the aim to optimize the SMART solution, in 2014 CEA signed a partnership with Eichrom Laboratories, one of the main European actor for radioactivity measurement labs. Indeed, with their 25 years experiences in radioactivity protocol development (Eichrom resins) and more than 10 years' experience in laboratory management, Eichrom appeared as the perfect partner for the functioning of the SMART.

III.A. Human resource

The collaboration with Eichrom Laboratories is the opportunity for the CEA to obtain quickly experienced and qualified technicians to operate in the SMART on various specific protocols (isotopic plutonium [5], ^{90}Sr [6] …). With the COFRAC (FRench ACcreditation COmity) accreditation (ISO 17025) of the stationary laboratory, available Eichrom' technicians to work in the SMART have followed a complete process of qualification recognized by a competent third party, COFRAC. Eichrom technicians are qualified in intern by training companionship and external training if necessary to complete the technical knowledge.

III.B. Eichrom know-how and expertise

25 years of Eichrom experience in radiochemistry and radioactivity measurement put to use for the SMART solution is the opportunity for SMART customer to obtain quickly and with the less effort to a high level of know-how and expertise in radioactivity measurement. The SMART solution appears as an easy solution to access to quick results in a short time. In this context, SMART solution benefits are the cost and time saving.

III.C. Quality Management (ISO 17025)

Since the beginning in 2003 and the realization of its first analyses, Eichrom laboratories (France, 35) are accredited ISO 17025. This recognition is the guarantee of quality of the internal organization and the technical competences judged by a third party and by one's peers. The high internal level of requirement imposed by Eichrom for the stationary laboratory is the same in the SMART. Eichrom does not know others requirement and that's why it uses every day the best laboratory practices for benefits of its customer.

III.D. Eichrom Laboratory as a back-up solution (support solution)

The support of the stationary laboratory in Rennes to the SMART solution presents specific advantages:

- To propose to customers a back-up solution in the aim to guarantee a business continuity and consequently the respect of the scheduled milestones.
- To propose to customers a supplementary analytical capacity of the SMART in case of unforeseen situation.

IV. APPLICATIONS

In France, and especially in CEA, decommissioning works are often carried out by specialized contractors, supervised by a project manager. The technical specifications specify everyone tasks and responsibilities. Most of the time, radiological characterization of the samples is managed by the CEA, either by a transfer to a local laboratory, if existing, or to an external subcontractor. But often, it's impossible to get the wished reactivity, and each day immobilization of the subcontractor is heavily charged to the CEA. Indeed, the aim of radiological in depth investigations is to locate accurately a contamination. In this situation, each sample analysis result determines the position and the number of the next samples.

In order to illustrate the advantages of the mobile laboratory, let's take a first example: the area located under the hall 03 of Building 31 in Fontenay-aux-Roses, has been the subject of radiological investigations. Indeed, an area contaminated by Ra-226 has been highlight in the soils of the front door, during remediation works in the neighborhood.

The origin of this pollution is known: the CEA of Fontenay-aux-Roses hosted the first French nuclear activities. Between its creation in 1945 and today, many changes happened in the center structure and research activities also. At the origin, the facilities were concentrated in the structures of the old military basement of Châtillon. It used to host the first European reactor ZOE, and several buildings in link with the production of yellow-cake and plutonium, as showed on Fig.9:

Fig. 9. Buildings linked for process of plutonium.

Nowadays, the center has a new structure and some old areas, cleaned-up with period criteria, has been covered to build new nuclear facilities. That's the case of the Building 31, which has been built on the old ditches of the fort, where the yellow-cake residues were discharged.

Fig. 10. Ancient and actual aerial view of CEA/FAR center

The aim of these investigations is to know if the radium contamination spreads under the Building 31, which has been cleaned-up and is going to be destroyed. These works has been performed by a subcontractor assisted by a radiation protection assistance company. Works are quite simple (two perpendicular trenches of 10 meters long, 1 meter wide and 3 meters depth), as shown on Fig 11.

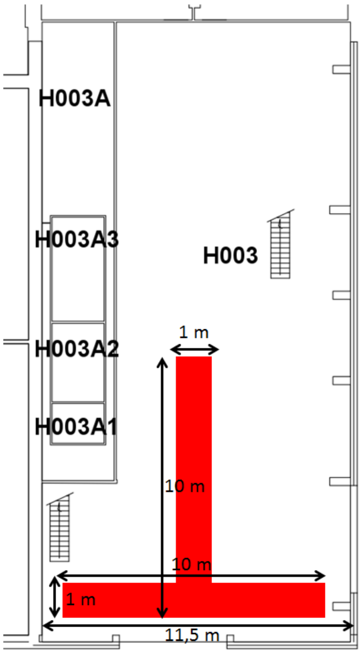

Fig. 11. ICAPP logo.

By the way, these works will generate 84 samples a day, which has to be analyzed by gamma spectrometry. Works continued as long as contamination was present. The highlight area contains soils contaminated by uranium 235, uranium 238, and Radium 226.Results had to be given 24h maximum after, in order to refill trenches with the same soils. Every delay would have stop all the works and cost a lot of money to the CEA.

These samples have been analyzed by the germanium detectors of SMART-1 and SMART-2 in an optimized acquisition time in order to reach detection limits of 100Bq/kg for Ra-226, U-235 and Pa-234m). Thanks to this organization and the sample loaders, results were get in time and cost and duration of investigations were respected. It would have been impossible to get results so quickly with an external laboratory. It is important to notify that for sending samples out of the center, a systematic gamma spectrometry has to be performed in order to organize the sample transport. With this mobile solution, radiological analysis can then be performed without the disadvantages of radioactive material transport, associated radioprotection controls, and administrative organization. It's also possible to control or change the priorities.

Another interesting example is the PETRUS project, consisting in radiological investigations under the Building 18 [7] in Fontenay-aux-roses. The PETRUS hot cell has been built in 1958 in the basement of the Building 18 in the CEA center of Fontenay-aux-Roses, in order to study the radiochemistry of trans-uranium elements. These hot cells were shut down at the end of the 80's. Nowadays, the Building 18 is under decommissioning. Radiological characterization has been asked to the clean-up section of the center in order to evaluate the radiological state of soils under the building and actions to be undertaken.

During the PETRUS project, a 10m depth well has been dug aside the facility. From this well, 12 drillings has been performed to get information about localization about contamination highlighted during the historical study.

Analyses were performed in the mobile laboratory on each sample in order to detect natural or artificial radioactivity. To this end, long time measurements with gamma spectrometry are realized. It provides limit of detection of 1Bq/kg in Np-237. Radiochemical analyses are also performed on each sample. Pu-238, Pu-239, Pu-240 and Sr-90 are researched over a detection limit of 1Bq/kg for the Pu-238 and 20Bq/kg for the Sr-90. With the SMART mobile laboratories, analyses were performed in real time, just after the sampling. Results had to be obtained the day after for the gamma spectrometry and the week after for the radiochemistry. This reactivity is essential because results can impact the position of the next drilling. Also, given all the mobilized staff in different companies (sampling, radioprotection, drillings, geologists), every work stop cost 7K€ to the CEA.

In this case, it's important to define an analytical strategy: A systematic long time gamma spectrometry (8h) was performed on every sample. Hot spots could be highlighted thanks to gamma emissions of Np-237, its daughter Pa-233, and also Am-241. A correlation was observed between these radionuclides. On these samples, a radiochemical analysis was performed in priority for Pu-238 and Sr-90 determination. Chemical protocols optimized by Eichrom (purification in cartridges in vacuum boxes), allowed to analyze 200 samples in 2 months. The SMART mobile solution allowed rising to this challenge, but also to:

-measure all samples on site, without transport out of the nuclear facility perimeter, in confidentiality;

-make sample selection, thanks to immediate gamma spectrometry and adapting detection limits in real time, in order to highlight hot spots for radiochemical complementary analysis;

-managing in real time the future drillings, in order to get a complete characterization while controlling the total costs.

V. CONCLUSIONS

In order to manage radiological investigation or remediation works, it's important to dispose of strong analytical tools to get an accurate radiological state of the aimed area.

Built with state of art technology, the first two SMART can run two radiochemistry batch in parallel (an alpha emitter and a beta emitter for example), with detection limits comparable to a stationary laboratory. SMART is a perfect answer to transport of samples, to avoid loss of time for different services and to avoid cost of radioactive transports. SMART is as well a perfect tool for project managers, as this movable laboratory is consecrated to one site: answers for drilling orientation are faster, decisions are quicker.

Combined with an accredited stationary laboratory as Eichrom laboratories, project manager has a solution for training of technicians, back up of crew, materials and analyses in case of unforeseen situations (more analyzes to perform, material issue, sickness issue…).

SMART-1 and SMART-2 are the two first mobile laboratories developed by CEA and exploited by Eichrom laboratories. Both are already working on the third one. Indeed, the meetings and exchanges with potential users highlighted new needs. SMART-3 may include analyzers for chemical elements as Mercury, Beryllium for example, or asbestos. Also, the SMART laboratory, with its sample loaders, could be really useful on the case of an emergency situation.

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