NucLab Marcoule: A Laboratory Facility Dedicated to Support Dismantling Operations – 14212

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ABSTRACT
Formerly dedicated to plutonium production support, NucLab was renovated to perform a wide range of analyses for dismantling, plant operation and process development activities mainly on Marcoule site and sometimes outside. The Laboratory is CEA entity from Nuclear Energy Division. It provides services to several industrial operators (nuclear process and power plant) in the fields of analytical chemistry, radioactivity measurements, in situ nuclear measurements, decontamination processes and industrial chemistry processes, waste treatments to meet the following analysis requirements. NucLab is able to support research, production and dismantling activities in all part of dismantling operations.

INTRODUCTION
Formerly dedicated to plutonium production support, NucLab has the capacities to perform a wide range of analyses for dismantling, plant operation and process development activities. It provides mainly services to the operators of the CEA Marcoule in the fields of analytical chemistry, metallographic examinations, radioactivity and radiochemistry measurements, in situ or online nuclear measurements, decontamination processes and industrial chemistry studies dedicated to dismantling operations and industrial processes.
With 50 years of experiments, the facilities are designed to analyze all types of nuclear industrial and research samples, being equipped with fume-hoods, glove boxes or shielded cells.

HISTORICAL CONTEXT
The Central Laboratory was established in 1956 in Marcoule, birthplace site of plutonium production in France. The laboratory was commissioned in 1958, in support of the Marcoule fuel reprocessing plant (Usine de Plutonium N°1 called UP1) and auxiliary units including the radioactive effluents and the solid waste treatment plants.
Its routine activity involved two or three 8 hour shifts related to quality and process control. Three extensions were added to the initial facility building between 1960 and 1986 to answer to the new needs and expectations of the site.
The range of activities included the following:
- Online process control (chemical and radiological) via a pneumatic transfer network between plutonium production units and the Laboratory.
- Quality control on raw materials and finished products.

Since the shutdown of UP1 in 1997, the Laboratory activities evolved; they shifted to support dismantling activities (i.e. UNGG reactors), beginning with the removal of radioactive material and rinsing of the process loops. These activities require large numbers of samples with chemical preparation followed by chemical and physical characterizations. The analysis results obtained in this step are fundamental for further waste treatments to guarantee safe decommissioning operations and waste decategorization. The treatment and conditioning of small quantities of exotic radioactive wastes such as organic effluents or highly contaminated wastes also began at that time.
In addition to this huge analytical demand, the laboratory has been reconfigured at the same time

in term of safety, taking into account the new rules (i.e. fire and earthquake resistance) and in terms of capabilities to support research & development and dismantling.
In 2012, CEA and AREVA decided to consolidate the laboratory in a partnership with a new organization called NucLab as a CEA entity operated by AREVA employees.

RESSOURCES AND CAPACITIES
The facilities are currently composed of 5 interconnected building wings covering a total ground area of about 4200 m2 (fig. 1):
The Laboratory is divided into:
- Unrestricted access zones dedicated for non-nuclear areas including the locker room, storage areas, yards and offices (1700 m2).
- Restricted access zone, the workplaces are distributed over 95 fume cupboards, 43 glove boxes and 35 shielded cells (2500 m2).

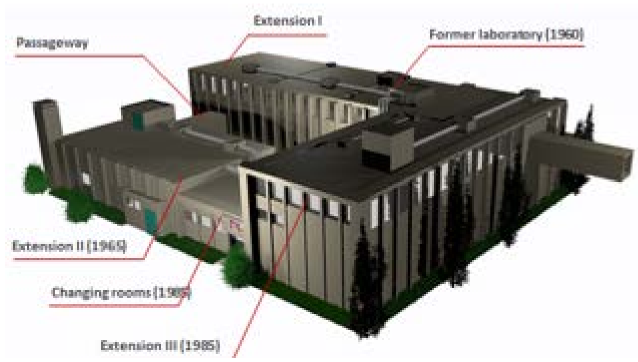

Fig.1: NucLab Building

NucLab is staffed by 75 persons including 65 analysts working in normal hours or 2x8 hour shifts.
The competences of the laboratory are :
- Central management entity (management, security, safety, quality, environment unit).
- Analytical Chemistry.
- Radioactive Measurement.
- Nuclear Measurement.
- Industrial and Chemical Engineering.

The analytical and study capacities of the laboratory can be illustrated by the following data for 2012:
- Treatment of 3500 samples
- 14000 analytical determinations
- Catalog of 329 industrial methods.

The NucLab provides industrial studies and analysis for many customers (mainly in the nuclear fields) including the CEA, AREVA, MELOX, EDF, SALVAREM, COMURHEX, and SOCODEI. NucLab collaborates with the other CEA R and D laboratories located in CEA Saclay, Cadarache and Marcoule in order to develop and new analytical expectations of the customers.

Analytical Chemistry
This activity covers a wide range of capabilities in analytical chemistry in support of research, development and dismantling applications:
- Elemental analysis of cations and toxic elements,

- Specific analyses: CN-, Cl-, F-,NO2-, NO3-, SO42-, PO43-, C2O42-, formiates, acetates, TBP, DBP, MBP, etc…
- Conventional chemical analysis: density, acidity, pH measurement, etc…
- Preparation and characterization: measurement standards, reactants.
- Pretreatment and preparation of samples before analysis: mineralization, dissolution (bitumen, concrete, etc.).
- Development of analytical procedures.
- Characterization of the evolution of TBP under irradiation.
- The analyses are performed by many devices in various environments:
- Monitored zone in conventional working conditions:
- X-ray fluorescence (table top and handled).
- XRD.
- ICP-AES.
- C/S analyzer.
- Atomic absorption spectroscopy.
- GC-MS and µ GC (x2).
- Optical and in situ Asbestos identification.
- Ignition point, flash point.
- Liquid chromatography.
- Ion chromatography.
- FTIR.

Fig. 2: XRF in conventional working environment

- Fume cupboard environments:
  - ICP-AES.
  - Liquid chromatography.
  - Atomic absorption spectroscopy.
  - UV-visible spectrometry.
  - Mineralization, dissolution.
  - TOC analyzer.
- Glove box environments:
  - ICP-AES.
  - C/S analyzer.
  - UV-visible spectroscopy.
  - Particle size analysis.
  - Mineralization, dissolution (fig.3).
  - Liquid chromatography, GC-FID.

Fig. 3: Mineralization in glove box

- Shielded line:
  - ICP-AES (fig.4).
  - Liquid chromatography.
  - UV-visible spectroscopy.
  - Spectrometry, specific electrodes, etc...
  - Mineralization.
  - Balance, microwave oven, density, etc…

Fig. 4: Analytical hot cells (ICP-AES)

Radioactive Measurements

this activity is dedicated to isotopic analysis, quantification of alpha, beta and gamma emitters:

- Uranium-plutonium balance.
- Isotopic compositions: U, Pu, B and Li.
- Determination of beta emitters : ^{3}H, ^{14}C, ^{36}Cl, ^{55}Fe, ^{59}Ni, ^{63}Ni, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{129}I, ^{151}Sm.
- Development of specific measurements (e.g. ^{99}Tc in bitumen-encapsulated waste, ^{129}I by ICP-MS).
- This activity is supported by analytical devices in various environments:
- 1 thermal ionization mass spectrometers.
- 2 liquid scintillators.
- 6 gamma spectrometers including one with sample changer.
- 4 alpha spectrometers with grid chamber detectors.
- 16 alpha spectrometers with semiconductor detectors.
- 1 ICP-MS in fume cupboard (fig. 5).

Fig. 5: ICP-MS in fume cupboard

Nuclear Measurements

This activity is dedicated to in situ radiological characterization of used equipment items (glove boxes, tanks, crushing machine, etc…), decontamination and dismantling monitoring of industrial set up and plants, waste packages. A specific methodology has been developed in Marcoule, adapted to industrial expectations (more particularly in UP1) since 1998 [1]. This topic was developed because the conventional radiological approach was not appropriate to the characterization of industrial devices due to large dimensions and complex shapes. So, if "samples" couldn't be analyzed in the laboratory, the laboratory has to find a solution to make measurements in situ on the site before dismantling.

This approach can be described as follows:

- In situ initial radiological survey of complex configurations (fig.6 and 7).
- Monitoring of dismantling operations.
- Local characterization of the waste.
- Modeling of the residual activity taking into account of the real geometry of the pieces to assess the dismantling strategy (fig. 8) in term of costs, schedule and waste specifications.
- Estimation of the Pu mass distribution with a correct uncertainty (1g up to 100g of Pu).
- Characterization of historical waste not well identified.
- Assessment of waste package management.

Fig. 6 a and b: Gamma Camera and In Situ Spectrometer System (ISOCS) Gamma probe positioned in UP1 plant

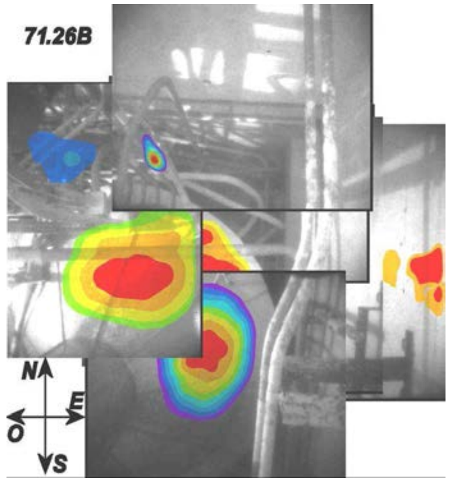

Fig. 7: Hot spot localization in a used evaporator: superimposed images of visible-light image and gamma image.

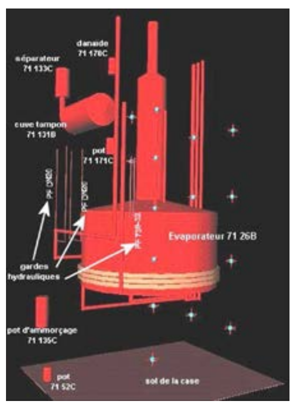

Fig. 8: Modeling of the evaporator to evaluate the residual activity

The analyses are performed using portable devices and spectral simulation software:

- Gamma camera (CARTOGAM).
- Four portable Ge gamma spectrometry systems (coupled with ISOCS and PASCALYS software).
- Six portable Cd-Te gamma spectrometry systems (coupled with PASCALYS).
- NaI portable probe for fast hot spot survey.
- Two dose rate measurement systems for pipes and other configurations.
- MERCURAD dose rate modeling software useful for dismantling scenario.

The specificity of this team is to group into a same entity most of the skills and know-how (radiologic and chemical measurements, modeling) needed to the monitoring of dismantling of industrial plant (about 100 equipment with Pu contamination treated at that time).

Industrial and Chemical Engineering

This activity is dedicated to the analytical development and control in support of industrial processes. It has a strong interaction with the analytical units in the Department.

The variety of the samples leads to work in 3 types of workstations:

- Ventilated hoods for low contaminating samples.
- Glove boxes for contaminating and low irradiating samples.
- Shielded cells for the most active samples (fig. 9).

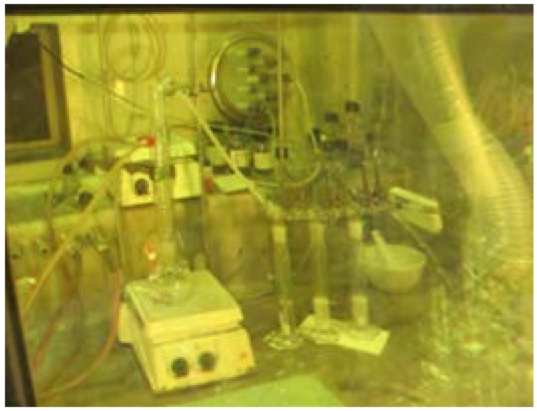

Fig. 9 a and b: Sample preparation in hot cell environment

This activity is supported by analytical devices in various environments:

- Characterization of active samples:
  - Sampling: tools design and assistance to the operator.
  - Macroscopic examination in shielded cells.
  - Mechanical treatment (cutting, grinding, sieving…).
  - Chemical treatment (acidic dissolution, purification, separation…).
  - Specific analytical means (µcalorimetry, DTA/MS-DSC, GC).
  - Physical characterization (densities, water rate, graphite rate…).
  - Study and validation of new chemical processes.
  - Recovery and recycling of Pu.
  - Co-precipitation and reactivity tests for acceptability of big quantities of liquid effluents in Marcoule's Treatment Station.
  - Cement encapsulation:
  - Validation of the formulation.
  - Quantification of gas releases (GC)
- Destruction of active organic matter:
  - Supercritical water oxidation process (fig. 10).
  - Treatment of Pu-charged resins through silver (II)-electrodissolution and oxalic conversion.
- Specific studies :
  - Dissolution of a irradiated UNGG fuel element.
  - Qualification of a nuclearized analytical device (LIBS) for future in situ measurements.
  - Decontamination of debased organic solvents.
  - In Situ Asbestos and elemental measurements with handled IR and XRF.

Fig. 10 a and b: Supercritical set-up for separation processes (blank test and oil storage in shielded cell)

Fig. 11: Electrolytic treatment device installed in shielded cell for ion exchange resin destruction

EXAMPLE OF THE INTEREST OF THE LABORATORY IN THE DECOMMISSIONING AND DISMANTLING OF UP1/MAR 200

A major program is in progress at Marcoule to dismantle the first French defence reprocessing complex UP1. This complex has been commissioned at the end of 50's and operated for 40 years. This extensive dismantling and waste recovery program must therefore cope with a large variety of waste and radiological situations. In many cases, operating events and poor documentation lead to a strong need of characterization activities prior to the studies or initiation of the dismantling operations. Those characterization programs are supported by the tools, analyses and expertise of NucLab. Most of the collected data are used in the safety reports and the measurement methods, calibration and qualification techniques are part of the licensing process of each dismantling or recovery operation.

Example of the MAR200 dissolution unit:
One of the examples that best illustrates this program is the cleanup and dismantling of the continuous dissolvers located in Building 117 of the UP1 complex. The building contained two

parallel dissolution lines (A and B).
For the line A dissolver, the initial analytical measurements revealed the lowest level of fissile material content in the deposits; these results allow to define the filtration parameters and to confirm the possibility of incorporating of the residues in the standard glass containment matrix. Based on these results and data, the safety report was approved. The recovery of the residues is now completed and the dismantling is in progress. All the residues have been completely vitrified.
For the line B equipment, preliminary inspection confirmed the existence of sludge and deposits at the bottom part of the dissolver. The in cell counters confirmed the presence of Pu and provided a rough estimate of the activity. A specific program to collect of 18 active samples was then defined and carried out.
Difficulties appeared in the sample collection as the sludge was located under the bottom support plate of the vessel. This perforated plate was about one inch thick and the preliminary rinsing test showed that the residues were partially blocked under the plate. In order to ensure fully representative sampling, the plate was removed by plasma cutting and specific systems were adapted to allow the collection of the samples for analysis (fig. 12 et 13). The results obtained by the laboratory were taken into account on line for the definition of the interventions and the corresponding safety reports. The assessment of Pu content and its chemical and physical forms were of prime importance for defining of the sludge recovery technique. They were also considered in the safety report prepared for the future elimination route for the resulting waste.
The analytical laboratory provided the project with more than 200 of physical, chemical and radiological data items under approved Q.A. conditions delivered in acceptable time. The required analyses included gamma spectroscopy, chemical analysis of U and Pu, quantification of hydrogen in Pu, Pu isotopic composition, 90Sr measurement, graphite quantification, chemical analysis of cations, anions, acetate, formiate as Pb, B, Ni, Cr total, Cr VI, As, Sb, Hg, Be, Se, Cd, CN.
The wide variety of available analytical techniques in NucLab was necessary to address the uncertainties on the sludge content, chemical composition and physical behavior under transfer and filtration conditions. Based on these results the recovery and filtration of the sludge is currently in progress.
Analytical techniques of NucLab are also used for on line monitoring of D&D operations. In such cases, they mostly contribute to the waste characterization and elimination process. For example, such techniques have been successfully implemented for sorting metallic waste previously stored in pits. The low level counting equipment was installed on site and operated remotely. This allowed a direct and qualified characterization of the waste as LLW (low level waste), the waste were grouted on line without no additional handling or transfer.

Fig. 12: perforation system to collect samples

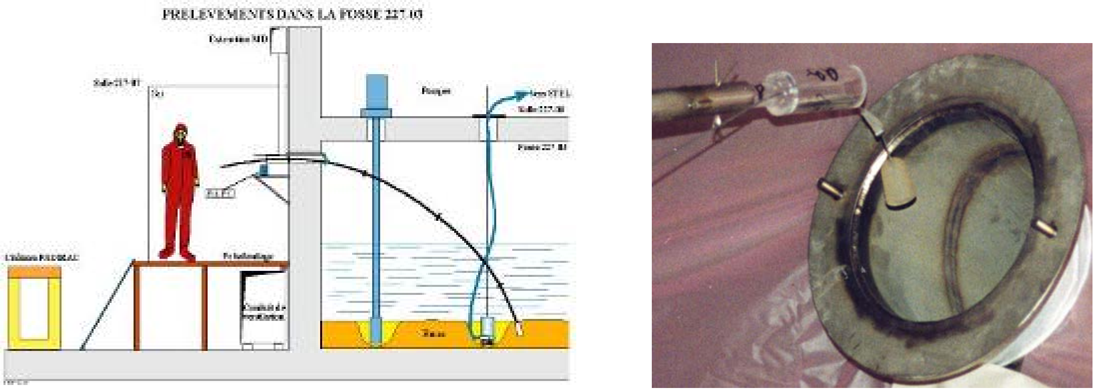

Fig. 13: specific device developed for collecting samples for analysis

CONCLUSIONS

This presentation has described the high technical potential and huge analytical capacity of Nuclab in the field of analyses, examinations, specific waste treatments and studies for dismantling operations and industrial processes. It allows treating analytical topics from laboratory to industrial scale. Even they were initially developed for Marcoule activities since 50 years, these activities are now be proposed to other nuclear industry entities

REFERENCES

1. S. Dogny, H. Toubon; Animma conference proceeding, Marseille (2009)