

HOT CELL DEVICES DESIGNED TO OPEN AND DISSOLVE IRRADIATED PROFIL R TARGETS

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ABSTRACT

Within the framework of the French LLRN (Long Life Radionuclides) transmutation program, several irradiation campaigns were launched in the Phenix fast neutron reactor in order to prove the transmutation efficiency^[1]. The PROFIL experiments are specifically devoted to the improvement of the effective cross-sections of various isotopes. One of the final goals is the decreasing of existing uncertainties on the capture-section of the fission products intervening in the loss of reactivity in the fast reactors. This program is separated into two parts: the PROFIL R irradiated in standard fast reactor spectrum and PROFIL M irradiated in moderated spectrum. The PROFIL R and PROFIL M irradiations were completed respectively in August 2005 and May 2008. For both irradiations more than hundred containers containing pure actinide and other element isotopes under different chemical forms, have to be characterized. Here, begun the technical challenge: how to recover, quantitatively and by selective dissolution less than 5 mg of isotope powder in a few millimeter sized containers by remote handling in hot cell facility. This paper presents the technical developments performed by several CEA laboratories to open the containers in hot cell and to recover quantitatively the powder before dissolution.

After the external target decontamination, three steps are performed:

- external metal cover opening
- internal metal cover opening
- powder recovery

Each step has required special device remote handling usable. The small size of the target imposed camera view to realize some steps.

The target opening and isotope powder dissolutions are realized in Cadarache Chicade Hot Cell Facility for other than actinides isotopes and in Marcoule Atalante Facility for actinide and iodine isotopes. The results obtained with non-irradiated target had shown the feasibility. The first results with irradiated target will be obtained in 2009.

1. Introduction

PROFIL-R covers a series of fundamental experiments to obtain basic nuclear data, typically cross-sections of specific isotopes in a fast neutron reactor spectrum for research on the transmutation of long-lived radionuclides [1, 2, 3]. The reaction rates are measured by the determination of the isotopic composition of pure isotope powders before and after irradiation, and thus require high-precision isotopic analysis. The experimental fluence is also determined by analysis of “fluence monitors” consisting of standard fuel pin segments or samples of well known separated isotopes, typically ^{235}U .

This article discusses the main developments in the high-activity laboratory for optimizing the opening of the container and the dissolution of isotope powders while minimizing the impact of the container structural materials. The following topics are addressed: sample containers and experimental pins, equipment developed for the separation of actinide powder specimens, and dissolution procedures for different types of deposits.

2. PROFIL-R pins and containers

PROFIL-R consists of two pins of identical geometry each comprising a stack of sample containers, inserted in a standard fuel assembly (figure 1) loaded in the 4th fissile ring of the Phenix reactor core occupying a neutronically clean zone without control rods. Axially distributed ^{235}U containers are used as fluence monitors for each pin. A standard fuel pin in the central assembly position is characterized to measure the fluence data.

The selected heavy nuclides and fission product cover all the applications for which more accurate cross-sections are necessary — not only the use of fast neutron reactors for minor actinide transmutation, but also for the dedicated reactor concept (thorium cycle isotopes). The heavy isotopes to be processed in the Atalante complex¹ are listed in table 1. Other isotopes, especially fission or activation products (Rb, Zr, Mo, Tc, Ru, Rh, Pd, Ag, I, Cs, Ce, Pr, Nd, Sm, Eu and Gd) will be processed in CHICADE² using the same techniques (table 2).

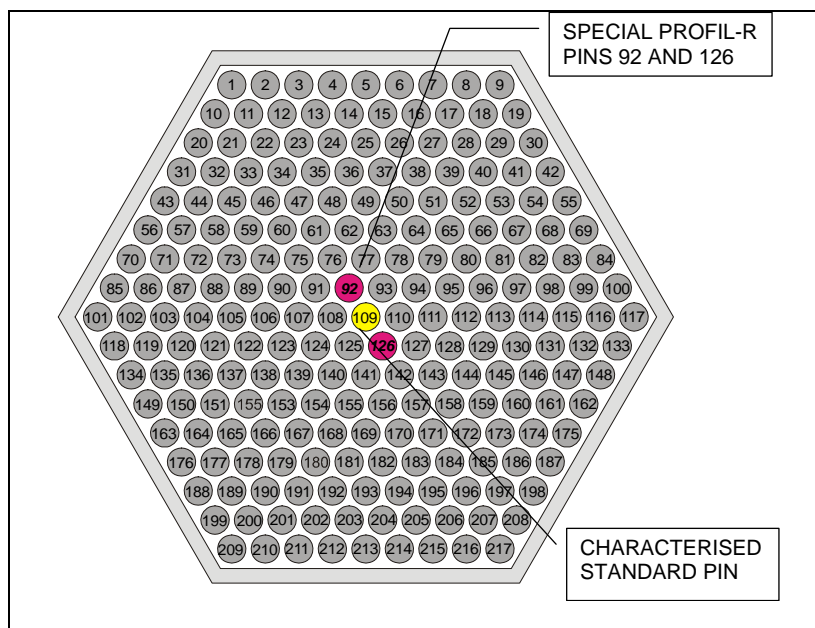


Fig. 1. Schematic layout of PROFIL-R assembly

Nature of deposit	Chemical compound	Isotope deposit mass per container (mg)	Isotope at%
²³² Th	ThO ₂	3.0	100.000
²³³ U	UO ₂	2.0	96.014
²³⁴ U	UO ₂	3.0	99.919
²³⁵ U	UO ₂	2.0	97.649
²³⁸ U	U metal	3.0	99.964
²³⁷ Np	NpO ₂	3.0	100.000
²³⁸ Pu	PuO ₂	2.0	94.942
²³⁹ Pu	PuO ₂	2.0	97.762
²⁴⁰ Pu	PuO ₂	3.0	99.841
²⁴² Pu	PuO ₂	3.0	99.125
²⁴¹ Am– ²⁴³ Am	AmO ₂ (Trace CmO ₂)	1.3 (²⁴¹ Am mass)	²⁴¹ Am = 49.829 ²⁴³ Am = 49.551

Table 1 . Nature and mass of actinide deposits.

Nature of deposit	Mass per container (mg)	Nature of deposit	Mass per container (mg)	Nature of deposit	Mass per container (mg)	Nature of deposit	Mass per container (mg)
pins 92				pins 126			
²³⁵ U	1.97	⁹⁹ Tc	4.98	²³⁵ U	1.97	⁹² Zr	4.829
¹⁰⁵ Pd	4.97	²³⁹ Pu	2	¹⁰⁹ Ag	4.95	⁹⁹ Tc	4.92
¹⁰⁴ Pd	4.93	¹⁰⁰ Mo	4.99	¹⁰⁷ Ag	4.99	⁹⁹ Tc	4.49
¹⁰³ Rh	4.98	²³⁸ Pu	1.99	⁹⁵ Mo	4.99	¹³³ Cs	2.99
B _{nat}	2.49	¹⁴⁰ Ce	4.99	⁹⁷ Mo	4.97	²³⁸ Pu	1.95
¹³³ Cs	2.97	²³⁷ Np	2.98	¹⁰⁰ Mo	4.99	¹⁴⁰ Ce	4.98
⁸⁷ Rb	2.87	²³⁷ Np	2.96	¹²⁹ I	2.97	¹⁴⁰ Ce	4.98
¹²⁹ I	2.97	¹⁴¹ Pr	4.99	¹⁰¹ Ru	4.99	²³⁷ Np	2.98
²⁴² Pu	2.95	²³² Th	2.99	²³⁸ U	3	¹⁴¹ Pr	4.98
¹⁰¹ Ru	4.88	²³² Th	2.99	¹⁰² Ru	4.97	²³⁵ U	1.97
²³⁸ Pu	1.95	¹⁴⁶ Nd	2.97	²³⁹ Pu	1.99	¹⁴⁶ Nd	3
¹⁰² Ru	4.9	²³⁴ U	2.93	¹⁰⁴ Ru	4.97	²³⁴ U	2.94
²⁴¹ Am	1.3	¹⁵⁷ Gd	2.95	²⁴⁰ Pu	2.99	¹⁴⁵ Nd	2.98
¹⁰⁴ Ru	4.96	²³³ U	1.96	¹⁰³ Rh	4.98	²³³ U	1.98
²³⁸ U	3	¹⁵⁶ Gd	2.96	¹²⁹ I	2.97	¹⁴³ Nd	2.97
²³⁸ U	2.89	²³⁵ U	1.88	¹⁰⁴ Pd	4.95	²³² Th	2.99
⁹⁵ Mo	4.9	⁹⁷ Mo	4.99	²⁴¹ Am	1.3	B _{nat}	2.51
²³⁵ U	1.97	¹⁴⁷ Sm	4.883	¹⁰⁵ Pd	4.97	¹⁴⁷ Sm	4.95
¹⁰⁷ Ag	4.814	¹⁴⁹ Sm	4.93	²³⁵ U	1.97	¹⁴⁹ Sm	4.96
²⁴¹ Am	1.3	¹⁵⁰ Sm	4.97	⁸⁷ Rb	2.88	¹⁵⁰ Sm	4.92
¹⁰⁹ Ag	4.95	¹⁵² Sm	4.95	⁸⁷ Rb	2.95	¹⁵² Sm	4.88
²⁴² Pu	3	¹⁵³ Eu	4.83	²⁴² Pu	2.96	¹⁵³ Eu	4.98
¹⁰⁶ Pd	4.969	¹⁵⁴ Gd	2.95	¹⁰⁶ Pd	4.99	¹⁵⁴ Gd	2.98
²⁴⁰ Pu	2.97	¹⁵⁵ Gd	3	¹⁰⁰ Mo	4.99	¹⁵⁵ Gd	2.95
⁹¹ Zr	4.98	¹⁴³ Nd	2.99	⁹¹ Zr	4.99	¹⁵⁶ Gd	2.96
²⁴⁰ Pu	2.97	¹⁴⁵ Nd	3	⁹¹ Zr	4.97	¹⁵⁷ Gd	3.02
⁹² Zr	4.754	²³⁵ U	1.97	²³⁷ Np	3	²³⁵ U	1.97
²³⁵ U	1.97			⁹² Zr	4.933		

Table 2 . Mass of powder deposits,.

Several mg of the isotope powder are conditioned in a 316L stainless steel inner container sealed by a TIG-welded cover and inserted in a 316L stainless steel outer container also sealed by a welded cover. The inner and outer container dimensions are indicated in Figures 2 and 3. The estimated steel mass is 150 mg for the inner container and 800 mg for the outer container. The presence of significant quantities of analytical interferences such as molybdenum in the structural steel led us to include a powder separation step prior to selective dissolution.

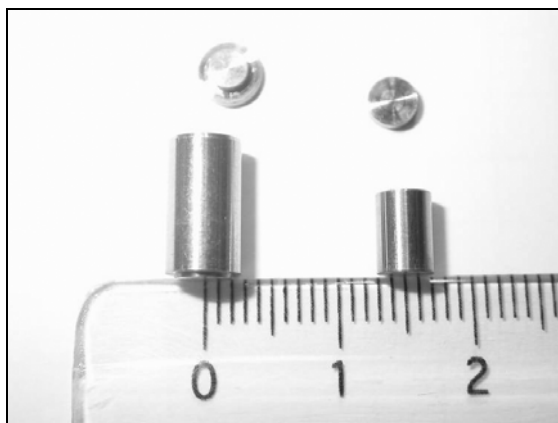


Fig. 2. Outer and inner containers

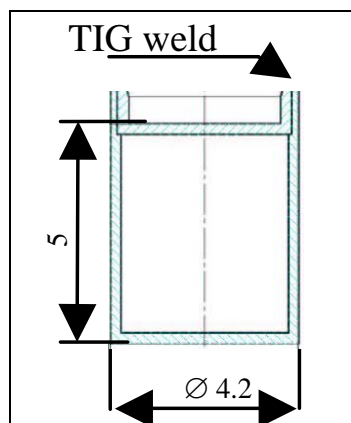


Fig. 3. Inner container

3. Powder recovery operations

Several steps are performed to recover and dissolve as much of the powder as possible after irradiation and to avoid pollution by any radioelements or natural element that could be present in the hot cell. The main difficulties arise from the size of the telemanipulated container and the very small quantity of powder recovered.

3.1 Identification

Each container is first identified (figure 4) using a video camera to read the number engraved on the outer container, then placed on a gripper to simplify subsequent handling operations. The container is decontaminated by ultrasound in 1 M nitric acid; after drying, it is weighed to within 0.1 mg.

3.2 Opening the Containers

The outer container is opened in an acidic medium by electrochemical machining of the TIG weld bead to release the cover. Figure 5 shows the electrolytic solution (approx. 1.5 M HNO_3) in an open beaker containing a stainless steel electrode connected to the negative pole of a generator, with the sample connected to the positive pole. The immersion depth is precisely adjusted with a micrometer screw to erode only the weld bead; the cover detaches, releasing the inner container which drops to the bottom of the cathode basket, and the electrical contact is broken by the loss of contact with the liquid.

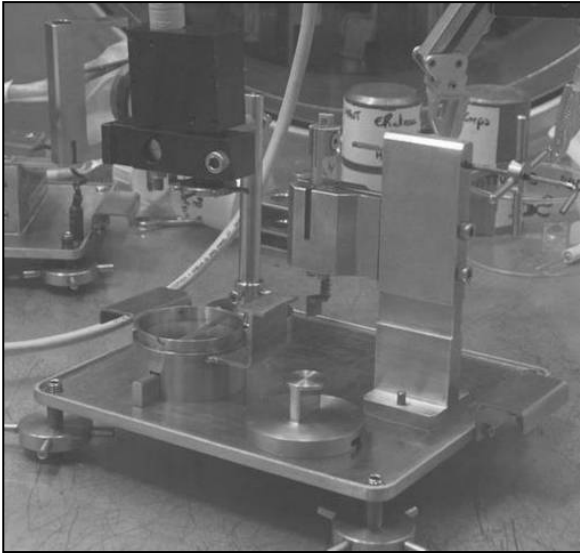


Fig 4. Identification machining unit

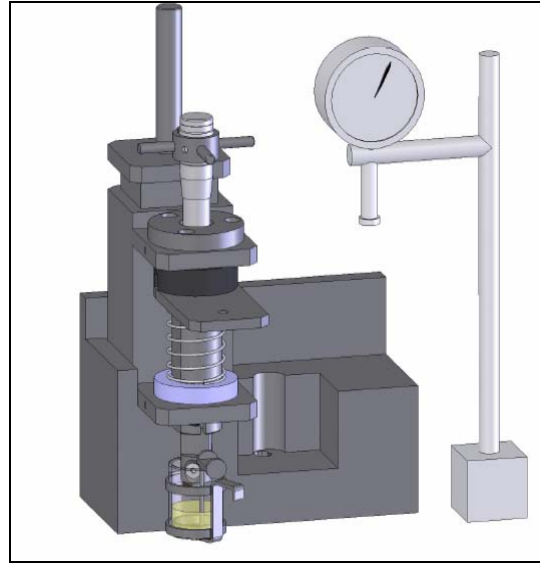


Fig 5. Acid electrochemical machining unit

After ultrasonic decontamination in nitric acid the inner container is also opened by electrochemical machining; this technique ensures that the inner container is opened without any mechanical stress loading that could distort it and hinder the recovery of its contents. Electroerosion is controlled by the duration and immersion depth to erode only the weld bead. The cover is secured mechanically to seal the inner container. After electroerosion and drying, a dowel is bonded to the inner container cover with heat-sensitive adhesive to serve as a grip during the powder recovery step.

The first container opened by electroerosion showed that a part of electrolytic solution was introduced in the container during the process in spite of the cover. Then soluble element contained in the powder was lost in the electrolytic solution. A new device has been designed to avoid this loss. The retained dry mechanical treatment (figure 5, 6) allows to abrade the weld bead by a mill. Then the cover can be easily removed, without using a bonded dowel as with the electroerosion technique, that simplifies the procedure.

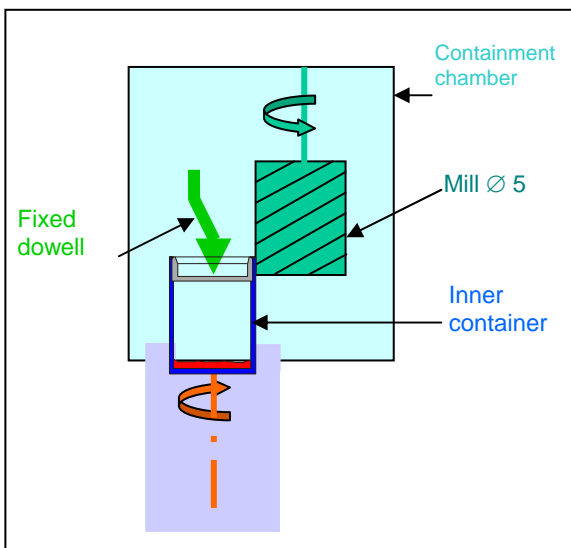


Fig 5. Mechanical erosion system

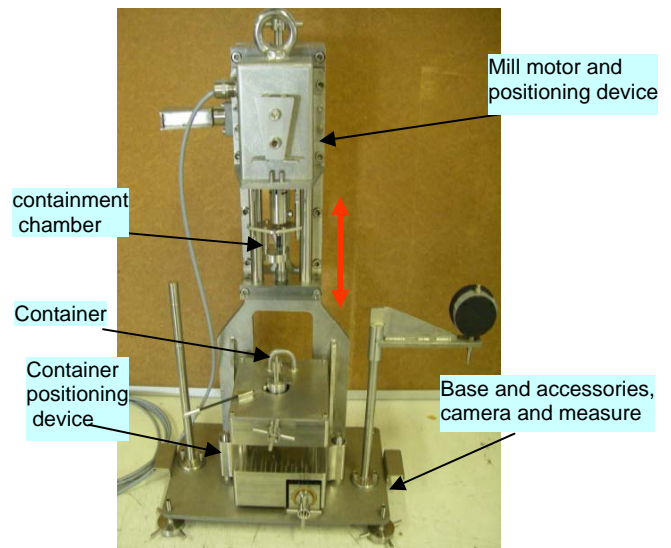


Fig 6. Mechanical erosion prototype

3.3 Powder Recovery

A special tool is used to recover the maximum quantity of isotope powder from each capsule while limiting the risk of material dispersion and external contamination.

The inner container mounted on its handling tool is fitted on the extraction device (Figure 7). The dowel bonded to the cover is gripped by a fork, and the inner container is opened in confined atmosphere under video surveillance. A scraper (Figure 7) mounted in the dissolution vial is inserted into the inner container and mechanically transfers the powder into the vial. The vial is then closed and disposed on the dissolution unit.

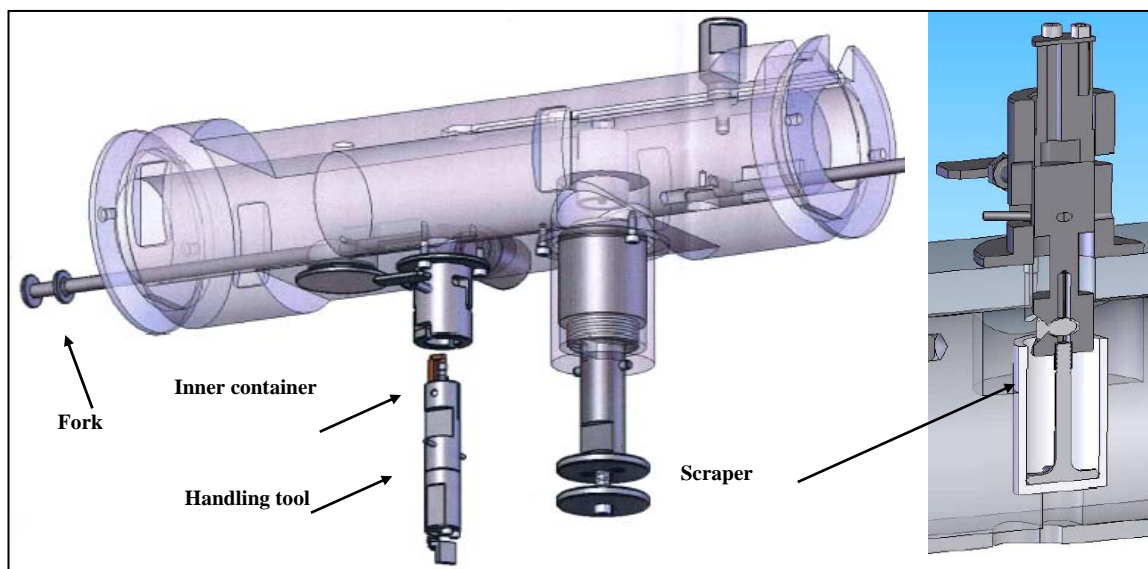


Fig 7. Powder recovery

3.4 Powder dissolution

After removal from the container the actinide and fission product powder is dissolved in the PTFE vial. The conditions of dissolution were specified and validated by previous tests performed on the non-irradiated powders. The resulting solutions do not contain suspended matter. Mass balance assays in the vial before and after dissolution show that no evaporation occurs during the process. The dissolution medium (Table 3) depends on the chemical element and its form (oxide or metal). Dissolution is performed at the boiling point with the closed vial placed in an alumina bath to homogenize the temperature and prevent hot spots. The samples are then transferred to the Laboratory of Nuclear Isotopic and Elemental Analysis in Saclay.

Nature of deposit	Chemical compound	Dissolution medium	Dissolution conditions
²³² Th	ThO ₂	HNO ₃ /HF – 11 M/0.1 M	10 h at boiling temp.
^{233,234,235} U	UO ₂	HNO ₃ – 4 M	1 h at room temp. + 1 h at boiling
²³⁸ U	U metal	HNO ₃ – 8 M	2 h at boiling temp.
²³⁷ Np	NpO ₂	HNO ₃ /HF – 11 M/0.1 M	10 h at boiling temp.
^{238,239,240,242} Pu	PuO ₂	HNO ₃ /HF – 11 M/0.1 M	10 h at boiling temp.
²⁴¹ Am– ²⁴³ Am	AmO ₂	HNO ₃ – 4 M	1 h at room temp. + 1 h at boiling
^{91,93} Zr	ZrO ₂	HNO ₃ /HF – 12 M/3 M	10 h at boiling temp
^{107,109} Ag	Ag metal	HNO ₃ – 14 M	24 h at boiling temp
^{101,102,104} Ru	metal	HCl – 10 M + KClO ₃	boiling temp

Table 3. Dissolution medium

4. Operating procedure

After two pins were fabricated and loaded in the reactor, an initial key step was completed in September 2005 when the irradiation campaign ended in the Phenix reactor at Marcoule. At the end of 2006 the pins were sent to the LECA-STAR facility at Cadarache for retrieval and sorting of the test containers. The repackaged containers were then transferred to CHICADE (fission products) and ATALANTE (actinides) where they were opened and the powder samples were dissolved. The time necessary for organizing and carrying out the transports was used to develop the container opening techniques and to build and test the equipment on unirradiated containers, to specify the dissolution protocols, and to develop analytical techniques for separation of the chemical elements and for isotopic analysis. The composition inventories for various types of deposits after irradiation were estimated using the DARWIN formulary for the maximum irradiation flux and after cooling times ranging from 1 to 10 years.

The key dates for this experiment are the following:

- Beginning of irradiation: 24 June 2003
- Removal from reactor: 4 September 2005
- Pin dismantling: 2006
- Fabrication and installation of the device in a shielded cell in Atalante (Figures 8 and 9)
- First dissolution of an actinide target: end of 2008
- Fabrication of the mechanical device: June 2009

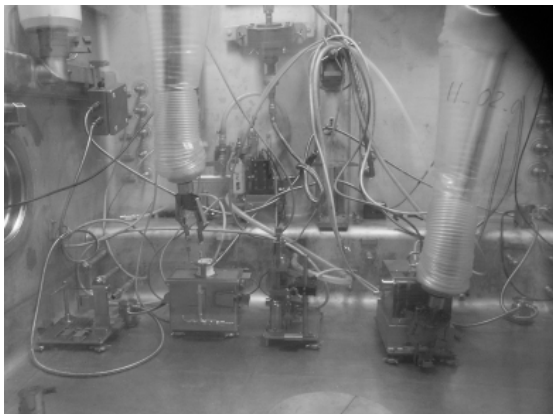


Fig 8. Target selection, weighing, external ultrasound cleaning apparatus, electroerosion opening system

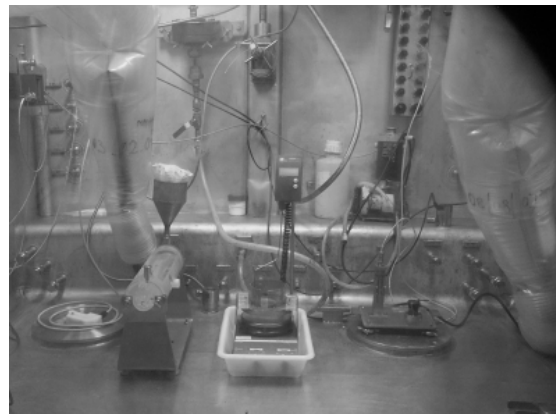


Figure 9. Powder recovery and dissolution

Potential device improvements were identified after the first irradiated targets were opened, but the dissolution solutions obtained were suitable for analysis. The separation and analytical techniques have been tested on actual solutions with promising results.

This ambitious research program involves technological and analytic challenges for several CEA laboratories. It has now entered the final phase and is expected to provide major results concerning the understanding of fast neutron reactors.

5. References

- [1] Phenix: the irradiation program for transmutation experiments”, J. Guidez et al., Eighth Information Exchange Meeting on Partitioning & Transmutation, Las Vegas, Nevada, USA, 9-11 November 2004
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