

ENSAR – 2nd period (31st August 2013)

Scientific report ActILab – JRA02 WP8 – Coordination: T. Stora – CERN)

No milestones or deliverables were defined for ActILab in this second reporting period. However already a number of Milestones have been anticipated and the related activities are reported in the following part.

TASK #1 (COORDINATION C. LAU – IPNO): SYNTHESIS OF NEW ACTINIDE TARGETS

M-JRA02-1.2 synthesis of nanostructured actinide targets (achieved)

At CERN, after an upgrade of the "class A" target laboratory, a systematic study has been carried out to synthesize an optimized UC_X material using CNT (Carbon NanoTubes). This study led by A. Gottberg has been achieved with the contribution of S. Tusseau-Nenez and N. Barré-Boscher from IPNO. In particular the best grinding parameters have been identified: the speed of the planetary mill, the suitable liquid with low surface tension, powder-WC ball ratio, the grinding time and the grinding cycle. The grain size of the starting standard uranium dioxide powder could be reduced more than two orders of magnitude, down to 160 nm (cf. Fig. 1).

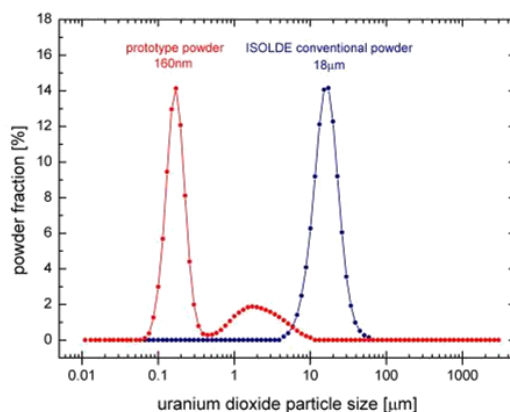


Figure 1: The reduction of the uranium-dioxide grain size obtained by the milling process developed. The result is compared to the conventional powder used at ISOLDE-CERN.

Such a powder has been combined with CNT and properly sintered to produce a novel actinide material having an efficient nanostructure. Based on BET, density evolution and particle size measurements, various physicochemical characterizations were achieved to define the final target prototype that was tested on-line at ISOLDE-CERN end of 2012.

When the target laboratory at IPNO became operational again, various UC_X prototypes were manufactured by using promising synthesis method. Different UC_X samples have been obtained from uranium oxide or uranium oxalate, combined with different type of graphite

including carbon microfibers and CNT. These samples and the samples sent from CERN have been characterized by Scanning Electron Microscope (SEM), X-Ray Diffraction (XRD), He pycnometry and Hg porosimetry. The different measurements achieved have been correlated to get a deeper understanding of the material properties.

M-JRA02-1.1 synthesis of actinide targets by sol-gel method - ‘chimie douce’ (achieved)

At INFN-LNL, S. Corradetti has taken the post-doc position in September 2012. Another grinding method has been investigated to grind uranium dioxide. For this purpose, the target laboratory has been equipped with a new micromill and sieves. A systematic study has also been performed. Repeating grinding and sieving processes the grain size distribution has been optimized. A distribution among the smallest grain size could be obtained. Then sintering has also been studied to obtain UC_X from green pellets made out of UO_2 and graphite. The thermal treatment has been studied as a function of the rising temperature steps. The outgassing during the reaction sintering has studied (cf. Fig. 2).

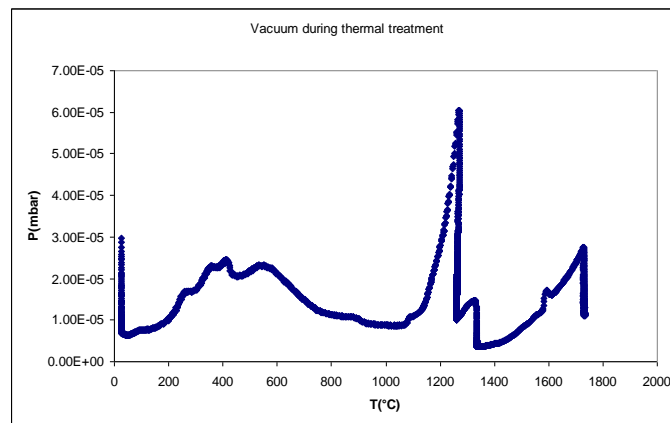


Figure 2: outgassing as a function of the heating temperature. The strong outgassing between 200 and 600 °C is due to H₂O and the decomposition of the binder. The strong outgassing peaks observed from 1 200 °C show the reaction sintering with the release of CO.

In 2013, UC_X prototype samples have been synthesized and sent to IPNO for physicochemical characterization. Some UO_2 powder obtained by the new grinding and sieving processes has also been sent. The samples have arrived at IPNO end of September 2013.

In parallel at IPNO, the target laboratory has been extended in April 2012 but the installation of the fluid circuits has been completed only by February 2013. Between April 2012 and February 2013, no radioactive material could be handled in the target laboratory. However a systematic study has been achieved using Lanthanum as a substitute of uranium. In this study, grinding as a function of the ball diameter and pressing the pellet as a function of the pressure have been investigated. In November 2012, UC and UO_2 +CNT samples from CERN have been successfully transferred to IPNO for further characterization.

TASK #2 (COORDINATION A. ANDRIGHETTO – INFN): CHARACTERIZATION OF NEW ACTINIDE TARGETS

In the framework of the ActiLab activity, several types of advanced materials to be used as ISOL (Isotope Separation On-Line) targets have been produced, characterized and tested, in order to find significant correlation between their microstructure and their capacity of releasing radioactive isotopes. The characterization has been focused on different aspects, which are described in the following paragraphs.

M-JRA02-2.1 Characterization of structures (achieved)

Each of the materials which was taken into consideration during the ActiLab activity was characterized with different techniques, in order to study in detail its composition and microstructure. The activity was conducted at IPNO (Institut de Physique Nucleaire d'Orsay), and consisted of: a) XRD (X-Ray diffraction) to characterize the composition of the synthesized materials, b) SEM (Scanning Electron Microscopy) to visually inspect the morphological characteristics of the microstructure of each material, c) Helium picnometry, to obtain information about the true density of the produced samples, and therefore to characterized into detail the type and amount of porosity generated during the synthesis process, d) Mercury porosimetry, to investigate the pore size distribution relative to different materials and synthesis.

The results of the aforementioned analysis techniques are very significant, since they clearly highlight the analogies and differences between the synthesized materials. In terms of composition (figure 3), different phases were found, such as UC, UC₂ and diverse types of carbon structures, depending on the stoichiometry chosen for the “oxide+carbon” reaction. The use of SEM (figure 4) allowed to have a visual evidence on the influence of the produced phases on the material microstructure. The combined use of picnometry and porosimetry gave important information about the type of porosity created during the synthesis process as well as the pore size distribution (figure 5).

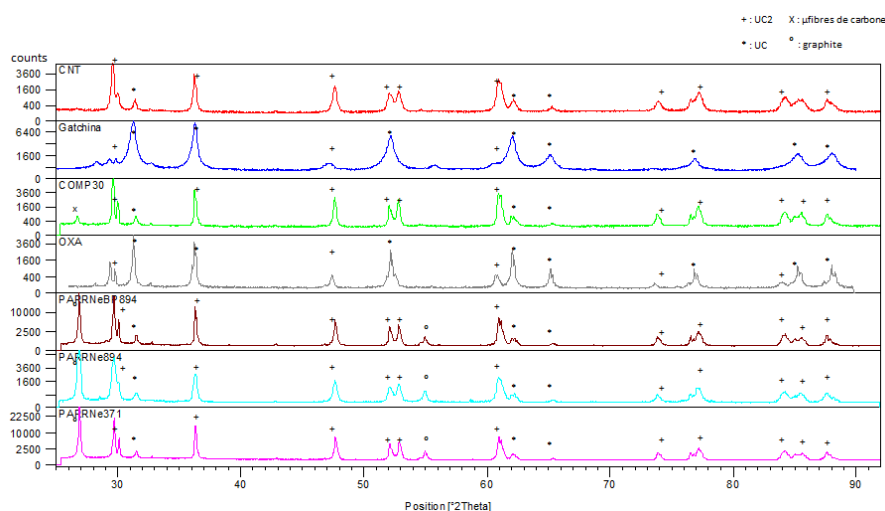


Figure 3. XRD patterns of different samples, highlighting the presence of UC, UC₂, graphite and carbon microfibres.

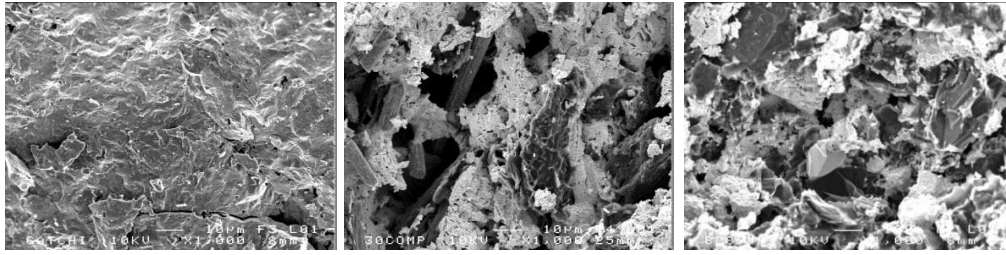


Figure 4. SEM images. From left to right: compact structure of high-density UC, open structure containing UC₂ grains and carbon fibers, open structure containing UC₂ grains and graphite residual clusters (black blocks).

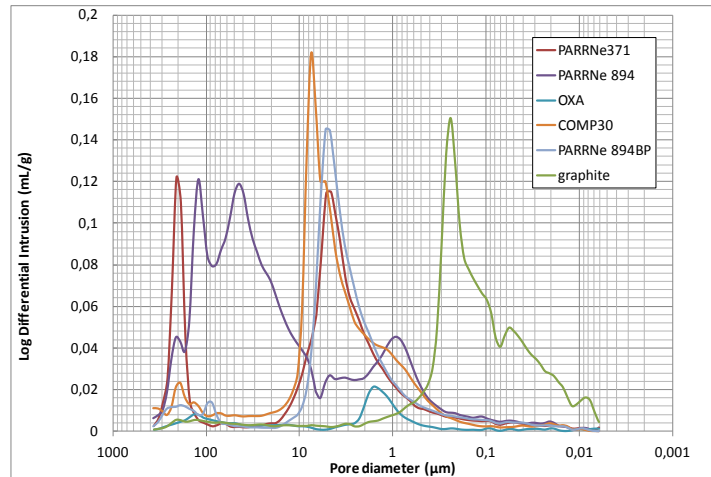


Figure 5. Pore size distribution of different samples using mercury porosimetry.

Based on systematic studies of LaO₂ and Carbon grinding as a function of the ball diameter and pressing the pellet as a function of the pressure, IPNO now focuses its efforts on the synthesis of nanostructured UC_x with the use of CNT (cf. Fig. 6).

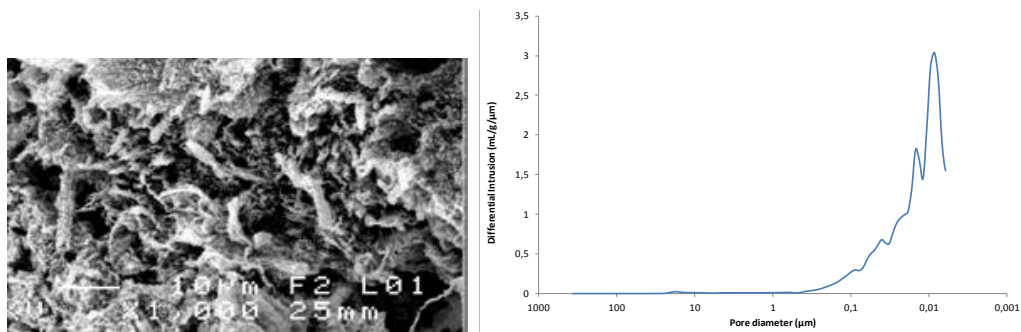


Figure 6: SEM picture of a UC_x synthesized with CNT correlated to a spectrum obtained by Hg porosimetry. These data show the porous structure of CNT and UC₂ (white color) growing on CNT.

The most characteristic samples used during the experiment at ALTO are being sent to INFN for thermal emissivity measurement. The safety procedure for shipment has been achieved.

M-JRA02-2.2 Characterization of thermal properties (in progress):



Recently, an experimental device for the evaluation of thermal emissivity by means of a dual frequency pyrometer has been developed at INFN-LNL (Istituto Nazionale di Fisica Nucleare – Laboratori Nazionali di Legnaro). The device is based on the use of a high vacuum furnace inside which a graphite crucible is heated by Joule effect (figure 7) and on top of which the test sample is directly placed. The system is now ready to perform emissivity measurements; it will be also used, with some modifications, to characterize the thermal conductivity of the newly synthesized uranium carbides, making use of inverse analysis calculations coupled to experimental thermal measurements.

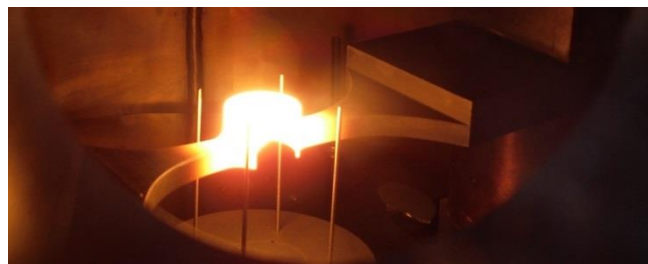


Figure 7. Graphite crucible to perform emissivity tests.

TASK #3 (COORDINATION M. MARTIN – PSI): ACTINIDE TARGETS PROPERTIES AFTER IRRADIATION

M-JRA02-3.1 Characterization of irradiated materials in hot cell (in progress):

Dismantling of the ISOLDE target container

The ISOLDE Target transportation was realized in January 2013. With respect to the dismantling, the target has been stored temporarily in PSI's hot cells.

On July 15th the ISOLDE container was opened and the target was separated from the container. In order to avoid oxygen diffusion into the target, after cutting the transfer line this tube has been closed, with glue. Figure 8 shows the opened container and the ISOLDE target with the closed transfer line (red mark).

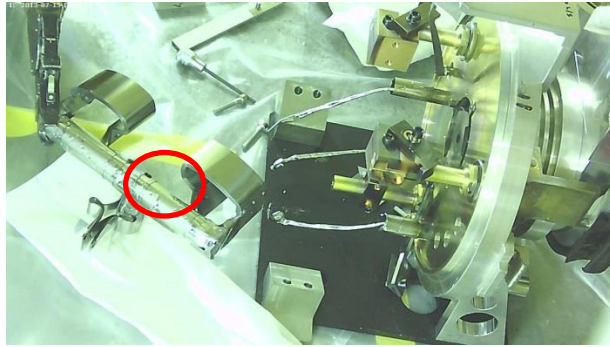


Figure 8: ISOLDE target with the closed transfer line (red mark) and the opened container

Opening of the target

Since the target handling became easier and Nitrogen flooding necessary, it was decided to use the EPMA preparation glove box.

At first a test cutting (Figure 9) has been performed on the unirradiated dummy target using a “Dremel”.



Figure 9: Dummy target and test cut on the target

The Dummy and the ISOLDE target have been cut, at the side where the carbon plug is located inside, as illustrated on Figure 10.

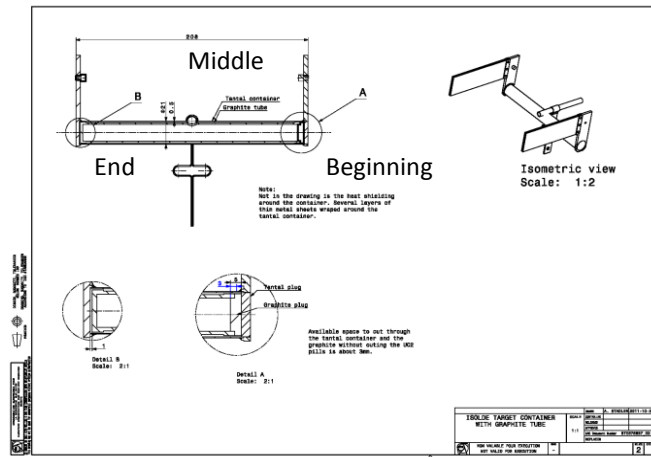


Figure 10: Target container cutting on side A

Pellet sample preparation

Contrary to the initial expectations, the carbon tube insert was stuck inside the tantalum tube and did not move out. For this reason the carbon plug had been removed. Subsequent to the cutting, the carbon plug was removed and the pellets were taken out, successively. Figure 11 shows the pellets partly sticking to each other, in groups of typically 3 to 7 pellets.

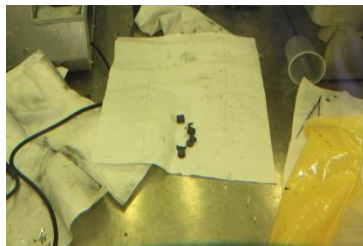


Figure 11: Pellet groups removed from the container (left) and one selected pellet group (right).

In order to separate the pellets individually from the group, a knife has been used. In table 1 the prepared specimens are listed, starting from the side of the carbon plug.

Table 1: specimen list of the different target areas counting from carbon plug, see Figure 3

Specimen name	Used technique	Notes
EPMA/Cer_B	EPMA	
XAS_BC	microXAS	Preparation of a FIB sample from this piece, center area
XAS_BR	microXAS	Preparation of a FIB sample from this piece, rim area
EPMA/Cer_M	EPMA	
XAS_MC	microXAS	Preparation of a FIB sample from this piece, center area
XAS_MR	microXAS	Preparation of a FIB sample

		from this piece, rim area
EPMA/Cer_E	EPMA	
XAS_EC	microXAS	Preparation of a FIB sample from this piece, center area
XAS_ER	microXAS	Preparation of a FIB sample from this piece, rim area

Characterization of irradiated materials

Unirradiated materials (a high density UC and the standard polyphasic ISOLDE UC_x were studied at the Swiss Light Source (SLS) of PSI (Paul Scherrer Institut), in particular with the micro spot fluorescence mapping, which allowed to perform elemental analysis on a small portion of the specimen. This technique allows to obtain elemental “maps” superimposed to a SEM image, as shown on figure 12. A scientific publication is ready for submission. Analysis of irradiated ISOLDE UC_x targets have started with the preparation of the samples to be investigated at the Swiss Light Source at PSI in October 2013.

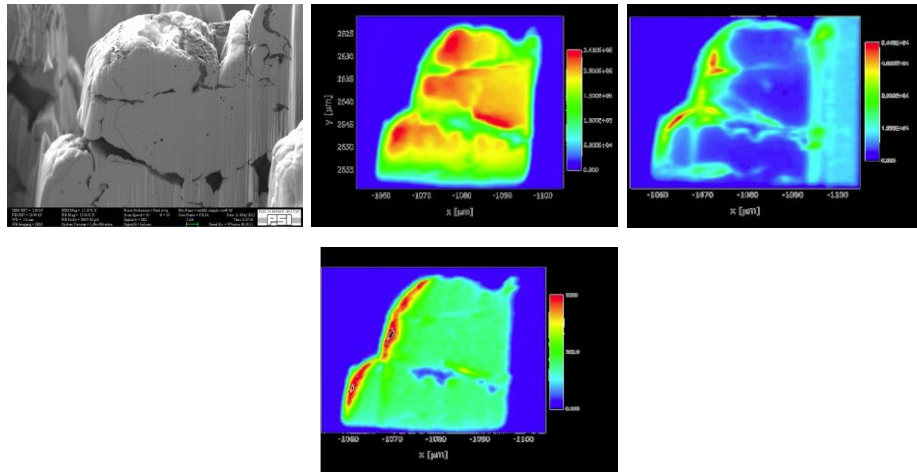


Figure 12. From left to right: SEM image of standard UC_x target, and X-ray fluorescence emission mapping of uranium, gallium and cesium concentrations seen with false colours.

TASK #4 (COORDINATION H. FRANBERG-DELAHAYE – GANIL): ONLINE TESTS OF ACTINIDE TARGETS

M-JRA02-4.1 Analysis of online tests of new actinide targets (achieved):

An experiment has been developed at IPNO to characterize the release kinetics of long-lived isotopes for the different UC_x synthesized target materials. The results obtained enable to correlate the physicochemical properties of the prototype UC_x pellets to the release kinetics of various isotopes of interest. The results are the object of an article in preparation.

Online tests of the material using small grained UO₂ powder together with Carbon Nanotubes prepared took place at CERN-ISOLDE in December 2012 with the target unit UC498-Re under the approved IS540 experiment. Preliminary yield estimates indicate a

higher isotope production rate are obtained for ^{11}Be , n-rich Cs and exotic ^{30}Na beams. Data are compared with those obtained with other types of UCx targets and shown on figure 13.

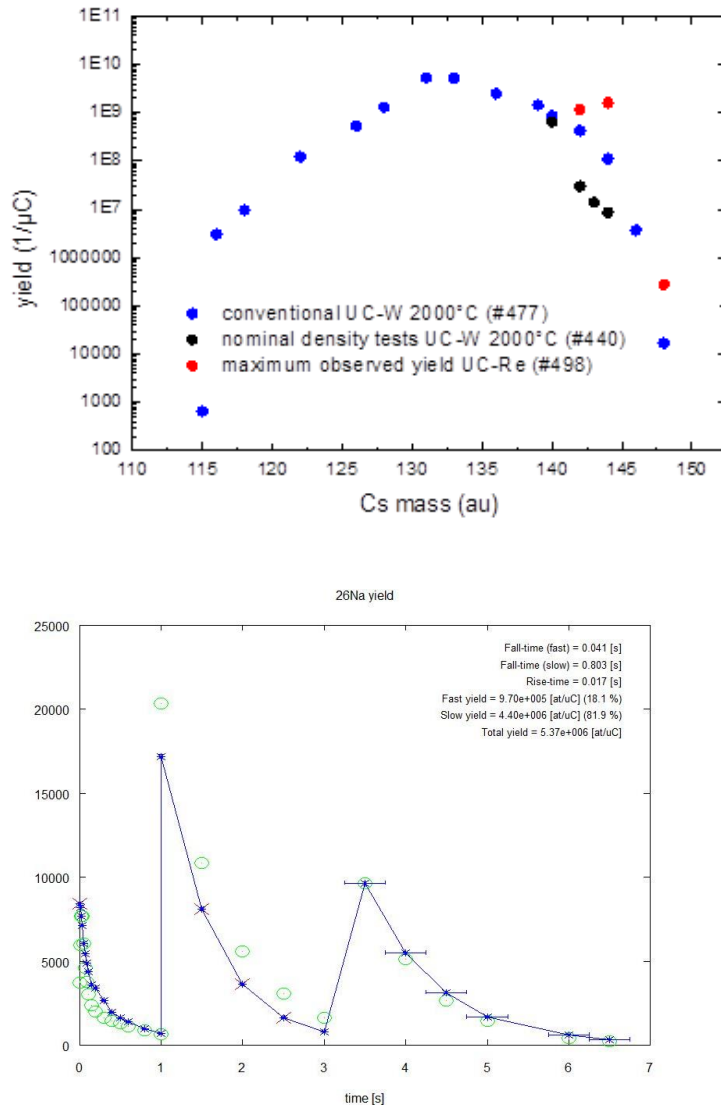


Figure 13. Upper figure: Comparison of yields of Cs beams at CERN-ISOLDE from a conventional UCx target a high density UC target and the target prepared with nano-powders for ActILab. Lower figure: Release properties of ^{26}Na from the UC498 target unit at 2000C.