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 two different aspects, which are described in the following paragraphs.

1. Characterization of structures

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 1), different phases were found, such as UC, UC_2 and diverse types of carbon structures, depending on the stoichiometry chosen for the "oxide+carbon" reaction. The use of SEM (figure 2) 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 3).
Based on systematic studies of lanthanum oxide 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 carbon nanotubes (CNTs) (cf. Fig. 4).
The most characteristic samples used during the experiment at ALTO were sent to INFN for thermal emissivity measurement (see Section 2 of this report).

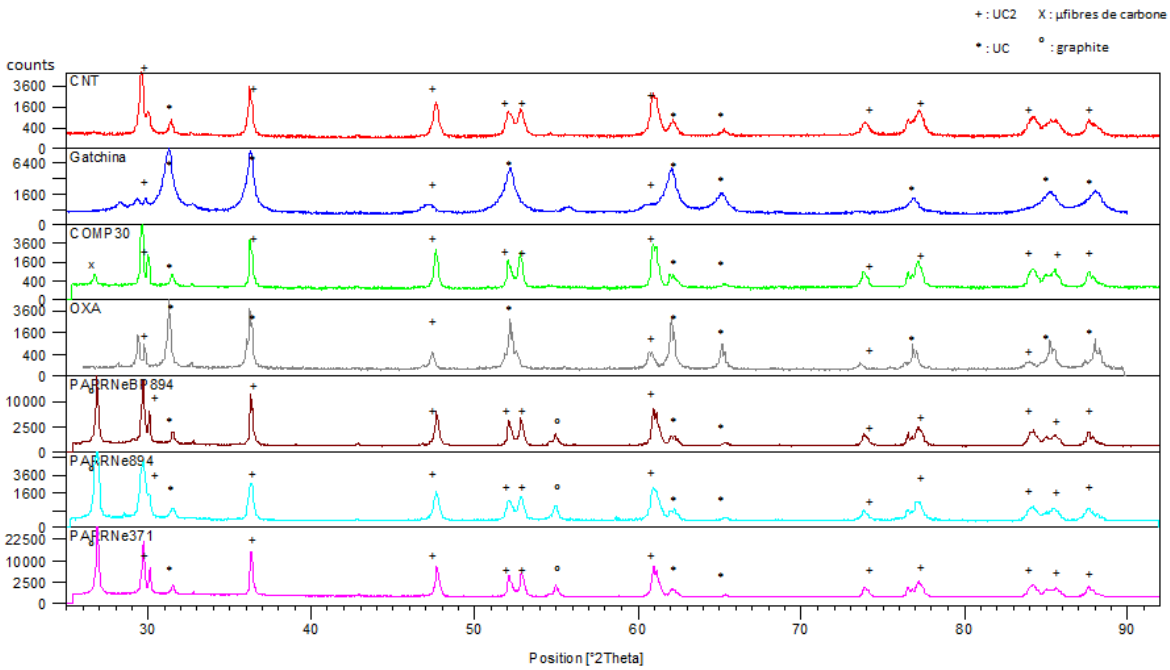

Figure 1. XRD patterns of different samples, highlighting the presence of UC, UC_2, graphite and carbon microfibres.

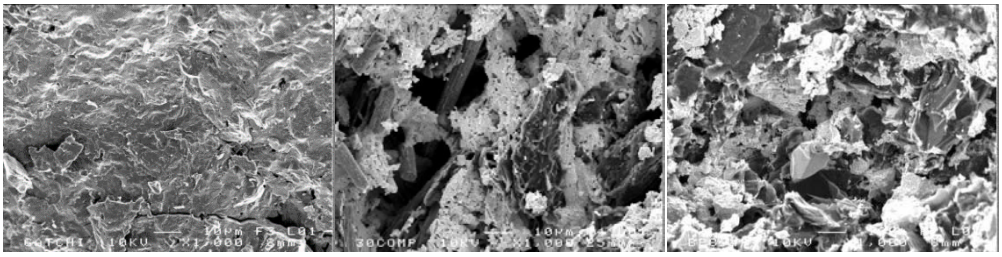

Figure 2. SEM images. From left to right: compact structure of high-density UC, open structure containing UC_2 grains and carbon fibers, open structure containing UC_2 grains and graphite residual clusters (black blocks).

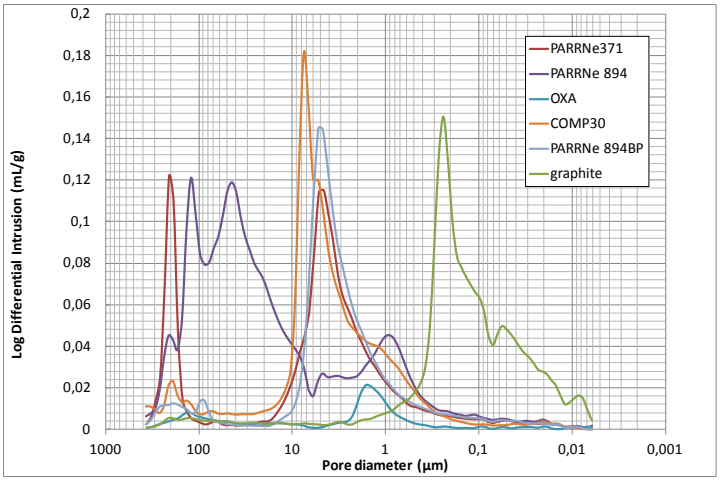

Figure 3. Pore size distribution of different samples using mercury porosimetry.

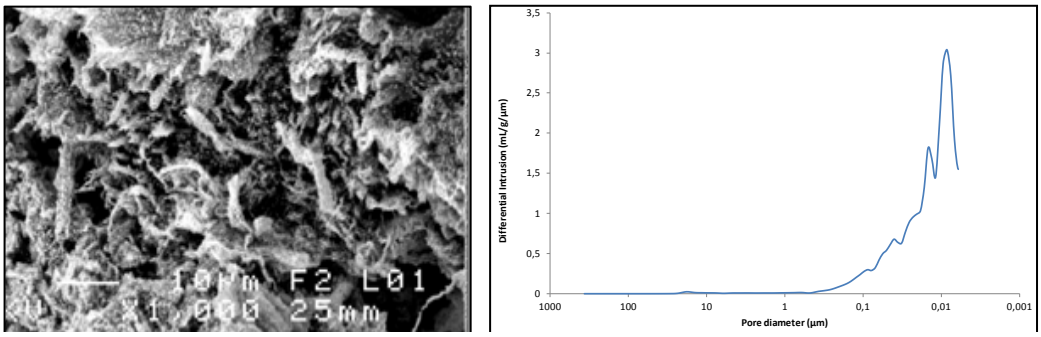

Figure 4: SEM picture of a UC_X synthesized with CNTs correlated to a spectrum obtained by Hg porosimetry. These data show the porous structure of CNTs and UC_2 (white color) growing on CNTs.

2. Characterization of thermal properties

Two types of measurements were carried out on different types of uranium carbides, relative to the determination of thermal conductivity and emissivity, respectively.

a) Thermal conductivity estimations were obtained with a method based on the one already reported by Manzolaro et al. [1], successfully applied for graphite, silicon carbide and lanthanum carbide SPES target prototypes at INFN-LNL. In order to make use of the method in the case of uranium carbide, a new setup was developed at Padova University in a dedicated actinide chemistry laboratory. The experimental technique is based on direct measurements of temperature and emissivity on a sample, under steady-state conditions, which are then converted to thermal conductivity data making use of the inverse analysis method.
From the experimental point of view, the method is based on the creation of a temperature gradient on the top surface of a thin disc of uranium carbide with diameter of about 30 mm, by irradiation of a hot graphite crucible placed at a certain distance from it, directly facing its bottom surface. The temperature gradient on the suspended disc is measurable by making use of a dual-frequency infrared pyrometer placed on the top of the vacuum chamber containing the setup. The data relative to two different analyzed positions (center and periphery) of the samples top surface are collected in separate heating cycles, with the same maximum current given to the crucible. Fig. 5 shows the experimental setup during operation. The inverse (optimization) problem is based on the minimization of the differences between the experimentally determined temperatures and those obtained numerically by a simulation carried out with ANSYS®, in two different positions of the heated sample. The objective of the optimization is the minimization of the residual function

$$J(\mathbf{f})=\sum_{i=1}^{N_{HC}} [T_{C\_COMP\_i}(\mathbf{f})-T_{C\_MEAS\_i}]^2+[T_{P\_COMP\_i}(\mathbf{f})-T_{P\_MEAS\_i}]^2 \quad (1)$$

where N_{HC} is the number of current steps used to power the heater and so to generate temperature gradients in the sample top surface, $T_{C\_COMP\_i}$ and $T_{P\_COMP\_i}$ are the numerically computed temperatures at the center and at the periphery of the sample disc, respectively, $T_{C\_MEAS\_i}$ and $T_{P\_MEAS\_i}$ are the correspondent measured values. **f** is the vector of the unknown coefficients:

$$\mathbf{f} = \{C_0, C_1\} \quad (2)$$

which characterize the linear dependency of the thermal conductivity on temperature:

$$k(T) = C_0 + C_1 * T \quad (3)$$

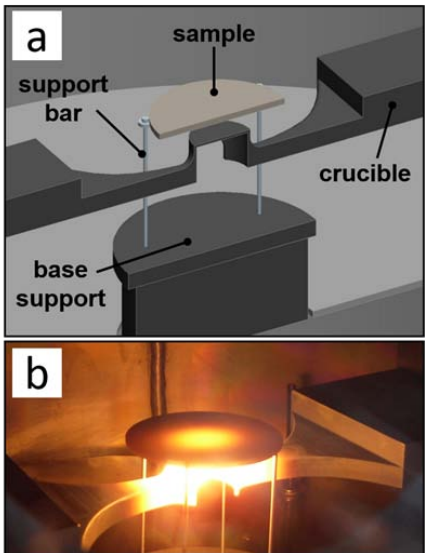

Figure 5: a) CAD view of the thermal conductivity estimation setup, b) sample heated by irradiation by the crucible, with the creation of a temperature gradient.

b) To carry out the emissivity direct measurements with a pyrometer, the samples to be tested were placed directly on top of the graphite crucible. This allowed reaching higher temperatures with respect to the thermal conductivity measurements case.

Table 1 shows the main geometrical and compositional properties of the samples. Only one of the samples (SPES MM), produced at Padova University, was used for thermal conductivity estimations. All the other samples, produced at IPNO, which possessed a diameter too small for these kind of tests, were used for the emissivity measurements.
In figs. 6 and 7, the comparison of the numerical and experimental temperatures and the thermal conductivity trend with respect to temperature are reported, respectively, for the sample SPES MM. Each reported temperature is the average value of five repeated measurements.

Table 1. Properties of the tested samples.

Sample	Production site	Reagents	Main phase	Diameter (mm)	Density (g/cm^3)
SPES MM	UNIPD	UO_2+graphite	UC_2	28.9	3.9
GATCHINA	PNPI	UC (provided by PNPI to ActiLab)	UC	13.2	12.9
ParrNe 894	IPNO	U_3O_8+graphite	UC_2	13.0	3.1
ParrNe BP	IPNO	U_3O_8+graphite	UC_2	12.6	4.4
OXA	IPNO	$U(C_2O_4)_2,2H_2O$ + graphite	UC	7.4	8.7
COMP30	IPNO	$U(C_2O_4)_2,2H_2O$ + graphite + carbon fibers	UC_2	8.3	4.5

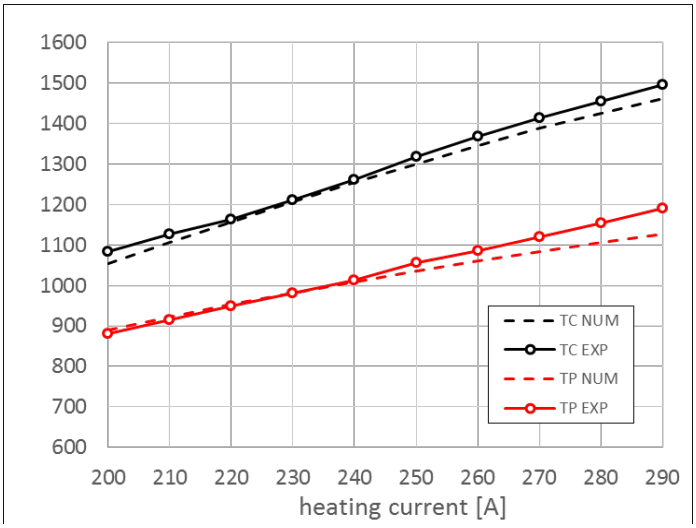

Figure 6. Experimental and numerical temperatures on center and periphery of a UC_X disc.

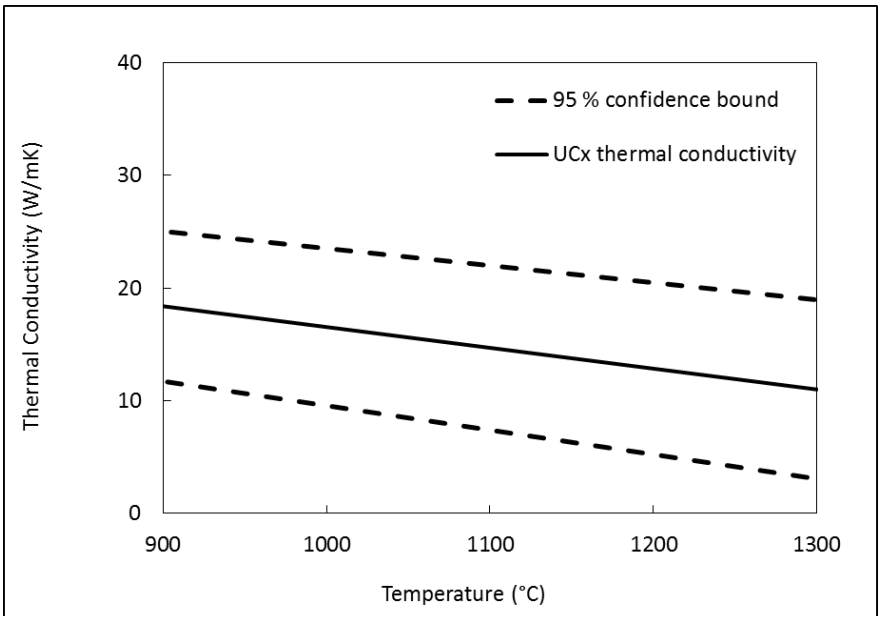

Figure 7. Thermal conductivity of UC_X.

Figure 8 shows the thermal emissivity trend with respect to temperature for the investigated materials. Different values and trends were found for different materials compositions. In particular, materials containing UC_2 as a main phase were found to have higher and more stable values of emissivity, whereas in UC-based ones a drop of emissivity at temperatures of about 1200 °C ÷ 1300 °C was observed, with any probability due to their reaction with the heating graphite crucible.

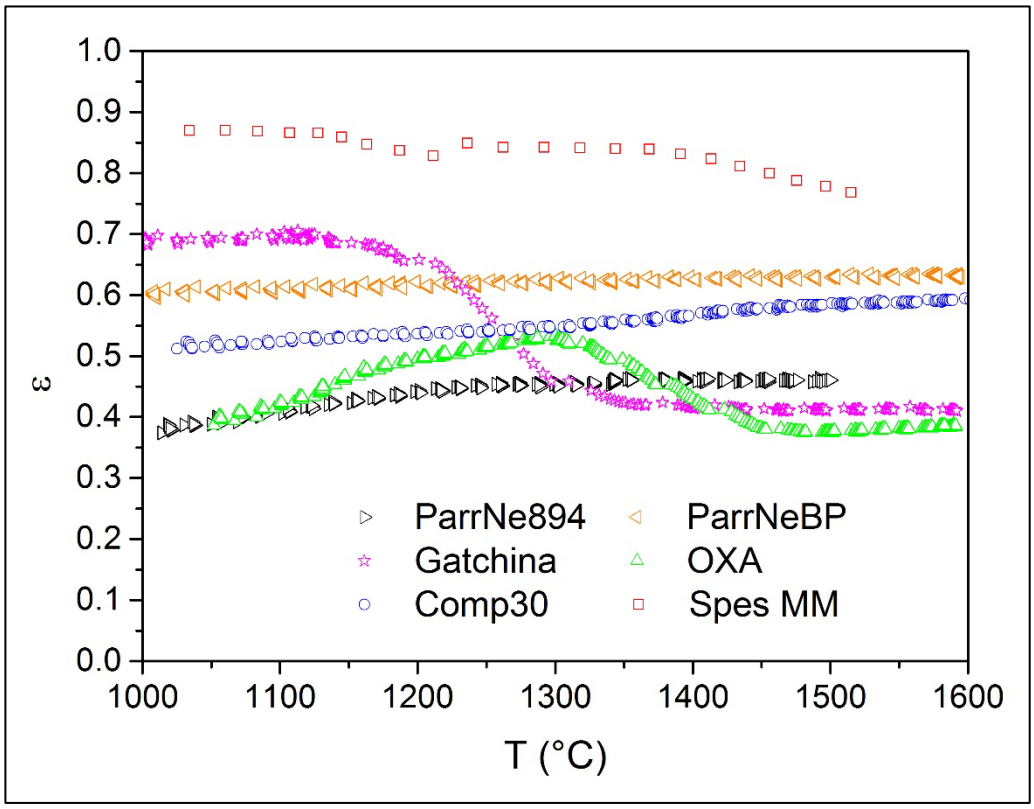

Figure 8. Thermal emissivity of different types of uranium carbide.

References

[1] M. Manzolaro, S. Corradetti, A. Andrighetto, L. Ferrari, A steady-state high temperature method for measuring thermal conductivity of refractory materials, Rev. Sci. Instrum. 84 (2013) 054902.