Novel synthesis of actinide targets

Various UC_x, including nanostructured, pellets have been worked out both at ISOLDE-CERN and at IPNO, in order to determine how the microstructure properties of the material depends on the synthesis parameters, and to correlate these properties to the release kinetics of the fission products at high temperature. First results recently obtained have been published [1]. Further pellet prototypes have been achieved to study the influence of the synthesis parameters on the physicochemical properties of the porous UC_x material. These parameters involve the grinding conditions, the type of carbon source used and the carburization temperature. An overview of the different materials is displayed on Table 1. Multi-walled carbon nanotubes (CNT) were tested as carbon source, as recently studied for other refractory ceramics [2, 3]. Materials are synthesized as pellets of typically 1-2 mm thickness and 10-20 mm diameter.

Sample	Uranium	Carbon	ratio C/U	Milling of uranium powder	Carburization
OXA	IPNO oxalate	Graphite	3	No	16h at 1770°C
COMP30	IPNO oxalate	Graphite +30 wt % of microfibres	3	No	16h at 1770°C
PARRNe 371	natural UO_2 depleted 0.3% - ref. MN371	Graphite	6	Mixer miller PM100 RETSCH	16h at 1770°C
PARRNe 894	natural UO_2 depleted 0.25% - ref. MN894	Graphite	6	Mixer	16h at 1770°C
PARRNe 894 BP	natural UO_2 depleted 0.25% - ref. MN894	Graphite	6	Planetary miller PM200 RETSCH	16h at 1770°C
CNT	Westinghouse UO_2	Carbone nanotubes	6	No	20 min at 1600°C

Table 1: List of the different UCx materias synthesized during ActILab.

Physicochemical characterizations were systematically performed to describe the structure and microstructure of the carburized pellets (Table 2).

Sample	Phase(1) and their relative proportion (wt %)	Effective density(2) ($g.cm^{-3}$, $\pm$ 0.2)	Open porosity(2) (%)	Closed porosity(2) (%)
OXA	UC / UC_2 and 70.5 / 29.5	12.2	26	7
COMP30	UC / UC_2 and 8.6 / 91.4	10.1	48	13
PARRNe 371	UC / UC_2 and 10.6 / 89.4	8.3	46	4
PARRNe 894	UC / UC_2 and 10.9 / 89.1	8.0	56	8
PARRNe 894 BP	UC / UC_2 and 5.8 / 94.2	8.2	44	6
CNT	UC / UC_2 and 14.7 / 85.3	8.5	77	5

Table 2 : Physicochemical characterizations obtained by X-Ray Diffraction XRD(1) and He pycnometry (2).

Additionally, SEM observations and mercury intrusion porosimetry were done to obtain a description of the pore size distribution. Figure 2 shows the results for PARRNe 371 (sample used up to now as ISOL target at ALTO facility) and CNT, the most promising sample.

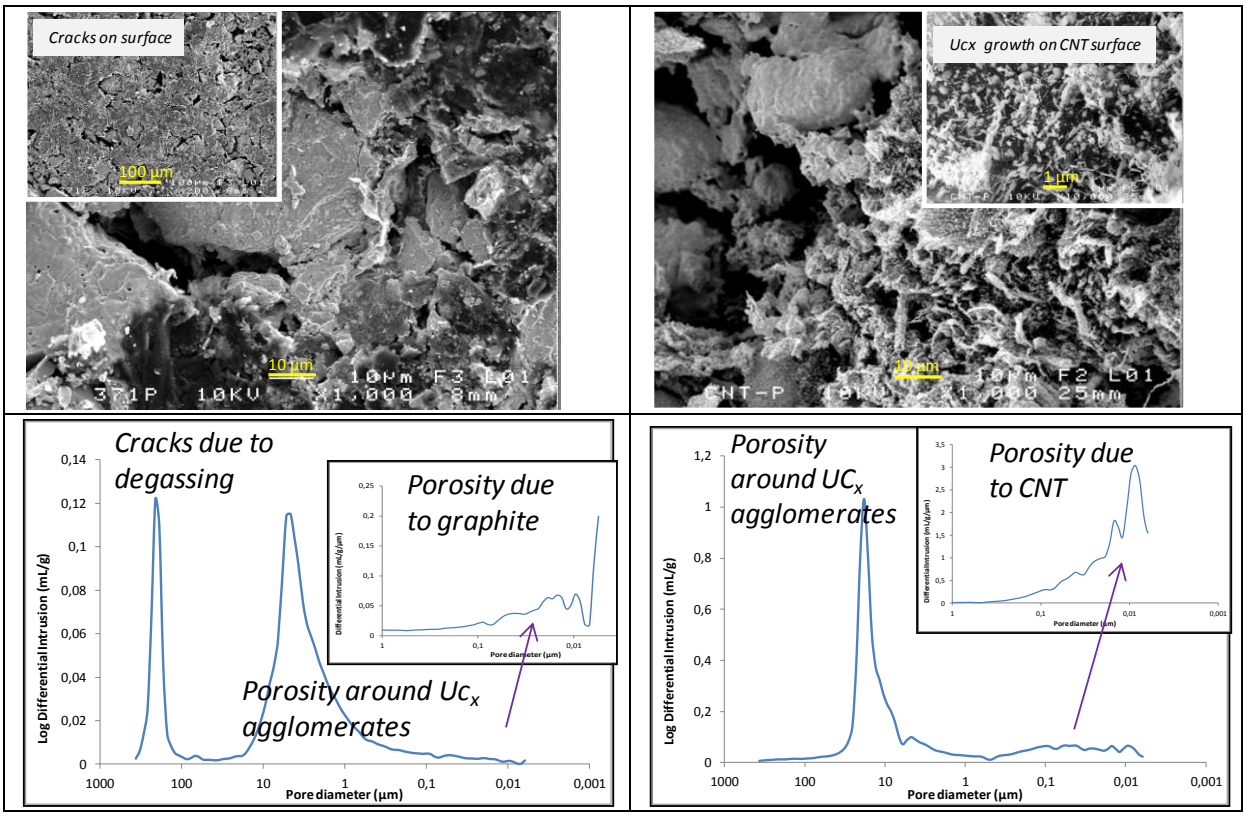

Figure 2: SEM observations and mercury intrusion porosimetry for (a) PARRNe 371 and (b) CNT

All of these physicochemical characterizations lead to conclude that:

- The grinding of uranium dioxide improves the homogeneity of the microstructure after carburization. Of course, the shrinkage is increased by 20% but the open porosity is still high. The carburization of grinded powder tends to stabilize mostly UC_2.
- Using uranium oxalate instead of uranium dioxide favors degassing during carburization but the closed porosity obtained is the highest.
- Using microfibers favors the formation of very large pores (cracks on surface) but the average open porosity is not increased in comparison with the samples made of graphite only.
- The use of CNT improves the open porosity. The microstructure of the pellet is more homogeneous with a limited grain growth. Nevertheless, it should be reminded that the carburization took place only 20 minutes, instead of 17 hours for the other samples. However, recent results obtained in CERN with targets made of lanthanum dicarbide and CNT [to be published] showed that a very weak grain growth occurs after a heating test at 1700°C for 6 days. An improvement of the synthesis is now necessary to obtain porous, homogeneous and nanostructured pellets with a reduced UCx grain growth.

References:

[1] Hy, B. et al. "An off-line method to characterize the fission product release from uranium carbide-target prototypes developed for SPIRAL2 project." *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 288 (2012): 34-41.

[2] Fernandes, S.et al., *"Microstructure evolution of nanostructured and submicrometric porous refractory ceramics induced by a continuous high-energy proton beam"; Journal of Nuclear Materials 416(1-2) (2011) 99-110*

[3] Ramos, J. P. et al., "*Intense Ar31-35 beams produced with a nanostructured CaO target at ISOLDE",* " *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* 320 (2014) 83-88