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Characterization of uranium carbide target materials to produce neutron-rich radioactive beams

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ABSTRACT

In the framework of a R&D program aiming to develop uranium carbide (UC_x) targets for radioactive nuclear beams, the Institut de Physique Nucléaire d'Orsay (IPNO) has developed an experimental setup to characterize the release of various fission fragments from UC_x samples at high temperature. The results obtained in a previous study have demonstrated the feasibility of the method and started to correlate the structural properties of the samples and their behavior in terms of nuclear reaction product release. In the present study, seven UC_x samples have been systematically characterized in order to better understand the correlation between their physicochemical characteristics and release properties. Two very different samples, the first one composed of dense UC and the second one of highly porous UC_x made of multi-wall carbon nanotubes, were provided by the ActILab (ENSAR) collaboration. The others were synthesized at IPNO. The systems for irradiation and heating necessary for the release studies have been improved with respect to those used in previous studies. The results show that the open porosity is hardly the limiting factor for the fission product release. The homogeneity of the microstructure and the pore size distribution contributes significantly to the increase of the release. The use of carbon nanotubes in place of traditional micrometric graphite particles appears to be promising, even if the homogeneity of the microstructure can still be enhanced.

1. Introduction

Finding optimal properties of the uranium refractory compound target materials constitutes a key ingredient for the production of a wide variety of isotope beams using the ISOL (Isotope Separator Online) technique. Different developments have been achieved in various facilities [1–3] and today uranium carbides with an excess of graphite (with variable stoichiometries, UC_x, since uranium dicarbide UC_2 and uranium monocarbide UC phases can be both stabilized depending on the amount of graphite added) are used by the different operating facilities throughout the world [4–8].

The optimization of UC_x targets is essential for the operation of the next generation facilities: EURISOL, HIE-ISOLDE, SPIRAL2 and SPES. It is also necessary for the existing facilities since higher intensities of short-lived nuclear beams are necessary in order to address important topics in nuclear and astrophysics. Recent experimental research aimed to correlate the submicron-scale porosity of materials with a significant improvement of their release properties. Firstly, very dense UC_x-based targets were developed by hot uniaxial pressing [9–12] and tested: the samples with 20 μm diameter grains of uranium monocarbide (UC) exhibited higher releases for all Rb and Cs isotopes than with a ISOLDE-type target. Then, porous structures were investigated, such as studies at SPES with the use of multi-wall carbon nanotubes (MWCNT) as carbon source [4]. The results showed that the yields were found to be, for most isotopes, lower than those obtained with a standard UC_x target. The microstructure was heterogeneous in term of UC_x grain size (micrometric) and porosity. MWCNT were also tested for other refractory ceramics

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[13,14]. Very recently, a new protocol was developed at ISOLDE and a homogenous nanostructure of LaC_2 was obtained, made of nanometric LaC_2 grains well dispersed in a carbon nanotube fiber matrix that could be stabilized even at high temperature [15].

Radioactive ions beams (RIB) intensities can be increased by improving the release efficiencies of UC_x fission targets which appear clearly correlated to the microstructure of the target material as we have previously shown in the framework of the SPIRAL2 project [7]. This study was dedicated to the development of dense and porous samples, two properties a priori antagonistic but necessary to increase respectively the amount of fission fragments produced and their diffusion out of the target. The quantity of graphite in excess was adjusted to stabilize UC rather than UC_2 in order to increase the density of the target.

The present study is focused on studying the releases out of single pellets of various fission products independently of the ^{238}U concentration. Seven very different UC_x samples ranging from a dense almost monophasic UC sample to highly porous UC_x composites synthesized with carbon nanotubes, instead of graphite, are characterized and compared. The two samples, labeled GATCHINA and CNT in the following, have been provided in the frame of the ActILab project (ENSAR Joint Research Activity within the European 7th Framework Program). The grinding of the uranium precursors and the nature of the carbon source (graphite, microfibers and carbon nanotubes) are taken into account. The whole experimental procedure aims to determine the impact of physico–chemical characteristics of a sample on the release properties. Therefore the correlation between the structural (quantity of phases) and microstructural properties of the samples (grain size, porosity and pore size distribution) and the fission-product release is studied.

2. Preparation and characterization of the samples

2.1. General remarks on techniques and methods used

X-ray diffraction (XRD) patterns were collected by an X-ray powder diffractometer (XRD, D8 Advance, Bruker AXS) in a Bragg–Brentano geometry (θ–θ) equipped with a Cu anode ($K_{\alpha 1}$ = 1.54178 Å). Samples were analyzed in air. Due to the use of a point scintillator detector, long time data acquisition was needed: from 10 to 90° 2θ, 0.02° step size, 16 s/step. Phase identification was performed with the DIFFRACPlus software (version 16, 2013, Bruker AXS) using powder diffraction files (ICDD PDF4+ 2013). The quantitative phase analysis was performed by using the MAUD software [16,17] in order to account for the Cu K_β line (Ni filter, no monochromator) and using the Crystallography Open Database [18]. The instrumental resolution was determined using a LaB_6 NIST standard (Standard Reference Material 660a, cell parameter = 0.41569162 nm ± 0.00000097 nm at 22.5 °C).

The effective density ρ_{eff} of the samples was measured with a helium pycnometer (Accupyc II 1340, Micromeritics). The apparent density ρ_{app} was determined by size measurement while the effective density corresponds to the volumetric mass of the pellet without open porosity [19]. Using these two values of densities, ρ_{eff} and ρ_{app}, the open porosity P_{open} of the samples can be assessed. Theoretical densities (ρ_{theo}) were estimated from the mass fraction of UC, UC_2 and UO_2 obtained by XRD and the estimated carbon in excess after carburization.

The pore size distribution was measured using a mercury intrusion porosimeter (AutoPore IV 9500, Micromeritics). Pressures were increased up to 228 MPa, corresponding to a pore size diameter of 5.5 nm. A surface tension constant of 0.485 N/cm, a contact angle between mercury and the pore wall of 130° were assumed and used in the Washburn equation [20].

The microstructural and morphological features were observed by scanning electron microscopy (SEM – JEOL 6301F) before irradiation. The secondary electron images (predominant topographic contrast) were taken using a 10 kV accelerating voltage. Samples, without metallization and polishing, were observed on both surface and fracture.

2.2. Characterizations of the raw powders

2.2.1. Uranium precursors

Depleted (0.3 wt% ^{235}U) uranium dioxide powder from CEA Cadarache (reference MN371) was used at the ALTO facility until March 2013. Hereafter AREVA depleted (0.25 wt% ^{235}U) uranium dioxide (MN894) is used; the powder consists of agglomerates made of spherical grains with an average size about 100 nm. The depleted uranium dioxide powder (0.31 wt% ^{235}U) used for samples prepared at ISOLDE-CERN is supplied by Westinghouse with an average particle size of 16 μm. The uranium oxalate powder, $U(C_2O_4)_2$, $2H_2O$, was prepared at IPNO [21,22] from uranium chloride obtained by dissolving natural metallic uranium in concentrated (4 M) hydrochloric acid; the powder consists of cuboid grains, with an average size of 2 μm.

2.2.2. Carbon precursors

The graphite powder (Cerac, purity = 99.5%, 325 mesh) was observed by SEM: it exhibits a large grain size distribution with a diameter less than 40 μm. SEM observations on carbon microfibers (Torayca) reveal a diameter of 5.2–7.5 μm with a length ranging from 55 to 310 μm, while the purchaser indicates a diameter of 7 μm and a length ranging from 10 to 150 μm. MWCNT were purchased from Nanocyl SA, (>95% C, <5% Metal Oxide, D = 9.5 nm, L = 1.5–10 μm) and used as received.

2.3. Synthesis of the pellets

The OXA samples were prepared from a mixture of uranium oxalate and graphite with a ratio of 1 mol of UO_2 for 3 mol of graphite. The COMP30 samples consisted of OXA samples in which 30 wt% of microfibers (μC) was added. Several mixtures were done separately and mixed together in order to obtain one homogeneous mixture for the preparation of the pellets. One batch for one pellet weighted about 1.3 g to obtain after sintering a pellet of about 1 g.

The UO_2 powders used at IPNO were ground before use with a mixer mill RETSCH PM200 or a planetary mill RETSCH PM100. The UO_2 for CNT samples provided by CERN was used as supplied.

GATCHINA pellets were obtained by powder metallurgy from uranium and graphite, as previously published [9–12]. The pellets used in this work presented an apparent density of 12.7 g cm^{-3} and an average grain size of 6 μm. Information about the raw powders was not available.

The experimental setups are summarized in Table 1. Table 2 summarizes the conditions of synthesis of the seven kinds of tested samples. At IPNO, UO_2 and graphite were mixed together manually in an agate mortar. At CERN, depleted UO_2 powder with an average particle size of 16 μm was mixed with MWCNT (9.5 nm thickness and 1.5–10 μm length) in a vibratory ball mill.

2.4. Pressing

A semi-automatic hydraulic press placed in a glove box was used (SPECAC Automatic Hydraulic Press) for OXA, COMP30 and PARRNe samples. The pressing protocol was performed by steps: from 0 to 3 t, waiting 1 min; from 3 to 5 t, waiting 3 min; from 5 to 7.5 t, waiting 1 min; from 7.5 to 10 t, waiting 15 min and after a slow decrease of the pressure. A 13 mm or 14 mm diameter pellet