Uranium Carbide for the SPES Project

INTRODUCTION

For the next generation of Radioactive Ion Beams (RIB) facilities robust production targets capable of withstanding irradiation with light ion beams at high power levels for long periods must be developed. These targets should be capable of releasing a broad spectrum of isotopes of a large number of elements, with short lifetime (less than 100 ms).

As tested in the past at ISOLDE [1] and recently studied at HRIBF [2] uranium dicarbide dispersed in excess graphite (namely UC\(_x\)) is the ideal candidate as target material for the production of RIB. Even though the release of nuclear reactions products has been extensively studied for RIB production [3], the reasons for the good release properties of such a material are not yet fully understood.

Since the process of isotopes release consists of diffusion within the grain and effusion towards the ionization source, the composition, the density and the operating temperature of the material constituting the target are important parameters to be considered.

In order to achieve the best performance, the target material should respond to stringent requirements, such as:
1. to have large cross section and diffusion coefficient;
2. to work at the highest possible operating temperature, defined as the temperature at which its vapour pressure begins to affect the ionization efficiency of the ion source used in the exotic ions generation;
3. to possess high emissivity and high thermal conductivity, responsible for heat dissipation during the target bombardment: a good heat dissipation of the target prevents its overheating and mechanical stresses;
4. to exhibit low density and high permeability to the effusion of the produced isotopic species. The presence of open porosity permits both diffusion and permeability [4].

Within the framework of the SPES project (Selective Production of Exotic Species) the research in the field of uranium carbide target materials is directed towards the development of materials which meets the requested properties able to maximize the target yield. In the following the activity of production of uranium carbide in the Radiochemistry Laboratory of the University of Padova will be shortly described.

EXPERIMENTAL PART

Porous UC\(_x\) samples were prepared by means of thermal treatment under vacuum of UO\(_2\), U\(_3\)O\(_8\), UO\(_3\) and graphite mixtures following the reaction scheme 1, keeping the x/n ratio equal to 1/4.

$$\text{U}_x\text{O}_y+(z+y+n)\text{C} \rightarrow \text{U}_x\text{C}_z + n\text{C} + y\text{CO} \quad (1)$$

The UO\(_2\) and U\(_3\)O\(_8\) materials were purchased from CERAC Inc. (Milwaukee, WI, USA) under powder form having size <300 \(\mu\)m. The graphite (powders size <45 \(\mu\)m) was instead taken from Sigma-Aldrich and all used as received. UO\(_3\) was prepared from uranyl nitrate hexahydrate (Sigma-Aldrich), according to the method proposed by Allen [5]. Samples prepared from UO\(_2\) and graphite mixtures were labeled as UO\(_2\)C, samples prepared starting from U\(_3\)O\(_8\) and graphite mixtures were labeled as U\(_3\)O\(_8\)C, samples prepared starting from UO\(_3\) and graphite mixtures were labeled as UO\(_3\)C. The reason for the use of this processing condition lies on:
1. the lower temperature needed by the carbothermic reaction, compared to the same process carried out under inert atmosphere;
2. the formation of pores, resulting from the CO released during the carbothermic reduction that occurs at \(T > 1000 \degree\)C.

The sintering process, which takes place at higher temperatures than the carbothermic reduction, can cause pores of small dimension to collapse if sintering is not appropriately controlled. This shortcoming makes it very difficult to properly control the amount and type of porosity generated by the thermal treatment. The thermal treatment was performed under high vacuum (\(10^{-4}-10^{-5}\) Pa) in a graphite crucible using the experimental set-up described in [6]. The heating schedule was designed in order to:
1. promote the carbothermic reaction (2\(^\circ\) C/min up to 1250 \(\degree\)C, 24 h at 1250 \(\degree\)C);
2. sinter the carburized powders (2 \(\degree\)C/min up to 1600 \(\degree\)C, 4 h at 1600 \(\degree\)C).

The cooling was performed slowly at a rate of 2\(^\circ\)C/ min. The heating process was monitored by means of a penning trap which measured the pressure inside the chamber (in
the $10^{-1}-10^{-5}$ Pa pressure range). The final samples were analyzed by X-Ray Diffractometry using Cu-Kα radiation (Philips PW 1710, XRD). During the analysis, the powders were protected from the environment by a PEEK film (VICTREX- UK, 25 μm thickness). After carburization and sintering, the samples were stored in a glove-box ($O_2$ and $H_2O < 1$ ppm). Samples were weighted and their dimensions were recorded. The bulk density ($\rho_{bulk}$) was measured by the weight over volume ratio. The theoretical density ($\rho_{th}$) was calculated as the weighted average, taking into account the values of 11.2 g/cm$^3$ and 1.9 g/cm$^3$ for the theoretical density of UC$_2$ and graphite, respectively.

**RESULTS AND DISCUSSION**

The pressure evolution versus temperature within the reaction chamber is reported in figure 1. As can be observed in the figure below the out-gassing in the three samples can be attributed to:

1. the elimination of adsorbed water from the powders of oxides and graphite;
2. the decomposition of the phenolic resin used as binder, which thermal gravimetric analysis indicates to occur in the 350 °C – 800 °C temperature range;
3. the reduction of $UO_3$ and $U_3O_8$ to $UO_2$, with release of $CO_2$;
4. The carbothermic reaction of $UO_2$ at $T > 1000$ °C.

**CONCLUSIONS**

Uranium carbide pellets of 13 mm diameter and 1 mm thickness were successfully produced and characterized in the Radiochemistry Laboratory of the University of Padova. The final composition of the samples was uranium dicarbide, graphite and a minor amount of uranium monocarbide. The samples bulk density and porosity were not affected by the uranium precursor.