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Thermal conductivity and emissivity measurements of uranium carbides

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ABSTRACT

Thermal conductivity and emissivity measurements on different types of uranium carbide are presented, in the context of the ActiLab Work Package in ENSAR, a project within the 7th Framework Program of the European Commission. Two specific techniques were used to carry out the measurements, both taking place in a laboratory dedicated to the research and development of materials for the SPES (Selective Production of Exotic Species) target. In the case of thermal conductivity, estimation of the dependence of this property on temperature was obtained using the inverse parameter estimation method, taking as a reference temperature and emissivity measurements. Emissivity at different temperatures was obtained for several types of uranium carbide using a dual frequency infrared pyrometer. Differences between the analyzed materials are discussed according to their compositional and microstructural properties. The obtainment of this type of information can help to carefully design materials to be capable of working under extreme conditions in next-generation ISOL (Isotope Separation On-Line) facilities for the generation of radioactive ion beams.

1. Introduction

The use of radioactive ion beams (RIBs) [1] in nuclear physics has been established as a fundamental method to both expand the current knowledge on nuclei very far from stability and provide a powerful tool for applications in several fields. Facilities which produce RIBs, or that are now being developed to do so in the near future, are traditionally divided into two main categories, depending on the way radioactive isotopes are generated and how the beam is formed and transported [2]: the in-flight method and the ISOL (Isotope Separation On-Line) technique.

In the first case [3], radioactive isotopes are created by the interaction (fragmentation or fission) of a primary beam of heavy ions with a thin target. The reaction fragments are ejected in the forward direction with respect to that of the incident beam and subsequently separated with a fragment separator and sent to the experimental areas. In the ISOL technique [4], spallation or fission reactions are obtained by bombarding targets made of heavy elements (mainly uranium), with intense beams of light particles (typically protons). The produced neutral radioisotopes diffuse out of the target and effuse towards an ion source, where they are ionized. After passing through different stages of separation and manipulation, the formed beam is post-accelerated to the experimental halls.

The core of an ISOL facility is represented by its target-ion source complex. In particular, the choice of the material constituting the target is vital to ensure excellent performances in terms of quantity and regularity of the isotopic yields over the duration of beam delivery. As stated above, during the operation of an ISOL facility the target material is involved in different processes:

- Generation of particles through nuclear reactions between the primary beam and the target nuclei.
- Diffusion of the produced isotopes inside the grains constituting the material.
- Effusion of isotopes from the grain surface inside the material porosity towards the target surface, and subsequent effusion towards the ion source.

Besides thermodynamical processes, chemical reactions of the produced isotopes with the surrounding materials (target and enclosures) are important factors which can affect their release. In order to increase the rates of diffusion and effusion, the target is kept at high temperature (2000 °C) in high vacuum (10^{-6} mbar or less) during operation [5]. This aspect, combined with the extreme thermomechanical stresses which are generated during the primary beam irradiation, makes the correct choice of the target material composition and properties even more fundamental.

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Due to its suitable nuclear (high cross section for the reaction between protons and ^{238}U) and thermomechanical (high critical temperature) properties, and the relative facility to synthesize it, uranium carbide is by far the most used material as a target for RIB production in ISOL facilities [5]. In most cases, targets are not made by fully dense uranium carbide, but instead contain a variable amount of graphite and residual open interconnected porosity, conditions which have been found to favor the isotopic release [6]. This material is commonly referred to as UC_X.

The ActiLab Work Package in ENSAR, a project within the 7th Framework Program of the European Commission, brought together different European laboratories (CERN, INFN-LNL, GANIL, IPNO and PSI) with the aim of studying and testing the synthesis and characterization of innovative materials based on uranium carbide [4]. In this paper, thermal conductivity and emissivity measurements performed on uranium carbides of different composition and microstructural properties, synthesized in the framework of this Work Package, are reported.

The obtainment of experimental data relative to these two properties is considered a fundamental step towards the development of future high power ISOL facilities, such as EURISOL [7], in which the target materials will be used in even more extreme environments with respect to the current ones.

In literature, very few data are available relatively to the thermal conductivity of uranium carbides. The most notable results were obtained by De Coninck et al. [8–10], who reported thermal conductivity values for UC, UC_2 and U_2C_3 by means of thermal diffusivity measurements using the laser-flash technique, and at the same time performed emissivity measurements using infrared pyrometers. More recently, thermal conductivity measurements on porous UC_2 were performed by Greene and co-authors [11] in the framework of the research on targets for ISOL facilities. Other data about the thermal conductivity of UC and UC_2 is available in [12–15]. A summary of the available data concerning thermal conductivity and emissivity of UC and UC_2 is reported in Table 1.

2. Thermal conductivity estimation using inverse analysis

The method here described for the estimation of thermal conductivity of uranium carbide is based on the one already reported by Manzolaro et al. [16], successfully applied for graphite, silicon carbide and lanthanum carbide SPES [17] 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, in which the research and development of materials for the SPES target is carried out. Details about the experimental device, specifically developed for the measurements here reported, are given in section 3. 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 [18].

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, heated by thermal radiation thanks to a hot graphite crucible placed at a certain distance from it, directly facing its bottom surface. A setup of this type is schematized in Fig. 1 [16]. Since the heating of the crucible is induced by Joule effect and the graphite resistivity depends on temperature, the thermal and the electrical problems controlling the system are coupled [16]. Moreover, since the measurements are conducted in high vacuum, the obtained thermal interaction between the crucible and the disc is a result of conduction and radiation only. This type of problem is characterized by the diffusion of energy within the solid region V and the radiative heat transfer between the surfaces forming the enclosure S_{enc}. The conductive problem can be expressed in general terms as [19]:

$$\frac{\partial}{\partial x}\left(k\frac{\partial T}{\partial x}\right)+\frac{\partial}{\partial y}\left(k\frac{\partial T}{\partial y}\right)+\frac{\partial}{\partial z}\left(k\frac{\partial T}{\partial z}\right)+\dot{q}=\rho c\frac{\partial T}{\partial t} \tag{1}$$

where $T(x,y,z)$ is the temperature field in the solid region V, t is the time, ρ is the density of the material constituting V, c is the specific heat, k is the thermal conductivity and $\dot{q}$ is the volumetric heat source.

The radiative part of the problem can be represented as [20]:

$$\sum_{i=1}^{N}\left[\frac{\delta_{ji}}{\varepsilon_i}-F_{j-i}\left(\frac{1-\varepsilon_i}{\varepsilon_i}\right)\right]\cdot q_{\mathrm{enc},i}=\sum_{i=1}^{N}(\delta_{ji}-F_{j-i})\cdot\sigma\cdot T_i^4 \tag{2}$$

where δ_{ji} is the Kronecker delta, ε_i is the hemispherical total emissivity of surface i, F_{j-i} is the radiation view factor, $q_{\mathrm{enc},i}$ is the net rate of radiative energy loss per unit area (flux) of surface i, σ is

Table 1

Thermal conductivity and emissivity data reported in literature for UC and UC_2. (a) Spectral emissivity at 0.65 μm, (b) spectral emissivity at 2.3 μm, (c) data relative to a porous sample containing UC_2 and graphite, all the other values reported in this table are relative to highly dense materials.

	UC	UC_2
Thermal conductivity (W/m °C)	23 ÷ 20 (650 ÷ 1150 °C) 20 ÷ 26 (1150 ÷ 2250 °C) [8]	13 ÷ 18 (600 ÷ 1740 °C) 19 ÷ 20 (1800 ÷ 2060 °C) [9]
	23 ÷ 21 (50 ÷ 450 °C) 21 ÷ 80 (450 ÷ 2300 °C) [12]	5 ÷ 8 (1500 ÷ 1880 °C) [11] (c)
		11 ÷ 20 (0 ÷ 1500 °C) [13]
	21 ÷ 17 (100 ÷ 400 °C) [14]	12 ÷ 21 (100 ÷ 2300 °C) [12]
	22 ÷ 24 (75 ÷ 2130 °C) [15]	
		7 ÷ 6 (200 ÷ 400 °C) [14]
Emissivity	0.49 ÷ 0.48 (1100 ÷ 2250 °C) [8] (a)	0.50 ÷ 0.53 (1040 ÷ 1740 °C) 0.55 (1800 ÷ 2060 °C) [9] (a)
		0.44 ÷ 0.45 (1040 ÷ 1740 °C) 0.50 (1800 ÷ 2060 °C) [9] (b)