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The Weak Acid Resin process: a dustless conversion route for the synthesis of americium bearing-blanket precursors

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Abstract

Mixed uranium-americium oxides are one of the materials envisaged for Americium Bearing Blankets dedicated to transmutation in fast neutron reactors. Conversion and fabrication processes are currently developed to make those materials in the form of dense and homogeneous oxide ceramic pellets or dense granulates incorporating uranium and americium. Their development points out the need of a simplified and optimized process which could lower hazards linked to dust generation of highly contaminating and irradiating compounds and facilitate material transfer in remote handling operations. This reason motivated the development of innovative "dustless" route such as the Weak Acid Resin route (WAR) which provides the oxide precursors in the form of sub-millimeter-sized microspheres with optimal flowability and limits dust generation during conversion and fabrication steps. This study is thus devoted to the synthesis of mixed uranium-americium oxide microspheres by the WAR process and to the characterization of such precursors. This work also deals with their application to the fabrication of dense or porous pellets and with their potential use as dense spherules to make Sphere-Pac fuel.

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Keywords: americium; uranium; oxide; microsphere; pellet; conversion; resin; transmutation.

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1. Introduction

Transmutation of minor actinides such as americium is a potential option considered for lowering radiotoxicity and thermal impact of ultimate waste in final depository in the case of a back-end nuclear fuel cycle including Pu recycling[1].

A specific process for the fabrication of americium-bearing fuel or blanket will then be needed in the mid-term[2]. R&D studies are currently ongoing to validate and optimize conversion and fabrication processes[3]. The main goal of those developments is to take into account the specificity of americium, which is a highly radioactive and contaminating radionuclide and which requires remote handling in shielded hot cell. Conversion and fabrication processes need to be simplified and optimized. One priority is to limit matter dissemination and facilitate material transfer during conversion and fabrication steps. In that context, the developments of processes which implement sub-millimeter-sized particles instead of micron-sized powders are highly recommended.

Conversion processes that may provide actinide oxide in the form of microspheres are good candidates to tackle this challenge: sol-gel processes[4], and sol-gel processes associated with infiltration methods[5] and processes based on the use of resin template microspheres such as the Weak Acid Resin process (WAR)[6] were tested on surrogate materials and progressively on actinide compounds[7,8]. In this work, the WAR process was revisited for the synthesis of oxide microspheres and validated for lanthanide[9] or uranium-based[10] oxides and finally americium-uranium mixed oxide[11]. The application of such a process concerns either dense or porous pellet fabrication but can also address the fabrication of Sphere-Pac fuel[12].

A focus will be given on the implementation of the Weak Acid Resin process in the Atalante facility and on the production of uranium-americium mixed oxide microspheres, which were used for the fabrication of dense mixed oxide pellets. The different stages of the microsphere precursor synthesis will be described as well as the characterization of such objects in terms of structure, morphology, microstructure, density and mechanical properties. The fabrication of dense or porous pellet from those precursors will also be discussed and their characterization will be presented. As a perspective, the adaptation of the process for making dense microspheres for Sphere-Pac fuel application will finally be discussed.

2. Experimental

2.1. Precursor synthesis

The WAR conversion process applied to the synthesis of mixed uranium-americium oxide microspheres is based on the fixation of uranyl and americium cations on beads of ion exchange resin, followed by the mineralization of the metal loaded resin by a first thermal treatment in an oxidative atmosphere and finally a second thermal treatment in a reductive atmosphere in order to adjust the oxygen stoichiometry of the uranium oxide.

The ion exchanger is a polyacrylic resin (Dow Chemicals, IMAC HP333) which is in the form of beads and which is sieved and washed before use (size distribution in the range 630-800µm). Exchange is performed on a column by recirculating an acid deficient uranyl-americium nitrate solution for a contact time of about 5 h. After fixation, the metal loaded resin is washed with deionized water and dried under air flow.

The beads are first collected in a crucible and calcined in a tubular furnace under synthetic air flow from room temperature up to 700°C at a rate of 2°C/min to give the intermediate metal oxide microspheres. Those intermediate oxides are secondly heat treated in a reducing atmosphere of Ar/H_2 (4 vol.%) at 700°C for 6h with a heating rate of 10°C min^{-1} to provide the uranium-americium mixed oxide at an oxygen stoichiometry close to 2.

2.2. Precursor characterization

Metals in solution were measured by Thermal Ionization Mass Spectrometry (TIMS) or UV-visible spectrophotometry (Agilent, Cary 6000i). Thermal conversion of the metal loaded resin was characterized by Thermal Gravimetric Analysis (TGA) (Netzsch STA99) coupled to micro-gas chromatography (µGC) (SRA). The crystalline structure of the resulting oxide was characterised by powder X-ray diffraction (XRD) using a θ-θ D8 advance BRUKER diffractometer equipped with a copper anticathode ($\lambda(K^{Cu}_{\alpha 1})$ = 1.54056 Å, $\lambda(K^{Cu}_{\alpha 2})$ = 1.54439