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Innovative oxide precursor synthesis dedicated to dustless process for minor actinide pellet fabrication

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Minor actinides (MA) are generated in UOX (uranium oxide) and MOX (mixed oxide) fuels during irradiation in nuclear reactors [1]. In the framework of their future management, their transmutation in fast neutron reactors through MABB (Minor Actinide-Bearing Blanket) irradiation represents a major research concern. In this context, due to its relatively high radiotoxicity in ultimate waste (if Pu multi-recycling option is considered), americium (Am) is the most studied MA for transmutation through the fabrication of $U_{1-x}Am_xO_{2\pm\delta}$ ceramic pellets. Commonly, these fabrications are based on powder metallurgy processes with ball-milling steps which generate large amounts of fine radioactive particles. Dustless processes are thus mandatory before envisaging an industrial deployment, and as a first step, for providing the fourth-generation-reactor project ASTRID (Advanced Sodium Technological Reactor for Industrial Demonstration). Answering this need, the development of an innovative route using micrometric spherical precursors has been initiated. Through an adaptation of the weak acid resin (WAR) process [2,3,4], the general approach consists in synthesizing micrometric and brittle spherical $U_{1-x}Am_xO_{2\pm\delta}$ mixed-oxide precursors. More particularly, oxide microsphere synthesis begins with the fixation of americium and uranium cations into ion exchange resin beads. The key step is then to study the ceramization of polymeric microspheres, through their thermal conversion under air. Then, spherical oxide precursors are compacted and sintered as dense ceramic pellets, answering the required AmBB specifications. The spherical geometry was chosen to facilitate the filling of the compaction chamber and subsequent pelletization step, as well as particle agglomeration into the microsphere tends to reduce particle dissemination.

In this context, the main objective of the study is to control oxide microstructure and mechanical properties to allow microspheric precursor pelletizing and sintering into dense pellets (theoretical density (TD) > 95%). Understanding $U_{1-x}Am_xO_{2\pm\delta}$ microsphere synthesis and particularly mechanisms

implied during calcination leading to polymer skeleton combustion and the formation of the mixed oxide solid solution hence appears fundamental. This is the purpose of this presentation combining in-situ results obtained through XRD (X-Ray Diffraction) and, for the first time on Am-based samples, XAS (X-Ray Absorption Spectroscopy).

Combination between the two in-situ technics will lead to the presentation of unpublished results on (U,Am)O₂ solid solution formation and its understanding. The determination of the different chemical structure taken by the elements during the mineralization were obtained thanks to XRD analysis, while the different valences taken by U and Am elements will be presented for the first time, with their temperature dependence. These data will be associated to SEM observations of the oxide microsphere microstructure at the different key step identified thanks to the in-situ characterizations. The entire work will be helpful for optimizing the thermal treatment required for oxide microspheres synthesis considering structural and chemical properties.

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