

A New Process for Dense $U_{1-x}Am_xO_{2+\delta}$ Fuel Fabrication by Conventional Sintering

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Abstract

In the frame of minor actinides, and more specifically americium, recycling via heterogeneous transmutation in FNR, MABB (Minor Actinide Bearing Blankets) fuel fabrication processes are currently being developed and optimised. Until now, these processes are based on those commonly used to prepare mixed-oxide fuels (such as $U_{1-x}Pu_xO_2$). They are thus based on a high temperature reactive sintering between two (at least) single-oxide (such as $UO_{2+\delta}$, $AmO_{2-\delta}$) precursors which can lead to defects (such as porosity or chemical/structural inhomogeneity) discrepant with requirements for such fuels. These defects are due to the competition between the two main phenomena constituting a reactive sintering, namely the solid solution formation, and the densification; the latter being inhibited as long as the former is not complete. In the case of $U_{1-x}Am_xO_{2+\delta}$ MABB fuels, the large difference in diffusion coefficient and oxygen potential between uranium and americium oxides renders the use of reactive sintering quite challenging for the fabrication of homogeneous and dense ($d > 94\%$ TD) $U_{1-x}Am_xO_{2+\delta}$ MABB fuels.

To overcome these limitations a new process, called UMACS (Uranium / Minor Actinide Conventional Sintering), has recently been developed at the LEMA (CEA Marcoule). This process is based on the separation of the two above-mentioned phenomena. Firstly, a solid state route is employed to synthesize a solid solution: a homogeneous powder mix of single dioxide is prepared, pelletized and heat treated. The thermal cycle and the atmosphere are chosen to favour the formation of the intended $U_{1-x}Am_xO_{2+\delta}$ oxide solid solution. A recent in-situ study on $UO_{2+\delta}/AmO_{2-\delta}$ interaction allows specifying the required thermal cycle. The atmosphere is composed of an $Ar/H_2/H_2O$ mixture to avoid any americium sublimation while maintaining uranium oxide in the fluorite-type-structure dioxide form. Secondly, a $U_{1-x}Am_xO_{2+\delta}$ powder is produced from those heat-treated pellets by the use of a grinding step. As-obtained monophasic powder is thus suitable for a conventional sintering. The solid solution formation/densification competition being avoided, the sintering is not impeded and highly dense samples can finally be achieved, without a significant amount of open porosity. As all the americium is already integrated in the solid solution, its sublimation is prevented, which allows varying the atmosphere in order to adjust the O/M ratio of the fuel.

The feasibility and reproducibility of this process will be exemplified by the recent fabrication and characterisation of highly dense $U_{1-x}Am_xO_{2+\delta}$ ($x = 0.075; 0.15; 0.2$) MABB fuels using the UMACS process.