Local structure of $Th_{1-x}MO_2$ solid solutions (M = U, Pu)

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Abstract - *X-ray absorption spectroscopy of $Th_{1-x}U_xO_2$ and $Th_{1-x}Pu_xO_2$ solid solutions was carried out on the Th, U L_3-edges, and Pu L_3 edge to study the local structure environment of actinide mixed oxides. Various compositions of $Th_{1-X}M_XO_2$ solid solutions have been prepared through the coprecipitation of the mixed oxalates from chloride or nitrate solutions: x = 0.11, 0.24, 0.37, 0.53, 0.67, 0.81, 0.91 and 1 for $Th_{1-x}U_xO_2$, and x = 0.13, 0.32, 0.66 and 1 for $Th_{1-x}Pu_xO_2$. They were characterized using X- ray diffraction. XRD analysis allowed to confirm that the variation of the lattice parameters varies linearly with the composition between the end members, suggesting that the atomic volume was conserved regardless of the details of the local distortions of the lattice, following the Vegard's law.*

Extending X-ray absorption fine structure (EXAFS) provides a direct characterization of the local distortions present in solid solutions. We found that opposite to the lattice parameter obtained by XRD, the interatomic distances given by EXAFS do not follow completely to neither the Vegard's law nor the virtual crystal approximation (VCA). However, the average lattice parameter obtained from EXAFS data for the first and the second shells agrees well with the one calculated from XRD data.