Thorium effect on the oxidation of uranium: Photoelectron spectroscopy (XPS/UPS) and cyclic voltammetry (CV) investigation on (U$_1$ − xTh$_x$)O$_2$ (x = 0 to 1) thin films

Abstract:
Thin films of U$_1$ − xTh$_x$O$_2$ (x = 0 to 1) have been deposited via reactive DC sputter technique and characterized by X-ray/Ultra-violet Photoelectron Spectroscopy (XPS/UPS), X-ray Powder Diffractometer (XRD) and Cyclic Voltammetry (CV) in order to understand the effect of Thorium on the oxidation mechanism. During the deposition, the competition between uranium and thorium for oxidation showed that thorium has a much higher affinity for oxygen. Deposition conditions, time and temperature were also the subject of this study, to look at the homogeneity and the stability of the films. While core level and valence band spectra were not altered by the time of deposition, temperature was affecting the oxidation state of uranium and the valence band due to the mobility increase of oxygen through the film. X-ray diffraction patterns, core level spectra obtained for U$_1$ − xTh$_x$O$_2$ versus the composition showed that lattice parameters follow the Vegard's law and together with the binding energies of U-4f and Th-4f are in good agreement with literature data obtained on bulk compounds. To study the effect of thorium on the oxidation of U$_1$ − xTh$_x$O$_2$ films, we used CV experiments at neutral pH of a NaCl solution in contact with air. The results indicated that thorium has an effect on the uranium oxidation as demonstrated by the decrease of the current of the oxidation peak of uranium. XPS measurements made before and after the CV, showed a relative enrichment of thorium at the extent of uranium at the surface supporting the formation at a longer term of a thorium protective layer at the surface of uranium-thorium mixed oxide.

URI:
Authors:
CAKIR Pelin
ELOIRDI Rachel
HUBER Frank
KONINGS Rudy
GOUDER Thomas
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