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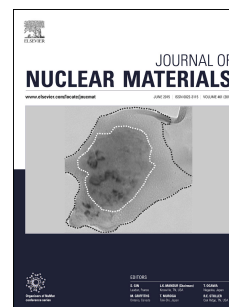
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Extended study on oxidation behaviors of $\text{UN}_{0.68}$ and $\text{UN}_{1.66}$ by XPS

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Abstract

The surface oxidation behaviors of $\text{UN}_{0.68}$ and $\text{UN}_{1.66}$ thin films are investigated by X-ray photoelectron spectroscopy (XPS), and the traditional U4f/N1s, O1s, valence band spectra as well as the unconventional U4d and U5d spectra are collected for the understanding of their oxidation behavior in-depth. Similar asymmetrical peak shape of the U4f spectra to uranium is observed for both uranium nitrides, despite of the slight shift to higher energy side for $\text{UN}_{1.66}$ clean surface. However, significant difference among the corresponding spectra of $\text{UN}_{0.68}$ and $\text{UN}_{1.66}$ during oxidation reveals the distinctive properties of each own. The coexistence of UO_{2-x} , UO_2 and $\text{UO}_{2-x}\text{N}_y$ on $\text{UN}_{0.68}$ surface results in the peculiar features of U4f spectra as well as the others within the XPS energy scale, where peaks of the oxidized species firstly shift to higher energy side compared to the clean surface, and then return towards those of stoichiometric UO_2 . For $\text{UN}_{1.66}$, the generation of U-N-O ternary compounds on the surface is identified with the symmetrical U4f peaks at 379.9 eV and 390.8 eV, which locate intermediate between UO_2 and $\text{UN}_{1.66}$, and gradually expanding to higher energy side during oxidation. Furthermore, the formation of N-O species on $\text{UN}_{1.66}$ surface is also detected as an oxidation product. The metallic character of $\text{UN}_{1.66}$ is identified by the intense signals at Fermi level, which is greatly suppressed by the increasing oxygen exposure and implies the weakening metallic properties of the U-N-O compounds. A multistage mechanism for $\text{UN}_{1.66}$ following the exposure to oxygen is discussed.

Key words: uranium nitride, XPS, oxidation, U-N-O ternary compound, UO_{2-x} , nitrogen oxide

Introduction

Uranium nitride is regarded as a proposal nuclear fuel for the IV generation nuclear reactor, and has been extensively studied experimentally since the 1960s. However, the properties of uranium nitride are less studied than for carbide and oxide fuel. Owing to the large variety of available valence states of uranium, the oxidation process of uranium contained system is very complex as known. A great deal of theoretical investigations have been carried out on studies of the electronic [1, 2], structural [1], thermodynamic [2, 3], mechanical [4, 5], magnetic properties [4] and surface chemisorption behaviors [6] of uranium nitride.

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