

## Study of Trivalent Cation doped Uranium Oxides through In-Situ X-ray Diffraction

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### 1. Introduction

For the confirmation of material components and structural formations, powder X-ray diffraction method has been generally used in the material research fields. [1] In the materials of nuclear fuel fields, structural deformation and compositional analyses of uranium oxides have been also used through powder X-ray diffraction technique. [2]

Uranium oxide materials have been of particular interest in broad nuclear fuel fields because of the structural changes derived from various oxidation state numbers such as UO<sub>2</sub>, U<sub>4</sub>O<sub>9</sub>, U<sub>3</sub>O<sub>8</sub> and UO<sub>3</sub>. Cation doped uranium oxide materials have drawn specially notice because of their applicable properties as semiconductors. [3-5]

Therefore, research of cation doped uranium oxide is important to understand their structure-characterization relationship. Because of the specificity of cation doped uranium oxide, the crystal structure is measured using neutron diffraction and X-ray diffraction after synthesis experiments of materials.

Recently, in-situ X-ray diffraction analysis method that can immediately confirm a structural change of a reactant has attracted attention. In addition, when a temperature control system is combined with X-ray diffraction measurement device, a structural change according to temperature can be confirmed in real-time.

In the case of nuclear fuel material, since the structure of the nuclear fuel can be observed during the reaction at high temperature, the difference of the nuclear fuel structure measured can be clearly identified at room temperature.

Lebreton's group has researched the Am doped UO<sub>2</sub> with in situ XRD at ~1973K. The results showed the performance of the lattice parameter, the crystallite size, and the strain up to ~1973K. [6] In this work, we report the study of trivalent cation (Gd<sup>3+</sup> and Nd<sup>3+</sup>) doped uranium oxide materials behaviors using in situ high temperature X-ray diffraction. We measure the variation of temperature on the size of crystalline, which is a cell parameter in the reaction process. And then,

the change of lattice parameters are calculated by Rietveld refinement method.

### 2. Experimental

Trivalent cation (Gd<sup>3+</sup> and Nd<sup>3+</sup>) doped uranium oxide powders were ground thoroughly with agate mortars and pestles and pressed into pellets. The pellets on the tungsten strip were evacuated and gradually heated to 1800 °C.

The In-situ X-ray powder diffraction patterns were collected on a Bruker D8-Advance diffractometer using Cu K $\alpha$  radiation with 40 kV and 40 mA and an Anton Parr HTK 2000 heating chamber. The scan ranges were 20-80° with a step size of 0.02°, and a step time of 0.1s. The temperature ranges were 25 °C - 1800 °C. The diffraction patterns were analyzed using Rietveld method with the TOPAS program. The Structural refinement of the materials was carried out in the space group *Fm-3m* (no.225) with a starting model based on the reported data of UO<sub>2</sub> [PDF#: 00-041-1422].

The lattice parameters were refined calculation methods, followed in subsequent iterations by the zero point error, unit-cell, peak shape, and temperature parameters.

### 3. Results and discussion

The results of the trivalent cation (Gd<sup>3+</sup> and Nd<sup>3+</sup>) doped uranium oxide at 1800 °C x-ray diffraction patterns are shown in Fig. 1. The crystal structure changes of the powders were revealed from the XRD patterns corresponding to 1800°C. The diffraction peaks move to the left with increasing amounts of Nd<sup>3+</sup> on the U<sup>4+</sup> site in the U<sub>0.9</sub>Nd<sub>0.1</sub>O<sub>2</sub>. However, in the case of U<sub>0.9</sub>Gd<sub>0.1</sub>O<sub>2</sub>, crystal structure mixtures of the powders were revealed from the XRD patterns corresponding to 1800°C. As shown in Fig. 1, the XRD patterns corresponding to the synthetic trivalent cation doped uranium oxide pellet at 1800°C.

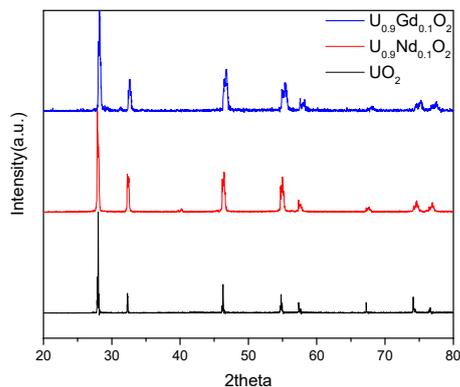


Fig. 1. In-situ high temperature powder X-ray diffraction patterns of the  $\text{UO}_2$ ,  $\text{U}_{0.9}\text{Nd}_{0.1}\text{O}_2$ , and  $\text{U}_{0.9}\text{Gd}_{0.1}\text{O}_2$  phases heated at  $1800^\circ\text{C}$  temperatures.

#### 4. Conclusions

The trivalent cation doped uranium oxide phases have been studied in situ X-ray diffraction with temperature equipment. We measured the effect of the temperature on the cell parameter using calculation method. In situ high temperature X-ray diffraction observed that structural change in materials. In further works, to follow various effects on the structure performances of  $\text{UO}_2$  depending on the doped content in the matrix with in situ high temperature X-ray diffraction.

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