Study of Metal doped Uranium Oxide Using In-Situ High Temperature X-ray Diffraction

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1. Introduction

Powder X-ray diffraction analysis has been widely used in the material development industry, such as the confirmation of material constituents and structural changes in materials. [1]

Among many research fields, structural modification and compositional analysis of uranium oxide in the nuclear fuel field, have also used through powder X-ray analysis. [2]

Uranium oxide materials containing U^{4+}, U^{5+}, and U^{6+} cations have been of especial interest in broad nuclear fuel fields because of the structural changes derived from various oxidation numbers.

In addition, rare-earth metal doped uranium oxide materials have drawn enormous attention because of their applicable characteristics such as semiconductors and catalysts. [3-6]

Therefore, research of the structural form of rare-earth metal doped uranium oxide is important to understand their characterizations.

Because of the specificity of metal doped uranium oxide, the crystal structure is measured using neutron diffraction and X-ray diffraction after synthesis experiments of materials.

Moreover, 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.

Prieur`s group has researched the Am doped UO_2 with in situ XRD at 25°C - 1300°C. The results showed the evolution of the lattice parameter, the crystallite size, and the strain up to 1300 °C. [6]

In this talk, we report the study of metal (Zr^{4+}, Gd^{3+}, and Nd^{3+}) 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 sintering process. And then, the change of lattice parameters are calculated using Rietveld refinement method.

2. Experimental

Metal (Zr^{4+}, Gd^{3+}, and Nd^{3+}) 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 1400 °C.

The X-ray powder diffraction patterns were collected on a Bruker D8-Advance diffractometer using Cu Kα radiation with 40 kV and 40 mA and an Anton Parr HTK 2000 heating chamber. The scan ranges were 20-110° with a step size of 0.02°, and a step time of 0.1s. The temperature ranges were 25 °C - 1400 °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_2 [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 metal (Zr^{4+}, Gd^{3+}, and Nd^{3+}) doped uranium oxide pellets at various temperatures x-ray diffraction patterns are shown in Fig. 1. At room temperature, the products were confirmed as UO_2, Gd_2O_3, ZrO_2, and Nd_2O_3 peaks showed in the XRD patterns, respectively.

Metal doped UO_2 crystal structures were retained after heating at 1300°C because the XRD patterns did not show any obvious differences compared with that corresponding from the R.T. However, the crystal structure changes of the powders were revealed from the XRD patterns corresponding to 1400°C. As shown in Fig. 1, the XRD patterns corresponding to the synthetic metal doped uranium oxide pellet at 1400°C.

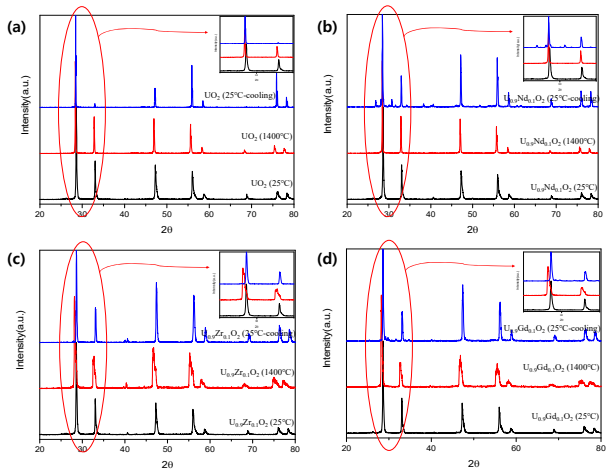

Fig. 1. In-situ high temperature powder X-ray diffraction patterns of the UO_2 and $U_{0.9}M_{0.1}O_{2-x}$ ($M=Zr^{4+}$, Nd^{3+}, and Gd^{3+}) phases heated at different temperatures. The results showed that the UO_2, and metal oxides crystal structures formed by heating metal doped uranium oxide pellet at 1400°C. (Insert: Powder X-ray diffraction data for results. The peak positons shift toward the left side.)

As seen in Fig. 1(insert), the diffraction peaks move to the left with increasing temperatures. The peak positions slightly shift toward the left side as temperature increase.

4. Conclusions

The Metal 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 material. In further works, to follow various effects on the properties of UO_2 depending on the doped content in the matrix with in situ high temperature X-ray diffraction.

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