In-situ High Temperature X-ray Diffraction for Structural Transition of Uranium Oxide

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1. Introduction

After nuclear power generation, amount of spent nuclear fuel is steadily increased, researchers have been increasingly interested in methods of disposal spent nuclear fuel. Various fission products such as Cs, Sr, Np and Am are generated and existed in the spent nuclear fuel. [1] Therefore, it is very important to structural changes according to temperature because various compounds between nuclear fuel and fission products can exist by heat generated from nuclear fuel. We are observed the structural behavior between UO_2 and fission product in spent nuclear fuel depending on temperature using powder x-ray diffraction. For the confirmation of spent fuel components and structural formations, powder x-ray diffraction (XRD) analysis has been usually used in the material research fields.

Especially, 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_2, U_4O_9, U_3O_8 and UO_3. [2]

In general, X-ray diffraction is too difficult to measure while controlling the temperature, we can the temperature by adding a special device, and acquire the powder diffraction pattern at the controlled temperature to check the structural change in real time.

Recently, for transmutation study of nuclear fuel, Lebreton`s group has researched the Am doped UO_2 with in situ X-ray diffraction at ~1973K. The results showed the performance of the lattice parameter, the crystallite size, and the strain up to ~1973K. [3]

In this work, we report the structure behaviors study of uranium oxide by different thickness 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 is calculated by Pawley method.

2. Experimental

Uranium oxide pellets were ground thoroughly with agate mortar and pestle and pressed into

pellets. The different thickness of 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 Kα 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 Pawley 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].

3. Results and discussion

The x-ray diffraction patterns of uranium oxide 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 temperature of UO_2. As shown in Fig. 1, the XRD patterns corresponding to the different temperature of uranium oxide pellet at 1800°C.

4. Conclusions

The 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 from cell parameter calculation and powder patterns. In further works, to follow various effects on the structure performances of UO_2 depending on the existed fission products in the matrix with in situ high temperature X-ray diffraction.

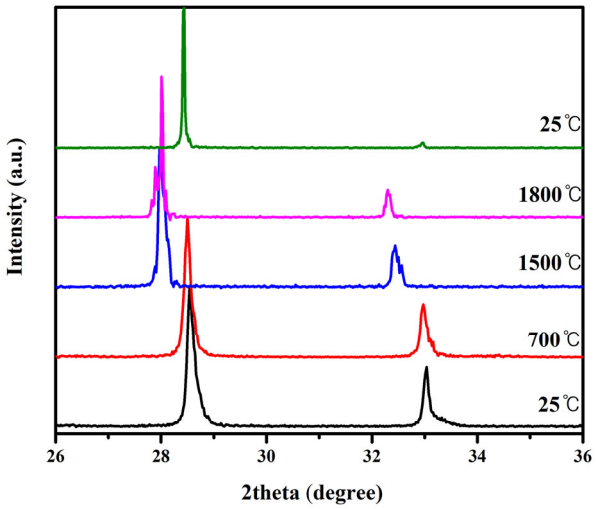

Fig. 1. In-situ high temperature powder X-ray diffraction patterns of the UO_2 phases heated at ~ 1800°C temperatures.

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REFERENCES

[1] J. Bruno, R. C. Ewing, Spent Nuclear Fuel. *Elements*, *2*, 343-349, 2006.

[2] D. A. Andersson, G. Baldinozzi, L. Desgranges, D. R. Conradson, S. D. Conradson, Density Functional Theory Calculations of UO_2 Oxidation: Evolution of UO_{2+x}, U_4O_{9-y}, U_3O_7, and U_3O_8. *Inorganic Chemistry*, *52*, 2769-2778, 2013.

[3] F. Lebreton, R. C. Belin, D. Prieur, T. Delahaye, and P. Blanchart, In Situ Study of the Solid-State Formation of $U_{1-x}Am_xO_{2\pm\delta}$. *Inorganic chemistry*, *51*, 9369-9375, 2012.