In situ High Temperature XRD Study of Uranium Oxide.

Dong Woo Lee [a], Jeongmook Lee [a], Jong-Yun Kim [a,b] and Sang Ho Lim [a,b,*]
[a]*Nuclear Chemistry Research Team, Korea Atomic Energy Research Institute, 111, Daedeok-daero 989 Beon-gil, Yuseong-gu, Daejeon, 34057, Republic of Korea*
[b]*Department of Radiochemistry & Nuclear Nonproliferation, University of Science and Technology, Gajeong-ro 217, Yuseong-gu, Daejeon, 34113, Republic of Korea*
**Corresponding author: slim@kaeri.re.kr*

1. Introduction

Researches of uranium oxides as nuclear fuels have been consistently investigate. As one of them, various types of uranium oxide structure are observed by effect of coordination environments of uranium cation such as UO_2, U_3O_8 and U_4O_9 [1]. Because of the specificity of uranium oxide, the crystal structure is measured using neutron diffraction and x-ray diffraction after sintering experiments [2]. However, to get the experimental results are in general time-consuming or not straightforward.

In-situ x-ray diffraction provides the needed crystal structure information. In addition, x-ray diffraction with controlled temperature, which is analyzed and synthesized at the same time.

Recently, Abril et el. [3] have studied the UO_2 nanoparticles with in situ x-ray diffraction at 30°C - 1200°C. The results showed the evolution of the crystallite size, the lattice parameter, and the strain up to 1200 °C.

In this work, we report the in situ high temperature XRD study of uranium oxide particles. We measure the effect of temperature on the size if crystalline, which is a cell parameter in the sintering process. And then, the variation of lattice parameter are calculated using Rietveld refinement method.

2. Experimental

Uranium oxide powders were ground thoroughly with agate mortars and pestles and pressed into pellet. The pellet on the tungsten strip were evacuated and gradually heated to 1300 °C

The X-ray powder diffraction data 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 - 1300 °C. The diffraction patterns were analyzed using Rietveld method with the TOPAS program [4]. 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 uranium oxide pellet at various temperatures x-ray diffraction patterns are shown in Fig. 1. At room temperature, the powders were confirmed as UO_2 since the (111) and (200) UO_2 characteristic peaks showed in the XRD pattern. And the UO_2 crystal structure was retained after heating at 700°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 800°C - 1300°C. As shown in Fig. 1, the XRD patterns corresponding to the synthetic uranium oxide pellet at 800 °C - 1300 °C are in agreement with that of $U_4O_{9.58}$ (PDF#: 01-074-4168)

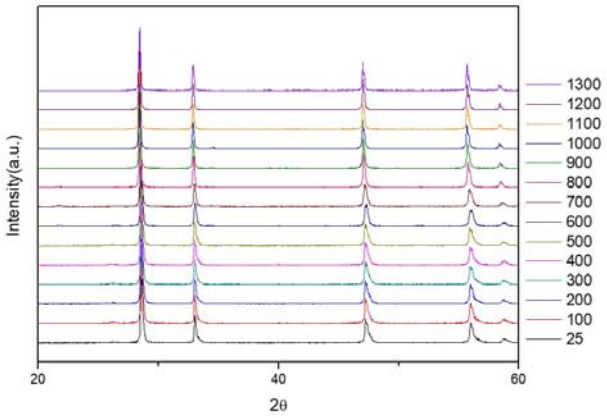

Fig. 1. In-situ high temperature powder X-ray diffraction patterns of the powder heated at different temperatures. The results showed that the UO_2, and $U_4O_{9.58}$ crystal structures formed by heating UO_2 pellet at 25°C - 700°C, and 800°C - 1300°C, respectively.

As seen in Fig. 2, the diffraction peaks move to the left with increasing temperatures. The peak positions slightly shift toward the left-hand side as temperature increase.

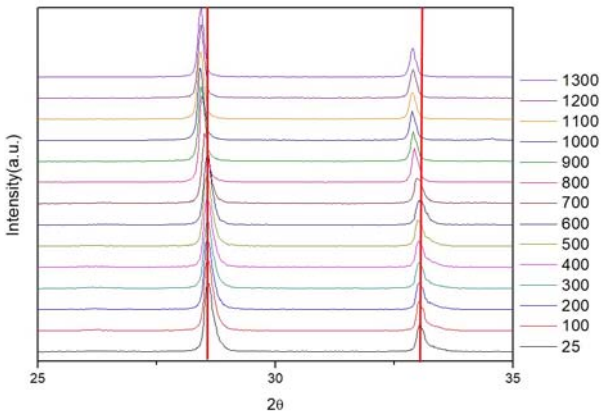

Fig. 2. Powder X-ray diffraction data for uranium oxide. The peak positons shift toward the left-hand side.

4. Conclusions

The UO_2 phases have been studied in situ x-ray diffraction with temperature device. We analyzed the effect of the temperature on the lattice parameter using calculation method. In situ high temperature x-ray diffraction showed that structural change in material. Further studies are planned, 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.

ACKNOWLEDGEMENT

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korean government (MSIP; No.2012M2A8A5025925).

REFERENCES

[1] R. J. McEachern, P. Taylor, A review of the oxidation of uranium dioxide at temperatures below 400°C, Journal of Nuclear Materials, Vol. 254, p.87, 1998.
[2] L. Desgranges, G. Baldinozzi, G. Rousseau, J-C Niepce, G. Calcarin, Neutron Diffraction Study of the in Situ Oxidation of UO_2, Inorganic Chemistry, Vol. 48, p. 7585, 2009.
[3] R. J. Abril, R. Eloirdi, D. Bouexiere, R Malmbeck, J. Spino, In situ high temperature X-ray diffraction study of UO_2 nanoparticles, Journal of Materials Science, Vol. 46, p. 7247. 2011.
[4] A. A. Coelhoa, TOPAS and TOPAS-Academic: an optimization program integrating computer algebra and crystallographic objects written in C^{++} Journal of Applied Crystallography. Vo. l51, p.210, 2013.