

Observation of UO_2 Powder Oxidation by X-ray Diffraction

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1. Introduction

In dry air, the oxidation of uranium dioxide pellets can be described as a two-step process ($\text{UO}_2 \rightarrow \text{U}_4\text{O}_9/\text{U}_3\text{O}_7 \rightarrow \text{U}_3\text{O}_8$). The principal oxidant is O_2 in air. The density of U_3O_8 is 23% less than that of UO_2 , which corresponds to a 36% net volume increase of oxidized pellets. As a result, the formation of U_3O_8 leads to fuel pellet powdering. This volume change by oxidation heavily affects the safety management for the long-time storage and disposal of spent fuel and the integrity of the fuel.

The oxidation kinetics of UO_2 powders is essential for evaluating long-time integrity maintenance when the dry storage of spent fuel be degraded.

In this study, the oxidation kinetics of UO_2 powder was analyzed using X-ray diffraction (XRD) systems.

2. Methods and Results

In this section, the oxidation experiments for UO_2 powder using XRD systems are described in detail.

2.1 Experimental Apparatus

Products were analyzed using the powder X-ray diffraction method with a Rigaku Type RINT 2000 diffractometer and a Ni filtered Cu $K\alpha$ irradiation (40 kV and 40 mA) equipped with a curved pyrolytic carbon. The phases of the products were then identified by comparing the obtained diffraction patterns with those from the JCPDS data.



Fig. 1. Rigaku Ultima IV

2.2 Experimental Procedures

Procedures for the oxidation experiment with UO_2 powder is performed as follows: to inject nitrogen in order to prevent after that to proceed with the experiment was to measure the exact weight of the UO_2 powder, from being oxidized while raising the temperature to put the XRD the UO_2 powder. Upon

reaching the desired temperature, 100 Nitrogen: starting the XRD measurement by injecting dry air at a rate of oxygen 25.

2.3 Experimental Results

The amount of time required will vary depending on the temperature U_3O_8 peaks can be confirmed by XRD graph to be increased more and more, a decrease of more UO_2 peaks. As can be seen in the following diagram, as the change oxidation progresses UO_2 peak appears is displayed peaks U_3O_8 gradually at first.

2.3.1 Oxidation Experiment at 300 °C

The results on the oxidation experiments with UO_2 powder using XRD systems are shown in Figure 2 below.

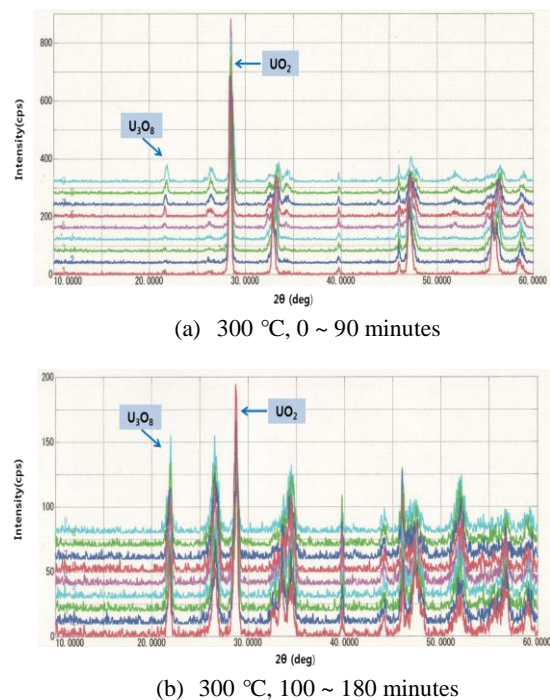


Fig. 2. X-ray diffraction peaks of UO_2 and U_3O_8 —(a) 300 °C, 0 ~ 90 minutes, and (b) 300 °C, 100 ~ 180 minutes—resulting from the oxidation experiment at 300 °C.

2.3.2 Oxidation Experiment at 350 °C

The results of the oxidation experiments with UO_2 powder using XRD systems are shown in the figure below.

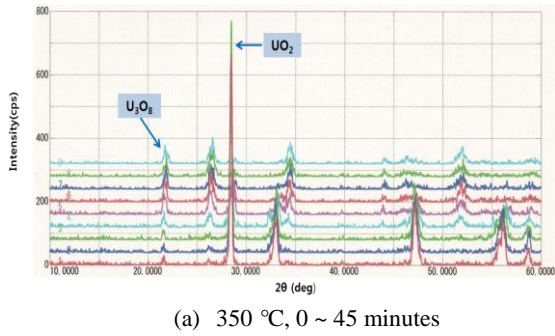


Fig. 3. X-ray diffraction peaks of UO_2 and U_3O_8 —(a) 350 °C, 0 ~ 45 minutes—resulting from the oxidation experiment at 350 °C.

2.3.3 Oxidation Experiment at 400 °C

The results of the oxidation experiments with UO_2 powder using XRD systems are shown in the figure below.

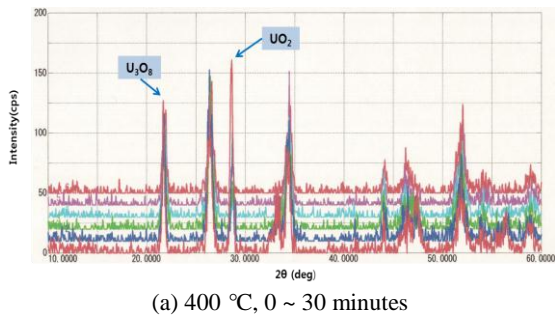


Fig. 4. X-ray diffraction peaks of UO_2 and U_3O_8 —(a) 400 °C, 0 ~ 30 minutes—resulting from the oxidation experiment at 400 °C.

2.4 Analysis of Oxidation Experiment

The results of the analysis of the oxidation experiments according to temperature can be explained as follows: Fig. 5 is a graph showing the percentage of U_3O_8/UO_2 over time through the oxidation experiments at various temperatures.

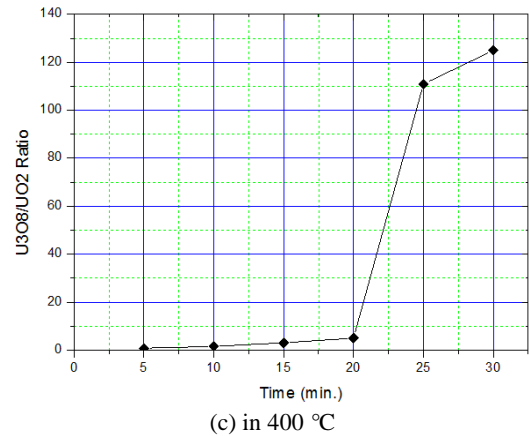
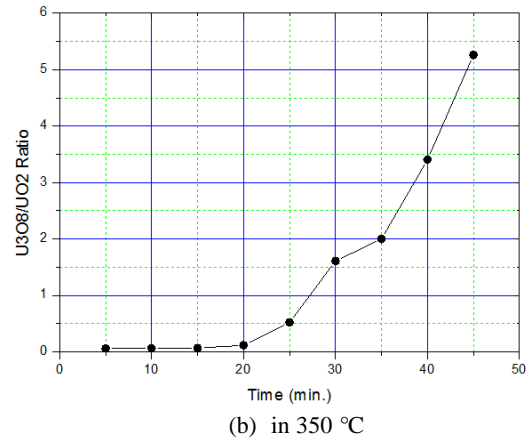
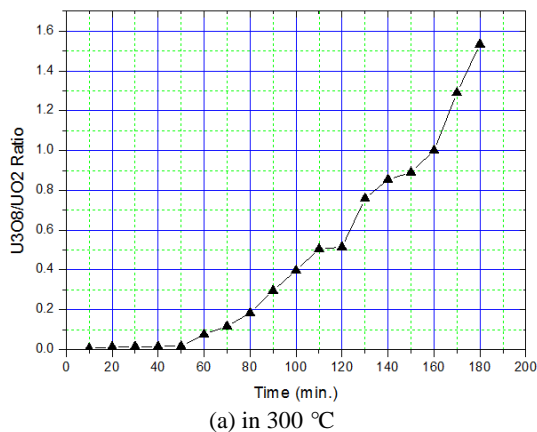


Fig. 5. Plot of experimental time vs. U_3O_8/UO_2 fraction based on the oxidation experiment (a) at 300 °C, (b) at 350 °C, and (c) at 400 °C.

3. Conclusions

In this paper, oxidation experiments with UO_2 powder using XRD systems were performed. The results confirm that the UO_2 oxidation proceeds and changes to U_3O_8 . Furthermore, Figure 5 shows that as the temperature increases, the oxidation rate increases, and the oxidation is produced more rapidly in U_3O_8 from UO_2 .

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