Accelerated Oxidation of Uranium Dioxide in Oxygen Plasma

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1. Introduction

Research and development (R&D) on the advanced nuclear fuel forms one of the most essential parts in the domestic industrialization and upgradation of nuclear technology in Korea along with R&D on next generation nuclear reactor In fact, the foreign pioneers in the nuclear fuel industry are attempting to introduce new technologies in the manufacturing process to improve the process and to enhance the fuel reliability. Plasma processing is one of new technologies in fuel manufacturing. As one of introductory research activities of new technologies a study on the accelerated oxidation by plasma gas is carried out. The objective of this study is to estimate the acceleration of uranium dioxide by oxygen plasma and to determine the mechanism of the acceleration process. And this oxidation technique is expected to applied to waste treatment such as spent resin produced in nuclear power plant and dry treatment of spent fuel.

2. Experimetal

Plasma reactor is a diode type and r.f. (13.56 MHz) power up to 600 W can be applied between the parallel electrodes. The distance between them remains 10 cm during the current experiments. Sample can be heated up to 1200℃ electrical heater inside the reaction chamber. Mass flow controller can control finely and flow rate remains 55sccm. Total gas pressure inside chamber is maintained at around 1 Torr during experiments.

2.1 Specimen Preparation

In experiments, thin disk type of natural uranium dioxide cut out of pellet using low-speed diamond cutter. Prior to the sample loading, the surfaces of the specimens are polished by grit 600 sand paper, cleaned by acetone and baked at 200°C for 10 minutes in a vacuum to evaporate the absorbed moisture on the surface. Reaction rate is determined by intermittent weight gain measurement.

2.2 Experimental Procedure

We remained as 50W in RF power, 1 Torr in pressure, at all experiments. First of all, oxidation rate of uranium dioxide is determined by intermittent weight gain measurement with an electro-micro balance (BP210D, Satorius) whose sensitivity limit is 10^{-5}g. Along with the oxidation measurement, OES (Optical Emission Spectroscopy) analysis is accompanied with the main experiments to diagnose and determine the plasma parameters and thus to understand the reaction mechanism.

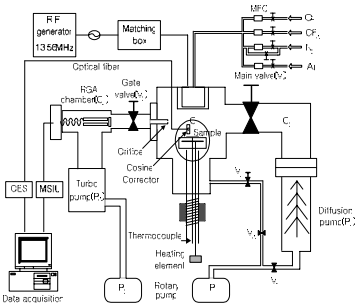

Figure 1. Schematic of plasma oxidation apparatus

3. Result and discussion

Figure 2. shows that oxidation of uranium dioxide in oxygen plasma enhanced about three times compared to that oxygen gas reaction at the same temperature and it is found that the reaction rate increase linearly with reaction time, which is in good agreement with earlier works.

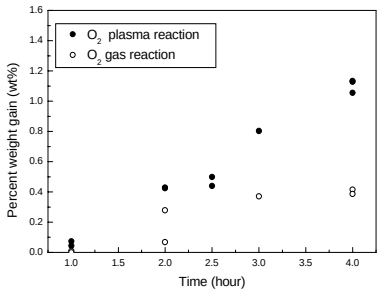

Figure 2. Oxidation of Uranium dioxide at 400℃

In order to find the more effective reaction rate at given temperature, reaction rates examined as mixture of oxygen gas and inert gas. For example, O_2 / Ar , O_2 / N_2, O_2 / He. gas mixture.

In fact, Oxygen radicals produced in the plasma play very significant role in this oxidation reaction. Thus, intensities of the oxygen radicals generated in the plasma are measured and analyzed by optical emission spectroscopy (OES) during experiment.

Figure 3,4,5. show OES diagnostics result of O_2 / Ar, O_2 /N_2, O_2 / He gas Plasma. These results show that intensities of oxygen in O_2 / N_2 gas plasma are increased about five times than in O_2 / Ar gas plasma. And intensities of oxygen in O_2 / He gas plasma are increased about seven times than in O_2 / Ar gas plasma.

Thus, We can anticipate that oxidation rate is most effective in O_2 / He plasma. At present, it is achieving experiments to verify this anticipation.

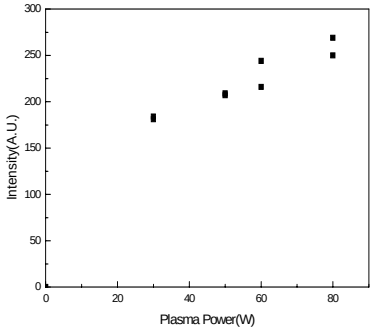

Fig. 3. Variation of Oxygen Intensity as a function of Power in O_2 / Ar Plasma

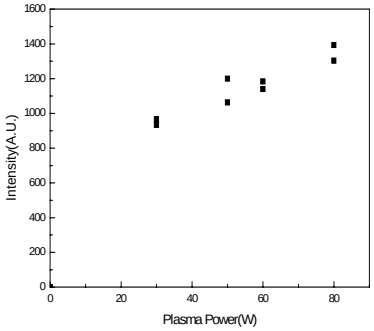

Fig. 4. Variation of Oxygen Intensity as a function of Power in O_2 / N_2 Plasma

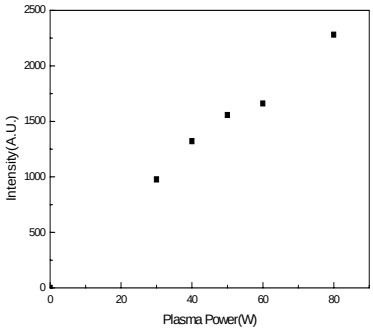

Fig. 5. Variation of Oxygen Intensity as a function of Power in O_2 / He Plasma

4. Conclusion

Experimental results confirmed that oxidation of UO_2 are enhanced about 70~300% in oxygen plasma reaction than oxygen gas reaction. Experimental finding that higher plasma power brings to higher oxidation rate suggest that the acceleration may be ascribed to the increased oxygen radical which is produced by plasma reaction.

Through OES diagnostics, It is expected to has more effective reaction rate in O_2 / He gas plasma.

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