8-3 Study on the Denitration Process of Uranyl Nitrate Solution by Microwave Heating
—Uranium Powder Production Process from Uranium Nitrate Solution—

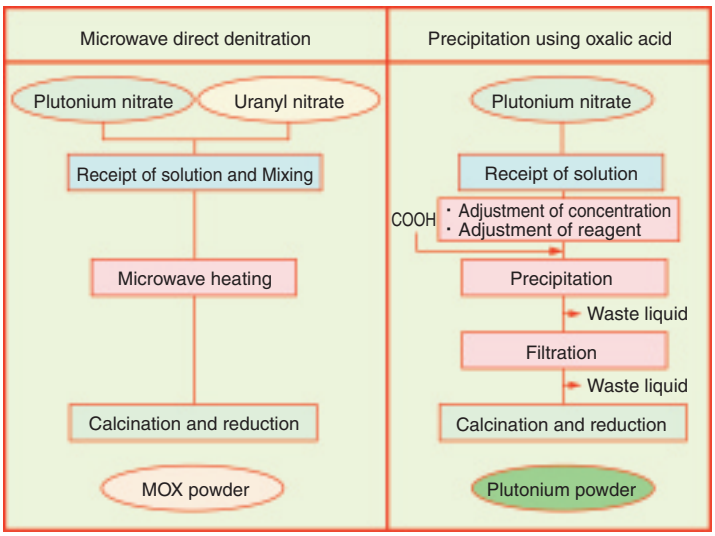

Fig.8-2 Comparison of conversion methods
Microwave denitration method was developed in Japan to directly produce MOX powder. There are three major merits compared to the conventional methods such as precipitation: 1) the process is simple and compact, 2) the produced powder has high homogeneity because plutonium and uranium are mixed in solution state and 3) volume of waste solution is extremely small because no reagent is used.

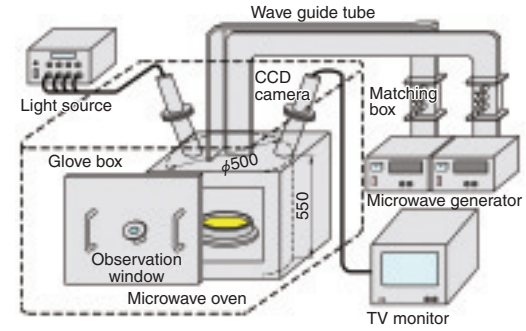

Fig.8-3 Schematic view of denitration testing apparatus
Microwave power is 3 kW.

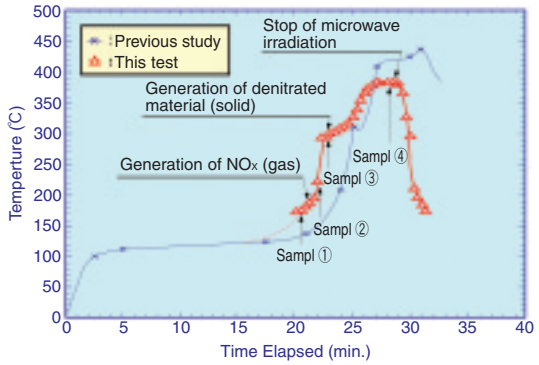

Fig.8-4 Temperature of uranium compounds during microwave irradiation
Crystallization water and nitrate start to be decomposed about 20 minutes after the start of irradiation, and finally only uranium oxide remains.

Spent fuel from nuclear power reactor is chemically treated in reprocessing plants, and then plutonium and uranium are recovered as reusable sources. In Europe, recovered plutonium is converted to the oxide powder by precipitation using oxalic acid and mixed with uranium oxide powder, followed by the mixed oxide (MOX) fuel fabrication. A new co–conversion method where plutonium and uranium are converted directly to MOX powder after mixing them in solution state was developed and adopted in Japan (Fig.8-2). This process has the feature of nuclear weapons proliferation resistance because there is no pure plutonium oxide powder in the process. This feature is exactly consistent with the policy of Japan that nuclear power should be used only for peaceful purposes. The microwave denitration method excites molecules by microwaves just like a household microwave oven, and nitrate is decomposed more rapidly by internal heating than by external heating. As a result, the produced powder is made up of fine particles which are suitable for sintering into high density pellets.

When mixed plutonium nitrate and uranium nitrate solution is heated, water and nitric acid evaporates at the first stage, then crystallization water of nitrates and nitrate itself are decomposed, and MOX remains at the end. In this study, we developed a special device to measure non-invasively and selectively the temperature of uranium material regardless of the moisture or NOx gas in the oven (Fig.8-3). The purpose of the development was that the conventional infrared thermometer could not measure temperature correctly because infrared rays were absorbed by moisture or NOx gas between the measured substance and the thermometer. The device was attached to the oven and temperature transition of the material was accurately measured. Then, samples were taken from each stage of reactions and thermometric analysis and X–ray analysis were carried out (Fig.8-4). As a result, we could confirm the chemical reactions formulae shown below :

$UO_2(NO_3)_2 \cdot 6H_2O \rightarrow UO_2(NO_3)_2 \cdot 3H_2O + 3H_2O$

$UO_2(NO_3)_2 \cdot 3H_2O \rightarrow UO_2(OH)NO_3 + HNO_3 + H_2O$

$UO_2(OH)NO_3 \rightarrow \beta\text{-}UO_3 + 0.5H_2O + NO_2 + 0.25O_2$

And/or $UO_2(OH)NO_3 \rightarrow \beta\text{-}UO_3 + HNO_3$

Reference

Kato, Y. et al., Reaction Mechanism of De-nitration of $UO_2(NO_3)_2$ by Microwave Heating, Nippon Genshiryoku Gakkai Wabun Ronbunshi, vol.4, no.1, 2005, p.77-83 (in Japanese).