

# Development of Pyrochemical Reprocessing for Spent LWR Fuels – To Produce Refined Uranium Metal from Simulated Spent Oxide Fuel –

## Background

The metal fuel cycle, consisting of metal fuel fast breeder reactors (FBRs) and pyrochemical reprocessing is a promising option that satisfies the requirements of economic advantage, environmental safety and high proliferation resistance. To supply metal fuel material to FBRs, application of pyrochemical reprocessing to spent light water reactor (LWR) fuels has been investigated. The process flow sheet schematically shown in Fig. 1 consists of pretreatment, electrolytic reduction and electrorefining. In previous studies on the electrolytic reduction,<sup>1,2</sup> CRIEPI demonstrated the feasibility of reducing  $\text{UO}_2$  and MOX ( $\text{UO}_2\text{-PuO}_2$ ) to metallic form and characterized optimum electrolysis conditions and reduction mechanisms.

## Objectives

The purpose of this study is to produce refined uranium metal from simulated spent oxide fuel through the pretreatment, electrolytic reduction and electrorefining processes by using electrolyzers suitable for practical use.

## Principal Results

### 1. Pretreatment

The starting material of simulated spent oxide fuel was oxidized in an air stream at  $1000^\circ\text{C}$  to give  $\text{U}_3\text{O}_8$  powder containing simulated fission product elements (FPs), assuming the fuel decladding by voloxidation. The oxide powder was compacted into green pellets and then sintered in a  $\text{N}_2\text{-13\%H}_2$  gas stream at  $1700^\circ\text{C}$  when  $\text{U}_3\text{O}_8$  was reduced to  $\text{UO}_2$ . As a result, porous  $\text{UO}_2$  pellets containing FPs were obtained as shown in Fig. 2.

### 2. Electrolytic reduction

About 100 g of the porous oxide pellets were charged in a cathode basket (Fig. 3(a)) and electrolysis was carried out for 7.8 h in a molten  $\text{LiCl-Li}_2\text{O}$  electrolyte at  $650^\circ\text{C}$ . The  $\text{UO}_2$  was converted into metallic uranium as shown in Fig. 3(b). It was verified that preparing the porous pellets was of great advantage to the electrolytic reduction process. The oxide reduction is facilitated because molten salt easily permeates inside the pellet and oxide ions diffuse outside, and the reduction rate is independent of the amount of pellets charged in the cathode. In addition, pellets can be handled much easier than powders.

### 3. Electrorefining

The reduction product was charged in an anode basket and electrolysis was carried out for 6.0 h in a molten  $\text{LiCl-KCl-UCl}_3$  electrolyte at  $500^\circ\text{C}$ . Most of the uranium metal was anodically dissolved into the electrolyte (Fig. 4(a)) and dendritic uranium metal with high purity was harvested on a stainless-steel cathode (Fig. 4(b)). FPs were separated from the uranium metal product as expected.

### 4. Applicability of the pyrochemical reprocessing to LWR fuels

The oxide reduction was successfully accomplished within 10 h that was the target decided as a practical throughput speed, and then refined uranium metal was collected with a high degree of efficiency. There might not be any essential problems to scale up the method. Consequently, it was recognized from the engineering point of view that pyrochemical reprocessing could be applicable to spent LWR fuels.

The present work is the result of “Application of electrochemical reduction to pyrochemical reprocessing for oxide nuclear fuel”, entrusted to CRIEPI by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

## Future Developments

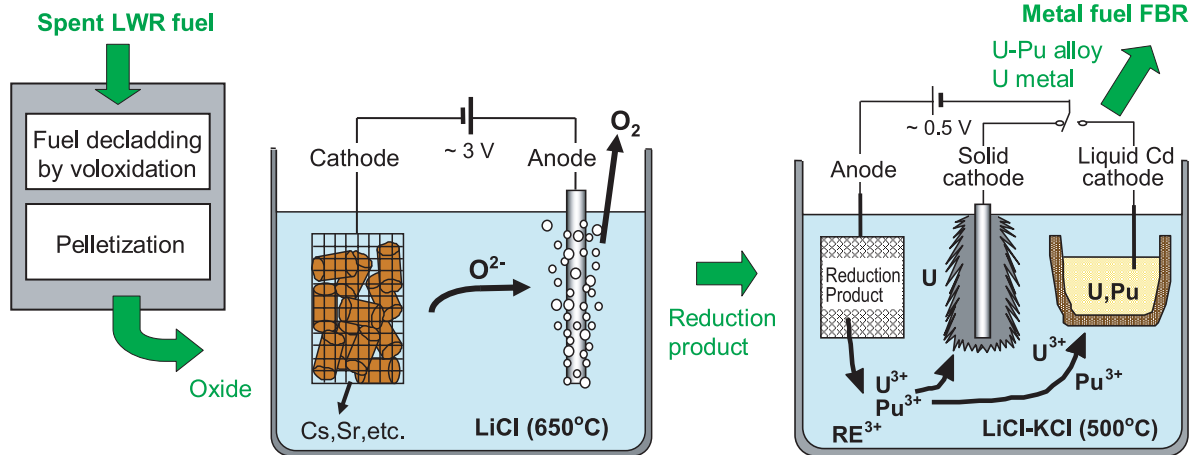
Full-scale  $\text{UO}_2$  reduction tests with 5-10 kg/batch will be demonstrated and electrode configuration investigated in order to improve the throughput and current efficiency even more.

**Main Researcher:** Yoshiharu Sakamura,

Senior Research Scientist, Advanced Nuclear Fuel Cycle Sector, Nuclear Technology Research Laboratory

## Reference

- 1 Y. Sakamura, “Development of Electrochemical Reduction Process for Oxide Nuclear Fuel – To Integrate Metal Fuel FBR Cycle with LWR Cycle –”, CRIEPI Annual Research Report 2006.
- 2 Y. Sakamura, M. Kurata and T. Inoue, “Electrochemical Reduction of  $\text{UO}_2$  in Molten  $\text{CaCl}_2$  or  $\text{LiCl}$ ”, J. Electrochem. Soc., 153, D31 (2006).



(1) Pretreatment  
Decladding and simple pelletization

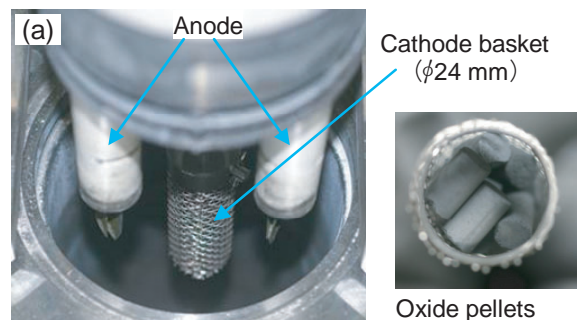
(2) Electrolytic reduction  
At the cathode, oxygen is ionized to dissolve in molten LiCl, and then U and Pu metals remain. O<sub>2</sub> discharges to form O<sub>2</sub> gas at the anode.

(3) Electrorefining  
At the anode, U and Pu metals dissolve into molten LiCl-KCl. U metal is collected on the solid cathode, and Pu metal accompanied by U is collected in the liquid cadmium cathode.

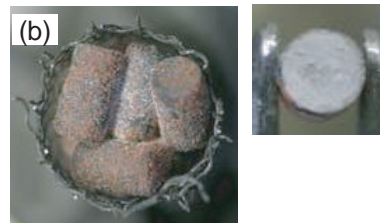
**Fig. 1** Pyrochemical reprocessing for spent LWR fuels



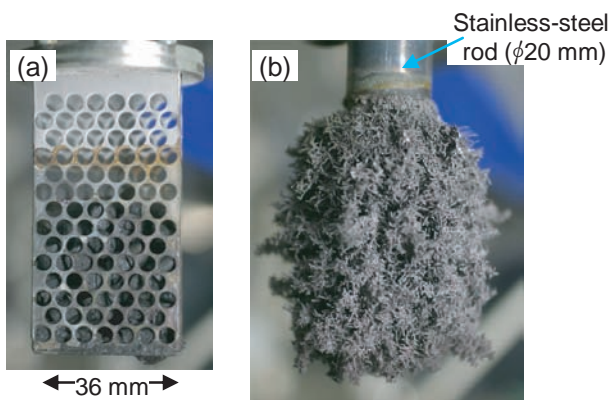
**Fig. 2** Oxide pellets with 32% of porosity prepared by pretreatment process: UO<sub>2</sub> containing 1 wt% of each simulated FP (Ce, Nd, Sm, Sr, Zr, Mo and Pd).



Electrolysis (7.8 h)



**Fig. 3** Results of electrolytic reduction test: (a) Before electrolysis, the electrodes were placed in the electrolyzer. The cathode basket with 101 g of the porous pellets was surrounded by three platinum anodes. (b) Inside of the cathode basket and typical cross-sectional pellet after electrolysis: UO<sub>2</sub> was reduced to metallic uranium.



**Fig. 4** Results of electrorefining test: uranium metal of the reduction product at the anode dissolved into the molten salt and deposited at the cathode. (a) Anode basket after electrolysis. (b) Dendritic uranium metal on solid cathode.