

## Principal Research Results

# Development of Electrochemical Reduction Process for Oxide Nuclear Fuel – To Integrate Metal Fuel FBR Cycle with LWR Cycle –

### Background

The metal fuel cycle, consisting of metal fuel FBR and pyrochemical reprocessing, is one of the promising options in the next generation nuclear fuel cycle technology. Since the LWR cycle is an oxide fuel technology, the oxide fuels of the LWR cycle have to be converted to metals for supplying them to the metal fuel FBR cycle. The electrochemical reduction process described in Fig.1 is expected to be the most efficient oxide reduction technique.

### Objectives

To characterize appropriate electrolysis conditions for  $\text{UO}_2$  reduction; to investigate parameters for designing engineering scale equipments; to demonstrate the production of uranium metal from  $\text{UO}_2$  through the electrochemical reduction process.

### Principal Results

#### 1. Appropriate electrolysis conditions

$\text{UO}_2$  reduction tests were conducted in  $\text{LiCl}$  (650°C),  $\text{LiCl-KCl}$  eutectic (600°C: low melting point) and  $\text{CaCl}_2$  (820°C: strong affinity for oxygen) salt baths. As shown in Fig.2, the  $\text{UO}_2$  sample was completely reduced to porous uranium metal in  $\text{LiCl}$ . But in  $\text{LiCl-KCl}$  eutectic and in  $\text{CaCl}_2$ , only the surface layer was reduced to the metal and the  $\text{UO}_2$  remained inside. It was suggested that the reduction rate should be determined by the transportation of oxide ion ( $\text{O}^{2-}$ ) from the inside to the bulk salt and that the affinity for oxygen was ineffective.<sup>1</sup>

The reduction will cause a gap formation in the  $\text{UO}_2$  sample because the density of  $\text{UO}_2$  is about 60% as much as that of uranium metal. Then the molten salt will permeate into the gap and  $\text{O}^{2-}$  will be discharged to the bulk salt through the molten salt pass. In  $\text{LiCl-KCl}$  eutectic and in  $\text{CaCl}_2$ , the molten salt pass was possibly closed with by-products (*i.e.*,  $\text{Li}_2\text{O}$  or uranium metal) and then the reduction did not progress inside.

#### 2. Production of uranium metal from $\text{UO}_2$

$\text{LiCl}$  was employed as the salt bath and electrochemical properties such as reduction potentials of  $\text{UO}_2$  and  $\text{LiCl}$  (Fig.3), anode reactions and current efficiency were clarified.<sup>1</sup> Then, it was demonstrated in the 100g-scale  $\text{UO}_2$  reduction under optimum conditions followed by the high temperature salt distillation that the uranium metal ingot free from  $\text{LiCl}$  could be produced from the  $\text{UO}_2$  feed with high efficiency, as shown in Fig. 4.<sup>2</sup>

It was concluded that practical electrochemical reduction equipments could be designed on the basis of the experimental results, where 5-10 kg of oxide fuel loaded in a cathode basket could be reduced to metal within 10 hours.

This work was entrusted by the Ministry of Education, Culture, Sports, Science and Technology (MEXT).

### Future Developments

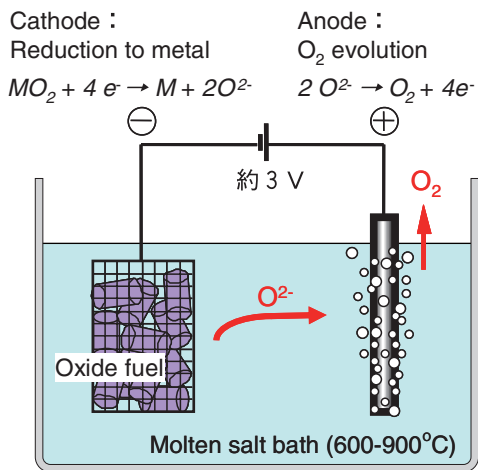
MOX ( $\text{PuO}_2\text{-UO}_2$ ) as well as  $\text{UO}_2$  was converted to metal in a small scale reduction test.<sup>3</sup> In future, more practical system will be investigated to demonstrate the process flow sheet.

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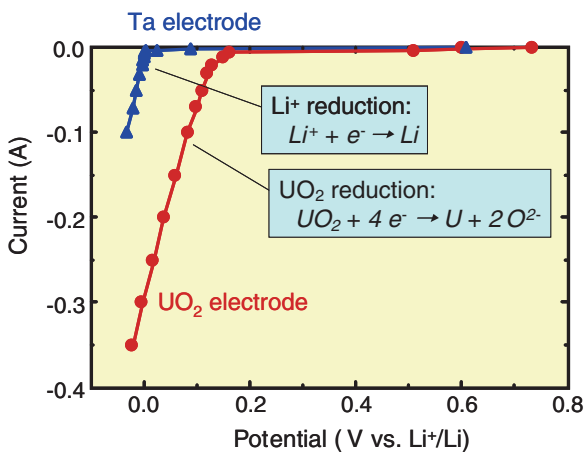
Senior Research Scientist, Advanced Nuclear Fuel Cycle Sector, Nuclear Technology Research Laboratory

### References

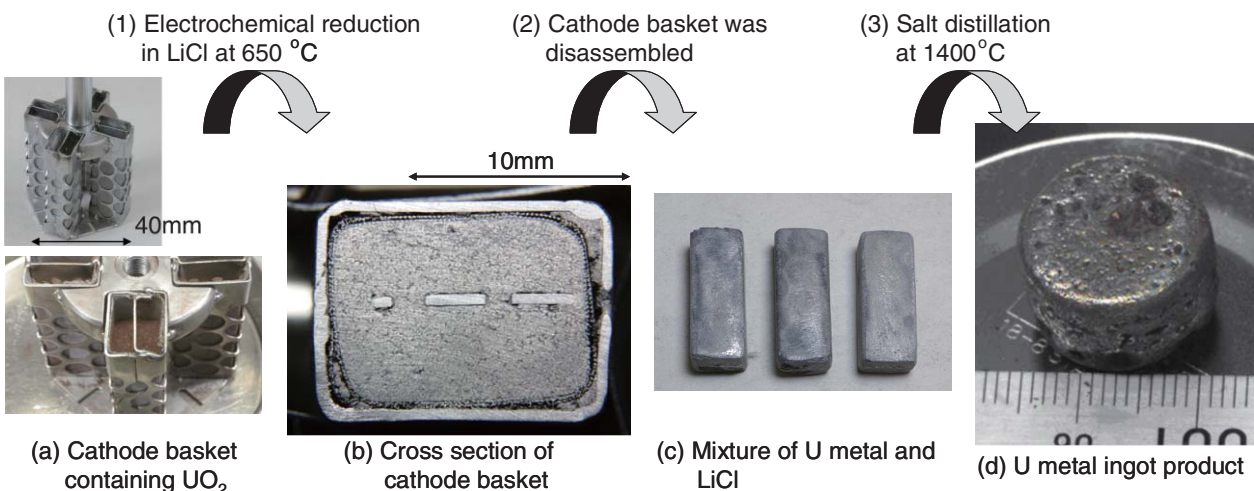
- 1 Y. Sakamura, M. Kurata and T. Inoue, "Electrochemical reduction of  $\text{UO}_2$  in molten  $\text{CaCl}_2$  or  $\text{LiCl}$ ", *J. Electrochem. Soc.*, 153 (3), D31-D39 (2006).
- 2 Y. Sakamura, T. Omori and T. Inoue, "Application of electrochemical reduction to produce metal fuel material from actinide oxides", submitted for publication in *Nucl. Technol.*
- 3 M. Iizuka, Y. Sakamura and T. Inoue, "Electrochemical reduction of  $(\text{U-40Pu-5Np})\text{O}_2$  in molten  $\text{LiCl}$  electrolyte", *J. Nucl. Mater.*, 359, 102-113 (2006).



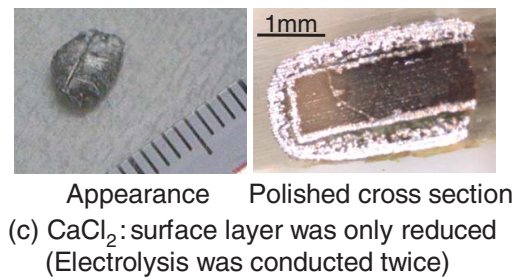
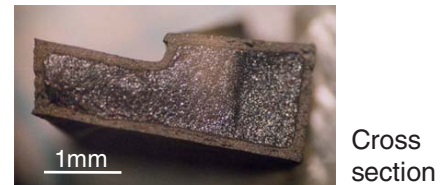
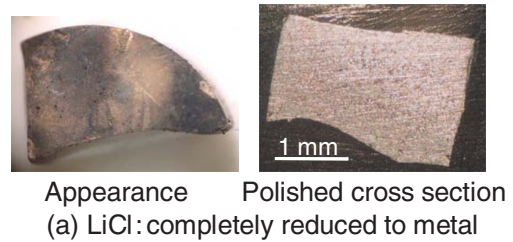
**Fig.1** Schematic diagram of electrochemical reduction process. During electrolysis, O<sup>2-</sup> is released into molten salt and actinide metal remains at the cathode. Oxygen gas is evolved at the anode



**Fig.3** Polarization curves of UO<sub>2</sub> and tantalum electrodes in LiCl at 650°C



**Fig.4** Production of U metal ingot from UO<sub>2</sub> through the sequence of electrochemical reduction and salt distillation (100g-scale test)



**Fig.2** UO<sub>2</sub> samples after electrochemical reduction in (a)LiCl, (b)LiCl-KCl eutectic and (c)CaCl<sub>2</sub> salt baths