Principal Research Results

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, CRIEPI demonstrated the feasibility of reducing UO2 and MOX (UO2-PuO2) 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°C to give U3O8 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 N2-13%H2 gas stream at 1700°C when U3O8 was reduced to UO2. As a result, porous UO2 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 LiCl-Li2O electrolyte at 650°C. The UO2 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 LiCl-KCl-UCl3 electrolyte at 500°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 UO2 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.

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Reference
5. Nuclear

Fig. 1 Pyrochemical reprocessing for spent LWR fuels

Fig. 2 Oxide pellets with 32% of porosity prepared by pretreatment process: UO₂ containing 1 wt% of each simulated FP (Ce, Nd, Sm, Sr, Zr, Mo and Pd).

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: UO₂ 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.