Background
Rokkasho reprocessing plant will start commercial operation within a few years. The R&D for the next step reprocessing is expected to be started after the stable operation of Rokkasho plant is achieved. Central Research Institute of Electric Power Industry has been developing pyro-reprocessing for metallic fuel since 1986 and, also, started R&D recently for a new pyro-processing that efficiently introduces heavy metals containing spent oxide fuels to metal fuel cycle. Based on preliminary studies using the pyro-metallurgical method shown in Fig.1 *, significant amount of U is estimated to be recovered from spent oxide fuels, and the form of U is confirmed to be purified UO₂.

Objectives
Based on various experimental studies, the process operation conditions and the process capability for the purification of UO₂ are elucidated.

Principal Results
1. Elucidation of anode dissolution condition of UO₂
Anode dissolution test of UO₂ was performed in various temperatures, U-concentrations in molten salt, and current density, etc. Consequently, the efficient dissolution of UO₂ from an anode to molten salt (>70%) was attained by controlling current density even for the case of powder or particles of UO₂, as shown in Fig.2.

2. Elucidation of cathode deposition condition of UO₂
Cathode deposition test of UO₂ was performed in various temperatures, U-concentrations in molten salt, and current density, etc. Consequently, UO₂ was recovered on a carbon cathode by electrolysis in 100% of efficiency. The most important thing to maintain the efficiency is to control the U-concentration at higher than approximately 8wt.%. In this condition, large size dense UO₂-particles can be collected on the cathode, as shown in Fig.3. By balancing the U-dissolution from the anode and the U-deposition on the cathode, the U-concentration in the molten salt can be maintained and, therefore, UO₂-product with stabilized quality can be recovered in the entire process.

3. Evaluation of decontamination factor of noble metals
In this process, there is a possibility of contaminating noble metals contained in spent fuels to recovered UO₂. The basic study for noble metals electrolysis was performed for their dissolution and deposition. The relationship between purity and recovery rate of UO₂ was evaluated as a function of current density. It was confirmed that degree of noble metal contamination is controlled by current density.

These results indicate that this process is feasible for the recovery of purified UO₂ from spent oxide fuels.

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Future Developments
The separation efficiency between U and noble metals will be confirmed by demonstration study. The electrode and crucible will be developed by large scale tests.

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Reference

C. Harmonization of energy and environment

(Fig. 1) Schematic image of UO$_2$-electrolysis process
(Spent oxide fuels are set on an anode and UO$_2$ is selectively dissolved by electrolysis. Some FP is dissolved in molten salt. High purified UO$_2$ is collected on a cathode.)

(Fig. 2) Progress in UO$_2$-dissolution on an anode
(Dissolution starts at grain-boundary and then grains separate each other.)

(Fig. 3) Relationship between morphology of deposited UO$_2$ and U-concentration in molten salt
(Provided that U-concentration in the molten salt is maintained larger than approximately 8wt.%, large size of UO$_2$-particle (>200$\mu$m) can be collected.)