

ENHANCEMENT OF CONFINEMENT CAPABILITY OF LONG-LIVED NUCLIDES
IN A SIMPLIFIED FUEL CYCLE

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ABSTRACT

Study on PARC(Partitioning Conundrum Key) process concept is under way using Back-End Cycle Key Elements Research Facility(BECKY) in NUCEF. Reprocessing should be performed economically and safely. Process simplification will be effective to improve economical competitiveness. And enhancement of confinement capability of long-lived nuclides will be effective to improve the safety standard and to ease the people's anxiety about reprocessing wastes. Those two measures are apparently contradictory each other. However, both measures are closely linked and interdependent in some cases. We are studying simplified reprocessing concept PARC which is expected to reduce risks due to long-lived nuclides in the reprocessing waste management, not only in the geological disposal but also in the environmental releases.

1. Introduction

It is important that the application of separation and transmutation functions of long-lived nuclides does not pose a substantial increase of economical burden on actual nuclear fuel cycle. According to the study until now, neptunium(Np-237), technetium(Tc-99), plutonium, americium and iodine(I-129) are assessed to take relatively significant role on potential hazard as long-lived elements in HLW.

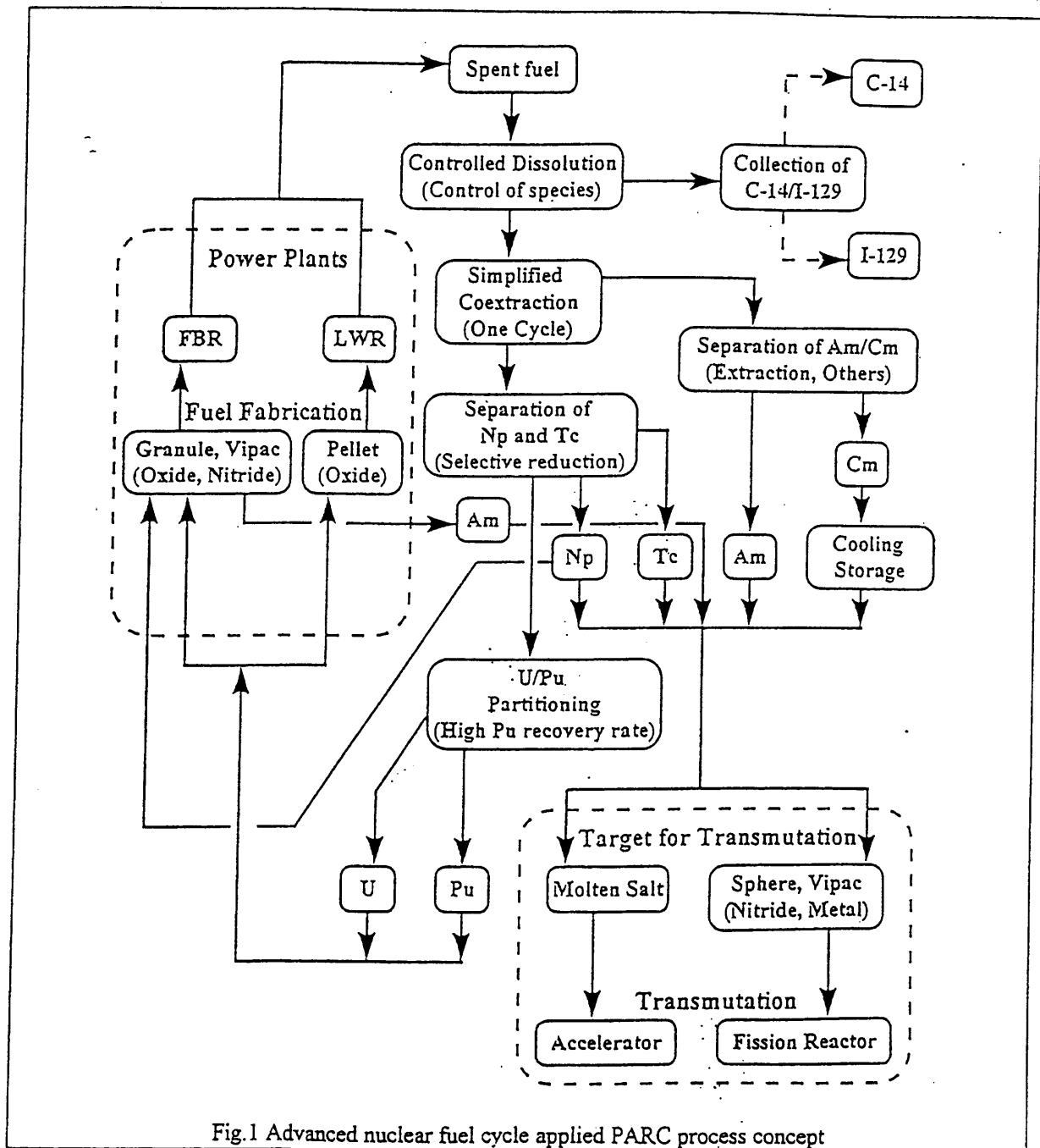


Fig.1 Advanced nuclear fuel cycle applied PARC process concept

Np and Tc are special elements which easily diffuse into products in reprocessing and migrate fast in geological matrix, too. This characteristic property makes those elements assessed as highly potentially

hazardous. Although Pu and the daughter Am do not migrate fast in geological matrix, the influences of these elements are estimated to be significant because of the increased amount recycled in the future fuel cycle. I-129 and C-14, the latter has been assessed from long ago to have a significant influence on the public health⁽¹⁾, are volatile and easy to be released into the environment. Those nuclides, alike Kr-85, are assessed to be relatively influential on the effective dose of inhabitants near reprocessing facility. Technologies for economical and effective separation of the above-mentioned long-lived nuclides are expected to be useful for enhancement of radioactivity confinement capability in reprocessing. Particularly, enhancement of separation efficiency for the elements which are susceptible of diffusing into products, such as Np and Tc, improves the performance of separation step. From these points of view, we are studying PARC concept of reprocessing incorporated with transmutation function of long-lived nuclides. Figure 1 shows our concept of fuel cycle in the future.

2. Back-End Cycle Key Elements Research Facility BECKY for Chemical Process Study

A research facility, NUCEF-BECKY, has been utilized in JAERI at 1995 to study nuclide behaviors in chemical processes and to study feasibility of advanced processes. Miniature scale experimental equipment has been installed in a small-scale $\alpha \gamma$ cell of BECKY with a capacity of 3kg/y of highly radioactive spent fuel. Main flow sheet of equipment for reprocessing study is shown in Fig.2.

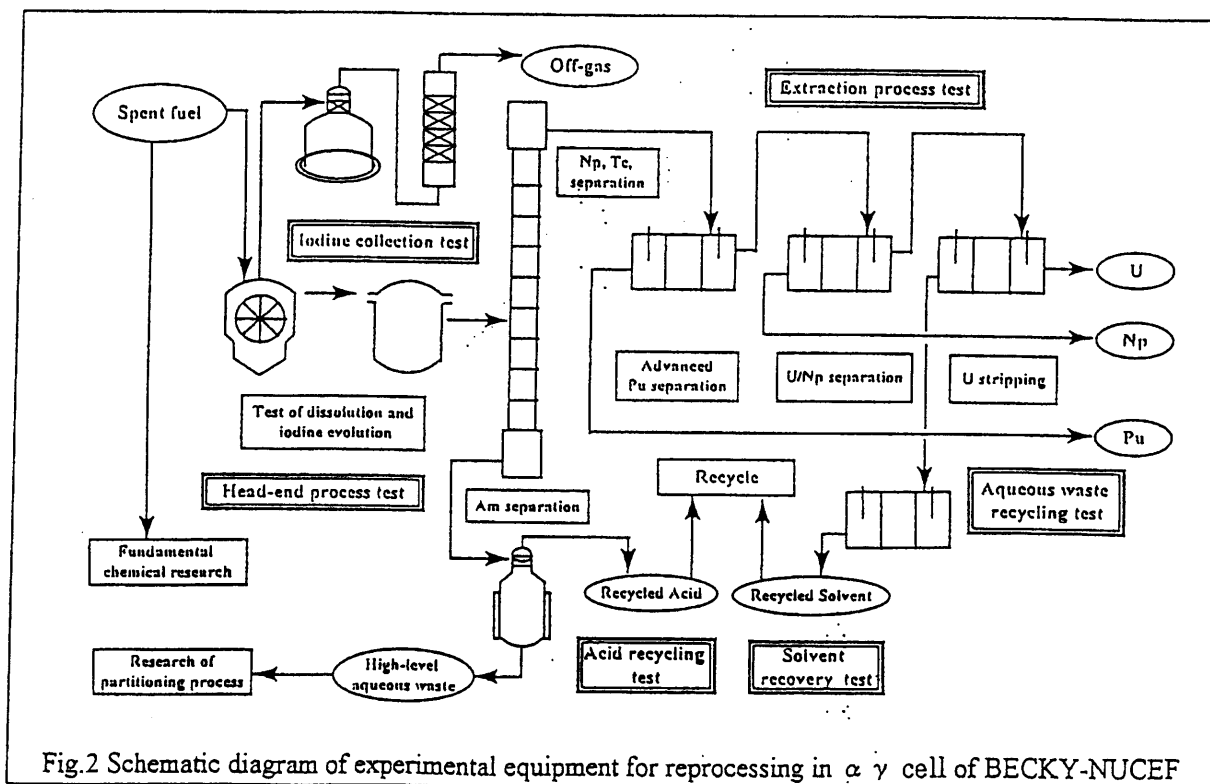


Fig.2 Schematic diagram of experimental equipment for reprocessing in $\alpha \gamma$ cell of BECKY-NUCEF

Experiments started with uranium mixed with simulated fission products for preparing actual hot test

with spent fuel around 1997. Data of dissolution, iodine distribution and extraction have been collected so far. Distribution profiles of iodine in the head-end process were measured using I-131 as a radioactive tracer. Concentration of nitrous acid were measured in continuous dissolver and pulsed columns. Chemical process study program consists of five subjects: (1) Kinetics study of spent fuel dissolution, (2) Feasibility of simplified extraction process, which separates Np and Tc as well as U and Pu, (3) Assessment of capability of minor radioactive nuclides confinement in the process, (4) Development and verification of process simulation code. (5) Adsorption technique of iodine and carbon-14 from dissolver off-gas.

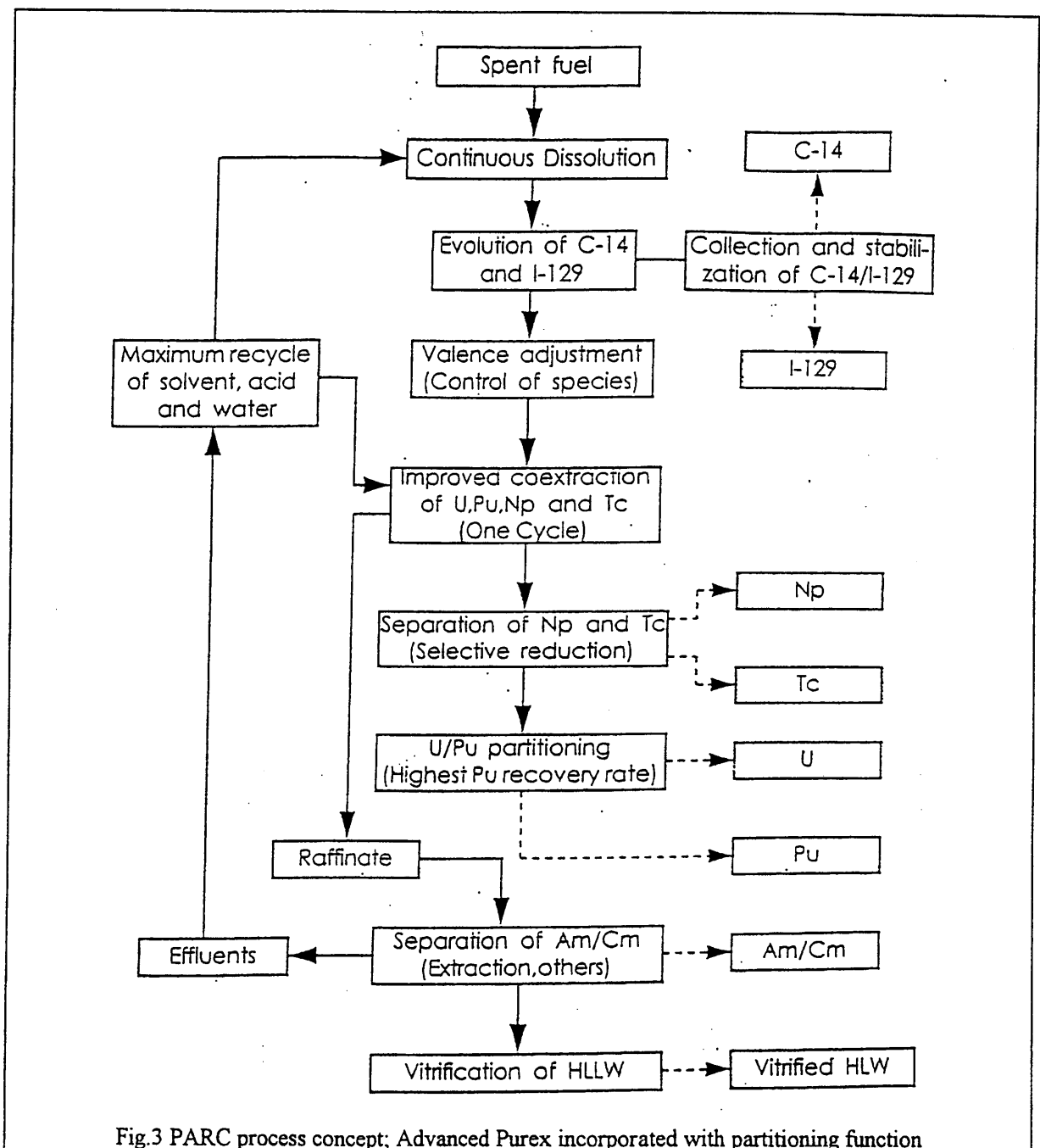
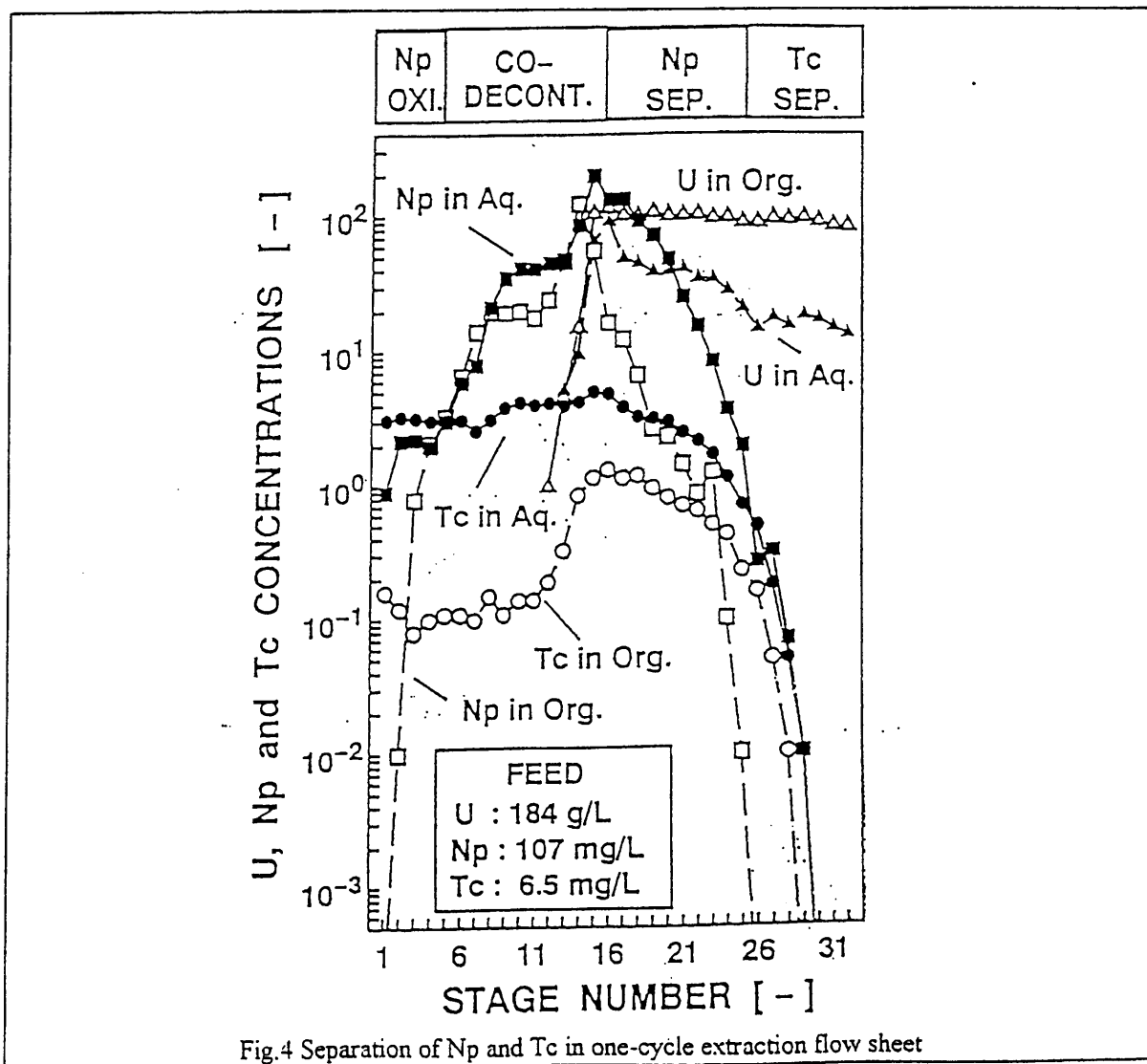


Fig.3 PARC process concept; Advanced Purex incorporated with partitioning function

3. Simplified Chemical Processing

We have proposed "PARC(Partitioning Conundrum Key) process"^(2,3), as shown in Fig.3. This is a challenging program to enhance confinement capability of long-lived nuclides in a simplified reprocessing. This process is designed to separate long-lived nuclides; diffusing Np-237 and Tc-99 in extraction, volatile C-14 and I-129 in head-end, and minor actinide Am from acidic raffinate. Used acid and organic solvent are recycled in the process. Principal features are as follows,

- Simplifying the chemical process
- Enhancement of separation efficiency of the extractable Np and Tc as well as Pu and U.
- Removal of C-14 and I-129 from the off-gas in head-end .
- Recovery of the inextractable minor actinides Am as much as possible
- Recycle use the used reagent at the maximum.



4. Separation Technique of Np-237 and Tc-99

Extraction flow sheet of PARC is designed as one-cycle process based on Purex technology as the primary option, since extractant TBP exhibits sufficiently high separation ability on the major elements; uranium and plutonium. Neptunium and technetium are separated from U and Pu before U/Pu separation step in one-cycle extraction by applying innovative valence control technique⁽⁴⁾. Figure 4 shows concentration profiles obtained in a flow sheet experiment using mixer-settlers. Very high recovery rate of Np, over 99.9%, has been demonstrated by conducting selective Np reduction with a decomposable reagent butyraldehyde for a Np, Tc and U solution system. 99.98% of Np, extracted together with U in the co-decontamination step, was reduced by n-butyraldehyde and separated from U in Np separation step. More than 99.8% of Tc was separated from U in Tc separation step using 5.5M nitric acid scrub solution. In a separately conducted experiment, Pu was separated from U using iso-butyraldehyde effectively without applying hydrazine for a Np, U and Pu solution system. Decomposition of butyraldehyde has been studied in order to get rid of side effect on the separation efficiencies in the down streams. Decomposition fraction calculated from measured concentrations of total organic carbon in aqueous solution is shown in Fig. 5. It was found that more than 99% of n-butyraldehyde was decomposed within 50 minutes with silver catalyzed electrochemical oxidation. Air bubbling without electrochemical oxidation decomposed app. 90% of n-butyraldehyde in 1 hr.

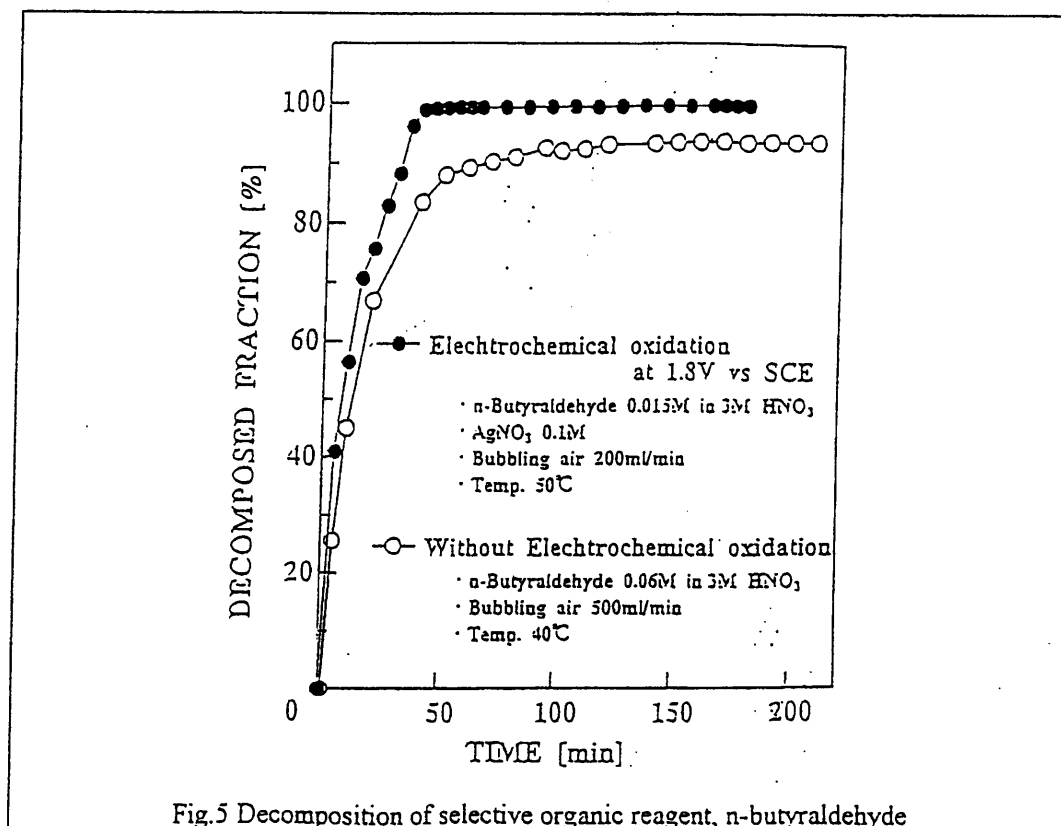


Fig.5 Decomposition of selective organic reagent, n-butyraldehyde

As the basis of one-cycle extraction process development, have been collected data of Np and Tc

behaviors under a standard flow sheet condition of a large scale Purex type reprocessing plant. It was found that app. 99% of Tc was extracted with U and Zr by TBP and only 1% of Tc was transferred into the raffinate of co-decontamination step. Approximately 30% of Np was transferred into the raffinate.

5. Technique for capturing I-129 and C-14

Iodine-129 is controlled to be released to dissolver off-gas(DOG) completely and removed by AgS(Silver impregnated Silica-gel). In order to assess confinement capability of volatile long-lived iodine I-129, iodine distribution in BECKY experimental equipment of head-end process(shown in Fig.2) was measured under continuous dissolution of unirradiated simulated spent fuel⁽⁵⁾. Pellets of the simulated spent fuel consist of uranium and non-radioactive fission product elements. A trace amount of radioactive iodine I-131 was added as KI to dissolver under operation. Nitrous acid concentration was high at app. 3×10^{-3} mol/l as shown in Fig.6. Most of iodine was released to DOG and captured completely by iodine adsorbent columns under condition of linear off-gas velocity up to 22 cm/s and adsorbent temperature at 150°C. Amount of iodine captured in columns is estimated to be app. 74% iodine loaded in dissolver. Less than 1% of iodine loaded was remained in fuel solution. The other iodine was distributed in HEPA filter by 11%, inside surface of pipes by 2%, insoluble residue and others by 12%, respectively. Colloidal formation with fission products such as AgI or PdI₂ is considered to be predominant retention mechanism under iodine concentration less than 10^{-6} mol/l in solution⁽⁶⁾.

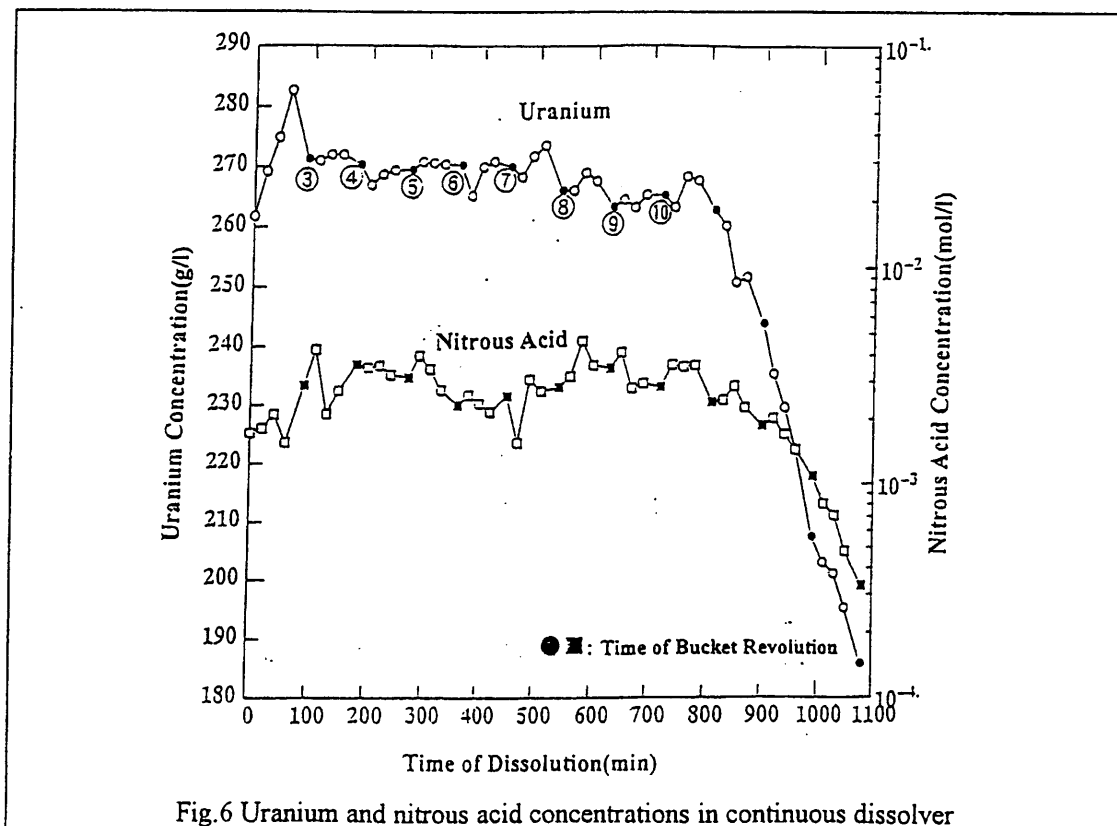


Fig.6 Uranium and nitrous acid concentrations in continuous dissolver

Major chemical form of C-14 is carbon dioxide(CO_2)⁽⁷⁾. CO_2 is released to DOG under dissolution of spent fuel. As an adsorbent, hydrogenated mordenite, which is capable of reversible adsorption-desorption of CO_2 , has been selected after screening test of various kinds of adsorbents. Breakthrough curves of CO_2 show adsorption capacity of hydrogenated mordenite is improved under lower temperature.

6. Development of Simulation Codes

Process simulation code system ARECS(Fig.7), associated with fundamental reprocessing data-base, has been developed to simulate chemical process system composed of dissolution, off-gas treatment and extraction process. This code system is capable of simulating the behaviors of not only U and Pu but also long-lived minor nuclides such as I-129, C-14; Np-237, Tc-99 which are important in the development of nuclear fuel cycle technology. And, a computer code has been developed to simulate generation and migration of very short-lived volatile nuclide such as I-131 by spontaneous fission of TRU nuclides. TRU nuclides such as Cm-244 generate various very short-lived volatile nuclide by spontaneous fission.

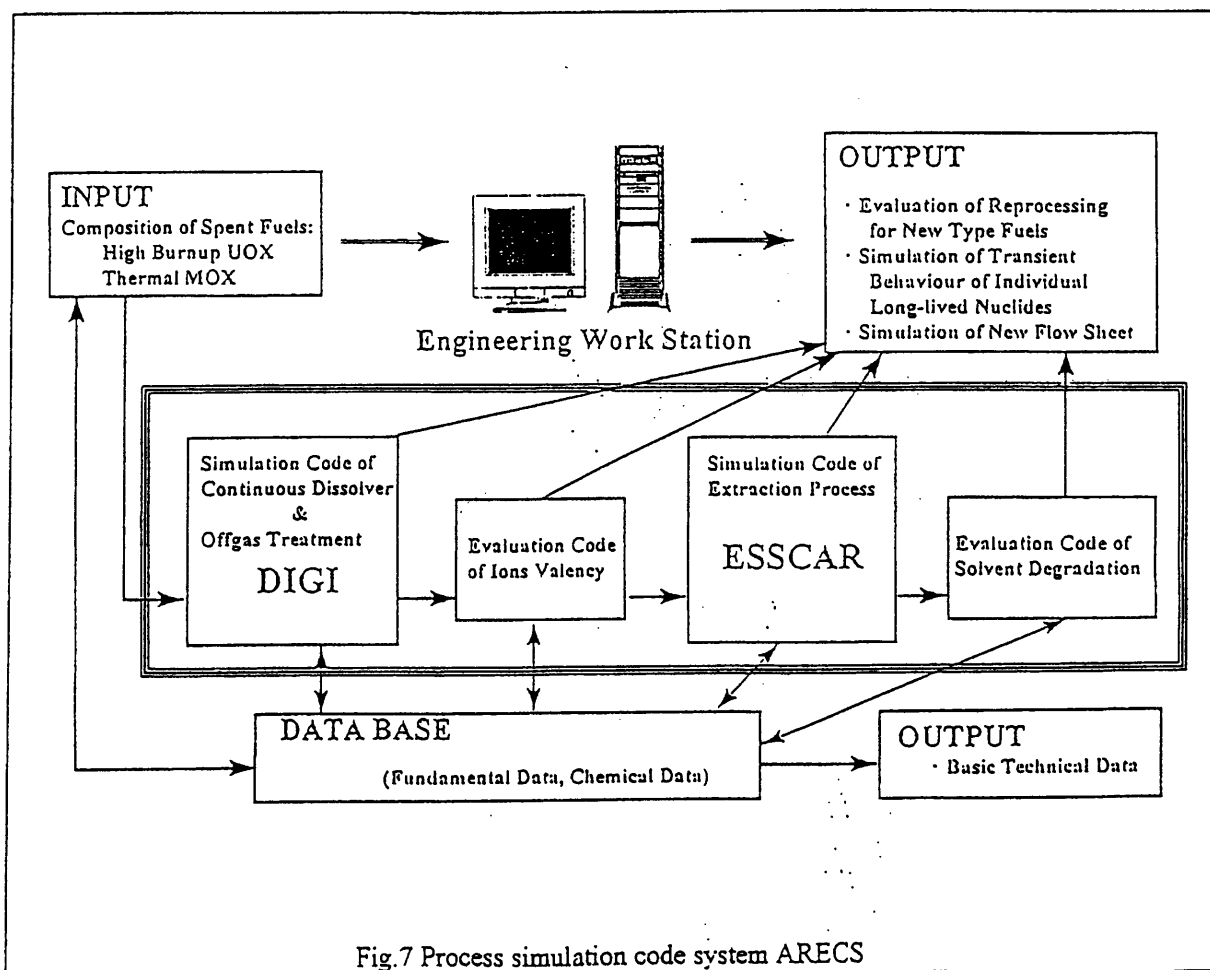


Fig.7 Process simulation code system ARECS

8. Conclusion

Development of chemical process technology for PARC concept is under way using Back-End Cycle Key Elements Research Facility(BECKY) in JAERI. High efficiency was demonstrated for separation of Np-237, Tc-99, I-129 and C-14 under a simulated condition. It is expected that technologies for enhancing confinement capability of long-lived nuclides improve the safety standard and to ease the people's anxiety about reprocessing wastes.

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