

“ORIENT-CYCLE” – EVOLUTIONAL RECYCLE CONCEPT WITH FAST REACTOR FOR MINIMISING HIGH-LEVEL WASTE

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Abstract

JNC has proposed an evolutionary concept of fast reactor recycle system for minimising high-level waste (HLW) by adopting an unconventional recycling scheme based on the idea of “rough removal of unnecessary elements” instead of a conventional one as “pure recovery of necessary elements”. The concept was named “ORIENT-cycle” (**O**ptimisation by **R**emoving **I**mpedimental **E**lements). “Unnecessary elements” to be removed were listed up from various aspects such as core design, front end and back end of the fuel cycle. Stable fission products that amount to about 60wt% of all FPs were identified as one category of key “unnecessary elements” from a neutronic aspect and also final disposal aspect because they were major parasitic neutron absorbers if recycled and were not required to be disposed in deep geologic media. Adaptable separation schemes and the neutronic feasibility of the multi-recycling were discussed. The production rate of HLW was reduced by a factor of around 10 compared with a conventional cycle.

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I. Introduction

In a past development of reprocessing technology, the fundamental principle had been “pure recovery of necessary elements” from spent fuels for further use in the fuel cycle. As generally known, fast reactors are less sensitive to impurities contained in a recycled fuel. It is considered that the insensitivity implies flexibility in structuring a fuel cycle scheme. To explore the flexibility and create a Japan-originated new recycle concept, the authors set up a fundamental principle of “rough removal of unnecessary elements”.

Assuming long-term use of nuclear power, the importance of back end of nuclear fuel cycle will increase more and more. Continuous use of fission reactors undoubtedly requires a series of repository constructions. Although transmutation technologies of long-lived nuclear waste by using fast reactors have been studied for several years among many research organisations, they are not necessarily linked to the reduction of HLW. Taking the political and sociological difficulties to find new repository sites into account, minimising HLW production to prolong a need of new repository is of major concern when considering a new recycle concept.

The purpose of this study is to propose a new recycle concept, namely ORIENT-cycle, based on the principle of “rough removal of unnecessary elements”. The authors especially focused on the point what reduction can be attained in HLW generation by making the full use of distinctive neutronic feature of fast reactors. In forming a new recycle scheme, only element-wise separation technologies are assumed.

II. Listing unnecessary elements

The meaning of “unnecessary” depends on the aspect we take. As an initial step, “unnecessary elements” were independently listed up from various viewpoints such as fuel fabrication, core design, waste conditioning and final disposal.

Front end aspect

The heat generation of low decontamination fuel material containing 5wt% of minor actinides and 2wt% of FP is 7 times as high as that of standard MOX fuel. High fuel temperature causes changes in O/M ratio, fuel powder fluidity, etc. However the increased heat generation of fuel seems to be possible to cope with by controlling atmosphere in the fuel fabrication cell, installing cooling equipment and dividing the lot. The main contributor for heat generation of the low DF (decontamination factor) fuel is Cm.

The radiation dose of the low DF fuel is higher than MOX fuel by the order of 4. The increased radiation dose of the recycled fresh fuel brings difficulties in the operation and integrity of equipment. In terms of radiation dose for operators, Eu is a concerned element. The radiation damage for equipment is ascribed to Pm, Eu and Y. In order to overcome these adverse influences, following measures are required to be taken: remote controlled cell, lot dividing and use of radiation-resistant material.

The impact of contaminated FP except for volatile elements on the MOX fuel fabrication capability could be negligible up to 3% of FP contamination. As for metal fuel, the contamination of minor actinides (MA) and rare earth (RE) into U-Pu-Zr alloy could be limited to about 2% because of the appearance of deposited phase.

The corrosive elements (I, Cs) could be picked up as unnecessary elements from the aspect of integrities of fuel and cladding. Furthermore it is likely that La causes eutectic reaction by depositing with metal and also that platinum group metals (Ru, Rh, Pd etc.) causes cracks in cladding by depositing metal layer.

Core design aspect

In fast reactors, actinides are all fissionable and contribute to produce energy. On the contrary fission products are not, so that all of fission products can be basically regarded as unnecessary elements for recycling in terms of energy production.

The neutronic impact by contaminated fission products in fresh fuel recycled into fast reactors is generally small due to the hard neutron spectrum. There are, however, some dominant fission products on the reactivity deterioration. Recycling such parasitic neutron absorbers into cores must be especially avoided from a neutronic viewpoint.

Figure 1 shows the reactivity impact of each FP isotope versus half-life. The FP composition examined is for 4 years cooling. It is roughly illustrated that longer life isotope have larger impact on the reactivity of the core. In generally, a neutron rich nuclide produced by fission reaction shows short half-life and small neutron capture cross-section. It decays out successively by emitting an electron into other nuclide having longer half-life and larger neutron capture cross-section, until reaching a stable nuclide. Accordingly lots of stable fission products are naturally contained in spent fuels after several years cooling and most of them would not be preferable for recycling.

Figure 1. Impact on reactivity of FP isotopes

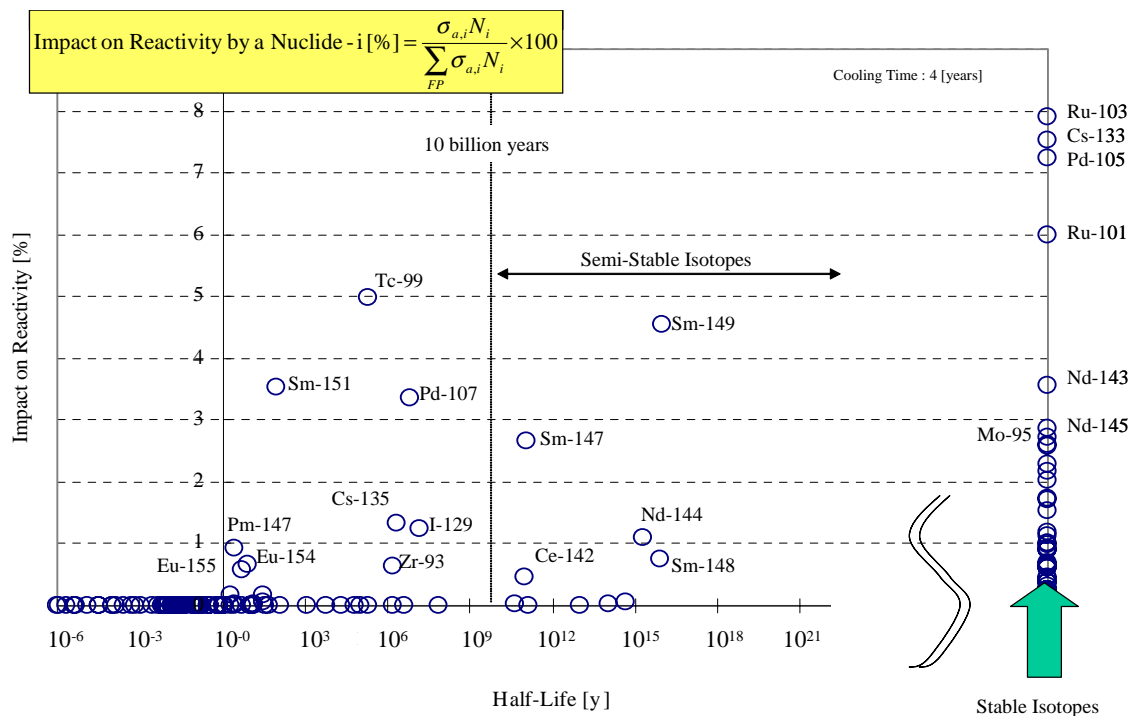
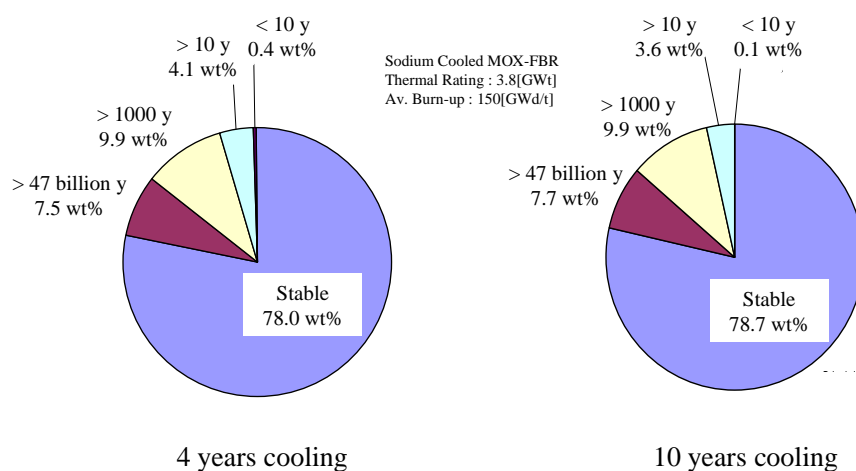


Figure 2. FP isotopes classification by half-life



The half-life classification for FP contained in spent fuels cooled for 4 or 10 years is indicated in Figure 2. The ratio of stable isotopes is about 78wt%. Besides, semi-stable isotopes ($T_{1/2} > 47$ billion years) is contained around 8wt%. These numbers are almost independent of the cooling length considered. Namely about 85wt% of FP can be classified as stable isotopes.

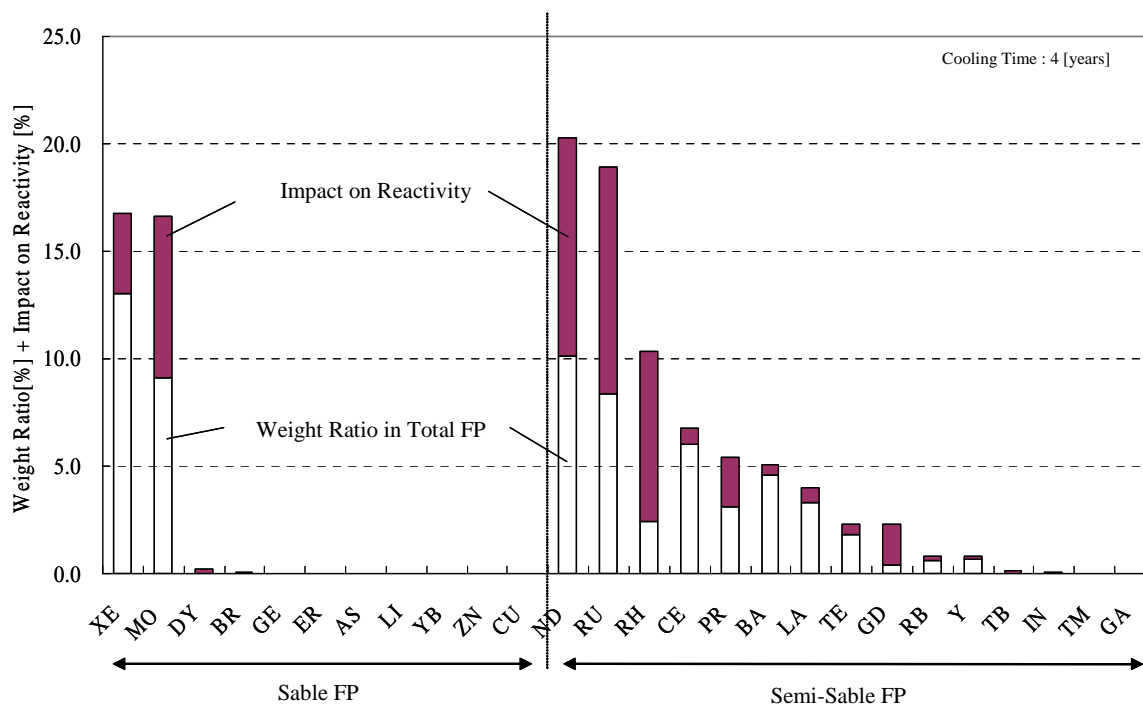
Then the authors examined to sort out the dominant FP having large neutron absorption ratio and large weight ratio based on an element-wise classification. Here the “stable element” and “semi-stable element” are defined as follows:

- Stable element: < 1 Bq/tonHM after 4 years cooling;
 semi-stable element: $T_{1/2} < 2$ years, or $T_{1/2} > 47$ billion years (for all isotopes included).

Figure 3 shows the dominant stable and semi-stable FP elements in terms of neutron absorption ratio and weight ratio. As stable elements, Xe and Mo were identified and as for semi-stable following elements were extracted: Nd, (Ru,) Rh, (Ce,) Pr, Ba, La, Te, Gd, Rb and Y. The parenthesised two semi-stable elements Ru and Ce could require longer cooling period than others to be treated as semi-stable due to residual radioactivity ascribed to ^{106}Ru ($T_{1/2}=1\text{y}$) and ^{144}Ce ($T_{1/2}=0.8\text{y}$), respectively.

These stable and semi-stable FP elements that amount to about 60wt% in total FP are not necessarily handled as radioactive waste nor disposed in deep geologic repository for isolation from the biosphere. Therefore they can be considered as “unnecessary elements” not only from neutronic viewpoint but also from final disposal viewpoint.

Figure 3. Dominant FP elements in weight and neutron absorption



Back end aspect

To reduce heat generation of the vitrified waste, separation of Cs and Sr is efficient. By separating them, shortening of necessary cooling time for HLW before the final disposal and reduction in size of the repository will be expected. The repository size, however, is also determined by the dynamic stability of the tunnel.

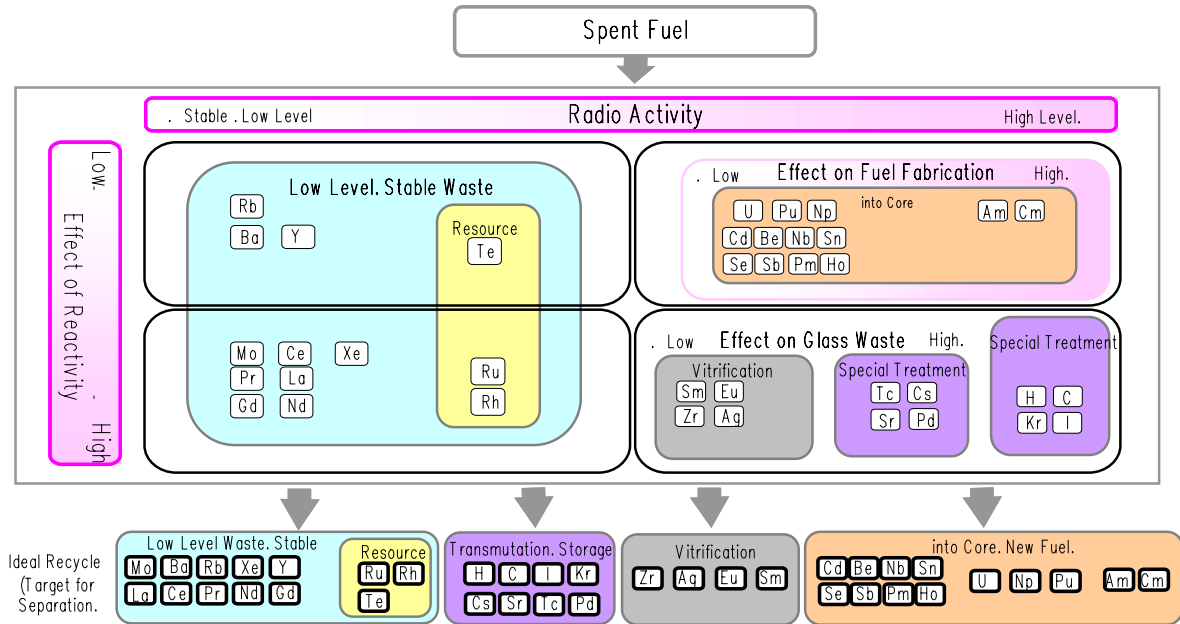
Separation of Mo, Ru and Pd is effective to improve the containment capability of vitrified waste. Because Mo causes soluble phase that deteriorates the chemical durability of the vitrified waste. Ru and Pd are the elements that disturb the operation of a melter for mixing high-level solution and glass materials.

Therefore the improved containment capability of vitrified waste by separating Mo, Cs, Sr, Ru and Pd from high-level solution would contribute to reduce the vitrified waste generation.

III. Definition of cycle scheme

Based on the examined results from various aspects of a total nuclear system, an ideal grouping pattern is illustrated in Figure 4. Elements existed in spent fuels are consequently divided into 5 categories as follows: 1) low-level waste, 2) valuable resources, 3) storage or transmutation, 4) vitrified waste and 5) recycled fuel.

Figure 4. Ideal categorisation based on the concept of “unnecessary material removal”



The cycle scheme of the ORIENT-cycle is illustrated in Figure 5 as a form of mass flow chart. High heat-generating FP, Cs and Sr, are separated and stored in the cooling/storage facility with other long-lived FP (I, Tc etc). The transmutation of I and Tc is supposed to be an option. Noble metals such as Pd are recovered for utilisation.

The stable and semi-stable FP elements (Xe, Mo, Nd, Ru, Rh, Ce, Pr, Ba, La, Te, Gd, Rb and Y) are selectively separated from spent fuels then disposed as low-level waste. The vitrified waste contains Zr, Ag, Eu, Sm etc. that are radioactive and inadequate for recycling because of the large neutron absorption cross sections. All actinides and other FP decontaminated in the fuel are recycled into the core.

IV. Separation flowsheet

In order to draw a conceptual separation flowsheet to materialise the ideal separation pattern as shown in Figure 4, several kinds of separation technologies were reviewed. By combining possible processes properly, new separation processes are designed for both aqueous process and pyro-process for reprocessing oxide fuel and metallic fuel, respectively. As for aqueous process, adoption of head-end voloxidation process for volatile FP removal is examined to make the following processes simple.

Aqueous separation process

By reviewing the present art of separation technology, we selected the processes suitable for aqueous reprocessing as follows:

Figure 5. Mass flow of the “orient-cycle”
(orient: optimisation by removing impedimental elements)

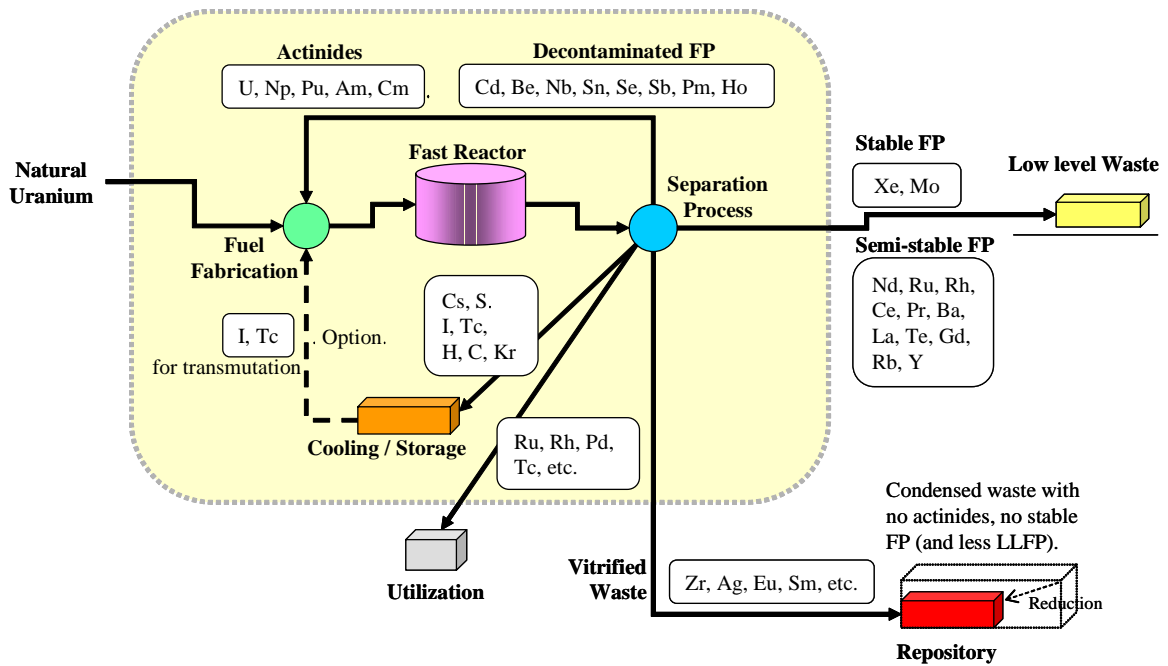


Figure 6. Conceptual flowsheet for aqueous separation processes

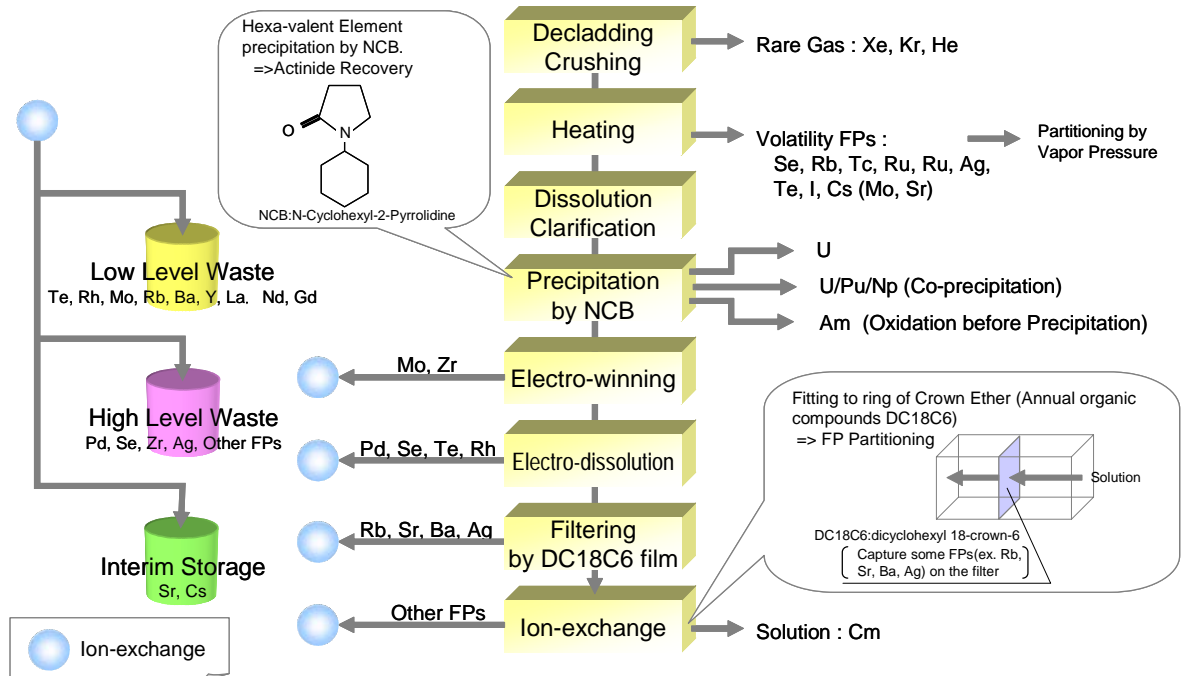
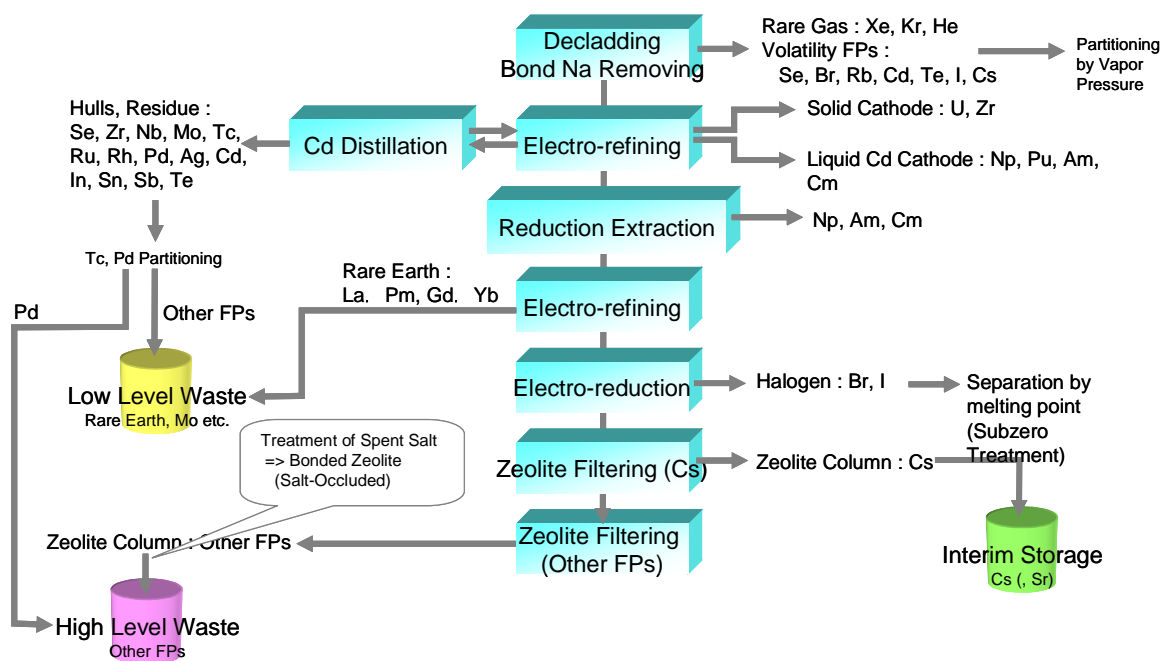


Figure 7. Conceptual flowsheet for pyro-separation processes



1. Voloxidation: removal of volatile fission products by high temperature oxidation or reduction.
2. NCB (N-cyclohexyl-2-Pyrrolidone) precipitation: separation of U, Np and Pu from residual solution by oxidising with ozone and separation of Am by electrolysis oxidation. [1]
3. Catalytic electrolysis extraction: electrochemical separation into three parts as rare elements (Pd, Se, Rh, Tc) deposited on the cathode, precipitation of Mo and Zr and other elements remained in nitric solution. [2]
4. Thin film separation: separation into two parts of remained FP using DC18C6 (Dicyclohexyl-18-crown-6) thin film.
5. Ion exchange: separation of Cm and purification of each element.

The conceptual flowsheet for aqueous separation processes is given in Figure 6. These processes are likely proven technologies except for the NCB precipitation process. However there are some problems to be solved as follows: the yield improvement for the volatile recovery method, the evaluation of impurities behaviour in electrolytic extraction stage and the development of equipment for film separation process.

Pyro-separation process

Figure 7 illustrates the conceptual flowsheet for pyro-separation processes. Most of noble metals shift to the Cd-pool in the electrolytic separation process. In this flowsheet considered the rare earth and halogen separations by electrolysis from FP which moves to spend salt conditioning stage. The processes to separate I by electrolytic process and Cs by using the zeolite column are included.

It is required to develop the separation technique of Tc and Pd from noble metal waste and also the separation technique of Sr and Se from spent salt.

V. Feasibility of the multi-recycling

Neutronic feasibility of the multi-recycling with the defined separation schemes was evaluated by using an equilibrium cycle calculation programme. In the burn up calculation, detailed reaction chain was considered for more than 1 200 FP isotopes because the recycled fuel contains many kinds of FP in the assumed recycle scheme.

The neutronic feasibility of the multi-recycling was judged by using a criticality index defined as follows:

$$\text{Criticality Index} = \frac{\sum_{i \in HM} \nu \sigma_{f,i} n_i \phi}{\sum_{i \in HM, FP} \sigma_{a,i} n_i \phi} \quad (1)$$

Table 1 summarises the results for both cases of aqueous process and pyro-process. Even in the equilibrium cycle with multi-recycling, accumulations of FP elements in the fuel was not more than 2wt% and the impact to the criticality index was less than 1%dk/kk'. The small impact on the reactivity by the contaminated FP could be attributed to the selective removal of the dominant neutron absorbers in the separation process.

Table 1. Neutronic feasibility of the multiple recycling

| | MOX fuel + Aqueous process | | Metallic fuel + Pyro-process | |
|--------------------------------|----------------------------|--------------|------------------------------|--------------|
| | No FP recycle | ORIENT-cycle | No FP recycle | ORIENT-cycle |
| FP Ratio in recycle fuel [wt%] | 0 | 0.7 | 0 | 1.9 |
| Criticality index | 1.209 | 1.201 | 1.287 | 1.282 |
| Reactivity impact [%dk/kk'] | – | -0.58 | – | -0.29 |

As one of parametric examinations, the feasibility of simplified multi-recycling by applying only voloxidation process was also examined. As a result, unacceptable FP accumulation in the fuel was observed and the value of the criticality index was far below unity.

The result showed that the impact of the contaminated FP in the recycled fuel was small enough to keep the core critical even after infinite recycling. Therefore it could be concluded that the multi-recycling of the assumed recycle scheme is neutronically feasible.

VI. Reduction of HLW

The production rate of HLW was evaluated to confirm the effect on the waste reduction by implementing the ORIENT-cycle.

By removing heat-generating elements Sr and Cs, and also Mo that causes soluble phase in glass, the chemical durability of the glass could be improved and the loading limitation of high-level liquid waste into glass could be expanded up to around 35wt% from the current value of 10-15wt%.

Table 2. Comparison of HLW production

| | MOX fuel + Aqueous process | | | Metallic fuel + Pyro-process | |
|-------------------------------------|-----------------------------|-------------------------|--|------------------------------|----------------------------------|
| | HLW Production | | Mass of Cs, Sr and Mo in HLW [g/GWh] | | Zeolite Waste Production [kg/HM] |
| | per unit fuel mass [kg/tHM] | per unit energy [g/GWh] | | | |
| Conventional-Cycle (Rokkasho Plant) | 44.1 | 120 | Cs: 9.6 Sr: 3.2 Mo: 12.4 | JNC Pyro-Cycle [4] | 420 |
| ORIENT-Cycle | 15.4 | 10.9 | Cs: 0.5 Sr: 0.07 Mo: 0.4 | ORIENT-Cycle | 244 |
| Reduction Ratio | about 1/3 | about 1/10 | Cs: about 1/20 Sr: about 1/40 Mo: about 1/30 | Reduction Ratio | about 4/7 |

The result of waste mass evaluation for the aqueous separation process indicated that the HLW produced per unit energy generation was reduced by about one order of magnitude compared with the conventional recycle procedure as shown in Table 2. This was caused mainly by removing high heat-producing elements and non-radioactive elements including Mo. It was roughly estimated that removing Sr, Cs and Mo meets the reduction factor of about 3 and removing stable and semi-stable elements leads further reduction with a factor of about 3.

As for the pyro-separation process, the volume of zeolite waste including high-level waste could be reduced by about half in case of the ORIENT-cycle.

VII. Conclusions

An evolutionary recycle concept “ORIENT-cycle” has been proposed based on the idea of “rough removal of unnecessary elements” instead of a conventional one as “pure recovery of necessary elements”. The main target was to minimise the HLW production.

“Unnecessary elements” to be removed were listed up from various aspects such as core design, front end and back end of the fuel cycle. As a result, stable and semi-stable fission products that amount to about 60wt% of all FPs were identified as a category of key “unnecessary elements” from a neutronic aspect and also final disposal aspect. Because they are parasitic neutron absorbers that deteriorates the criticality of the core and simultaneously they are non-radioactive elements that are not required to be disposed in deep geologic media for long-term isolation. The stable and semi-stable elements include Xe, Mo, Nd, etc. In order to reduce the volume of HLW, Cs, Sr and Mo were also defined as “unnecessary elements” for the vitrified waste.

Conceptual separation flowsheets were developed for aqueous and pyro-separation processes and the neutronic feasibilities of the multi-recycling were evaluated based on the mass flow data for the defined recycle schemes. The impact of the contaminated FP in the recycled fuel was so small that the multi-recycling could be neutronically feasible.

The result of waste mass evaluation for the aqueous separation process indicated that the HLW produced per unit energy generation was reduced by about one order of magnitude compared with the conventional recycle procedure. This is attributed to the removal of high heat-producing elements and non-radioactive elements including Mo.

It turns out that the ORIENT-cycle is feasible to be formed as a fast reactor recycle system and effective to reduce HLW production. The flowsheets optimisation of the separation processes must be performed so as to minimise the secondary waste and the cycle cost as a future work.

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