# Perspectives on Application and Flexibility of LWR Vitrification Technology for High Level Waste Generated from Future Fuel Cycle System

M. Shiotsuki, A. Aoshima, S. Nomura
Japan Atomic Energy Agency (JAEA)
4-49 Muramatsu, Tokai-mura, Ibaraki 319-1184
Japan

## ABSTRACT

Achievement of reliable technologies on solidification and disposal of the HLW from future fuel cycle systems such for high burnup LWR, Pu-thermal (MOX), fast breeder reactor (FBR) and their transient stages is one of the most important issues to establish such advanced fuel cycle systems. In this paper, applicability and flexibility of the current vitrification technology for LWR fuel cycle to HLW from the future fuel cycle systems were reviewed by examining characteristics of the HLWs. The current developed vitrification technology is expected to have an advantage for applying to the solidification process of the HLW generated from future fuel cycle systems with some modification/optimization of the melting condition, etc. Moreover, it is thought that the advance aqueous reprocessing system developed for future FBR cycle has the potential which can contribute to the further reducing the number/volume of the HLW. It is also confirmed that development efforts on countermeasure for accumulation of noble metals, which JAEA has been carrying out aiming to accomplishing more stable and reliable operation of the vitrification process and extending the melter's life, will be able to contribute in the future fuel cycle system furthermore.

## CURRENT STATUS ON VITRIFICATION TECHNOLOGY IN JAPAN

As the solidification technology of the high-level radioactive waste (HLW) generated from the LWR reprocessing process, development of the vitrification technology with the borosilicate glass has been progressed and actual plants have already been established and in operation in France, United Kingdom, Belgium, Russia and Japan.

### Tokai Vitrification Facility (TVF) [1]

A vitrification facility called "TVF (Tokai Vitrification Facility)" at the Tokai Reprocessing Plant (TRP) site had been constructed and has been operating by Japan Atomic Energy Agency (JAEA, former "JNC"). In the TVF, highly radioactive liquid waste from the main process of the TRP is vitrified into glass matrix with a liquid fed joule-heated ceramic melter (LFCM). The first TVF melter had initiated the hot operation in 1995 and was operated until 2002. The melter being operated now is the second one. Total number of produced canisters was over 200 canisters (110-litre) until November, 2005.

The TVF has characteristic feature in maintenance system design: the fully remote maintenance system, which makes equipment exchange work so saving comparing to direct maintenance. In the fully remote

maintenance system, all main equipment besides the melter is mounted in racks, which is installed in the large cell ("the vitrification cell") and the all equipment is remotely maintained by using in-cell cranes and the two-armed servo manipulators. The melter is also installed in the vitrification cell by bolting on the base framework.

The melter is designed to exchange every five years (equivalent to 500 canisters) in the maximum operation mode due to limitation of corrosion of the structural materials such as a ceramic wall and electrodes. Glass fiber cylindrical cartridge is used as a glass additive for melting. The HLLW is soaked into a cartridge before it is fed into the melter. The glass melted at the temperature of 1150 is poured into a canister periodically through a discharge nozzle located at the bottom of the melter. Fig. 1 indicates schematic view of the TVF melter.

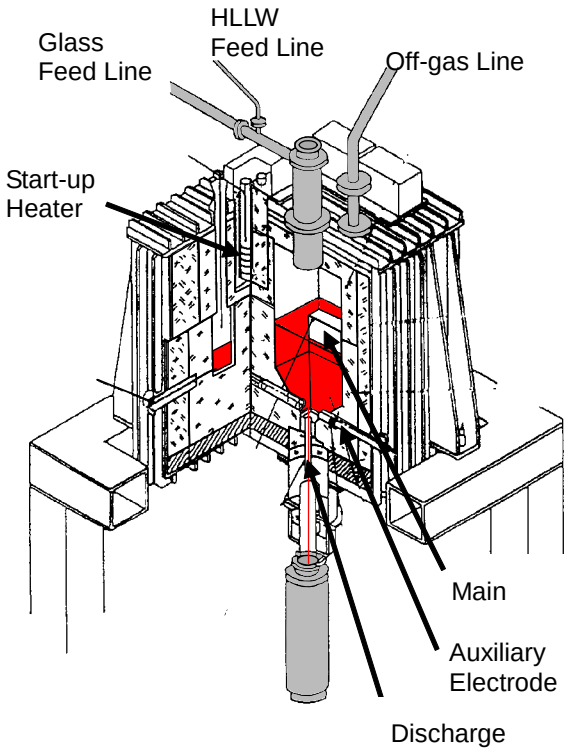


Fig. 1. The TVF melter

The bottom part of the melter was designed to have the 45° sloped bottom aiming to have good draining ability of sedimented noble metals in molten glass. During the pouring, the weight and glass level are successively measured by load cells and a gamma-ray device, respectively. The filled canister is subsequently cooled before transfer to the welding position and the lid is welded by a TIG welder to seal the canister. After being decontaminated by high pressurized water jet spray and wire brushing, inspection of contamination and containment are carried out. Finally, canisters are stored in the storage pits.
Melter off-gas is cleaned by a submerged bed scrubber (SBS), a venturi scrubber, a perforated plate type water scrubber, a high efficiency mist eliminator (HEME), and subsequent filter process such as a ruthenium absorber, an iodine absorber and HEPA filters.

Through the hot test operation, the special operation method, "the low temperature operation mode" which keeps temperature of glass at the melter bottom around 800~900 was evaluated. This special operation mode was aimed to avoid accumulation of noble metals to the melter bottom by increasing the viscosity of the glass. However, gradual decrease of electric resistance between main electrodes was observed exceeding 50 canister productions due to accumulation of noble metals at the melter bottom. After 130 canisters production in 2002, damage of the main electrode had occurred during the noble metals flush-away operation by adding sodium carbonate. The exhausted air temperature from one of the main electrodes was suddenly down due to clogging of glass into an air path inside of the electrode.

Consequently, the first melter had removed and the second melter was installed with improvement of configuration of the bottom part in order to prevent accumulation of noble metals. The second melter was restarted in operation on October, 2004. Since then, TVF has been operating with stable condition up to now.

## Ongoing R&Ds on Vitrification Technology in JAEA

JAEA has been performing R&Ds on vitrification technology in parallel to the operation of TVF aiming to reducing waste volume or number, extending melter life time and achieving more stable operation; volume reduction technology, advanced melter design with basic experiments.

## Volume reduction technology

At present, the solidified glass loads about 25 wt% waste containment which composes 10% sodium dioxide and 15% metal elements (fission products, actinides etc). If the containment is increased to 30 % (e.g. 12 % sodium nitrate and 18 % metal elements), the number of canisters decreases 23%. Such effect may help cost saving for its transportation, storage and disposal as well. Critical indications, which were determined waste load ratio, have been considered as molybdenum concentration in glass matrix and heat production of the canister. Much molybdenum concentration (e.g. 3wt%) had been evaluated to generate some salt phase soluble to water, which might reduce performance of the solidified glass. A high containing radionuclide canister can be easily understood as relatively higher heat production than a reference loading canister. The heat level influences on storage capability at the production of the canister and storage period or disposal design (e.g. distance between disposal pits). Additionally, operational parameter may be influenced by the high waste loading due to changes of viscosity and/or electric resistance of the melted glass. JAEA has started the R&D of the volume reduction technology in collaboration with utilities. The R&D started with basic experiments to re-evaluate above-mentioned indications and will be progressed in a small scale melter and a full scale cold mock-up tests.

## Advanced melter design

Principal aims of advancement of the melter are to extend melter life-time and to increase robustness/flexibility of operational performances for prevention of noble metal accumulation, composition changes for future recycle systems, etc.

As mentioned above, the current melter is designed as five-year life time because of ceramic wall and electrode corrosion. Once the melter is exchanged, costs for removal and dismantling of the retired melter and for manufacturing new melter would demand enormous economical burden. Additionally, the exchanging work interrupts its operation for beyond several months and its dismantling work results to generate much volume of the waste which requires another need of future storage and disposal. In spite of application of the new bottom design in the second TVF melter, it is desired to put further effort to establish the radical countermeasure against noble metal accumulation. To support the development, JAEA has re-started basic experiments regarding noble metal behavior such as generation, particle growth and deposition, and systematic understanding of corrosion of ceramic and electrode. Simulation system

has been also under development to analyze coupled features and phenomena such as temperature, heat generation, electrical potential, current density as well as noble metal particle distribution (Fig. 2).

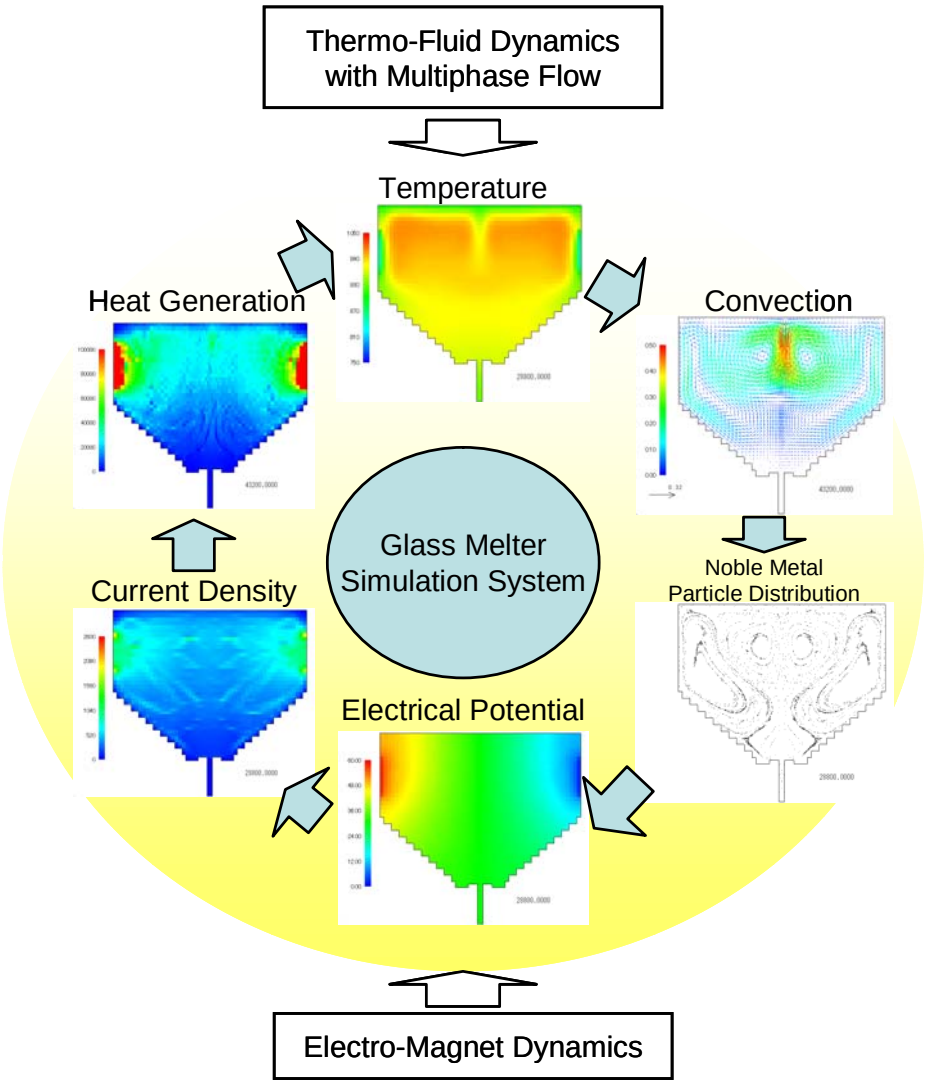


Fig. 2. Concept of the developed simulation system

## CHARACTERISTICS OF HLW GENERATED FROM ON-GOING LWR CYCLE SYSTEMS

First Japanese commercial reprocessing plant, which is now under experimental operation using uranium, was designed as average burnup of 45GWD/t. The vitrification process of the reprocessing plant and reference HLW composition are also considered with this burnup. To increase efficiency of use of nuclear fuel, higher burnup operation has already been applied in Japan. In BWR, the maximum burnup to 55 GWD/t has been in practical use. Such burnup is planned also in PWR. From a viewpoint of improvement in efficiency of uranium use, it is recognized as reprocessing of Pu-thermal (MOX) spent fuel being important. Study on the Measure to be taken for the reprocessing is planned to start in around 2010.

In this session, the influence of high burnup (55 GWD/t) was evaluated using ORIGEN2 code regarding changes of important compositions (noble metals and molybdenum) for vitrification process and of heat generation of the canisters [2]. Moreover, evaluation for MOX (55 GWD/t) was also tried by same methodology.
In these evaluations, a reference HLW canister of 45 GWD/t burnup (1.25 canisters/tU) was considered and numbers of other evaluated canisters were converted by proportions of their burnup. Same element fractions, designed in the current PUREX process, from spent fuels to the HLWs were taken into account.

Result of changes of compositions of noble metals and molybdenum is shown in Table I.

Table I. Changes of Compositions of Noble Metals and Molybdenum by High Burnup and MOX

| HLW | Composition at vitrification, kg/canister | | | | |
|---|---|---|---|---|---|
| | Noble Metals | | | | Mo |
| | Ru | Rh | Pd | Sub-total | |
| Reference (LWR45) | 2.38 | 0.45 | 1.46 | 4.29 | 3.64 |
| High-burnup (LWR55) | 2.44 | 0.41 | 1.59 | 4.44 | 3.62 |
| / | ***1.03*** | ***0.91*** | ***1.09*** | ***1.04*** | ***0.99*** |
| Pu-thermal (MOX55) | 2.95 | 0.80 | 2.98 | 6.73 | 3.30 |
| / | ***1.24*** | ***1.77*** | ***2.04*** | ***1.57*** | ***0.91*** |

Increasing of burnup in LWR uranium fuel allows generation of fission products (FP) proportionally without any obvious difference on fractions of elements, which might influence on conditions of the current vitrification process such as noble metals (Ru, Rh and Pd) and molybdenum. Since FP content was adjusted by restrictions of both a waste loading ratio and heat generation of the vitrified waste fundamentally, the influence is expected to be negligible.

Contents of noble metal elements would be increasing remarkably in MOX recycle, opposite to that of molybdenum would be slightly decreased. Such changes would require more careful operation against

noble metal behavior. This result suggests that it is required to pursue R&D on noble metal in vitrification especially in the MOX fuel cycle.

Results of changes of heat generation at vitrification and at suggested disposal time (50 years storage is considered after vitrification) are shown in Table II. Increasing of burnup in LWR uranium fuel makes heat generation slightly enhanced at vitrification. Although the heat generation increases in actinide indicates remarkable, the influence is still small since the ratio in the whole radionuclide is small. At the time after 50 years cooling, high burnup effect becomes negligible.

Table II. Changes of Heat Generation of Canisters by High Burnup and MOX

| HLW | Heat, kW/canister | | | | | |
|---|---|---|---|---|---|---|
| | at Vitrification | | | after 50years | | |
| | Actinide | FP | total | Actinide | FP | total |
| Reference (LWR45) | 0.12 | 2.19 | 2.31 | 0.04 | 0.31 | 0.35 |
| High-burnup (LWR55) | 0.19 | 2.19 | 2.38 | 0.05 | 0.30 | 0.35 |
| / | ***1.53*** | ***1.00*** | ***1.03*** | ***1.18*** | ***0.98*** | ***1.00*** |
| Pu-thermal (MOX55) | 2.15 | 1.89 | 4.04 | 0.58 | 0.22 | 0.80 |
| / | ***17.64*** | ***0.86*** | ***1.75*** | ***13.50*** | ***0.73*** | ***2.29*** |

On the contrary, heat of HLW canister generated from MOX fuel cycle is remarkably increased due to higher heat generation by actinide both at vitrification and at 50years cooling. Such result may require modification of the storage capability and the disposal concept. When heat would be decreased at the level of 0.35kW, which reference HLW indicate after 50years cooling, over 100 years cooling storage may required for the HLW from MOX fuel cycle. Such result may influence on economical burden. It is suggested that the MA recovery process planned to be applied in FBR fuel cycle (see. Chapter 3.1) is also effected in the future MOX fuel cycle.

## CHARACTERISTICS OF HLW GENERATED FROM FUTURE FBR CYCLE SYSTEMS

### Advanced Aqueous Reprocessing System

Feasibility study on commercialized FBR cycle systems in Japan has been performed considering the design requirements for safety, economic competitiveness, reduction of environmental burden, efficient use of resources, and enhancement of nuclear non-proliferation. In the framework of Phase II (JFY2001-2005) study, two advanced aqueous reprocessing systems for oxide fuel, the new extraction system for TRU recovery (NEXT) and the supercritical fluid direct extraction process (Super-DIREX), and two pyrochemical systems, the oxide electrowinning for oxide fuel and the metal electrorefining for metallic fuel, have been designed conceptually by reflecting the latest results of research and development. As a

result of comprehensive evaluations on these systems from the viewpoints of the five design requirements; safety, economic competitiveness, reduction of environmental burden, efficient use of resources and enhancement of nuclear non-proliferation, the NEXT system was confirmed as the most promising candidate concept for industrial reprocessing of oxide fuel.

The NEXT system consists of a simplified solvent extraction process with the addition of a U crystallization step and a MA recovery process by the extraction chromatography. Salt-free reagents such as hydrazine oxalate and hydrazine carbonate are also adapted to easily decomposed and released as gas phase. A flow diagram of the NEXT is shown in Fig. 3 [3].

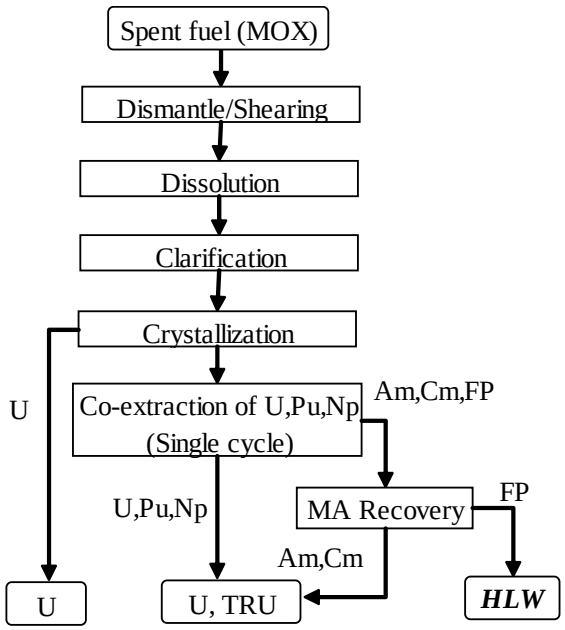


Fig. 3. Process flow diagram of the NEXT system

## HLW Generated from the Candidate FBR Recycle System

Effects and influences for the vitrification process and the HLW in the FBR fuel cycle system were evaluated by investigating the fuel composition using a reference core case for the composition of spent fuel (153 GWD/t) and a flow sheet of the NEXT system.

Results of changes of compositions of noble metals and molybdenum in HLW and heat generation at vitrification are shown in Table III and IV, respectively.

Its spent fuel produces fission products with slightly difference fraction from those of the reference HLW. Both compositions of noble metals and molybdenum indicate similar increase tendency shown in HLW from MOX fuel cycle (see chapter 2). Such changes would require more careful operation against noble metal behavior. And it is confirmed to be important to promote R&D on noble metal in vitrification process. Due to adding minor actinide (MA) recovery process, not only MA nuclides but also residual uranium and plutonium provided in the HLW are remarkably reduced. It enables heat and long-term toxicity of the HLW to be decreased, which are expected to reduce burden of the management and disposal of the HLW. Less containing actinide elements would also contribute to reduce the number/volume of the HLW.

Also, according to an ideal design basis of the flow sheet, no sodium salt is produced in the reprocessing plant. Such change has a high potential for reducing the number/volume of the HLW, since around half of the waste loading is originated by sodium nitrate in the current HLW. However, sodium salts would help for maintaining suitable viscosity and electric resistance of the melted glass. Thus, optimization of the vitrification process and/or modification of the composition of the glass would be required.

Additionally, it might be necessary to take into consideration regarding the content of corrosion products due to changes of the fuel cladding materials and the reprocessing process in the FBR fuel cycle system for detail evaluation.

Table III. Changes of Compositions of Noble Metals and Molybdenum by FBR Fuel Cycle

| HLW | Composition at vitrification, kg/canister | | | | |
|---|---|---|---|---|---|
| | Noble Metals | | | | Mo |
| | Ru | Rh | Pd | Sub-total | |
| Reference (LWR45) | 2.38 | 0.45 | 1.46 | 4.29 | 3.64 |
| FBR core (FBR153) | 3.18 | 0.94 | 2.62 | 6.74 | 3.36 |
| / | ***1.34*** | ***2.07*** | ***1.80*** | ***1.57*** | ***0.92*** |

Table IV. Changes of Heat Generation of Canisters by FBR Fuel Cycle

| HLW | Heat, kW/canister | | |
|---|---|---|---|
| | at Vitrification | | |
| | Actinide | FP | total |
| Reference (LWR45) | 0.12 | 2.19 | 2.31 |
| FBR core (FBR153) | --- | 1.68 | 1.68 |
| / | | ***0.77*** | ***0.73*** |

## HLW Generated from an Evolutional Fuel Cycle Concepts

A further evolutional recycle concept of FBR has been studied aiming to minimizing 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" (Optimization by Removing Impedimental Elements) [4]. "Unnecessary elements" to be removed were listed up from various aspects such as core design, front end and back end of the fuel cycle. In this concept, stable fission products that amount to about 60 wt% 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. To reduce burden for storage and/or disposal of the vitrified waste, separation of Cs and Sr, which indicate major heat elements in FPs, was considered. Also, noble metals and molybdenum are also removed from the vitrified waste in the cycle. The grouping pattern of the ORIENT cycle is illustrated in Fig. 4. Only four elements were identified to be vitrified; samarium, europium, zirconium, and silver. Since these elements have long half lives, vitrification technology can be assigned as adequate solidification system to be applied in the cycle. Consequently, volume of vitrified waste in the ORIENT cycle is expected to be reduced by a factor of around 10 compared with a conventional cycle.

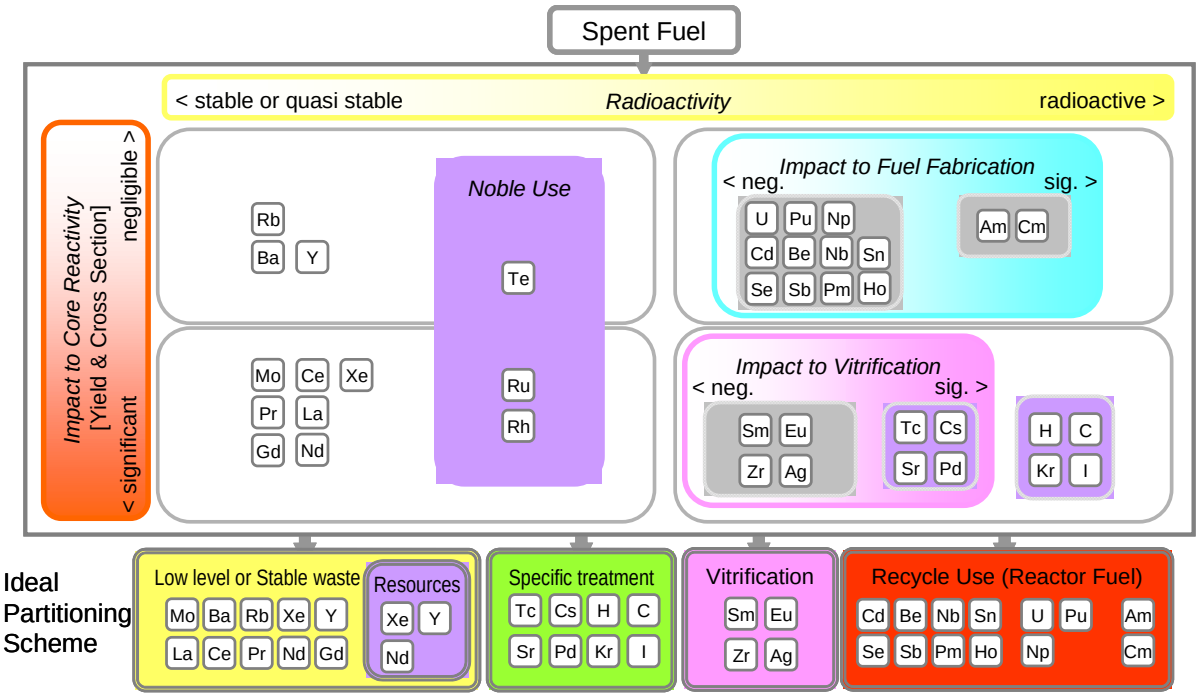


Fig. 4. Ideal categorization of elements considered in the ORIENT cycle

## CONCLUSION

The current developed vitrification technology is expected to have a potential for applying to the solidification process of the HLW generated from future fuel cycle systems with some modification/optimization of the melting condition, etc. Moreover, it is thought that the advance aqueous reprocessing system developed for future FBR cycle has the potential which can contribute to the further reducing the number/volume of the HLW. It is also confirmed that development efforts on countermeasure for accumulation of noble metals, which JAEA has been carrying out aiming to accomplishing more stable and reliable operation of the vitrification process and extending the melter's life, will be able to contribute in the future fuel cycle system furthermore. Moreover, since volume reduction of low level wastes is also one of the important development items in the current reprocessing process, it might be adequate to consider possibility and flexibility of application of the vitrification technology to an advanced treatment process for low-level liquid waste with high condensation.

## REFERENCES

1 Aoshima, A., Kozaka, T., Tanaka, K., *Glass Melter Replacement and Melter Technology Development in the Tokai Vitrification Facility*, Proc. of ICONE-12, Virginia, USA, Apr. 25-29, pp28-33 (2004).

2 Croff, A.G. (1980). *ORIGEN2 –A Revised and Updated Version of the Oak Ridge Isotope Generation and Depletion Code*, Ork Ridge National Laboratory, ORNL-5621

3 Sato, K., Koma, Y., et al., *Conceptual Design Study and Evaluation of Advanced Reprocessing Plants in the Feasibility Study on Commercialized FR Cycle System in Japan*, Proc. of Global 2005, Tsukuba, Japan, Oct. 9-13, Nr. 502 (2005).

4 Nagaoki, Y., Nomura, K., Nakajima, Y., Ogata, T., Namba, T., *Technical Approach of the Reasonable Nuclear Energy System for a New Era*, Ttans Am Nucl Soc. Vol. 91, pp.529-530, 2004