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Nuclear energy and waste management – pyroprocess for system symbiosis

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Abstract

The actinide management has become a key issue in nuclear energy. Recovering and fissioning transuranium elements reduce the long-term proliferation risks and the environmental burden. The better way of waste management will be made by system symbiosis: a combination of light-water reactor and fast reactor and/or accelerator-driven transmutation system should be sought. The new recycling technology should be able to achieve good economy with smaller plants, which can process fuels from different types of reactors on a common technical basis. Ease in handling the higher heat load of transuranium nuclides is also important. Pyroprocesses with the use of molten salts are regarded as the strong candidate for such recycling technology. In JAEA, the first laboratory for the high-temperature chemistry of Am and Cm has been established. The fundamental data will be combined with the computer code for predicting the molten-salts electrolytic processes.

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1. Introduction

The actinide management has become a key issue in nuclear energy due to increasing proliferation concern and long-term environmental perception. Partitioning and transmutation (P–T) of transuranium elements, particularly plutonium, reduce the long-term proliferation risks and the environmental burden. However, it apparently costs less to dispose of the spent fuel without reprocessing [1]. On the other hand, the economical penalty of P–T may not be prohibitively large, considering relatively small impact of the fuel cycle cost on the electricity generating cost.

According to OECD/NEA [1], the likely cost of nuclear fuel cycle is 4.28–6.30 mills/kW h (1991 money) for the case of direct disposal of spent fuel; 5.17–7.06 mills/kW h for the case of reprocessing/recycling with disposal of vitrified high-level waste. The fuel cycle cost is about 15% of the cost of electricity production. Recent analyses indicate that the introduction of P–T may double the fuel cycle cost [2], which will result in the increase of the cost of electricity production by about 15%. Rather the

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policy makers and power companies have become aware of the financial risks of installing a huge capacity of recycling technology. At the same time, a plant of huge capacity would fix the choice of nuclear technology of a nation for a long time.

The past experience in the reactor technology development seems to indicate that no single type of reactors, however successfully it may be developed, would completely replace the existing reactors, which has reached a mature stage with good economical returns. In the foreseeable future, uranium would not run short, although price swings may be inevitable. In such situation, the better way of waste management will be made only by system symbiosis: a combination of LWR and fast reactors (FR) and/or accelerator-driven transmutation systems (ADS) should be sought. The nuclear fuel cycle, which incorporates this type of waste management, has been called 'double-strata' fuel cycle [3].

The requirements for the new recycling technology should be, therefore:

- Achieving good economy with smaller plants.
- Capable to process fuels from different types of reactors on a common technical basis.
- Ease of handling higher heat load of transuranium nuclides (Pu-238, Cm-244, etc.).

Pyroprocesses with the use of molten salts are regarded as the strong candidate for such recycling technology. Recent efforts in developing pyroprocesses for nuclear fuels in Japan will be discussed.

In JAEA, the first laboratory for the high-temperature chemistry of transuranium elements, mainly Am and Cm, has been established. The fundamental data on the molten-salt chemistry of transuranium oxides and nitrides will be combined with the computer code for predicting the molten-salts electrolytic processes. These studies will be supplemented by the molecular dynamics simulation of relevant molten-salts systems.

2. Management of transuranium elements in symbiotic system

A light-water reactor of 1 GWe discharges 30 t of spent fuels each year. Those spent fuels contain 0.2 t of plutonium and 0.02 t of minor actinides (MA: neptunium (Np), americium (Am) and curium (Cm)), depending on the fuel burnup and the cooling time after the fuel discharge. There are industrial experiences in handling large mass of plutonium, but nearly nothing about MA. In view of fuel cycle technology, larger decay heat and higher fast-neutron emission of MA pose significant difficulty, particularly in fabrication and transportation of fuel assemblies. According to Pillon [4], adding 1.2% of MA to the MOX fuel with 20% plutonium for a fast reactor will increase the decay heat by a factor of 5–6 and the fast-neutron source strength by three orders of magnitude.

When we cannot expect that the nuclear energy constantly expands in near future in Japan, we can assume at best that the change is very gradual. Realistically, we assume that nuclear power capacity is capped at a constant value, say 70 GWe in Japan. Actually the industry rather predicts that it saturates at even lower value. In this situation, the problem of managing plutonium and minor actinides may be solved by a symbiotic system shown in Fig. 1.

The system in Fig. 1 manages plutonium and minor actinides in separate strata of a fuel cycle. The LWR is gradually replaced by FR with low breeding ratio, which utilizes plutonium for power production. The ADS is deployed to manage MA and optionally long-lived fission products. The second stratum of the fuel cycle dedicated to MA burning is one hundredth in terms of annual heavy-metal mass compared with the first stratum of commercial fuel cycle. With ADS of 800 MW t/unit, 9 units for all LWR case and 16 units for all FR case would be required to support 70 GWe (~70 units) of commercial power production.

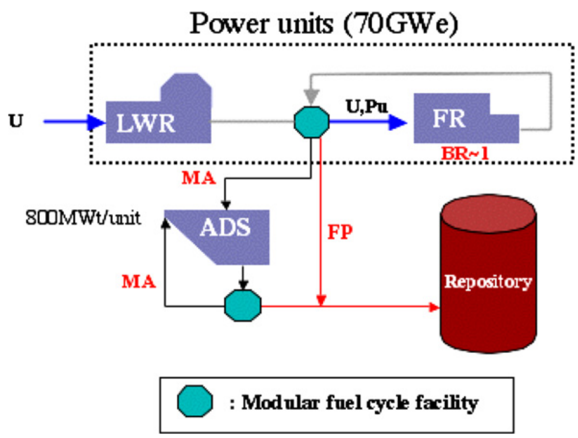

Fig. 1. An example of symbiotic system consisting of power reactors, where LWR is gradually replaced by FR, and ADS.

The evolution of this symbiotic system and its effect on the MA management are shown in Fig. 2. In this model calculation, the nuclear power is assumed to saturate at 70 GWe in 2020. The FR begins to replace LWR from year 2040 by one unit every two years. In this model case, the water-cooled fast reactor is assumed for calculating the system mass balance. The ADS is introduced from year 2040 by one unit every two years [5].

Minor actinides may be managed along with plutonium in a type of fast reactor. But considering the difficulty of handling MA, it may be better managed in a different stratum of the fuel cycle, which is smaller by two orders of magnitude than the first-stratum fuel cycle utilizing plutonium. The symbiotic approach does not exclude either option.

The key to realize this symbiotic approach is the recycling technology. Weakness of current PUREX reprocessing is that it relies too much on the economy of scale. Existing commercial plants are designed with a capacity of 800–1200 t HM/y. The drawback of this huge-plant approach is, besides the financial risk for the private investment, is to fix the nuclear technology for a long period of time. The huge reprocessing plant is inevitably the first-of-a-kind and the last-of-a-kind design for almost a half century. It also fixes the choice of the nuclear power system; flexible symbiotic approach becomes out of question.

What is needed is a recycling process that can be economical even at a smaller production scale. Modular plants based on such a process may be also improved by learning-by-doing beyond the first-of-a kind construction. At present we cannot say that pyroprocess meets this ideal. At the same time there might be some aqueous processes that are suitable to the modular plant. The development of both types of processes, we believe, should be made with this aspect in view.

3. Research and development on molten-salts electrorefining in JAEA

The pyroprocess using molten salts as solvent is regarded as a strong candidate to handle the MA-bearing fuel in a small-scale plant combined with ADS. There are several types of MA-bearing fuels that are being studied. The state-of-the-art fuel technology for burning MA is reviewed in an OECD/NEA report [6]. In JAEA, the study on the ADS fuel focuses on the use of MA nitrides. Also JAEA has been participating in a joint study of pyro-processes with JAPC, TEPCO and Tohoku Electric Power for MOX (mixed oxides) fuels, and with CRIEPI for metal fuels. These pyroprocesses are based on the molten-salts electrorefining. Those for metal and nitride fuels are very similar with each other.

The research program in JAEA aims at improving predictive capability of the behavior of transuranium elements in molten-salts electrorefining. The greater efforts are directed to (1) install a capability to study the high-temperature chemistry of MA, (2) collecting structural data of molten salts and (3) develop the technology for process simulation. The structure of the study is depicted in Fig. 3. The experiments with MA are made in a dedicated

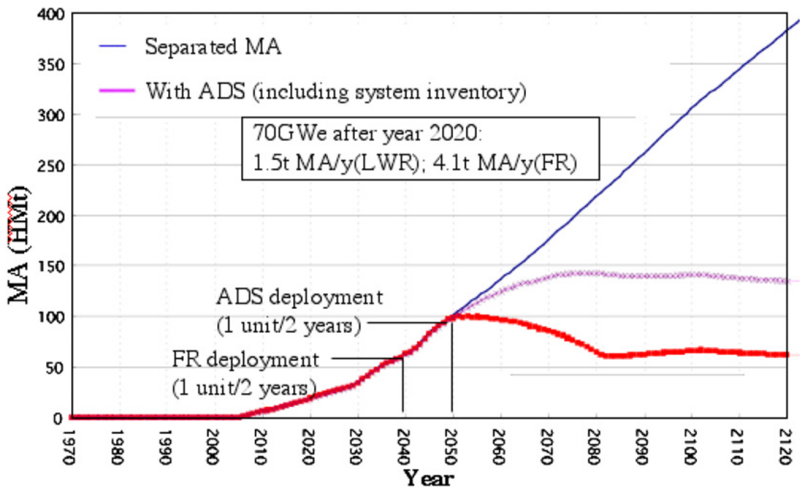

Fig. 2. The evolution of symbiotic system as depicted in Fig. 1.

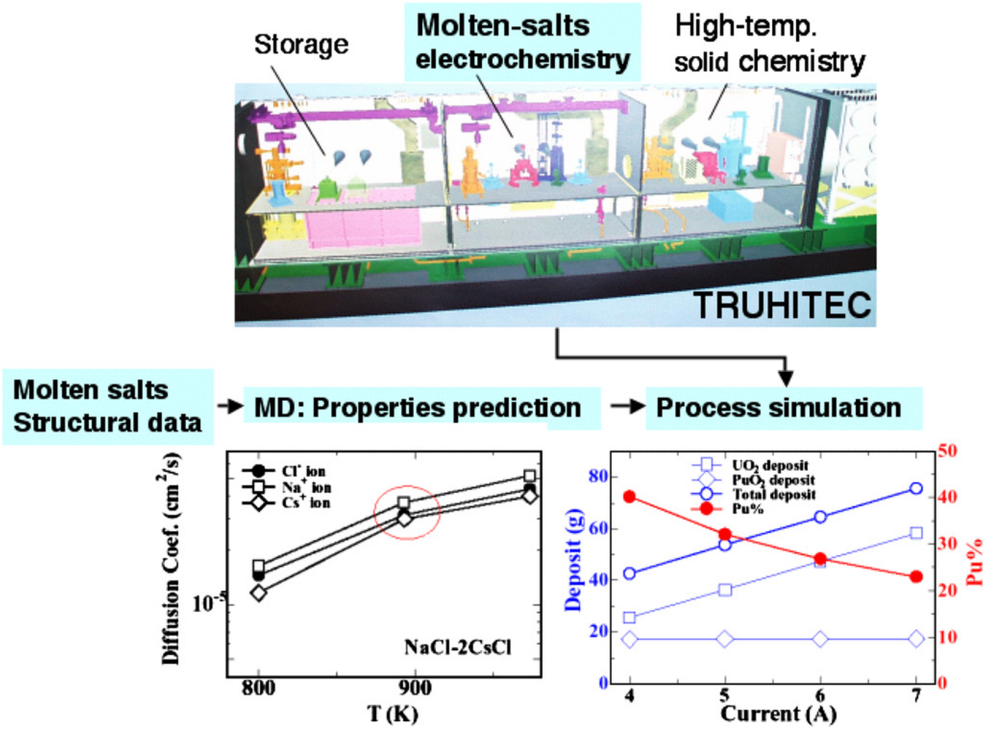

Fig. 3. Structure of pyroprocess study in JAEA [7].

facility, TRUHITEC. The data are employed in the process simulations. The experimental data are also complemented by the molecular dynamics simulations (MD), which generate physical properties of molten salts. Thus, the simulation consists of two parts:

- Property prediction by a massive MD with structurally optimized potential models.
- Process simulation of electrorefining based on the local equilibria at electrodes and the diffusion in boundary layers. Process simulation program is being expanded to handle two-dimensional geometry.

So far, the structural data on molten salts are limited to those of binary chlorides with uranium or lanthanides. It is hoped in future to install a capability in SPring-8, 8 GeV synchrotron orbital ray, to handle molten salts containing MA.

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