Development of Pyro-processing Fuel Cycle Technology for Closing Actinide Cycle

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Abstract

Pyro-processing fuel cycle technology is a promising technological candidate to realize advanced fuel cycle with reduced burdens of proliferation risk and long life radioactive waste. CRIEPI have been studying pyro-processing and metal fuel technology since 1986 for obtaining basic properties, process chemistry and engineering technologies. In this century, CRIEPI and JRC-ITU have jointly demonstrated the recovery of MAs from both irradiated metal fuels and spent MOX fuels. Since 2009, accumulation of the design basics through tests with engineering-scale pyro-processing equipment is carried out by CRIEPI. According to the results obtained in these studies, feasibility of pyro-processing fuel cycle technology was evaluated through estimation of Technology Readiness Level and cycle cost based on the design study of fuel cycle facility of 40 tonHM/y throughput.

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1. Introduction

The importance of safe and peaceful use of nuclear energy shall be kept unchanged as carbon free energy source to meet future energy demand in the world. Accordingly, the development of innovative nuclear fuel cycle technology that has the advantages of economic and safety power generation in
addition to proliferation resistance is strongly expected. Pyro-processing fuel cycle technology is studied in many countries for realizing advanced fuel cycle with reduced burdens of proliferation risk and long live radioactive waste [1-2]. Hence Japanese government has recognized the metal fuel cycle which consists of metal fueled fast breeder reactor (FBR) and pyro-reprocessing as a “sub-concept” of FACT project for commercialization of FBR fuel cycle in mid-21st century[3], because the metal fuel cycle has a potential advantages to MOX fuel cycle that consists of mixed oxide fueled FBR and advanced aqueous reprocessing technology (NEXT) in the core performances such as breeding ratio and fuel cycle cost. The metal fuel cycle technology will increase flexibility in the future nuclear deployment. For example, partitioning and transmutation (P&T) scenario for the long-lived nuclides in high level waste from aqueous reprocessing of light water reactors and double strata concept with nitride fuel cycle[4] can be realized by applying the pyro-reprocessing technologies of metal fuel cycle concept.

Central Research Institute of Electric Power Industry (CRIEPI) has been studying pyro-processing and metal fuel technology since 1986 with domestic and international collaborations[5]. Since 1994, CRIEPI and Japan Atomic Energy Research Agency (JAEA) have jointly started studying basics of actinide behaviour in pyro-process, and expanded the joint study to carry out the integrated pyro-processing test and the metal fuel fabrication test for irradiation in JOYO reactor[6]. Since 2009, accumulation of design data through tests with engineering-scale pyro-processing fuel cycle equipment is underway as a project entrusted from Japanese government (MEXT)[6]. In the joint study between CRIEPI and JRC-ITU, the irradiation integrity of MA containing metal fuels up to 10 at.%BU and the recovery of MAs from both irradiated metal fuels and spent MOX fuels have been demonstrated[6]. According to the results obtained in these studies, pyro-processing fuel cycle technology was recognized as a promising option for FBR Cycle and Partitioning & Transmutation of MAs. After the Fukushima daiichi accident, applicability of pyro-processing to treat the damaged spent fuel debris is under study[6]. In this paper, the technological achievements of these studies are overviewed.

2. Process Chemistry Development

2.1. Measurement of Basic Properties

As the basics of pyrochemistry, standard potentials and thermodynamic properties have been assessed for relevant elements such as actinides elements and FPs. We recommend the values in Table I as the most reliable standard potentials and reduction potentials of actinides and lanthanide elements in LiCl-KCl[7]. Excepting the reduction potentials of Pu and Np to Cd cathode, the potentials in this table were all measured by ourselves.

In addition to the electrochemical potentials, equilibrium distribution factor is an important basic data for

<table>
<thead>
<tr>
<th>Inert electrode</th>
<th>Standard potential $V_{\text{Cl}/\text{Cl}^-}$</th>
<th>Cd electrode</th>
<th>Reduction potential $V_{\text{Cl}/\text{Cl}^-}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>U(III)/U(0)</td>
<td>-2.468</td>
<td>U(III)/U-Cd</td>
<td>-2.557</td>
</tr>
<tr>
<td>Np(III)/Np(0)</td>
<td>-2.674</td>
<td>Np(III)/Np-Cd</td>
<td>-2.580</td>
</tr>
<tr>
<td>Pu(III)/Pu(0)</td>
<td>-2.773</td>
<td>Pu(III)/Pu-Cd</td>
<td>-2.564</td>
</tr>
<tr>
<td>Am(III)/Am(0)</td>
<td>-2.327</td>
<td>Am(III)/Am-Cd</td>
<td>-2.576</td>
</tr>
<tr>
<td>Cm(III)/Cm(0)</td>
<td>-2.990</td>
<td>Cm(III)/Cm-Cd</td>
<td>-2.665</td>
</tr>
<tr>
<td>Fr(III)/Fr(0)</td>
<td>-3.040</td>
<td>Fr(III)/Fr-Cd</td>
<td>-2.631</td>
</tr>
<tr>
<td>Np(III)/Np(0)</td>
<td>-3.047</td>
<td>Np(III)/Np-Cd</td>
<td>-2.633</td>
</tr>
<tr>
<td>Ce(III)/Ce(0)</td>
<td>-3.038</td>
<td>Ce(III)/Ce-Cd</td>
<td>-2.636</td>
</tr>
<tr>
<td>Y(III)/Y(0)</td>
<td>-3.068</td>
<td>Y(III)/Y-Cd</td>
<td>-2.753</td>
</tr>
<tr>
<td>La(III)/La(0)</td>
<td>-3.103</td>
<td>La(III)/La-Cd</td>
<td>-2.661</td>
</tr>
</tbody>
</table>

* The reduction potentials of elements in LiCl-KCl eutectic salt into liquid cadmium alloy were derived when the concentration of the elements in the salt and in the liquid cadmium are the same (0.001 mole fraction).
predicting separation behavior in molten salt / liquid metal extraction process. Figure 1 shows the distribution factors of the actinide and lanthanide elements between molten LiCl-KCl salt and liquid cadmium obtained by the joint study between CRIEPI and JRC-ITU[8]. The data agreed well with literature values, and suggests the possibility to separate Am and Cm from other lanthanides.

2.2. Process Chemistry Development

In order to develop process flowsheet reliable to evaluate an industrial applicability, experiments to evaluate detail material balance of the elements have been carried out with using unirradiated fuels and irradiated fuels. As a joint study between CRIEPI and JRC-ITU, tests to recover actinides from irradiated fuels are underway using Ar atmosphere hot cell shown in Fig. 2. Electroreduction of LWR irradiated MOX fuels followed by recovery of U and Pu-MA-U with electrorefining was carried out as Fig. 2[9] This is the first demonstration of MA recovery from spent LWR-MOX, which leads to reduction of the environmental burden of radioactive waste.

As for the irradiated metal fuels, U-Pu-Zr with minor actinides, U-19Pu-10Zr with 2%MA and 2%RE, with 5%MA and 5%RE, and without MA and RE, irradiated at PHENIX reactor were served for the electrorefining experiments. Recovery of actinides has been demonstrated with keeping reliable material balance. Separation of actinides from lanthanides agreed well with the experiments with unirradiated materials. Another important finding was that the behavior of insoluble materials in the anode residue shown in Fig. 3. Behaviour of noble metal follows that of zirconium as predicted from the measured data[10].

3. Engineering-scale Fuel Cycle Tests

Reduced fuel cycle cost of the pyro-reprocessing can be attained even in a small-scale plant of a few tons per year (t/y) capacity, which is not the case with the aqueous process. A high capacity plant is realized by a multiple installation of the process equipment. The demonstration of an engineering-scale
equipment of a few t/y capacity is, therefore, a key step toward the commercialization of the pyro-reprocessing. According to the development described in former sections, integrated tests to demonstrate the technological feasibility of pyro-reprocessing has been entrusted from government. The main process apparatuses – reduction to metal, electrorefining, solid cathode processing, liquid Cd cathode processing, 6 stages counter current contactor and fuel casting – equivalent to 1 t-HM/y capacity were designed and manufactured. Repeated recycle tests using uranium is underway with the apparatus shown in Fig. 4 for demonstrating the system integrity and reliability[11]. Through the test, the equipment performance, the amount and characteristics of the uranium loss, and the additional treatment possibly required in the practical process flow sheet, are estimated. As sequential operation of the steps starting from the electroreduction and finishing with the injection casting, which corresponds to introduction of the current LWR oxide fuels to the metal fuel fast reactor cycle, has been completed. Currently, repeated cycle operation for the U-Zr fuel is in progress, which simulates the closed metallic fuel fast reactor fuel cycle. Consequently, simulated metallic fuel (U-Zr alloy rods) was successfully fabricated using UO₂ as the starting material. The electrorefining, product transfer, salt distillation and injection casting equipment were operated satisfactorily as shown in Fig. 5 & 6. However, regarding the electroreduction, problems like the stagnancy of Li₂O and the parasitic generation of lithium metal at the cathode emerged with increase of the equipment capacity. Consequently, the total amount of uranium charged in the whole processes agreed well with that recovered in the fuel casting process[10].

Fig. 4 Engineering-scale fuel cycle test apparatuses installed in two Ar glove boxes.

Fig. 5 Cathode deposit obtained in electrorefining

Fig 6 Injection casted U-Zr alloy rods
4. Evaluation of Feasibility of Pyro-processing Technology

4.1. Technology Readiness Level

The technology readiness level (TRL) developed by U.S. NASA for the selection of the space technology is modified to apply to the evaluation of the maturity of the pyro-reprocessing fuel cycle. The technology readiness level (TRL) developed by U.S. NASA for the selection of the space technology is modified to apply to the evaluation of the maturity of the pyro-reprocessing fuel cycle technology.

Table 2 shows the definition of TRL for the metallic fuel cycle compared with NASA’s original one. In the definition, TRL is increased as the technology is getting matured and becomes 9 when it is widely used as commercial plant. It becomes 5, when the technology is confirmed to work in the engineering scale. Similar evaluation was performed in GNEP by U.S. DOE and in Japan, for the comparison of separation and transmutation technologies. According to the pioneering work by US (ANL and INL) and our works described above, TRL of each process component was evaluated.

Technologies for electrorefining, cathode processing, metal waste consolidation and injection fuel casting are evaluated as TRL=5, where validation of the process by small scale tests with spent fuels and engineering-scale test with U or simulants are completed. On the other hand, the technologies for TRU extraction, zeolite column and salt waste consolidation are evaluated as TRL=4, where small tests with U, Pu, MA and FP simulants have been completed. Hence the necessary technology to be developed for engineering-scale hot tests is to design reliable equipment adapted for remote operation for TRU extraction, zeolite column and salt waste consolidation.

4.2. Design and cost estimates of Fuel Cycle Facility

With the updated flowsheet and technological developments, conceptual design of the pyro-processing fuel cycle facility for 40 tHM/y throughput of metal fuel was carried out. In addition to detailed design of each apparatus, safety issues such as loss of cooling system were incorporated to the design consideration. As shown in the bird’s eye view in Fig. 10, the designed facility has a size of 50mW x 103mL x 34mH, and consists of 4 floors. Hot cells for fuel fabrication apparatuses and waste treatment apparatuses were
installed in the basement. Hot cells for the reprocessing apparatuses such as two electrorefiners and 6 cathode processors were installed in 1st floor along with in-cell transport equipment and stock areas, while a 6-staged contactor and two zeolite columns were installed in the 2nd floor. Utilities were located in the 3rd floor. Based on this design, capital cost of this facility was roughly estimated, and relatively low fuel cycle cost was obtained; 500,000 yen/kgHM for reprocessing and 280,000 yen/kgHM for fuel fabrication. As the construction cost of the Metal Fuel FBRs is expected to be the same as that of the Oxide Fuel FBRs which is reported about 1.9 yen/kWh [16], the total cost of electricity from the Metal Fuel FBRs and its Fuel Cycle facility is expected to be about 2.7 yen/kWh which shows better potential than the other energy sources.

5. Conclusion

Recent progress of pyroprocessing fuel cycle technology is summarized. Successful results were obtained for both the small-scale experiments with irradiated materials and the engineering-scale tests with uranium or simulants. Conceptual design study of a fuel cycle facility of 40 tHM/y capacity showed feasibility to meet process requirements and safety requirements within acceptable fuel cycle cost. Any technological problems impossible to be overcome have not been found yet. Consequently, necessary data for the next step, engineering-scale experiments with irradiated materials, have almost been assessed.

Acknowledgements

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