

Material Attractiveness Evaluation of Actinides in Pyroprocessing Facility for Partitioning and Transmutation Cycle

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ABSTRACT

Research and development of partitioning and transmutation (P&T) cycle with accelerator-drive systems (ADSs) transmuted minor actinides (MAs) separated from the commercial cycles has been continuously conducted to reduce the high-level radioactive waste (HLW) contained in spent fuel discharged from nuclear power plants. The Japan Atomic Energy Agency has proposed a pyrochemical process for reprocessing ADS spent fuel with high decay heat and radioactivity due to the large amount of MA. Since the chemical form and composition of the fuels are different from those of the current commercial cycles, it is necessary to examine the inspection goal of the safeguards (SGs) and the design level of physical protections (PPs) which are required for the P&T cycle. In this study, the material attractiveness was evaluated assuming the diversion of the Cd cathode and the nitride powder from the pyroprocessing in terms of non-proliferation. Additionally, they were compared with the material attractiveness of the MOX fuel assemblies (fresh and spent fuels) for a conventional boiling water reactor (BWR). The Cd cathode used to recover actinides from ADS spent fuel by molten salt electrolysis in the pyroprocessing facility of P&T cycle was less attractive than the MOX fuel assembly for the BWR because the Cd cathode included ²³⁸Pu having high decay heat. The nitride powder electrorefined from the ADS spent fuel was also less attractiveness than the MOX fuel assembly for the BWR because of the same reasons of the Cd cathode.

INTRODUCTION

Research and development of partitioning and transmutation (P&T) cycle with accelerator-drive systems (ADSs) transmuted minor actinides (MAs) separated from the commercial cycles has been continuously conducted to reduce the high-level radioactive waste (HLW) contained in spent fuel discharged from nuclear power plants [1-5]. The Japan Atomic Energy Agency has proposed a pyrochemical process for reprocessing ADS spent nitride fuel with high decay heat and radioactivity due to the large amount of MA.

In the pyroprocessing for the ADS spent nitride fuel, the electrorefining step is carried out using a LiCl-KCl eutectic melt at 773 K to recover actinides (Ans) separated from the fission products (FPs), which is based on the latest electrorefining design for reprocessing of spent metal fuel [6]. A basket filled with the ADS spent nitride fuel serves as an anode. A liquid Cd is used as

cathode to recover Ans. Ans recovered in the liquid Cd cathode via electrorefining are converted to nitride by heating in a $^{15}\text{N}_2$ gas stream [7,8]. Ans nitride powder produced via the electrorefining is mixed with MA nitrides fabricated from the partitioned MA generated in a commercial reactor fuel cycle and used as the ADS fuel for the next burn-up cycle [9].

Since the chemical form and composition of the fuels are different from those of the current commercial fuel cycles, it is necessary to examine the inspection goal of the safeguards (SGs) and the design level of physical protections (PPs) which are required for the P&T cycle. In this study, the material attractiveness was evaluated assuming the diversion of some items in the pyroprocessing facility. On the basis of the results, quantitative components based on the fundamental material property were created as an important factor for deciding the inspection goal for SGs and the design level for PPs required for the pyroprocessing facility. In addition, each material attractiveness of some items in the pyroprocessing facility was compared with that of the MOX fuel assemblies (fresh and spent fuels) for a conventional boiling water reactor (BWR).

EVALUATION METHOD

The material attractiveness evaluation for nuclear non-proliferation was conducted for the inherent features of the ADS fuel in pyroprocessing using the method developed by Aoki et al [10], which had been extended based on that for nuclear security developed by Bathke et al [11]. Here, it was assumed that the adversary would eventually achieve the manufacture of an NED made of pure Pu (Pu-NED). Additionally, the attractiveness of mixed oxide (MOX) fuel assemblies stored in the commercial boiling water reactor (BWR) facility for the Pu-NED manufacturing case was evaluated for comparison with that of the materials in the pyroprocessing. State actor was assumed as an adversary in this study to evaluate the material attractiveness in terms of non-proliferation. For material attractiveness assessment for state actors, a proliferating state intending to divert the safeguarded nuclear material into Pu-NED was assumed to have the following characteristics: (1) having advanced technology, well-developed industries, and abundant capital; (2) having no natural U resources; (3) accepting of the Non-Proliferation Treaty, comprehensive safeguards agreement, and additional safeguards agreements; (4) requiring 50% reliability for the NED; and (5) requiring production of one NED [12,13]. In the evaluation for state actors, the physical properties of important nuclear materials were evaluated in each phase, i.e., processing and utilization, as shown in Figure 1. In the processing phase, the conversion times defined by IAEA [14] were used as indicators for the evaluation in the processing phase. The indicators of the utilization phase were the bare critical mass (BCM), heat content per BCM (HC), and the spontaneous fission neutron per BCM (SFN). The BCM was calculated using the Monte Carlo calculation code MCNP-6.1.1 [15] with the nuclear data library JENDL-4.0 [16] as well. The MCNP calculations employed 1,050 generations of neutrons, with 500,000 histories per generation. The first 50 generations were excluded from the statistics for each case, yielding

500,000,000 active histories in each calculation. The standard deviation with respect to the multiplication factor was 0.03%. For the radius, the number of effective digits after the decimal point was set as 3, and the case exceeding the criticality was surveyed while increasing the radius in increments of 0.001 cm. The BCM was calculated using the radius immediately before the multiplication factor exceeded 1.00001. The HC and SFN were obtained from calculation using the isotope generation and depletion calculation code ORIGEN2.2 [17] with the set of cross section library ORLIBJ40 [18]. Table 2 present a metric of categorization for the material attractiveness for state actors [10,11,19]. The metric of attractiveness of SFN was defined with thresholds of 10% and 30% of the doping fraction of ^{240}Pu to ^{239}Pu [19].

In this study, the evaluation target items were Cd cathode and nitride powder including Ans. The Pu compositions of the ADS burnup calculation result [20] were used for the material attractiveness evaluation regarding the Pu-NED. The isotopic ratios of ^{238}Pu and ^{239}Pu reached equilibrium after the 5th cycle, and there was no significant fluctuation. Thus, the Pu isotopic fractions of the 1st cycle of the launching stage and the 5th cycle in which the composition was in the equilibrium stage were the attractiveness evaluation targets of the Pu-NED (Table 1). In addition, the MA and rare-earth elements (REs) are always conducted in fuel component during pyroprocessing. So, the removing the MA and RE are also evaluation target in processing phase.

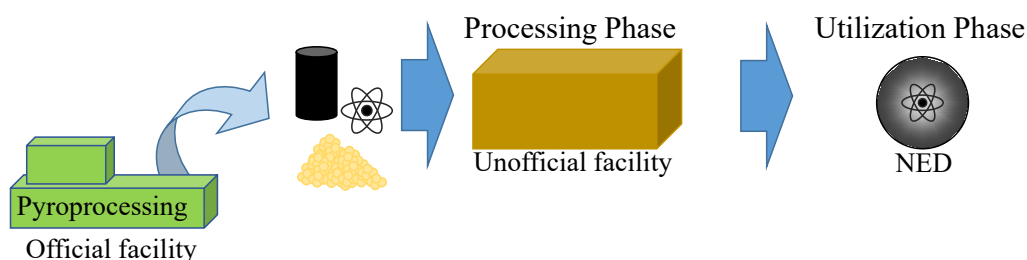


Figure 1. Processing and Utilization phases.

Table 1. Pu isotopic fraction [20].

Nuclide	Atomic ratio (at.%)	
	Spent fuel of 1 st cycle	Spent fuel of 5 th cycle
Pu-238	26.2	49.6
Pu-239	35.8	15.0
Pu-240	24.1	23.4
Pu-241	5.8	2.2
Pu-242	8.1	9.7

Table 2. Metrics of categorization for material attractiveness for state actors [10,11,19].

Class	Processing phase	Utilization phase		
	Conversion time	BCM (kg)	HC* (kW/BCM)	SFN* (neutrons/s/BCM)
1 (Preferred)	1 week (Non-irradiated metal)	<80	<0.45	$<9.89 \times 10^5$
2 (Potentially)	1–3 weeks (Non-irradiated compound)	80–800	0.45–4.5	$9.89 \times 10^5 - 3.58 \times 10^6$
3 (Impractical)	1–3 months (Irradiated material)	800– 4000	>4.5	$>3.58 \times 10^6$
4 (Impossible)	3–12 months (Low enriched U)	>4000		

* HC and SFN are indices calculated under the assumption of the α phase.

RESULTS

The evaluation results for the material attractiveness for nuclear non-proliferation are presented in Table 3 [21]. First, the processing times and complexities of the Cd cathode including components obtained by the ADS spent fuel of the 1st and 5th cycle were categorized into the same class as those of the BWR spent fuel. While alkali metals (ALs) and alkaline earth metals (ALEs) in FPs remained in the molten salt, REs and MAs are recovered in the Cd cathode. Extracting pure Pu from a mixture of RE and Ans requires complex processing steps whereas separation of Cd is relatively easy due to the difference in boiling point between Cd and the others. Second, the BCM of the Cd cathode including components obtained by the ADS spent fuel of the 1st and 5th cycle were categorized into the same class as those of the BWR fresh and spent fuels although there is small difference in exact BCM value owing to ²³⁸Pu. Third, the HC of the Cd cathode including components obtained by the ADS spent fuel of the 1st and 5th cycle were categorized into class 2, which was lower than those of the BWR fresh and spent fuels owing to the accumulation of ²³⁸Pu having a large decay heat. Fourth, the category of SFN of the Cd cathode including components obtained by the ADS spent fuel of the 1st and 5th cycle were class 3, which was same as those of the BWR fresh and spent fuels although there is small difference in calculated SFN value owing to ²³⁸Pu. Finally, the evaluation results of the nitride powder will be explained in the same way as the Cd cathode. The separation of nitrogen from nitride fuel is as complex as that of oxygen from MOX, but the extraction of pure Pu from materials containing REs and MAs requires more complex processing steps. So, the processing times and complexities of the nitride powder including components obtained by the ADS spent fuel of the 1st and 5th cycle were categorized into class 3, which is the same class as those of the BWR spent fuel. In the evaluation results of

the other indicators, BCM, HC, and SFN, the nitride powder electrorefined from the ADS spent fuel of the 1st and 5th cycle presented the same tendency as the Cd cathode for the same reasons. Therefore, in terms of the nuclear non-proliferation regarding Pu, each material attractiveness of the Cd cathode and the nitride powder electrorefined from the ADS spent fuel was smaller than that of the BWR.

Table 3. Evaluation results for the material attractiveness for nuclear non-proliferation [21].

Theft target			Processing phase	Utilization phase		
Facility	Chemical form of theft item	Cycle	Processing time and complexity	BCM (kg)	Heat content (kW/BCM)	SFN (neutron /sec/BCM)
BWR	MOX	(Fresh)	[2]*	13.6 [1]	0.14 [1]	4.8x10 ⁶ [3]
PYRO	An-RE-Cd alloy	1st	[3]	13.5 [1]	2.03 [2]	1.4x10 ⁷ [3]
		5th	[3]	13.3 [1]	3.75 [2]	2.3x10 ⁷ [3]
	An-RE-N powder	1st	[3]	13.5 [1]	2.03 [2]	1.4x10 ⁷ [3]
		5th	[3]	13.3 [1]	3.75 [2]	2.3x10 ⁷ [3]
BWR	MOX	(Spent)	[3]	15.8 [1]	0.30 [1]	8.3x10 ⁶ [3]

* The number in [] indicates the attractiveness class.

CONCLUSIONS

Considering the diversion of the Cd cathode and the nitride powder from the pyroprocessing facility, the material attractiveness was evaluated with regard to non-proliferation. On the basis of the result, quantitative components based on the fundamental material properties, such as BCM, HC, and SFN, were created as an important factor for deciding the inspection goal for SGs and the design level for PPs required for the pyroprocessing facility. In addition, the material attractiveness of the Cd cathode and the nitride powder in the pyroprocessing facility compared with that of the MOX fuel assemblies (fresh and spent fuels) for the BWR because the chemical form and composition of the fuels in P&T cycle were different from those of the current commercial cycles. The Cd cathode and the nitride powder were less attractive than the MOX fuel assembly for the BWR because they included a large amount of ²³⁸Pu having high decay heat.

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REFERENCES

- [1] H. Oigawa, K. Tsujimoto, K. Nishihara, et al., “Role of ads in the back-end of the fuel cycle strategies and associated design activities,” the case of Japan. *J. Nucl. Mater.* 415, 229–236 (2011). <https://doi.org/10.1016/j.jnucmat.2011.04.032>.
- [2] K. Tsujimoto, T. Sasa, K. Nishihara, et al., “Neutronics design for lead-bismuth cooled accelerator-driven system for transmutation of minor actinide,” *J. Nucl. Sci. Technol.* 41, 21–36 (2004). <https://doi.org/10.3327/jnst.41.21>.
- [3] K. Tsujimoto, H. Oigawa, N. Ouchi, et al., “Research and development program on accelerator driven subcritical system in JAEA,” *J. Nucl. Sci. Technol.* 44, 483–490 (2007). <https://doi.org/10.3327/jnst.44.483>.
- [4] K. Nishihara, K. Iwanaga, K. Tsujimoto, et al. “Neutronics design of accelerator-driven system for power flattening and beam current reduction,” *J. Nucl. Sci. Technol.* 45, 812–822 (2008). <https://doi.org/10.3327/jnst.45.812>.
- [5] K. Nishihara, S. Nakayama, Y. Morita, et al., “Impact of partitioning and transmutation on LWR high-level waste disposal,” *J. Nucl. Sci. Technol.* 45, 84–97 (2008). <https://doi.org/10.1080/18811248.2008.9711418>.
- [6] K. Kinoshita, T. Koyama, T Kobayashi, et al., “Material balance evaluation and plant design of pyroprocessing for metal fuel.” Komae-shi (Japan): Central Research Institute of Electric Power Industry; 2012. (Report No. L11009) [in Japanese].
- [7] H. Hayashi, H. Shibata, M. Akabori, et al., “Electrolysis of AmN in LiCl–KCl eutectic melts and reinitridation of Am recovered in liquid Cd cathode.” *Electrochemistry.* 2009;77:673–676.
- [8] Y. Arai, T. Iwai, M. Akabori, et al., “Synthesis of actinide nitrides in molten cadmium.” *Nucl Technol.* 2008;162:244–249.
- [9] H. Hayashi, T. Nishi, M. Takano, et al., “Recent progress and future R&D plan of nitride fuel cycle technology for transmutation of minor actinides.” Proceedings of the 13th Information Exchange Meeting on Actinide and Fission Product Partitioning and Transmutation; 2014 Sep 23–26, Seoul, Korea. p.370–377.
- [10] T. Aoki, H. Sagara, C. Y. Han., “Material attractiveness evaluation of inert matrix fuel for nuclear security and non-proliferation.” *Ann. Nucl. Energy* 126, 427–433 (2019). <https://doi.org/10.1016/j.anucene.2018.10.063>.
- [11] C. G. Bathke, N. Inoue, B. B. Ebbinghaus, et al., “Summary of a joint US-Japan study of potential approaches to reduce the attractiveness of various nuclear materials for Use in a nuclear explosive device by a terrorist group.” Sep 29-Oct 3, Salt Lake City (USA). Proc. Global 2013.
- [12] R. Bari, P. Peterson, J. Whitlock, et al., “Evaluation Methodology for Proliferation Resistance and Physical Protection of Generation IV Nuclear Energy Systems Revision 6.” The Generation IV International Forum, GIF/ PRPPWG/2011/003 (2011).
- [13] R. Bari, P. Peterson, I. Therios, et al., “PR&PP Evaluation: ESFR Full System Case Study Final Report.” The Generation IV International Forum, GIF/ PRPPWG/2009/002 (2009).
- [14] International Atomic Energy Agency, “IAEA Safeguards Glossary 2001 Edition.” International Atomic Energy Agency, International Nuclear Verification Series No.3 (2002).
- [15] T. Goorley, M. James, T. Booth, et al., “Initial MCNP6 Release Overview.” *Nucl. Technol.*, 180, 298–315; (2012) <https://doi.org/10.13182/NT11-135>.
- [16] K. Shibata, O. Iwamoto, T. Nakagawa, et al., “JENDL-4.0: a new library for nuclear science and engineering.” *J. Nucl. Sci. Technol.* 48, 1–30 (2011).
- [17] S. B. Ludwig, A. G. Croff, “Revision to ORIGEN2 – Version 2.2.” Transmittal memo of

CCC-0371/17, Oak Ridge National Laboratory (2002).

[18] K. Okumura, K. Sugino, K. Kojima, et al., “A Set of ORIGEN2 Cross Section Libraries Based on JENDL-4.0: ORLIBJ40.” Technical Report JAEA-Data/Code 2012-032 (2013).

[19] Y. Kimura, M. Saito, H. Sagara, et al., “Evaluation of proliferation resistance of plutonium based on spontaneous fission neutron emission rate.” Ann. Nucl. Energy 46, 152–159 (2012).

<http://dx.doi.org/10.1016/j.anucene.2012.03.032>.

[20] T. Sugawara, R. Katano, K. Tsujimoto, et al., “Impact of impurity in transmutation cycle on neutronics design of revised accelerator-driven system,” Ann. Nucl. Energy 111, 449–459

(2018). <https://doi.org/10.1016/j.anucene.2017.09.017>.

[21] A. Oizumi, H. Sagara et al., “Material attractiveness evaluation of fuels in pyroprocessing facility for partitioning and transmutation cycle for nuclear security and non-proliferation,”

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