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Linkage options between unit process to enhance proliferation resistance of pyroprocessing

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ABSTRACT

Pyroprocessing technology has been actively developed at KAERI as one of the options to address the national spent fuel management issue. As much as the unit process development itself, the linkage method between each unit process is also of importance. There may be different linkage options between a set of two process units, or different options caused by different technologies for one of two unit processes connected to each other. The linkage method may affect the loss of nuclear materials and distribution of fission products, which are essential from the safeguards and proliferation resistance points of view. In this study, different technology options for some of the major process steps and possible linkage options were identified, and aspects of their proliferation resistance including the impact on the safeguards are discussed. Several options for decladding and feeding to electro-reduction, dross and the residual salt treatment of reduced materials as well as feeding to electro-refining, and salt transfer from electro-refining to electro-winning were identified and their technical and proliferation resistance aspects were analyzed. This study does not conclude any ranking of each option or suggest which option is the best. It is important to note that the objectives to review the process and linkage options are to give the basic characteristics and technical information related to safeguards measures and proliferation resistance and to suggest a conceptual idea for certain options to investigate further and enhance those aspects.

1. Introduction

Pyroprocessing technology has been actively developed at KAERI as one of the options to address the national spent fuel management issue (Lee et al., 2013). This technology has an inherent advantage over an aqueous process in terms of proliferation resistance due to non pure plutonium stream, high heat and radioactivity, and compact batch mode equipment and operation. Since 2006, KAERI has carried out the concept development, bench scale testing, and demonstration of the laboratory scale key unit process. As much as the unit process development itself, the linkage method between each unit process is also of importance. It may have different linkage options between a set of two process units (e.g. between electro-refining and electro-winning), or different options caused by different technologies for one of the two unit processes connected to each other (oxidative decladding versus mechanical decladding followed by electro-reduction).

The linkage method may affect the loss of nuclear materials and distribution of fission products. This is essential from a safeguards and proliferation resistance point of view. In this study, different technology options for some of the major process steps (Lee et al., 2013; Williamson and Willit, 2011; NEA, 2012) and possible linkage options were identified, and their proliferation resistance aspects including the impact on the safeguards are discussed. Most of the discussion is in a qualitative manner, but based on quantitative characteristics, which is the concentration of each actinide and fission product, heat and radiation dose estimated by the ORIGEN code and MCNPX simulation using a reference PWR spent fuel (55GWd/MTHM of burn-up, 4.5% of initial enrichment, 10 years of cooling).

2. Linkage between decladding and electro-reduction

There are two applicable decladding technologies for the pyroprocess head-end process, oxidative decladding and mechanical decladding. Both have their own technical advantages and drawbacks. Oxidative decladding generates relatively fine spent fuel oxide powder with much fewer volatile fission products (Cs, Tc, I, etc). The decladding efficiency is more than 99% and nearly up to 100% depending on the operation temperature and fuel burn-up (Lee et al., 2013). Mass equivalent to 1 significant quantity of

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plutonium is about 770 kg of HM_3O_8, and the volume is in the range of 220–310 cm^3 at an estimated density of 2.5–3.5 g/cm^3. The decay heat is 1.9 W/kg and the dose rate at 1 meter from the surface of a 50 kg (batch)-HM is 290 rad/hr. Nuclear material from oxidative decladding is fine powder with a particle size range of tens of microns to millimeters (Westphal et al., 2012). Therefore, the critical issue for the next process step is to minimize the material loss.

Mechanical decladding results in crushed spent fuel with various sizes and hulls with a non-negligible amount of residual nuclear material. A mass equivalent to 1 significant quantity of plutonium is about the same with oxide powder from oxidative decladding, but the volume is half due to the high density (5.3–6 g/cm^3) of a crushed pellets. The decay heat is 2.1 W/kg and the dose rate at 1 meter from the surface of 50 kg (batch)-HM is 250 rad/hr. Even though it has slightly higher heat generation isotopes and gamma-ray emitters, the higher density makes the dose rate for the same amount of material slightly lower, and less volume for 1 SQ makes material handling easier. Critical issues from a safeguards point of view are the representativeness of sampling and minimizing the loss of fine materials into salt.

For both decladding technologies, there are five options to transfer nuclear materials to the electro-reduction process (Table 1).

2.1. Option 1: Pelletizing after oxidative decladding

The first option is to fabricate pellets in an appropriate size and feed them to the electro reduction step. The additional process step may be a drawback in terms of operation and economics, but it is the most practical solution to minimize the material loss into the LiCl salt in an electro-reduction system. KAERI considers this option seriously and performs relevant tests (Lee et al., 2013). From the stand point of proliferation risk, an additional process step can add a possible diversion point and needs monitoring. However, the benefit from decreased loss of nuclear materials will be greater than the additional monitoring. In particular, adapting a homogenizing process before the pellet fabrication will increase the accuracy of the input material accountancy (Budlong-Sylvester et al., 2003). The increased density of pellets makes the transfer volume into an argon cell decrease. A small transfer container is better for the operation and proliferation resistance.

2.2. Option 2: Mechanical decladding with partial voloxidation

The second option is for a mechanically de-cladded crush pellet. The first step is to select only large crushed material (>1 mm) and feed it into the electro-reduction process. The other fine materials and residue materials on the hulls can be fabricated into a pellet form after low temperature voloxidation. This also needs an additional process step and equipment, which is technically the same as oxidative decladding, but the operation frequency may be much less or the equipment size much smaller. The material accounting scheme can be more complicated due to the divided inventory measurement points of the input feed material with a different oxide factor of the original spent fuel (HMO_2) and voloxidized powder (U_3O_8). Even though there is a chance to obtain a representative sample of voloxidized powder by homogenizing, it is small fraction of the material processed. The majority of decladded material still needs a better way to take representative sample or to account by code calculation.

In particular, this option can be modified as below:

(1) Mechanical decladding of a flat burn-up region in the axial direction of spent fuel (65–70%): homogeneous with the burn-up measurement signature (Cs-134, 137).
(2) Voloxidation of both ends of the fuel rods (30–35%), hull residual and fine materials: subject to homogenizing and pelletizing.

Modified option can make the homogenization process much smaller than Option 1, keeping two separated homogeneous strata to enhance the input accountability.

2.3. Option 3: Mechanical decladding with partial chlorination

The third option is similar with Option 2 except that the other fine materials and residual material attached on hulls are converted into chlorinate form using $ZrCl_4$ in LiCl-KCl salt, and then fed directly into the electro-refining process. This raises the burden of an additional salt process. Chlorinated nuclear material in salt is homogeneous enough to give a destructive analysis sample for material accountancy, but is bad form for a non-destructive measurement due to increased (α, n) neutrons. A material accounting

Table 1
Linkage options between decladding and electro-reduction processes and their technical and proliferation resistance aspects.

Option		Technical aspect (Rating)		Safeguards and proliferation resistance aspect (Rating)	
1	(Homogenization +) Pelletizing	-Practically most feasible -Additional process step	H	- Chance to apply homogenization to get representative sample - Need additional process monitoring and/or measurement - Single stratum of small volume (small transfer container) into Ar cell, increased proliferation resistance	H
2	Large particle: Direct into Anode basket Small particle & Hulls: Oxidation and pelletizing	-Same process and equipment with Option 1, but less operation number or smaller equipment size	H	- Difficult to get representative sample (but there is possibility to get two homogeneous strata) - Better burn-up measurement signature (Cs-134, 137) - Additional material accounting and monitoring scheme due to divided stratum with different oxide factor (HMO_2, HM_3O_8).	M
3	Large particle: Direct into Anode basket Small particle & Hull residue: Chlorinate (using $ZrCl_4$ in LiCl-KCl) & feed into ER	-Additional salt process	H	- Difficult to get representative sample - Better burn-up measurement signature (Cs-134, 137) - Bad form for non-destructive measurement due to increased (α,n) neutrons - Complex material accounting and monitoring scheme due to divided stratum with different chemical form (Oxide, Chlorinate)	M
4	New Cathode Basket concept to minimize loss of material in salt	-Lack of detailed concept	L	If possible, minimize diversion risk without any loss of nuclear materials into process (with homogenization)	VH
5	Recovery of precipitated oxide (loss) from the bottom part of LiCl salt, chlorinate using $ZrCl_4$ in LiCl-KCl, and feed into ER	-Need detailed technology to effectively recover -Additional process step	L	- Complex material accounting and monitoring scheme due to divided stratum with different chemical form	M