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A STUDY OF SAFEGUARDS SYSTEMS ON DRY REPROCESSING FOR FAST BREEDER REACTORS

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Abstract

A "Feasibility Study on the Commercialized Fast Breeder Reactor (FBR) Cycle System" is underway at Japan Nuclear Cycle Development Institute (JNC). Concepts to commercialize the FBR fuel cycle are being created together with their necessary research and development (R&D) tasks. "Dry," non-aqueous, processes are candidates for FBR fuel reprocessing. Dry reprocessing technology takes advantage of proliferation barriers, due to the lower decontamination factors achievable by the simple pyrochemical processes proposed. The concentration of highly radioactive impurities and non-fissile materials in products from a dry reprocess is generally significantly larger than the normal aqueous (Purex) process. However, the safeguards of dry reprocesses have not been widely analyzed. In 2000, JNC and Los Alamos National Laboratory (LANL) initiated a joint research program to study the safeguards aspects of dry reprocessing. In this study, the safeguardability of the three options: metal electrorefining, oxide electrowinning, and fluoride volatility processes, are assessed.

FBR spent fuels are decladded and powdered into mixed oxides (MOX) at the Head-End process either by oxidation-reduction reactions (metal electrorefining and fluoride volatility) or mechanically (oxide electrowinning). At the oxide electrowinning process, the spent MOX fuel powder is transferred to chloride in molten salt and nuclear materials are extracted onto cathode as oxides. For metal electrorefining process, on the other hand, the MOX fuel is converted to chloride in molten salt, and nuclear materials are extracted onto cathode as a metal form. At the fluoride volatility process, the MOX fuel powder is converted to UF_6/PuF_6 (gaseous form) in a fluidized bed; plutonium and uranium fluorides are separated by volatilization properties and then are converted to oxides.

Since the conceptual design of a dry reprocessing plant is incomplete, the operational mode, vessel capacities, residence times, and campaigns are not fully defined. Preliminary estimates of the longest acceptable campaign length while still meets loss detection goals were made using typical measurement errors and annual throughputs of plutonium within the facility. For all reprocessing facilities, both in-process inventory and the input/output materials measurements must be determined for closing the materials balance. Usually, operations are to be shut down periodically and plants are to be completely cleaned out to recover all materials in measurable forms during inventories. If there is no cleanout between campaigns, fluctuations of in-process inventory have to be monitored. We conclude that the three dry reprocessing methods will have adequate safeguardability, if limited to small-scale campaigns or to low annual throughputs. For a large scale, e.g,. 50 t(HM)/y FBR fuel reprocessing plant, there remain challenges to be addressed through process development in JNC and safeguards R&D study with LANL.

Keywords: advanced nuclear fuel cycle, spent fuel, pyroprocess, separation, nuclear safeguards, material accountability

1. Introduction

Japan Nuclear Cycle Development Institute (JNC) and the electric utilities in Japan have established a new organization to develop a commercialized fast breeder reactor (FBR) cycle system.[1] Since July 1, 1999, feasibility studies have been undertaken to determine the promising concepts and to define the necessary R&D tasks.

In the first two years, a number of candidate concepts have been selected from various options for recycle technologies. Dry reprocessing methods are selected because of viability. Conceptual designs of dry reprocessing facilities are examined to evaluate the possibility of practical usage. And the feasibility studies should also guide the necessary R&D to commercialize FBR cycle system. To obtain information for the evaluation, JNC and Los Alamos National Laboratory (LANL) have studied the safeguardability of proposed dry reprocessing plants based on conceptual studies of the proposed processes.

2. Design Base For The Study

2.1 Conceptual Design Study of Facilities

A basic specification of the reprocessing facilities to be evaluated with respect to safeguardability is shown below. This specification[2] is common to all of the reprocessing methods with target fuel of FBR spent fuel, Core and Blanket containing burn up 150 GWd/t Core Average; annual throughput of 50 ton HM/y (approximately 4.8 ton Pu/y); annual operating days of 200 days/year; target recovering rate of HM 99% for Pu, U; and impurities in product of 2 wt% of products (low decontamination product).

2.2 Characteristics of Reprocessing Methods

1) "Oxide-Electrowinning": Transform mixed oxide (MOX) fuel (powder) to chloride in molten salt, and nuclear materials are extracted onto cathode as an oxide form as shown in Fig. 1.

The spent fuel is decladded and powdered at the Head-End process mechanically. The fuel powder is set on an anode, and a part of the uranium is deposited on a cathode by electrolysis operation. The remaining fuel, U, Pu, fission products (FPs), on anode is dissolved into molten salt with the chlorine gas. Noble metals, which disturb MOX deposition, are collected to cathode by electrolysis. Afterwards, U・Pu is collected on the cathode in the form of MOX. Minor Actinides (MA) are collected to the cathode.

2) "Metal-Electrorefining": Transform MOX fuel (powder) to chloride in molten salt, and nuclear materials are extracted onto cathode as a metal form. Metals are converted to oxide, if necessary (Fig. 2).

The spent fuel is decladded and powdered at the Head-End process by oxidation-reduction reactions. The blanket fuel is processed similarly as for the DUPIC process. The fuel powder of the spent core fuel is dissolved into molten salt with chlorine gas. Noble metals are collected into metal Cd. U metal is then collected to the cathode by the electrolysis operation. After changing cathode to liquid Cd, U/Pu/MA are collected to the Cd cathode by electrolysis. Some of the FPs that remain in the salt are extracted to liquid metal with reduction reagent.

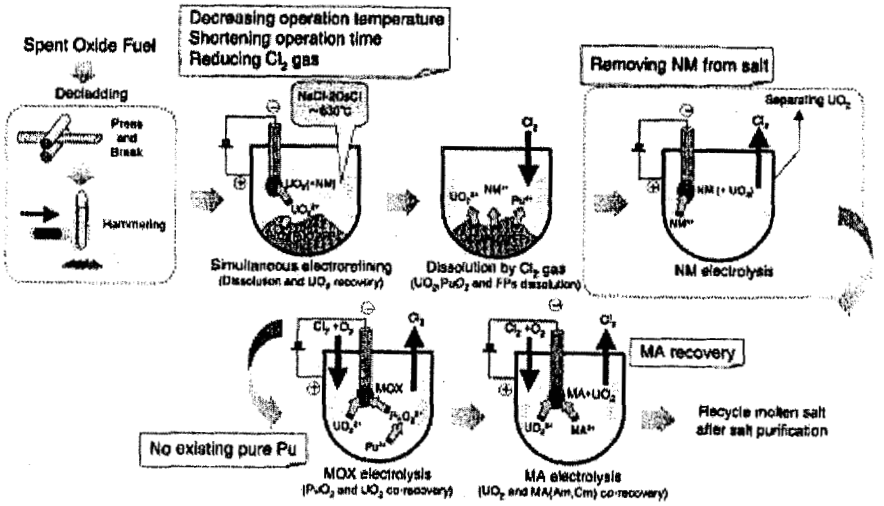

Fig. 1. "Oxide-Electrowinning" process.[3]

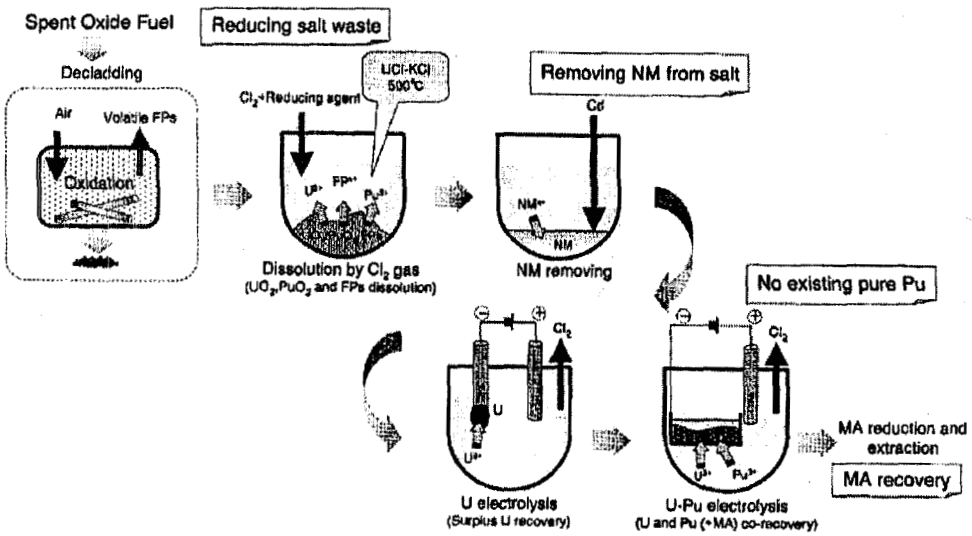

Fig. 2. "Metal-Electrorefining" process.[3]

3) "Fluoride volatility": Transform MOX fuel (Powder) to UF_6/PuF_6(Gas); Pu and U are separated by volatilization properties. Then fluoride nuclear materials are converted to oxides as shown in Fig. 3.

The spent fuel is decladded and powdered at the Head -End process by oxidation-reduction reactions. The fuel powder is converted to the fluoride in the fluidized bed and most of UF_6 is volatilized for separation. The operational condition is changed to volatilize $(U + Pu)F_6$ and to separate it from FPs. The separated fluoride fuels are collected and converted to oxide forms.

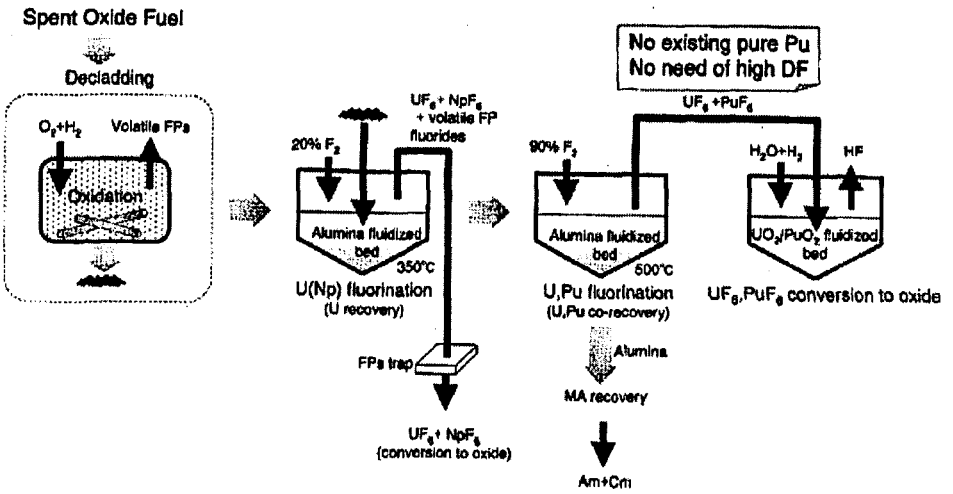

Fig. 3. "Fluoride Volatility" process.[3]

3. Evaluation of Safeguardability for Dry Methods

3.1 Material Balance Closure

For closing periodic material balances (MB), for all processes, it is necessary to measure the input materials (feed) and the output materials (product, waste, and side streams). In the dry processes, the spent fuel oxide powder is produced at the head-end of the process. This powder could be homogenized and sampled for destructive analysis, providing a measurement uncertainty of approximately 1%. If the powder is not homogenized, the input accountancy measurement is the burnup calculation on the spent fuel; this has an anticipated uncertainty of approximately 10% averaged over a core load.

In addition to the input and output, it is necessary to account for all materials in process at the time of a physical inventory. Two approaches are available, clean-out of all materials or measurement of in-process materials.

Usually, for annual inventories, the process is shut down and completely cleaned out. However, this is very disruptive of processing operations and does not provide timely detection of loss. For reprocessing facilities, more frequent material balance may be required for a higher certainty in the material balance. More frequent material balances may be possible by measuring materials in process at the time of material balance closure. The process may be stopped, but with materials remaining within process equipment; the process may be stopped at a point when materials become accessible; or the inventory can be performed "on demand" while the process is in operation.

3.2 In-Process Inventory Measurements

In-process inventory measurements would be desirable because the concept of near-real-time accountancy (NRTA) cannot be implemented unless in-process inventory can be measured or estimated. Being able to compute a MB at any time ("on demand") is essential to modern NRTA systems. In-process measurements are not likely to be feasible for any of the three options without extensive R&D because of the high-temperature vessels, "mist" Pu, presence of fission products and other complications.

If there is no clean-out between campaigns (not yet determined), or if NRTA is desired, then fluctuation in process holdup will be an issue. Similarly, because of internal recycling and potential holdup, batch tracking would be challenging. Salts, electrodes (and other process

equipment), and fluidized beds are reused, introducing any holdup Pu into the next batch of spent fuel.

3.3 Design Verification

Design verification could be a challenge for all three methods. The main issue is the ability of the operator to change the process conditions to recover various Pu concentration materials. Given the large measurement uncertainties expected, this would be difficult to detect with materials accounting alone. Therefore, an R&D priority is to design some way to verify that process conditions have not changed during the time since the last inspection.

3.4 Measurement Uncertainty and Loss Detection

Processing campaigns have not yet been decided upon, but annual throughput is expected to be a maximum of approximately 4800 kg of Pu, 24 kg of Pu per day, in each method. For discussion purposes, we assume here that each process would be operated in 1-2 week campaigns with a cleanout between each campaign. The IAEA detection goal for Pu is to detect a loss of 1 SQ of Pu with 95% detection probability and a 5% false alarm probability. To achieve this goal, σ_{MB} must satisfy $\sigma_{MB} <= 8/3.3 = 2.42$ kg. If we can measure the input material with $\sigma_{Total} = 1\%$ and output with $\sigma_{Total} = 0.5\%$, then σ_{MB} becomes 1.1%. This result suggests that it should be possible to meet typical IAEA detection goals for campaigns having 9 or fewer 24-kg batches per campaign (because $9 \times 24 \text{ kg} \times .011 = 2.376$ kg, which is less than the $\sigma_{MB} = 2.42$-kg goal). This calculation is a preliminary estimate that is expected to be modified as more information becomes available about measurement performances. The estimate is intended to provide initial guidance about suitable campaign lengths.

We anticipate that a reasonable goal for any of the three facilities is to achieve σ_{MB} of approximately 1% of throughput, and that therefore IAEA loss detection goals could be met over approximately 10-day campaigns with clean-outs between campaigns.

4. Evaluation Of The Three Candidate Dry Reprocessing Options

There is no distinguishable difference in accountancy among the three processes. Impressions for three dry methods are summarized below.

1) If the powdered spent fuel oxide is homogenized, sampled, and measured by chemical analysis, then that head end has an advantage over the process head ends that require input measurement of the spent fuel assemblies. Measurement uncertainty for chemical analysis of powdered oxide from spent fuel should be significantly lower than NDA of the spent fuel assemblies or burnup estimates.
2) All processes have to use a measurement of a MOX powder (containing recycled U) as the output accountability measurement.
3) All three processes may have batch identity and batch tracking problems. This is further complicated by Pu in holdup, entrained in process equipment, or in side streams.
4) All have in-process inventories that are impossible to measure with current technology. Nondestructive assay R&D will be required to address these materials.

All have design verification issues, in that the degree of separation of Pu and U can be changed without detection by the IAEA. Facility/process monitoring R&D is therefore required.

5. Non-Destructive Assay (NDA) Methods For Reprocessing Safeguards

The extremely high levels of neutron and gamma-ray emissions from dry reprocessing will make traditional NDA techniques for Purex reprocessing very difficult to apply. To implement to dry reprocessing, safeguards R&D on NDA are required.

5.1 Input Accountability

At a Purex plant, for example, clarified dissolver solutions are the input to the main chemical process area where separation and purification occurs. Input accountability measurements at a reprocessing plant are essential for effective safeguards. Analytical items required at this point are uranium concentrations, plutonium concentrations, and isotopic compositions. The Hybrid K-Edge/X-Ray Fluorescence Densitometry (HKED)[4] and Isotope Dilution Gamma-Ray Spectrometry (IDGS)[5] methods may be applied to measure input samples for dry reprocesses if the samples are uniform and can be completely dissolved. The HKED measures the uranium element concentration by using K-edge densitometry (KED) and the plutonium element concentration by using x-ray fluorescence (XRF). IDGS is a novel technique recently developed for the simultaneous measurements of concentrations and isotopic compositions for both plutonium and uranium in highly irradiated spent-fuel dissolver solutions by using high-resolution low-energy gamma-ray spectrometry.

5.2. The Curium Monitoring Approach Reprocessing Safeguards

5.2.1. Introduction

Curium can be a useful signature for safeguarding spent fuel at the head-end of reprocessing plants, high-level waste vitrification plants, and bulk-handling facilities such as the direct use of PWR spent fuel in CANDU Reactors (DUPIC). The direct measurement of the plutonium and the uranium in spent fuel is extremely difficult because of the high-radiation levels associated with the fission product gamma rays and the curium neutrons. After several years of post-irradiation cooling, the ^{244}Cm becomes the dominant source (>95%) of neutrons and it decays with an 18.1 year half life. For many spent fuel storage, process, and handling activities, the ratio of the curium to plutonium is invariant because there is no chemical process that can change the relative amounts of the actinides. For these cases, the ^{244}Cm can be used as a tag for the plutonium mass if the Cm/Pu ratio is known.[6,7] The same curium ratio approach can be used to measure uranium and other actinides.

5.2.2. Curium Ratio Concept

The Cm/Pu ratio can be directly measured at key measurement points (KMP) such as the input accountability tank in the high activity waste input tank for vitrification, and in the blended mixed oxide (MOX) powder for a dry process. Under current safeguards procedures, the plutonium concentration is routinely measured at these three KMP.

When using the curium balance approach, the ^{244}Cm would be measured from the same sample that was used for the plutonium determination using destructive analysis (DA) or nondestructive analysis (NDA). If the accuracy or cost of the curium DA is prohibitive, the ^{244}Cm mass of a small sample (1-5 ml) can be easily measured from the neutron emission rate from the sample's ^{244}Cm.

The primary use of the curium balance or tagging approach is to determine the plutonium mass in the waste and product flow streams that are otherwise unmeasurable. These streams include

- the hulls,
- compacted cladding,
- vitrified high level waste containers,
- input spent fuel assemblies.

The high yield and excellent penetrability of the fast-neutrons from the spontaneous fission allow the complete assay of bulk samples that might weigh several hundred kilograms (e.g., a vitrified waste canister) and yet have a sensitivity to measure samples as small as one gram (e.g., accountability powder samples).

5.2.3. Curium Measurement Methods

The neutron detectors that are used for measuring the curium must be designed to withstand the high gamma-ray levels from the fission products that usually accompany the curium. Lead shielding is used for this purpose because the neutrons can penetrate the lead with negligible attenuation. Also, we have developed ^{3}He neutron detectors that can operate in gamma fields that are an order of magnitude higher than was previously possible. The neutron signals can be measured in gamma fields of Sv/h.

The spontaneous fission neutrons from curium are the dominant source of neutrons from most categories of spent fuel and associated wastes. In general, the higher the burnup, the higher the ^{244}Cm fraction in spent fuel assemblies.[6] In the high-level waste after the plutonium separation, the mass ratio of the Pu/Cm is ~5-10 but the neutron emission rate per gram from ^{244}Cm is 10^{4} times more than for ^{240}Pu. To obtain the plutonium mass, we count the ^{244}Cm neutrons and calculate the plutonium from the Pu/Cm ratio.

The curium concentration can be measured at several different locations in a reprocessing facility such as Spent Fuel Assembly, Accountability Tank powder, Hot-Cell, High Level Waste Tank, and Input To Vitrification Plant.

5.2.2. Dual Use of Curium for Monitoring

In addition to using the Cm/Pu ratio to measure the plutonium, the ^{244}Cm neutrons provide an effective radiation monitoring signal. This unattended continuous monitoring signal can provide an effective supplement to the C/S system. In general, the ^{3}He based neutron sensors are more reliable and robust than the video systems.

The dual use (NDA and C/S) of the curium neutrons is being implemented at reprocessing facilities such as the TRP leached hull monitor and the TVF canister monitor. The software to support this dual use function is reported elsewhere.[8]

The curium concentration measurement can be performed in the unattended mode with remote transmission to the IAEA. These measurements could provide an integrated process monitoring function that would supplement an in-depth safeguards approach for enhanced reprocessing plant safeguards.

5.3. Summary

The ^{244}Cm neutrons provide a convenient signal to measure other actinides such as U, Pu, Am, etc. The fast-neutrons from ^{244}Cm are very penetrating so they can be used to measure bulk samples including fuel assemblies, waste drums, and vitrified glass canisters.

For waste streams, the prolific neutron yield of the ^{244}Cm provides a sensitivity for the plutonium determination that is about two orders of magnitude better than could be obtained from a direct Pu measurement.

From a safeguards viewpoint, a primary benefit of using the curium balance approach is the improved C/S performance from the dual use of the signal. The leached hulls monitor at the TRP hot cell portal provides a valuable C/S function for the entire head-end area. The continuous radiation information at the key measurement areas provides a transparency for the facility operation that was not possible before implementing the curium NDA sensors.

6. Conclusion

The purpose of this report is to evaluate which one of three candidate dry reprocessing options is the most "safeguards friendly." The three options, which all produce MOX fuel, are (1) oxide electrowinning, (2) fluoride volatility, and (3) metal electrorefinening. All three options process approximately 24 kg of Pu per day, 207 kg of U per day, plus alternate nuclear

material such as Np and Am. These methods would have safeguards challenges if operated on a large scale (200 kg of Pu or more per year) that will have to be addressed through process development and safeguards R&D. However, if the use is limited to small-scale campaigns (10 days) or to low annual throughput (50 kg or less of Pu per year), then any of the three methods is probably adequately safeguardable (and able to meet IAEA detection goals).

We believe the curium measurement method is a good approach for reprocessing safeguards. However, with all options, we anticipate the need for R&D of assay methods for input spent fuel, waste streams, and in-process inventories, as well as process and/or facility monitoring, because none of the three methods are very safeguards friendly when operated at high throughput.

As more detailed information is developed on process descriptions, a more detailed assessment of batch or campaign material balance closures can be performed. In the current stage of development of each of the subject processes, it is not yet possible to assess the relative safeguardability of the three processes, such as detection capability of frequent material balances when materials are still within the process. This will have to be assessed at later stages of process development.

Acknowledgments
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