

Assessment of the Limit of Errors on the Nuclear Material Accounting in Advanced Spent Fuel Conditioning Process

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Abstract

A preliminary study on the safeguardability of ACP (advanced spent fuel conditioning process) was conducted. The proposed ACP concept is an electrometallurgical treatment technique to convert oxide-type spent nuclear fuels into metal forms, which can achieve significant reduction of the volume and heat load of spent fuel to be stored and disposed of. For the safeguardability analysis of the ACP facility, sub-processes and their KMPs (Key Measurement Points) were defined first, and then their material flows were analyzed. Finally, the limit of errors in the inventory difference (ID) value of the facility was estimated with assumption by assuming international target values for the uncertainty of measurement methods and their uncertainty. From the preliminary calculation, we concluded that if the assumptions regarding measurement instruments can be achieved in a safeguards system for the ACP facility, the safeguards goals of International Atomic Energy Agency (IAEA) could be met.

1. Introduction

The question of “how to manage the spent fuel discharged from reactors” has been a key factor to be considered, as a part of the sustainable supply of nuclear energy policy in Korea like other countries. Approximately 6,000 metric tons of spent nuclear fuel from reactor operation has been accumulated in Korea. It is also expected that more than 30,000 metric tons of spent nuclear fuel would be accumulated by the end of 2040.

Significant reduction of the volume and heat load of spent fuel by use of lithium reduction process could lighten some burden of final disposal in terms of disposal size, safety and economics. With those perspectives, Korea Atomic Energy Research Institute (KAERI) has been developing a pyro-metallurgical technique, so-called lithium reduction process, to convert the oxide fuels into metallic form since the year 1997. In addition to this, pyro-metallurgical process with more economic and efficient advantages has been developed by KAERI since 2001.

In the lithium reduction process, lithium dissolved in molten LiCl reduces the oxide components of the spent fuel, yielding the corresponding metals and Li₂O. The metallic product is collected separately and will be held in interim storage until its ultimate disposition is decided. In this considering process, some fission product elements with high heat load such as cesium and strontium are dissolved in lithium chloride molten salt, and are separated from the spent fuel product.

The goals of the ACP is to recover more than 99.8% of the Actinide elements and to minimize the volume and heat load of spent fuel to be placed in interim storage and geological repository. The ACP concept will also give enhancement of the long-term safety as well as economical benefit for spent fuel disposal.

This paper summarized the preliminary results of joint research project by KAERI and Los Alamos National Laboratory (LANL) to study the safeguardability of the ACP concept. Since the conceptual design of an ACP facility is incomplete and related parameters are not fully defined, some assumptions were made for the safeguardability assessment.

2. Description of the ACP Technology

2.1 Main Process Concept

The ACP technology developed by KAERI is based on the pyrochemical process that was done in the 1960s and 1970s. The reference concept consists of six major sub-processes. These sub-processes are illustrated by the reference flow diagram shown in Fig. 1. They include:

- (1) Dismantling the fuel assemblies, cutting fuel rods, and removal of the Zircaloy cladding,
- (2) Thermal oxidation of UO₂ to the form of U₃O₈,
- (3) Reduction of the oxide fuel to metals, using a suitable reductant in a molten salt,
- (4) Regeneration of the reductant metal by electrolysis of its oxide to allow recycling it and to minimize the waste generation,
- (5) Smelting of metalized fuel,
- (6) Casting of metalized fuel in a form that is suitable for interim storage and deposition.

Based on the reference technology, an alternative concept has been developed to simplify the process and to increase the reduction performance. In the alternative concept, the lithium is produced electrolytically at the uranium oxide cathode and this lithium reduces oxide spent fuel to metal. Consequently, separate lithium recovery process is no longer needed in this concept.

In the reference lithium reduction process, the oxide fuel elements are chopped into segments and are voloxidized, and the resultant oxide powder is loaded into porous magnesia basket. The baskets are charged into a reduction vessel, where the fuel is reduced with lithium dissolved in molten LiCl at 650 °C. A layer of molten lithium floating on the surface of the salt maintains the lithium activity at unity. The different elements of the fuel (actinides and fission products) react

with the lithium in various ways, depending on their chemical characteristics.

The actinide oxides, designated generically as MO_2 , are reduced to metal according to the reaction:



The reduced metal remains in the basket.

The fission product oxides can be divided into four groups based on their reaction with lithium. Fission products that form chlorides are classified as the “FPA” group. The alkali and alkaline earth elements are in this category. They are dissolved in the salt phase after converted to their chlorides according to these reactions:



Also in the FPA group is europium, which behaves like the alkaline-earth metals and forms $EuCl_2$. The rare earth (RE) elements’ behavior is complicated. Thermodynamically, they are not reduced and remain as oxides. However, some of the rare-earth oxides will interact with Li_2O according to the following typical equilibrium:



The $LiNdO_2$ has limited solubility in the salt, but Nd_2O_3 is virtually insoluble. Thus, elements in the RE group are distributed between the basket and the salt. Fission products that react with lithium to form salt-soluble compounds, such as LiI and Li_2Te , are designated as the “FPB” group. This group includes Te, Sb, Se, Br, and I. The balance of the fission products, the noble metal (NM)

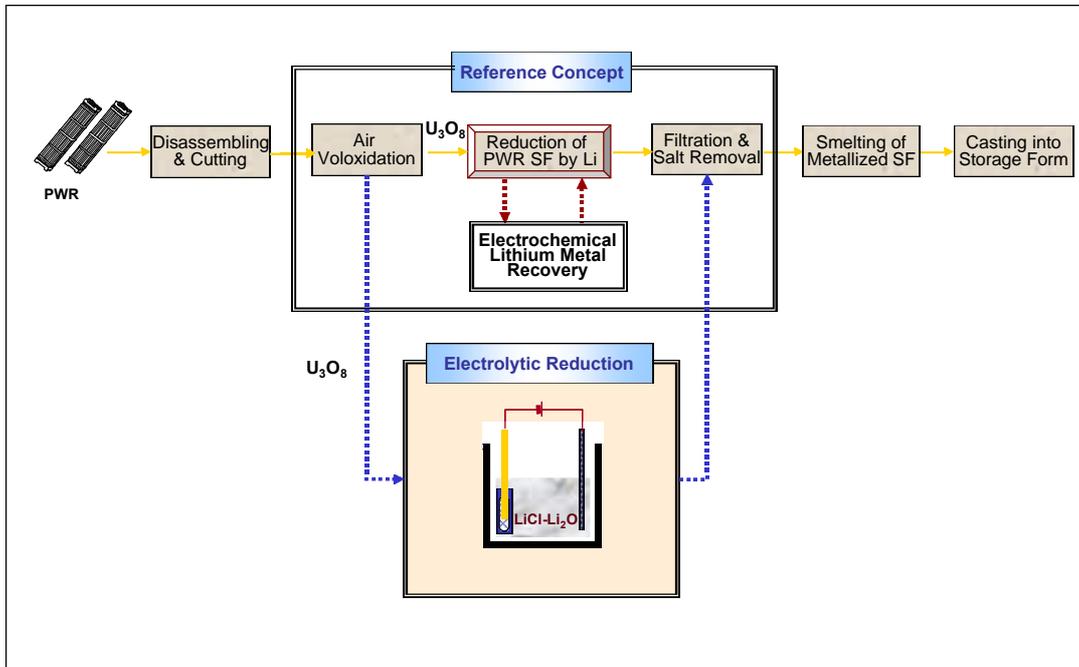


Fig. 1. Flow Diagram of Lithium Reduction Process.

group, is reduced to metal along with the actinides and remains in the fuel basket.

Effective reduction is possible at temperatures as low as 500 °C when lower melting point eutectics such as LiCl-KCl or LiCl-LiF are used. However, the LiCl-KCl combination proved incompatible with the mineral waste form and the LiCl-KCl combination was undesirable because of potassium vaporization at 525 °C. In addition, the solubility of Li₂O in LiCl-KCl is very low at 500 °C. These considerations lead to the selection of LiCl as the process salt and 650 °C as the process temperature. While the solubility of the Li₂O in LiCl at 650 °C is 8.7 wt%, it was reported that the Li₂O concentration must be kept below 3 wt% to obtain efficient actinides reductions [3]. The reason is apparently the equilibrium reaction:



To keep the Li₂O concentration at an acceptable value and to recover the lithium for reuse after the reduction step, the Li₂O is electrochemically decomposed to liberate oxygen and lithium:



This process is known as Electrowinning. Equation (6) is the net result of the following two reactions; one is taking place at the anode and the other at the cathode:



The degree of reduction of the TRU elements is a primary concern because of the goal to recover greater than 99% of the actinides in the PWR spent fuel. Table 1 is a summary of the results achieved in many reduction experiments for Li reduction concept [4].

The Electrowinning step also serves to reduce the concentration of FPB and RE fission products dissolved in the salt. The rare-earth oxides that dissolved in the salt during the reduction step precipitate as the Li₂O concentration is reduced. These precipitated oxides are filtered from the salt before the recovered salt is returned for use in another reduction step. The FPA group fission products are unaffected and remain with the salt. The concentration of these fission products is

Table 1. Reduction Yield Achieved for Lithium Reduction Concept.

Oxide Reduced	Reduction Yield
UO ₂	100.000
PuO ₂	99.996
NpO ₂	99.998
AmO ₂	99.982
CmO ₂	99.986

controlled when it reaches the design limit by discarding a small portion of the salt as part of the mineral waste form. In the Electrolytic reduction concept, the lithium Electrowinning is conducted in the uranium oxide cathode simultaneously and there is no process for salt recovering.

2.2 Facility Description

2.2.1 Facility Concept

For safeguardability analysis of the ACP facility, a pilot scale ACP facility with a capacity of 50 MTHM/year was assumed. The facility is a physically stand-alone facility, and administratively isolated from reactors and interim spent-fuel storage facilities. Main process of the facility is assumed alternative electrolytic reduction concept shown in Fig. 1, which has no need of lithium recovery system. The facility availability is assumed 60 %, which is equivalent to 219 full operating calendar days per year. The process consists mainly of three parts: spent fuel handling area (spent fuel disassembling and rod extraction), main hot cell (decladding, reduction, smelting, casting etc.), and metal ingot handling area (loading metal rods into storage cask and temporary storage). The reference fuel used in the ACP facility is Korean Yong-Gwang Unit 1 & 2's 17 × 17 standard PWR spent fuel assemblies with a minimum 10 years of cooling time after discharge from the reactor.

2.2.2 Process Flow and Description

The ACP consists of several process steps as shown in Fig. 2. In the process, various solid wastes will be generated and all wastes containing nuclear material shall be managed for safeguarding.

Table 2. Basic Specifications of Pilot Scale ACP Facility.

Target fuel	Korean Yong-Gwang Unit 1 & 2's 17x17 standard PWR spent fuel assemblies with a minimum of 10 years of cooling time after discharge from the reactor.
Facility throughput	50 MTHM/y (approximately 0.45 MT-Pu/y)
Facility availability	60% plant production availability (i.e., equivalent 220 full operating calendar days/year)
Impurities in product	2 wt% of products (low decontamination product)
Main process of the facility	Electrowinning reduction process (reduction rate: 99.8% for all actinides)

1. Disassembling: The spent PWR fuel disassembling is performed in dry cells or in pool using commercially available technology. Some non-fuel bearing structural components are generated and volume reduction process of structural components is added.
2. Rod cutting: A conventional mechanical process such as shear cutter is used and cutting wastes are generated.
3. Decladding: A conventional mechanical slitting technology is used for decladding. Cladding hull is generated and some nuclear material is embedded in the hull.
4. Voloxidation: The purpose of this process is to increase the reduction rate in consequent process. UO_2 powder is converted to U_3O_8 with an oxidation temperature of 500 .
5. Reduction: This reduction process is the key process in this ACP. The U_3O_8 powder is

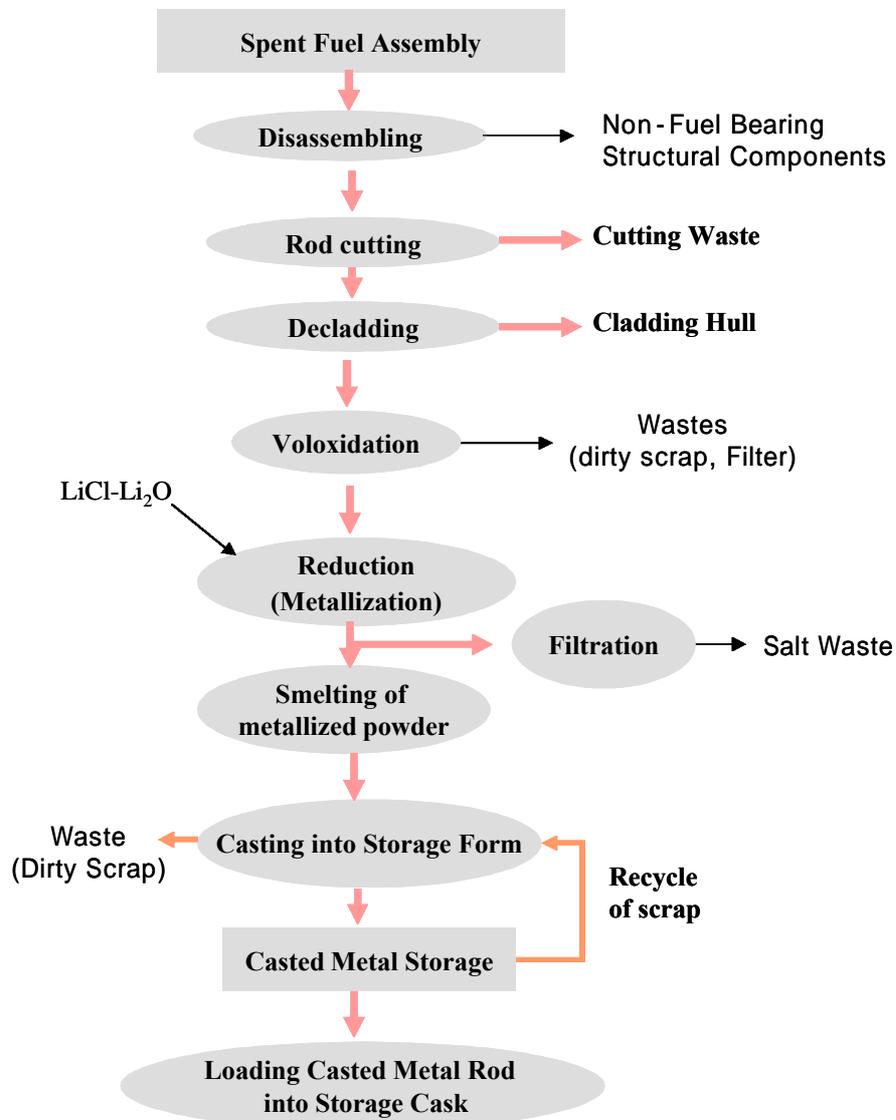


Fig. 2. Process Flow of Pilot Scale ACP Facility

reduced to a metallic form with 99.8% reduction yield for typical actinides. Further study to decide optimum operating condition to maximize the reduction rate and to minimize the process time shall be required. The generated salt waste can be recycled for several times.

6. Smelting/casting: Smelting and casting process are required to get an appropriate storage form of metal rod of 20 kg. Some dirty and clean scrap (rejected ingot) is generated in this process. The clean scrap is treated for recycling.
7. Loading metal rod into storage cask: Storage cask for metal rod is provided and stored temporarily in storage area. After that, metal casks can be transported to interim storage area.

3. Material Control and Accountability

In the absence of facility-specific information for the ACP facility, the facility design features like material balance area (MBA), material flow pattern, key measurement points (KMPs), and inventory during material balance closing were assumed for conceptual facility. The assumptions necessary to calculate the detection sensitivity of the operator's materials accounting system are presented below. Details of material flows, materials inventories, and material measurements, which determine the detection sensitivity, are also given. Fig. 3 identifies MBA boundaries, KMPs, and locations of inventories at material balance closings. A basic specification of the conceptual ACP facility to be evaluated with respect to safeguardability is summarized in Table 2. It is assumed that this facility operates 220 days/calendar year and the facility closes material balances once every 3 months (or once after 54 days of operation). For this analysis, It is also assumed that the present IAEA detection goals for spent LWR fuels would apply to materials within the ACP facility. Nuclear material contents for material balance were calculated using the fuel depletion code ORIGEN-ARP and the resulting material contents are summarized in Table 3.

3.1 Assumed Facility Design and Material Flow

Fig. 3 is abbreviated representation of the major process areas of this conceptual facility. As shown in this illustration, the fuel conditioning facility is considered as a two MBA. The facility operator does material accounting based on some declared values for feed materials; destructive chemical analyses for mixed oxides; and NDA measurements for U metals, recyclable scraps, and disposable waste streams. IAEA verification is preceded by a shutdown and cleanout of all major process areas and the accumulation of inventories at a few locations shown in Fig. 3 as KMPs.

IAEA verification will employ attributes and variables measurements, preferably NDA measurements. The facility closes material balances once every three months and plans to have the IAEA inspections coincide with this schedule for plant shutdown, cleanout, and material balance closing. The large inventories of feed materials and products (MBA-A and KMP-J in Fig. 3) are maintained as "items" for inventory purposes and are stored in separate storage locations. The

cleanout operation before material balance closing recovers almost the entire residual process holdup. Therefore, inventory of plutonium as process holdup is negligible.

3.2 Key Measurement Points

The KMPs associated with both transfer and inventories at the conceptual ACP facility are also illustrated in Fig. 3. The transfer KMPs are represented by numbers and the inventory KMPs are designated by letters. Some details of transfer and inventories at bulk counting area (MBA-B) can be summarized as follows.

- ◆ KMP-1: Receiving. Feed input to MBA-B is spent PWR fuel rods. Approximately 7,500 fuel rods are received in 3 months. Because of the difficulty of independently measuring the plutonium content of spent PWR fuel rods, the facility uses declared values of SNM content (based on burnup values of PWR fuel assemblies) as input measurements.
- ◆ KMP-2: Shipping. Product output is from the facility. Approximately 620 newly fabricated U metals are shipped out during 3-month period. Approximately 10 U metals are produced each day. The facility uses the plutonium-content assay of reduced U metal powder to calculate the plutonium contents of metals. Each metal contains approximately 0.18 kg of plutonium in total.
- ◆ KMP-3: Shipping. This output stream is measured waste forms, such as non-nuclear components of PWR fuel pin claddings contaminated with fuel residues,

Table 3. Actinide Content of Reference Spent Fuel.

Isotopes	g/initial MTU	wt % of HM
²³⁴ U	201	0.021
²³⁵ U	7,626	0.789
²³⁶ U	3,974	0.411
²³⁸ U	944,400	97.760
²³⁷ Np	479	0.050
²³⁸ Pu	141	0.015
²³⁹ Pu	5,251	0.544
²⁴⁰ Pu	2,119	0.219
²⁴¹ Pu	769	0.080
²⁴² Pu	463	0.048
²⁴¹ Am	509	0.053
²⁴³ Am	92	0.009
²⁴⁴ Cm	18	0.002
total Pu	8,743	0.905
total	966,041	100.000

miscellaneous discardable materials, filters, salt, and other disposable trash. It is assumed that these discards are measured with some NDA techniques to estimate the fissile content of discards and to declare them as measured discards. Approximately 0.5 Mt of waste material containing 1.3 kg of plutonium are discarded as waste during a 3-month period.

- ◆ KMP-A: Disassembled PWR fuel rods in trays. A total of 10 rods/tray are assumed. At inventory time, there are six trays at this location containing approximately 100 kg of fuel rods. The facility estimates the plutonium content with NDA technique and the declared values of the PWR fuel assembly.
- ◆ KMP-B: Discardable cutting waste of mixed oxide after the rod cutting. Approximately 0.007 kg/rod of materials mostly containing uranium as UO_2 is produced by rod cutting. At the time of material balance closing, it is assumed that this location has approximately 25 kg of cutting waste in two containers. The facility performs destructive chemical analysis to determine the plutonium content.
- ◆ KMP-C: Non-nuclear cladding materials of PWR fuel rods. The decladding ratio is assumed 99.9%. At inventory time, it is assumed that this location has approximately 13 kg of heavy metals that contains 0.11 kg of plutonium. It is assumed that these discards are measured with NDA technique to declare them as measured discards.
- ◆ KMP-D: Storage of mixed oxides after the rod cutting and decladding stage. Approximately 100 kg of materials mostly containing uranium as UO_2 is located at inventory closing. These are in five containers, each containing approximately 20 kg. The plutonium fraction is assumed 0.00905. The facility performs destructive chemical analysis to determine the plutonium content.
- ◆ KMP-E: U_3O_8 and associated powder residues from Voloxidation process. About 13 kg of this material remain at this location at inventory time. These are in one container and the facility performs destructive chemical analysis to determine the plutonium content.
- ◆ KMP-F: Extracted mixed oxides after the controlled Voloxidation stage. Approximately 100 kg of materials mostly containing uranium as U_3O_8 . These are in five containers, each containing approximately 20 kg. The plutonium fraction is assumed 0.00905. The facility performs destructive chemical analysis to determine the plutonium content.
- ◆ KMP-G: Salt waste that is used for actinides reduction. It is assumed that the reduction ratio is 99.8% and the $LiCl-Li_2O$ reductant is reused continuously for processing five batches. The salt ratio to reduce SNM is assumed three and approximately 60 kg of salt is needed for each reduction process. At inventory time, it is

assumed that this location has approximately 500 kg of salt waste in five containers, each containing 100 kg. The plutonium fraction is assumed 0.00046. The facility performs NDA technique to determine the plutonium content.

- ◆ KMP-H: Uranium ingot after the controlled reduction stage. Approximately 100 kg of

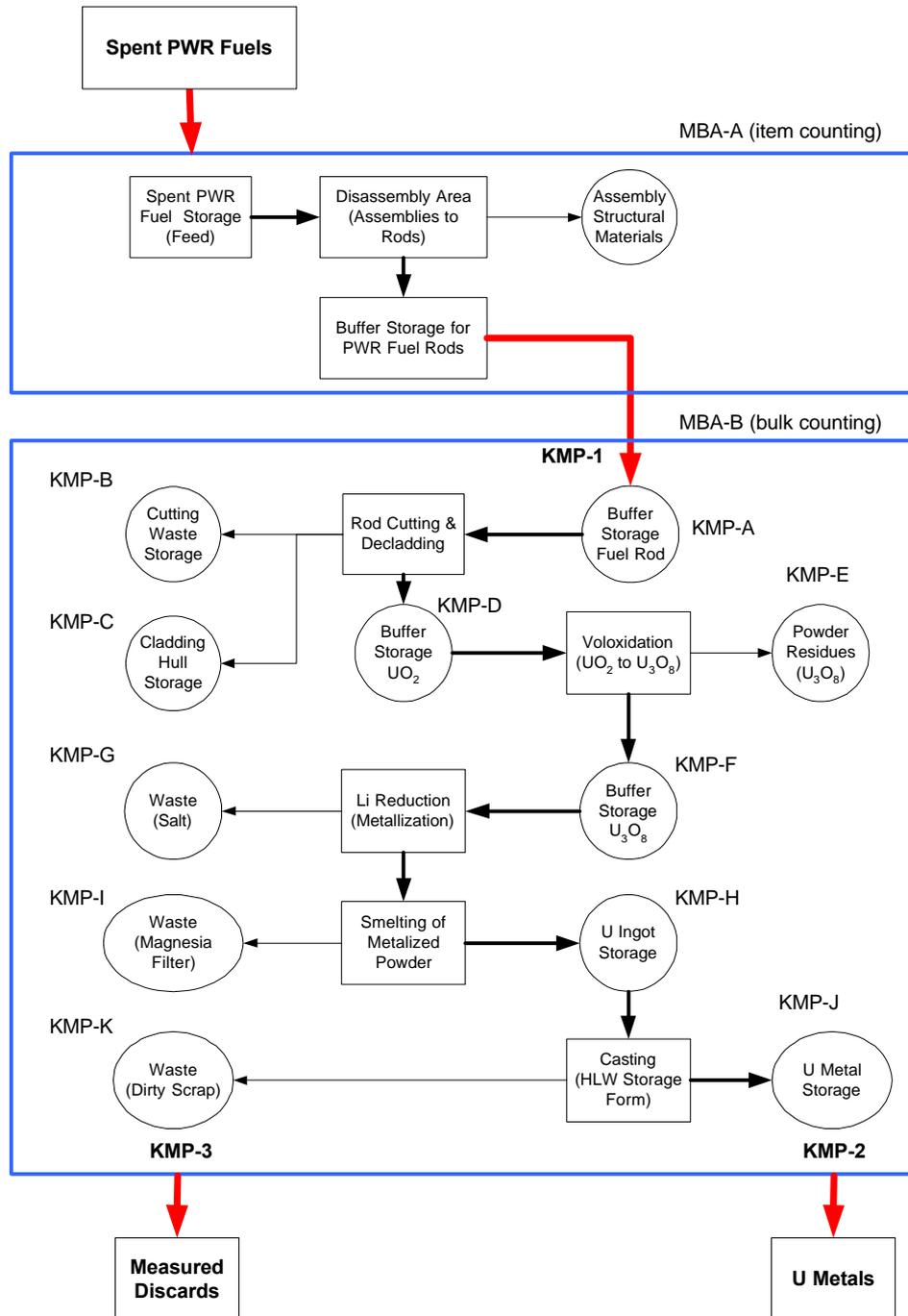


Fig. 3. Material Flow and Key Measurement Points at a Conceptual ACP Facility.

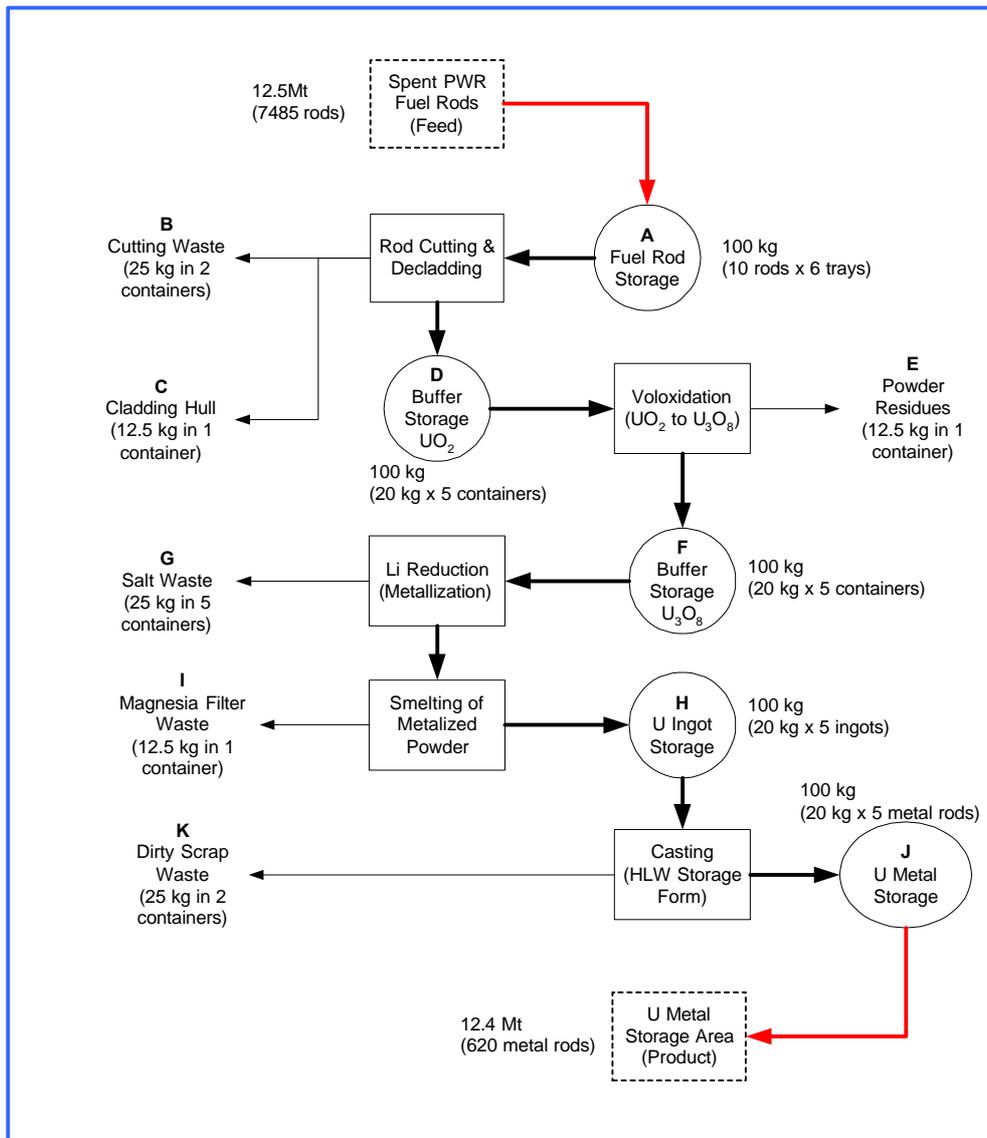


Fig. 4. Inventory at a Conceptual ACP Facility at Material Balance Closing.

uranium metals are located as five ingots of 20 kg at inventory time. The plutonium fraction is assumed 0.00905. The facility uses the NDA technique to estimate the plutonium content of metal ingot.

- ◆ KMP-I: Magnesia filter waste that is used for Li reduction and treated as waste after smelting of metalized powder process. It is assumed that approximately 0.1% of the metalized powders are remaining in the filter waste. At inventory time, it is assumed that this location has approximately 30 kg of waste materials, containing

0.11 kg of plutonium. The facility uses the NDA technique to estimate the plutonium content of the waste.

- ◆ KMP-J: Fabricated U metals. A total of 100 kg of U metals are located as a form of disposition storage. The plutonium fraction is assumed 0.00905. The facility uses the NDA technique to estimate the plutonium content of the metals. It is assumed that the IAEA may sample and verify the metals using a conventional neutron coincidence counter.
- ◆ KMP-K: Dirty scrap of U metal after metal casting process. The generation ratio of these scrap waste is assumed to be 0.2%. At inventory time, it is assumed that this location has approximately 25 kg of waste materials, containing 0.23 kg of plutonium. The facility uses the NDA technique to estimate the plutonium content of the waste.

3.3 Measurement Uncertainty and ID Evaluation

Material accountancy requires quantitative knowledge of material, 1) present in the material balance area at the beginning and ending of the accountancy period and, 2) transferred into and out of the area during the period. Concrete results require characterization of process operations and related material flows. It is also necessary to characterize the accounting system, facility measurement procedures, and related uncertainties. Material accountancy performed over multiple zones, and therefore over multiple process operations, does not require knowledge of the material that is transferred between processes.

Inventory for the bulk-handing area (excluding long-term storage) is shown in Fig. 4 and Table 5. From the standpoint of facility accounting, it is important that all items in inventory be associated with measured values. Such measured values should be obtained in a way compatible with efficient operation. The destructive assay (DA) measurements for plutonium concentration are made on a batch basis. It is unnecessary, time consuming, and costly to obtain a sample from each individual container of powder. Instead, samples are drawn from containers deemed representative of other containers. It is assumed that two concentration measurements per five containers are adequate for oxide powder. Power residues have one concentration measurement for two containers.

The uncertainties of quantitative measurements used in the conceptual accounting have the assumed uncertainties as Table 5 [2]. Because nuclear material processed in ACP facility is contained in many types and forms, material accountability requires that the nuclear material content of all flows entering and exiting a material balance area and the quantities of nuclear material in the ending inventory be known. The ID (Inventory Difference) is defined as the difference between the measured inventory and what is expected to be in the inventory based on the previous inventory and measured flows into and out of the process. The ID is calculated via the following equation,

$$ID = BI - EI + TI - TO \quad (9)$$

where BI and EI are the beginning and ending inventories and TI and TO are the transfers of nuclear material into and out of the material balance area, respectively. Because measurement errors will occur, the actual amount of material measured will differ somewhat from zero, the expected quantity, creating a non-zero ID. The probability of detecting the loss of a given quantity of material depends upon the uncertainty associated with the determination of the ID. ACP facility's material control and accountability methods propagate all measurement and sampling uncertainties to give a standard error.

The IAEA detection goal for Pu is to detect a loss of one SQ of Pu with 95% detection probability and a 5% false alarm probability. To achieve this goal, s_{MB} must satisfy $s_{MB} \leq 8/3.3 = 2.424 \text{ kg}$ (this assumes that the ACP facility tests for loss, not for gain of Pu, so the statistical testing is one-sided and the alarm limit is at $1.65\sigma_{ID}$). For two-sided testing, the upper alarm limit for the inventory difference is $2\sigma_{ID}$ and for one-sided testing, as assumed here, the alarm limit is $1.65\sigma_{ID}$. The control limit of $1.65\sigma_{ID}$ is such that error means the measured ID has a 95% probability of being less than $1.65\sigma_{ID}$, assuming that the true ID is zero, and assuming that all materials have been measured and accounted for and all sources of error are used in determining the limit of error.

Even though the processing campaign of ACP facility is not decided, the annual throughput is expected to be a maximum of approximately 450 kg of Pu, 2 kg of Pu per day. For this preliminary study, it is assumed that the facility closes material balances once every three months. Using uncertainty values, the result for the s_{inv} (bulk-handing inventory) is 0.063 kg of elemental plutonium, assuming no data falsification. The corresponding value for s_{thru} (bulk-handing throughput) is 2.361 kg of plutonium. This result suggests that it should be possible to meet typical

Table 4. Inventory for the Bulk-Handling Area.

Inventory KMP	Material Form	Pu(kg)	No. of Item	Accounting Method
A	Fuel Rods	0.91	60	Burnup, NDA
B	Powder (UO ₂)	0.23	2	DA + weight
C	Cladding hull material	0.11	1	NDA
D	Powder (UO ₂)	0.91	5	DA + weight
E	Powder residues (UO ₂)	0.11	1	DA + weight
F	Powder (U ₃ O ₈)	0.91	5	DA + weight
G	Salt waste	0.23	5	NDA
H	U ingot	0.91	5	NDA
I	Magnesia filter waste	0.11	1	NDA
J	U metal	0.91	5	NDA
K	Dirty metal scrap	0.23	2	NDA
Total		4.30	92	

Table 5. Assumed Measurement Uncertainties for Facility Accounting.

Measurement	So-called Random	So-called Systematic	Sampling	Instrument	Notes
Burnup value for plutonium	1.0%	1.0%	-	FRSC	1)
NDA (Non-nuclear material)	4.0%	1.5%	-	HLNC	2)3)
Weights	0.05%	0.05%	-	EBAL	
Concentration (UO ₂)	0.2%	0.2%	0.2%	IDMS	4)5)
Concentration (U ₃ O ₈)	0.2%	0.2%	0.2%	IDMS	4)5)
Concentration (Salt Waste)	0.2%	0.2%	0.2%	IDMS	4)5)
NDA (U metal)	2.0%	1.0%	-	HLNC	2)3)
NDA (Nuclear material)	4.0%	1.5%	-	HLNC	2)3)

1) In case of U-235 mass measurement.

2) Measurement time 300 sec.

3) Isotopic determination by mass spectrometry and alpha spectrometry.

4) Materials typically encountered in the nuclear fuel cycle.

5) Under conditions of sufficiently different isotopic compositions of spike and sample and near-optimum sample.

IAEA detection goals for campaigns having 3-months or fewer. This calculation is a preliminary estimate that is expected to be modified as more information becomes available about measurement performance.

4. Summary

A preliminary assessment of the safeguardability of ACP (advanced spent fuel conditioning process) was conducted in this study. The ACP concept is electrometallurgical treatment technique to convert PWR spent nuclear fuel into a single set of metal disposal form, reducing the volume and simplifying the qualification process. An objective assessment of the safeguardability of the ACP requires simultaneous consideration of its inherent proliferation resistance and the ability to design an integrated system of technical measures designed to guard against diversions and be capable of timely detection. At this research stage, most of the process data cannot be clearly defined. Therefore, the preliminary conceptual design to examine safeguardability of the ACP facility is based on the open literatures for a similar process considered.

Based on the assumptions we have made during this preliminary evaluation, the conceptual ACP fuel conditioning facility credibly meets reasonable diversion resistance and safeguards goals. Therefore, it is reasonable to conclude that (1) using the options proposed for Phase-I study, ACP fuel conditioning facility can be designed, built, and operated to meet the goals of IAEA safeguards; and (2) additional technologies necessary to make this possible can be developed in a timely fashion so that an inventory of the ACP facility materials can be verified by both the state and international safeguards inspectors. This conclusion regarding safeguardability assessment should be considered only as a preliminary evaluation. Some modifications are needed as more

reliable information on technical parameters is available.

Acknowledgement

This study was supported by the National Nuclear R&D Program at Korea Atomic Energy Research Institute and was done under the auspices of the Korea Ministry of Science and Technology.

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