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사용후핵연료 차세대관리공정 개념 설계에 대한 핵물질안전조치성 예비분석 보고서

PRELIMINARY ASSESSMENT OF SAFEGUARDABILITY ON THE CONCEPTURE DESIGN OF ADVANCED SPENT FUEL CONDITIONING PROCESS

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본 보고서를 2003 년도 사용후핵연료 특성계량화 기술개발 과제의 기술보고서로 제출합니다.

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요 약 문

한국에서는 현재 약 6,000 톤의 사용후핵연료가 누적되어 있으며, 2040 년까지 30,000 톤 이상의 사용후핵연료가 발생될 것으로 예상되고 있다. 상용 원자로에서 발생되는 사용후핵연료의 처리 문제는, 한국을 비롯한 대부분의 원전 보유국에서의 지속적인 원자력 에너지 정책 결정을 위한 핵심적인 사안이 되고 있다.

최근, 국내에서는 핵확산 저항성이 있는 사용후핵연료의 건식재처리 및 이를 바탕으로 한 후행핵주기 개념이 새로이 제안되고 있다. 사용후핵연료의 핵비확산성 건식처리 공정은 사용후핵연료의 부피 감소 및 발열량 저감 효과를 기대할 수 있으며, 이를 통해 사용후핵연료 처분장 부지 문제, 안전성 및 경제성의 부담을 경감시킬 수 있을 것으로 예상된다. 이와 같은 관점에서, 한국원자력연구소에서는 Li 용융염을 이용한 사용후핵연료의 금속전환 기술을 개발해 오고 있다.

본 연구에서는, 미국 로스알라모스 국립연구소와 공동으로 사용후핵연료 차세대관리공정 (Advanced Spent Fuel Conditioning Process, ACP)의 개념 설계 자료에 대한 핵물질안전조치성을 예비 분석하였다. 차세대관리공정은 산화물 형태의 사용후핵연료를 전기야금 기술을 통해 금속형태로 전환시키는 개념으로서, 처분 또는 저장되는 사용후핵연료의 부피와 발열량 감소를 기대할 수 있다. 본 보고서에서는 ACP 시설에 대한 핵물질안전조치성 분석을 위해 차세대관리공정의 하부 공정과 물질 흐름 및 주요 측정 지점 (Key Measurement Points, KMP) 등을 개념 설계하였다. 또한, 각 핵물질 흐름에서의 핵물질 계량 방안을 제시하였으며, 각 계량관리 기법에 대한 ITV (International Target Value)를 적용하여 재고차 평가에 대한 오차 한계를 계산하였다. 이와 같은 개념 설계 자료에 근거한 핵물질안전조치성 예비 분석 결과, ACP 시설에 적용될 핵물질 계량장비의 측정 정밀도를 만족시키는 경우에는 IAEA 의 핵물질안전조치 목표를 달성할 수 있는 것으로 분석되었다. 향후, 본 연구의 실질적인 활용을 위해서는 ACP 시설의 시설 용량과 MBA 기간 및 측정 오차와 같은 인자들을 고려한 민감도 분석 등의 추가적인 연구가 진행되어야 할 것으로 판단된다.

SUMMARY

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 this report, a preliminary study on the safeguardability of ACP (advanced spent fuel conditioning process) was conducted with Los Alamos National Laboratory. 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 standard deviation of 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. In the second phase of this study, further study on sensitivity analyses considering various factors such as measurement errors, facility capacities, MBA periods etc. may be needed.

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I. 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.

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Fig. 1 shows main strategies on spent fuel management by use of the pyro-metallurgical process that is developed by KAERI. In the lithium reduction process, lithium dissolved in molten LiCl

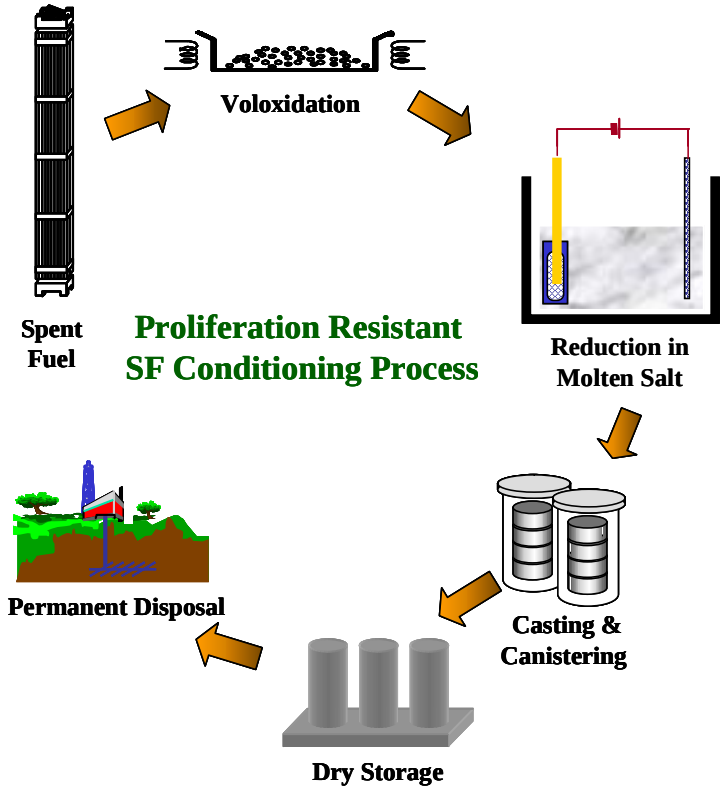

Fig. 1. Proliferation Resistant Advanced Spent Fuel Conditioning Process.

reduces the oxide components of the spent fuel, yielding the corresponding metals and Li_2O. 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 progress report 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.

II. DESCRIPTION OF THE ACP TECHNOLOGY

A. 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. 2. They include:

(1) Dismantling the fuel assemblies, cutting fuel rods, and removal of the Zircaloy cladding,

(2) Thermal oxidation of UO_2 to the form of U_3O_8,

(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 as shown in Fig. 3. Consequently, separate lithium recovery process is no longer needed in this concept.

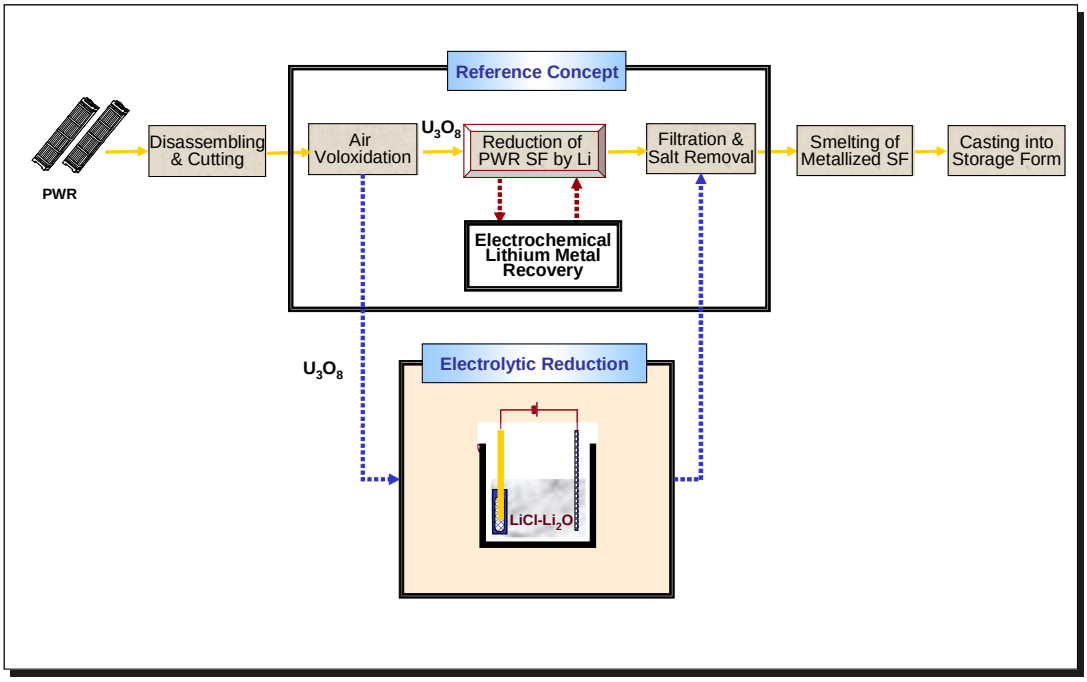

Fig. 2. Flow Diagram of Lithium Reduction Process.

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 ℃. 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:

$$MO_2 + 4Li \rightarrow M + 2Li_2O \quad (1)$$

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:

$$Cs_2O_2 + 2LiCl \rightarrow 2CsCl + Li_2O \quad (2)$$

$$SrO + 2LiCl \rightarrow SrCl_2 + Li_2O \quad (3)$$

Also in the FPA group is europium, which behaves like the alkaline-earth metals and forms

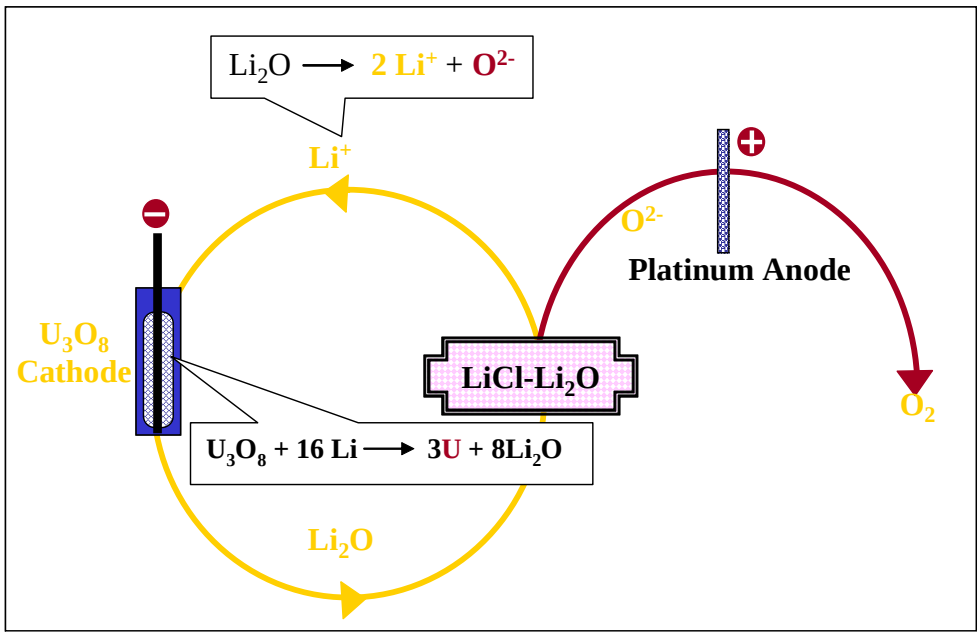

Fig. 3. Schematic Drawing of Electrolytic Reduction Mechanism.

$EuCl_2$. 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:

$$Nd_2O_3 + Li_2O \rightarrow 2LiNdO_2 \qquad (4)$$

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

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

Effective reduction is possible at temperatures as low as 500℃ 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℃. In addition, the solubility of Li_2O in LiCl-KCl is very low at 500℃. These considerations lead to the selection of LiCl as the process salt and 650 ℃ as the process temperature. While the solubility of the Li_2O in LiCl at 650 ℃ is 8.7 wt%, it was reported that the Li_2O concentration must be kept below 3 wt% to obtain efficient actinides reductions [3]. The reason is apparently the equilibrium reaction:

$$2Pu + 3Li_2O \rightarrow Pu_2O_3 + 6Li \quad (5)$$

To keep the Li_2O concentration at an acceptable value and to recover the lithium for reuse after the reduction step, the Li_2O is electrochemically decomposed to liberate oxygen and lithium:

$$Li_2O \rightarrow 2Li + \frac{1}{2}O_2 \quad (6)$$

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:

$$O^{-2} \rightarrow \frac{1}{2}O_2 + 2e^- \quad \text{(Anode)} \quad (7)$$

$$2Li^+ + 2e^- \rightarrow 2Li \quad \text{(Cathode)} \quad (8)$$

Table 2. Typical Specifications of Reference Spent Fuel.

Final burnup	33,000 MWd/MtU
Specific Burnup	37.5 MW/kgU
Nominal Enrichment	3.2 wt. % U-235
No. of Spacer grid	2 (Inc-718) + 6(Zry-4).
Pellet Density	10.4 g/cm^3
Total weight of fuel assembly	660 kg
U-weight/Assembly	440 kg
No. of Fuel Rods/Assembly	264
Pellet OD	8.05 mm
Active Length	365.8 cm
Cladding OD	9.5 mm
Cladding ID	8.22 mm
Cladding Thickness:	0.64 mm

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 Li_2O 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 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.

B. Facility Description

B.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. 3, 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. The design parameters of the reference fuel are shown in Table 2.

B.2. Process Flow and Description

The ACP consists of several process steps as shown in Fig. 4. In the process, various solid wastes will be generated and all wastes containing nuclear material shall be managed for safeguarding.

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

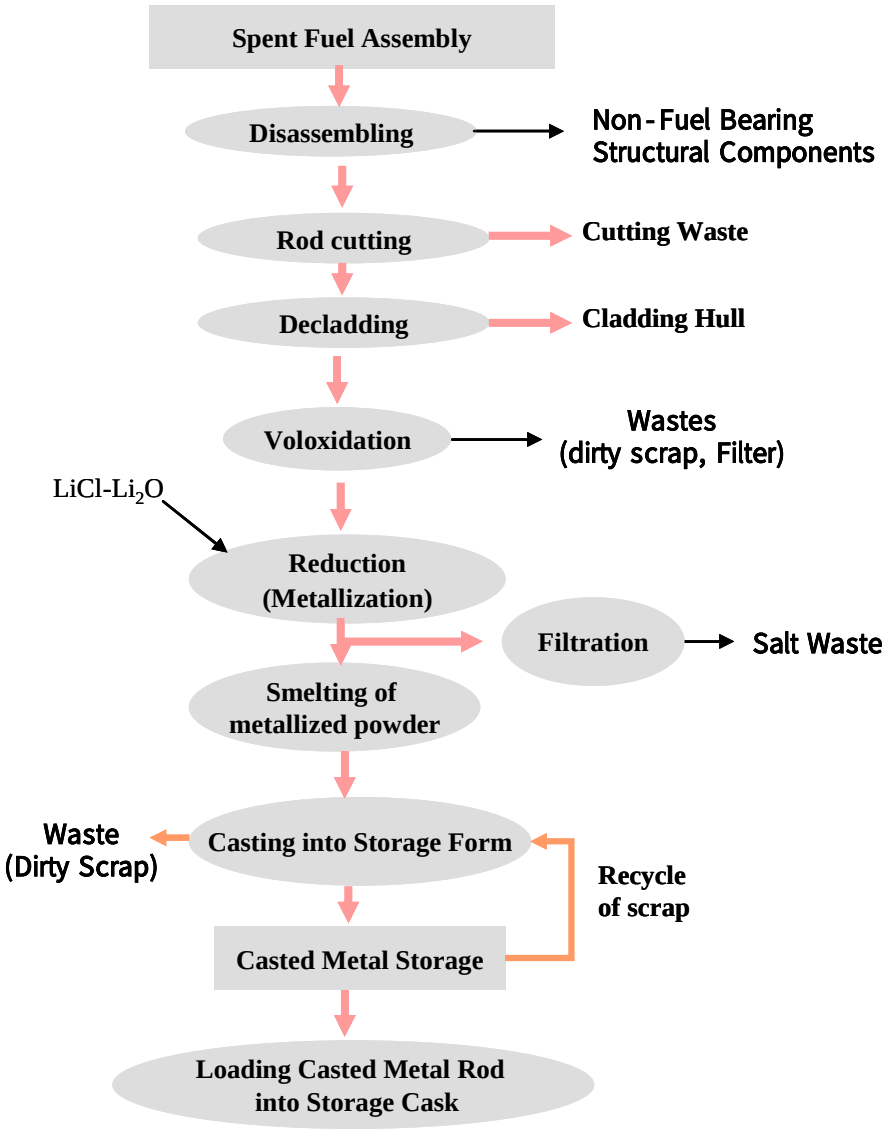

Fig. 4. Process Flow of Pilot Scale ACP Facility

generated and volume reduction process of structural components is added.

2. Rod cutting: A conventional mechanical process such as shear cutter is used and [m1]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 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 temporally in storage area. After that, metal casks can be transported to interim storage area.

III. PROLIFERATION RESISTANCE FEATURES OF ACP

In the lithium reduction process, as its name implies, lithium dissolved in molten LiCl reduces the oxide components of the fuel, yielding the corresponding metals and Li_2O. The key process of this system is a metallization process that employs lithium metal as a reductant and lithium chloride (LiCl) as a solvent. Most of actinide oxides will be reduced to metal in the process. In the reduction step, alkali and alkali-earth fission products would be converted to chloride whereas rare earth fission products would remain as oxide form. Some part of rare earth fission products as well as alkali and alkali-earth fission products remain in the salt but other fission products including noble metal go along with the actinide reduction metal. The lithium is recovered for reuse in subsequent reductions by electrolytically decomposing the Li_2O to form Li and O_2. The ACP has the following inherent features of proliferation resistance.

- Due to the nature of lithium reduction process, no fissile material can be separated in pure form. Plutonium, for example, is co-deposited together with miner actinides and some fission products. Therefore, the material requires further chemical reprocessing to separate pure fissile elements. This involves longer warning times and requires a technology standard in order to obtain material suitable for weapon purpose.
- The decay heat and radioactivity of the metalized spent fuel ingot are about 25% of the initial spent fuel. The presence of some fission products leads to high dose rate arising from the material.
- The lithium reduction process has to be located in heavily shielded hot cell due to its high radiation characteristics. The processing is self-contained, and there is no transport of intermediate materials outside of the facility. Therefore, access to the sensitive materials is extremely difficult. This feature may be concordant with the PIPEX concept as was proposed during the INFCE [1]. The PIPEX approach to reduce access to nuclear materials at the reprocessing and conversion stage would be to make use of the heavy concrete shielding that provides protection against radiation in reprocessing plants to give a physical barrier against

Table 3. 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)

diversion.

- The lithium process operates on a batch mode, which allows fundamentally easier material accounting than do continuous flow systems like PUREX system. The batch processing nature, combined with appropriate material sampling and physical security, can effectively assure against theft.
- The batch-type process readily supports near real time accounting (NRTA). Movement of material is controlled remotely with movements and weights recorded in real time by the NRTA system. When material is moved from one process step to the next, it is moved as a discrete mass in a labeled container, and weighed before shipment from one station and after receipt at the next station. Waste and scrap also are handled as discrete and weighted items.

IV. 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. Figure 5 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 3. 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 are calculated based on the reference fuel in Table 2, and the resulting material contents are summarized in Table 4.

A. Assumed Facility Design and Material Flow

- Figure 5 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. 5 as KMPs.
- IAEA verification will employ attributes and variables measurements, preferably NDA measurements.

Table 4. 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

- 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. 5) 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.

B. Key Measurement Points

The KMPs associated with both transfer and inventories at the conceptual ACP facility are illustrated in Fig. 5. 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, 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.

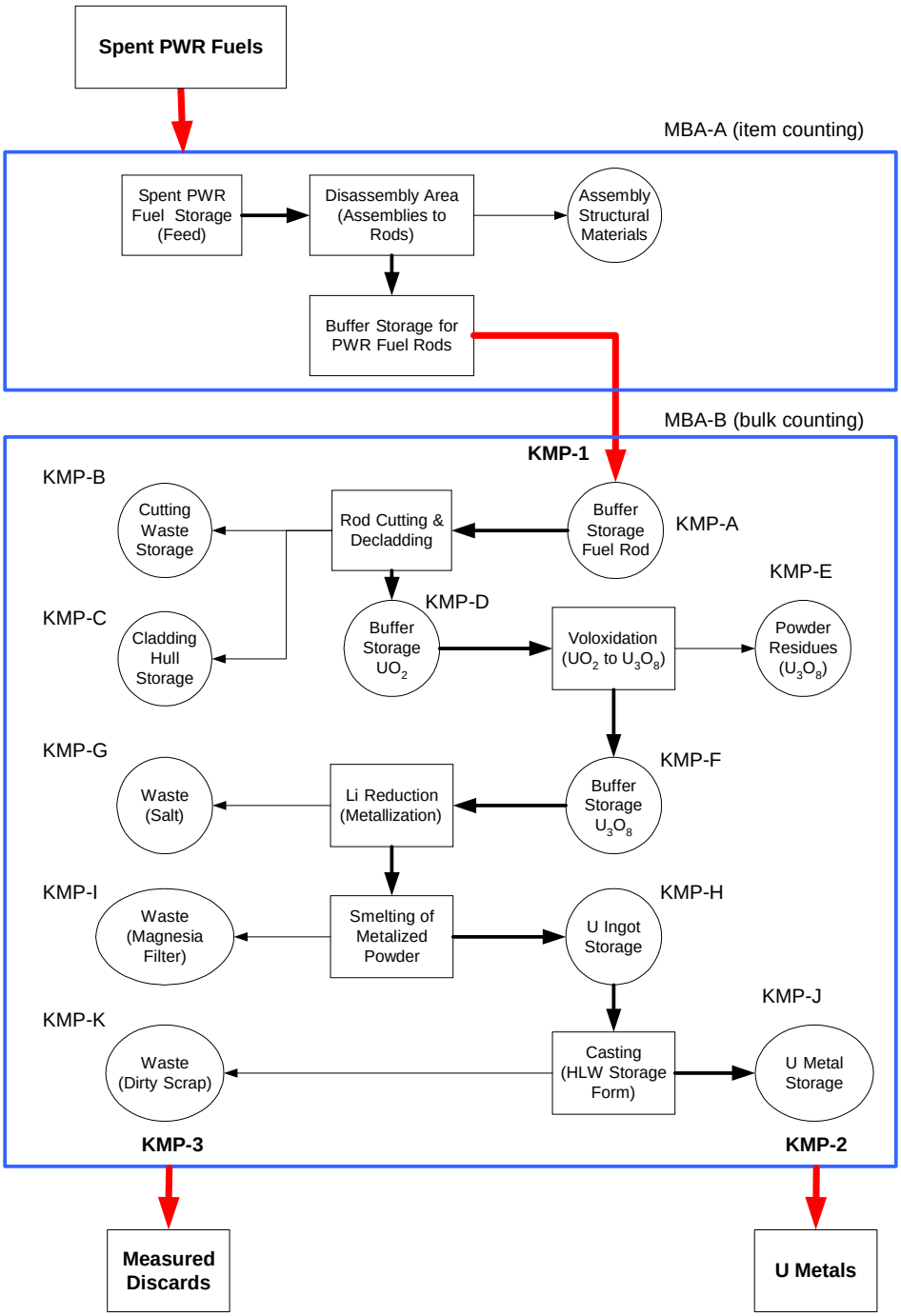

Fig. 5. Material Flow and Key Measurement Points at a Conceptual ACP Facility.

- 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 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

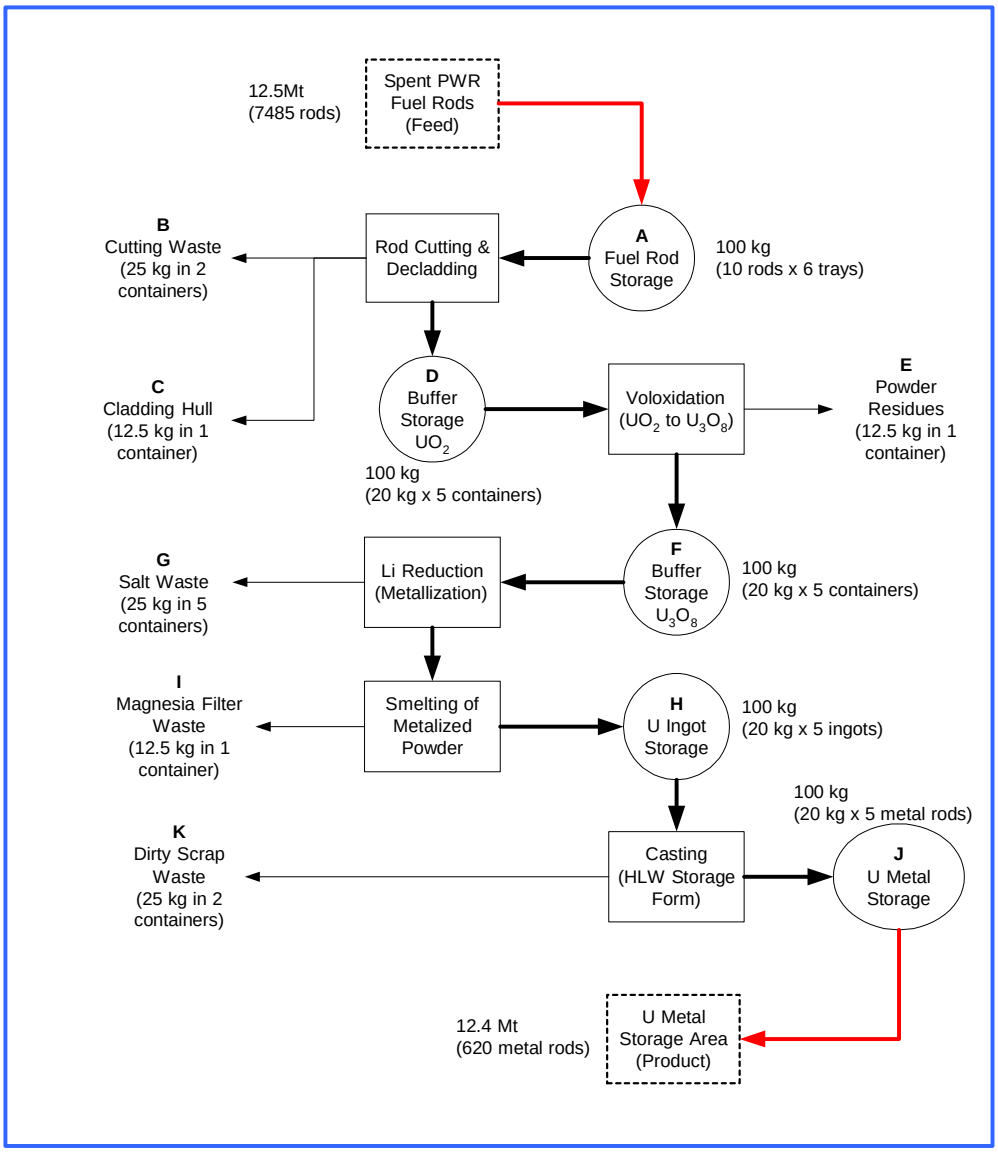

Fig. 6. Inventory at a Conceptual ACP Facility at Material Balance Closing.

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.

Table 5. 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_3O_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	

- 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.

C. 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. 6 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.

Table 6. 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_3O_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.

The quantitative measurements used in the conceptual accounting have the assumed uncertainties as Table 6[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,

$$ID = BI - EI + TI - TO \tag{9}$$

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, σ_{MB} must satisfy $\sigma_{MB} \le 8/3.3 = 2.424\ kg$ (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 σ_{inv} (bulk-handing inventory) is 0.063 kg of elemental plutonium, assuming no data falsification. The corresponding value for σ_{thru} (bulk-handing throughput) is 2.361 kg of plutonium. This result suggests that it should be possible to meet typical 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.

V. NDA SYSTEM FOR MATERIAL ACCOUNTING

NDA system must have following requirements for Advanced Spent Fuel Conditioning Process (ACP) facility.

① NDA system could treat the solid type of metal ingot, powder and salt waste.

② NDA system should be the radiation hard system, so they do not contain any weak material for radiation, especially gamma rays.

③ NDA system can determine Pu mass with consideration that the reduction rates of corresponding actinides change during the metallization and voloxidation process.

④ The assay chamber can be measured for bundle and metal ingot. The bundle has a diameter of 10.0 cm and a length of 25 cm, and the metal ingot has a diameter of 10.0 cm and a length of 13.0 cm with 20.0 kg mass. The neutron efficiency must be uniform at least 25.0 cm along the sample cavity.

Detail technical points for analysis for ACP materials and NDA system design process are described at Appendices A and B.

VI. SAFEGUARDS R&D REQUIREMENTS

Most of the technologies necessary to safeguard spent nuclear fuels at reactor facility and during transportation are now available. However, because of the unique nature of

electrometallurgical concept and the radiation characteristics of material associated with the ACP facility, not all technologies and system necessary to safeguard the fissile materials are presently available.

Known principles of NDA techniques and destructive chemical analysis techniques can be adapted to design equipment and procedures to meet the requirements of safeguarding nuclear materials at the ACP facility. During and following the selection of a ACP option for engineering demonstration, parallel efforts must be directed at developing systems for material accounting, measurements, containment and surveillance, and verification of the flow and inventories of materials at the ACP facility. Some of the details of new systems and technologies that are desirable to establish a modern safeguards regime for the ACP facility are the following.

1) A fully developed flow sheet for the processes and material flow across the ACP facility.
2) A computer model for the process operations and material flow to evaluate safeguards consequences well before actual operation of facility.
3) A modern computerized nuclear materials accountancy system at the facility level capable of automated data recording at strategic points. Such data using authenticated recording systems can be shared with the IAEA to facilitate periodic inspections and detailed physical inventory verifications.
4) Facility-specific designs of instruments for NDA of inventories at the ACP facility.
5) A variety of material verification measurement approaches should be examined before deciding on an optimal plan for developing ACP specific NDA instruments. Research and development requirements for developing such instruments include;
    (a) developing neutron multiplicity techniques to separate signals from ^{244}Cm and plutonium for spent PWR fuel;
    (b) investigating the feasibility of using remote neutron monitoring to safeguard spent PWR fuel during conditioning process to U-metal;
    (c) investigating the use of LA-MS (Laser Ablation Mass Spectrometry) for measuring the Pu:U:Cm ratio in U-metal;
    (d) investigating the feasibility of using LIBS (Laser Induced Breakdown Spectroscopy) for measuring the Pu:U:Cm ratio in U-metal;
    (e) Developing authentication methods for unattended NDA of spent PWR fuel and fabricated U-metal.

VII. 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.

In section IV, we analyzed a conceptual process and a material flow using our experiences with conventional fuel cycles and known measurement methods and their uncertainties. The measurement uncertainties we used are based on open literatures related with safeguard issues. In conclusion, in case of the assumptions regarding measurement instruments can be achieved in a safeguards system for the ACP facility, International Atomic Energy Agency (IAEA) safeguards goals could be met. The assumptions made in this study may be too optimistic for ACP facility because of the radiation characteristics of the materials. As we get information that is more reliable on measurements and verification approaches, these data and calculations should be modified.

In section V, we identified the requirements for developing necessary NDA instrument to verify the materials accountancy system at the ACP facility. Isotopic analysis for ACP materials with respect to mass distribution, total dose rate, neutron production rate and heat recommends that curium-monitoring method could be available if relative Pu amount to Cm is known.

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.

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APPENDIX A

ANALYSIS AND RECOMMENDATION OF NDA SYSTEM FOR THE SAFEGUARDS OF LAB SCALE ACP FACILITY

1. Analysis of ACP materials.

Overview of ACP is shown in Fig 1. To understand isotope contents of the ACP material, we analyzed material flow from input spent fuel (point 1) from final ingot (point 9). These process materials have different Cm-ratios for Pu, U, and U-235 through voloxidation and metallization process.

For further understanding of ACP material, we examined mass(Fig. 2), neutron production(Fig. 3) and heat distribution(Fig. 4) of the process material in order to get available non-destructive nuclear material accounting method, for example, gamma-ray spectroscopy, neutron counting and calorimetry and so on.

(1) Isotopic composition analysis:

Fig. 2 is the isotopic mass distribution except U-238 that is over 95% in ACP. Measuring weight gives the mass of uranium, especially U-238 concentration.

(2) Gamma-ray identification method:

Gamma-ray NDA method could identify the isotopic concentration. The availability of gamma-ray method depends on that the strength of characteristic gamma rays of and NDA system the special nuclear material (SNM) overcome those of fission fragments.

In general, the characteristic gamma rays of the SNM have lower energies less than 200-keV. Spent fuel (point 1) has many fission fragments. They give relatively strong gamma rays. Under the environment of such a strong fission fragment gamma rays, the detection of characteristic gamma-rays of SNM is very difficult. The radiation doses of ingot (point 9) are dominated by Pu. However, most gamma-ray emissions are from fission products rather than from Pu. To apply the gamma-ray technique, first of all, the radiation hardened detector sensor need to be developed. It means that the adoption of gamma ray NDA could be difficult for ACP nuclear material accounting.

(2) Neutron accounting method

Fig. 3 is the distribution of neutron production rate in ACP materials. Cm-244 is the major nuclear material to produce neutron signature in ACP. Actually, neutron-counting method could give high accuracy of nuclear material accounting if sample is pure. ACP material is complex and suffered a change of the relative ratios among the actinide, especially, Pu, U, U-235 and Cm. Unfortunately, neutron counting could not identify any isotopes in sample.

(3) Calorimetric Method

Calorimetric method (Fig. 4) can determine the SNM mass when the sample composition is known or the sample is pure. However, in case of the spent fuel and ACP material cases, the composition could not be determined within reasonable measurement uncertainty. Hence, this method is not good for ACP materials.

2. Analysis of NDA system for ACP safeguards

As a result of isotopic analysis for mass, neutron production rate and heat, the most available NDA method is thought to be the neutron NDA based on the Cm monitoring[1, 2]. Curium can be useful signature for safeguarding spent fuel at bulk-handling facilities such as the direct use of PWR spent fuel in CANDU reactor(DUPIC). Before the metallization process (point 3 in Fig. 1), the ratio of the curium to plutonium is invariant because there is no chemical process that can change the relative amount of the actinides. For these cases, the Cm-244 can be used as a tag for the plutonium mass if the Cm/Pu ratio is known. After metallization process of ACP, the relative amount of Pu to Cm-244 can be changed according to the reduction rates for corresponding actinides. It is known that the reduction rates for actinides can be different to measure in vitro. To imply curium-monitoring approach, R&D for determining the Cm-ratios for the ACP materials is required. When sampling is available from process flow, the Hybrid K-Edge/X-ray Fluorescence densitometry (HKED), Isotopic Dilution Gamma-ray Spectrometry (IDGS) [3] methods may be applied to measure Cm/Pu ratio. If the sampling is not available such as metal ingot and salt waste, then the laser ablation inductive coupled plasma mass spectroscopy (LA-ICP-MS) [4], Laser induced breakdown spectroscopy (LIBS) [5] method may be used as promising method, which are under development to identify isotopic composition in spent fuel.

3. Curium-ratio determination recommendation for lab-scale ACP

Curium monitoring method is a recommended technology for lab-scale ACP. The nuclear material accounting and control process was shown in Fig. 5. The lab-scale ACP is designed to treat five batches of 20 kg-U, i.e., totally 100 kg of spent fuel.

Therefore, the off-line chemical analysis will be available to determine the Cm-ratios among actinides. However, for the pilot facility with high capacity, an on-line analysis system will be needed and technology development is still opened.

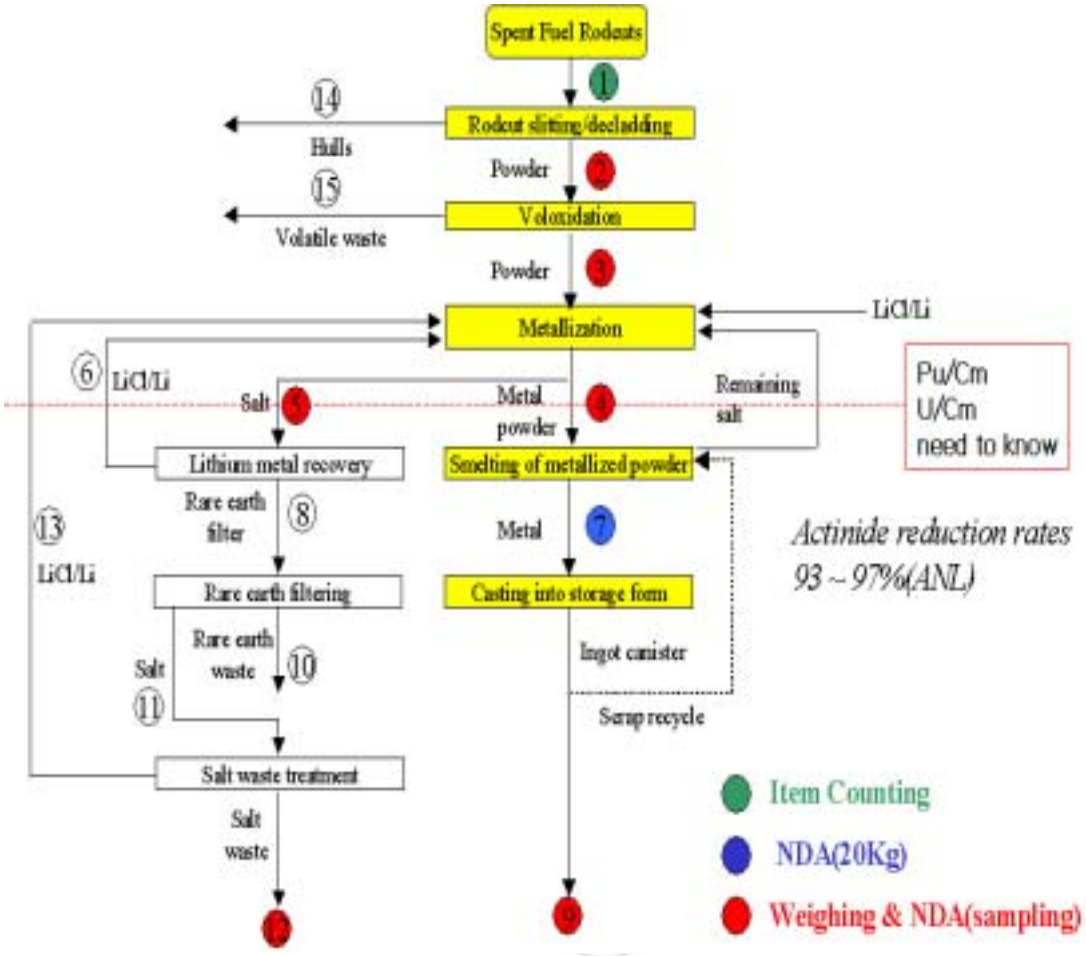

Figure 1. Overview of ACP material flow and the proposed nuclear material accounting method.

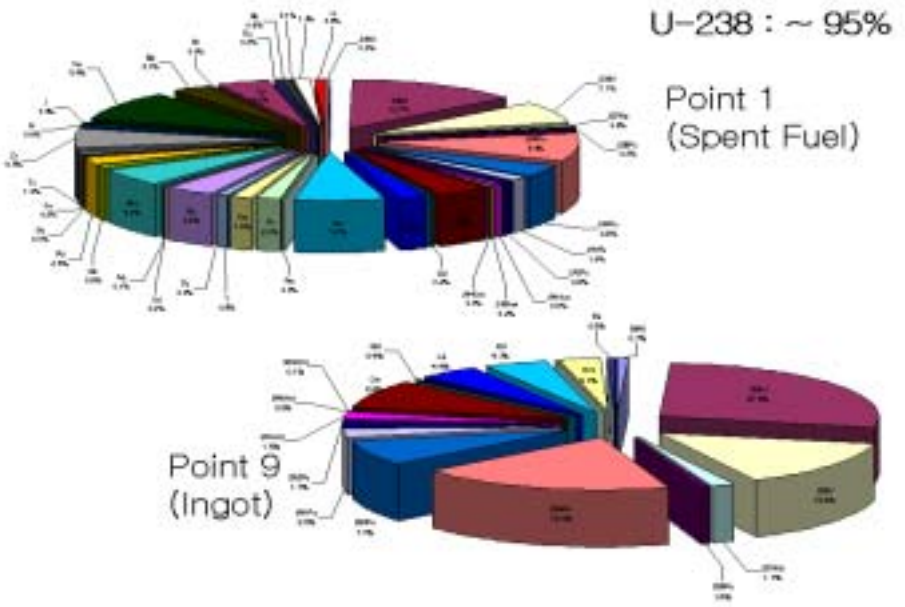

Figure 2. Mass distribution for spent fuel and ingot.

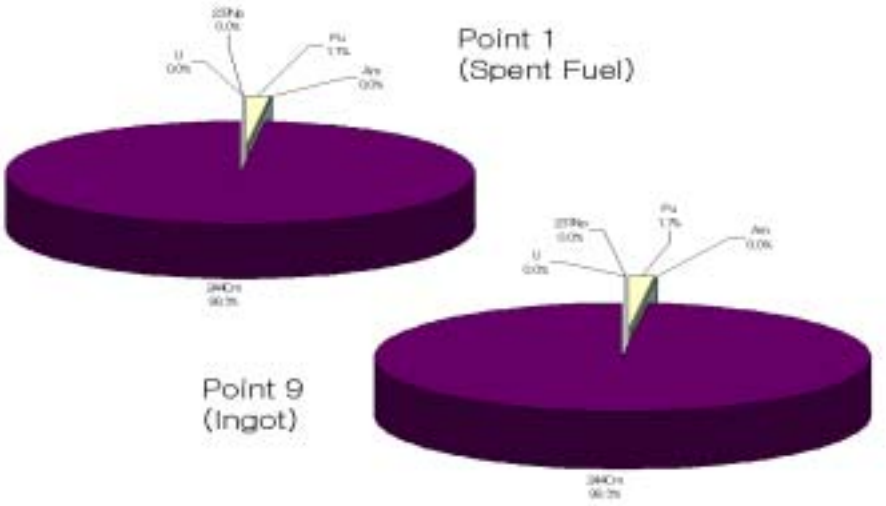

Figure 3. Neutron production rate distribution of ACP materials.

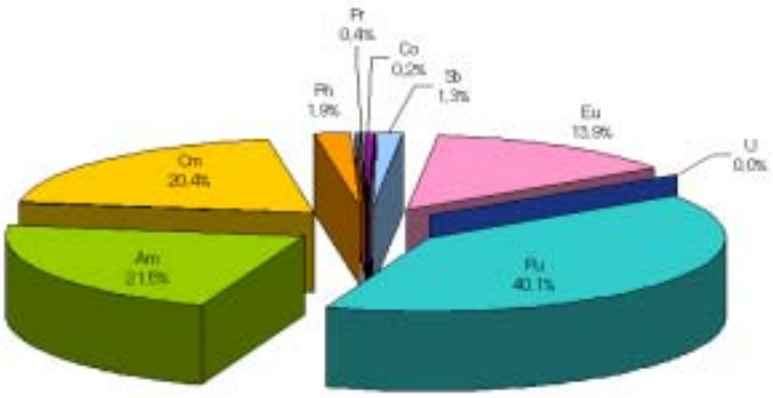

Figure 4. Heat distribution of ingot of ACP (point 9).

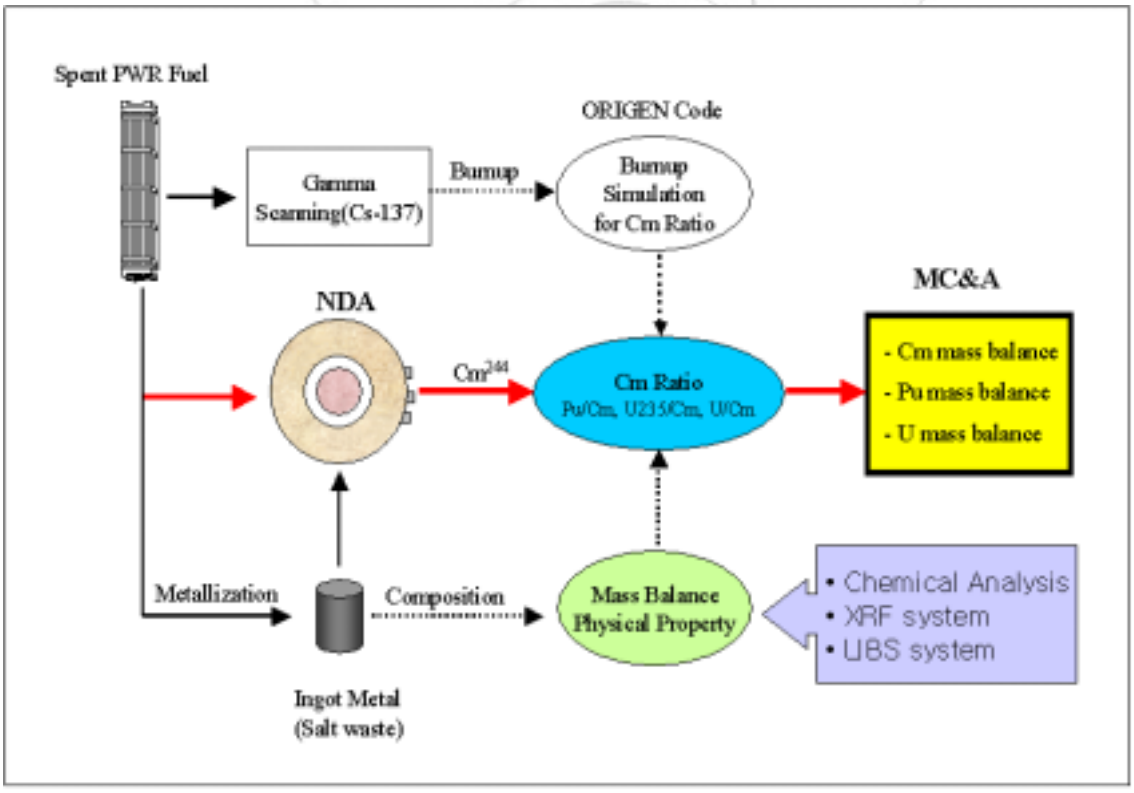

Figure 5. The proposed nuclear material accounting for ACP.

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APPENDIX B

MCNP MODEL STUDY FOR ACP NDA SYTEM

Two types of NDA detectors were considered. One is 30cm length of He-3 tube and other is 50 cm as shown in Fig. 6. The major consideration for MCNP simulation is that the proposed specification could keep the uniform neutron detection efficiency along sample cavity at least 25cm.

1. Simulation using 30cm of He-3 tube

For saving the space, we used NDA system using the 30cm of He-3 tube with adequate Cd thermal neutron cylinder absorber. However, this case could not satisfy the requirement. Fig. 7 and 8 are the result of MCNP simulation. Fig. 7 is the result of Cd cylinder without any neutron reflector. Fig. 8 is result of four kinds of neutron reflector placed at bottom side without any neutron absorber. This case did not used Cd absorber to have higher detection efficiency. Reflectors were assumed Ni, C, CH2 and C/CH2 with 10 cm thickness. The CH2 reflector system showed about 10 cm of uniform detection efficiency region.

In conclusion, any of 30 cm tube system could not satisfy the requirement described in section V.

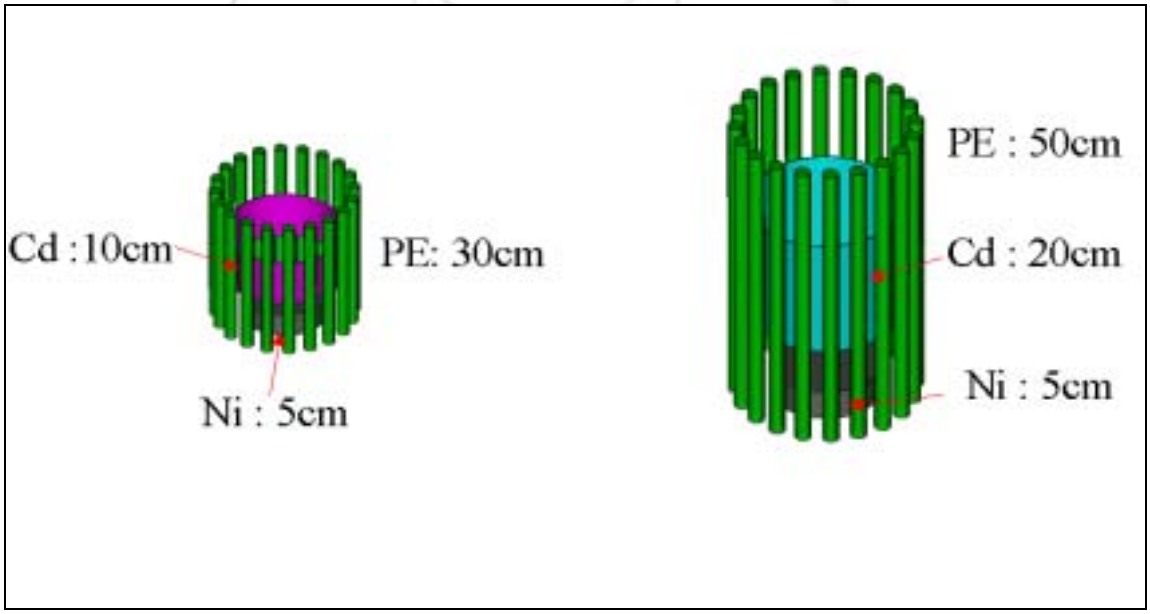

Figure 6. MCNP model for 30 and 50 cm of He-3 tube.

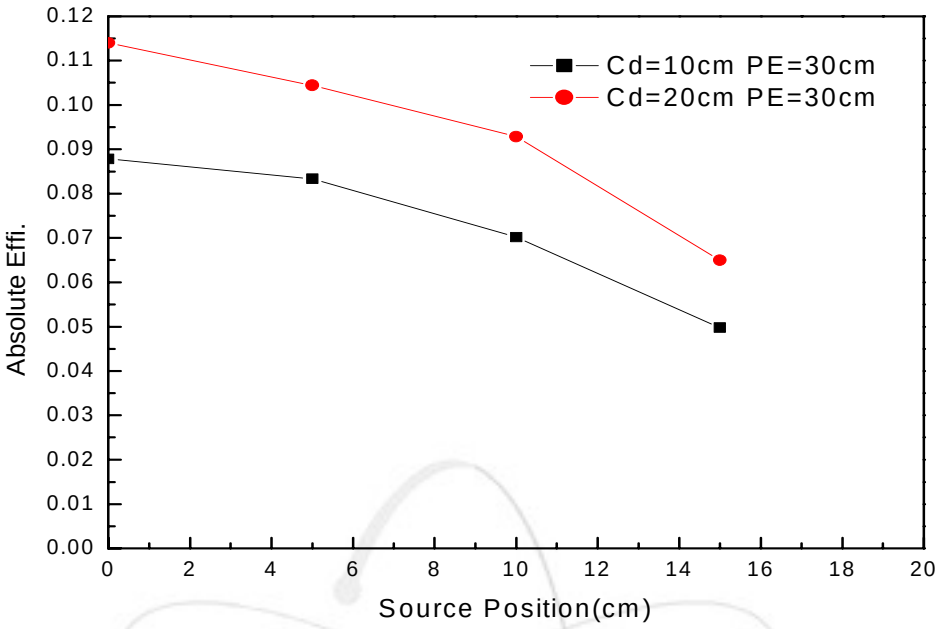

Figure 7. Neutron detection efficiency for 30cm He-tube system.

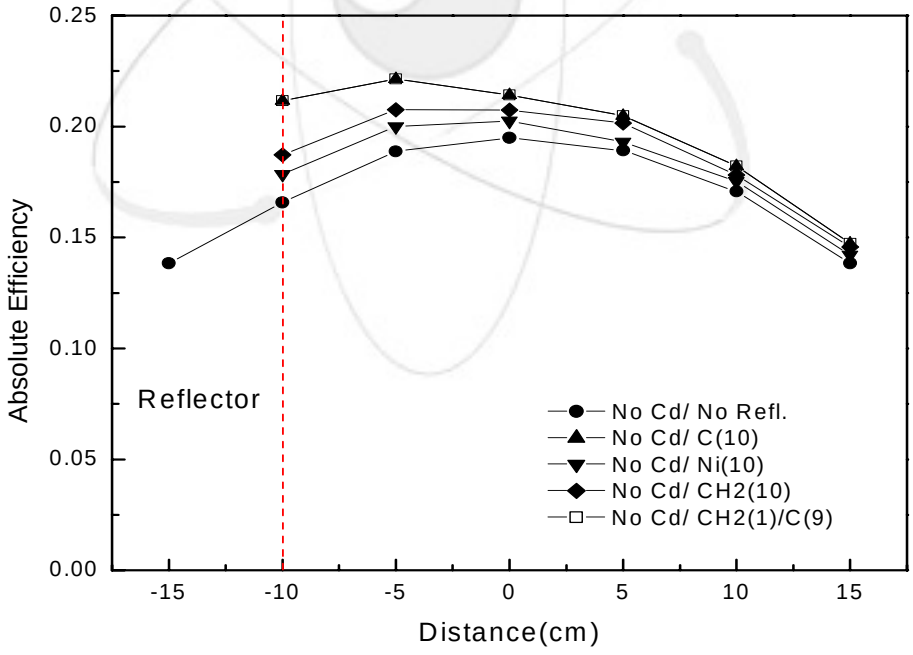

Figure 8. Reflector study for 30 cm of He-3 tube system.

2. Simulation using 50 cm of He-3 tube

Other MCNP simulation using 50cm He tube with Cd absorber was performed. In this case, the 20cm of Cd absorber system with cylindrical shape showed uniform detection efficiency over 20 cm lengths as shown in Fig. 9.

In order to expand over 25 cm uniform efficiency region, several kinds of neutron reflectors were considered. Fig. 10 is the preliminary result for optimization using 50 cm of He-3 tube. The 5 cm of bottom reflector consisted of Ni, C and CH2 were studied. Simulation study informed that reflectors functioned to increase the detection efficiency near reflector side by reuse of the reflected neutron.

As a result, reflector is a useful component to increase neutron detection efficiency near bottom region. Next study will be focused on to satisfy the uniform neutron efficiency over 25 cm using reflector component.

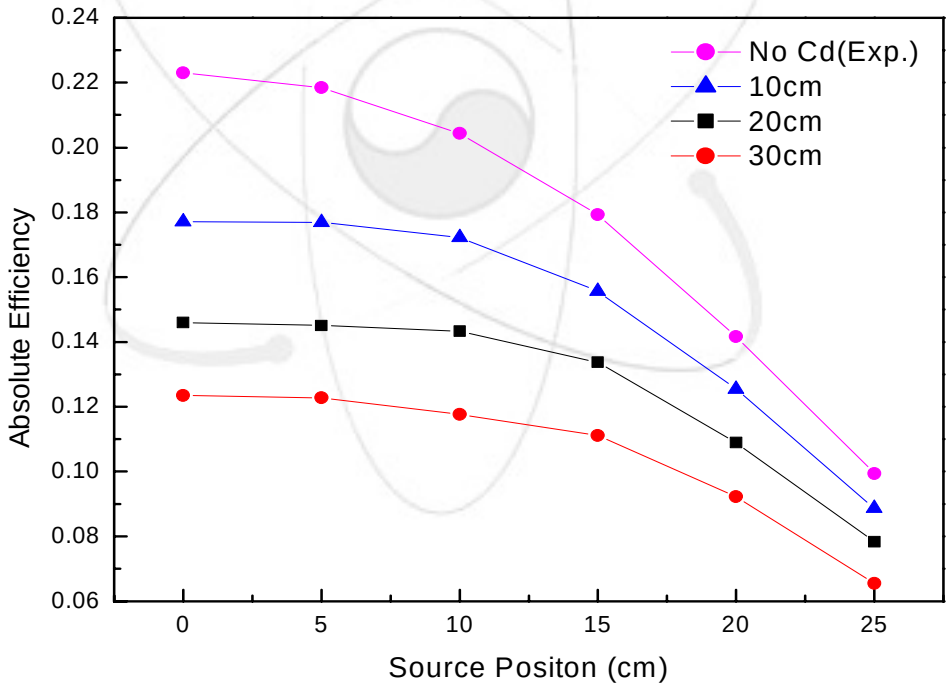

Figure 9. Neutron detection efficiency for 50cm He-3 tube system.

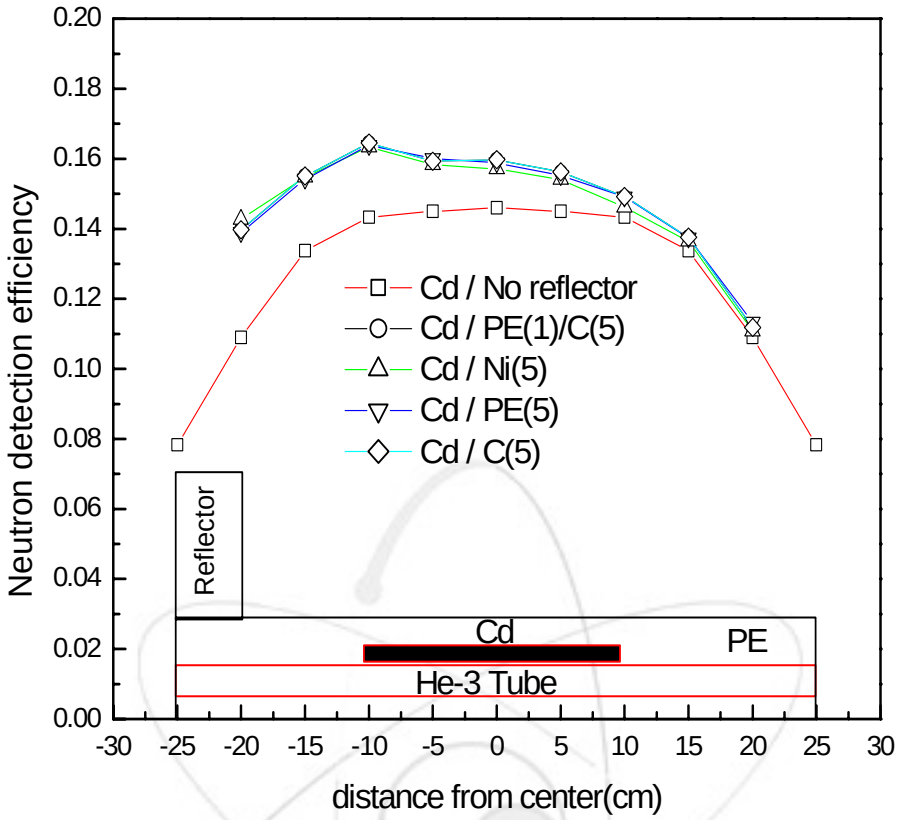

Figure 10. Reflector study for 50 cm of He-3 tube system.

3. Specification of the conceptual neutron NDA system.

Based on MCNP study, the specification of the conceptual neutron NDA system was determined tentatively as followings:

- He-3 tube: 1”D x 20”L x 20EA positioned at diameter of 240mm
- HDPE: inner diameter 177mm/ outer diameter 381mm
- Cd absorber: diameter 240 x 200L x 1.57mmT

4. Die away time and Multiplicity distribution simulation

Using the MCNP-X, the die-away time and multiplicity distribution for conceptual design system were simulated. Fig. 12 and 13 are the corresponding results.

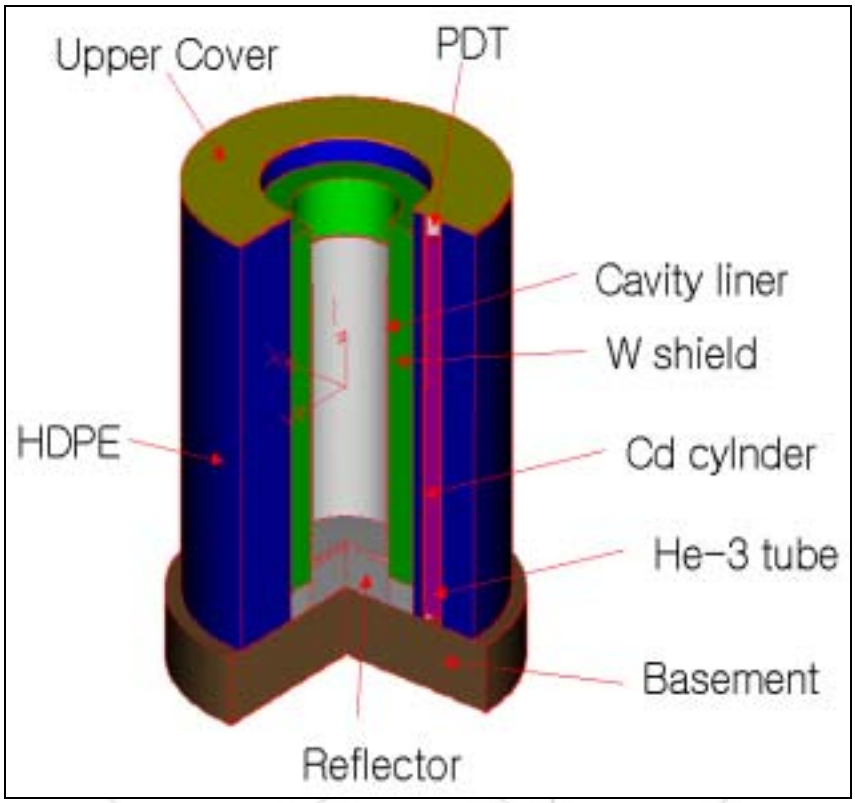

Figure 11. Conceptual Design of ACP NDA system.

5. Other studies for NDA system techniques

a. NDA system calibration

NDA system calibration is needed according to the newly determined parameters [1]. As a result, the neutron production rate is increased 2.2% from 1.36x 10^5 to 1.39 x 10^5 counts/sec gr-Cm244.

b. Cd neutron absorber geometry and placement

In present study, NDA system had simple cylindrical Cd absorber. MCNP simulation gives a good accordance for the system requirement. However, in order to have more degree of freedom for the correction, the Cd absorbers may be needed to place at surfaces of each He-3 tubes.

c. Background study for NDA performance using DSNC data

The operation data of DSNC system that has similar specification to ACP-NDA system were collected. Fig. 14 shows the background results during the operation of last two years since 2000.1.1. From this study, we did not extract pure DSNC operation

performance data because data showed only changes of outside radiation levels according to feed into the SNM in hot cell.

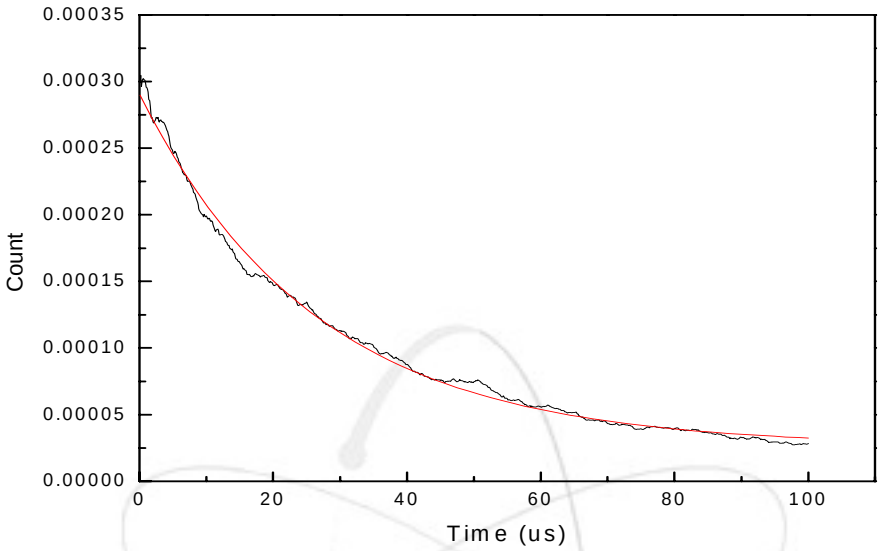

Figure 12. Die-away time simulation using MCNP-X.

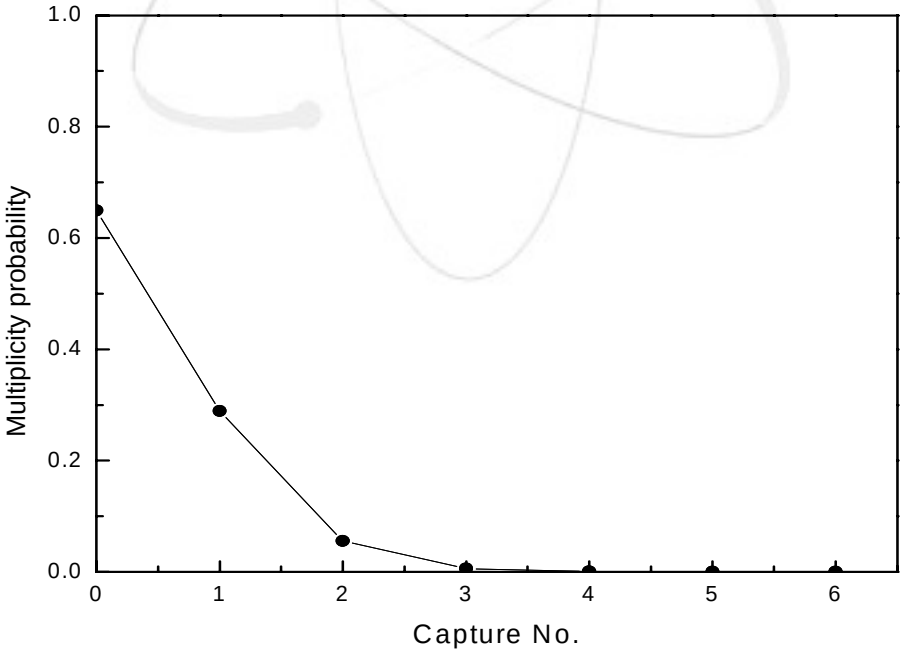

Figure 13. Multiplicity distribution simulation using MCNP-X.

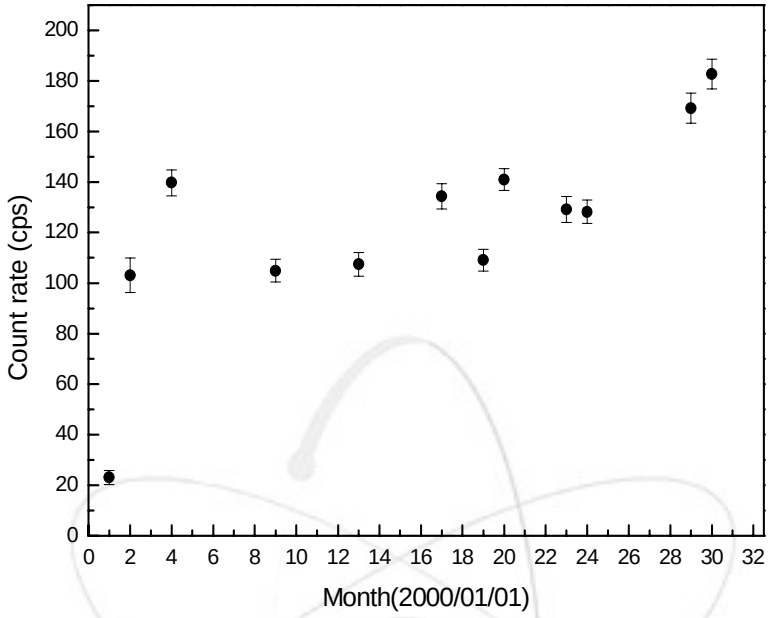

Figure 14. Background level of DSNC from 2000.1.1.

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서지정보양식

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초록 (15-20 줄 내외)

본 연구에서는, 국내에서 수행되고 있는 Li 용융염을 이용한 사용후핵연료의 금속전환체 연구의 원활한 진행에 있어서 핵심 사항인 핵물질안전조치성 평가를 위한 예비 분석을 진행하였다. 본 보고서에서는 ACP 시설에 대한 핵물질안전조치성 분석을 위해 차세대관리공정의 하부 공정과 물질 흐름 및 주요 측정 지점 (Key Measurement Points, KMP) 등을 개념 설계하였다. 또한, 각 핵물질 흐름에서의 핵물질 계량 방안을 제시하였으며, 각 계량관리 기법에 대한 ITV (International Target Value)를 적용하여 재고차 평가에 대한 오차 한계를 계산하였다. 이와 같은 개념 설계 자료에 근거한 핵물질안전조치성 예비 분석 결과, ACP 시설에 적용될 핵물질 계량장비의 측정 정밀도를 만족시키는 경우에는 IAEA 의 핵물질안전조치 목표를 달성할 수 있는 것으로 분석되었다. 향후, 본 연구의 실질적인 활용을 위해서는 ACP 시설의 시설 용량과 MBA 기간 및 측정 오차와 같은 인자들을 고려한 민감도 분석 등의 추가적인 연구가 진행되어야 할 것으로 판단된다.

주제명 키워드
(10 단어 내외)

사용후핵연료, 차세대관리공정, 핵물질안전조치, 핵물질계량관리, MUF 평가

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Project Manager and Department	Lee, Sang-Yoon (Spent Fuel Technology Development)
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Abstract (15-20 Lines)

In this report, a preliminary study on the safeguardability of ACP (advanced spent fuel conditioning process) was conducted with Los Alamos National Laboratory. 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 standard deviation of 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. In the second phase of this study, further study on sensitivity analyses considering various factors such as measurement errors, facility capacities, MBA periods etc. may be needed.

Subject Keywords (About 10 words)

ACP, Spent Fuel, Nuclear Safeguards, MUF, Safeguardability