

# Status of Development of Pyroprocessing Safeguards at KAERI

## 한국원자력연구원 파이로 안전조치 기술개발 현황

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The Korea Atomic Energy Research Institute (KAERI) has developed a safeguards technology for pyroprocessing based on the Safeguards-By-Design (SBD) concept. KAERI took part in a Member-State Support Program (MSSP) to establish a pyroprocessing safeguards approach. A Reference Engineering-scale Pyroprocessing Facility (REFP) concept was designed on which KAERI developed its safeguards system. Recently the REFP is being upgraded to the REFP+, a scaled-up facility. For assessment of the nuclear-material accountancy (NMA) system, KAERI has developed a simulation program named Pyroprocessing Material Flow and MUF Uncertainty Simulation (PYMUS). The PYMUS is currently being upgraded to include a Near-Real-Time Accountancy (NRTA) statistical analysis function. The Advanced Spent Fuel Conditioning Process Safeguards Neutron Counter (ASNC) has been updated as Non-Destructive Assay (NDA) equipment for input-material accountancy, and a Hybrid Induced-fission-based Pu-Accounting Instrument (HIPAI) has been developed for the NMA of uranium/transuranic (U/TRU) ingots. Currently, performance testing of Compton-suppressed Gamma-ray measurement, Laser-Induced Breakdown Spectroscopy (LIBS), and homogenization sampling are underway. These efforts will provide an essential basis for the realization of an advanced nuclear-fuel cycle in the ROK.

Keywords: Pyroprocessing, Safeguards, REFP, PYMUS, ASNC, HIPAI, LIBS

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한국원자력연구원은 IAEA에서 권고하고 있는 안전조치기반설계(SBD)에 입각하여 파이로 안전조치 기술을 개발하고 있다. 한국원자력연구원은 파이로 안전조치접근방안 개발을 위한 IAEA 회원국지원프로그램(MSSP)을 수행하였다. IAEA 회원국 지원프로그램을 통하여 기준파이로시설(REPF) 개념을 설계하고, 이 시설에 대한 안전조치시스템을 개발하였다. 최근에 기준파이로시설은 용량이 증대된 REPF+로 업데이트 되고 있다. 핵물질계량관리시스템 성능평가를 위하여 전산코드 PYMUS를 개발하였으며, PYMUS는 전용탐지회로 통계평가 방안을 포함하여 업그레이드하고 있다. 파이로 입력물질 계량을 위한 비파괴분석장비로 ASNC가 개발되고 있으며, 파이로 출력물질인 U/TRU 잉곳을 계량하기 위한 비파괴분석장비로 HIPAI가 개발되고 있다. 또한 컴프톤 억제 감마선분광기술, LIBS 기술, 균질화 공정의 샘플링 오차에 대한 평가도 진행 중이다. 이러한 노력들은 국내에서 선진핵주기기술 실현에 크게 기여할 것이다.

중심단어: 파이로, 안전조치, 기준파이로시설, ASNC, HIPAI, 레이저 유도 플라즈마 분석법

## 1. Introduction

KAERI has developed a pyroprocessing to reduce the volume, radioactivity, and heat load of light water reactor (LWR) spent fuel. Group recovery of TRU makes pyroprocessing more proliferation-resistant than conventional reprocessing. However, the pyroprocessing facility under IAEA safeguards is not yet operational, and safeguards criteria as well as the overall safeguards approach of the facility remain to be fully established.

In parallel with process technology development and facility design, KAERI currently is developing pyroprocessing safeguards technology. Herein, we introduce the recent pyroprocessing safeguards R&D activities at KAERI.

## 2. Development of Safeguards for Pyroprocessing

Since pyroprocessing differs from the conventional reprocessing in respect of the characteristics of material flow and plutonium-bearing materials, a tailored safeguards approach has to be developed. To that end, the ROK, on behalf of KAERI and other institutes, has contracted with the IAEA's MSSP titled Support for Development of a Safeguards Approach for a Pyroprocessing Plant. As the key initial component of that effort, an REPF concept was de-

veloped [1]. The REPF's input material is LWR spent fuel, and its output materials are U/TRU ingots, U ingots, and waste. Its throughput is 10 MTHM/year.

The main processes of the REPF are receipt and storage of spent fuel, the head-end process, the electrolytic reduction process, the electro-refining process, the electro-winning process, and the waste process. Additionally included is a homogenization process for obtainment of representative input-material-accounting samples.

Three Material Balance Area (MBA)s were identified in the NMA system of the REPF: MBA-1, the spent-fuel storage and head-end process; MBA-2, the main process area in argon cells, and MBA-3, the storage area for products and waste. Flow and inventory KMPs also were identified, and possible accounting methods for each KMP were specified. The important process materials for which accurate accounting is necessary were homogenized as input spent-fuel powder, U/TRU ingots, and U ingots. The REPF's NRTA system's NDA methods are based mainly on neutron balance using the Pu/Cm ratio [2, 3]. A Sigma-MUF evaluation of the NMA system concluded that the overall REPF safeguards system meets the IAEA's detection goal.

Recently, the REPF is being upgraded to the REPF+, a 30 MTHM-throughput facility, in order to investigate the scale-up effect on the safeguards. One of key features of

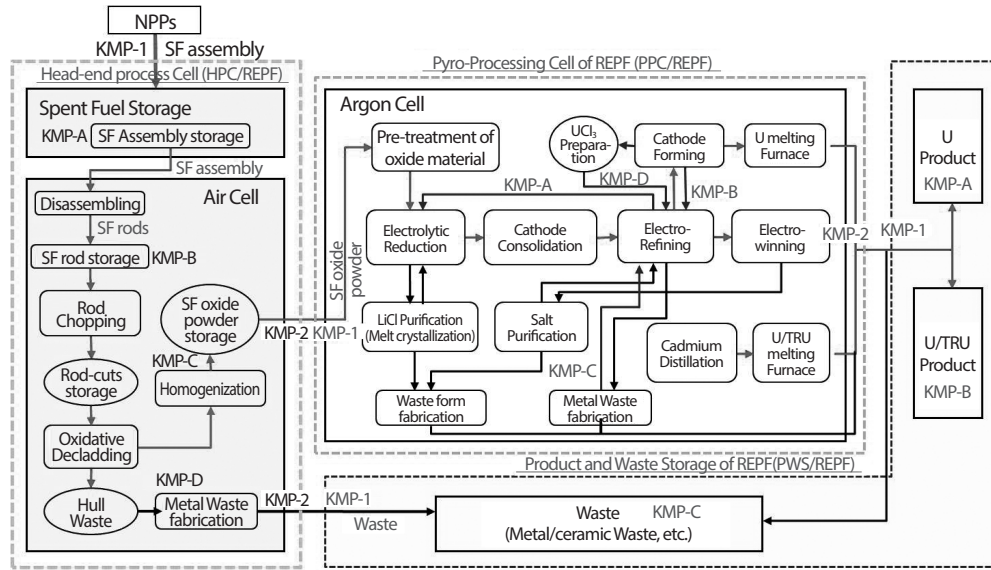


Fig. 1. PYMUS Simulation: Material Flow and KMPs of REPF.

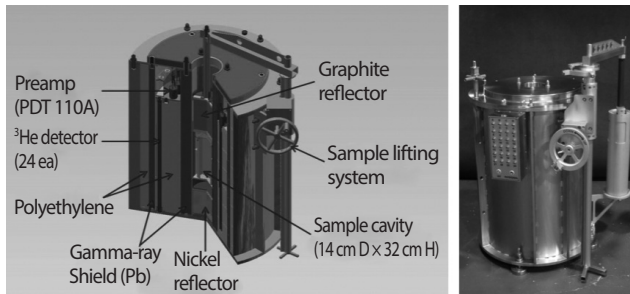


Fig. 2. Upgraded ASNC. In this version, the sample can be loaded vertically with the remote sample-lifting system.

the REFP+ is its allowance for unlimited nuclear-material mixing between campaigns, which was limited in the REPF.

For analysis of nuclear-material flow and calculation of MUF uncertainty, a simulation program, PYMUS, has been developed (see Fig. 1) [1, 4]. PYMUS currently is being upgraded for evaluation of detection probability based on statistical testing for various diversion scenarios.

For NMA of the process material of an Advanced spent-fuel Conditioning Process Facility (ACPF), an ASNC was developed. Its performance was successfully tested with spent-fuel-rod cuts in 2007 [5-7]. More recently, the ASNC

has been upgraded for enhanced remote maintenance capability (see Fig. 2) [8]. Some detector parameters of the ASNC were measured for a <sup>252</sup>Cf standard source after installation into the hotcell. The measured values were 24.3%, 64 μs, 62.4 μs, and 0.5102 for detection efficiency, gate length, die-away time, and doubles gate fraction, respectively. Based on the results of an ASNC performance test, the measurement error of NDA-based input accountability will be determined.

Most of the plutonium processed in pyroprocessing is accumulated in U/TRU-ingot form. Unfortunately, the complexity of U/TRU compositions - specifically the significant neutron multiplications by induced-fission events - complicates precise measurement of U/TRU-ingot Pu mass by NMA. In order to enable measurement of U/TRU-ingot Pu mass without using the Cm/Pu ratio, novel prototype NMA equipment, namely the HIPAI, has been developed [9, 10]. This is in fact a hybrid system combining two measurement techniques based on fast- and thermal-neutron-induced fission events, respectively. The fast-neutron-based technique is Fast-Neutron Energy Multiplication (FNEM), and the thermal-neutron-based technique is Passive Neu-

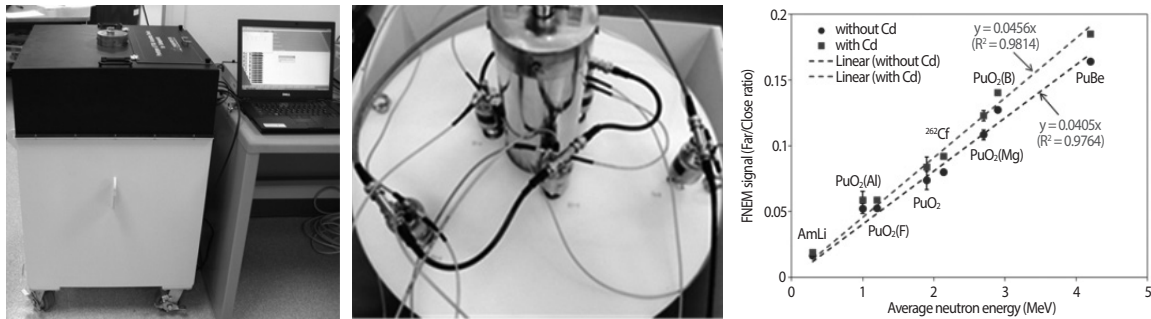


Fig. 3. HIPAI for U/TRU-ingot Pu accounting.

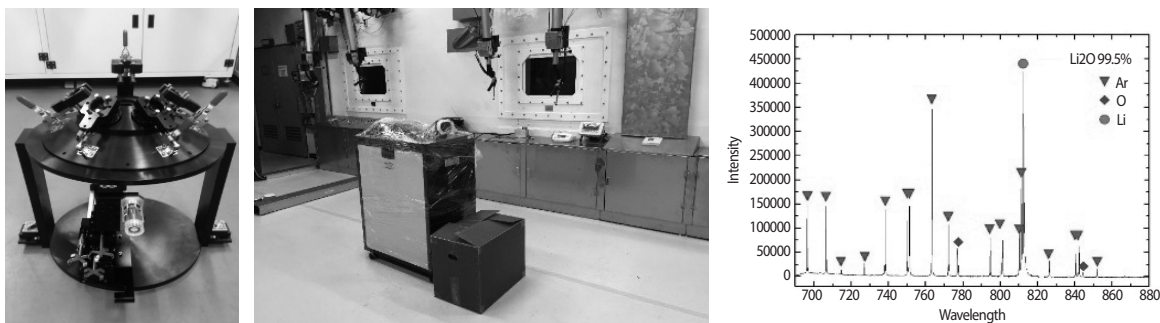


Fig. 4. LIBS installed at ACPF.

tron Albedo Reactivity (PNAR). By HIPAI's effective combination of these two methods, the Pu-mass signature is significantly enhanced.

Fig. 3 shows the prototype HIPAI. It was fabricated using 6 <sup>3</sup>He tubes, and its performance was evaluated using AmLi, PuBe, <sup>252</sup>Cf, and Pu sources. The measurement results were also shown in Fig. 3. The error bar represents the measurement precision, i.e.,  $\pm 1\sigma$ , which is mainly dependent on the detection efficiency of the far ring detectors. The measured signal showed good linearity for with and without the Cd liner cases ( $R^2 \approx 0.98$ ).

Another complication typically encountered is high-energy gamma-rays' large Compton background, which prevents identification of meaningful peaks in the energy spectrum of spent fuel or pyroprocessing material containing a large portion of rare earth elements. However, the Compton background can be reduced by the Compton-suppression technique. A Compton-suppressed detector system based

on analog signal processing has been installed at KAERI's Post-Irradiation Examination Facility (PIEF) for effective measurement of X-ray and Gamma-ray energy spectra [11]. Further, the List-mode Data Acquisition System (DAQ) based on digital signal processing has been incorporated into the Compton-suppression detector system, the performance of which will soon be tested.

KAERI also is developing LIBS for safeguards applications. LIBS is a promising technique, as it makes possible the analysis of material without any preliminary, time-consuming sample preparation. Fiber-Optic LIBS, which delivers laser energy to the target and collects the reflected plasma light through optical fiber [12], is one options applicable to hot-cell environments. The performance of the FO-LIBS system for measurement of process-material Pu/U ratio has been installed at the air cell of the ACPF, and its performance will be tested as spent fuel becomes available.

A proposed method for precise input-material accoun-

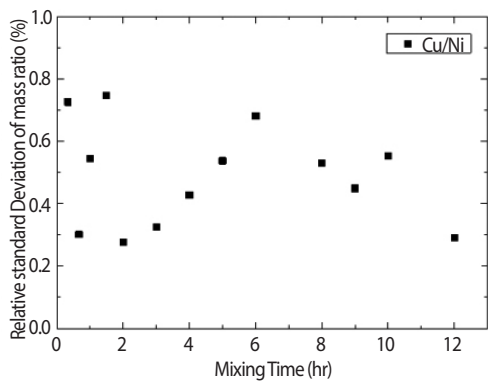
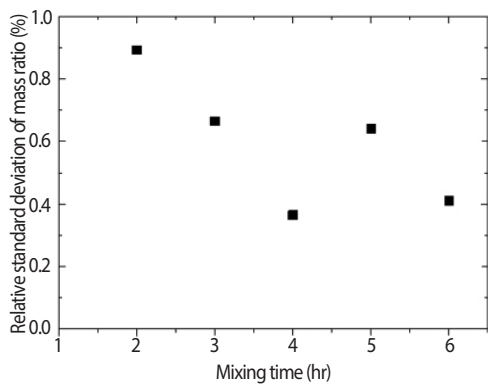
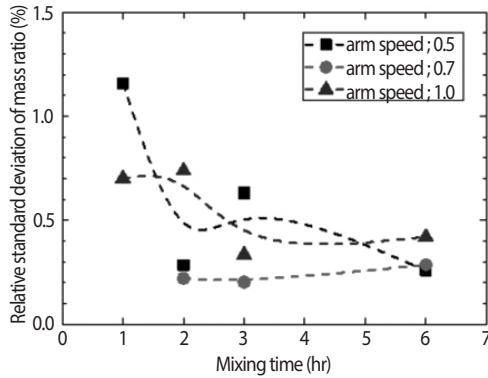


Fig. 5. The measured Relative Standard Deviation (RSD) of the mass ratio from the mixing test [Top: Nauta, Bottom left: 10 kg tumbler, Bottom right: 50 kg tumbler].

tancy at a pyroprocessing facility is Pu-mass measurement of sample obtained from a homogenization mixer. For this purpose, KAERI has proposed a large-sized Nauta mixer that can mix one assembly at once. This mixer has limitations such as difficulties in remote-repair and control for

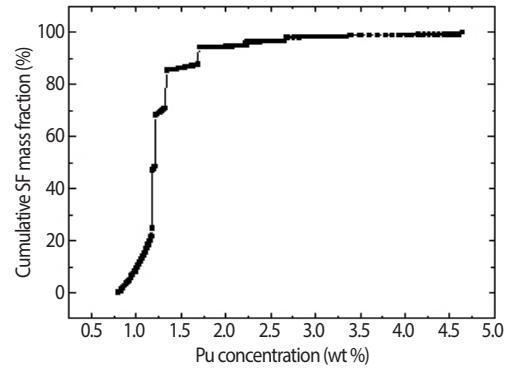


Fig. 6. Cumulative spent fuel mass fraction as a function of Pu concentration.

hot cells. Recently, a two-stage mixing process has been introduced. In the 1<sup>st</sup> stage, the homogeneity of the output material is less than adequate. Part of the output materials from the 1<sup>st</sup> state are collected, and they are mixed again in the 2<sup>nd</sup> stage for higher homogeneity. Moreover, the mixer sizes in the two-stage mixing process are smaller, as is suitable for hot-cell operation. A Nauta mixer with a capacity of 10 kg, and two mixing containers with a capacity of 10 kg and 50 kg were fabricated and tested with surrogate materials in the identical condition. 45  $\mu$ m CuO and NiO powder were charged into each mixer, and mixed for several hours. 1 g samples were taken at each hour, and the uncertainty of the powder mass ratio was analyzed by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) whose instrumental uncertainty was 0.5% in ANOVA analysis. The analyzed results are shown in Fig. 5 in which the uncertainty (RSD) of the mass ratio is less than 0.8% after 2 hours mixing irrespective of the mixer type and size.

In order to estimate the Pu heterogeneity based on test result, the powder heterogeneity in a binary mixture of CuO/NiO was expanded to the powder heterogeneity as a function of powder mass fraction in a multi-component mixture having various types of spent fuel powder with different Pu concentrations [13]. And as shown in Fig. 6, the distribution of Pu concentration of a 59 GWd/tU spent fuel



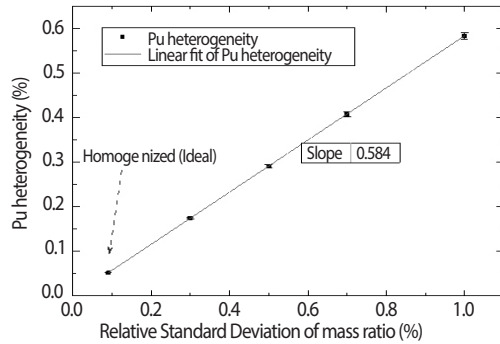


Fig. 7. The estimated Pu heterogeneity.

was calculated using a rod axial gamma scan profile and a RAPID program to simulate burn-up, U, and Pu contents in a radial direction. In this calculation, there are three assumptions; 1) The size of all powder particles is same about 45  $\mu\text{m}$  after mechanical decladding and low temperature oxidation, or small difference of powder particle size does not affect on mixing performance. 2) The powder heterogeneity between CuO/NiO powder and spent fuel powder is same even though theoretically spent fuel powder can be more heterogeneous than CuO/NiO powder because of a higher density of spent fuel powder. 3) The distribution of Pu concentration in spent fuel powder after decladding and low temperature oxidation process is same with the distribution of Pu concentration in a spent fuel rod. Pu mass in a 1g sample is  $\sum(m_p \times \rho_{Pu})$ , in which  $m_p$  is each spent fuel particle mass (or mass of each mass fraction group) and  $\rho_{Pu}$  is the Pu concentration of each powder particle, thus as shown in Fig. 7, a Pu heterogeneity is a function of the powder heterogeneity ( $\sigma_{mp}$ ) because  $m_p$  is only an error term. The Pu of spent fuel powder mixed by the Nauta and tumbler mixer is more heterogeneous than homogenized powder as shown in Fig. 7. However, the Pu heterogeneity (RSD) at the mass ratio uncertainty of 1% is lower than 0.6% which does not significantly affect the Pu accounting uncertainty. The Pu accounting uncertainty in the two-stage mixing process is affected not only by the Pu heterogeneity, but also by the mass uncertainty charged into the 1<sup>st</sup> stage mixer and

the sampling mass uncertainty in the 1<sup>st</sup> stage mixer. A Pu accounting uncertainty lower than 1% can be accomplished with the mass uncertainty lower than 2% and sampling mass uncertainty lower than 5% in the 1<sup>st</sup> stage mixer. The detailed calculation procedure is explained in [13].

The goal of the safeguards system design is effective and efficient implementation of the safeguards to meet the IAEA technical objectives, and the safeguards system can be assessed by estimating the MUF uncertainty or Pu diversion detection probability of the NMA system. The precise input material accountancy is very important to the effectiveness of the safeguards, and the NDA technologies such as ASNC, HIPAI, and Compton-suppressed gamma-ray measurement system are very important to the efficiency of the safeguards system. LIBS is one of the promising technologies especially to enhance the timely detection of the misuse of the facility.

### 3. Summary

KAERI has developed a pyroprocessing for the recycling of the spent fuel. In parallel, it has developed a safeguards approach for a particular pyroprocessing facility (the REPF), along with the relevant technology, through the IAEA's MSSP. Prototype safeguards instrumentation such as the hybrid NDA technique and LIBS has been developed, and their performances will be tested. In the meantime, close international collaboration on the pyroprocessing safeguards continues. All of these efforts will support the implementation of effective and efficient safeguards for pyroprocessing facilities in the ROK.

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