Research Paper https://doi.org/10.7733/jnfcwt.2024.026

Radiation Monitoring of Nuclear Material in Process for Reducing Environmental Burden

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(Received April 25, 2024 / Revised May 20, 2024 / Approved June 3, 2024)

A procedure for minimizing the environmental burden and maximizing the efficiency of storage sites used for the final disposal of spent fuel has been proposed. In this procedure, fission products (highly mobile and producing heat) are collected, and uranium and TRU-RE (transuranium-rare earth) oxide are independently stored. The possibility and applicability of radiation measurement for monitoring the nuclear materials effectively throughout the process has been simulated and evaluated. For the simulation, the properties of the chemical processes were analyzed, the major radiation emitters were determined, and the production of nuclear materials by chemical reactions were evaluated. In each process, the content of nuclear material was changed by up to 20% to represent abnormal conditions. The results showed that the plutonium peak was matched with the change in the TRU content and the measured signal was changed linearly with respect to the content change of the plutonium. From the neutron measurement, a linear response of the TRU content variation was obtained. In addition, a logic diagram was developed for the nuclear monitoring. The integration of radiation detections is recommended for monitoring the process effectively and efficiently.

Keywords: Nuclear material, Monitoring, Fuel process, Measurement, Radiation, Spent fuel

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1. Introduction

As nuclear energy will play an important role in reduction of global carbon emissions and energy supply, a steady increase in nuclear power plant is also expected in Korea. However, due to the long-term operation of nuclear power plants, wet storage within the power plant is reaching the point of saturation for spent fuel. As an alternative, temporary use of a new power plant storage pool and construction of a dry storage facility near the power plant are planned, however, ultimately, a final disposal site is required. Approximately 750 tons of spent nuclear fuel is generated annually in Korea; with approximately 400 tons from light-water reactors and 350 tons from heavy-water reactors. Storage facilities are expected to be saturated starting in 2021 for the heavy water reactor, Wolseong, and for the light water reactor in that order: Hanbit ’30, Hanul ’30, Kori ’32, and Shin Wolseong ’42. Moreover, following the decision to dismantle Kori Unit 1, there is an urgent need for a plan to export and manage the spent nuclear fuel stored in wet storage. Therefore, it is necessary to quickly prepare a final disposal site for spent fuel and develop core technologies for the safe management of spent fuel.

In order to maximize the economy, efficiency, and stability of spent nuclear fuel management, a research is being conducted on various technologies to minimize the environmental burden during final disposal, through burn of major toxic materials using the nuclear fuel cycle and chemical process for high heat production and mobile materials [1]. In developing the chemical process, it is necessary to secure monitoring technology that can evaluate nuclear materials in the process. A nuclear material monitoring that reflects non-proliferation requirements from the early development stage of the process is also a recommendation in the international nuclear non-proliferation system. Additionally, according to the U.S. 10CFR, nuclear materials and high-level waste must be subject to verification and monitoring of major nuclides.

The possibility of measurement of emitted radiations from nuclear material was investigated for application of nuclear monitoring in the selected chemical process. Major gamma rays and corresponding nuclei are well defined property and neutron emitters are also well known information for spent fuel [2, 3], even though the information depends on initial enrichment and burnup. To simulate monitoring of nuclear materials in the process, the nuclear materials produced in the chemical process were evaluated, and the major nuclides and measurable radiations were determined for each salt process. The appropriate location of detectors for radiation monitoring was determined. The radiation signal was obtained and analyzed for normal and abnormal conditions. The spectrum analysis was performed when abnormal processes occur. The relationship of the measurement signals for the change of nuclear material contents was investigated and the linearity of signals was analyzed at the selected energy and total detection sum for all processes. For the final product, fuel block, radiation measurement technology was also applied to determine continuous of nuclear material information. In addition, an algorithm for predicting the operation status of the process was developed.

The radiation measurement is relatively simple and convenient way in monitoring nuclear materials for the process, because major gamma and neutron emitters are prominent and TRU-RE is conserved in the process. Therefore, if radiation monitoring system is accompanied by current monitoring technologies [4-8], it will be very effective and efficient in monitoring nuclear materials for the process.

2. Process for Reducing Environmental Burden

Various chemical processes are being developed to reduce the environmental burden when disposing of spent nuclear fuel [1]. In the development, safe management of storage site was also considered in final disposal of spent fuel. In the selected process [1] for the simulation, the main

Table 1. Spent fuel composition for the process (4.5%, 55 GWd·MTU^{-1}, 10 yrs cooling)

Nuclide (ZZAAA)	Content (wt%)	Nuclide (ZZAAA)	Content (wt%)	Nuclide (ZZAAA)	Content (wt%)
8016	1.16410×10^{-1}	60147	1.85244×10^{-16}	92233	2.50776×10^{-11}
36083	7.44654×10^{-7}	61147	1.21899×10^{-5}	92234	7.90677×10^{-7}
42095	6.87372×10^{-4}	62147	2.17084×10^{-4}	92235	7.71366×10^{-3}
45103	4.27620×10^{-4}	60148	3.35702×10^{-4}	92236	3.83065×10^{-3}
46105	3.40362×10^{-4}	61148	2.20648×10^{-18}	92238	8.14496×10^{-1}
45105	2.80119×10^{-17}	61149	2.13509×10^{-17}	93237	3.30917×10^{-4}
46108	1.20277×10^{-4}	62149	3.31162×10^{-6}	94238	9.30968×10^{-5}
47109	7.68853×10^{-5}	62150	2.71226×10^{-4}	94239	4.62566×10^{-3}
48000	7.94086×10^{-8}	62151	1.23013×10^{-5}	94240	1.92605×10^{-3}
53127	7.49858×10^{-7}	62152	1.14834×10^{-4}	94241	7.01519×10^{-4}
54131	7.40779×10^{-6}	63151	9.97031×10^{-7}	95241	4.46742×10^{-4}
55135	6.27974×10^{-6}	63153	1.05279×10^{-4}	95242	3.89381×10^{-7}
53135	2.49381×10^{-19}	63154	1.36336×10^{-5}	94242	4.41929×10^{-4}
54135	3.76187×10^{-19}	63155	7.94059×10^{-7}	95243	9.15009×10^{-5}
60143	7.24303×10^{-4}	64155	1.03129×10^{-5}	96244	4.40000×10^{-7}
60145	6.16509×10^{-4}	64157	3.74738×10^{-8}		

heat production materials, Cs and Sr, and the highly mobile materials, I and Tc, are separately collected and disposed as a waste. The selected process stores TRU-RE and uranium oxide independently from spent fuel. The characteristics of internal process were analyzed and the process products, salt substances, and major radioactive nuclides are evaluated and determined [1]. In the process, uranium oxide is recovered in LiCl−KCl, which contains most of the content of spent nuclear fuel, and uranium block is fabricated to manage storage site independently. TRU-RE chloride is produced in NH_4Cl and LiCl−KCl. TRU-RE phosphate is finally collected in Li_3PO_4. It is fabricated as a block form to dispose at the final storage site. Therefore, uranium and TRU-RE are not mixed in the final disposal. From the independent disposal, the process has the advantage of maximizing the capacity of the final disposal site. Moreover, the process has the feature that TRU-RE is always consistent in the internal process, without separation and extraction. Therefore, plutonium is conserved within TRU in the entire process. The simulation was performed to determine the applicability of radiation monitoring and the feasibility of measurement signals on the process. Moreover, the development of the process is expected to contribute not only to the management of spent nuclear fuel, but also to increasing safety in the final disposal of high-level waste at nuclear fuel cycle.

3. Radiation Monitoring

3.1 Process Property

3.1.1 Head-end process

The nuclides emitting main gamma-ray from spent fuel, such as ^{60}Co, ^{95}Zr, ^{95}Nb, ^{105}Rh, ^{65}Zn, ^{137}Cs, ^{134}Cs, ^{144}Pr, and ^{154}Eu [2, 3], are used for measurement. The structure is disassembled from the spent fuel assembly in the head-end process, and the major gamma-ray emitting nuclides from

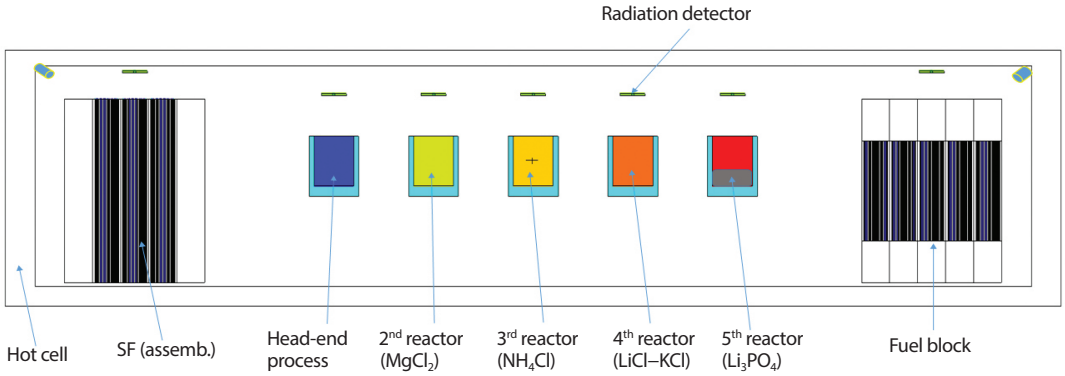


Fig. 1. Schematic view of radiation monitoring.

the structure material are removed through the decladding process. Cesium, a representative and volatile nuclide emitting intense gamma rays [2, 3] from spent nuclear fuel, is all collected as the process temperature increases and disposed of as waste. Therefore, the monitoring simulations were performed by assuming that there would be no interference in gamma ray measurement by cesium isotopes in further process. In the head-end process, the form of oxide powder is produced and supplied to the $MgCl_2$ salt process. Iodine, technetium, and krypton are captured and disposed of as waste. The possibility of measuring ^{239}Pu and ^{235}U from the oxide powder, including uranium, TRU, and fission products, was simulated. Almost all of the major rare-earth gamma-ray emitting nuclides are present, and ^{244}Cm, the major neutron emitting nuclide, remains constant along with TRU. To perform the radiation monitoring, the composition of spent nuclear fuel was obtained from 4.5% enrichment, 55 $GWd{\cdot}MTU^{-1}$ burnup, and the 10 years of cooling [9]. Table 1 shows the spent fuel composition for the simulation.

3.1.2 Salt process

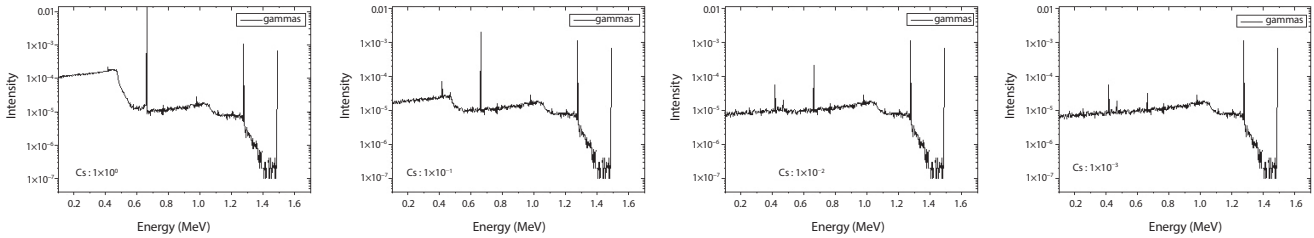
Since nuclear materials and rare-earth materials coexist in the TRU-RE oxide and chloride product during the chemical reactions, the possibility of measuring gamma ray emission from ^{239}Pu and ^{235}U along with ^{144}Pr and ^{154}Eu was evaluated. Due to the presence of TRU in all processes, measurement of spontaneous fission neutron emission by ^{244}Cm was also evaluated for nuclear monitoring. The major product by chemical reaction in the internal process is summarized [1];

- Uranium and (TRU+RE) oxide is present and strontium and barium are collected in $MgCl_2$ salt
- The oxidized nuclear material is separated into chloride of (TRU-RE) and oxide of uranium by applying NH_4Cl and uranium oxide is selectively recovered in LiCl−KCl
- Finally, (TRU+RE) phosphate is obtained from (TRU+RE) chloride in Li_3PO_4 salt

The schematic view of material flow, reactors, chemical salt and detectors for radiation monitoring are summarized in Fig. 1.

3.2 Simulation on Abnormal Condition

In order to estimate the possibility of applying nuclear material monitoring through radiation measurement, the condition was determined that can cause abnormal internal processes. The measured signals for major nuclides were analyzed by changing nuclear material content, up to 20% [10, 11, 12]. Since the nuclear materials generated in each process exist in the form of oxides and chlorides, and with the salts, simulations were performed to suit the characteristics of the nuclear materials in each process. In



(a) with Cs (b) Cs 10 times down (c) Cs 100 times down (d) Cs 1,000 times down

Fig. 2. Measurement influence by Cs in the head-end process (gamma).

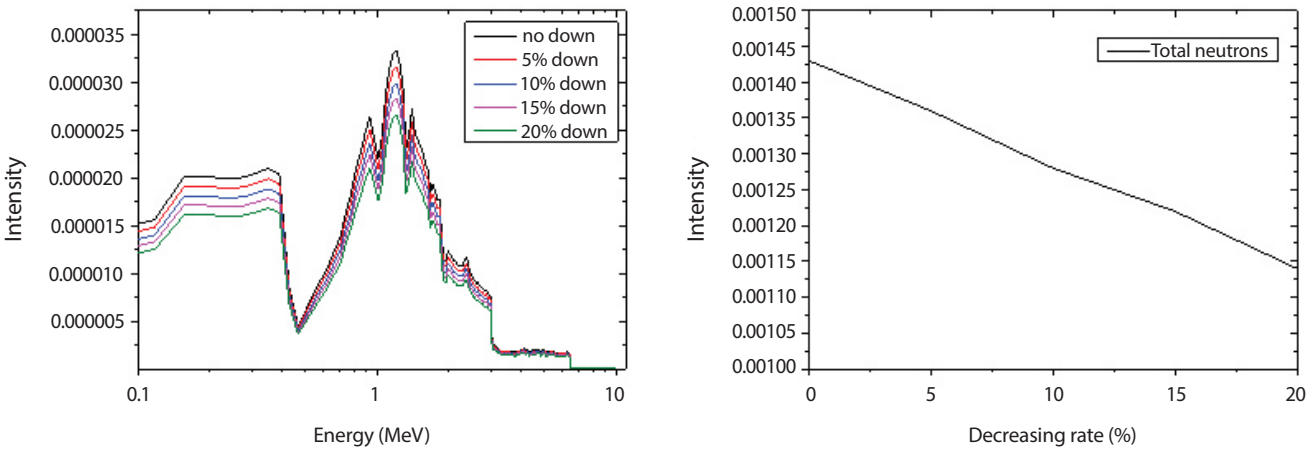
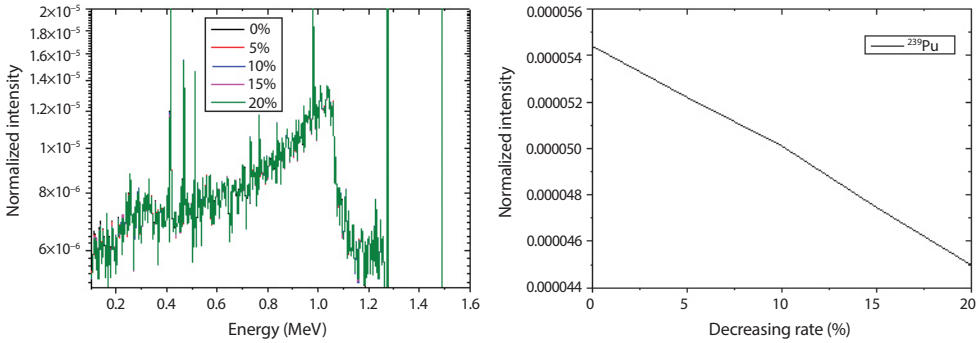


Fig. 3. Neutron measurement in the head-end process (change up to 20%).

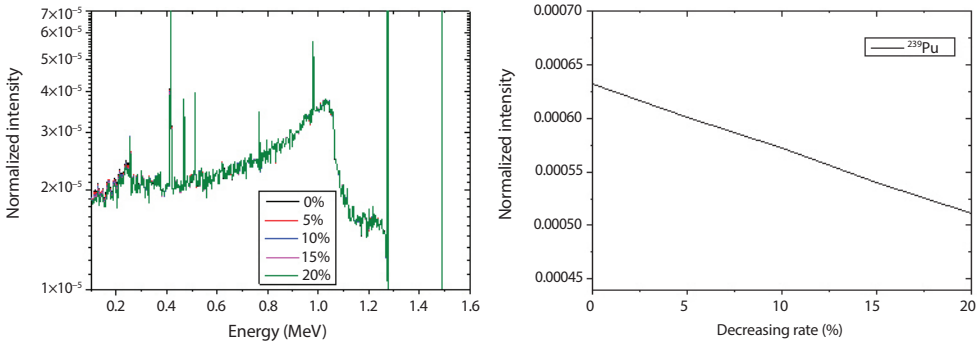
the simulation, the detection possibility in the event of an abnormal condition, the relationship between signal and content change in nuclear material, and the linearity of the measurement signal were analyzed. The gamma-ray emitting nuclides, their energies and intensities, were determined, and the energy spectrum of the neutron source was utilized for the measurements. Alumina with a diameter of 40 cm, a thickness of 5 cm and a height of 100 cm was used as the chemical reactor. The amount of spent nuclear fuel processed in the reactor was assumed to be 2 kg. The density of the salt in the process was 1.5 to 4 $g \cdot cc^{-1}$ depending on the type of salt, and the density of oxidized nuclear fuel block was 5.6 $g \cdot cc^{-1}$ in the simulation [13]. At the final product, solidified blocks of 5 cm in diameter and 10 cm in length were created for the fuel block. By considering disposal site, the total length of the block was assumed to be 100 cm. In addition, the assembly of fuel blocks was composed of 5×5 by considering the equivalent volume of spent fuel assembly.

3.2.1 At head-end-process

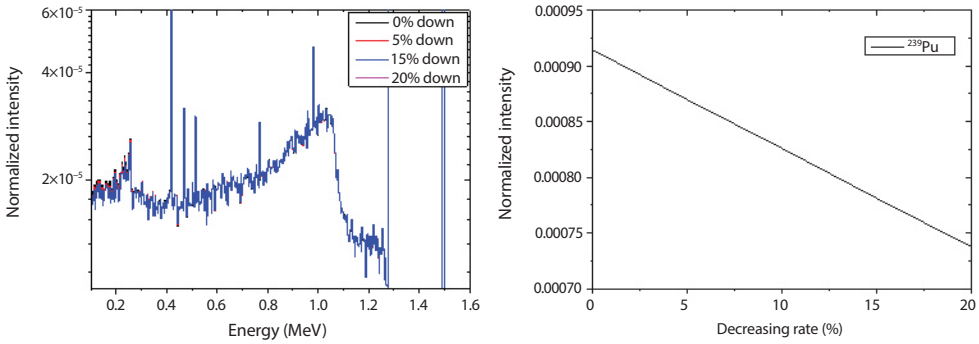
Cesium directly affects the measurement of plutonium and may cause signal distortion in the measurement of plutonium. Therefore, the possibility of measuring plutonium according to changes in Cs content (10, 100, 1,000 times reduction based on spent nuclear fuel) was examined and the relationship in the measurement signal was analyzed. To measure emitted gamma rays, the CdZnTl (1 inch in dia, 2 inches long) was used and a 10 cm thick lead shielding material was applied around the detector. The detector was located above the center of the chemical reactor. From the measurements, the plutonium peak started to appear at a



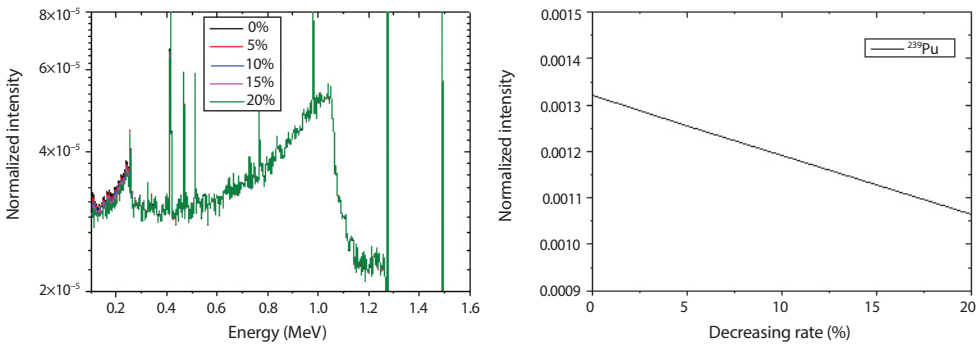
(a) (TRU-RE) and U oxide (at $MgCl_2$)



(b) (TRU-RE) chloride and U oxide (at NH_4Cl)

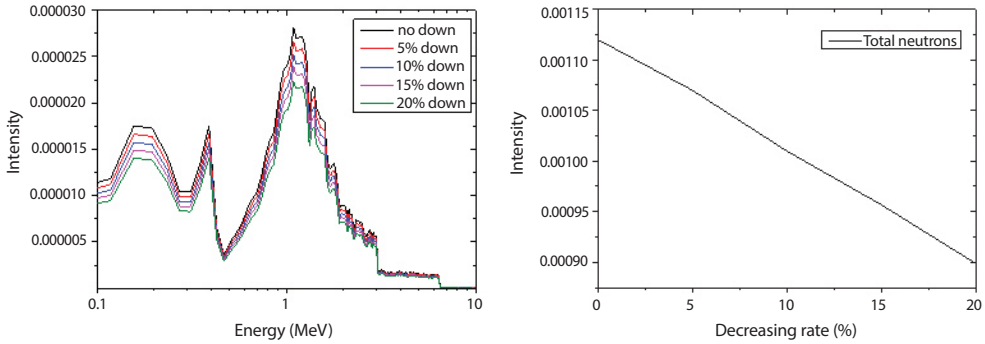


(c) (TRU-RE) chloride and U oxide (at LiCl−KCl)

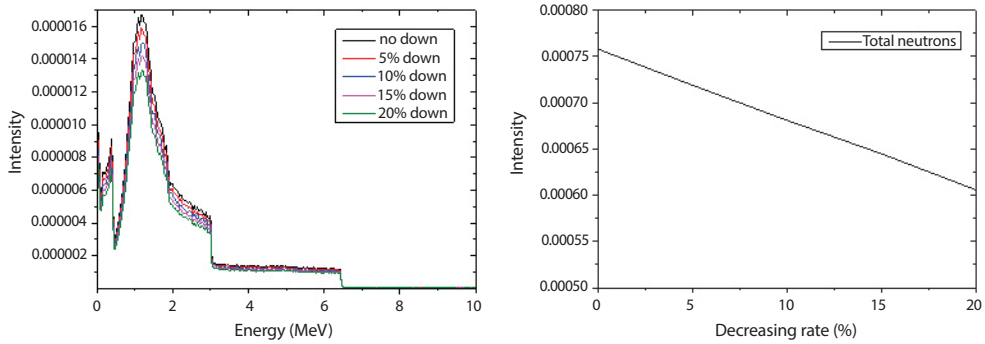


(d) (TRU-RE)PO_4 sediment (at Li_3PO_4)

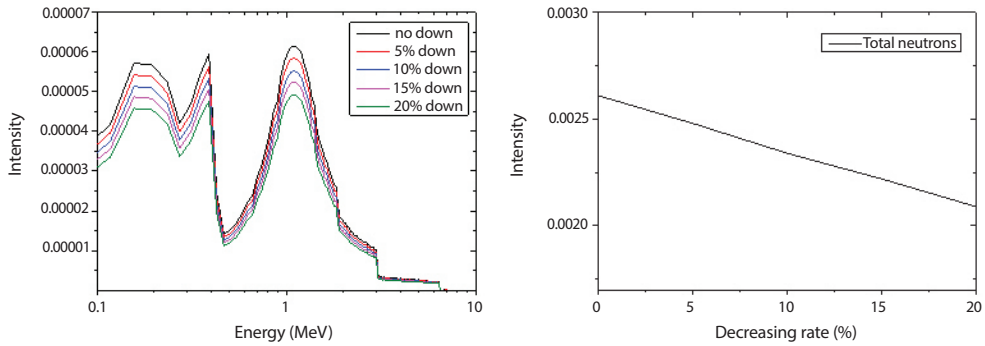
Fig. 4. Detection and linearity in the salt process (~20wt% change).



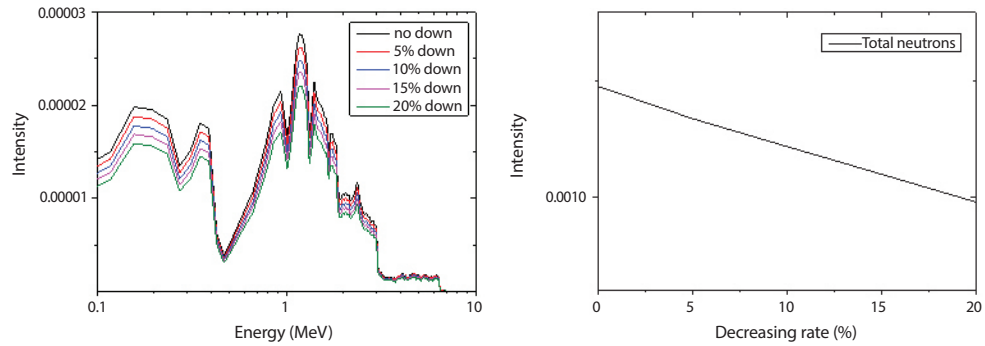
(a) in $MgCl_2$



(b) in NH_4Cl



(c) in LiCl−KCl



(d) in Li_3PO_4

Fig. 5. Neutron measurement and linearity at processes (~20wt% change).

10 times reduction of Cs, but the plutonium peak becomes evident at a 100 times reduction of Cs, as shown at Fig. 2. Moreover, changes in cesium content affect the overall measurement probability in the low-energy region. Therefore, even though it is a small amount presence of cesium, it influences on the plutonium measurement. At the head-end process, a clearance of cesium is very important for further measurements.

Additional simulations were performed for the application of nuclear monitoring through neutron emission measurements. The major source neutron is emitted from ^{244}Cm by spontaneous fission for TRU. The ^{4}He detector was used in neutron measurement (1 inch in diameter and 2 inch long), and a 10 cm thick polyethylene shielding material was used around the detector, which was located above the center of the chemical reactor. The ^{4}He has the characteristic of having a reaction threshold with neutrons at high energy, therefore, it is suitable for measuring fast neutrons. The detection signal in the simulation was obtained by using the MCNP code [13]. The detected signal is simply expressed as

$$\varepsilon \int_A \int_E \varphi\,(r,E,t)\,\sigma_{reaction}) \; dEdA \qquad (1)$$

where φ(r,E,t) is the source neutron arriving at detector, depending on position, energy and time, $\sigma_{reaction}$ is the energy dependent reaction cross section at detector, A is the detector area, and ε is the detector efficiency. The reaction cross section is totally dependent on detector material. The detected signal was classified by the neutron energy. In the simulation, the change in the measurement signal was analyzed while reducing the nuclear material content by 5, 10, 15, and 20%. Considering the characteristics of the instrument, a relatively high energy region, above 0.1 MeV, was used. The reduction in neutron signal due to the decrease in nuclear material content was clearly shown in the detected energy spectrum, at Fig. 3. Moreover, for practical neutron measurement, the total energy sum of the measured signals was also investigated. The result shows that the measurement signal decreases linearly as the nuclear content decreases. Therefore, it was shown that it is possible to detect signals according to changes in nuclear material content even from the measurement of neutron signals.

3.2.2 At salt process

For the nuclear monitoring of salt processes, simulations were performed when the nuclear material content changes in the salt process. The linearity between the signal and the content change was verified and the possibility of determining whether there is an abnormality in the process was evaluated. The simulation on the detection signal was performed using the MCNP code [13]. The detected signal is simply expressed as

$$\varepsilon \int_t \int_E h(E)\,\varphi\,(r,E,t)\; dEdt \qquad (2)$$

where h(E) is energy deposition at the detector, φ(r,E,t) is the photon arriving at the detector and ε is detector efficiency.

From the measurement results in Fig. 4, there was an energy peak of gamma ray emitted by plutonium from the uranium and TRU oxide mixture in the $MgCl_2$ salt, and linearity according to content reduction was satisfied. It shows the reliability of the detection of changes in plutonium due to changes in TRU content. In the NH_4Cl reaction, the measured spectrum was differentiated according to the change in the content of nuclear material, and the linearity of the signal change was also shown to be satisfactory. In the LiCl–KCl process, a linear signal for the change in plutonium content was obtained through measurement using TRU-RE chloride. In the Li_3PO_4 additional process, a linear signal for the change in plutonium content from the TRU-RE phosphate sediment was also obtained. Therefore, detection of plutonium in precipitates and chloride forms within the process was possible. However, the simulation results show limitations in measuring the uranium signal. Therefore, there is a restriction to uranium monitoring of oxide and chloride forms in all salt reactions.

Neutron measurement was also investigated for process

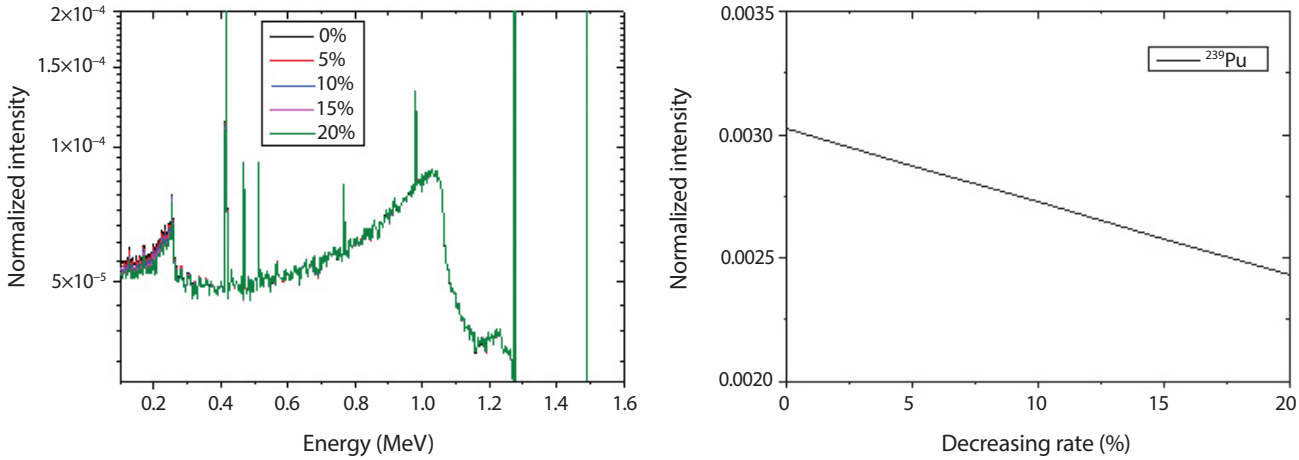


Fig. 6. Measurement by plutonium content change (~20wt% change).

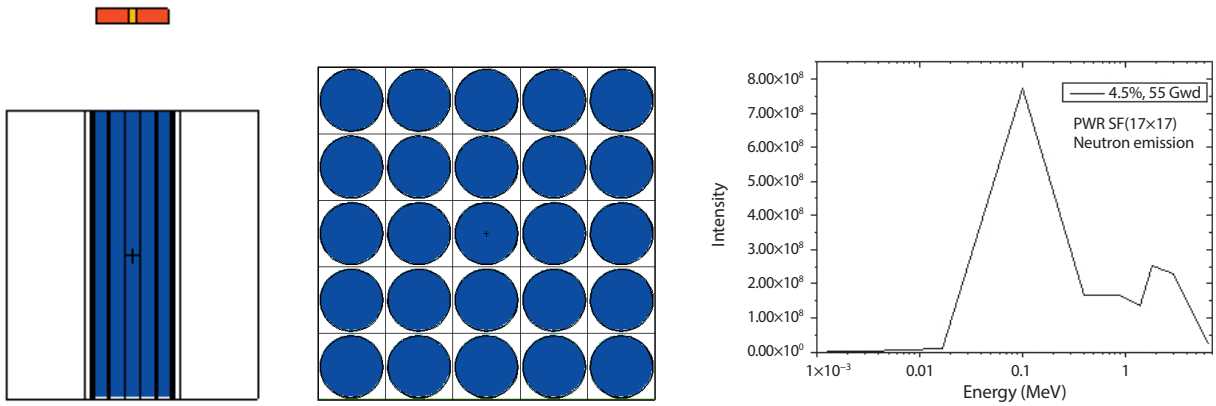


Fig. 7. Geometry of fuel block and source neutron spectrum.

monitoring. The relationship between neutron signals and content change of nuclear material was analyzed as well. In order to reduce the detection contribution from scattering of incident neutrons, an energy of 0.1 MeV or above was used when a shielding material was present. The decrease energy spectrum due to the decrease in nuclear material content was obtained in each process and the total sum of measurement signals shows a linear decrease in content change, at Fig. 5. Therefore, it represents that process monitoring can be applied by neutron measurement.

3.2.3 At fuel block

The TRU-RE phosphate block is fabricated to dispose in final storage site. To evaluate the radiation monitoring on the fuel block, gamma and neutron measurement was simulated. To detect changes of the plutonium content in the fuel block, the plutonium was measured by changing it up to 20%. In the spectrum analysis, the peak of plutonium existed in the change and the linearity of the plutonium signal according to content reduction was satisfied, as shown in Fig. 6. However, since the solidified body has a high density and is composed of TRU and rare-earth materials, there is a limit to the measurement of plutonium gamma rays beyond a certain depth (~7 cm) inside. Therefore, the accuracy of measurement can be improved only when the homogeneous condition of the solidified body is satisfied.

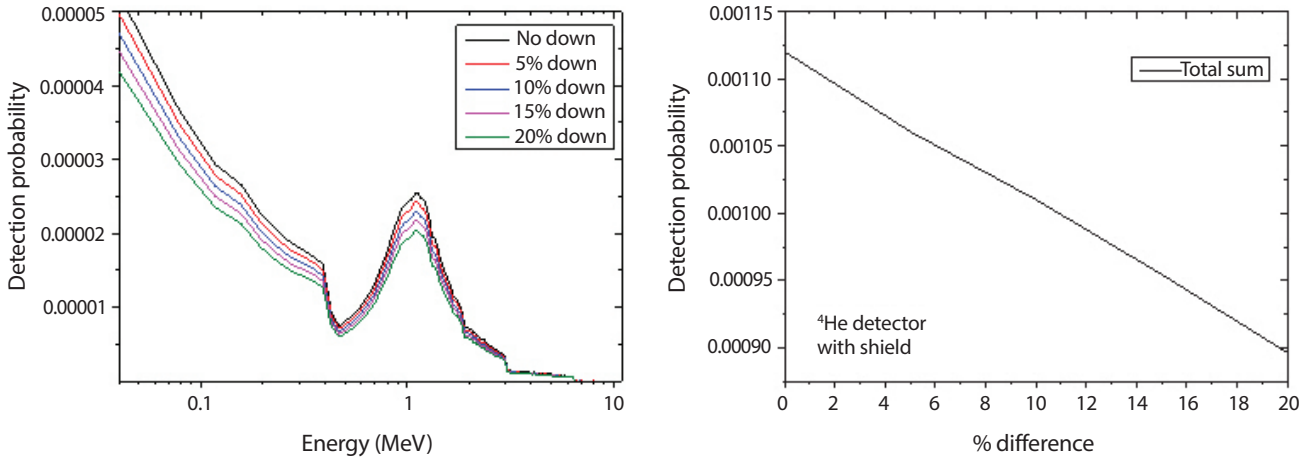


Fig. 8. Neutron measurement at fuel block (5, 10, 15, 20% change).

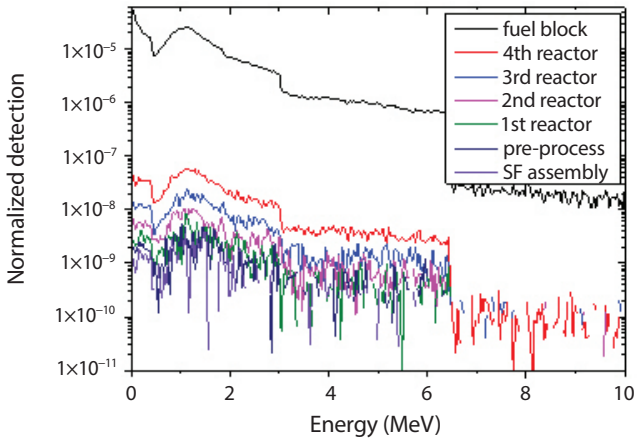


Fig. 9. Detection influence by adjacent neutrons at fuel block.

However, for the fuel block measurement, homogeneous condition was assumed.

Simulation of detecting nuclear material by measuring fast neutrons was also performed. Fig. 7 shows the geometry of fuel block assembly, detector location and source neutron energy spectrum for simulation. The uniform source distribution was assumed for all fuel block.

Fig. 8 shows the result on energy spectrum change and linearity of plutonium content in the detection. The measurement energy spectrum shows decrease according to the reduction of the source intense, and the sum of the measurement signals for all energies also shows a linear decrease with the change in content decrease. Therefore, it shows that detection of nuclear materials from neutron measurements will be also possible when a change in nuclear material content occurs, up to 20%. From the simulation results, the combination of gamma and neutron detection is recommended and very effective to monitor the plutonium in the product. In addition, it is suggested that it would be preferable to transfer nuclear materials to the next step after the current process is completed and to introduce new spent fuel rods after one campaign is done for appropriate monitoring of nuclear materials within the process.

In the process, various radiations exist from the spent fuel. Therefore, to improve the accuracy of nuclear material detection, the influence by adjacent radiation sources should be analyzed. At measurement of fuel block, the neutron influence from spent fuel assembly, head-end process, and several chemical reactors was evaluated, because all internal processes involve TRU-RE and several fission products. In the case of gamma ray measurements, lead block remove all interference, and in the case of neutrons, direct measurement from the fuel block show more than 100 times contribution than that of the surrounding influence. Fig. 9 shows the neutron detection contribution by surrounding sources. Therefore, when measuring direct fast neutrons in the high-energy region, the role of a shielding

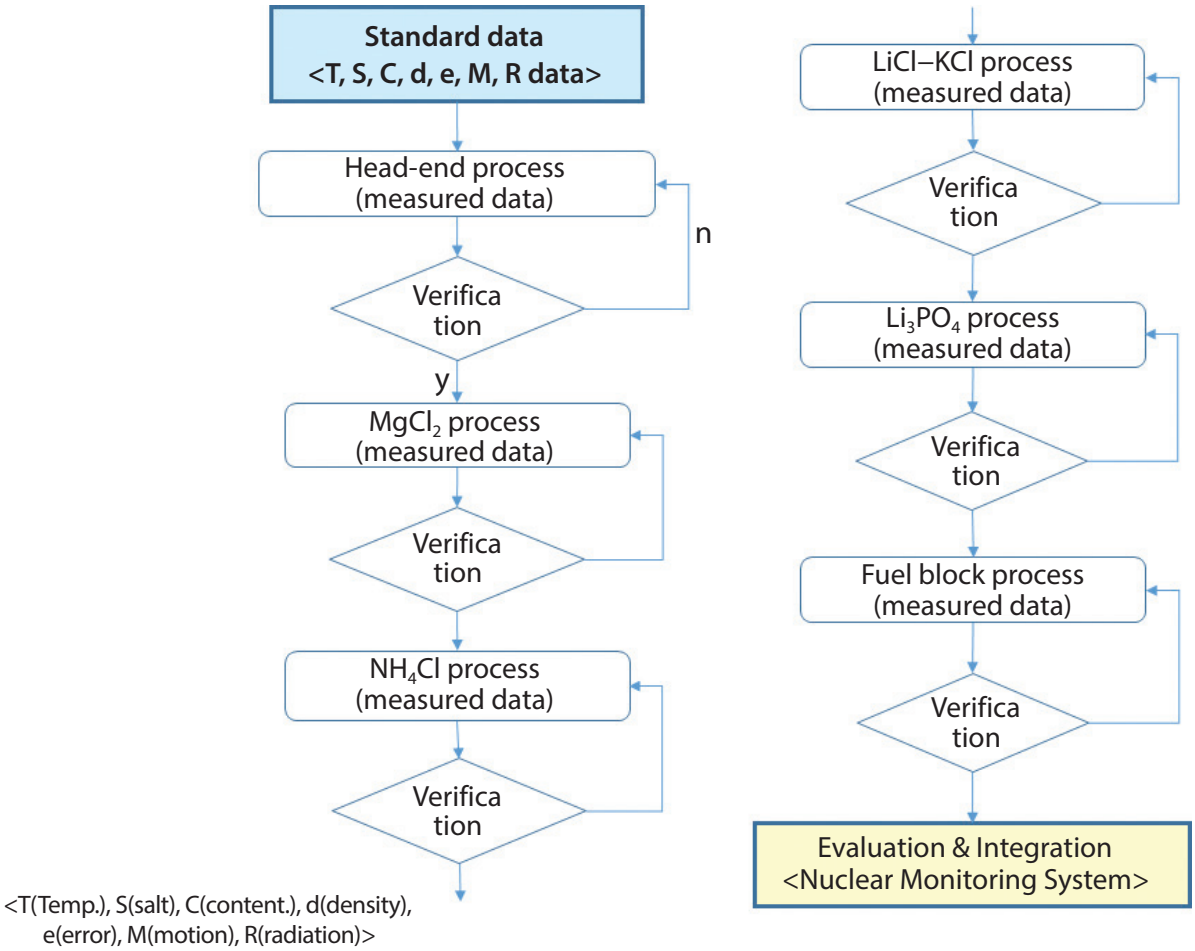


Fig. 10. Logic diagram of algorithm for monitoring system.

material to reduce the energy of scattered neutrons is important to increase detection accuracy.

3.3 Conceptual Design of Radiation Monitoring

The conceptual design for monitoring nuclear material in the process was developed using measured data including radiation detection. As the TRU-RE material is conserved in the process, the characteristic radiation measurement could be applied to verify nuclear materials effectively and efficiently. For obtaining standard signals for normal condition, the characteristics of each internal process are analyzed. The conventional methodologies are determined to clarify the reaction in the process as well. The radiation measurement is additionally performed. An anomaly operation is traced by the measured data. Fig. 10 represents the schematic diagram for monitoring system. Logic concept for algorithm development is summarized below;

1) Determine the input nuclear material content into the process and establish detectors for monitoring
2) Obtain operating parameters for standard process
3) In the head-end process, measure temperature, density, chemical form, content, error data, and radiation, check process operation by Cs and perform radiation monitoring
4) In the salt and fuel block process, verify the measured

data through comparative analysis with standard data for nuclear material

5) Complete the evaluation of nuclear material monitoring for the process through integration of all measurement data.

4. Results and Conclusion

The process is under development to minimize the environmental burden of spent fuel storage. The process is to increase the efficiency of the disposal site by separating and collecting uranium and TRU-RE oxide and disposing of them independently. Additionally, by collecting highly heat-generating and mobile nuclides during the process, the area of the disposal site will be dramatically reduced. Regarding the development of the process, the applicability and feasibility were verified on the nuclear monitoring through real-time and effective radiation measurements.

The condition for abnormal state was analyzed by changing the content of nuclear materials, up to 20%, and the linearity of the measurement signal was verified when an abnormal process occurred. From several simulations, it was possible to detect plutonium in the change of nuclear material content in sediments, chloride, and oxide form. It was also expected to be possible to determine whether there was an abnormality in the process through the radiation detection. However, since there are limitations in measuring uranium in all salt processes, it will be difficult to derive a uranium content from the measurement. Therefore, measurement through an active method using an external source is proposed for uranium assay. For the fuel block, TRU-RE phosphate form, the linearity of the measurement signal was satisfied according to the change in content.

From simulations, it was evaluated that radiation monitoring of the process would be more effective through the combination of neutrons and specific gamma ray measurement. In addition, it is believed that the development of an integrated system with existing technologies and radiation measurement will be very helpful in completing process monitoring of nuclear material.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

Acknowledgements

This work was supported by the Nuclear Research Foundation of Korea (NRF) grant funded by the Korea government (MSIP) (NRF-2021M2E3A3040093).

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