Transactions of the Korean Nuclear Society Autumn Meeting
Changwon, Korea, October 30-31, 2025

Operation Estimation Based on Monitoring Signals from Nuclide Management Process

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Keywords : Safeguards, Monitoring, Nuclide management process, Abnormal operation

1. Introduction

The nuclide management process is under development and operated based on the chemical reactions. This process has been developed to reduce the disposal area by selectively separation of the fission products with long-lived, high-heat generating, and high-mobility nuclides from spent fuel [1,2]. To ensure nuclear non-proliferation and implement effective safeguards, it is essential to integrate safeguards considerations from the conceptual design phase of novel technologies.

In this study, the behavior of nuclear material and associated signal variations under both normal operation and potential diversion scenarios were analyzed for each unit process within the nuclide management process, reflecting the current status of process technology development and anticipated operating conditions. The analysis was conducted using HSC Chemistry 10 software. Furthermore, the potential of signal-based monitoring technologies to detect quantitative deviations from normal operation was evaluated.

Reflecting the current status of process technology development, this study was conducted by considering 2 nuclide management process options. The major different unit processes between the 2 options are as following:

- Option 1: Sr (1st) chlorination and TRU/RE (2nd) chlorination using salt reaction (separate processes)
- Option 2: Sr chlorination and TRU/RE chlorination using NH_4Cl gas-solid reaction (using same vessel)

2. Analysis of nuclide behaviors for major processes

2.1 1st chlorination process using salt reaction

Under normal operating conditions, an analysis was conducted on the behavior of nuclear materials during the Sr chlorination process using $MgCl_2$ as the reagent, assuming an operating temperature of 750 °C. Under the normal conditions, Pu and U remain in the form of PuO_2 and UO_2 without reacting with $MgCl_2$ after the operation, while Sr and Ba are chlorinated to form $SrCl_2$ and $BaCl_2$, respectively.

In the 1st chlorination process, a key operating condition that can affect the abnormal behavior of Pu is the use of reagents such as UCl_3 or $ZrCl_4$-typically used in the 2nd chlorination process-instead of $MgCl_2$. Under abnormal operating conditions, the amount of $Cl_2(g)$ generated increases compared to that under normal conditions. Under abnormal conditions, the relationship between the amount of $Cl_2(g)$ generated and the reduction of PuO_2 is shown in Fig.1. This correlation enables the estimation of changes in Pu content based on the measured amount of $Cl_2(g)$.

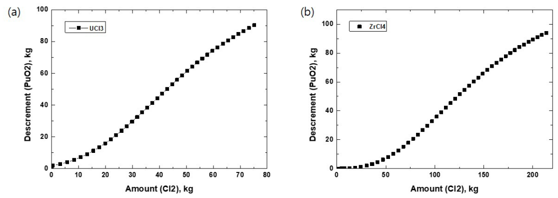

Fig. 1. Reaction behavior under the 1st chlorination process at 750 °C (a) correlation curve between $Cl_2(g)$ generation and PuO_2 reduction using UCl_3 as the reagent, (b) correlation curve between $Cl_2(g)$ generation and PuO_2 reduction using $ZrCl_4$ as the reagent

2.2 2nd chlorination process using salt reaction

In the 2nd chlorination process based on salt reactions, the use of UCl_3 and $ZrCl_4$ as reagents is being considered, with an operating temperature of 500 °C. Under normal operating conditions with a sufficient supply of reagents, PuO_2 is chlorinated to form $PuCl_3$. A potential outcome of abnormal operation in the 2nd chlorination process is that PuO_2 remains undissolved and residues along with uranium oxides, resulting in its inclusion within uranium block. Unlike the 1st chlorination process based on salt reactions, the behavior of Pu during the 2nd chlorination reaction is significantly influenced by temperature. The Fig.2 illustrates the phase transition of Pu as a function of temperature during reactions with 300 kg of UCl_3 and $ZrCl_4$, respectively. As temperature increases, the chlorination reaction of PuO_2 becomes less favorable. Thus, the operating temperature can be utilized as a key monitoring parameter in the 2nd chlorination process.

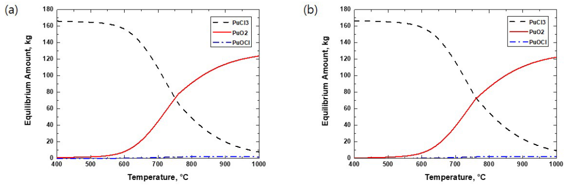

Fig. 2. Temperature-dependent behavior of Pu in the 2nd chlorination reaction (a) 300 kg UCl_3, (b) 300 kg $ZrCl_4$

2.3 Chlorination process using gas-solid reaction

The chlorination based on a gas-solid reaction is applied in Option 2. NH_4Cl is being considered as the reagent, and both 1st and 2nd chlorination reactions are designed to occur simultaneously within a single vessel. In the case of fission products, a stable behavior was observed beyond a certain threshold of NH_4Cl quantity and temperature. In contrast, the behavior of Pu was found to be highly sensitive to both the amount of NH_4Cl and the operating temperature. An operating temperature range of 380 ºC to 480 ºC is being considered as the normal operation condition. Accordingly, the analysis was conducted at 380 ºC, where the proportion of Pu existing as $PuCl_3$ is at its maximum within the considered temperature range. The target nuclides for 1st chlorination, fission products, undergo a phase change from oxides to chlorides when reacted with a sufficient amount of NH_4Cl at 380 ºC. Under conditions with a sufficient amount of NH_4Cl, the amount of $PuCl_3$ formed decreases as the temperature increases relative to the normal operating condition. Accordingly, in abnormal conditions involving temperature variations, temperature monitoring can be used to estimate changes in Pu behavior by exploiting the correlation between temperature and the decrease in $PuCl_3$.

3. Conclusions

This study investigated the behavior of nuclear materials and the corresponding signal changes during both normal and abnormal operations in key unit processes of nuclide management process. In particular, it evaluated the feasibility of signal-based monitoring technologies to quantitatively identify deviations from standard operational signals. Two options for the nuclide management process are currently under development, among which 3 key unit processes are identified as particularly significant from the perspective of safeguards monitoring. In the 1st chlorination process based on the salt reaction, the behavior of Pu is influenced by the type of reagent used, leading to the generation of $Cl_2(g)$, which differs from that observed normal conditions. In the 2nd chlorination process using salt reaction, the behavior of Pu is affected by temperature. Similarly, in the chlorination using gas-solid reaction, the amount of PuO_2 converted to $PuCl_3$ is dependent on temperature. These all clearly show the correlation between Pu changes relative to normal condition and signal.

Acknowledgements

This work was partly supported by a National Research Foundation of Korea (NRF) grant funded by the Korean government (MSIP) (NRF-2021 M2E3A3040093)

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