Proceedings of the Korean Nuclear Society Spring Meeting, Kwangju, Korea, May 2002

Proliferation Resistance of the Lithium Reduction Process

W.I.Ko, J.H.Ha, S.Y.Lee, D.Y.Song, H.D.Kim and S.W.Park
Korea Atomic Energy Research Institute
P.O. Box 105, Yusong, Taejon, Korea

Abstract

This paper addresses the characteristics of proliferation resistance of the lithium reduction process and international domestic safeguarding methods. In addition to dealing with qualitative features of the proliferation resistance, this study is emphasizing on the quantitative analysis of radiation barrier, which could be a significant accessibility barrier if the field is high enough to force a theft to shield the object during a theft. From the radiation barrier analysis, it is indicated that whole-body radiation dose is about 20 rem/hr at one meter of smelt and ingot metal of 40 kgHM, which could be considered to be a significant reduction in risk of theft. For safeguarding of this process, we propose a NDA concept for nuclear material accounting which is to measure the amount of curium in the reduction metal and associated process samples using a neutron coincidence counter and then to convert the curium mass into special nuclear material with predetermined curium ratios. For this, a well-type neutron coincidence counter with substantial shielding to protect the system from high gamma radiation is conceptually designed.

I. Introduction

With the volume reduction perspective of the spent fuels to be stored and/or disposed of, a lithium reduction process has been developed in Korea [1]. The safeguards systems, however, have not been developed and implemented, so it is not known how easily international safeguards could be implemented. Generally, it is known that the dry process like the lithium process is more proliferation resistance than the current plutonium-uranium extraction process (PUREX) because the lithium process has low fission product decontamination, and plutonium is also inherently commingled with minor actinides. Those features increase apparently inherent proliferation resistance but it may make nuclear material control and accounting (MC&A) more difficult, which may be one of the safeguardability concerns of the process. In this respect, this paper addresses international and domestic safeguarding methods of the lithium reduction process and its characteristics of proliferation resistance.

With respect to the MC&A, there are well established-analytical methods like isotope dilution spectrometry or inductively coupled plasma mass spectrometry. However, those methods need sample preparation process and is necessary to remove the matrix and to obtain pure fractions in order to avoid isobaric interference. It is relatively labor intense and requires a careful study of the chemical recoveries of the element of interest. In order to overcome the difficulty of the MC&A, a non-destructive analysis (NDA) technique is proposed here. As a result, a well-type neutron coincidence counter with substantial shielding to protect the system from high gamma radiation is conceptually designed. The measurement efficiency of the concept is calculated by use of MCNP (Monte Carlo N-Particle Transport) Code [2]. In addition, this study adds the quantitative analysis of radiation barrier, which could be a significant accessibility barrier if the field is high enough to force a theft to shield the object during a theft.

II. Proliferation Resistance Features of the Lithium Reduction Process

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 LiO_2. The key process of this system is a metallization process which 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 LiO_2 to form Li and O. Those processes have the following inherent features of proliferation resistance.

- Due to the nature lithium process, no fissile material can be separated in pure form. Plutonium, for example, is co-deposited together with miner actinides and some fission products such as lanthanides. So the material requires further chemical reprocessing. 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 metallized spent fuel ingot are about 25% of the initial spent fuel. The presence of some fission products lead to a high dose rate arising from the material.
- The lithium reduction process has to be done in a heavily shielded hot cell due to its highly radioactive materials. 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 [3]. The PIPEX approach to reducing 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 diversion.

- The lithium process operate on a batch mode, which allows fundamentally easier material accounting than do continuous flow systems such as 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.

Even though the lithium process has those features of the proliferation resistance, some are concerned about that the electrometallogical process could be modified to produce separated plutonium[4].

III. **Radiation Field Analysis**

The immediate health consequence of high doses of whole body radiation represents a significant barrier to theft of highly radioactive materials. Operations involving such materials require heavy shielding and remote handling equipment. The shielding material, being heavy and cumbersome, and/or remote handling would force the thief to use lifting equipment during the thief and to haul away a significantly lager mass than just the stolen object.

In order to see the performance of radiation barrier in reduction metal of 40 kg, we have calculated the radiation dose at 1 m from the surface of the metal ingot at the mid-plane. For this, photon spectrums obtained from ORIGEN code and gamma shielding code, Microshield[5], are used.

The table 1 shows the radiation doses at 1 m from the surface of the materials at the mid-plane. The third column of the table means the dose rate for obtaining 1 SQ Pu at a time. It is indicated from the table that metal ingots for obtaining 1 SQ Pu at a time have good radiation barriers for theft. The metal ingot of 40 kg batch with dose rate of 11~20 rem/hr has a moderate radiation barrier for theft.

Table 1 Radiation Doses at 1 m from the Surface of the Materials at the Mid-plane

Original Fuel	Dose rate (rem/hr) per metal ingot (20kg batch)	Dose rate (rem/hr) per metal ingot (40kg batch)	Dose rate (rem/hr) for obtaining 1 SQ
43,000 MWD/MTU, 10 years cooling (contains 1.07wt%Pu))	9.8	19.6	366
33,000 MWD/MTU, 10 years cooling (contains 0.91wt%Pu)	5.6	11.2	246

It is important to note how much radiation fields is enough to force a thief to shield the spent fuel during a theft. The IAEA considers all materials above 100 rem/hr at one meter to be highly radioactive and self-protecting [6]. As described in figure 1, DOE considers whole body doses above 15rem/hour at one meter to cause a significant reduction in risk of theft and 100 rem/hour at one meter to essentially rule out theft as a principle risk consideration [7].

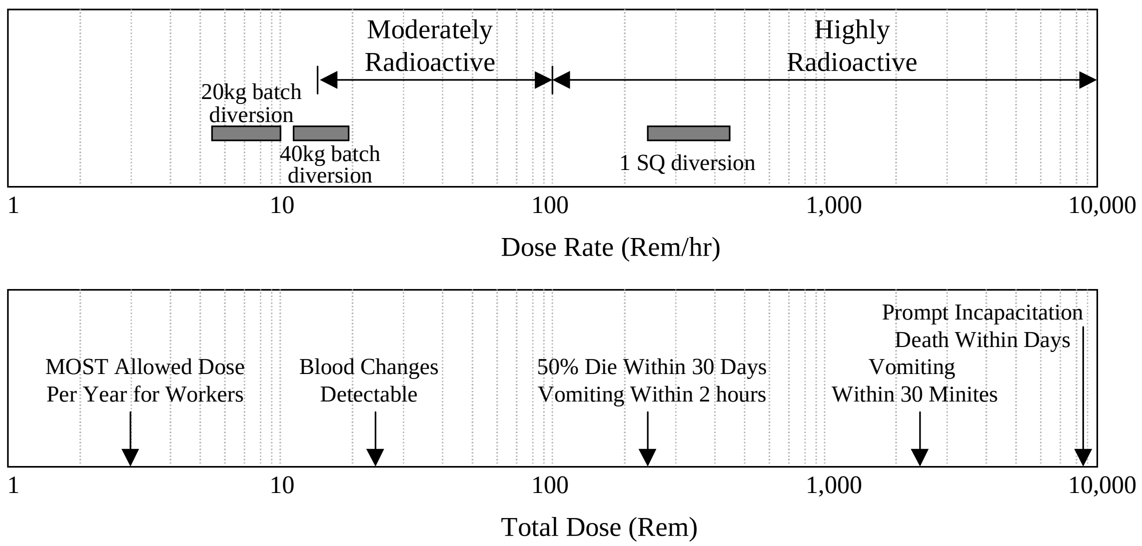

Figure 1 Whole Body Doses and Its Effect

It is indicated that whole-body radiation dose is about 20 rem/hr at one meter of smelt and ingot metal of 40 kgHM, which could be considered to be a significant reduction in risk of theft defined by DOE.

IV. Proposed Non-Destructive Assay Method

A well-type neutron coincidence counter was conceptually designed as shown in the figure below. The concept is adopting a curium-based safeguards approach, which has been used in DFDF (DUPIC Fuel Demonstration Facility) in Korea [8]. The Cm244 becomes the dominant source of neutron when burnup is higher than ~25 GWd/MTU and cooling time is over ~3 years. The proposed NDA concept for nuclear material accounting is to measure the amount of curium in the process material using a neutron coincidence counter and then to convert the curium mass into special nuclear material with predetermined curium ratios.

As shown in figure 2, substantial shielding was added to protect the He3 tube/electronics from the intense gamma ray of the process material. A cylindrical tungsten shield of ~4 cm in thickness was arranged around the central cavity. A total of 24 3He tubes were located symmetrically in the high density polyethylene moderator. The MCNP code had been used to optimally design and about 20 % of measurement efficiency could be obtained.

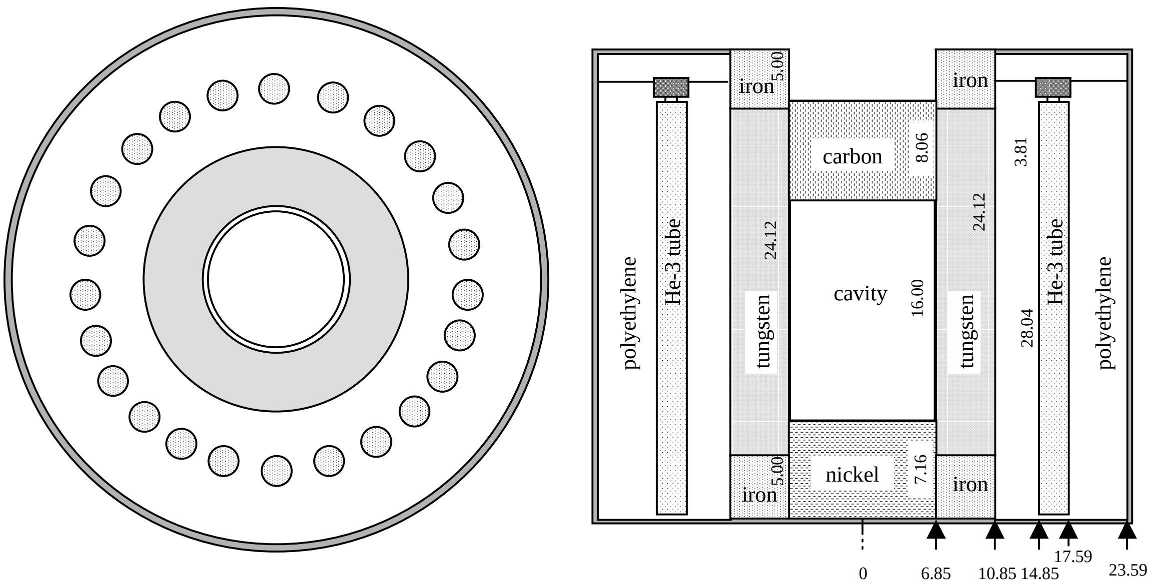

Figure 2 Concept of NDA System for Lithium Reduction Process

V. Conclusion

The lithium process has various inherent features of proliferation resistance. From the radiation barrier analysis, it is indicated that whole-body radiation dose is about 20 rem/hr at one meter of smelt and ingot metal of 40 kgHM, which could be considered to be a significant reduction in risk of theft. It concludes tentatively that the lithium reduction process not only has inherent features of proliferation resistance but also could be safeguardable.

References

1. J.S.Seo et. al., "Ion Exchange Properties of Zeolite in LiCl Moten Salt", Proceedings of the Korean Nuclear Society Spring Meeting, Korea, May 2000.
2. J.F.Briesmester, "MCNP - A General Monte Carlo N-Particle Transport Code", LA-13709-M, Los Alamos Laboratory (2000).
3. IAEA, "INFCE Working Group 4 Report", INFCE/PC/2/4, International Atomic Energy Agency, (1980).
4. NAS National Research Council, "An Assessment of Continued R&D into an Electrometallogical Approach for Treating DOE Spent Nuclear Fuel", National Academy of Science (1995)
5. "Microshield Version 3 Manual", Grove Engineering, Inc., Rockville, MD (1987).
6. IAEA, The Physical Protection of Nuclear Material. INFCIRC/Rev.3 (1993)
7. US DOE, Guide for the Implementation of DOE Order 5633.3b, Control and Accountability of Nuclear Materials.
8. Y.G.Lee et. al., "Development of DUPIC Safeguards Neutron Counter", Proceedings of 38th INMM Meeting (1997).