Prospects and Status of Chemical Separation of Radionuclides from Decommissioning Radioactive Waste in Korea

Tae-Hyeong Kim [a*], Jung Bo Yoo [a], Chang Je Park [b], Hong Joo Ahn [a], Kwang Yong Jee [a]
[a]*Nuclear Chemistry Research Division, Korea Atomic Energy Research Institute, 34057, 111 Daedeok-daero-989, Yuseong-gu, Daejeon, Republic of Korea*
[b]*Department of Nuclear Engineering, Sejong University,143-747, 209 Neungdong-ro Gwangjin-gu, Seoul, Republic of Korea*
[*]*Corresponding author: thkim@kaeri.re.kr*

1. Introduction

World widely, 448 nuclear power reactors are operating, 61 reactors are under construction, while 160 reactors are permanently shut down[1]. The average age of the nuclear reactors operating world-wide are close to 30 years[1]. For the case of those plants over 30 years, it can be subjected for decommissioning due to political and economic reasons. Therefore, simultaneous dismantling of plants are expected within 20 to 30 years. International Atomic Energy Agency (IAEA) estimates that global nuclear decommissioning process will create a significant market size over the course of the years ahead[2]. In June of 2017, permanent shut down of Kori Unit 1 was done in Korea for the first time, making decommissioning of nuclear power plants actualize. Because lifespan expectancy of 11 plants will come to an end by 2029, securing disassembly techniques is an important issue.

The Nuclear Safety and Security Commission (NSSC) notice states that radionuclides composed over 95% of the total radioactivity must be identified[3]. However, some of radionuclides emitting alpha-, beta-, and very low energy gamma radiation must be individually separated for radioactivity measurements. During decommissioning of nuclear power plants, great deal of various types of radioactive waste is created, however, it is unknown which radionuclides must be identified for disposal within the various types of the waste. In addition, use of chemical and purification methods depends on the target nuclides and types of the waste. Therefore, we examined the prospects and status on chemical separation of radionuclides from decommissioning radioactive waste.

2. Prospects and status on chemical separation of radionuclides

2.1. Current status on decommissioning of nuclear power plant

World widely, 160 nuclear power plants are shut down permanently, and this number includes those that are already decommissioned, are decommissioning, and will be in the future. Of those, only 19 units have been completely decommissioned while the United States is the only country with experience in decommissioning of commercial power plants. In particular, the United States have decommissioned the most power plants with many experience, including 15 power plants and of those, eight are commercial power plants. Korea has experience in decommissioning two research power plants (MARK II and MARK III) and one uranium conversion facility, however no decommissioning experience of commercial power plant or even large components has been done.

IAEA prospects the amount of dismantled radioactive waste when decommissioning one typical PWR plant (900 - 1300 MWe) as Table 1[4]. The number of drums were calculated based on the metal density to be 7.0 g/cm^3, concrete density to be 2.0 g/cm^3, dry active waste (DAW) density to be 0.8 g/cm^3, in addition to filling rate of 85 %.

Table 1. Typical radioactive material generated from decommissioning[4] (900-1300 MWe PWR)

Radioactive material	Weight (t)	Volume (m^3)	Drum
Activated steel	650	93	546
Activated concrete	300	150	882
Contaminated ferrite steel	2,400	343	2,017
Steel likely to be contaminated	1,100	157	924
Contaminated concrete	600	300	1,765
Contaminated lagging	150	21	126
Dismantling DAW	1,000	1,250	7,353
Total	6,200	2,314	13,613

However, the amount dismantled radioactive waste may vary within power plants. For the case of Maine Yankee plant in the United States, great amount of contaminated soil was found, which resulted in increase of the radioactive waste. Here, the radioactive waste was composed of 64% concrete, 23% soil, and 3% large components[5].

During decommissioning of nuclear power plants, massive amounts of radioactive waste are generated, which constitutes a significant portion of the cost of

decommissioning process. Ministry of Trade, Industry and Energy estimates that 41% of the decommissioning budget is responsible for processing with radioactive waste disposal.

2.2. Chemical separation of radionuclides

According to the notice of NSSC No. 2014-03, concentrations of radionuclides containing 95 % of total radioactivity and concentrations of the following radionuclides must be determined: ^{3}H, ^{14}C, ^{55}Fe, ^{58}Co, ^{60}Co, ^{59}Ni, ^{63}Ni, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{129}I, ^{137}Cs, ^{144}Ce, and gross alpha. Of these, ^{60}Co and ^{137}Cs in bulk samples can be easily determined by gamma spectrometry. However, alpha-, beta-, and very-low-energy gamma-emitting radionuclides, such as ^{55}Fe, $^{59/63}Ni$, ^{90}Sr, ^{94}Nb and ^{99}Tc must be individually separated for radioactivity measurements due to a high self-absorption of alpha particles in the sample matrix and a poor energy resolution of beta spectrometry. Thus, a separation of the difficult-to-measure (DTM) radionuclides is necessary to provide reliable analytical results.

Sequential separation and purification methods, including precipitation, anion exchange, and extraction chromatography, of alpha-, beta-, and very-low-energy gamma-emitting radionuclides such as ^{55}Fe, $^{59/63}Ni$, ^{90}Sr, ^{94}Nb and ^{99}Tc have been developed by Nuclear Chemistry Research Division from Korea Atomic Energy Research Institute (KAERI) and adopted for the evaluation of radionuclide inventory in the radioactive waste. The average recoveries of Tc, Sr, Fe, Nb, and Ni were 99.3 $\pm$ 0.3 %, 94.8 $\pm$ 0.6 %, 93.3 $\pm$ 1.2 %, 95.3 $\pm$ 1.9 %, and 92.6 $\pm$ 0.6 %, respectively[6].

However, when the relative ratio of Ca/Sr exceeds 100, the recovery tends to decrease when using the current method. Therefore, a new method to analyze samples with high calcium concentration, such as concrete, is being developed. With such method, higher recovery rate can be seen even when Ca/Sr relative ratio is over 500[7]. Thus, the modification of chemical separation and purification method is crucial for various types of radioactive wastes.

2.3 Prediction of radionuclide inventory

Activated or contaminated concrete is one of the major radioactive waste from decommissioning. In order to figure out the effect of impurities on activities in the concrete, the isotopic activities were calculated with ORGIEN-S code. We have assumed that the reactor was fully operated for 40 years and cooled for 10 years and the neutron flux to be 10^{10} neutrons/cm^{2}s. The composition of the concrete was same as the typical Portland concrete. Concentrations of cobalt and europium as impurity were assumed to be 100 part per million (ppm).

If the concrete does not contain any impurities, ^{39}Ar and ^{55}Fe make up 99.1% of the total radiation; however, if it contains impurities, such as cobalt and europium, ^{152}Eu, ^{154}Eu, and ^{60}Co make up 98.0% of the total radiation rather than the ones mentioned above. Because the small amount of impurities existing in the waste may count for most of the total radiation, identification and calculation of radionuclide in various composition and activation condition are necessary and therefore it is ongoing.

3. Conclusions

For final disposal of radioactive wastes generated during decommissioning of nuclear power plants, the radionuclides composing 95% of total radiation must be identified. Since DTM radionuclides must be individually separated for radioactivity measurements; therefore, a method to separate and purify radionuclides has been developed by KAERI and adopted to evaluate radionuclide inventory. Because separation recovery is effected by the composition of samples, modification of the chemical separation method is needed to analyze various types of radioactive waste generated from decommissioning. In addition, like concrete, dominant radionuclides may vary due to impurities, which may require a prediction model to identify the dominant radionuclides in various conditions. These developments would allow more accurate analysis of radioactivity in decommissioning radioactive waste, resulting in more safe, economical, and reliable disposal of the waste.

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