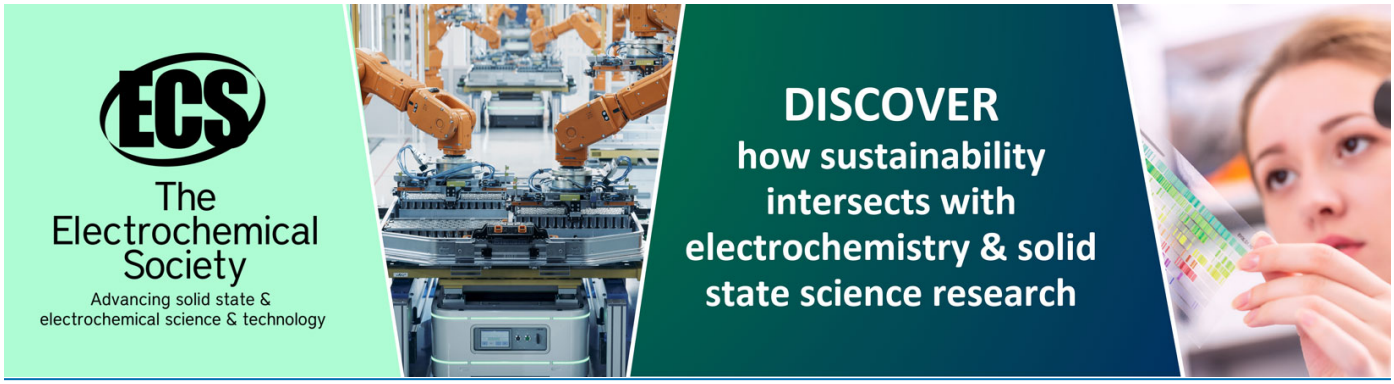
IOP Conference Series: Earth and Environmental Science

Radioactive Fission Waste from Molybdenum-99 Production and Proliferation Risks



Radioactive Fission Waste from Molybdenum-99 Production and Proliferation Risks

Aisyah, Pungky Ayu Artiani*, Jaka Rachmadetin

Center for Radioactive Waste Technology, National Nuclear Energy Agency, South Tangerang-Banten 15314, Indonesia

*email address: pungky@batan.go.id

Abstract. Molybdenum-99 (^{99}Mo) is a parent radioisotope of Technetium-99m (^{99m}Tc) widely used in nuclear diagnostics. The production of this radioisotope by PT. INUKI generated radioactive fission waste (RFW) that theoretically contains ^{239}Pu and ^{235}U, posing a nuclear proliferation risk. This paper discusses the determination of radionuclides inventory in the RFW and the proposed strategy for its management. The radionuclides inventory in the RFW was calculated using ORIGEN 2.1 code. The input parameters were obtained from one batch of ^{99}Mo production using high enriched uranium in PT. INUKI. The result showed that the RFW contained activation products, actinides, and fission products, including ^{239}Pu and ^{235}U. This result was then used for consideration of the management of the RFW. The concentration of ^{235}U was reduced by a down-blending method. The proposed strategy to further manage the down-blended RFW was converting it to U_3O_8 solid form, placed in a canister, and eventually stored in the interim storage for high-level waste located in The Radioactive Waste Technology Center.

1. Introduction

Radioisotopes are widely used for medical purposes, among which is ^{99m}Tc used in diagnostic imaging of many organs in the human body. This radioisotope accounts for approximately 80% of all nuclear medicine diagnostics, such as diagnosing brain disorder, lungs blood clots, bone cancer, coronary artery and thyroid disease, kidney, liver, and gallbladder function [1]-[3]. Due to its short half-life of 6 hours, ^{99m}Tc is produced from ^{99}Mo, its parent radionuclide. The half-life of ^{99}Mo is 66 hours, and about 88% of its decay is beta decay which produces metastable ^{99m}Tc [4], [5].

PT. Industri Nuklir Indonesia (PT. INUKI), located in Nuclear Serpong Area, South Tangerang, is the only ^{99}Mo producer in Indonesia. In the past, the state-owned company produced ^{99}Mo radioisotope using 92.7% of high enriched uranium (HEU) in the GA. Siwabessy Multipurpose Research Reactor [6]. The ^{99}Mo production used the CINTICHEM process, which involved dissolution, filtering, separation, purification, and some conditioning processes, as shown in Figure 1 [4], [7]. Several reagents were used, and different types of radioactive waste containing various radionuclides were generated, one of which is the radioactive liquid waste called Radioactive Fission Waste (RFW) [6]-[8]. However, the production of ^{99}Mo by this method has been ceased because the use of HEU targets in ^{99}Mo production generated RFW, which contained more fissile radionuclides, such as ^{239}Pu and ^{235}U, posing a risk of nuclear proliferation.

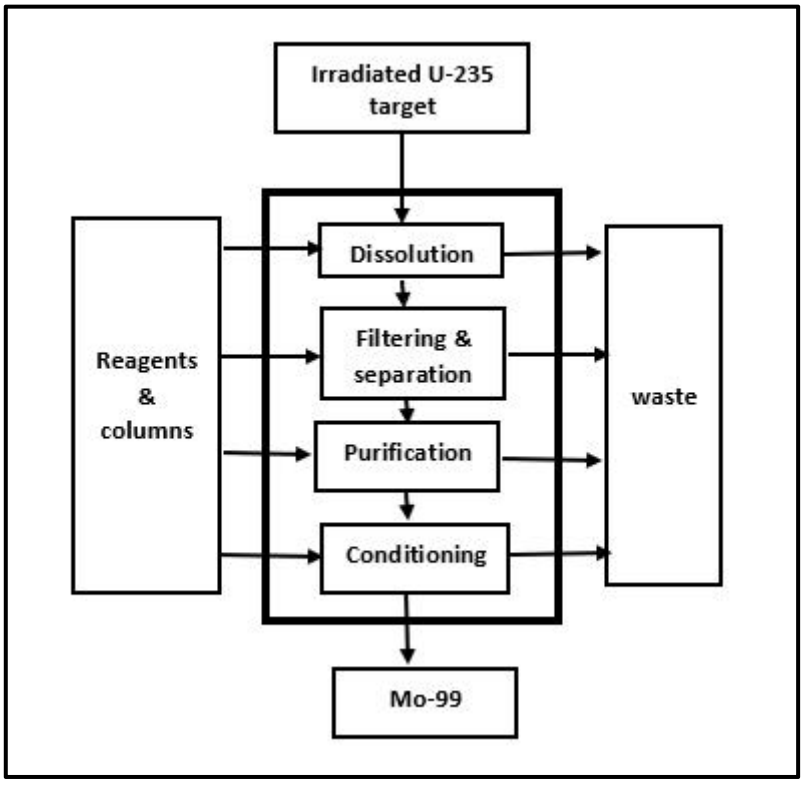


Figure 1. The CINTICHEM process scheme on the ^{99}Mo production.

The remaining concern regarding the ^{99}Mo production using HEU, as previously mentioned, is the management of the legacy RFW. Based on Government Regulation No. 61/2013 regarding Radioactive Waste Management, the RFW is classified as intermediate-level radioactive waste, which requires high-level safety in its management. Various methods for treatment of the RFW generated from ^{99}Mo production have been studied in some countries. The separation of U from ^{99}Mo liquid production waste has been studied using 15% Aliquat-336 in Toluene as an extractant solution [9]. Direct immobilization of the RFW has been studied using cement [10], glass, ceramics [11], and synroc [12], [13]. Among these immobilization methods, synroc developed by the Australian Nuclear Science and Technology Organization (*ANSTO*) has the highest volume reduction, up to 70%, compared to using cement, glass, or ceramics [12]. The higher volume reduction is beneficial because it will lead to a more efficient storage capacity. Another method that has been studied is precipitation and calcination of the liquid RFW to the U_3O_8 solid phase [14].

This study aims to determine radionuclides inventory in the RFW from the ^{99}Mo production in PT. INUKI and propose the strategy for the management of the RFW. The radionuclides inventory in the RFW was calculated using ORIGEN 2.1 code because laboratory characterization is difficult due to its high radioactivity level. The result from this calculation was then used to determine the strategy for the management of the RFW. The proliferation risk was evaluated based on the calculated ^{235}U and ^{239}Pu content in the RFW. The current status of the management of the legacy RFW was discussed. Finally, a strategy for the management of the RFW was proposed by reviewing the methods developed in other countries as mentioned above, considering their applicability in Indonesia.

2. Methodology

The radionuclides inventory in the RFW generated by ^{99}Mo production from HEU was calculated using ORIGEN 2.1 code with the following equation [15]-[17]:

$$\frac{dX_i}{dt} = \sum_{j=l}^{N} l_{ij}\lambda_l\chi_j - \emptyset\sum_{k=l}^{N} f_{ik}\sigma_k\chi_k - (\lambda_i + \emptyset\sigma_i + r_i)\chi_i + F_i, \qquad i = 1..., N \tag{1}$$

where x_i is the atomic density of nuclide i (atom/cm^3); N is the number of nuclides (atom); l_{ij} is the fraction of the radioactive disintegration by nuclide j, which resulted in the nuclide i formation (fractional); λ_i is the radioactive decay constant (sec^{-1}), ϕ is the space and energy-averaged neutron flux (n/cm^2 sec); f_{ik} is the fraction of the neutron absorbed by nuclide k, resulting in the formation of nuclide i (fractional); σ_k is the spectrum-averaged neutron absorption cross-section of nuclide k (barn); r_i is the continuous removal rate of nuclide i from the system (atom/sec), and F_i is the continuous feed rate of nuclide i (atom/sec).

The calculation using ORIGEN 2.1 code was based on one batch production data of ^{99}Mo from a target containing HEU, which was carried out in PT. INUKI. The HEU was electroplated in a cylindrical stainless steel AISI 304 welded at both ends. The target dimensions are 457.2, 28, and 27 mm for length, outside diameter, and inside diameter. The thermal neutron flux was set to 1.12x10^{14} n/cm^{2}s. This value was obtained from the irradiation history of the target at the Center Irradiation Position (CIP) at the position of D6 or E7 at GA Siwabessy Multipurpose Research Reactor. The calculation of the specific activity of radionuclides was carried out after the extraction process of the target up to 50 years of decay time. Hundreds of radionuclides were obtained from the calculation by ORIGEN 2.1 code. Those of which significantly contributed to the waste management were selected based on their half-life and clearance levels. The risk of proliferation was analyzed based on ^{239}Pu and the remaining ^{235}U in the RFW.

3. Results and Discussion

3.1. Actinides in the RFW and Proliferation Risk

Table 1 listed the selected radionuclides inventory in the RFW classified into three groups, i.e., actinides, activation, and fission products, together with their respective specific activities, obtained from the calculation using ORIGEN 2.1 code. As can be seen from the result, the fission products account for most of the total specific activity after 50 years of decay. The calculated specific activity of this group of radionuclides was 3.01×10^9 Bq/g, more than four orders of magnitude higher than those values of actinides and activation products. However, fission products are relatively short-lived compared to the radionuclides in the actinides group. For example, the half-life of ^{238}U and ^{235}U are 4.47×10^9 and 7.04×10^8 years, respectively. Therefore, in the long-term, the specific activity of the RFW will be predominantly by that of actinides.

Table 1. Inventory of selected radionuclides and their specific activities in the RFW obtained from the calculation using ORIGEN 2.1 code

Group	Radionuclides	Specific activity (Bq/g) for 50 years decay
Actinide	^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu	1.2x10^5
Activation product	^{3}H, ^{14}C, ^{55}Fe, ^{59}Ni, ^{60}Co, ^{63}Ni, ^{108M}Ag, ^{151}Sm ,^{152}Eu, ^{154}Eu, and ^{155}Eu	7.52x10^4
Fission product	^{85}Kr,^{90}Sr,^{99}Tc,^{106}Ru,^{25}Sb, ^{137}Cs, ^{147}Pm, ^{151}Sm, ^{155}Eu	3.01x10^9
Total		3.01x10^9

As shown in Table 1, the total specific activity of the actinides in the RFW from the calculation using ORIGEN 2.1 code was 1.2×10^5 Bq/g which contains seven radio-actinides, i.e., ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Pu. Figure 2 further shows the specific activities of radio-actinides up to 50 years of decay time. Figure 2 shows that the actinide in the RFW with the lowest specific activity after 50 years decay time was ^{241}Pu. Its half-life of 14.4 years is relatively low compared to the other radio-

actinides in the RFW. On the other hand, the actinide with the highest specific activity after 50 years of decay was ^{235}U. This is because only approximately 6% of the ^{235}U in the target undergo fission processes to produce ^{99}Mo, whereas the remaining 94% of which is discarded in the RFW. Furthermore, ^{235}U has a half-life of 7.038×10^{8} years, making it the predominant actinide in the RFW after 50 years of decay time.

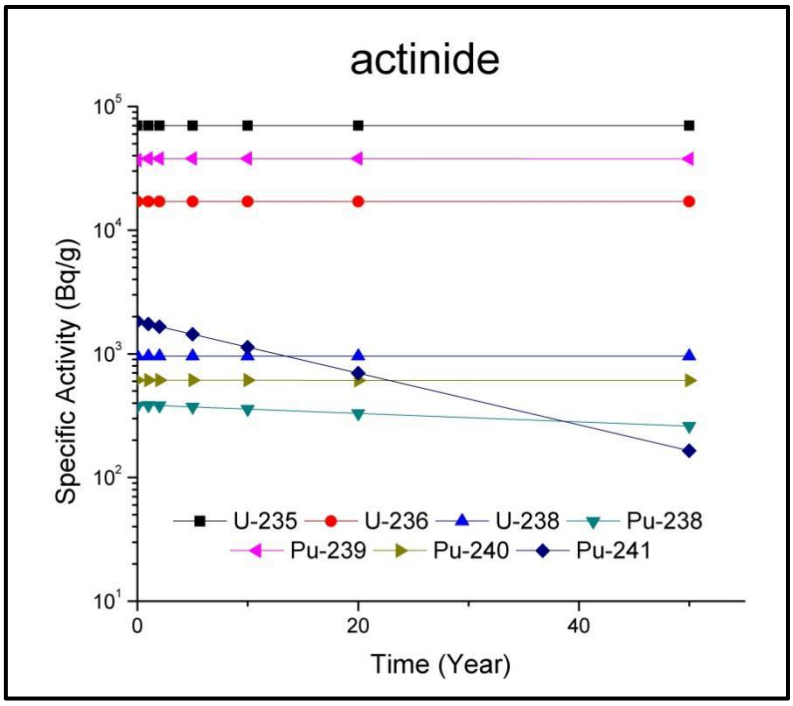


Figure 2. The specific activities of the actinides contained in the RFW.

The result in Figure 2 also shows that the second-highest actinide in the RFW was ^{239}Pu. This actinide was generated from the transmutation of ^{238}U when the ^{238}U in the target was irradiated in the nuclear reactor. A neutron was absorbed by ^{238}U and changed to ^{239}U. The ^{239}U then underwent β^- decay to produce ^{239}Np, which further undergone the same decay and transformed into ^{239}Pu. The transmutation of ^{238}U to produce ^{239}Pu is described in the following reaction [18]:

$$^{238}_{92}U + ^{1}_{0}n \rightarrow ^{239}_{92}U \xrightarrow[23.5\ min]{\beta^-} ^{239}_{93}Np \xrightarrow[2.356\ d]{\beta^-} ^{239}_{94}Pu$$

The resulting ^{239}Pu tends to absorb an additional neutron which further produces ^{240}Pu and ^{241}Pu. Therefore, the RFW contains all those three Pu radionuclides, as shown in the calculation result.

The presence of ^{239}Pu and ^{235}U in the RFW make tight security and safeguard necessary for the RFW management to comply with the non-proliferation treaty that Indonesia has ratified. The content of ^{239}Pu is one of the major concerns in the risk of nuclear proliferation because it can be used as a fuel for the thermal-spectrum reactor and nuclear weapon [19]. The probability for fission of ^{239}Pu is higher than that of ^{235}U. Furthermore, a larger number of neutrons produced per fission event make ^{239}Pu has a smaller critical mass due to this nuclear proliferation concern, PT. INUKI, under the partnership with International Atomic Energy Agency, carried out the conversion from HEU to low enriched uranium (LEU) in the ^{99}Mo. This uranium target conversion leads to the higher generation and different properties of the RFW. Therefore, further study is required for the management of the RFW from the LEU target.

3.2. Current status and proposed management of the RFW

In 2017, PT. INUKI received support from the U.S. Department of Energy (U.S. DOE) in the management of RFW. The mission was to support the knowledge and experience in RFW management and disposal, including technological and financial support. Under this program, the RFW was down-blended to convert its HEU content to LEU by mixing the RFW with natural uranium [20]. All of the legacy RFW in PT. INUKI has been down-blended to LEU and is stored in the hot cell until the present. This down-blended RFW waste should be removed from the hot-cell to ensure long-term safety and reduce the risk of nuclear proliferation.

Based on The Act No. 10/1997 on Nuclear Energy and The Government Regulation No. 61/2013 regarding Radioactive Waste Management, The Radioactive Waste Technology Center (RWTC) is responsible for managing all radioactive waste in Indonesia. Regarding the management of the RFW, cordial partnership between PT. INUKI and the RWTC are necessary. Consequently, the strategy proposed for the management of the RFW shall consider the resources and the available facilities in the two institutions.

Among the methods previously mentioned for the treatment of the RFW, conversion to U_3O_8 is considered the most feasible option considering the resources in both PT. INUKI and the RWTC. Other methods, such as radionuclides separation, cannot be applied in PT. INUKI or the RWTC due to restriction of HEU content in the RFW. This method is similar to that of spent fuel reprocessing in a closed fuel cycle. Likewise, direct immobilization using glass, ceramics, or synroc cannot be the option because such facilities are unavailable in the RWTC. The proposed method of calcination of the RFW to U_3O_8 is similar to that has been studied in the Argonne National Laboratory, as described in Figure 3. The down-blended liquid RFW is converted to a solid phase by precipitation followed by calcination to produce U_3O_8. The U_3O_8 is then packaged in a canister and stored in a storage facility.

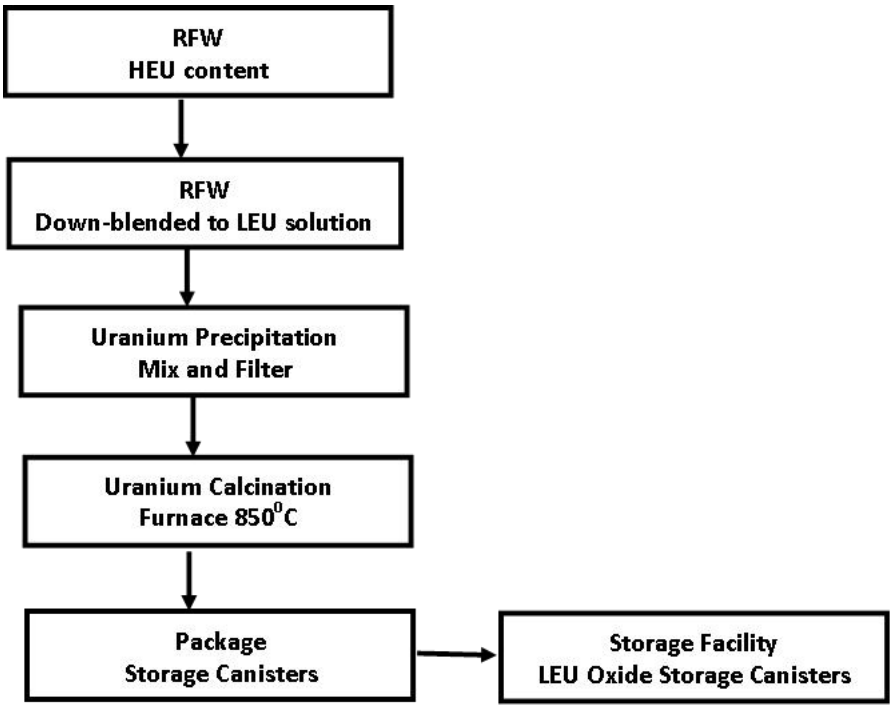


Figure 3. Conversion processes of the RFW solution to U_3O_8.

The canister used for the U_3O_8 package is made of stainless steel with a capacity of 60 liters, as shown in Figure 4. The U_3O_8, packaged in canisters, is stored inside storage pits in the high-level waste storage facility at the RWTC. This facility has 20 cylindrical and two cuboidal storage pits; all are 4.5 m deep below the surface, as shown in Figure 5. The cylindrical storage pits have diameters,

and wall thicknesses of 60 and 10 cm, respectively, with the capacity of each pit being seven canisters. This design is expected to provide safety for the storage of the U_3O_8 canisters in terms of high radiation protection. Safety from human intrusion is provided by the control of the facility implemented by the RWTC. More detailed safety aspects of this storage strategy will be assessed in further study.

Figure 4. Stainless steel canister for U_3O_8 package as the final waste form of the RFW from ^{99}Mo production.

Figure 5. High-level waste storage facility at the RWTC. The red color shapes are the top lid of storage pits. The yellow color is the concrete floor of the facility.

4. Conclusion

Characterization of radionuclides inventory of the RFW generated from ^{99}Mo production in PT. INUKI has been studied by calculation using ORIGEN 2.1 code. The result showed that the RFW contains activation products, actinides, and fission products, including ^{239}Pu and ^{235}U. Due to the content of ^{239}Pu and ^{235}U, the risk of nuclear proliferation was considered in the RFW management. The content of ^{235}U in the RFW has been reduced by the down-blending method. The down-blended RFW is stored in the hot cell at PT. INUKI until the present. By reviewing the available treatment methods, the calcination of the RFW to U_3O_8 solid phase was considered the most feasible option. After the calcination, the proposed strategy is to pack the U_3O_8 solid phase in a canister. Subsequently, the canister package is stored in a storage pit in the high-level waste storage facility at the RWTC.

5. Acknowledgment

This work was supported by government funding through DIPA of Radioactive Waste Technology Center BATAN, Budgetary Year 2021.

References

[1] Lee S. K., Lee S., Kang M., Woo K., Yang S. W., and Lee J. 2020 *Nucl. Eng. Technol.* **vol 52** pp 1517–1523
[2] Gumiela M. 2018 *Nucl. Med. Biol* **vol 58** pp 33–41
[3] Raposio R., Thorogood G., Czerwinski K., Rozenfeld A. 2019 *Appl. Radiat. Isot.* **vol 148** pp 225–231
[4] Lee S K., Beyer G. J., and Lee J. S. 2016 *Nucl. Eng. Technol.* **vol 48** pp 613–623
[5] Ohta M., Kwon S., Sato S., Ochiai K., Suzuki H. 2018 *Nucl. Mater. Energy* **vol 15** pp 261–266
[6] Wisnubroto D.S., Zamroni H., Sumarbagiono R., Nurliati G. 2021 *Nucl. Eng. Technol.* **vol 53** pp 549-561
[7] Aisyah, Artiani P. A., Purwanto Y. 2018 *Ganendra* **vol 99** pp 71–82
[8] Rao A., Sharma A. K., Kumar P., Charyulu M. M., Tomar B. S., Ramakumar K. L. 2014 *Appl. Radiat. Isot.* **vol 89** pp 186–191
[9] Mokhine N. D., Mathuthu M., Stassen E. 2020 *Phys. Chem. Earth Parts A/B/C* **vol 115** p 102822
[10] Shon J. S., Kim G. Y., Im J. 2019 *Ann. Nucl. Energy* **vol. 124** pp 342–348
[11] Stewart M. W. A. et al. 2013 *Sci. Technol. Nucl. Install.* **vol 2013**
[12] Cheung C. K. W. et al. 2012 *Procedia Chem.* **vol 7** pp 548–553
[13] Gregg D. J., Farzana R., Dayal P., Holmes R., and Triani G. 2020 *Synroc technology: Perspectives and current status (Review)* **vol. 103**
[14] Bakel A. J., Quigley K. J., and Vandegrift G. F. 2003 Argonne National Laboratory (ANL) progress in minimizing effects of LEU conversion on calcination of fission product ^{99}Mo acid waste solution *International RERTR Meeting* (Illinois)
[15] Croff A. G. 1983 *Nucl. Technol.* **vol 62** pp 335–352
[16] Croff A. G. 1980 *A User's Manual for the ORIGEN2 Computer Code* (Tenesse : Oak Ridge National Laboratory)
[17] Aisyah, Mirawaty, Saputra D.L.I., Setiawan R., Artiani P A, Ratiko, Nasruddin 2021 Sains Malaysiana **vol 2** pp 525-536
[18] Akyurek T., Tucker P.L., and Usman S. 2013 *Ann. Nucl. Energy* **vol 69** pp 278–291
[19] Yoo H., Seo J. H., Lee N. Y., Hyun J. L., Koh M. S., and Ahn S. H. 2017 *Prog. Nucl. Energy* **vol 100** pp 309–315
[20] Jagmin-Brookins J. and Pfennigwerth G. L. 2017 *Annual Waste Management Symposium* (Phoenix : WM Symposia)