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Original Article

# Monte Carlo simulations of criticality safety assessments of transuranic element storage in a pyroprocess facility

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ABSTRACT

In this study, criticality safety assessments of the potential for storing transuranic element (TRU) ingots via a pyroprocess were evaluated to determine the appropriate TRU storage design parameters, in this case the ratio of the TRU ingot height to the radius and the number of TRU ingot canisters stacked within a container. Various accident situations were modeled over a modeling period of 5 years for a cumulative inventory of TRU ingots with various water densities in submerged containers and with various pitches between the containers in the facility. Under these combinations, we calculated the threshold of TRU height and radius ratio depending on the number of canisters in a container to keep the stored TRU in a subcritical state. The ratio of the TRU ingot height to radius should not exceed 4.5, 1.1, 0.5, 0.3, and 0.2 for two, three, four, five, and six levels of stacked canisters in a container, respectively.

## 1. Introduction

Nuclear power plants generate spent fuels (SFs) as ineluctable byproducts. SFs are considered types of waste and a long-lasting potential hazard to the public. Therefore, a number of countries want to bury these materials underground permanently without any reprocessing. However, SFs still possess valuable nuclear materials such as $^{238}$U and the group of transuranic elements (TRUs), which can be recycled for use in a sodium-cooled fast reactor. Therefore, the reprocessing of SFs to extract them could be economically feasible in the future. However, the reprocessing of these materials introduces a risk when extracting $^{239}$Pu separately from SFs as $^{239}$Pu can be used to create atomic bombs. Recently, the idea of pyroprocessing was suggested to reprocess SFs to extract TRU together with Pu in a combined state to ensure that the separation of Pu would be fundamentally impossible. In other words, pyroprocessing must satisfy the requirements of international nuclear nonproliferation treaties. Secondary advantages of pyroprocessing are that it can reduce volume and radiotoxicity of high-level wastes and shorten the management period of the waste repository.

The products of pyroprocessing are metal fuels of the type used in sodium-cooled fast reactors. Because it is not possible to separate TRU with high fission cross sections, such as Pu, from TRU materials, pyroprocessing is regarded as a secure and peaceful means of SF recycling. In relation to this, the Korea Atomic Energy Research Institute proposed a conceptual pyroprocess facility, termed the Reference Engineering-scale Pyroprocess Facility, where 10 tons of heavy metal (tHM) SFs can be processed per year [1]. The Korea Atomic Energy Research Institute is currently revising the Reference Engineering-scale Pyroprocess Facility with the goal of realizing a facility on a larger scale capable of treating 30 tHM per year.

From a safety perspective, the criticality of TRU ingots for storage in a pyroprocessing facility is one of the major issues that must be evaluated. Although many issues related to the safeguarding of the hot cells used in pyroprocessing have been discussed, the criticality safety issue in relation to TRU storage has not yet been investigated [2–5]. On the other hand, there have been several studies regarding criticality safety with burnup credits for spent nuclear fuels [6–9]. Nonetheless, there have been no in-depth studies on the safety of SFs in a reformed state, such as TRU ingots generated from pyroprocessing [10].

TRU storage involves filled containers arranged at a certain pitch, as shown in Fig. 1. Under normal operating conditions, the

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inside of the storage is filled with argon. For the simulations, the outer edges of the storage are assumed to be vacuum. A container can be packed with several canisters of TRU ingots, which are stacked upward. Fig. 1B shows an example of one such container that is composed of three levels of stacked canisters. The criticality of a TRU container depends not only on the burnup credit but also on the geometry, i.e., the height-to-the radius ratio of the TRU ingot (TRU H/R ratio) and the number of stacked canisters in a container.

The aim of this study is to find the ideal TRU H/R ratios and the number of stacked canisters such that TRU storage does not reach a supercritical state under any extreme accidental conditions. By doing so, it will become possible to estimate the effective multiplication factor ($k_{eff}$) of the storage facility using Monte Carlo N-Particle transport code, version 6–1.0 (MCNP6). Details pertaining to configurations and methods of the simulations are described in the following sections.

## 2. Materials and methods

### 2.1. Description of TRU ingot

A TRU ingot is the metal-form product generated from the SF assemblies used in the pyroprocess. A 16 × 16 PLUS7 fuel assembly serves as the basis for the fuel unit model in this work. The initial composition of a discharged pressurized water reactors (PWR) assembly with a $^{235}$U enrichment rate of 4.5 w/o, burnup of 55 GWd/MTU, and cooling time of 10 years was applied to the ORIGEN-ARP code [11]. Unit process yields in the pyroprocess were taken into account when determining the composition of the radionuclides of the TRU ingot. The primary radionuclides are shown in Table 1. The TRU ingot is cylindrical in shape. The mass of the TRU ingot was assumed to be 4.0 kg, and the density was 19.82 g/cm$^3$. The total production of TRU ingots per year is 150 pieces based on an annual throughput of 30 tHM.

### 2.2. Description of TRU ingot canister and container

The TRU ingot was assumed to be placed within a stainless steel (STS304) canister, and the canisters were stacked vertically inside a stainless steel (STS304) container, as shown in Fig. 1B. In our calculations, it was assumed that the number of stacked canisters could vary from two to six. The number of containers in the TRU storage area could vary accordingly; the range of the array diameter was from 1.9 to 2.9 m. The array of containers was assumed to have a hexagonal lattice pattern, as shown in Fig. 1A.

**Table 1**
Primary radionuclides of the TRU ingot obtained by using the ORIGEN-ARP code and applying unit process yields.

| Radionuclide | $^{238}$U | $^{237}$Np | $^{239}$Pu | $^{240}$Pu | $^{241}$Pu | $^{242}$Pu | $^{241}$Am |
|---|---|---|---|---|---|---|---|
| W/O (%) | 18.4 | 4.0 | 29.6 | 14.0 | 5.5 | 4.8 | 3.7 |

TRU, transuranic element; W/O, weight percent.

The thicknesses of the canister and the container were assumed to be 5 mm and 10 mm, respectively. The inner radius and the height of the canister were determined based on the possibility of the inner surface of the canister being placed 2 cm away from the surface of the TRU ingot. The container dimensions are just large enough to hold the canisters.

### 2.3. Postulation of accidental situations

The TRU storage area must be designed to comply with the criticality safety requirements recommended in 10CFR60.131. Thus, $k_{eff}$ should not exceed 0.95 under any extreme accidental condition. This value must include an allowance for bias and uncertainty in the calculation. Various accidental situations were postulated as described in the following to determine the most reactive configuration; three unlikely, independent, and concurrent accidental situations were taken into account. These situations are not controllable because they are caused by unexpected events such as natural disasters, accidents, or intentional manipulation.

- Owing to a flood or the activation of sprinklers due to fire, water or steam can leak into a canister (or container), and the submerged canister can be fully reflected by water. Consequently, all possible water densities from 0.1 to 1.0 g/cm$^3$ were considered (0.1 g/cm$^3$ intervals) [12].
- Owing to earthquakes or instances of intentional manipulation, the pitch between the containers can vary. In this study, the assumptions are that the canisters are in contact with each other and that the pitch would increase up to 50 cm at 1 cm intervals.
- The inventory of TRU ingots can accumulate and be left in TRU storage for an extended period due to unexpected circumstances. For the purposes of this work, the inventory was assumed to accumulate for up to 5 years.

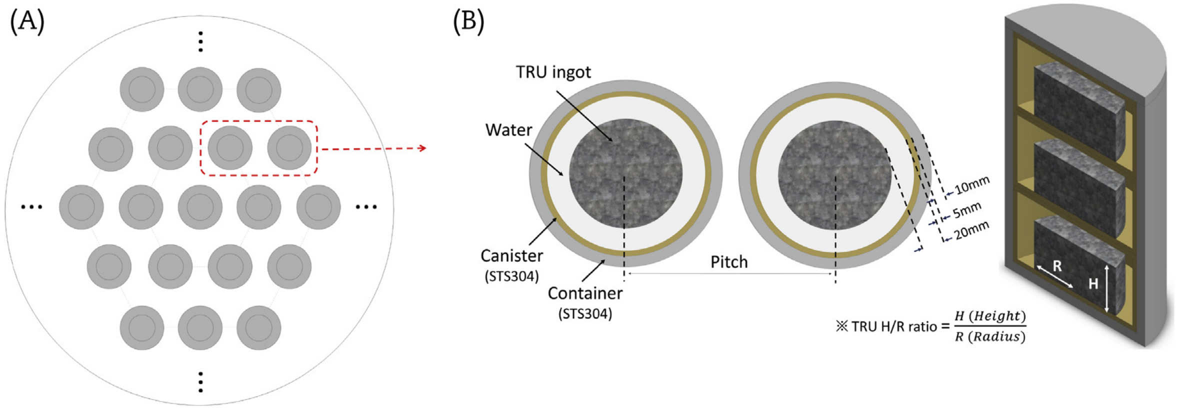

**Fig. 1.** TRU storage configuraiton (A) TRU storage filled with containers arranged in a hexagonal lattice pattern. (B) A schematic of a horizontal cross-sectional view of TRU ingots in a canister, showing the pitch on the left, and a schematic of a vertical cross-sectional view of three TRU ingots in three canisters in a container on the right.
H/R, height-to-the radius; TRU, transuranic element.

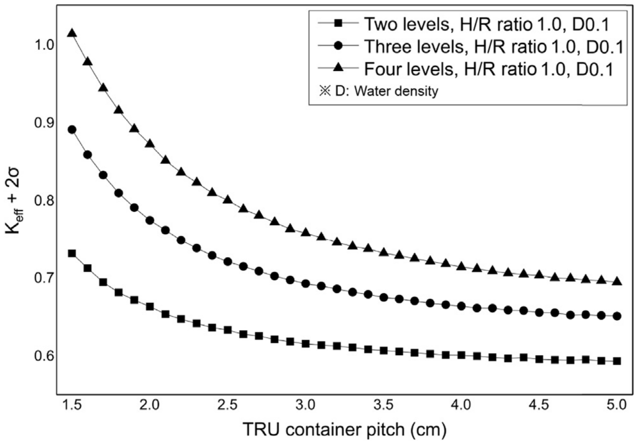

**Fig. 2.** Plot of $K_{eff}$ values according to the TRU container pitch for different combined situations.
H/R, height-to the radius; TRU, transuranic element.

### 2.4. Criticality calculation

The criticality evaluation in this study was carried out via MCNP6, which is a widely used tool in various validation processes and which allows for the possibility of using continuous energy and the latest nuclear cross-section libraries. Evaluated Nuclear Data File (ENDF)/B-VII.1 was used, and S (α,β) thermal neutron cross sections were applied to hydrogen in a water molecule.

All criticality calculations used 10,000 neutrons per cycle. Moreover, the initial 1,000 cycles were discarded for convergence before the $k_{eff}$ tallies, and 500 active cycles were run to prevent bias in the $k_{eff}$ calculations [13,14].

## 3. Results and discussion

To determine the most extreme accidental situations according to the TRU H/R ratio and the number of stacked canisters, $K_{eff}$ values were calculated while varying the pitches and water densities of the canisters submerged in the container. Fig. 2 shows an example

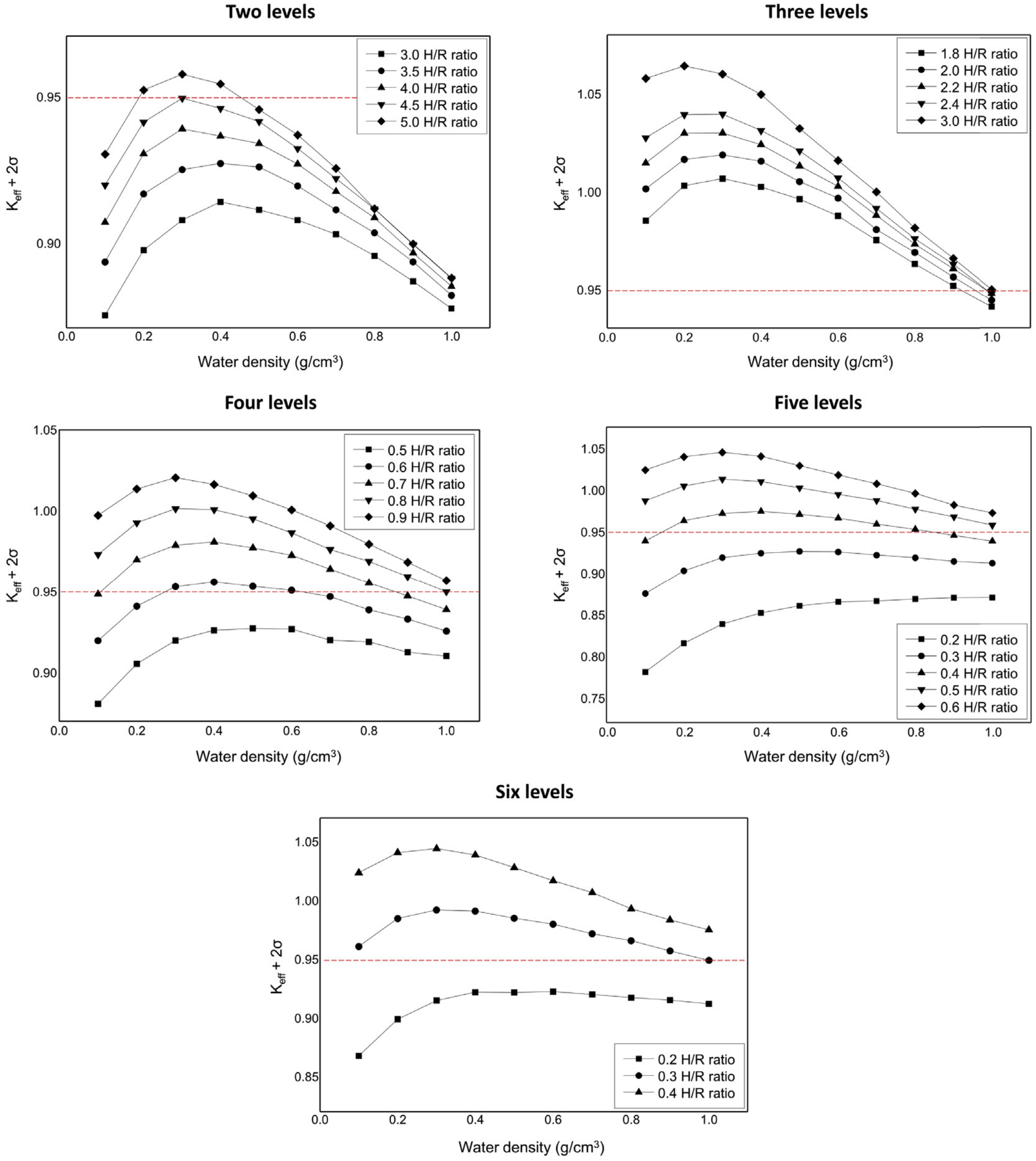

**Fig. 3.** Plot of $K_{eff}$ values as a function of the water density for different TRU H/R ratios and the number of stacked canisters.
H/R, height-to the radius; TRU, transuranic element.

**Table 2**
The highest $k_{eff}$ values according to the TRU H/R ratio and the number of stacked canisters among the various postulated accidental situations tested here. The water densities shown in the table correspond to the highest $k_{eff}$ values.

| TRU H/R ratio | Two levels | | Three levels | | Four levels | | Five levels | | Six levels | |
|---|---|---|---|---|---|---|---|---|---|---|
| | $k_{eff}+2\sigma$ | ρ (g/cm$^3$) | $k_{eff}+2\sigma$ | ρ (g/cm$^3$) | $k_{eff}+2\sigma$ | ρ (g/cm$^3$) | $k_{eff}+2\sigma$ | ρ (g/cm$^3$) | $k_{eff}+2\sigma$ | ρ (g/cm$^3$) |
| 0.2 | | | | | 0.81018 | 1 | 0.87007 | 1 | 0.92232 | 0.6 |
| 0.3 | | | | | 0.85682 | 1 | 0.92586 | 0.5 | 0.99183 | 0.3 |
| 0.4 | | | | | 0.89483 | 0.6 | 0.97384 | 0.4 | 1.04400 | 0.3 |
| 0.5 | | | 0.83556 | 1 | 0.92728 | 0.5 | 1.01245 | 0.3 | 1.08368 | 0.2 |
| 0.6 | | | 0.85940 | 0.7 | 0.95602 | 0.4 | 1.04466 | 0.3 | | |
| 0.7 | | | 0.87928 | 0.7 | 0.98081 | 0.4 | 1.06793 | 0.2 | | |
| 0.8 | | | 0.89871 | 0.6 | 1.00136 | 0.3 | | | | |
| 0.9 | | | 0.91352 | 0.6 | 1.02050 | 0.3 | | | | |
| 1 | | | 0.92956 | 0.5 | | | | | | |
| 1.1 | 0.80883 | 1 | 0.94303 | 0.4 | | | | | | |
| 1.2 | 0.81765 | 0.8 | 0.95609 | 0.4 | | | | | | |
| 1.3 | 0.82752 | 0.8 | 0.96626 | 0.4 | | | | | | |
| 1.4 | 0.83518 | 0.7 | 0.97517 | 0.3 | | | | | | |
| 1.5 | 0.84293 | 0.6 | 0.98325 | 0.4 | | | | | | |
| 1.6 | 0.84926 | 0.6 | 0.99132 | 0.4 | | | | | | |
| 1.7 | 0.85561 | 0.6 | 0.99963 | 0.3 | | | | | | |
| 1.8 | 0.86183 | 0.7 | 1.00640 | 0.3 | | | | | | |
| 1.9 | 0.86726 | 0.7 | 1.01432 | 0.3 | | | | | | |
| 2 | 0.87351 | 0.5 | 1.01860 | 0.3 | | | | | | |
| 2.2 | 0.88300 | 0.5 | 1.02973 | 0.3 | | | | | | |
| 2.4 | 0.89228 | 0.5 | 1.03940 | 0.3 | | | | | | |
| 3.0 | 0.91313 | 0.4 | 1.06408 | 0.2 | | | | | | |
| 3.5 | 0.92631 | 0.4 | | | | | | | | |
| 4.0 | 0.93813 | 0.3 | | | | | | | | |
| 4.5 | 0.94864 | 0.3 | | | | | | | | |
| 5.0 | 0.95690 | 0.3 | | | | | | | | |

H/R, height-to the radius; TRU, transuranic element.

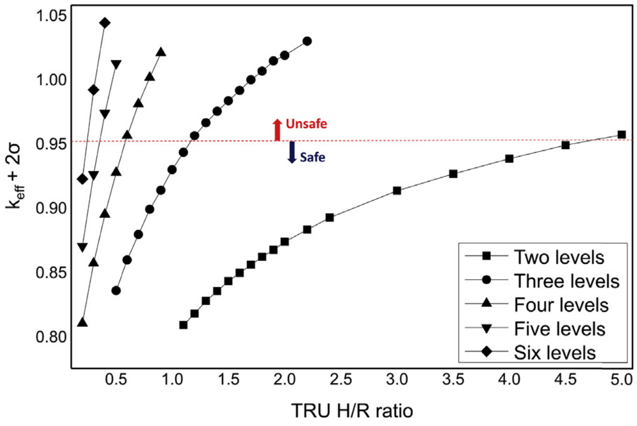

**Fig. 4.** Plot of the highest $k_{eff}$ value according to the TRU H/R ratio and the number of stacked canisters among the various postulated accidental situations tested here. H/R, height-to the radius; TRU, transuranic element.

of $K_{eff}$ as a function of the pitch in various combined situations. This figure indicates that the highest $K_{eff}$ arises when the containers are in contact with each other, and $K_{eff}$ decreases as the pitch increases. This phenomenon applies to all results calculated in this study.

Fig. 3 shows a plot of the $K_{eff}$ values calculated as a function of the water density for various TRU H/R ratios and numbers of stacked canisters. As shown in this figure, $K_{eff}$ tends to increase with a decrease in the water density of up to 0.2 g/cm$^3$. The maximum value of $K_{eff}$ is generally reached at a water density of 0.2–0.5 g/cm$^3$. This increase causes the TRU storage to transform from a subcritical state into a supercritical state under certain accidental situations. For three levels of stacked canisters, the difference in $K_{eff}$ from lowest to highest is greater than that for other levels of stacked canisters because of the water density.

Because the density of water for the highest $K_{eff}$ differs depending on the TRU H/R ratio and the number of stacked canisters, this value should be determined. Table 2 shows the highest $k_{eff}$ according to the TRU H/R ratio and the number of stacked canisters among various postulated accidental situations. The water density corresponding to the highest $K_{eff}$ is shown in this table. These $k_{eff}$ values are plotted as a function of the TRU H/R ratio in Fig. 4. Here, we found that the TRU H/R ratios should not exceed 4.5, 1.1, 0.5, 0.3, and 0.2 for two, three, four, five, and six levels of stacked canisters, respectively, so that TRU storage can be maintained in a subcritical state under extreme accidental conditions. In this figure, as the TRU H/R ratio increases, the $K_{eff}$ values increase for all stacked canister levels. In addition, the $K_{eff}$ values increase with the increasing number of stacked canisters. In other words, variation in the height has a greater effect on the variation of $K_{eff}$ than on the variation of the radius because increasing the height increases the probability of a fission reaction when neutrons move vertically. Decreasing the array size by reducing the radius, however, has a relatively small effect on the $K_{eff}$ values.

## 4. Conclusion

10CFR60.131 recommends that storage facilities should remain in a subcritical state even under extreme accidental conditions. Therefore, to determine the appropriate design parameters for TRU storage, a criticality safety analysis of a TRU storage area in a pyroprocessing facility was conducted using MCNP6. Various accidental situations were postulated for different water densities in submerged containers and for different pitches between the containers for a 5-year cumulative inventory of TRU ingots. In this work, we presented the threshold of the TRU H/R ratio depending on the number of stacked canisters so that the TRU storage area would remain in a subcritical state under extreme accidental conditions. The TRU H/R ratios should not exceed 4.5, 1.1, 0.5, 0.3, and

0.2 for two, three, four, five, and six levels of stacked canisters, respectively.

## Conflicts of interest

All authors have no conflicts of interest to declare.

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## Appendix ASupplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.net.2018.03.021.

## References

[1] H.D. Kim, H.S. Shin, D.Y. Song, T.H. Lee, B.Y. Han, S.K. Ahn, S.H. Park, Application of Safeguards-by-design for the Pyroprocessing Facilities in ROK, XL, 2012, pp. 24–31.

[2] R.A. Borrelli, Use of curium spontaneous fission neutrons for safeguardability of remotely-handled nuclear facilities: fuel fabrication in pyroprocessing, Nucl. Eng. Des. 260 (2013) 64–77, https://doi.org/10.1016/j.nucengdes.2013.03.025.

[3] R.A. Borrelli, Use of curium neutron flux from head-end pyroprocessing subsystems for the high reliability safeguards methodology, Nucl. Eng. Des. 277 (2014) 166–172, https://doi.org/10.1016/j.nucengdes.2014.06.028.

[4] R.A. Borrelli, Functional components for a design strategy: hot cell shielding in the high reliability safeguards methodology, Nucl. Eng. Des. 305 (2016) 18–27, https://doi.org/10.1016/j.nucengdes.2016.05.010.

[5] S.I. Moon, S.J. Seo, W.M. Chong, G.S. You, J.H. Ku, H.D. Kim, Identification of safety controls for engineering-scale pyroprocess facility, Nucl. Eng. Technol. 47 (2015) 915–923, https://doi.org/10.1016/j.net.2015.08.005.

[6] A. Mohammadi, M. Hassanzadeh, M. Gharib, Shielding calculation and criticality safety analysis of spent fuel transportation cask in research reactors, Appl. Radiat. Isot 108 (2016) 129–132, https://doi.org/10.1016/j.apradiso.2015.12.045.

[7] V. Barkauskas, R. Plukiene, A. Plukis, Actinide-only and full burn-up credit in criticality assessment of RBMK-1500 spent nuclear fuel storage cask using axial burn-up profile, Nucl. Eng. Des. 307 (2016) 197–204, https://doi.org/10.1016/j.nucengdes.2016.07.012.

[8] J.J. Herrero, A. Vasiliev, M. Pecchia, H. Ferroukhi, S. Caruso, Review calculations for the OECD/NEA burn-up credit criticality safety benchmark, Ann. Nucl. Energy 87 (2016) 48–57, https://doi.org/10.1016/j.anucene.2015.08.014.

[9] M. Rezaeian, J. Kamali, Effect of a dual-purpose cask payload increment of spent fuel assemblies from VVER 1000 Bushehr nuclear power plant on basket criticality, Appl. Radiat. Isot. 119 (2017) 80–85, https://doi.org/10.1016/j.apradiso.2016.10.008.

[10] S.I. Moon, W.M. Chong, G.S. You, J.H. Ku, H.D. Kim, Y.K. Lim, H.S. Chang, Preliminary safety study of engineering-scale pyroprocess facility, Nucl. Eng. Technol. 46 (2014) 63–72, https://doi.org/10.5516/NET.06.2013.018.

[11] S.M. Bowman, I.C. Gauld, OrigenArp primer: How to Perform Isotopic Depletion and Decay Calculations with SCALE/ORIGEN, Oak Ridge Natl. Lab., 2010, pp. 1–85. ORNL/TM-20.

[12] E. Tomlinson, C. Brown, Nuclear criticality safety considerations in design of dry fuel assembly storage arrays, Nucl. Technol. 63 (1983) 347–350.

[13] F. Brown, A Review of Best Practices for Monte Carlo Criticality Calculations, Los Alamos Natl. Lab., 2009, pp. 1–10. LA-UR-09-03136.

[14] R. Brewer, Criticality Calculations with MCNP5 TM : A Primer, Los Alamos Natl. Lab., 2009, pp. 0–201. LA-UR-09-00380.