

Assessment of Th-227 Production from Ra-226 Targets in a Commercial PHWR Neutron Field

Hongmin Jeon^a, Saehyun Choi^a, Taehoon Park^a, Pilhyeon Ju^a, Sungyeol Choi^{a,b,c*}

a Department of Nuclear Engineering, Seoul National University,

1 Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea

b Nuclear Research Institute for Future Technology and Policy, Seoul National University,

1 Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea

c Institute of Engineering Research, Seoul National University,

1 Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea

*Corresponding author: choisys7@snu.ac.kr

Keywords : Th-227, CANDU, MCNP, Medical Radioisotope, SCALE/ORIGEN

1. Introduction

Th-227 has recently drawn attention as a therapeutic alpha emitter [1,2]. The recovery of Th-227 from an Ac-227 parent is an established practice [1], so the supply of Th-227 through this route rests on the production of the Ac-227 parent.

Commercial power reactors offer an intense thermal-neutron environment in which the Ac-227 parent can be produced using existing infrastructure, as a complement to established production routes [3]. In this context, Ra-226 is the irradiation target for Ac-227 production, and is activated mainly by the (n,γ) capture reaction in the predominantly thermal neutron field of a commercial PHWR, followed by the subsequent decay chain:

Ra-226 (n,γ) Ra-227 (β⁻, 42.2 min) → Ac-227

(Decay) Ac-227 (β⁻, 21.8 y) → Th-227 (α, 18.7 d) → Ra-223 (α, 11.4 d) → ... → Pb-207 (stable)

(Capture) Ac-227 (n,γ) Ac-228 (β⁻, 6.15 h) → Th-228 (α, 1.91 y) → Ra-224 (α, 3.63 d) → ... → Pb-208 (stable)

Ac-227 is formed by a single capture on Ra-226, whereas Th-228 is co-produced through a second capture on the accumulated Ac-227 (via the short-lived Ac-228), so the two inventories evolve differently with irradiation time. Since Th-228 is an isotope of the product element and cannot be removed chemically, its co-production determines the isotopic quality of the recovered thorium.

This work evaluates how the irradiation period governs the Ac-227 parent inventory, the Th-227 product and the co-produced Th-228 – and thus the isotopic quality of the recovered thorium – for a Ra-226 target in a commercial PHWR. The effect of the post-irradiation decay period on the Th-227 inventory and

the Th-228/Th-227 activity ratio is then examined, and the irradiation window corresponding to a given isotopic specification is identified.

2. Method

2.1. Neutron field

A three-dimensional full-core model of a commercial PHWR (CANDU-6) was constructed with MCNP6.2 using ENDF/B-VIII.1 continuous-energy cross sections. The Ra-226 target was modeled as a pellet located in the heavy-water moderator region, and the neutron flux over the target volume was tallied in a 252-group structure and collapsed to one-group cross sections by the ORIGEN build_lib function. Normalized to nominal full-power operation, total flux over the target volume was $3.31 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$, of which 78 % lay below 0.625 eV.

2.2. Depletion and decay

The target mass is small enough that its effect on the core reactivity is negligible, so the neutron field obtained from MCNP was held fixed during depletion. This allows the burnup to be solved by point depletion, at negligible computational cost compared with Monte Carlo depletion, so that several irradiation periods can be scanned. Point-depletion calculations were performed with SCALE 6.3.3 ORIGEN for a Ra-226 target of 3.6933 g. The irradiation period was the only variable in this study: six values— 2 h, 1 d, 2 d, 10 d, 50 d and 120 d — were calculated, each followed by an identical 90 d decay period during which the whole target was left intact without chemical separation. The Ac-227 activity was evaluated 1 d after discharge, by which time the residual Ra-227 had essentially decayed to Ac-227. The Th-227 and Th-228 inventories were compared at the common 90 d post-irradiation point.

Since a chemical separation of thorium cannot alter the Th-228/Th-227 ratio, the ratio obtained for the whole target is that of the recovered thorium. The 90 d

period was chosen to cover the growth of Th-227 toward its quasi-equilibrium level with Ac-227 (3.32 half-lives of Th-227, at which 90% of the equilibrium level is reached, corresponds to 62 d). The 90 d point is used as a common comparison time for all cases; the full time evolution is given in Fig. 2, so that the recovery time can be chosen against any given requirement rather than fixed here.

3. Results and Discussion

3.1. Dependence on the irradiation period

Table 1 summarizes the calculated inventories. The irradiation period sets a trade-off between Th-227 yield and the isotopic quality of the thorium. A short irradiation gives a small absolute inventory: 0.080 Ci of Th-227 for the 1 d case against 2.71 Ci for the 50 d case. It also gives far less Th-228. The Th-228/Th-227 ratio was 0.087 at 1 d, 4.99 at 50 d and 12.6 at 120 d, increasing almost in proportion to the irradiation period. The mechanism is a second neutron capture. Th-228 requires a capture on the accumulated Ac-227 (via 6.15-h Ac-228), so it builds up faster than the single-capture products as irradiation continues. The same second capture also consumes part of the accumulated Ac-227. At 120 d the Th-228 activity reached 51 Ci against 4.1 Ci of Th-227.

Table 1. Ac-227, Th-227 and Th-228 activities and the Th-228/Th-227 activity ratio for the six irradiation periods. The last column is the marginal exchange rate, the increase in Th-227 activity divided by the increase in Th-228 activity relative to the preceding row ($\Delta\text{Th-227}/\Delta\text{Th-228}$).

Irradiation	Ac-227 (Ci)	Th-227 (Ci)	Th-228 (Ci)	Th-228/Th-227	Exchange rate
2 h	7.08×10^{-3}	6.69×10^{-3}	2.0×10^{-5}	3.4×10^{-3}	—
1 d	8.43×10^{-2}	7.97×10^{-2}	6.92×10^{-3}	8.67×10^{-2}	10.6
2 d	1.67×10^{-1}	1.58×10^{-1}	2.87×10^{-2}	1.81×10^{-1}	3.61
10 d	7.80×10^{-1}	7.42×10^{-1}	7.01×10^{-1}	9.46×10^{-1}	0.87
50 d	2.83	2.71	13.5	4.99	0.15
120 d	4.21	4.05	51.2	12.6	0.036

The trade-off is monotonic. No irradiation period is optimal in itself; the choice follows from the isotopic specification imposed on the product.

Read in that direction, Table 1 maps a specification onto an operating window in both yield and purity. Limits of 0.1, 1 and 10 on the Th-228/Th-227 ratio correspond to irradiations of about 1 d, 10 d, and between 50 d and 120 d. Each tenfold relaxation of the limit buys about a tenfold gain in Th-227 up to a ratio of 1, and less beyond it.

The exchange also becomes progressively unfavourable. Beyond about 10 d, each additional unit of Th-227 costs more than one unit of Th-228. The reason is that the Ac-227 inventory saturates while Th-

228 keeps rising: extending from 50 d to 120 d (2.4 times longer) yielded only 1.5 times more Ac-227 (Fig. 1).

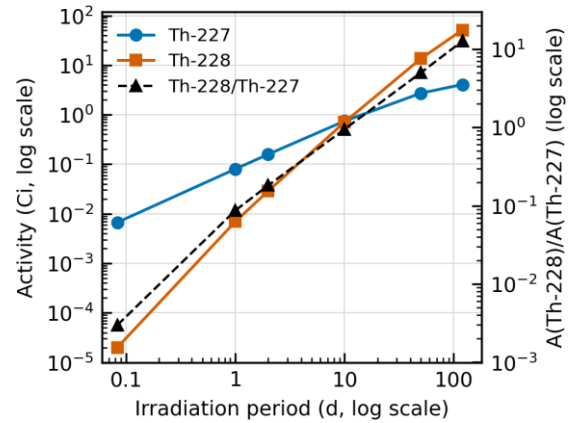


Fig. 1. Th-227 and Th-228 activities (left axis, log) and Th-228/Th-227 activity ratio (right axis) versus irradiation period; log x-axis, 0.08 d (2 h) – 120 d.

3.2. Post-irradiation changes of the thorium inventories

Two features shape the evolution. First, neutron-induced production of Ac-228 ceases at the end of irradiation (EOI). The residual 6.15-h Ac-228 then decays to Th-228 during the first few days, raising the Th-228 inventory by roughly a factor of two for the 1 d case but by only about 1% for 50 d. Th-228 is then essentially constant on the time scale of interest (its half-life is 1.9 y). Second, Th-227 grows continuously from the long-lived Ac-227 parent toward transient equilibrium, on the scale of its own 18.7 d half-life. The ratio therefore decreases monotonically with decay time in every case (Fig. 2). For the 50 d case it falls from 12.7 at EOI to 5.0 at 90 d. The separation between the curves for different irradiation periods, more than three orders of magnitude between 2 h and 120 d, persists throughout. The isotopic composition at EOI is therefore not representative for short irradiations, and the product quality should be assessed only after the residual Ac-228 has decayed. Longer decay before the first thorium recovery increases the Th-227 inventory while simultaneously improving the isotopic composition; the choice of recovery time can then be made against whatever composition requirement applies. The gain is bounded, however. Th-227 reaches 90% of its equilibrium level with Ac-227 at 62 d. Beyond that the ratio falls only through the difference between the 1.9 y half-life of Th-228 and the 21.8 y half-life of the Ac-227 parent, an effective half-life of about 2.1 y. Bringing the 50 d case down to the ratio reached by the 1 d case would therefore take about 12 years. The attainable isotopic quality is set by the irradiation period. The decay period only determines how quickly it is approached. Like the irradiation period it is fixed by the specification rather than by an optimum.

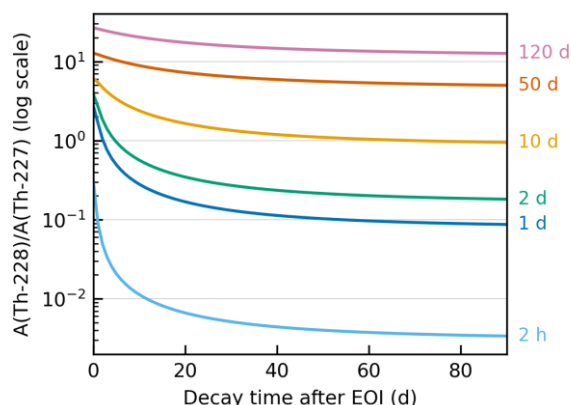


Fig. 2. Th-228/Th-227 activity ratio as a function of decay time after the end of irradiation, for the six irradiation periods. Decay time 0–90 d; logarithmic ordinate.

4. Conclusion

The production of Th-227 from a Ra-226 target in a commercial PHWR neutron field was evaluated with MCNP6.2 and SCALE/ORIGEN. Six irradiation periods from 2 h to 120 d were calculated, each followed by a 90 d post-irradiation period. Long irradiation converted the accumulated Ac-227 into Th-228, degrading the Ac-227 yield per irradiation time and the thorium isotopic composition simultaneously. Short irradiation preserved an almost proportional Ac-227 build-up while substantially suppressing Th-228 co-production. This ordering between irradiation periods held at every point of the evaluated post-irradiation interval. Because the dependence is monotonic, no irradiation period is optimal in itself. The results instead map an isotopic specification onto the corresponding operating window, with a Th-228/Th-227 limit of 0.1, 1 and 10 corresponding to irradiations of about 1 d, 10 d and between 50 d and 120 d respectively. These results identify the irradiation period as the primary variable controlling the isotopic quality of the thorium obtainable from this route. The acceptable Th-228 content and the actual separation and target-handling cycle were not defined in this work. Work in progress extends the six discrete conditions to a dense grid of irradiation and decay times, so that the yield and the isotopic ratio can be read as a continuous operating map. It also introduces the production rate per unit cycle time as the objective. Under a specified isotopic limit this objective admits a genuine optimum, which the present batch-yield formulation does not.

Acknowledgements

This work was supported by KOREA HYDRO & NUCLEAR POWER CO., LTD. (A24LP07).

REFERENCES

- [1] R. Eychenne, M. Chérel, F. Haddad, F. Guérard and J.-F. Gestin, Overview of the Most Promising Radionuclides for Targeted Alpha Therapy: The "Hopeful Eight", *Pharmaceutics*, Vol. 13, No. 6, 906 (2021).
- [2] V. Di Iorio et al., Recommendations on the Clinical Application and Future Potential of α -Particle Therapy: A Comprehensive Review of the Results from the SECURE Project, *Pharmaceutics*, Vol. 18, No. 10, 1578 (2025).
- [3] A. R. Hanu, J. Atanackovic, C. Boyd, E. M. Johnston and A. J. Waker, Characterization and Mapping of the Neutron Fields around Bruce Power's ^{177}Lu Isotope Production System, *Applied Radiation and Isotopes*, Vol. 208, 111284 (2024).