

ASSESSING THE OTTO OPTION: THORIUM-CYCLE EXPERIMENTAL POWER REACTOR SPENT FUEL CHARACTERISTICS

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ABSTRACT

ASSESSING THE OTTO OPTION: THORIUM-CYCLE EXPERIMENTAL POWER REACTOR SPENT FUEL CHARACTERISTICS. Two main refuelling option considered for use in a High Temperature Gas-cooled Reactor (HTGR) are multi-pass and once through-then-out (OTTO) cycle. The former possesses superiority in term of fuel utilisation whilst the latter is considered better in term of system simplicity. HTGR-based Experimental Power Reactor (Reaktor Daya Eksperimental/RDE) is supposed to adopt multi-pass refuelling scheme. However, OTTO-scheme shall also be considered for the simplicity it offers and thus potentially lower cost. Due to different neutronic and burnup profile between the two, the resulting spent fuel characteristic is also different and possibly requires different handling mechanism. This paper assesses the characteristics of OTTO-scheme RDE spent fuel using thorium fuel cycle to provide preliminary data and insight for its spent fuel management. The assessment is performed employing ORIGEN2.1 code. At day 30 of cooling after determined end-of-cycle (EOC), each spent fuel yields 234.9 Curies of radioactivity, emitting 66.26 neutrons/second, 1×10^{13} photons/second, and releasing 0.7675 watts of decay heat. These numbers must be taken into consideration regarding spent fuel management and spent fuel cask design. TI-208 isotope characteristics, whose existence is unique to thorium fuel cycle, were also determined. It is found to be yielding 3.42×10^{-3} Curie of radioactivity and releasing 1.2×10^8 photons/second at its peak. Understanding its high-energy gamma release, proper radiation protection mechanism must be implemented.

Keywords: RDE, spent fuel, thorium, OTTO scheme, ORIGEN2.1

ABSTRAK

PENILAIAN OPSI OTTO : KARAKTERISTIK BAHAN BAKAR NUKLIR BEKAS SIKLUS-THORIUM PADA REAKTOR DAYA EKSPERIMENTAL. Dua opsi refueling yang dipertimbangkan dalam High Temperature Gas-cooled Reactor (HTGR) adalah multi-pass dan once through-then-out (OTTO). Siklus multi-pass lebih superior dari segi pemanfaatan bahan bakar, sementara OTTO dianggap lebih baik dari simplisitas sistem. Reaktor Daya Eksperimental (RDE) yang berbasis HTGR direncanakan untuk menggunakan skema multi-pass. Namun, skema OTTO layak pula untuk dipertimbangkan mengingat kesederhanaan sistem yang ditawarkannya, sehingga biaya yang dibutuhkan dapat lebih rendah. Mengingat perbedaan profil netronik dan derajat bakar antara kedua skema tersebut, karakteristik bahan bakar bekas yang dihasilkannya juga berbeda dan mungkin membutuhkan penanganan berbeda pula. Makalah ini menilai karakteristik RDE dengan skema OTTO menggunakan siklus bahan bakar thorium dengan tujuan menghadirkan data awal untuk manajemen bahan bakar bekasnya. Penilaian dilakukan menggunakan kode komputer ORIGEN2.1. Pada hari ke-30 pendinginan setelah end-of-cycle (EOC), tiap bahan bakar memancarkan radioaktivitas sebesar 234,9 Curie, 66,26

neutron/detik, 1×10^{13} foton/detik, dan melepaskan 0,7675 watt panas peluruhan. Angka-angka ini harus dipertimbangkan terkait manajemen bahan bakar bekas dan desain kanister bahan bakar bekas. Karakteristik isotop TI-208, yang eksistensinya hanya ada pada siklus bahan bakar thorium, juga ditentukan. TI-208 diketahui memancarkan radioaktivitas sebesar $3,42 \times 10^{-3}$ Ci dan melepaskan $1,2 \times 10^8$ foton/detik pada puncaknya. Mengingat lepasan gamma energi tinggi dari TI-208, harus diimplementasikan mekanisme proteksi radiasi yang tepat.

Kata kunci : RDE, bahan bakar nuklir bekas, thorium, skema OTTO, ORIGEN2.1

1. INTRODUCTION

Development of nuclear reactor technology is currently gearing into Generation IV designs, one of them being high temperature gas-cooled reactor (HTGR). The design is superior over the conventional light water reactor (LWR) in term of its passive safety system and higher operational temperature. Its low power density, along with TRISO-coated micro-sized fuel kernels, ensure passive cooling capability and better fission product retention during both normal and accident condition (1–4).

HTGR technology was adopted by National Nuclear Energy Agency of Indonesia (Badan Tenaga Nuklir Nasional/BATAN) as the design for Experimental Power Reactor (Reaktor Daya Eksperimental/RDE), a 10 MWth non-commercial power reactor expected as the reference plant for the indigenous nuclear power plant programme (5,6). Pebble bed core was chosen over prismatic core due to its perceived design superiority (7). Thus, the TRISO-coated fuel particles will be further encapsulated within a tennis sized-graphite ball, called pebble. The pebble fuel, combined with aforementioned features, ensure that HTGR is meltdown-proof (8).

Two main refuelling schemes are available for pebble bed HTGR, namely multi-pass scheme and once-through-then-out

(OTTO) scheme. *Peu-a-peu* scheme, while also available, is rarely considered (9,10). Multi-pass scheme recirculates the fuel pebble several times until it reaches the desired burnup, accompanied with cooling period between the recirculation. This option offers better fuel utilisation, better neutron economy, and flatter core power peaking (11).

However, the fuel recirculation system is relatively complicated and therefore contributes to cost increment. OTTO scheme omits such system, the fuel is burned within the core until it reaches its desired burnup, then directly discarded. This option offers simplicity and possibly lower cost than multi-pass scheme. The trade-offs are worse neutron economy and higher power peaking. Lack of cooling period in-between also results in lower burnup compared to multi-pass scheme, although the difference may not be significant (11–13).

Although RDE opted for multi-pass refuelling scheme for its design, OTTO scheme may also be considered for simpler design with relatively small trade-off. Neutronic calculation of HTGR with OTTO scheme was discussed previously and even suggested as the preferable design for RDE (10–15). As previously mentioned, different refuelling scheme affects the burnup and thereby different spent fuel characteristics. Assessing the characteristics of spent fuel

from OTTO scheme RDE is a necessity to properly design its spent fuel management, from the fuel handling system, interim storage, and its final disposal.

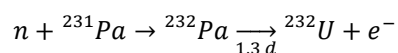
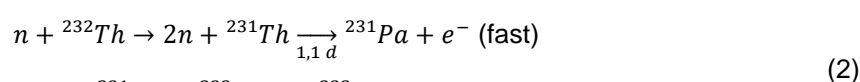
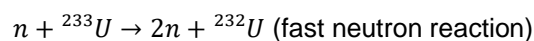
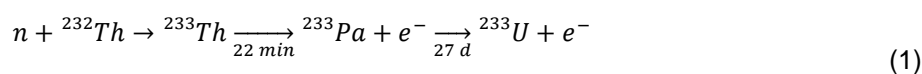
Likewise, despite RDE will most likely use low-enriched uranium (LEU), other fuel cycle such as uranium-thorium (U-Th) or even mixed oxide (MOX) is also possible to be used (16–18). Different fuel cycle also yields different spent fuel characteristics. The purpose of this paper is to characterise the spent fuel of thorium-fuelled RDE with OTTO scheme, in order to provide preliminary data and insight for its spent fuel management, from handling until disposal. The characterisation is performed using radionuclide burnup and decay code ORIGEN2.1. HTGR-specific cross-section library is adopted to yield accurate calculation output. The result will be compared with multi-pass RDE fuel, both LEU and thorium-based.

Theory

One pebble fuel contains thousands of TRISO-coated fuel kernels, dispersed randomly within a graphite sphere. The initial design proposal employs 17%wt-enriched LEU in oxide form as the fuel. As the alternative fuel cycle, thorium cycle employing U-233 and thorium will replace LEU in this study.

Thorium cycle possesses many advantages over LEU cycle in thermal reactor (19). Among them are lower initial inventory, larger fuel breeding, lower transuranic generation, and more proliferation resistant. The latter is contributed by the existence of U-232, a 68.9-year half-life isotope which decays into Tl-208, a strong 2.6 MeV gamma ray emitter. A few hundred ppm of U-232 contamination within U-233 is sufficient to make the latter hazardous and consequently requiring specialised handling system in order to divert it for weapon (20–22).

U-232 is formed primarily from two-step reaction involving (n,2n) reaction (20). Its formation is explained in Equation 1 and 2.



The existence of U-232 makes thorium fuel cycle more proliferation resistant, but at the same time also complicates the fuel management due to potentially intense gamma emission. Thus, to safely manage the

thorium RDE spent fuel, U-232 inventory is necessary to be evaluated.

Computer code used in this study is ORIGEN2.1, a development from ORIGEN programme originally developed for calculating source term for various reactor

materials and reactor types. ORIGEN2.1 works by employing Bateman equation and one-group spectrum-averaged cross section library. The build-up and decay of isotopes

$$\frac{dX_i}{dt} = \sum_{j=1}^N l_{ij}\lambda_j X_j + \varphi \sum_{k=1}^N f_{ik}\sigma_k X_k - (\lambda_i + \varphi\sigma_i + r_i)X_i + F_i, \quad i = 1, \dots, N \quad (3)$$

where X_i denotes density of nuclide i , N is the number of nuclides, l_{ij} is radioactive disintegration fraction by other nuclide that leads to formation of species i , j is iterations number from $j = 1$ to $j = N$, φ is neutron flux averaged in position and energy, f_{ik} is neutron fraction absorbed by other nuclides that leads to formation of species i , σ_k is averaged neutron absorption cross section of nuclide k , r_i is nuclide i continuous removal rate out from the system, and F_i is nuclide i continuous feed rate.

ORIGEN2.1 requires specific reactor-dependent cross section in order to solve the equation properly. Thus, HTGR-specific one-group averaged cross section, codenamed RDE.LIB, was employed. The library was generated for HTGR reactors and has been validated for use in 10MWt HTGR. As the calculation result, ORIGEN2.1 yield actinides, fission products, and activation products for amount of 130, 850, and 730 nuclides, respectively (25–27).

Radionuclide calculation for HTGR reactors using ORIGEN have been performed (27–29), including spent fuel characteristics for RDE and AVR spent fuel (26,30,31). However, none of them discussing radionuclide characterisation of OTTO scheme HTGR. Similarly, spent fuel characteristics of thorium-based fuel

are represented by differential equation shown in Equation 3 (23,24).

discussed by Dwijayanto et al (30) was calculated for multi-pass scheme.

2. METHODOLOGY

The burnup and decay simulation of thorium fuelled, OTTO scheme RDE is performed mirroring LEU fuelled, multi-pass scheme RDE in most of the parameters (26). For exception, obviously, is that the fuel stays within the reactor core without decay period during fuel recirculation. Since ORIGEN2.1 performs the calculation as a point reactor, it is not feasible to simulate the whole core. Instead, fuel burnup and decay are calculated for one pebble.

Fuel loading is also different from regular RDE fuel. Several neutronic calculations for thorium-based HTGR fuel (11,32,33) are available. This study opted for fuel loading used in Reference (11), since the fuel loading mentioned in the article is dedicated for OTTO-scheme, thorium-cycle RDE.

Thorium is widely known to be performing better than uranium in thermal spectrum. Thus, it is expected that the thorium cycle pebble fuel is able to stay longer within the core, as mentioned in several references (11,34). However, for direct comparison with LEU RDE fuel, burnup time is set at 1080 effective full power days

(EFPD), exactly the same with previous simulation.

After irradiated, decay calculation is performed until 50 years after discharge. Since the short-lived radionuclides are yet to decay right after discharge, decay pattern for the first 180 days is also calculated, 30 days for each cycle.

Pebble fuel parameters used in this calculation is shown in Table 1.

Table 1. Pebble fuel parameters

Parameter	Value
Fuel type	(U,Th)O ₂
Fuel thermal power	3.7 kW
Heavy metal loading	18.23 g
Fissile enrichment	7.2%
Fuel pebble mass	200 g
Fuel loading scheme	OTTO
Fuel residence time	1080 EFPD

In many neutronic/burnup calculations involving thorium fuel cycle, fissile U-233 is usually assumed to be pure from any contaminations (11,32,35,36). In reality various degrees of impurity in form of higher isotopes of uranium, namely U-234, U-235, and U-236, as well as lower isotope U-232, often exist as contaminants for U-233 (37,38). Thus, as realistic assumption, this study assumed that fissile U-233 is contaminated with aforementioned isotopes. Isotopic composition of uranium from Heuer et al (37) is adopted in this study. This is a slight deviation from the reference fuel, where it was assumed pure.

Uranium isotopic composition is shown in Table 2.

Table 2. Isotopic Composition of Thorium-cycle RDE Fuel (37)

Isotope	Mass (g)	Isotope	Mass (g)
U-232	0.00049	Th-232	16.91744
U-233	1.12868	O-16	2.51371
U-234	0.15502	C-12	176.6301
U-235	0.02201	Si-28	2.62616
U-236	0.00636		

3. RESULTS AND DISCUSSION

The calculation result was compiled and divided into four categories, namely radioactivity, photon release, neutron emission, and lastly heat release. TI-208 is specifically discussed for its radioactivity and photon release. All values are for one fuel pebble. Results shown and discussions are centred after the fuel is discharged from the core.

Radioactivity

ORIGEN2.1 calculates radioactivity in three categories, namely activation products, actinides along with its daughter products, and fission products (referred hereinafter as AP, ACT, and FP, respectively). Right after discharge, fuel radioactivity is extremely high due to the absence of fuel cooling period in the fuel recirculation system. Its immediate activity, thus, cannot be directly compared to those of multi-pass spent fuel. Decay calculation for the first 180 days after discharge was then performed. The pattern is shown in Figure 1 and Table 3.

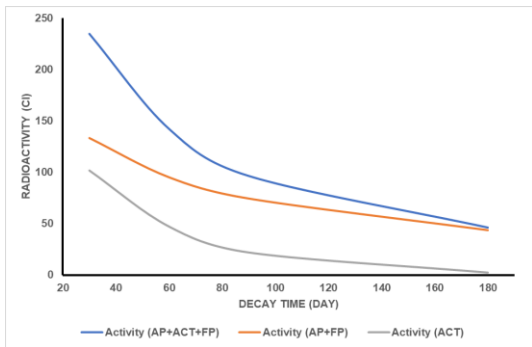


Figure 1. Total radioactivity decay over 180 days

Table 3. Total radioactivity value over 180 days of decay

Time (days)	Radioactivity (Ci)
0 (EOC)	2,296
30	234.9
60	142.2
90	96.35
180	46.01

Spent fuel radioactivity decreased to around 10% its initial activity in just 30 days, and further decreased to 2% at day 180. Such steep decrease is understood as the short-lived ACTs and FPs are vanishing quickly. FPs are the first to disappear. Yttrium, strontium, iodine, xenon, barium, and lanthanum isotopes decayed into less than 10% their EOC radioactivity after 30 days of decay, whilst zirconium and niobium isotopes both decayed into 13.2% and 12.4% their EOC activity, respectively. The aforementioned isotopes account for 60.6% of EOC FPs activity, which explains FPs sharp decrease of radioactivity share.

ACTs initially decay more slowly, noting that its share of radioactivity increased to 43.25% at day 30. However, its share diminished into 4.85% after 180 days, since the short-lived Th-233 completely vanished

and Pa-233 decayed to 10% its initial activity. The rest of ACTs do not contribute for total radioactivity in noticeable amount.

Generally, the radioactivity of OTTO and multi-pass thorium-based fuel is higher compared to multi-pass LEU-based fuel. LEU-based fuel radioactivity at discharge is comparable with that of OTTO thorium-based fuel at day 90 after discharge. Such high activity is caused by the existence of Pa-233, the precursor of U-233, whose half-life (27.2 days) is longer than the precursor of Pu-239 exists in LEU-cycle. This must be taken into account when designing the spent fuel cask regarding its decay heat removal, especially during the first year after discharge.

Further decay calculation was performed until 5 years after discharge. The result is shown in Figure 2.

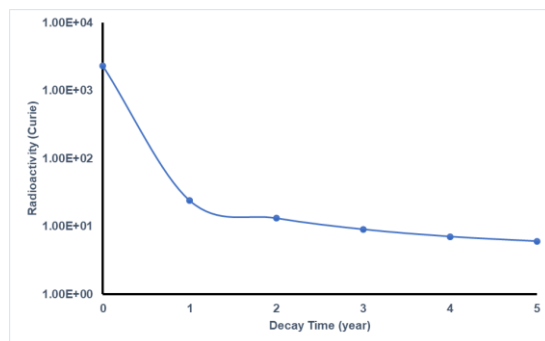


Figure 2. Radioactivity decay over 5 years

The remaining FPs and ACTs at the fifth-year amount for 6.03 Ci compared with 2,296 Ci at the EOC, or about 0.263% of the latter. The fifth-year radioactivity is slightly lower than multi-pass LEU-based fuel, around 6% less, but 23.5% higher than thorium-based, multi-pass RDE fuel. The difference between thorium-based fuel is most likely due to different fuel loading and

thus different burnup characteristics. In term of long-term heat removal, thorium-fuelled RDE spent fuel with OTTO scheme may not need different treatment with its regular LEU fuel cycle.

Nevertheless, the radiation protection aspect is not necessarily similar between LEU and thorium-fuelled spent fuel, due to TI-208 activity. Radioactivity of TI-208 along with its parent, U-232, are shown in Figure 3.

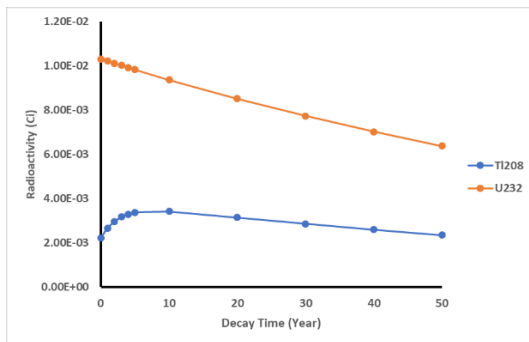


Figure 3. U-232 and TI-208 decay over 50 years

U-232 activity decreased continuously over time, but different pattern is apparent in TI-208. Instead of decreasing, its activity increased rather drastically for about 52.9% in the first 5 years. The activity continues to

increase until peaked in tenth year, albeit only increased 1% from the fifth year. Afterwards, the radioactivity decreased steadily, reduced to 68.8% of its peak activity at year 50.

Although TI-208 radioactivity is fairly low, its 2.6 MeV gamma emission may be necessary to put into consideration for radiation protection in spent fuel handling. Its peculiar radioactivity behaviour should also be noted as its gamma emission peaked in tenth year and yet to fall below EOC activity even after 50 years of cooling. In OTTO scheme, this is especially important since the radioactivity of both isotopes are twice as high as its multi-pass counterpart.

Photon Release

Photon released by the spent fuel are mostly in form of gamma emission. Thus, both terms are used interchangeably.

ORIGEN2.1 is able to yield 18 photon energy groups, ranging from 0.01-9.5 MeV. The following figure shows the 18-group gamma release at the EOC and after 1, 5, and 10 years of decay.

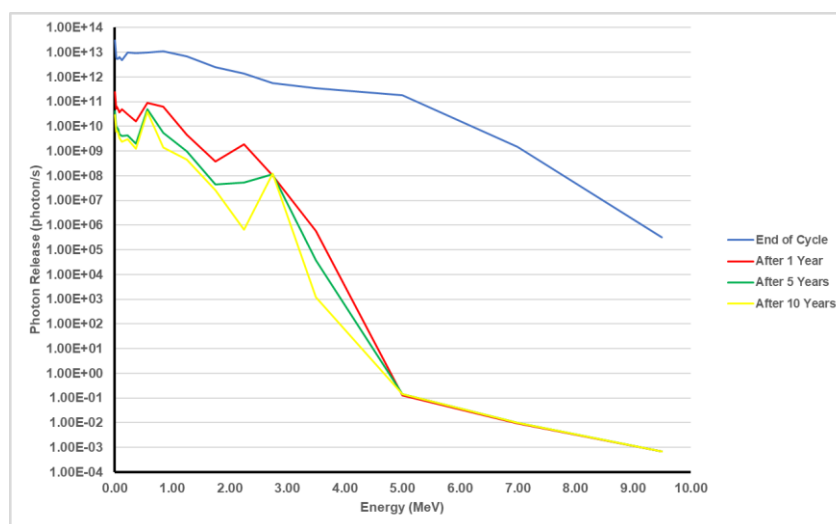


Figure 4. Gamma release at EOC and over 10 years of decay

Gamma release at the EOC is understandably much higher than the rest, amounting 1.11×10^{14} photons/second release rate. Therefore, its value does not necessarily represent the mandatory requirement in spent fuel handling. Calculation for gamma release after 30 days of decay was then performed and found to be releasing approximately 1×10^{13} photons/second. This value is around three times higher than LEU-based RDE spent fuel at discharge and 21.85% higher than its multi-pass counterpart. Meaning, higher

radiation protection is necessary after fuel discharge.

Generally, release rate is decreasing as the energy gets higher, except for one high-energy 2.75 MeV group apparent at fifth and tenth year of decay. This particular energy group released higher gamma ray than previous group, before dropping in the next group. The blip shows non-negligible release of gamma ray from TI-208, daughter product of U-232 decay chain.

Better perspective of TI-208 gamma release is shown in the following ACT-specific photon release graph.

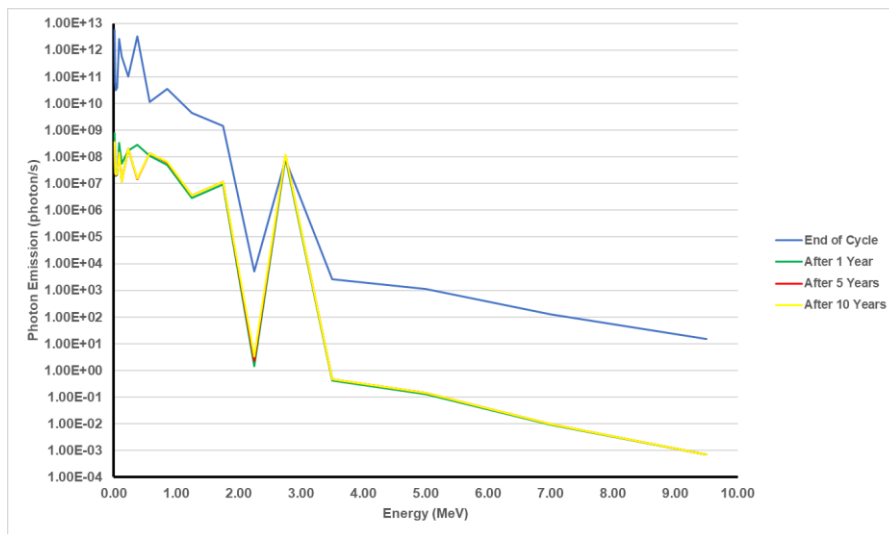


Figure 5. ACTs gamma release at EOC and over 10 years of decay

The figure shows clearly the sharp blip in 2.75 MeV energy group, even at the EOC. At the fifth and tenth year of decay, the 2.75 MeV energy group is also among the highest emitting group. This, along with the previous figure, suggest that despite being low in activity, TI-208 gamma release is not necessarily negligible in radiation protection

aspect. Especially considering that TI-208 activity peaked at the tenth year of decay.

Neutron Emission

Main source of neutron emission in LEU-cycle spent fuel is Pu-240 and Cm-242, due to their high probability of spontaneous fission. Pure thorium fuel cycle generates significantly less transuranic elements, thus

neutron emission from spontaneous fission is lower compared to LEU fuel cycle. In total, neutrons released by spontaneous fission amount for 0.228 neutrons/second. This is thousands of times smaller than neutrons released by LEU-cycle spent fuel, since Cm-242 is virtually absent.

Apart from spontaneous fission, ORIGEN2.1 also calculated neutron emission by (alpha, n) mechanism. This path released more neutrons than spontaneous fission, amounting for 65.06 neutrons/second. U-232 daughter products, Po-212 and Po-216, are the largest neutron emitter, accounting for 11.47 and 11.02 neutrons/second respectively. Nevertheless,

neutron emission from (alpha, n) mechanism is still remarkably low. In total, each thorium cycle spent pebble fuel released 65.29 neutrons/second at EOC. Its neutron emission also roughly twice as large as multi-pass thorium-based RDE fuel.

Since the main neutron emitter are U-232 daughters, it is also expected that the neutron emission will increase over time. This proves to be true, as neutron emission from those isotopes increased at day 30 of decay. However, the increase is too trivial of any significance.

Table 3 provides detailed neutron emission from OTTO thorium cycle RDE spent fuel.

Table 3. Neutron emission at EOC and after 30 days of decay

EOC				30 days of decay			
(alpha, n)		Spontaneous		(alpha, n)		Spontaneous	
Bi-212	5.41	U-232	5.22x10 ⁻⁴	Bi-212	5.51	U-232	5.22x10 ⁻⁴
Po-212	1.15x10 ¹	U-233	6.50x10 ⁻⁴	Po-212	1.17x10 ¹	U-233	6.54x10 ⁻⁴
Po-213	7.33x10 ³	U-234	6.66x10 ⁻⁴	Po-213	7.25x10 ⁻³	U-234	6.66x10 ⁻⁴
Po-216	1.10x10 ¹	Pu-238	2.20x10 ⁻¹	Po-216	1.12x10 ¹	Pu-238	2.21x10 ⁻¹
At-217	5.43x10 ⁻³	Pu-240	3.81x10 ⁻⁴	At-217	5.37x10 ⁻³	Pu-240	3.81x10 ⁻⁴
Rn-220	9.01	Cm-242	5.97x10 ⁻³	Rn-220	9.18	Cm-242	5.31x10 ⁻³
Fr-221	4.21x10 ⁻³	Cm-244	2.59x10 ⁻⁴	Fr-221	4.16x10 ⁻³	Cm-244	2.59x10 ⁻⁴
Ra-224	6.59			Ra-224	6.72		
Th-228	5.60			Th-228	5.71		
U-232	8.75			U-232	8.75		
U-233	5.22			U-233	5.26		
U-234	6.19x10 ⁻¹			U-234	6.19x10 ⁻¹		
Pu-238	1.35			Pu-238	1.35		
Total			65.29 neutrons/second	Total			66.26 neutrons/second

Heat Release

Spent fuel still releases heat as the result of radionuclides decay. In agreement with previous subsections, AP contribution in

decay heat release is too low to be regarded of significance, even at the EOC.

Right after discharged, rapid decay of short-lived FPs and ACTs ensures the heat

release is large. Again, this is incomparable with multi-pass, LEU spent fuel. Heat release calculation was then performed until 90 days

after discharge, 30 days for each cycle. Calculation result is provided in Table 4.

Table 4. Heat release for each radionuclide category over 90 days of decay

Category	Decay heat release (watt)			
	EOC	30 days	60 days	90 days
AP	8.48×10^{-4}	4.99×10^{-9}	3.01×10^{-9}	2.54×10^{-9}
ACT	1.07	2.32×10^{-1}	1.09×10^{-1}	5.14×10^{-2}
FP	2.32×10^1	5.35×10^{-1}	3.68×10^{-1}	2.88×10^{-1}
Total	2.42×10^1	7.67×10^{-1}	4.77×10^{-1}	3.39×10^{-1}

FPs consistently being the largest contributor of decay heat release. Due to short-lived FPs decay, its share reduced to 69.71% at day 30 of decay from 95.58% at EOC. Its share increased again to 84.83% at day 90 as longer-lived ACTs decay.

At day 60 of decay, the heat released from OTTO thorium-cycle RDE spent fuel is comparable with that of multi-pass LEU-cycle spent fuel, despite the latter being decayed for only 40 days. Similar with the previous radioactivity case, larger heat release of the former is due to Pa-233, whose half-life is longer than Np-239, the precursor of Pu-239 exists in LEU-cycle, and thus vanished more slowly. Meanwhile, compared to multi-pass thorium-based fuel, OTTO thorium-based fuel has slightly higher heat release but still comparably similar.

Peculiar behaviour shown in U-232 daughter products discussed previously was analysed concerning its heat release. Decay heat calculation is performed further until 50 years of decay. For the first ten years, the increase is caused by U-232 daughter products, such as Tl-208, Po-212, Po-216,

Rn-220, Ra-224, and Th-228, build up and peaked.

After the tenth year, those radionuclides start to decay, but in slower rate than FPs such as krypton, rhodium, antimony, tellurium, cerium, and praseodymium isotopes. Thus, ACTs share still continue to increase. In the fiftieth year, ACTs share in heat release accounts for 29.96%. Nonetheless, such behaviour did not result in halting the decay heat decrease let alone crank up the heat release, so its effect on heat release can be ignored.

4. CONCLUSION

Thorium-cycle RDE spent fuel shows several distinct, potentially non-negligible, characteristics compared to LEU-cycle fuel. The first and possibly most important is the existence of Tl-208 in significantly higher amount, thanks to U-232 generation inherent to thorium fuel cycle. Tl-208 activity and subsequently gamma release peaked at the tenth year of decay and need to be addressed properly in the spent fuel management due to its strong gamma release. Its neutron emission is also

considerably lower due to the absence of transuranic elements, whilst its heat release during the first 60 days being higher due to the existence of Pa-233.

Compared to its multi-pass counterpart, OTTO thorium-based spent fuel has generally higher value in all of evaluated parameters, although it may also be influenced by the different fuel loading used. Future works should also calculate spent fuel radionuclide characteristics for its full EFPD estimation to provide necessary data for its spent fuel management.

5. ACKNOWLEDGEMENTS

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