
CHARACTERISING U-232 AND TL-208 BUILDUP AND DECAY ON THORIUM-FUELLED RGTT200KR. Andika Putra Dwijayanto^{1,2}¹ Department of Nuclear Engineering and Physics Engineering, Engineering Faculty, Universitas Gadjah Mada, Jl. Grafika No. 2, Yogyakarta 55281² Centre for Nuclear Reactor Technology and Safety, National Nuclear Energy Agency, Building No. 80 Puspiptek Area, Setu, South Tangerang 15314**ABSTRACT**

CHARACTERISING U-232 AND TL-208 BUILDUP AND DECAY ON THORIUM-FUELLED RGTT200K. Thallium-208 (Tl-208), a decay daughter of uranium-232 (U-232), is a strong 2.6 MeV gamma emitter present in significant amount in thorium fuel cycle. Its existence enhances the anti-proliferation characteristics of thorium fuel cycle, but at the same time complicates the fuel handling system. In order to ensure that radiation hazard is properly contained, the buildup and decay characteristics of both U-232 and Tl-208 need to be understood. This paper aimed to provide a characterisation on U-232 and Tl-208 buildup in the thorium-fuelled RGTT200K, a 200 MWt very high temperature reactor (VHTR) developed by BATAN, using ORIGEN2.1 depletion code. Pure and impure U-233 were used as the fissile nuclide for comparison. The result showed that U-232 buildup rate is faster in pure U-233, but its Tl-208 buildup is slower. Nonetheless, pure U-233 always has its U-232 and Tl-208 activity lower than impure U-233. Accordingly, both U-232 and Tl-208 radioactivity post-discharge in pure U-233 are lower than impure U-233, although the difference become somewhat negligible after 300 years of decay. Tl-208 activity peaked after 10 years of decay, necessitating different approach in managing post-discharge fuel management.

Keywords: RGTT200K, thorium, Tl-208, U-232, VHTR

ABSTRAK

MENGGARAKTERISASI BANGKITAN DAN LURUHAN U-232 AND TL-208 PADA RGTT200K BERBAHAN BAKAR THORIUM. Thallium-208 (Tl-208), anak luruh dari uranium-232 (U-232) merupakan pemancar gamma kuat 2.6 MeV yang terdapat secara signifikan pada daur bahan bakar thorium. Keterdapatannya menguatkan karakter anti-proliferasi daur thorium, tetapi juga membuat sistem penanganan bahan bakar jadi lebih rumit. Pada *high temperature gas-cooled reactor* (HTGR) tipe *pebble bed multi-pass*, potensi bahaya Tl-208 terdapat setidaknya pada sistem resirkulasi bahan bakar dan kanister bahan bakar bekas. Artikel ini ditunjukkan untuk mengkarakterisasi bangkitan dan luruhan U-232 dan Tl-208 pada RGTT200K, *very high temperature reactor* (VHTR) yang dikembangkan oleh BATAN berdaya 200 MWt berbahan bakar thorium dengan menggunakan perangkat lunak deplesi nuklida ORIGEN2.1. U-233 murni dan terkontaminasi digunakan sebagai perbandingan. Hasil simulasi menunjukkan bahwa laju bangkitan U-232 lebih cepat pada U-233 murni, tetapi bangkitan Tl-208 lebih lambat. Namun, U-233 murni selalu memiliki aktivitas U-232 dan Tl-208 lebih rendah daripada U-233 terkontaminasi. Demikian pula, radioaktivitas setelah bahan bakar keluar reaktor lebih tinggi pada U-233 terkontaminasi, walau perbedaannya menjadi tidak terlalu signifikan setelah 300 tahun peluruhan. Aktivitas Tl-208 memuncak setelah 10 tahun peluruhan, sehingga meniscayakan perlunya pendekatan berbeda dalam manajemen bahan bakar nuklir bekas.

Kata kunci: RGTT200K, thorium, Tl-208, U-232. VHTR

INTRODUCTION

Thorium fuel cycle is currently being pursued as an alternative to uranium fuel cycle. It offers several advantages, one of them being strong anti-proliferation characteristics. The main anti-proliferation component of thorium fuel cycle is the presence of U-232 in the fuel. It is formed during thorium irradiation inside a nuclear reactor, then decays successively into Tl-208. The latter is a strong 2.6 MeV gamma emitter; a few hundred ppm of its parent isotope U-232 is adequate to prevent the attempt of proliferating thorium fuel cycle [1, 2].

U-232 is practically ever-present in thorium fuel cycle, and its concentration increases to the burnup. The longer the nuclear fuel resides within a reactor core, more U-232 is built up and consequently causing higher gamma emission from Tl-208. Apart from being an effective proliferation barrier, U-232 existence poses another issue in fuel handling, especially after discharge. High-energy gamma emission complicates post-discharge fuel handling as it requires heavier shielding and remote operation. Thus, any reactor run in thorium fuel cycle must consider this in the radiation protection system [3–5].

Among the reactors that can employ thorium fuel cycle is RGTT200K [6]. This pebble bed very high temperature reactor (VHTR) was once studied using thorium fuel cycle. What has not been addressed in the

study, and most of researches concerning thorium fuel cycle, is the characteristics of U-232 and Tl-208 buildup during fuel burnup. The characterisation is crucial in the spent fuel management, as both U-232 and Tl-208 exist in significant amount only in thorium fuel cycle and thereby may require different treatment. Several studies have been done in term of characterisation of radionuclides in HTGR-type reactors [7–9], including thorium-fuelled ones [10, 11]. But none of them were directed specifically on U-232 and Tl-208.

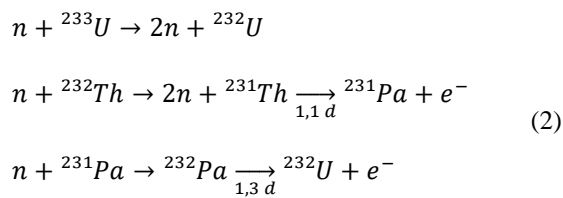
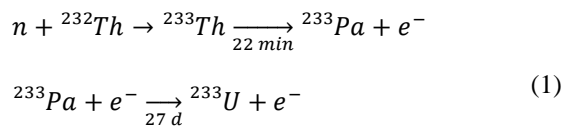
The buildup and decay characteristics of U-232 and Tl-208 in thorium-fuelled RGTT200K will be addressed in this article. The buildup and decay are simulated using ORIGEN2.1 computer code with HTGR-specific neutron cross section library. The characteristics are addressed both during irradiation inside the reactor core and after the fuel is discharged.

THEORY

RGTT200K is a very high temperature reactor (VHTR) designed by National Nuclear Energy Agency of Indonesia (Badan Tenaga Nuklir Nasional/BATAN) prior to the development of Experimental Power Reactor (Reaktor Daya Eksperimental/RDE) [12]. Unlike its successor, RGTT200K was designed to operate at temperature of 950°C with a thermal power of 200 MWt. The high temperature allowed RGTT200K for various cogeneration purposes, such as hydrogen

generation, coal gasification, and seawater desalination [13, 14]. Its design adopted pebble bed core with multi-pass fuel recirculation scheme. The pebble fuel consists of thousands of TRISO-encapsulated fuel kernels enclosed in a graphite sphere [15]. In this study, the fuel is in form of homogeneous (U,Th)O₂.

Main characteristics of thorium fuel cycle is the low transuranic elements generation and the presence of U-232. The latter is formed through two possible (n,2n) reactions [2].



U-232 buildup through Eq. 1 is more dominant at the beginning of cycle (BOC), whilst Eq. 2 has the highest share of buildup after Pa-231 is accumulated during irradiation [2]. The energy threshold of (n,2n) reaction is 6 MeV, thus the reaction depends on fast neutron flux [3, 16]. U-232 then decays with a half-time of 69 years, successively into Tl-208.

ORIGEN2.1 computer code is employed to calculate the buildup and decay of the aforementioned nuclides. It was developed to calculate the buildup and decay of various radionuclides for fuel composition analysis and its radiological characteristics.

ORIGEN2.1 works using Bateman Equation and one group-averaged neutron cross-section library, represented by Eq. 3 [7].

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

where X_i denotes nuclide i density, N is the nuclides number, l_{ij} is fraction of radioactive disintegration by other nuclide that leads to formation of i , j is iterations number from $j = 1$ to $j = N$, φ is position and energy averaged neutron flux, f_{ik} is fraction of neutron 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, and F_i is nuclide i continuous feed rate.

To perform buildup and decay calculation, ORIGEN2.1 requires a specific one group-averaged neutron cross section library. LIBRDE neutron cross section library is employed in this calculation. This library was verified for use in HTGR-type reactor [8, 9].

METHODOLOGY

Calculating the buildup and decay of Tl-208 and U-232 was performed using ORIGEN2.1 code with HTGR-specific cross section library. The limitation of ORIGEN2.1 is that it calculates buildup and decay of nuclides as a point reactor. It is therefore unsuitable to perform the calculation as a whole core. One pebble fuel is simulated

instead, using constant power as its depletion mode.

RGTT200K fuel parameters were adopted from Zuhair et al [6]. Its general parameters are shown in Table 1. Although the fuel used in the referenced study was using pure U-233, this calculation will simulate both pure and impure U-233 as a comparison. The latter has its U-233 fissile nuclide contaminated by other uranium isotopes. The isotopic vector of impure U-233 is adopted from Heuer et al [17], and provided in Table 2.

The RGTT200K reference did not provide the effective full power days (EFPD) of the chosen optimal fuel composition. Therefore, this study will adopt the fuel recirculation times from HTR-PM [18], of which has the closest power level. The fuel is recirculated for a total of 15 times with the EFPD of 1050 days.

Table 1. RGTT200K fuel parameters [6, 18]

Pebble thermal power (kW)	5.56
Fuel type	(U,Th) ₂ O ₂
U-233 enrichment	8%
Heavy metal loading (g)	6
Pebble mass (g)	200
Fuel loading scheme	Multi-pass
Fuel recirculation (times)	15
EFPD (days)	1050

Table 2. U-233 isotopic vector (%wt)

Isotope	Pure	Impure
U232	0	0.04
U233	100	85.99
U234	0	11.81
U235	0	1.68
U236	0	0.48
U238	0	0.00

RESULT AND DISCUSSIONS

U-232 buildup and decay

As implied from Table 2, the RGTT200K fuel with impure U-233 already contains U-232 in the fresh fuel. Theoretically, its buildup must be larger than then pure one. Although Fig. 1 proved that it was true, a certain pattern must be addressed.

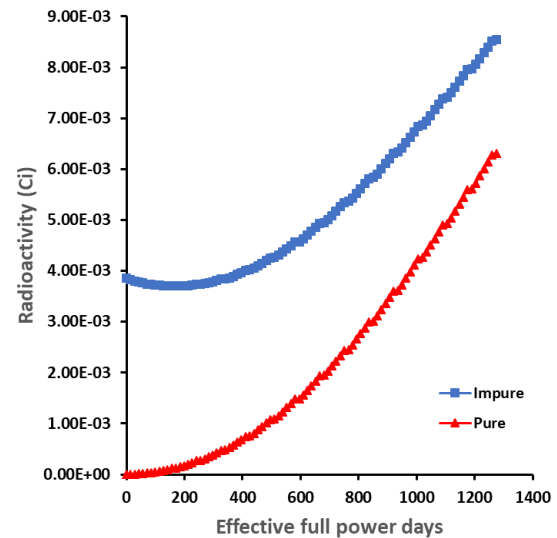


Fig. 1. U-232 buildup pattern

Whilst U-232 buildup in RGTT200K fuel with pure U-233 showed a regular increasing pattern, fuel with impure U-233 appeared to be different before day 200. Instead of increasing, U-232 radioactivity was slightly decreasing. Instead of accumulating even further, some of the preexisting U-232 were incinerated instead. This would mean that the U-232 buildup rate from thorium is lower than U-232 annihilation rate by neutron absorption at the beginning of irradiation.

The reason for this behaviour can be explained as follows. It was previously suggested that (n,2n) reaction that leads into

U-232 formation is predominantly through Eq. 1 at the beginning of irradiation. The key is the (n,2n) reaction in U-233. It offers shorter pathway to generate U-232, simply by one-step reaction. Meanwhile, Eq. 2 depended on Pa-231 as the main pathway to generate U-232, which took longer steps to form. As Pa-231 is non-existent at the BOC, it is clear that (n,2n) reaction in U-233 should dominate U-232 buildup. However, the (n,2n) cross section of U-233 is comparably smaller than thorium, resulting in lower U-232 formation through Eq. 1. Consequently, U-232 buildup through Eq. 1 can be lower than U-232 absorption and/or decay, resulting in its mass decrease and, accordingly, radioactivity decrease. Only after Pa-231 is sufficiently accumulated from (n,2n) reaction of thorium, and U-233 content is reduced enough to increase (n,2n) reaction from thorium, U-232 buildup exceeded its absorption and/or decay, resulted in increase of U-232 radioactivity.

Such behaviour caused U-232 buildup rate in impure U-232 to be lower than pure U-232, despite the radioactivity at the EOC is still higher. Fig. 1 suggested that the U-232 activity in pure U-233 is possible to exceed impure U-233 if the RGTT200K fuel is irradiated for longer time. However, such conclusion cannot be convincingly drawn just yet.

After irradiation, the fuel was discharged and let decay for 300 years. The decay of U-232 (see Fig. 2) is in accordance

to normal decay pattern. As of year 300 after decay, the difference in radioactivity was much less apparent than the initial difference. For longer decay time, it can be envisioned that the U-232 radioactivity will end up not too dissimilar to each other.

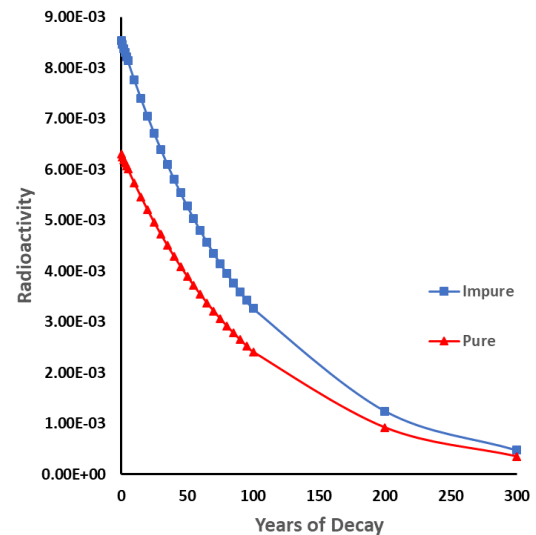


Fig. 2. U-232 decay pattern

Tl-208 Buildup and Decay

For Tl-208 calculation, it was assumed that its activity is zero at BOC for each type of fuel. Pre-existing Tl-208 that should be present within impure U-233 was ignored. This approach was used as there is no data or method to determine the initial Tl-208 activity in an impure U-233, especially for the composition vector used in this study.

Tl-208 buildup pattern can be seen in Fig. 3.

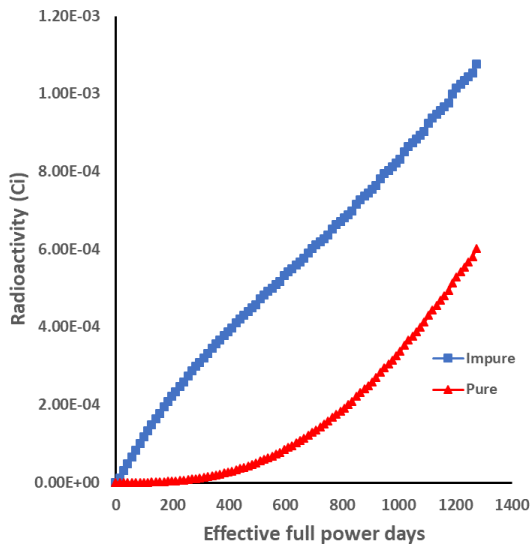


Fig. 3. Tl-208 buildup pattern

The buildup pattern was different between fuel with pure and impure U-233. The initially steeper buildup in impure U-233 was in line with the initial decrease of U-232 radioactivity, especially for the first 200 days of irradiation. It implied that most of U-232 were decayed into Tl-208 before sufficient Pa-231 was formed. As U-232 accumulated, its decay into Tl-208 slowed down due to competing nature of U-232 decay and neutron absorption, as the latter has decent fission and capture cross section in thermal spectrum. Tl-208 buildup got steeper once again nearing the EOC, as fission products build up and competed with U-232 for neutron capture, practically reducing U-232 absorption and increasing its decay.

Tl-208 buildup in RGTT200K fuel with pure U-233 behaved just like its parent isotope. The buildup was slower at the beginning and got steeper throughout the irradiation time. U-232 was not present at the BOC, and thus nothing to be decayed into Tl-

208. Similar to U-232, Tl-208 activity in pure U-233 can potentially exceed impure U-233 if the fuel was irradiated in even longer time. However, as the radioactivity difference of Tl-208 isotopes were larger than U-232 isotopes, the fuel may have reached its desired burnup before it happens. This would be even more unrealistic in real condition, as Tl-208 would have already existed in impure U-233, and thus the activity difference can actually be higher.

Tl-208 hazard is more apparent after the fuel is discharged from RGTT200K core after achieving targeted burnup. Thereby, the most important Tl-208 characteristic to be analysed is its decay pattern. As its gamma release was proportional to its activity, only Tl-208 radioactivity will be displayed here in Fig. 4.

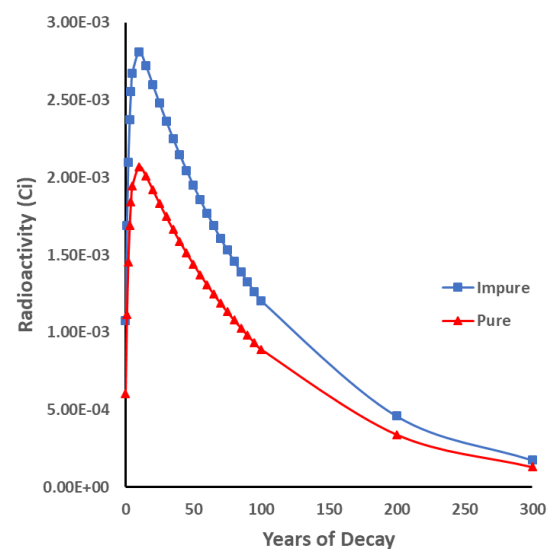


Fig. 4. Tl-208 decay pattern

The key issue in Tl-208 was its peculiar behaviour in the first 10 years of decay, of which its activity is increased instead of decreased. This behaviour was discussed in

previous publications [10, 11], and considered to be the main difference of spent fuel management in thorium fuel cycle. Understanding the nature of Tl-208 gamma emission, additional radiation protection mechanism must be enacted especially for the first 10 years or even beyond, as Tl-208 radioactivity decrease is much slower than its buildup.

Despite pure U-233 has lower Tl-208 activity at all times compared to impure U-233, for the first 10 years, the difference of radioactivity was decreased from around 44% to 26%. The difference remains at the year 300 after decay. This characteristic must also be taken into consideration in the radiation protection scheme of the thorium RGTT200K spent fuel.

CONCLUSIONS

The buildup and decay of U-232 and Tl-208 in thorium-fuelled RGTT200K showed some characteristics not already discussed in previous analyses. In term of absolute value, RGTT200K fuel with impure U-233 certainly resulted in higher U-232 and Tl-208 radioactivity at the EOC. However, U-232 buildup rate in fuel with pure U-233 is higher, showing larger difference between BOC and EOC radioactivity. The difference in Tl-208 radioactivity difference was also decreased after 10 years of decay. From this, it can be drawn that even fuel with pure U-233 can still pose significant radiation hazard after discharge, and thus must be treated

accordingly. Further works must translate the gamma release of Tl-208 after discharge to calculate the required shielding to protect the worker from Tl-208 hazard.

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