The Potential Role of the Thorium Fuel Cycle in Reducing the Radiotoxicity of Long-Lived Waste – 13477

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ABSTRACT

The thorium (or more accurately the Th-232/U-233) fuel cycle is attracting growing interest world wide and one reason for this is the reduced radiotoxicity of long-lived waste, with the Th-232/U-233 fuel cycle often being justified partly on the grounds of low radiotoxicity for long cooling times. This paper considers the evolution of heavy metal radiotoxicity in a Molten Salt Fast Reactor (MSFR) operating a closed Th-232/U-233 cycle during different operational phases. The paper shows that even in the MSFR core, the equilibrium radiotoxicity of the thorium fuel cycle is only reached after almost 100 years of operation. MSFR was chosen because it has many theoretical advantages that favour the Th-232/U-233 fuel cycle. Conventional solid fuel systems would be expected to behave similarly, but with even longer timescales and therefore the MSFR cycle can be used to define the limits of what is practically achievable. The results are used to argue the case that a fair approach to justifying the Th-232/U-233 breeder cycle should not quote the long term equilibrium radiotoxicity, but rather the somewhat less favourable radiotoxicity that could be achieved within the operational lifetime of the first generation of Th-232/U-233 breeder reactors.

INTRODUCTION

The thorium fuel cycle is attracting growing interest world wide and one reason for this is the reduced radiotoxicity of long-lived waste. Th-232 is a fertile nuclide that is converted to fissile U-233 on capturing a neutron. U-233 has a high fission cross-section and a relatively low neutron capture cross-section such that only very small quantities of heavier nuclides are produced. Once the fission products in spent fuel or vitrified high level waste (VHLW) have decayed, after a cooling time of about 500 years, the radiotoxicity falls well below that of the equivalent spent fuel or VHLW from the uranium fuel cycle. While the radiotoxicity of the fission products from the thorium and uranium fuel cycles are very similar, the radiotoxicity of the uranium fuel cycle at 500 years' cooling is dominated by plutonium and the higher actinides. The very low inventories of plutonium and higher actinides, which results from the combination of a starting point 5 atomic mass units lower and the small capture cross-section of U-233, is what distinguishes the thorium fuel cycle in terms of long term radiotoxic burden.

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There is no denying that in a self-sustained thorium fuel cycle with U-233 breeding, the radiotoxicity expressed in units of Sieverts per Giga-Watt-year (Sv/GWy) is very low compared with the uranium fuel cycle. The difficulty is that in practice the self-sustained condition takes a long time to establish. The fact that Th-232 is a fertile nuclide implies the thorium fuel cycle must initially be driven using neutrons produced by U-235 or Pu-239 fissions. Essentially, the fully self-sustaining Th-232/U-233 equilibrium fuel cycle can only be established after a long period during which it is driven towards equilibrium using neutrons from the uranium fuel cycle and these neutrons carry with them a higher radiotoxicity burden. A Th-232/U-233 breeder fuel cycle would pass through three operational phases:

- PHASE I: This is the *start-up phase* or *dependent phase* in which the first generation of reactors are maintained critical using an external supply of fissile material, which might be low enriched uranium (LEU), high enriched uranium (HEU), plutonium recycled from Light Water Reactors (LWRs) or a mix of plutonium and minor actinides. The start-up phase continues until such time as U-233 production within the system is sufficient to maintain criticality with no external fissile material. In practice, the start-up dependent phase typically lasts for many years, during which the system gradually reduces its dependence on the external fissile source.
- PHASE II: This is the *autonomous phase* that covers the period when the first generation of reactors become self-sufficient in U-233, requiring no external fissile material. The first generation reactors would continue to operate to the end of their lifetimes.
- PHASE III: This is the *equilibrium phase* where a new generation of reactors starts operation using the U-233 left over from the first generation of plants. The equilibrium phase can continue indefinitely with subsequent generations of new plants being built and operated as long as there is a requirement for nuclear energy.

In Phases II and III, if the system has a positive breeding gain (BG), the number of reactors in the system can expand if necessary, using the excess U-233 produced, but limited by the doubling time of the system.

The three phases of operation as defined here apply generally to all Th-232/U-233 breeder systems and they provide a useful frame of reference for further discussion.

All fission reactors generate a radiotoxicity legacy in the form of spent fuel or high level waste that is initially dominated by fission product activity for a few hundred years and then by the radiotoxicity of transuranics (Pu, Np, Am and Cm). Though the yields of fission products produced by U-233, U-235 and plutonium fissions are slightly different, the yields are nevertheless sufficiently similar that the fission product radiotoxicities do not vary to any great extent. Therefore the fission product radiotoxicity burden of the Th-232/U-233 fuel cycle is roughly the same irrespective of which of the above operational phases apply. After few hundred years, the fission products contributing to total radiotoxicity decay away, leaving the transuranics

as the main contributors. Figure 1 shows the time dependence of radiotoxicity for a Pressurised Water Reactor (PWR) for conventional LEU and Th/Pu fuel assemblies. This shows how the radiotoxicity (normalized to electrical energy production) of the Th/Pu fuel falls below that of the LEU fuel about 500 years after discharge and remains lower until the two curves cross just before 10^5 years, at which point in-growth of U-233 daughters dominates. It is this reduction in radiotoxicity at about 500 years that is considered one of the main benefits of the thorium fuel cycle. In this particular example, the benefit is very modest, because the PWR system is not capable of operating as a U-233 breeder and is reliant on an external fissile feed in the form of LEU, never progressing beyond Phase I.

Once a Th-232/U-233 fuel cycle is established in the autonomous or equilibrium operation (Phases II and III) its radiotoxicity burden after 500 years is very much lower because of the small inventory of transuranics. It is very common to see the radiotoxic burden of the Th-232/U-233 fuel cycle quoted for these two phases, without any account of Phase I, which is an unavoidable step prior to Phase II or III becoming established. Phase I will unavoidably generate an increased radiotoxicity burden and the purpose of this paper is to consider what impact Phase I has on the radiotoxicity of the thorium fuel cycle and whether it is reasonable and fair to focus on just Phase II or Phase III.
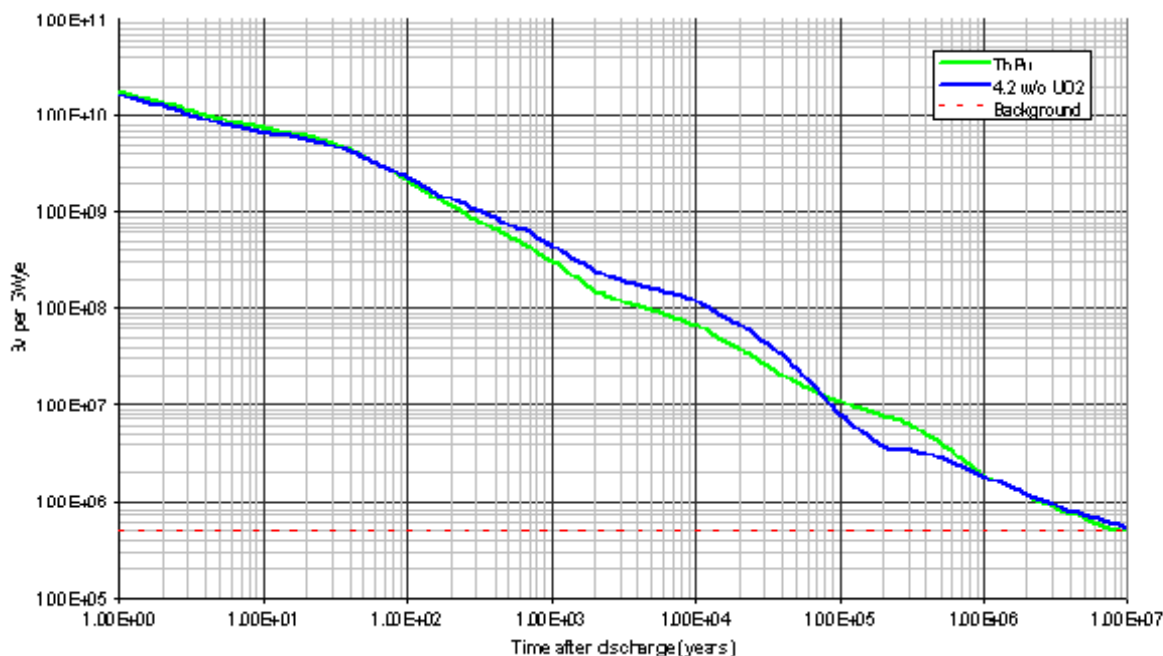

Figure 1: Radiotoxicity of Th-Pu fuel compared with UO₂ fuel irradiated in an LWR

The evolution of the radiotoxicity burden during the three phases is examined here in the context of the molten salt fast reactor (MSFR). The molten salt reactor was chosen because it has a number of theoretical advantages over conventional solid fuel reactors that will allow it to define the limits of what is possible in any system, limited only by the underlying neutron cross-sections in the fast neutron spectrum. Choosing a fast spectrum reactor also allows the best possible result in terms of burning up the residual transuranics remaining from Phase I.

DESCRIPTION

The MSFR core analysed in this study is based on a non-moderated thorium molten salt reactor (TMSR) described recently by French researchers [1, 2], with a thermal power output of 2.5 GW. The molten salt composition consists of either or $LiF-ThF_4-(Pu-MA)F_3$ (which is used in Phases I and II) and $LiF-ThF_4-UF_4$ (used in Phase III). Here MA denotes minor actinides - the Phase I and II external fissile source is assumed to be plutonium and minor actinides recycled from LWR spent fuel reprocessing. The reactor is unmoderated and therefore has a fast neutron spectrum and the driver core has a breeding gain close to zero. The reactor has a separate fertile blanket which increases the net breeding gain to 12%, but this study concentrates only on the performance of the driver core. The initial mass of the driver core is approximately 35 tHM (tonnes Heavy Metal), with an equilibrium U-233 loading of approximately 3.5 tHM.

The reference PUMA core starts up at the beginning of Phase I with an initial load of 14.4 tHM of plutonium and minor actinides (PUMA) taken from LWRs and 20.6 tHM of Th-232. The operating strategy assumes that fission products are removed continually, while all the heavy nuclides remain in the core. 1 tonne of Th-232 is added to the core every year to make up for the loss of heavy metal mass through fissions. The PUMA load provides the fissile material for Phase I operation and as U-233 builds up from Th-232 fertile captures, the U-233 gradually becomes the dominant fissile material in Phases II and III.

For comparison, a U-233 driver core was also modelled. At initial start-up this core contains 3.7 tHM of U-233 and 31.4 tHM of Th-232 and operates in the same way as the PUMA core. The U-233 core is representative of 2nd or later generation MSFR core that operates using U-233 produced in a preceding MSFR generation. The U-233 core provides an indication of radiotoxicities in Phase III (equilibrium) operation.

The two cores were modelled using the ECCO-ERANOS [3] nuclear design codes, using the R-Z geometry option in 33 neutron energy groups, with key resonance nuclides modelled in 1968 energy groups in the ECCO cell model. The cores were each depleted for 100 full power years, assuming periodic removal of fission products and Th-232 makeup. The ERANOS heavy metal inventories at 20, 50 and 100 years were transferred to the FISPIN inventory code [4] and decayed for cooling times from 1 year to 107 years, from which the radiotoxicities of heavy metals were calculated using the ICRP-72 effective dose coefficients [4].

A comparison of the ERANOS inventories with the equivalent inventories reported in [1] showed consistently close agreement. The two ERANOS models also show sensible behaviour of the core multiplication factor k-eff, which always remains quite close to 1.0, with minor deviations accounted for by the approximations made in modelling fission product removal and Th-232 makeup.

RESULTS

Figures 2 and 3 show normalized radiotoxicities in Sv/GWye for heavy metals only for the PUMA and U-233 cores respectively as a function of cooling time after the reactor has been shutdown. The radiotoxicities have been normalized by dividing by the integrated electrical energy output in GWye. The curves show the radiotoxicity burden that would be generated if the reactor was permanently shutdown after 20, 50 or 100 years. Normalizing to the integrated electrical energy output puts the three time snapshots on a comparable basis and gives the only meaningful comparison.

Superimposed on Figures 2 and 3 is the corresponding normalized radiotoxicity curve for a Pressurised Water Reactor (PWR) reactor fuelled with UO2 (LEU) with a mean discharge burnup of 45 GWd/t. This is representative of the current LWR fleet and provides a reference with which to compare the closed Th-232/U-233 fuel cycle.
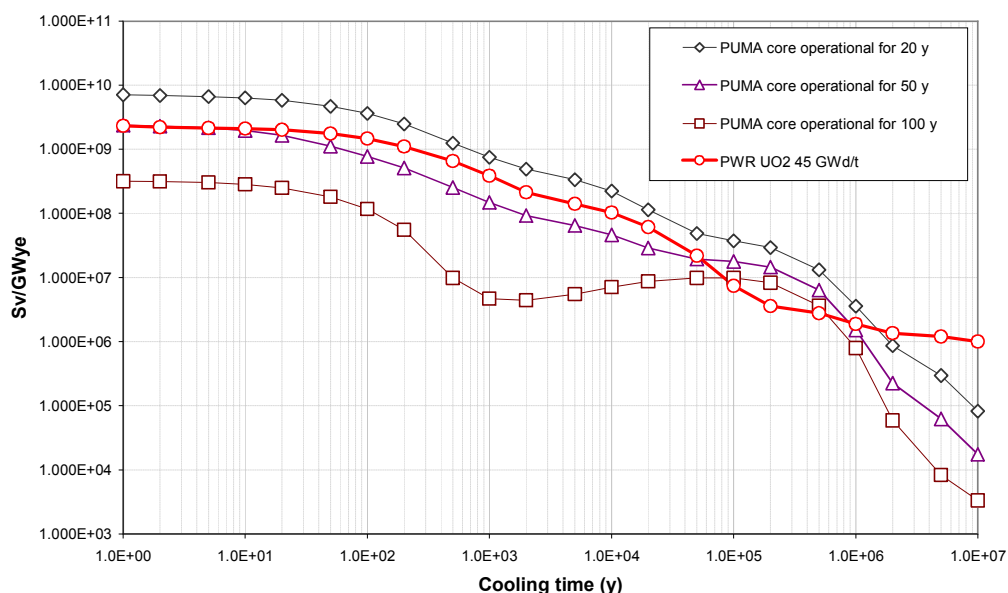

Figure 2: Heavy metal normalized radiotoxicity for the PUMA core

The PUMA core evolves between Phase I (start-up, reliant on external fissile feed) to Phase II (autonomous, self-sustaining fissile) over a period of 20 to 30 years, during which time the U-233 inventory builds up gradually towards its eventual equilibrium level. As the U-233 builds up, it provides an increasing fraction of the total fission power, until by 50 years it is very close to equilibrium. There is no sharp boundary between Phases I and II, but by 20 years U-233 is contributing to about 70% of total fissions, which seems a sensible point to assign the boundary. After 20 years of operation, there is still a significant proportion of the initial PUMA inventory remaining. The residual PUMA inventory still contributes to fission power and also makes a significant contribution to heavy metal radiotoxicity, as evident from the upper (grey) line in Figure 2. Indeed, this line is actually higher than the PWR UO_2 reference case, the reason being the residual PUMA inventory.

There is still a significant residual PUMA inventory after 50 years of operation, though in this case the radiotoxicity has fallen below that of the PWR UO_2 reference. In the 50 year case, although the contribution from the PUMA inventory to total fissions is very small, there nevertheless remains a significant inventory of transuranics with very small fission cross-sections that have yet to be fissioned and it is these which are dominating the heavy metal radiotoxicity.

After operating for 100 years, the heavy metal radiotoxicity is now finally down to the level of the equilibrium Th-232/U-233 fuel cycle, with the transuranic inventory now very low. The radiotoxicity profile in this case now shows the pronounced dip between 400 year and 10000 years that is characteristic of thorium cycles. The peak at 10^5 years is also characteristic of the thorium cycle and occurs due to the buildup of daughter products from U-233 decay (1.6×10^5 year half-life). At very long cooling times (10^7 years) the heavy metal radiotoxicity for the 100 y case is the much lower than the PWR UO_2 reference.
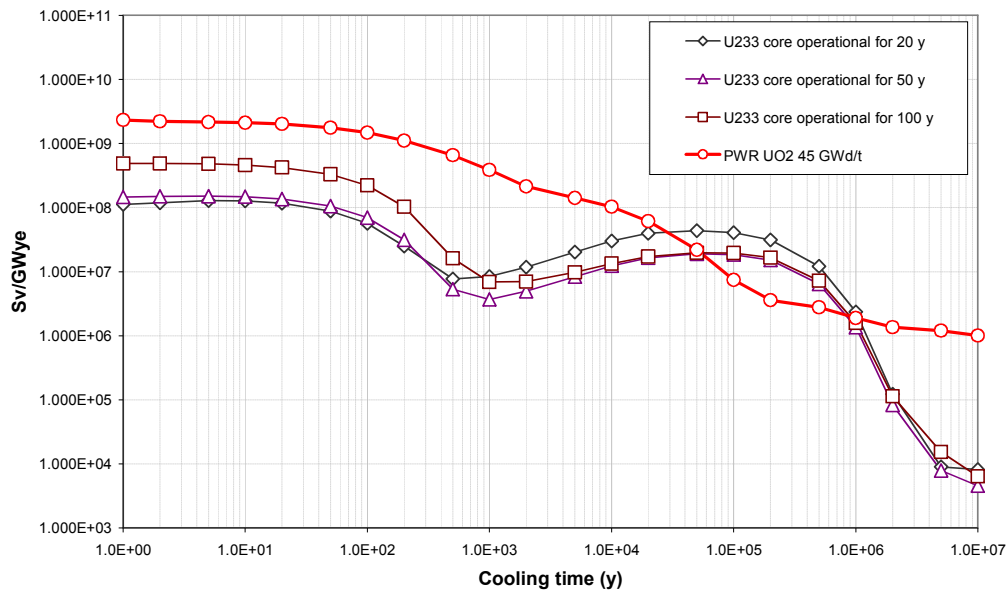
Figure 3: Heavy metal normalized radiotoxicity for the U-233 core

Figure 3 shows the corresponding radiotoxicity plot for the U-233 core, which is representative of the Phase III (long term equilibrium operation) . In this case the three cases are all quite close to each other and at cooling times 1000 years or greater, the 50 and 100 operational years U-233 cases coincide closely with the PUMA case for 100 years operation. This illustrates that the system will eventually converge to the same equilibrium, irrespective of the start-up core fissile composition. It is interesting to note that for short operating times the radiotoxicity of the U-233 core starts off very low, but builds up as the core continues to operate. This is because the transuranic inventory starts off at zero and takes a very long time to build up because of the large number of neutron captures needed.

DISCUSSION

This analysis illustrates how the heavy nuclide radiotoxicity profiles vary through the three different phases. By focusing on MSFR, the results are intended to define the best possible outcome for the thorium fuel cycle. There are three key features of MSFR that are relevant to this assertion: The first is that the system is capable of operating as a U-233 breeder, with no external fissile requirement. The second is that the recycle time is effectively zero in a molten salt reactor, which speeds up the rate at which the system is able to transition between Phases I and II and then to Phase III. Any conventional solid fuel system will be affected by very significant recycle times that will slow down these transitions. Finally, the MSFR has a fast neutron spectrum that

maximizes the burn-down of the transuranics from the initial PUMA core. Thermal spectrum Th-232/U-233 systems will be unable to achieve the same burn-down because of the less favorable fission/capture ratios.

Nevertheless, the results demonstrate that the transition from Phases I to II takes about 20 years and that the contribution of the PUMA initial inventory to radiotoxicity does not burn down completely until the system has been operational for approaching 100 years. This is very significant for the debate about the radiotoxicity of the Th-232/U-233 cycle, because it demonstrates that the true equilibrium position cannot be reached except on very long timescales of the order of 100 years. At this point the discussion ceases to be strictly technical and necessarily crosses over into societal issues.

The important point is that the equilibrium timescale is larger than the lifetimes of current generation reactors (many of the current generation of LWRs are expected to be given lifetime extensions to 60 years). Innovative new reactors cannot necessarily be assumed to be capable of such long lifetimes and the question is whether it is fair to quote radiotoxicities at the equilibrium level when it will very probably require a commitment to at least two reactor generations to achieve it. It would be fairer to claim only the radiotoxicity that can be achieved within a single reactor lifetime, which Figure 2 shows will be significantly less favorable. Stakeholders could be expected to seek approval for the deployment of a new reactor system with a lifetime of 30 years or more. But no stakeholder could reasonably be expected to commit to successive generations of new reactors, with timescales of possibly 100 years or more, in order to deliver the promised low radiotoxicities.

CONCLUSIONS

The results shows that even in the MSFR core, the equilibrium radiotoxicity of the thorium fuel cycle requires approaching 100 years of operation to attain. MSFR was chosen because it has many theoretical advantages that favour the Th-232/U-233 fuel cycle. Conventional solid fuel systems would be expected to behave qualitatively the same, but with even longer timescales and therefore the MSFR cycle can be used to define the limits of what is practically achievable. The results show the effects of the initial start-up fissile loading, in this case assumed to be a mix of plutonium and minor actinides (PUMA). Although the MSFR core becomes largely self-sustaining through U-233 breeding after 20 years of operation which corresponds to the transition between Phase I (start-up) to Phase II (autonomous), it takes nearly 100 years before the initial PUMA inventory burns down to the equilibrium level. This means that the very low radiotoxicity of the Th-232/U-233 fuel cycle cannot be achieved within the lifetime of the first generation of MSFR plants.

This result leads to an attempt to define some ground rules for assessing the radiotoxicity benefits in a realistic and fair manner. One obvious rule would be to limit the time horizon for initial justification to the design lifetime of the fist generation of thorium reactors. Taking credit from a more extended operational period, such as allowing for further generations of thorium reactors, involves stakeholders making a commitment beyond generations that cannot be reasonably justified. If is it possible to demonstrate a significant reduction in radiotoxicity within the design lifetime of the first generation of reactors, this should be the maximum benefit cited. Any additional benefits from subsequent generations of reactors should then be regarded as a bonus, but not used in the initial justification. With this approach, the paper has established a more realistic and defensible justification position for thorium as a low radiotoxicity option.

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