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Studies of MYRRHA using thorium fuel

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Abstract

The problem of long-lived radioactive waste has yet to find an acceptable solution, with political opposition to geological storage remaining strong. One possible solution which has often been discussed is to use ADSR systems for its incineration. The advantages of thorium as a fuel in this process are obvious, but few detailed numerical studies have been performed. We investigate the potential for using a thorium fuelled subcritical reactor for the incineration of long-lived minor actinide radioactive waste, based on the well-developed MYRRHA reactor design, operating in sub-critical mode. We examine the neutron fluxes and spectra in the reactor, particularly in the In-Pile Section (IPS) regions that would be appropriate for such transmutation, comparing the result from thorium fuel rods with those of the standard uranium/plutonium fuel. From this we present the burn-up rates that would be achieved, both initially and in the longer term as ²³³U is formed.

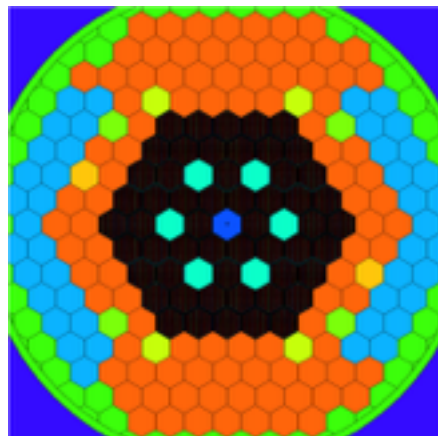
Keywords: ADSR, MA waste, thorium, transmutation

1. Introduction

The disposal of the long-lived Minor Actinide (MA) elements has yet to find an acceptable solution. Fission products in nuclear waste have half-lives such that their safe storage is not unthinkable, and U and Pu can be partitioned and recycled as fuel, but several isotopes of Am (americium) and Cm (curium), in particular, are more problematic. Given the difficulties, both practical and political, of geological disposal, transmutation by further exposure to neutrons in a reactor is an increasingly attractive possibility. Clearly this requires that the reactor fuel used in this incineration does not generate more MA waste than it destroys. One way of boosting the net consumption of MA nuclei is the use of fast neutrons, as the relative sizes of the (n,γ) and (n,f) cross sections means that above ~1 MeV a neutron is more likely to cause the MA nucleus to fission, whereas at lower energies it is far more likely to be absorbed. A second way is the use of thorium rather than uranium as the fertile/fissile fuel component: exact numbers depend on the cross sections and decay rates involved, but basically ²³²Th has six fewer nucleons than ²³⁸U, and the chance of a nucleus absorbing enough neutrons to get to an isotope like ²⁴¹Am is just much smaller.

Studies of the use of ADS thorium reactors for this have been done, especially by the Rubbia group[1] and the Aker/Jacobs ADTR design [2] though they are primarily concerned with power generation. More details can be found in the book by Nifenecker and colleagues [3] and a comprehensive comparison of ADS and fast critical reactors has been done by the OECD [4]. The main reason for performing similar studies with the MYRRHA system [5] is that this design has been developed in more detail than any other, and this refinement will continue as the system is constructed, in contrast to other ADSRs which, unfortunately, exist only on paper. A secondary motivation is the hope that MYRRHA may one day be used with thorium fuel, although this is not part of the current proposed programme.

Figure 1. Plan view of the MYRRHA geometry used, with cells of different types denoted by different colours, as explained in the text. Cells measure 10.45 cm between opposite faces.



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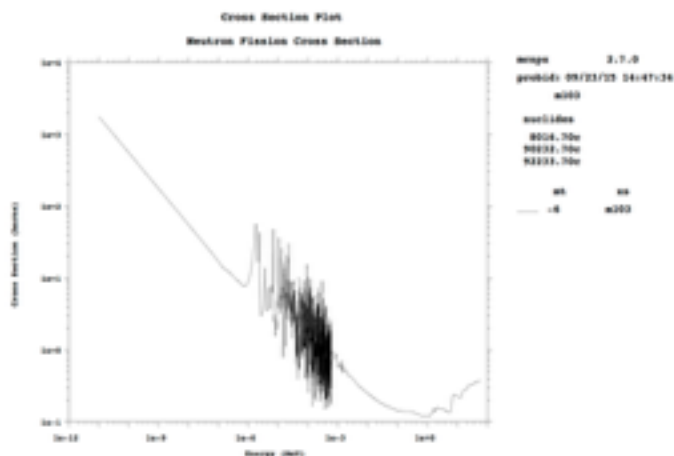
The geometry of MYRRHA used in this study is shown in plan view in Figure 1; it comprises a set of hexagonal cells with 10.45 cm between opposing faces. The central cell (deep blue) is for the beam and the target, which is a molten Lead Bismuth Eutectic (LBE). The ring of 6 cells around it (black) are all fuel cells, made out of pins containing the fissile and fertile fuel. In the next ring of 12 cells half are fuel cells and half (turquoise) are In Pile Sections (IPS) for materials testing. Two further rings of fuel cells, bringing the total to 54, complete the core. The next ring is mostly just LBE filled cells (brown) although 4 are control rods (yellow-green), in the out position for this configuration. The next ring is similar: LBE except for six cells for radioisotope production: two for Molybdenum (light brown) and four for Actinium (green). Later rings include beryllium-loaded reflectors (light blue) and stainless steel shielding (green). Vertically the rods are divided into three regions, with a central active part 65 cm in length. The geometry and the material composition [6] are described in considerable detail, and the file extends to some 2000 lines.

We considered three fuel mixtures

- The first, denoted U/Pu, is the standard MYRRHA MOX fuel mixture. It contains the 3 natural uranium isotopes, ^{234}U , ^{235}U and ^{238}U , a range of plutonium isotopes, and a small amount of americium.
- The second, denoted Th/Pu, has all the uranium replaced by ^{232}Th , representing a possible initial thorium fuel composition.
- The third, denoted Th/U, also has all the Pu and Am is replaced by ^{233}U . This is used to illustrate a final thorium fuel composition, though for exact calculations one would have to include Pa and other isotopes produced in the fertile to fissile conversion.

We use the MCNPX program [7], as it is a particle transport code well-established in nuclear simulations. The ENDF/B-VII cross section libraries [8] were used, as being the latest available. This did not let us incorporate cross sections at the anticipated running temperatures, and we used the standard 300K values. This is not expected to cause differences that are significant at the level of this study. The fission cross section for the third fuel mixture is shown in Figure 1. For the other two mixtures they are broadly the same, but differ in detail. Samples of 10,000 protons were used. Variation in the results of runs appeared compatible with the statistical errors (noted below as appropriate) so we do not believe that unexpected variations arise from the sharpness of these resonances, or any other source

Figure 2. Fission cross sections, as a function of neutron energy, for the Th/U fuel mixture.



For the new mixtures we replaced the old elements by an equal proportion of the new. However this affected the reactivity. This can be evaluated by a standard MCNPX calculation: for the original mix it was 0.95, which is a sensible level for a subcritical reactor. For mixture 2 it fell to 0.91. This loss of reactivity could be for a number of reasons, but the fact that ^{238}U has a larger (though small) fission cross section than ^{232}Th is probably responsible. For mixture 3 the reactivity rose to 1.17, in line with the superior fissile properties of ^{233}U . In order to compare like with like, and also to make the reactor subcritical, the proportions of fissile and nonfissile isotopes were adjusted to bring all three reactivities close to the same 0.95 level (actually 0.951, 0.940 and 0.959 for the 3 fuels types, all with an error 0.003).

*Table 1. Properties of the spallation+ fission process for the 3 fuels
Statistical errors are at the level of the final digit*

Fuel	Fission heat in MeV/g	Number of fissions/proton	Fission neutrons/proton	Neutrons/fission	Escape neutrons/proton
U/Pu	0.22	318	932	2.93	1.9
Th/Pu	0.19	262	774	2.95	1.6
Th/U	0.32	496	1256	2.53	2.5

Samples of 10,000 beam protons were used for simulations in this study. Some of the results are shown in Table 1. The fission heat figures shown are those given by MCNPX. The fuel in the 54 FA cells comprises 128 rods of radius 0.27 cm, 65 cm long, giving volume of 0.1 cubic metres, and a density of 10.5g/cm³, giving a total of 1,080 kg of fuel. For a beam current of 2.5 mA, the numbers in table 1 can be converted into the fission power in MW by multiplying by a factor of $1.08 \cdot 10^6 \times 2.5 \cdot 10^{-3} = 2.7 \cdot 10^3$.

2. Neutron spectra

Figure 3. Neutron spectra at various locations in the reactor (1) averaged over the fuel, (2) in the inner ‘IPS’ cells used for irradiation (3) for the outer ‘Molybdenum production’ cells and (4) for the ‘Actinium production’ cells. The three different fuel types are indicated by the colour, as described in the legend.

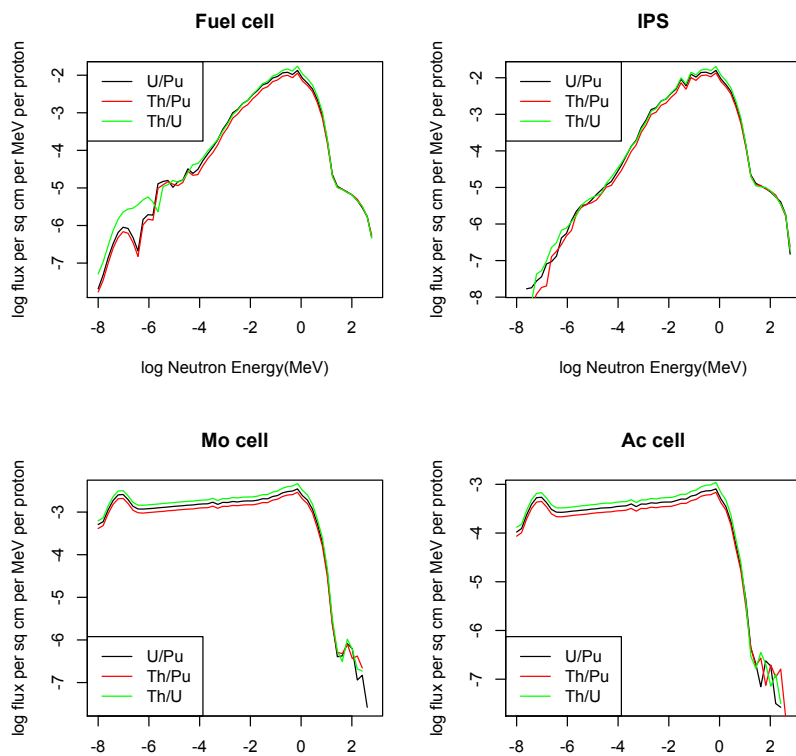
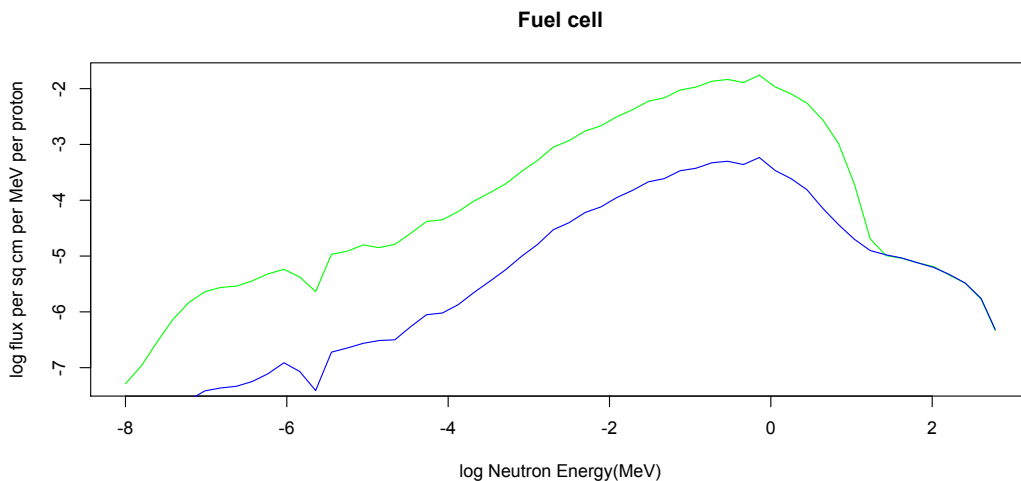


Figure 3 shows the neutron flux and spectrum, from simulation of 10000 protons. The first plot is averaged over all fuel cells. The three curves are shown for the three fuel mixtures and are broadly similar. Any differences between the first two must be ascribed to different absorption, as the fissile Pu isotopes are the same in both. (The presence or absence of the ²³⁵U made no discernible difference.) The spectrum peaks around 1 MeV but extends all the way down to thermal neutrons (~10⁻⁸ MeV). The flux in the six IPS, which are near the centre, is similar to the fuel average, but there are fewer slow/thermal neutrons, as shown in the second plot. By contrast in the outer irradiation cells, shown in the bottom two plots, the flux is both lower and less energetic, as more neutrons are produced near the centre, and they undergo more collisions in reaching the outer cells.

Figure 4. Neutron spectra in the fuel cells for type 3 fuel: the blue curve show the component due to sources other than fission. Roughly half are from spallation and half from (n,xn) reactions



The spectra in the fuel and IPS displays a large peak with a high energy shoulder above 10 MeV. It is tempting to ascribe the peak to the fission neutrons and the shoulder to spallation, but the true picture is more complicated. This is shown in Figure 4, which displays the fuel cell spectrum for fuel type 3, as before, together with the spectrum obtained when the simulation is run with no fission processes. This does indeed populate the high energy shoulder, but the low energy peak is still present: the size is smaller but the shape is the same. The similarities in shape show that the features are not statistical fluctuations but are due to the structure in the energy dependence of the cross sections.

3. Incineration of Minor Actinides

Table 2. Minor Actinide incineration integrated flux numbers
 Statistical errors are at the level of the final digit for the inner figures,
 for the outer figures they are larger, of order 10%

Isotope	Fuel	Fission inner	Absorption Inner	Ratio	Fission outer	Absorption Outer	Ratio
²³⁹ Pu	U/Pu	0.24	0.066	3.8	2.69	1.40	2.0
	Th/Pu	0.20	0.050	4.0	2.19	1.12	2.0
	Th/U	0.28	0.073	4.1	3.32	1.69	2.0
²⁴⁰ Pu	U/Pu	0.055	0.064	0.64	0.005	5.87	0
	Th/Pu	0.047	0.050	0.76	0.004	4.81	0
	Th/U	0.068	0.069	0.73	0.006	7.28	0
²⁴² Pu	U/Pu	0.039	0.086	0.59	0.002	0.88	0
	Th/Pu	0.033	0.062	0.64	0.002	0.71	0
	Th/U	0.049	0.094	0.69	0.003	1.08	0
²⁴¹ Am	U/Pu	0.039	0.24	0.16	0.018	2.85	0.01
	Th/Pu	0.033	0.19	0.17	0.015	2.32	0.01
	Th/U	0.048	0.26	0.19	0.023	3.51	0.01
²⁴³ Am	U/Pu	0.028	0.20	0.14	0.0029	1.36	0
	Th/Pu	0.024	0.16	0.15	0.0024	1.10	0
	Th/U	0.035	0.1	0.16	0.0037	1.66	0

The program computes the normalised integrated weighted flux, given by Equation 1,

$$F = \frac{1}{V} \int_0^{E_{max}} \phi(E)\sigma(E)dE \tag{Eq(1)}$$

for the fission and (n,gamma) absorption cross sections for a number of significant actinide species, for both the inner cells, using the IPS, and the outer cells, using the cells nominally used for actinium production. This was done for the three fuel mixtures described earlier. The MCNPX calculation makes no allowance for the MA elements in the cells, however these effects should be small compared to the levels of precision here. Results are shown in Table 2.

From the relative sizes of these results we may draw several conclusions:

- Fuel 3 generally shows a higher burn-up rate than fuel 1, but the differences are not large
- ²³⁹Pu is generally more likely to fission rather than absorb neutrons. This is true in the outer cells but even more so in the inner cells, where the neutron spectrum is harder. ²³⁹Pu is a special case, and can be regarded as a fuel rather than a waste product.
- For the other isotopes, the ratio of fission to absorption is respectable in the inner cells, but in the outer cells it is negligible. Exposing such isotopes in the IPS cells will transmute some fraction of them to short-lived fission products and some other fraction to a higher atomic weight by absorbing a neutron. This daughter product will in turn have a chance of fission or absorption, with the added possibilities of beta and alpha decay.

As to absolute sizes: MCNPX [7] computes the total neutron track length in the cell in cm, weights it by the appropriate cross section in barns, and divides by the volume in cm³. The numbers shown are thus in units of barn cm / cm³, and are probabilities, multiplied by 10²⁴. Thus, taking the number in the top left as an example, if there is a ²³⁹Pu nucleus anywhere in the IPS, and the fuel is mixture 1, the probability that it will be fissioned as a result of one proton striking the target is 0.24 x 10⁻²⁴.

To find the rate one multiplies by the number of protons per second, which for a 2.5 mA beam is 1.56 10¹⁶, giving a rate of 3.8 10⁻⁹ s⁻¹ or 0.12 y⁻¹. A conversion lifetime of 8y is promising, but the more interesting isotopes are down on this by an order of magnitude.

Figure 5 Incineration of ^{241}Am in the FPS with fuel mixture 2

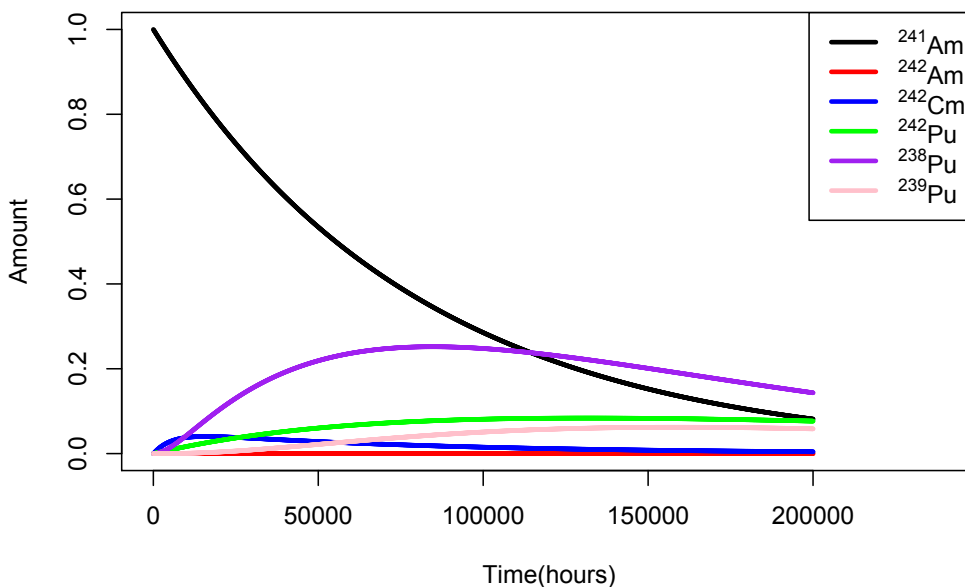


Figure 5 shows, as an example, the solution (using stepwise integration) of the Bateman equations for ^{241}Am in the IPS, for fuel mixture 2 over a 23 year period. A simplified form was used, in that the operation was taken as continuous, and the only processes considered were single-absorption, and, where relevant, decay. The ^{241}Am is consumed by absorption rather than fission, and the main product in the process is ^{238}Pu , from the successive decays of ^{242}Am and ^{242}Cm . Further isotopes (^{240}U , ^{240}Pu , ^{243}Pu , ^{243}Am and ^{243}Cm , with their daughter products) are also produced, but in small quantities.

What this suggests is that MYRRHA is not itself going to solve the long term waste problem - but this is not surprising as it is only meant as a demonstrator, a prototype for large scale systems operating at much higher currents. What the numbers in Table 2 indicate is that MYRRHA will be able to convert measurable amounts of waste isotopes, and prove the concept for an industrial transmutation system.

4. Evolution of the fuel

The processes we consider are conversions of the fertile fuel species through the (n,gamma) reactions to intermediate nuclei. For fuel 1 this is ^{238}U to ^{239}U , for fuels 2 and 3 it is ^{232}Th to ^{233}Th . In both cases this is followed by a rapid beta decay, to ^{239}Np and ^{233}Pa respectively. With half lives of 23 and 21 minutes these can be considered as instantaneous. They are then followed by a second beta decay. For fuel 1 this is ^{239}Np to ^{239}Pu , still relatively rapid with a half life of 2.4 days, but for fuels 2 and 3 it is the relatively slow ^{233}Pa to ^{233}U decay, with a half life of 27 days. If ^{233}Pa absorbs a neutron this takes it along a non-productive route, so the extent to which this occurs is a serious design consideration. This is a well known problem for thorium reactors.

*Table 2. Neutron absorption for isotopes in the fuel cells
Statistical errors are at the level of the final digit*

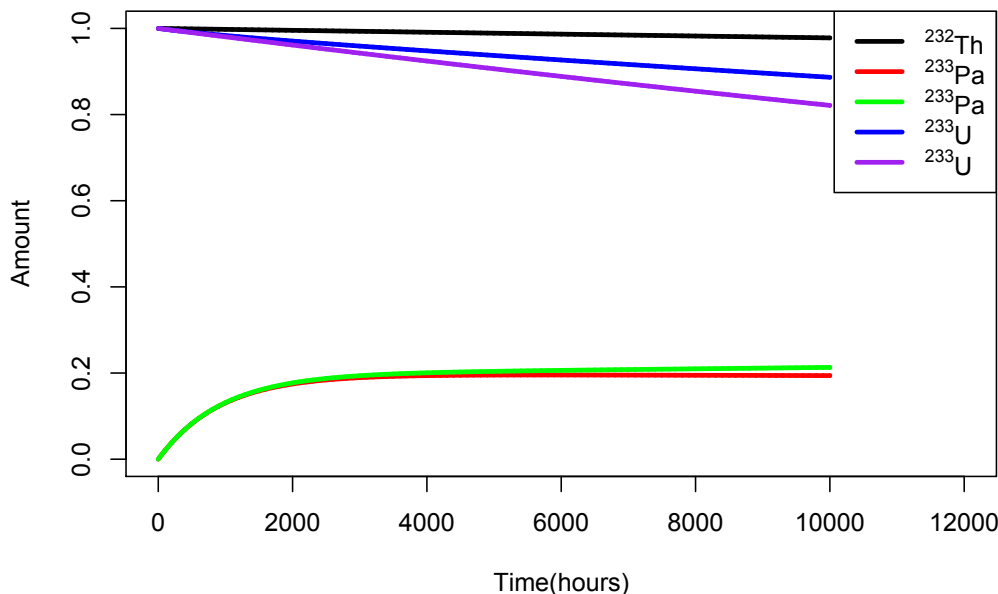
Fuel	^{238}U	^{232}Th	^{233}Pa
U/Pu	0.028	0.045	0.18
Th/Pu	0.036	0.028	0.15
Th/U	0.046	0.039	0.19

Table 2 shows the MCNPX integrated fluxes for the (n,gamma) process on the relevant isotopes. Again, this is in barn-cm/cm³, and the probability of a nucleus being converted as a result of a single beam proton is given by these numbers scaled by 10⁻²⁴. From the relative values we can conclude that there are some differences between conversion rates for the fuel mixtures: as the composition changes the probability of a fertile to fissile thorium conversion increases from 0.028, the same as standard ^{238}U conversion, to a higher value of 0.039. (The numbers for ^{232}Th in U/Pu, and for ^{238}U in Th/U, are suspect as the fuel component is not included in the simulation.)

Atom for atom, the probability of a Pa nucleus absorbing a neutron and being lost from the fuel chain is several times greater than the probability of a Th nucleus entering: this must be balanced by the fact that the fuel contains relatively little Pa at any time.

For a simple illustrative calculation we ignore the effect of the changes of the neutron flux with time as the composition changes. Figure 6 shows the change in composition for fuel 3, assuming continuous operation at 2.5 mA. The ^{233}U fuel is used up (purple curve) but this is partially replaced by the conversion from ^{232}Th via ^{233}Pa (blue). That this is only partial can be understood as the MYRRHA fuel is moderately enriched (~20%) so there is not enough conversion taking place, relative to the amount of fuel already present, for full replacement. The ^{233}Pa soon stabilises; the effect of neutron absorption in its intermediate state, which is a worry at high flux, is small.

Figure 6: Evolution of the fuel with time. The thorium and uranium are normalised to their original amount. The blue and purple curves show the development of the uranium with and without fertile-to-fissile conversion. The protactinium is normalised to the amount of uranium, and scaled by 100 for visibility. The red and green curves show the effect of including losses from neutron absorption by the Pa.



This is a simple model with only the most important reactions included. A full analysis, including the production of MA isotopes in the fuel, will be published later.

5. Conclusions

MCNPX can be used to analyse a thorium-fuelled MYRRHA system, and compute the neutron fluxes and the conversion probabilities for different isotopes in the fuel or in cells dedicated to irradiation. MYRRHA can be used for pilot studies of MA incineration schemes, on which industrial strength transmutation systems can be built.

Measurable quantities of MA waste can be transmuted in the IPS: enough for research though not enough in itself to make a significant impact. There is no simple dominant process and a fuller analysis of the absorption/decay reaction chains among the heavy nuclei will be needed.

Future studies will develop this detailed model of isotope production and destruction, and build a complete system of Bateman equations describing the evolution of the composition, particularly a study of the products produced by ^{232}Th nuclei which escape being transformed to ^{233}U and then fissioned, to verify that only small quantities of MAs are produced. These should include cross sections for more processes, and include the effects of changes in the neutron flux and spectra due to the actinides being irradiated, and the evolution of the fuel components. Furthermore, to avoid overdependence on one simulation code, the calculations will be repeated using the Geant4 program.

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