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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 ^{233}U is formed.
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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,gamma) 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 ^{232}Th has six fewer nucleons than ^{238}U, and the chance of a nucleus absorbing enough neutrons to get to an isotope like ^{241}Am 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

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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.

The geometry of MYRRHA used in this study is shown in plan view in Fig. 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) is 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.

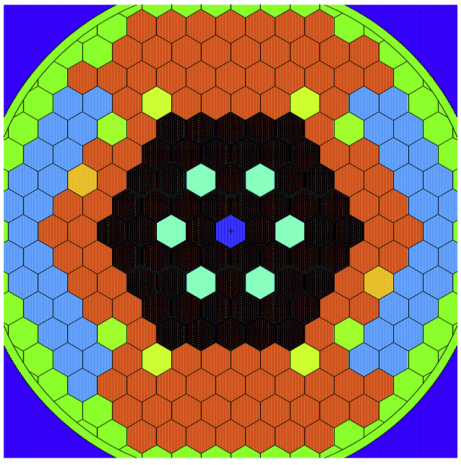

Fig. 1 – Plan view of the MYRRHA geometry used, with cells of different types denoted by different colours, as explained in the text. Cells measure 10.45 cm between opposite faces. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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 300 K 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 Fig. 2. 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.

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).

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 10.5 g/cm^3, giving a total of 1080 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 $1.08\ 10^6 \times 2.5\ 10^{-3} = 2.7\ 10^3$.

Neutron spectra

Fig. 3 shows the neutron flux and spectrum, from simulation of 10,000 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 ^{235}U 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

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