Potential of MYRRHA with Thorium fuel as an actinide burner

Roger Barlow *in place of*
Asiya Rummana,
Huddersfield University and Ibra College of Technology, Oman

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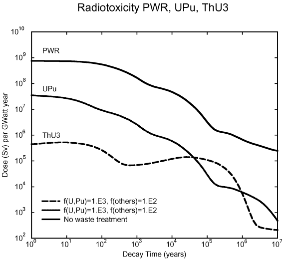
Contents

Based on Asiya Rummana's PhD Thesis

The problem of Minor Actinides

The isotopes of neptunium, americium and curium

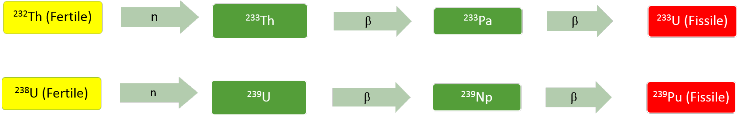
MA lifetimes of $\sim 100,000y$
Can be converted to short(er) lived fission products by exposure to fast neutrons
Strong motivator for ADSRs



Problem: Fast reactors generate more MAs due to n absorption on $^{238}\mathrm{U}$
Solution: Use of thorium rather than uranium as fuel
In studies consider $^{241}\mathrm{Am}$ as typical example of a problem MA.

The Thorium Cycle

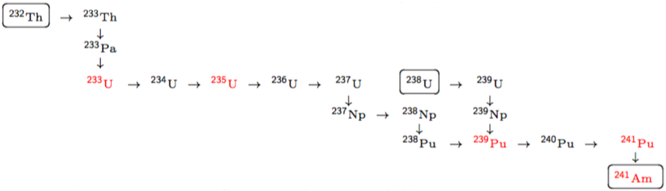
Thorium is essentially pure $^{232}\mathrm{Th}$.
Fertile not fissile - analogue of $^{238}\mathrm{U}$.



NB $^{233}\mathrm{Pa}$ half-life 27 days compare to 2.3 days days for $^{239}\mathrm{Np}$

Lower MA production for 2 reasons:

1) more steps needed



2) Fission:absorption cross section ratio ~ 10 for $^{233}\mathrm{U}$, ~ 2.5 for $^{239}\mathrm{Pu}$

MYRRHA+Thorium+Geant4

Interesting to study Thorium fuelled ADSR
Move from ideal reactor (spherical, uniform density) to realistic model
Use MYRRHA as a worked-out detailed design
(Details supplied - many thanks!)
Validate calcuations using two simulation programs

- MCNPX - the standard
- Geant4 - modern, flexible, originally for particle physics detectors

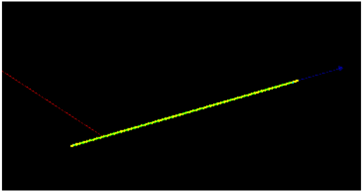
Consider 3 fuel mixtures (in oxide form)

1. Standard fuel mixture. U + MOX. 0.95 criticality
2. Replace U by Th. Represents initial Th fuel mix with fissile ‘starter’. Proportions adjusted for 0.95 criticality
3. Replace MOX by $^{233}\mathrm{U}$. Again adjust proportions for 0.95 criticality

MYRRHA geometry

Dimensions and composition provided by MCNPX deck[1] read at run-time.
Top-down structure using ‘universes’

Geant4 defines structure at compile time in bottom-up way.
Fuel pins, then Fuel Assemblies, then the reactor



[1] Shows the age of the program!

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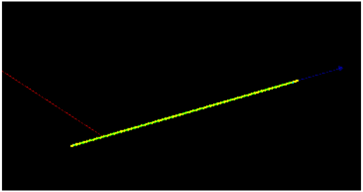

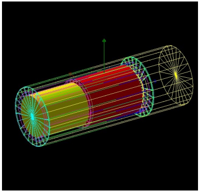

Fuel pin to scale, and in 'enlarged' view with x and y dimensions increased. Useful aid to visibility.

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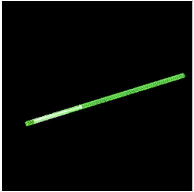
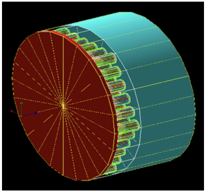
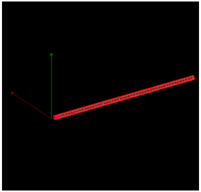
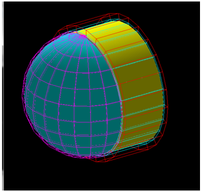
MYRRHA geometry - putting it together

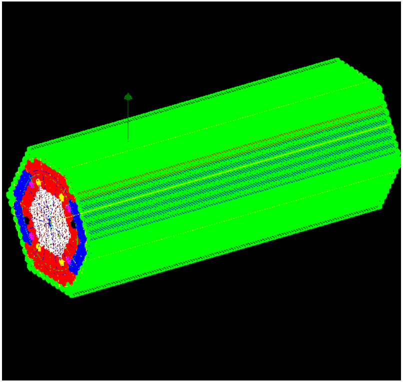



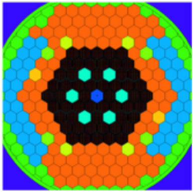
Fuel assembly made of 91 fuel pins - lots of function calls

Target unit in the centre is a one-off

MYRRHA geometry - putting it together

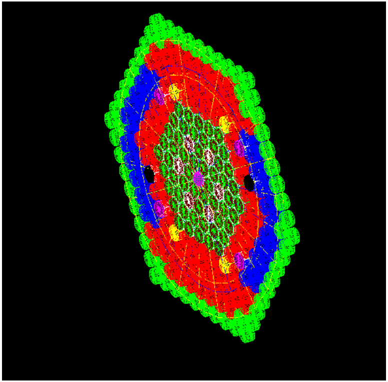


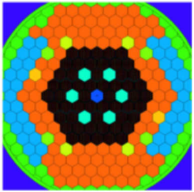


Compare MCNPX version

- Target
- Fuel Cells
- In Pile Section. (IPS) cells for high flux studies
- Control rods (not used in ADS mode)
- Mo and Ac cells: like IPS but lower flux, for medical isotopes
- Shielding and reflectors

MYRRHA geometry - putting it together





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Geant4 - changes to the code

Version Geant 4.10.1. 100K protons/run. Physics list QGSP_BIC_HP
Package only includes elements 1 - 92. Need to change cross section library from G4NDL to JEFF3.1 (using program of Mendoza et al)

Occasional crashes

```
Called G4PiNuclearCrossSection outside parametrization
***G4ElectroNuclearCrossSection::GetFunctions:
A=‘‘<<244.064<<"(?).  No CS returned!
```

Occurs in calculations for pions and muons, which could not handle isotope nuclei with $Z > 92$. $600 MeV > 289 MeV$ pion threshold
The relevant source code files: `G4KokoulinMuonNuclearXS.cc`, `G4KokoulinMuonNuclearXS.hh` , `G4PiNuclearCrossSection.cc` modified to use $Z = 92$ in their formulae for nuclei with $Z > 92$.

Approximation, but small effect: few targets ($\sim$ percent) and a very few particles (10K protons gave 9830K neutrons and only 438 charged pions) and EM interaction of a $\pi^{\pm}$ with Am not very different to U.

Results: Neutron Fluxes

Flux is readily given in MCNPX by a tally card
For Geant4, sum up total neutron track length using `SteppingAction`

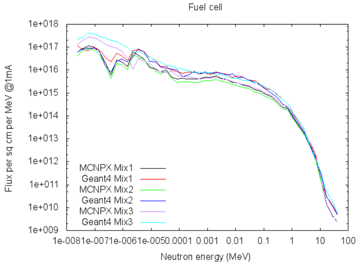
Location	Mix1		Mix2		Mix3	
	G4	MX	G4	MX	G4	MX
Fuel	6.28	7.08	6.37	5.80	8.62	8.41
IPS	5.19	8.48	8.76	6.98	8.34	9.95
Mo cell	2.8	4.8	5.07	3.91	6.94	6.04
Ac cell	1.17	1.09	0.93	0.90	1.98	1.37

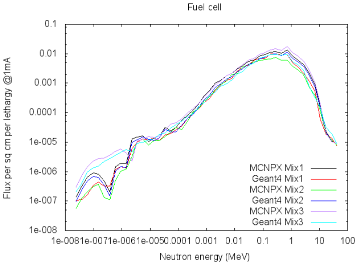
Table: Average flux values (units are 10^{14} neutrons/cm^2/s) for a 1 mA proton beam

Note 'Fuel' is averaged over all fuel elements

- Reasonable agreement between 2 codes.
- high flux in IPS, as expected
- lower flux in isotope cells - but Mo higher than Ac
- Mixture 3 has higher fluxes. More fissions, less absorption

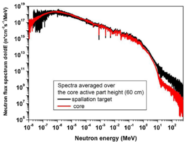
Energy Spectra
In the fuel

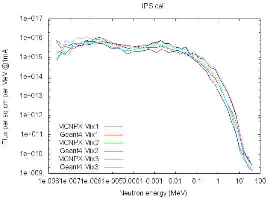


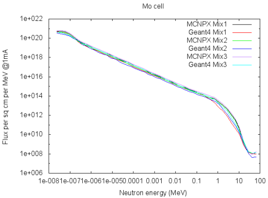


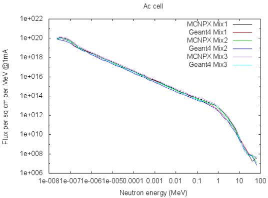
Average neutron flux in fuel, per unit energy(left) and lethargy (right)

Two programs agree well, though not perfectly
No sign of thermalisation
Some differences between fuel mixtures
Compares with simulations of Sarotto et al









Spectrum in IPS cells similar to fuel cells (not surprising)

Spectra in isotope cells significantly softer

Fuel mixes 1-3 similar

Programs consistent

Using the spectra

Consider the three dominant processes

- Neutron absorption increases A by 1
  Convolute neutron spectrum with cross section to find absorption rate
- Beta decay increases Z by 1. Lifetime λ from data tabes
  Decay of short-lived isotopes (e.g. $^{233}\mathrm{Th}$) considered as instantaneous
- Fission removes the nucleus

Bateman Equations

$\frac{dN_{A,Z}}{dt} = (-\lambda_{A,Z} - Q_{A,Z} - F_{A,Z})N_{A,Z} + \lambda_{A,Z-1}N_{A,Z-1} + Q_{A-1,Z}N_{A-1,Z}$

F, Q factors accessible in MCNPX using F4 Tally cards

(Target nucleus need not actually be in simulation, so approximation)

For Geant4, spectrum has to be histogrammed and then convoluted by hand with cross section, interpolating when the binning didn't match

Solve Bateman equations exactly using eigenvectors, and also numerically as a check. (Does not consider changes in spectrum with composition (would be possible), or fission products (would be very hard))

Input Numbers for Bateman Equations

Isotope	Fuel	Inner IPS					
		Fission MCNPX	Fission GEANT4	Absorption MCNPX	Absorption GEANT4	Ratio MX	Ratio G4
$^{239}\mathrm{Pu}$	U/Pu	0.23	0.12	0.04	0.02	5.57	4.92
	Th/Pu	0.19	0.24	0.03	0.04	5.82	5.64
	Th/U	0.27	0.16	0.05	0.03	6.09	4.73
$^{240}\mathrm{Pu}$	U/Pu	0.06	0.02	0.04	0.02	1.49	1.00
	Th/Pu	0.06	0.07	0.03	0.05	1.62	1.43
	Th/U	0.06	0.04	0.05	0.03	1.15	1.19
$^{242}\mathrm{Pu}$	U/Pu	0.05	0.02	0.05	0.03	0.98	0.62
	Th/Pu	0.04	0.05	0.04	0.05	1.02	1.02
	Th/U	0.06	0.02	0.06	0.04	1.15	0.59
$^{241}\mathrm{Am}$	U/Pu	0.05	0.02	0.21	0.12	0.24	0.15
	Th/Pu	0.04	0.05	0.16	0.19	0.26	0.23
	Th/U	0.06	0.02	0.23	0.16	0.28	0.14
$^{243}\mathrm{Am}$	U/Pu	0.04	0.01	0.20	0.12	0.18	0.11
	Th/Pu	0.03	0.03	0.15	0.18	0.20	0.17
	Th/U	0.05	0.02	0.21	0.15	0.21	0.10

Units are probability for a particular nucleus in the volume to undergo that process for one incident proton, times 10^{24}

Thorium Fuel Evolution

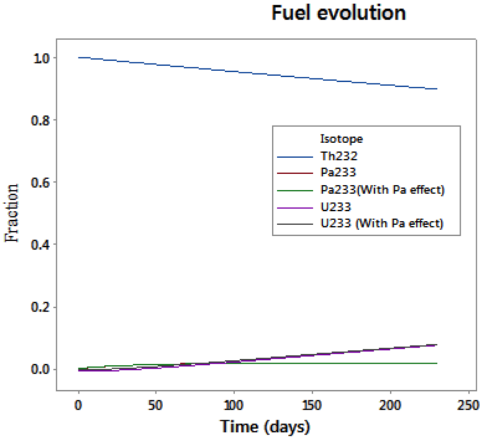
Showing that the Protactinium effect is negligible

Fertile thorium becomes fissile uranium

1 mA beam,

Geant4 numbers.

Fissioning of $^{233}\mathrm{U}$ not included



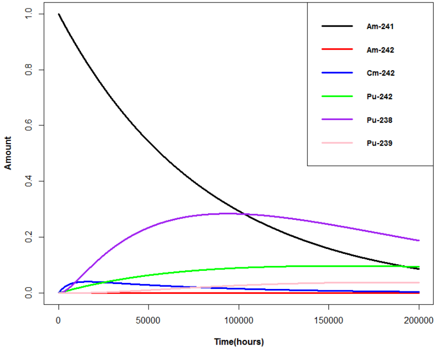
Intermediate $^{233}\mathrm{Pa}$ has half-life 27 days and has a chance of absorbing a neutron before it decays. Including this makes very little difference

Isotope incineration

$^{241}\mathrm{Am}$ consumed.

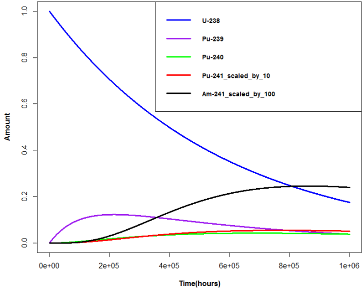
IPS cells,

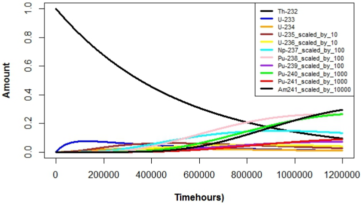
Geant4 numbers



Converted to fission products and relatively benign $^{238}\mathrm{Pu}$.
Rate too slow to be economically useful (100,000 hours = 11 years), but will validate deployment of larger ADSRs

Further MA production





MA production from $^{238}\mathrm{U}$ (left) and $^{232}\mathrm{Th}$ (right)
1 mA beam. Geant 4 numbers (MCNPX similar)

$^{238}\mathrm{U}$ gives some $^{241}\mathrm{Am}$ (note scale factor of 100)
$^{232}\mathrm{Th}$ gives much less $^{241}\mathrm{Am}$ (note scale factor of 10,000)
Timescales unrealistic for MYRRHA - indicative for high current ADSRs

Conclusions

- We have a Geant4 model of the MYRRHA geometry
- We have made the necessary modifications to the program to handle transuranic materials and beam energies above the pion production threshold
- Thorium fuel mixtures give similar neutron fluxes and spectra to uranium ones, assuming reactivity the same
- Placing samples of MA such as $^{241}\mathrm{Am}$ in the IPS cells will give measurements to justify future incineration facilities
- MA production from thorium fuel will be very small