OPTIMISATION STUDIES OF ACCELERATOR DRIVEN FERTILE TO FISSILE CONVERSION RATES IN THORIUM FUEL CYCLE

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Abstract

The need for proliferation-resistance, longer fuel cycles, higher burn up and improved waste form characteristics has led to a renewed worldwide interest in thorium-based fuels and fuel cycles. In this paper the GEANT4 Monte Carlo code has been used to simulate the Thorium-Uranium fuel cycle. The accelerator driven fertile to fissile conversion rates have been calculated for various geometries. Several new classes have been added by the authors to the GEANT4 simulation code, an extension which allows the state-of-the-art code to be used for the first time for nuclear reactor criticality calculations.

INTRODUCTION

Worldwide energy use will increase in the next couple of years and sustainable energy sources may not be able to meet energy needs. Nuclear fission could potentially provide a significant fraction of the world's energy as fuel is readily available and waste is very small and not technically difficult to handle. Realizing the potential of fission energy will require high-conversion reactors and the recycling of fissionable atoms, which in turn will require some technical challenges to be met.

The initial enthusiasm on thorium fuels and fuel cycles was not sustained, due to new discovery of uranium deposits and their improved availability. However, in recent times, the need for proliferation-resistance, longer fuel cycles, higher burn up, improved waste form characteristics and reduction of plutonium inventories has led to renewed interest in thorium-based fuels and fuel cycles [1]. Thorium is three to four times more abundant than uranium, and thorium fuels complement uranium fuels to ensure long term sustainability of nuclear power. Thorium exists in nature in a single isotopic form ^{232}Th which decays very slowly.

Thorium 232 can be used as a nuclear fuel, the fuel cycle being an attractive way to produce long term nuclear energy with low radiotoxicity waste as a much less quantity of plutonium and long-lived actinides are formed compared to the ^{238}U-^{239}Pu fuel cycle. In the Th-U cycle, the ^{232}Th is converted by neutron capture to thermally fissile ^{233}U. The conversion of ^{232}Th to ^{233}U is a three-step process involving neutron capture by ^{232}Th to produce ^{233}Th which decays to ^{233}U via two successive β decays.

THE GEANT4 SIMULATION CODE

In the present work, the GEANT4 simulation code [2] has been used to simulate the neutron interaction inside the Thorium fuel rods and the fertile to fissile conversion processes. GEANT4 provides an extensive set of hadronic physics models, both for the intra-nuclear cascade region and for modelling of evaporation.

One of the best models available is the Liege intra-nuclear cascade model coupled with the independent evaporation/fission code ABLA, which has been validated against experimental data for spallation processes in many different heavy elements [3]. The Liege model is largely free of parameters and is preferred by validation, and it is more data driven. However this model does not include pre-equilibrium: the INCL cascade is directly "coupled" to equilibrium de-excitation handled by ABLA and therefore it does not describe well enough low energy reactions (where nuclear structure effects start to play their role). INCL/ABLA works very nicely only above 100 MeV, being one of one of the best models available.

On the other hand, the other two models available in GEANT4, Bertini and Binary cascade, do incorporate the pre-equilibrium model. The Binary cascade model has been recently improved following a validation study against the TARC experiment data, in order to improve several shortcomings in applying these models to neutron spallation processes in heavy metals [4]. All these recent developments have been considered and implemented in our code.

In the simulations presented in this paper, the improved Binary Cascade model was selected. For neutron energies below 20 MeV, the high-precision models were selected. These models use the ENDF/B-VI(VII), JEFF and JENDL neutron data libraries. The S(α, β) coefficient which takes into corrected treatment for neutron scattering on chemically bound elements in the thermal region has also been implemented in the GEANT4 physics list used for this study.

The default fission model in GEANT4 does not describe accurately the spontaneous fission processes, it describes well only the neutron induced fission. However, since the GEANT4 release 4.9.0, a new module for Livermore LLNL neutron-induced and spontaneous fission model is available in GEANT4. This new model was used in all simulations.

Three new classes have been written and added to GEANT4: G4SDTimeFilter, G4SDParticleWithTimeFilter and G4SDParticleWithVolumeFilter [5]. These new classes allow the user to simulate the time evolution of the number of different isotopes present inside the nuclear fuel for any input parameters: the proton beam size and energy, the fuel composition and finally the target size and material.
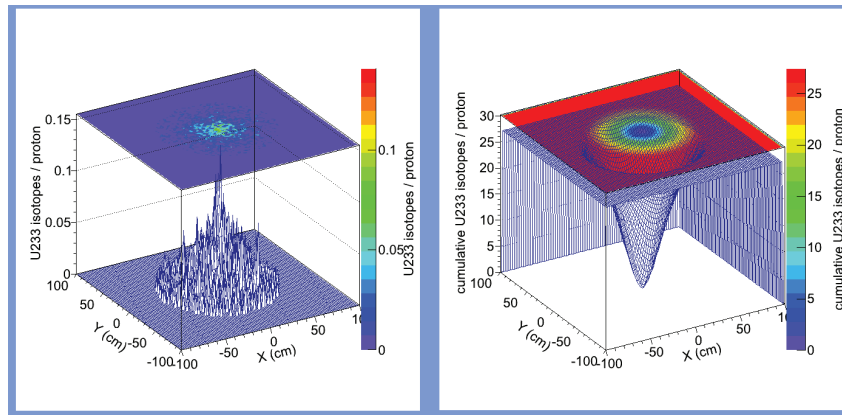
Figure 1: The distribution of ^{233}U inside the transverse X-Y plane normalized per incident proton.
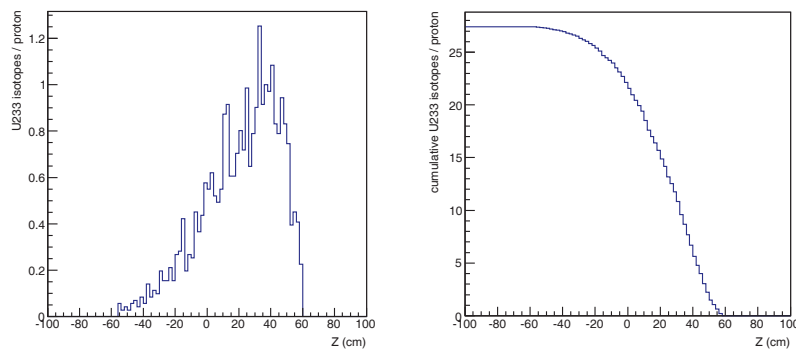

Figure 2: The distribution of ^{233}U along the longitudinal Z-axis normalized per incident proton.

THORIUM CONVERSION RATE FOR DIFFERENT FUEL RODS GEOMETRIES

Starting with an initial fuel composition the instantaneously production rates of individual isotopes are calculated and then these rates are used to predict the fuel composition after a given period of time which has to be short enough to ensure that the rates remain constant. And then the impact of the proton beam on the new reactor fuel is simulated. Different geometries were simulated in order to determine the optimum radius and height for the "pure" ^{232}Th fuel rod with respect to the ^{233}U yield.

Previous results [6] suggested a ^{232}Th to ^{233}U conversion rate of up to 1 conversion per incident proton. But those simulations were done before the fix of the bug found by the authors in the GEANT4 neutron HP capture model [7].

The overall thorium conversion rate turned out to be a strong function of the target radius. Table 1 shows the number of ^{232}Th to ^{233}Th conversions per incident proton, for 120 cm long Th targets and 1 GeV protons. It can be seen that a careful optimization of the fuel rods geometry can result in thorium conversion rates as high as 27 conversions per each incident proton.

For a target radius of 60 cm, the spatial distributions of the ^{233}U isotopes produced are shown in Fig. 1 for the

Table 1: Number of ^{232}Th to ^{233}U Conversions Per Incident 1 GeV Proton

Target radius	^{232}Th - ^{233}U conversions/proton
1 cm	0.1 ± 0.0004
10 cm	5.7 ± 0.03
20 cm	13.2 ± 0.05
30 cm	18.8 ± 0.06
40 cm	22.6 ± 0.06
50 cm	24.8 ± 0.06
60 cm	27.3 ± 0.07

transverse X-Y plane and Fig. 2 along the Z-axis.

Previous results [5] indicated that in order for a subcritical reactor to be close to criticality the level of ^{233}U has to be ~ 1.8 %. The diameter of the fuel rods was set to 1 cm. The fuel rods have been implemented into the GEANT4 simulation, being surrounded by a heavy water moderator, as shown in Fig. 4. Starting with pure ^{232}ThO$_2$ rods, we simulated the fuel evolution during the exposure to a beam of 1 GeV protons with an intensity of 1 mA for a period of 300 days.

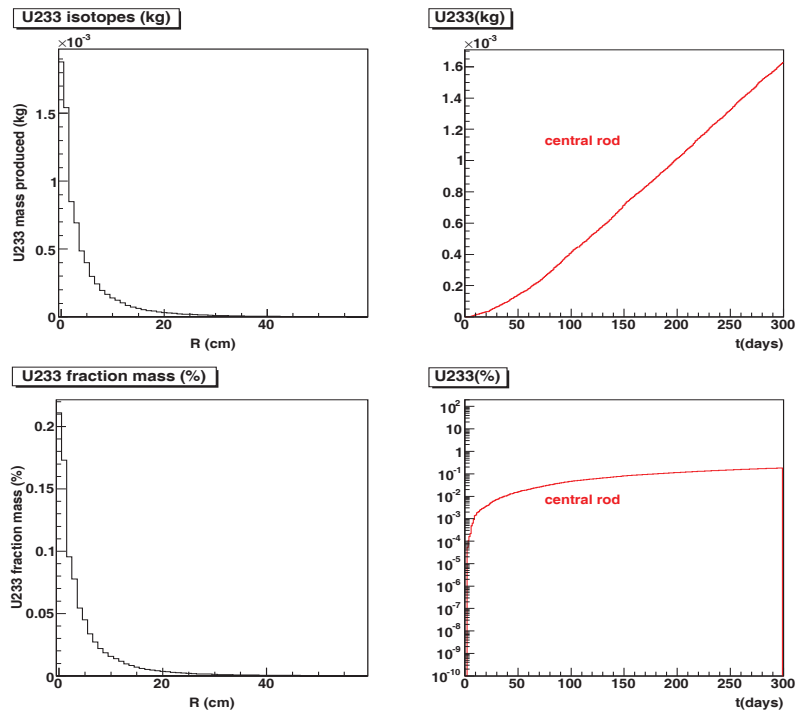The yield of ^{233}U has been averaged over all the fuel

Figure 3: The production of ^{233}U following a 300 days exposure of an assembly of 1 cm diameter fuel rods to the 1 GeV proton beam.
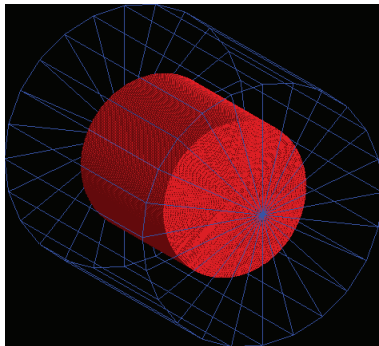

Figure 4: GEANT4 simulation of an assembly of 1 cm diameter fuel rods inside a D_2O moderator.

rods at equal radius and it is shown in Fig. 3 for all the rods at a given radius, after the 300 days of exposure to the beam. Figure 3 also shows the time evolution of the ^{233}U concentrations inside the central rod over the same period.

CONCLUSION

The time evolution of the number of ^{233}U isotopes following the continuous exposure to a proton beam has been simulated for different target geometries. The ^{232}Th to ^{233}U conversion rate increases with the target radius, for 60 cm reaching the value of 27 conversions per incident proton. The fraction mass of ^{233}U reached after 300 days of continuous exposure to a 1 mA, 1 GeV proton beam, increased up to 0.2%.

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