GEANT4 STUDIES OF THE THORIUM FUEL CYCLE

Cristian Bungau, Roger Barlow, University of Manchester, UK
Adriana Bungau, Robert Cywinski, University of Huddersfield, UK

Abstract

Thorium fuel has been proposed as an alternative to uranium fuel in nuclear reactors. New GEANT4 developments allow the Monte Carlo code to be used for the first time in order to simulate the time evolution of the concentration of isotopes present in the Thorium fuel cycle. A full study is performed in order to optimise the production of Uranium-233 starting with "pure" Thorium fuels, leading to levels of Uranium-233 which ensure the operation of the nuclear reactor in a regime close to criticality.

COMPUTATION DETAILS

GEANT4 provides an extensive set of hadronic physics models, both for the intra-nuclear cascade region and for modelling of evaporation. There are many different (data based, parametrized and theory-driven) models using different approximations and each has its own applicable energy range. The Liege intra-nuclear cascade model was selected together with the independent evaporation/fission code ABLA. This model has been validated against experimental data for spallation processes in many different heavy elements [1]. The Liege model is largely free of parameters and is preferred by validation and, compared to the other theoretical models available in GEANT4, 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 [2]. All these recent developments have been considered and implemented in our code.

In the simulations presented in this paper, the Liege model was selected to simulate interactions for energies above 150 MeV, while for lower energies the 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 $S(\alpha, \beta)$ 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 [3]. 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.

METHODOLOGY

Due to limited computational power available, no Monte Carlo code can simulate the continuous time evolution of the isotopes involved in the ^{232}Th - ^{233}U fuel cycle. Instead, the standard procedure is to simulate the evolution of the fuel cycle in discrete time steps. 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 time step which has to be short enough to ensure that the rates remain constant. At the end of the selected time step, the impact of the proton beam on the new reactor fuel is simulated and the procedure described above is repeated.

However after each time step in the simulation all the isotopes are initialised at the beginning of their lifetime. Since it is not possible to generate isotopes at different stages during their lifetime, a different approach was proposed.

Instead of running the simulation in time steps adding up to several days, the new approach is to run the simulation in two stages. First the fuel rod is exposed to the proton beam for 1 day. In the simulation one proton is generated every 5 minutes for 24 hours and the time evolution of each present isotope is recorded. The simulation now enters the second stage. The proton beam is switched off and the time evolution of each isotope continues to be recorded. The ^{233}Th produced by neutron capture on ^{232}Th decays into ^{233}Pa, while the ^{233}U will be continuously produced by the decay of ^{233}Pa. After $\sim$ 100 days there will be almost no more ^{233}Pa left. In order to calculate the new fuel composition, the results have to be scaled up to the equivalent of an initial exposure for 1 day to a proton beam with an intensity of 1 mA. The same procedure is then repeated for the new fuel composition, until the fraction mass of ^{233}U reaches the required value.

A proton beam with an energy of 1 GeV and 1 mA intensity was used in all simulations.

RESULTS

Previous results [3] indicated that in order for the subcritical (slow-neutrons) reactor to be close to criticality the level of ^{233}U has to be ~ 1.8 %.

The time evolution of the fraction masses of ^{233}Pa and ^{233}U inside the fuel rod after a continuous exposure for one day of a ^{232}Th fuel rod to the proton beam is shown in Fig. 1.

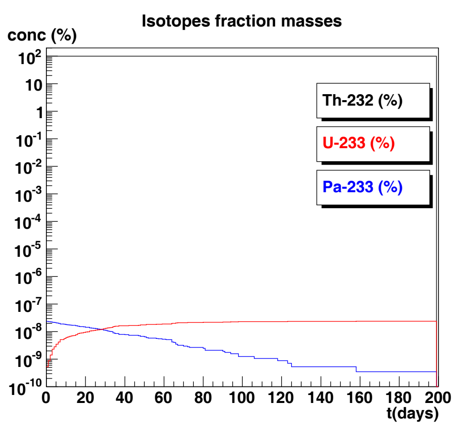

Figure 1: Time evolution of the fraction masses of ^{233}Pa and ^{233}U inside the fuel rod.

These production rates for ^{233}U indicate that it is basically impossible in practice to reach the value of ~ 1.8 % ^{233}U starting with a "pure" ^{232}Th fuel rod. However additional ^{235}U or Pu would increase the production rate.

Careful investigation of the simulation output showed that while in the majority of the neutron capture events the de-excitation gammas were correctly generated, the resulting $(A + 1)$ isotope was not generated by the simulation. This new bug has been reported to the GEANT4 developers team and a fix has been proposed [4].

RESULTS FOLLOWING THE BUG FIX

Following the discovery and fix of the GEANT4 code bug the simulations of the 1 day exposure of the "pure" ^{232}Th fuel rod were repeated. The results are shown in Fig. 2 and 3.

In the previous results there was the same number of neutron capture events on ^{232}Th, however the production of ^{233}Th was suppressed and therefore the yield of ^{233}U was significantly reduced. Following the bug fix, the ^{233}U yield increased by almost 4 orders of magnitude.

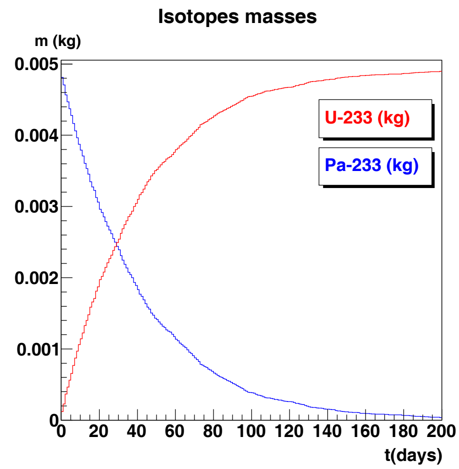

Figure 2: Correct time evolution of the masses of ^{233}Pa and ^{233}U inside the fuel rod after 1 day continuous exposure to the proton beam.

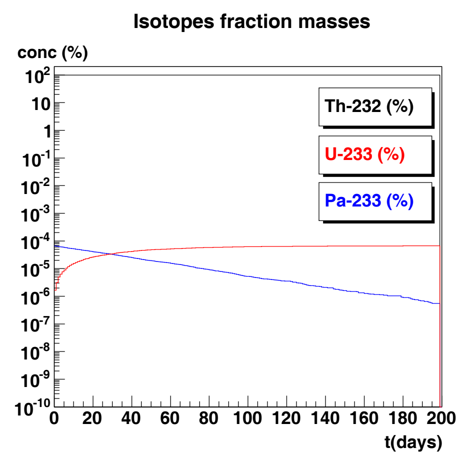

Figure 3: Correct time evolution of the fraction masses of ^{233}Pa and ^{233}U inside the fuel rod after 1 day continuous exposure to the proton beam.

SIMULATION OF DIFFERENT ^{232}TH FUEL RODS GEOMETRIES

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. The results are shown in Fig. 4. The mass of ^{233}U produced increases constantly with both fuel rod radius and height, however no significant gain can be achieved by going above 60 cm in height. The fraction mass of ^{233}U produced is inverse proportional to the fuel rod radius, and it does not depend much on the fuel rod height over the range investigated in this paper.

The simulations have been repeated, in order to show the effect of several days of exposure to the proton beam. The yield and fraction mass of ^{233}U produced are shown in Fig. 5 and 6 for different target dimensions.

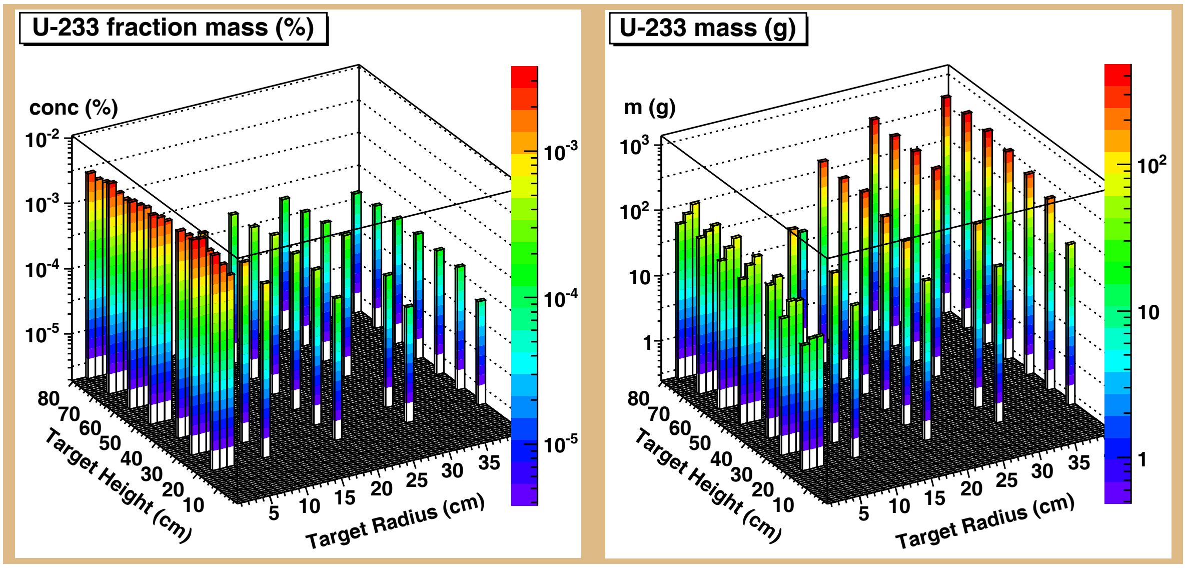

Figure 4: ^{233}U yields after 1 day continuous exposure to the proton beam.

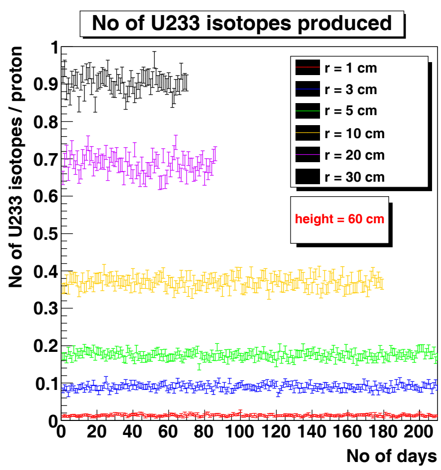

Figure 5: No of ^{233}U isotopes produced per incident proton after (no of days) exposure to the proton beam.

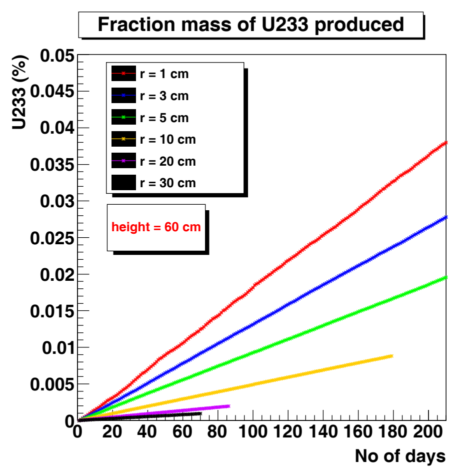

Figure 6: Fraction mass of ^{233}U produced after (no of days) exposure to the proton beam.

The yield of ^{233}U is constant in time for the first 200 days of exposure to the proton beam. This is because the fraction mass of ^{233}U remains very low for the entire period and therefore the initial parameters remain almost the same.

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 fraction mass of ^{233}U produced is maximum for the smallest radius considered of 1 cm, while the total yield of ^{233}U increased with the target radius, for 30 cm reaching the constant value of almost one ^{233}U isotope produced per incident proton.

REFERENCES

[1] A. Heikkinen *et al.*, "A Geant4 physics list for spallation and related nuclear physics applications based on INCL and ABLA models", 17th International Conference on Computing in High Energy and Nuclear Physics (CHEP09), IOP Publishing, Journal of Physics: Conference Series 219 (2010) 032043.

[2] A. Howard *et al.*, "Validation of Geant4 against the TARC benchmark and new developments in PreCompound/De-Excitation modelling", 1^{st} Workshop on Accelerator Radiation Induced Activation, October 2008, PSI, Switzerland.

[3] C. Bungau *et al.*, "Study on neutronics design of an accelerator driven subcritical reactor", IPAC'10, Kyoto, May 2010, MOPEA040, p. 160 (2010), `http://www.JACoW.org`.

[4] Bug found in GEANT4 neutron capture model `http://bugzilla-geant4.kek.jp/show_bug.cgi?id=1155`.