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FAST ACCELERATOR DRIVEN SUBCRITICAL SYSTEM FOR ENERGY PRODUCTION: NUCLEAR FUEL EVOLUTION

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ABSTRACT

Accelerators Driven Systems (ADS) are an innovative type of nuclear system, which is useful for long-lived fission product transmutation and fuel regeneration. The ADS consist of a coupling of a sub-critical nuclear core reactor and a proton beam produced by a particle accelerator. These particles are injected into a target for the neutrons production by spallation reactions. The neutrons are then used to maintain the fission chain in the sub-critical core. The aim of this study is to investigate the nuclear fuel evolution of a lead cooled accelerator driven system used for energy production. The fuel studied is a mixture based upon ^{232}Th and ^{233}U. Since thorium is an abundant fertile material, there is hope for the thorium-cycle fuels for an accelerator driven sub-critical system. The target is a lead spallation target and the core is filled with a hexagonal lattice. High energy neutrons are used to reduce the negative reactivity caused by the presence of protoactinium, since this effect is most pronounced in the thermal range of the neutron spectrum. For that reason, such material is not added moderator to the system. In this work is used the Monte Carlo code MCNPX 2.6.0, that presents the the depletion/ burnup capability. The k_{eff} evolution, the neutron energy spectrum in the core and the nuclear fuel evolution using ADS source (SDEF) and kcode-mode are evaluated during the burnup.

Key Words: ADS, thorium, MCNPX, burnup.

1. INTRODUCTION

The Accelerator Driven Subcritical System (ADS) is an innovative nuclear reactor which has been studied to produce energy and transmute radioactive wastes, or as a starter to breed the required ^{233}U in a thorium based type of fuel. A great number of works on the ADSs and the relative neutronics has been reported in the scientific literature [1], [2], [3], [4], [5], [6], [7]. The concept of ADS combines a particle accelerator with a sub-critical core. The sub–criticality enables that a fuel with a large fraction of fuel and/or minor actinides can be loaded into the sub–critical system without any reactivity induced safety problems, this system can be controlled safely and easily because the sub–critical systems need an external source of neutrons to start the nuclear reactions. An ADS consists of three parts: (1) particle accelerator, (2) spallation target and (3) subcritical reactor core. A high intensity continuous wave proton beam with an

energy of around 1 GeV and a current of several tens milliamperes is injected into a target of heavy metal resulting in a spallation reaction that emits neutrons. Such neutrons are used to maintain the fission chain in the subcritical core.

At the present rate of consumption, the known uranium resources on earth will be insufficient to continue its use as fuel for nuclear energy production, beyond some 200 more years. Accelerator-driven systems are an interesting solution to this problem since they could use as a starter to breed the required ^{233}U in a thorium based type of fuel [4].

There are many reasons for the resurgence of interest in the thorium fuel cycle nowadays. The phenomenal increase in the price of uranium and thorium's abundance is about three times of uranium are probably the most compelling reasons. A better neutronic characteristic of ^{232}Th and the regenerated ^{233}U are attractive because several previous studies have shown some promising reactor physics performances of thorium-fueled core due to those characteristics. Thorium-fueled reactor is also an attractive tool to produce long-term nuclear energy with low radiotoxicity waste [8].

In this work we report results obtained from the simulation of an ADS for regeneration fuel and durable energy production. Our goal has been to evaluate the feasibility of the concept of accelerator- driven subcritical reactors for the ^{233}U production, by performing an analysis of the fuel composition throughout the operation period. For this simulation the Monte Carlo code MCNPX 2.6.0 is used. Such version is quite interesting for the ADS simulation because it allows to obtain the nuclear fuel evolution during the operation using ADS source (SDEF) and kcode- mode. The burnup capability of this code is directed for the description of systems with values of k_{eff} close to one, since for very small values of system criticality the error tends to get larger. However, it allows a reasonable approximation for the qualitative analysis proposed in this initial work. The results provided in this paper allow a general analysis of the nuclear fuel evolution and further indicate that fuel cycle based on thorium can be used for long-time nuclear energy generation in accelerator-driven reactors.

The paper is organized as follows. The parameters of the system simulated are shown in Section 2; the calculational procedure is presented in Section 3; the time evolution of the neutron flux and fuel composition are evaluated and the results are described in Section 4 and, finally, Section 5 brings up our conclusions.

2. ACCELERATOR DRIVEN SUBCRITICAL SYSTEM

This study presents the nuclear fuel evolution of a typical cylindrical accelerator driven subcritical for energy production system loaded with $^{232}ThO_2 + 15\%\ ^{233}UO_2$. Fig. 1 shows a schematic view of the hybrid reactor design.

Table 1 summarizes the geometrical and operational parameters of the system simulated in this study. The accelerator tube has a radius of 1.5 cm, and the axial position center of the target. The simulated system consists of four different sections: (i) the spallation neutron target (central cylinder), (ii) the subcritical core, (iii) the reflector zone.

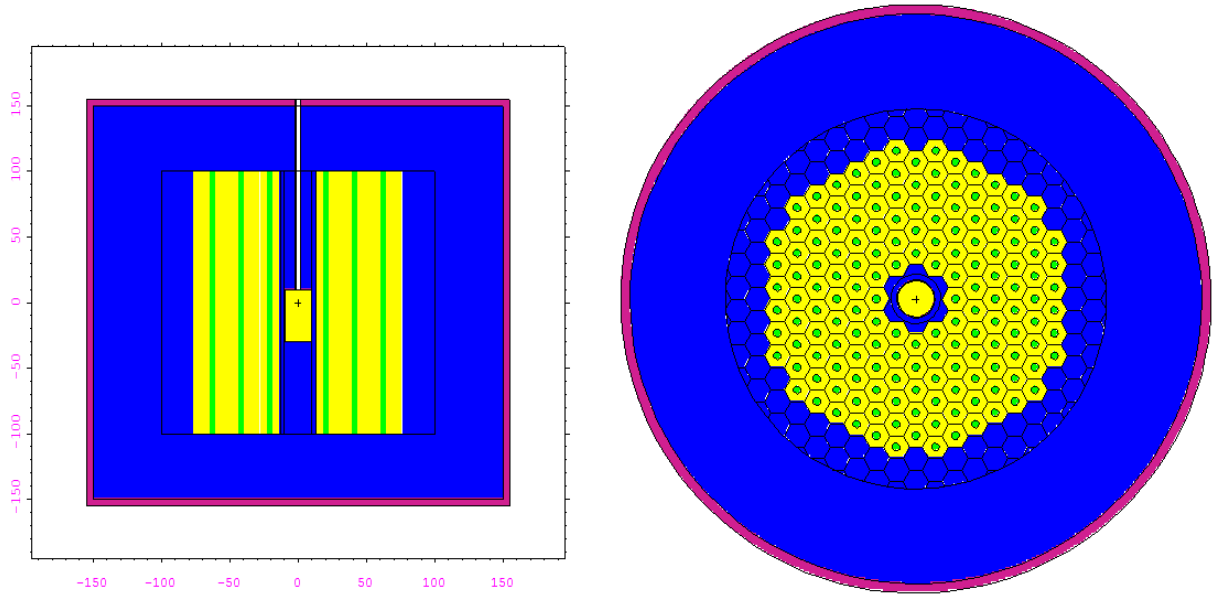

Figure 1: ADS geometry (X-slice for x=0) and (Z-slice for z=0): fuel (green), Pb of target and core (yellow), Pb of the reflector(blue) and iron of the tank(lilac).

Table I: Main parameters of the system.

^{232}Th/ ^{233}U system	
Core volume	6.0 m^3
Fuel mixture (initial)	$^{232}ThO_2$ + 15% $^{233}UO_2$
Fuel mass	3.92 t
Thermal power	515 MW_t

2.1 The spallation neutron target

Due to their higher neutron yields only heavy targets are considered practical. Lead [3], or more often, lead-bismuth [9], are proposed as liquid targets. The use of only lead for the target prevents the radiological hazards from ^{210}Po, maintaining a high neutrons production by spallation, since the values of spallations cross sections for the two materials are very close. Since the Lead has a good heat transfer rate, it also is used as a coolant. The Pb cylindrical target has 9.5 cm radius and 39 cm height.

2.2 The subcritical core

The core is a cylinder of 6.0 m^3 filled with a hexagonal lattice loaded with $^{232}ThO_2$ + 15% $^{233}UO_2$. The volumetric fractions of coolant (55%) and fuel (7%) were adjusted to obtain initial $k_{eff} = 0.95$. The inner diameter of the fuel rods is 2 cm and the number os fuel rods is 156. The coolant used is natural Lead. This design offers many advantages like convective cooling, passive

safety and small neutron absorption cross section [10]. The structural material composing the remaining 38% is Pb of the reflector and iron of the tank.

Another advantage of Lead is that it is not a neutron moderator. This is important because the protactinium effect, which limits the achievable values of k_{eff}, is less severe for fast spectra. In general reactor control is easier with fast spectra, especially for the thorium based cycle. For solid fuels, due to smaller capture cross-sections of fission products, the variations of k_{eff} are less severe for fast than for thermal spectra. However, the inventory of ^{233}U is much larger in fast reactors (about 7 times) , with the associated larger breeding times and inventory radiotoxicity. Because of the high melting temperature of lead it has been proposed to use eutectic lead-bismuth as coolant. However, due to the high working temperature of the coolant necessary to obtain a good thermodynamical efficiency, the ^{210}Po evaporation may become a severe problem. The cost of bismuth is much higher than that of lead, and it is not clear that the bismuth reserves will be abundant enough to provide a large pool of reactors with the required quantities. [10]

2.3 The reflector zone

The Lead reflector thickness was kept fixed at 50 cm.

3. CALCULATIONAL PROCEDURE

The MCNPX 2.6.0 code [11] was used to simulate the geometrical and operational characteristics of the system. For all simulations in this study, the k_{eff} values have been obtained by running the MCNPX calculation in the KCODE-mode, and the fuel compositions and the radial thicknesses have been adjusted to obtain $k_{eff} = 0.95$. In the simulation it used a combination of ADS source (SDEF) and kcode-mode.

In the burnup calculations, Monte Carlo code MCNPX 2.6.0, which is a combination of LAHET [12] and MCNP, was used. The depletion/burnup capability is based on CINDER90. MCNPX depletion is a linked process involving steady-state flux calculations by MCNPX and nuclide depletion calculations by CINDER90. The code runs a steady-state calculation to determine the system eigenvalue, 63-group fluxes, energy-integrated reaction rates, fission multiplicity (ν), and recoverable energy per fission (Q values). CINDER90 then takes those MCNPX-generated values and performs the depletion calculation to generate new number densities for the next time step. MCNPX takes these new number densities and generates another set of fluxes and reaction rates. The process repeats itself until after the final time step specified by the user [13].

In this work were used 73 time steps of 50 days (total time = 10 years). The thermal power of operation throughout this period was 515 MW_t.

4. RESULTS

Fig. 2 shows the multiplication factor as a function of time for the ^{232}Th/^{233}U hybrid system running with its original fuel over 10 years. The k_{eff} with its standard deviation for the first step was 0.95456 ± (0.00041) and $k_{eff} = 0.46909 \pm (0.00024)$ for a last step. The drop of the k_{eff}

value is mainly due to the increased poisoning caused by the accumulation of fission products having large neutron capture cross-sections and, of course, the constant energy generation by the system.

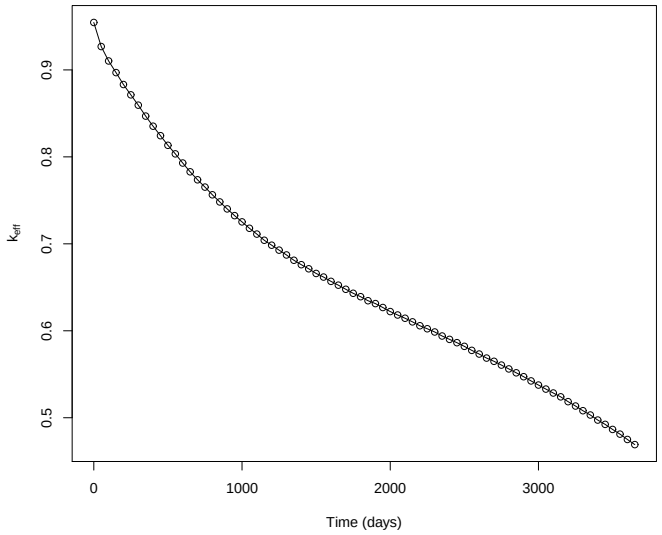

Figure 2: Evolution of the ^{232}Th/^{233}U system reactivity .

Fig. 3 shows the flux as a function of the energy in the fuel, and in the Lead of the core (coolant). Both spectrum have their maximum around 250 keV. This value will be used for the fuel evolution analysis.

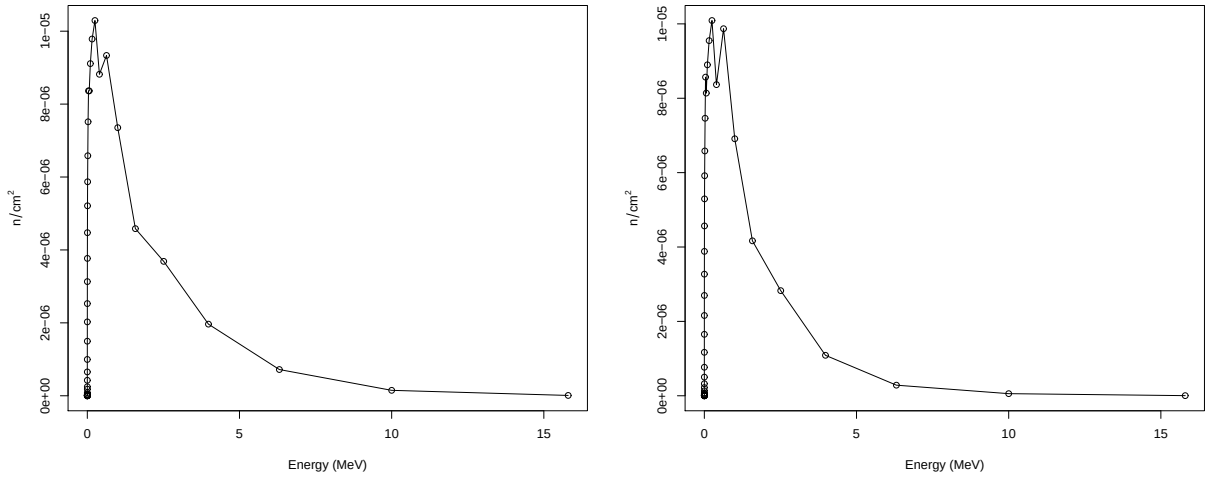

Figure 3: Neutron flux in the fuel (left) and in the coolant (right).

4.1 Analysis of the ^{232}Th concentration in the operational time

Fig. 4 describes the ^{232}Th concentration during the 10 years of operation. It can be verified that the concentration of this isotope is reduced by about 30% during the operation time. It is the result of the considerable capture cross section of this isotope, which allows the ^{233}U production. The total cross sections and capture (n, gamma) cross section for ^{232}Th are shown

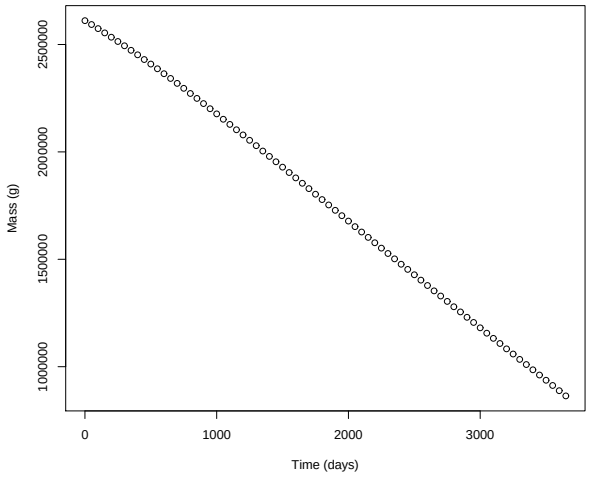

Figure 4: Mass variation of the ^{232}Th.

in Fig. 5. From these figures we can see that the most probable value of neutron energy for our system (0.251 MeV) is located after the resonances region. The total cross-section of ^{232}Th for this neutrons energy is around 10 barns, while the cross section for radiative captures is close to 1 barn. Around 10 % of the neutron interactions with this isotope generates radiative capture. This captures lead to ^{233}Th formation. In addition to the generation of ^{233}U, it further contributes weakly to the system criticality through fission reactions, since the cross section of fission of this nucleus (Fig. 6) to the energy of interest is very low (less than 10^{-6} barns).

4.2 Analysis of the ^{233}Pa concentration in the operational time

A relevant question in the neutron spectrum choice is the protactinium effect. ^{233}U is formed when ^{232}Th captures a neutron, and soon then undergoes two β decays:

$$^{232}Th + n \longrightarrow {}^{233}Th \xrightarrow[22.3\ \mathrm{min}]{\beta^-} {}^{233}Pa \xrightarrow[26.97\ \mathrm{d}]{\beta^-} {}^{233}U \tag{1}$$

The presence of protactinium imposes limits on the admissible neutron flux when using solid fuels. This limitation is due to two detrimental effects of protactinium [10]:

- Protactinium captures neutrons, and, thus, decreases the reactivity of the reactor.

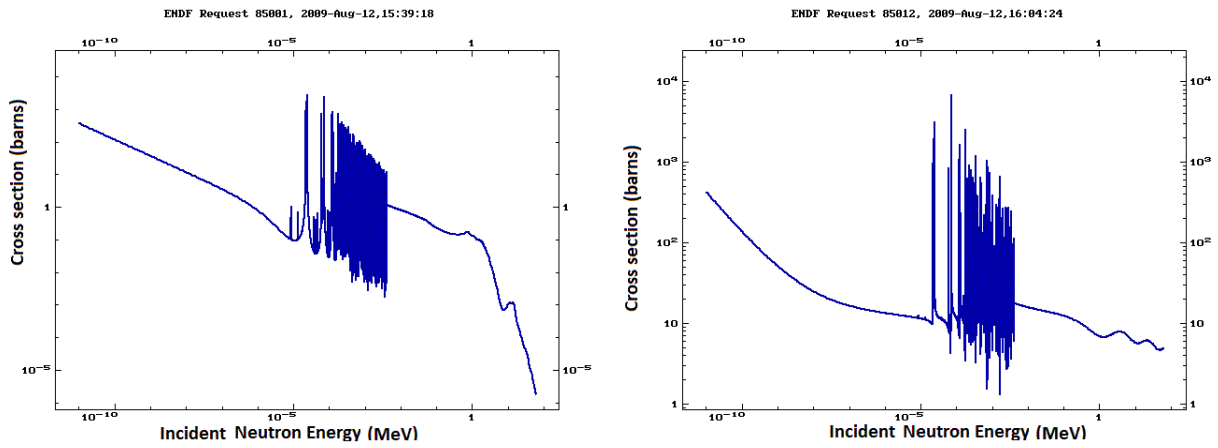

Figure 5: Total cross section (left) and capture cross section (right) for ^{232}Th.

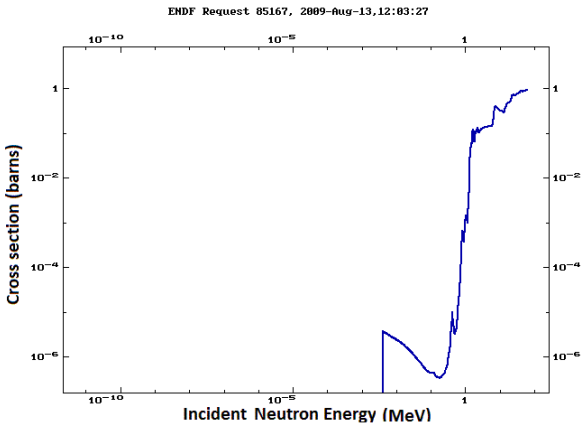

Figure 6: Fission cross section for ^{232}Th.

- After a reactor stop, the ^{233}Pa inventory transforms into ^{233}U, which leads to an increase of the reactivity. This increase may lead to criticality of the reactor. The characteristic time for such evolution is of the order of the life-time of ^{233}Pa, i.e., about one month. Corrective actions could be easily taken by insertion of a negative reactivity. However the advantage of passive safety of hybrid systems would be lost. It is, thus, interesting to keep the system sub-critical in all circumstances.

Fig. 7 shows the ^{233}Pa concentration during the 10 years of operation. It may be noted that this isotope is formed in large scale which contributes negatively to the reactivity.

Fig. 8 shows the total neutron cross section and neutron capture cross section for ^{233}Pa. The ^{233}Pa can, through the neutron capture, to generate the isotope ^{234}Pa. When this happens, it does not decay (via beta decay) for ^{233}U, what is a disadvantage for a system that aims to produce this isotope.

From Fig. 8 it can be observed that the capture cross section for the region of interest is relatively

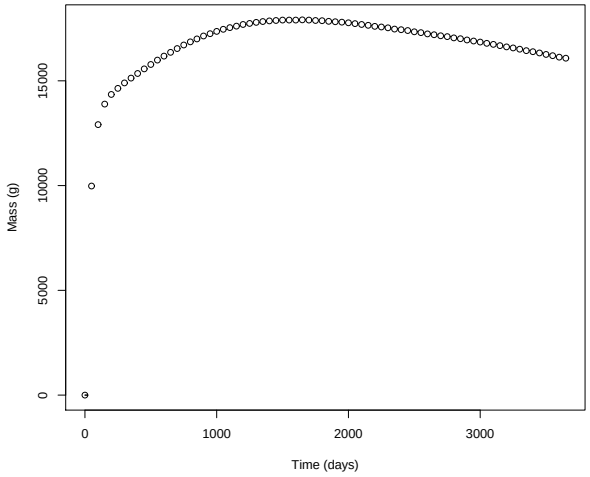

Figure 7: Mass variation of the ^{233}Pa.

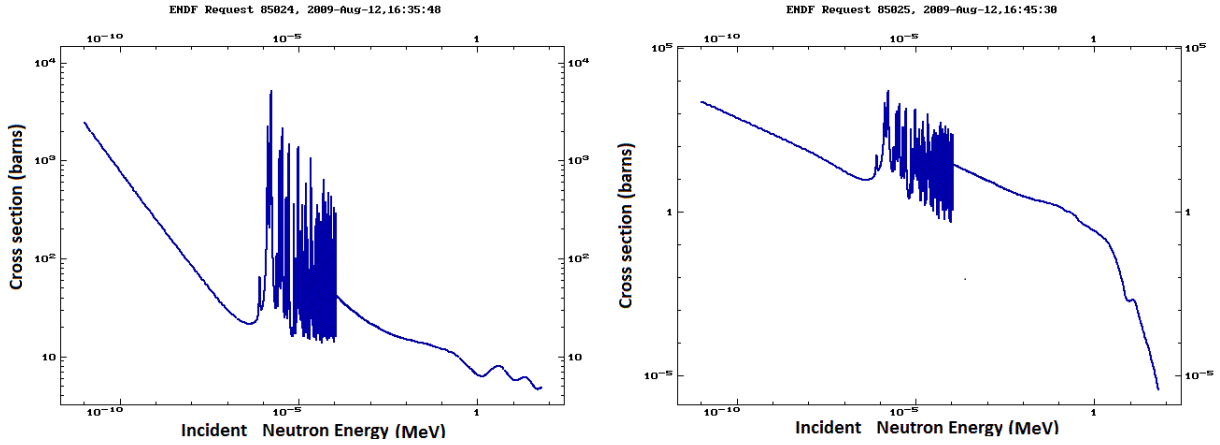

Figure 8: Total cross section (left) and capture cross section (right) for ^{233}Pa.

low (around 1 barn), representing about 10 % of the also small total cross-section (around 10 barns). It can be verified that non-use moderator facilitates the fuel regeneration and the system reactivity maintenance, since the total cross-section for the ^{233}Pa in the fast region of the neutron spectrum is clearly lower than in the thermal region. So, the negative effect of protactinium in the system reactivity (and thus in the fuel regeneration) is less pronounced than if are used low-energy neutrons.

4.3 Analysis of the ^{233}U concentration in the operational time

Fig. 9 presents the ^{233}U concentration during the operation time. As can be verified in this figure the concentration of this isotope decreases with the time. This isotope is produced by the

regeneration of the ^{232}Th, but it is constantly consumed for power generation, since the initial loading is used to generate 515 M W_t thermal power for 10 years. After all, the ^{233}U is the main fissile isotope of the system.

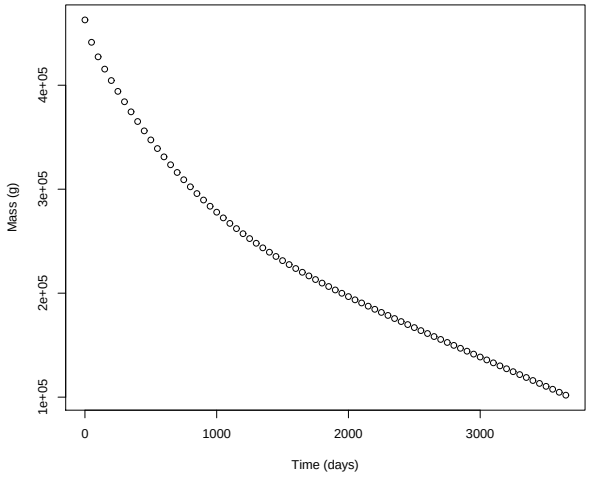

Figure 9: Mass variation of the ^{233}U.

The total number of fission occurring per second in the system is:

$$\text{Fission rate} = P_{\text{MW}} \times \frac{10^6 J}{MWs} \times \frac{\text{fission}}{191\ \text{MeV}} \times \frac{\text{MeV}}{1.60 \times 10^{-13} J} \times \frac{86400 sec}{\text{day}} \tag{2}$$

Resulting in a burnup rate of:

$$\text{burnup rate} = 2.83 \times 10^{21} \times 515 \times 233/6.022 \times 10^{23} = 561.35 \ \ g/day \tag{3}$$

For ten years of operation the consumption is approximately 2049 kg of this isotope. It can be verified that the concentration of this isotope ^{232}Th is reduced by about 360 kg during the 10 years of operation. This proves that not all the ^{233}U spent for the power generation comes from initial loading system, but it comes from regeneration of the ^{232}Th.

The ^{233}U can, through the neutron capture, to generate the isotope ^{234}U (which can give rise to ^{235}U). However, as can be verified in the Fig. 10 and Fig. 11, the total cross section for 0.250 MeV is around 10 barns and the fission and capture cross sections are around 4 and 1 barn, respectively. That is, fissions occur more than captures forming of ^{234}U.

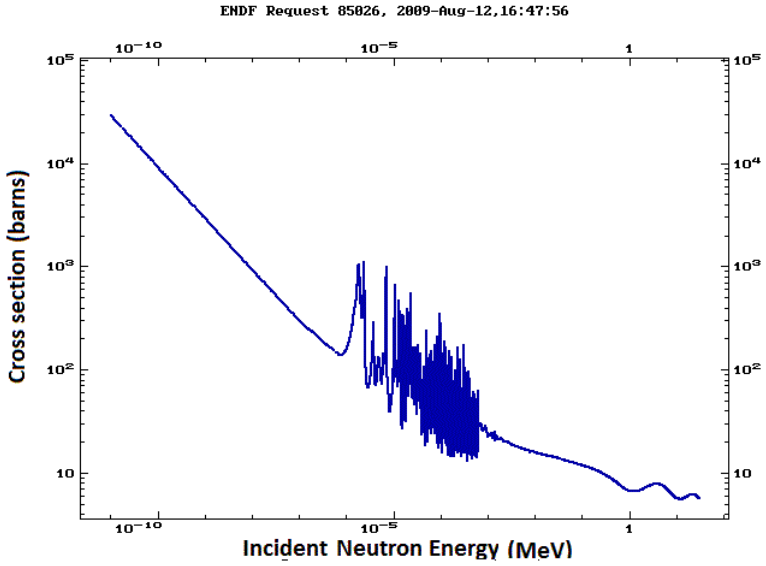

Figure 10: Total cross section for ^{233}U.

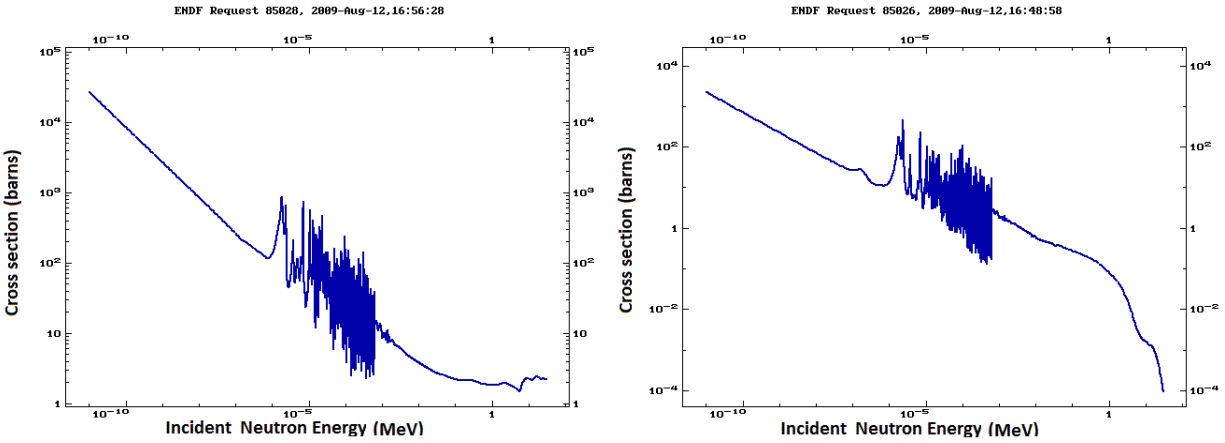

Figure 11: Fission cross section (left) and capture cross section (right) for ^{233}U.

4.4 Analysis of the ^{235}U concentration in the operational time

Fig. 12 describes the ^{235}U concentration during the 10 years of operation. It may be noted that this isotope is formed in large scale, because, as we see in the Fig. 13, about 10 % of the nuclear reactions of the ^{234}U leads to the ^{235}U formation, since the total cross-section is nearly 10 barns and capture is around 1 barn. The ^{234}U formation is quite pronounced (this isotope is produced from the ^{233}U) resulting in a high production of ^{235}U.

However, as shown in Fig. 14, this isotope does not contribute significantly to the system reactivity due to its low fission cross section in the region of interest.

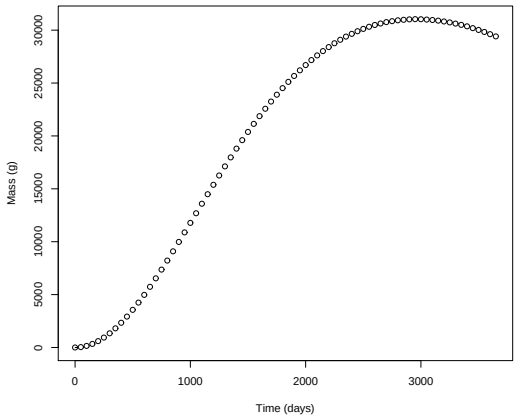

Figure 12: Mass variation of the ^{235}U.

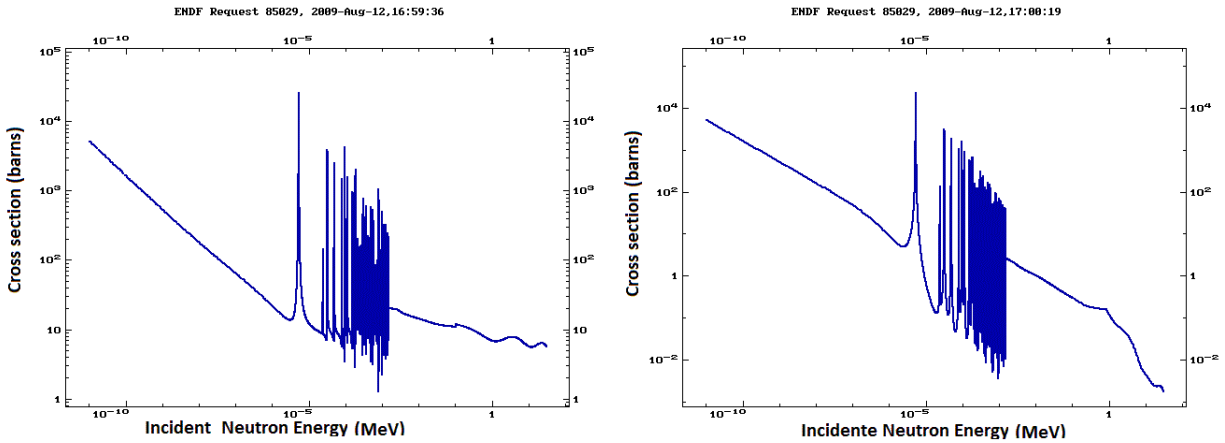

Figure 13: Total cross section (left) and capture cross section (right) for ^{234}U.

5. CONCLUSIONS

In this paper we have presented calculations of fast subcritical hybrid system using fuel based on thorium. The main conclusions have been:

- Fuel cycle based on thorium can be used for long-time nuclear energy generation in accelerator-driven reactors.
- The use of fast neutrons is privileged with respect to the use of thermal neutrons in systems that operate in the Th /U cycle.
- Despite the low k_{eff} it is possible to generate electricity and regenerate fuel.

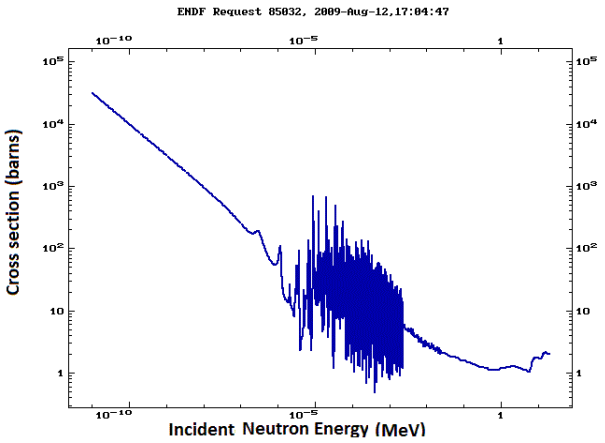

Figure 14: Fission cross section for ^{235}U.

Future work will involve increasing of the volumetric fraction of fuel in the core, with consequent increase in the k_{eff} values. Furthermore, a better fit of the volumetric fraction will allow the use of smaller values of fuel enrichment.

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