Preventive way of relatively low radioactive waste energy production by thorium-uranium fuel cycle application

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Abstract

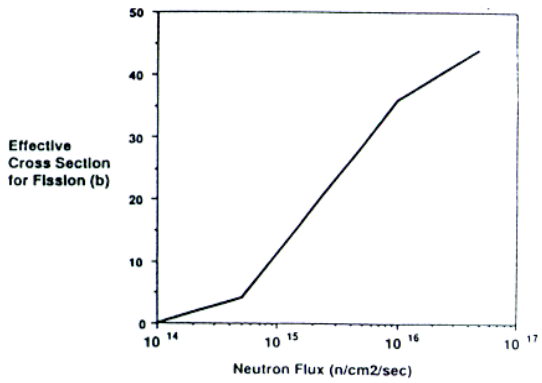
The paper examines the thorium-uranium fuel cycle as a prophylactic way of energy production where the radio-toxicity of the wastes is about three orders of magnitude smaller than in the case of classical PWR reactors. Once-through thorium based fuel cycle analysis in the EPR reactor for energy production was undertaken as the first step of studying thorium application feasibility in the existing light water reactors with minimal modifications in order to exploit them. Monte Carlo methodology calculations were applied in the analysis. Since the analysis of thorium based fuel application in the EPR reactor has shown that once-through thorium fuel cycle can be reached only with difficulty, we have focused on the accelerator driven system (ADS), where the effect of breeding can be used more easily in a more flexible way for higher burn-up of the fuel having an impact on economy improvement. Reviewing the available literature concerning the ADS allowed us to include the analysis of optimal spallation neutron target sizes, which enables obtainment of the required ion beam current of the value achievable with today's technology.

Introduction

An analysis of the possible ways of reduction of radioactive wastes by transmutation of radioactive long-lived fission products such as ^{99}Tc, ^{129}I and ^{135}Cs and by the burning up of transuranic nuclides implies that the reactor core should consist of three zones with fast, epithermal and thermal neutron spectra.

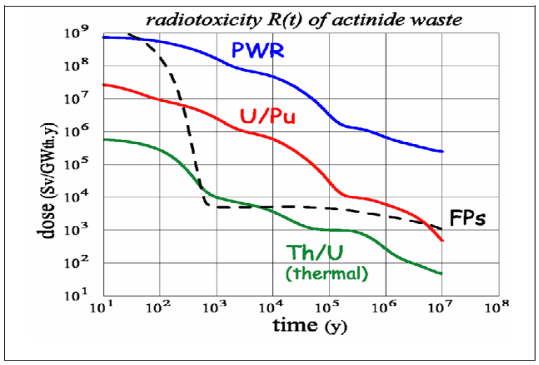
It is also known that the high flux thermal neutron environment ($\geq 10^{16}$ n/cm^2·s) is expected as the best way for the transmutation of most of the radioactive waste to stable or short-lived nuclides and for increasing the probability for fission such actinides as ^{237}Np and ^{238}Np. As an example, Figure 1 shows an effective neptunium cross-section of fission as a function of an intense thermal neutron flux [1]. It should be noted that there is not sharp threshold, but the advantage of 10^{16} n/cm^2 s is very significant (about 35 b). For comparison, the fission cross-section of ^{237}Np in a fast neutron is about 1.5 – 2 barn.

Figure 1: Neptunium fission in an intense thermal neutron flux [1]



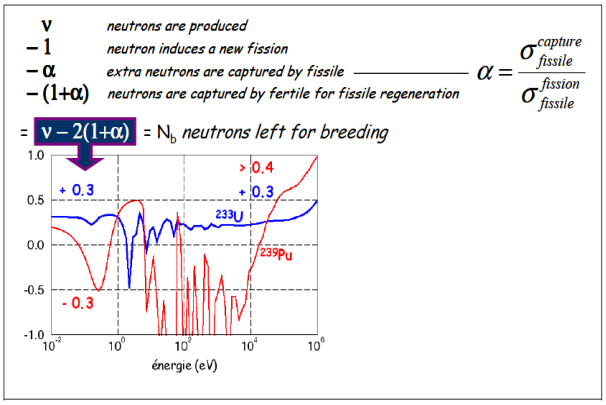
Since we are not able to construct such a reactor core with the three zones and we are not able to reach such an intense thermal neutron flux ($\geq 10^{16}$ n/cm^2·s) in terms of technical feasibility and in view of the inefficiency of actinides incineration for a low intensity thermal neutron flux, the focus is on the thorium-uranium fuel cycle as a prophylactic way of energy production, where the radiotoxicity of the wastes is about three orders of magnitude smaller than in the case of classical PWR reactors [2] (see Figure 2).

Figure 2: Radiotoxicity for various cycles [2]



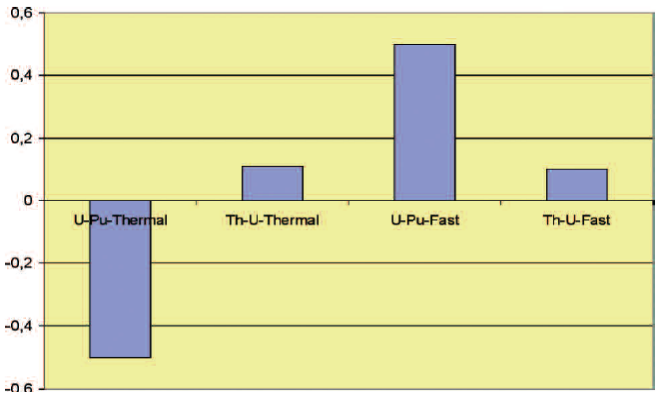
While the ^{238}U 238-^{239}Pu fuel cycle requires fast neutrons to be sustainable, the ^{232}Th-^{233}U fuel cycle is sustainable with either thermal neutrons or fast neutrons. The ^{232}Th – ^{233}U fuel cycle allowed us to obtain the breeding of fissile atoms both in fast, epithermal and thermal neutron spectra. The number of neutrons available for breeding (Nb) is plotted in Figure 3 for the two fertile elements (^{232}Th, ^{238}U) as a function of neutron energy. In the whole neutron energy spectrum for ^{233}U, N_b is always equal 0.3 [2] (see Figure 3).

Figure 3: Available neutrons for breeding both for ^{233}U and ^{239}Pu [2]



However, the number of neutrons available for breeding (Nb) presented in paper [3] is equal to about 0.1 (see Figure 4).

Figure 4: Number of neutrons available for breeding (Nb) in the uranium-plutonium and the thorium-uranium cycles with thermal and fast neutron spectra



Breeding is impossible for negative values of (Nb) [3].

Thorium-based fuels have benefits in terms of radiotoxicity. There are, however, challenges in terms of reprocessing the spent thorium-based fuel. That is why, as a first step of studying the thorium application feasibility, once-through thorium-based fuel cycle analysis for energy production and radioactive waste transmutation were undertaken, which do not require technically difficult reprocessing of the spent fuel.

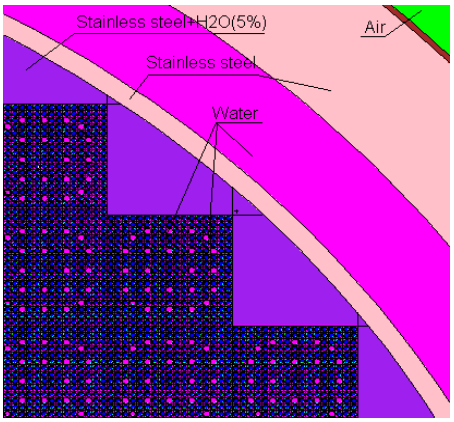
The idea is to analyse the thorium-based fuel application in the open cycle feasibility study in the existing light water reactors with minimal modifications in order to exploit them.

A preliminary analysis of thorium-based fuel application in the EPR reactor has shown that once-through thorium fuel cycle can be reached only with difficulty. It is inferred that the ^{233}U concentration tends to saturation value which does not depend on power density while the kinetics of reaching the saturation value depends on it.

Preliminary analysis of thorium-based fuel application in the EPR reactor

The main aim of this analysis is to show dependence of thorium burn-up in different places of the reactor core. It is needed to determine optimal conditions of ^{333}U productions (position in reactor core and burning period of time). To investigate this problem a European pressurised reactor EPR was used [4-5]. Figure 5 presents the vertical cross-sections of EPR core fragment.

Figure 5: Vertical cross-section of EPR core fragment presenting several UO_2 fuel assemblies neighbouring the reflector [stainless steel +H_2O(5%)]

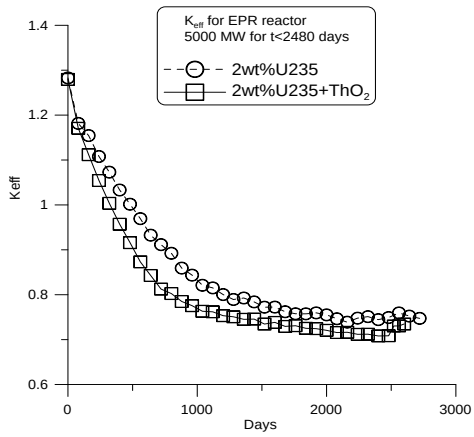


Applying the Monte Carlo methodology (MCNPX 2,6) the neutron multiplication factor k_{eff} has been calculated as a function of burn-up for the reactor core loaded with 241 uranium fuel assemblies (189 uranium and 52 thorium assemblies placed in a peripheral part of the reactor core), as is shown in Figure 5, the uranium assemblies are exploited in constant power of 5 000 MW_{th} (see Figure 6).

It is clearly seen that the impact of the peripheral thorium fuel element assemblies on the neutron multiplication factor in the EPR reactor is insignificantly small for the zero burn-up.

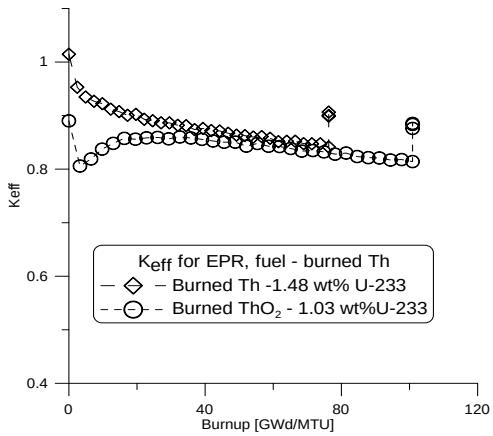
However, conversion of the fertile thorium ^{232}Th into fissile uranium ^{233}U in the peripheral part of the reactor core is ineffective since the neutron flux is comparatively small. The average concentration of fissile uranium ^{233}U in the thorium dioxide rods reached the value of 1.03 wt% after 2 480 days of exploitation on 5 GW_{th} power.

Figure 6: Variation of k_{eff} as a function of irradiation time for the core configurations loaded with 241 uranium fuel assemblies (189 uranium and 52 thorium assemblies placed in a peripheral part of the reactor core) and solely the uranium assemblies (189) are exploited on thermal power 5 000 MW_{th}



Assuming that all the 241 thorium assemblies with 1.03 wt% concentration of uranium ^{233}U were loaded into the EPR reactor core, a computer simulation of neutron multiplication factor k_{eff} was performed as a function of burn-up for the 5 GW_{th} power and compared with the same simulation but for the uranium ^{233}U concentration equal to 1.48 wt% (see Figure 7).

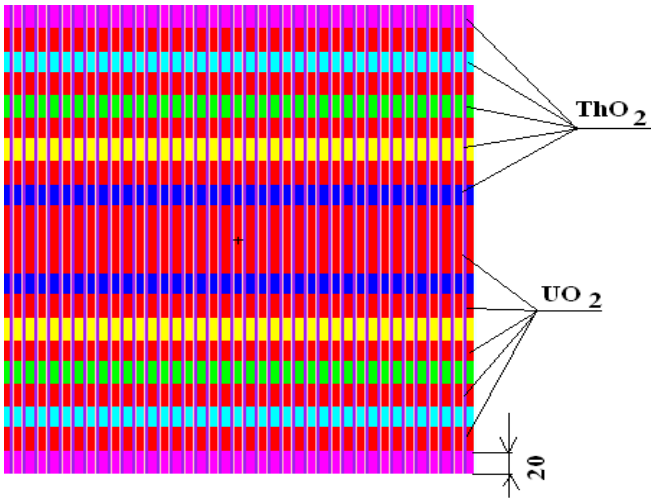
Figure 7: Neutron multiplication factor k_{eff} as a function of burn-up for thorium fuel assemblies with 1.03 wt% and for 1.48 wt% ^{233}U concentration of EPR reactor core exploited on the 5 GW_{th} power



It should be noted that the initial uranium ^{233}U concentration once is above and once is below the saturation concentration, which explains the different shapes of these two curves.

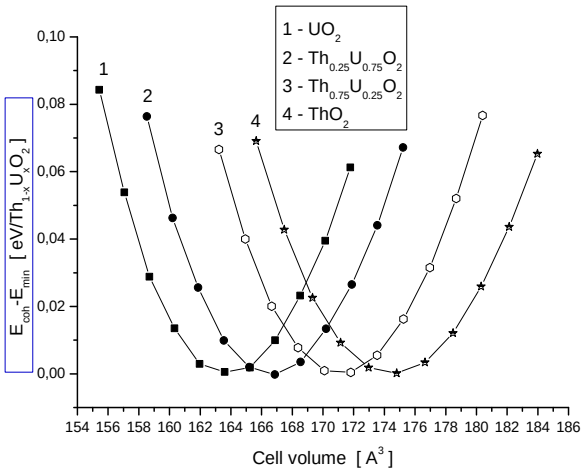
In order to clarify the different shapes of neutron multiplication factor k_{eff} presented in Figure 7 and to investigate the uranium^{233}U concentration dependence on fission rate and time of irradiation, it can be concluded that the EPR fuel rod is filled with UO_2 and ThO_2 forming in the reactor core several fuel layers alternately located. Symmetrically from the fuel rod centre, five thorium dioxide layers 20 cm thick are located in the following ranges 30-50 cm, 70-90 cm, 110-130 cm, 150-170 cm, 190-210 cm. The remaining ranges are filled with enriched 2.25 wt% uranium dioxide (see Figure 8).

Figure 8: Longitudinal cross-section of EPR core and different size fuel layers in the reactor core



"Ab initio" calculations (Figure 9), compared with the experimental data, show that the lattice constants in the $Th_{1-x}U_xO_2$ compound decrease linearly with increasing mole ratio *x*, which means that the uranium dioxide lattice constants are smaller than the thorium dioxide lattice constants. This leads to the conclusion that the uranium dioxide and thorium dioxide-based fuel pellets can be in different proportions and alternately located in the fuel rod because ^{233}U increase in the thorium pellet causes a decrease in its volume, which, in turn does not generate stresses between the rod clad [7].

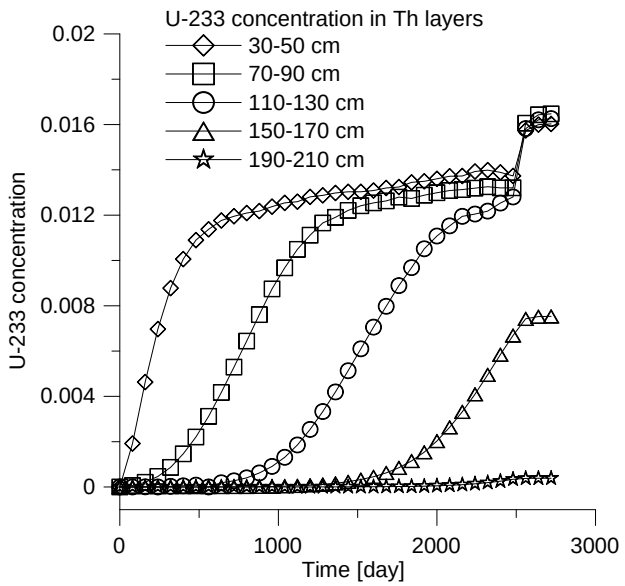
Figure 9: Variation of the cohesive energy with cell volume for $Th_{1-x}U_xO_2$ [7]



The concentration of ^{233}U tends to the saturation value with reactor operation time and sharply increases after the power is cut off (Figure 10). In this reactor the saturation concentration of ^{233}U amounts to about 0.0129 during its work. During the burning time equalling 2 500 days only three thorium layers (30-50 cm, 70-90 cm and 110-130 cm) achieved saturation concentration. The saturation value of ^{233}U concentration practically does not depend on the power density (Figure 10), while the kinetics of reaching the saturation value strongly depends on the power density (neutron flux). The higher the neutron flux is, the more rapid increase of ^{233}U concentration is observed. The five fertile

thorium fuel layers alternately located with the uranium dioxide in the fuel rod (see Figure 8) are irradiated with different neutron flux. The closer the fertile thorium fuel layer is to the centre of the reactor core, the higher is the neutron flux. The final concentration of ^{233}U (Figure 10) consists of two parts: a ''power on'' concentration and a ''power off ''one.

Figure 10: The uranium^{233}U concentration versus reactor operation time for five thorium dioxide layers 20 cm thick as in Figure 8



It should be noted that the ^{233}U concentration tends to saturation value which does not depend on power density while the kinetics of reaching the saturation value depends on it. For initial concentration of ^{233}U equal to saturation value (1.29 wt%) the EPR reactor does not reach criticality, scarcely for 1.5 wt% the reactor becomes slightly overcritical (see Figure 6), which permits exploitation of the reactor during a certain time. This concentration value is a sum of the saturation concentration with the "power off" concentration. It means that once-through thorium fuel cycle can be reached only with difficulty.

While the available neutron numbers for breeding in the thorium-uranium fuel cycle is only a little above zero (0.1 – 0.3, the value is not given precisely in the open literature), we have focused on the accelerator-driven system (ADS). A review of the available literature concerning the ADS lets us include the analysis determination of optimal spallation neutron target sizes, which enables obtainment of the required ion beam current of the value achievable with today's technology.

Required proton beam current for an ADS

The accelerator beam current I (in units of mA) for thermal output power of an ADS, $(P_o)_{th}$ (in units of MW_{th}) is given by:

$$(P_o)_{th} = I\, E_i \cdot G \qquad (1)$$

where E_i is incident proton energy in units of GeV.

The energy gain G of an ADS is given by Equation [8]:

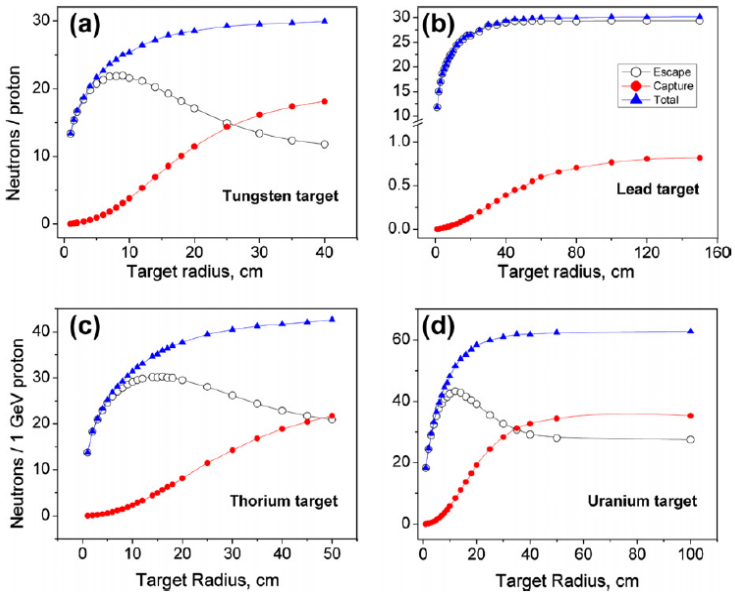
$$G = N_{ne} \cdot \frac{\varphi k_{ef}}{\nu\,(1 - k_{ef})} \cdot E_f \qquad (2)$$

where N_{ne} - the mean number of the neutrons that escape the target per incident primary ion (proton), in units of neutrons per GeV, k_{eff} - effective neutron multiplication factor of the subcritical assembly in the absence of source neutrons, ν – the average number of neutrons released per fission, E_f - the recoverable energy released per fission (in units of GeV) and φ - the importance of the source neutrons, G- the energy gain, refers to thermal output power of the ADS.

Optimal spallation target sizes

The optimal target size refers to target dimensions where for a given incident ion energy the largest number of neutrons escape the target surfaces. Calculations of neutrons that escape and those that are absorbed for different target materials are shown in Figure 11 [9].

Figure 11: Variation of the escaping, captured and total neutron yield as a function of the target radius in the irradiation of range-long targets of ^{nat}W, ^{nat}Pb, ^{232}Th and ^{238}U with 1.0 GeV protons [9]



The optimal dimensions (radius and length) of the cylindrical targets of tungsten, lead, thorium and uranium for two projectile beams of 1 GeV protons and 1.5 GeV deuterons are given in Table 1. Table 1 also presents the numbers of escaping neutrons per incident primary ion.

Table 1: Optimal radius and length and number of the neutron leakage per incident ion for some cylindrical targets for 1 GeV proton and 1.5 GeV deuteron irradiations [9]

Target material	1 GeV proton			1.5 GeV proton		
	Radius (cm)	Length[a] (cm)	Escaping neutrons N_{ne}	Radius (cm)	Length (cm)	Escaping neutrons
W	8.6	31	22.0	9	42	38.2
Pb[b]	~60	54	29.3	~60	73	50.8
Th	14.2	54	30.2	15.8	73	51.2
U	11.7	34	43.1	12.6	45	72.6

[a] Target length is rounded up.
[b] Target radius for lead is approximate as seen in Figure 11b.

Having the optimal target size and number of the neutron leakage and using the Equations (1) and (2) the required accelerator beam current was calculated (see Table 2) on the assumption that $\varphi = 1$.

Table 2: The energy gain and required accelerator beam current for an ADS with keff = 0.98 with optimal targets of ^{nat}Pb, ^{na}tW, ^{232}Th and ^{238}U when they were irradiated by 1 GeV proton and 1.5 GeV deuteron beams

Ion + target	E_i	N_{ne} Source neutron per ion	G energy Gain	Required beam current for 1.5 GW_{th}	Required beam current for 3.0 GW_{th}
	GeV			mA	mA
p+ ^{nat}W	1	22.0	86,2	17.4	34.8
d+ ^{nat}W	1.5	38.2	99.8	10.0	20.04
p+^{nat}Pb	1	29.3	114.9	13.1	26.20
d+^{nat}Pb	1.5	50.8	132.8	7.5	15.0
p+^{232}Th	1	30.2	118.4	12.7	25.4
d+^{232}Th	1.5	51.2	133.7	7.5	15.0
p+^{238}U	1	43.1	169.0	8.9	17.8
d+^{238}U	1.5	72.6	189.8	5.3	10.6

From Table 2 it is evident that by using 1.5 GeV deuteron beams instead of 1 GeV protons, the increased energy gain, and thus the output power is higher (by a factor of 2.8-3.6) than the power required to operate the coupled accelerator, in all four target materials studied.

It is shown that for a modular ADS with uranium target and output power of 1 500 MWth a 1.5 GeV deuteron beam of current 5.3 mA is required, which is achievable with today's technology.

Conclusions and remarks

When applying the thorium-uranium fuel cycle, the radiotoxicity of the wastes is about three orders of magnitude smaller than in the case of classical PWR reactors.

Conversion of the fertile thorium ^{232}Th into fissile uranium ^{233}U in the peripheral part of the reactor core is ineffective since the neutron flux is comparatively (relatively) small.

For initial concentration of ^{233}U equal to saturation value (1.29 wt%) the EPR reactor does not reach criticality, scarcely for 1.5 wt% the reactor becomes slightly overcritical, which permits the exploitation of the reactor during a certain time. This concentration value is a sum of the saturation concentration with the "power off" concentration. It means that once-through thorium fuel cycle can be reached only with difficulty.

The saturation value of ^{233}U concentration practically does not depend on the power density, while the kinetics of reaching the saturation value strongly depends on power density (neutron flux).

While the available neutron numbers for breeding in the thorium-uranium fuel cycle is only a little above zero (0.1 – 0.3, the value is not given precisely in the open literature), we have focused on the accelerator-driven system (ADS), where the effect of breeding can be utilised more easily in a more flexible way for higher burn-up of the fuel having an impact on economy improvement.

For a modular ADS with uranium target and output power of 1 500 MW_{th} a 1.5 GeV deuteron beam of current 5.3 mA is required, which is achievable with today's technology.

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