XX International Baldin Seminar on High Energy Physics Problems – "Relativistic Nuclear Physics & Quantum Chromodynamics"; Russia, Dubna, October 4 – 9, 2010.

Once through thorium based fuel cycle analysis of accelerator driven system for energy production and radioactive waste transmutation – impact on economy improvement.

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Once through thorium based fuel cycle analysis of accelerator driven system for energy production and radioactive waste transmutation – impact on economy improvement.

1. Introduction
- 2. Neutron energy range of interest for the transmutation of long living fission products and incineration of minor actinides
- 3. The bases of energy balance of nuclear power station driven by the proton accelerator.
- 4. Improvement of ADS economy by utilization the fertile Th232.
- 4.1 The idea of research the particular safety problem.
  4.2 Calculation results
- 5. Conclusions

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1. Introduction

- Long range objective:- fuel economy in the sub-critical assembly of ADS in terms of : LLFP transmutation, MA incineration, energy production and thorium based fuel application.
- To fulfill this the sub-critical assembly of ADS should consist of three zones
- The requirement comes out of the fact that each radioactive isotope to be reduced is to be located in a different spectrum of thermal, resonance , and high energy neutron fluxes.
- Moreover high flux thermal neutron environment ($\geq 10^{16}$ n/cm^3·s) is convenient for transmutation most of the radioactive waste both LLFP and MA

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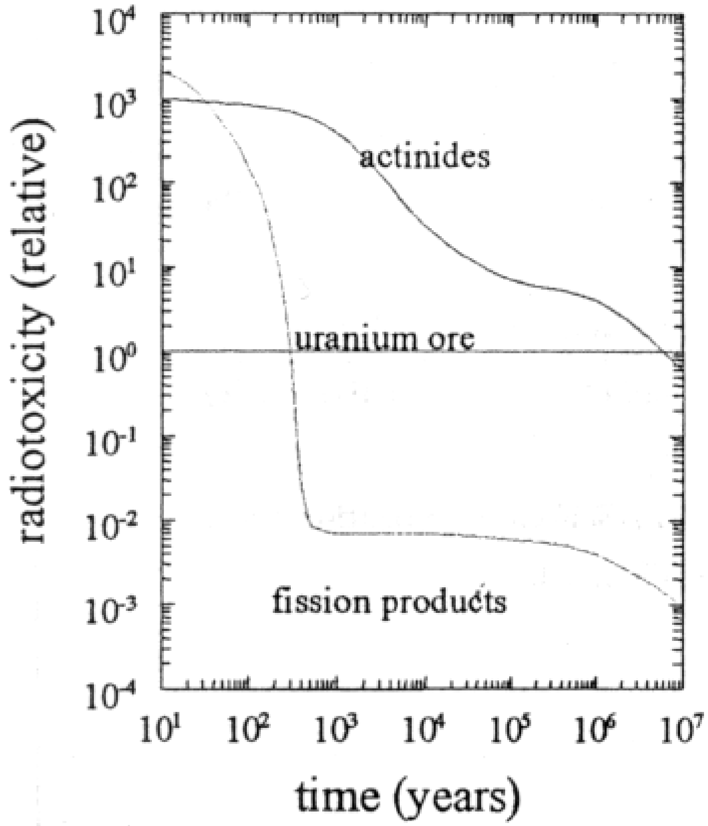
2. Neutron energy range of interest for the transmutation of long living fission products and minor actinides

- The potential radio-toxicity of fission products can be neglected after 250 years.
- However the potential radio-toxicity of actinides remain yet high after million years.
- **This is clearly demonstrated on the Fig. 1. beside.**



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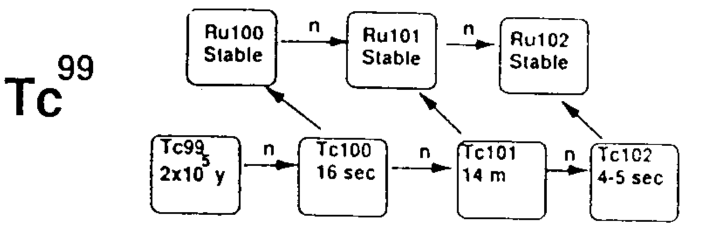
2. Neutron energy range of interest for the transmutation of long living fission products and minor actinides

- The following fission products Tc-99 and I-129 constitute 95 % of total activity of the long lived fission products.
- salts of these two elements are soluble in water and could contaminate the biosphere
- The long leaving iodine ^{129}I after neutron capturing becomes an iodine ^{130}I which decays to the stable xenon ^{130}Xe according to the reaction:
- $\mathbf{^{129}I\ (T_{1/2} = 1.57 \cdot 10^{7}\ y) + n \Rightarrow {}^{130}I\ (T_{1/2} = 12.4\ h) \Rightarrow {}^{130}Xe\ (stable)}$



Isotope	σ (thermal)	σ (fast)
Tc99	20 barns	0.2 barns
I129	31	0.2
Sr90	1	(0.1)
Cs137	0.25	(<0.01?)

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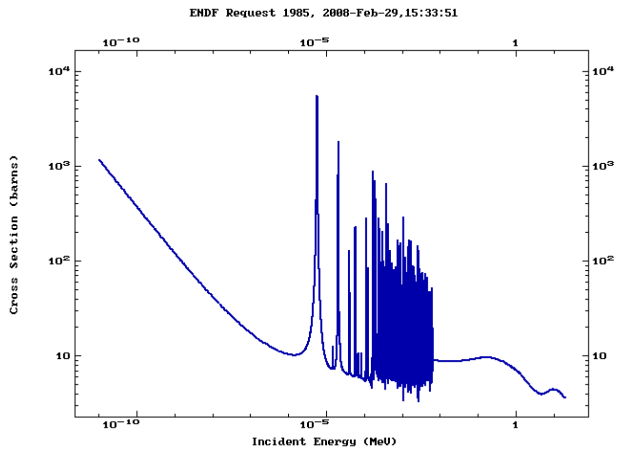
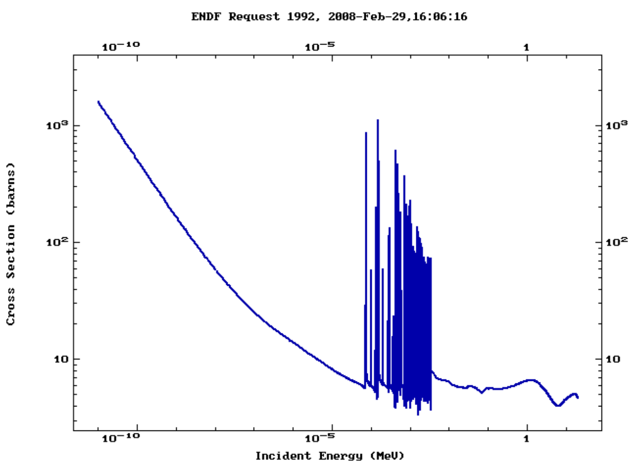
2. Neutron energy range of interest for the transmutation of long living fission products and minor actinides

- In fact, the neutron absorption cross-sections in function of energy for the fission products ^{99}Tc and ^{129}I have resonance complex structure of cross sections.
- The resonance region lies in the neutron energy range from 1 eV to 10 keV. This range is very important for the effective transmutation.
- **This is clearly seen on the figs.2 and 3 of ^{99}Tc(upper) and ^{129}I(lower) presented beside.**





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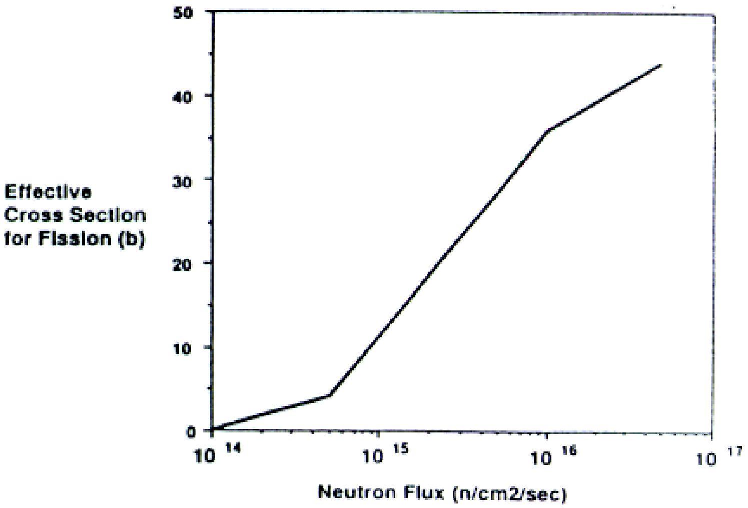
2. Neutron energy range of interest for the transmutation of long living fission products and minor actinides

- The problem is that the actinide waste, which consists of neptunium and higher atomic number elements except plutonium, is thought not to transmute well in a thermal flux of the typical commercial power reactor (10^{14} n/cm^2 s.)
- However, for high enough thermal neutron flux (higher than 10^{14} n/cm^2 s) the effective neptunium cross section for fission increases in function of thermal neutron flux.
- **Fig. 4 of neptunium fission cross section beside shows it.**



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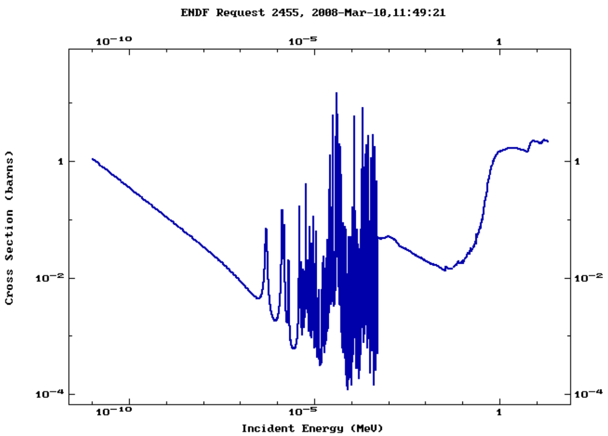
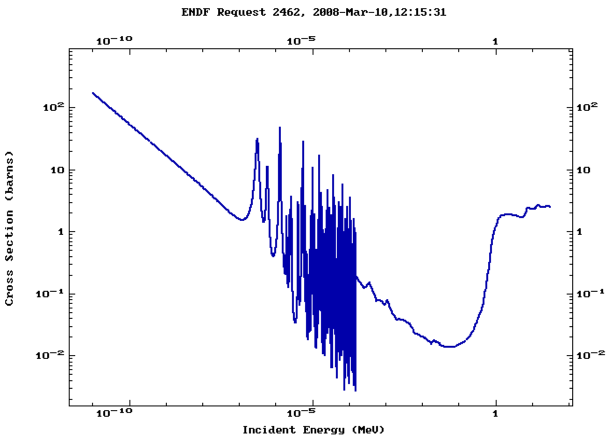
Preliminary analysis of thorium based fuel application in the accelerator driven systems for energy production and radioactive waste transmutation.

2. Neutron energy range of interest for the transmutation of long living fission products and minor actinides

- Since we are not able easily to reach such an intense thermal neutron flux in view of the inefficiency of actinides incineration for a low intensity thermal neutron flux, we focus our attention on fast flux zone of the facility, where the fission probability is more favorable
- **This is clearly seen on the figs 5 and 6 of ^{237}Np(upper) and ^{241}Am(lower) fission neutron cross section presented beside.**



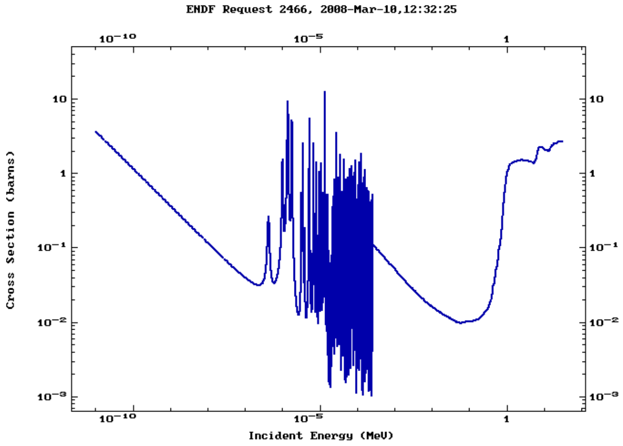


Preliminary analysis of thorium based fuel application in the accelerator driven systems for energy production and radioactive waste transmutation.

2. Neutron energy range of interest for the transmutation of long living fission products and minor actinides

- **Also fig. 7 of ^{243}Am fission neutron cross section versus energy presented beside supports it.**

- Finally, the neutron range of interest for the transmutation of LLFP is the range from 1 eV to 10 keV and for incineration of MA is the range of fast neutrons used.



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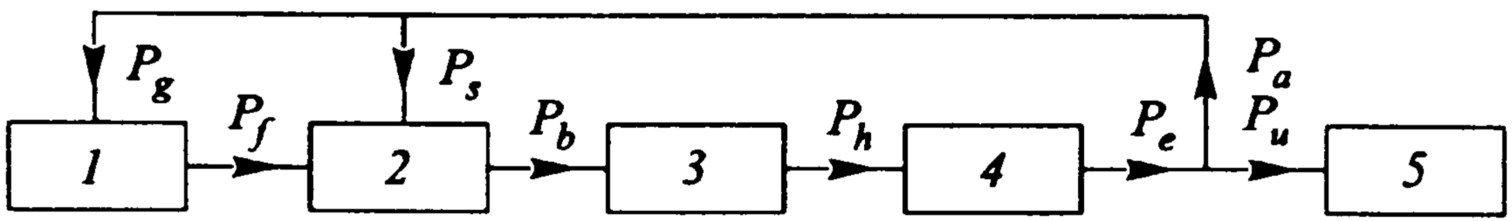
3. The bases of energy balance of nuclear power station driven by the proton accelerator.



- **Fig. 8. Simplified schema of nuclear power station driven by proton accelerator; 1 – high frequency generator, 2 – accelerator, 3 – atomic reactor, 4 – electric power station, 5 – user**
- Pb – proton beam power; Ph – thermal power of subcritical assembly, Pe –generated electric power, Pa- electric power consumed by the accelerator, Pu – electric power directed to the user, Pg – electric power used to supply the high frequency generator, Ps – electric power to supply the vacuum facility, cooling of the magnets and resonators, control system and other systems, Pf - high frequency power.

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3. The bases of energy balance of nuclear power station driven by the proton accelerator.

- $P_h = kP_b$ (1) where k – energy amplification factor.
- Thermal power is transformed into the electric power Pe with efficiency ηe. Now then the electric power is described by the equation:
- $P_e = k\eta_e P_b$ (2)
- Part of the electric power is consumed by the accelerator P_a, and part of it is directed to the user P_u:
- $P_e = P_a + Pu = P_s + P_g + P_u$ (3)
- Here the power of industry grid (net) is to be transformed into the high frequency power P_f:
- $P_f = \eta_g P_g$ (4) where η_g - transformation factor.

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3. The bases of energy balance of nuclear power station driven by the proton accelerator.

- The high frequency power P_f directed to resonators in order to creation the accelerating electric field and acceleration of proton beam is transformed into the proton beam power:
- $P_b = \eta_b P_f$ (5)
- Where η_b – transformation factor:
- Finally, using the equations (2) – (5) and using the elementary techniques we then arrive at:
- $P_s + P_u = P_e - P_g = k\eta_e P_b - P_b/\eta_g\eta_b = (k\eta_e - 1/\eta_g\eta_b)\, P_b$ (6)
- Of course, the last term of equation (6) is reasonable if the expression in the brackets is additive (positive), it means when:
- $k\eta_e\eta_g\eta_b > 1$ (7)
- In the equation (7) two factors experimentally evaluated are following $\eta_e = 0.3$ and $\eta_g = 0.65 – 0.7$ under condition that the high frequency generator works on frequency equal to 30 – 40 MHz. Now then only the factors k i η_b can be changed in the equation (7).

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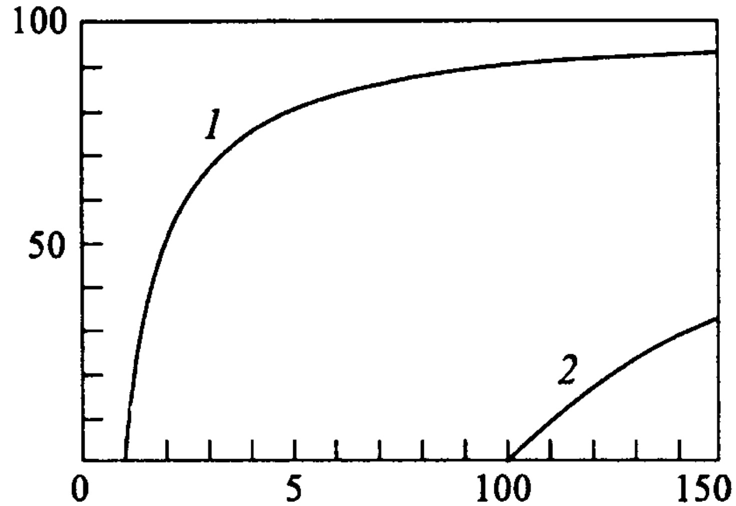
3. The bases of energy balance of nuclear power station driven by the proton accelerator



- **Fig.9. The ηb factor in function of accelerator power for two different accelerators.**
- The η_b factor for two accelerators:the curve 1 for a cyclotron and the curve 2 for a linear accelerator.
- As it is seen in the Fig., the ηb factor of cyclotron reaches the value higher than 0.95 for power equal to100MW, while the ηb of linear accelerator reaches only the value about 0.30 for power equal to 150 MW. So, the analysis of economy in the sub-critical assembly of the accelerator driven system (ADS) we should limit to the cyclotron.

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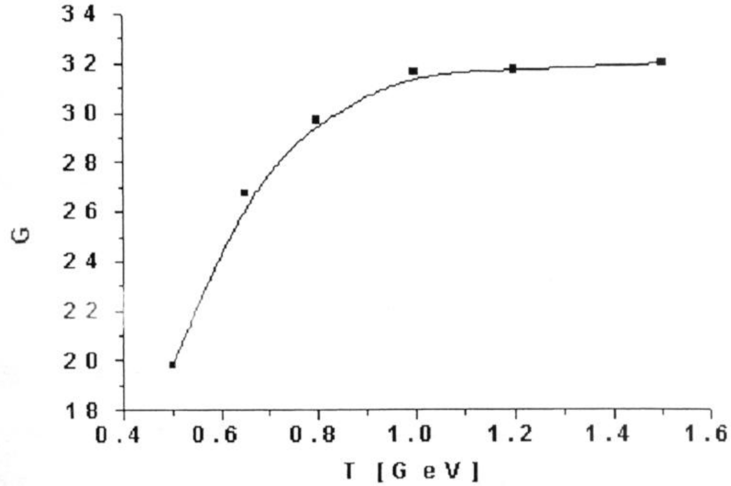
3. The bases of energy balance of nuclear power station driven by the proton accelerator



- **Fig.10. Energetic amplification G in function of proton energy for kef= 0.945±0.003**
- It is clearly seen that the energetic amplification G is equal to about 30 for the proton energy equal to 1GeV. This means that the thermal power of the ADS is about 3000 MWth if the beam proton power is 100 MW (proton current equal to 100 mA at proton energy equal to 1GeV). The evaluated thermal power of 540 MWth as own needs amounts to about18 % of the nominal ADS power. Adding to this the own needs of electric power station equal to about 10 -15% of the nominal ADS power we see that about 1000 MWth is used for the own needs.
- While according to the report nea3109-ads for an ADS with a neutron multiplication factor of 0.95 their own needs is equal to about 12%.

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3. The bases of energy balance of nuclear power station driven by the proton accelerator.

The above short analysis of energy balance for an ADS with a neutron multiplication factor of 0.95, proton accelerator, proton energy of 1 GeV and energy amplification factor k of 30 is summed up in the Table 1 below:

Table 1.

Proton beam power [MW]	Minimal factor k	Minimal thermal power [MW_{th}]	Minimal electrical power [MW_{el}]	Nominal thermal power [MW_{th}]	Own needs [%]	
100	5.4	540	162	3000	18	
50	6.41	320	96	1500	21.3	
30	9.32	279	84	900	31	

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4. Improvement of ADS economy by utilization the fertile thorium 232

- The energy conversion part of an accelerator-driven nuclear power system is similar to that of a normal power plant. Except for the sub-critical state, the core is very similar to that of a critical reactor. It can be designed to operate either with a thermal or fast neutron spectrum.
- However, in the accelerator-driven system, the electrical energy which is recycled to the accelerator reduces the net electrical efficiency of the system. In order to improve the ADS economy, utilization of fertile thorium 232 for breeding ^{233}U fuel can be a solution.
- ^{232}Th is a better fertile material than ^{238}U in thermal reactors because of the three times higher thermal neutron absorption cross-section of ^{232}Th (7.4 barns) as compared to ^{238}U (2.7barns). Thus, conversion of ^{232}Th to ^{233}U is more efficient than that of ^{238}U to ^{239}Pu in thermal neutron spectrum though the resonance integral of ^{232}Th is one third of that of ^{238}U.
- Figures below presenting the neutron absorption cross section of ^{232}Th and neutron absorption cross section of ^{238}U show it.

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4. Improvement of ADS economy by utilization the fertile thorium 232

Fig. 11. Neutron capture cross section of ^{232}Th

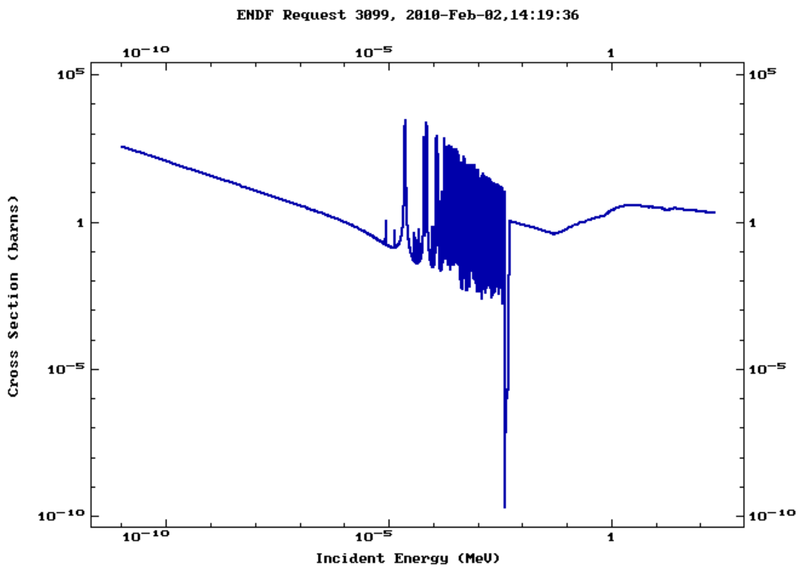
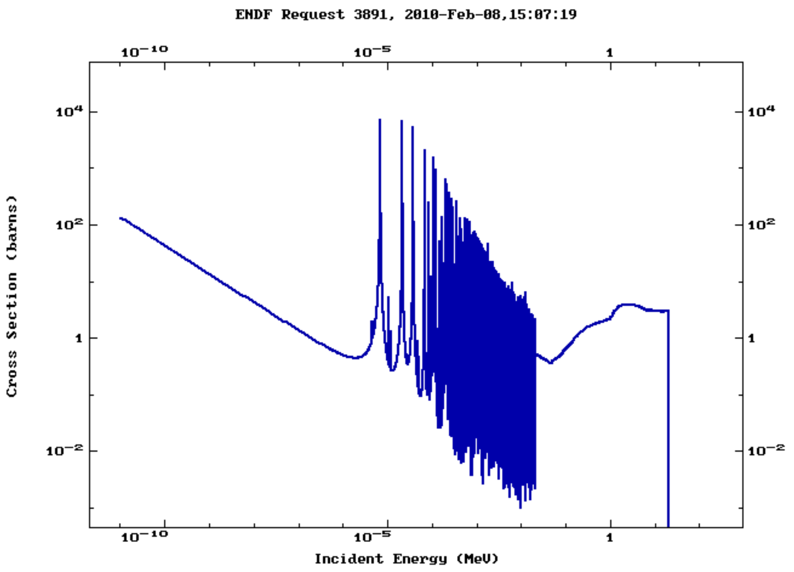


Fig. 12 Neutron capture cross-section of ^{238}U



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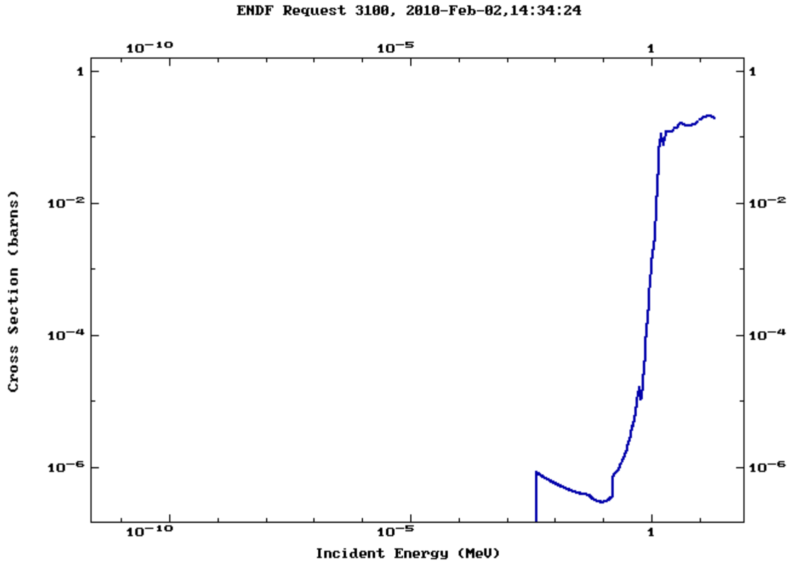
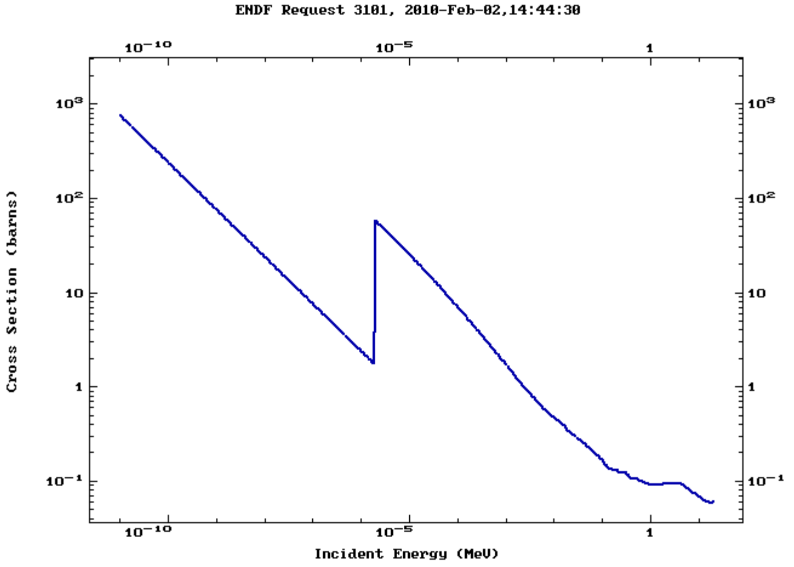
4. Improvement of ADS economy by utilization the fertile thorium 232

Fig. 14.Neutron fission cross-section of ^{232}Th

15.Neutron fission cross-section of ^{233}Th





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4. Improvement of ADS economy by utilization the fertile thorium 232

- Practically ^{232}Th do not undergo fissions except for higher energy than 1 MeV (see Fig.14).
- ^{233}Th undergoes fission (see Fig.15) and undergoes transmutation to ^{234}Th with high probability (1500 barns) what decreases the effectiveness of obtaining the ^{233}U (see Fig. 16).
- Protactinium ^{233}Pa obtained from ^{233}Th by β decaying undergoes transmutation to protactinium ^{234}Pa by neutron capture (20 barns) what in turn decreases the effectiveness of obtaining the ^{233}U (see Fig. 17). Obtained ^{233}U from ^{233}Pa by β decaying undergoes transmutation to uranium ^{234}U by neutron capture (46 barns) what in turn also decreases the amount of ^{233}U (see Fig. 18) which can undergo fission.
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4. Improvement of ADS economy by utilization the fertile thorium 232

Fig. 16. Neutron capture cross-section of ^{233}Th

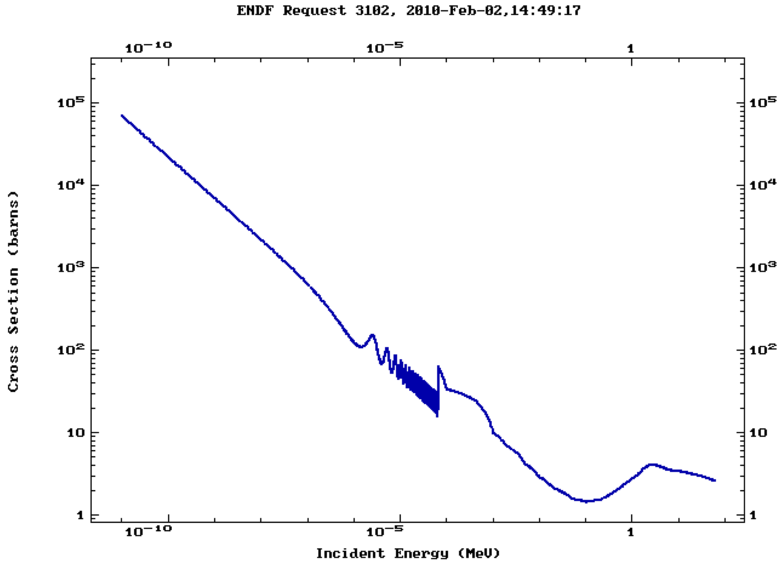
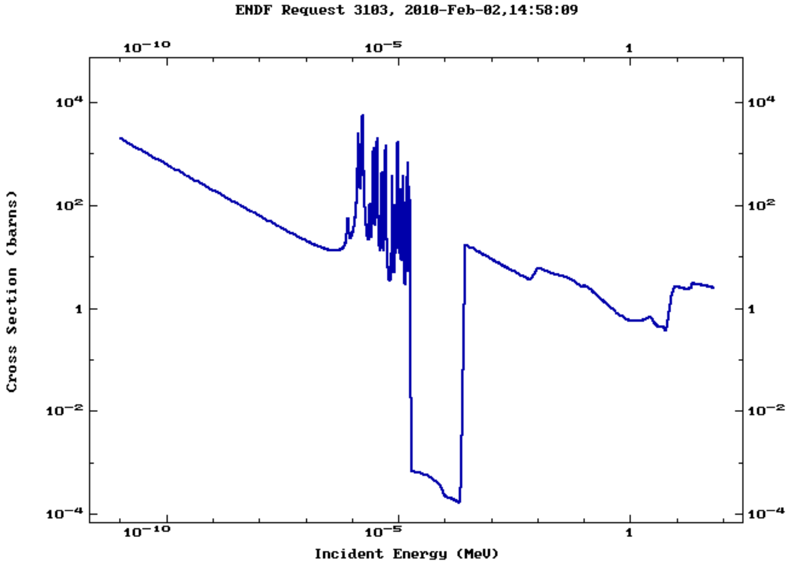


Fig. 17.Neutron capture cross-section of ^{233}Pa



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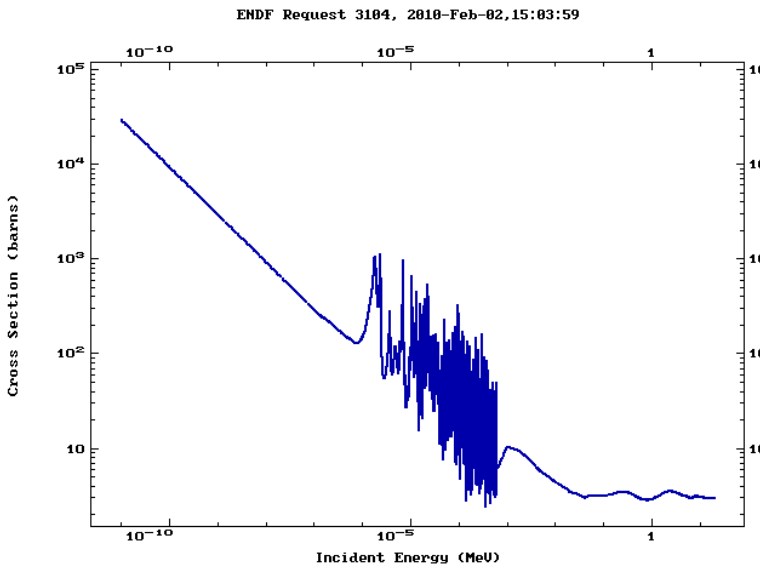
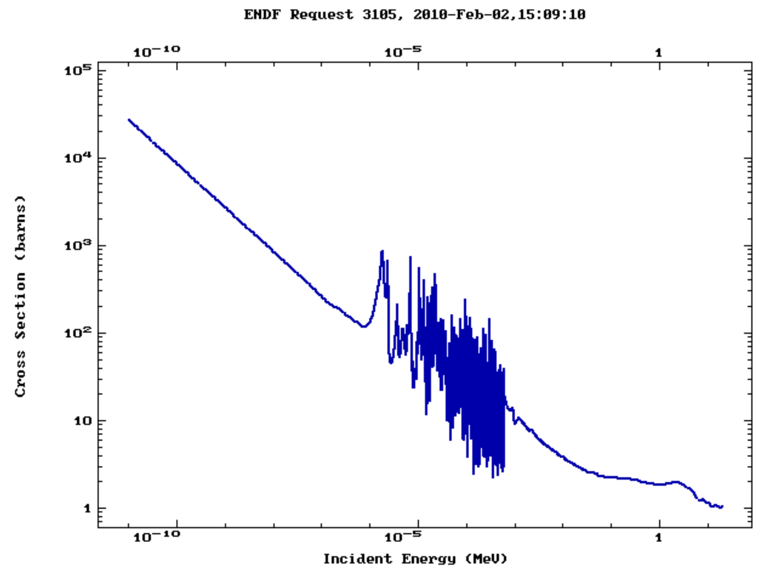
4. Improvement of ADS economy by utilization the fertile thorium 232

18. Neutron capture cross-section of ^{233}U

Fig. 19. Neutron fission cross-section of ^{233}U





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4. Improvement of ADS economy by utilization the fertile thorium 232

- The smaller is the neutron energy the higher is the fission probability of ^{233}U (see fig. 19). This let us infer that thorium based fuel is suitable to build thermal breeder reactors.
- this infer is supported by the fact that for the fissile ^{233}U, the number of neutrons liberated per neutron absorbed (represented as η) is greater than 2.0 over a wide range of thermal neutron spectrum, unlike ^{235}U and ^{239}Pu (see Fig. 20). The capture cross-section of ^{233}U is much smaller (46 barns) than the ^{235}U (101 barns) and ^{239}Pu (271 barns) for thermal neutrons, while the fission cross-section of all the three isotopes is of the same order (525, 577 and 742 barns for ^{233}U, ^{235}U and ^{239}Pu respectively). Thus, non-fissile absorption leading to higher isotopes (^{234}U, ^{236}U and ^{240}Pu respectively) with higher absorption cross-sections is much less probable.
- Taking under consideration the above data we can see (note) that the ^{232}Th – ^{233}U fuel cycle let us obtain breeding of fissile atoms (material) both in fast, epithermal and thermal neutron spectra.

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4. Improvement of ADS economy by utilization the fertile thorium 232

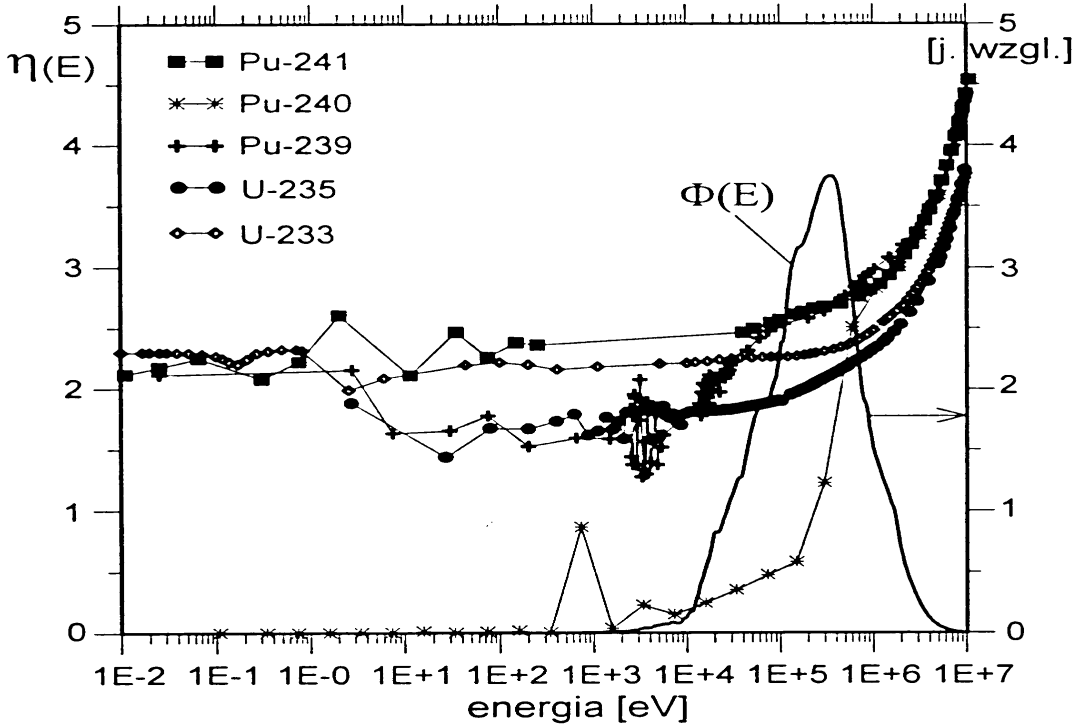

Fig. 20. Number of neutrons η(E) liberated per neutron absorbed by the ^{233}U, ^{235}U ^{239}Pu, ^{240}Pu, ^{241}Pu actinides (Φ(E) – neutron spectrum in the lead cooled system)

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4. Improvement of ADS economy by utilization the fertile thorium 232

- This inference that the thermal breeder reactor can be constructed is clearly seen in the Fig.20a [7]. The authors [7] underline that as long as the available neutron numbers for breeding(N_b) is always slightly larger than 0, breeding is possible. This is the case for the whole neutron energy spectrum for uranium 233 where N_b is always equal 0.3

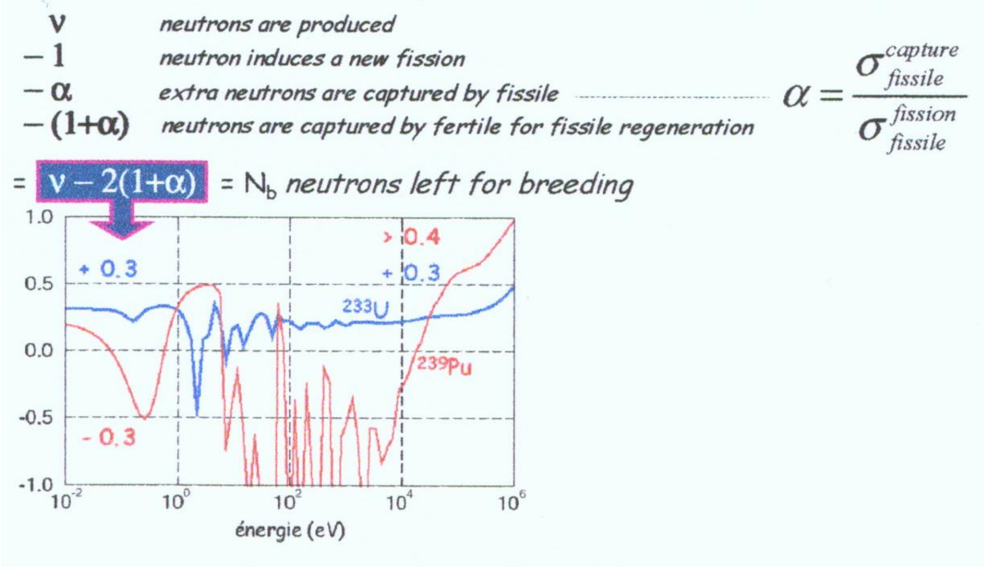

Fig.20a. Available neutrons for breeding both for ^{233}U and ^{239}Pu.

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4. Improvement of ADS economy by utilization the fertile thorium 232

- Regarding safety aspects, the prominent feature of the ADS is its reduced potential for reactivity induced accidents. From the other side it is underlined that, the ADS design must certainly find a way to reduce burn-up reactivity loss per unit energy release. This can be reached if we [8]:
- - Compensate for declining reactivity with frequent partial core refuelling as burn-up occurs.
- - Load excess fissile material and then compensate for loss of reactivity by withdrawing
- external neutron-absorbing control rods.
- - Increase the source strength as burn-up occurs.
- - Increase the source effectiveness as burn-up occurs.
- - Some combination of the above.

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4. Improvement of ADS economy by utilization the fertile thorium 232

- The first step – which clearly should be taken – is to increase the critical mass by lowering k_∞ to nearly unity and requiring a larger and less leaky lattice of maximum fissile inventory.
- The second approach is to refuel the core in parts and provide for an adjustable parasitic neutron absorber (a control rod) which can be moved so as to hold sub-critical reactivity constant as fissile content is burned out.
- The third potential approach is to adjust the proton beam current and resulting source strength.
- The better effectiveness of the spallation neutron source can be obtained by moving the spallation target from top of core to core centre.

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4. Improvement of ADS economy by utilization the fertile thorium 232

- In the case when thorium based fuel is applied in the subcritical core which is operated so that the burn-up reactivity loss is reduced by withdrawing external neutron-absorbing control rods keeping the core very slightly subcritical all the time in order to hold the source strength constant, the reactor core can become overcritical during several tenths of days after switching off the ADS.
- The above safety problem and the neutron source strength increase as burn-up occurs and as well as creation of fissile material ^{233}U in function of irradiation time can be preliminary considered by using the layout of YALINA THERMAL (the thermal subcritical core) and application the Monte Carlo methodology.

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4.1 The idea of research the particular safety problem.

- The idea is to choose four configurations (geometry) of the Yalina Thermal subcritical core loaded with a fixed number of 217 fuel rods of EK-10 type.
- **The first configuration** consists solely of 217 fuel rods of EK-10 type (see Fig. 1), **the second configuration** consists of 217 fuel rods of EK-10 type surrounded by 148 thorium rods which are assumed as the EK-10 type with the difference that the uranium dioxide is replaced by the thorium dioxide (see Fig. 2). **The third configuration** also consists of 217 fuel rods of EK-10 type and 148 thorium rods which are placed near about the center of the subcritical core (see Fig.3).
- The fixed number of EK-10 type fuel was chosen in order to facilitate interpretation of the calculation results performed by using the Monte Carlo methodology.
- **The fourth configuration** consists solely of 365 (217 +148) thorium rods (see Fig. 4). The fourth configuration is thought over to consider if the core loaded solely with thorium rods could be operated in the ADS.

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4.1 The idea of research the particular safety problem.

Fig.1. Core configuration with 217 U rods –geometry 1; red dots- EK-10 type fuel**.**

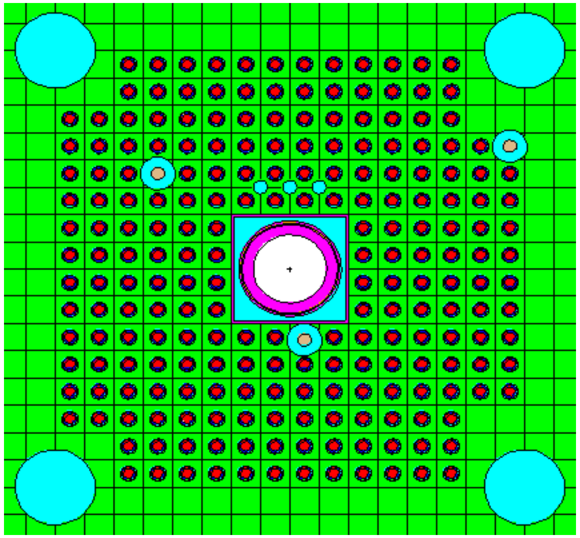
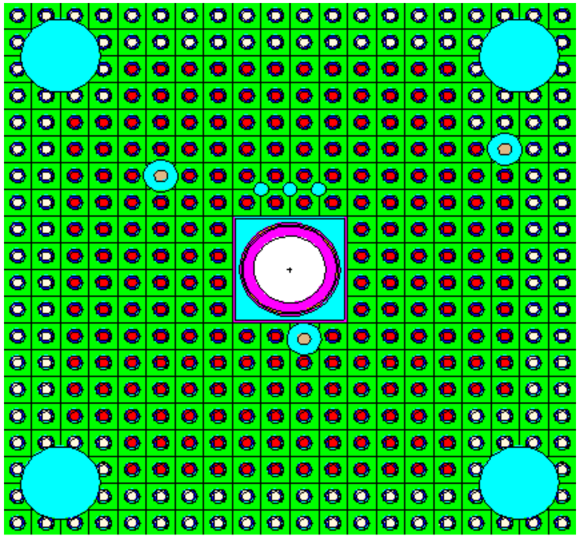


Fig. 2. Core configuration with 217 U rods and 148 Th rods – geometry 2; red dots – EK-10 type fuel; white dots – Th232



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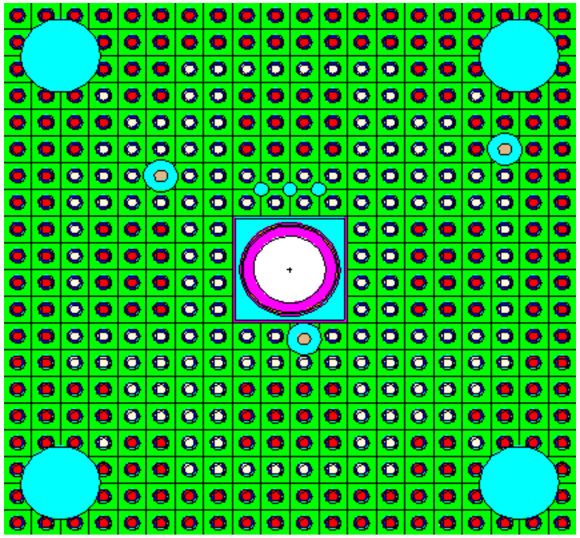
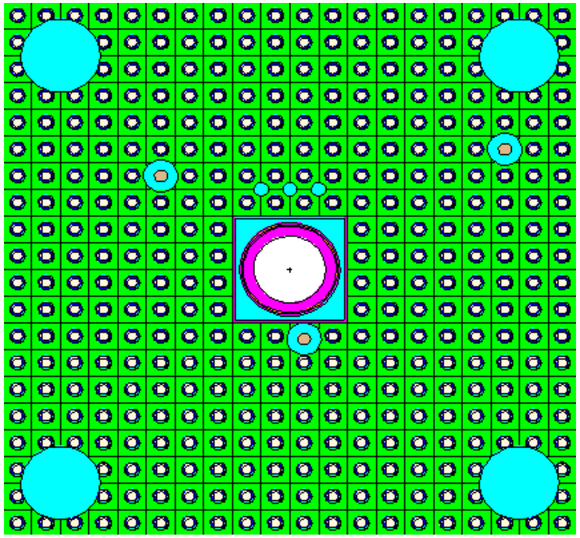
4.1. The idea of research the particular safety problem.

Fig. 3. Core configuration with 217 U rods and 148 Th rods – geometry 3; red dots – EK-10 type fuel; white dots – Th232

Fig. 4. Core configuration with 365 Th232/U233 rods –geometry 4; white dots- Th232/U233





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4.2. Calculation results.

- In order to make analysis of the safety problem mention above we have to compare the multiplication factors K_{eff} change in function of irradiation time for the four configurations. (Fig. 1. – Fig. 4).
- Running the MCNPX code we have assumed to keep constant the power equal to 5 MW what enforced the neutron flux and its increase in function of fissile material burn-up what in turn extort to increase the neutron source strength. The fuel of EK-10 type were designed to operate at the level of power (neutron flux above 10^{13} n/cm^2·s).
- The external neutron source was assumed ^{252}Cm.
- These assumptions are completely theoretical and could not be realized in practice but let us to make the analysis mentioned above from the physical point of view.
- Simultaneously we have got information about breeding the fissile material ^{233}U from the fertile material ^{232}Th.

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4.2. Calculation results.

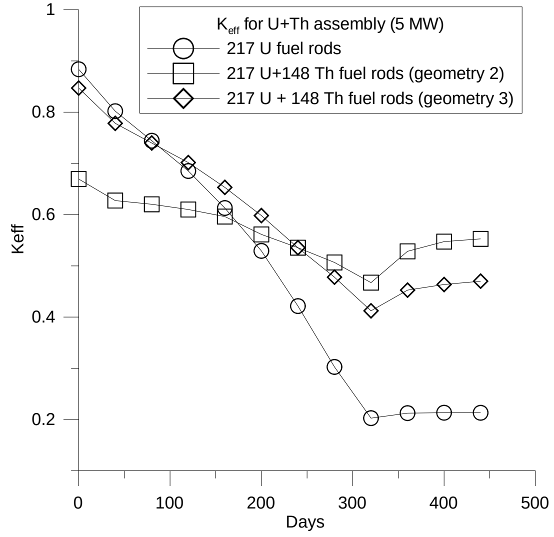

Fig.5. Multiplication factor K_{eff} in function of time for three configurations (Fig. 1, Fig. 2, Fig. 3). Constant power of 5 MW during 320 days and after the power is zero.

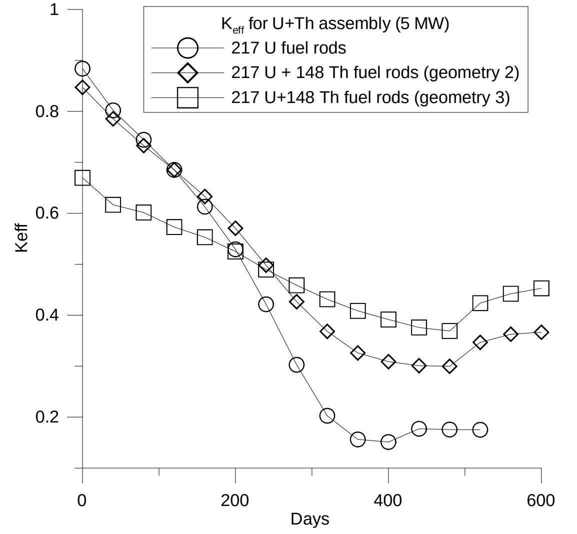

Fig. 6. . Multiplication factor K_{eff} in function of time for three configurations (Fig. 1, Fig. 2, Fig. 3). Constant power of 5 MW during 480 days and after the power is zero

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4.2. Calculation results.

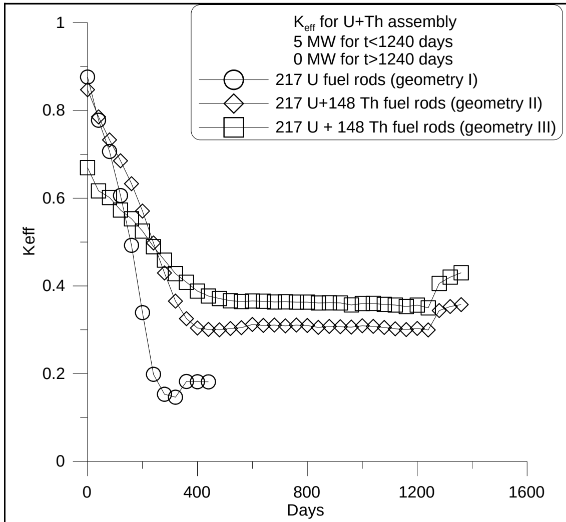

Fig. 7. Multiplication factor K_{eff} in function of time for three configurations (Fig. 1, Fig. 2, Fig. 3). Constant power of 5 MW during 1240 days and after the power is zero.

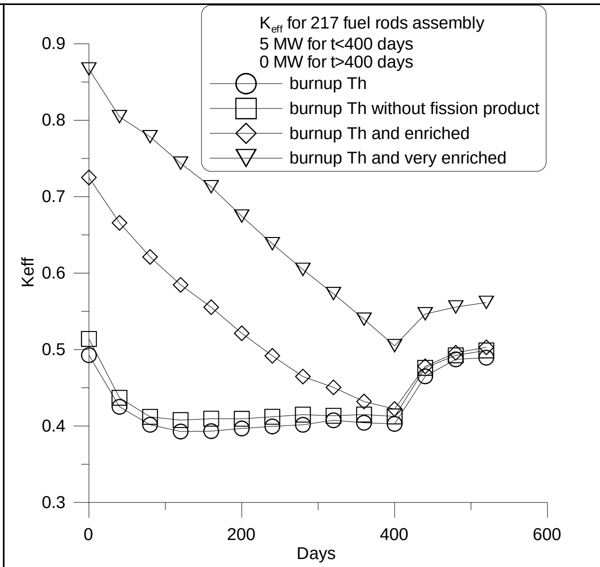

Fig. 8. Multiplication factor K_{eff} in function of time for three different amount of ^{233}U in fuel rod (3.42, 6.73,9.95 g) for configuration from Fig. 1. Constant power of 5 MW during 400 days and after the power is zero

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4.2. Calculation results.

- Figures 5,6 and 7 show that the effective multiplication factor K_{eff} of geometry 2 does not differ significantly from the K_{eff} of geometry 1. It means that 148 thorium rods located in the reflector have small impact on K_{eff}. This location can be a good one for generation the ^{233}U. However, the near about core center location of the 148 thorium rods (geometry 3) have significant negative impact on K_{eff}. Furthermore, after about 400 days of irradiation on power 5 MW the multiplication factor K_{eff} stop to decrease what means that the fission rate of ^{233}U is equal to the production rate of the fissile material.
- Four curves in Fig. 8 simulate change of multiplication factor K_{eff} in function of irradiation time (burn-up). Three of the curves refer to the chemically separated ^{233}U fuel from the used fuel and used in a new nuclear fuel with three different weights 3.42, 6.73 and 9.95 g in a rod. The fourth curve marked with circles refers to the ^{233}U of weight 3.42 g in a rod and utilized in situ. This curve after 120 days reaches the lowest K_{eff} value and increases very slightly what we can infer that the number of nuclei that fission is smaller than the number of fissile nuclei created by neutron capture on the fertile nuclei. It means that the condition of breeding is obtained.

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4.2. Calculation results.

- Quite surely the steady state of protactinium density assign one of the breeding condition. Kinetic of reaching the fissile uranium steady state assigns the kinetic of obtaining the breeding condition. The calculations are done for the assumption that into the configuration from Fig. 1 are loaded solely the thorium based fuel rods and that the power remains constant 5 MW during 400 days and after the power is equal zero. The multiplication factor K_{eff} decreases with burn-up and after switching off the power the factor increases. There is very small difference between the change of multiplication factor K_{eff} for the ^{233}U generated utilized in situ and the chemically generated fuel. It means that a once through fuel cycle (open fuel cycle) can be applied.
- Alike simulation as in Fig. 8 was done with the difference that the configuration from Fig. 4 are loaded solely the thorium based fuel rods. In this case the core became over critical for the ^{233}U weight of 9.95 g in a rod.

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4.2. Calculation results.

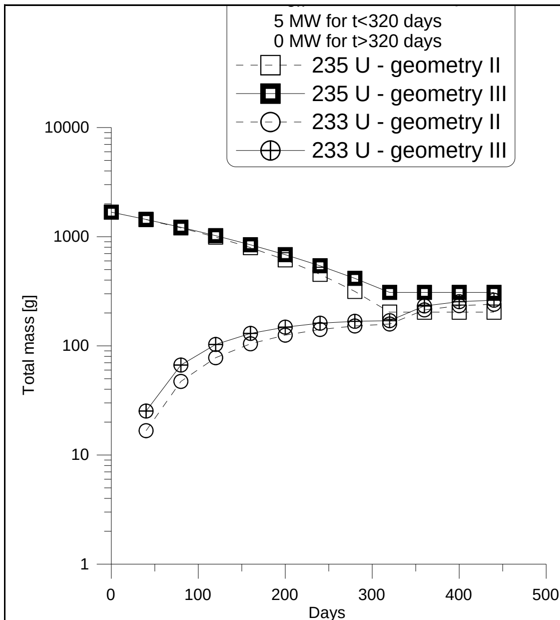

Fig. 9. Total mass of ^{235}U and ^{233}U in function of time for two configurations (Fig. 2, Fig. 3). Constant power of 5 MW during 320 days and after the power is zero.

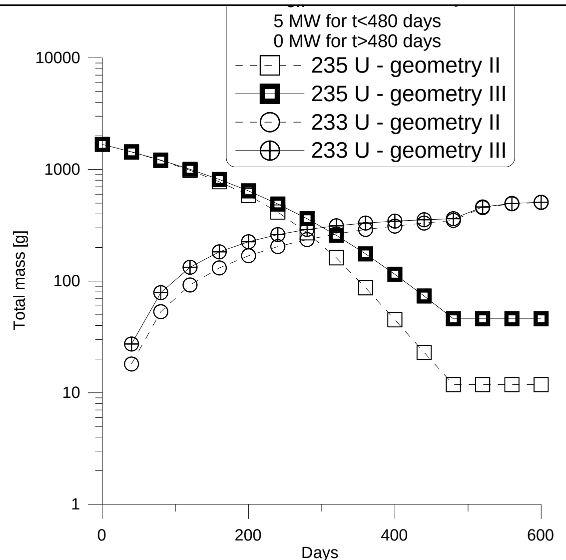

Fig. 10. Total mass of ^{235}U and ^{233}U in function of time for two configurations (Fig. 2, Fig. 3). Constant power of 5 MW during 480 days and after the power is zero.

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4.2. Calculation results.

- Figs 9 and 10 presents the burn-up of ^{235}U and the generation of ^{233}U in function of irradiation time for the configurations from Fig, 2 and Fig. 3 for different times of irradiation 320 days and 480 days.
- The generation rate of ^{233}U in function of irradiation time for different location of the thorium rods in the core shows that is similar. Remembering however that the location of the thorium rods in the core have a big impact on the value of multiplication factor K_{eff} it is profitable to generate the thorium based fuel in the reflector location.

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4.2. Calculation results.

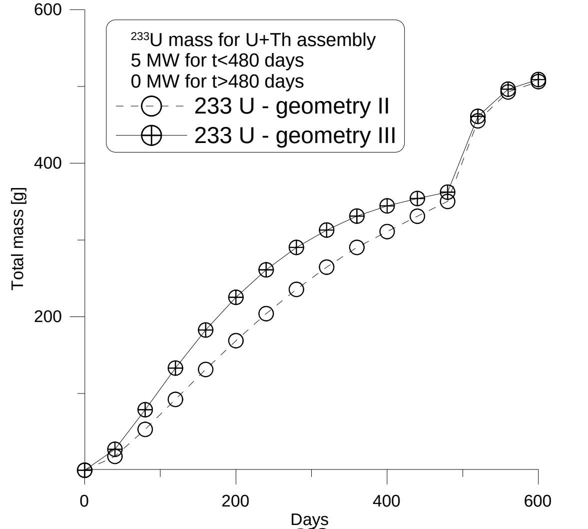

Fig. 11. Total mass of ^{233}U in function of time for two configurations (Fig. 2, Fig. 3). Constant power of 5 MW during 480 days and after the power is zero

The generation of ^{233}U in function of irradiation time for the configurations from Fig, 2 and Fig. 3 for the time of irradiation 480 days presented in Fig. 11 in linear-linear diagram shows that the ^{233}U increases from 360 g to 500 g after switching off the power. The increase is about 38% what in the case of keeping the core very slightly subcritical during operation does not guarantee safety of the ADS after switching off the power.

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4.2. Calculation results.

- Another observation is that the generation of ^{233}U per rod reaches 3.37g/rod while the fresh fuel rod of EK-10 type have 8g/rod of ^{235}U.
- This suggests that the generated ^{233}U fuel can be utilized as improving the exploitation economy in a once through fuel cycle by shuffling the fuel element assemblies in the core.
- Forming the new nuclear fuel properly enriched with ^{233}U and mixed with thorium let to obtain higher burn-up in the thermal nuclear powers.

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5. Conclusions

- The sub-critical assembly of ADS should consist of three zones for transmutation of radioactive wastes: thermal, resonance and high energy neutron.
- Moreover high flux thermal neutron environment ($\geq 10^{16}$ n/cm$^3\cdot$s) is convenient for transmutation most of the radioactive waste both LLFP and MA
- If the ADS, beside the transmutation of fission products and incineration of actinides, is to be used for production of electric power then it should be a big power unit because the own needs of the accelerator and the power unit are significant.
- The ^{232}Th – ^{233}U fuel cycle let us obtain breeding of fissile atoms both in fast, epithermal and thermal neutron spectra.
- Thermal breeder reactor can be constructed
- In the case when thorium based fuel is applied in the subcritical core which is operated so that it is keeping the core very slightly subcritical all the time the reactor core can become overcritical after switching off the ADS during several tenths of days.

- Thank you for the attention.