NEUTRON-PHYSICAL CHARACTERISTICS OF BLANKET OF HYBRID FUSION NEUTRON SOURCE BASED ON SOLUTION OF THORIUM NITRATE AND MINOR ACTINIDES IN HEAVY WATER

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Abstract

In the Russian Federation, the creation of a fusion neutron source (FNS) is a priority in the implementation of the federal target program for the development of new-generation nuclear energy technologies. The Kurchatov Institute considers the Tokamak T-15MD as a prototype source of thermonuclear neutrons. The development of a hybrid blanket for a fusion neutron source is an important part of the tasks being solved. In this paper, the possibilities of using a hybrid blanket consisting of a solution of thorium nitrate in heavy water are investigated. The obtained estimates of the neutron-physical characteristics show that thorium nitrate enriched with the isotope ^{15}N is able to replace thorium dioxide, which has a high abrasiveness. The production of uranium-233 and tritium in a solution of thorium nitrate and minor actinides makes it possible to achieve fuel self-sufficiency of the machine in operation mode with a guaranteed high degree of safety (keff ~ 0.95). Due to the high proportion of thermal neutrons in the energy spectrum of FNS, transmutation in the radiation capture reaction makes it possible to completely ignite the loading of neptunium-237 and americium isotopes in a safe mode at keff ~ 0.95 in 1.5–2 years. At the same time, valuable elements used in industry and medicine are formed. If the enrichment of the blanket with uranium-233 is increased to keff ~ 0.98, then these isotopes can be burned out within a year. At keff ~ 0.95, the machine has an electrical capacity of about 80~130 MW, which corresponds to the parameters of a low-power reactor.

1. INTRODUCTION

The Russian program for the development of new-generation nuclear energy technologies includes the creation of fusion neutron sources (FNS) [1–8]. The use of neutron sources for burning minor actinides and producing fuel for nuclear power is considered in Russia as one of the promising ways to obtain faster returns from research in the field of controlled thermonuclear fusion and the use of energy in real conditions for practical demonstration of the energy potential of thermonuclear reactions. In the Russian Federation, the Kurchatov Institute considers the Tokamak T-15MD as a prototype source of fusion neutrons [9].

In a hybrid fusion neutron source using fusion and fission reactions, the plasma is surrounded by a blanket filled with thorium-232. When thorium-232 is irradiated with neutrons, uranium-233 is formed. As a fuel for a nuclear reactor, it has an advantage over uranium-235, since it does not lead to the accumulation of long-lived minor actinides (MA) with a half-life of hundreds of thousands of years, which have to be disposed of for burial. The amount of actinides formed from uranium-235 in thermal nuclear reactors can be burned in the same blanket. The result is nuclides with a half-life of only hundreds of years, and these nuclides will become safe fairly quickly. In addition, lithium can be converted to tritium in a blanket. The hybrid system does not need either a full-fledged nuclear or fusion reactor. The tokamak in it serves only as a source of neutrons, which trigger the nuclear decay of the fuel in the external blanket. There is no need for a stable thermonuclear reaction, so there is no need to comply with the Lawson criterion anymore, and it is enough to heat the deuterium-tritium plasma to relatively moderate temperatures, 30–50 million degrees. Neutrons are formed due to the interaction of beams of deuterium atoms accelerated in injectors with this plasma. The nuclear part of the hybrid is also simplified. The decay of fuel in it should not be self-sustaining, it is stimulated by neutrons coming out of the deuterium-tritium plasma.

2. THE PURPOSE AND OBJECTIVES OF THE STUDY

The main purpose of the work is to study the possibilities of transmutation of minor actinides and the production of nuclear fuel (uranium–233 and tritium) in a FNS blanket consisting of a solution of thorium nitrate in heavy water.

In order to achieve this goal, the following tasks were identified in the framework of this study:

- Justification of the use of thorium nitrate solution in heavy water as a FNS blanket material;
- Creation of a three-dimensional neutron-physical model of a FNS machine with an aqueous blanket containing thorium nitrate to perform Monte Carlo calculations;
- Comparison of the calculated neutron-physical characteristics of a heavy-water blanket with thorium dioxide and a blanket with thorium nitrate;
- Calculation of the production of nuclear fuel (uranium-233 and tritium) in the FNS model when loading minor actinides;

5. Calculation of the transmutation of minor actinides in an aqueous blanket.

3. CALCULATION METHOD AND CALCULATION MODEL OF FNS

The calculations were performed using the MCNP-4 code [10] based on the Monte Carlo method. Nuclear data from the ENDF-B-VII point section library was used [11].

The calculation model of FNS with an aqueous blanket is shown in Fig. 1–2.

The model of the neutron source and the geometry of the machine are based on the design parameters of the thermonuclear neutron source presented in [12].

The outer contour of the model has the shape of a cylinder with a radius of 201.8 cm and a height of 301.6 cm. Inside the circuit there are: a source of thermonuclear neutrons (plasma); a central column of copper, simulating an inductor; a vacuum chamber surrounded on all sides by the first wall; a solid–state blanket of lead, which is a multiplier of fast neutrons; coils of a poloidal magnetic field (PMFC); a blanket of thorium nitrate solution in heavy water in a ratio of $^{232}Th(NO_3)_4$ (61.43 g) + D_2O (100 g), which generates fissile nuclides and heat; a shell made of lithium $^6Li_2O$ oxide, a shell made of lithium ^6Li isotope along the outer contour of a model for the production of tritium, and neutron field characteristic detectors.

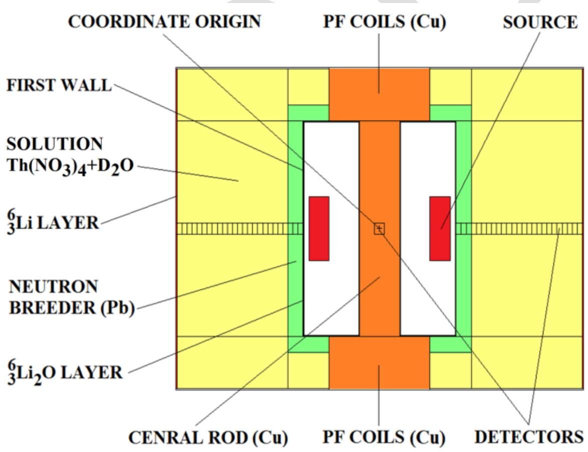


FIG. 1.Vertical section of the calculation model.

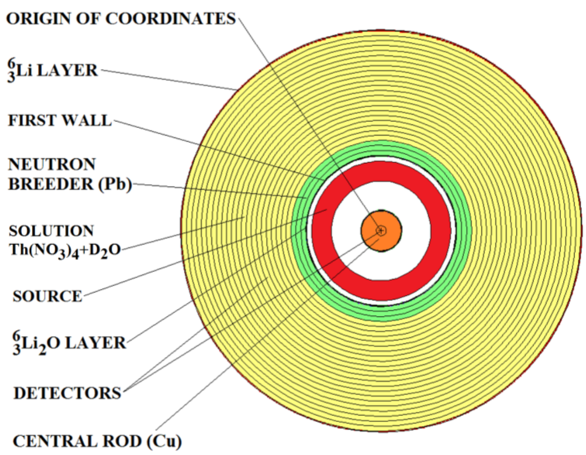


FIG. 2. Horizontal section of the calculation model.

The spatial distribution of neutrons over the source volume is assumed to be uniform. The source is defined as monoenergetic with the neutron energy of the D-T reaction equal to 14.1 MeV. The radiation intensity is 1.775×10^{18} n/s, which corresponds to a nuclear fusion power of 5 MW. It is assumed that the angular distribution of the source is isotropic.

4. JUSTIFICATION OF CHOICE OF AQUEOUS BLANKET MATERIAL

In an early version of the FNS model, a slurry suspension of thorium dioxide Th_2O in the ratio $^{232}ThO_2$ (34.95 g) + D_2O (100 g) was used in a heavy-water blanket [12]. Thorium dioxide is insoluble in water, so it was assumed that its finely dispersed phase would be created. However, there is no evidence of obtaining a stable suspension of this composition [13], which means that its use during the operation of FNS can lead to accelerated destruction of equipment due to the abrasive action of thorium dioxide particles.

Instead of thorium dioxide in the FNS aqueous blanket, it is proposed to use thorium nitrate enriched in the isotope ^{15}N. Thorium nitrate is the most water-soluble salt of thorium. Its maximum solubility is 190.7 g per 100 g of water at 20 °C, which ensures the creation of a homogeneous solution [14, 15].

Thus, the use of thorium nitrate in a heavy-water blanket solves the problem of increased wear of equipment, which is characteristic of a suspension of thorium dioxide, which has a high abrasiveness.

The thermal decomposition of thorium nitrates has been studied in [16, 17].

Dehydrated thorium nitrate $Th(NO_3)_4$ decomposes at a temperature of 400 °C to form thorium dioxide ThO_2, nitrogen oxide NO_2 and oxygen O_2.

When heating $Th(NO_3)_4 \times 4H_2O$ hydrate in air at 110–120 °C, thorium oxynitrate hydrate $ThO(NO_3)_2 \times 0.5\ H_2O$ is formed. At temperatures above 360 °C, thorium oxynitrate decomposes to form thorium dioxide ThO_2.

The operating temperature range of the FNS aqueous blanket is comparable to the temperature range of the coolant of the VVER reactor. The temperature of the coolant in the VVER-1000 reactor is: at the reactor inlet – 289.7 °C, at the outlet – 320 °C [18]. Thus, thorium dioxide is not formed in the operating temperature range of the coolant typical for the VVER-1000.

The isotope ^{15}N has a uniquely low absorption cross section for both thermal and fast neutrons. Its use in thorium nitrate $Th(^{15}NO_3)_4$ significantly increases the proportion of radiation neutron capture by thorium as a result of suppression of the competing process on nitrogen. This makes it possible to significantly improve the neutron balance of the hybrid blanket and radically reduce the amount of radioactive carbon-14 production, which will increase the radiation safety of FNS.

A comparison of the data in Table 1 shows that the neutron-physical characteristics of $^{232}Th(NO_3)_4$ (61.43 g) + D_2O (100 g) blanket with suppressed fission (without ^{233}U enrichment) differ for the better from the corresponding parameters of a blanket with a suspension of $^{232}ThO_2$ (34.95 g) + D_2O (100 g). Minor actinides were absent in the composition of both blanks.

For a nuclear fusion power of 5 MW, the production of ^{233}U is 14.43 kg/year in the $^{232}ThO_2$ blanket and 15.88 kg/year in the $^{232}Th(NO_3)_4$ one, which is 10% more. The production of tritium is 1.079×10^{-1} kg/year in the $^{232}ThO_2$ blanket and 1.880×10^{-1} kg/year in the $^{232}Th(NO_3)_4$ one, which is 74% more. At the same time, the production of tritium in $^6Li_2O$ in the model with the $^{232}Th(NO_3)_4$ blanket is 1.652×10^{-1} kg/year that is 1.5 times greater than for the blanket with $^{232}ThO_2$ (9.967×10^{-2} kg/year). In the ^6Li shell, the production of tritium for a blanket with $^{232}Th(NO_3)_4$ (8.221×10^{-2} kg/year) is 7.74 times less than for a $^{232}ThO_2$ blanket (1.062×10^{-2} kg/year).

TABLE 1. REACTION RATES AND PRODUCTION OF NUCLEAR FUEL IN FNS MODELS WITH $^{232}THO_2$ AND $TH(^{15}NO_3)_4$ BLANKET (0% ^{233}U) WITHOUT MINOR ACTINIDES (MA). THE POWER OF THE SOURCE OF D-T NEUTRONS IS 1 N/S FOR REACTION RATES AND $1,775\times10^{18}$ N/S FOR FUEL GENERATION

Material of the aqueous blanket	$^{232}ThO_2$ (34.95 g) + D_2O (100 g)	$^{232}Th(NO_3)_4$ (61.43 g) +D_2O (100 g)
Volume of the aqueous blanket, m^3	15.6987	
Total amount of ^6Li, kg	341.255	
^{233}U enrichment (wt. %)	0	0
Loading of MA, kg	0	0
Loading of ^{232}Th, kg	5.137×10^3	5.116×10^3
Loading of ^{233}U, kg	0	0

Neutron yield per 1 fission of the ^{232}Th nucleus	3.189	3.232
k_{eff}	<<1	<<1
(n, γ) – ^{232}Th	8.278×10^{-1}	8.728×10^{-1}
(n, f) – ^{232}Th	1.015×10^{-3}	1.171×10^{-3}
$(n, \nu f)$ – ^{232}Th	3.237×10^{-3}	3.811×10^{-3}
$(n, 2n) + (n, 3n)$ – ^{232}Th	3.784×10^{-3}	4.479×10^{-3}
(n, t) – $^{6}Li_2O$	3.670×10^{-1}	5.925×10^{-1}
(n, t) – ^{6}Li	3.027×10^{-2}	3.810×10^{-2}
(n, t) – total	3.973×10^{-1}	6.310×10^{-1}
Leakage	1.097×10^{-2}	6.849×10^{-3}
^{3}H production, kg/year ($^{6}Li_2O$)	9.967×10^{-2}	1.652×10^{-1}
^{3}H production, kg/year (^{6}Li)	8.221×10^{-2}	1.062×10^{-2}
^{3}H production, kg/year (total)	1.079×10^{-1}	1.760×10^{-1}
^{3}H consumption, kg/year	$\sim3.000\times10^{-1}$	$\sim3.000\times10^{-1}$
Reproduction rate of ^{3}H	3.598×10^{-1}	5.867×10^{-1}
^{233}U production, kg/year	1.443×10^{1}	1.874×10^{1}
^{233}U consumption, kg/year	0	0
Reproduction rate of ^{233}U	0	0
Thermal power of the ^{232}Th blanket, MW	1.679	7.151×10^{-1}
Electrical power, MW	5.597×10^{-1}	2.384×10^{-1}
Thermal power density, MW/m3	1.070×10^{-1}	4.555×10^{-2}
Maximum statistical uncertainty, %	0.08	0.4

5. TRANSMUTATION OF MINOR ACTINIDES AND PRODUCTION OF NUCLEAR FUEL IN A FNS BLANKET WITH THORIUM NITRATE SOLUTION IN HEAVY WATER

In the thorium fuel cycle, which is used in FNS, minor actinides are formed as follows. When irradiated with a thermal neutron, uranium-233 can capture it and form a new transuranic element, uranium-234. The isotope uranium-234, in turn, will capture a neutron and turn into a fissile isotope uranium-235. As a result of transmutations of uranium-235, neptunium-237 and heavy isotopes of plutonium are formed, which partially transform into americium and curium.

In order for new actinides to form, thorium needs to capture 5 neutrons, which is five times more than for uranium-238. The probability of such capture is 8% [19], therefore, transuranic elements in a thorium reactor are produced in small quantities compared to a reactor using uranium as fuel. These MA can be disposed of in a closed nuclear fuel cycle.

Heavy metal nitrates, including minor actinides, are highly soluble in water [20].

In the blanket spectrum with a solution of thorium nitrate $^{232}Th(NO_3)_4$ in heavy water, effective MA transmutation with the production of nuclear fuel and the accumulation of rare isotopes is possible. When a blanket is enriched with an isotope of uranium-233, a noticeable proportion of thermal neutrons is present in the energy spectrum (Fig. 3). Table 2 shows the results of transmutation of minor actinides in the aqueous blanket for one year (365 days) of operation of the machine at a plasma neutron source power of 5 MW (1.775×10^{18} n/s), depending on the degree of enrichment level of uranium-233. A graphical representation of the change in the mass of MA burned in 365 days is shown in Fig. 4. The initial MA load was determined by the amount of minor actinides produced by the average VVER-1000 reactor per year (365 days of operation) [21].

TABLE 2. PRODUCTION OF NUCLEAR FUEL AND TRANSMUTATION OF MINOR ACTINIDES IN $^{232}TH(NO_3)_4$ HEAVY-WATER BLANKET DEPENDING ON THE DEGREE OF ^{233}U ENRICHMENT FOR 1 YEAR (365 DAYS) OF MACHINE OPERATION AT A NEUTRON SOURCE POWER OF 5 MW (1.775×10^{18} N/S)

The mass of minor actinides in a thorium blanket with a total load of 21 kg is 9.345 kg of ^{237}Np, 10.214 kg of ^{241}Am, 1.281 kg of ^{243}Am, 4.2×10^{-3} kg of ^{243}Cm, 1.554×10^{-1} kg of ^{244}Cm				
Material of the aqueous blanket	$^{232}Th(NO_3)_4$ (61,43 g) + D_2O (100 g)			
Volume of the aqueous blanket, m^3	15.6987			
Total amount of ^{6}Li, kg	341.255			
^{233}U enrichment (wt. %)	0	1.6	1.7	1.8
Loading of ^{232}Th, kg	5.095×10^{3}	5.014×10^{3}	5.009×10^{3}	5.004×10^{3}

Loading of ^{233}U, kg		0	8.153×10^{1}	8.662×10^{1}	9.172×10^{1}
k_{eff}		0.004	0.926	0.952	0.983
Reactivity $\rho = (k_{eff} - 1)/k_{eff}$		—	–0.079	–0.050	–0.017
Neutron yield per 1 fission of the ^{233}U nucleus		—	2.498	2.498	2.498
Burning of minor actinides, kg/year	^{237}Np	6.872×10^{-1}	4.793	7.672	1.946×10^{1}
	^{241}Am	2.909	1.991×10^{1}	3.184×10^{1}	8.075×10^{1}
	^{243}Am	8.458×10^{-2}	7.081×10^{-1}	1.145	2.922
	^{243}Cm	1.138×10^{-3}	7.492×10^{-3}	1.196×10^{-2}	3.025×10^{-2}
	^{244}Cm	2.728×10^{-3}	2.445×10^{-2}	3.955×10^{-2}	1.012×10^{-1}
^{233}U production, kg/year		1.588×10^{1}	$1{,}097\times10^{2}$	$1{,}755\times10^{2}$	$4{,}449\times10^{2}$
^{233}U consumption, kg/year		0	1.087×10^{2}	1.842×10^{2}	4.926×10^{2}
Reproduction rate of ^{233}U		0	1.009	0.953	0.903
^{3}H production, kg/year		0.188	0.417	0.553	1.117
^{3}H consumption, kg/year		$\sim3.000\times10^{-1}$	$\sim3.000\times10^{-1}$	$\sim3.000\times10^{-1}$	$\sim3.000\times10^{-1}$
Reproduction rate of ^{3}H		0.627	1.390	1.843	3.273
Thermal power of the ^{232}Th blanket, MW		1.931	2.442×10^{2}	4.123×10^{2}	1.100×10^{3}
Electrical power, MW		6.437×10^{-1}	8.141×10^{1}	1.374×10^{2}	3.667×10^{2}
Thermal power density, MW/m^{3}		1.230×10^{-1}	1.556×10^{1}	2.626×10^{1}	7.007×10^{1}
Maximum statistical uncertainty, %		0.1	0.8	1	3

The data in Table 2 show that at a ^{233}U content of 1.6–1.7%, the reproduction coefficient of uranium-233 is close to 1. With such blanket enrichment, the machine can reach an electrical power of about 80–130 MW, which corresponds to the one of a low-power reactor. At the same time, complete reproduction of tritium is achieved. With an increase in the content of ^{233}U in the aqueous blanket, the amount of all minor actinides burned increases:

— ^{237}Np − when uranium-233 is enriched at 1.6% per year (365 days), 51% of the total neptunium content is burned. With a further increase in the ^{233}U content, the amount of the isotope burned increases rapidly. At 1.7% of uranium-233, 82% of neptunium is burned per year. When 1.8% enrichment is reached, 19 kg of neptunium is burned per year, with its initial loading of 9 kg. This means that the entire initial ^{237}Np download is burned in 172 days;
— ^{241}Am – already at 1.37% of uranium-233, the combustion of all americium loaded into a blanket (10 kg) is achieved per year. With a maximum enrichment of 1.8%, the entire actinide load can be burned in 46 days;
— ^{243}Am – 55% is burned per year at 1.6% of uranium-233, 55%, and 89% is burned at 1.7%. At 1.8% ^{233}U, the entire 1.281 kg load is burned in 160 days;
— ^{243}Cm and ^{244}Cm – an increase in the content of ^{233}U leads to a different result of the combustion of these isotopes. The entire ^{243}Cm (4 g) isotope load is completely burnt out in a year at 1.48% enrichment. At maximum enrichment (1.8% of ^{233}U), ^{244}Cm does not burn completely, the amount of actinide burned per year is 65% (100 g of the initial load of 155 g).

The peculiarity of the burning of MA in the thermal part of the spectrum is the high cross-section of the radiation capture reaction (n,γ) relative to the cross-section of the fission reaction (n,f). The ratio of burnt actinide in the capture reaction and the fission reaction for some MA isotopes is shown in Fig. 5.

It can be seen from the graphs that for all isotopes except curium-243, the rate of the capture reaction is several times higher. This allows the minor actinide to be ignited faster using the thermal spectrum. The isotope curium-243 has a higher rate of (n,f) reaction, it belongs to fissile isotopes.

The isotopes ^{238}Pu, ^{242}Am, ^{244}Am, ^{244}Cm, and ^{245}Cm are formed in the FNS blanket through the radiation capture reaction. These are expensive artificial radioactive, including fissile isotopes, which are in demand in space research and medicine. The cost of some of them exceeds the market value of precious metals by an order of magnitude [20].

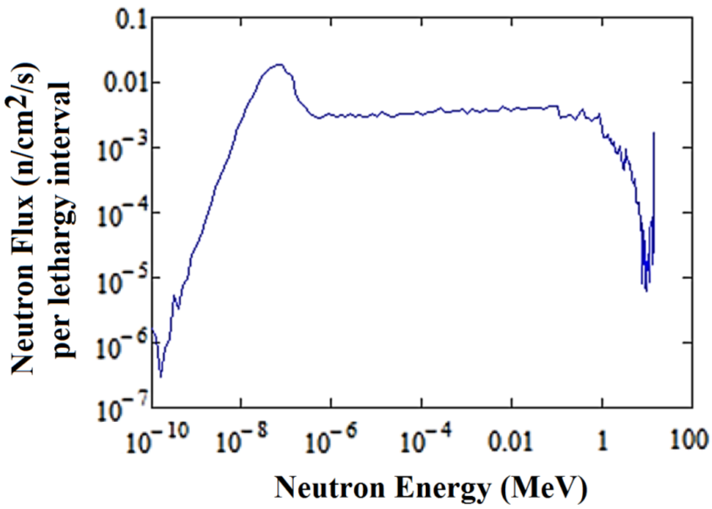


FIG.3. The averaged neutron flux energy spectrum in the heavy water blanket ($Th(NO_3)_4 + D_2O$) at keff = 0.95 (1.7% ^{233}U at a neutron source power of 1 n/s.

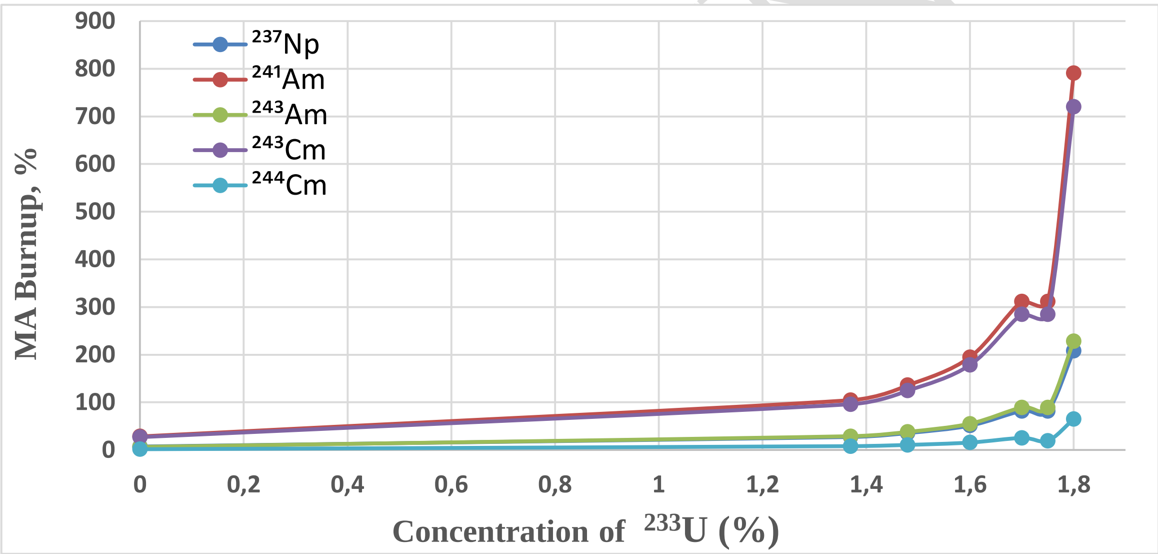
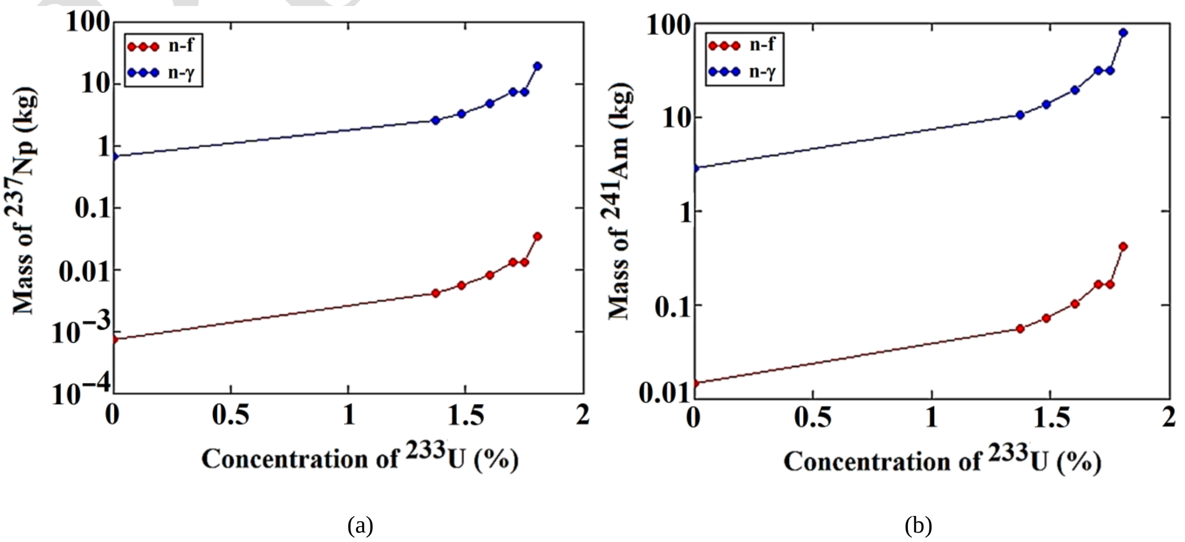
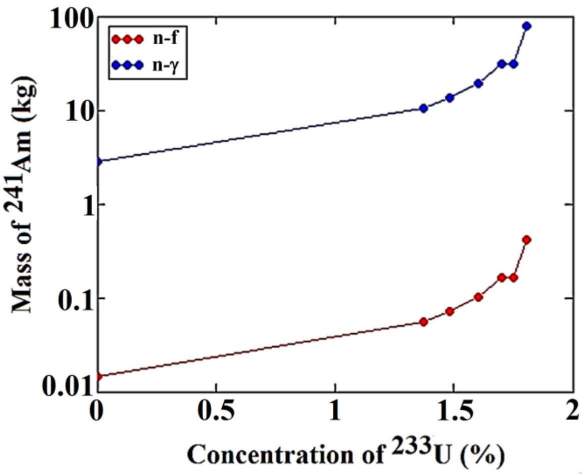


FIG.4 MA burnup in 365 days at a neutron source power of 5 MW (1.775×10^{18} n/s) depending on the degree of enrichment of the blanket with uranium-233.





(c)

FIG.5. Mass of ^{237}Np (a) ^{241}Am (b), and ^{243}Cm (c) isotope burned up as a result of radiation capture and fission reaction in 365 days at a neutron source power of 5 MW (1.775×10^{18} n/s) with an initial load of 9 kg of ^{237}Np, 10 kg of ^{241}Am, and 4 g of ^{243}Cm.

6. CONCLUSION

A comparison of the neutron-physical characteristics of a heavy-water blanket with a solution of thorium nitrate and a suspension of thorium dioxide shows that thorium nitrate enriched with the isotope ^{15}N is able to successfully replace thorium dioxide, which has a high abrasiveness.

The production of uranium-233 and tritium in a solution of thorium nitrate and minor actinides in the mode of the deep subcritical state of the blanket (keff ~ 0.95) makes it possible to achieve fuel self-sufficiency.

At keff ~ 0.95, the machine has an electrical power of about 80–130 MW, which corresponds to the one of a low-power reactor.

Transmutation in the FNS spectrum with a high proportion of thermal neutrons makes it possible to completely ignite the loading of neptunium-237 and americium isotopes in a safe mode (keff ~ 0.95) in 1.5–2 years. The values improve if the enrichment of the blanket with uranium-233 is increased.

The ^{244}Cm isotope does not completely burn up. The calculations show that in the thermal spectrum, the ^{244}Cm load can be burned by 65%. As a result of α-decay with a $T_{1/2}$ period of 18.1 years, ^{244}Cm turns into plutonium, which can be returned to the fuel cycle [22].

The utilization of MA in a fusion hybrid plant makes it possible to successfully ignite actinides due to the high thermal neutron flux density, which can be changed by varying the power of nuclear fusion. It is also possible to regulate the energy spectrum of neutrons by changing the ratio of fuel and moderator in the blanket. The state of the blanket (core) is controlled not by delayed neutrons, but by neutrons emitted by the plasma, which eliminates the loss of control over the blanket's thermal power.

During the burning of MA, isotopes valuable in industry and medicine are formed in the radiation capture reaction. There are many fissionable ones among them, which can be ignited by recycling.

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