COMPUTATIONAL STUDY ON DISTRIBUTION FEASIBILITY OF RADIOACTIVE WASTE TRANSMUTATION IN ACCELERATOR DRIVEN SYSTEM

by

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In this paper, the feasibility of high-level radioactive waste transmutation in accelerator driven system sub-critical reactor assembly, has been studied for two zone's model and with three different core configurations. The inner zone has a fast neutron spectrum and the outer one has a thermal neutron spectrum. The subcritical core is coupled with external neutron source of energy 14 MeV (D-T source). The effects of high level waste isotopes sample (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{244}Cm, and ^{245}Cm) distribution on the neutron spectrum and burnup performance in the inner zone have been investigated and discussed, by proposed three core configurations non-uniform, uniform, and spiral. The burnup calculations have been performed for one-year operation cycle for all the all proposed models. This work shows that one can effectively transmute most of the actual minor actinides isotopes in the inner fast spectrum zone of the proposed system, with optimal distribution of these isotopes.

Key words: burnup, neutron spectrum, actinide, waste transmutation, accelerator driven system, fission rate

INTRODUCTION

The rapid development of nuclear power in many countries (for example, China, India, Korea) [1], will lead to more accumulation of spent nuclear fuel in the future, and the major contributors to the discharged fuel radioactivity are fission products, including the heavy nuclides generated in the core, isotopes of plutonium and isotopes of the so called minor actinides (MA), neptunium, americium and curium. In a closed fuel cycle, U and Pu are recycled and the MA become, in due time, the major source of radioactivity, that long-term radioactivity generated in nuclear reactors must be destroyed or safely deposited. Until now, the only and the main way of environmental protection is deposition of spent fuel. One of the promising ways to decrease radioactivity of fuel cycle and consequent risks, is based on transmutation. During transmutation, radioactive nuclides are transformed under neutron irradiation into short-lived or stable nuclides [2-8].

Transmutation of radioactive waste, especially heavy nuclides in fast neutron spectrum [9] and accelerator driven system [10, 11] is promising and gives rise to the idea of radioactive equivalence of handling with radioactive wastes [9, 12-15].

Usually, the effectiveness of transmutation can be characterized by the value of α which is defined as the ratio of the neutron capture cross-section to the fission cross-section of a given isotope MA: $\alpha = \sigma_c/\sigma_f$ [16]. If this value is less than unity, the transmutation can be considered as effective. A good example of such conception is presented for three Pu isotopes in fig. 1 which were prepared using IAEA' Nuclear Data Services [17]. While ^{239}Pu and ^{241}Pu, which are fissile nuclides, have values of α less than unity, practically in the whole neutron spectrum, the isotope ^{240}Pu has $\alpha < 1$ only for neutron energy exceeding the mean energy of fission spectrum, so, ^{240}Pu can be effectively transmuted only in a system which has a neutron spectrum with a large part of extremely fast neutrons with energy >5 MeV. Usually such spectra appear in ADS which have the external neutron source

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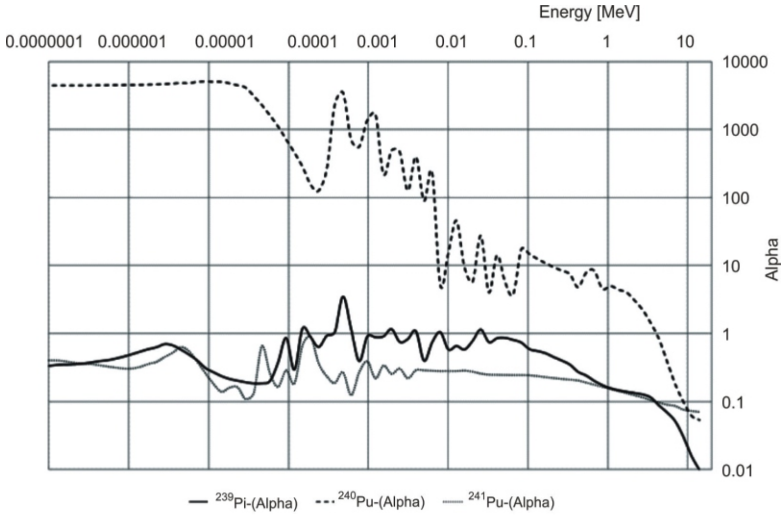

Figure 1. The distribution of the value of *alpha* depending on the neutron energy of plutonium isotopes (^{239}Pu, ^{240}Pu, and ^{241}Pu)

with energy exceeding 10 MeV (D-T source – 14 MeV, spallation source – about 300 MeV in dependence on primer particle energy). Practically the same situation can be observed for all other transuranium isotopes.

On the contrary, long-lived fission products need a large quantity of thermal neutrons to provide an effective transmutation due to capture reaction with a following conversion to a short-lived, or stable isotope. Therefore, the effectiveness of transmutation of a spent fuel is determined not only by the values of α but also by the neutron spectrum of a given facility.

While the MA homogeneous loading in critical reactors has to be limited to avoid the deterioration of some safety related parameters (*e. g.* delayed neutron fraction, Doppler and coolant void coefficients), the subcriticality of the accelerator driven system (ADS) allows operating with rather high MA loadings. In the last decade, within the 5-7th Framework Programmes (FP) of the European Atomic Energy Community (EURATOM), Accelerator Driven Systems were investigated as the *reference* solution for MA transmutation [18].

In the present study, the options for the efficient transmutation of transuranic elements will be discussed, so the focus will be on Pu, Am, and Cm mainly, due to their long-term contribution to the radiotoxicity of the spent nuclear fuel.

All Pu isotopes are radioactive and decay by emitting particles and gamma radiation [13]. The Am is generally considered as a prime candidate for transmutation because it is present in relatively large amounts in the spent fuel and is a significant contributor to gamma activity and radiotoxicity, especially after about 500 years cooling time when the contribution of most fission products has decreased by several orders of magnitude. The ^{241}Am and ^{243}Am have reasonably large neutron cross-sections and are amenable to destruction in an intense and fast neutron flux by a combination of neutron captures and fissions. In irradiated nuclear fuel, ^{241}Am is the dominant nuclide, though there are small but significant quantities of ^{242}Am, and ^{243}Am [19]. Note also that the quantity of ^{241}Am is increased in time due to β-decay of ^{241}Pu (half-life is 14.38 year).

The Cm makes a significant contribution to gamma activity and radiotoxicity and is also a major contributor for neutron emissions. Although ^{242}Cm has a short half-life (163 days), it is continually generated in irradiated fuel from the decay of ^{243}Am (141 year half-life) [20].

It should be noted that there is another contribution to long term radiotoxicity – long-lived fission products such as ^{129}I, ^{99}Tc and other. They also could be transmuted in neutron field by neutron capture with further radioactive decay. But the greatest probability of such processes can be achieved, of course, in thermal neutron spectrum. So we have some contradiction: in order to transmute all components of the spent nuclear fuel we need to have fast and thermal neutron spectra simultaneously. It happens that such situation could be created in a single nuclear facility – a two- zone subcritical system.

It is well known that the two-zone ADS of special configuration has some advantages compared to one-zone system. In particular, the two-zone system, or in general, multi-zone systems amplify neutron flux more effectively from an external source [21-24]. Usually, the design of such a system consists of neutron source in the centrum of the system coupled with accelerator. This neutron source is surrounded by the subcritical reactor core which contains two parts: inner zone (the closest to the neutron source part) and outer zone (physically separated from the inner zone by some membrane). Such configuration allows creating two different neutron spectra in the considered zones if the inner zone contains sufficiently high-enriched fuel with no neutron moderator. The outer zone can contain the usual composition for thermal reactor.

The presented scheme of a two-zone subcritical system driven by a D-T neutron generator is studied as the basis for development of a research subcritical reactor, which will be used in particular to study the transmutation of the entire spectrum of nuclear waste (MA and long-lived fission products). Regarding the choice of external neutron sources, it is well-known fact that proton accelerators are very expensive. The option of

D-T neutron generator is a good solution for designing a real research subcritical reactor, to be used for experiments and testing of different schemes for radioactive waste transmutation, taking into account the possibility to develop powerful neutron generator with intensity 1 10^{-14} s^{-1} [25, 26]. Moreover, the presented scheme of a two-zone subcritical system driven by a D-T neutron generator will have a relatively small cost, and it will be possible to be built and operated by a country with a small gross domestic product such as Ukraine and Egypt.

The purpose of this paper is to study the radioactive waste transmutation in the inner region of the subcritical core modeled into two regions, inner zone and outer zone. Besides, it aims to investigate the feasibility of distribution of the Pu, Am, and Cm samples inside the inner zone by proposing three different configurations and calculating the burn up for each model with the use of Monte Carlo N-particle transport code MCNPX.

DESCRIPTION OF THE SUBCRITICAL CORE

Since the operational life of the VVR-M research nuclear reactor (Kyiv, Ukraine) is near completion, the Academy of Sciences of Ukraine is considering different concepts of research nuclear reactors to replace the VVR-M in future. One option is a subcritical reactor driven by an external neutron source. Main geometrical and material characteristics are based on the previous investigations and papers performed at Institute for Safety Problems of NPP (Kyiv, Ukraine) and Institute for Nuclear Research (Kyiv, Ukraine) [27, 28].

The fast neutron region (Inner zone), with 15 cm radius and 16 cm as external radius surrounding the central tube, is contained in a tank made of stainless steel; this region is composed of shortened fuel pins from VVER-1000 reactor [29] and it uses He as a coolant, the fuel element pitch is 1275 cm, radius of pin's cladding is 0.455 cm, radius of pin's fuel is 0.393 cm; specifically, the fuel in the fuel elements is uranium dioxide with 20 % enriched ^{235}U, the fuel density is 10.96 gcm^{-3}, cladding material of fuel elements is zirconium +1 % niobium, the diameter of fuel element is 0.786 cm, the diameter of cladding material is 0.91 cm.

Inner zone is surrounded by the thermal region that is also composed of shortened fuel pins of VVER-1000 reactor. The enriched uranium dioxide with 4 % enrichment level serves as a fuel in the thermal neutron region; density of the fuel is 10.96 gcm^3, the fuel element pitch in this region is 6 cm, radius of pin's cladding is 0.455 cm, radius of pin's fuel is 0.393 cm. The cladding material of fuel elements is zirconium + 1 % niobium. The square lattice of fuel elements is used in both zones. Around each fuel element there is a gas space that helps to remove the heat. Let the value of the outer zone radius be equal to 63 cm. So, in this case H = 126 cm, because $R_{\text{out}} = H/2$ (in this case we will have minimum leakage). So we obtained k_{eff} = 0.97   0.00063 for our base model.

For the source definition, the charged particles (deuterons) move from top to bottom of the central tube of the system and finally hit the titanium target saturated with tritium. Fusion of a deuterium and a tritium nucleus (D-T nuclear reaction) results in the formation of a He-4 nucleus and a neutron with a kinetic energy of approximately 14.1 MeV. The intensity of neutron source was chosen to be 3.2 s^{-} which was shown in [26] that the D-T neutron generator can be created with approximately such intensity and the power of the proposed ADS model is 4 MW.

COMPUTER TOOLS AND MODEL

Subcritical core has been modeled using the Monte-Carlo radiation transport code MCNPX [30]; two-region model has been constructed as illustrated in fig. 2. The inner zone consists of 392 fuel elements; 52 of which have been replaced by Pu, Am, and Cm samples (Pu, Am, Cm) O_2 within MgO, with a density of 6.077 gcm^{-3} [31]. The selected initial composition of actinides element is presented in tab. 1.

Three core models have been considered. In these models, the selected actinide sample (Pu, Am, and Cm) elements are loaded inside the inner zone in three different distributions as:

- Non-uniform distribution model. The selected actinides sample elements surround the target at the core center as shown in fig. 3(a).
- Uniform distribution model. The elements are distributed evenly in the core as shown in fig. 3(b).
- Spiral distribution model [32]. The elements are distributed spirally around the target as shown in fig. 3(c).

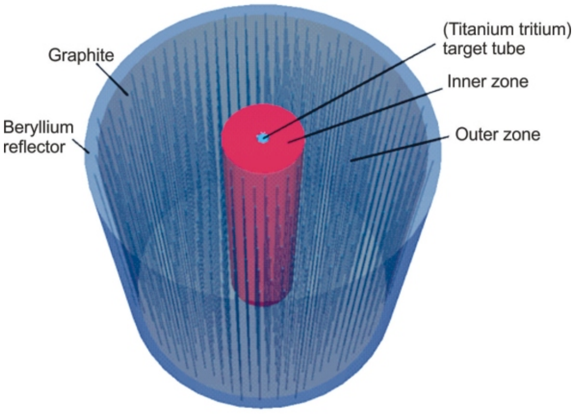

Figure 2. The 3-D view of MCNPX model for two regions

Table 1. Initial composition selection of actinide samples elements [30]

Element	wt.%	Isotope	wt.%	Half-life (y)
Plutonium	23.48	^{238}Pu	5.06	8.77 10^1
		^{239}Pu	37.91	2.41 10^4
		^{240}Pu	30.31	6.56 10^3
		^{241}Pu	13.21	1.43 10^1
		^{242}Pu	13.51	3.7 10^5
Americium	30.63	^{241}Am	66.67	4.33 10^2
		^{243}Am	33.33	7.37 10^3
Curium	6.12	^{244}Cm	90	1.81 10^1
		^{245}Cm	10	8.50 10^3
Magnesium	19.37	Mg-nat.	100	–
Oxygen	20.39	O-nat.	100	–

RESULTS AND DISCUSSIONS

The neutron flux characteristics and capture-to-fission ratio

The probability that an absorbed neutron causes fission is very important indicator/parameter to investigate the feasibility of Pu, Am, and Cm transmutation. In terms of cross-sections, this probability is defined as, $\alpha = \sigma_c/\sigma_f$, which is referred to the capture-to-fission ratio, as was mentioned in the introduction section. The lower ratio simply means that an absorption reaction will result in the fission rather than in the radioactive capture. The ratio depends strongly on the incident neutron energy. In the fast neutron region, the ratio decreases. So the feasibility of MA transmutation should be described by the ratio of capture-to-fission reaction rate (rather than the ratio of cross-sections only) which is taken into account for the neutron spectra

$$\alpha_R = \frac{\int_0^\infty N\sigma_c(E)\,\varphi(E)\,dE}{\int_0^\infty N\sigma_f(E)\,\varphi(E)\,dE} \qquad (1)$$

where N is the number density of a given nuclide, $\varphi(E)$ – the neutron flux energy dependence (neutron spectrum), and σ_c and σ_f – the corresponding cross-sections respectively.

The neutron spectra are determined by the neutron source and by presence or absence of the moderator. In subcritical system, the distance to external neutron source plays also the essential role. In two-zone subcritical system, the outer zone can influence the spectrum of inner zone in dependence of the material of the membrane which separates inner and outer zones. If this membrane can absorb the thermal neutrons diffusing from outer zone to the inner zone (for *e. g.* Cd), we could achieve harder spectrum in the inner zone and therefore more effective MA transmutation. For the preliminary investigation of the proposed model, the neutron energy spectra have been described for the two zones to demonstrate the difference of the thermal and fast neutron region as illustrated in fig. 4. Then, neutron spectra for different configurations of loading (uniform, non-uniform, spiral), for different distances from central source of neutrons with the energy 14 MeV before the irradiation (beginning of cycle – BOC) and after irradiation (end of cycle – EOC), are

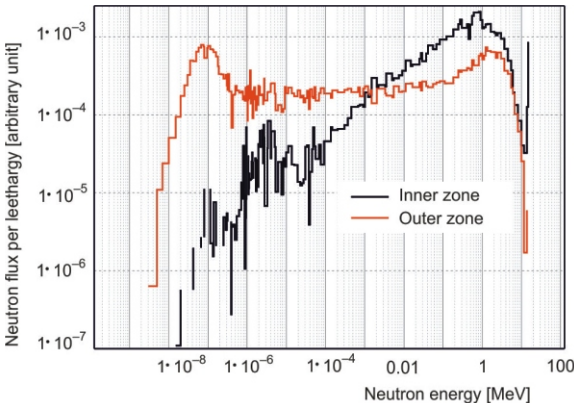

Figure 4. Neutron energy spectra at inner and outer zone

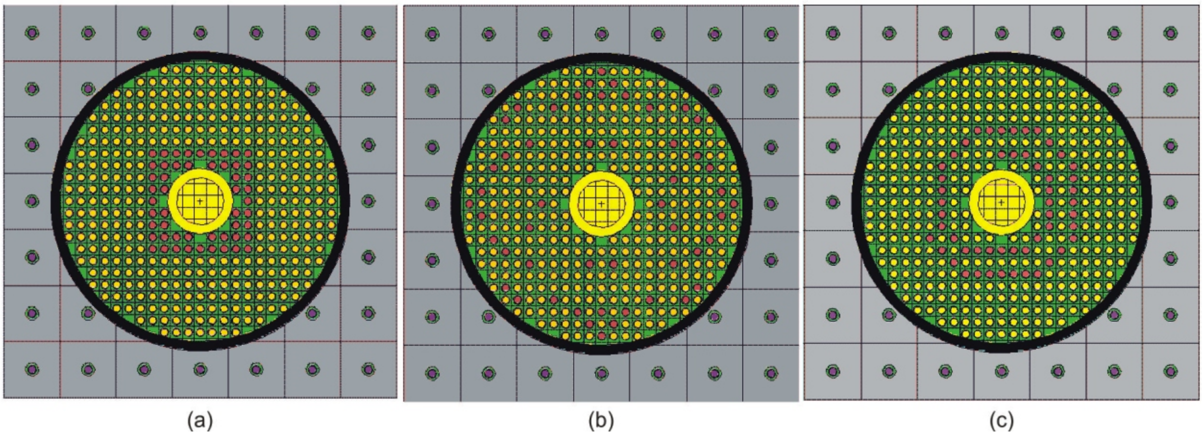

Figure 3. Schematic view of a horizontal cross-section in three models (from left to right): (a) non-uniform, (b) uniform and (c) spiral

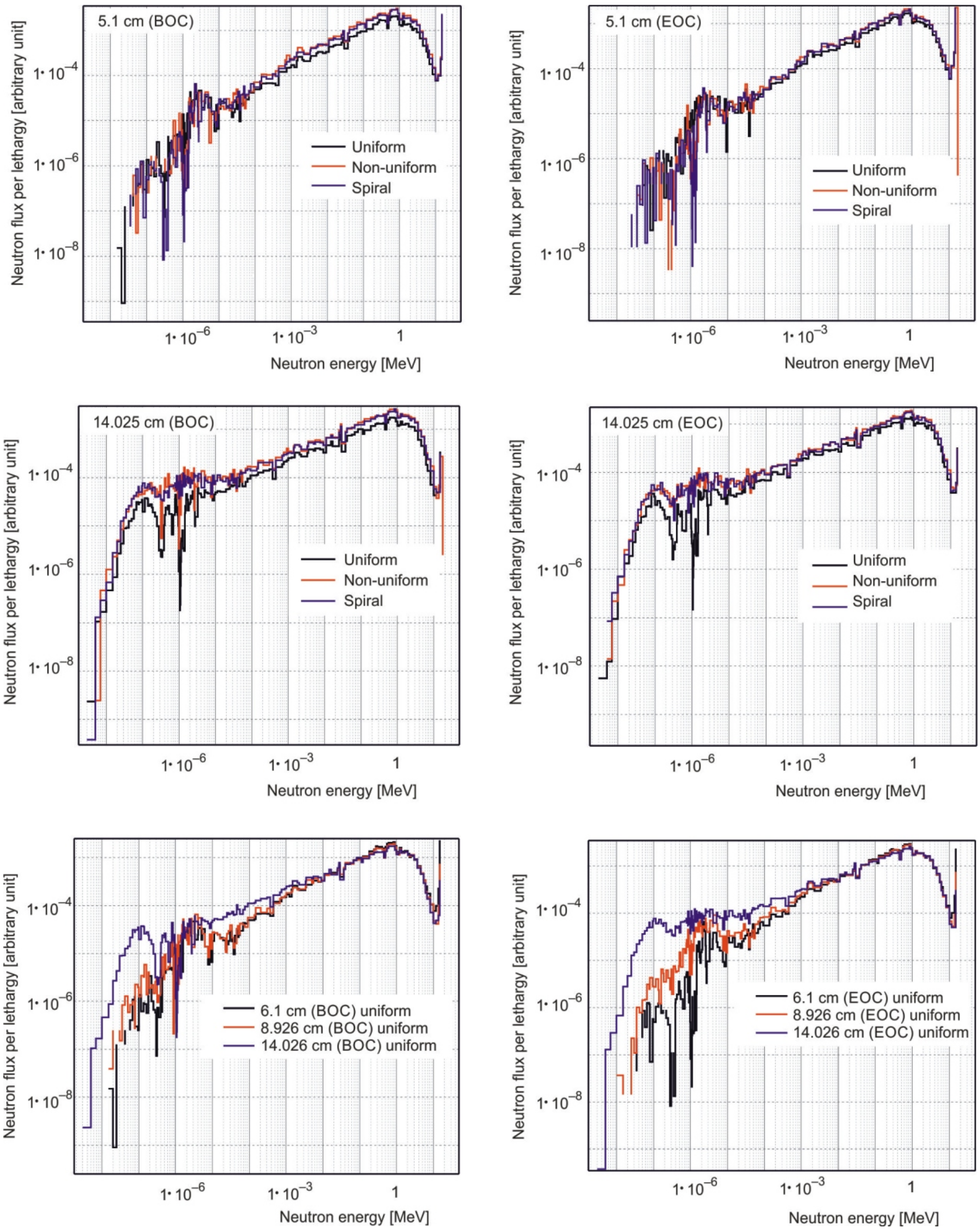

Figure 5. Neutron spectra at BOC and EOC for different configurations and different locations in inner zone

shown in fig. 5. All spectrum results, in all cases, are normalized for one source neutron, and the flux from MCNPX tally results is divided by the logarithm of energy difference, which is known as the flux per unit lethargy.

One can observe the noticeable differences in the spectra for different loading configurations only in the region of resonances, which are insufficient for transmutation problems, when the high energy part of the spectrum plays the main role. One can observe also the insufficient differences in the spectra before and after irradiation. The distance to the source plays more important role, and one can observe the visible increase of thermal neutrons at the large distance

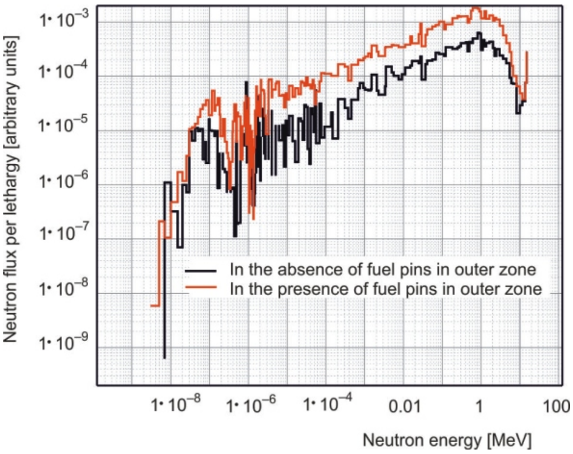

Figure 6. Neutron spectra in the inner zone in cases of presence and absence of outer zone fuel pins

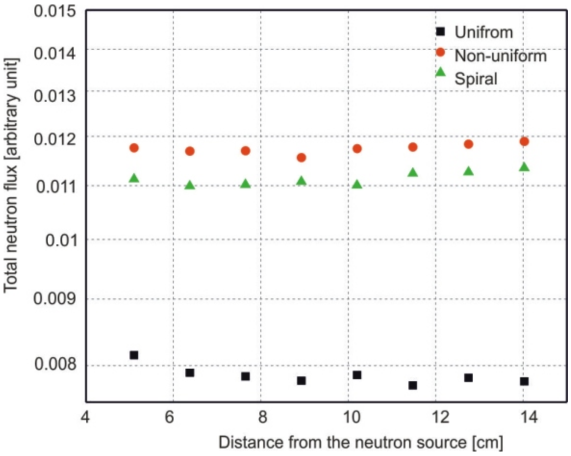

Figure 7. Total neutron flux in 8 pins in the inner zone through the *x*-direction of non-uniform, uniform, and spiral configurations

(14.025 cm) to the center of the core in spite of the moderator absence. This effect can be explained by the penetration of the thermal neutrons from the outer zone because the stainless steel membrane between outer and inner zone is practically transparent for thermal neutrons. This explanation is confirmed by calculating the neutron spectra in the inner zone for two cases, one with the presence of fuel pins in the outer zone and the other in the case of absence of these fuel pins. The neutron flux, changed as we can see in fig. 6, which means an observable contribution exists from the outer zone to the inner one. Naturally, the study of the neutron spectra could not allow us to make the conclusion about the optimal configuration of loading without performing detailed calculations of the quantity of transmuted nuclides. For further investigation, the total neutron flux distribution in all pins in the inner zone and through *x*-direction from the center, has been calculated, as well as for all three configurations (uniform, non-uniform, and spiral). The results are shown in fig. 7, and the relative error was less than 0.03. Based on these calculation results, it's clear that the non-uniform configuration provides a higher flux compared to the other two distributions, this means a higher transmutation potential of actinides sample in this configuration.

The ratio of capture to fission reaction rate for each isotope and at different locations (distance from the central tube), has been calculated and illustrated in tab. 2. The results show that the ratio depends on the distance from the core center and the isotope, as observed in general view for all the isotopes: the ratio of capture to fission rate increases when the target position is far from the center, in the case of ^{238}Pu the fission rate slightly exceeds the capture at 5.1 and 6.375 cm, after these points the ratio exceeds unity. While the isotopes ^{239}Pu and ^{241}Pu show less than unity values for the ratio at all chosen distance points, the probability to undergo fission is low for isotopes ^{240}Pu and ^{242}Pu, as can observed from the ratio values.

Burnup performance

The burnup-k_{eff} curves are a very important factor in the choice of the best model that satisfies the reactivity and control requirements in the subcritical system during the whole fuel cycle. So, in the results of this calculation, as is shown in fig. 8, it is clear that every model has practically the same k_{eff} during one-year cycle, but the uniform model appears as the most convenient for the reactivity and reactor safety issues due to the smallest initial value of k_{eff}. It is also evident that the non-uniform and spiral configurations are practically the same as for their influence to k_{eff} and to transmutation effectiveness due to practically the same neutron spectra. In addition, the burnup of the transuranic element sample, with operation

Table 2. Ratio of capture to fission reaction rate (α_{R}) for each isotopes, of Pu, Am, and Cm in inner zone

R_{in} [cm]	^{238}Pu*	^{239}Pu*	^{240}Pu*	^{241}Pu*	^{242}Pu*	^{241}Am*	^{243}Am*	^{244}Cm*	^{245}Cm*
5.1	0.90	0.43	23.98	0.26	16.79	10.96	22.81	4.92	0.12
6.375	0.97	0.43	24.28	0.25	16.57	12.12	23.86	2.79	0.13
7.65	1.04	0.43	27.17	0.26	20.54	12.33	27.64	4.39	0.13
8.925	1.16	0.47	28.67	0.28	20.67	14.42	36.13	6.39	0.13
10.2	1.27	0.50	36.46	0.29	28.19	16.68	31.83	7.09	0.13
11.475	1.67	0.52	49.47	0.30	27.42	20.00	38.12	7.57	0.13
12.75	2.16	0.53	80.47	0.32	32.90	25.76	41.76	8.25	0.13
14.025	3.30	0.55	89.80	0.33	32.54	35.74	51.07	10.58	0.14

*Statistical relative error less than 0.07

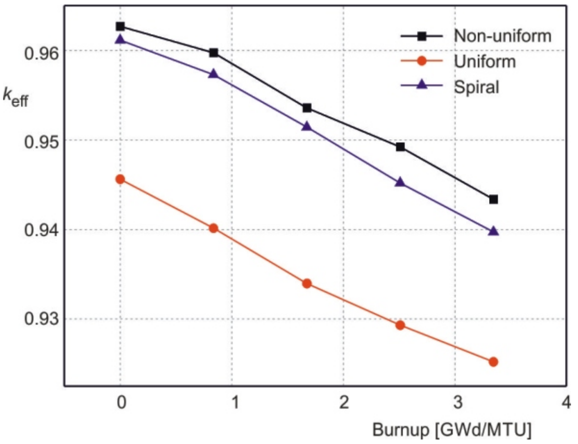

Figure 8. The k_{eff} variation with burnup for non-uniform, uniform, and spiral actinides distribution mode

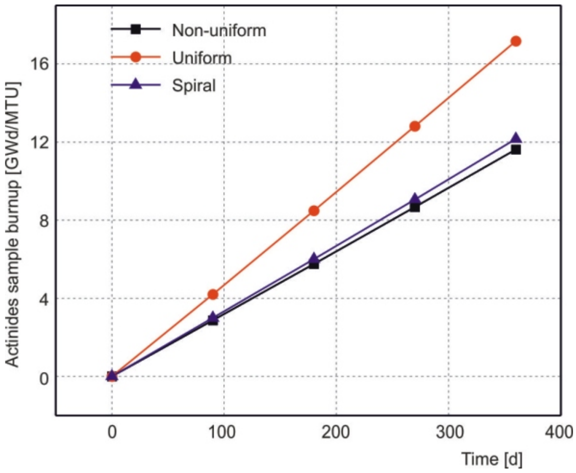

Figure 9. Burnup of an actinides sample for non-uniform, uniform, and spiral models in one-year operation cycle

time for the three models, has been calculated and the results are illustrated in fig. 9. One can see that the rate of increase of burnup over the operation cycle for the uniform case is larger than in the other two cases. This preference for the uniform model through the burnup operation is due to the symmetric distribution of the fuel and the actinides samples in the inner zone, which allows to benefit from wide spectrum of neutrons to burn the fissionable isotopes of Pu, Am, and Cm (see the large part of thermal neutrons at 14.025 cm in fig. 5).

The change in mass through the burnup processes of Pu, Am, and Cm nuclides for the three configuration models, non-uniform, uniform, and spiral, has been calculated for the period of one year, and is expressed in terms of transmutation/accumulation rate, as illustrated in tab. 3. The results show the following.

Table 3. The relative change of mass for each isotope in three models, non-uniform, uniform, and spiral respectively, for one year operation cycle

Isotope	Configuration	Initial mass [g]	Final mass [g]	$\frac{M_{in} - M_{fin}}{M_{in}}$ [%]
^{238}Pu	Non-uniform	235	240.3	–2.26
	Uniform	235	247.5	–5.32
	Spiral	235	241.1	–2.60
^{239}Pu	Non-uniform	1761	1750	0.61
	Uniform	1761	1738	1.31
	Spiral	1761	1749	0.68
^{240}Pu	Non-uniform	1408	1446	–2.70
	Uniform	1408	1445	–2.63
	Spiral	1408	1445	–2.63
^{241}Pu	Non-uniform	613.6	583.2	4.95
	Uniform	613.6	584	4.82
	Spiral	613.6	583.6	4.89
^{242}Pu	Non-uniform	627.6	630.2	–0.41
	Uniform	627.6	633.7	–0.9
	Spiral	627.6	630.4	–0.45
^{241}Am	Non-uniform	4039	4033	0.15
	Uniform	4039	4009	0.74
	Spiral	4039	4030	0.22
^{243}Am	Non-uniform	2019	2001	0.89
	Uniform	2019	1991	1.39
	Spiral	2019	2000	0.94
^{244}Cm	Non-uniform	1090	1063	2.48
	Uniform	1090	1073	1.56
	Spiral	1090	1065	2.29
^{245}Cm	Non-uniform	121.1	122.2	–0.91
	Uniform	121.1	122.2	–0.91
	Spiral	121.1	122.3	–0.99

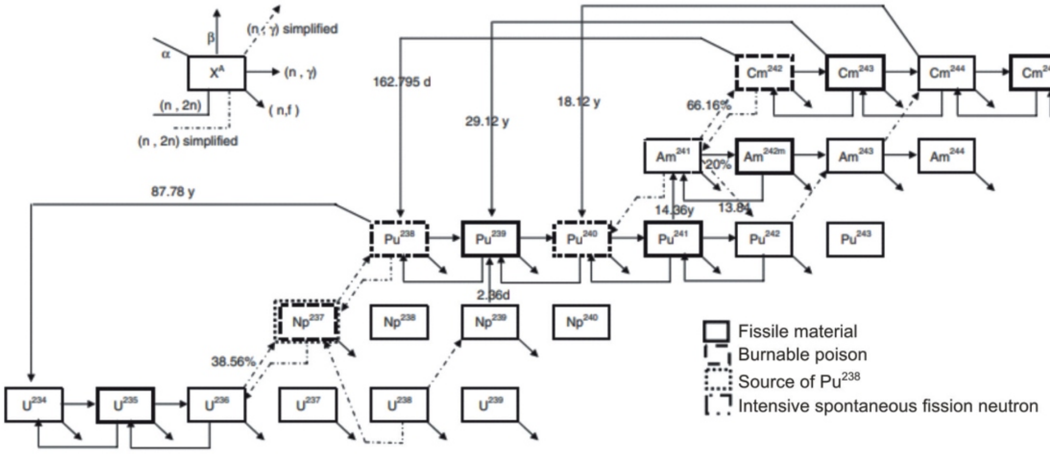

Figure 10. Nuclides chain [33]

Plutonium Isotopes. The ^{239}Pu is usually considered as the major isotope which contributes to the radiotoxicity and proliferation risk, which has a relatively large amount and a half-life of 24100 years. The results in tab. 3, show the transmutation rate in all the proposed models, but the best value is the case of uniform model where 1.306 % of the total mass (the initial mass 1761 gm) has been transmuted. These results are also identical with capture-to-fission ratio results (tab. 2), which gives the initial indication that ^{239}Pu has high probabilities to fission. Of course, this is a well known conclusion which is indicated in every handbook on nuclear engineering, and a big percentage of ^{239}Pu transmutation in the case of uniform model can be explained by the large part of thermal and epithermal neutrons at 14.25 cm and a very big fission cross-section for ^{239}Pu in epithermal region.

Also in the case of ^{241}Pu we have a good percentage for transmutation rate, but there is no significant difference among the three models. While in the case of ^{238}Pu, ^{240}Pu, and ^{242}Pu, the accumulation of extra amount of these isotopes was dominant, with different rates in the three models (the negative sign in the tab. 3 indicates accumulation). The non-uniform distribution for ^{238}Pu and ^{242}Pu achieved low accumulation with 2.255 % per year and 0.414 % per year, respectively. On the other hand, there are no significant differences between the three distributions regarding ^{240}Pu. The accumulation can be easily explained from the heavy metal transmutation chains [33] shown in fig. 10. In addition to the ^{239}Pu (n, fission) reaction mentioned above, we can conclude also, based on the chain diagram that the significant amount of ^{239}Pu converted to ^{238}Pu through the pathway ^{239}Pu (n, 2n), where we have a high accumulation rate of ^{238}Pu (5.319 %) in the case of uniform model.

Americium isotopes. The two chosen isotopes ^{241}Am and ^{243}Am in this study, are the prime candidates to transmutation, as we have mentioned previously. From the calculation results in tab. 3, the mass of both isotopes, ^{241}Am and ^{243}Am, has been decreased in all models, but the uniform is the best option for the transmutation, where the high rates of 0.743 % per year and 1.387 % per year, respectively, are achieved.

Curium isotopes. Are also major contributors to gamma and neutron emissions, especially the isotopes ^{244}Cm and ^{245}Cm, where they appear in a relatively large amount in spent fuel, for instance, in this study we have 1090 g of ^{244}Cm, and 121.1 g of ^{245}Cm. The non-uniform model achieves a high transmutation rate for ^{244}Cm, on the other side, we have a little bit accumulation for 2^{45}Cm, although the capture- to fission ratio indicates a high probability of fission. Of course, this accumulation for ^{245}Cm outcome from transmutation part of ^{244}Cm to ^{245}Cm due to initial mass of ^{244}Cm is five times greater than ^{245}Cm, and by the trajectory ^{244}Cm (n, γ), as we can see from the diagram in fig. 10.

The exact behavior of transmutation efficiency remains ambiguous, as it is hypothesized that several factors contribute to the MA transmutation efficiency at inner (fast) zone of two-zone subcritical system including energy of neutron source, distance to external neutron source, the impact of outer (thermal) zone on the periphery of the inner (fast) zone, *etc*. Nevertheless, from the point of view of transmutation efficiency for two-zone subcritical system, non-uniform and spiral configurations of fuel elements with MA are more effective. One should remember that it is possible to transmute the long lived fission products in the outer zone of such a subcritical system. The effectiveness of such transmutation will be studied in the next article. Also, the final conclusion on effectiveness of transmutation/accumulation of different components of thermal reactor spent fuel will be made in the next article, taking into account the accumulation and transmutation of MA in the working fuel of both zones of considering ADS.

CONCLUSIONS

The main goal of present paper is to study the possibilities of transuranic element transmutation in a two-zone subcritical system, consisting of an inner zone with a fast neutron spectrum and an outer zone with a predominant thermal neutron spectrum. An additional goal is the study of transmutation with different configurations of fuel elements with MA inside the inner (fast) zone.

The MA transmutation has been studied repeatedly in various fast spectra with different neutron sources in subcritical systems. A feature of this work is the rather low energy of the neutron source and the presence of an outer (thermal) zone at the periphery of the fast zone. The effect of this zone, which lies in the transfer of thermal neutrons into inner (fast) zone is substantial enough, and it affects largely on the configuration of fuel rods with a uniform MA. This is confirmed by the faster burnup of fissile isotopes near the border with the outer (thermal) zone and the practical absence of transmutation of even plutonium isotopes. Therefore, from the point of view of transmutation efficiency, non-uniform and spiral configurations of fuel elements with MA are more effective. It should be noted that the introduction into the outer (thermal) zone of fuel elements, with fission products that well absorb thermal neutrons, can change the conclusions about the efficiency of MA transmutation, especially if the absorbing fuel elements are placed near the boundary between the outer (thermal) and inner (fast) zones.

Based on the aforementiones results, the following conclusions can be made:

In terms of transmutation efficiency, non-uniform and spiral configurations are more beneficial for the case of two-zone subcritical system. In other words, MA transmutation in harder neutron spectrum is preferable, which is an evident result.

The inner (fast) zone of two-zone subcritical system can transmute most of the actual MA isotopes in the inner zone of two zone system, driven by usual neutron generator with sufficiently small neutron energy ~14 MeV. Thus, this subcritical system can be effectively used for investigation of the MA transmutation processes in the inner zone with a fast neutron spectrum.

Such a small neutron energy is proved to be insufficient to transmute even isotopes of Pu – they are accumulated rather than transmuted, especially taking into account the accumulation of plutonium isotopes in the usual fuel of a given subcritical system.

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AUTHORS' CONTRIBUTIONS

A. A. Al Qaaod, was responsible for constructing model, calculation and analysis, and writing the original draft. V. M. Pavlovych, contributed to conceptualizing the research, improving the manuscript writing and supervised the research. D. Ridikas, did improving the manuscript writing, review the results, and editing. V. Gulik, contributed to conceptualizing the research, formal analysis, writing, review, and editing. E. Amin, was in charge of review of the results, software and validation.

REFERENCES

[1] ***, International Atomic Energy Agency. Nuclear Power Reactors in the World, Reference Data Series No. 2. (2019), p. 80

[2] Salvatores, M., Palmiotti, G., Radioactive Waste Partitioning and Transmutation Within Advanced Fuel Cycles: Achievements and Challenges, *Progress in Particle and Nuclear Physics, 66* (2011), 1, pp. 144-166

[3] Sagara, H., *et al*., Denaturing of Plutonium by Transmutation of Minor-Actinides for Enhancement of Proliferation Resistance, *Journal of Nuclear Science and Technology, 42* (2005), 2, pp. 161-168

[4] Shahbunder, H., *et al*., Effects of Pu and MA Uniform and Nonuniform Distributions on Subcritical Multiplication of TRIGA Mark II ADS Reactor, *Annals of Nuclear Energy, 94* (2016), 8, pp. 332-337

[5] Magill, J., *et al*., Impact Limits of Partitioning and Transmutation Scenarios on the Radiotoxicity of Actinides in Radioactive Waste, *Nuclear Energy, 42* (2003), 5, pp. 263-277.

[6] Mueller, A. C., Transmutation of Nuclear Waste and the future MYRRHA Demonstrator, *Journal of Physics: Conference Series, 420* (2013), 1, 012059

[7] Shahbunder, H., *et al*., Investigation of Subcritical Multiplication Parameters in TRIGA Mark II Accelerator Driven System, *Annals of Nuclear Energy, 81* (2015), 7, pp. 336-341

[8] Yang, C., *et al*., Minor Actinide Transmutation Analysis of Fusion-Driven Subcritical System, *Yuanzineng Kexue Jishu/Atomic Energy Science and Technology, 47* (2013), 6, pp. 250-254

[9] Kazansky, Y. A., Romanov, M. I., Transmuting Minor Actinides with Thermal Reactor Neutrons, *Nuclear Energy and Technology, 1* (2015), 3, pp. 208-212

[10] Rubbia, C., *et al*., Conceptual Design of a Fast Neutron Operated High Power Energy Amplifier, Technical Report CERN/AT/95-44(ET), CERN, 1995

[11] Bowman, C. D., *et al*., Nuclear Energy Generation and Waste Transmutation Using an Accelera-

tor-Driven Intense Thermal Neutron Source, *Nuclear Instruments and Methods in Physics Research, A, 320* (1992), 1-2, pp. 336-367

[12] Al Qaaod, A. A., *et al*., Transmutation Performance of Uniform and Nonuniform Distributions of Plutonium and Minor Actinides in TRIGA Mark II ADS Reactor, *Annals of Nuclear Energy, 121* (2018), pp. 101-107

[13] Gulevich, A., *et al*., Comparative Study of ADS for Minor Actinides Transmutation, *Progress in Nuclear Energy, 50* (2008), 2-6, pp. 359-362

[14] Xoubi, N., Neutronic Design Study of Accelerator Driven System (ADS) for Jordan Subcritical Reactor as a Neutron Source for Nuclear Research, *Applied Radiation and Isotopes, 131* (2018), 4, pp. 71-76

[15] Al Qaaod, A., *et al*., ^{226}Ra Irradiation to Produce ^{225}Ac and ^{213}Bi in an Accelerator-Driven System Reactor, *Nuclear Science and Techniques, 31* (2020), 44

[16] Salvatores, M., The Physics of Transmutation in Critical or Subcritical Reactors, *Comptes Rendus Physique, 3* (2002), 7-8, pp. 999-1012

[17] Chadwick, M. B., *et al.*, ENDF/B-VII.0: Next Generation Evaluated Nuclear Data Library for Nuclear Science and Technology, *Nuclear Data Sheets, 107* (2006), 12, pp. 2931-3060

[18] Malambu, E., *et al*., Neutronic Analysis of Minor Actinide Transmutation in MYRRHA, *NEA/NSC/R (2015), 2* (2015), pp. 238-246

[19] Fairhall, G., Minor Actinide Transmutation Position Paper. Natl. Nucl. Lab. (2011), pp. 1-20

[20] Science, N., Minor Actinide Burning in Thermal Reactors, NEA No. 6997, (2013), 78

[21] Daniel, H., Petrov, Y. V., Subcritical Fission Reactor Driven by the Low Power Accelerator, Nuclear Instruments and Methods in Physics Research, *Section A., 373* (1996), 1, pp. 131-134

[22] Babenko, V. O., *et al*., Modeling of Two-zone Accelerator-Driven Systems, *Nuclear Physics and Atomic Energy, 13* (2012), 3, pp. 266-275

[23] Gulik, V., Tkaczyk, A. H., Cost Optimization of ADS Design: Comparative Study of Externally Driven Heterogeneous and Homogeneous Two-Zone Subcritical Reactor Systems, *Nuclear Engineering and Design, 270* (2014), 4, pp. 133-142

[24] Gulik, V. I., Simulation of Fuel Distribution in the Inner Fast Zone for Two-Zone Subcritical Reactor, *Nuclear Physics and Atomic Energy, 18* (2017), 4, pp. 331-340

[25] Kovalchuk, V. D., *et al*., Activation Study at SNEG-13 Facility of the Candidate Ferritic Steel 10X9VFA, *Fusion Engineering and Design, 42* (1998), 1-4, pp. 343-348

[26] Verbeke, J. M., *et al*., Development of a Sealed-Accelerator-Tube Neutron Generator, *Applied Radiation and Isotopes, 53* (2000), 4-5, pp. 801-809

[27] Babenko, V. A., *et al*., On the Subcritical Amplifier of Neutron Flux Based on Enriched Uranium, *NATO Security through Science Series B: Physics and Biophysics,* (2006), pp. 253-263

[28] Babenko, V. O., *et al*., The Research Subcritical Reactor, *Nuclear Physics and Atomic Energy, 9* (2008), 1, pp. 56-61

[29] Galchenko, V. V., *et al*., Computational Benchmark for Fuel Assembly of VVER-1000 Using the Monte Carlo Serpent Code, *Nucl Technol Radiat, 33* (2018), 1, pp. 24-30

[30] Hendricks, J. S. *et al*., MCNPX Extensions Version 2.5.0, LA-UR-05-2675, (2005), p. 79

[31] Lemehov, S. E., *et al*., Progress and Advancements in Fuel Performance Modelling for MYRRHA/XT-ADS, International Topical Meeting on Nuclear Research Applications and Utilization of Accelerators, 4 May, 2009, Vienna, p. 8

[32] Kuwagaki, K., *et al*., Concept of Breed and Burn Reactor with Spiral Fuel Shuffling, *Annals of Nuclear Energy, 127* (2019), 5, pp. 130-138

[33] Meiliza, Y., *et al*., Protected Plutonium Breeding by Transmutation of Minor Actinides in Fast Breeder Reactor, *Journal of Nuclear Science and Technology, 45* (2008), 3, pp. 230-237

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ПРОРАЧУН РАСПОДЕЛЕ ИЗВОДЉИВОСТИ ТРАНСМУТАЦИЈЕ РАДИОАКТИВНОГ ОТПАДА У СИСТЕМУ СА АКЦЕЛЕРАТОРСКИМ ПОГОНОМ

Проучена је изводљивост трансмутације високорадиоактивног отпада у поткритичном реакторском склопу покретаном акцелераторским системом – за двозони модел са три различите конфигурације језгра. Унутрашња зона има спектар брзих неутрона, а спољашња спектар термичких неутрона. Поткритично језгро повезано је са спољашњим неутронским извором енергије 14 MeV (D-T извор). На основу претпостављене три основне конфигурације неуниформне, униформне и спиралне, истражени су и размотрени ефектри расподеле изотопа (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{244}Cm, и ^{245}Cm) на расподелу неутронског спектра и перформансе изгарања у унутрашњој зони. За све предложене моделе извршени су прорачуни изгарања за једногодишњи оперативни циклус. Овај рад показује да се може ефикасно трансмутовати већина стварних споредних изотопа актинида уз оптималну дистрибуцију ових изотопа у унутрашњој зони брзог спектра претпостављеног система.

Кључне речи: изгарање, неутронски спектар, актинид, трансмутација отпада, систем покретан акцелератором, јачина фисије