V. I. Gulik[1], V. M. Pavlovych[2]

[1]Institute for Safety Problems of Nuclear Power Plants, NAS of Ukraine, 12, Lysogirska st., Kyiv, 03028, Ukraine
[2]Institute for Nuclear Research of NAS, 47, Nauky av., Kyiv, 03680, Ukraine

The Development of Neutron-Physical Model for Two-Zone Research Subcritical Reactor for Nuclear Waste Transmutation

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subcritical system,
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optimization of subcritical core.

The creation of small-scale research subcritical reactors is necessary, in particular, for the development of the technology for nuclear waste transmutation in Accelerator Driven Systems. The construction of such facilities will allow the development of the technology of nuclear waste transmutation without constructing an expensive industrial scale subcritical reactor. The low-cost neutron generator can serve as a driver for such research subcritical reactors. A two-zone model of subcritical system driven by high-intensity neutron generator is proposed in this work. The proposed system can use two separated cores with different neutron spectra: fast and thermal. This paper also represents the main stages of the proposed model of two-zone subcritical reactor development. The results of simulations, aimed at optimizing of the geometry and fuel composition of the two-zone subcritical system, performed in Serpent and MCNP codes are presented. An overview of the analysis of different facilities that can be used as an external neutron source for subcritical reactor is shown, however a high-intensity neutron generator based on D-T reaction was chosen as the optimal neutron source for low-cost research subcritical reactor for investigation of nuclear waste transmutation. Generally, it is observed that the two-zone subcritical system can effectively amplify neutron flux from external neutron sources.

Introduction

After Three Mile Island accident in the United States (1979), the Chornobyl disaster in Ukraine (1986) and Fukushima Daiichi disaster in Japan (2011), the safe operation of nuclear reactors has become a very controversial topic. Since the mid-twentieth century the huge quantity of spent nuclear fuel and vitrified nuclear waste obtained by the operation of conventional nuclear reactors has been accumulated. Currently, many countries have still not implemented a strategy for dealing with spent nuclear fuel and nuclear waste management. On the other hand, there are also some countries (such as Sweden and Finland) which adopted a strategy for the final disposition of spent nuclear fuel in deep geological repositories, but so far there is no existing storage of spent nuclear fuel or nuclear waste for a long-term period with the exception of the experimental facility in Sweden.

Over the past twenty years the possibility of solving nuclear waste problem by means of nuclear waste transmutation has been heavily investigated [1–3]. The nuclear waste can be divided into two main categories: minor actinides and long-lived fission products [4]. The minor actinides include isotopes such as: ^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm etc. The transmutation of minor actinides occurs mainly as a result of fission reaction. As shown in number studies, the most favorable conditions for minor actinides transmutation are observed in fast neutron spectrum [5]. Therefore, the fast reactors or the subcritical systems driven by an external neutron source with sufficiently high neutron energy (Accelerator Driven Systems (ADS)) are considered to be the most realistic scheme

for minor actinides transmutation. On the other hand, the long-lived fission products include some of the following isotopes: [129]I, ^{99}Tc, [99]Zr, [135]Cs, [126]Sn etc. The long-lived fission products transmutation occurs mainly as a result of capture reaction. It has been found in several studies that the most favorable conditions for transmutation of long-lived fission products are observed in the thermal neutron spectrum [6, 7]. However, the transmutation of long-lived fission products in the traditional critical reactors is complicated due to a number of technical issues. Therefore, the subcritical systems with thermal neutron spectrum are a great option to investigate long-lived fission products transmutation.

The development of neutron-physical model of two-zone research subcritical reactor driven by high-intensity neutron generator for investigations of minor actinides and long-lived fission products transmutation has been described in the present paper.

Model development

High-intensity D-T neutron generator as a driver for subcritical core

The analysis of the latest research of subcritical systems driven by external neutron source (Fig. 1) helps to summarize what type of facilities of external neutron source could be used:

1. The proton (cyclotron or linear) accelerator that uses spallation reactions to produce neutron flux in the subcritical reactor [8]. Advantage: high intensity of neutron source; disadvantages: high cost and difficulties in beam-window construction. MYRRHA project in the European Union [9] can be given as an example to this type of facility.

2. Electron accelerators that use photon-neutron reactions to produce neutron flux in the subcritical reactor [10]. Advantage: the possibility to obtain sufficiently high current of electrons and therefore the relatively high intensity of neutron source; disadvantage: high cost. An example of this type of facility is the KIPT neutron source facility in Kharkiv, Ukraine [11].

3. High-intensity neutron generator and plasma focusing (PF) devices that use deuterium-tritium (D-T) reactions to produce neutron flux in the subcritical reactor [12–14]. Advantages: relatively low cost and well-known technology; disadvantages: relatively low intensity of neutron source and necessity to have tritium producing facility. A good example is the YALINA-booster facility in Belarus [15].
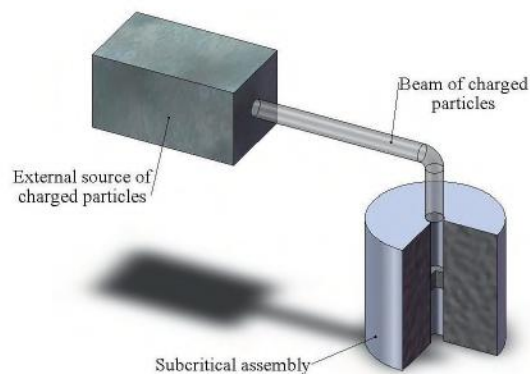

Fig. 1. Simple scheme of ADS

4. Fusion neutron sources based on the fusion reaction to produce neutrons in the subcritical reactor [16]. Advantage: high intensity of neutron source; disadvantages: still under development, so not ready for immediate use.

In the condition of limiting funds, such as in Ukraine at present time, it is impossible to develop and construct the expensive high current proton and electron accelerators, so the option of D-T neutron source is an optimal solution for the development of experimental facilities for the investigation of nuclear waste transmutation in such cases.

Another useful feature of the proposed subcritical reactor is the existence of two cores with different neutron spectra in a single facility (fast and thermal). There are two major advantages in such a two-zone subcritical system: 1) the ability of more effectively enhancing the neutron flux from the external source [17]; 2) the fact that it is very convenient to investigate transmutation of minor actinides and long-lived fission products, due to the existence of fast and thermal neutron spectra in a single facility [5, 6].

The using of MCNP Monte Carlo code for model development

The investigations of two-zone subcritical system driven by high-intensity neutron generator were performed in Institute for Nuclear Research in Kyiv, Ukraine, whose scientists had a big experience in high intensity neutron generators design and production [18], as well as in study of neutron flux amplification in subcritical assemblies of different types and different material composition [19–22]. The neutron and energy amplification factor were selected as basic parameters for this study.

The neutron codes MCNP-4c and SCALE-5 as well as ENDF/B-VI nuclear data library were used for the simulation stage [23].

Thereafter, the research of amplifying abilities of two-zone subcritical system, geometrical optimization and optimization of material composition of such systems were performed [24–27]. It has been found that two-zone subcritical system can more effectively amplify the neutron flux from external source than one-zone subcritical system, from the point of view of neutron economy. The optimal geometric and material characteristics for inner and outer zones of subcritical reactor also were established.

Nuclear waste transmutation was simulated using codes MCNP, ORIGEN and Monteburns for developed MCNP model of two-zone subcritical reactor. The transmutation of main minor actinides in the inner zone with fast neutron spectrum was carried out along with the transmutation of main long-lived fission products in the outer zone with thermal neutron spectrum. The main results for the modeling of nuclear waste transmutation were presented in our previous publications [28, 29].

The diagram of the final version of MCNP code model is presented in Fig. 2. The overall conditions for the present MCNP model were:

1) geometry and material parameters of fuel elements for VVER-1000 reactor;

2) the fuel in pins – uranium dioxide with different enrichments for ^{235}U (in inner zone – 15 %, in outer zone – 4 %);

3) fuel density – 10.96 g/cm³;

4) cladding material – zirconium +1 % niobium;

5) hexagonal lattice of fuel elements was used in both zones;

6) coolant of inner zone is helium whereas coolant of outer zone is light water;

7) the effective neutron multiplication factor is 0.97. The ENDF/B-VI nuclear reaction database containing evaluated (recommended) cross sections was also used for MCNP calculations.

In addition, there are also MCNP visualizations for the developed two-zone subcritical model which are presented in Figs. 3–6.

Serpent Monte Carlo code for model development

The second stage of the presented work was the model development of the two-zone research subcritical reactor driven by high-intensity neutron generator
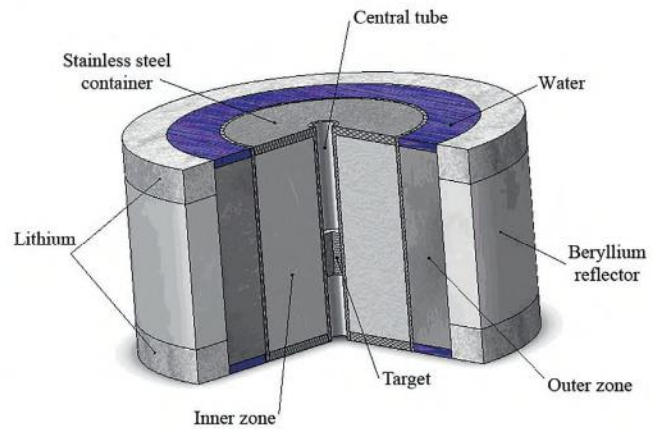

Fig. 2. Diagram of the two-zone subcritical reactor model developed by MCNP code
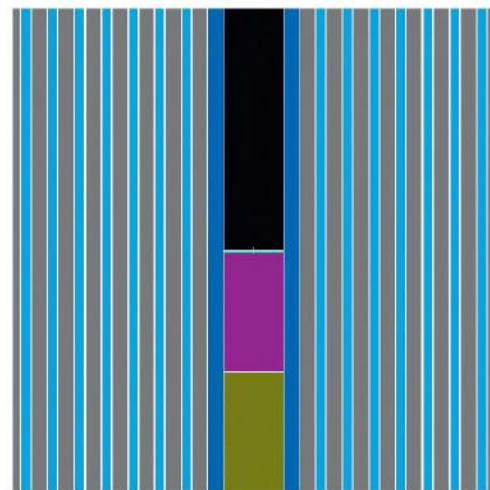

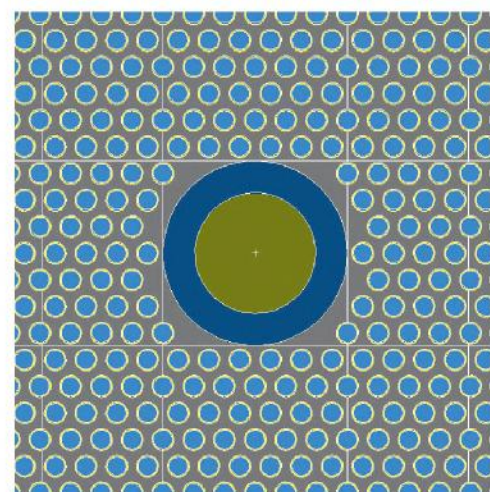Fig. 3. Vertical cross-section view of central part for calculated model in MCNP code

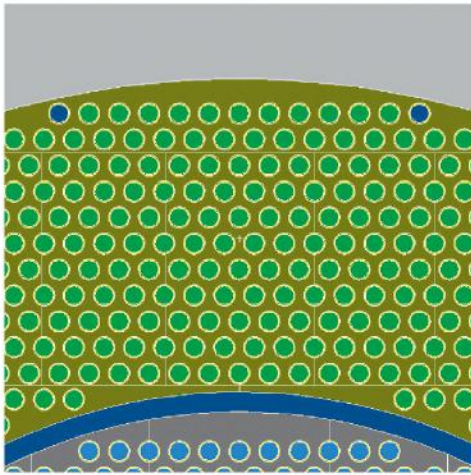Fig. 4. Horizontal cross-section view of central part for calculated model in MCNP code

Fig. 5. Horizontal cross-section view of outer zone
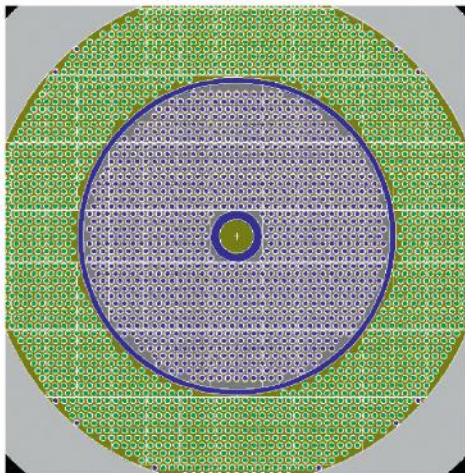for the calculated model in MCNP code

Fig. 6. Horizontal cross-section view of the whole core
for the calculated model in MCNP code

using new Monte Carlo Serpent code [30]. Serpent code has been developed in Finland in VTT Technical Research Centre since 2004. There are two main advantages of studying the subcritical systems in Serpent Monte Carlo code: i) the possibility of modeling of a subcritical system with external neutron source and ii) the possibility of burnup calculation for the simulation of nuclear waste transmutation.

The first modelling results of the two-zone subcritical system with uranium dioxide for homogeneous case with the help of the Serpent code were presented in our previous publication [31]. Additional calculation results of the modelled homogeneous subcritical system with uranium metallic fuel without coolant and cladding materials are also presented in this paper. The diagram of the final version of Serpent code model is shown in

Fig. 7 and visualization Serpent code is displayed in Fig. 8. The typical conditions for the present Serpent model were: 1) homogenizing fuel – uranium metallic with different enrichments for ^{235}U (in inner zone – 20 %, in outer zone – 4 %); 2) fuel density – 19.07 g/cm³; 3) effective neutron multiplication factor set to 0.97. The ENDF/B-VII nuclear data library was used for Serpent calculations.

The optimization results of the two-zone subcritical system with heterogeneous fuel were presented in our previous work [32] with the help of Serpent simulation.
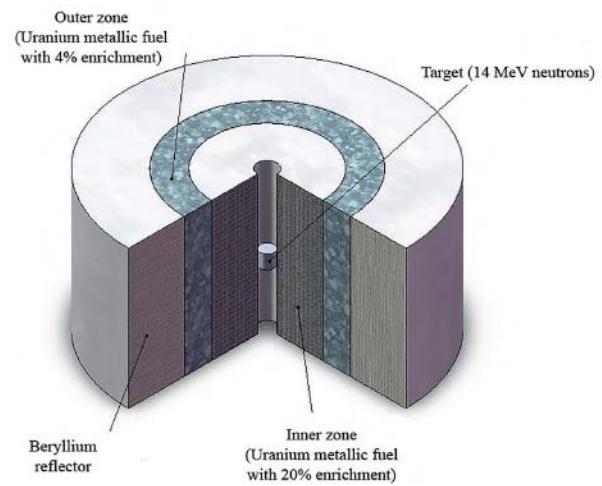

Fig. 7. Diagram of the two-zone subcritical reactor
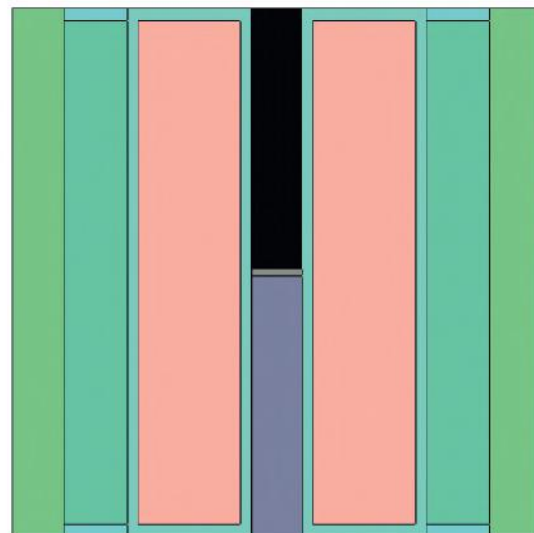model for homogeneous systems

Fig. 8. The scheme of the calculated homogeneous
model in Serpent code

MCNP and Serpent modelling results

The modelling of transmutation characteristics for two-zone subcritical system

Homogeneous mixture of uranium metallic enriched in ^{235}U for 20 % in the inner zone and a homogeneous mixture of uranium metallic with 4 % enrichment in ^{235}U in the outer zone were considered as fuel material in the present model. Zones have neither coolant nor cladding materials, therefore both zones have fast neutron spectrum in neutron-physical model. Other characteristics for this case are presented in section 2.3. The Serpent code was used for this modelling.

The following are some of the additional parameters in the developed model:

diameter of the target is 5 cm;

thickness of stainless steel container wall is 1 cm;

thickness of the beryllium reflector is 5 cm;

diameter-to-height ratio of subcritical core is 1 for minimization of the leakage of neutrons;

neutron source is isotropic point sources with 14 MeV monoenergetic neutrons;

effective neutron multiplication factor is 0.97;

total number of source neutrons is 50,000;

number of batches is 500.

The seven two-zone subcritical systems with different geometrical parameters were analyzed as well as the one-zone subcritical system that was calculated for comparison. Every two-zone system has varied radius of the inner zone given ranging from 9.5 to 15.5 cm. The radius of the outer zone selected in such a way that k_{eff} of all system is equal to 0.97 and in a way to provide a diameter-to-height ratio equal to 1. Radioactive isotopes ^{237}Np and ^{243}Am were chosen as the examples of nuclear waste for the investigation of transmutation characteristics. The volume normalized neutron flux integrated over space and energy (*Flux*), fission microscopic cross-section (σ_f), capture microscopic cross-section (σ_c) and ratio of these cross-sections (α) are used as main parameters in this work [5]. The fission microscopic cross-section (σ_f) is the microscopic cross-section of fission reaction for the chosen isotopes (^{237}Np and ^{243}Am) averaged by inner or outer zones. The capture microscopic cross-section (σ_c) is the microscopic cross-section of capture reaction for the chosen isotopes (^{237}Np and ^{243}Am) averaged by inner or outer zones. In addition, the value (α) is the ratio of the average capture to fission cross section for the chosen isotopes (^{237}Np and ^{243}Am) in the inner or outer zones [5]. Finally, the value (α) is useful for un-

derstanding transmutation potential of subcritical systems that are being analyzed. The optimal conditions of the two-zone subcritical system with respect to the transmutation efficiency (*Flux* and α) were defined as the main goal for the present calculation results.

Figs. 9–22 represent the modelling results for homogeneous mixture of uranium metallic in the inner zone enriched to 20 % and in the outer zone enriched to 4 % for ^{235}U. The data in the figures are laid out with the data on the ordinate and the radius of the inner zone in the abscissa, in the two-zone subcritical system. The certain square marker (point) on figures correspond to the one-zone subcritical system and they are represented in the figures for comparison.

First, we analyzed the change of the neutron flux in inner and outer zones (see Figs. 9 and 10) in case of changing the inner zone radius (and as the result, the volume of more enriched uranium).

Fig. 10 represents a natural increase for the neutron flux in outer zone in case of increasing the volume of inner zone due to increasing the amount of more enriched uranium (20 % for ^{235}U) in inner zone. And Fig. 9 shows the peak of the neutron flux with the radius of inner zone of approximately 14 cm. This indicates that it is possible to choose the ratio of the volumes of both zones so that the neutron fluxes in inner and outer zones would be maximum and, as a result, the number of neutrons that can be used for the transmutation of minor actinides would be higher.

Then we investigated the transmutation characteristics of inner and outer zones with respect to the chosen isotopes which belong to minor actinides: ^{237}Np and ^{243}Am. We attributed the fission microscopic cross section, as the cross section of the most important process
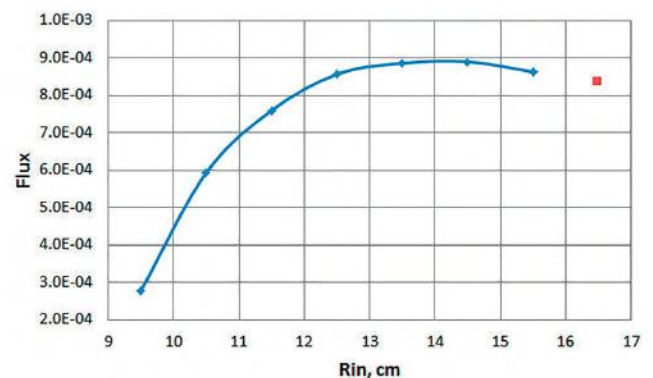

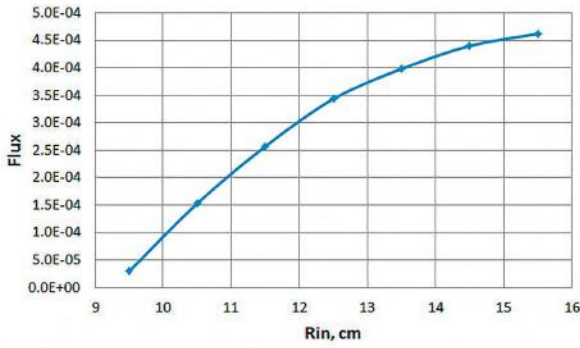Fig. 9. The volume normalized neutron flux integrated over space and energy for inner zone

Fig. 10. The volume normalized neutron flux integrated over space and energy for outer zone
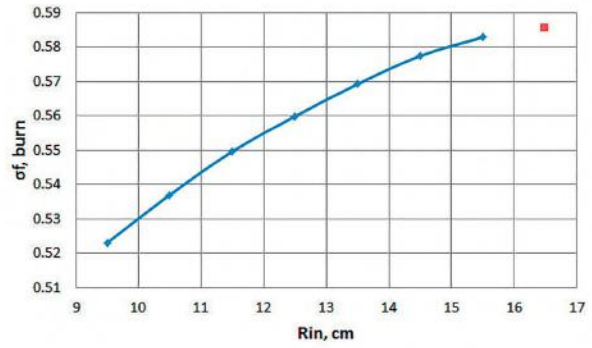

Fig. 11. The fission microscopic cross-section of ^{237}Np for inner zone

for the transmutation of minor actinides, to the transmutation characteristics. The capture microscopic cross section is important since the capture reaction plays the role of a competing process with respect to fission and significantly affects the "economy" of neutrons. The ratio of these two cross sections is an excellent indicator of the transmutation potential of the reactor; it was proposed by Massimo Salvatores in [5].
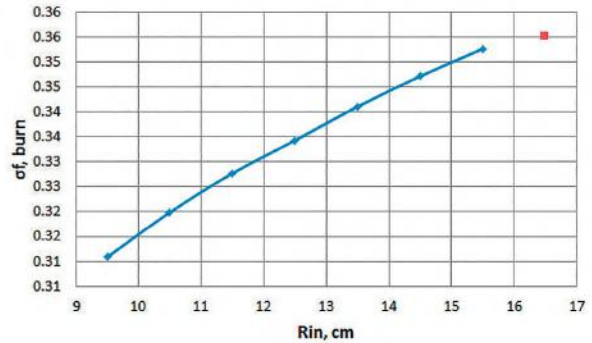
Microscopic cross sections of fission and capture in inner zone for ^{237}Np and ^{243}Am are presented in Figs. 11–14. In case of increasing the volume of inner zone with more enriched uranium, the possibility of fission naturally increases for both isotopes. For capture cross section it is possible to choose the optimal volume ratio of both zones at which the probability of a capture reaction will be minimal. For both isotopes, the optimal volume ratio of inner and outer zones occurs when the radius of inner zone is approximately 13 cm.
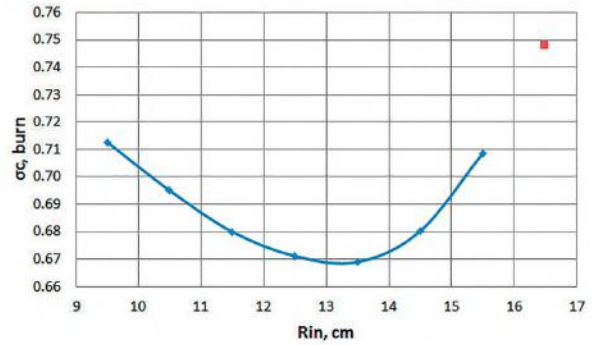
Figs. 15–18 show fission and capture microscopic cross sections in outer zone for ^{237}Np and ^{243}Am. The fission and capture cross sections for outer zone increase in case of increasing the volume of inner zone. As a result, a change in the geometric parameters of outer zone does not allow optimizing the transmutation characteristics of the two-zone subcritical system.
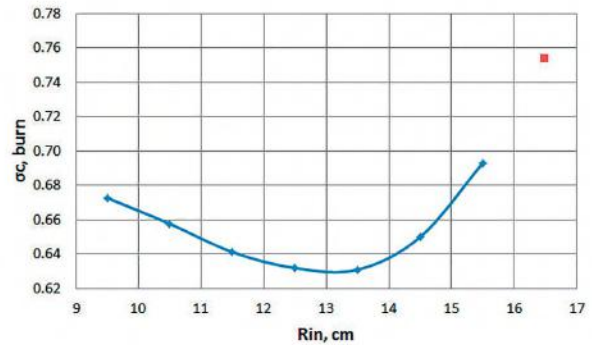
Figs. 19–22 show the ratios of capture and fission microscopic cross sections in inner and outer zones for ^{237}Np and ^{243}Am. Figures 19 and 20 show that it is possible to choose such a ratio of the volumes of inner and outer zones at which the probability of transmutation for both ^{237}Np and ^{243}Am will be maximum. This optimal volume ratio is observed when the radius of inner zone is approximately 13–14 cm for both isotopes. For outer zone, it is slightly different (Figs. 21 and 22), here in case of decreasing the volume of outer zone, the probability of transmutation of the chosen isotopes will decrease. But at the same time, with optimal radii for inner zone, it is possible to
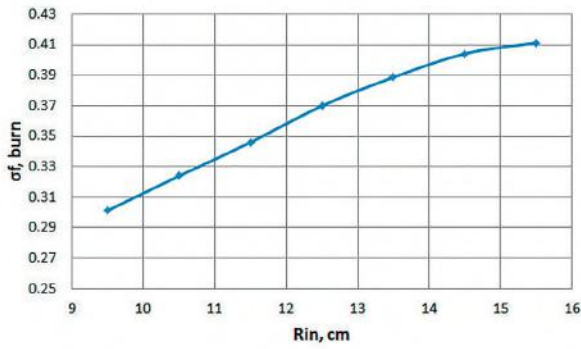

Fig. 12. The fission microscopic cross-section of ^{243}Am for inner zone

Fig. 13. The capture microscopic cross-section of ^{237}Np for inner zone

Fig. 14. The capture microscopic cross-section of ^{243}Am for inner zone

Fig. 15. The fission microscopic cross-section
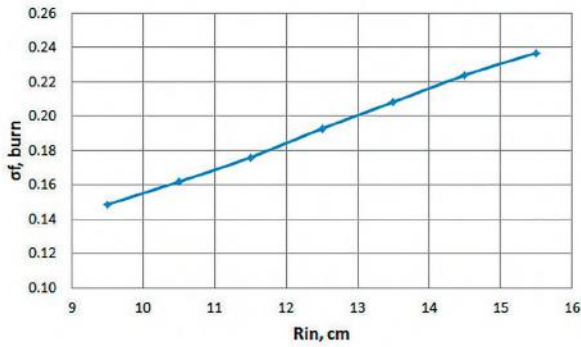of ^{237}Np for outer zone

Fig. 16. The fission microscopic cross-section
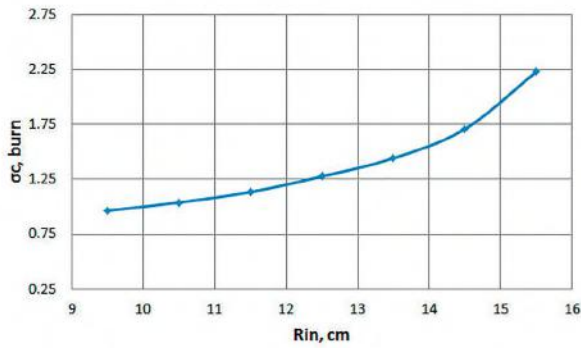of ^{243}Am for outer zone

Fig. 17. The capture microscopic cross-section
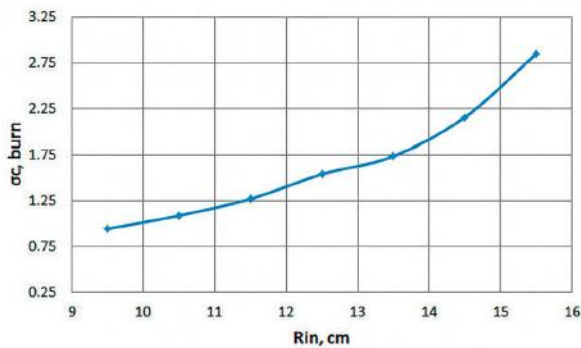of ^{237}Np for outer zone

Fig. 18. The capture microscopic cross-section
of ^{243}Am for outer zone

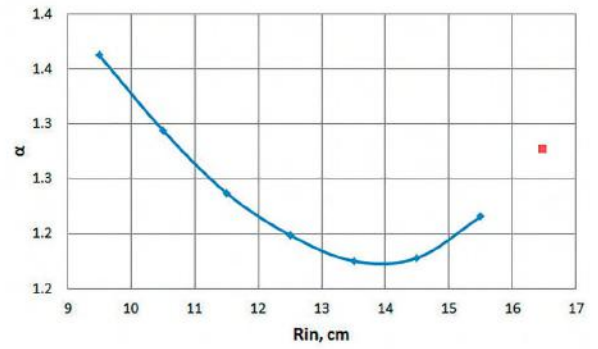Fig. 19. The ratio of the average capture to fission
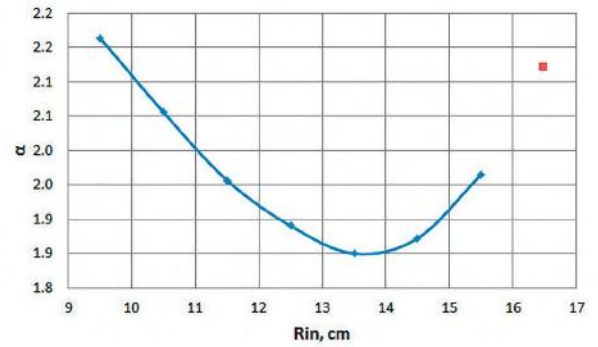cross section of ^{237}Np in the inner zone

Fig. 20. The ratio of the average capture to fission
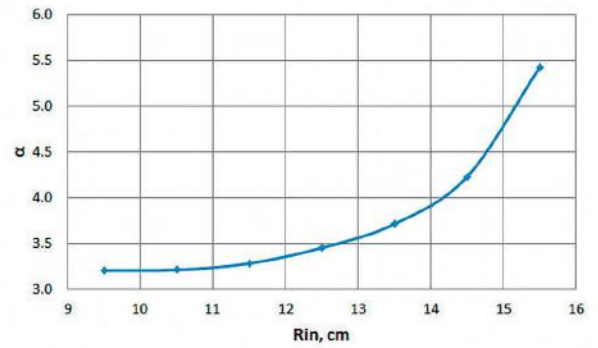cross section of ^{243}Am in the inner zone

Fig. 21. The ratio of the average capture to fission
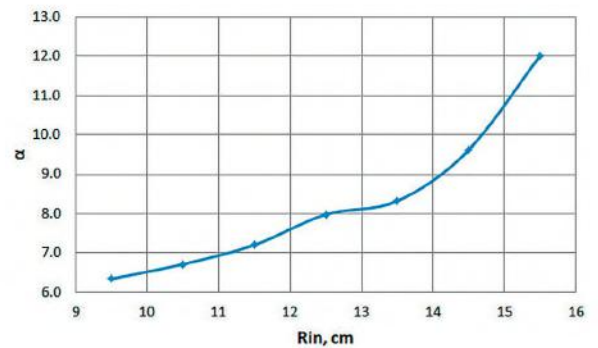cross section of ^{237}Np in the outer zone

Fig. 22. The ratio of the average capture to fission
cross section of ^{243}Am in the outer zone

maintain acceptable values of the ratios of capture and fission microscopic cross sections for outer zone.

The presented modelling results show that even a simple distribution of materials with different nuclear properties (different enrichment for ^{235}U) in the core can have an effect on the transmutation characteristics of the system. In this case, it is possible to choose the ratios of the volumes of different materials in such a way that these characteristics are optimal. Also, such a distribution of different materials may, if necessary, reduce the cost of a subcritical reactor due to a more efficient arrangement of highly enriched uranium. The presented example of division into subzones with different materials can be applied in the internal "fast" zone of the subcritical reactor shown in Fig. 2.

The Figs. 10, 14, 15, 20 and 21 display the optimal fuel composition for two-zone subcritical reactor from a viewpoint of transmutation efficiency for minor actinides. The optimization research presented in our previous publication [32] showed that the optimal fuel composition could be obtained for two-zone subcritical reactor from fuel cost perspective. The presented calculation results for two-zone homogeneous subcritical system without coolant and cladding materials could be a good basis for the development of effective heterogeneous model for future research in subcritical reactor systems.

The energy-release calculations for two-zone subcritical system

If we return to the model of a reactor with separate zones with fast and thermal neutron spectra, then it is of interest to consider the distribution of energy-release in the active core composed of two zones with different neutron spectra.

The energy-release calculations for two-zone subcritical system described in Chapter 2.2 were estimated with the help of the developed MCNP model. The total energy-release within the entire subcritical system was found to be 412.67 kW (for intensity of neutron generator = $1 \cdot 10^{13}$ neutron/s), while the energy-release in inner zone was found to be 70.85 kW and the energy-release in outer zone was 341.82 kW. The energy-release (Q) for each pin located on x axis from the target to the beryllium reflector (see Figs. 3–6) is presented in Fig. 9. The energy-release (Q) in subcritical reactor was calculated with taking into account total neutron fissions in the core, averaged energy release from fission (about 200 MeV) and intensity of neutron generator (in this case = $1 \cdot 10^{13}$ neutron/s).

From Fig. 23 we can identify which pin's layer has the maximum and minimum of energy-release. It is clear that pins in outer zone possess higher energy-release due to thermal neutron spectrum. In addition, it can also be noted that the thermal neutron spectrum has influence on inner zone where pins 13 and 14 have higher energy-release compared with the other pins in inner zone.

From Fig. 23 we can see that the average energy-release, and as a result, the fuel burn-up in outer ("thermal") zone is about five times higher than the average energy-release in inner ("fast") zone. This may conduct additional difficulties in the operation of the future two-zone subcritical reactor.
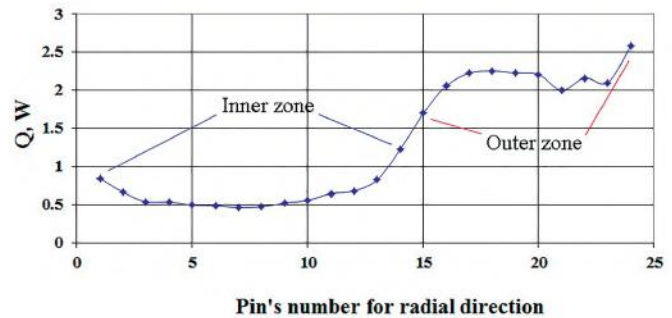

Fig. 23. The energy-release for individual pin located on x axis for radial direction versus the pin's number

Conclusions

This paper presents the idea behind the development of the two-zone subcritical research reactor driven by high-intensity neutron generator for investigation of nuclear waste transmutation along with the methodology and results of the performed simulations by various Monte Carlo codes.

The neutron-physical model of the presented subcritical reactor with the help of MCNP and Serpent Monte Carlo codes was developed and the energy-release calculations with the help of MCNP model were carried out.

The microscopic fission cross-section, microscopic capture cross-section and ratio of these cross-sections for ^{237}Np and ^{243}Am were estimated for the developed model. It was found that there is an optimal value of ratio of inner zone volume to outer zone volume from the viewpoint of transmutation efficiency for minor actinides.

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В. І. Гулік[1], В. М. Павлович[2]

[1]*Інститут проблем безпеки АЕС НАН України, вул. Лисогірська, 12, м. Київ, 03028, Україна*
[2]*Інститут ядерних досліджень НАН України, просп. Науки, 47, м. Київ, 03680, Україна*

Розробка нейтронно-фізичної моделі двозонного дослідницького підкритичного реактора для трансмутації ядерних відходів

Створення невеликих дослідницьких підкритичних реакторів необхідно, зокрема, для розробки технології трансмутації ядерних відходів у підкритичних системах, що керуються зовнішнім джерелом нейтронів. Будівництво таких установок дає змогу розробити технологію трансмутації ядерних відходів без будівництва дорогого промислового

підкритичного реактора. Недорогий генератор нейтронів може служити зовнішнім джерелом нейтронів для таких дослідницьких підкритичних реакторів. У роботі запропоновано двозонну модель підкритичної системи, що керується генератором нейтронів високої інтенсивності. Така система може використовувати дві окремі зони реактора з різними нейтронними спектрами: швидким та тепловим. Представлено основні етапи розробки запропонованої моделі двозонного підкритичного реактора. Результати моделювання виконано в кодах Serpent і MCNP з метою оптимізації геометрії і складу палива у двозонній підкритичній системі. Також представлено огляд різних установок, які можуть бути використані як зовнішнє джерело нейтронів для підкритичного реактора. Однак оптимальним джерелом нейтронів для недорогого дослідницького підкритичного реактора для дослідження трансмутації ядерних відходів був обраний D-T генератор нейтронів високої інтенсивності. У цілому спостерігається, що двозонна підкритична система може ефективно посилювати потік нейтронів від зовнішніх джерел нейтронів.

Ключові слова: підкритичні системи, що керуються прискорювачами, підкритичний реактор, трансмутація радіоактивних відходів, оптимізація підкритичної зони.

В. И. Гулик[1], В. Н. Павлович[2]

[1]*Институт проблем безопасности АЭС НАН Украины, ул. Лысогорская, 12, г. Киев, 03028, Украина*
[2]*Институт ядерных исследований НАН Украины, просп. Науки, 47, г. Киев, 03680, Украина*

Разработка нейтронно-физической модели двухзонного исследовательского подкритического реактора для трансмутации ядерных отходов

Создание небольших исследовательских подкритических реакторов необходимо, в частности, для разработки технологии трансмутации ядерных отходов в подкритических системах, управляемых внешними источниками нейтронов. Строительство таких установок позволит разработать технологию трансмутации ядерных отходов без строительства дорогостоящего промышленного подкритического реактора. Недорогой генератор нейтронов может служить внешним источником нейтронов для таких исследовательских подкритических реакторов. В данной работе предложена двухзонная модель подкритической системы, которая управляется генератором нейтронов

высокой интенсивности. Данная система может использовать две отдельные зоны реактора с разными нейтронными спектрами: быстрым и тепловым. Представлены основные этапы развития предлагаемой модели двухзонного подкритического реактора. Результаты моделирования выполнены в кодах Serpent и MCNP с целью оптимизации геометрии и состава топлива двухзонной подкритической системы. Представлен обзор анализа различных установок, которые могут быть использованы в качестве внешнего источника нейтронов для подкритического реактора. Однако в качестве оптимального источника нейтронов для недорогого исследовательского подкритического реактора для исследования трансмутации ядерных отходов был выбран D-T генератор нейтронов высокой интенсивности. В целом показано, что двухзонная подкритическая система может эффективно усиливать поток нейтронов от внешних источников нейтронов.