V Y T A U T A S M A G N U S U N I V E R S I T Y

I N S T I T U T E O F P H Y S I C S

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EVOLUTION OF TRANSURANIUM ISOTOPIC COMPOSITION IN POWER REACTORS AND INNOVATIVE NUCLEAR SYSTEMS FOR TRANSMUTATION

The summary of the doctoral thesis

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This work was performed at the Nuclear and Environmental Radioactivity Research Laboratory of the Institute of Physics (Lithuania) in 1999-2000 / 2002-2003 and in CEA Saclay (France), DSM/DAPNIA/SPhN Nuclear Physics Division, Mini-Inca group in 2000-2002.

GENERAL REMARKS

Topicality of the subject

The nuclear energy started some 50 years ago as a promising source of cheap and clean energy supply for rapidly growing new, energy-intensive technologies. Reactor accidents at Three Mile Island (Pennsylvania) in 1979 and in Chernobyl (Ukraine) in 1986 undermined public confidence in reactor technologies, and the nuclear energy growth slowed. Recently, improved technology with safe operating performance of nuclear power plants and its environmental advantages renewed interest in nuclear energy. Generation IV (from 2020) nuclear energy systems are supposed to provide sustainability, competitive economics, safety and proliferation resistance [1]. However, another serious problem will remain – long-lived highly radioactive nuclear waste, already generated during last 5 decades. Indeed, nuclear waste from present commercial power plants and dismantled strategic materials contain large quantities of plutonium, other actinides (neptunium, americium and curium) and long-lived fission products (iodine, technetium). Although these isotopes contribute only 1% to the total spent nuclear fuel, they will determine the highest radiotoxicity and hazard. If one assumes the same level of global nuclear power generation for the future as it exists today – there will be more than 250 000 tons of spent nuclear fuel worldwide by 2015.

Disposal of discharged fuel or other high-level radioactive residues in a geological repository is the preferred choice in most countries. However, this solution suffers from the uncertainties of the safety and impregnability of the storage over hundreds of thousands of years. Hence the possibilities of partitioning and transmutation of long-lived radioactive waste into stable or short-lived isotopes, which could then be surface-stored with little/no proliferation value, are now under investigation. In addition, transuranium elements could serve as fuel for transmutation systems. Several solutions of long-term radiotoxicity reduction by transmutation in new transmutation systems have been studied in the current years. The idea of combining a powerful accelerator (external neutron source) with a sub-critical reactor core for transmutation purposes is evolved [2]; new critical reactors to burn nuclear waste are also being studied [3,4].

In principle, any intensive neutron source could be used for waste transmutation. The main parameters defining the transmutation process are the neutron energy spectrum and neutron fluxes of the system. The more intensive neutron flux determines the shorter lifetime of a certain nucleus in the system flux. On the other hand, transmutation depends on neutron capture and the neutron-induced fission cross sections. A unique solution is impossible because capture and fission cross sections vary considerably from one isotope to another. Looking for a new efficient and economically viable transmutation system the main consideration is the neutron flux and spectrum characteristics of a particular system.

In order to investigate the dependence of transmutation efficiency on the neutron spectrum, in this work two systems with different neutron properties, mainly GT-MHR (with variable thermalized neutron flux) and a fusion-fission hybrid system (with epithermal-fast neutron flux), were analyzed. In the case of a fusion-fission hybrid system both the materials and technology development are still needed. However, in the first stage of the system analysis the neutronic characteristics study in the transmutation blanket is of great relevance [5]. Recently the worldwide interest in HTR (high temperature reactor) technology is experienced [3,6,7]. Transmutation concept in GT-MHR relies on the use of high burnup particle fuel and gradually thermalized neutron spectrum in the graphite moderated system. GT-MHR complicated structure causes the problem of exact core modelling via analytical methods [6]. Detailed study of the main performance parameters of GT-MHR and technological solutions are the main presumptions for implementation of this reactor in the future. The processes in microscopic coated particles and nuclide evolution in the 3D reactor core can be assessed by Monte Carlo method.

The quality of computer code simulations (based on Monte Carlo techniques) depends on the quality of the existing nuclear data libraries. Most of the data for minor actinides contained in the main nuclear data libraries are contradictory due to the lack of experimental data. For this reason, the integral experiments are very important for nuclear data development and code validation purposes [8, 9].

Objectives of the thesis

This thesis is dedicated to reduction of transuranic part of nuclear waste by transmutation. The general aim is the pioneering development of physical basics of transmutation analysis, the contribution to transmutation studies in Lithuania and the progress worldwide.
The main tasks of the thesis are:

1. Investigation and optimization of the neutronic characteristics of the sub-critical fusion-fission hybrid system transmutation blanket for effective transuranic element transmutation.
2. Development of the critical high temperature helium cooled reactor model and investigation of neutron spectrum properties and main performance parameters as a function of different active core geometry.
3. Investigation of the final RBMK spent nuclear fuel composition transmutation feasibility and evaluation of burning rate in the considered transmutation systems (1 and 2 as above).
4. Comparison of reliability of different data libraries by modelling high temperature reactor and identification of the major isotopes for which a significant deviation between different data files exists.
5. Investigation of neutron cross section for the ^{241}Am, ^{243}Am and ^{242}Pu isotopes in the high intensity thermal neutron flux.

The scientific novelty

In this work two different transmutation systems, a fusion-fission hybrid system and a high temperature modular helium cooled reactor (GT-MHR), were analyzed in order to investigate the transmutation process in different neutron energy spectrum range (GT-MHR – thermalized variable neutron spectrum during the burnup, a fusion-fission hybrid system – epithermal-fast neutron spectrum).

For the first time an exact GT-MHR core was modelled using a Monte Carlo method which permits evaluation of the neutron interaction processes in microscopic ceramic-coated fuel and burnable poison particles.

The possibility to transmute different transuranium isotopic composition was studied in this work. For different types of existing nuclear reactors the amount of Pu and minor actinides formed in the spent nuclear fuel is not the same. It seems that a chemical element separation does not cause a big problem any longer. On the other hand, to perform an isotopic separation is a much more complicated task with today's technology. In this work the sensitivity due to different isotopic fuel composition of the considered transmutation systems was analyzed.

The major differences between different nuclear data libraries in terms of averaged one-group cross sections by modelling GT-MHR were investigated. The isotopes with contradictory neutron cross sections were identified. New experimental nuclear data for ^{241}Am, ^{243}Am ^{242}Pu were obtained in the high thermal neutron flux.

The scientific and practical significance

Implementation of partitioning and transmutation technology is intended to reduce the inventories of actinides in nuclear waste. The study of GT-MHR and a fusion-fission hybrid system enhances the development of new generation transmutation systems. The detailed study of neutronic characteristics of a fusion-fission hybrid system will help to optimize the molten salt transmutation blanket and to achieve better system performance parameters. The simulation

results with a Monte Carlo approach including an exact GT-MHR geometry are intended to serve as a reference calculation for other (less detailed) methods as a deterministic approach.

RBMK-1500 SNF plutonium and MA transmutation scenarios in GT-MHR and a fusion-fission hybrid system are the first investigation of this type, which may be useful for the final decision of nuclear waste strategies in Lithuania.

The identification of isotopes with considerable nuclear data discrepancies and experimental measurements of ^{241}Am, ^{243}Am and ^{242}Pu neutron capture-fission cross sections will serve for the nuclear data library improvement and new evaluations.

Publications on the subject of the thesis and approbation

Articles:

A1. E. Kimtys, A. Plukis, R. Plukienė, G. Bražiūnas, P. Goberis, A. Gudelis, R. Druteikienė and V. Remeikis, "Analysis of plutonium isotopic ratios using the SCALE 4.4A code package", Environmental and Chemical Physics, Vol. 22, No 3-4, (2001), pp. 112-116.

A2. R. Plukiene, D. Ridikas, E.T. Cheng, "Fusion – Fission Hybrid System with Molten Salt Blanket for Nuclear Waste Transmutation", Proc. of the *XIXth Int. Autumn School on New Perspectives with Nuclear Radioactivity (NRAD2001),* Portugal, (2001). Preprint CEA Saclay, (2002), pp.1-14. http://www-dapnia.cea.fr/Doc/Publications/Archives/dapnia-02-04.pdf

A3. D. Ridikas, L. Bletzacker, O. Deruelle, M. Fadil, G. Fioni, A. Letourneau, F. Marie, R. Plukiene, "Comparative analysis of ENDF, JEF and JENDL data libraries by modelling high temperature reactors and plutonium based fuel cycles", Journal of Nuclear Science and Technology, supplement 2, (2002), pp.1167-1170.

A4. G. Fioni, O. Deruelle, M. Fadil, A. Letourneau, F. Marie, R. Plukiene, D. Ridikas, I. Almahamid, D.A. Shaughnessy, H. Faust, P. Mutti, G. Simpson, I. Tsekhanovich, "The Mini-Inca project: experimental study of the transmutation of actinides in high intensity neutron fluxes", Journal of Nuclear Science and Technology, supplement 2, (2002), pp.876-879.

A5. R. Plukiene and D. Ridikas, "Modelling of HTRs with Monte Carlo: from a Homogeneous to an Exact Heterogeneous Core with microparticles", Annals of Nuclear Energy, 30/15, (2003), pp. 1573 – 1585.

Aprobation of the results:

B1. R. Kavaliauskaitė, A. Plukis, V. Remeikis, „Pu izotopinės sudėties tyrimas γ-spektrometriniu metodu 20-60 keV energijų srityje", *Vilniaus universiteto 33-oji Lietuvos nacionalinė fizikos konferencija*, Programa ir pranešimų tezės, Vilnius, (1999), 204 p.

B2. R. Plukiene, D. Ridikas, E.T. Cheng, "Fusion – Fission Hybrid System with Molten Salt Blanket for Nuclear Waste Transmutation" Proc. of the *XIXth Int. Autumn School on New Perspectives with Nuclear Radioactivity (NRAD2001),* Lisbon, Portugal, (2001).

B3. D. Ridikas, L. Bletzacker, O. Deruelle, M. Fadil, G. Fioni, A. Letourneau, F. Marie, R. Plukiene, "Comparative Analysis of ENDF, JEF and JENDL Data Libraries by Modelling High Temperature Reactors and Plutonium Based Fuel Cycles: Military Pu case", Proc. of the *Int. Conference on Nuclear Data for Science and Technology (ND2001)*, Tsukuba, Japan, (2001).

B4. D. Ridikas, L. Bletzacker, O. Deruelle, M. Fadil, G. Fioni, A. Letourneau, F. Marie, R. Plukiene, "Comparative Analysis of Different Data Libraries for the Performance of High Temperature Reactors (GT-MHR): Civil Pu case", Proc. of *the Int. Conference on Accelerator Applications/Accelerator Driven Transmutation Technology and Applications* (AccApp/ADTTA'01), Reno, Nevada, USA, (2001).

B5. G. Fioni, O. Deruelle, M. Fadil, A. Letourneau, F. Marie, R. Plukiene, D. Ridikas, I. Almahamid, D.A. Shaughnessy, H. Faust, P. Mutti, G. Simpson, I. Tsekhanovich, "The

Mini-Inca project: experimental study of the transmutation of actinides in high intensity neutron fluxes", Proc. of *the Int. Conference on Accelerator Applications/Accelerator Driven Transmutation Technology and Applications* (AccApp/ADTTA'01), Reno, Nevada, USA, (2001).

B6. O. Deruelle, M. Fadil, G. Fioni, A. Letourneau, F. Marie, R. Plukiene, D. Ridikas, "Measurement of neutron capture cross sections relevant for nuclear waste transmutation, by alpha and gamma spectroscopy", *The 11th International Symposium on Capture Gamma-Ray Spectroscopy and Related Topics,* Prague, Czech Republic, (2002).

B7. D. Ridikas, R. Plukiene, E.T. Cheng, "Fusion – Fission Hybrid System for Minor Actinides Transmutation", *International workshop on Fast Neutron Physics*, Dresden, Germany,(2002).

B8. D. Ridikas and R. Plukiene, "Modelling of HTRs: from Homogeneous to Exact Heterogeneous Core with Monte Carlo", Proc. of *the Int. Conference PHYSOR 2002,* Seoul, Korea, (2002).

B9. R. Plukienė, D. Ridikas, A. Plukis, V. Remeikis, „Aukštatemperatūrių reaktorių modeliavimas: skirtingos kuro izotopinės sudėties įtaka", *35-oji jubiliejinė Lietuvos nacionalinė fizikos konferencija*, Programa ir pranešimų tezės, Vilnius, (2003), 153 p.

B10. R. Plukienė, D. Ridikas, A. Plukis, V. Remeikis, „RBMK ir LVR radioaktyviųjų atliekų transmutavimo scenarijai branduolinės sintezės-dalijimosi sistemoje", *35-oji jubiliejinė Lietuvos nacionalinė fizikos konferencija*, Programa ir pranešimų tezės, Vilnius, (2003), 154 p.

B11. D. Ridikas and R. Plukiene, „Comparative Analysis of ENDF, JEF & JENDL Data Libraries by Modelling the Fusion-Fission Hybrid System for Minor Actinide Incineration", *Workshop on Nuclear Data for the Transmutation of Nuclear Waste*, GSI-Darmstadt, (2003).

Personal contribution:

The modelling of transmutation systems and spent nuclear fuel composition of power reactor as well as interpretation of the results described in the thesis has been performed by the author. The experimental measurements and determination of actinides cross sections at ILL Grenoble (France) have been obtained when participating in the experimental Mini-Inca group activities.

Overview of the thesis

The thesis consists of an introduction, 5 chapters, conclusions and a list of references (158 items). The material of the thesis is presented in 123 pages, including 59 illustrations and 39 tables. Topicality of the subject, tasks, scientific novelty and practical significance are formulated. The list of the scientific publications and contributions at the conference is presented.

Chapter 1. REVIEW OF THE LITERATURE ON TRANSMUTATION

In **chapter 1** the review of transmutation methods for solving long-lived radioactive waste problem is presented. The way of generation of nuclear waste in the reactors is briefly introduced at the beginning. Different strategies for dealing with nuclear waste are discussed. A general consensus indicates that only through nuclear reactions one can eventually eliminate the long-term uncertainties of geological repositories. Indeed, the implementation of partitioning and transmutation (P&T) technology may decrease the hazards associated with long-lived nuclear waste. The stages of transmutation process and the various scenarios for closing fuel cycles in different countries (Japanese, French, Russian, CERN-Rubbia and American approaches) are described including the scenarios, associated scientific problems and achievements [10]. These scenarios are built around single or double strata concepts, according to the way the radionuclides are recycled in the existing nuclear facilities (single stratum) or in specific

facilities (fast neutron reactors or hybrid systems). The transmutation process, a term covering all transformations of nuclei by nuclear reactions, is seeking to transform long-lived radioactive nuclei from spent nuclear fuel into shorter-lived elements [2]. The main trends in transmutation are: the recycling of MOX (*Mixed Oxide Fuel*) in the LWR fuel cycle or the secondary use of LWR fuel in CANDU reactors [11,12], optimization of Fast Reactors with the objective to maximize plutonium and actinide consumption and feasibility studies of new systems (critical or sub-critical) able to significantly reduce the inventories of long-lived isotopes and development of advanced reprocessing techniques [12,13].

The neutronic parameters governing the transmutation process of the critical and sub-critical systems are explained. Any type of transmutation is a function of the neutron energy and corresponding cross section dependence. If long-lived fission products can be transmuted only by neutron capture, there are two possibilities for actinides: a) to promote direct fission or b) to promote neutron capture leading to fissionable daughter isotopes. The preferred physics process is a direct fission: the conversion of initial nucleus into relatively short-lived fission products, no parasitic capture and the end of chain without production of minor actinides. The competition between the capture and fission processes is then of high relevance. The average capture and fission cross section rates ($\alpha = \overline{\sigma_c} / \overline{\sigma_f}$) for most of actinides are preferred (they are smaller) in fast neutron spectra [14]. On the other hand, transmutation depends not only on the microscopic cross sections but also on the neutron flux intensity. The higher neutron flux is available – the shorter effective lifetime of a certain nucleus is in this flux. The neutron cross sections for capture and fission reactions decrease significantly as a function of neutron energy. The large cross sections of thermal neutrons provide for high specific destruction rates and together with the relatively low radiation damage, they allow for higher burnups to be achieved than in fast or epithermal spectra [3].

Most of the proposed transmutation systems reflect the need for variety of different spectra in order to have a more efficient and economically viable transmutation. Unluckily, a unique solution is impossible, simply because capture and fission cross sections vary considerably from one isotope to another. The new generation nuclear reactors and particular systems for transmutation are reviewed. The neutron excess can be provided by an external neutron source. Accelerator driven sub-critical systems (ADS) for radioactive waste transmutation and energy production for the first time were proposed by C. Rubbia [15] and C.D. Bowman [16]. For implementation of ADS, the spallation target (neutron production) and proton beam optimization are needed together with development of new spent fuel partitioning technique and materials technology. It seems that a plasma-based fusion device could provide as intense neutron source as a high power accelerator, say, up to 10^{19} n/s. This fusion based system is at the very first stage of development, and the studies on the blanket concept for destroying the actinides are of great relevance [5,17]. Research studies on the critical reactor are carried out to evaluate the transmutation potential of long-lived waste in appropriate reactor configuration based on current technologies (LWR, CANDU) as well as innovative reactors (like GT-MHR, AMSTER) [3,4,11,12]. Evaluations of the theoretical feasibility of the Pu consumption and waste transmutation from the point of view of the reactor core physics and of the isotopic compositions of the fuels insure the “scientific” part of the transmutation feasibility.

In this work the main focus was on new systems for transmutation: the sub-critical fusion-fission hybrid system and a new generation critical high temperature reactor (GT-MHR).

Chapter 2. CALCULATION METHODS AND CODES

For the investigation of nuclear processes and calculation of main performance parameters of the nuclear reactors, different simulation codes are used. The simulation of the behavior of a reactor involves two distinct steps: evaluation of neutron transport in the reactor (neutron spectrum, the mean interaction cross sections averaged over the neutron spectrum) and

evaluation of fuel evolution over a time short enough so that the neutron spectrum and the mean cross sections are approximately constant. The obtained new composition of the medium is the starting point for a new neutron transport evaluation in the reactor. By repeating these two steps as many times as necessary, the behavior of the reactor over long time periods can be obtained.

A great number of simulation codes with different modelling approaches exist by now. The modelling methods and code packages are chosen depending on the research needs and the goal of the particular investigation. The deterministic methods usually are used for numerical modelling of the critical reactor, while, Monte Carlo is used for modelling of both critical and sub-critical systems. Deterministic codes and code packages like HELIOS, SCALE 4.4a [19], WIMS could be adapted for different power plant modelling. The neutron flux spectrum and corresponding neutron cross sections are obtained by solving a simplified Boltzmann equation for each neutron group for the cell network. In practice, experiments are needed to validate the group cross-sections for each type of reactors.

To obtain the RBMK-1500 reactor spent fuel composition SCALE 4.4a (*Standardized Computer Analyses for Licensing Evaluation*) code package was used [18]. For each time depended isotopic composition of nuclear fuel, the SCALE 4.4a program sequence SAS2 performs analysis of 1D neutron flux for the reactor fuel assembly using a two-part procedure with two separate cell models.

In brief, particle transport using the Monte Carlo technique consists of following each of many particles from a source throughout its life to its death (by absorption or escape). Probability distributions are randomly sampled using transport data. These distributions are used to determine the type of interaction, energy of particles when they scatter, leakage of particles and the number of neutrons produced if fission occurs. The most frequently used code to describe these processes is MCNP (*Monte Carlo N-Particle Code*) [19]. The fluxes of the neutrons determined in this way can be used to tally a wide variety of information (reaction rates, heating rates, criticality eigenvalue etc.). MCNP is well suited to solve complicated three-dimensional, time-dependent problems.

Monteburns [20] is an automatic-cyclic coupling of MCNP [19] and ORIGEN2 [21] codes, which was employed at different stages of the transmutation systems (fusion-fission hybrid system and GT-MHR) simulations. The main function of Monteburns is to transfer one-group cross section and flux values from MCNP to ORIGEN2, and then to transfer the resulting material composition (after irradiation and/or decay) from ORIGEN2 back to MCNP in a repeated, cyclic fashion. ORIGEN2 performs burnup calculations using the matrix exponential method in terms of time-dependent formulation, destruction and decay concurrently. The advantage of the Monteburns code is that it can use different nuclear data libraries as long as they are compatible with MCNP format. The quality of the results obtained by computer codes depends on nuclear parameters of isotopes involved in the incineration process.

Chapter 3. ANALYSIS OF THE FUSION-FISSION HYBRID SYSTEM

In **subchapters 3.1 and 3.2** the neutronic model of the fusion-fission hybrid system with a molten salt (LiF-BeF_2-$(HN)F_4$ – "flibe") blanket was presented including simulation details.

In order to transmute efficiently the nuclear waste, the high intensity neutron source is needed. An inertial confinement fusion (ICF) device (based on $D+T={}^4He+n$ (14 MeV) nuclear reaction) could provide with a powerful neutron source - 1 MW fusion power gives $\sim 4\times10^{17}$ n/s [16]. A molten salt blanket, surrounding this neutron source, then could serve as a medium for transuranic actinides (TRU) to be burned. Flibe also has a function of both coolant and carrier of tritium breeding material (^6Li in this case). A well known advantage of the molten salt is a possibility of both refueling of burned TRU and extraction of fission products (FP) on-line. A simplified fusion-fission hybrid system was modelled with the main geometry and material parameters given in Table 1. Fig. 1 presents a simplified geometry setup used in calculations.

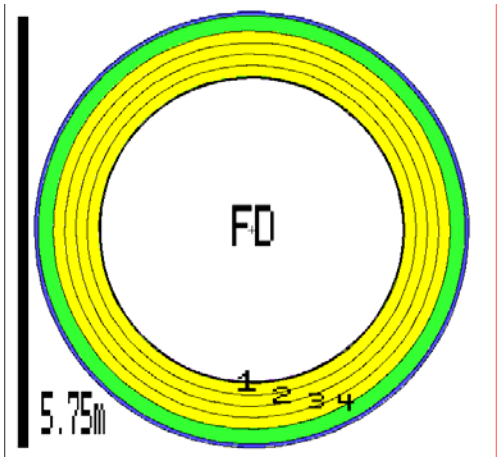


Fig. 1. A simplified geometry model of the fusion-fission hybrid system used in the calculations. FD stands for fusion device – the place where d+T reactions occur.

Table 1. Neutronics model: zone dimension and material compositions for the molten salt transmutation blanket .

Zone Name	Radius R_i- R_{i+1}, cm	Thickness, cm	Material composition
Cavity	0 – 200	200	Void
Liquid wall	200 – 201	1	Flibe (a)
Metallic wall	201 – 201.3	0.3	SS316, 50%
Graphite	201.3 –202.3	1	Graphite
Blanket			
Region 1	202.3 –217.3	15	Flibe (b)
Region 2	217.3 –232.3	15	Flibe (b)
Region 3	232.3 –247.3	15	Flibe (b)
Region 4	247.3 –262.3	15	Flibe (b)
Reflector	262.3 –282.3	20	Graphite
Envelope	282.3 –287.3	5	SS316, 50%

Flibe (a) contains 0.1% of ^{6}Li compared to the total amount of Li. Flibe (b) density is 2 g/cm^3 and corresponding TRU density is 0.0074 g/cm^3. 0.6 % of ^{6}Li enrichment for flibe (b) was taken as a reference case [17].

All calculations on the flibe-based actinide transmutation blanket were made employing Monteburns code system. MCNP was used to obtain k_{eff} of the blanket due to fission of TRU without an external source in the mode of a criticality eigenvalue problem, and also to estimate neutron flux as well as k_{scr} (the total neutron multiplication coefficient) in its external source mode. In this case k_{scr} is defined as:

$$k_{scr} = (M_n - 1)/(M_n - 1/\nu),$$

where ν is the average number of neutrons per fission and M_n is a total neutron multiplication factor of the system.

For all structure materials and actinides we have chosen ENDF data files [22] being most frequently employed, while for fission products JENDL data library [23] was taken due to the largest number of fission products available (~200). In all cases the fission power was kept constant ($3GW_{th}$), which corresponds to a variable fusion power of the fusion device. In our simulations this implies a renormalization of the absolute neutron flux if an effective neutron multiplication coefficient of the system is changed.

In **subchapter 3.3** a typical molten salt (flibe) blanket, dedicated to burn actinides extracted from the LWR spent nuclear fuel, is analyzed. The investigation of the main performance parameters (k_{scr}, burnup, fluxes and equilibrium conditions), depending on different treatment of fission products as well as different refueling of already destroyed actinides in the molten salt blanket, was performed.

Table 2. Averaged neutron spectrum over all flibe regions, presented in different energy groups (%) as a function of burnup in case of different TRU feeding.

Time,d.	Φ_n, % (**Discrete**)					Φ_n, % (**Continuous**)				
	[0.,1.] eV	[1.,100.] eV	[.1,100.] keV	[.1,1.] MeV	[1.,20] MeV	[0.,1.] eV	[1.,100.] eV	[.1,100.] keV	[.1,1.] MeV	[1.,20] MeV
0	0.16	14.58	62.49	12.28	10.45	0.04	14.58	62.49	12.28	10.45
400	0.16	15.37	62.07	12.02	10.33	0.03	14.84	62.55	12.24	10.19
1200	0.14	15.43	61.87	12.11	10.42	0.04	15.12	62.12	12.2	10.38
2000	0.16	15.33	61.72	12.14	10.62	0.03	14.93	62.22	12.22	10.47

The initial load of the fuel and subsequent feeding material is a typical TRU vector defined by the 30 MWd/kg burnup of LWR spent fuel (to be given below in Table 3). The initial TRU inventory is 3.04 tons, and feeding is 3.1 kg TRU + 0.6 kg of ^{6}Li.

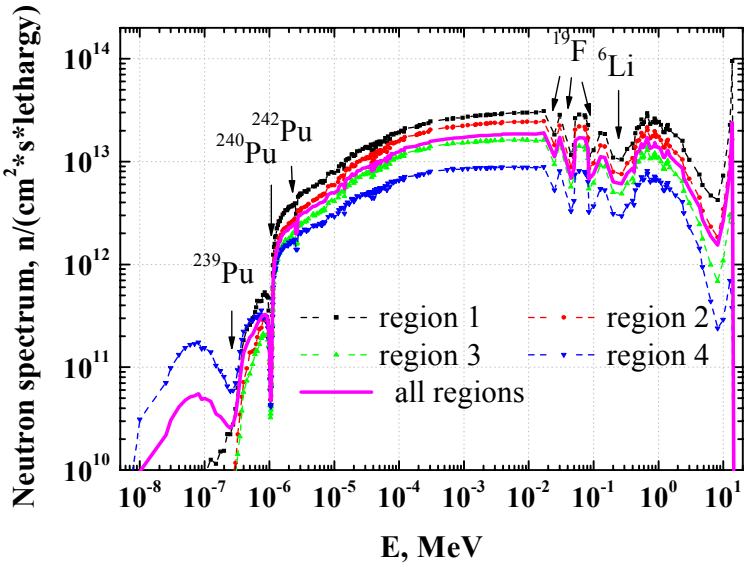


Fig. 2. Profile of the neutron spectrum in different flibe regions. A solid line indicates the average neutron flux over all regions.

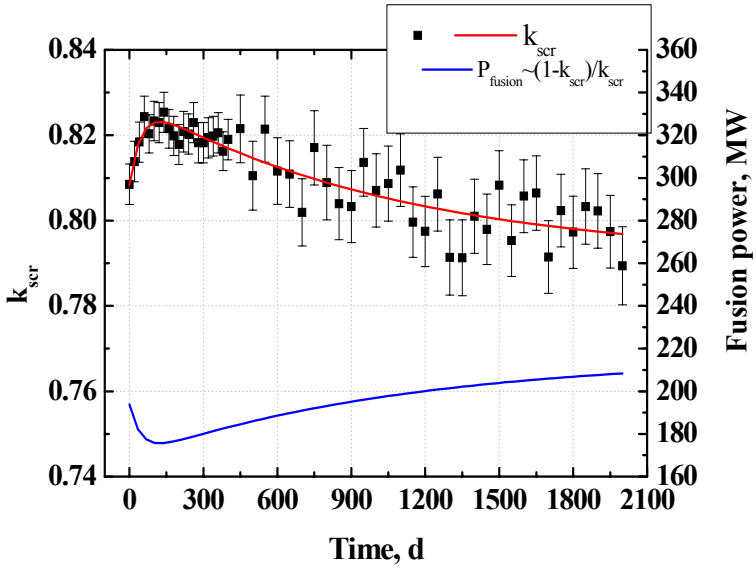


Fig. 3. k_{scr} and fusion power as a function of irradiation time in continuous TRU feeding and removal of FP case.

Fig. 2 presents the neutron spectra in different regions of the molten salt transmutation blanket. The averaged neutron flux is very high (1.48×10^{15} n/s) and corresponds to the flux typical only for high flux reactors. In addition, the amount of thermal neutrons is very small (<1%). Most of the neutrons are epithermal or fast, which could favor the transmutation of actinides in the resonance region. Different replenishing of the destroyed TRU material was tested (continuous and discrete feeding). Table 2 gives a neutron distribution in different energy groups during irradiation time. It is clearly seen that the neutron spectrum remains stable and does not depend on either continuous or discrete feeding of a fresh fuel is simulated.

The criticality of the system depends on actinides isotopic composition and fission product (FP) concentration in the blanket. A corresponding behavior of k_{scr} and required fusion power P_{fusion} at a constant fission power of 3000 MW_{th} for continuous feeding and FP removal are presented in Fig. 2. The increase of k_{scr} occurs mainly due to the mass conversion of ^{240}Pu into ^{241}Pu during the early stage of irradiation (see Fig. 4 and Fig. 5 for detailed). The variation of fusion power is between 175 MW and 210 MW.

A typical evolution of the TRU isotopic composition is presented in Fig. 4. This is the case when the system is refueled continuously. In addition, the fission products are also continuously removed on-line at the 100% rate. Since not all actinides are stabilized even after 2000 days of operation (Cm isotopes in particular), we assume that the system reaches equilibrium when the variation of Pu mass is less than 4% with respect to the actual plutonium mass. According to this assumption we conclude that the equilibrium concentration for most of the TRU materials in the blanket can be achieved after about 3 years of exposure. When the equilibrium situation is reached, about 3.1 kg of TRU is burned per day, which corresponds to nearly 1.1 tons of TRU burned annually with an output of 3 GW_{th} fission power. In equilibrium, a significant amount of curium isotopes is to be accumulated, while for the rest of actinides fully equilibrated concentrations are reached.

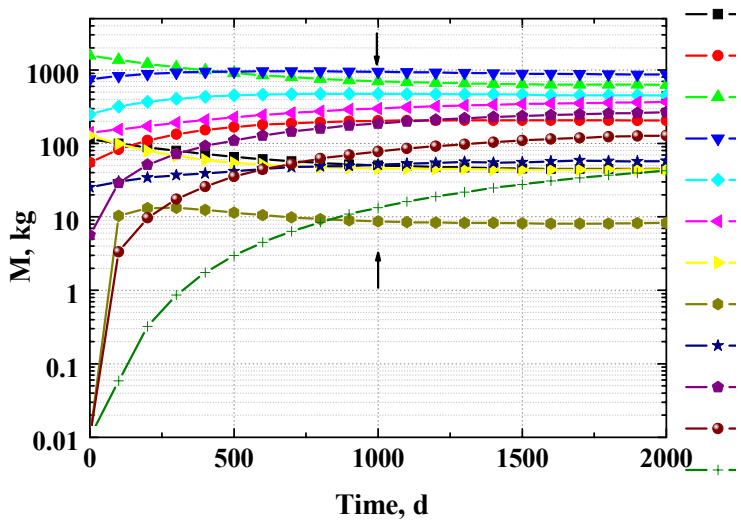


Fig. 4. TRU mass dependence on irradiation time: continuous feeding with 3.1 kg/d of TRU and continuous removal of all fission products.

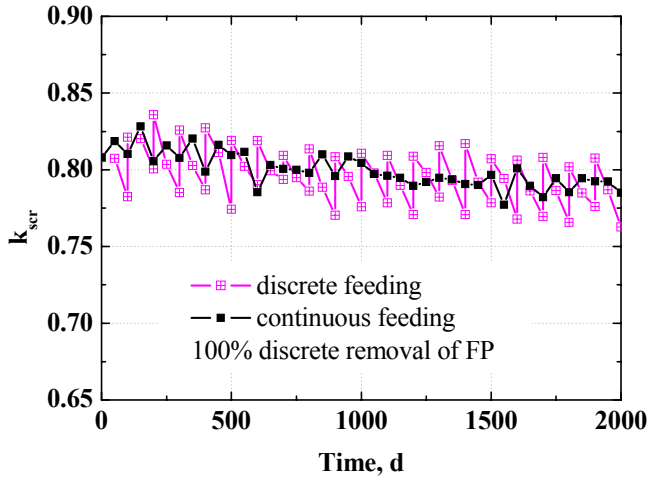


Fig. 5. k_{scr} as a function of discrete and continuous TRU feeding and with discrete removal of all (100%) fission products at the end of steps.

In general, it is preferred that an effective neutron multiplication factor is as stable as possible for any nuclear reactor being either critical or sub-critical. Fig. 4 illustrates the effect of different feeding options on k_{scr}. Discrete feeding determines sharp variation of k_{scr} (consequently, possible sharp variations of the reactor power), the situation is improved considerably if continuous feeding option is chosen. On the other hand, a different TRU mass in the blanket in equilibrium was obtained (3100 kg and 2770 kg with continuous and discrete feeding, respectively). Although a smaller TRU mass in equilibrium is preferred due to safety reasons, a more stable system performance is obtained in the case of continuous feeding. The way fission products are removed continuously or discretely (as long as they are removed 100%) has little influence on the performance of the system. On the other hand, the percentage of removed FP relative to the total mass of fission products present in the blanket becomes crucial as shown in Fig. 6.

Four different scenarios were considered with the following FP removal options: 100%, 50%, 20% and 0%, respectively. 0% corresponds to the case when all fission products are left in the molten salt flibe, while in the case of 100% - all fission products are removed. In the first case, k_{scr} is rather stable during the irradiation time. In this case we conclude that at least 50% of the fission products have to be taken away from the flibe blanket. When only 20% of fission products are removed, due to the accumulation of fission products, k_{scr} decreases from 0.81 to 0.76, and from 0.81 to 0.65 when all the fission products are left. This would result in a corresponding increase of the external neutron source to keep the reactor fission power constant.

The fission product (FP) influence was estimated by analyzing the creation/destruction and removal ratios of the stable fission product ^{132}Xe. It was found that ^{132}Xe reaches equilibrium at different times and at different quantities depending on the fission product removal rate. Rather similar situation will be with other stable or long-lived and with similar capture cross sections sections fission products, the equilibrium may be reached much faster and even without removal.

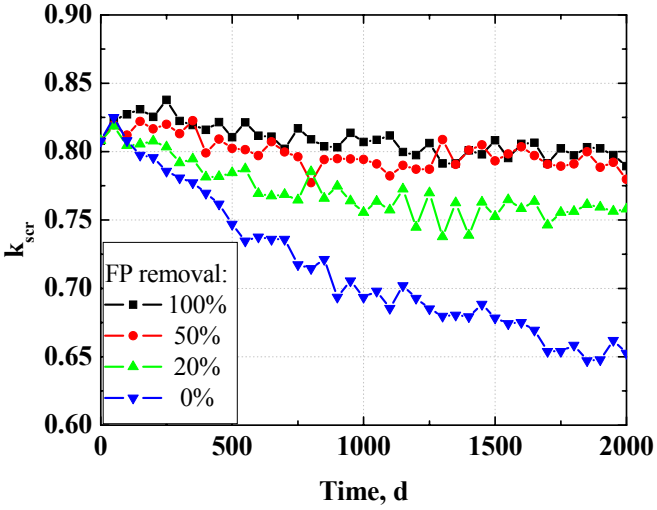


Fig. 6. k_{scr} as a function of different percentage of fission products removed from the flibe.

Detailed burnup calculations show that equilibrium conditions of the fuel concentration in the flibe transmutation blanket could be achieved provided that fission products were removed at least in part and fresh spent fuel TRU was added during burnup on-line.

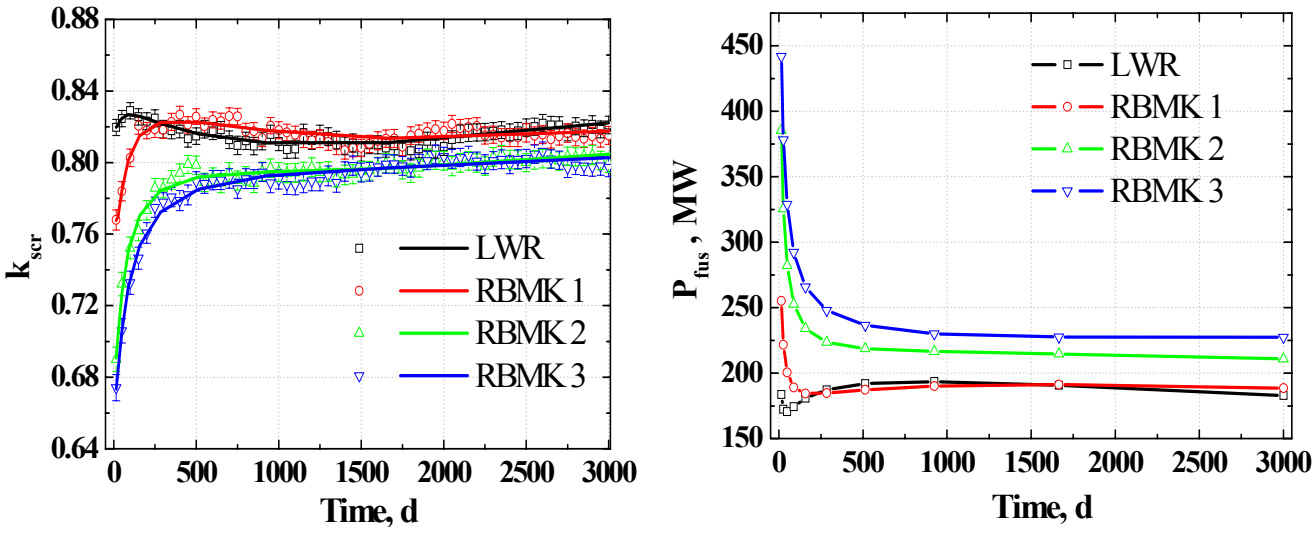
In **subchapter 3.4** the possibilities to incinerate the actinides separated from different power plants spent nuclear fuel (LWR and RBMK) in the fusion-fission hybrid system were investigated. The hybrid system transmutation blanket k_{scr}, $\Phi_n(E)$, equilibrium conditions and tritium breeding ratio were calculated for each transuranium case. The comparative analysis of LWR and RBMK SNF transuranium incineration rate was performed.

The considered TRU isotopic compositions in the molten salt are presented in Table 3. "LWR" represents 30 MWd/kg burnup LWR (PWR) reactor SNF transuranium vector after 5 years cooling time. RBMK-1500 SNF composition strongly depends on the initial RBMK reactor fuel enrichment and burnup rate [A1]. Three representative RBMK TRU vectors after 50 years cooling time were evaluated using SCALE4.4a code package: "RBMK 1" – 2% ^{235}U enrichment, 14 MWd/kg; "RBMK 2" – 2.4% ^{235}U enrichment, 22 MWd/kg; "RBMK 3" – 2.6% ^{235}U enrichment, 26 MWd/kg.

Initial conditions of the hybrid system were the same as described in 3.3 for all TRU isotopic vectors considered: P_{therm} – 3 GW, initial TRU mass 3.04 t, the continuous TRU and ^{6}Li feeding and continuous FP removal are performed. In Table 3 it is clearly seen that ^{240}Pu part is considerably larger in all RBMK TRU composition cases compared with LWR. The smaller k_{scr} values are obtained in RBMK TRU cases mainly due to a strong neutron absorption by ^{240}Pu and relatively smaller fissile isotope concentration compared with the LWR TRU case (Fig. 7(a)). Correspondingly larger fusion power is needed at the beginning of irradiation as it is presented in Fig. 7(b) to sustain 3 GW thermal power of the system.

Table 3. Initial TRU compositions (%) in the molten salt blanket for different SNF cases.

Isotope	LWR	RBMK 1	RBMK 2	RBMK 3
^{237}Np, %	**3.78**	**2.07**	**3.32**	**3.82**
^{238}Pu, %	1.81	0.19	0.51	0.67
^{239}Pu, %	51.64	61.18	49.62	46.08
^{240}Pu, %	24.61	27.56	31.09	31.86
^{241}Pu, %	8.22	0.64	0.94	0.98
^{242}Pu, %	4.61	1.92	4.70	6.06
Total Pu, %	**90.89**	**91.49**	**86.86**	**85.64**
^{241}Am, %	4.21	6.32	9.30	9.74
^{243}Am, %	0.83	0.12	0.51	0.78
Total Am, %	**5.04**	**6.44**	**9.81**	**10.52**
^{244}Cm, %	**0.19**	**0.005**	**0.01**	**0.02**



(a) (b)

Fig. 7. (a) k_{scr} as a function of irradiation time in the molten salt blanket for different TRU cases, (k_{scr} calculated with <1σ >= 0.004). (b) Corresponding P_{fus} is needed to compensate neutron balance and sustain 3 GW thermal power for different TRU cases.

Table 4. Actinides concentrations (%) at the equilibrium stages for different TRU cases.

	LVR	**RBMK 1**	**RBMK 2**	**RBMK 3**
^{237}Np, %	**1.6**	**0.8**	**1.3**	**1.4**
^{238}Pu, %	6.5	4.5	6.6	7.1
^{239}Pu, %	22.6	25.1	20.1	18.5
^{240}Pu, %	30.9	34.9	32.9	31.8
^{241}Pu, %	15.2	14.9	14.3	13.9
^{242}Pu, %	9.8	7.7	9.4	10.4
Total Pu, %	**85.0**	**87.1**	**83.3**	**81.7**
^{241}Am, %	1.5	1.9	2.6	2.6
^{242m}Am, %	0.3	0.4	0.5	0.5
^{243}Am, %	1.7	1.4	1.6	1.8
Total Am, %	**3.5**	**3.7**	**4.7**	**4.9**
^{242}Cm, %	0.7	0.9	1.2	1.2
^{243}Cm, %	0.2	0.3	0.5	0.5
^{244}Cm, %	6.0	4.7	5.9	6.7
^{245}Cm, %	2.6	2.1	2.6	3.0
^{246}Cm, %	0.4	0.4	0.5	0.6
Total Cm, %	**9.9**	**8.4**	**10.7**	**12.0**

By comparing LWR and RBMK transuranium transmutation cases in terms of the main performance parameters of the hybrid system, a number of important advantages was obtained in favour of the LWR isotopic composition: the k_{scr} (0.82) and P_{fus} (180MW) are almost stable comparing the beginning and equilibrium stages, the equilibrium is reached after ~3 years of the system operation, and TBR is sufficient to supply tritium for the fusion device (TBR=1.25). In the case of RBMK 1, the equilibrium is reached after ~3.3 years, k_{scr} (0.77) is less at the beginning but later it approaches to LWR TRU case. In equilibrium k_{scr}=0.814, P_{fus}=190MW, TBR=1.25. In the case of RBMK 2 and RBMK 3 transmutation in the molten salt k_{scr} at the beginning of irradiation is 0.69 and 0.67, respectively, and 370 MW and 440 MW fusion power

is needed for corresponding cases, the tritium breeding rate is not sufficient for such fusion power (TBR<1 0.73 and 0.67). When equilibrium for main isotopes concentrations is reached in both cases the situation is improved: k_{scr} – 0.795 and 0.797, P_{fus} – 220 MW and 230 MW, TBR 1.11 and 1.04 for corresponding cases.

The neutron spectrum in the molten salt blanket is independent of the transuranium isotopic composition and is stable during the irradiation time. Most of the neutrons are epithermal (about 80%). In all TRU isotopic vector cases the hybrid system incinerates about 1.13 tons per year of TRU with 3 MW thermal power output. The initial TRU vector influences the composition of actinides at the equilibrium stage. The actinides isotopic composition at equilibrium stage is presented in Table 4. The smallest amount of Cm is accumulated in RBMK1 case. ^{237}Np, ^{239}Pu, ^{241}Am are incinerated most effectively – about 90% of these isotopes are transmuted. The transmuted part of ^{240}Pu is 76%, ^{241}Pu ~60% and ^{243}Am ~40%, the lowest transmutation rate was obtained for ^{44}Cm. These results are valid for all TRU cases considered.

TRU radiotoxicity (inhalation) before and after transmutation in the hybrid system (including 10 years cooling time) was investigated in detail for LWR and RBMK 2 cases. The radiotoxicity of RBMK 2 at the beginning was considerably smaller comparing with LWR. This was determined by the smaller quantity of ^{241}Pu ($T_{1/2}$=14.2 y) and ^{238}Pu ($T_{1/2}$=87.7 y) due to longer RBMK SNF cooling time (50 years) in RBMK 2 case. At the end of transmutation the isotopic composition in both cases becomes more or less similar and same does the radiotoxicity. The final waste radiotoxicity decreases considerably (4-6 times) in the period from 1 thousand to 100 thousand years. In the molten salt transmutation blanket a large amount of Cm is accumulated, but contribution of this isotope to the total waste radiotoxicity is minor.

In **subchapter 3.5** the minor actinides transmutation scenarios in the fusion-fission hybrid system molten salt blanket were investigated. To optimize the neutronic characteristics of the blanket and to improve the minor actinides transmutation process, two different molten salt compositions were tested. The first molten salt blanket (2LiF-BeF_2-$(HN)F_4$) – the molten salt used in previous calculations – consists of: 28.57% - ^{6}Li+^{7}Li, 14.29% - Be, 57.13% - F (F – molten salt), the second one is the modified molten salt blanket, where half of the blanket volume is occupied by Be: 14.29% - ^{6}Li+^{7}Li, 57.14% - Be, 28.57% - F (Be – molten salt). The starting transuranium composition in the molten salt was Pu and minor actinides separated from LWR spent nuclear fuel of 30 MWd/kg burnup, the subsequent feeding - only minor actinides from the same spent fuel. Initial TRU mass is 3.04 tons in F molten salt blanket and 1.52 tons in Be molten salt blanket. The fission products have been removed continuously during the irradiation.

Moreover the influence of ^{6}Li enrichment in Li on both molten salt blanket characteristics was tested. ^{6}Li ($^{6}Li(n,\alpha)T$) is very important to the fusion device, but at the same time has strong influence on k_{eff} and k_{scr} of the transmutation blanket. TBR=1 means that every source neutron produces 1 tritium nucleus for fusion reaction. Small part of tritium is produced in the molten salt blanket via $^{7}Li(n,n'\alpha)T$ reaction. The hybrid system performance parameters with F molten salt blanket containing 0% and 0.6% of ^{6}Li in lithium with different ^{6}Li feeding modes, and with Be molten salt blanket containing 0%, 0.2% and 0.6% of ^{6}Li in lithium with different ^{6}Li feeding modes were analyzed. k_{scr} behavior in the different molten salt blanket cases with different ^{6}Li treating options is presented in Fig. 9 (a) and (b). The best system performance results in terms of k_{scr} and accordingly fusion power stability were obtained in case of F molten salt blanket with 0.6% ^{6}Li without ^{6}Li feeding (k_{scr} max fluctuation during irradiation [0.74-0.81], P_{fus} [188-301 MW], at equilibrium stage k_{scr}=0.79±0.004, P_{fus}~230 MW) and in case of Be- molten salt blanket with 0.2% ^{6}Li without ^{6}Li feeding (k_{scr} max fluctuation [0.83-0.91], P_{fus} [100-210 MW], at equilibrium stage k_{scr} 0.86±0.003, P_{fus}~164 MW).

The k_{scr} behavior for these cases can be explained by fissile isotope quantity and interaction with neutron properties. In Fig. 8 averaged macroscopic fission cross sections are presented for ^{238}Pu, ^{239}Pu,^{241}Pu isotopes, MA and all TRU in (a) F molten salt blanket and (b) Be molten salt

blanket. At the beginning stage ^{239}Pu is a dominant fissile isotope in F molten salt blanket. Due to ^{239}Pu incineration k_{scr} is decreasing up to the moment when ^{241}Pu and some other fissile nuclei start dominating in the blanket (Fig. 8(a)). In the Be molten salt blanket the sharp variation in k_{scr} at the early stage of operation is explained by fast plutonium isotope mass equilibration process. At the beginning k_{scr} increases due to ^{240}Pu conversion to ^{241}Pu, and decreases later on due to intensive plutonium isotopes burning (^{239}Pu, ^{240}Pu ^{241}Pu). Before equilibrium stage k_{scr} increases again due to ^{238}Pu accumulation from different chains (^{237}Np(n,γ)→^{238}Np(β-)→^{238}Pu, and (^{241}Am(n,γ)→^{242}Am(β-)→^{242}Cm) ^{242}Cm(α)→^{238}Pu. ^{238}Pu converts to ^{239}Pu by (n,γ) reaction and in such a way ^{239}Pu disappearance is compensated.

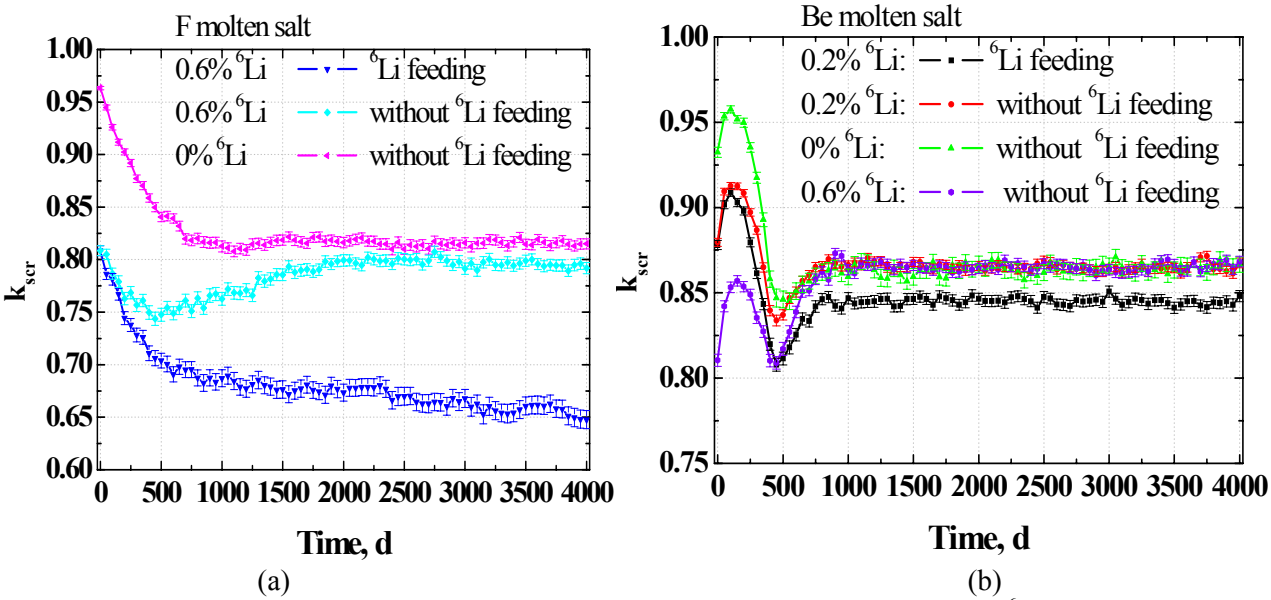


Fig. 8. The behavior of k_{scr} (a) in F molten salt and (b) in Be molten salt with different ^{6}Li quantity and feeding modes.

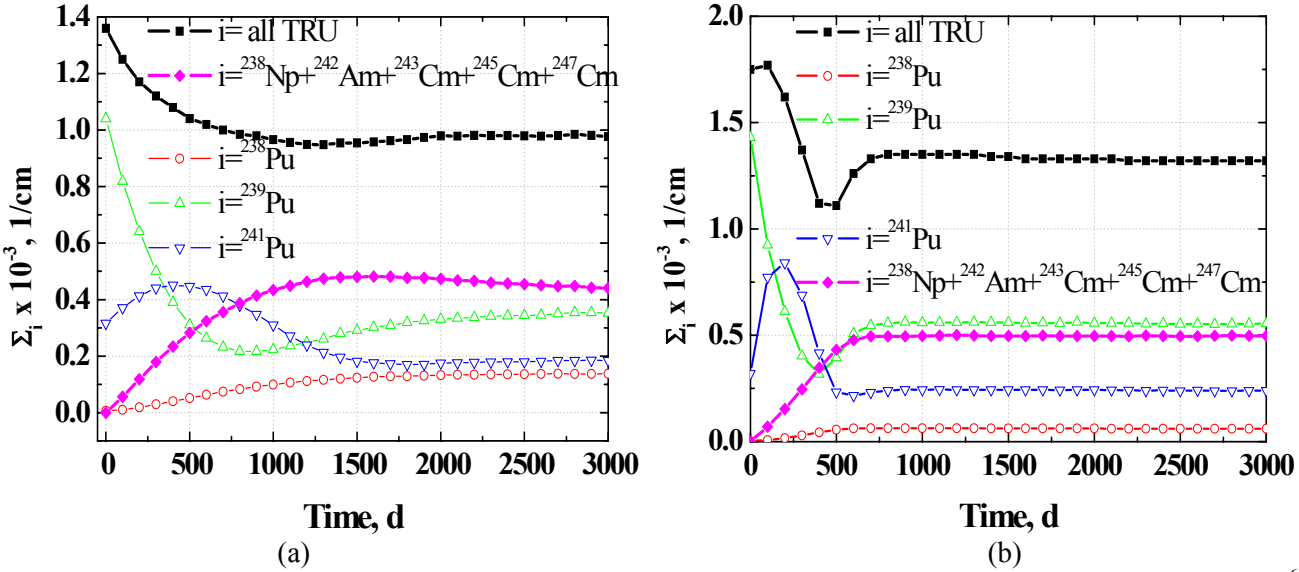


Fig. 9. Averaged macroscopic fission cross sections for fissile isotopes (a) in F molten salt with 0.6% ^{6}Li without Li feeding and (b) in Be molten 0.2% ^{6}Li without Li feeding.

In both molten salt cases the same incineration rate (1.1 tons/year) of actinides was calculated, but in the case of Be molten salt the equilibrium stage is reached after 2 years and in F molten salt case – after 5 years.

The reason for different plutonium incineration in F and Be molten salt blankets is different neutron spectrum. The neutron spectra for both transmutation blankets are presented in Fig. 9.

The thermal neutron contribution in the neutron spectrum of Be molten salt medium is ~10 times larger at the beginning of irradiation and at equilibrium stage as compared with F molten salt.

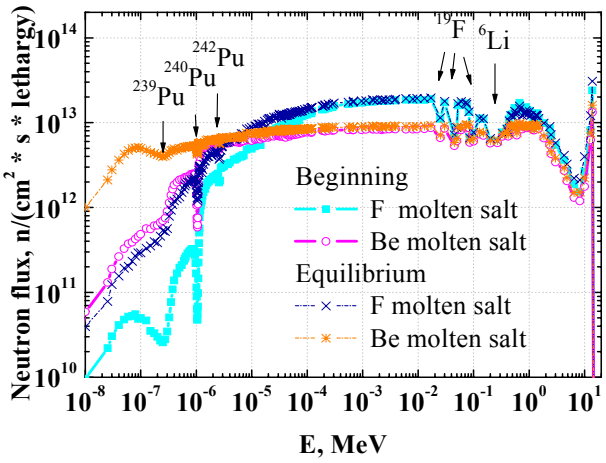


Fig. 10. Averaged neutron energy spectra at the beginning of irradiation and at equilibrium stage for F and Be molten salt blankets.

By comparing two molten salt transmutation media, better hybrid system performance parameters were obtained in Be molten salt blanket case: a lower fusion power to sustain 3 GW thermal power is needed, equilibrium is reached faster, the total mass of transuranium in the blanket is smaller, so the criticality safety and radiation protection concerns are of somewhat smaller scale. In addition, the transuranium are incinerated more effectively.

Be molten salt blanket with different fission products removal options was analyzed as well. It was pointed out that in order to stabilize the hybrid system performance, at least 30% of fission products should be removed continuously from the blanket.

Chapter IV. HIGH TEMPERATURE HELIUM COOLED REACTOR MODELLING

In **subchapter 4.1** the overview of GT-MHR project and construction features is presented. GT-MHR is an advanced nuclear power plant that in principle could provide an increased safety, high thermal efficiency and deep burnup rates [3,6,7]. The GT-MHR features are: the helium coolant, which is inert and remains in a single phase under all conditions; the graphite core, which provides high heat capacity, slow thermal response, and structural stability at high temperatures; the refractory coated particle fuel, which allows high burnup, retains fission products during operation at high temperatures and retains their integrity in a repository environment for hundreds of thousands of years. GT-MHR can be used to burn all types of fuel and offers significant advantages in accomplishing transmutation of plutonium isotopes and high destruction of ^{239}Pu in particular [7]. It utilizes natural erbium as a burnable poison with the capture cross section having a resonance at a neutron energy that ensures a strong negative temperature coefficient of reactivity. The lack of interaction of neutrons with coolant (helium gas) ensures that temperature feedback of fuel and graphite is the only significant contributor to the power coefficient. A gas-cooled high temperature reactor or a separate irradiation zone in the centre of GT-MHR assembly, coupled to an accelerator, could also provide a fast neutron environment due to the same reason - the helium coolant is essentially transparent to neutrons and does not change neutron energies. Since fertile nuclei (^{240}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{242}Cm, ^{244}Cm) have significant fission cross sections in a fast neutron energy spectrum, one could consider an additional fast stage, following a thermal stage, in order to eliminate the remaining actinides [3].

In **4.2 subchapter** a critical GT-MHR, dedicated to burn Pu isotopes in the once-through fuel cycle was studied by comparing performance characteristics such as k_{eff}, length of the fuel cycle, burnup rates, discharged fuel composition, one-group cross sections, etc. with geometry representation as a parameter, namely homogeneous HTR1 (see Fig. 11), single-heterogeneous HTR2 (Fig. 12) and double-heterogeneous HTR3 (Fig. 13). This work was performed in order to

examine and validate/reject a simplified GT-MHR geometry description, which would be preferred due to less time-consuming calculations and 2D (deterministic modelling possibility). For the first time an exact and finite GT-MHR core (HTR3), containing 310μm ceramic-coated fuel and burnable poison particles, was modelled using the Monte Carlo method. The MCNP code was employed for this purpose. The main parameters used for modelling of three geometry designs are summarized in Table 5. The case of a critical GT-MHR loaded with Pu isotopes from military applications (94.0 % of ^{239}Pu, 5.4 % of ^{240}Pu and 0.6 % of ^{241}Pu) was analyzed. In HTR3 case the core contains 1.87×10^{10} fuel particles, which corresponds to 701 kg of PuO_2, and 3.4×10^9 of natural Er_2O_3 (mass – 293.6 kg) particles.

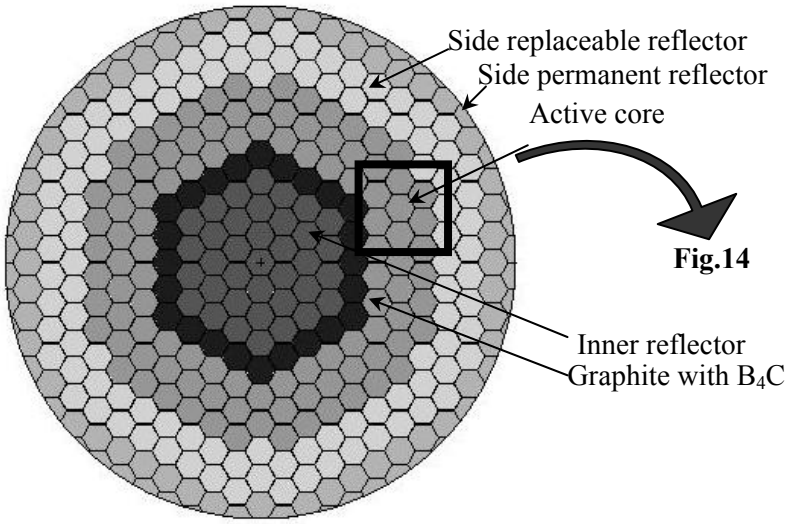


Table 5. Basic GT-MHR parameters [7].

Power, MW_{th}	600
Active core size: - height, cm - area, m^2	 800.0 11.5
Active core volume, m^3:	91.9
Graphite mass in reactor, t:	616.3
Averaged temperature, ^oC: - active core - inner reflector - side, top and bottom reflectors	 800 730 500

Fig. 11. A cross section of the GT-MHR homogeneous core with hexagonal structure (HTR1). All material compositions are distributed homogeneously over corresponding zones and characterized by specific material densities. An active core consists of 102 hexagonal prism columns located around the inner reflector (37 columns) in addition to a separate zone with B_4C (24 columns).

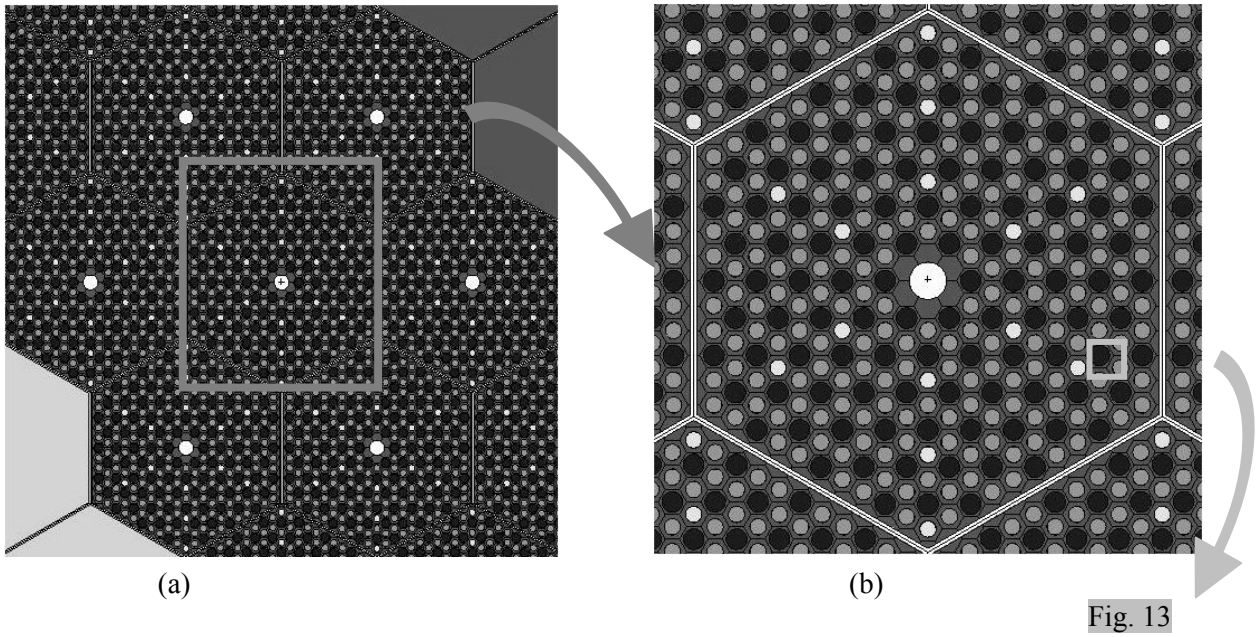


Fig. 12. Fragments of single-heterogeneous GT-MHR (HTR2): a) an active core structure: three rings of hexagonal fuel columns; b) magnified view of a separate fuel assembly. Fuel and erbium materials are homogeneously placed in the silicon – graphite matrix in the form of homogenized compacts. Fuel compacts are presented in small grey circles, burnable poison compacts – in light grey. Bigger diameter holes stand for He channels, while the rest material represents the graphite matrix.

In this work we used the same MCNP code to obtain k_{eff} - eigenvalues and neutron fluxes. As soon as k_{eff} is less than 1 by 2-3σ the length of the fuel cycle is determined in the case of a critical system.

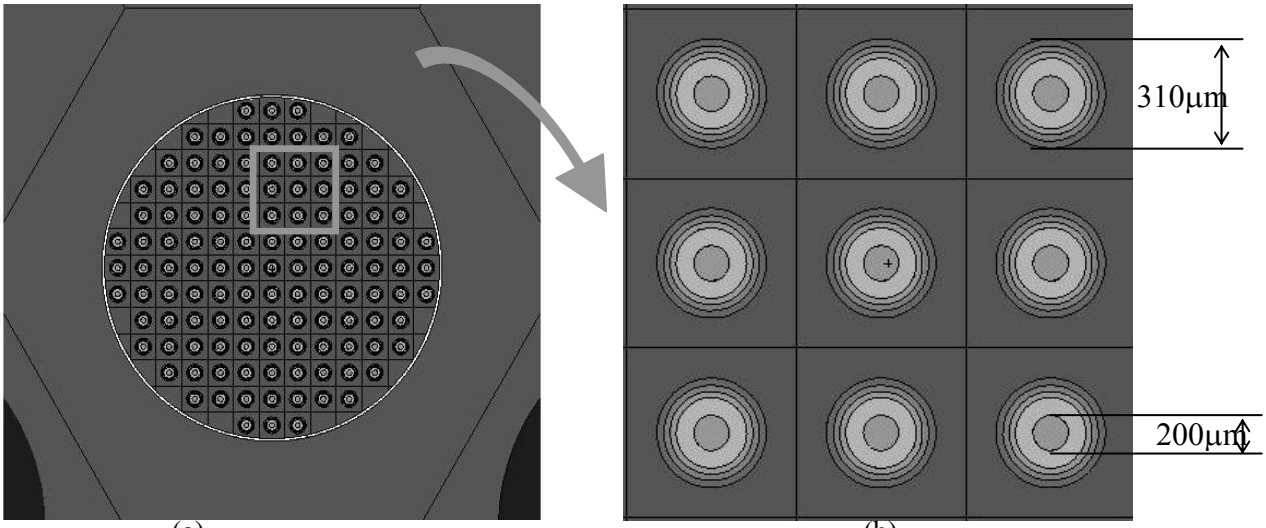


Fig. 13. Fragments of double-heterogeneous GT-MHR (HTR3): a) fuel element (compact) cross section with coated fuel particles; b) magnified view of coated fuel particles: spherical kernels of PuO_{2-x} are surrounded by protective coatings made of pyrocarbon and SiC layers (TRISO coating). The same structure is valid for particles containing burnable poison - natural Er_2O_3.

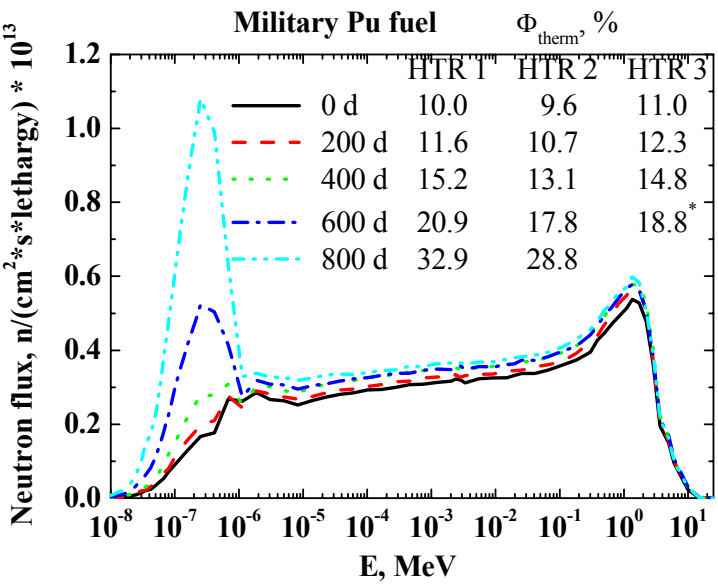


Fig. 14. Typical change of the average energy spectra of neutrons in the active core of GT-MHR for different fuel burn-up (expressed in full power days - fpd). The thermal neutron contribution is presented in the legend for each geometry configuration.

* Note that the fuel cycle of HTR3 ends after 550 days.

A typical feature of GT-MHR is that its neutron spectrum changes considerably during the burnup. The neutron spectrum evolution for military Pu fuel poisoned with natural Er is shown in Fig. 14. The observed increase of the thermal flux from 10% to 30% for different geometry cases is due to the loss of ^{239}Pu and ^{240}Pu in addition to the loss of the burnable poison during the operation. We note also that at the constant reactor power typical averaged GT-MHR neutron fluxes in the active core may increase typically by 50-100 %, i.e. from ~1×10^{14} n/(cm^2 s) to ~2×10^{14} n/(cm^2 s) at the beginning and at the end of the fuel cycle, respectively.

A significant difference was found in k_{eff} between HTR1, HTR2 and HTR3 both at the beginning of the fuel cycle and in its evolution as a function of burnup (Fig. 15). In addition, HTR3 fuel cycle is much shorter compared to the other two cases. A different behaviour of k_{eff} is explained by different modelling of burnable poison (homogeneous versus heterogeneous distribution) and also by a significant self-shielding effect in micro particles (single-heterogeneous versus double-heterogeneous geometry), see Table 6 and Fig. 16(b) for details.

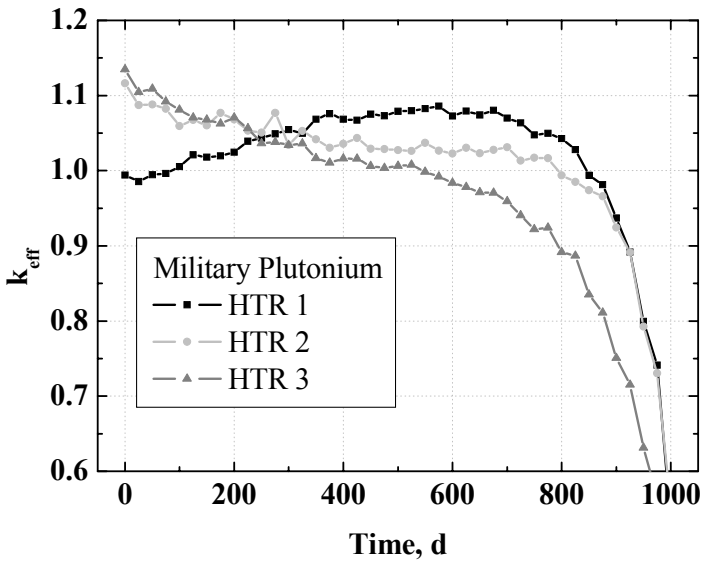


Fig. 15. A behaviour of k_{eff} for different GT-MHR modelling cases with military plutonium fuel.

Consequently, the burnup of erbium and changes in isotopic fuel composition (e.g., ^{240}Pu and ^{241}Pu) are rather different during the fuel cycle (Fig. 17). The spatial and energy shielding of the neutron flux even in such small particles cannot be neglected for isotopes with high resonance cross sections as ^{240}Pu and ^{167}Er. We have shown that it is not possible to obtain reliable isotopic composition of the discharged fuel by simplifying the reactor model. An exact modelling of the fuel particles in the compacts (HTR3), in comparison with single-heterogeneous (HTR2) and homogeneous distributions (HTR1), had minor influence on the neutron spectrum in the corresponding cells than in the case of burnable poison. Indeed, in 200 μm fuel kernels the self-shielding effect for the ^{239}Pu ~0,3eV resonance cross section is negligible (Table 6 for ^{239}Pu σ_f and Fig 18(a) for σ_c).

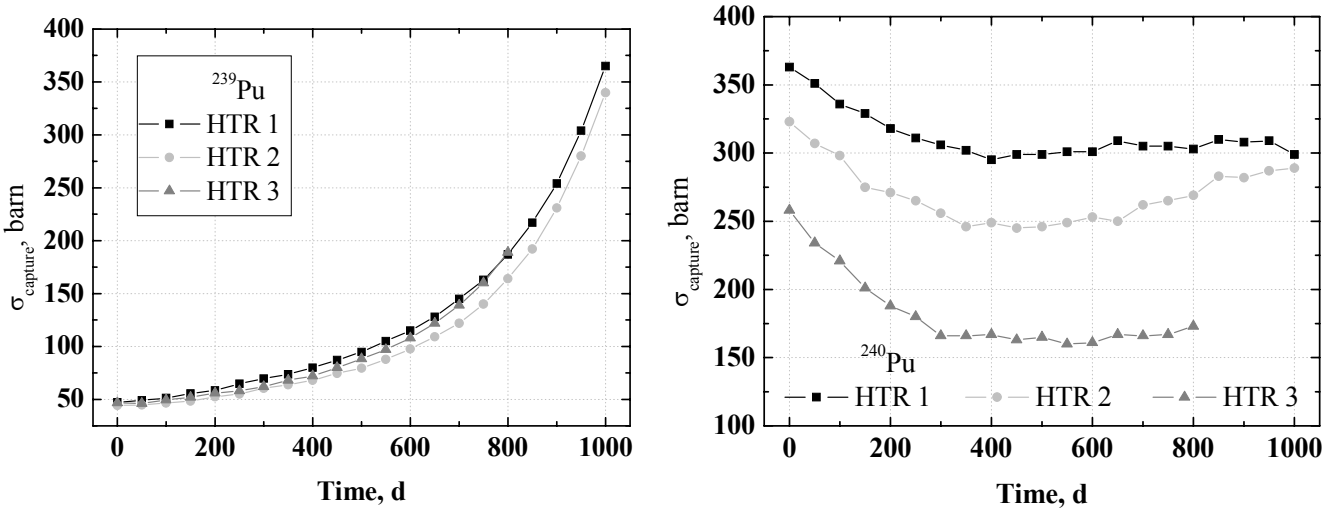


Fig. 16. (a) ^{239}Pu capture cross sections for different GT-MHR geometry configurations, (b) same as (a) but for ^{240}Pu.

Table 6. Effective capture and fission cross sections for different GT-MHR geometry configurations

Isotope	^{167}Er σ_c, barns			^{239}Pu σ_f, barns		
Time, d	HTR1	HTR2	HTR3	HTR1	HTR2	HTR3
0	252	145	164	77.7	72.6	78.3
200	284	172	201	95.6	85.6	92.7
400	338	227	258	131	111	119
600	416	320	368	188	159	178
800	563	493	578	305	267	308

The microscopic ^{239}Pu capture cross sections are similar for all three geometry configurations as a function of time. That is why the corresponding evolution of ^{239}Pu is very similar as shown explicitly in Fig. 17. Consequently, the evolution of ^{239}Pu at a constant reactor power and comparable neutron fluence is very similar for all three geometry configurations. Therefore, the destruction rate of ^{239}Pu could also be successfully estimated using a simplified GT-MHR model as HTR1 or HTR2.

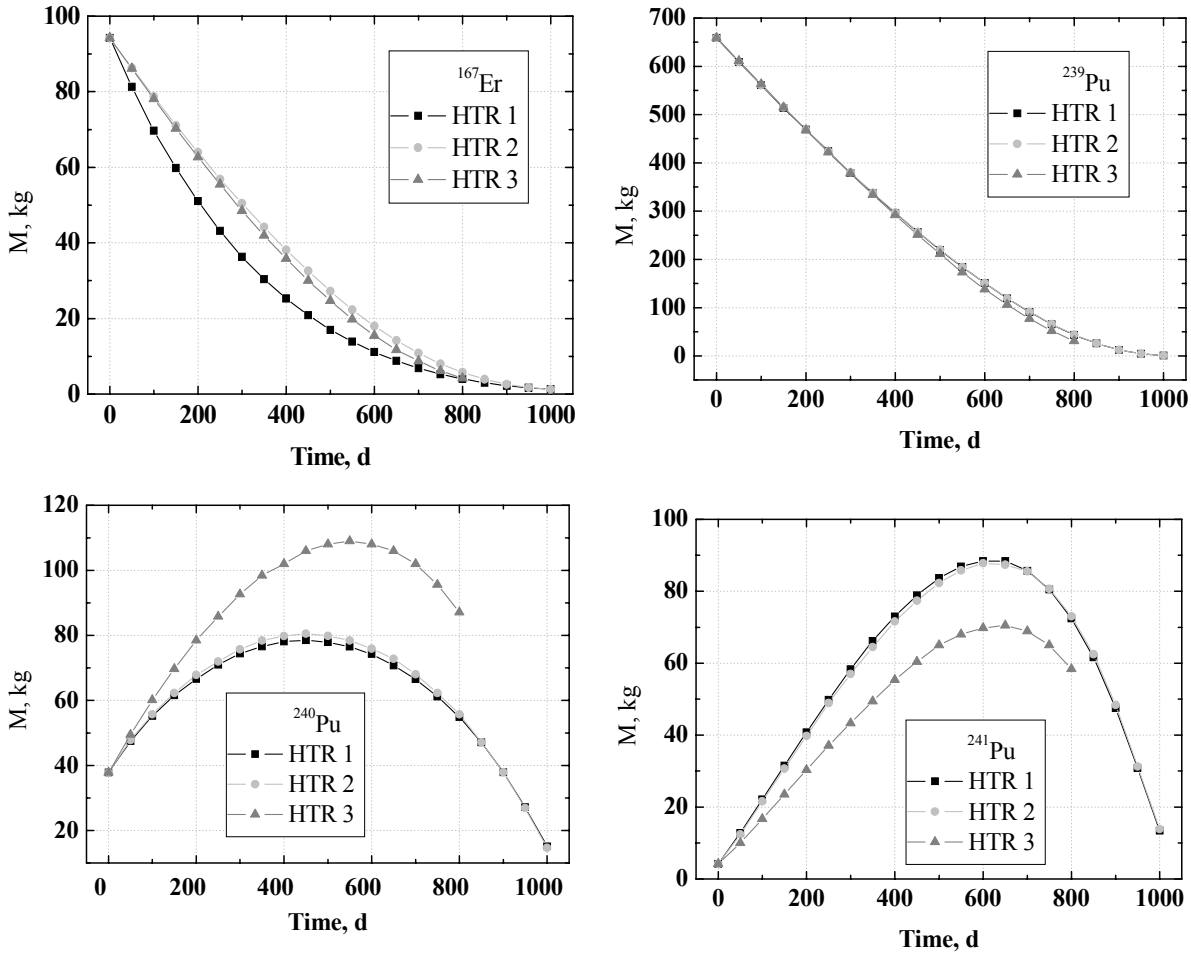


Fig. 17. Evolution of mass of ^{167}Er and Pu isotopes for different GT-MHR geometry configurations.

In **subchapter 4.3** GT-MHR reactor operating with different fuel isotopic composition: plutonium from military applications, plutonium extracted from LWR and RBMK spent nuclear fuel was analyzed. The main goal was to investigate the influence of initial plutonium isotopic composition on the GT-MHR performance parameters in the once-through cycle using double-heterogeneous reactor core geometry within the Monte Carlo approach. The possibility to transmute plutonium separated from RBMK-1500 SNF in high temperature helium cooled reactor was investigated for the first time. The initial fuel and burnable poison compositions are presented in Table 7. "Military Pu" stands for plutonium from military applications, "LWR Pu" – plutonium from 42 MWd/kg burnup LWR SNF. Since RBMK-1500 SNF isotopic composition strongly depends on the initial ^{235}U enrichment fuel and its final burnup for RBMK plutonium several cases were tested: "RBMK Pu1" – 2% ^{235}U initial enrichment, 14 MWd/kg burnup plutonium composition, "RBMK Pu2" – 2% ^{235}U initial enrichment, 20MWd/kg burnup; "RBMK Pu3" – 2.4% ^{235}U initial enrichment, 22 MWd/kg burnup and "RBMK Pu4" – 2.6% ^{235}U initial enrichment, 26 MWd/kg burnup.

Table 7. Different GT-MHR fuel and erbium load at the beginning of the fuel cycle considered in this work. *Note that in RBMK Pu1 case GT-MHR performance parameters were investigated two times – with burnable poison erbium (Fig. 18), and without erbium (Fig. 19).

Fuel	Military Pu	LWR Pu	RBMK Pu1*	RBMK Pu2	RBMK Pu3	RBMK Pu4
Isotopic Pu composition, kg						
^{238}Pu	--	32.4	2.5	6.0	7.0	9.5
^{239}Pu	659.0	661.0	802.1	673.3	685.5	652.2
^{240}Pu	37.8	277.0	361.8	443.1	429.6	439.0
^{241}Pu	4.2	142.0	8.4	12.4	13.0	14.1
^{242}Pu	--	87.6	25.2	65.2	64.9	85.2
Total plutonium, kg :	701.0	1200.0	1200.0	1200.0	1200.0	1200.0
Isotopic Er_2O_3 composition, kg:						
^{166}Er	132.0	16.8	18.0	--	--	--
^{167}Er	94.2	11.4	12.2	--	--	--
^{170}Er	61.4	7.5	8.0	--	--	--
Total erbium, kg :	293.6	35.7	38.2	--	--	--

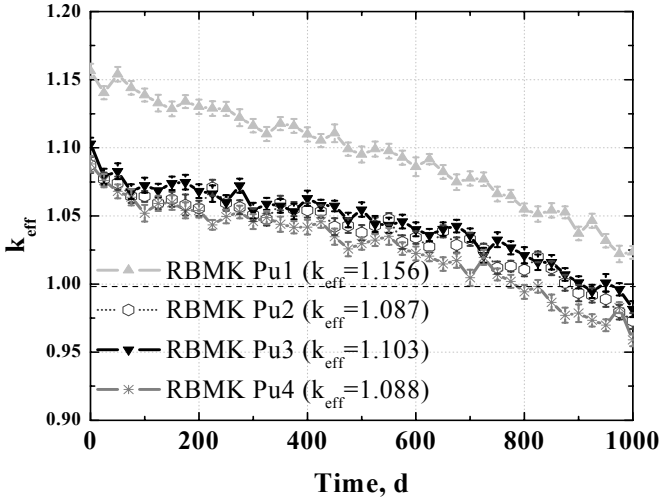


Fig. 18. A behaviour of k_{eff} for different RBMK plutonium cases without burnable poison in GT-MHR.

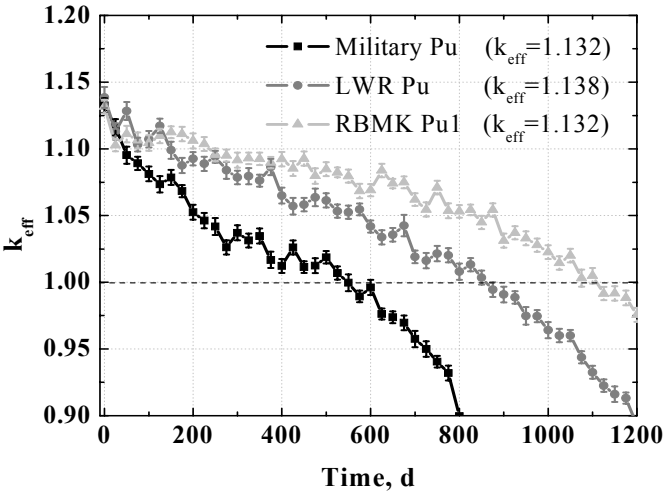


Fig. 19. A behaviour of k_{eff} for Military, LWR and 14MWd/kg burn-up RBMK plutonium with burnable poison in GT-MHR.

The first intention was to check the RBMK plutonium as GT-MHR fuel feasibility. For this purpose GT-MHR performance parameters were analyzed for four RBMK fuel cases without burnable poison. The k_{eff} eigenvalues as a function of time for RBMK Pu1, Pu2, Pu3 and Pu4 compositions are presented in Fig. 18. The most attractive RBMK plutonium composition is that of 14MWd/kg burnup (up to now there is about 15% of all INPP SNF), but the use of other burnup RBMK SNF plutonium in GT-MHR is also possible. In RBMK Pu3 case 90% of ^{239}Pu transmutation rate was obtained. RBMK isotopic composition fuel in GT-MHR due to a higher amount of ^{240}Pu can support rather long operation time thanks to the formation of ^{241}Pu which compensates the decrease of ^{239}Pu. In practice, one would need 2 GT-MHR's units with 10 fuel loads every 3 years (assuming that the reactor life is ~30 years) to burn all INPP SNF plutonium. The outcome of the once-through GT-MHR fuel cycle of RBMK-1500 plutonium transmutation would be ~127 TWh of the electric power.

Since RBMK Pu2, Pu3 and Pu4 cases do not differ very much from the point of view of isotopic composition, for further comparative analysis only RBMK Pu3 was employed.

In order to obtain a relative fuel cycle length for different GT-MHR fuel cases, the appropriate quantity of burnable poison was added to achieve the same starting k_{eff}. Unfortunately, RBMK Pu3 k_{eff} was too low at the beginning and it was impossible to add burnable poison, but RBMK Pu1 composition with burnable poison was compared with the LWR Pu and military Pu as it is shown in Fig. 19. A significant difference was found in k_{eff} evolution as a function of burnup, in length of the fuel cycle and in the transmutation efficiency between military, LWR, and RBMK plutonium cases. Starting with the same k_{eff} value, military plutonium k_{eff} is decreasing continuously and already after 550 days the fuel cycle end is reached with 73% of ^{239}Pu burnup. LWR fuel cycle continues 850 days and 82% of ^{239}Pu burnup is obtained. GT-MHR performance with 14MWd/kg RBMK plutonium is much better: longer fuel cycle (1100 days), very efficient ^{239}Pu burning (93%) and relatively small accumulation of minor actinides at the end of the fuel cycle.

Thermal neutron contribution to the neutron flux spectrum at the beginning of the fuel cycle inside fuel particles is quite similar, i.e. 9-10% for all cases considered. The observed increase of the thermal flux at the end of the fuel cycle is up to 22%.

Table 8. Reactivity coefficient ($\Delta k_{eff}/\Delta T$) dependence on fuel, graphite and fuel / graphite temperature for different GT-MHR fuel cases (Δk_{eff} expressed in pcm, and ΔT in K)

Fuel	Pu temperature effect: ΔT_{Pu}=300 K ($T_{Graphite}$=1200 K)	Graphite temperature effect: ΔT_G=400 (T_{Pu}=1500 K)	Graphite & Pu temperature effect ΔT_{Pu}=300 K ΔT_G=400 K
RBMK Pu1	-3.6±0.9	-9.2±0.7	-12.8±1.1
RBMK Pu3	-2.4±0.9	-9.1±0.7	-11.5±1.1
LWR Pu	-2.1±0.9	-8.5±0.7	-10.6±1.1
Military Pu	0.9±0.9	-9.6±0.7	-8.7±1.1

It seems that the TRISO fuel kernel is of right dimensions (200 μm diameter) to provide a negative temperature coefficient by enhanced resonance neutron absorption of ^{240}Pu nuclei in all cases considered. The values are presented in Table 8. Both the moderator and fuel temperature increase were taken into account for these estimates. Even fuel load without burnable poison (RBMK Pu3 case) does not affect the GT-MHR safety as long as its temperature coefficient is concerned – it remains negative in all cases.

Chapter V. NEUTRON CROSS SECTIONS

In **chapter 5** the nuclear data problem is analyzed. The most often used nuclear data libraries (ENDF/B, JEF, JENDL) present large discrepancies for a number of actinides, wich play a dominant role in transmutation cores as Am, Cm and rare Pu isotopes.

In **subchapter 5.1** the influence of different sets of data libraries on the GT-MHR performance parameters when modelling codes and conditions are the same. The simulations of main performance parameters of GT- MHR (k_{eff} eigenvalues, the length of the fuel cycle, neutronic characteristics and the evolution of fuel composition) with LWR and military plutonium fuel were performed. The results have shown that in general the performance of GT-MHR is not considerably influenced by the choice of data libraries employed. Nevertheless a number of major differences among ENDF/B-VI [22], JEF 2.2 [24] and JENDL 3.2 [23] data files are identified and quantified in terms of averaged one group cross sections both for military and civil plutonium based fuel cycles (see table 9). The considerable discrepancies were found for Am and Cm isotopes. These results should provide the first guidelines for the experimental programmes where measurements of fission and capture cross sections are planned.

Table 9. Relative deviations of one group fission cross sections in the case of ENDF/B-IV, JEF 2.2 and JENDL 3.2 data files obtained by simulating GT-MHR LWR and Military plutonium fuel cycles.
The folowing notation is used: ENDF/JEF=(σ(ENDF)-σ(JEF))/σ(JEF)

Isotope	LWR Pu				Military Pu			
	ENDF/JEF %	ENDF/JENDL %	JEF/JENDL %	Error	ENDF/JEF %	ENDF/JENDL %	JEF/JENDL %	Error
^{238}U	4.8	-1.2	-5.7	0	2.1	-3.8	-5.8	0
^{237}Np	2.8	-13.7	-16.1	1	3.8	-15.2	-18.3	1
^{238}Pu	-4.6	-6.5	-2.0	0	-5.6	-6.0	-0.5	0
^{239}Pu	-4.4	-2.8	1.6	0	-1.4	1.9	0.3	0
^{240}Pu	-12.8	1.2	15.9	1	-13.1	1.6	17.0	1
^{241}Pu	-2.1	-1.0	1.2	0	-0.7	3.8	4.5	0
^{242}Pu	-12.0	-1.8	11.6	1	-12.9	-2.8	11.6	1
^{241}Am	-10.9	7.0	20.1	2	-11.3	4.5	17.8	1
^{242}Am	148.6	148.0	0.0	2	162.9	167.2	1.6	2
^{243}Am	26.1	2.4	-18.8	2	23.5	-0.7	-19.6	2
^{242}Cm	-66.1	-78.6	-37.0	2	-65.9	-78.4	-36.6	2
^{243}Cm	16.8	25.6	7.5	2	14.9	23.9	7.9	2
^{244}Cm	-2.2	37.2	40.3	2	-3.0	45.9	50.4	2
^{245}Cm	7.2	9.1	1.8	0	6.3	14.9	8.2	1
^{246}Cm	6.9	4.2	-2.6	0	-5.2	4.8	10.6	1
^{247}Cm	-5.5	1.4	7.3	0	-5.1	19.7	26.2	2
^{248}Cm	-4.1	-18.3	-14.7	1	-5.2	-16.4	-11.8	1

In **subchapter 5.2** the Mini-Inca integral experiments at Laue-Langevin Institute (ILL) Grenoble (France) are reviewed. The first experiments were dedicated to the ^{241}Am, ^{243}Am and ^{242}Pu cross section measurements in the high intensity 6×10^{14}n/(cm^2·s) thermal neutron flux (98%). Several of low mass (~10μg) high purity actinides samples were irradiated and analyzed off-line by different techniques such as nuclear spectrometry and mass spectrometry.

The ^{241}Am total neutron capture cross section (^{241}Am σ_c=(696±48) b) confirms the possibility to incinerate this isotope in the thermal neutron flux. The branching ratio of ^{241}Am(n,γ) has been estimated for the first time (BR=0.914±0.007). The value of ^{241}Am(n,γ)^{242gs}Am=(330±50) b is comparable with the value given by ENDF/B-VI (252 b)).

The ^{242}Pu capture cross section (^{242}Pu(n,γ)^{243}Pu=22.7±1,1 b) differs slightly from the previous existing data but is rather reliable due to simple analysis thanks to a high level of neutron thermalization available at the high flux reactor of ILL.

Finally, in the case of ^{243}Am (σ_c =(82.5±3.9) b), the obtained total capture cross section is by 8% larger than ENDF/B-VI, JEF2.2, JENDL3.2 (76 b), this means 8% larger minor actinides formation in the thermal neutron flux if obtained by using the existing nuclear data libraries. ^{243}Am(n,γ)^{244gs}Am=(5.2±1.7) b is comparable with other data libraries.

CONCLUSIONS

1. The analysis of neutron processes in the fusion-fission hybrid system transmutation blanket have shown that isotopic composition of transuranium and feeding mode have no effect on actinide transmutation effectiveness, but these parameters influence equilibration of actinide concentrations in the blanket. k_{scr} stability in the transmutation blanket strongly depends on the concentration of fission products.

2. Optimization of neutronic characteristics of the fusion-fission system molten salt blanket for minor actinides incineration scenario has demonstrated that Be molten salt blanket is more favorable for transmutation.
3. For the first time an exact and finite GT-MHR core, containing microscopic ceramic-coated fuel and burnable poison particles, was modelled using the Monte Carlo method (MCNP code). It has been demonstrated that the spatial and energy shielding of the neutron flux in small fuel and erbium particles cannot be neglected for isotopes with high resonance cross sections as ^{240}Pu and ^{167}Er.
4. For the first time the possibilities of plutonium and minor actinides separated from RBMK-1500 spent nuclear fuel transmutation in the fusion-fission hybrid system and GT-MHR reactor were investigated. The detailed studies have shown that ^{237}Np, ^{239}Pu, ^{241}Am isotopes are incinerated most effectively in the fusion-fission hybrid system and the greatest burnup rates of ^{239}Pu are obtained using GT-MHR.
5. By modelling GT-MHR with different evaluated nuclear data libraries (ENDF, JEF and JENDL) the considerable discrepancies (>20%) were identified for $^{241-243}$Am and $^{242-244}$Cm isotopes in terms of averaged one group capture and fission cross sections.
6. During Mini-Inca integral experiments the following thermal neutron cross sections were obtained: ^{241}Am σ_c=(696±48) b and ^{243}Am σ_c=(82.5±3.9) b. These results have improved the existing data and confirm the possibility to incinerate Am isotopes in a thermal neutron flux. In addition, for the first time ^{241}Am branching ratio BR=0.914±0.007 was determined experimentally.

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