The Transmutation of Nuclear Waste in the Two-Zone Subcritical System Driven by High-Intensity Neutron Generator - 12098

V.O. Babenko**, V.I. Gulik*, V.M. Pavlovych*
*Institute for Nuclear Research, pr. Nauky 47, Kyiv, 03680, Ukraine
**Bogolyubov Institute for Theoretical Physics, Metrolohichna str. 14-b, Kiev, 03680, Ukraine

ABSTRACT

The main problems of transmutation of high-level radioactive waste (minor actinides and long-lived fission products) are considered in our work. The range of radioactive waste of nuclear power is analyzed. The conditions under which the transmutation of radioactive waste will be most effective are analyzed too. The modeling results of a transmutation of the main radioactive isotopes are presented and discussed. The transmutation of minor actinides and long-lived fission products are modeled in our work (minor actinides – Np-237, Am-241, Am-242, Am-243, Cm-244, Cm-245; long-lived fission products – I-129, Tc-99). The two-zone subcritical system is calculated with help of different neutron-physical codes (MCNP, Scale, Montebarn, Origen). The ENDF/B-VI nuclear data library used in above calculations.

INTRODUCTION

Contemporary nuclear power industry is a result of more than 50 years of technological innovation and development. This technology, which has become an industrially mature one over these years, is now a reliable source of electricity. The first research and power nuclear reactors were put into operation in the forties and fifties, and then nuclear power engineering developed very quickly over a period of the sixties and seventies. A great number of nuclear reactors of different types were studied and built over that period [1]. However, after the nuclear accidents at the Chernobyl Nuclear Power Plant (NPP) (reactor runaway with the prompt neutrons, 1986) and the Three Mile Island Nuclear Generating Station (loss-of-coolant accident with melting of the core of nuclear reactor, 1979), the development of nuclear power was suspended. Most of the nuclear power countries have blocked their nuclear energy programs, while other countries have refused to construct new NPP's [2].

Nevertheless, the interest in nuclear power has considerably risen in recent 20 years, which is caused by the fact that nuclear power has no alternative as a powerful source of electricity from the viewpoint of economics, non-proliferation of the environmental pollution and insufficiency of nonrenewable energy sources. Hence, a new development of nuclear power has begun in many countries, which is called the Nuclear Renaissance. Since about 2000 the term Nuclear Renaissance has been used to refer to a nuclear power industry revival. However, at the present time, nuclear energy suffer from a number of shortcomings that must be resolved. These are the ones: possibility of military use of nuclear energy and danger of nuclear weapons proliferation; danger of serious accidents; the problem of nuclear radioactive wastes. These reasons block further development of nuclear energy in the world. Considerable progress has been made in the past 20 years towards the problems of nuclear weapon proliferation and safety of operating NPP's, while the problem of radioactive waste disposal is still an enormously difficult problem which to date no country has fully solved.

THE PROBLEM OF NUCLEAR WASTE

The nuclear waste problem is totally unresolved up to now. The major problem of nuclear waste is what to do with it. Many of the isotopes are very radioactive for a very long time before they decay and thus methods of their disposal remain one of the most pressing problems facing the nuclear sector. There are only two realistic approaches to the nuclear waste problem at the present time: 1. Direct disposal of radioactive waste in the underground storage without reprocessing, or partial reprocessing of nuclear waste with further use of uranium and plutonium in the form of mixed oxide fuel (MOX). 2. Complete reprocessing of the spent nuclear fuel (SNF) with separation of the most dangerous long-lived high-level radioactive wastes for their further transmutation by electronuclear systems and fast neutron reactors.

The first approach, however, suffer from a number of grave shortcomings, namely:

- It's impossible to make a long-term forecast of safe storage because data is absent on long-duration strength of structural materials for a period of more than one hundred years;
- A huge amount of radioactive wastes has been stockpiled up to now, but only a few SNF storage facility units are under construction in the world. And the greatest one is Yucca Mountain Nuclear Waste Repository in Nevada, USA. The repository has a statutory limit of 77000 tons. To store this amount of waste requires 65 km of tunnels. The U.S. Department of Energy was to begin accepting spent fuel at the Yucca Mountain repository by January 31, 1998. However, 13 years after this deadline, the repository at Yucca Mountain is still over a decade away from being opened, and the opening date continues to be delayed [3]. Yucca Mountain as a repository now is off the table. It is also worthy of note in this connection that NPP's of the world produce 8000 tons of SNF each year;
- An extremely large amount of expensive materials is required to provide disposal of nuclear wastes;
- Methods of nuclear-waste repository safety analysis are absent;
- There is a high probability for long-lived radionuclides to escape from the storage facility and enter biological environment for over a period of thousands of years;
- Special geological explorations of large amounts are needed.

Considering practical aspects of radioactive waste disposal in stable geological formations, average price of SNF storage in such repository is US $ 1000 per kg. Thus, storage of high-level radioactive wastes needs US $ billion (US $ 109) per WWER-1000 unit for its forty-year operating period.

At the beginning of nuclear era it was planned to solve the problem of nuclear waste, as well as the problem of natural uranium shortage, by the implementation of fast neutron reactors. But the world's known uranium resources increased much since then, while the growth of nuclear power industry has slowed down. Therefore expected implementation of fast reactors didn't happen. It is also worthy of note that fast reactors are effective only for transmutation of transuranium elements (actinides) such as the isotopes of plutonium, neptunium, americium, and curium, while their use for transmutation of fission products such as technetium-99 leads to decrease in multiplication factor of nuclear fuel and to deterioration of physical properties of nuclear reactor which are connected with its safety [6].

Many countries, including Ukraine, use a single-phase fuel cycle. SNF in this case is assembled in a spent fuel storage pool at the NPP unit or in intermediate storage facilities for spent fuel. Some countries started reprocessing of SNF by making use of PUREX technology for separation of uranium and plutonium that was available from military studies [7]. PUREX is an acronym standing for Plutonium-URanium EXtraction — de facto standard aqueous nuclear reprocessing method for the recovery of uranium and plutonium from used nuclear fuel. It is based on liquid-liquid extraction ion-exchange.

A part of separated plutonium is used as MOX fuel in common nuclear reactors, while the remainder mixture of secondary actinides is prepared for final disposal. The majority of nuclear power countries today have growing stocks of SNF or separated plutonium and high-level radioactive wastes in the form of glass units, and further future of these materials is quite uncertain.

Geological disposal of radioactive waste is an acceptable decision now for safety of the people and environmental protection. However, difficulties and problems associated with design, development and licensing of waste disposal facilities, as well as social protest against nuclear waste, suspend the implementation of this technology.

POSSIBILITY OF NUCLEAR WASTE TRANSMUTATION

Nuclear transmutation processes and their possible application to the reprocessing and reduction of radioactive nuclear wastes are widely discussed in the last decades. Nuclear transmutation is the conversion of one chemical element or isotope into another. This occurs either through nuclear reactions (in which an outside particle reacts with a nucleus), or through radioactive decay (where no outside particle is needed). Natural transmutation occurs when certain radioactive elements spontaneously decay over a period of time, transforming into other elements. Artificial transmutation may occur in devices that have enough energy to cause changes in the nuclear structure of the elements. However, some studies show possibility of artificial transmutation in biological systems without any additional input of energy. Nevertheless, this effect of low-temperature nuclear transmutation of isotopes in microbiological systems is under question [8].
Considering high-level radioactive wastes contained in SNF, it becomes clear that two main groups of the elements need transmutation. Transuranium elements, such as plutonium and minor actinides, form the first group. Nuclear fission products form the second group. The main elements and isotopes contained in SNF are presented in table I.

Table I. Average composition of spent nuclear fuel (main radionuclides/1 GWel·year) [9].

Actinides				Fission products			
Nuclide	$T_{1/2}$, (a)	Mass, (kg)		Nuclide	$T_{1/2}$, (a)	Mass, (kg)	Isotope fraction, (%)
		U(U-235)	MOX				
U-235	$7.0 \cdot 10^8$	280	50	Kr-85	10.8	0.4	
U-236	$2.3 \cdot 10^7$	120	20	Sr-90	29	14	
U-238	$4.5 \cdot 10^9$	$2.8 \cdot 10^4$	$2.7 \cdot 10^4$	Cs-137	30	32	
Np-237	$2.1 \cdot 10^6$	15	10	Sm-151	93	0.3	
Pu-238	88	6 (2 %)	25 (3.5 %)	Se-79	$6.5 \cdot 10^4$	0.2	10
Pu-239	$2.4 \cdot 10^4$	170 (57 %)	350 (47.5 %)	Zr-93	$1.5 \cdot 10^6$	23	20
Pu-240	6600	70 (23 %)	200 (27 %)	Tc-99	$2.1 \cdot 10^5$	25	100
Pu-241	14	40 (13 %)	80 (11 %)	Pd-107	$6.5 \cdot 10^6$	7	16
Pu-242	$3.8 \cdot 10^5$	15 (5 %)	80 (11 %)	Sn-126	$1.0 \cdot 10^5$	1	31
Am-241	430	7	30	I-129	$1.6 \cdot 10^7$	6	75
Am-242	141	0.1	0.2	Cs-135	$2 \cdot 10^6$	10	14
Am-243	7370	3	25				
Cm-244	18	0.7	15				
Cm-245	8500	0.1	3				

Let us consider the process of high-energy artificial transmutation (first of all neutron transmutation) in more detail. In essence, each type of transmutation is a function of neutron cross sections. Fission is the most evident nuclear reaction for the process of minor actinide transmutation, therefore competition between nuclear fission and neutron capture processes is highly important. Hence, special ratio $\alpha = \sigma_c/\sigma_f$ is defined, where σc is the neutron capture cross section and σ_f is the nuclear fission cross section. The value of ratio α for different isotopes is presented in table II. Fast neutron spectrum is seen to be much more efficient than the thermal one for transmutation of these isotopes (the value of ratio α is the least).

Table II. Fusion, caption cross section, and the ratio for different elements. Average cross section (barn) [10].

Isotope	PWR spectrum			Fast neutron spectrum		
	σ_f	σ_c	α	σ_f	σ_c	α
Np-237	0.52	33	63	0.32	1.7	5.3
Np-238	134	13.6	0.1	3.6	0.2	0.05
Pu-238	2.4	27.7	12	1.1	0.58	0.53
Pu-239	102	58.7	0.58	1.86	0.56	0.3
Pu-240	0.53	210.2	396.6	0.36	0.57	1.6
Pu-241	102.2	40,9	0.4	2.49	0.47	0.19
Pu-242	0.44	28.8	65.5	0.24	0.44	1.8
Am-241	1.1	110	100	0.27	2.0	7.4
Am-242	159	301	1.9	3.2	0.6	0.19
Am-242m	596	137	0.23	3.3	0.6	0.18
Am-243	0.44	49	111	0.21	1.8	8.6
Cm-242	1.14	4.5	3.9	0.58	1.0	1.7
Cm-243	88	14	0.16	7.2	1.0	0.14
Cm-244	1.0	16	16	0.42	0.6	1.4
Cm-245	116	17	0.15	5.1	0.9	0.18
U-235	38.8	8.7	0.22	1.98	0.57	0.29
U-238	0.103	0.86	8.3	0.04	0.3	7.5

In principle, the process of transmutation may take place in practical industrial nuclear power reactors of LWR type, but fast nuclear reactors are more effective for this purpose. Considerations suggesting that transuranium elements, including plutonium, can in fact be burned up in fast reactor operating in a self-regulation regime were also presented (the so-called traveling wave fission reactor operating in a self-adjustable neutron-nuclear mode) [11]. However, this proposal is at the beginning of its elaboration. Accelerator driven systems (ADS) are believed to be the most safe and effective solution for radioactive waste transmutation at the present time [4, 12, 13].

THE MODELING OF NUCLEAR WASTE TRANSMUTATION IN ADS DRIVEN BY HIGH-INTENSITY NEUTRON GENERATOR

In [14-16] we proposed effective setup of two-zone subcritical research reactor driven by high-intensity neutron generator. This reactor has regions with fast and slow neutron spectrum. Here we examine possibility of nuclear waste transmutation in such a system by modeling of physical processes of accumulation and transmutation of radioactive nuclides. Charged particles (deutrons) move from top to bottom by the central tube of the system (see fig.1) and finally hit the titanium target saturated with tritium. Fusion of a deuterium and a tritium nucleus (D-T nuclear reaction) results in the formation of a He-4 nucleus and a neutron with a kinetic energy of approximately 14.1 MeV. The titanium target lies on a copper supporting stand cooled by water. The water coolant tubes run under the target. The fast neutron region surrounding the central tube is contained in a tank made from stainless steel. This region is composed of shortened fuel pins from WWER-1000 reactor and it uses liquid helium as a coolant. The enriched uranium dioxide with 20% enrichment level serves as a fuel in the fast neutron region. The latter one is surrounded by the thermal region that is also composed of shortened fuel pins from WWER-1000 reactor, but the thermal region uses light water as a coolant. The enriched uranium dioxide with 4% enrichment level serves as a fuel in the thermal neutron region.

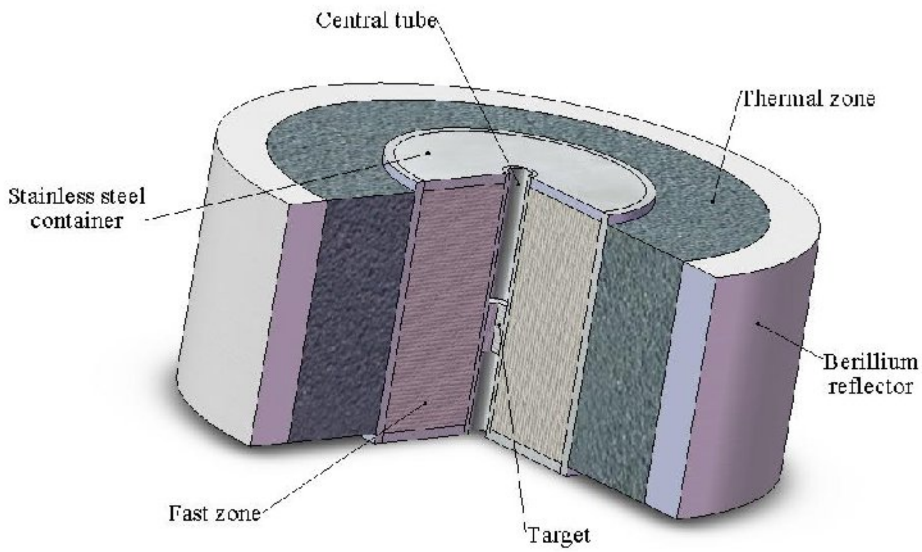

Fig. 1. The simplified subcritical assembly

PROBLEM DEFINITION

The well-known neutron-physical Monte Carlo transport code MCNP-4c [17] was employed for problem modeling and calculations. We also use MONTEBURNS code [18] that is an automated multi-step Monte Carlo burnup code system. MONTEBURNS calculates coupled neutronic/isotopic results for nuclear systems and produces a large number of criticality and burnup results based on various material feed/removal specifications, power(s), and time intervals. MONTEBURNS is a fully automated tool that links the MCNP Monte Carlo transport code with a radioactive decay and burnup code ORIGEN2. Calculations were made for subcritical reactor (k_{eff} = 0.97) depicted in fig. 1 that has a point source of 14 MeV neutrons at the center. Large numbers of 14 MeV neutrons can be produced using the $T(d,n)^4He$ reaction, as was mentioned. ENDF/B-VI nuclear reaction database containing evaluated (recommended) cross sections, fission product yields and other data, with emphasis on neutron-induced reactions, was also employed to provide calculations with basic nuclear data.

According to the analysis in previous sections, transmutation rates of Np-237, Am-241, Am-242, Am-243, Cm-244 and Cm-245 isotopes were calculated at the center of fast region. This region has nearly optimal conditions for the minor actinide transmutation, i.e. there is an extremely high fast neutron flux there. The periphery of the thermal region is the best area for transmutation of long-lived fission products, thus I-129 and Tc-99 isotopes were put into this region.

In our simulations we modeled transmutation of all the amount of nuclear waste produced by 1 GWt nuclear reactor of LWR type after a one-year operating period (see table 1). Thus, Np-237 (32 fuel elements), Am-241 (21 fuel elements), Am-242 (1 fuel element), Am-243 (9 fuel elements), Cm-244 (2 fuel elements) and Cm-245 (1 fuel element) isotopes were put into the fast region. The long-lived fission products I-129 (50 fuel elements) and Tc-99 (91 fuel elements) were put into the thermal region, as discussed above. Pins with radioactive nuclear waste were placed in the reactor core instead of usual fuel pins.

There is no substantial change in criticality of the system after the replacement of ordinary fuel pins with minor actinide pins in the fast region. But placement of long-lived fission products, especially technetium, leads to essential decrease in criticality level of the system. Thus, all the minor actinides and I-129 isotope were put into the system, but only 7 pins with Tc-99 were put into the thermal region in order to preserve the given level of subcriticality at k_{eff} = 0.97.

RESULTS OF NUCLEAR WASTE TRANSMUTATION MODELING

Modeling of nuclear waste transmutation was performed assuming that power of the system is 0.5 MWt. It corresponds to the external neutron source strength of $1 \cdot 10^{13}$ n/sec. Choice of the external neutron source is discussed in [19]. Period of nuclear waste transmutation modeling was chosen to be 10 years. In addition to isotope transmutation in the reactor core, we also calculated production of radioactive isotopes in the whole subcritical system. Calculation results of nuclear waste transmutation modeling in the subcritical system are presented in table III

Table III. The modeling results of transmutation of nuclear waste.

Isotope	Initial load, g	Transmuted, g	Transmuted in regard to initial load, %	Additionally, accumulated throughout the system, g	Effectiveness of transmutation, g
Np-237	14700	49.275	0.335	117.08	-67.8
Am-241	6960	140.671	2.02	0.673	139.99
Am-242	332	16.99	5.12	4.24	12.75
Am-243	2980	17.009	0.57	0.135	16.87
Cm-244	656	209.4	31.92	9.36	200.043
Cm-245	328	2.133	0.65	0.485	1.648
I-129	5980	44.42	0.743	24.44	19.976
Tc-99	1950	7.23	0.121	112.88	-105.65

These results show possibility of transmutation processes even in such a low-power experimental electronuclear system. We shall note the following results for transmutation of different isotopes:

- Np-237. Production of this isotope is the largest among the minor actinides for LWR reactor, but transmutation rate of Np-237 is quite low. Only 50 g of Np-237 were transmuted, while its initial quantity is equal to 15 kg. Also additional 117 g of Np-237 were produced in the reactor core;
- Am-241. 140 g of Am-241 were transmuted, while 7 kg of this isotope were loaded. And 108 g of it transforms into Np-237 that has a longer half-life and lower transmutation rate;
- Am-242. Transmutation rate of this isotope is quite high, while its quantity is quite low;
- Am-243. Only 17 g of Am-243 were transmuted, while its initial quantity is equal to 3 kg. Additional 0.13 g of this isotope were produced in the reactor core. Transmutation rate of this isotope is quite low;
- Cm-244. One third of this isotope was transmuted, while 656 g of Cm-244 is the initial loading. Cm-244 has the highest transmutation rate among the minor actinides;
- Cm-245. Only 2 g of Cm-245 were transmuted, while its initial quantity is equal to 328 g. Additional 0.485 g of this isotope were produced in the reactor core. Transmutation rate of this isotope is quite low;
- I-129. Only 44 g of I-129 were transmuted, while its initial quantity is equal to 6 kg. Additional 24.44 g of this isotope were produced in the reactor core. Transmutation rate of this isotope is quite low, while quite large amount of this isotope is produced in the reactor core during operating time of electronuclear system;
- Tc-99. Transmutation rate of this isotope is low, while the additional amount of this isotope produced in the reactor core is the highest.

These results show possibility of nuclear waste transmutation in two-zone subcritical system driven by a middle-intensity neutron source. Further study and calculations are also needed to establish optimal isotope composition at every core loading, layout of fuel, conditions of irradiation and so on.

CONCLUSION

Thus, radioactive wastes can be divided into two main groups that need to be transmuted. The minor actinides form the first group and the long-lived fission products form the second one. For the purpose of effective transmutation these isotopes must be extracted from the spent nuclear fuel with the help of either PUREX technology or pyrometallurgical technology.
The two-zone reactor system with fast and thermal regions is more effective for nuclear waste transmutation than the one-zone reactor. Modeling results show that nearly all radioactive wastes can be transmuted in the two-zone subcritical system driven by a high-intensity neutron generator with the external neutron source strength of $1 \cdot 10^{13}$ n/sec. Obviously, transmutation rate will increase with a rise of the external neutron source strength. From the results above we can also see that the initial loading of radioactive isotopes into the reactor system should exceed by mass those isotopes that are finally produced.

REFERENCES

1. Barabashev S.V., Zibnicky R.G., Shimchev S.A. The world of nuclear energy / edited (by) Barabashev S.V. - Zaporozhye: Dikoe pole, 2007. – 112 p.
2. Babenko V.A. Jenkovszky L.L. Pavlovich V.N. Nuclear Power: Tendencies in the World and Especially Ukraine, Physics of Particles and Nuclei, 2007, Vol. 38, No. 6, pp. 795–826.
3. Bznuni S.A. et al. Promising ADS // The physics of elementary particles and atomic nucleus. - 2003. - v. 34. – pp. 977-1032.
4. Kadi Y., Revol J.P. Design of an Accelerator-Driven System for the Destruction of Nuclear Waste: Proceeding // Workshop on Hybrid Nuclear Systems for Energy Production, Utilization of Actinides and Transmutation of Long-Lived Radioactive Waste. - Trieste, Italy: ICTP, 3-7 September 2001.
5. Bomko V.A. et al. The fast neutron incineration of nuclear waste in ADS – alternative of geological disposal for Ukraine // Problems of atomic science and technology. - 2004. - №4, Series: Plasma Electronics and New Methods of Acceleration. – pp. 43-50.
6. Chmelev A.N. et al. Nuclear Energy Systems: Safety, Transmutation of Radiowastes and Fuel Self-Sustaining // Progress in Nuclear Energy. - 1995. - Vol. 29. - P.439-443.
7. Sokolova S.D. Advanced and innovative technology reprocessing // Nuclear technology foreign, - 2009, - №6, - pp. 3 – 15.
8. Vysotskii V.I., Kornilova A.A. Nuclear fusion and transmutation of isotopes in biological systems. - Moscow: Mir, 2003. – 304 p.
9. Taczanowski S. Transmutations of nuclear waste in accelerator-driven subcritical systems // Applied Energy. - 2003. - Vol. 75. - P.97-117.
10. Salvatores M. The physics of transmutation in critical or subcritical reactors // C.R. Physique. - 2002. - Vol. 3. - P.999-1012.
11. Pavlovich V.N., Rusov V.D., Khotyaintsev V.M., Khotyaintseva O.M. Slow nuclear fission wave reactor // 10th international sac seminar on advanced nuclear fuel cycle for the XXI century.2007, Nizhny Novgorod. P.111
12. Chmelev A.N. et al. Nuclear Energy Systems: Safety, Transmutation of Radiowastes and Fuel Self-Sustaining // Progress in Nuclear Energy. - 1995. - Vol. 29. - P.439-443.
13. Beastle L.H. Application of Partitioning/Transmutation of Radioactive Materials in Radioactive Waste Management: Proceeding // Workshop on Hybrid Nuclear Systems for Energy Production, Utilization of Actinides and Transmutation of Long-Lived Radioactive Waste. - Trieste, Italy: ICTP, 3-7 September 2001.
14. Babenko V.A., Gulik V.I., Jenkovszky L.L., Pavlovich V.N., Pupirina E.A. Study of one-zone subcritical amplifiers of neutron flux involving enriched uranium // Problems of Atomic Science and Technology, Series: Nuclear Physics Investigations (45). – 2005. - v. 6. - p. 122-126.

15. Babenko V.A., Gulik V.I., Pavlovich V.N., Pupirina E.A. Two-zones subcritical nuclear reactors // Problems of NPP' Safety and of Chornobyl/No 6, 2006, pp.8-15
16. Babenko V.A., Gulik V.I., Pavlovich V.N. Research subritical reactor // Nuclear physics and energy. – 2008. - № 1(23). - pp. 56-61.
17. Harmon C.D. et al. Criticality Calculation with MCNP. A Primer / L-I-12827-M, Los Alamos. – 1994.
18. Briesmeister J. MCNP General Monte Carlo Code N–Particle Transport Code Version 4C / LA–13709–M, Los Alamos. – 1994.
19. Babenko V.A., Gulik V.I., Pavlovych V.M. Two-zone Sub-critical Reactor Driven by a High-intensity Neutron Generator as a Research Facility for the Nuclear Waste Transmutation // Proc. of Waste Management Conference (WM2010). - Phoenix, USA. - 7-11 March, 2010.