BEAM TECHNOLOGIES FOR INCINERATION AND TRANSMUTATION OF THE NUCLEAR WASTE

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The problem of nuclear wastes is accounted from the viewpoint of 3 aspects: ecological expediency, influence on the environment, and safety with respect to explosion. The most efficient method is burning of the wastes in the Energy Amplifier, which is based on the complex of the reactor-accelerator. The goal of this paper is to scope the development of complex techniques of the transuranic elements (TRU) incineration, and transmutation most hazardous long-lived radionuclides, fragments of nuclear fission, eliminated with the heat removal through the natural convection of air. The main part of the work is associated with setting up the problem for Ukrainian nuclear energetics: the nuclear waste incineration as an alternative to the geological disposal.

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1 INTRODUCTION

The increasing amount of the nuclear waste from nuclear reactors is a problem that in a short time may become one of the main reasons for refusal from nuclear energetics. Such a situation takes place in several leading countries. This occurs in spite of the severe danger of deterioration of the equilibrium of atmospheric processes due to the vast amount of CO_2 released during incineration of the mineral hydrocarbon fuel. The threat of environmental pollution became a political problem as it badly irritates the wide public. The amount of the radioactive waste produced at nuclear reactors of total electric power of 400 GW by 2010 will have reached 300000 tones.

The most abundant and highly active radionuclides are those produced at the reactors with nuclear fission, and transuranic elements (TRU) arising in nuclear reactions on slow neutrons. Radionuclides, fission fragments (FF), the products of the uranium fission, produced in amount of 1.8 tone per year at one LWR block have radioactivity one order higher than radioactivity of TRU during first 50 years after the waste discharge. Later their contribution to the total radioactivity drops but some radionuclides have an extremely high half-life and their disposal in geological repositories is very expensive.

The transuranic elements produced at one reactor in amount of 0.48 tone per year are very long-lived, and their radioactivity after 1000 years is 10^5 higher than the fission product activity. And, though all transuranic elements possess reasonably high fissionability it is impossible to use them in present slow neutron reactor.

The goal of this paper is to scope the development of complex techniques of the TRU incineration, and most transmutation hazardous long-lived radionuclides, fragments of nuclear fission. The most important reasons for that are economical feasibility, the minimal affect on the environment and safety, as all the processes of reprocessing, incineration, and storage occur under sub-critical conditions.

The main part of the work is associated with setting up the problem for Ukrainian nuclear energetics: the nuclear waste incineration as an alternative to the geological disposal.

2 NUCLEAR WASTE IN UKRAINE

Currently at the Ukrainian power stations there are 11 reactors under operation of the LWR-1000 and 2 reactors of the LWR-440 type [2]. Their total electric power is 12 GW. For a 3-year fuel cycle (2 reloads and 1 discharge) a year, 23.3 tones of waste are discharged from one reactor. The total amount of fuel units (FU) that simultaneously placed at 12 reactors in Ukraine (for simplicity we will take that 2 LWR-440 reactors are equal to one unit of LWR-1000) is 1956. They contain 840 tones of fuel in uranium enriched to 4.4% in ^{235}U for year. Accounting the weight of the constructions (31%), with 3-year fuel cycle it will be produced 406 tones of activated waste. As the operation life of such reactors is 40 years it will be accumulated 16240 tones of radioactive waste. Composition of nuclear waste produced in Ukraine annually and in 40 years of operation of 12 reactors of LWR type is given in the Table 1.

Table 1 Composition and quantity of nuclear waste produced at reactors in Ukraine

	%	t/year	t/40 years
U	94.771	264.536	10581.420
Np	0.059	0.165	6.165
Pt	0.951	2.663	106.512
Am	0/092	0.257	10.304
Cu	0.00217	0.006	0.243
F F	4.124	11.547	461.888

In the Fig. 1 radiotoxicity of all waste components are given in absolute values (Sievert) as a function of time (left scale) and related to radiotoxicity of coal necessary to be burnt to produce the same energy (right scale). The example of nuclear waste accumulated in amount of 2527 tones. To estimate the value of radiotoxicity from 12 nuclear reactors operating in Ukraine it is necessary to multiply all values given in the Fig.1 by 6.426.

As one can see from the Fig.1 at first (during approx. 100 years) radiotoxicity of fission fragments dominants. ^{137}Cs and ^{90}Sr isotopes play a significant role. In 1000 years with their decay the contribution of TRU (in amount of only 1 % of the total waste mass) will be about 99.995% of the total radiotoxicity. The further TRU decay will last for million years.

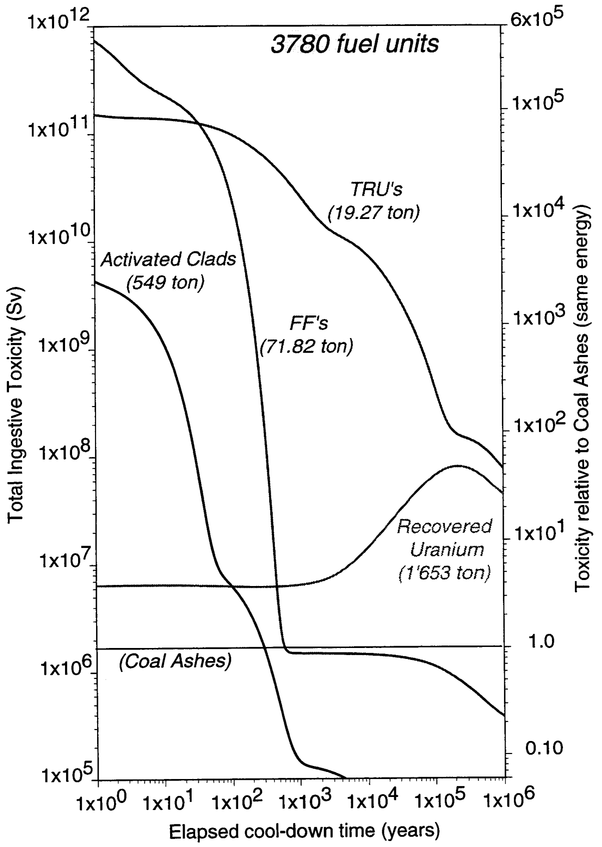

Fig. 1. Radiotoxity in absolute units and referred to coal ashes for the same delivered energy of LWR waste.

The present practice of handling with the spent nuclear fuel lies in its keeping at power plants during 30-50 years, and, after that, in its disposal in the geological repositories. The necessity of its reprocessing with separation of uranium, transuranic elements, radionuclides - products of fission, and activated clads is disputed because of technological difficulties and for economical reasons. However, this practice has the essential drawbacks:transuranic elements having a large energy story and half-lives comparable with geologic are buried; it is unreal to provide retention during 10^5 years. Radionuclides have high penetration ability. The economical consideration of the process of disposal in the geological repositories under controlled conditions of storing gives an averaged estimation of about 800 $US per a kilogram of waste. Therefore, the geological disposal of 16240 t waste requires the giant amount of 13 Billion $US.

The TRU incineration process consists in transformation of their nuclei through the fission reaction. In the case of fast neutrons the probability of neutron capture with nuclei is much higher than for the thermal neutrons. In thermal neutron reactors TRU incineration is practically absent.

A small amount of fast neutron reactors operating in over-critical mode use a mixture of uranium and plutonium as a fuel. However, they have not received wide recognition because the process of plutonium incineration only occurs when the concentration of plutonium exceeds 15%. At a lower concentration, the process of the further production (breeding) of plutonium takes place. However mode of operation with high concentration of fissionable plutonium under over-critical mode is associated with the danger of the critical mass.The proposition of Russian scientists [4] about a possibility of plutonium incineration in the fast neutron reactors in self-controlled neutron-nuclear mode (neutron-fission wave) is well known. However, this proposition is at the first stage of development.

In several countries development of intense proton linear accelerators (the average beam power of about 100 MW) is under way. They can perform transmutation of radionuclides from nuclear waste [5-8].

The most efficient and safe alternative of plutonium and other higher actinides incineration is electronuclear energetic system proposed by the CERN group under supervision of Carlo Rubbia and named "Energy Amplifier" (EA). The detailed description of the installation is presented in the references [9-13].

The Energy Amplifier lies at the cross of accelerating technologies and technologies of energy production in the process of nuclear fission. The conceptual peculiarities of EA consists of 4 basic novelties:

1. Sub-criticality. In the basis of EA lies a sub-critical nuclear reactor with a neutron multiplication factor k=0.97-0.98. This factor is a guarantee of the absolute safety of nuclear energetics and eliminates a possibility of emergency.
2. The deficiency in neutrons necessary for the chain reaction is compensated due to the neutrons generated in the spallation-reaction on nuclei with large mass number irradiated with a proton beam accelerated to the energy of 1-1.5 GeV. The initial spectrum of these neutrons is very hard.
3. The neutron moderator is lead. The process of neutron dempingproceeds adiabatically, by their multiple scattering on the lead nuclei. Lead in the EA has several function simultaneously: generating spallation-neutrons, their moderation, energy transfer by natural convection, it serves as a environment for fuel elements positioning. Finally it is an excellent shielding material and most the radiation produced by EA core is readily and promptly absorbed.
4. As a fuel for the EA various mixtures of fission materials may serve. The most effective are mixtures of monoisotopic thorium with transuranic elements produced in nuclear reactors on slow neutrons, and mixtures of thorium with ^{235}U or ^{233}U, weapon plutonium.

Besides the immediate problem of the creation of new sources of cheap and safe nuclear energetics operating on available and practically unexaustible fuel (thorium) the EA could radically solve other challenging problem of nuclear energetics - incineration of radioactive nuclear waste. All of them are introduced into a thorium matrix of fuel elements and the process of their incineration is accompanied by generation of 233U according to the reaction chain 232Th + n → 233Th → 233Pa → 233U. The latter may be used for initiation of the EA new cycles or in theLWR reactors.

Plutonium and other transuranic elements are completely incinerated in several cycles. The process is accompanied by release of a great amount of energy, 940 MW·days per 1 kg of TRU. In practice, this release rises due to energy of fission fragments (several percents) and fission reaction of ^{233}U generated from ^{232}Th.

It follows from the given information that one EA with the heat power of 1500 MW operating on the TRU-thorium fuel mixture will produce 547.5 GW·days of heat power annually, or with electric power of 625 MW - about 6 billion kW·hours of electric energy annually. With that 420 kg of TRU would be liquidated.

Thus for incineration of 123 tones of TRU 293 EA-cells x years or 8 EA units that will work during about 40 years are required. Incineration of that amount TRU would allow to obtain addition energy about 40% of the total energy, produced at LWR. The cost of additional energy is estimated as 80 Billion $US.

3 TRANSMUTATION OF LONG-LIVED FISSION FRAGMENTS

The main goal of EA operation in the mode of incineration of nuclear waste is elimination of TRU that gives the decisive contribution to radiotoxicity after the LWR operation cycle. However, in the case of successful incineration there left several long-lived radionuclides that give the basic contribution to the residual waste radiotoxicity, therefore it is necessary to consider a possibility of their transmutation.

The fission fragments capture neutrons and decay with transformation into other short-lived and then into a stable element. This method requires high isotopic purity, otherwise other, mainly stable, isotopes will capture a large number of neutrons and will be transformed into active elements again.

Radiotoxicity of the fission fragments as a function of time is presented in the Fig. 2 [1]. As one can see, the are six FF which have constant activities during long time comparable with geologic time. Complete data on composition and characteristics of long-lived fission fragments are given in the Table 2.

Table 2. Composition and characteristics of long-lived fission fragments for a standard LWR after 40 years of operation [11]

	Initial mass, kg	Half-life, years	Activity, Cu	Disp. vol., m^3
^{99}Tc	843	$2.11\cdot10^5$	14455	48181
^{129}I	196.02	$1.57\cdot10^7$	34.7	4.327
^{93}Zr	810.4	$1.53\cdot10^6$	2040.1	583
^{135}Cs	442.2	$2.3\cdot10^6$	510.1	510
^{126}Sn	29.48	$1.0\cdot10^5$	838.1	239
^{79}Se	6.57	$6.5\cdot10^4$	458.6	131

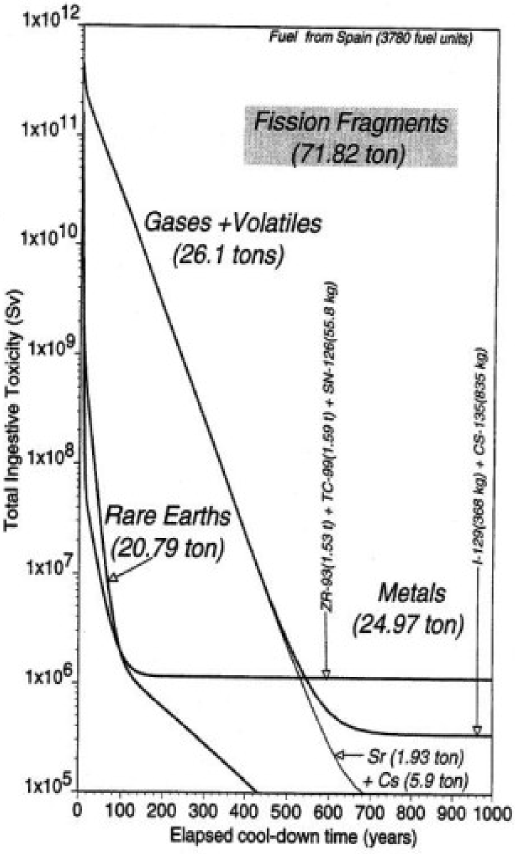

Fig. 2. Ingestive radiotoxicity of the fission fragments.

The presented consideration shows that ^{99}Tc and ^{129}I should undergo transmutation first. As a result of their removal the volume of long-lived waste disposal in A-class would be lowed by a factor of 37 (from 53971 m^3 to 1463 m^3).

In practice, a possibility of transmutation of long-lived waste requires the availability of an intense neutron source. The minimal number of neutron necessary for complete transmutation of 3 most dangerous isotopes ^{99}Tc, ^{129}I, and ^{79}Se generated during 40 years at a standard LWR will take 11.29 kg of neutron (1 kg of neutron contains $5.97\cdot10^{26}$ neutrons).Energy Amplifier will be generated 106.2 kilograms of neutrons. A part of the flux ("flowing away") will be used for the transmutation.

Currently experimental investigations in physics of fast neutrons generated in the process of the spallation-reaction, their transformation in the process of multiple scattering are carried out intensively and the characteristics of the process of the neutron transmutation are studied. The most typical result was obtained at the CERN [16] in the experiment with the initial proton beam accelerated at the proton synchrotron (PS) to the energies of 2.5 and 3.57 GeV. The method of transmutation (named TARC - Transmutation by Adiabatic Resonance Crossing) based on the application of resonant capture of neutrons by nuclei with adiabatic reduction of the neutron energy in the lead environment gives a possibility to increase essentially neutron capture efficiency.

From the Fig. 3 [16] it is seen that in the process of the spallation-reaction neutrons are generated within the energy range 10^4-10^7 eV. The energy of about 14% of them exceeds 20 MeV. In the process of the elastic neutron scattering the adiabatic decrease of their energy from 10^7 eV to thermal occurs. On this background the resonant peaks of the neutron capture by ^{99}Tc nuclei is clearly seen. Particularly sharp resonance occurs at the energy of 5.6 keV (400 barn), which neutrons cross in three steps. The resonant integral of the neutron capture by ^{99}Tc nuclei is 310 barn whereas the transverse cross-section of the thermal neutron capture is about 20 barn. The neutron captured by ^{99}Tc nuclei (τ = 2.1·10^5 years) generate ^{100}Tc, which decays into stable ^{100}Ru. In such a manner the transmutation with adiabatic intersection of resonances appears to be an order more effective than on the thermal neutrons.

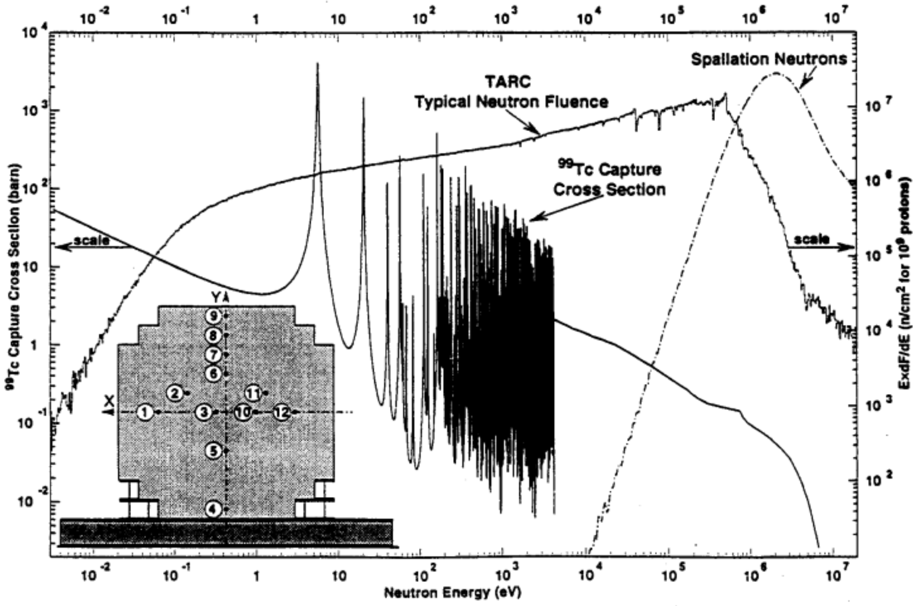

Fig. 3. ^{99}Tc neutron capture cross-section and neutron fluence energy distribution as a function of neutron energy for 3.57 GeV protons.

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