DISPOSAL OF RADIOACTIVE WASTE BY MEANS OF NUCLEAR CONVERSION

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Nowadays radioactive waste (RW) are being buried in geologic rocks. There is an alternative method of waste disposal. It is a transmutation of radioactive isotopes into stable ones by nuclear reactions with the use of accelerators with a particle energy of 50-1000 MeV. We have shown that neutrons from spallation reactions have no advantages in comparison with a hard neutron spectrum reactor technology. We have shown that during reprocessing the Th-U reactor RW, where there is a lack of actinide group, the effectiveness of proton beams and neutrons from the spallation reaction for RW disposal may be at the required level.
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Radioactive products are accumulated as a result of U nucleus fission in the reactors and successive radiation neutron captures. The problem of disposal of radioactive waste from nuclear power plants has been discussed in literature since the end of the 60th. On the one hand it is the most important problem the solution of which determines the development of the environmentally safe nuclear engineering on the Earth. A number of variants for this problem be solved has been proposed. On the other hand, each of suggested solutions is rather complex from the mechanical point of view and the successful realization causes so many problems that up till now none of proposals has been realized seriously in the world. There is only the well-known conception of radioactive waste disposal by burring them in stable geological rocks after hardening as glass or ceramics. The tectonic stability of these formations over at least 1000 years should permit to decrease the potential danger to the acceptable level. And though the areas required for their storage are relatively small (3000 m^2 for 1 GW per year), the social opinion considers such a solution of this problem unacceptable and it is the moving force to search new more effective methods of radioactive waste transmutation. The solution of the problem is especially urgent in the Ukraine therefore 13 power reactors of 12 GW produce about 30% of all the electrical energy.

The single reliable and safe method of long-lived radioactive nuclear waste disposal is the transmutation i.e. the conversion them into stable isotopes by neutron [1] or other radioactive radiation. Many of nuclear physical laboratories of the world are engaged now in basing this practically admissible method of transmutation but up to date none of suggested methods has been used. The difficulty is due to that the quantity of radioactive nuclei in waste of the atomic industry to be transmutated is rather high and transmutation of every kind of radioactive nuclei requires definite individual conditions, and the assertion of the type that it is enough to place these fission fragments into a nuclear reactor with a high neutron density to expose them to effective transmutation evidences on misunderstanding of this process. And the main condition is that the effective transmutation of radioactive nuclei is possible only on pure isotopic targets. Therefore all these variants of transmutation begin to work after waste fuels are disintegrated into separate elements and isotopes. In NSC KIPT over many years a new original method of ion mass separation is developed and in our opinion this problem should be solved in the cooperation with other Institutes of the Ukraine.

In [1] the problem of using fission reactors with high-energy spectrum for lowering content of isotopes such as Krypton-85, Strontium-90 and Cesium-137 was considered. In [2] the same problem was researched supposing that generators of neutrons are protons of the electronuclear installation. The application of a thermonuclear reactor for transmutation was considered in [3]. All the above mentioned technologies were generalized and studied in the well-know work [4] where following conclusions have been made.

Disposing of fission products such as Strontium-90, Cesium-137, Krypton-85, long-lived radioactive nuclei (LLRN) with transmutation as a result of multiple cycles of irradiation in existing now and being designed constructions of nuclear reactors is impossible due to insufficiently high neutron flows for the significant lowering of the effective half-life decay of the aggregates of these nuclides.

Attainment of this aim requires to create neutron flows of about 10^{17} neutron/cm^{2}sec in special reactors. Possible variants may be only electronuclear reactors, since even in thermonuclear reactors the neutron flow density will be of order lower. Moreover, the latter has not been realized and electronuclear reactors require as minimum the realization of a wide program of developments. However, investigations carried out in NSC KIPT [5] show there is the inaccuracy in BNL papers quoted. For burning Cesium-137 in the intensive neutron flow with a density of 10^{17} neutron/cm^{2}sec in the spectrum of neutrons formed on the target-convertor of the linear accelerator the radioactive capture cross-section will be strongly suppressed by parallel nuclear reactions and therefore the burning time of this isotope increases from 2 to 10 years, that is absolutely unacceptable. Additional studies carried out has shown that Cesium-137 isotope can be sufficiently effectively transmutated in direct reactions with protons of rather low

energies. That conclusion obtained by theoretical methods gives the practical basis for all the problem of LLRN transmutation with proton beams of moderate energies. The idea of radioactive waste transmutation has the authorship certificate [6].

It was shown earlier [6,7] that there was the principal possibility for transmutation of long-lived nuclide of nuclear fuel cycle (NFC) into short-lived stable ones as a result of nuclear reactions (p, xn yp) under irradiation radiation of nuclear fuel fission products with protons of the energy lower than 50 MeV. To estimate the technological possibility of transmutation (burning) using the proton accelerators of low energies (Ep<100 MeV) it is necessary to have information about nuclear constants of proceeding nuclear physical processes. In preliminary estimations the results of calculations for cross-sections of long-lived radioactive fission product burning are given and the value of the energy consumption for NFC fission product transmutation with low-energy protons is considered.

To carry out the analysis of fission product transmutation with proton beams of the energy less than 50 MeV the calculations for functions of excitation reactions (*p, xn, yp*) of on nuclei ^{85}Kr, ^{88}Sr, ^{90}Sr, ^{99}Tc, ^{107}Pd, ^{133}Cs, ^{137}Cs, ^{151}Sm have been fulfilled. Calculations were made according to the statistical model of the compound nucleus taking into account the preequilibrium decay [8]. From results obtained it follows that when ^{85}Kr, ^{90}Sr, ^{167}Pd are irradiated with proton beams of the energy Ep<50 MeV, stable and short-lived nuclides are formed. ^{99}Tc irradiation with Ep<25 MeV proton beam results in formation of radioactive nuclei ^{97}Tc, ^{96}Tc and ^{93}Mo with half-life $4.6\cdot10^6$ years and $3.5\cdot10^3$ years, respectively.

During irradiation of ^{137}Cs with Ep>30 MeV the reaction channels (p, 5n) and (p, p2n) are opened leading to production of ^{133}Ba ($T_{1/2}$=10.54 y.) and ^{135}Cs ($T_{1/2}=2.3\cdot10^6$ y.), and for ^{151}Sm in all the energy range the production of long-lived nuclides takes place.

In the process of transmutation of fissile products without isotope separation the stable isotopes of a given element will transmutate into other nuclides often being radioactive ones.

Calculations results for functions of excitation of reactions proceeding on stable isotopes ^{88}Sr and ^{133}Cs show that the maximum value of $T_{1/2}$ nuclides formed during proton interaction with ^{88}Sr is 106.6 days, and in case of ^{133}Cs it is 10.54 yeas, i.e. an additional quantity of long-lived nuclides is produced that lowers transmutation effectiveness. Total reaction cross-sections of ^{88}Sr and ^{133}Cs approach to those of radioactive nuclides of strontium and cesium that results in the additional expense of an accelerated beam for reprocessing. Generally, independently on the transmutation method the burning of NFC long-lived fission products without isotope separation will lead to increasing the power consumption, to say nothing of possibility of new nuclide production. Under these conditions in some cases it can be more advantageously to carry out transmutation on isotopically enriched targets.

Let us consider the energy consumption for transmutation with low-energy protons (<50 MeV). In above-mentioned energy range the relation of probability of nuclear interactions and ionization is $<10^{-2}$. Energy consumption for transmutation of only one nucleus will be of order of 10^2 Ep/η_y, where Ep is the accelerated beam energy, η_y is the efficiency of the accelerator itself. Formation in a power reactor of one nucleus gives the useful energy equal to $E_f\eta_p/Y$, where E_f is the decay energy, η_p is the efficiency of a reactor, Y is the output per one fission. The expedient transmutation from the viewpoint of power will be defined as follows:

$$E_f\eta_p/Y>10^2*Ep/\eta_y.$$

Hence we obtain the limit on a cumulative NFC fission product yield:

$$E_f\eta_p\eta_y/10^2Ep > Y.$$

Assuming E_f=200 MeV, η_p=0.3, η_y=0.5 we obtain Y < 0.3/Ep.

This equation shows that it is advantageous to use a lower energy of accelerated beam. In < 50 MeV energy range the optimal transmutation energy value is 15-20 MeV, that range where for NFC fission products the maximum of (p, 2n) reaction yield takes place and its value is near the total proton absorption cross-section. Taking Ep=20 MeV one obtains the output for which the energy balance is reached, Y=1.5%, i.e., product transmutation is energetically profitable with Y<1.5%.

The result obtained is valid only for a monoisotopical target. In case of transmutation without isotope separation the limitation for transmutated nuclide yield will be

$$Y < 1.5\cdot10^{-3}\sigma_T c_T/(\Sigma c_i\sigma_i),$$

where σ_T is the transmutation cross-section, $c_т$ is the transmutated nuclide concentration and c_i and σ_i are the concentrations and total reaction cross-sections of other isotopes in the mixture.

We have shown [6] that during irradiation of nuclear fuel segments with protons of the energy less than 50 MeV as a result of (p, xn yp) nuclear reactions long-lived nuclides transmutate into short-lived and stable ones with the total transmutation cross-section of 1 barn. So, for ^{137}Cs proton beam energy of 20 MeV there are following reactions leading to stable and short-lived nuclides:

^{137}Cs(p, n)^{137}Ba; ^{137}Cs(p, 2n)^{136}Ba; ^{137}Cs(p, 3n)^{135}Ba;

^{137}Cs(p, pn) ^{136}Cs ⟶ (β^-, 13 сутоκ) ^{136}Ba;

^{137}Cs(p, αn) ^{133}Xe ⟶ (β^-, 5,29 сутоκ) ^{133}Cs.

The calculation of these reaction cross-section made on the statistical model of the compound nucleus taking into account the preequilibrium decay gives the total cross-section of above mentioned reactions 1.03 barn.

Under irradiation of radioactive nuclei with a particle beam of the flow density φ there are two processes leading to decreasing the nucleus-target number: the process of the natural radioactive decay λ and nuclear reactions transmutating the initial nuclei into other nuclides with the cross-section σ. In this case a half-decay period of the initial nuclide will be as

$$T_{1/2} = \ln2/(\lambda + \sigma\varphi).$$

At the proton 20 MeV beam density the on a cesium

target $2\cdot10^{17}$p/cm^{2}s (about 32 mA/cm^2) we obtain $T_{1/2}$=0.106 year. Since the initial amount of ^{137}Cs (and ^{90}Sr), contained in one ton of a spent fuel in 1000 times exceeds the activity of 1 T natural uranium then the irradiation time for decreasing the activity in 1000 times will be 10 $T_{1/2}$, i.e., in the given case it will be 1.06 year, that corresponds to the optimal time of transmutation as is mentioned above.

The number of nuclei transmutated during the time t for the initial quantity N_o is

$$N = N_o[1 - \exp(-\sigma\varphi t)].$$

We take N_o equal to the production of ^{137}Cs in reactors WWER of 1000 MW (el.)–$3.3\cdot10^{26}$ nuclei per year. Then under irradiation during a day we have the amount of transmutated nuclei equal to $5.8\cdot10^{24}$. Production of ^{137}Cs during twenty four hours is $9\cdot10^{23}$ nuclei. Thus, such an approach permits, in principle, to transmutate long-lived waste with processing both newly formed and storaged ones.

The energy consumption is the essential moment. In the above proton energy range the relation of probability of nuclear interaction is about 10^{-2}, i.e. to transmutate one ^{137}Cs nucleus it is necessary to accelerate about 100 protons. Then the energy consumption for transmutation of one nucleus is $W_{exp.}$=20·100=2 GeV.

So, inasmuch as for fission of one nucleus ^{235}U in the reactor the ^{137}Cs nucleus is formed with a probability $6\cdot10^{-2}$, then the «useful» energy released in reactor during formation of one ^{137}Cs nucleus we define as $W_{f.}$=200/$6\cdot10^{-2}$=3.3 GeV. Taking into account the accelerator efficiency (<50%) and the reactor efficiency (30%) we have the relation $W_{exp./}W_{f.}$>4.

The possibility to decrease the energy consumption for transmutation can be achieved: in the process of recuperating the energy of the beam passed through the target (the thickness of which must be less than the accelerator proton path) using an accumulation ring proposed by Ado et al. [9], where after passing the target the beam is accelerated again for compensation of the energy loss. It is the realization of the target as a plasma of sufficiently high density. The choice of more exceptable method of decreasing the energy losses is possible only after the comparative analysis of the above methods.

Therefore to solve the problem of NPP long-lived waste transmutation with low- energy proton beams one should solve the following questions:

- theoretical analysis of processes of proton interaction with long-lived nuclear fission products with the aim to choose an optimal beam parameters;
- development of a method of decreasing unproductive losses of the beam energy in the process of transmutation;
- experimental investigations of transmutation processes to correct the theoretical estimations of nuclear reaction cross-sections in the process of transmutation.

Finally, radioactive waste of nuclear power plants can be transmutated with the use of accelerators into stable isotopes excluding the actinide group. For this purpose two group of accelerators are necessary – accelerators of the electric nuclear breading (proton accelerators with the energy of 1-1.5 GeV and average current of 0.1 A) and accelerators the construction of which is possible already nowadays (proton accelerators with the energy of 100-300 MeV and mean current 0.001 A). However, transmutation with accelerators is possible only on monoisotopic targets therefore together with accelerators high- efficiency separators of radioactive waste should be created. Theoretical and experimental investigations have been organized on each trend and at a physical level promising results were obtained.

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