TRANSMUTATION OF RADIOACTIVE WASTE ON LOW-ENERGY PROTON ACCELERATORS

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The problem of nuclear power-generating plant radioactive waste transmutation is considered in the literature approximately since 1958. On the one hand, it is a major problem the solution of which determines the development of ecologically pure nuclear energy on the Earth. There are several solutions of this problem. On the other hand, each of the proposed solutions is so complicated in respect to engineering, and its successful realization is arranged with so many` conventions, that till now any of propositions is not seriously implemented in the world. There is an only settled concept of radioactive waste removal by their burial in stable geological structures after solidification in the form of glass or ceramics. The tectonic stability of these structures during at least 1000 years will allow to reduce the potential hazard to an acceptable level. And though areas required for this storage, are rather small (about 3000 sq.m. for 1 GW electric power per year) the public judgment believes such a solution unacceptable and it the motivation in searching new, more effective methods of transmutation of radioactive waste.

Analysis of the content of radioactive isotopes in the irradiated fuel of a LWR reactor shows that radioactive nuclides are characterized by a different physical property, their quantity among the fission products is various and they produce different danger for the environment. The majority of nuclei even with a very high activity (yttrium - 90, barium - 137, cerium - 144 etc.) have a small half-life period and their quantity will decrease up to a safe level in time about 2-3 years. Such radioactive fissile products form a group of short-lived radioactive (SLRN) and they do not represent serious danger for the environment. Other nuclei have a half-life period about 10-30 years and a high activity (strontium - 90, cesium - 137) and they decay up to a safe level in time about 1000 years. They are to be disposed or transmutated without fall by nuclear-physical methods and are termed as long-lived radioactive nuclides (LLRN). The intermediate group is made with fissile nuclei with a half-life period of about one year (ruthenium-106, cerium - 144, promethium - 147, europium - 154), transmutation of which, in our opinion, is possible also at moderate energy accelerators (hundreds of MeV).

In article [1] the problem of using fission reactors with a high-energy neutron spectrum for reducing the content of isotopes of krypton - 85, strontium-90 and cesium - 137 in radioactive waste is considered. In [2] the same problem was explored in the assumption that neutron generators of are the protons of the electronuclear installation. Application of a thermonuclear reactor for transmutation was considered in [3]. The numbered technologies are summarized and investigated in the well- known paper [4] where the following conclusions are made:

The destruction fission products, such as strontium -90, cesium - 137, and krypton - 85 (LLRN) by transmutation as a result of multiple irradiation cycles at nuclear physical reactors of existing and designed constructions is impossible because of lack of high- neutron fluences sufficient for significant lowering the effective half-life period of the majority of these nuclides. The reaching of such a purpose requires making in special reactors of the neutron density of about 10^{17} neutron/cm^{2}s. Possible variants can be only electronuclear reactors, since in thermonuclear reactors the density of neutron fluences is less by one order of magnitude. However studies which have been carried out in NSC KIPT, [5], evidence that in BNL papers we refer the inexactness is supposed. In the process of burning out cesium - 137 in an intensive neutron fluence with a density of 10^{17} neutron/cm^{2}s in a spectrum of neutrons generated in the target - converter of a linac, the section of radioactive capture will be strongly suppressed by the parallel nuclear reactions and, consequently, the time of a burning out of this isotope increases from two till ten years, that is completely unacceptable. Additional investigations we have carried out have shown, that the isotope cesium - 137 can rather effectively transmute in direct reactions with low-energy protons. This conclusion obtained by theoretical methods, puts on practical ground all the problem of LLRN transmutation using the moderate- energy proton beams.

To carry out analysis of the process of fission product transmutation with the beam of protons of an energy less than 50 MeV, the excitation functions for (p, xn yp) reactions on nuclei ^{85}Kr, ^{88}Sr, ^{90}Sr, ^{99}Tc, ^{107}Pd, ^{133}Cs, ^{137}Cs, ^{151}Sm were calculated. The calculations were carried out on a statistic model of a compound nucleus with taking into account the preequilibrium decay [7].

From our results it follows that ^{85}Kr, ^{90}Sr, ^{107}Pd under irradiation with the E_p < 50 MeV proton beams the stable or short-lived nuclides are formed. In the case of ^{99}Tc the irradiation with > 25 MeV protons leads to formation of radioactive nuclei ^{97}Tc, ^{96}Tc and ^{93}Mo with half-life periods of $4.6\cdot10^6$ years and $3.5\cdot10^3$ years, respectively.

Under irradiation of ^{137}Cs at E_p > 30 MeV the channels of (ð, 5n) and (p, p2n) reactions are opened, that allows one to produce ^{133}Ba ($T_{1/2}$=10.54 years) and ^{135}Cs ($T_{1/2}=2.3*10^6$ years), and for ^{151}Sm over total energy range the production of long-lived nuclides takes place.

The results of calculation of excitation functions for reactions on stable isotopes ^{88}Sr and ^{133}Cs show that the peak value $T_{1/2}$ of nuclides generated during interaction between protons and ^{88}Sr is 106.6 days, and in the case of ^{133}Cs it is 10.54 of year, i.e. the additional quantity of long-lived nuclides is yielded that reduces the efficiency transmutation. The total reaction cross-sections of ^{88}Sr and ^{133}Cs are close to cross-sections of radioactive nuclides of strontium and cesium, that results in additional consumption of the accelerated beam for reprocessing. Generally, independently on the

transmutation method, "burning out" of long-lived fission products of NFC without isotopic separation will increase the energy consumption, let alone the opportunity to form new nuclides. These conditions can lead to that in some of cases it may be more advantageous to carry out the transmutation on isotopic enriched targets.

We have shown in [6], that under irradiation of nuclear fuel fission fragments with protons of an energy less than 50 MeV, as a result of (p,xn yp) nuclear reactions, the transmutation of long-lived nuclides in short-lived ones and stable ones with summed transmutation cross-section of about 1 barns. takes place. So, at the energy of proton beam 20 MeV for ^{137}Cs the following reactions giving stable or short-lived nuclides take place:

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

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

^{137}Cs (p, pn) ^{136}Cs $\Rightarrow$ (β^-, 13 days) ^{136}Ba;

^{137}Cs (p, αn) ^{133}Xe $\Rightarrow$ (β^-, 5,29 days) ^{133}Cs;

The calculation of these reaction cross-sections carried out using the statistical model of a compound nuclei with taking into account the preequilibrium of decay gives the summed cross-section of numbered reaction equal to 1.03 barns.

Under irradiation of radioactive nuclei with a particle beam of a fluence density φ, there are two processes leading to decreasing the numbers of nuclei – targets: process of a natural radioactive decay λ with a decay constant and nuclear reactions transmuting the initial nucleus in other nuclides with the cross-section σ takes place. In this case the half-life period of an initial nuclide looks as

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

At a density of a 20 MeV proton beam on a cesium target $2\cdot10^{17}$ p/cm^{2}s (about 32 mA/cm^2) $T_{1/2}$=0.106 year is gained. As the initial quantity of ^{137}Cs (and ^{90}Sr), contained in one ton of spent fuel exceeds in 1000 times the activity 1T of natural uranium, the time of irradiation for reducing the activity in 1000 times is 10 $T_{1/2}$, i.e. in this case it makes 1.06 years, that corresponds to the optimum transmutation time, as it was mentioned above.

The number of nuclei being transmuted during the time t at the initial quantity N_o is

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

No is taken equal to the annual yield of ^{137}Cs in 1000 MW WWER reactors (el.)i.e. $3.3\cdot10^{26}$ nuclei. Then under irradiation within one day we gain a number of nuclei ^{137}Cs equal to $5.8\cdot10^{24}$. The daily yield of ^{137}Cs in this reactor makes - $9\cdot10^{23}$ nuclei. Thus, such approach allows, basically, transmutation of long-lived waste with reprocessing of newly generated as well as and stored waste.

The important moment is expenditure of energy for transmutation.. In the mentioned proton energy range the relation of probability of a nuclear interaction resulting in transmutation, to ionization interaction makes about 10^{-2}, i.e. for transmutation of one nucleus ^{137}Cs about 100 protons should be accelerated. Then energy consumption for transmutation of one nuclei will be $W_{expen} = 20\cdot100=2$ GeV.

As at fission of one nucleus ^{235}U in the reactor, the nucleus ^{137}Cs is formed with a probability of $6\cdot10^{-2}$, the "useful" energy released in the reactor during formation of one nuclei ^{137}Cs is determined as $W_{tot} = 200/6\cdot10 - 2=3.3$ GeV. Taking into account the efficiency of accelerator (< 50%) and of reactor (~30 %) we obtain the ratio $W_{expen}/ W_{tot.} > 4$.

The opportunity to decrease the energy consumption for transmutation can be reached: in the process of energy beam regeneration passed through a target (in this case the thickness of a target should be less than the run of the accelerated proton) for example, using a ring of a type proposed by Ado [8], where after passage of a target a beam is accelerated once more to compensate the liberated energy, the realization of a target in the form of the plasma having sufficiently high density. The choice of the most acceptable way to reduce the energy consumption is possible only after the comparative analysis of above-mentioned opportunities.

CONCLUSION

The radioactive waste of the nuclear power industry can be transmuted in stable isotopes, except for group of actinides. For this one needs two groups of accelerators: electronuclear breeding accelerators (proton accelerators with the energy (1 - 1.5) GeV and medium current 0.1A) and accelerators which can be constructed today (proton accelerators with the energy (100 - 300) MeV and medium current 0.001A). However, only the monoisotopic targets can be transmuted on accelerators, therefore, simultaneously with accelerators high-performance radioactive waste separators should be created.

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