POSSIBILITY OF USING OF COLD FUSION FOR THE TRASMUTATION OF NUCLEAR WASTE PRODUCTS

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The possibility of using cold fusion for nuclear waste products transmutation is investigated in this paper. In generally a method is based on saturation of the titanium by a mixture of deuterium and air. Possible nuclear fusion reactions are discussed. Their "burning out" sections, effective half-life periods and intensity of neutron beams are evaluated. The applicability of the method for a transmutation of the nuclear waste containing cesium-137 is considered.

1. Introduction

Existing methods of reprocessing of radioactive waste products (RWP) do not solve a problem of their final removal from the biosphere. Fission products with long decay half-lives result in high activity of used nuclear fuel. The main contributions are from strontium-90 (half-life period $T_{1/2} = 29.1$ years), cesium-137 ($T_{1/2} = 30.17$ years),[1] krypton-85 ($T_{1/2} = 10.8$ years), technetium-99 ($T_{1/2} = 213{,}000$ years), and transuranium elements with very long half-life periods. Water dissolution technologies are as a rule used for reprocessing the used nuclear fuel. RWP transform to a liquid condition as a result. One ton of used nuclear fuel produces 2.4 tons, or 2.4 cm^3, of liquid waste products, which include 0.1 tons: highly active, 1.5 tons: medial active; 0.8 tons: low active wastes. At present, the main ways of neutralization of the RWP include various methods of concentrating of them into a solid state[2] to store it in mountain structures, with an expectation of spontaneous decay. For example, ^{90}Sr and ^{137}Cs must be stored in controllable conditions for about 300–600 years.

There is an alternative way to transform RWP into stable or short-lived isotopes. It is possible due to use irradiation of the RWP by beams of neutrons and/or the charged particles. This is known as nuclear transmutation.

The main goal at the present is the development of a suitable conventional method, or the creation of a new special physical device, for nuclear transmutation. In different science centers such kind of devices are considered. Technology for "burning off" long-living fission products of uranium in fluxes of thermal neutrons was proposed in the 60th year of past century[3], and then developed in the beginning of 80th year,[4] and in the beginning of 90th year[5] as well. At that time, different types of particle accelerators were used for the same purpose. There exists an elec-

tronuclear technology for transmutation of the RWP.[6] The electronuclear device is a hybrid of the particle accelerator and breeder zones of the nuclear power reactor. It allows energy production from "non-combustible" isotopes of uranium and thorium without self-sustaining chain reactions. The device allows the utilization of its own radioactive wastes and RWP from several other nuclear power stations. There is a project for a high-intensity neutron source based on initiation of thermonuclear fusion reactions.

2. Calculation of Transmutation Data

Artisjuk *et al.*[5] have calculated that "burning off" cross sections of ^{90}Sr and ^{137}Cs are maximal for neutrons with energy 14 and 16 MeV and have 1500 and 1800 mb of magnitude, respectively. At the same time, "burning off" cross sections of ^{137}Cs for neutron energies from 1 to 5 MeV vary from 40 up to 3 mb.[4] The energy range from 0 to 5 MeV represents an energy spectrum of instantaneous fission neutrons of a fast reactor. The maximal number of neutrons occurs at an energy of 0.45 MeV. The maximal neutron flux density has a value of 3.7×10^{15} neutron/cm^2 s for the fast reactor BOR-60. Under the formula for an effective half-life period of radioactive nuclide,[4] it is possible to calculate the half-life period for neutrons of a fast reactor in a case of ^{137}Cs.

$$T_{1/2\,\mathrm{fr}} \;=\; \frac{\ln 2}{\lambda + \sigma(E) + \varphi(E)} \;=\; 6.48\,\mathrm{years}, \tag{1}$$

where λ is a constant of decay, $\sigma(E)$ the cross section of radiation capture, $\varphi(E)$ the density of flux of neutrons, and $\sigma(E)\ \varphi(E)$ is numerically integrated on energy of neutrons. The specific activity of ^{137}Cs as a function of natural half-life period is

$$A_{\mathrm{natural}} \;=\; 0.693\,\mathrm{N}/T_{1/2} \;=\; 3.202 \times 10^{15}\,\mathrm{Bq/kg} \tag{2}$$

where N is the number of nucleus of the given matter capable to decay.

According to radiation safety rules accepted in 1999 by the Russian Ministry of Health,[7] a specific activity of ^{137}Cs of 10 Bq/g, or 10^4 Bq/kg, is not dangerous in a working place. In this case, the effective half-life period for one year of an irradiation of the RWP with ^{137}Cs is equal to $T_{1/2\mathrm{min}} = 0.026$ years. Taking into account that the decay constant for ^{137}Cs is $\lambda = 0.022969838$ per year, we find a value of the product $\sigma(E)\ \varphi(E) = 0.014785279$ per year. The corresponding density of neutron flux for fast reactor is $\varphi(E) = 6.5 \times 10^{14}$ neutron/cm^2 s. This is two orders less than the integrated density of flux of neutrons accepted at calculations in Ref.[4]. It is comparable to the neutron flux density in fast reactors.

We shall consider an opportunity of use of new source of nuclear radiation for a transmutation of nuclear waste products performed in "Method of nuclear fusion and the device for it realization" the Russian patent No. 2145123.[8]

3. Experimental Data

Data obtained in [9] shows registration of neutron pulses with energies 2.5, 4.5, 13 and 17 MeV, and a flux density of about 10^4 neutron/cm^2 s and γ-radiation with energy up to 4.5 MeV. We used ^{3}He detector of neutrons. It is possible to assume that, as a result of the realization of the method under the patent[8], neutrons with higher energy were obtained. It is known[10] that full cross sections of ^{3}He for neutrons with energy 2.4 and 14 MeV differ by almost three times. Cross sections of (n, p) reactions differ by six times. The sensitivity of a neutron detector is much less for neutrons with energy more than 2.4 MeV.

Therefore, we recalibrated this neutron detector using neutron source with higher energy. The ^{239}Pu-α-Be source has been chosen for this purpose with radiation $2.12 \times 10^5 \alpha$-particle/s in 2π angle and with the surface square $S_{source} = 160\,cm^2$. We obtained a flux of about 500 neutrons/s in Ref. [8] according to californium source ^{252}Cf of neutrons. In our case ^{239}Pu-α-Be source has average energy of neutrons $E_n = 4$ MeV. It is possible to determine from our experimental data, what actual flux of neutrons was registered.

The calculation of the neutron detector sensitivity for ^{239}Pu-α-Be source is carried out using the following formula:

$$\chi = \frac{N - N_b}{P} \tag{3}$$

where N is the average value of the account of pulses of the detector for ^{239}Pu-α-Be source, pulses/s, N_b the average value of the account of background pulses of the detector, pulses/s and power of the source evaluated by the formula

$$P = \frac{I_{source}\, Q_{source}}{2S_{source}} \tag{4}$$

where I_{source} is the external radiation of a source in 2π angle, α-particle/s, Q_{source} the output of neutrons from Be is equal to 40 neutrons/10^6 of α-particles, and the factor of 2 takes into account radiation in π angle. As a result, the sensitivity of the neutron detector for ^{239}Pu-α-Be source is equal to $\chi = 5.974$ pulses cm^2/s.

The calculation of the neutron flux from the source is carried out using

$$I = \frac{4\pi(N_{calculation} - N_b)S}{\Omega\chi} \tag{5}$$

The share of solid angle occupied with the detector of neutrons is

$$4\pi/\Omega = \sqrt{L^2 + b^{2/4} + h^{2/4}} \tag{6}$$

Here L is the distance from source of neutrons up to the detector of neutrons, which is equal to 9.5 cm, b the width of the detector of neutrons 56 cm and h is the height of the detector of neutrons 29 cm. $N_{calculation}$ is the number of pulses

per second registered in experiment, $N_{\rm b}$ the average value of pulses per one second at measurement of background and S is the square occupied with the detector $1624\,{\rm cm}^2$. For the ^{239}Pu-α-Be source, a neutron flux $I = 1.68 \times 10^5$ neutron/s was obtained.

Assume now that we will irradiate the RWP inside of our sample. In our case the size of the sample is: diameter 0.9 cm and length 3.2 cm. We can calculate neutron flux density on the surface of this sample. According to formula for isotropic distribution of the radiation from [11], density of flux of neutrons due to proposed method will be $\varphi(E)_{\rm cf} = 6.525 \times 10^4$ neutron/cm^2 s.

Possible nuclear reactions of deuterium with isotopes of nitrogen and oxygen contained in air are described in [12]. It is probable that neutrons with energies 3.3, 5, 5.7, 9.9 MeV can be obtained in such kind of reactions. We shall evaluate what amount of isotopes of oxygen ^{17}O and ^{18}O contained in deuterium with air mixture at realization of the patented method.[8] The calculations show that amount of oxygen will be 7.821×10^{-6} g ^{17}O and 4.312×10^{-5} g ^{18}O or 0.00515 and 0.0268 cm^3, accordingly. The oxygen content in a mixture with deuterium can be increased up to 4% without danger of explosion. In our case it is possible to increase of volume of oxygen up to 188 cm^3. It means, that the quantity of an isotope ^{17}O may be increased in 36,450 times, and the quantity of an isotope ^{18}O may be increased in 7003 times. The density of neutron flux is proportional to quantity of isotopes of oxygen. So, we can calculate the neutron flux density if a mixture of ^{17}O or ^{18}O with deuterium is used. In [12], it was found that intensity of radiation of neutrons is proportional to weight of a sample. Therefore, if the sample weight is increased 10,000 times, i.e. up to 70 kg , then the neutron flux density will be increased by four orders of magnitude. In this case, we have the following results: $\varphi(E)_{\rm cf} = 2.38 \times 10^{13}$ neutron/cm^2 s for ^{17}O and $\varphi(E)_{\rm cf} = 4.57 \times 10^{12}$ neutron/cm^2 s for ^{18}O.

We obtain following values for effective half-life periods after numerical integration of expression (1) on the maximal "burning off" cross sections for high energy neutrons: $T_{1/2{\rm cf}} = 3.07$ years for ^{17}O and $T_{1/2{\rm cf}} = 11.20$ years for ^{18}O. This implies that for a neutron flux density of about 10^{13} neutron/cm^2 s it is necessary irradiate the RWP including ^{137}Cs for 117.5 years.

In the mixture investigated in [8], the ^{15}N content was 0.1875 cm^3. The ^{15}N contents in a mixture with deuterium will be limited to deuterium quantity only. The ^{15}N quantity should be equal to quantity of deuterium at least. Let us calculate neutron flux density for sample of 70 kg weight as it was made for isotopes of oxygen. In this case it is necessary to use the mixture of 4.7×10^7 cm^3. The ^{15}N volume will be 2.35×10^7 cm^3. So, the ^{15}N quantity will increase in 1.25×10^8 times. The neutron flux density will be $\varphi(E)_{\rm cf} = 8.18 \times 10^{12}$ neutron/cm^2 s, and an effective half-life period $T_{1/2{\rm cf}} = 7.484$ years for ^{15}N. Having estimated neutron flux from the source (patent [8]), we can evaluate mean rates of prospective reactions. These are 6.06×10^{-12} 1/s per d + ^{17}O pair, 1.16×10^{-12} 1/s per d + ^{18}O pair and 1.67×10^{-13} 1/s per d + ^{15}N pair.

4. Discussion of Results

The values of neutron flux densities obtained in the above calculations are close to each other in the case of isotopes of oxygen and isotope of nitrogen, but 27 times less than value of density of flux of neutrons for effective "burning off" in a fast reactor. A gas mixture of volume $47\,\mathrm{m}^3$ is necessary to saturate the sample of 70 kg weight. The working volume will be $1.47\,\mathrm{m}^3$ if pressure of the mixture is 32 atmospheres. Also it is possible to increase working pressure up to 50 atmospheres and working volume up to $10\,\mathrm{m}^3$. In this case we can use quite heavy sample 700 kg. We can expect the neutron flux density density to be 10 times greater and the effective half-life of ^{137}Cs will decrease to $T_{1/2\mathrm{cf}} = 0338$ years.

So, it will be necessary to irradiate the RWP with ^{137}Cs during 12.9 years. Similar technical characteristics of the device and technology are close to become real. Improvement of these characteristics can be done due to determination of channels of nuclear reactions by means of spectrometry of neutrons, detection of other products and study of influence of different parameters of the process on mean rates of these nuclear reactions. It should be mentioned, the probabilities of the prospective reactions are far from unity. High-energy α-particles and protons will also give a contribution in the process of "burning off" the RWP. In particular, high-energy α-particles with energy 5.5, 7, 8, 10, 12, and 14 MeV were registered in the paper[13] in result of deuterium desorption from Au/Pd/PdO:D. These results confirm a possibility of realization of similar process in titanium sample.

In conclusion it should be stressed the method patented in [8] is applicable for transmutation of the RWP, in particular, for ^{137}Cs.

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