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Transmuting minor actinides with thermal reactor neutrons

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Abstract

The closure of uranium-based fuel cycle assumes that Uranium and Plutonium are extracted from the spent fuel. Among remaining heavy nuclides, the most radioactive are minor actinides. One of the concepts to reduce the reprocessed fuel radioactivity is the repeated long-term irradiation of minor actinides (this process is referred to as transmutation). There are works stating the necessity of hard spectrum neutrons for irradiation, for which special reactors-transmuters and accelerator-driven systems are to be included into the fuel cycle.

In this work, practicability of minor actinide transmutation in thermal spectrum reactors is considered. The main factor representing the transmutation practicability that is used in this work, is the ratio of radioacitivities with transmutation, and without, $\xi(t)$. This ratio as a function of time can be greater or less than 1, and its values define to the most extent the transmutation practicability. We computed functions $\xi(t)$ for minor actinides. The conclusions are (from the reduction of radioactivity point of view): Neptunium transmutation is hardly necessary, since the order of magnitude reduction in radioactivity is first reached in hundred thousand years; rather small effect (the order of magnitude radioactivity reduction in 500 years) is found for Curium; the Americium radioactivity is reduced by factor of 10–100 in 300 years after irradiation. It is shown that the best values of $\xi(t)$ are reached at 70–08% burnup of minor actinides.

The final conclusion about the practicability of Americium and Curium transmutation must be drawn by taking into account in the considered scenarios the difference in probability of the environmental release, the difference of biological effect and the transmutation efficiency of minor actinides continuously fed to spent fuel storages by the operating nuclear energy industry.

Keywords: Minor actinides; Nuclear fuel cycle; Transmutation; Spent fuel.

Problem definition

A large-scale nuclear energy industry that relies on the natural Uranium is only possible with fast neutron reactors and with the closed fuel cycle. This has been realized for sufficiently long time [1], and was reflected in our country during the last decade in the federal target program devoted to the development of nuclear energy industry with fast reactors and the closed fuel cycle [2].

The radioactivity of spent nuclear fuel exceeds by many orders of magnitude the radioactivity of fresh fuel. During the first hundreds of years, the major contributor to the discharged fuel radioactivity are fission products, and later on – heavy nuclides generated in the core – isotopes of Plutonium and isotopes of the so called minor actinides (MA) – Neptunium, Americium and Curium.

In a closed fuel cycle Uranium and Plutonium are recycled and the minor actinides become in due time the major source of radioactivity.

When isolated from the environment, radioactivity sources are harmless; however there is always a nonzero probability of their release to the environment. In this way a risk of damage arises that is proportional to the level of radioactivity and to the probability of the release. Risks of spent fuel storage are studied in many works [3].

There is another threat – theft of MA with the following assembly of a nuclear explosive device; for example, critical mass of the most abundant Neptunium isotope amounts

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to about 60 kg. If this threat prevails, Neptunium must be destroyed (transmuted) regardless of problems with its radioactivity.

Long-term radioactivity generated in nuclear reactors must be destroyed or safely deposited. Until now, the only and the main way of environmental protection is deposition of spent fuel; its general review is given, for example in the monograph [4].

One of the promising ways to decrease radioactivity of fuel cycle (and, consequently, risks) is based on transmutation. During transmutation, radioactive nuclides are transformed under neutron irradiation into short-lived or stable nuclides (as well as into long-lived nuclides with half-life time above millions of years). Transmutation of fission products is proved to be ineffective in general [5–7]. Transmutation of heavy nuclides in fast neutron spectrum, however, is promising, and gives rise to the idea of radioactive equivalence of handling with radioactive wastes [8–10]. Large-scale transmutation of heavy nuclei to fission products in the present time and in the near decades will be possible only thermal reactors, therefore we consider in this work transmutation only in the thermal neutron spectrum.

Necessity, possibility and practicability of transmutation is a complex multiple-function problem with the radioactivity playing one of the key roles. It should be also noted that damage assessment should take into account differences in probabilities of radioactive nuclide release into the environment from storage and during chemical reprocessing and irradiation in a nuclear reactor. The latter is considerably larger.

As a result of transmutation, radioactivity can considerably increase so that for the current population the risk increases, but for future generations the risk of damage will be lower. Arguments to choose acceptable time intervals and amplitudes of radioactivity grow must be found. Thus, before arguing about the practicability of heavy nuclides transmutation, periods of time must be defined, where risk increase is acceptable.

One of the necessary and sufficient conditions for an accelerated destruction of a radioactive nuclide with decay constant λ in the reaction of neutron absorption can be written as

$$\sigma_a \varphi/\lambda > 1 \tag{1}$$

where σ_a is the nuclide's average absorption cross-section in the neutron flux φ (integrated over energy).

In the problem of radioactivity reduction, condition (1) is necessary but not sufficient. This problem has been realized for a long time and some results of calculations can be found in [4,6]. As a matter of fact, during transmutation nuclides with different decay times are generated and as a result, radioactivity can increase as well.

When heavy nuclides are burned into fission products, the necessary and sufficient condition can be written as

$$\sigma_f \varphi/\lambda > 1 \text{ and } f(t) < \lambda \exp(-\lambda t), \tag{2}$$

where $f(t)$ is the time-dependent radioactivity of fission products, which form depends on the fuel irradiation conditions (irradiation time, neutron flux intensity).

The specified condition implies that the specimen is irradiated continuously with a constant flux.

Let us consider an ideal destruction scheme of particular heavy nuclides. Consider a nuclide that decays into stable products with decay constant λ. We investigate two options: in the first one – the nuclide under consideration is stored in a storage since time $t = 0$; in the second one – the same amount of the nuclide is transmuted into fission products (this assumption is valid when the fission cross-section of the nuclide exceeds considerably its radiative capture cross-section). We represent the radioactivity of fission products by the function $f(t)$ that corresponds to a three-year fuel irradiation in the VVER-1000 core. Fig. 1 shows the ratio of fission products radioactivity to the radioactivity due to the nuclide decay. This ratio ξ we refer to as transmutation efficiency:

$$\xi(t) = 2f(t)/\exp(-\lambda t). \tag{3}$$

Note that the lower $\xi(t)$, the more reasonable transmutation to fission products is, from the point of view of radioactivity, i.e. the better transmutation effect will be reached.

The assumption that the nuclide under consideration decays only in stable nuclei, results in underestimation of $\xi(t)$. The assumption that under neutron irradiation the nuclide undergoes only fission reaction results in underestimation of $\xi(t)$ as well. In other words, the data of Fig. 1 give the lowest possible (the best efficiency) values of $\xi(t)$.

Depending on the heavy nuclides half-life time, the transmutation efficiency as function of time can exceed 1 (transmutation has no sense) or fall below 1 (transmutation has sense). When for all heavy nuclides the radioactivity of fission products exceeds the parent nuclide's radioactivity as irradiation starts ($\xi > 1$), than the essential role plays T_ξ, the time when ξ becomes lower than 1. In the ideal case (see Fig. 1), the value of T_ξ is connected to the heavy nuclide's half-life time. For half-life times from thousand to million years, the values of T_ξ increase from 100 to 500 years.

When considering transmutation practicability of minor actinides – the major contributors to the radioactivity of the spent fuel (Neptunium, Americium, Curium), conditions are more complex as compared to that shown on Fig. 1. In this case, quantitative analysis with the help of numerical modelling is necessary.

Software and results of calculations

We used the ORIGEN2 code [11], modified to take into account fission products of all nuclides, rather than some chosen by default. In particular, the list of fission product parents was augmented with Americium, Curium and Neptunium isotopes. Their fission product radioactivity parameters – time dependence and absolute yield was set to that of ^{235}U.

To investigate the minor actinides transmutation, we followed a small-specimen experiment: ampoules with the initial material are placed into a core with stable irradiation conditions. The time dependence of radioactivity of Neptunium, Americium and Curium elements, irradiated in the VVER-1000 neutron spectrum with intensity of $3.65\,10^{14}$ 1/cm s, and