The main problems of transmutation of actinides and long-lived fission products from NPP spent fuel

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Abstract. The paper formulates the main problems associated with research on transmutation, which should be paid attention to by today's young researchers. The processes of production of hazardous nuclides during transmutation in reactor facilities are considered. The goals of transmutation and the choice of nuclides to be transmuted are discussed. The concept of radiotoxicity is explained as a measure of the radiological hazard of radioactive nuclides, based on the maximum permissible concentration of nuclides according to the IAEA standards. The problem of the formation of secondary radioactive nuclides in nuclear fuel during generation of neutrons for transmutation is discussed. The advantages and disadvantages of various methods of transmutation in nuclear installations are considered: inclusion of transmutable nuclides in nuclear fuel in fast reactors, transmutation in specialized thermal and fast transmutation reactor installations and ADS systems. The problem of accumulation of highly radioactive actinides in a transmutation facility during long-term transmutation and the problem of a potential hazard of the transmutation facility itself are discussed. The unacceptability of application of common-type power reactors for the transmutation of long-lived fission products is demonstrated.

1. Introduction
The development of industrial technologies the world requires an increase in power production. It is assumed that in the future, nuclear power will play a significant role in power production. The future development of nuclear power engineering is associated with the transition to a closed nuclear fuel cycle. It is assumed that the main type of reactors in a closed fuel cycle will be fast reactors [1]. An important part of the closed fuel cycle are processes of radiochemical reprocessing of spent nuclear fuel, manufacturing of new fuel and separation of the part that is considered as radioactive waste. At the current stage of the nuclear power industry, when the closed fuel cycle has not yet been implemented, spent nuclear fuel is accumulated in special storage facilities. The part of spent fuel is reprocessed to separate uranium and plutonium, which can be returned to the fuel cycle. With an increase in the total capacity of nuclear power, the amount of spent fuel and radioactive waste will increase.

An important question that agitates both specialists and the world community is how to deal with radioactive waste?

There are several approaches to the management of spent nuclear fuel and radioactive waste. One of them is nuclear transmutation, designed to reduce the level of accumulated radioactivity. The term "transmutation" means the purposeful process of neutron exposure to long-lived radioactive nuclei, as a
result of which radioactive nuclei are ultimately converted into stable nuclei. This result is achieved through a combination of two types of nuclear reactions. First, by means of neutron capture, radioactive nuclei are converted into other isotopes, which in turn decay into stable nuclei in a relatively short time. Second, heavy radioactive nuclei are transformed into other heavy isotopes through one or several successive captures of a neutron, and a fission reaction occurs in the process of these transformations.

The transmutation of radioactive waste is considered by the world scientific community as an integral part of the future nuclear fuel cycle. It allows the transformation of long-lived radioactive nuclides into stable or nuclides with a shorter half-life, reducing the amount and hazard of waste to be finally disposed of, and easing the requirements for long-term storage. It is important that the transmutation of radioactive waste, showing real ways to reduce their hazard, contributes to the creation of a favorable image of large-scale nuclear power from the point of view of public opinion.

The main hazard of radioactive waste is associated with long-lived fission products and actinides from spent nuclear fuel. Actinide transmutation can be effectively performed in installations based on nuclear reactors. Transmutation of long-lived fission products differs from that of actinides. It requires a large number of neutrons. Reactor installations cannot provide the amount of excess neutrons and neutron fluxes required for the transmutation of even the main long-lived fission products. Accelerator driven systems and thermonuclear installations, in which neutrons are produced not in fission reactions, but in other reactions, can serve as an alternative to reactors.

The active development of nuclear power in our country and around the world is accompanied by the following changes in approaches to the development and design of reactor systems. First, the existing technologies are being improved and new technologies appear which will lead to the possibility of implementing reactors of new types and new reactor technologies, first of all, to the transition to a closed nuclear fuel cycle based on fast reactors. Secondly, the development of computational tools makes it possible to perform calculations in the reactor area with more detailed descriptions of processes and higher accuracy due to the transition to higher-level software tools. This allows transmutation research to be carried out at a modern scientific and engineering level.

In the future nuclear power, the transmutation of produced hazardous nuclides should be an important part of the nuclear fuel cycle. Although the greatest interest in the study of transmutation fell on the previous decades [2], at present, due to the improvement of nuclear technologies and computational facilities, research on individual issues of transmutation is being carried out, including by the staff and students of NRNU MEPhI, at a new, more advanced level.

In this paper, the authors would like, based on the accumulated experience, to formulate the main problems that, to one degree or another, accompany any research on transmutation, and which should be paid attention to by today’s young researchers.

2. Transformations of nuclides in the reactor. Formation of dangerous actinides and fission products

Transmutation cannot be considered as an alternative to underground disposal of radioactive waste. It is assumed that the use of transmutation will reduce the long-term hazard of radioactive waste to a value at which deep underground disposal becomes safe.

What nuclides should be transmuted? How does the formation of hazardous nuclides occur during reactor operation?

In power reactors with a thermal spectrum of neutrons on uranium fuel with low enrichment (PWR), a chain of transformations starting with $^{235}$U leads to the formation of $^{237}$Np and $^{238}$Pu. The main uranium isotope $^{238}$U is the source of formation of all isotopes of plutonium, beginning with $^{239}$Pu. The last significant isotope of plutonium is $^{242}$Pu.

Americium is represented by two isotopes $^{241}$Am and $^{243}$Am. The isotope $^{241}$Am is formed in the reactor by the decay of $^{241}$Pu (half-life 14.4 years). The same process occurs when the spent fuel is placed outside the reactor in a storage facility. Further, the isotope $^{241}$Am in the reactor is ultimately converted into $^{238}$Pu.
The isotope $^{241}\text{Am}$ is formed from $^{242}\text{Pu}$ through the short-lived intermediate $^{243}\text{Pu}$. A small part of it is formed from $^{241}\text{Am}$ through the metastable isomer $^{242m}\text{Am}$. Upon absorption of neutron, $^{242}\text{Am}$ transforms into $^{244}\text{Cm}$ through an intermediate short-lived $^{244}\text{Am}$. A small part of $^{244}\text{Cm}$ is formed from $^{241}\text{Am}$, which, in turn, is formed from $^{242}\text{Am}$.

The isotope $^{244}\text{Cm}$ is a very important nuclide in the actinide chain. It is $\alpha$-radioactive. A relatively short half-life of 18.11 years is responsible for its high activity and radiation hazard in spent nuclear fuel. When decaying, it is transformed into $^{240}\text{Pu}$. The reaction rate of the radiative neutron capture of $^{244}\text{Cm}$ is sensitive to epithermal neutrons. The thermal cross section of the reaction is small. When irradiated in a reactor, it is transformed into $^{245}\text{Cm}$.

An important characteristic of $^{243}\text{Cm}$ is its high fission cross section. However, not all $^{245}\text{Cm}$ nuclei are fissioned in the reactor. The radiative neutron capture cross section is not small although it is significantly less than the fission cross section. Therefore, heavier isotopes of curium $^{246}\text{Cm}$, $^{247}\text{Cm}$, $^{248}\text{Cm}$ are successively formed from $^{245}\text{Cm}$ as a result of neutron capture. The production cross sections for these nuclides are small, and their number decreases with increasing atomic mass. In addition, they can accumulate in appreciable amounts only in the case of repeated multiple irradiation of the initial actinides in the reactor, and this is not so much related to nuclear fuel, but to the transmutation of actinides. With further transformations, isotopes of berkelium, californium, einsteinium, and fermium are formed in very small amounts.

During long-term storage in storage facility, $^{241}\text{Am}$ is formed due to the decay of $^{241}\text{Pu}$, and $^{238}\text{Pu}$ is formed due to the decay of $^{242}\text{Cm}$ through the intermediate $^{244}\text{Cm}$. These nuclides, which were not present in the unloaded fuel, are radioactive to some extent, and their contribution to the overall radiation hazard must be taken into account.

3. Choice of nuclides for transmutation. Purpose of transmutation

So, the most significant hazardous minor actinides that accumulate in fuel are $^{241}\text{Am}$, $^{243}\text{Am}$, $^{244}\text{Cm}$. Among them, $^{241}\text{Am}$ accumulates to the greatest extent, and the main radiological hazard in the initial storage period is created by $^{244}\text{Cm}$. After 100 years of storage, the radiotoxicity of $^{241}\text{Am}$ forms more than 90% of the total radiotoxicity of actinides in storage facility. Attention is now being paid to the priority transmutation of $^{241}\text{Am}$. Fission products in long times, more than 100 years, are much less dangerous than minor actinides. The hazard of fission products is determined by $^{99}\text{Tc}$ and $^{129}\text{I}$. Their transmutation is desirable, but not a primary problem. Those two dangerous nuclides that accumulate in large amounts, namely $^{90}\text{Sr}$ and $^{137}\text{Cs}$, have a half-life of about 30 years and decay completely in about 100 years and do not require transmutation. The rest of the long-lived fission products from the spent fuel decay in less time and also do not require transmutation [3].

What is the purpose of transmutation? The ideal goal of transmutation is to transform hazardous nuclides into harmless ones. But, since this is impossible for various reasons, one have to set realistic goals. Among them: partial transmutation, which reduces the requirements for final geological disposal; involvement of actinides in the nuclear fuel cycle based on their fission; multiple transmutation in specialized reactor facilities. Let’s consider each of them in more detail.

Partial transmutation is performed in specialized solid-fuel transmutation reactors. The purpose of such transmutation before final storage is to incinerate part of the actinides and convert the other part into new actinides, providing a low level of radiation hazard [4]. Partial transmutation should be correspondingly short-term, since the mode of long-term continuous irradiation practically cannot be realized in solid-fuel reactors. It cannot be longer than the operation time of the reactor until the exhaustion of its resource.

The involvement of actinides in the nuclear fuel cycle can be considered primarily due to the fact that fast reactors will be the main type of reactors in a closed fuel cycle. In the fast spectrum, practically all actinides formed during the operation of the reactor from the initial nuclear fuel are fissioned. Therefore, there is reason to believe [5] that the addition of a small fraction of actinides to the main nuclear fuel of fast reactors will lead to their burning.
Multiple transmutation in specialized reactor facilities assumes the construction of a number of such facilities, the only purpose of which will be the transmutation of actinides. Multiple transmutation implies a cyclic transmutation process with unloading, radiochemical processing, adding new actinides, loading for the next transmutation cycle. When the resource of the transmutation installation is exhausted, a new installation must be built to replace it.

4. Measure of radiological hazard. To what extent can one transmute?
At the initial stage of work on transmutation, the mass of nuclides, activity, and energy release were taken as a measure of danger. The most profitable were measures based on biological hazards using the maximum permissible level of external exposure (sievert) or the permissible amounts of nuclides in water or air according to IAEA standards (radiotoxicity).

Radiotoxicity is associated not with external exposure, as a radiation hazard, but with internal exposure of the human body due to the ingestion of radionuclides with inhaled air or drinking water. Depending on the way of ingestion of radionuclides into the body, a distinction is made between an inhalation hazard when ingested by inhalation (radiotoxicity by air) and an oral hazard when ingested with water and food (radiotoxicity by water). They are based on the maximum permissible concentrations of the individual nuclides in the inhaled air or drinking water. These limit concentrations are determined by the Russian norms NRB-99/2009 [6], which are in agreement with the international norms of the International Commission on Radiation Protection. Radiotoxicity by air for a given nuclide is determined as the volume of air in m$^3$, in which this nuclide will be diluted so that its concentration becomes equal to the maximum permissible concentration. Radiotoxicity by water in kg of water is determined similarly.

One of the main questions of transmutation is to what limit should one transmute? There is still no consensus. As an example, in the design of the BREST fast reactor, the concept of radiation equivalence was put forward [7], which states that the radiation hazard of waste that is buried in the ground after transmutation should be approximately equal to the radiation hazard of uranium mined from the ground. In other words, as much danger has been extracted, as much danger has been buried.

5. Consumption of neutrons, nuclear fuel, electricity, formation of new actinides and fission products
The neutrons for transmutation are produced in a nuclear reactor. In this case, new actinides and fission products are formed in the fuel. And this must be taken into account. For example, during the transmutation of long-lived fission products, the process of neutron generation is accompanied by the formation of secondary radioactivity of fission products formed in the fuel of a nuclear reactor. The most significant secondary fission products in terms of radioactivity are $^{90}$Sr and $^{137}$Cs. If we consider only transmutation of the most significant long-lived $^{99}$Tc and $^{129}$I, the resulting secondary activity due to $^{90}$Sr and $^{137}$Cs turns out to be 1500 times higher than the activity of the incinerated $^{99}$Tc and $^{129}$I [8]. However, this result should not be considered as serious restriction to the transmutation of long-lived fission products. The point is that we compare activities of incinerated and newly formed nuclides, of which the new $^{90}$Sr and $^{137}$Cs nuclides are relatively short-lived (30 years). Their activity should be high. The result should be interpreted as follows. For the possibility of incineration long-lived fission products, one has to pay with the accumulation of secondary highly active fission products, which, however, will decay after about 100 years.

Transmutation into accelerator driven systems (ADS) requires separate consideration. For the correct approach to the balance of incinerated and newly produced nuclides when using electricity for transmutation in ADS, this electricity must be accounted for as if it were produced by nuclear power plant. Simply put, when transmuting in a reactor, new secondary nuclides are formed in the same reactor, and when transmuting using ADS, new nuclides are formed in a different place, in the fuel of the nuclear power plant that produced electricity for ADS.
6. Separation of radioactive waste
In the absence of a closed nuclear fuel cycle, one of the preferred methods for storing spent fuel is to store it in the form of spent fuel assemblies without any reprocessing. If it is necessary to reuse spent fuel, radiochemical reprocessing is required. In this case, the separation of radioactive waste is a necessary stage of processing. Separation of radioactive waste is useful for the following reasons. Different groups of nuclides have different level of danger and require different storage conditions. Part of the nuclides can be sent to the production of secondary fuel. Those nuclides that are directed for transmutation may require a different spectrum and neutron flux. However, it should be keep in mind that separation, as with any radiochemical processing, generates a large amount of low-level waste.

7. Types of nuclear installations for transmutation
The first candidates are now fast power reactors with the inclusion of transmutable actinides in the composition of nuclear fuel. The advantage of a fast spectrum is that almost all intermediate actinides in transformation chains undergo fission. The disadvantage is that the reaction rates are low, and this leads to large equilibrium amounts of intermediate actinides. In addition, despite fission, there are still radiation capture reactions, due to which a slow accumulation of distant actinides occurs. In addition to being included in nuclear fuel, transmuted nuclides can be placed in fast reactors in the form of separate targets in special moderating zones, in which a thermal spectrum for transmutation is created locally.

Specialized transmutation reactors can be fast reactors and thermal reactors. The thermal spectrum at high flux gives high reaction rates due to large cross sections. However, the chains of nuclide transformations are longer due to the fact that not all nuclides undergo fission.

In molten salt reactors, transmuted actinides can be homogeneously incorporated into the composition of nuclear fuel.

A separate type of transmutation units are ADS - accelerator driven systems with proton accelerator. Their main advantage is higher safety. They can contain zones with different neutron spectra. The disadvantage is the need for a high-current proton accelerator with proton energy of about 1 GeV and a neutron-producing target.

8. Radiation hazard contained in transmutation installations
When the initial transmuted actinides are irradiated with neutrons in a reactor or other transmutation installation, other radioactive nuclides are formed, the next are formed from them along the chain of nuclide transformation, etc. As a result, during prolonged irradiation with regular feed with new actinides, a certain stationary mode of operation is established. This mode is characterized by a stationary isotopic composition of actinides and their radiation hazard. In addition, nuclear fuel must be consumed to support such a process. The stationary mode, as a rule, is characterized by a higher radiation hazard than the loaded actinides [9].

The amount of actinides in a stationary mode depends on the absolute rates of reactions. In the fast spectrum, the stationary amount of hazardous actinides is greater due to low reaction rates, and the radiation hazard is greater. In the thermal spectrum with a high neutron flux, the reaction rates are higher, and the radiation hazard in the stationary mode is lower. However, it should be remembered that the isotopic composition in the stationary mode in the fast spectrum differs from the isotopic composition in the thermal spectrum, because not all actinides are fissionable in the thermal spectrum.

The most important thing is that the transmutation facility can be considered as a kind of repository of very dangerous actinides, not only the initial ones, but also all others that make up the stationary isotopic composition. Moreover, all of them are kept in conditions of high power intensity, typical for nuclear reactors. Therefore, transmutation facilities have a high degree of potential danger. This distinguishes a transmutation reactor from a storage facility for long-term storage of radioactive waste under controlled and less dangerous conditions.
9. Application of common-type power reactors and specialized reactors for transmutation

Targets with transmutable actinides or fission products can be placed in the core with minimal modifications of fuel assemblies. The addition of actinides to common-type power reactors may be undesirable due to changes in reactor operating conditions and safety problems. The addition of fission products for their transmutation leads to decrease in power production due to the fact that part of the fuel assembly is occupied by targets, and part of the neutrons is spent on transmutation. Thus, when simulating the transmutation of $^{99}$Tc and $^{129}$I in VVER-1000 with the rate of their production in the reactor, the power production decreases by 10-13% [10]. Thus, the economic efficiency of the reactor is significantly reduced, which can cause a negative attitude from power generating companies.

Specialized reactors for transmutation can be both high-flux thermal and fast reactors. Their advantage is that they do not have the task of producing power. Therefore, they can operate in less power-intensive conditions. Their disadvantage is the consumption of nuclear fuel and the production of new actinides in it. In the case of fast reactors, the transmuted actinides themselves can serve as nuclear fuel [11].

10. Conclusions

It should be noted that all works on transmutation have the character of scientific research works. The accelerator for ADS has not yet been created, the problems with the neutron-producing target have not been solved. It is not clear how long it will take before the actual creation of at least a demo installation. During this time, new sources of neutrons, for example, thermonuclear ones, may be developed, and it may turn out that they are more suitable for transmutation.

In conclusion, it should be said that this paper is addressed primarily to specialists starting their path in the field of nuclear power engineering, it briefly formulates the main problems that today's young researchers should pay attention to.

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