Prospective methods for optimization of minor actinides transmutation technology

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Abstract. Among the various international policies for the management of spent nuclear fuel and radioactive waste (RW), an important issue is the need to reprocess this waste. The aim of this work is to analyze current modern methods for transmutation of minor actinides (MA) in the reactor cores of thermal, fast (with homogeneous and heterogeneous placement) and in accelerated driven systems in order to identify factors limiting the efficiency of transmutation. This work is an analytical review and it defines the main methods for removing the limitations of the effectiveness of the transmutation process. On the basis of MA formation sources data and MA decay chains, namely americium, neptunium and curium, as well as transmutation methods in the latest reactor systems, factors limiting the efficiency of the transmutation process and methods of reducing their influence are investigated. The main factors are: an increased level of gassing during transmutation, industrial restrictions and the influence of the transmutation process on the processes occurring in the core during transient processes. The main current problem of transmutation is the high activity in terms of heat release level and neutron emission of irradiated assemblies due to the presence of curium isotopes.

1. Introduction
Since the beginning of the 21st century, nuclear energy has spread widely throughout the world, due to the high environmental friendliness of nuclear power plants, provided that they are operated without accidents. However, the widespread use of nuclear power leads to the need for a large volume of R&D and implementation of expensive technological projects related to the reprocessing of spent nuclear fuel (SNF). Spent nuclear fuel of a nuclear power plant is one of the types of high-level radioactive waste, which includes fuel assemblies with fuel elements removed from the reactor core.

Among the various international policies for the management of spent nuclear fuel and radioactive waste (RW), an important issue is the need to reprocess this waste. Within the framework of this issue, the analysis of current factors and features limiting the efficiency of transmutation of minor actinides (MA) in the cores of thermal reactors, fast reactors (with homogeneous and heterogeneous placement) and in accelerated driven systems is carried out.

2. Policy on SNF and RW management
One of the main factors influencing the current state of scientific research in the field of SNF and RW reprocessing, including MA transmutation, is the adopted policies for SNF and RW management in different countries. The issue of processing radioactive waste and spent nuclear fuel affects many related
industrial, environmental and economic issues. Further in the work, statistical and reporting data on the level of accumulation of spent nuclear fuel and radioactive waste both in Russia and around the world are presented. Based on these data, the problem and the reasons for the need for R&D in this area are updated.

2.1. SNF and RW management policy in the world
Countries around the world have different policies related to the reprocessing of spent nuclear fuel. In particular, in countries such as the USA, Canada, Sweden, Spain, Finland and some others, spent nuclear fuel is classified as radioactive waste (RW) and is either subject to direct disposal without reprocessing, or is subject to long-term storage. Based on the analysis of recent reports from the member countries of the OECD-NEA and the IAEA [1-10], it can be concluded that at the moment only Great Britain, France, Japan and Russia are engaged in the processing of spent nuclear fuel on an industrial scale. Also, we can conclude that the value of the projected SNF volumes is growing every year.

2.2. SNF and RW management policy in Russia
According to the public annual report «Results of the activity of the State Atomic Energy Corporation «Rosatom» for 2017 and 2018 [11,12,13], the volume of accumulated spent nuclear fuel in Russia is increasing by almost a thousand tons. Figure 1 shows a graph of SNF accumulation in the Russian Federation in accordance with [13]. At the same time, only a small amount of spent nuclear fuel was reprocessed, as can be seen from the graph in figure 2.

Figure 1. Accumulation of spent nuclear fuel in the Russian Federation (tons) [13].

Figure 2. SNF removal for storage and processing (tons) [13].
2.3. Justification of the need for R&D to improve the efficiency of SNF and RW processing

When analyzing the data presented above, it can be seen that the amount of spent nuclear fuel and radioactive waste is growing every year. The policy for the disposal of spent nuclear fuel or radioactive waste not only leads to the need for the construction and maintenance of waste storage facilities in deep geological formations (or storage facilities of other types), which leads to necessary and constantly increasing economic costs, but also creates a danger for future generations. This danger is associated with the possible leakage of radioactive isotopes into groundwater or surface water bodies and, subsequently, the ingress of highly active nuclides into living organisms, including humans, which will lead to a large-scale environmental and health disaster. The likelihood of such events is extremely low, but the composition of radioactive waste disposed of in storage facilities is such that its half-life reaches up to millions of years, which places a burden on hundreds of thousands of future generations. Due to such a long storage period of waste, even the slightest possibility of violation of the conditions of detention becomes relevant. Consequently, R&D activities are needed to reduce the half-life of disposed radioactive waste to the shortest possible time.

Taking into account the low volume of reprocessing of the total volume of annually produced spent nuclear fuel, with unchanged technological capacities of reprocessing plants, there remains only the scenario of increasing the efficiency of the transmutation process. This increase in efficiency can be achieved through reducing the influence of limiting factors on the transmutation process. Let's consider these limiting factors in more detail.

3. Features of transmutation of minor actinides

When considering a closed nuclear fuel cycle, when plutonium is reprocessed many times in fast reactors, minor actinides, namely americium, neptunium and curium, make a major contribution to the long-term radiotoxicity of spent nuclear fuel after aging for several centuries. On this basis, these three nuclides will be considered as reference MA in the future. In this work, radiotoxicity means the ability of a radioactive substance to cause radiation damage. This term was adopted in this work, due to the wide use of this indicator in many foreign scientific works, including in the International Commission on Radiological Protection, data from the reports of which will be used further.

4. Factors limiting the efficiency of minor actinides transmutation

During the analysis of various works, three main types of limiting factors were identified. These factors are associated with the level of gas production during the operation of a nuclear reactor with MA loading into the core, with the influence of MA loading into the core of a nuclear reactor on its behavior during some transient processes, and the third group of factors is associated with the limitation on the level of heat release and neutron power of irradiated assemblies. The first of the factors considered in this work is the limitation on the level of heat release and neutron power of radioactive waste and spent nuclear fuel for transportation and processing.

4.1. Limitation on the level of heat release and neutron radiation for irradiated assemblies with minor actinides loading

Reprocessing of blanket assemblies containing minor actinides is difficult mainly due to the high heat release level and powerful neutron radiation from irradiated blanket assemblies, which are significantly higher than that of conventional fuel assemblies, due to the high activity of minor actinides. In order to analyze the impact of irradiated blanket assemblies on the nuclear fuel cycle, their heat release level will be compared with various manufacturing constraints based on the calculated mechanical and thermal characteristics of the various handling devices calculated by the CEA and also used in [14], namely:

- Heat release limit of 40 kW for work with assemblies inside the reactor vessel;
- A hypothetical upper limit of 7.5 kW for the removal of residual sodium in an assembly prior to storage under water;
• A hypothetical lower limit of 2.5 kW for sodium flushing, which is consistent with the use of improved overhead flush technology.

The neutron radiation rate can affect the dose level when working with transport containers or in processing facilities, as well as affect the radiolysis of the solvent during processing. However, these considerations are highly dependent on the industrial approach used.

The analysis of these factors was carried out in several studies with homogeneous and heterogeneous placement of MA in the core. To analyze the heterogeneous approach, the design of an assembly with a bundle of fuel rods with a fuel volume fraction of 38.6% and oxide fuel with 80% depleted uranium and 20% loading of minor actinides will be considered.

When considering the results of calculations with a heterogeneous approach, it should be taken into account that neptunium has a lower heat release of decay, since it is formed mainly from PU-238, which has a low specific heat of decay. Therefore, the target assembly can be reprocessed without any restrictions (if we assume a 40 kW limit for short-term handling). Likewise, for the 7.5 kW limit, with several weeks required to reach this limit. However, if this limit is lowered to 2.5 kW, the cooling time will increase to over 60 years due to the relatively long half-life PU-238 (87 years). The graph of the change in the level of heat release for the FAs with the curium, americium and neptunium loadings with the cooling time in the heterogeneous approach is shown in Figure 3. A similar graph for the neutron radiation power of the assemblies is shown in Figure 4.

![Figure 3. Change in heat release level of spent target assemblies loaded with neptunium, americium, and curium.](image-url)
Figure 4. Change in the neutron radiation power of spent target assemblies loaded with neptunium, americium and curium.

The main conclusion that follows from these data is that neptunium is not a problem for the end stage of the nuclear fuel cycle, while a fresh assembly with curium is almost thirty times "hotter" than an assembly with americium, while its neutron emission level is equivalent to that at a freshly irradiated American target. Transporting fresh assemblies from blankets containing curium seems to be impossible compared to transporting assemblies from blankets containing americium. With regard to neutron radiation, as shown in Fig. 4, targets loaded with neptunium do not emit neutrons, since they do not contain neutron source nuclei. On the other hand, since CM-244 and heavier isotopes of curium are the main sources of neutrons in blankets, it is obvious that the most powerful neutron radiation is observed in the case of loading curium. With this load, the neutron radiation from the assembly after five years of cooling is almost fifty times higher than that of a standard MOX assembly, making it much more difficult to handle. As in the case of heat release level, the limitations on the power of the neutron source should also limit the total mass of curium that can be loaded.

4.2 Restriction on the gassing level in assemblies with loading of minor actinides during operation of a nuclear reactor

The next limiting factor considered relates to gas production during irradiation, which plays a significant role in the design of blanket assemblies [15]. To assess the sources of gas formation, a diagram of the sources of gas formation during MA transmutation was prepared, shown in Figure 5.
Figure 5. Sources of gas formation during MA transmutation.

In [16] work, CEA researchers have obtained numerical estimates of the level of gas formation calculated for a homogeneous model of the core of the SFR V2B reactor with a capacity of 3600 MW when loaded with Np, Am and Cm [16]. These results are shown in Table 1.

Table 1. Gassing levels when loading different minor actinides, calculated in operation [16].

| Gas production       | Neptunium loading | Americium loading | Curium loading |
|----------------------|-------------------|-------------------|----------------|
| Gases – fission      | 0.99              | 0.84              | 3.35           |
| products (cm$^3$/g) |                   |                   |                |
| Helium (cm$^3$/g)   | 0.26              | 4.19              | 3.83           |
| Total (cm$^3$/g)    | 1.25              | 5.02              | 7.18           |

As can be seen from Table 1, the gas production in the case of neptunium loading is relatively small compared to the cases of americium or curium loading. In general, the volume reserve of the assembly structure for curium loading case is more limited due to the increased volumes of gas formation. In the case of neptunium loading, gas formation is not a design problem. Thus, the design of assemblies for blankets containing curium is likely to be further limited by the need to account for increased gas production, which would require limiting the fuel volume fraction and increasing the height of the lower and upper expansion chambers. Thus, this factor primarily limits the efficiency of curium transmutation.

Limitation associated with the influence of the loading of minor actinides on the behavior of the core during transient processes in a nuclear reactor.

The third limiting factor considered in this work will be the influence of the loading of minor actinides on the behavior of the core during transient processes. The most widely studied processes in terms of the influence of minor actinides were selected as the considered transient processes. In addition, these transients have been chosen because they are considered traditional, in the sense that they represent...
hypothetical situations of high damage and are thus a good measure of the core behavior during any such transients. In total, three transition processes were chosen, namely:

- unprotected loss of coolant (ULOF) nuclear reactor accident, which corresponds to stopping the main pumps without inserting control rods;
- unprotected loss of heat sink (ULOHS) accident, which corresponds to the shutdown of the auxiliary pump, but while maintaining the operability of the main pumps, effectively carrying out the heat removal of the core without introducing control rods;
- unprotected rod run-out transient over power (UTOP) accident, that corresponds to an increase in reactivity in the core. In this case, researchers usually consider a slow increase in reactivity due to a malfunction of the control element drive mechanism and the subsequent removal of the control element without introducing others.

Investigating scientific works [17-20] on the effect of different loads of minor actinides on the behavior of the core during various transient processes, we can draw some conclusions. Americium has a positive effect on the ULOHS transient, practically does not affect the ULOF, and has a significant effect on the UTOP transient. Neptunium behaves similarly with americium in this respect. This is consistent with the influence of americium and neptunium loading on integral feedback coefficients. More interesting is the behavior of the core with curium loading, where performance degradation in the ULOHS and ULOF processes is observed along with a smaller decrease in performance during UTOP and with a limited change in integral feedback coefficients. This is consistent with the actual increase in the Doppler effect caused by the inclusion of curium in the fuel. From this analysis, it can be assumed that the production of curium during irradiation has a positive effect on the behavior of the core during transient processes, as it behaves as fissile material and eliminates some of the restrictions imposed by the americium loading. However, it seems that the influence of the loading of minor actinides on the core, considered here, is very small.

5. Ways to reduce the influence of limiting factors on the process of minor actinides transmutation in a nuclear reactor

The first restrictions considered in this work were related to the level of heat release level and the level of neutron radiation power in irradiated assemblies loaded with minor actinides. Since these restrictions are associated with problems during the transportation and processing of spent nuclear fuel, they can be designated as “industrial restrictions”. Based on the scientific work associated with this limiting factor [14], the following technologies have been developed or confirmed when handling fresh assemblies. Since these restrictions mainly concern assemblies with curium, it is necessary to pre-cure blankets with curium loading before transportation, which is currently being carried out by most enterprises. Technologies for separating curium in high-level assemblies are being developed. The most effective way to reduce the influence of this limiting factor on the transmutation process is to limit the total mass of curium loaded into the target assembly.

The limiting factor associated with the level of gassing has been studied in several works [21]. The main result of these studies can be considered the developed recommendations for changing the design of target assemblies for blankets containing curium, since they are additionally limited by the need to take into account increased gas production, which requires either limiting the volume fraction of fuel in an assembly with MA loading, or increasing the height of the lower and upper expansion chambers.

Since the study of the influence of the loading of minor actinides on the behavior of the core during transient processes of the core was mainly carried out by the CEA [16], most of the recommendations were developed by the researchers of this organization. Specifically, these recommendations are to limit the amounts of MA and to use a homogeneous transmutation approach, which does not entail a significant negative impact on the behavior of the AZ during transient processes.
Conclusion
As a result of the analysis of the limiting factors of the transmutation process, taking into account the peculiarities of modern methods of transmutation, it is possible to draw some conclusions about the preferred method of transmutation of specific actinides. Regardless of the chosen transmutation technique, the main efficiency limitations remain factors associated with the nuclear fuel cycle. Thus, neptunium is the best candidate for heterogeneous transmutation in fast reactors, its irradiated assemblies are the least active and do not emit neutrons, and gas production is minimal. Unfortunately, the transmutation of neptunium in a thermal reactor is ineffective. Americium shows high efficiency of transmutation in thermal reactors, while possessing a sufficiently high level of heat release from irradiated assemblies. Curium is the worst candidate for transmutation in fast reactors. Curium possesses a high level of thermal load, level of neutron radiation and gassing of fresh and irradiated assemblies.

The main current problem of transmutation is the high activity in terms of heat release level and neutron emission of irradiated assemblies due to the presence of curium isotopes. To carry out the radiochemical reprocessing of spent nuclear fuel, cooling is required for many years, which extremely limits the industrial potential of reprocessing. The main task of researchers at the moment is to find a way to separate curium from other minor actinides in SNF, taking into account the possible effect of curium on transportation processes (high heat release level) and possible radiolysis of chemical components during radiochemical processing (high level of neutron radiation).

The possibility of combining work with other modern and relevant research presented in the works is being considered [22-26].

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