ON NECESSITY OF CREATION OF ACCELERATOR DRIVEN SYSTEM WITH HIGH DENSITY OF THERMAL NEUTRON FLUX FOR EFFECTIVE TRANSMUTATION OF MINOR ACTINIDES

A.S. Gerasimov, G.V. Kiselev, L.A. Myrtsymova
State Scientific Centre of the Russian Federation
Institute of Theoretical and Experimental Physics (RF SSC ITEP)
25, B. Cheremushkinskaya, 117259 Moscow, Russian Federation

Abstract

The opportunity for using a high thermal neutron flux for the effective incineration of fission products and minor actinides is studied in this paper. Incineration rates of main long-lived fission products in various fluxes are presented. An efficiency of actinide transmutation is studied for three types of installations: thermal power reactor with neutron flux $5 \cdot 10^{13}$ neutr/(cm^{2}s), fast neutron power reactor with neutron flux $5 \cdot 10^{15}$ (neutr/cm^{2}s), and homogenous heavy-water blanket of ADS with thermal neutron flux $5 \cdot 10^{15}$ neutr/(cm^{2}s).

1. Introduction

The role of accelerator driven systems (ADS) in promising atomic power engineering can be very important. It is mainly explained by the fact that they decrease the amount of long-lived radioactive waste. Specific properties of ADS permit to use different kinds of nuclear fuel such as uranium, plutonium, thorium, minor actinides (and their combinations), and then to use various nuclear fuel cycles. In ADS, long-lived fission products and actinides can be effectively transmuted.

Characteristics of transmutation process depend on parameters of the transmutation installation. The main characteristics are neutron flux and spectrum and excess of neutrons, which can be used for transmutation. High thermal neutron fluxes make possible to obtain high transmutation rates and provide low loads of incinerated nuclides in the installation. ADS can provide also rather high excess neutrons for transmutation. Unfortunately, there is no possibility to increase significantly the neutron flux in fast neutron spectrum installations. However, fast neutron spectra could be preferable from the view point of neutron balance.

The application of high-flux ADS for transmutation of long-lived fission products and actinides is studied in this paper.

2. Fission product transmutation

Experts of different institutions came to an agreement about a list of long-lived fission products that should be transmuted rather than ultimately stored. The main nuclides are ^{99}Tc and ^{129}I. Their half-life is too long, and accumulation in spent nuclear fuel is rather high. Other radioactive fission products either could not be transmuted effectively or have a low importance.

In Table 1, masses (in grams) of nuclides for transmutation of ^{99}Tc are presented for irradiation in constant neutron flux $\Phi = 10^{14}$ neutr/(cm^{2}s) and spectrum hardness $\gamma = 0.4$ typical for a light water blanket of an ADS (spectrum hardness is the share of epithermal neutrons in neutron flux). Data are normalised by 1 gram of initial ^{99}Tc. In Table 2, the same data are presented for $\Phi = 10^{15}$ neutr/(cm^{2}s) and spectrum hardness $\gamma = 0.1$ typical for heavy water blanket of ADS.

In Tables 3 and 4, analogous data for transmutation of ^{129}I are given. They are normalised by 6.35 grams of initial ^{129}I and 1.40 gram of initial ^{127}I. Iodine isotopes are accumulated in spent fuel of light water power reactors in those amounts on account of power produced 1 MW-year.

Table 1. **Transmutation of ^{99}Tc with $\Phi = 10^{14}$ neutr/(cm^{2}s) and $\gamma = 0.4$**

T, year	^{99}Tc	^{100}Ru	^{101}Ru	^{102}Ru	^{103}Rh	^{107}Pd
0	1	0	0	0	0	0
1	6.11-1	3.83-1	5.94-3	2.85-4	4.89-7	2.14-14
2	3.74-1	6.05-1	1.93-2	1.95-3	5.23-6	4.15-12
3	2.28-1	7.30-1	3.57-2	5.68-3	1.85-5	8.25-11
4	1.40-1	7.96-1	5.26-2	1.16-2	4.25-5	6.49-10
5	8.53-2	8.26-1	6.86-2	1.98-2	7.77-5	3.09-9

Table 2. **Transmutation of ^{99}Tc with $\Phi = 10^{15}$ neutr/(cm^{2}s) and $\gamma = 0.1$**

T, year	^{99}Tc	^{100}Ru	^{101}Ru	^{102}Ru	^{103}Rh	^{107}Pd
0	1	0	0	0	0	0
0.1	8.43-1	1.55-1	1.52-3	2.19-5	2.56-8	2.37-17
0.2	7.11-1	2.83-1	5.65-3	1.65-4	3.39-7	5.37-15
0.5	4.26-1	5.43-1	2.83-2	2.18-3	8.08-6	5.83-12
1	1.82-1	7.25-1	8.00-2	1.33-2	6.63-5	9.08-10
2	3.31-2	7.29-1	1.70-1	6.56-2	3.86-4	1.02-7

Table 3. **Transmutation of ^{129}I with $\Phi = 10^{14}$ neutr/(cm^{2}s) and $\gamma = 0.4$**

T, year	^{129}I	^{130}Xe	^{131}Xe	^{132}Xe	^{133}Cs	^{134}Cs	^{135}Cs	Ba
0	6.35	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1	5.57	7.40-1	2.51-2	1.32-2	2.26-5	2.31-6	2.12-7	6.08-7
2	4.89	1.32	6.55-2	7.65-2	2.44-4	4.05-5	7.97-6	2.33-5
3	4.29	1.77	1.02-1	1.94-1	8.60-4	1.79-4	5.57-5	1.68-4
4	3.77	2.11	1.32-1	3.57-1	1.95-3	4.61-4	2.00-4	6.19-4
5	3.30	2.36	1.54-1	5.55-1	3.50-3	8.98-4	5.05-4	1.61-3

Table 4. **Transmutation of ^{129}I with $\Phi = 10^{15}$ neutr/(cm^{2}s) and $\gamma = 0.1$**

T, year	^{129}I	^{130}Xe	^{131}Xe	^{132}Xe	^{133}Cs	^{134}Cs	^{135}Cs	Ba
0	6.35	0.0	0.0	0.0	0.0	0.0	0.0	0.0
0.1	5.76	5.60-1	2.09-2	4.08-3	2.90-6	1.03-7	8.31-9	2.61-8
0.2	5.23	1.02	6.64-2	2.75-2	3.85-5	2.21-6	3.73-7	1.18-6
0.5	3.92	1.94	2.22-1	2.69-1	8.94-4	7.92-5	3.71-5	1.22-4
1	2.42	2.48	3.64-1	1.11	6.57-3	1.66-3	1.63-3	6.91-4
2	9.21-1	2.05	3.40-1	3.09	2.85-2	9.37-3	2.12-2	1.24-2

These data show that in a middle-range neutron flux, the rate of ^{99}Tc and ^{129}I transmutation is not high. In transmutation of ^{99}Tc, isotopes of Ru, Rh and Pd are produced, while in transmutating ^{129}I, isotopes of Xe, Cs and Ba are produced. High flux provides much higher rate of transmutation.

3. Actinide transmutation

As for MA incineration, there are various viewpoints concerning the most preferable neutron spectrum. The choice of MA transmutation conditions is important to define a type of reactor or ADS installation. From this point of view, it is convenient to compare an efficiency of actinide transmutation in different facilities. In Table 5, calculated characteristics of transmutation modes in 3 types of transmutation facilities are presented: thermal power reactor PWR with neutron flux $5\cdot10^{13}$ neutr/(cm^{2}s), fast neutron power reactor with neutron flux $5\cdot10^{15}$ neutr/(cm^{2}s), and homogeneous heavy-water blanket of ADS with thermal neutron flux $5\cdot10^{15}$ neutr/(cm^{2}s). These data were obtained by experts of ITEP and

MEPI within the framework of Project of ICST #17 [1]. Two types of feed by actinides were considered: total 39 kg/year with isotopic composition 45% ^{237}Np, 44% ^{241}Am, 8.5% ^{243}Am, 2.2% ^{244}Cm, and total 83 kg/year with isotopic composition 57% ^{241}Am, 30% ^{243}Am, 11.5% ^{244}Cm. Note that second feed is typical for actinides from spent uranium-plutonium fuel.

Table 5. **Transmutation characteristics**

Type of installation	**Thermal neutrons**		**Fast neutrons**		**High flux ADS**	
Neutron flux, neutr/(cm^{2}s)	$5{\cdot}10^{13}$	$5{\cdot}10^{13}$	$5{\cdot}10^{15}$	$5{\cdot}10^{15}$	$5{\cdot}10^{15}$	$5{\cdot}10^{15}$
Feed by actinide, kg/year	39	83	39	83	39	83
Time of equilibrium, year	50	50	40	40	0.5	0.5
Equilibrium actinide mass, kg	700	2 300	880	2 100	2.7	2.3
Equilibrium respective radiotoxicity	1	3.1	1.6	3.4	0.03	0.1
Reference time, year	250	170	400	200	4	2

The main feature of MA transmutation is that the radiotoxicity of incinerated actinides is increased in the initial period of irradiation because of higher radiotoxic nuclide production. After some period, equilibrium is established. In equilibrium, the rate of actinide incineration is equal to actinide feed. In the installation, there is an equilibrium actinide mass and radiotoxicity. The time of equilibrium achievement is almost the same both for thermal and fast neutron reactors. The high thermal flux installation has an obvious advantage, because the time of equilibrium achievement is significantly (up to 100 times) shorter than for other types of installation. Radiotoxicity is presented in respective units, it is normalised by initial value for thermal neutron installation for a first type of feed. Equilibrium actinide mass and radiotoxicity is also much more preferable for high flux ADS installation, it is 40-50 times less than for common-type thermal or fast neutron installation.

An interesting parameter is the reference time. It is a parameter for comparison of processes of transmutation and storage. If actinides are located into transmutation installations with constant rate (feed) then in some time equilibrium radiotoxicity in the installation is established. If actinides are located in storage with the same rate then radiotoxicity of actinides in storage uniformly increases. Reference time in Table 5 is a time when a radiotoxicity would be equal to that of in transmutation installation. So, for common-type installation, reference time makes 200-400 years. It means that during 200-400 years, simple actinide storage with uniform addition in storage facility gives us less radiotoxicity than that of established in transmutation installation. However, transmutation in high flux ADS installation will be preferable after 2-4 years of transmutation.

Table 6. **Respective radiotoxicity in irradiation of plutonium**

T, days	**10^{14} neutr/(cm^{2}s)**	**10^{15} neutr/(cm^{2}s)**
0	1	1
100	1.9	5.4
200	2.0	14
300	1.8	18
500	2.1	13
700	2.7	7.1
1 000	5.4	2.2

Another important problem concerning to radiotoxic actinide accumulation is weapon-grade plutonium utilisation. In Table 6, respective radiotoxicity of actinides in process of almost pure ^{239}Pu irradiation is presented. Neutron flux 10^{14} and 10^{15} neutr/(cm^{2}s) and neutron spectrum of heavy-water ADS blanket are considered. There is an increase of radiotoxicity during a long period of irradiation. It is caused in an initial period by accumulation of ^{241}Pu and then by accumulation of ^{244}Cm. A major part of radiotoxicity is produced by ^{244}Cm. Maximal radiotoxicity is 18 times higher than that of initial plutonium.

4. Conclusion

The data presented show that both long-lived fission products and actinides can be successively transmuted in ADS installations. For transmutation of fission products, a most important feature of ADS is high amount of excess neutrons, which can be captured in transmuting nuclides. Low equilibrium amount nuclides does not play so important role as in transmutation of actinides.

For actinide transmutation, a high flux ADS installation is needed. The low equilibrium radiotoxicity of incinerated actinides and high level of nuclear safety preventing reactivity accident are the main features of ADS important for transmutation facility.

REFERENCES

[1] B. Bergelson *et al.*, *Transmutation of Minor Actinides in Different Nuclear Facilities*. Proceedings of the International Workshop “Nuclear Methods for Transmutation of Nuclear Waste”, Dubna, Russian Federation, 29-31 May 1996. World Scientific Publishing Co., 1977, pp. 67-76.