INIS-FR--396

FR0108167

HIGH FLUX TRANSMUTATION OF FISSION PRODUCTS AND ACTINIDES

A. GERASIMOV, G. KISELEV, L. MYRTSYMOVA, SSC RF ITEP

25, B.Cheremushkinskaya, Moscow 117259, Russia

Key words: Spent Fuel, Radwaste, Transmutation

Long-lived fission products and minor actinides accumulated in spent nuclear fuel of power reactors comprise the major part of high level radwaste. Their incineration is important from the point of view of radwaste management. Transmutation of these nuclides by means of neutron irradiation can be performed either in conventional nuclear reactors, or in specialized transmutation reactors, or in ADS facilities with subcritical reactor and neutron source with application of proton accelerator. Different types of transmutation nuclear facilities can be used in order to insure optimal incineration conditions for radwaste. The choice of facility type for optimal transmutation should be based on the fundamental data in the physics of nuclide transformations.

Nuclides which must be transmuted

The most dangerous long-lived fission products of spent nuclear fuel are Sr-90 and Cs-137 with half-life of about 30 years. They are accumulated in significant amounts in spent fuel of power reactors. They will remain of radiological danger during a hundred years because of high radioactivity. Other long-lived fission products Tc-99, I-129, Se-79, Zr-93, Pd-107, Cs-135, Sn-126 have a half-life in the range of 10^5 to 10^7 years. Nuclides Tc-99, Zr-93, Cs-135 have maximum or close to maximum yield per fission of nuclear fuel. I-129 is accumulated in smaller amounts and its danger is mostly due to the ability to migrate and to be accumulated in human body.

Sr-90 and Cs-137 have very low neutron cross sections. Extremely high neutron fluxes would be required for their incineration with a rate close to the rate of decay. Therefore, transmutation of Sr-90 and Cs-137 is ineffective in existing conventional facilities or in those which can be envisaged in the near future. These nuclides could be stored in long-term controllable storage during several hundred years till almost complete decay. Among other nuclides, Tc-99 and I-129 should be transmuted primarily because of high yield of Tc-99 and danger of I-129. Their neutron cross sections are in the range of several dozens barns, and these nuclides could be incinerated in reactor or ADS facility with middle-range or high neutron flux.

Other long-lived fission products Se-79, Zr-93, Pd-107, Sn-126 are less dangerous from radiological point of view. They require huge number of neutrons to be transmuted. Therefore, their transmutation cannot be considered a target for the near future.

Minor actinides accumulated in spent fuel are much more dangerous than long-lived fission products. And it is rather difficult to indicate more or less dangerous nuclides as their respective role varies during the long-term storage. Actinides Pu-241, Pu-238, Cm-244 are most dangerous during dozens of years after spent fuel unloading. Both Pu-241 and Cm-244 decay almost completely in about 200 years, while Pu-238 decays in 1000 years. Am-241 is most dangerous during 3000 years after fuel unloading. After that Pu-240 and Pu-239 become most dangerous. Therefore, all minor actinides have to be transmuted.

Transmutation of Tc-99 and I-129

The process of fission product transmutation is rather simple because radioactive nuclide is transformed in stable nuclide by capture of one neutron. However even transmutation of Tc-99 and I-129 presents certain problems. The efficiency of transmutation can be estimated from the neutron balance. The efficiency is calculated as mass of nuclide transmuted during one year per 1 GW of power of transmutation facility. Tc-99 is the only long-lived isotope of technetium extracted from spent fuel. Stable iodine isotope I-127 is found together with I-129 in long-lived fission products. Therefore, the number of neutrons required for incineration of one nucleus of Tc-99 is about 1.1-1.2 as some of the neutrons are captured in products of Tc-99 transmutation. 1.3-1.4 neutrons are required for incineration of I-129 as additional neutrons are captured in I-127 which is irradiated together with I-129. The main characteristic of a certain transmutation facility is the number of free neutrons which could be used for transmutation. This number could be close to one neutron per fission in nuclear fuel. Such value is typical for specialized thermal neutron transmutation facility with high enrichment of nuclear fuel and middle-range neutron flux. In high-flux facilities, the number of free neutrons is lower because of higher respective role of structural materials in neutron balance. The number of free neutrons for transmutation is proportional to the facility power. Therefore, transmutation facility with the power of 1 GW can incinerate Tc-99 and I-129 produced in power reactors with total thermal power of about 12 GW.

Transmutation is performed stepwise. The time of one stage of irradiation could be equal to one or several fuel lifetimes of transmutation facility. If typical lifetime of heavy-water facility with thermal neutron flux of 10^{14} neutr/cm^{2}s is about 0.5 years, incinerated part of Tc-99 for this time is about 8% and that of I-129 is about 5% with respect to initial amount. Thus, burnup of incinerated nuclides is rather low and therefore loading of Tc-99 and I-129 should be respectively high. Next stage of irradiation begins with the addition of new portion of Tc-99 and I-129. In the case of I-129, intermediate extraction of transmutation products is required after several stages since long-lived Cs-135 is produced from I-129 by 6-fold neutron capture.

When high-flux facility with the same power is used, characteristics of the transmutation process are different. In comparison with the middle-range neutron flux, the fuel load of the high-flux facility as well as the fuel lifetime and the number of free neutrons are lower. The duration of the irradiation stage and transmuted nuclide loading are also lower. The number of fuel reloadings and transmuted nuclide reloadings per year is higher than for middle-range neutron flux. This leads to an increase of the number of chemical reprocessing connected with reloading but total reprocessed or reloaded mass of fuel or that of transmuted nuclides per year remains the same. The efficiency of transmutation decreases only because of the decrease of the number of free neutrons. Half-incineration time which is defined as time of incineration of a half of initial amount of a transmuted nuclide decreases in higher neutron flux. Therefore, facility with middle-range flux could be used for current transmutation of long-lived fission products and high-flux facility would be required the back end of nuclear power for quick incineration of accumulated waste.

Transmutation of actinides

Process of actinide transmutation differs considerably from fission product transmutation. The main difference lies in the fact that fission products are transformed to stable nuclides while actinides are transformed to other actinides and finally to fission products. Radiological danger of fission products

decreases in the course of transmutation. Danger of transmuted actinides increases as more dangerous actinides are produced in the initial stage of transmutation. Minor actinides of spent fuel of power reactors are neptunium, americium, and curium isotopes. Most dangerous are Am-241 and Cm-244. New isotopes are produced as follows. Np-237 is transformed to Pu-238 in the course of neutron irradiation. Am-241 is transformed at first to short-lived Am-242, then to Cm-242 which is subsequently transformed into Pu-238. Am-243 is transformed to Cm-244. These processes occur both in thermal and fast neutron spectrum nuclear facilities. We shall use radiotoxicity as a measure of radiation danger of nuclides. Radiotoxicity of individual nuclide is inversely proportional to its maximum permissible activity in air or in water according to radiation safety standards. It is safe to drink water with maximum permissible activity of nuclide. Radiotoxicity increase during transmutation is illustrated by table 1. Transmutation of americium and curium isotopes extracted from annual unloading of power VVER-1000 type reactor was considered. Transmutation facility with thermal spectrum has a neutron flux $5 \cdot 10^{13}$ neutr/cm^{2}s and spectrum of light-water reactor. Fast neutron facility resembles power fast neutron sodium-cooled BN-800 type reactor.

Transmutation facility	Time, year				
	0	1	2	3	5
Thermal neutron spectrum	55	136	96	55	18
Fast neutron spectrum	55	94	91	83	35

Table 1. Radiotoxicity during transmutation of americium and curium, 10^{14} kg water.

Radiotoxicity increases during first several years of irradiation in both thermal and fast neutron spectrum due to Pu-238 production from Am-241 via Cm-242. Radiotoxicity decreases to an initial value after 2-3 years of irradiation. Rather quick reduction of initially increased radiotoxicity is typical of the process of irradiation of americium and curium without addition of nuclides during irradiation. If neptunium is introduced to initial nuclides, time of radiotoxicity increase to initial value is much longer because of additional production of Pu-238 from Np-237.

High flux transmutation

A serious problem in actinide incineration is to reduce radiotoxicity during transmutation. This problem is most important in modes with periodical addition of new portions of actinides during transmutation. In this mode, radiotoxicity increases during initial period of irradiation and then it reaches an equilibrium value when the rate of actinide incineration is equal to actinide feed. The time for which equilibrium is achieved is almost the same both for thermal and fast neutron spectrum. Equilibrium radiotoxicity for middle-range neutron flux is much higher than radiotoxicity of actinides loaded during one year. It can be reduced using high neutron flux.

Characteristics of transmutation modes in middle-range and high flux facilities are presented in table 2. Thermal neutron light water facility with neutron flux $5 \cdot 10^{13}$ neutr/cm^{2}s, fast neutron facility like sodium-cooled fast neutron reactor with neutron flux $5 \cdot 10^{15}$ neutr/cm^{2}s, and homogeneous heavy-water blanket of ADS with thermal neutron flux $5 \cdot 10^{15}$ neutr/cm^{2}s are considered. These data were published in [1]. Continuous introduction of actinides at a rate of 39 kg/year with isotopic composition 45% Np-237, 44% Am-241, 8.5% Am-243, 2.2% Cm-244, typical of spent fuel of PWR reactor was considered.

Type of facility	Thermal neutr.	Fast neutrons	High flux ADS
Neutron flux, neutr/cm^{2}s	$5 \cdot 10^{13}$	$5 \cdot 10^{15}$	$5 \cdot 10^{15}$
Time of equilibrium, year	50	40	0.5
Equilibrium actinide mass, kg	700	880	13
Equilibrium radiotoxicity	1	1.6	0.03
Reference time, year	250	400	4

Table 2. Transmutation characteristics

The high thermal flux facility has an obvious advantage, as the time for which equilibrium is achieved is significantly (up to 100 times) shorter than that of other types of facilities. Radiotoxicity is presented in respective units, it is normalized by value for thermal neutron facility. Equilibrium actinide mass and radiotoxicity is also much more preferable for high flux ADS facility, it is 40-50 times lower than that of middle-range flux facility with thermal or fast neutron spectrum. The reference time presented at the

bottom of table 2 is a parameter for comparison of processes of transmutation and storage with equal rate of actinide addition both to transmutation and to storage. An equilibrium radiotoxicity in facility is reached in a certain time while radiotoxicity in storage uniformly increases. Reference time is the time when radiotoxicity in storage becomes equal to that of transmutation facility. Thus, for conventional nuclear facility, reference time is 200-400 years. It means that during 200-400 years, simple actinide accumulation in storage with constant feed provides less radiotoxicity than that of established in transmutation facility with the same feed. However, transmutation in high flux ADS facility will be more preferable after 4 years of transmutation.

Conclusion

Transmutation of minor actinides leads to the increase of radiotoxicity during irradiation. It takes significant time compared to the lifetime of reactor facility to achieve equilibrium without effective transmutation. High flux nuclear facilities allow to minimize these draw-backs of conventional facilities with both thermal and fast neutron spectrum. They provide fast approach to equilibrium and low level of equilibrium mass and radiotoxicity of transmuted actinides. High flux facilities are advantageous also for transmutation of long-lived fission products as they provide short incineration time.

Reference

[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, Russia, 29-31 May, 1996. World Scientific Publishing Co., 1977, pp.67-76.