Comparative Study of ADS-burners with Thermal, Intermediate and Fast Neutron Spectrum for Transmutation of Minor Actinides

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Abstract. The problem of minor actinides (MA) handling in the closed nuclear fuel cycle (NFC) is one of the most important for future nuclear power. There are several approaches for MA transmutation but there are no common criteria for the comparison of their efficiency. In paper [1] we turned out the attention to the importance of taking into account the duration of the closed NFC in addition to the usual criterion of the neutron economy. In accordance with these criteria the different ADS-burners are considered: LBE-cooled reactor (fast neutron spectrum), molten-salt reactor (intermedium spectrum) and heavy water reactor (thermal spectrum). In all the cases the time of transmutation is more then 20 years.

I. Introduction

We define MA as a mixture of isotopes Np, Am and Cm and the process of MA transmutation as the direct and indirect fission of MA in the closed fuel cycle. The direct MA fission is the fission of the loaded MA, and the indirect one is the fission of transuranium (TRU) isotopes (including Pu ones) originated by the loaded MA in its transformation chains, as well as TRU produced by the other fissile elements (e.g. Pu) which are necessary for the ADS subcriticality level maintaining Therefore the mass of the nontransmuted TRU in NFC is equal to the mass of all TRU originated by MA and Pu loading in reactor [2]. The inclusion of TRU produced by Pu in the total balance of TRU certainly worsens the overall MA transmutation efficiency but it makes all consideration more realistic.

II. Time of transmutation

The transmutation of any TRU is a stochastic process which can be characterized by the probability density function ω (t). In the closed fuel cycle (without taking into account the small reprocessing fuel losses) the function ω(t) is normalized by the condition:

$$\int_0^\infty \omega(t)dt = 1, \qquad (1)$$

and the mean time τ_{in} of the TRU transmutation in reactor is

$$\tau_{in} = \int_0^\infty t \cdot \omega(t) dt . \tag{2}$$

As an example the functions $\omega_{MA}(t)$ and $\omega_{Pu}(t)$ for Pu and MA (including Np) for thermal reactor with molten salt fuel and heavy water as neutron moderator (its main parameters are given in Table 1) are presented on the Fig.1 [3].

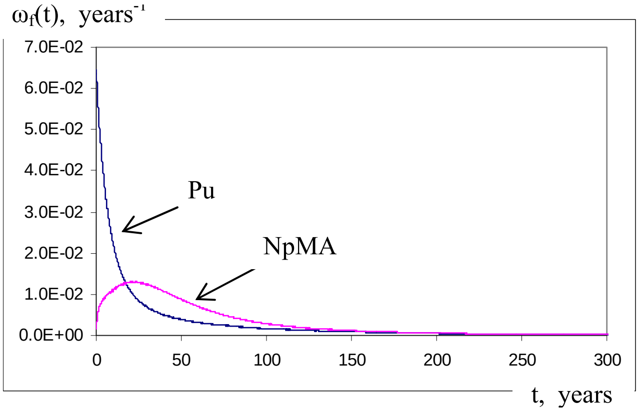

FIG.1. The transmutation probability density functions for ADS with thermal spectrum. In this case $\tau_{in}^{MA} = 67\,years$; $\tau_{in}^{PU} = 46\,years$.

There are two approaches to MA transmutation: the first one considers all Pu isotopes as the radioactive waste which should be burned together with all MA; in the second approach Pu is the valuable nuclear fuel which must be eliminated from the burning TRU balance. In what follows we calculate function $\omega(t)$ for the set of TRU originated in reactor by MA and Pu loading, i.e. for all isotopes of Np, Am, Cm, and all isotopes of Pu. We choose the initial MA composition which corresponds to those in the PWR spent fuel after 30 years of storage.

The transmutation time τ_{in} can also be determined via the equilibrium mass G_{TRU}^{in} of TRU in reactor and the rate of MA feeding q_{MA} [kg/year]. The probability $\xi(t)$ that TRU generated by MA and Pu loaded at the time moment t = 0 will be transmuted (i.e. fashioned directly or indirectly) at the time moment t is equal

$$\xi(t) = \int_0^t \omega(\mathrm{t}) d\mathrm{t} . \tag{3}$$

The nontransmuted TRU mass $G_{TRU}(t)$ accumulated to the time moment t due to MA loading with the rate q_{MA} is equal to

$$G_{TRU}^{in}(t) = q_{MA} \cdot \int_0^t \eta(t) dt, \quad \eta(t) = 1 - \xi(t). \tag{4}$$

Transforming $Eq.(4)$ and taking into account $Eq.(1\text{-}3)$ we get:

$$\int_0^t \eta(t)dt = t\cdot\eta(t)\Big|_0^t + \int_0^t t\cdot\omega(t)dt \underset{t\to\infty}{=} \tau_{in}\,. \tag{5}$$

The total equilibrium mass of TRU $G_{TRU}^{in} = G_{TRU}(\infty)$ is equal to:

$$G_{TRU}^{in} = q_{MA}\cdot\tau_{in}\,. \tag{6}$$

The equilibrium TRU mass G_{TRU}^{equ} originated by both MA and Pu loading in the closed NFC is equal

$$G_{TRU}^{equ} = q_{MA}\cdot\tau\ , \tag{7}$$

where $\tau = \tau_{in} + \tau_{out}$ and τ_{out} is the duration of outer NFC, i.e.

$$\tau_{in} = \frac{G_{TRU}^{in}}{q_{MA}};\ \tau = \frac{G_{TRU}^{equ}}{q_{MA}} = \frac{G_{TRU}^{in}}{q_{MA}}\left(1+\frac{\tau_{out}}{\tau_{in}}\right), \tag{8}$$

$$G_{TRU}^{equ} = G_{TRU}^{in} + G_{TRU}^{out};\ \ \frac{G_{TRU}^{in}}{\tau_{in}} = \frac{G_{TRU}^{out}}{\tau_{out}}, \tag{9}$$

where G_{TRU}^{out} is the amount of TRU in the outer NFC.
In the equilibrium closed NFC the MA feeding rate q_{MA} is equal to the MA transmutation rate considered usually as a main parameter in the all previous estimations of the reactor-burner efficiency. But in the closed NFC one more parameter G_{TRU}^{equ} is essential, which can be represented by the transmutation time τ. The third parameter $\varepsilon = q_{Pu}/q_{MA}$ is the ratio of Pu and MA feeding. In our opinion it is more informative than the number of neutrons, necessary for one MA nucleus transmutation, which is usually discussed.

TABLE 1. CHARACTERISTICS OF ADS-BURNERS AT k_{eff} =0.95*)

Reactor characteristics	**Neutron spectrum**		
	Fast	***Intermediate***	***Thermal***
Subcriticality Δk	0.05	0.05	0.05
Accelerator power, MW	10	10	10
ADS power, *MW*	330	760	300
Neutron flux Φ, $n/cm^2 \cdot s$	$1.5 \cdot 10^{15}$	$4 \cdot 10^{14}$	$4 \cdot 10^{13}$
Equilibrium Pu/Ma in the cycle loading, *tonn*	1.1/1.3	10.8/2.7	0.9/0.5
Pu/MA feeding content ε	0.01	2.9	2.5
Ttransmutation MA rate q_{MA}, *kg/year*	120	55	25
Transmutation time τ_{in}, *years*	~20	~200	~ 55

*) *In all the cases the accelerator power is equal 10 MW (1 GeV, 10 mA).*

TABLE 2. CHARACTERISTICS OF ADS-BURNERS AT k_{eff} =0.97

Reactor characteristics	**Neutron spectrum**		
	Fast	***Intermediate***	***Thermal***
Subcriticality Δk	0.03	0.03	0.03
Accelerator power, MW	6.0	5.9	10.0
ADS power, *MW*	330	760	500
Neutron flux Φ, $n/cm^2 \cdot s$	$1.3 \cdot 10^{15}$	$4 \cdot 10^{14}$	$3 \cdot 10^{13}$
Equilibrium Pu/MA in the cycle loading, *tonn*	1,5/1,9	12.0/2.9	1.5/0.9
Pu/MA feeding content ε	0,01	2.9	3.0
Ttransmutation MA rate q_{MA}, *kg/year*	120	55	40
Transmutation time τ_{in}, *years*	~30	~220	~60

III. Number of the fuel reprocessing cycles

As a rule the duration τ_c of a single fuel cycle, which consists of the time of fuel irradiation in reactor and refabrication in the outer NFC is much less than MA transmutation time τ. This means that TRU initiated by MA loading in reactor have to be reprocessed many times. The number of the single reprocessing cycles is equal to ratio

$$n_{rep} = \frac{\tau}{\tau_c}\,. \tag{10}$$

The total fraction Q_{loss} of MA loosed due to the fuel reprocessing is

$$Q_{loss} = n_{rep} \cdot \chi_c, \tag{11}$$

where χ_c is the relative MA losses per one reprocessing cycle.
At τ_c ~5-10 years the number of reprocessing is n_{rep} ~10 in most of the cases and this fact leads to sharp restrictions for the MA losses Q_{loss} in the closed NFC. Particularly the value $Q_{loss} \leq 10^{-3}$ can be achieved at $\chi_c < 10^{-4}$ only. Such a level of the spent fuel purification is discussed now in the molten salt technology only.

IV. DISCUSSION.

For illustration of the presented approach we have chosen three types of ADS-burners:

- Heavy water ADS (thermal neutron spectrum) which was developed for Th-U fuel cycle [3];
- Molten salt ADS (intermedium spectrum) with cascade scheme of neutron flux multiplication [4];
- Lead-bismuth ADS (fast neutron spectrum) [5] based on Russian submarine reactor as prototype [6].

The main characteristics of these reactors are presented in Tables 1 and 2. In Table 1 the accelerator power (W_a = 10 MW, I_a = 1 mA, E_a = 1 GeV) is fixed. In all the cases the initial fuel composition consist of the spent fuel of VVER-1000 after 30 years storage), enriched with ^{239}Pu to the level necessary for keeping the fixed subcriticality Δk.
As it is clear from Tables 1 and 2 the reduction of subcriticality does not enhance the efficiency of ADS- burners (but reduce the accelerator power. Among the considered systems only ADS based on LBE fast reactor has the acceptable characteristics ($\tau_{in} \sim 20 years,\ \varepsilon = 0.01$), but it should be taken into account that in the contemporary NFC $\tau_{out} / \tau_{in} \sim 3$, i.e. $\tau \sim 4\tau_{in}$. For the molten salt ADS $\tau \approx \tau_{in}$ and the nuclear fuel reprocessing for MSR is essentially simplified.

It is clear that minimal amount of TRU in closed NFC can not be less than G_{TRU}^{equ} which essentially exceeds q_{MA} during the all period of the MA burner functioning. In this sense reactor-burner can be considered also as a storage of TRU. The elimination of G_{TRU}^{equ} is the special problem of the back end of nuclear power.

V. CONCLUSION

The transition to the closed NFC requires the modification of the general approach to the efficiency estimations of the different reactor-burners and their NFC. Particularly in this case it is essential to turn attention not only on the MA transmutation rate q_{MA} but also on the equilibrium mass G_{TRU}^{equ} in total NFC which can be represented via transmutation time τ.

For comparison of the MA transmutation efficiency of ADS-burners in the closed NFC we suggest to use three basic parameters:

- rate of MA transmutation, q [kg/year];
- transmutation time, τ [year];
- fraction of fissile nuclides (e.g. Pu), $\varepsilon = q_{Pu} / q_{MA}$.

The final conclusion concerning the transmutation efficiency of the considered ADS reactor-burners should be completed by the comparison with the critical reactor –burners efficiency as well as the analysis of the outer NFC, especially of MA losses during the fuel refabrication, economical restriction etc.

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Appendix 1: References

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