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Effect of minor actinides doping on plutonium produced in large-scale FBR blanket

Yoshitalia Meiliza*, Shigeo Ohki, Tsutomu Okubo

Japan Atomic Energy Agency (JAEA), 4002 Narita, Oarai, Ibaraki 311 1393, Japan

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ABSTRACT

The present study focuses on the effect of minor actinides (MAs) addition into the FBR blanket as ways of increasing fraction of even-mass-number plutonium isotopes, especially ^{238}Pu, aiming at enhancing the proliferation resistance of plutonium produced in the blanket. The MA loading potential to enhance the proliferation resistance of plutonium is investigated, with considering actual design constraints on the fuel decay heat from the fuel handling and fabrication points of view, as MAs considerably generate decay heat. It reveals that depending on doping quantity of MAs, it is possible to denature produced plutonium by MA transmutation. MA addition in the blanket gives a significant increment in ^{238}Pu fraction of generated plutonium but less effect on other even-mass-number plutonium isotopes. However, it is important that MA compositions should be adequately controlled to satisfy both the proliferation resistance requirements and the decay heat constraints for fuel handling.

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1. Introduction

Plutonium recycling in FBRs is important from the viewpoint of effective utilization of energy resources. Proliferation resistance of plutonium is one of the important aspects in FBR strategy, especially plutonium produced in the FBR blanket that includes a large amount of ^{239}Pu isotope and only a small amount of even-mass-number plutonium isotopes. One of the efforts to deter plutonium proliferation is to change physical characteristic of plutonium itself with regard to material barrier aspect. (Massei and Schneider, 1982; Wydler et al., 1980)

Characteristics of proliferation resistance properties indicate how difficult is the material conversion to nuclear explosive devices (NEDs) from a particular fissile material once the material is available in an "acceptable" chemical form. The most difficult barrier against proliferation is the isotopic barrier which incorporates with critical mass, spontaneous fission neutron generation, decay heat, radiation dose, etc. (Beller and Krakowski, 1999; Department of Energy, 1997; DeVolpi, 1982).

In the future commercialization stage where a number of FBRs are deployed, there is a possibility that the FBR design gives a high priority to proliferation resistance in terms of material barriers. The present study focuses on the effect of MAs addition into the FBR blanket as ways of increasing fraction of even-mass-number plutonium isotopes, especially ^{238}Pu, aiming at enhancing the proliferation resistance of plutonium produced in the blanket. The intrinsic proliferation resistance property of plutonium is evaluated based on the decay heat and spontaneous fission neutron generation.

2. Intrinsic features of plutonium proliferation resistance

The physical characteristics of plutonium such as the decay heat, spontaneous fission neutron generation and critical mass are the main constituents of isotopic barriers against proliferation. Among plutonium isotopes, ^{238}Pu has the smallest critical mass but it has a high decay heat and a large spontaneous fission neutron generation. The impurity of ^{240}Pu in ^{239}Pu was considered as undesirable material due to its large spontaneous fission neutron not due to large critical mass. The neutron source enhances the probability of pre-ignition, (Mark, 1993) thus leading to "fizzle yield", a minimum possible yield of explosion for implosion-type NEDs, while heat and radiation complicate the manufacturing and maintaining NEDs.

Denaturing of plutonium leads to higher content of even-mass-number plutonium isotopes which enhances proliferation resistance of plutonium due to high decay heat dominated by ^{238}Pu as well as large spontaneous fission neutrons governed by even-mass-number plutonium isotopes (^{238}Pu, ^{240}Pu and ^{242}Pu). Two pathways for producing ^{238}Pu by MA transmutation are through β^- decay from ^{238}Np ($T_{1/2} = 2.36$ d), which is produced from ^{237}Np neutron capture

* Corresponding author. Tel.: +81 29 267 4141; fax: +81 29 267 1676.
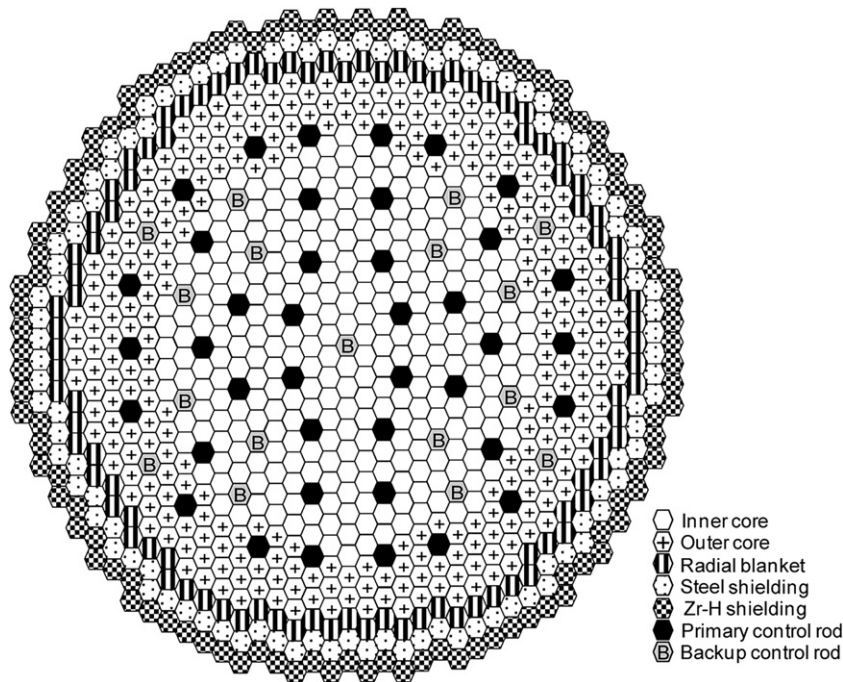E-mail address: yoshitalia.meiliza@jaea.go.jp (Y. Meiliza).

Fig. 1. Core configuration of FaCT reference core.

reactions, and through α decay from ^{242}Cm ($T_{1/2}$ = 162.79 d), which is the β^- decay daughter of ^{242}Am produced from ^{241}Am neutron capture reactions. The role of MAs in the generations of ^{240}Pu and ^{242}Pu are through α decay from ^{244}Cm ($T_{1/2}$ = 18.1 y), which is the β^- decay daughter of ^{244}Am produced from ^{243}Am neutron capture reactions, and through electron capture from ^{242}Am ($T_{1/2}$ = 16 h), which is produced from ^{241}Am neutron capture reactions.

The properties of plutonium isotopes have been studied as indicators to specify proliferation resistant plutonium. Thus far, there are one official criterion and several proposals on this isotopic barrier of plutonium. International Atomic Energy Agency (IAEA), 1972 through Information Circulation No. 153 (INFIRC 153) has mentioned that plutonium containing 80% of ^{238}Pu or more is exempted from proliferation concerns. The categorization of plutonium protection based on the ^{238}Pu isotopic ratio was studied by Kessler et al., 2008. He examined thermal analysis for implosion-type NEDs and determined that plutonium containing more than 9% ^{238}Pu isotopic fraction in reactor-grade plutonium was necessary to get material regarded as proliferation resistant.

Pellaud, 2002 proposed a plutonium classification scheme based on spontaneous fission neutron generation. He summarized the suitability of plutonium mixture for explosive purposes by the ^{240}Pu isotopic content and described that reactor-grade plutonium, plutonium with more than 18% ^{240}Pu, is conceivably usable material for use in explosive purposes. Recently, the Protected Plutonium Production (P^3) concept has been proposed. (Saito, 2004; Meiliza et al., 2008) In this concept, MAs were utilized as a resource of ^{238}Pu to produce the proliferation-resistance of plutonium.

Since the definitive criteria as the world standard for proliferation resistance of plutonium has not been established yet, provisionally, the present paper uses 9% ^{238}Pu isotopic fraction as the target for plutonium denaturing.

3. Methodology

In the transition state from LWRs to FBRs, there is a possibility of MA utilization in the FBR core and blanket due to the recycling of LWR spent fuel. A large-scale FBR core, i.e. 3530 MWth, based on the representative core concept in the Japanese Fast Reactor Cycle Technology Development (FaCT) Project (Ohki et al., 2008) was chosen as the reference core. The core layout adopted in the present calculations is shown in Fig. 1. A breeding ratio of 1.1 can be obtained with the following specifications: core height 100 cm, axial blanket thickness 40 cm, the number of fuel pin per subassembly 255, fuel pin diameter 10.4 mm, and operation cycle length around 800 days (with 4 fuel exchange batches). From the viewpoint of fuel handling and fabrication, the decay heat limit of a subassembly is 2.6 kW and 4 kW for fresh fuel and spent fuel cooled for 4 years, respectively. The former value is corresponding

Table 1
Major design condition for FaCT reference core (low breeding core).

No	Item	Value
1	Plant condition	
	Power output [MW$_e$/MW$_{th}$]	1500/3530
	Coolant temp. [ºC] (outlet/inlet)	550/395
	Shielding region diameter [m]	< ~ 7.0
2	Core performance targets	
	Breeding ratio	~1.1
	Discharge burnup [GWd/t]	
	Core	150
	Total	> 80
	Operation cycle length [month]	> ~ 24
3	Safety requirement	
	Sodium void reactivity [$]	< ~ 6
	Core (active) height [cm]	< 100
	Average core specific heat [kW/kg-MOX]	> ~ 40
	Subassembly concept for recriticallity free	FAIDUS*
4	Fuel integrity limit	
	Max. linear heat rate [W/cm]	< ~ 430
	CDF** (steady state)	< 0.5
	Max. fast neutron fluence (E$_n$>0.1 MeV) [n/cm^2]	< ~ 5 x 10^{23}
5	Decay heat limit Kawaguchi and Namekawa, 2007	
	Fresh fuel [kW/Subassembly]	< 2.6
	Spent fuel*** [kW/Subassembly]	< 4.0

* Fuel Assembly with Inner Duct Structure
** Cummulative Damage Fraction
*** after 4 years of cooling time