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Evaluation of proliferation resistance of plutonium based on spontaneous fission neutron emission rate

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ABSTRACT

The proliferation resistance of plutonium can be enhanced by isotopic denaturing and one of the features of Pu isotopic denaturing is increasing of the spontaneous fission neutron emission rate. A high spontaneous fission neutron emission rate causes "predetonation" in a nuclear explosive device, and it makes the designing and building of a nuclear explosive device more complicated. In this paper, the enhancement of Pu proliferation resistance by isotopic denaturing was evaluated based on spontaneous fission neutron emission rate using a simple nuclear explosive device model. The feature of spontaneous fission neutron was evaluated with probabilistic approach, and a new Pu categorization which makes enable to categorize plutonium by its isotopic compositions was proposed based on the evaluation results. The Pu categorization was applied to an evaluation of the proliferation resistance of plutonium produced in LWRs and FBRs using the evaluation function, *"Attractiveness"* for two types of plutonium; plutonium produced in typical LWRs and FBRs, and denatured plutonium produced by transmutation of MAs.

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1. Introduction

Nuclear proliferation issue is a serious concern in future nuclear energy use and proliferation resistance has been defined as a technology goal for next generation nuclear energy systems. Potential proliferators must obtain a sufficient amount of an appropriate weapon-usable material for manufacturing a nuclear explosive device (NED) and plutonium is one of the most concerned materials in the proliferation issue. Plutonium separated from spent fuel contains various Pu isotopes having different nuclear properties such as critical mass, decay heat, spontaneous fission neutron emission rate, and radioactivity. A higher content of fissile plutonium increases a serious concern in the possibility of the Pu diversion. Since early 1980, some non-proliferation efforts to deter the diversion of plutonium from peaceful use have been carried out, for instance, to denature plutonium in regard to Pu isotopic composition against the proliferation risk (Devolpi, 1982; Waltz et al., 1980; Massey and Schneider, 1980). Furthermore, Saito et al. (2006) recently proposed the concept of *Protected Plutonium Production* (P^3). In this concept, minor actinides (MAs) such as ^{237}Np and ^{241}Am are utilized as the resources for ^{238}Pu production to enhance the proliferation resistance of plutonium in the fuel. The production of even-mass-number Pu isotopes (especially, ^{238}Pu) by transmutation of MAs has been verified with irradiation experiments (Saito et al., 2006; Saito, 2009; Koyama et al., 2010; Sagara, 2005).

Plutonium isotopic barrier is the most important features of intrinsic feature for proliferation resistance of nuclear fuel cycle system. Table 1 shows the nuclear properties of Pu isotopes related to the isotopic barrier (Matsunobu et al., 1991). Decay heat is one of the key nuclear properties for the Pu isotopic barrier. A higher decay heat makes plutonium handling or NED manufacturing technically difficult. It has been mentioned that plutonium containing more than 15% ^{238}Pu is technically unfeasible for a NED from the view point of decay heat (Kimura et al., 2011). Another important nuclear property for the isotopic barrier is spontaneous fission neutron (SFN) emission. Higher neutron source by SFN emission may cause premature initiation of a chain reaction before the fissile plutonium is fully compressed and hinders from assembling plutonium inside a NED. This is known as "predetonation" and it reduces the explosive yield of the NED. To avoid predetonation a NED requires more technical sophistication and it makes designing and building of the NED technically difficult. Therefore, a plutonium with high SFN emission rate becomes unattractive for weapon use.

In earlier publications, the predetonation caused by a high SFN emission of reactor-grade plutonium has been evaluated with the probabilistic approach (Devolpi, 1982; Mark, 1993). It has been mainly focused on Pu denaturing by increasing the isotopic fraction of ^{240}Pu emitting a high SFN because plutonium with a high content of ^{240}Pu can be easily produced in typical light water reactors (LWRs). Aside from ^{240}Pu, other even-mass-number Pu isotopes, ^{238}Pu and ^{240}Pu should also contribute to the enhancement of Pu proliferation resistance since they have higher SFN emission

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Table 1
Nuclear properties of Pu isotopes.

Isotope	Decay heat (W/kg)	Spontaneous fission neutron rate (n/g/s)	Bare critical mass[a] (kg)
^{238}Pu	567	2660	13.1
^{239}Pu	1.93	0.0226	14.8
^{240}Pu	7.06	1030	44.8
^{241}Pu	3.4	0.0493	17.6
^{242}Pu	0.12	1720	87.8

[a] δ-phase of plutonium metal (15.8 g/cm^3).

rates than ^{240}Pu as shown in Table 1. Furthermore, Sagara et al. (2010) discussed the effect of coupled doping of the even-mass-number Pu isotopes to ^{239}Pu on the number of SFN emission with a simple bare critical mass model. They concluded that the effect of coupled doping plays a very important role in the Pu denaturing by increasing of both SFN rate and the required mass (in this case, bare critical mass).

In this paper, the enhancement of Pu proliferation resistance by SFN emission was evaluated with the probabilistic approach using a simple NED model. The non-predetonation probability was mainly studied as an indicator for the evaluation. The features of Pu denaturing by all even-mass-number Pu isotopes and also the effect of coupled doping were studied parametrically. As a summary of the probabilistic evaluation, a new Pu categorization which makes enable to categorize plutonium by its isotopic compositions were proposed. The present Pu categorization was applied to evaluate the proliferation resistance of plutonium produced in typical LWRs and fast breeder reactors (FBRs) blanket along with an evaluation function "*Attractiveness*" (Saito et al., 2007) as the case studies.

2. Evaluation methodology

2.1. Predetonation probability

The proliferation resistance of plutonium by the SFN emission was evaluated based on the predetonation probability of a fission core in NED of which the theory was described in the earlier publications (Devolpi, 1982; Mark, 1993) with a ramp increase in criticality given by

$$\Delta k(t) = \Delta k_{max}\left(\frac{t}{T}\right), \tag{1}$$

$$\alpha(t) = \frac{\Delta k_{\max}}{l}\left(\frac{t}{T}\right), \tag{2}$$

where $\Delta k(t) = k_{\text{eff}}(t) - 1$ is the supercriticality, which increases from a prompt criticality to a maximum ($\Delta k_{\max}$) during a compression time of T (described as a reactivity insertion duration in the further discussions) at time t, $\alpha(t)$ is Rossi-alpha (corresponding to the reciprocal of time constant of nuclear power excursion in one point reactor kinetics) and l is the prompt neutron lifetime (Devolpi, 1982; Mark, 1993). The fission chain is initiated at $t \geqslant T$ in a normal NED and for a strong SFN source, the cumulative probability that persistent fission chains have occurred before time t_i is given by Hansen (1960):

$$P(t_i) = 1 - \exp\left[-\frac{2Sl}{\nu\Gamma_2}\int_0^{t_i}\alpha(t')dt'\right] = 1 - \exp\left[-\frac{S}{\nu\Gamma_2}\frac{\Delta k_{\max}}{T}t_i^2\right], \tag{3}$$

where ν is average number of neutrons per spontaneous fission (assumed as 2.3 from Matsunobu et al. (1991)), $\Gamma_2 = 0.714 + 0.035\,\nu$ is Diven factor (Diven et al., 1956), and S is total number of SFN emitted per second in a fission core which is the product of SFN emission rate and mass of the core. After the fission chain reaction is initiated, the core starts explosion at approximately e^N fissions ($N = 40$ (Sandmeire et al., 1972; Kessler et al., 2008) was used in this paper), the time, $t = t_{ex}$, described as

$$t_{ex} = \left(t_i^2 + \frac{2lNT}{\Delta k_{\max}}\right)^{\frac{1}{2}}. \tag{4}$$

According to the empirical relation of explosive yield (Serber's relation) (Mark, 1993; Serber, 1992), the explosive yield is proportional to $(\Delta k)^3$ at the explosion start. Using this relation, Eqs. (1) and (4) can give the relative value of the reduced yield (Y) to the design one (Y_0) as

$$\frac{Y}{Y_0} = \left[\left(\frac{t_i}{T}\right)^2 + \frac{2lN}{\Delta k_{\max}T}\right]^{\frac{3}{2}}. \tag{5}$$

From Eqs. (3) and (5), the non-predetonation probability ($P_{\text{non-predet}}$) and the probability of achieving an explosive yield ($Y/Y_0 > X$) can be given by

$$P_{\text{non-predet.}} = \exp\left[-\frac{S}{\nu\Gamma_2}\Delta k_{\max}T + \frac{2S}{\nu\Gamma_2}Nl\right], \tag{6}$$

$$P\left(\frac{Y}{Y_0} > X\right) = \exp\left[-\frac{S}{\nu\Gamma_2}\Delta k_{\max}TX^{\frac{2}{3}} + \frac{2S}{\nu\Gamma_2}Nl\right]. \tag{7}$$

The minimum explosive yield ($Y_{\min}$) is given by

$$\frac{Y_{\min}}{Y_0} = \left(\frac{2Nl}{\Delta k_{\max}T}\right)^{\frac{3}{2}}. \tag{8}$$

Thus, the predetonation probability is a function of the maximum supercriticality $\Delta k_{\max}$, the prompt neutron lifetime l, the reactivity insertion duration T and the total number of SFN emitted per second in a fission core S. Obviously, the total number of SFN emitted per second in a fission core, S, should be the most important parameter but other two parameters also needs to be considered to determine the predetonation probabilities. The latter two parameters become important especially in the case of doping of ^{240}Pu and ^{242}Pu.

2.2. Nuclear explosive devise model and compression condition

The simple NED model in the present paper was defined combining the existing NED models mentioned in earlier studies, so that all features of Pu denaturing by all-even-mass-number Pu isotopes doping can be considered. A core part of Hypothetical Nuclear Explosive Device (HNED) model proposed by Kessler (2007) was employed as an NED model which consists of a metallic Pu-sphere and a metallic uranium (U) reflector with 5 cm in thickness as shown in Fig. 1. The required mass of the Pu-sphere was determined by $k_{\text{eff}} = 0.98$ with the reflector. This means the dimension of the NED model was changed by the isotopic composition of plutonium. The required mass of Pu-sphere is described as a subcritical mass (SCM) in the succeeding discussions. In earlier studies, the required parameters for predetonation probabilities except to SFN emission rate have been assumed as constant for simple discussions. For instance, Mark (1993) defined the maximum supercriticality as unity and the factor $\Delta k_{\max}/\nu\,\Gamma 2$ in Eqs. (6) and (7) becomes 12. Mark (1993) also assumed the mass of Pu-sphere as constant, 8 kg. In this paper, all parameters were changed by giving a specific compression condition to the simple NED model with SCM Pu-sphere.

The maximum supercriticality $\Delta k_{\max}$ was determined by a homogeneous compression ratio (ρ/ρ_0) of 4 (Devolpi (1982) and

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