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Proliferation Resistant Fuel for Pebble Bed Modular Reactors

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Abstract—*We show that it is possible to denature the plutonium produced in pebble bed modular reactors by doping the nuclear fuel with either 3050 ppm of ^{237}Np or $\sim$2100 ppm of isotopic composition of Am. A correct choice of these isotope concentrations yields denatured plutonium with isotopic ratio $^{238}Pu/Pu \geq$ 6%, for the entire fuel burnup cycle. The penalty for introducing these isotopes into the nuclear fuel is a subsequent shortening of the fuel burnup cycle, with respect to a nondoped reference fuel cycle, by $\sim$40 and 20 full-power days, respectively, which correspond to 4.1 and 2.0 GW(d)/ton reduction in fuel discharge burnup.*

I. INTRODUCTION

Proliferation of nuclear weapons produced with power reactor plutonium has always been a major problem of the nuclear energy industry. This includes the pebble bed modular reactor (PBMR), which is a specific design of a Generation IV high-temperature reactor (HTR), mainly due to its online refueling feature and the small element size of the pebble, which makes it simpler to divert the pebbles, which may be misused for the production of weapons grade plutonium. However, it should be noted that to obtain a significant quantity of weapons-grade plutonium, many pebbles must be diverted at low burnup levels. Many low-level-burnup pebbles means reduction in neutron economy, and it is more detectible due to its effect on the power plant operation. Some other suggestions have been made for making PBMRs more proliferation resistant.[1–4]

A promising approach to preventing the proliferation of power reactor plutonium is to denature the plutonium by increasing the ratio of ^{238}Pu to total Pu in the spent fuel.[5] The ^{238}Pu isotope is characterized by a high heat rate ($\sim$567 W/kg) due to the alpha decay of the ^{238}Pu with half-life of 87.74 years, in addition to its high–spontaneous fission neutron emission, which is higher than that of ^{240}Pu. Thus, the presence of ^{238}Pu in Pu considerably complicates the design and construction of nuclear weapons based on Pu, owing to these characteristics of ^{238}Pu. Recent papers[6,7] show that a Pu mixture is proliferation resistant if the ^{238}Pu fraction of the total Pu is larger than 6%. In this paper we have studied a feasible technique for ensuring that the ^{238}Pu-to-Pu ratio, in the Pu produced in PBMRs, is >6% during the entire fuel cycle.

Contamination of the spent fuel with ^{238}Pu may be achieved by doping the nuclear fuel with either ^{241}Am or ^{237}Np (Refs. 8 through 17). The ^{238}Pu isotope is obtained from both ^{241}Am and ^{237}Np by a neutron-capture reaction and the subsequent decay of the reaction products, as seen in Fig. 1.

The ^{237}Np isotope is by itself a potential weapons-grade material. However, its large critical mass of 57 $\pm$ 4 kg (Ref. 18) and the difficulty of extracting it from irradiated fuel elements make it impractical for weapons purposes. On the other hand, the critical mass of ^{241}Am is smaller, i.e., 34 to 45 kg. However, with decay heat production of 114W/kg, the critical mass becomes a heat source of 3.9 to 5.1 kW, which makes ^{241}Am unsuitable for weapons applications.[7] As a result, it is a nonproliferating material.

Another advantage of introducing ^{241}Am into the fuel is that during the (n, γ) reaction, another isotope is
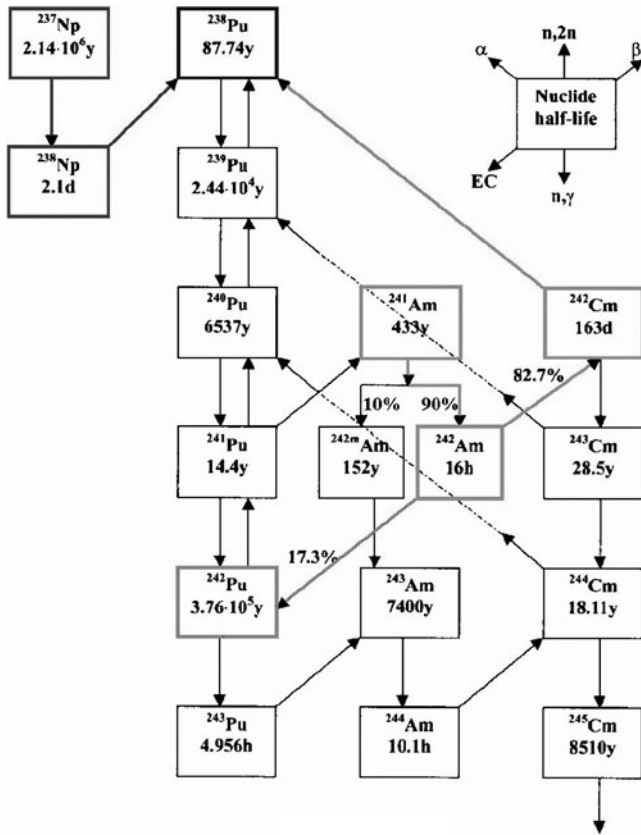
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Fig. 1. Plutonium-238 production paths.[17]

produced, i.e., ^{242m}Am, which is highly fissile with extremely high spontaneous fission neutrons emission rate. Because of its high fission cross section, the presence of ^{242m}Am reduces the residual poison reactivity penalty resulting from the introduction of ^{241}Am into the fuel. The branching ratio of the ^{241}Am(n, γ) reaction generally depends on the neutron energy spectrum in the reactor. However, in the thermal region this branching ratio is independent of the neutron energy. Since PBMR is a thermal reactor, this ratio is averaged and taken to be 10% and 90% to ^{242m}Am (metastable) and ^{242g}Am (ground state), respectively.[19]

It should be noted that either ^{241}Am or ^{237}Np are obtained by reprocessing of spent fuel. Obtaining ^{241}Am or ^{237}Np is less proliferating than obtaining Pu in the same way. In addition, reprocessing of spent fuel is being carried out already in order to produce mixed-oxide (MOX) fuel.

In this paper we study the doping requirements of both ^{241}Am and ^{237}Np in order to guarantee a proliferation-resistant Pu isotopic composition in the spent fuel during the entire fuel burnup cycle in PBMRs. An important result of this study is that the introduction of either ^{241}Am or ^{237}Np into the nuclear fuel is accompanied by a penalty with respect to fuel burnup cycle duration.

II. PBMR DESCRIPTION

The PBMR has been developed with the intention to improve safety, economics, and proliferation resistance. It has a vertical steel reactor pressure vessel, which contains the core barrel and the annular pebble fuel core.[20] The fuel pebbles are 60 mm in diameter, weighing 210 g, with graphite inner part encasing coated particles (TRISO) with total of 9 g of uranium enriched to 9.6% ^{235}U, as illustrated in Fig. 2. They cycle continuously through the reactor, where each pebble cycles through the core about six times with a single-cycle duration of 3 months, until they are expended after ~3 years. At this point, the fuel average enrichment of a pebble is 4% to 5%, and its average burnup is 80 GW(d)/ton. Operational cycles are expected to last 6 years between shutdowns.[20,21] Other calculations estimate a discharge burnup of a little over 90 GW(d)/ton (Ref. 22).

The PBMR has many advantages, such as helium as a coolant (radioactively inert, not corrosive, and can be fed directly into a turbine), online refueling, passive cooling mechanisms, and low–power density core such that no meltdown scenario could physically occur, even in the case of a loss-of-coolant accident.[21] In the case of a pebble coating breach, only a small amount of radioactive nuclides would be released, due to the pebble fuel being divided over 15 000 particles, individually coated with ceramic materials.

Another advantage, from a proliferation point of view, is that pebble reprocessing is currently very difficult.[21] However, recent studies have demonstrated that pebble reprocessing is possible.[23–25]

III. ANALYSIS METHODOLOGY

In our analysis we consider a widely used PBMR unit cell approximation,[26] as illustrated in Fig. 3. The unit cell is cubic and represents a body-centered cubic (bcc) lattice with eight 1/8 fuel pebbles at the corners and one fuel pebble centered in the middle (a total of two fuel pebbles per unit cell). By averaging over the entire core, we set the unit cell packing factor to 0.61 (Refs. 27 and 28), whereas the maximum packing factor for bcc lattice is 0.68. Throughout the calculations we used reflective boundary conditions for all six cubic faces, such that the calculated multiplication factor is the infinite multiplication factor k_∞. Whenever beginning of cycle (BOC) was calculated, we used fresh fuel. The parameters of the specific PBMR design being analyzed are summarized in Table I.

In our model we have assumed that a given pebble is surrounded by pebbles with the same burnup. As a result, this model is less accurate, compared to a model that considers pebbles with nonuniform burnups. However, the difference in k_{eff} between uniform burnup distribution and sinusoidal burnup distribution is ~1000 pcm
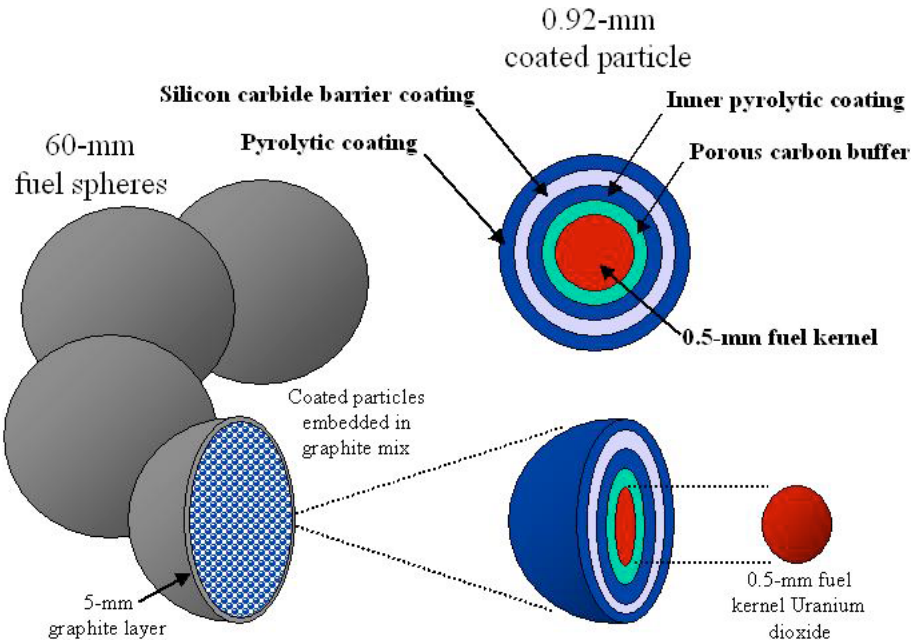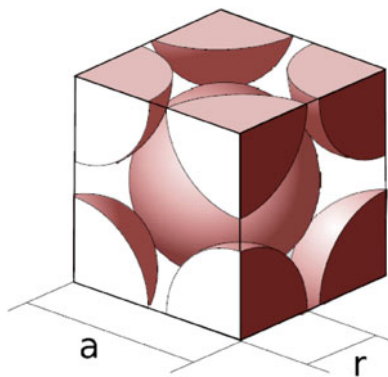
Fig. 2. Schematic drawing of fuel pebble structure.[21]

Fig. 3. A bcc lattice unit cell.

JEFF-3.1 evaluated data files. The TRISO coated particles in the fuel kernel (see Fig. 2) are positioned in space according to a three-dimensional cubic mesh with a specific choice of the edge so that the total mass of fuel per pebble is conserved and no clipped particles are present.[26]

IV. RESULTS

Two different scenarios are considered for doping the fuel. In the first one, an isotopic composition of Am is introduced into the nuclear fuel at BOC. The Am isotopic composition considered (referred to as Am) is 85.6% ^{241}Am, 0.08% ^{242m}Am, and 14.32% ^{243}Am, which corresponds to the discharged fuel of a typical pressurized water reactor.[35] This given composition is used throughout the analyses below. The total amount of Am in the reactor, corresponding to a concentration of 2100 ppm, is ~9.7 kg (0.02 g/pebble). In the second scenario, ^{237}Np is introduced into the fuel at BOC. The total amount of ^{237}Np in the reactor, corresponding to a concentration of 3050 ppm, is ~14.0 kg (0.03 g/pebble).

To estimate the accuracy of our calculations, we have compared k_∞ BOC as well as the discharge burnup at $k_\infty =$ 1 with similar results calculated by Çolak and Türkmen.[36] Çolak and Türkmen calculated a unit cell of HTR using a model similar to the one we used (bcc). They utilized a 44−energy group library compared to our continuous library (JEFF-3.1) and SERPENT for neutron transport and burnup calculations, where we have used BGCore (Ref. 33). We obtained results for k_∞ at BOC of 1.437, the

(Ref. 29). This difference has a small effect on the general trend of our results.

The calculations are performed using the BGCore software package,[33] which provides a comprehensive computer simulation of nuclear reactor systems and their fuel cycles. BGCore interfaces with the MCNP Monte Carlo particle transport code[34] with a SARAF module for calculating in-core fuel composition and spent-fuel emissions following discharge. The SARAF module was independently developed at Ben-Gurion University of the Negev. In the BGCore system, the SARAF module receives the relevant data from MCNP, executes the depletion time step, and feeds back the updated fuel composition for the next MCNP time step. The SARAF data library required for these calculations is based on the

TABLE I

PBMR Data Used in the Calculations*

Data	Value
Thermal power [MW(thermal)]	400
Electrical power [MW(electric)]	165
Discharge burnup [GW(d)/ton]	80
Coolant	Helium
Mean power per pebble (W)	890
Number of fuel pebbles in core	450 000
Fuel pebble diameter (mm)	60
Graphite outer layer thickness (fuel-free zone) (mm)	5
Pebble graphite matrix density (g/cm^3)	1.74
TRISO coated particles per fuel pebble	15 000
Average core packing factor	0.61
Square pitch between pebbles (mm)	71.842
Fuel	UO_2
Fuel density (g/cm^3)	10.4
Fuel kernel diameter (mm)	0.5
Fuel enrichment	9.6%
Uranium mass per pebble (g)	9
Square pitch between TRISO coated particles (mm)	1.6145
Coating layer materials (inner to outer)	C, C, SiC, C
Coating layer thicknesses (inner to outer) ($\times 10^{-3}$ mm)	95, 40, 35, 40
Layer densities (inner to outer) (g/cm^3)	1.05, 1.9, 3.18, 1.9

*References 27 through 32.

same value obtained in Ref. 36. Furthermore, the discharge burnup at $k_\infty = 1$ was found to be 82 GW(d)/ton compared to 81 GW(d)/ton obtained in Ref. 36. Thus, the comparison which used two independent codes, as well as discrete vs. continuous cross section library, indicates that both methods for calculating HTR agree within 100 pcm for k_∞ and within ~1 GW(d)/ton for discharge burnup.

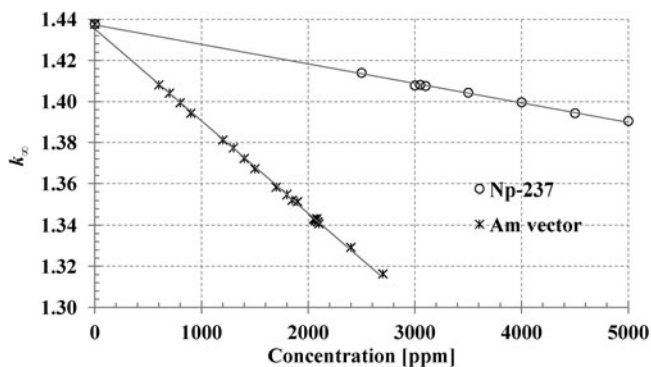The dependence of the PBMR k_∞ (of a unit cell) on the concentration of either Am or ^{237}Np at BOC is illustrated in Fig. 4. The concentration is given in units of ppm—parts per million of the UO_2 weight. In both scenarios, k_∞ exhibits a linear dependence on the concentration. The difference between the two scenarios is due to the high absorption cross section of ^{241}Am with respect to ^{237}Np.

Several burnup calculations are carried out to find the minimal concentration of the Am or ^{237}Np needed in order to meet the requirement ^{238}Pu/Pu $\geq$ 6% during the entire burnup cycle. According to Figs. 5 and 6, the minimal concentrations are 2100 and 3050 ppm of Am and ^{237}Np, respectively.
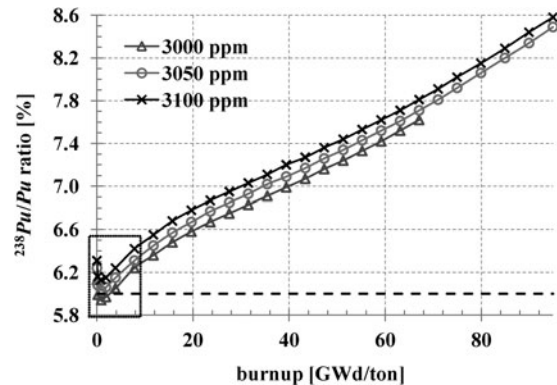

Fig. 4. Change in PBMR k_∞ at BOC as a function of ^{237}Np and Am concentrations.

Fig. 5. Weight ratio of ^{238}Pu to total plutonium as a function of fuel burnup for ^{237}Np doping. The area enclosed within the dotted frame is enlarged in Fig. 7b.
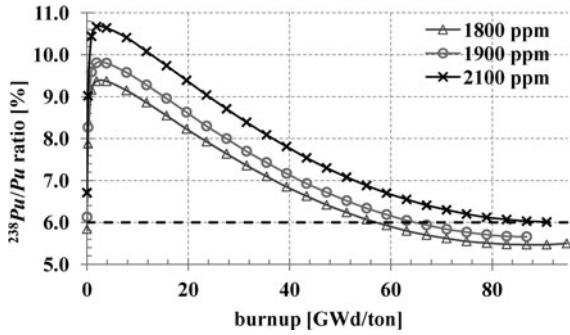
Fig. 6. Weight ratio of ^{238}Pu to total plutonium as a function of fuel burnup for Am doping.

However, the qualitative behavior of the ratio ^{238}Pu/Pu with respect to burnup is different for the two scenarios, as illustrated in Fig. 7. In the ^{237}Np doping, this ratio initially decreases on a very fast time scale [~10 full-power days (FPDs)], after which it starts to monotonically increase (at a slower rate), attaining and then exceeding its initial value after ~60 FPDs. In the Am doping, on the other hand, this ratio exhibits a very sharp initial rise, reaching a maximum after ~40 FPDs, after which it monotonically decreases until it reaches an asymptotic value toward the end of the cycle.

From a proliferation point of view, it is possible to use even lower concentrations of ^{237}Np. Decreasing the concentration of the ^{237}Np below 3050 ppm results in a decrease of the ^{238}Pu/Pu ratio below 6%, but only for a short period at BOC, as illustrated in Fig. 7b for ^{237}Np doping at 3000 ppm. This ratio reaches a minimum of 5.9% after 10 FPDs and climbs back up to 6.0% after 30 FPDs. The total amount of plutonium produced during the first 10 and 30 FPDs is 2.0 and 5.8 kg, respectively.[a]

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[a]After 20 FPDs the ^{238}Pu/Pu weight ratio is 6.0% and the total amount of plutonium is 3.9 kg.

The total amount of Pu during burnup is calculated for both scenarios and for a reference PBMR (without fuel doping), as illustrated in Fig. 8. The total amount of Pu produced at end of cycle [after 80 GW(d)/ton] is rather similar for both ^{237}Np and Am doping (79.0 and 78.0 kg, respectively). However, there is an increase of 11% (^{237}Np doping) and 9% (Am doping) of the total amount of Pu with respect to the reference PBMR. This excess Pu is mainly due to the increase in ^{238}Pu amount for the doped fuel.

After determining the minimal concentrations of Am or ^{237}Np required for obtaining ^{238}Pu/Pu $\geq$ 6%, we calculate the penalties in reactor performance due to the introduction of these isotopes. The discharge burnup of fuel from a PBMR is taken to be 80 GW(d)/ton (see Table I). We calculate the behavior of the effective multiplication factor k_{eff} during burnup under the assumption that $k_\infty - k_{eff} = 5080$ pcm due to leakage,[37] as illustrated in Fig. 9, and find that at 80 GW(d)/ton, $k_{eff} = 0.959$ for the reference PBMR. The requirement that the doped fuel burnup cycle end at the same k_{eff} as that of the reference reactor results in the shortening of the cycle by 40 and 20 FPDs for the ^{237}Np and Am doping, respectively. This corresponds to reduction of 4.1 and 2.0 GW(d)/ton in fuel discharge burnup.

Another aspect of ^{241}Am doping is the shielding requirements during fuel fabrication. To address this aspect and estimate the effects of radiation hazards that will result from the ^{241}Am doping of the pebble fuel, we have compared it with fresh light water reactor (LWR) MOX-type fuel. The average plutonium content in commercial LWR MOX is 9.5% (Ref. 38) with 10% of ^{241}Pu of the total plutonium amount. The amount of ^{241}Am in MOX fuel is ~0.2% of the total fuel weight, when considering 5 years' storage between separation of plutonium and fabrication of the MOX fuel. For the pebble fuel, the amount of ^{241}Am during fabrication is 0.2% of the total fuel weight. Thus, the amount of ^{241}Am in doped pebbles is the same as in fresh commercial LWR MOX fuel. Consequently, shielding requirements
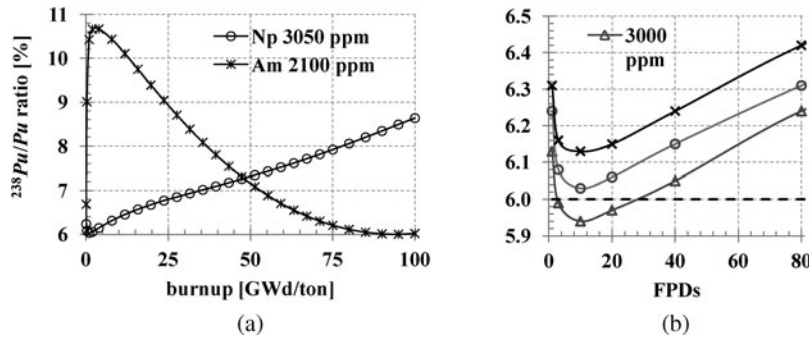

Fig. 7. (a) Weight ratio of ^{238}Pu to total plutonium as a function of fuel burnup for ^{237}Np and Am doping. (b) Zoom-in on the first 80 FPDs of the fuel cycle for the ^{237}Np doping at three different concentrations.
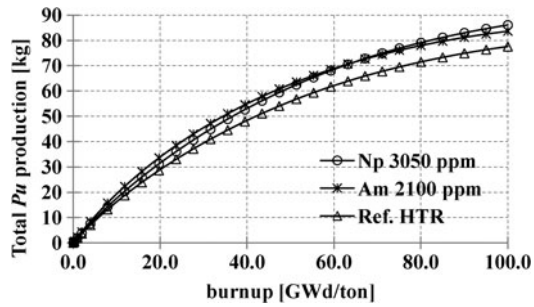
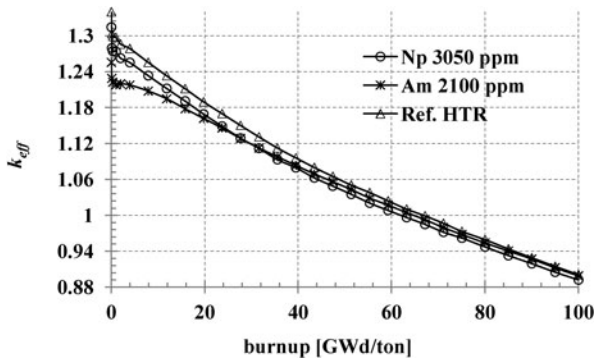Fig. 8. Total amount of plutonium produced during fuel burnup.

Fig. 9. k_{eff} as a function of fuel burnup.

for pebble fuel fabrication will be comparable to those of MOX fabrication, considering only ^{241}Am-related radiation hazards.

V. CONCLUSIONS

Denatured plutonium with the ratio ^{238}Pu/Pu $\geq$ 6% for the entire fuel burnup cycle can be achieved by adding either Am or ^{237}Np to the nuclear fuel with minimum concentrations of 2100 and 3050 ppm, respectively. However, ^{237}Np is more favorable than ^{241}Am for nuclear weapons production,[35,39] and the penalty it imposes on the fuel burnup cycle length is larger than that of Am. Note that the total amount of plutonium produced is increased by ~9% to 11% compared to a reference PBMR, mainly due to an increase in ^{238}Pu production. We conclude that ^{241}Am is a superior choice (to ^{237}Np) in doping PBMR fuel for its lower minimal concentration, reduced penalty on the fuel burnup cycle, and proliferation considerations.

We find that the qualitative behavior of the ratio ^{238}Pu/Pu with respect to burnup is different for the two scenarios. In the case of ^{237}Np, the fast initial decrease and then a monotonic increase of this ratio en-

ables us to use doping concentration smaller than 3050 ppm. For example, when using 3000 ppm of ^{237}Np, the production of plutonium with isotopic ratio ^{238}Pu/ Pu < 6% requires fuel discharge after ~10 to 20 days. The corresponding amount of plutonium produced is ~2 to 4 kg, which essentially makes proliferation impractical.

The penalty for ^{237}Np or Am fuel doping is the shortening of the fuel burnup cycle, compared to the reference PBMR, by ~40 and 20 FPDs, respectively. This corresponds to reduction of 4.1 and 2.0 GW(d)/ton in fuel discharge burnup. In addition, a large amount of spent fuel should be reprocessed in order to obtain the required ^{237}Np or Am amount. This will produce an additional amount of separated plutonium, which might increase the proliferation risk.

Considering only ^{241}Am-related radiation hazards, shielding requirements for pebble fuel fabrication will be the same as the requirements for the MOX fabrication. If one considers radiation hazards from additional elements in MOX, the shielding requirement for MOX fabrication turns out to be more demanding.

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