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Novel homogeneous burnable poisons in pressurized water reactor ceramic fuel



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ABSTRACT

Due to excess reactivity, fresh nuclear fuel often contains burnable poisons. This research looks at six different burnable poisons and their impacts on reactivity, material attractiveness, and waste management. An MCNP simulation of a PWR fuel pin was performed with a fuel burnup of 60 GWd/MTHM to determine when each burnable poison fuel type would decrease below a k_{∞} of 1. For determining the plutonium material attractiveness in each burnable poison fuel type, the plutonium isotopic content of the used fuel was evaluated using Bathke's Figure of Merit formula. For the waste management analysis, the thermal output of each burnable poison fuel type was determined through ORIGEN decay simulations at 100 and 300 years after being discharged from the core. The performance of all six burnable poisons varied over the three criteria considered and no single burnable poison performed best in all three considerations.

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1. Introduction

For neutron economy in a pressurized water reactor (PWR), at the beginning of a core cycle, burnable poisons are loaded in the fresh fuel for controlling core reactivity, in addition to using boric acid in the coolant. This approach enables a long-term reactivity control in a nuclear reactor without using excessive amounts of boric acid. The goal of using burnable poisons is to burn the poison (absorb neutrons) when the fuel is new and there is a large amount of excess reactivity and for the burnable poisons to be mostly consumed at the end of the fuel's life when excess reactivity is small. Common materials that are used as burnable poisons are boron and gadolinium [1,2]. These materials fit the overall properties of a burnable poison: having large absorption cross sections; burning out before the fuel is consumed; producing isotopes that have small absorption cross sections through neutron capture; and having the ability to not cause physical damage to the fuel or cladding [3].

Other materials have also been studied for use as burnable poisons such as erbium, americium, neptunium, cadmium, samarium, plutonium, and curium [2,4,5]. These materials have been studied for various reasons including reducing nuclear waste

and heat for long-term storage, attempting to produce specific isotopes, and improving the volume ratio of burnable poison to fuel in future fuel designs.

This paper will investigate six different types of burnable poisons and evaluate them using three different criteria: long-term reactivity of the fuel, plutonium material attractiveness, and waste heat in nuclear waste disposal. The long-term reactivity analyses were used to determine if the burnable poison could extend the life of the fuel while allowing the reactor to operate under normal conditions. The material attractiveness perspective focused on the plutonium isotopes created in the reactor and the ease of using this plutonium for nefarious purposes. The nuclear waste disposal study focused on the waste heat that is produced by the used fuel in long-term storage and the isotopes present in the used fuel.

2. Methodology

The Monte Carlo N-Particle (MCNP) radiation transport code version 6.1 was used to calculate the infinite multiplication factor, k_{∞} , of each burnable poison and the isotopic content of the used fuel. A typical PWR ceramic fuel rod was modeled in MCNP. The fuel model contained 4.5% enriched uranium at a temperature of 900 K inside a Zircaloy-IV cladding with a helium gas gap (900 K) separating the fuel and cladding. The cladding (600 K) was surrounded

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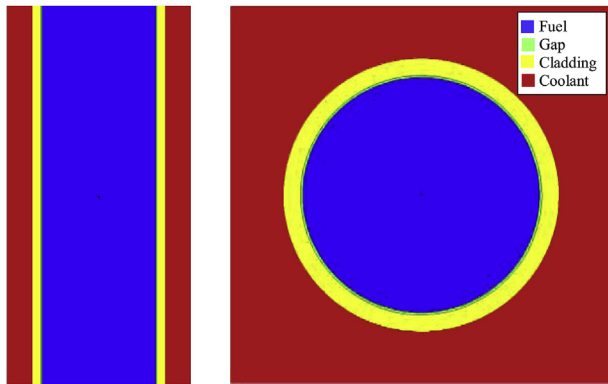


Fig. 1. Cross-sectional (left) and top-view (right) for the modeled PWR fuel.

by pure water (H_2O) at a temperature of 600 K. The cross-sectional and top-view are shown in Fig. 1 with the materials annotated by the legend. The dimensions for the model are listed in Table 1. The sides of the water channel were modeled as reflective boundaries thus causing the model to represent an infinitely large reactor.

Six burnable poisons were identified that were then homogeneously doped into the fuel portion of the fuel rod. The six burnable poisons used were two traditional elements: natural boron (B), natural gadolinium (Gd), and four novel materials: neptunium (Np), americium (Am), plutonium from used low-enriched uranium fuel (Pu LEU), and plutonium from used mixed oxide fuel (Pu MOX). The isotopic weight percentages used for the burnable poisons are shown in Table 2.

The MCNP model was used to perform a k_∞ analysis on each burnable poison fuel type. The weight percent of each poison doping in the fuel was modified until each fuel had an initial k_∞ of 1.250 ± 0.0002 . The weight percent concentrations of each burnable poison used throughout the rest of the MCNP simulations can be found in Table 5, in the results section.

Each burnable poison fuel type was burned at a power level of 96.87 kW in MCNP for 1100 days (two 18-month cycles) with the time steps as 0.25, 0.5, 0.75, 1, 8, 16, 32, 64, 100, 100, 100, 100, 100, 100, 100, 100, and 78 days. This burnup of 60 GWd/MTHM was used to assess the extension of the fuel lifetime beyond the typical 45 GWd/MTHM.

2.1. k_∞ vs Burnup

To evaluate the performance of each burnable poison fuel type, three different analyses were performed. The first analysis was to determine the time dependent change in k_∞ for each burnable poison fuel type and at what burnup the fuel became spent ($k_\infty = 1$). These results were also compared to the no-poison fuel type case. Table 3 highlights several of the burnable poison nuclides of interest and their most likely neutron absorption chain.

2.2. Material attractiveness

The second analysis was to assess the material attractiveness of the produced plutonium vectors. A commonly accepted analysis for determining material attractiveness combines the heat generation, spontaneous fission neutron production rate, critical mass of nuclear material, and dose rate to determine the attractiveness of the material [13]. This material attractiveness is represented by a Figure of Merit (FOM) value that determines whether the material falls within the ranges of attractive and unattractive, as shown in Table 4. This analysis is based on four commonly accepted limiting

Table 1
Dimensions for the modeled PWR fuel [6].

Material	Measurement (cm)
Fuel pin radius	0.3922
Helium gap thickness	0.00785
Cladding thickness	0.0572
Fuel pin height	380
Fuel rod (pin) pitch	1.26

cases: the threshold for low enriched uranium, the 80 atom percent ^{238}Pu limit, a “self-protecting” dose rate of 500 rad/h, and a spontaneous fission neutron production rate of reactor grade plutonium (6.8×10^6 n/s).

This analysis is separated into two cases. The first case considers technologically advanced proliferant states. This case excludes the spontaneous fission neutron production rate factor because it is believed by some that most technologically advanced states would be equipped to manage it. Equation (1) shows the relationship between the FOM value and the bare critical mass in units of kg (M), the heat generation in units of W/kg (h), and the dose rate of one fifth of the bare critical mass at 1 m in units of rad/h (D).

$$FOM = 1 - \log_{10} \left(\frac{M}{800} + \frac{Mh}{4500} + \frac{M}{50} \left(\frac{D}{500} \right)^{\frac{1}{\log_{10} 2}} \right) \quad (1)$$

The second case considers lesser technologically advanced proliferant states. This case includes all four limiting factors. Equation (2) shows the relationship between the FOM value and the bare critical mass of the metal in units of kg (M), the heat generation in units of W/kg (h), the spontaneous fission neutron production rate in units of n/s/kg (S), and the dose rate of one fifth of the bare critical mass at 1 m in units of rad/h (D).

$$FOM = 1 - \log_{10} \left(\frac{M}{800} + \frac{Mh}{4500} + \frac{MS}{6.8(10)^6} + \frac{M}{50} \left(\frac{D}{500} \right)^{\frac{1}{\log_{10} 2}} \right) \quad (2)$$

It should be noted that the plutonium vectors analyzed all came from fuel burned until k_∞ was approximately equal to 1.

2.3. Waste management

The third analysis focused on waste management. It was carried out to determine the net thermal power generated by the burned fuel rod for each burnable poison fuel type as the addition of actinides to the fuel impacts the heat produced by the used fuel. The heat produced by the used fuel is a long-term waste management issue for permanent geological storage that can be reduced by transmuting (burning) the long-lived high-heat producing actinides, such as plutonium and americium isotopes [14]. For estimating the heat produced by the used fuel at different decay times, ORIGEN decay simulations were performed. The actinide content at $k_\infty \approx 1$ from the MCNP simulations was used as the input and was decayed for 100 years and 300 years. This time period is considered by some to be the approximate duration a permanent geological repository would be externally cooled before being sealed [14]. The thermal power level of the used fuel when external cooling of a permanent geological repository is stopped determines the maximum fuel content safely allowed in the repository. The heat production results obtained by ORIGEN were used to calculate the thermal power produced by the fuel. In order to determine the net increase in long term thermal power for each burnable poison fuel type, the thermal power produced by the actinide poison (if any) in the fresh fuel was subtracted from the final thermal power

Table 2
Isotopic weight percent for the six different burnable poisons.

Poison	Isotopic Percentages	Reference
B	¹¹ B 80%, ¹⁰ B 20%	[7]
Gd	¹⁵² Gd 0.2%, ¹⁵⁴ Gd 2.18%, ¹⁵⁵ Gd 14.8%, ¹⁵⁶ Gd 20.47%, ¹⁵⁷ Gd 15.65%, ¹⁵⁸ Gd 24.84%, ¹⁶⁰ Gd 21.86%	[8]
Np	²³⁷ Np 100%	[9]
Am	²⁴¹ Am 58.66%, ^{242m} Am 0.22%, ²⁴³ Am 41.12%	[9]
Pu (LEU)	²³⁸ Pu 1%, ²³⁹ Pu 60%, ²⁴⁰ Pu 25%, ²⁴¹ Pu 9%, ²⁴² Pu 5%	[10]
Pu (MOX)	²³⁸ Pu 4%, ²³⁹ Pu 37%, ²⁴⁰ Pu 32%, ²⁴¹ Pu 16%, ²⁴² Pu 12%	[11]

Table 3
Neutron absorption chain and thermal neutron absorption (σ_a) and fission (σ_f) cross-section data for burnable poisons of interest [12].

¹⁰ B ($\sigma_a = 3838$ b)	→	¹¹ B ($\sigma_a = 0.005$ b)	
¹⁵⁵ Gd ($\sigma_a = 60,889$ b)	→	¹⁵⁶ Gd ($\sigma_a = 2.188$ b)	
¹⁵⁷ Gd ($\sigma_a = 254,078$ b)	→	¹⁵⁸ Gd ($\sigma_a = 2.496$ b)	
²³⁷ Np ($\sigma_a = 164.6$ b)	→	²³⁸ Pu ^a ($\sigma_a = 558.2$ b)	→ ²³⁹ Pu ($\sigma_a = 1017.7$ b)
($\sigma_f = 0.02249$ b)		($\sigma_f = 17.89$ b)	($\sigma_f = 747.4$ b)
²³⁸ Pu ($\sigma_a = 558.2$ b)	→	²³⁹ Pu ($\sigma_a = 1017.7$ b)	
($\sigma_f = 17.89$ b)		($\sigma_f = 747.4$ b)	
²⁴⁰ Pu ($\sigma_a = 289.4$ b)	→	²⁴¹ Pu ($\sigma_a = 1373.5$ b)	
($\sigma_f = 0.05877$ b)		($\sigma_f = 1012$ b)	
²⁴² Pu ($\sigma_a = 18.79$ b)	→	²⁴³ Am ^b ($\sigma_a = 78.62$ b)	
($\sigma_f = 0.002557$ b)		($\sigma_f = 0.1161$ b)	
²⁴¹ Am ($\sigma_a = 603.4$ b)	→	²⁴² Cm ^c ($\sigma_a = 21.54$ b)	→ ²⁴³ Cm ($\sigma_a = 747.6$ b)
($\sigma_f = 3.018$ b)		($\sigma_f = 5.064$ b)	($\sigma_f = 617.4$ b)
²⁴³ Am ($\sigma_a = 78.62$ b)	→	²⁴⁴ Cm ^d ($\sigma_a = 16.10$ b)	→ ²⁴⁵ Cm ($\sigma_a = 2347$ b)
($\sigma_f = 0.1161$ b)		($\sigma_f = 1.037$ b)	($\sigma_f = 2001$ b)

^a The half-life of ²³⁸Np is 2.117 days.

^b The half-life of ²⁴³Pu is 4.954 h.

^c The half-life of ²⁴²Am is 16.02 h.

^d The half-life of ²⁴⁴Am is 10.1 h.

Table 4
FOM weapons utility and material attractiveness range [13].

FOM	Weapons Utility	Attractiveness
>2	Preferred	High
2–1	Attractive	Medium
1–0	Unattractive	Low
<0	Unattractive	Very Low

Table 5
Weight percent for each burnable poison in the fuel that yields a k_{∞} of 1.250 ± 0.0002 .

Poison	Weight Percent (%)
B	0.0222
Gd	0.0135
Np	0.945
Am	0.362
Pu (LEU)	4.25
Pu (MOX)	1.47

produced by the used fuel. It should be noted that the actinide content of the used fuel all came from fuel burned until k_{∞} was approximately equal to 1.

3. Results

The first set of MCNP simulations on the modeled PWR fuel rod was carried out to determine the weight percent of poison doping for each identified burnable poison that would yield a k_{∞} of 1.250 ± 0.0002 . Table 5 lists the estimated weight percent for each poison material in the fuel. The weight percent for boron and gadolinium are significantly smaller than that of the actinides. This is due to the large thermal neutron absorption cross-section of

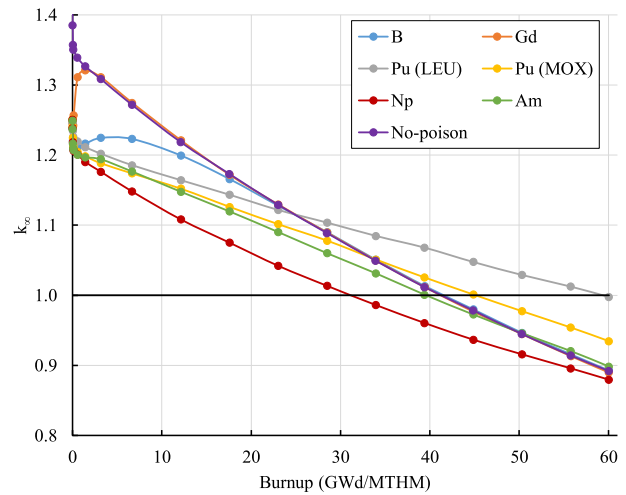


Fig. 2. The k_{∞} as a function of burnup for poison materials in Table 3 and no-poison fuel.

these materials, ¹⁰B (3838 b), ¹⁵⁵Gd (60,889 b), and ¹⁵⁷Gd (254,078 b). The Pu LEU fuel had the smallest effect on criticality due to its large ²³⁹Pu fissile content. For reference, the k_{∞} value of the fresh fuel rod without any poisons was 1.401 ± 0.0002 .

3.1. k_{∞} vs Burnup

After determining the burnable poison concentrations to get all the fuels at a starting k_{∞} value of 1.250, all the burnable poison fuel types (and the no-poison case) were burned for 60 GWd/MTHM. The results of the fuels k_{∞} value with respect to burnup can be seen in Fig. 2.

Table 6
Plutonium isotopic compositions (weight percent) for investigating the perceived material attractiveness.

Isotopes	No-poison	B	Gd	Np	Am	Pu (LEU)	Pu (MOX)
²³⁸ Pu	1.60	1.61	1.62	28.08	9.47	2.00	3.17
²³⁹ Pu	57.42	57.25	57.25	48.66	52.50	41.26	43.70
²⁴⁰ Pu	22.84	22.84	22.90	13.61	20.36	27.59	22.24
²⁴¹ Pu	13.59	13.67	13.63	8.03	12.11	19.24	16.98
²⁴² Pu	4.56	4.61	4.61	1.61	5.57	9.91	13.91
Fuel burnup [GWd/MTHM]	39.42	39.42	39.42	28.51	39.42	60.00	44.88

Table 7
FOM values for multiple plutonium vectors.

	No-poison	B	Gd	Np	Am	Pu (LEU)	Pu (MOX)
FOM Eq. 1	1.85	1.85	1.85	1.19	1.53	1.94	1.77
FOM Eq. 2	0.73	0.73	0.73	0.48	0.63	0.69	0.62

From Fig. 2 it can be seen that both traditional poisons, boron and gadolinium, exactly follow the no-poison case after they burnout. This is due to the fact that as they absorb neutrons they do not produce fissile material. While both of these materials suppress the initial k_{∞} value they have no impact on the long term criticality lifespan of the fuel. The curves of the four actinide poisons in Fig. 2 are all less steep than the no-poison case. This is attributed to the ability of these materials to become fissile after absorbing a neutron, e.g. $n + {}^{240}\text{Pu} \rightarrow {}^{241}\text{Pu}$. Both neptunium and americium (²⁴¹Am and ²⁴³Am) shorten the criticality lifespan of the fuel compared to the no-poison case. The attributed cause for this reduced lifespan is that all three of these nuclides require two neutron captures before becoming a highly fissionable nuclide (see Table 3). At extrapolated burnups of 60 GWd/MTHM, the neptunium and americium poisons have transmuted enough atoms into highly fissionable nuclides to compensate for the remaining burnable poison nuclides, resulting in a k_{∞} value similar to that of the extrapolated no-poison case. The addition of the plutonium burnable poisons increased the criticality lifespan of the fuel compared to the no-poison case. This is attributed to the fact that the burnable poisons ²³⁸Pu and ²⁴⁰Pu become highly fissionable nuclides after only one neutron capture. The Pu (MOX) case was less effective at extending the criticality lifespan of the fuel than Pu (LEU) due to its higher content of ²⁴²Pu, which requires three neutron captures before becoming the highly fissionable nuclide ²⁴⁵Cm.

3.2. Material attractiveness

The k_{∞} analysis showed how the presence (or production) of plutonium isotopes can affect the reactivity in the core. Additionally, the production of plutonium is a proliferation concern, especially following the fuel removal from a nuclear reactor. To quantify the perceived material attractiveness of each burnable poison fuel

Table 8
Contributions of each term in the FOM evaluation.

FOM terms	No-poison	B	Gd	Np	Am	Pu (LEU)	Pu (MOX)
$\frac{M}{800}$	0.028	0.028	0.028	0.024	0.028	0.032	0.032
$\frac{Mh}{4500}$	0.113	0.114	0.114	0.619	0.267	0.083	0.136
$\frac{MS}{6.8(10)^6}$	1.701	1.708	1.710	2.703	2.077	1.909	2.236
$\frac{1}{50} \left(\frac{D}{500} \right) \log_{10} 2$	3.28×10^{-17}	3.35×10^{-17}	3.41×10^{-17}	2.61×10^{-13}	1.03×10^{-16}	1.03×10^{-16}	4.65×10^{-16}

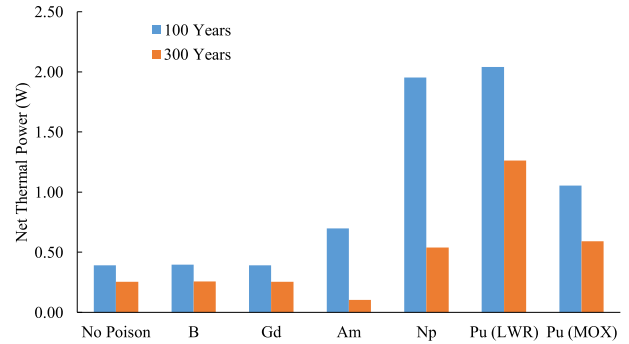


Fig. 3. Net thermal power produced by the actinides for each burnable poison fuel type per fuel rod.

type, an analysis using Bathke's method was conducted. The plutonium vector from each of the burned poison fuel types is listed in Table 6.

Table 7 shows the FOM values for the plutonium vectors shown in Table 6. From the perspective of a technologically advanced proliferant state (Eq. (1)), all eight plutonium vectors can be considered attractive, but not preferred. However, the neptunium doped fuel FOM is noticeably close to 1 due to its high concentration of ²³⁸Pu. For all of the fuel types, ²³⁸Pu content is the dominant factor in determining the FOM value due to the minimal contributions to radiation dose and variations in material criticality from the plutonium isotopes. From the perspective of a less technologically advanced proliferant state (Eq. (2)), all seven plutonium vectors are unattractive. This is primarily due to the spontaneous fission neutron production rate caused by the concentration of the even plutonium isotopes. A breakdown of how each of the four terms in the FOM formula (Eq. (2)) contribute to the overall FOM value are shown in Table 8.

3.3. Waste management

The net thermal power of the actinides in the used fuel, adjusting for the thermal power of the added poison in the fresh fuel, is shown in Fig. 3 at 100 years and 300 years when fuel placed in permanent geological storage might be sealed from external

Table 9

Masses of actinides of interest from each fuel type at 100 years of cooling time. Values shown are per fuel rod and have units of g.

Actinide	No-poison	B	Gd	Np	Am	Pu (LEU)	Pu (MOX)
²³⁸ Pu	0.143	0.147	0.145	3.02	1.16	0.708	0.530
²³⁹ Pu	10.6	10.6	10.5	11.5	11.4	27.3	14.6
²⁴⁰ Pu	4.20	4.24	4.21	3.20	6.04	19.5	8.40
²⁴¹ Am	2.22	2.26	2.22	1.68	2.62	11.6	5.14
²⁴³ Am	0.146	0.150	0.147	0.051	1.10	2.03	1.478
²⁴⁴ Cm	1.07×10^{-3}	1.10×10^{-3}	1.09×10^{-3}	2.45×10^{-4}	3.82×10^{-2}	3.44×10^{-2}	2.37×10^{-2}
Fuel burnup [GWD/MTHM]	39.42	39.42	39.42	28.51	39.42	60.00	44.88

Table 10

Masses of actinides of interest from each fuel type at 300 years of cooling time. Values shown are per fuel rod and have units of g.

Actinide	No-poison	B	Gd	Np	Am	Pu (LEU)	Pu (MOX)
²³⁸ Pu	0.030	0.030	0.030	0.621	0.239	0.146	0.109
²³⁹ Pu	10.5	10.5	10.4	11.4	11.3	27.2	14.5
²⁴⁰ Pu	4.12	4.16	4.13	3.13	5.95	19.1	8.25
²⁴¹ Am	1.63	1.65	1.63	1.23	1.92	8.52	3.76
²⁴³ Am	0.143	0.147	0.144	0.050	1.08	2.00	1.45
²⁴⁴ Cm	5.05×10^{-7}	5.19×10^{-7}	5.12×10^{-7}	1.16×10^{-7}	1.80×10^{-5}	1.62×10^{-5}	1.12×10^{-5}
Fuel burnup [GWD/MTHM]	39.42	39.42	39.42	28.51	39.42	60.00	44.88

cooling. The tabulated mass values of actinides of interest are shown in Table 9 at 100 years and Table 10 at 300 years.

The boron and gadolinium cases produce the same thermal power as the no-poison case. This is expected since boron and gadolinium do not produce any thermal power themselves and have little effect on the number of neutron capture events in the uranium. The americium case shows a net decrease in thermal power at 300 years compared to the no-poison case but an increase at 100 years. This is attributed to the fact that at 300 years of cooling time ²⁴¹Am is the primary source of thermal power in used fuel. By using the net thermal power as the waste management metric, ²⁴¹Am that is transmuted during the fuel burnup is subtracted from the final amount of heat produced in the used fuel. It should be noted that analyses using this metric implicitly assume that some fraction of americium destined for permanent geological storage can be recycled into fresh PWR fuel. The increase in thermal power at 100 years is caused by an increased content of ²³⁸Pu due to neutron capture reactions with ²⁴¹Am followed by a beta (16 h) and alpha (163 days) decay to ²³⁸Pu. The neptunium case shows more than a doubling in thermal power compared to the no-poison case. This is attributed to the large buildup of ²³⁸Pu in the used fuel from neutron captures by ²³⁷Np followed by beta decay (see Table 3). The Pu (LEU) case has the highest thermal power of all the cases. This is attributed to the buildup of ²⁴¹Pu in the used fuel which nearly all decays to ²⁴¹Am after 300 years ($T_{1/2}$ of ²⁴¹Pu = 14.35 years).

4. Conclusions

The k_{∞} vs burnup analysis of the burnable poisons highlighted that traditional poisons such as boron and gadolinium have no effect on the long term criticality lifespan of the fuel compared to the no-poison case and that the rate of criticality loss per unit burnup is the same after 20 GWD/MTHM. While all the actinide burnable poisons did reduce the rate of criticality loss with respect to burnup, both the americium and Pu (MOX) case had minimal change in the burnup at a k_{∞} value of 1 compared to the no-poison case. The neptunium burnable poison reduced the criticality life-span by approximately 25% and the Pu (LEU) case increased it by approximately 50%.

The material attractiveness analysis on the burned fuel types revealed that the neptunium doped fuel produces a plutonium vector that is the least attractive by both FOM equations. The FOM

values determined using Eq. (1) are primarily effected by ²³⁸Pu content (heat production) and Eq. (2) values are primarily effected by the content of even plutonium isotopes (neutron emission rate).

The waste management analysis showed that using americium as a burnable poison minimizes the 300 year thermal power heat load in a permanent geological repository and that neptunium, Pu (LEU), and Pu (MOX) all increase the heat load. The burnable poisons boron and gadolinium had no noticeable effect.

Overall, this research investigates a novel set of burnable poisons that can impact the reactivity control, material attractiveness, and waste management requirements while comparing them against the traditional no-poison fuel case. While each identified burnable poison offered a unique result that can be beneficial depending on the objective, the best extension of fuel burnup is Pu (LEU), neptunium produces the least attractive plutonium for proliferation, and americium reduces the heat burden for a permanent geological repository. Boron and gadolinium had no effect on any of the three evaluation criteria.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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