THORIUM BLENDED AND REGULAR MOX BURN-UP STUDIES FOR FAST REACTOR FUEL CYCLE SAFEGUARDS

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ABSTRACT

Fast reactor fuel cycle (FRFC) is regaining importance because of its vital role in the long term development of nuclear power. The safety issues associated with the fast reactors are now well understood and newer designs can address those from the lessons learned in the past. The same cannot be ascertained about their safeguards issues. Primary reason being the presence of special nuclear material (SNM) in very high concentrations inside fresh and spent fuel of fast reactors, making it more attractive to the Proliferators and leads to less proliferation resistance (PR). Advantageously changing the fuel material without adversely affecting the normal reactor operations could improve the PR and in turn enhance the safeguards of the FRFC. The intrinsic PR characteristics estimated through computational fuel burn-up analysis of a regular and a thorium blended MOX for a typical fast reactor system is presented here. Detailed core physics computations showed that, employing either of these fuel types has insignificant effect on the reactor operations. However, build-up of denaturing material ^{232}U (1300 to 1500 PPM by weight of ^{233}U) along with the fissile material ^{233}U during the burn-up of thorium blended MOX makes the spent fuel relatively less attractive to the Proliferators. The daughter products in the decay chain of ^{232}U are significant gamma emitters and in particular, ^{208}Tl emits gamma rays with energy of 2.6MeV, which not only makes the fissile material low grade but also pose handling difficulties. It is observed that the photon emission rate in the range of 2.25MeV to 2.75MeV from the actinides of a thorium blended spent fuel is higher by a factor of ~4000 compared to the regular MOX spent fuel. These gamma rays provide a distinctive signature that can be used to detect and track SNM. Also, thorium blended fresh fuel will have 2.6MeV signature because ^{208}Tl is a daughter product in the natural decay chain of ^{232}Th. It is concluded from the study that deployment of thorium blended MOX fuel in place of regular MOX fuel in a fast reactor should help in improving the PR characteristics and enhancing the safeguards of a FRFC.

INTRODUCTION

Varieties of nuclear reactors with fast neutron spectrum are in the research and development stages for future deployment and can play a vital role in the long term development of nuclear power [1] [2] [3]. Nuclear proliferation resistance (PR) characterization of spent fuel discharged from one such fast reactor (FR) is the subject matter of this paper. The PR characterization is necessary to develop safeguards approaches for the fast reactor fuel cycle (FRFC). Fuel assembly design selected for the study is that of the Indian FR, based on the information available in open literature [2] [3]. Indian FR core utilizes two kinds of fuel assemblies, namely inner and outer core assemblies. The difference is in the amount of reactor grade plutonium contained in them. Inner core fuel assemblies have less amount of plutonium compared to the outer core fuel assemblies to achieve power flattening. Both, inner and outer core fuel assemblies are made from mixed oxides (MOX) of PuO_2 and depleted UO_2. The ratios of PuO_2:UO_2 are 21:79 and 28:72 for the inner and outer core assemblies respectively. Abundance of thorium in India and the better neutronic

characteristics will make its deployment into the Indian FR cores in near future [4]. Computational investigations of neutronic and nonproliferation characteristics were carried out by modifying the Indian FR design by blending in a reasonable amount of thorium in the core without bringing in any major changes to the engineered safety features [5]. Each modified fuel assembly contains 90 (PuO_2-ThO_2) MOX fuel pins in the two outermost layers, which replaces the (PuO_2-UO_2) MOX fuel pins, with no changes to the plutonium content. There are 217 MOX fuel pins in each assembly. Figure 1 illustrates the configuration of fuel pins in the original Indian FR design **(case-1)** and the modified one employed in the present study **(case-2)**. Current paper presents a detailed proliferation resistance analysis of a typical spent fuel assembly focusing on the outer core fuel assembly, as it contains more plutonium.

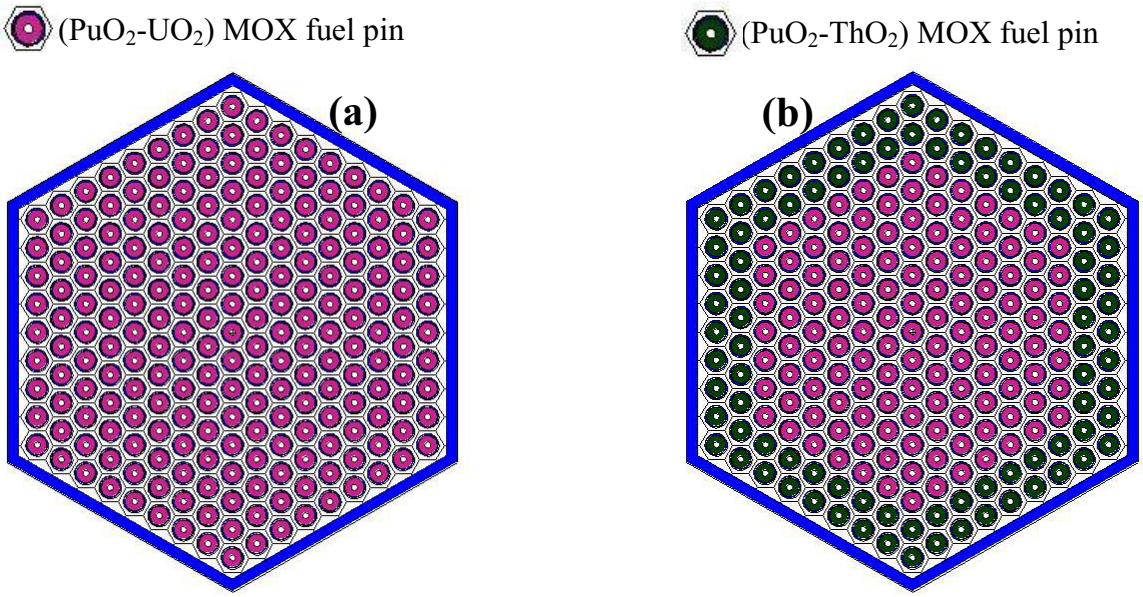

Figure 1: Fuel pin configurations of FR assemblies used for the burn-up case studies (a) original Indian design: **case-1** (b) modified for the present study: **case-2**

FUEL ASSEMBLY BURN-UP COMPUTATIONS

Original (case-1) and thorium blended (case-2) configurations of an outer core fuel assembly were subjected to burn-up analysis using depletion/burn-up code, ORIGEN2.2 [6]. Both fuel assemblies underwent a burn-up of ~70GWd/t, which is about the average fuel burn-up at discharge in the FR core under investigation. There are a large number of possible cross-section data bases in ORIGEN2.2 computer code library since the one-group cross sections employed in the code are highly reactor- and fuel type specific. Hence, the cross-section library for the uranium-plutonium cycle LMFBRs (liquid metal fast breeder reactor) was selected for the present study. Output tables generated for the spent fuel assembly included photon source strengths (in 18 representative energy groups), isotope wise spontaneous neutron source strength, nuclide depletion table as a function of burn-up, etc. Data on inventory of key nuclides in fresh and spent fuel assembly for both cases are shown in Table 1. It is noticeable from the Table 1 that either fresh or spent fuel assembly, in both cases, contain 1 significant quantity (SQ) of plutonium; one SQ being the approximate amount of

nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded [7]. One SQ for plutonium and ^{233}U is 8kg.

Table 1: Key nuclide inventory data of fresh and spent fuel assembly for case-1 and case-2

Name of nuclide	Case-1		Case-2	
	Fresh Assembly (grams)	Spent Assembly (grams)	Fresh Assembly (grams)	Spent Assembly (grams)
^{238}U	3.188E+04	3.004E+04	1.886E+04	1.775E+04
^{235}U	7.891E+01	5.135E+01	4.728E+01	3.136E+01
^{233}U	0.000E+00	7.463E-06	0.000E+00	6.164E+02
^{232}U	0.000E+00	1.320E-05	0.000E+00	7.779E-01
^{238}Pu	0.000E+00	3.186E+00	0.000E+00	2.785E+00
^{239}Pu	8.416E+03	6.903E+03	8.257E+03	6.228E+03
^{240}Pu	3.022E+03	3.201E+03	2.964E+03	3.118E+03
^{241}Pu	6.488E+02	5.706E+02	6.419E+02	5.615E+02
^{242}Pu	1.672E+02	1.906E+02	1.575E+02	1.822E+02
^{232}Th	0.000E+00	1.446E-07	1.259E+04	1.170E+04

ANALYSIS

(a) Photon emission spectra obtained in 18 energy groups due to actinides and its daughters present in the spent fuel assembly for cases 1&2 and for subsequent cooling periods of 1 year, 5 years and 50 years are shown in Figure 2. Similar spectra obtained for a typical pressurized water reactor (PWR) spent fuel assembly with a burn-up of 33GWd/t is shown in Figure 3 for comparison purposes. From Figure 2, it can observed that the significant difference between the two cases is that the photon emission rate in the range from 2.25MeV to 2.75MeV for thorium blended spent fuel (case-2) is higher by a factor of ~4000 compared to the regular MOX spent fuel (case-1). This is due to the build-up of ^{232}U (~1300ppm) along with the fissile material, ^{233}U, in those 90 peripheral (PuO_2-ThO_2) MOX pins of case-2 fuel assembly. ^{232}U acts as a denaturing material for the fissile material ^{233}U. The daughter products in the decay chain of ^{232}U are significant photon emitters. In particular, ^{208}Tl emits photons with energy of 2.6MeV. These energetic photon emissions could be advantageously utilized for the detection of spent fuel assembly movements. However, the presence of photon emissions from fission products of the spent fuel assembly can be a challenge in the photon spectrometry of spent fuel assemblies (refer Figure 4).

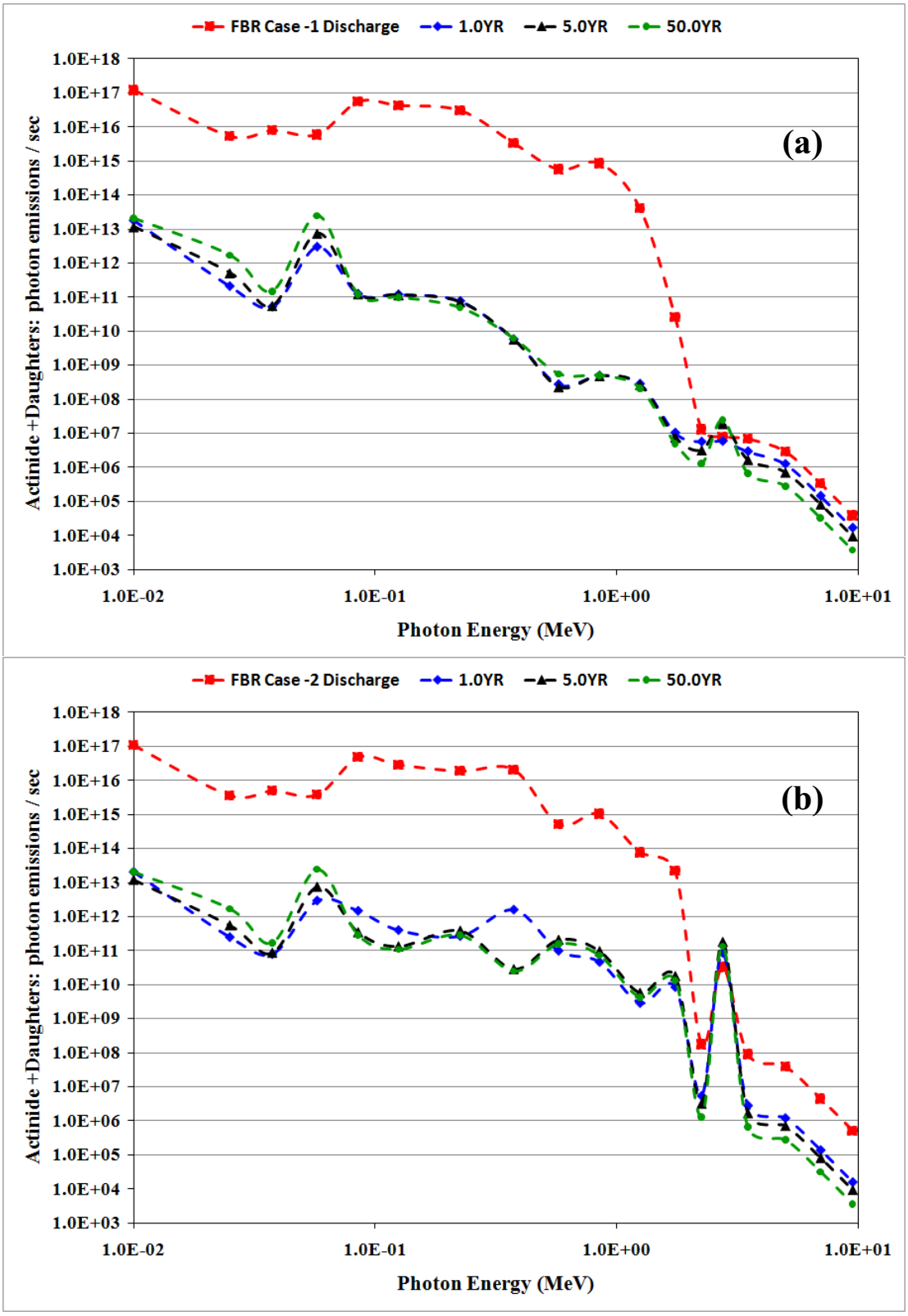

Figure 2: Photon emission spectra in 18 energy groups due to actinides + daughters of spent fuel assembly (a) case-1 (b) case-2 and for subsequent cooling periods of 1year, 5 years and 50 years

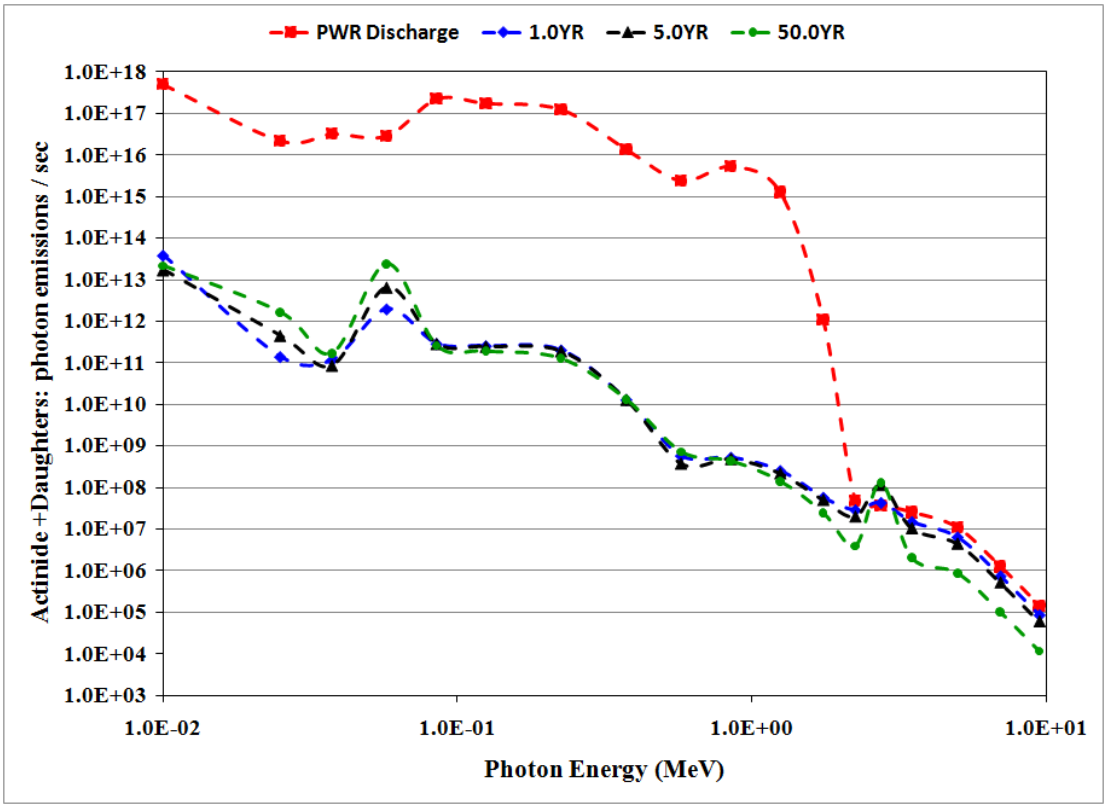

Figure 3: Photon emission spectra in 18 energy groups due to actinides + daughters of a typical PWR spent fuel assembly (33GWd/t) burn-up and for subsequent cooling periods 1year, 5 years and 50 years

(b) Total spontaneous fission neutron emission rates (neutrons/second) for case-1 and case-2 spent fuel assemblies are 6.22E+07 and 6.20E+07, they are quite the same in magnitude. These emissions mostly are by ^{242}Cm, ^{244}Cm, ^{240}Pu and ^{242}Pu in both cases. Again for the purpose of comparison for the PWR spent fuel assembly, the spontaneous fission neutron emission rate is 2.42E+08neturons/second. Hence, the detection of spent fuel assembly movements utilizing the spontaneous fission neutron emission will yield same results irrespective of regular MOX or thorium blended MOX configurations studied here.

(c) Spent fuel is usually recycled in FRFC after reprocessing it by which fission products and higher actinides are practically removed to obtain plutonium and uranium separately. Plutonium obtained from spent fuel reprocessing can still be tracked from its spontaneous fission neutron emissions. However, tracking of ^{233}U obtained from reprocessing the case-2 spent fuel assembly is not feasible using spontaneous fission neutrons because they are sparse. But, tracking can be done with the help of ^{232}U contamination accompanying ^{233}U in amounts ranging from 1300 to 1500ppm. The daughter products in the decay chain of ^{232}U are significant photon emitters. In particular, ^{208}Tl emits photons with energy of 2.6MeV. The effect of fission product photon emissions would have been practically eliminated at the reprocessing stage and should not interfere in photon spectrometry based tracking of $^{233+232}$U.

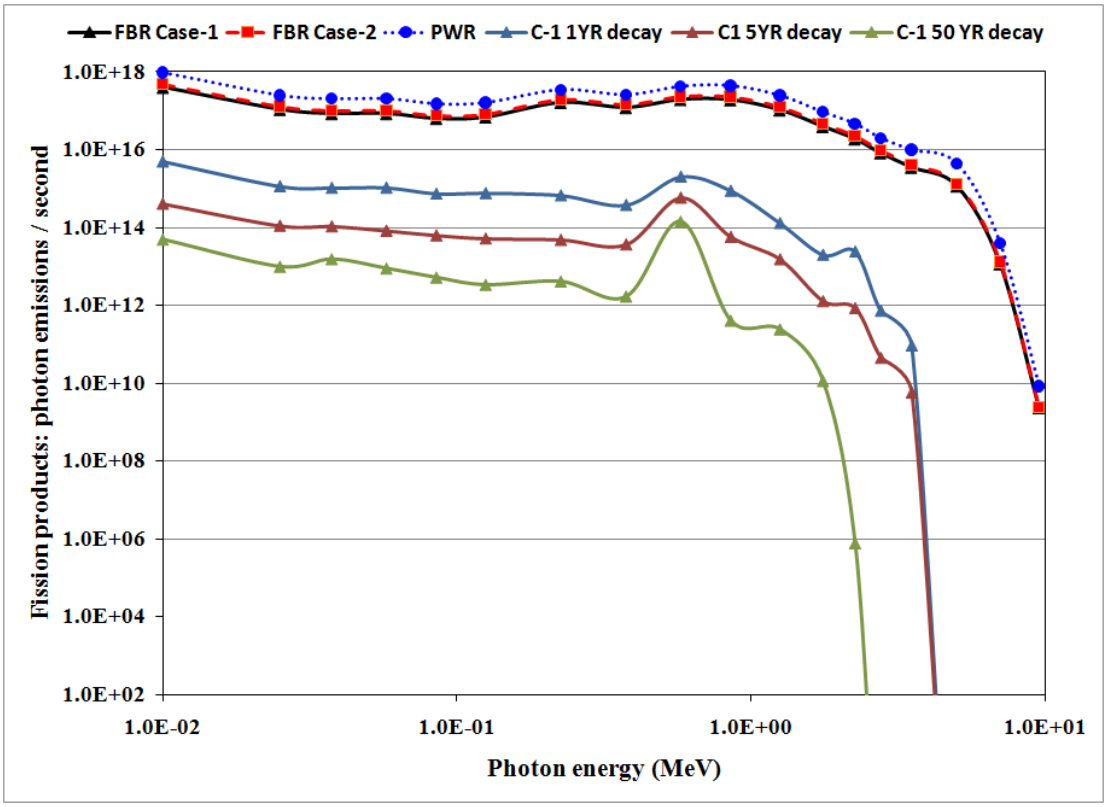

Figure 4: Photon emission spectra in 18 energy groups due to fission products of spent fuel assembly

(d) For the purpose of proliferation resistance comparison, dose rates from 8 kg (1 SQ) of reactor grade Pu and 8 kg (1 SQ) of ^{233}U denatured with 1500ppm of ^{232}U both derived from reprocessing 2 years cooled FR spent fuel assembly were computed at a distance of 1m using Monte Carlo radiation transport code, MCNP5 [8]. The dose rates obtained are 0.045mSv/h and 140mSv/h respectively (refer Table 2). Similar dose rates have been reported by Kang *et al* [9]. Even though the denatured ^{233}U is not self protecting as per IAEA definition of 1000mSv/h [10], it is better compared to reactor grade plutonium from this point of view.

(e) Apart from the dose rates at a distance of 1m, other proliferation resistance characteristics for one SQ sphere of plutonium and ^{233}U are compared in Table 2. Plutonium has about 4% higher alpha radio-toxicity compared to ^{233}U denatured with 1500ppm of ^{232}U. The decay heat (MeV/s) for reactor grade plutonium is only 1.7E+11 whereas it is 7.8E+12 for denatured ^{233}U with 1500ppm ^{232}U. Spontaneous fission neutron emission is very low for 1 SQ of ^{233}U.

(f) Diversion of 1 spent fuel assembly from an equilibrium FR core is sufficient to derive 1SQ of plutonium for case-1 without ^{232}U contamination in it. If 1 SQ of plutonium is to be diverted for case-2, still only one assembly needed to be diverted, but it contains ^{232}U contamination as an extra handling difficulty for the proliferators. Also, for second case it needs to undergo both PUREX and THOREX spent fuel reprocessing. To obtain 1 SQ of ^{233}U from case-2, about 1200 thorium blended fuel pins from 13 spent fuel assemblies need to be diverted, which in turn increases the probability of detection.

Table 2: Proliferation resistance characteristics for 1SQ of FBR grade spent plutonium* and 1 SQ of $^{233}U^{+}$

Fissile Material (2 years decayed)	**Proliferation resistance characteristic**			
	Gamma Dose Rate at 1m (mSv/h)	Spontaneous fission neutron emission (neutrons/sec)	Alpha radioactivity (MBq)	Decay heat (MeV/s)
1 SQ of Pu*	4.50E-02	2.39E+06	3.80E+07	1.70E+11
1 SQ of $^{233}U^{+}$	1.40E+02	1.84E+01	3.65E+07	7.8E+12

* $^{238}Pu/^{239}Pu/^{240}Pu/^{241}Pu/^{242}Pu$ wt%: 0.03/63.52/29.45/5.25/1.75

+ contains 1500ppm of ^{232}U

CONCLUSIONS

Fuel burn-up computations and analyses of Indian fast reactor (PuO_2-UO_2) MOX fuel assembly design and a modified one with (PuO_2-ThO_2) in its two outer layers of peripheral fuel pins were carried out. The results of the burn-up computations allowed performing the proliferation resistance characterization for both of these fuel assemblies for safeguards approaches for the fast reactor fuel cycle. Following key observations are made from the characterization:

- Spontaneous fission neutrons or fission product photon emissions can be utilized to track the movement for both types of fuel assemblies because their magnitudes are not significantly different. However, Proliferators will have more handling difficulty with respect to the diversion of thorium blended fuel assembly. This is because of the enhanced radiation dose rates from the daughter products of ^{232}U contamination in the peripheral fuel pins of the fuel assembly.
- Fissile materials plutonium and ^{233}U obtained from spent fuel reprocessing of each type have its own distinctive intrinsic proliferation resistance signatures. 1 SQ of plutonium has a spontaneous fission neutron emission rate of 2.39E+06neutrons/sec, where as it is negligible for 1 SQ of ^{233}U. Instead, 1 SQ of ^{233}U denatured with 1500ppm of ^{232}U has a photon emission rate of 1.67E+12photons/sec in the energy range of 2.25MeV to 2.75MeV, mostly by the decay of ^{208}Tl, a daughter product in the decay chain of ^{232}U.
- The future deployment of thorium in fast reactors will only enhance the intrinsic proliferation resistance of fuel assemblies/fissile materials with respect to their handling difficulty to the proliferators and higher probability of detection.

ACKNOWLEDGEMENTS

The work reported here is taken up under the sponsorship of USDOE National Nuclear Security Administration's office of Non-proliferation and International Security as per the contract No.DE-FG52-06NA27606. Special thanks are due to Prof. William S. Charlton of Texas A&M University

for the fruitful technical discussions the authors had with him for the accomplishment of the work reported here.

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