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Modelling of HTRs with Monte Carlo: Sensitivity due to Different Isotopic Fuel Composition

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The gas turbine modular helium-cooled reactor (GT-MHR) is a potential candidate for the maximum plutonium destruction in once-through cycle. A particular feature of GT-MHR is that its refractory coated fuel (TRISO particles) is supposed to provide an impermeable barrier to the release of fission products and, at the same time, to resist very deep burn-up rates (more than 90% for ^{239}Pu).

In this work we performed detailed Monte Carlo simulations of the GT-MHR operation by loading with different plutonium fuel vectors: plutonium from military applications, plutonium from LWR and RBMK spent nuclear fuel. The comparison of the main GT-MHR performance parameters: k_{eff} eigenvalues, the length of the fuel cycle, neutron characteristics and the evolution of fuel composition in particular were obtained. We show that the performance of GT-MHR may be considerably influenced by the plutonium isotopic composition vector used as initial fuel material. This is the first time when incineration possibility of the RBMK-1500 based plutonium isotopic composition was tested using high temperature reactor. Finally, the propagation of statistical errors for the fuel burn-up was examined in detail showing no uncertainty accumulation.

KEYWORDS: High Temperature Reactor, Monte Carlo, Pu Incineration

1. Introduction

In principle, GT-MHR can be used to burn all types of fuel and offers significant advantages in accomplishing the transmutation of plutonium isotopes and high destruction of ^{239}Pu in particular [1]. The GT-MHR features are: the helium coolant, which remains in a single phase under all conditions; the graphite core, which provides high heat capacity, slow thermal response, and structural stability at high temperatures; the refractory coated particle fuel, which allows high burn-up, retains fission products during operation in high temperatures and retains their integrity in a repository environment for hundreds of thousands years [2]. It utilizes natural erbium as a burn-able poison with the capture cross section having a resonance at a neutron energy such that ensures a strong negative temperature coefficient of reactivity. The lack of interaction of neutrons with coolant (helium gas) makes sure that temperature feedback of fuel and graphite is the only significant contributor to the power coefficient. These features are combined with a closed Brayton energy conversion cycle giving a net efficiency in the range of 45-47 % [2].

In this work we performed detailed simulations of GT-MHR operation by loading it with different Pu fuel vector: plutonium isotopes separated from LWR and RBMK-1500 spent nuclear fuel and plutonium from military applications. The main goal of our study is to compare the main GT-MHR reactor performance parameters as k_{eff} - values, the length of the fuel cycle, neutron characteristics, temperature coefficients, and the evolution of fuel composition as a function of different initial fuel isotopic composition. The modelling of GT-MHR and plutonium based fuel cycles within a Monte

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Carlo approach was made using double-heterogeneous reactor core geometry including micro-particles [3]. The propagation of Monte Carlo errors was addressed in detail in the last section of this paper. This is the first time when transmutation possibility of the plutonium, originating from the RBMK-1500 spent nuclear fuel (Ignalina nuclear power plant in Lithuania) was tested using GT-MHR.

2. Modeling details

The investigation of transmutation feasibility of different isotopic plutonium composition in GT-MHR reactor is based on 3D double-heterogeneous reactor core geometry with micro-particles as described in our recent studies [3, 4]. In brief, the main feature of a double-heterogeneous geometry is an exact active core structure, where fuel and erbium compacts include ceramic - coated fuel and erbium particles mixed uniformly in a graphite matrix. Each coated fuel particle consists of spherical kernel of PuO_{2-x} (200 μm diameter) surrounded by triple protective coatings from pyrocarbon and SiC (so called TRISO coating). The same structure is valid for particles containing burn-able poison - natural erbium in the form of Er_2O_3.

In this work we used the MCNP code and associated data libraries to obtain k_{eff}, neutron fluxes and temperature coefficients. As soon as k_{eff} is smaller than 1 (within statistical errors) the length of the fuel cycle is determined. In the more realistic description of GT-MHR, control rods would need to be modeled. Here the control rods were not designed since their effect would be very similar in the case of a comparative analysis (relative comparison) if modeled-omitted exactly in the same way. For burn-up calculations we employed the Monteburns code (MCNP+ORIGEN) [5]. ENDF/B-VI data library (as the most often employed with MCNP) was used for the fuel and structure materials, JENDL-3.2 data files (having the biggest number of fission products) were employed for fission products. Neutron spectrum in GT-MHR changes considerably during the fuel burn-up [3]. In this work the neutron fluxes and cross sections were recalculated in every fifty days for the follow-up ORIGEN burn-up calculations.

As it was mentioned above, our major interest in this study was to look at the GT-MHR performance parameters by investigating different plutonium fuel isotopic configuration, i.e. plutonium originating from: a) the spent nuclear fuel of LWR (light water reactor), b) the spent nuclear fuel of RBMK - 1500 (graphite - moderated water - cooled reactor), and c) plutonium found in military applications (see Table 1).

Table 1. Different GT-MHR fuel and erbium load at the beginning of the fuel cycle.*Note that in RBMK Pu1 case GT-MHR performance parameters where investigated two times – with burnable poison erbium (in Figure 1), and without erbium (in Figure 2).

Fuel	Military Pu	LWR Pu	RBMK Pu1*	RBMK Pu2	RBMK Pu3	RBMK Pu4
Pu composition, kg					7.0	9.5
^{238}Pu	--	32.4	2.5	6.0	685.5	652.2
^{239}Pu	659.0	661.0	802.1	673.3	429.6	439.0
^{240}Pu	7.8	277.0	361.8	443.1	13.0	14.1
^{241}Pu	4.2	142.0	8.4	12.4	64.9	85.2
^{242}Pu	--	87.6	25.2	65.2		
Total plutonium, kg :	701.0	1200.0	1200.0	1200.0	1200.0	1200.0
Er composition, kg :						
^{166}Er	132.0	16.8	18.0	--	--	--
^{167}Er	94.2	11.4	12.2	--	--	--
^{170}Er	61.4	7.5	8.0	--	--	--
Total erbium, kg :	293.6	35.7	38.2	--	--	--

Since RBMK-1500 SNF (Spent Nuclear Fuel) isotopic composition strongly depends on the initial

^{235}U enrichment and its final burn-up, for the RBMK plutonium several cases were tested:
"RBMK Pu1" – 2.0 % ^{235}U initial enrichment, 14 MWd/kg burn-up,
"RBMK Pu2" – 2.0 % ^{235}U initial enrichment, 20 MWd/kg burn-up,
"RBMK Pu3" – 2.4 % ^{235}U initial enrichment, 22 MWd/kg burn-up,
"RBMK Pu4" – 2.6 % ^{235}U initial enrichment, 26 MWd/kg burn-up.

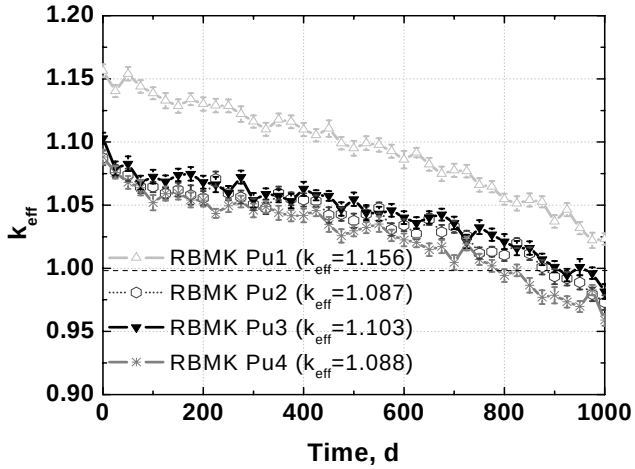


Figure 1. A behaviour of k_{eff} for different RBMK plutonium cases without burnable poison in GT-MHR.

RBMK-1500 reactor spent fuel composition was calculated using SCALE 4.4a code package (with ENDF/B-V nuclear data library) [6].

The corresponding GT-MHR plutonium fuel masses for each composition are presented in Table 1. 1200 kg for civil plutonium cases and 700 kg for military plutonium were the optimal fuel loads for GT-MHR. The burnable poison (erbium) in natural isotopic composition was added to reactor core to achieve the same k_{eff} eigenvalue at the beginning of the fuel cycle for LWR Pu, RBMK Pu1 and Military Pu. A high load of ^{240}Pu in other three cases Pu2, Pu3 and Pu4 limited the possibility to add burnable poison (see next section).

3. Burn-up results

We perform our calculations by simulating the once-through fuel cycle at the constant 600 MW_{th} power. Our primary goal was to check the RBMK plutonium as an initial GT-MHR fuel load. For this purpose GT-MHR performance parameters were analyzed for four RBMK fuel cases without burnable poison. The $k_{eff} \pm 1\sigma$ time dependence (consequently, its dependence on burn-up) for RBMK Pu1, Pu2, Pu3 and Pu4 compositions are presented in Figure 1. The most attractive RBMK plutonium composition is that of 14 MWd/kg burn-up (up to now there is about 15% of all SNF), but the use of other burn-up RBMK SNF plutonium in GT-MHR is also possible. In RBMK Pu3 case more than 90% of ^{239}Pu transmutation was obtained (see the discussion below).

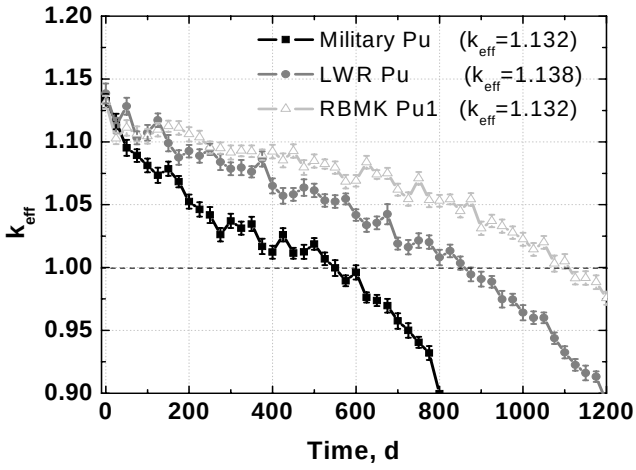


Figure 2. A behaviour of k_{eff} for Military, LWR and 14 MWd/kg burn-up RBMK plutonium with burnable poison in GT-MHR.

The GT-MHR with military plutonium composition was tested in our previous studies [3, 4]. In order to obtain relative fuel cycle length for different GT-MHR fuel cases the appropriate quantity of burnable poison was added

to achieve the same starting k_{eff}. Unfortunately for the higher burn-up SNF from RBMK k_{eff} was too low and it was impossible to make a comparative analysis. Therefore, only the RBMK Pu1 composition with added burnable poison was compared with LWR Pu and military Pu. Figure 2 presents k_{eff} evolution in time for these three cases.

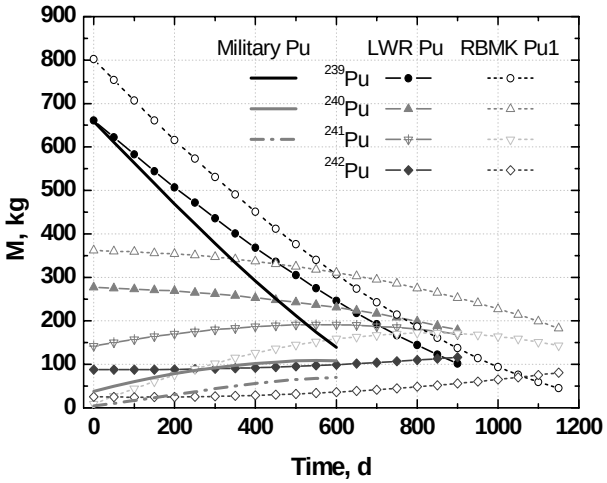


Figure 3. Evolution of mass of Pu isotopes for GT-MHR fuel cases: Military Pu, LWR Pu and RBMK Pu1.

In brief, a significant difference was found in k_{eff} evolution as a function of burn-up, in length of the fuel cycle and correspondingly in the incineration efficiency between military, LWR, and RBMK Pu1 plutonium cases. Starting at the same k_{eff} value the military Pu k_{eff} is decreasing continuously and already after ~550 days the fuel cycle ends with 73% of ^{239}Pu burn-up. LWR fuel cycle continues for ~850 days and 82% of ^{239}Pu burn-up is obtained. GT-MHR performance with RBMK Pu is even better: it gives longer fuel cycle (~1100 days), very efficient ^{239}Pu burning (93%) and relatively small accumulation of minor actinides. One can observe that in RBMK Pu1 case the decreasing of k_{eff} is slightly slower in the first part of GT-MHR operation comparing with LWR case. RBMK isotopic composition fuel in GT-MHR due to a higher amount of ^{240}Pu can support rather long operation time thanks to the formation of ^{241}Pu which compensates the decrease of ^{239}Pu. That means what GT-MHR with RBMK fuel can support operating itself and the TRISO fuel kernel is of right dimensions to provide stable k_{eff} by enhanced ^{240}Pu resonance neutron absorption.

Plutonium evolution for Military Pu, LWR Pu, and RBMK Pu1 cases is presented in Figure 3. The fastest burn-up of ^{239}Pu was obtained in Military Pu case, slowest in LWR Pu case. We also note significant differences for ^{240}Pu and ^{241}Pu evolution. In military plutonium case the production of ^{240}Pu from ^{239}Pu is most intensive because ^{239}Pu in this fuel isotopic composition dominates. The greatest ^{240}Pu destruction and ^{241}Pu formation was in RBMK Pu1 case.

After decommissioning of Ignalina NPP about 22.5 tons of plutonium will be accumulated in SNF. The above analysis shows that in practice one would need 2 GT-MHRs units with 10 fuel loads each every 3 years (assuming that reactor's life is ~30 years) to burn all INPP SNF plutonium. The outcome of the once-through GT-MHR fuel cycle of RBMK plutonium transmutation would be ~127 TWh of electric power.

It is also important to note that in all cases considered a negative temperature coefficient is obtained as it is shown in Table 2. Even the absence of burnable poison in RBMK Pu3 case does not change reactivity feedback – the negative $\Delta k_{eff}/\Delta T$ values in the GT-MHR were obtained for fuel, graphite and both fuel and graphite temperature increase. The only exception is military Pu case, where with increase in fuel temperature a slightly positive reactivity coefficient was predicted. On the other hand, this value is not statistically significant.

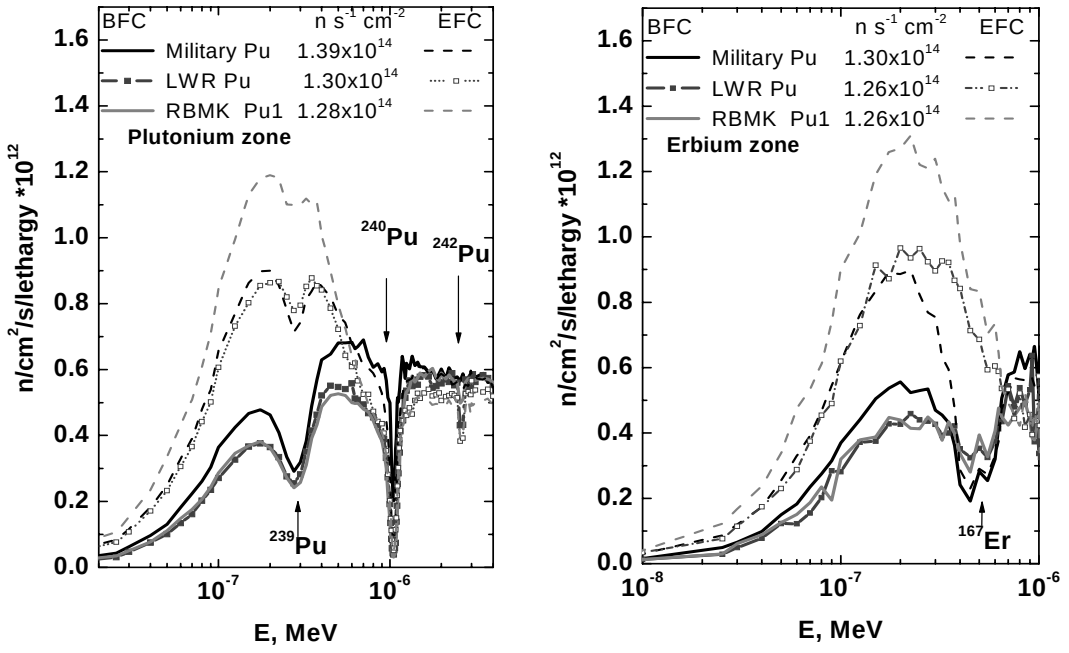
In Figure 4 we present averaged neutron fluxes in micro-particles for three isotopic fuel composition considered both at the beginning (BFC) and at the end of fuel cycle (EFC). Neutron fluxes "seen" by Pu and Er particles are presented in separate graphs on the left (4a) and on the right (4b) respectively. Thermal neutron contribution to neutron spectrum at the beginning of the fuel cycle inside fuel particles is quite similar, i.e. 9-10% for all cases considered. The observed increase of the thermal flux at the end of the fuel cycle increases up to 22%. One can observe, what in the resonance region neutrons are

equally suppressed by the presence of ^{240}Pu in civil plutonium cases both at the beginning and at the end of irradiation cycle. The neutron absorption at resonance 0.3 eV in ^{239}Pu decreases with time due to ^{239}Pu burn-up at the end of the fuel cycle. At the end of irradiation the bigger quantity of ^{242}Pu is accumulated in civil plutonium cases and the neutron absorption in the 2.67 eV resonance is more pronounced.

Table 2. Reactivity coefficient ($\Delta k_{eff}/\Delta T$) dependence on fuel, graphite and fuel / graphite temperature for different GT-MHR fuel cases (Δk_{eff} expressed in pcm, and ΔT in K).

Fuel	Pu temperature effect: ΔT_{Pu}=300 K ($T_{Graphite}$=1200 K)	Graphite temperature effect: ΔT_G=400 K (T_{Pu}=1500 K)	Total temperature effect: ΔT_{Pu}=300 K, ΔT_G=400 K
RBMK Pu1	-3.6±0.9	-9.2±0.7	-12.8±1.1
RBMK Pu3	-2.4±0.9	-9.1±0.7	-11.5±1.1
LWR Pu	-2.1±0.9	-8.5±0.7	-10.6±1.1
Military Pu	0.9±0.9	-9.6±0.7	-8.7±1.1

The neutron flux spectrum presented in Figure 4b shows, that at the beginning the most intensive neutron resonance capture in erbium particles is in military plutonium case. It is determined by almost 8 times bigger Er mass and somewhat higher thermal neutron flux when compared to other cases. Neutron flux spectrum in Er particle in both civil plutonium cases is similar. On the other hand, in GT-MHR with military plutonium fuel there is some erbium (~20kg) left at the EFC.



(a) (b)

Figure 4. a) Neutron flux spectrum in fuel material particles for different GT-MHR fuel cases at the beginning and at the end of the fuel cycle. b) Same as in (a) but in the burn-able poison particles.

4. Propagation of errors

The Monteburns code does not supply information about the statistical errors on its burn-up predictions. Nevertheless, we performed an indirect sensitivity test in two different ways: a) by modifying the starting pseudorandom number of the Monte Carlo code, and b)

by increasing the number of Monte Carlo histories by a factor of two. Finally, 4 possible combinations of the above variables (a-b) were examined.

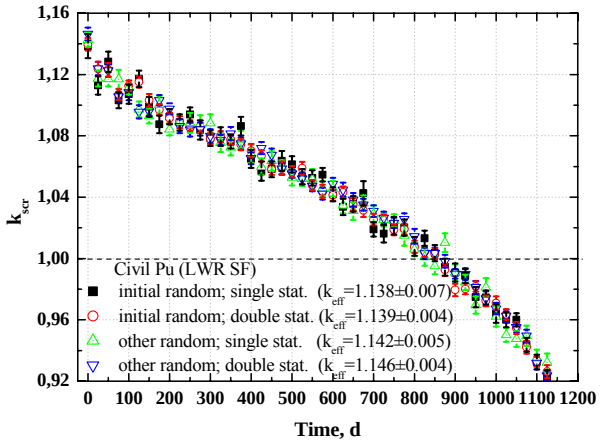


Figure 5. Evolution of $k_{eff} \pm 1\sigma$ as a function of burn-up and also as a function of different statistics-random number combinations (see the legend and Table 3 for details).

Our results on the evolution of k_{eff} as a function of burn-up are presented in Figure 5. In brief, one can clearly see that either by changing the initial random number seed, or by increasing statistics, or by using different combinations of both the k_{eff} eigenvalue varies only within its standard deviation. In addition, similar examination of the above input parameters did not have a considerable influence on the final burn-up results as presented in detail in Table 3. We conclude that as long as the convergence of the k_{eff} eigenvalue is obtained (correspondingly, the neutron flux value convergence), no significant influence on the final burn-up results should appear.

Table 3. Reload of the civil plutonium isotopic composition at the end of the fuel cycle as a function of different statistics-random number combinations (also see Figure 5).

LWR SF at the EOC m, kg					
	Initial load	single statistics; initial random	double statistics; initial random	single statistics; another random	double statistics, another random
Pu-238	32.4	29.2	29.2	29.2	29.2
Pu-239	661.0	122.0	123.0	123.0	121.0
Pu-240	277.0	189.0	189.0	191.0	189.0
Pu-241	142.0	176.0	176.0	175.0	175.0
Pu-242	87.6	113.0	113.0	113.0	113.0
Am-241		9.6	9.6	9.6	9.5
Am-242		0.1	0.1	0.1	0.1
Am-243		21.8	22.0	21.6	21.7
Cm-242		3.3	3.3	3.3	3.3
Cm-243		0.1	0.1	0.1	0.1
Cm-244		14.9	14.9	14.7	15.0
Cm-245		1.2	1.1	1.1	1.1
Cm-246		0.1	0.1	0.1	0.1
Actinides	**1200.0**	**680.2**	**681.4**	**681.7**	**678.1**

5. Conclusions

The influence of different initial fuel isotopic composition on GT-MHR performance was investigated using the 3D reactor core geometry with ceramic-coated fuel and burnable poison particles within the

Monte Carlo approach. The once-through fuel cycle was examined in all cases. The following conclusions are drawn:

- a significant difference was found in k_{eff} evolution, in the length of the fuel cycle and, correspondingly, in the transmutation efficiency between military, LWR, and RBMK plutonium fuel cases;
- it was demonstrated that TRISO fuel kernel (200 μm) is of optimal dimensions to provide negative temperature coefficient. Even the absence of burnable poison did not degrade the GT-MHR temperature reactivity coefficient;
- RBMK SNF isotopic composition in GT-MHR due to higher amount of ^{240}Pu can support rather long operation time, thanks to the formation of ^{241}Pu which compensates the disappearance of ^{239}Pu;
- 14 MWd/kg burn-up RBMK plutonium composition is the most attractive for use in GT-MHR (up to 93% of ^{239}Pu could be further incinerated). The use of higher burn-up RBMK SNF plutonium in GT-MHR is also possible: 90% of ^{239}Pu transmutation was obtained in 22 MWd/kg burn-up RBMK Pu case;
- a statistical convergence of the burn-up calculations was examined indirectly and showed no uncertainty accumulation due to the Monte Carlo method.

The possibility to transmute RBMK-1500 plutonium separated from SNF in high temperature helium cooled reactor was investigated for the first time. To burn all Ignalina NPP SNF plutonium one would need 2 GT-MHRs units with 10 fuel loads each every 3 years (assuming that reactor life is ~30 years). The outcome would be ~127 TWh of electricity produced.

Finally we note that the above burn-up rates still could be improved with the help of some dedicated fuel shuffling strategies. Studies along these lines should be definitely continued.

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