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Enriched Gadolinium Burnable Poison for PWR Fuel – Monte Carlo Burnup Simulations of Reactivity

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Additional information is available at the end of the chapter

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1. Introduction

Nuclear power plant utilities have to make business in a very competitive market. Therefore, some of them may have interest to increase the competitiveness through the use of nuclear fuels which could achieve very high burnups, beyond 70 GWd/Ton. In order to reach and surpass this value may be necessary to work with uranium enrichments higher than the traditional 5% limit. Make real such nuclear fuels demands heavy investments in R&D.

Advanced PWR nuclear fuels that will be able to burn well beyond 70 GWd/Ton necessarily need to have, among other things, uranium enrichments higher than 5%, thus leading to the necessity of using extremely efficient burnable poisons. Gadolinium oxide, also known as Gadolinia (Gd_2O_3) is currently used as burnable poison for PWR fuel. The fabrication processes, properties and behavior of Gadolinia mixed in UO_2 fuel are well known and well established. Seven Gadolinium isotopes naturally exist on earth. From these, only two isotopes, the Gd-155 and Gd-157 have extremely high thermal neutron absorption cross sections.

Burnable poisons containing gadolinium have the undesirable effect of reducing the thermal conductivity of UO_2 - Gd_2O_3 fuel and thus leading to higher temperature profiles in the fuel. In order to avoid such hot spots the currently available PWR rods use lower U-235 enrichment in all fuel pellets containing gadolinium. The use of gadolinium enriched in the most important isotopes, Gd-155 and Gd-157, to absorb neutrons, may permit to reduce the content of Gadolinia (Gd_2O_3) in the pellets and thus to improve the thermal conductivity of the fuel rods. This could permit to use the same U-235 enrichment in both types of fuel pellets, the ones with UO_2 - Gd_2O_3 and those with only UO_2 .

This Chapter has the objective to theoretically investigate the neutronic behavior of PWR fuels that would use $\text{UO}_2\text{-Gd}_2\text{O}_3$ compositions in which the gadolinium content is enriched from the natural abundance to a 100% Gd-155 and Gd-157 isotopes. The Monte Carlo burn-up codes system *Monteburns* [1] is used to perform the simulations. They will evaluate only the effects that modifications in the fuel composition produce over the curves of reactivity \times burnup. Indeed, the infinite neutron multiplication factor (k_{inf}) is used to plot the curves instead reactivity. The first set of these simulations aims to estimate the effects of variations in the contents of natural gadolinium and in the uranium enrichment. In the next set, the gadolinium content and the uranium enrichment are fixed, and the gadolinium isotopic compositions are modified by changing the enrichment of Gd-155 and Gd-157 until 100% to each isotope. This procedure is repeated to uranium enrichments going from 2.85% to 15%. After all, one last set of simulations evaluates the influence of the 100% Gd-155 and 100% Gd-157 enriched Gadolinia content. All the results are presented graphically and discussed. Finally, the optimized fuel composition is presented.

2. Physical model

2.1. Geometry and materials data

Figure 1 shows the 3x3 lattice of infinite fuel pin cells used in the simulations. One fuel rod containing the burnable poison ($\text{UO}_2\text{-Gd}_2\text{O}_3$) is located at the centre of the lattice and it is surrounded by seven rods containing standard fuel (UO_2) and one empty position (control rod guide tube). This geometry is kept to all the simulations. Only the isotopic compositions of the UO_2 and $\text{UO}_2\text{-Gd}_2\text{O}_3$ pellets are changed to the several cases simulated. As the 3x3 lattice is infinite the reflexive boundary condition was assumed.

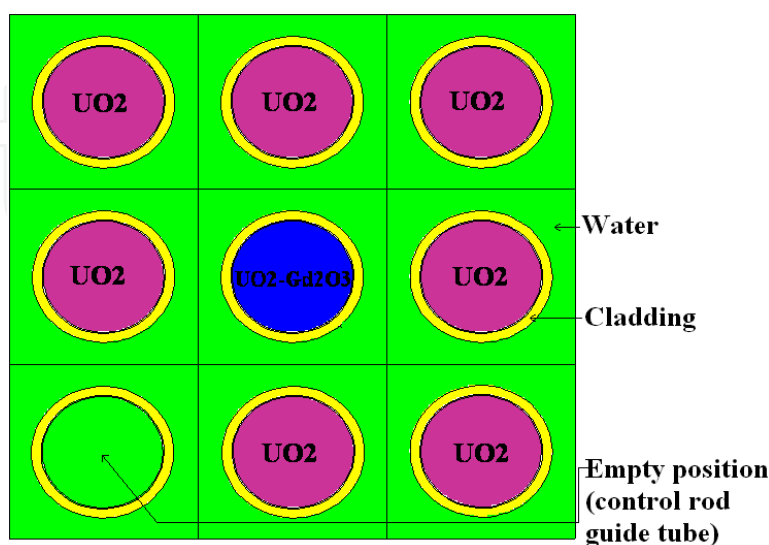


Figure 1. Infinite fuel pin cells 3x3 lattice.

The simulations were done considering that the fuel rods have the same geometric characteristics of the rods used in the AREVA FOCUS 16x16 fuel elements. Design data for this fuel is presented in [2]. Table 1 show some of the characteristics of the fuel rods adopted in the simulation model. Table 2 shows the thermal power and the uranium mass loaded in a typical 1300 MWe PWR reactor. These parameters are necessary to calculate the average specific power used in the burnup calculations. Table 3 presents the atomic number densities used in the model for water and cladding material (Zircalloy-4 was used instead of PCA-2B due the lack of information for this alloy).

Parameter	Unit	Value
Fuel Rods		
Outer diameter of cladding	cm	1.0750
Inner diameter of cladding	cm	0.9300
Active length	cm	390.00
Pitch	cm	1.430
Cladding Material		PCA-2B
Fuel Pellets		
Outer diameter	cm	0.9110
Guide tubes		
Outer diameter	cm	1.38
Thickness	cm	0.0700
Cladding Material		PCA-2B

Table 1. Geometry data of the fuel rods and guide tubes.

Parameter	Value
Thermal power (MW)	4000
Uranium mass (Ton)	~ 111

Table 2. Thermal power and uranium mass of a typical 1300 MWe PWR reactor

MATERIAL	Atoms/barn.cm
Nuclide	
ZIRCALOY-4	
Natural Cromium	7.5891e-05
Natural Iron	1.4838e-04
Natural Zirconium	4.2982e-02
WATER	
H-1	6.6242e-02
O-16	3.3121e-02

Table 3. Atomic number densities of cladding and cooling materials.

Finally, Table 4 presents the theoretical compositions of the several simulated fuels. These fuels are the alloys in which the uranium and gadolinium enrichments and the contents of Gadolinia and uranium dioxide are modified. In the simulations these compositions refer to the $\text{UO}_2\text{-Gd}_2\text{O}_3$ rod in the center of the 3x3 lattice and the UO_2 rods surrounding it. The calculation of the atomic number densities of the nuclides of Uranium, Gadolinium and Oxygen in fresh fuels to the several inputs was automatized by a MS Excel electronic spreadsheet.

Important to mention that the inputs 0050 (0.0% Gadolinia and 5.0% U235 enrichment for $\text{UO}_2\text{-Gd}_2\text{O}_3$ rod) and 7028 (7.0% gadolinia and 2.8% U235 enrichment for $\text{UO}_2\text{-Gd}_2\text{O}_3$ rod) refer to real (commercial) FOCUS [2] fresh fuels and are always used to compare the behavior of the others theoretical fuels simulated.

Some studies regarding sensitivities of the neutron cross sections to temperatures were performed and presented in [3, 4]. These studies led to the conclusion that the MCNP [5] pointwise neutron cross sections data library.66c can be used satisfactorily by MonteBurns to simulate the fuel burnup of the lattice in spite of this set of cross sections data be processed at room temperature (293 K). As shown in [4] the results for overall production and destruction of the main nuclides during the burnup are similar if using library.66c or other libraries processed at higher temperatures.

2.2. Burnup intervals, important nuclides and neutron cross sections

In the simulations the rods were irradiated until around 90 GWd/ton. This total burnup was divided into several intervals with the burnup step values increasing since 0.5 until 25 days. Nuclides which the atom fraction, weight fraction, fraction of absorption, and fraction of fission greater than 10^{-4} were considered important nuclides and therefore its one group cross section were calculated for each burnup step. Sensitivities studies were performed to determine this *importance fraction*. However, there are a few nuclides with the importance fraction greater than 10^{-4} to which the one group cross sections were not calculated due to the lack of data in the MCNP standard libraries. The MCNP.66c pointwise neutron cross sections data were used preferably.

Input Name	UO ₂ -Gd ₂ O ₃		UO ₂	
	Gd ₂ O ₃ Content(%)	Composition	(%U-235)	(%U-235)
0050	0	Natural	5.0	5.0
1050	1.0	Natural	5.0	5.0
2045	2.0	Natural	4.5	5.0
3038	3.0	Natural	3.8	5.0
4035	4.0	Natural	3.5	5.0
5033	5.0	Natural	3.3	5.0
6031	6.0	Natural	3.1	5.0
7028	7.0	Natural	2.8	5.0
hgd4015	2.0	40%Gd-155 and 15% Gd-157	2.8	5.0
hgd6010	2.0	60%Gd-155 and 10% Gd-157	2.8	5.0
hgd8005	2.0	80%Gd-155 and 05% Gd-157	2.8	5.0
hgd9900	2.0	100%Gd-155 and 0% Gd-157	2.8	5.0
hgd1540	2.0	15%Gd-155 and 40% Gd-157	2.8	5.0
hgd1060	2.0	10%Gd-155 and 60% Gd-157	2.8	5.0
hgd0580	2.0	05%Gd-155 and 80% Gd-157	2.8	5.0
hgd0099	2.0	0%Gd-155 and 100% Gd-157	2.8	5.0
gd4015	2.0	40%Gd-155 and 15% Gd-157	5.0	5.0
gd6010	2.0	60%Gd-155 and 10% Gd-157	5.0	5.0
gd8005	2.0	80%Gd-155 and 05% Gd-157	5.0	5.0
gd9900	2.0	100%Gd-155 and 0% Gd-157	5.0	5.0
gd1540	2.0	15%Gd-155 and 40% Gd-157	5.0	5.0
gd1060	2.0	10%Gd-155 and 60% Gd-157	5.0	5.0
gd0580	2.0	05%Gd-155 and 80% Gd-157	5.0	5.0
gd0099	2.0	0%Gd-155 and 100% Gd-157	5.0	5.0
gdx4015	2.0	40%Gd-155 and 15% Gd-157	6.0	6.0
gdx6010	2.0	60%Gd-155 and 10% Gd-157	6.0	6.0
gdx8005	2.0	80%Gd-155 and 05% Gd-157	6.0	6.0
gdx9900	2.0	100%Gd-155 and 0% Gd-157	6.0	6.0
gdx1540	2.0	15%Gd-155 and 40% Gd-157	6.0	6.0
gdx1060	2.0	10%Gd-155 and 60% Gd-157	6.0	6.0
gdx0580	2.0	05%Gd-155 and 80% Gd-157	6.0	6.0
gdx0099	2.0	0%Gd-155 and 100% Gd-157	6.0	6.0
gdd4015	2.0	40%Gd-155 and 15% Gd-157	10.0	10.0
gdd6010	2.0	60%Gd-155 and 10% Gd-157	10.0	10.0
gdd8005	2.0	80%Gd-155 and 05% Gd-157	10.0	10.0
gdd9900	2.0	100%Gd-155 and 0% Gd-157	10.0	10.0
gdd1540	2.0	15%Gd-155 and 40% Gd-157	10.0	10.0
gdd1060	2.0	10%Gd-155 and 60% Gd-157	10.0	10.0

Input Name	UO ₂ -Gd ₂ O ₃		UO ₂		
	Gd ₂ O ₃ Content(%)	Composition	(%U-235)	UO ₂ Content (%)	(%U-235)
gdd0580	2.0	05%Gd-155 and 80% Gd-157	10.0	98.0	10.0
gdd0099	2.0	0%Gd-155 and 100% Gd-157	10.0	98.0	10.0
gdq4015	2.0	40%Gd-155 and 15% Gd-157	15.0	98.0	15.0
gdq6010	2.0	60%Gd-155 and 10% Gd-157	15.0	98.0	15.0
gdq8005	2.0	80%Gd-155 and 05% Gd-157	15.0	98.0	15.0
gdq9900	2.0	100%Gd-155 and 0% Gd-157	15.0	98.0	15.0
gdq1540	2.0	15%Gd-155 and 40% Gd-157	15.0	98.0	15.0
gdq1060	2.0	10%Gd-155 and 60% Gd-157	15.0	98.0	15.0
gdq0580	2.0	05%Gd-155 and 80% Gd-157	15.0	98.0	15.0
gdq0099	2.0	0%Gd-155 and 100% Gd-157	15.0	98.0	15.0
1551510	1.0	100%Gd-155	15.0	99.0	15.0
1551515	1.5	100%Gd-155	15.0	98.5	15.0
1551520	2.0	100%Gd-155	15.0	98.0	15.0
1551525	2.5	100%Gd-155	15.0	97.5	15.0
1551530	3.0	100%Gd-155	15.0	97.0	15.0
1551540	4.0	100%Gd-155	15.0	96.0	15.0
1571510	1.0	100% Gd-157	15.0	99.0	15.0
1571515	1.5	100% Gd-157	15.0	98.5	15.0
1571520	2.0	100% Gd-157	15.0	98.0	15.0
1571525	2.5	100% Gd-157	15.0	97.5	15.0
1571530	3.0	100% Gd-157	15.0	97.0	15.0
1571540	4.0	100% Gd-157	15.0	96.0	15.0

Table 4. Enrichments and Gadolinia contents of the fuels.

3. Results and analysis

The results of the various simulations will be graphically presented in the next pages. The subtitles, on right side of the graphs, indicate the input simulated as showed in Table 4. The focus of the analyses is to estimate the behavior of the fuel reactivity as a function of the burnup for the 3x3 lattice of fuel rods. Figure 2 shows the evolution of the infinite neutron multiplication factor (k_{inf}) for fuels with natural Gadolinia content from 0 to 9.9% and different uranium enrichment. As expected, the higher reactivity peak found is to the fuel without any Gadolinia (input 0050) and the peak occurs at zero burnup. The greater is the content of Gadolinia in the fuel the smaller is the reactivity of the fresh fuel. In addition, for all fuels containing Gadolinia the reactivity peak is observed after some days of irradiation and not at zero burnup condition. These peaks moves to the right (to higher burnups) as the Gadolinia content increases.

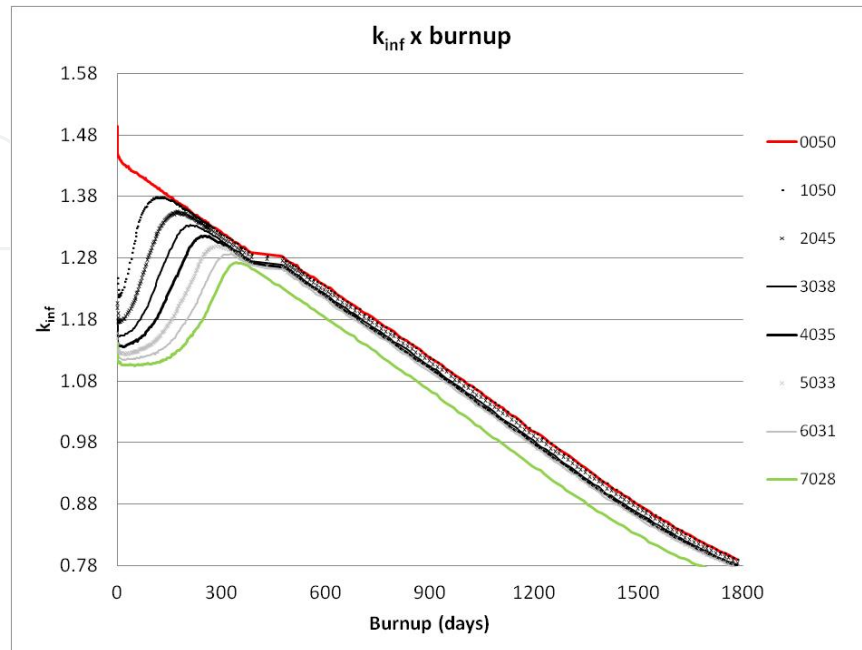


Figure 2. Infinite neutron multiplication factor as a function of burnup – changing Gadolinia content and %U-235.

3.1. Evolution of the infinite neutron multiplication factor as a function of the burnup – Gadolinia content fixed in 2% and changing the enrichments of Gd155, Gd157 and U235

The results presented in this section refer to Gadolinia contents in fuels lower than 7%. Usually this content is the limit to commercial fuels in order to avoid stronger effects affecting the thermal conductivity. As a starting point is used 2.0% Gadolinia content like recommended in [6]. In addition, the natural composition of the gadolinium in the simulations is modified to represent enrichments of Gd-155 and Gd-157 in the range from 5.0% to 100%.

Figure 3 (zoomed to the first 450 days of burnup in Figure 4) shows the evolution of k_{inf} as a function of burnup to the 3x3 lattice to: UO_2 - Gd_2O_3 fuel rod, enriched 2.8% in U-235, and the seven UO_2 fuel rods surrounding it enriched 5.0% in U-235. The zoomed Figure 4 shows clearly that all simulated fuels have the reactivity peak lower than the peak observed to the reference fuel 0050 that is free of Gadolinia. The lowest reactivity peak was found to the other reference fuel (7028 - natural Gadolinia) and its curve is very close to the curve of the fuel with 100% Gd-155. Such results are very exciting to proceed simulating fuels having higher U-235 enrichments until to find a composition to which the reactivity peak is close to the 0050 fuel. Figures 5 to 15 show the results of these simulations to U-235 enrichments in the range of 5.0% to 15.0% (subtitles are always as Table 4).

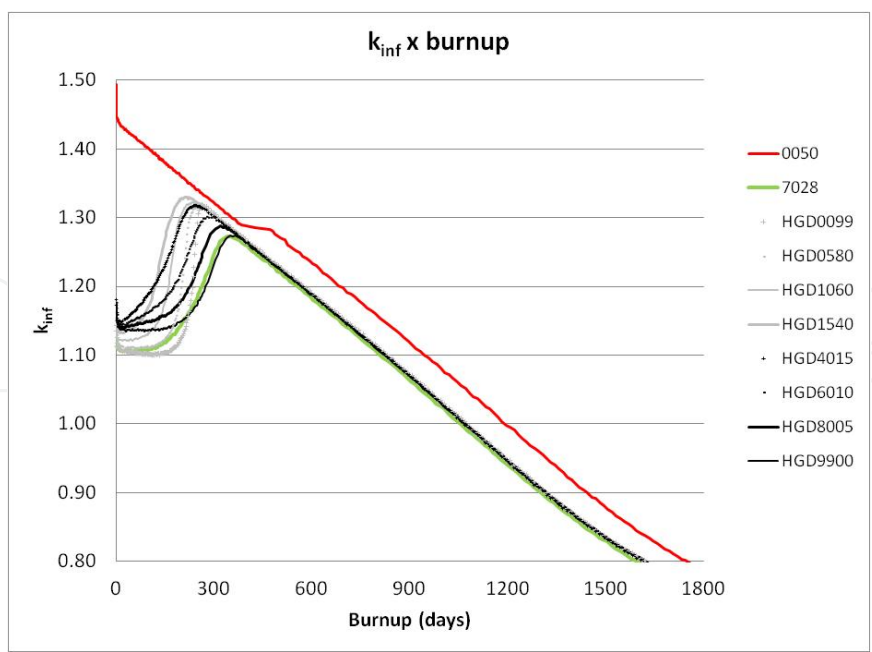


Figure 3. Infinite neutron multiplication factor as a function of burnup – 2.0% Gd₂O₃, 2.8% U-235, changing Gd-155 and Gd-157 enrichments.

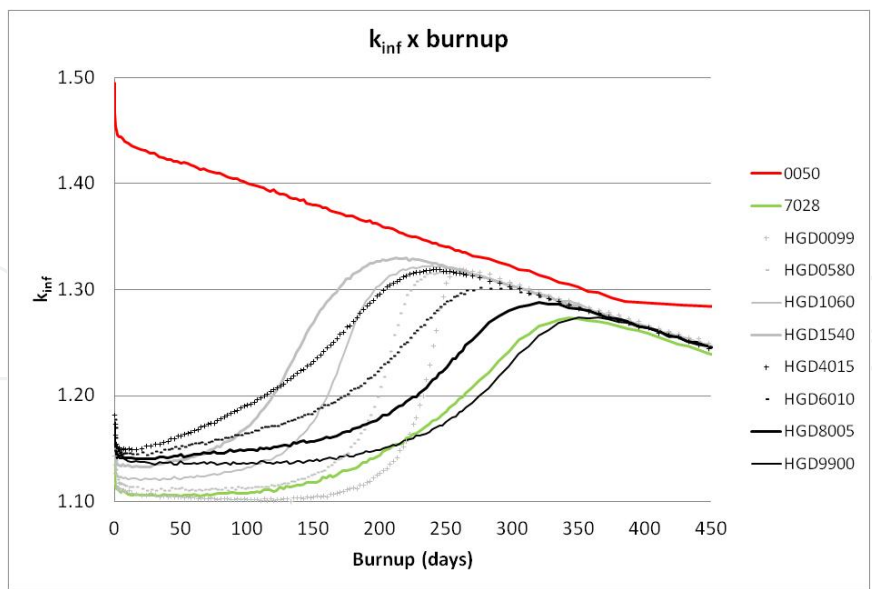


Figure 4. Zoom of Figure 3 (beginning of burnup interval).

Increasing the U-235 enrichment of the $\text{UO}_2\text{-Gd}_2\text{O}_3$ fuel rod to 5.0%, Figures 5 and 6 are obtained. One can notice a slight growth in reactivity of the compositions simulated and the curves for the eight inputs, after the peaks region, started to decouple from 7028 curve towards the 0050 curve.

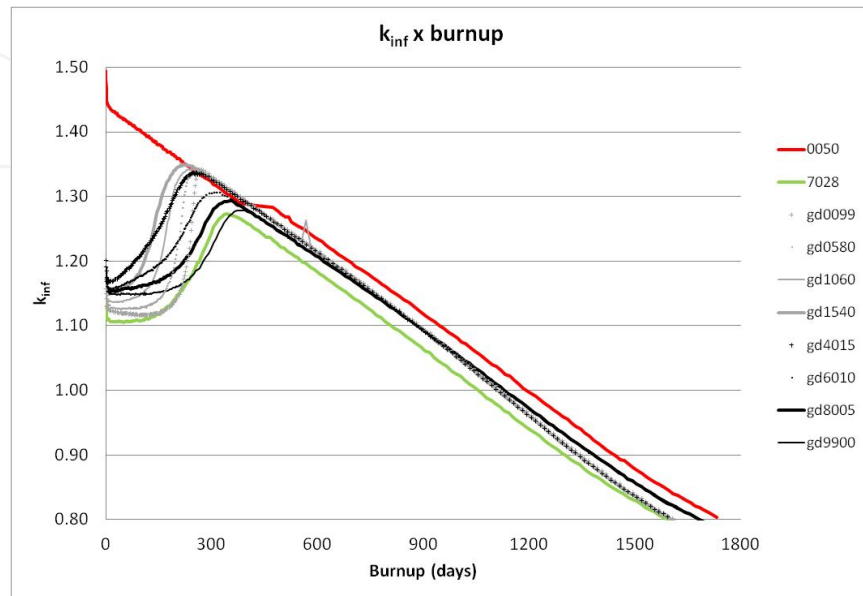


Figure 5. Infinite neutron multiplication factor as a function of burnup – 2.0% Gd_2O_3 , 5.0% U-235, changing Gd-155 and Gd-157 enrichments.

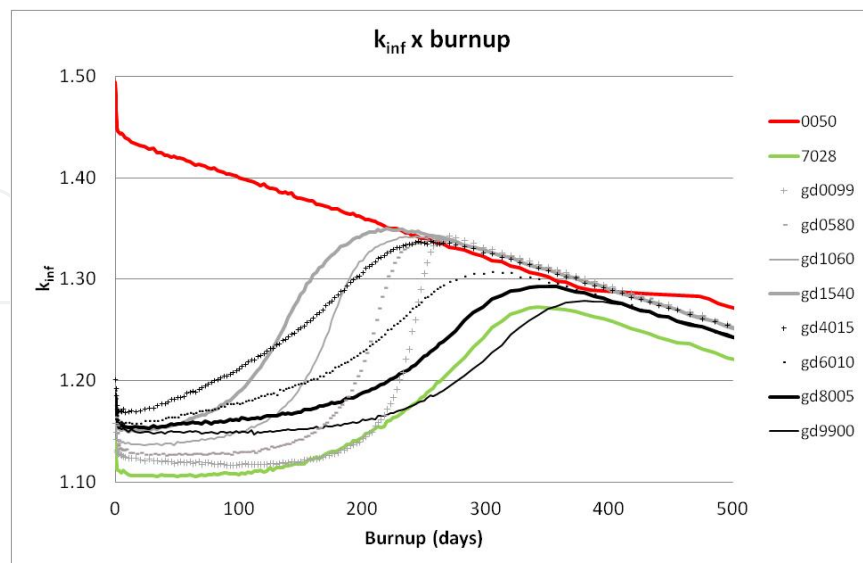


Figure 6. Zoom of Figure 5 (beginning of burnup interval).

Next step of the simulations considered 6.0% of U-235 enrichment in the fuel. Figure 7 plots the results (Figure 8 zoomed it until 600 days). As expected, the reactivities of the fuels containing enriched gadolinium continue growing and surpass the 0050 curve after a few hundred days of burnup. However, the reactivity peak for the 0050 is, so far, the greatest.

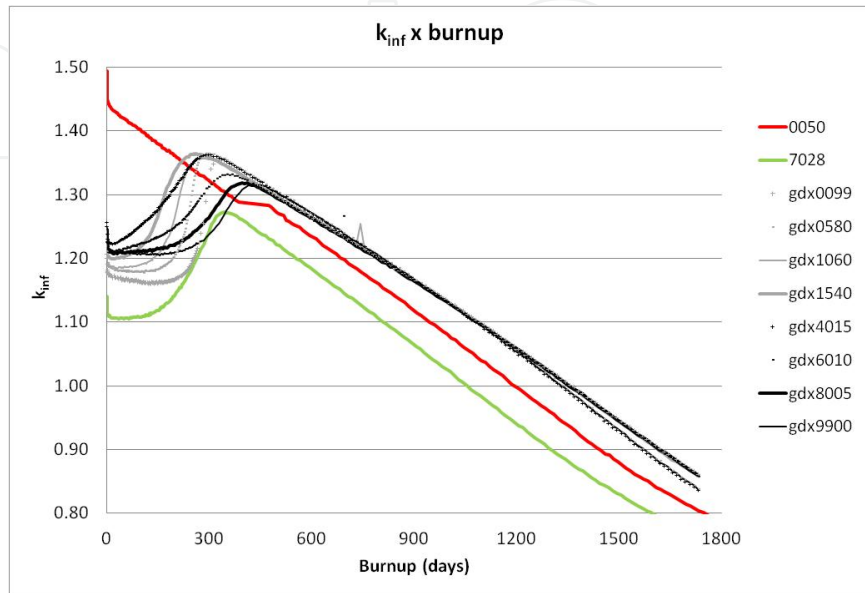


Figure 7. Infinite neutron multiplication factor as a function of burnup – 2.0% Gd₂O₃, 6.0% U-235, changing Gd-155 and Gd-157 enrichments.

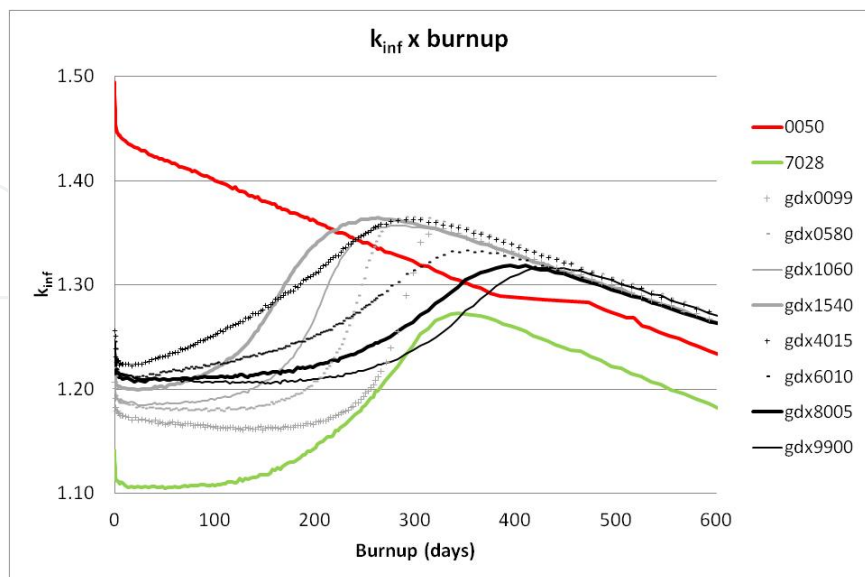


Figure 8. Zoom of Figure 7 (beginning of burnup interval).

The simulations are going to continue while the reactivity peak of the 0050 reference fuel is not exceeded. Figure 9 (and Fig. 10) shows the results for fuel compositions with 10% U-235. The reference fuel 0050 still has the greatest reactivity peak but the others gadolinium enriched compositions are getting closer. Furthermore, it is clearly evident the reactivity gain of the gadolinium enriched fuels for higher burnups as well as the flattening of its reactivity peaks.

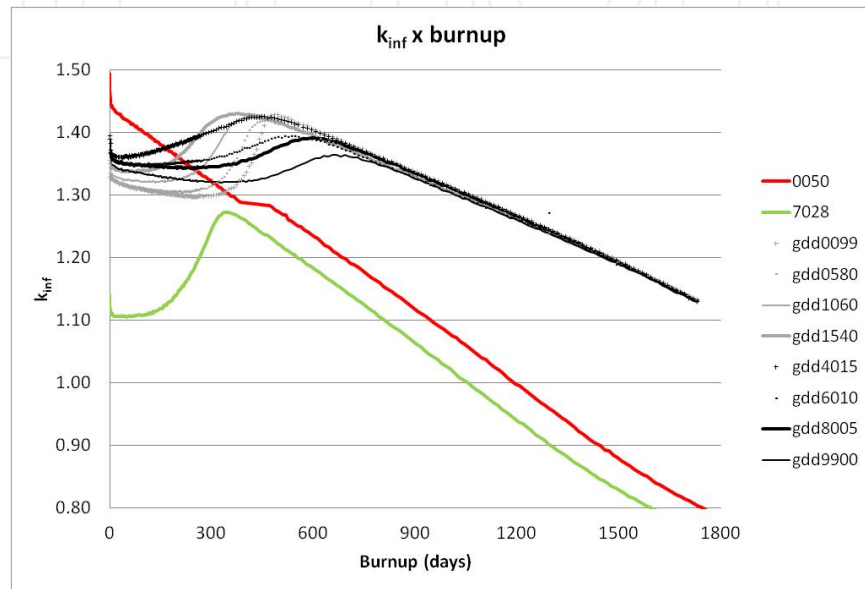


Figure 9. Infinite neutron multiplication factor as a function of burnup – 2.0% Gd_2O_3 , 10.0% U-235, changing Gd-155 and Gd-157 enrichments.

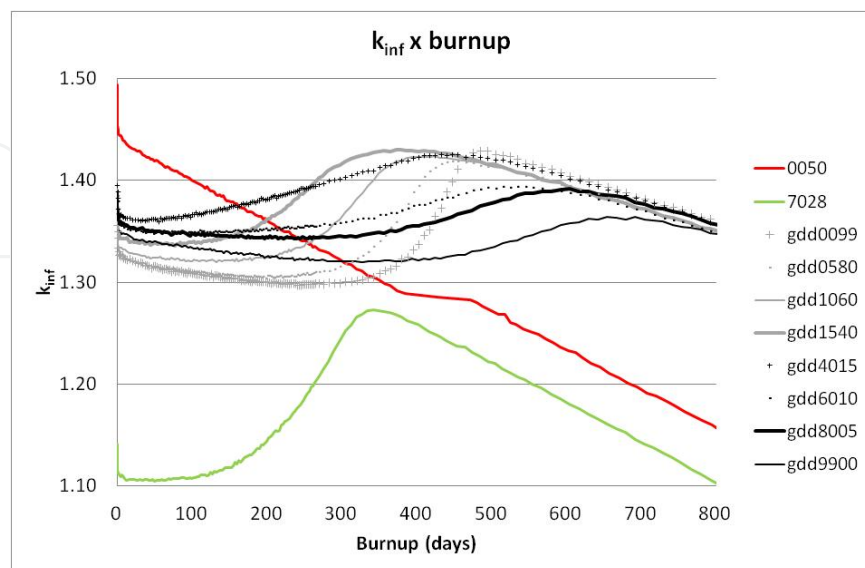


Figure 10. Zoom of Figure 9 (beginning of burnup interval).

The next three figures are plots of the gadolinium enriched fuels having 15.0% of U-235. Figure 11 shows the whole burnup interval, and the Figures 12 and Fig. 13 are the zoomed, respectively, in the beginning and at the end of the burnup interval. They show that the reactivity peaks of the gadolinium enriched fuels finally come close to the 0050 peak. In addition, Figure 11 shows very clearly the reactivity gain of these fuels, compared to the reference fuels, in the whole burnup interval. Reference fuel 0050, for instance, after around 500 days of burnup, has its reactivity reduced to a value that the gadolinium enriched fuels will achieve only after around 1700 days. Finally, it is extremely important to notice that the fuel compositions with higher enrichments in Gd-155 have the reactivity peak more flattened than the others and the reactivity peaks occurs at zero burnup, just like the fuels without burnable poison. Therefore, the next analyses are going to focus in compositions with 100% Gd-155 (compositions with 100% Gd-157 will be studied too, just for comparison).

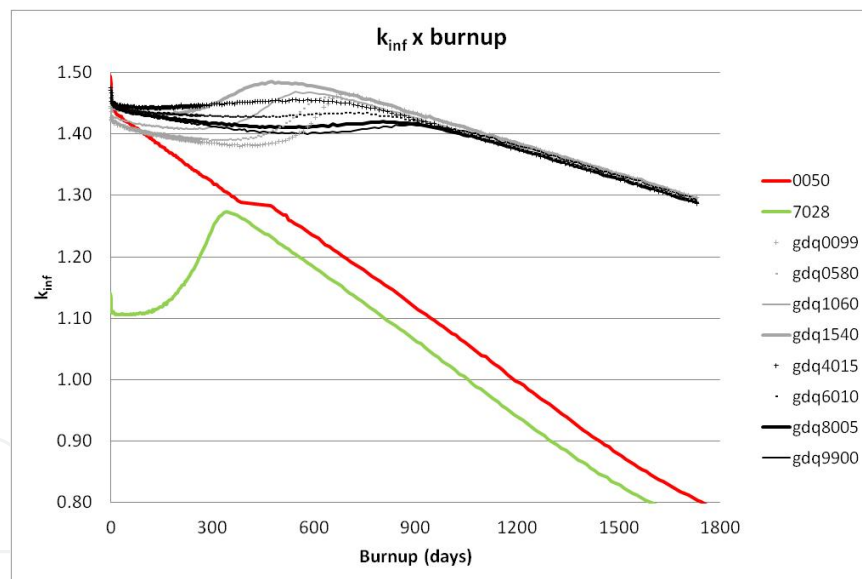


Figure 11. Infinite neutron multiplication factor as a function of burnup – 2.0% Gd_2O_3 , 15.0% U-235, changing Gd-155 and Gd-157 enrichments.

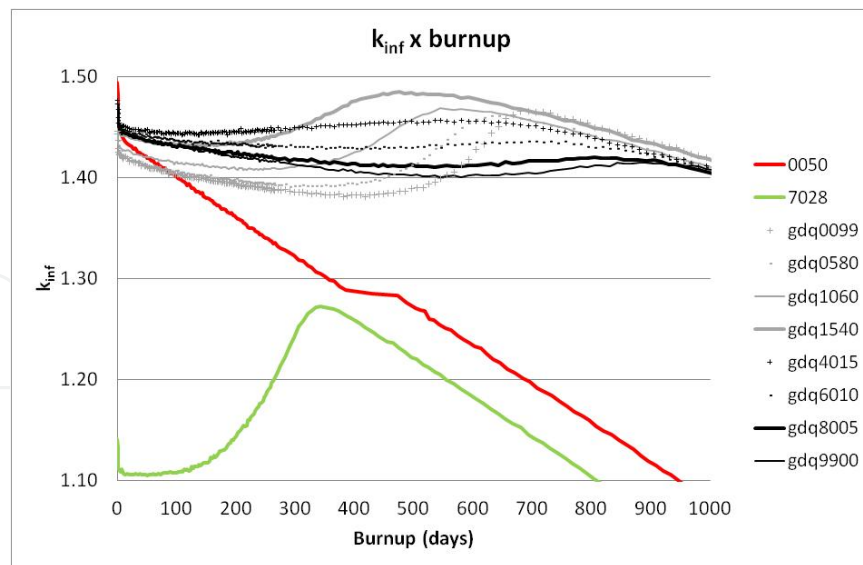


Figure 12. Zoom of Figure 11 (beginning of burnup interval).

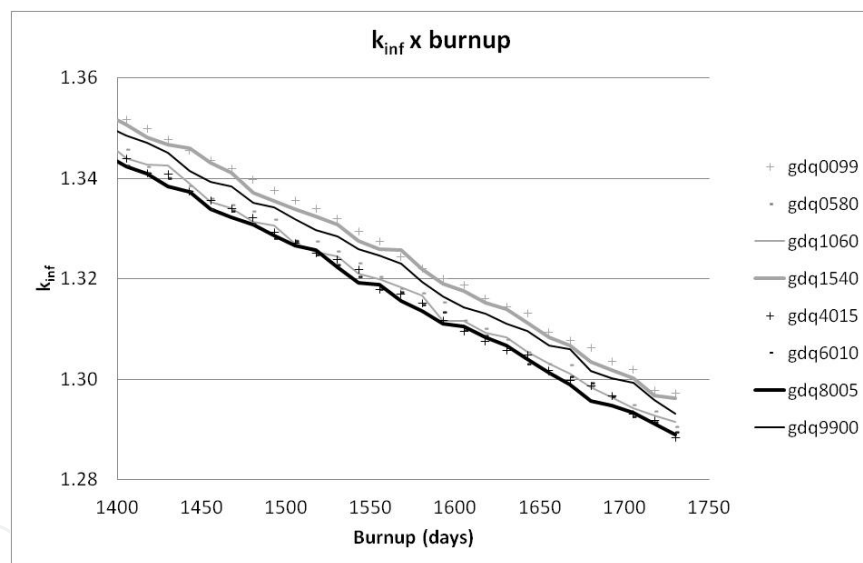


Figure 13. Zoom of Figure 11 (end of burnup interval).

3.2. Evolution of the infinite neutron multiplication factor as a function of the burnup – Fixed 15% U235, 100% Gd155 and Gd157 enrichments; Gadolinia contents change

Thus far, the studies performed have shown that a fuel composition with 2% Gd_2O_3 and 98% UO_2 , enriched at 15% U-235 and at 100% Gd-155 is able to keep high reactivities at very long burnups. Furthermore, the reactivity peak of this composition occurs at zero burnup and it is a bit lower than the peak for the reference fuel. Nevertheless, this 2% Gadolinia content was settled based in [6]. Therefore, it is necessary to evaluate changes in such parameter.

The Figure 14 presents the evolution of the reactivity for compositions with 100% Gd-155 and 15.0% U-235 enrichments and Gadolinia contents from 1.0% to 4.0%, as listed in Table 4. Figure 15 shows the same results for a composition with 100% Gd-157. It is observed from these figures that when the gadolinium content is reduced to 1% the reactivity peaks surpass the maximum value achieved by the reference fuel 0050. However, this reactivity peak is not exceeded by fuel compositions with Gadolinia content within 1.5% to 4.0%. Moreover, it is very evident the advantage of using enrichments of 15% U-235 and 100% Gd-155 (instead of 100% Gd-157) due to the absence of reactivity peaks after the fresh fuel condition.

Strictly from the neutronic point of view, fuel compositions having the Gadolinia content within 1.5% to 4.0%, high Gd-155 enrichment (~100%) and percentage of U-235 around 15% can be technologically very attractive to keep the reactivity at high and stable levels, even for very long burnups. Nevertheless, this study is incomplete for neither has thermal-hydraulic nor thermo-mechanical analyses of these fuels. The physical model may also be improved towards a whole reactor core instead of a simple lattice. It is also absent any economic analysis regarding uranium and gadolinium at these levels of enrichment. These problems can be addressed in future works.

Finally, Figure 16 is presented to show clearly the evolution of fuel compositions having 100% Gd-155 and 2.0% Gd_2O_3 in different U-235 enrichments.

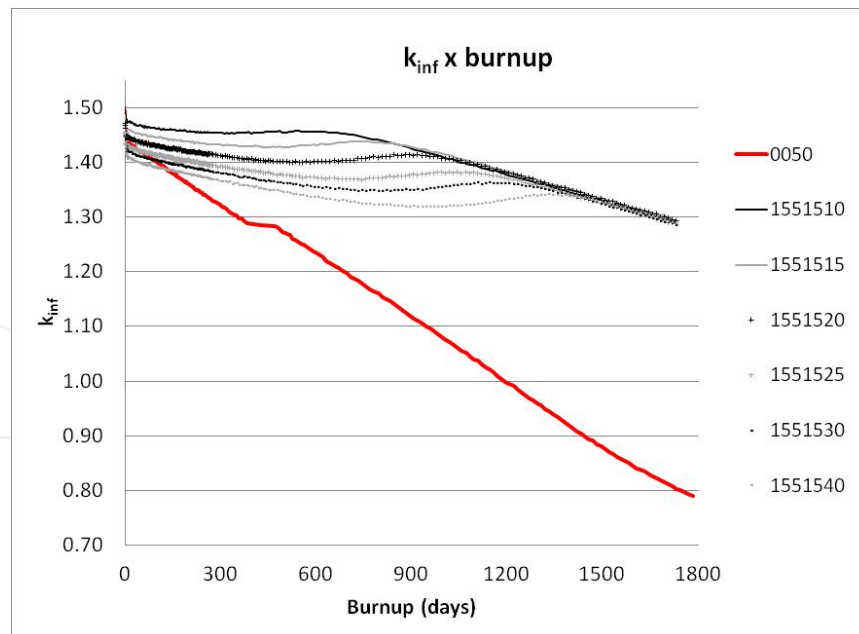


Figure 14. Infinite neutron multiplication factor as a function of burnup – 100% Gd-155, 15% U-235 and changing content of Gd_2O_3 .

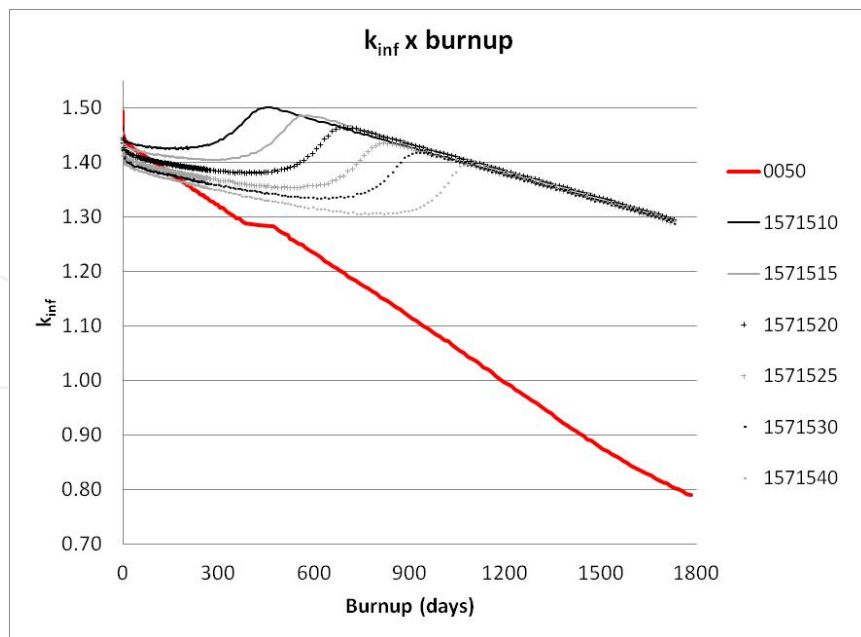


Figure 15. Infinite neutron multiplication factor as a function of burnup – 100% Gd-157, 15% U-235 and changing content of Gd₂O₃.

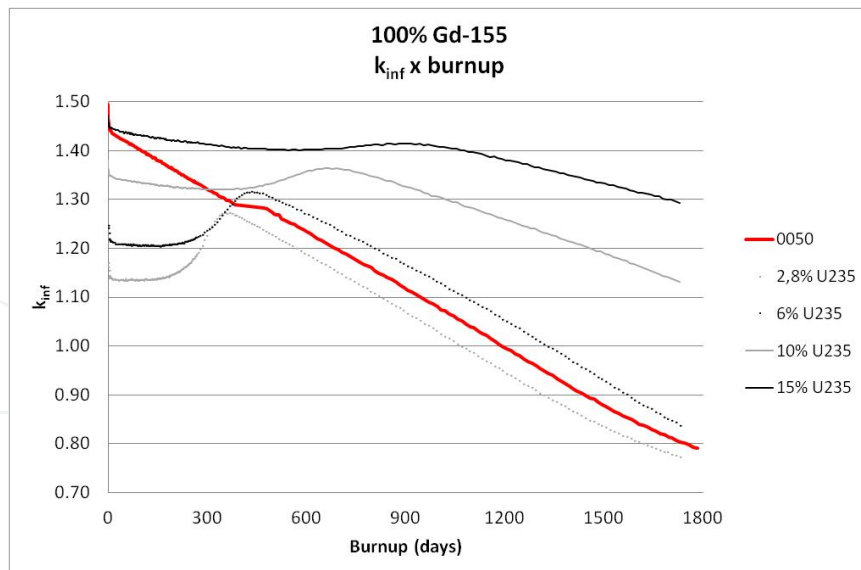


Figure 16. Infinite neutron multiplication factor as a function of burnup – fuel compositions having 100% Gd-155, 2.0% Gd₂O₃ and different U-235 enrichments.

4. Conclusions

Monte Carlo Burnup codes system Monteburns was used to simulate 3x3 lattices of rods containing several fuel compositions with different enrichments and contents of uranium and gadolinium oxides. The greater is the content of Gadolinia in the fuel the smaller is the reactivity of the fresh fuel. In addition, for fuels containing Gadolinia the reactivity peak is observed after some days of irradiation and not at zero burnup condition. These peaks moves to higher burnups as the Gadolinia content increases. The rise of the uranium-235 enrichment in the fuel can compensate the reduction of reactivity due to the use of Gadolinia as burnable poison and even lead to higher burnups and longer cycles. It was found that fuel compositions having the Gadolinia content within 1.5% to 4.0%, 100% Gd-155 enrichment and 15% U-235 enrichment may keep high and stable levels of reactivity for very long burnups. In addition, the reactivity peak occurs at zero burnup, just like the fuels without any burnable poison, and the Gadolinia content is within a range that can be able to avoid the undesired effect in the fuel thermal conductivity.

Thus far, strictly from the neutronic point of view, fuel compositions having the Gadolinia content within 1.5% to 4.0%, high Gd-155 enrichment (~100%) and U-235 enrichment around 15% can be technologically very attractive. Nevertheless, additional analyses of thermal-hydraulic and thermo-mechanical properties for these fuels are needed. These problems can be addressed in future works as well as an economic analysis regarding uranium and gadolinium at these levels of enrichment and the physical model can be improved towards a whole reactor core model.

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