

Physics of hydride fueled PWR

Francesco Ganda* and Ehud Greenspan
University of California at Berkeley, 4105 Etcheverry Hall,
94720 Berkeley, CA

Abstract

Thermal-hydraulically optimal U-ZrH_{1.6} fuelled PWR cores have positive coolant and fuel temperature coefficients of reactivity (RC) when using IFBA and soluble boron to compensate for excess reactivity. It was found possible to make negative all RC by either replacing the IFBA by erbium or by replacing some of the ZrH_{1.6} by ThH₂. The latter approach is preferred as it offers longer core life as well as reduced TRU inventory in the discharged fuel.

KEYWORDS: PWR, zirconium hydrides, reactivity coefficients, thorium

1. Introduction

Over the last few years, UC Berkeley researchers have been studying the physics of infinite lattices and finite fuel assemblies of PWR and BWR cores fueled with hydride rather than oxide fuel [1-5]. This study was part of a broader study that considered also thermal-hydraulics, transient, fuel performance, and economic analyses the purpose of which was to assess the feasibility of improving the performance of PWR and BWR cores by designing them to operate on hydride fuel. Hydride fuel was found to offer a number of substantial improvement possibilities, including significantly more effective plutonium recycling possibilities [4, 5] and significantly higher power density in BWR cores of simplified design. [6]

The reference hydride fuel considered is U-ZrH_{1.6} – the fuel type used in TRIGA type research reactors. For a given lattice pitch, P, and fuel rod outer diameter, D, hydride fuel is characterized by a significantly larger hydrogen-to-heavy metal (H/HM) ratio than uranium dioxide fuel. A comprehensive scoping study has been performed to define the acceptable D-P design space of hydride versus oxide fuel in PWR and BWR cores, and the discharge burnup attainable for dozens of combinations of D, P and for a number of uranium enrichment levels. For a given geometry (D – P pair) to be acceptable, all reactivity coefficients of a core made of such unit cells have to be negative. The reactivity coefficients considered are the fuel temperature coefficient (FTC) of reactivity, the coolant temperature coefficient (CTC) of reactivity and the reactivity effect of voiding 5% and 90% of the water.

Due to the relatively large H/HM ratio, it was found more difficult to obtain a negative CTC for hydride than for oxide fuel in PWR. In particular, the CTC and FTC of reactivity of the geometry that offers the maximum power density was found to be positive; the latter beyond a relatively low burnup. A couple of approaches were investigated for avoiding the positive reactivity coefficients – use of burnable poisons and addition of thorium.

The purpose of this paper is to describe the approaches explored for obtaining negative reactivity coefficients (RC) and to provide an in-depth analysis of ThH₂ containing hydride fuel.

* Corresponding author, Tel. 510-643-9281, E-mail: gandaf@nuc.berkeley.edu

2. Reference Unit cell and Computational Method

The reference unit cell fuel is U-ZrH_{1.6} having 45^w% uranium of 12.5% enrichment and without burnable poisons; all the excess reactivity is compensated by soluble boron. The unit cell geometry, defined in Table 1, is that providing peak power density [7]. The BOL composition is shown in Table 2. The average fuel temperature is assumed 723 K. The achievable burnup is 129.28 GWD/MTiHM, calculated assuming a three-batch checkerboard-reloading scheme.

Table 1: Details of the geometry.

Clad outside radius (cm)	0.42
P/D	0.685
Fuel radius (cm)	0.359
Clad inside radius (cm)	0.367

Table 2: Atomic densities at BOL.

Nucleus	at/b-cm
H-ZrH ₂	0.047116013
Zr-ZrH ₂	0.029447508
U-235	0.001176708
U-238	0.008227539

All results were obtained with the NEWT module of the SCALE 5.1 package using a special procedure described in [8]. The adequacy of this system has been verified by benchmarks with XSDRNPN (part of the SCALE package as well) and with MCNP. The cross sections are derived from the 44 group library, pre-treated with BONAMI and NITAWL for un-resolved and resolved resonance treatment, respectively.

The burnup-dependent CTC and FTC of reactivity calculated for the reference hydride fuel lattice are shown in Figures 1 and 2. The CTC is calculated from the Δk_{∞} at 324°C and the nominal operating temperature of 314°C; the corresponding water density is 0.66831 g/cc and 0.69483 g/cc. The FTC is calculated from the Δk_{∞} at 550°C and the nominal operating temperature of 450°C. These fuel temperatures are very low as compared with typical oxide fuel temperatures because the thermal conductivity of hydride fuel is approximately 5 times larger.

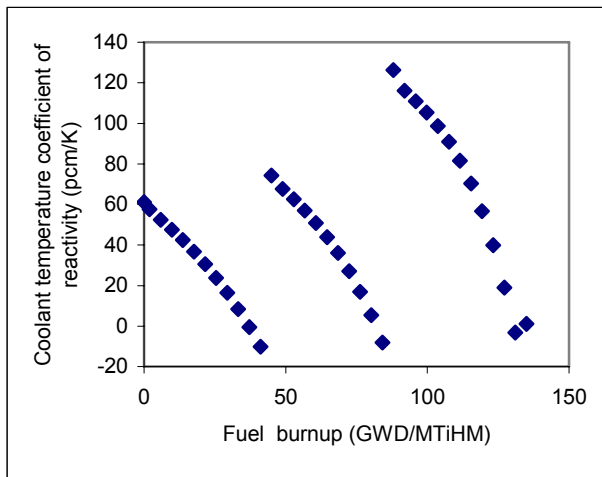


Figure 1: Batch-wise CTC of reactivity as a function of burnup

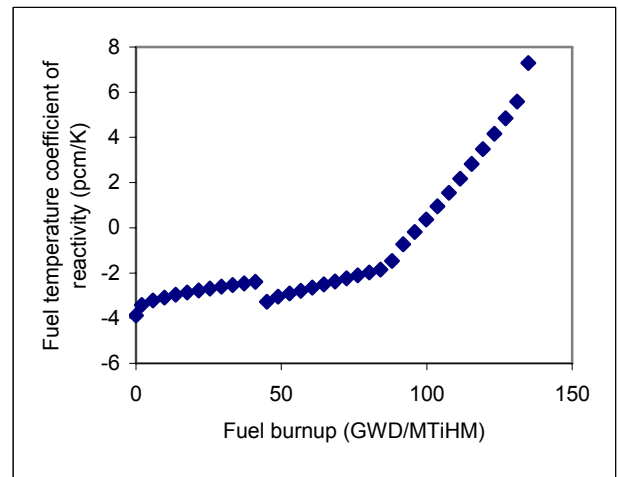


Figure 2: Batch-wise FTC of reactivity as a function of burnup

The CTC of reactivity is positive over most of each cycle due to the relatively large amount of soluble boron— about 3870 ppm, required to compensate for the excess reactivity at BOL. Detailed neutron balance analysis shows that the boron in the water is responsible for the major contribution to the CTC of 63.4 pcm/K, followed by the hydrogen in the water which account for another 8.5 pcm/K. ²³⁸U and ²³⁵U insert negative contributions of -10.2 and -2.1 pcm/K respectively. The reduced absorption rate of ¹⁰B is mainly due to the reduced density of the coolant; the spectrum hardening due to water temperature increase reduces the effective one-group absorption cross section of ¹⁰B from 321 barns in the nominal case to 315 barns.

The fuel temperature coefficient of reactivity (Figure 2) - although more negative than that of UO₂ at BOL - increases with burnup, and becomes positive at around 100 GWD/MTiHM. In the following we will focus on the FTC at 127 GWD/MT – close to EOL. At this burnup k_∞ is 0.80385 at nominal condition and 0.8067 after 100K fuel temperature increase, giving a FTC of about +4.8 pcm/K. The critical soluble boron is almost zero.

Table 3 gives the neutron balance at nominal and perturbed conditions, normalized to one fission neutron emission. It shows that the biggest individual contribution – about -6.72 pcm/K, comes from ²³⁹Pu; the only other substantial negative contribution comes from ²³⁸U. However, most of the nuclei present in the system at EOL give a positive contribution to the FTC of reactivity, the biggest coming from ²³⁵U, followed by hydrogen in the water, ¹³⁵Xe, hydrogen in the fuel and soluble boron.

Table 3: Fractional absorption at EOL: nominal versus perturbed fuel temperature.

Nucleus ID	Fission neutron emission	Absorption	Delta abs (pcm)
²³⁵ U	2.76E-01	1.36E-01	447.788
¹ H in H ₂ O	0.00E+00	6.16E-02	115.2687
¹³⁵ Xe	0.00E+00	1.96E-02	103.0139
¹ H in ZrH _{1.6}	0.00E+00	3.06E-02	98.2866
¹⁰ B in H ₂ O	0.00E+00	4.09E-02	76.56
¹⁴³ Nd	0.00E+00	2.16E-02	73.0709
¹⁵¹ Sm	0.00E+00	6.81E-03	48.0361
Zr in ZrH _{1.6}	0.00E+00	1.85E-02	33.3509
²³⁸ Pu	9.01E-04	6.66E-03	32.5728
¹⁴⁹ Sm	0.00E+00	7.88E-03	32.5313
¹⁰³ Rh	0.00E+00	1.44E-02	17.9113
...		...	
²³⁶ U	1.40E-03	1.39E-02	-0.6092
¹⁴⁸ Pm	0.00E+00	1.03E-03	-1.7122
⁶⁰¹ Pm	0.00E+00	2.89E-03	-1.7211
²⁴¹ Am	2.02E-05	7.88E-04	-3.0154
²⁴¹ Pu	1.49E-01	6.92E-02	-3.8201
²⁴⁰ Pu	6.16E-04	6.27E-02	-9.2563
¹⁵⁵ Eu	0.00E+00	5.74E-03	-18.6463
²³⁸ U	2.47E-02	1.67E-01	-86.8128
²³⁹ Pu	3.49E-01	1.88E-01	-672.766

Figure 3 shows the neutron spectra in the fuel, normalized per one fission neutron emission, while Figure 4 shows the difference between the perturbed and nominal spectra. The largest spectral variations occur below $\sim 1\text{eV}$; at higher energies the flux slightly decreases due to the fact that the effective fission cross section of ^{239}Pu increases due to the flux amplitude increase in the vicinity of its strong $\sim 0.3\text{eV}$ resonance. This phenomenon is illustrated and quantified in Figures 5 and 6.

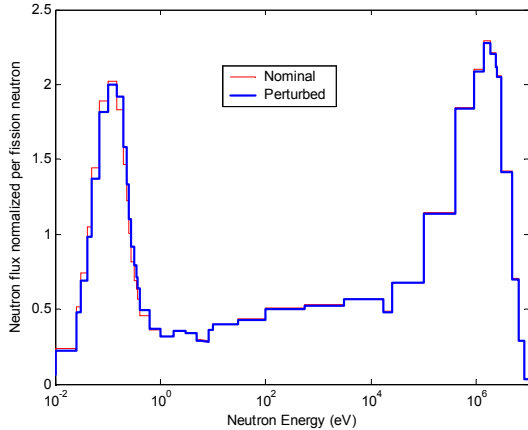


Figure 3: Neutron spectra in the fuel at EOL.

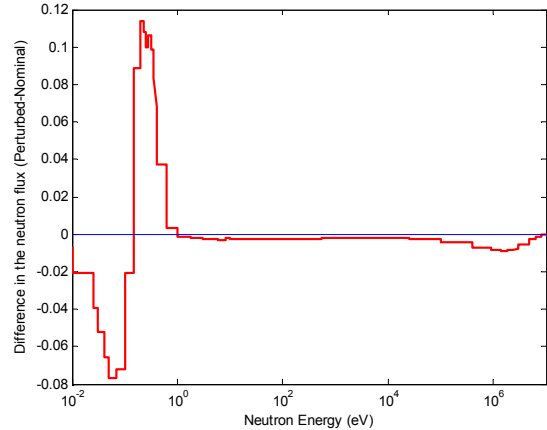


Figure 4: Difference in the fuel neutron spectra (perturbed - nominal) at EOL.

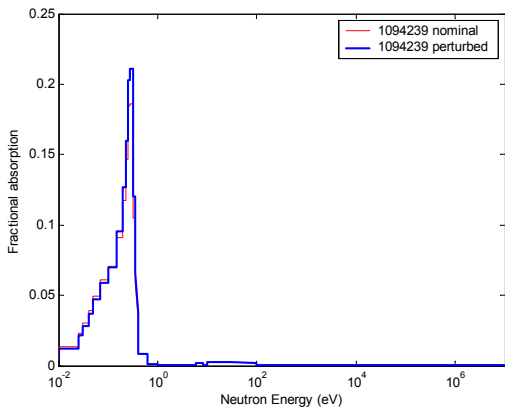


Figure 5: Fractional absorptions of ^{239}Pu at EOL for $\text{UZrH}_{1.6}$.

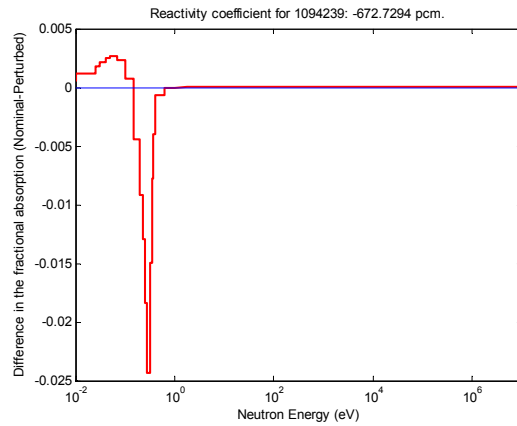


Figure 6: Difference in the ^{239}Pu fractional absorption at EOL for $\text{UZrH}_{1.6}$.

The substantial spectrum hardening due to fuel temperature increase is unique to hydride fuel; it is due to the presence of hydrogen in the fuel. This spectrum hardening is responsible for positive reactivity effect from the majority of the nuclei present in the reactor at EOL. As an example consider ^{235}U . Figure 7 shows the energy-dependent fractional absorption of ^{235}U , while Figure 8 shows how it varies between the nominal and perturbed conditions. The dominant contribution comes from the neutron spectral shift below 1eV in which energy range the ^{235}U absorption cross section behaves close to $1/v$ with a sharper drop around its $\sim 0.3\text{eV}$ resonance.

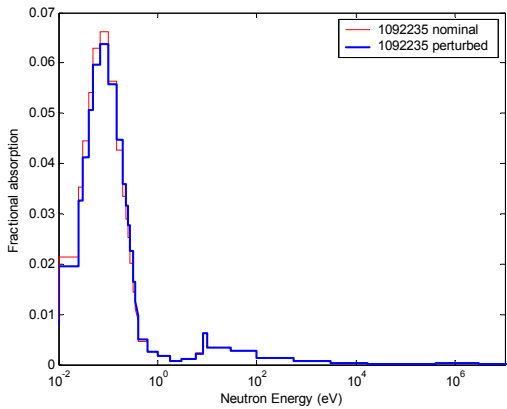


Figure 7: Fractional absorption of ^{235}U at EOL for $\text{UZrH}_{1.6}$.

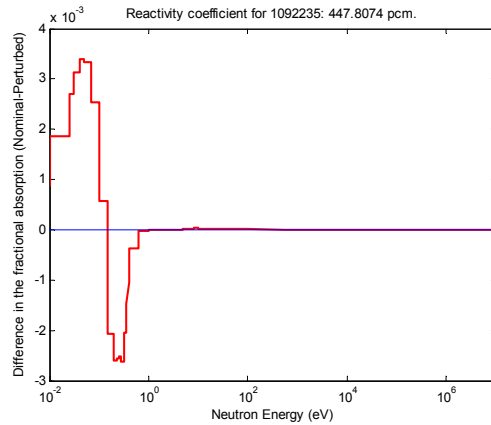


Figure 8: Difference in the ^{235}U fractional absorption at EOL for $\text{UZrH}_{1.6}$.

Why, then, thermal spectrum hardening does not make the FTC of reactivity positive also at lower burnups? In fact, at BOC the FTC of hydride fuel is even more negative than that of oxide fuel. Figures 9 and 10 show the BOL fuel spectra and spectra difference. Compared with Figures 3 and 4 it is observed that the thermal neutron flux and flux difference are significantly smaller at BOL than at EOL. This is due to the significantly higher boron and ^{235}U concentrations at BOL. However, the ratio of the area of flux amplitude increase (in the few tenth eV energy range) to flux amplitude decline (below ~ 0.1 eV) is significantly larger than at EOL (Figure 4). This difference is primarily due to the fact that at BOL the primary fissile isotope is ^{235}U the cross section of which dips around ~ 0.3 eV thus increasing the flux amplitude for a given fission rate, whereas towards EOL the ^{239}Pu makes a significant contribution to the fission rate and its fission cross section peaks around ~ 0.3 eV, thus reducing the flux amplitude. As a result, the fractional absorption per fission neutron in all the core constituents increases with fuel temperature increase at BOL, making the FTC negative. Of the -3.86 pcm/K BOL FTC, only -2.22 pcm/K BOL FTC are due to ^{238}U Doppler effect. The rest -1.74 pcm/K is contribution from fuel spectrum hardening that is unique to hydride fuel; -1.10 pcm/K from ^{10}B , -0.30 pcm/K from ^{235}U and -0.15 pcm/K from ^1H .

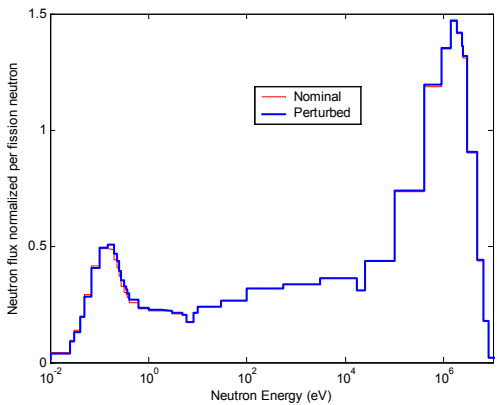


Figure 9: Neutron spectra in the fuel at BOL

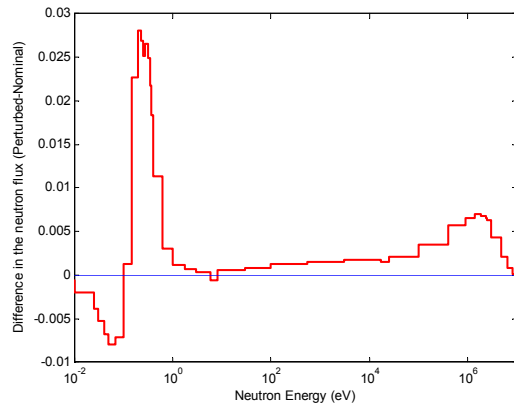


Figure 10: Difference in the fuel neutron spectra (perturbed - nominal) at BOL due to fuel temperature increase

3. Use of Burnable Poisons

Five burnable poisons (BP) were considered for making the RC negative – gadolinium, IFBA, natural erbium, enriched erbium (i.e., ^{167}Er 100%) and dysprosium. The amount of BP added was such as to achieve a core-average k_{inf} of 1.05 at the minimum of the fuel life. Figures 11 and 12 illustrate the effect of different burnable poisons on the CTC and FTC of reactivity. Erbium, either natural or enriched, was found the most effective; it makes the FTC of reactivity negative even at EOL due to its pronounced capture resonances in the few eV energy range. However, its use imposes some penalty on the attainable discharge burnup; $\sim 10\%$ for geometries of interest. Hence, even though BP can make all the RC negative throughout the cycle, we found use of thorium hydride as part of the hydride fuel a more desirable design approach.

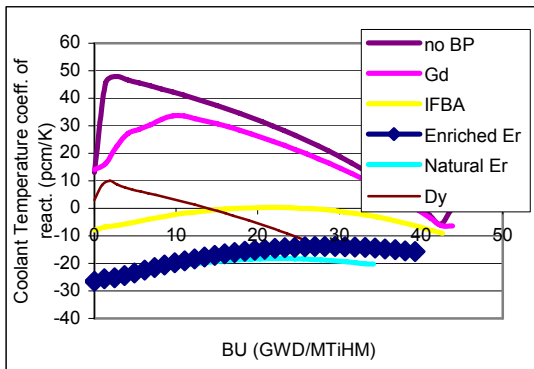


Figure 11: Effect of burnable poison on the CTC of reactivity

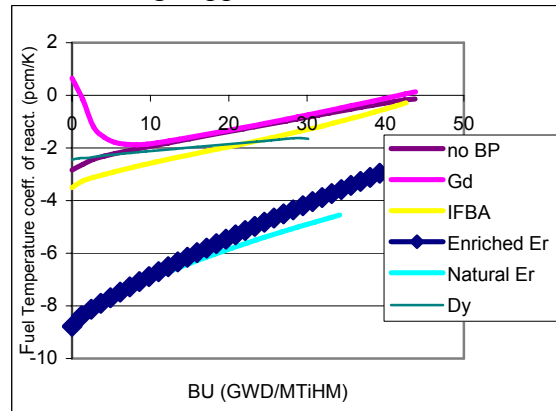


Figure 12: Effect of burnable poison on the FTC of reactivity

4. Use of ThH₂

A parametric study was performed for the reference unit cell trading different volume fraction of $\text{ZrH}_{1.6}$ by ThH_2 while maintaining constant the amount of 12.5% enriched uranium. Figure 13 shows the ThH_2 volume fraction effect on the attainable cycle length; it peaks around 16.5%. Figure 14 shows the coolant and fuel temperature RC as a function of burnup when the hydride

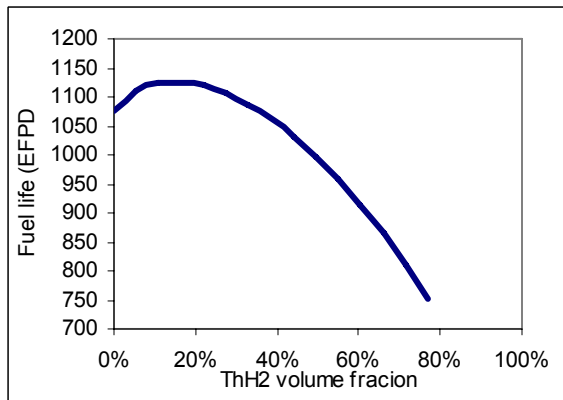


Figure 13: Attainable cycle length as a function of ThH_2 volume fraction.

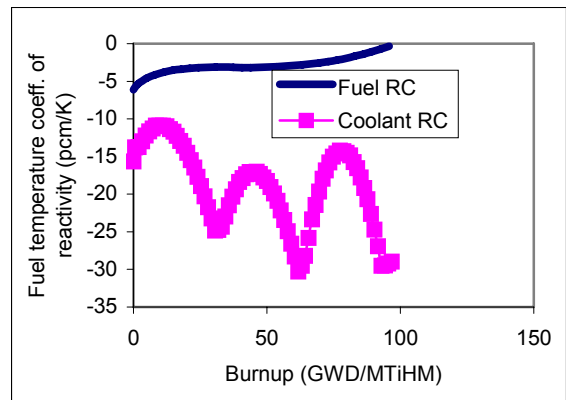


Figure 14: Coolant and fuel temperature RC as a function of burnup for $\text{U-ZrH}_{1.6}\text{-ThH}_2$ 16.5% ThH_2 with IFBA.

fuel contains 16.5 % ThH₂ and when IFBA is used to compensate for the majority of the excess reactivity. Both temperature coefficients are negative throughout the cycle. If soluble boron was the only means used to compensate for the excess reactivity, the amount of ThH₂ required for getting negative RC is about 77 %. This would have implied a substantial cycle length penalty. In the following we'll explore how the thorium, along with IFBA, makes the RC negative

The least negative CTC of reactivity is reached at around 10 GWD/MT: it is about -10 pcm/K. At this point the majority of the absorptions are in ²³⁵U, followed by ¹⁰B in the IFBA, ²³⁸U, ²³²Th and ²³⁹Pu; the absorption in soluble boron is negligible. As a result, coolant density drop has only a small positive contribution to the CRC +9.7 pcm/K and +7.3 pcm/K by, respectively, the coolant H and ¹⁰B, that are augmented by positive spectral contributions by ¹⁰B of the IFBA (+6.6 pcm/K) and ¹³⁵Xe (+2.1 pcm/K). These are more than compensated by the negative contributions from ²³⁸U (-20.4 pcm/K), ²³²Th (-8.9 pcm/K), ²³⁹Pu (-2.9 pcm/K), zirconium in the fuel and clad (-1.4 and -0.6 pcm/K) and ²⁴⁰Pu (-0.9 pcm/K).

The least negative FTC is obtained at EOL, i.e. at about 92 GWD/MT (Figure 14); its value is about -0.743 pcm/K, versus -6 pcm/K at BOL. Most of the absorptions are in ²³⁹Pu, followed by ²³⁸U, ²³⁵U, ²³²Th, ²³³U and the other isotopes of plutonium. The difference between the nominal and the perturbed condition, in pcm, is shown in the 4th column of Table 4: the largest positive contributions are given by ²³⁵U, ²³³U and ¹³⁵Xe, whereas the largest negative contributions come from ²³⁹Pu, ²³⁸U, other plutonium isotopes and ²³²Th.

Table 4: Fractional absorption at EOL: nominal versus perturbed fuel temperature.
U-ZrH_{1.6}-ThH₂ 16.5 %

Nucleus ID	Fission neutron emission	Absorption	Delta abs (pcm)
²³⁵ U	2.86E-01	1.42E-01	282.6347
²³³ U	1.81E-01	8.08E-02	123.3958
¹³⁵ Xe	0.00E+00	2.06E-02	77.1065
¹ H in ZrH _{1.6}	0.00E+00	2.12E-02	41.3195
¹ H in H ₂ O	0.00E+00	4.52E-02	40.025
¹⁴³ Nd	0.00E+00	1.85E-02	38.5605
¹⁵¹ Sm	0.00E+00	5.58E-03	28.2721
¹⁴⁹ Sm	0.00E+00	7.53E-03	21.4997
²³⁸ Pu	6.38E-04	4.12E-03	13.6333
		...	
²³² Th	2.56E-03	8.70E-02	-22.4038
²⁴¹ Pu	1.16E-01	5.36E-02	-36.656
²⁴⁰ Pu	6.00E-04	5.27E-02	-37.7121
²³⁸ U	2.47E-02	1.49E-01	-175.532
²³⁹ Pu	2.84E-01	1.54E-01	-536.039

Comparing Table 4 with Table 3 it is observed that the addition of thorium is responsible for a reduction in the fraction of neutrons absorbed in ²³⁹Pu, ²⁴⁰Pu and ²⁴¹Pu and to a significant increase in the fraction of neutrons absorbed in ²³⁵U and ²³³U as well in the primary fertile isotopes – ²³⁸U and ²³²Th. It is a combination of different isotopes and of different neutron spectrum that makes negative the FTC of reactivity of thorium-containing hydride fuel.

Figure 15 shows the spectrum in the fuel at EOL in the nominal and perturbed cases while **Figure 16** shows the difference between the nominal and perturbed spectra. Due to the ^{232}Th and ^{233}U contributions, the thermal component of the spectrum with thorium (Figure 15) is smaller than that without thorium (Figure 3). Compared against Figure 4, the positive-to-negative spectral difference component of Figure 16 is larger than of Figure 4 but not as large as of Figure 10.

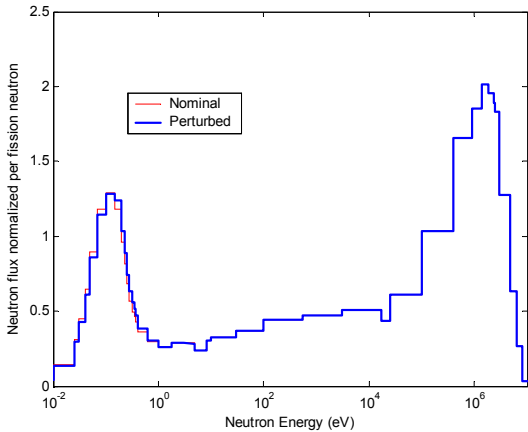


Figure 15: Flux spectra in the fuel at EOL; nominal and fuel-perturbed cases.

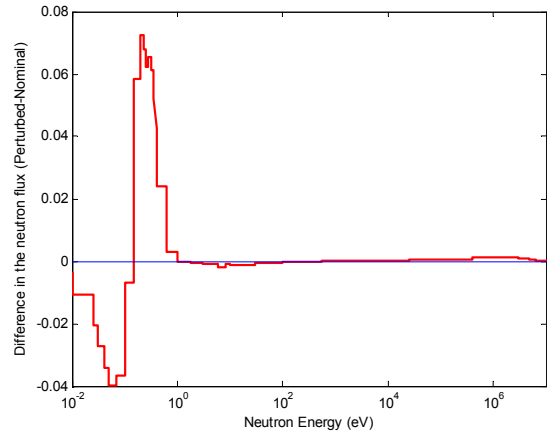


Figure 16: Difference in the fuel flux spectra (nominal - perturbed) at EOL.

The fractional absorption energy dependence in nominal conditions and the difference in this fractional absorption between the perturbed and nominal conditions are shown for ^{239}Pu , ^{235}U , ^{233}U and ^{238}U in respectively, Figures 17-18, 19-20, 21-22 and 23-24.

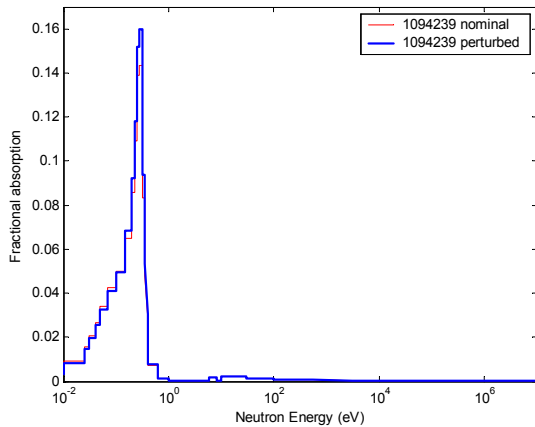


Figure 17: Fractional absorption of ^{239}Pu at EOL.

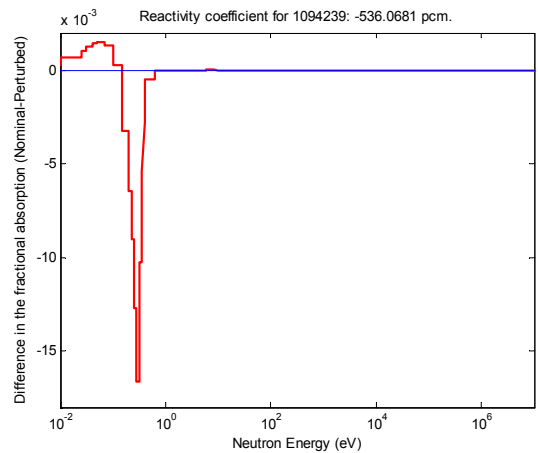


Figure 18: Difference in the fractional absorption (nom-pert) of ^{239}Pu at EOL.

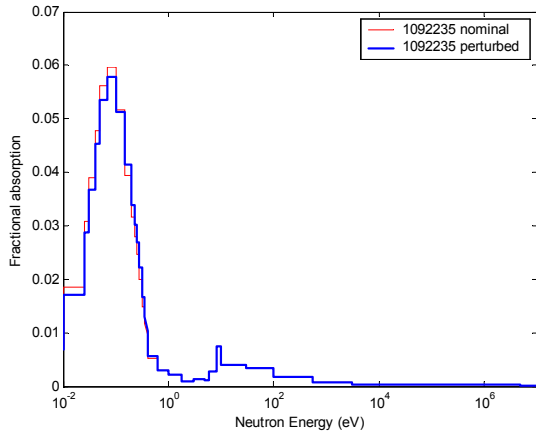


Figure 19: Fractional absorption of ^{235}U at EOL.

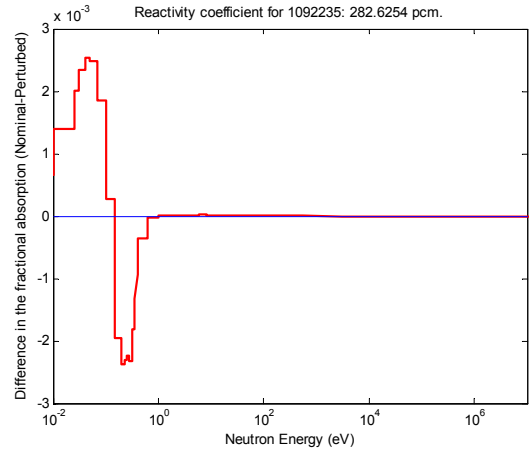


Figure 20: Difference in the fractional absorption (nom.-pert.) of ^{235}U at EOL

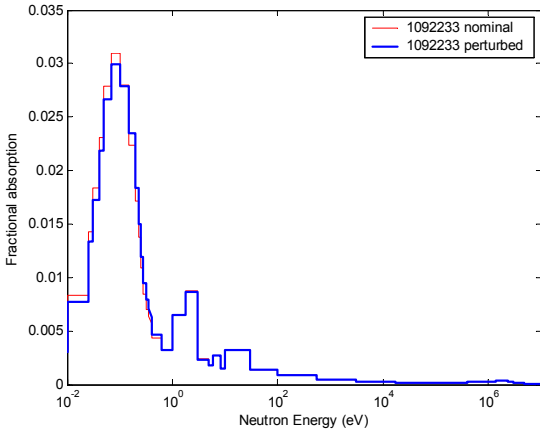


Figure 21: Fractional absorption of ^{233}U at EOL.

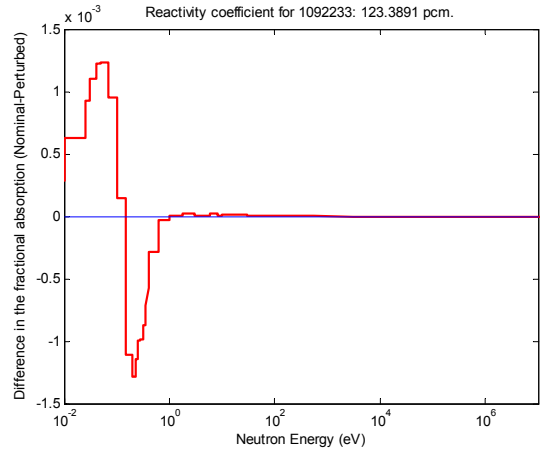


Figure 22: Difference in the fractional absorption (nom.-pert.) of ^{233}U at EOL

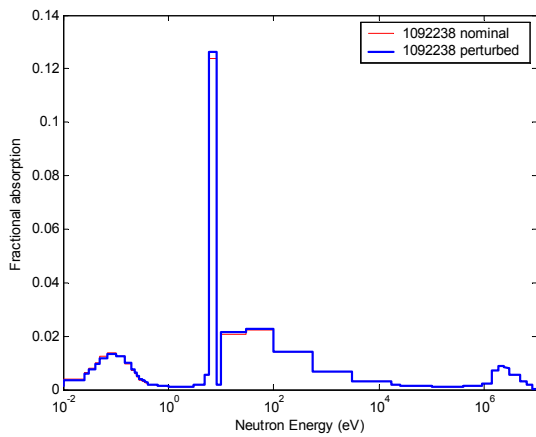


Figure 23: Fractional absorption of ^{238}U at EOL.

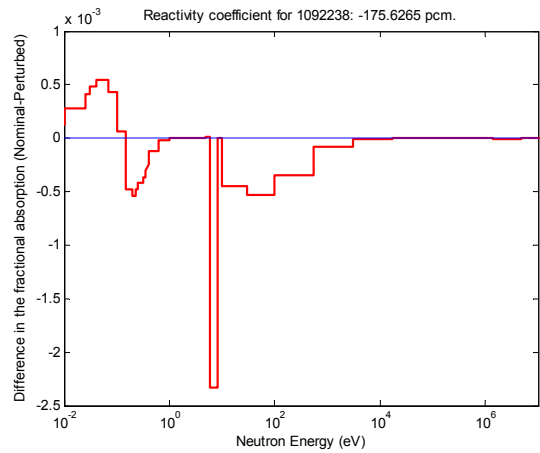


Figure 24: Difference in the fractional absorption (nom.-pert.) of ^{238}U at EOL

5. Conclusion

Despite of their large H/HM ratio relative to oxide fuel unit cells of identical geometry, hydride fuelled PWR cores can be designed to have negative coolant and fuel temperature coefficient of reactivity over their entire life. The preferred design approach identified is to replace, in 16.5 volume percent of the fuel, the zirconium hydride by thorium hydride and use as much IFBA as practical to compensate for the excess reactivity along with soluble boron in the water. Additional benefits provided by the use of thorium are:

- Increase the attainable core life through conversion to ^{233}U ;
- Reduced TRU inventory in the discharged fuel.

Samples of uranium, zirconium hydride and thorium hydride fuels have been successfully produced, characterized and underwent irradiation experiments [11].

Acknowledgement

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