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ASSESSMENT OF BURNABLE ABSORBER FUEL DESIGN BY U_WB_1 DEPLETION CODE

Martin Lovecký[a,*], Jana Jiřičková[a], Radek Škoda[a,b]

[a] *Regional Innovation Centre for Electrical Engineering, University of West Bohemia, Univerzitní 8, 306 14 Plzeň, Czech Republic*

[b] *Faculty of Mechanical Engineering, Czech Technical University, Technická 4, 160 07 Praha 6, Czech Republic*

[*] corresponding author: lovecky@rice.zcu.cz

Abstract. U_WB_1 depletion code is being developed as a fast computational tool for the study of burnable absorbers in University of West Bohemia in Pilsen, Czech Republic. The research of fuel depletion aims at a development and introduction of advanced types of burnable absorbers in nuclear fuel. Burnable absorbers compensate for the initial excess reactivity and consequently allow for lower power peaking factors and longer fuel cycles with higher fuel enrichments.

The paper describes the depletion calculations of CANDU, PWR and SFR nuclear fuel doped with rare earth oxides as burnable absorber. Uniform distribution of burnable absorber in the fuel is assumed. Based on performed depletion calculations, rare earth oxides are divided into two groups, suitable burnable absorbers and poisoning absorbers. Moreover, basic economic comparison is performed based on actual stock prices.

Keywords: burnable absorber, fuel depletion, Monte Carlo.

1. Introduction

U_WB_1 nuclear fuel depletion code [1, 4] is being developed as a fast fuel depletion code to conduct burnable absorber research. The goal of the research is to optimize new materials as burnable absorbers (BA) in nuclear fuel. BAs compensate for the initial excess reactivity and consequently allow for lower power peaking factors and longer fuel cycles with higher fuel enrichments. Research of advanced types of burnable absorbers (BA) in nuclear fuel requires fast depletion code that would be able to calculate broad range of elements, nuclides or their combination. State-of-art depletion codes require large amount of computational time, therefore, decision to develop fast depletion code was made.

Currently developed U_WB_1 depletion code comprises of transport and burnup solver that works in 2sPC depletion scheme in order to perform fast calculation of nuclear fuel depletion. In order to achieve high precision in U_WB_1 code, Monte Carlo solver for lattice cell calculations is used to solve transport equation. Bateman equations are solved in special depletion scheme that relies on nuclide-based predictor-corrector method in both transport and burnup part of fuel depletion.

The paper describes the depletion calculations of CANDU, PWR and SFR nuclear fuel doped with rare earth oxides as burnable absorber. Uniform distribution of burnable absorber in the fuel is assumed. Based on performed depletion calculations, rare earth oxides are divided into two groups, suitable burnable absorbers and poisoning absorbers. Moreover, basic economic comparison is performed based on current stock prices.

2. U_WB_1 depletion code

First version of the newly developed U_WB_1 fast nuclear fuel depletion code [1] significantly reduced calculation time by omitting the solution step for the Boltzmann transport equation. However, estimation of multiplication factor during depletion was not sufficiently calculated [2]. Moreover, 1-group effective cross sections for strong absorber models like gadolinium showed disagreement between the U_WB_1 tested code and the SERPENT reference code [3].

Monte Carlo transport solver for U_WB_1 code was introduced [4] in order to improve code accuracy and remove pre-calculated case-dependent data libraries, therefore, eliminate constant effective cross section assumption. Nuclear data from ENDF/B-VII.1 library is used for U_WB_1 Monte Carlo solver. Standard ray-tracing algorithm for neutron random walk is a part of the solver. Two-dimensional fuel pin models geometry, where fuel lattice model is assumed. The speed of the solver is higher than for the MCNP6 reference code. For light water reactor models, U_WB_1 Monte Carlo solver is on average 10 times faster.

Two-step predictor-corrector method (2sPC) was developed for U_WB_1 code. The idea is to change the coupling of transport and burnup solvers by omitting major fraction of transport solver callings, because transport solver is, by orders of magnitude, slower than burnup solver. Both transport and burnup variables are calculated for predicted states and corrected with more precise values as the two parts of fuel depletion are coupled. Only three transport solver solutions are used, the initial fuel state and predicted and corrected states for final burnup state. Effective cross sections are evaluated during fuel depletion by

assuming nuclide-based non-linear dependency. Multiplication factors in the depletion steps other than the first and the last one are estimated by neutron production to absorption ratio that is calculated without the need to call the transport solver.

Employing 2sPC depletion scheme leads to a significant reduction of calculation time by a factor of 10. With the Monte Carlo speed-up against the reference code, fuel depletion with U_WB_1 code is around 100 times faster than with MCNP6 reference code.

3. Burnable absorbers

In order to mark a material as a good burnable absorber, two properties are desired. These are a high neutron absorption cross section with low daughter nuclide's neutron absorption cross section. High neutron capture cross section causes neutrons to be absorbed, therefore, multiplication factor of the fuel is decreased and initial excess of reactivity is compensated. The absorber is burnable if the nuclide resulting from neutron absorption has a lower neutron absorption cross section, therefore, reactivity worth of inserting burnable absorber is decreasing during fuel depletion. The higher the differences between cross sections, the faster the burnable absorber will be burned. Typical absorption reaction for burnable absorbers is (n,γ), although another reactions are possible, e.g. (n,α) for light elements like boron.

Present nuclear fuels use mainly gadolinium, europium and erbium oxides. Gadolinium oxide is burned faster than the two others and because of the very high absorption cross section of Gd-157 nuclide, initial reactivity compensation for gadolinium higher than for other materials.

Gadolinium nuclides with interesting burnable properties are Gd-155 and Gd-157 with natural abundances 14.80 at% and 15.65 at%. Europium has natural abundance of 47.81 at% Eu-151 and 52.19 at% Eu-153. Both europium nuclides behaves as a good burnable absorber nuclide, both nuclides have have high neutron capture cross sections with end-product nuclide characterized by low capture cross section. Erbium nuclides with interesting burnable properties are Er-166 and Er-167 with natural abundances 33.50 at% and 22.87 at%. Total cross section for thermal energy 0.0253 eV are: 60 801 barns for Gd-155, 253 929 barns for Gd-157, 9190 barns for Eu-151, 367 barns for Eu-153, 31 barns for Er-166 and 652 barns for Er-167.

4. Rare earth oxides

Burnable absorber research will be conducted in two phases. The first phase will be represented by parametric study of various types of BA materials. The U_WB_1 depletion code will be used in order to calculate a large number of cases, employing its fast calculation scheme. Promising materials will be used in the second phase for a deeper neutronic analysis with state-of-art codes, possible discrepancies or inaccuracies of the U_WB_1 code will be removed. The output of the research is to sort materials by their suitability as BA from neutronics point of view. After that, another types of analyses, like chemical compatibility, thermal conductivity, economic evaluation and other relevant areas will be performed.

The paper focuses only on rare earth oxides as burnable absorbers. These are often used or planned to use as BA in PWR nuclear fuel. The most common is gadolinium oxide, followed by europium and erbium oxides. Rare earth oxides were briefly analyzed in [5] for VVER nuclear fuel, in this paper, PWR, CANDU and SFR nuclear fuels are compared.

Rare earth elements are available as metals or oxides. Thermal reactors and some of proposed fast reactors use uranium oxide fuel, therefore, rare earth oxides were evaluated. Seventeen rare earth elements are abundant in nature (scandium, yttrium and 15 lanthanides from lanthanum to lutetium). Most of them are available in oxides with X_2O_3 stechiometry, in order to introduce the same normalization for all elements, masses of all other rare earth cases were recalculated to have X_2O_3 stechiometry.

5. Calculation cases

CANDU, PWR and SFR nuclear fuel depletion was calculated. Of seventeen available rare earth oxides, 16 were calculated. Promethium, having only radioactive nuclides, was excluded from the study. It the first part of the study, criticality calculations with U_WB_1 code were performed in order to determine rare earth oxides content in the fuel (wt% X_2O_3). The target multiplication factor was selected to be around 0.05 lower than for the fresh fuel without BA, namely 1.10 for CANDU fuel, 1.25 for PWR fuel and 1.15 for SFR fuel (fresh fuel multiplication factors are 1.15, 1.32, 1.20). In the second part, fuel depletion with U_WB_1 code were calculated and multiplication factor dependency on fuel burnup was analyzed. Finally, economic comparison was performed.

U_WB_1 code requires simple fuel cell geometry. Recently, the option to include arbitrary number of concentric cylinders with either square or triangular lattice was included. CANDU fuel was homogenized and modeled as 18 concentric cylinders that represents four fuel rings of the total number of 37 pins. Natural uranium oxide fuel with final burnup 10 000 MWd/MTU was assumed. Fuel depletion was divided into 31 steps with constant power 32.4364 MW/MTU. Standard CANDU materials were used in the model – Zircaloy-4 for fuel cladding, Zr2.5Nb alloy for pressure tube that is divided by CO_2 gap from Zircaloy-2 calandria tube, heavy water with density 0.8179 g/cm^3 and purity 99.11 wt% as coolant in the pressure tube and heavy water with density 1.086 99 g/cm^3 and purity 99.97 wt% as moderator between calandria tube.

PWR nuclear fuel was modeled as 5.0 wt% U-235 enriched uranium oxide with Zircaloy-4 cladding and light water moderator with 600 ppm boric acid. Fuel radius 0.4025 cm and cladding radius 0.4750 cm was

used, lattice half pitch 0.6295 cm was assumed as typical PWR dimensions. The fuel was depleted with power 40.0 MW/MTU in 34 steps up to final burnup 50 000 MWd/MTU.

SFR nuclear fuel was modeled as 15.0 wt% U-235 enriched uranium oxide with 7.9 g/cm^3 iron cladding and 0.88 g/cm^3 sodium coolant. Fuel radius 0.28 cm and cladding radius 0.33 cm was used, lattice half pitch 0.42 cm were used as typical SFR dimensions (BN-800 reactor was assumed as typical SFR reactor). The fuel was depleted with power 130.0 MW/MTU in 74 steps up to final burnup 150 000 MWd/MTU, three times higher than of PWR fuel.

6. Criticality results

CANDU, PWR and SFR nuclear fuel criticality calculation results from U_WB_1 code are presented in Fig. 1 to Fig. 3. The figures show dependency of multiplication factor for fresh fuel with various BA content (mass fraction) on logarithm scale.

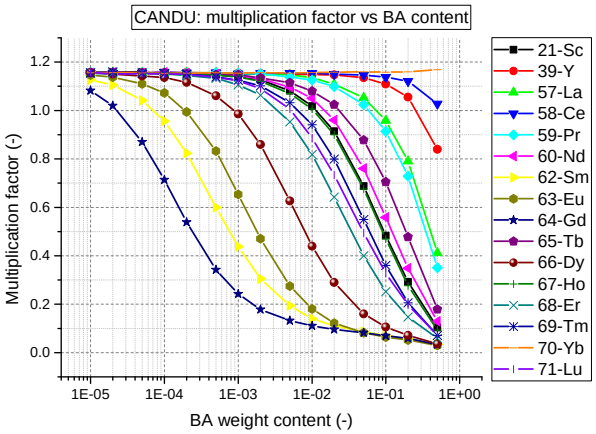

Figure 1. CANDU multiplication factor vs BA content.

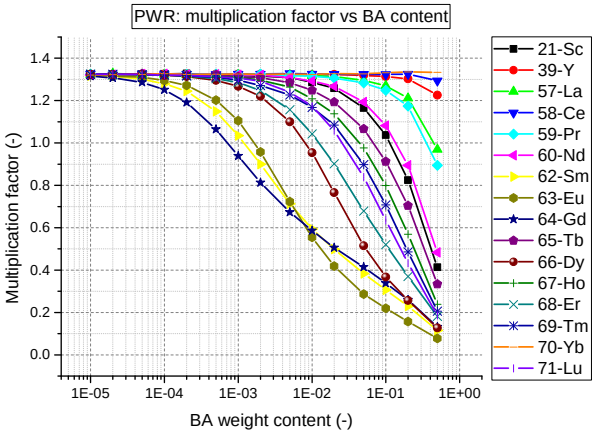

Figure 2. PWR multiplication factor vs BA content.

Multiplication factor reduction from BA insertion into the fuel is higher for CANDU fuel that needs only a small BA content to achieve target multiplication factor. PWR fuel is similar to CANDU, however, due to higher fuel enrichment, more BA content is needed. Lastly, SFR with fast neutron spectra and high fuel enrichment requires considerably higher BA content than previous fuel types in order to achieve desirable initial reactivity compensation.

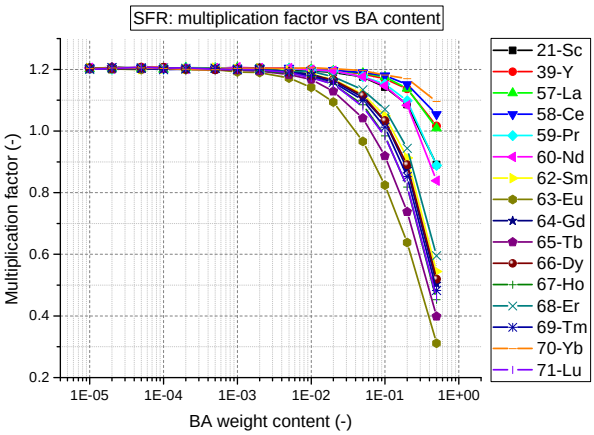

Figure 3. SFR multiplication factor vs BA content.

Based on performed criticality calculations and selected target multiplication factor values 1.10 for CANDU fuel, 1.25 for PWR fuel and 1.15 for SFR fuel, BA weight fraction was calculated. Linear interpolation between selected BA contents (3 values for each logarithm order) was used. Calculated BA content is shown in Fig. 4 to Fig. 6.

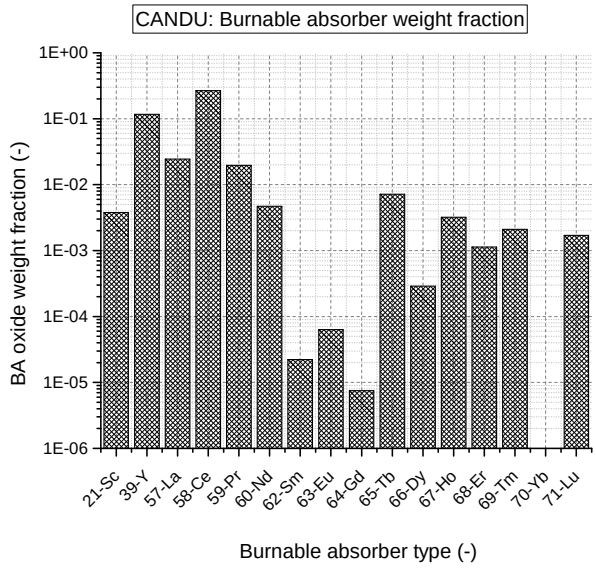

Figure 4. CANDU burnable absorber weight fraction.

In the case of both CANDU and PWR fuel, even maximum considered BA content (50 wt% X_2O_3) was not enough for dropping the multiplication factor to the target one. For PWR fuel, 58-Ce content was also too high. For SFR fuel, it was possible to achieve desired target multiplication factor for all rare earth oxides BA.

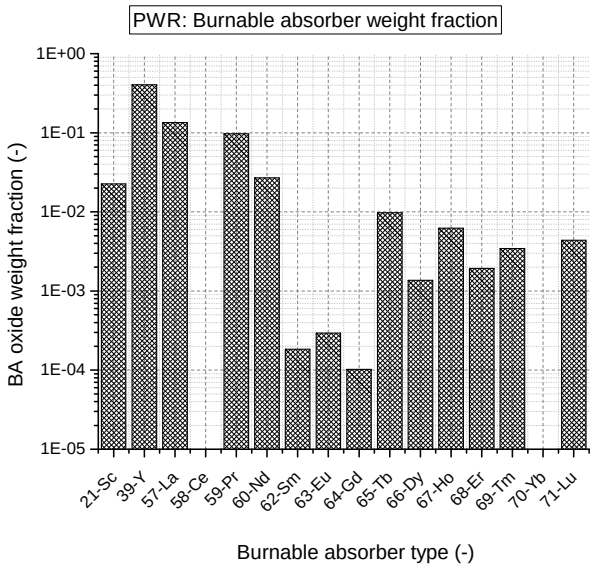

Figure 5. PWR burnable absorber weight fraction.

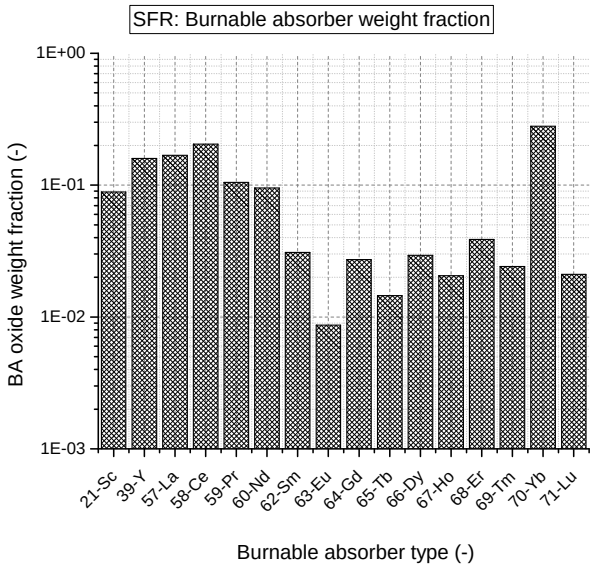

Figure 6. SFR burnable absorber weight fraction.

Minimum BA content was calculated for gadolinium oxide for CANDU and PWR fuel (7.52×10^{-6} and 1.02×10^{-4} weight fraction) and for europium oxide for SFR fuel (8.67×10^{-3} weight fraction).

For most of rare earth oxides, CANDU and PWR fuel requires BA content from 0.1 wt% X_2O_3 to 1.0 wt% X_2O_3. For SFR fuel, required BA content mostly varies from 1.0 wt% X_2O_3 to 10.0 wt% X_2O_3, i.e. 10 times higher than of thermal spectra fuels.

Maximum BA content was calculated for 58-Ce. For CANDU fuel, 26.8 wt% Ce_2O_3 was determined. Slightly lower conten of 20.4 wt% Ce_2O_3 was calculated for SFR fuel. PWR fuel required more than 50.0 wt% Ce_2O_3 and cerium oxide was excluded, making 39-Y the case of maximum BA content for PWR fuel (40.5 wt% Y_2O_3).

7. Depletion results

CANDU, PWR and SFR nuclear fuel criticality calculation results from U_WB_1 code are presented in Fig. 7 to Fig. 9.

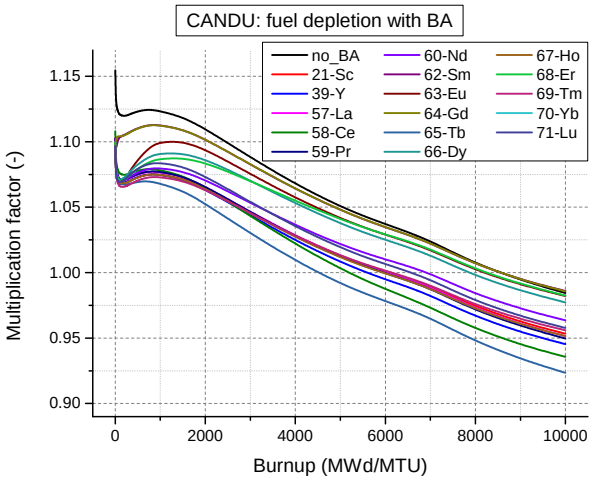

Figure 7. CANDU multiplication factor during fuel depletion.

CANDU figure shows that from macroscopic view, only 63-Eu and 64-Gd compensate initial reactivity in the way that initial reactivity transient due to fission product build-up almost disappears. For other BA, initial reactivity is compensated, however, initial reactivity transient is still presented. For good burnable absorbers, residual poisoning (decrease of multiplication factor compared to no BA case) is negligible, other BA have residual poisoning that, for some cases, is even higher than initial reactivity compensation.

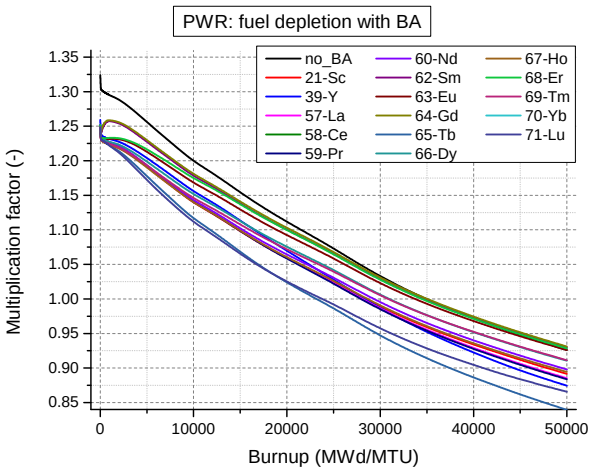

Figure 8. PWR multiplication factor during fuel depletion.

For PWR fuel, similar conclusions as for CANDU fuel can be deduced. Only 63-Eu and 64-Gd oxides are able to change the shape of multiplication factor dependency on burnup, i.e. remove initial reactivity transient. However, for PWR fuel, initial reactivity transient is not only removed, but the effect is reversed,

multiplication factor increases in the first days of fuel operation.

For SFR fuel, all rare earth oxides show similar behavior, burning of the oxides is very slow and reactivity compensation remains relatively constant, therefore, rare earth oxides behaves mostly as non-burnable absorbers.

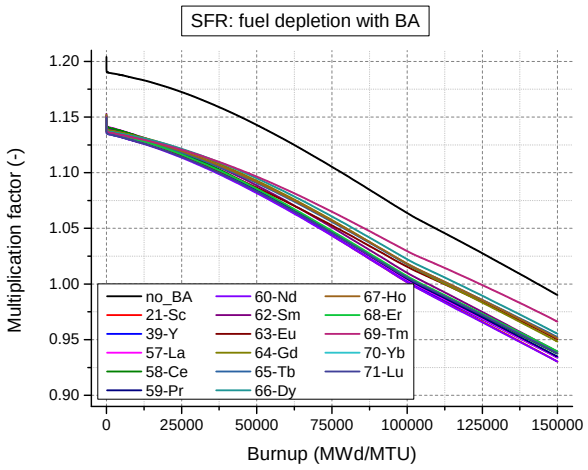

FIGURE 9. SFR multiplication factor during fuel depletion.

Based on performed depletion calculations and multiplication factor progress during fuel depletion, BA worth was calculated. BA worth is defined as reactivity differences between two cases, the one with BA in the fuel and the one without BA. Results are depicted in Fig. 10 to Fig. 12.

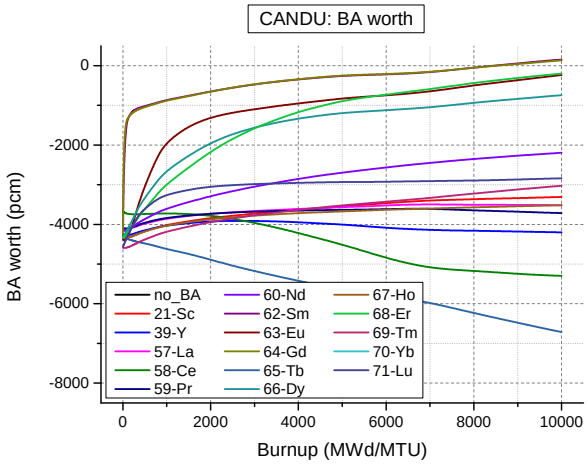

FIGURE 10. CANDU burnable absorber worth during fuel depletion.

From BA worth curve variation during fuel depletion, it can be stated which rare earth oxides behaves like more or less suitable burnable absorbers and which behaves like poisoning absorbers. The latter group can be divided into constant poisoning absorbers (BA worth is constant function of burnup) and increasingly poisoning absorbers (BA worth is decrasing function of decrasing function of burnupburnup, i.e. diverges

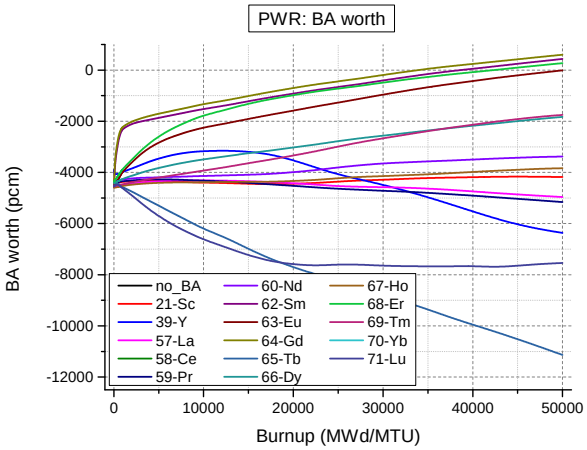

FIGURE 11. PWR burnable absorber worth during fuel depletion.

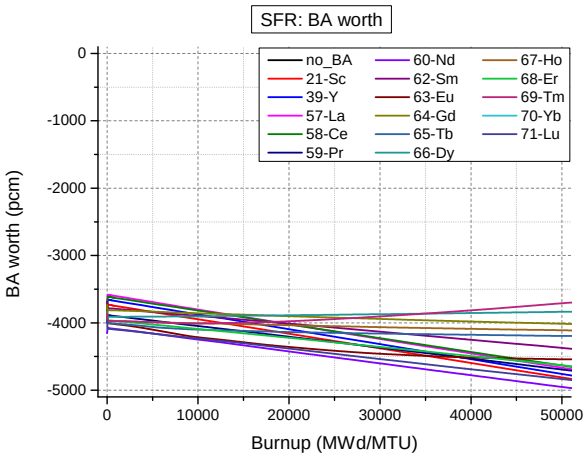

FIGURE 12. SFR burnable absorber worth during fuel depletion.

from zero reactivity worth, rather than converging behaviour of burnable absorbers).

Because of the fact that BA worth curve does not change its derivation sign (constant, increasing or decreasing function), it is possible to define ratio of EOC to BOC reactivity worth and compare it to unity, see final results in Fig. 13.

CANDU and PWR fuel behaves similarly, burnable absorbers are more depleted for final burnup in the case of CANDU fuel, therefore, residual poisoning is lower for CANDU and burnable absorbers are more suited for CANDU fuel.

Total number of 9 out of 16 rare earth oxides are marked as burnable absorbers for both CANDU and PWR fuels, namely 21-Sc, 60-Nd, 62-Sm, 63-Eu, 64-Gd, 66-Dy, 67-Ho, 68-Er and 69-Tm. Total number of 4 out of 16 rare earth oxides are marked as burnable absorbers only for CANDU fuel, but behaves as poisoning absorbers for PWR fuel. These are 39-Y, 57-La, 59-Pr and 71-Lu. The rest 3 out of 16 rare earth oxides, 65-Tb, 58-Ce and 70-Yb, are poisoning absorbers for both CANDU and PWR fuel.

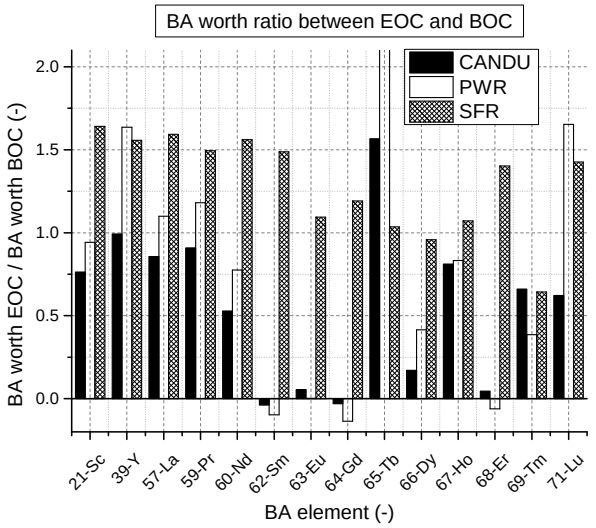

Figure 13. Rare earth oxides as BA - EOC/BOC worth ratio.

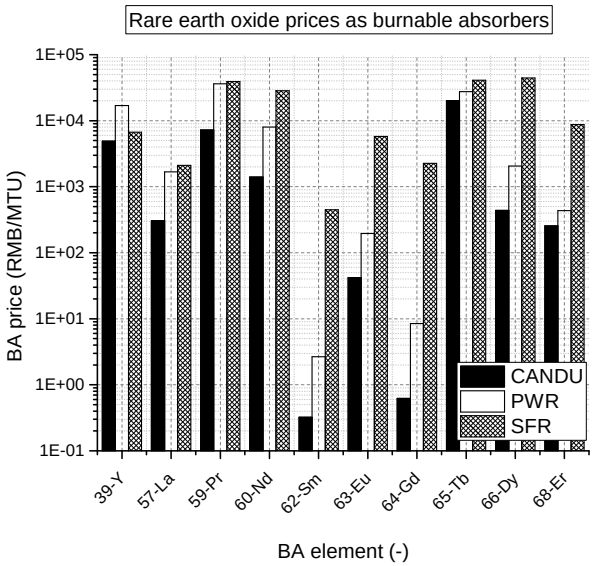

Figure 14. Rare earth oxides as BA – economic comparison.

Situation for SFR fuel is simpler than for both thermal spectra fuels. Only 2 out of 16 rare earth oxides, 66-Dy and 69-Tm, are suitable as burnable absorbers. Thulium oxide is a far better BA for SFR than dysprosium oxide that burns very slowly. However, significant fraction of thulium oxide remains in the fuel at EOC.

8. Economic comparison

Economic comparison of rare earth oxides as burnable absorbers is based on prices from early 2016 [6], however, rare earth oxides are mined mainly in one country and prices are volatile. Table 1 (or Fig. 14) summarizes the results. Input oxides prices (RMB/MT) were converted to match X_2O_3 stechiometry and multiplied by target BA content (weight fraction) to evaluate oxides prices for unit uranium mass (RMB/MTU). Total fuel price comprises of BA price, uranium price and other factors, only BA price is compared in this study.

Element	Price (RMB/MT)
39-Y	34 000
57-La	13 250
58-Ce	13 900
59-Pr	429 000
60-Nd	322 500
62-Sm	17 000
63-Eu	1900
64-Gd	87 500
65-Tb	4300
66-Dy	1915
68-Er	265 000

Table 1. Rare earth oxides as BA – economic comparison.

For the best burnable absorbers, the economic comparison showed the price of rare earth oxide in MTU vary from 1 RMB to 1000 RMB for CANDU and PWR fuels and approximately 100 times higher prices for SFR fuels.

9. Conclusions

U_WB_1 depletion code was used to assess burnable absorber design in the nuclear fuel. CANDU, PWR and SFR fuel lattice criticality and depletion calculations were performed for rare earth oxides as burnable absorbers uniformly distributed in the fuel.

Rare earth oxides content needed to achieve target initial reactivity compensation was evaluated. Based on multiplication factor behavior during fuel depletion and BA reactivity worth ratio for final burnup and inital state, rare earth oxides were divided into two groups, burnable absorbers and poisoning absorbers.

CANDU and PWR fuel behaves similarly. Total number of 9 out of 16 rare earth oxides are marked as burnable absorbers for both CANDU and PWR fuels, namely 21-Sc, 60-Nd, 62-Sm, 63-Eu, 64-Gd, 66-Dy, 67-Ho, 68-Er and 69-Tm. Total number of 4 out of 16 rare earth oxides are marked as burnable absorbers only for CANDU fuel, but behaves as poisoning absorbers for PWR fuel. These are 39-Y, 57-La, 59-Pr and 71-Lu. The rest 3 out of 16 rare earth oxides, 65-Tb, 58-Ce and 70-Yb, are poisoning absorbers for both CANDU and PWR fuel.

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For the best burnable absorbers, the economic comparison showed the price of rare earth oxide in MTU

vary from 1 RMB to 1000 RMB for CANDU and PWR fuels and approximately 100 times higher prices for SFR fuels.

Further analysis of possible burnable absorbers with broad range of elements, nuclides or their combination is in progress. The research aims to a quantitative comparison of materials that could be used as burnable absorbers. The resulting data are planned to be utilized in subsequent fuel design analyses including thermal conductivity, chemical compatibility, irradiation stability, economic evaluation etc.

List of symbols

BOC beginning of cycle (irradiation)
EOC end of cycle (irradiation)
MT metric ton
MTU metric tons of uranium
RMB renminbi (Chinese currency system)

Acknowledgements

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