Neutronic analysis of mixed thorium-uranium fuel bundle for CANDU reactors

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Abstract. The paper shows neutronic analysis of the CANDU (CANada Deuterium Uranium) nuclear reactor fuel channel with mixed thorium-uranium fuel bundles. The numerical model of the fuel channel was designed using The Monte Carlo Continuous Energy Burn-up Code – MCB developed at the AGH University of Science and Technology, Faculty of Energy and Fuels, Department of Nuclear Energy. The super-computer Prometheus available in the frame of the PI-Grid Infrastructure at the Academic Computer Centre Cyfronet AGH was used for multi-scale calculations. The fuel bundles are composed of two clusters of fuel rods. The neutronic analysis considers detailed numerical simulation of neutron transport in fully heterogeneous geometry of the fuel channel. Moreover, burnup simulations were performed using Transmutation Trajectory Analysis method implemented in the MCB code. In the analysis we mainly consider time evolutions of neutron multiplication factor, fissile $^{233}$U, $^{235}$U, $^{239}$Pu and fertile $^{238}$U and $^{232}$Th. The simulations were performed for eight scenarios with various fuel composition.

1. Introduction

The concept of using heavy water (deuterium oxide, D$_2$O) as a moderator in nuclear reactors was investigated since the very beginning of commercial utilization of nuclear power. The research on heavy water reactors were conducted in Canada, France, Germany, Italy, Japan, Sweden, Switzerland, the UK, the USA and the former USSR. All of the research and prototype reactors used heavy water as a moderator, however the designs were different, e.g. pressure tube heavy water cooled, pressure tube light water cooled, pressure vessel heavy water cooled and pressure tube gas cooled.

In the end only Canada reached the stage of commercial operation with its heavy water moderated heavy water cooled pressure tube reactor named CANDU. The first one of this type was Nuclear Power Demonstration (NPD), which was ran successfully from 1962 to 1987 in Rolphton, Ontario, generating 22 MW$_c$. The second CANDU with electric power of about 200 MW was Douglas Point reactor and it has started its operation in 1968. After some improvements of the design and further power increase the first multi-unit station was constructed in Pickering, Ontario, with 8 CANDU reactors, each generating above 500 MW$_c$. Last units came in operation in 1983. The Pickering design was the base for CANDU-6 reactor that is operated successfully in Canada and other countries since that time.
With its unique and reliable reactor design Canada entered the international nuclear market. It has assisted the Indian Department of Atomic Energy (DAE) in the construction of two 200 MW$_e$ reactors (Douglas Point type) - Rajasthan 1 and 2. However, Indian first atomic bomb detonation in 1974 ended the nuclear cooperation between these countries. Since then India worked on its own CANDU-like designs and it now has about 20 heavy water reactors in operation. Canada has sold the CANDU design to Argentina, Romania, South Korea, Pakistan and China, which results in 10 such reactors being operated in these countries. In Canada there are 19 CANDU reactors running, most of them in Ontario [1].

2. Technology
The main purpose of using heavy water instead of common light water as a moderator is reduced neutron absorption rate in deuterium oxide. Thanks to that lower concentration of fissile atoms is required to maintain the chain reaction and natural uranium fuel can be used as well as some alternative fuels like MOX (mix of uranium and plutonium oxides), RU (“recovered uranium” from used Light Water Reactor’s fuel (LWR)) or thorium. Using natural uranium eliminates the cost of enrichment process and the risk of proliferation. However, additional cost is generated by the need for pure heavy water to fill the core and heat transfer system. What is more, D$_2$O is less effective as a moderator and greater amount of water is required for the same amount of fuel. Natural or recovered uranium fuel has to be reloaded more frequently.

To deal with the specificity of heavy water coolant and moderator a different constructions of the reactor core were designed in comparison to the typical LWR. In CANDU the core is contained in a cylindrical austenitic stainless steel containment called Calandria where the heavy water is maintained in low temperature (~80˚C) and low pressure (~0.1 MPa) – see (Figure 1). In such containment there are about 380-480 horizontal pressure tubes, each containing 12 half-meter fuel assemblies lying end to end. Placing the fuel in individual tubes allows for online refuelling which improves the capacity factor [2].

![Figure 1. Schematic diagram of a CANDU reactor [3].](image)

3. Thorium fuel cycle
Thorium occurs in nature in a single form of $^{232}$Th and is about three times more abundant than uranium. It is a fertile isotope which captures a neutron and transmutes to the fissile $^{233}$U. For this reason thorium fuel has to be combined with another fissile material able to provide sufficient neutron flux, such as $^{233}$U, $^{235}$U or $^{239}$Pu. Introduction of thorium to the general use in nuclear power plants would increase fuel diversification and safety of supply. It would also vastly prolong the sufficiency of estimated conventional nuclear fuel, which is about 100 years for today. The advantages of thorium fuel cycle together with its drawbacks comparing to traditional uranium cycle are presented below in Table 1 [4, 5].
Table 1. Advantages and disadvantages of thorium fuel cycle.

| Advantages | Disadvantages |
|------------|---------------|
| Good chemical and physical properties: higher melting point, higher thermal conductivity and chemical stability, lower coefficient of thermal expansion, less gas fission products released. | Difficult and costly fuel fabrication for potential close fuel cycle because of high γ radiation from $^{232}$U. |
| Less minor actinides and plutonium in spent fuel. | Likelihood of using $^{233}$U for nuclear weapon. |
| Proliferation resistance: irradiated fuel contaminated with easily detectable $^{232}$U. | Neutron source required to produce fissile $^{233}$U from $^{232}$Th. |
| Thorium is more abundant, safer and more effective in mining. | Relatively long time of $^{232}$Th breeding to $^{233}$U due to $^{233}$Pa half-life of 27 day. |
| Possibility of breeding fuel in thermal reactors. | Need of many costly researches and licensing processes before starting industrial use. |

The CANDU reactors are capable of operating on many types of nuclear fuels [6]. Such versatility makes them unique among other types of nuclear reactors. The need for proliferation-resistance, burnup extension, reduction of plutonium inventory as well as in situ use of bred fissile material has followed to interest in thorium based fuel cycles [7]. The channel design, excellent neutron economy, on-power refuelling, and simple fuel bundle design of CANDU reactors significantly facilitate the introduction of thorium fuel in this nuclear system. To sum up, the CANDU reactors offer a proven, safe and reliable reactor technology for thorium fuel cycles.

4. Methodology

The MCB code available at the supercomputer Prometheus of the Academic Computer Centre Cyfronet of the AGH University was used for all numerical simulations [8, 9]. The code was validated with post-irradiation assay of the PWR spent fuel assembly, which proofed its reliability for calculations of isotopic changes in neutron flux [10]. In addition, the code was merged with other numerical tools, especially for coupled neutron-thermo-hydraulic-burnup calculations [11]. Firstly, the 3D fully heterogeneous model of the CANDU-6 fuel channel was designed using available data [12]. Then, the model was verified in the series of short-time numerical simulations. All detected bugs in coding were corrected and the checked input file was saved for further research. Eight cases listed in Table 2 were simulated using established numerical set-up. The first case without thorium component in fuel - just with natural uranium is a reference case because reflects behaviour of the real fuel channel of CANDU-6 reactor. The fuel channels contains 12 fuel bundles with 37 fuel rod each. The length of the fuel equals about 50 and the dimeter of the pin about 1.2 cm. The mass of fuel per bundle depends on density and fraction of all fuel components i.e. ThO$_2$ and UO$_2$. The fuel bundle was divided into two fuel regions: the outer region containing 30 fuel rods and inner region containing 7 fuel rods. The division allows implementation of different fuel types in each region. Therefore, it was possible to design the considered fuel bundle using so called seed-blanket concept, where more reactive driver fuel (outer region) provides neutrons for fertile fuel (inner region). The fuel channel was surrounded numerically by the reflective boundary conditions, which means that in numerical simulations we do not take into account neutron leak age outside fuel channels, and thus we estimated infinite neutron multiplication factor ($K_{inf}$) instead of effective neutron multiplication factor ($K_{eff}$). The fuel channel from both ends was surrounded by stainless steel walls with inner tube filled with D$_2$O, which is a good approximation of side reflector. In practice its design in extremely complicated due to structural elements necessary for operation of fuelling machine. In the simulations we consider isotopic changes due to neutron interactions in nuclear fuel for one year of irradiation on average power of 5.4 MW$_{th}$, divided into 12 steps of 30 days except of case C1 were additional discretisation of 10 days periods for first 100 days were applied. Figure 2 shows cross section of the numerical model plotted by the VISED utility available with MCNP family codes. The MATLAB software were
used for data post-processing. The ENDF 7.1 libraries were linked to the Monte Carlo solver. The average time for 3·10^6 neutron histories equals about 3h using 2 nodes of 24 cores each (Xeon E5-2680v3 12C 2.5 GHz), which gives precision of K_{inf} of about 25 pcm.

### Table 2. Investigated scenarios.

| Index | Case       | Description                                      |
|-------|------------|--------------------------------------------------|
| C1    | NAT        | Natural uranium in both zones. Reference case.    |
| C2    | 5-U        | 5 % enriched uranium in both zones.              |
| C3    | NAT-TH     | Natural uranium in outer zone. Thorium in inner zone. |
| C4    | 5-TH       | 5 % enriched uranium in outer zone. Thorium in inner zone. |
| *C5   | 20-TH      | 20 % enriched uranium in outer zone. Thorium in inner zone. |
| C6    | NAT-NAT-TH | Natural uranium in outer zone. 40% natural uranium and 60% thorium in inner zone. |
| C7    | 5-40-5-TH  | 5% enriched uranium in outer zone. 40% of 5% enriched uranium and 60% thorium in inner zone. |
| *C8   | 20-40-20-TH| 20% enriched uranium in outer zone. 40% of 20% enriched uranium and 60% thorium in inner zone. |

* The maximal allowed enrichment level for commercial nuclear reactors equals 5% while for research reactors 20%.

### Figure 2. Cross sections of designed numerical model.

5. Results

In the analysis we consider time evolutions of infinite neutron multiplication factor K_{inf}, fissile isotopes $^{233}$U, $^{235}$U, $^{239}$Pu, fertile isotopes $^{235}$U, $^{232}$Th and dependences between presented parameters. Figure 3 presents evolution of K_{inf} for eight considered scenarios. The initial value of K_{inf} depends strongly on the content of fissile and fertile material in the fuel. The higher the initial content of fissile material is the higher is K_{inf}, which is shown in Table 3. The K_{inf} drops almost linearly except of first month of irradiation. During this time its behavior depends on mass and type of fertile material. The initial increases for cases C3 and C6 is a result of high mass of fertile material and low mass of fissile material in the initial fuel. The increase is strictly related to the enhanced breeding of $^{239}$Pu from $^{238}$U and $^{233}$U from $^{232}$Th, which is shown in Figure 4. In general, the breeding of fissile material depends on the content of fertile material in the initial fuel – see Table 4. As it is shown in Figure 4, even in case C2, where enrichment equals 5% the $^{239}$Pu breeding is strongly limited comparing to scenarios with natural uranium fuel and uranium-thorium fuel. The worst breeding capabilities were achieved for the high enriched fuels(C5 and C8) because the initial mass of fissile $^{235}$U is sufficient for the system operation. To sum up, the value of the K_{inf} depends on the ratio of fissile to fertile material. The low magnitude of the ratio corresponds to the good breeding and thus to low value of K_{inf} and vice versa. In addition, in Figure 5 we present the initial decrees in K_{inf} and increase at about 40 days of
irradiation for case C1. The first effect occurs due to the formation of absorbing $^{135}$Xe form fission while second from enhanced breeding of plutonium called plutonium peak. The breeding of fissile isotopes depends on the content of fertile isotopes in the initial fuel. The highest production of $^{239}$Pu and $^{233}$U was obtained for cases C3 and C6 with natural uranium and thorium components. In the first case 700 grams of fissile material was produced while in the second one about 600 grams after one year of irradiation. The conversion ratio for the considered nuclear system is below unity because no surplus mass of fissile isotopes was produced. The lowest difference between initial and final mass of fissile isotopes was obtained for case C3. In principle, fuel for cases C2, C5, C7, C8 is too reactive, which will cause problems in reactivity control while fuel for case C3 shows subcriticality. The mentioned cases are the numerical cases necessary in the first stage of parametric studies to assess system neutronic characteristics for proper choice of nuclear fuel for further investigations.

### Table 3. Masses of fissile isotopes.

| Case         | $^{235}$U [g] | $^{239}$Pu [g] | $^{239}$Pu+$^{233}$U [g] | $^{235}$U [g] | Fissile Material [g] |
|--------------|---------------|----------------|--------------------------|---------------|----------------------|
|              | EOL | EOL | EOL | BOL | EOL | DIF | BOL | EOL | DIF | BOL | EOL | DIF |
| C1 NAT       | NA  | 458 | 458 | 1647 | 478 | 1168 | 1647 | 936 | 711 |
| C2 5-U       | NA  | 363 | 363 | 11292 | 8961 | 2331 | 11292 | 9324 | 1968 |
| C3 NAT-TH    | 318 | 374 | 691 | 1335 | 344 | 991 | 1335 | 1035 | 300 |
| C4 5-TH      | 116 | 319 | 434 | 9155 | 6857 | 2298 | 9155 | 7291 | 1864 |
| C5 20-TH     | 48  | 201 | 250 | 36683 | 34145 | 2538 | 36683 | 34395 | 2288 |
| C6 NAT-NAT-TH | 202 | 406 | 608 | 1450 | 392 | 1058 | 1450 | 1000 | 450 |
| C7 5-40-5-TTH | 68  | 337 | 406 | 9952 | 7637 | 2314 | 9952 | 8043 | 1909 |
| C8 20-40-20-TTH | 29  | 210 | 238 | 39870 | 37328 | 2542 | 39870 | 37566 | 2304 |

BOL – Beginning of life  
EOL – End of life  
DIF=BOL-EOL

### Table 4. Masses of fertile isotopes.

| Case         | $^{238}$U [g] | $^{232}$Th [g] | Fertile Material [g] |
|--------------|---------------|----------------|----------------------|
|              | BOL | EOL | DIF | BOL | EOL | DIF | BOL | EOL | DIF | BOL | EOL | DIF |
| C1 NAT       | 226930 | 224920 | 2010 | NA  | NA  | NA  | 226930 | 224920 | 2010 |
| C2 5-U       | 217190 | 216570 | 620  | NA  | NA  | NA  | 217190 | 216570 | 620  |
| C3 NAT-TH    | 184000 | 182150 | 1850 | 38510 | 37907 | 603  | 222510 | 220057 | 2453 |
| C4 5-TH      | 176100 | 175510 | 590  | 38510 | 38360 | 150  | 214610 | 213870 | 740  |
| C5 20-TH     | 148310 | 148030 | 280  | 38510 | 38452 | 58   | 186820 | 186482 | 338  |
| C6 NAT-TH    | 200000 | 198090 | 1910 | 24177 | 23799 | 378  | 224177 | 221889 | 2288 |
| C7 5-40-5-TH | 191410 | 190820 | 590  | 24178 | 24091 | 87   | 215588 | 214911 | 677  |
| C8 20-40-20-TH | 161190 | 160910 | 280  | 24198 | 24163 | 35   | 185388 | 185073 | 315  |

BOL – Beginning of life  
EOL – End of life  
DIF=BOL-EOL
Figure 3. Evolution of infinite neutron multiplication factor.

Figure 4. Evolution of $^{239}$Pu and $^{233}$U.
6. Conclusions
In the study we have presented neutronic analysis of the CANDU-6 reactor fuel channel with mixed thorium-uranium fuel. The designed numerical model in a good way describes neutronic effects, which occurs in the heavy-water moderated systems. The results were verified with the results obtained by other scientific groups and the general outcomes shows good consistency [13,14]. The study gives outlook on the neutronic characteristic of the fuel channel, especially:

- The fuel with large fertile material fraction shows better breeding capabilities of $^{239}$Pu and $^{233}$U.
- The best breeding capabilities were obtained for mixed natural uranium and thorium fuel (C3).
- The initial evolution of $K_{inf}$ depends on the fissile to fertile ratio in the initial fuel.

Additionally, the obtained evolutions of $K_{inf}$ as well as fissile and fertile isotopes show some hints for improvements towards effective utilisation of thorium-uranium fuels. The enhanced parametric study on fuel density, fraction of fissile and fertile material, geometry of fuel bundle and reactivity coefficients is recommended. Thus, the study cannot be treated as a final approach towards modelling of CANDU-type systems at the level of the fuel channel. Therefore some improvements may be outlined towards future research:

- modelling with reactivity control devices.
- modelling of fuel bundle on-power refuelling.
- modelling on full core level.

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