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Neutronic Analysis of an Incorporated Thorium-Uranium Breeder-Booster in CANDU-6

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Table of Contents

1.0	Abstract		.3
2.0	Introd	uction	.3
3.0	The Design		.4
4.0	Metho	odology	.5
5.0	Simula	ation of a Nominal Reactor with the Addition of Thorium Channels	.7
5.1	Sim	ulation without refuelling	.7
5	5.1.1	Reactivity vs. time	.7
5	5.1.2	Thorium Mass vs. time:	. 8
5	5.1.3	Uranium-233 mass vs. time:	.9
5.2	Sim	ulation with refuelling	.9
6.0	Conclu	usion	12
7.0	Refere	ences	12

1.0 Abstract

This paper describes a design to utilize potential thorium fuel cycle for the CANDU-6 reactor. The design consists of three additional clusters of channels placed vertically into the calandria and spanning the diameter of the circular face. The clusters will be placed along the axial plane: one at the centre; and the remainder spaced towards both ends of the reactor. The cluster that passes through the centre of the calandria will be filled with a thorium solution and will be the breeder. The other clusters on both sides of the calandria vessel will contain the bred uranium nitrate solution.

2.0 Introduction

Current nuclear reactors are fuelled with uranium dioxide (UO2). However, within the next fifty years, it is projected that the price of uranium will increase substantially, thus, alternative fuel cycles, which will extend our nuclear resources, are of significant interest. In particular, the use of thorium-232 (232 Th) to breed fissile uranium-233 (233 U) has long been proposed as one fuel cycle which could be exploited. According to the following reaction;

$$^{232}Th + {}^{1}n \rightarrow {}^{233}Th \xrightarrow{22 mins} {}^{233}Pa \xrightarrow{27 mins} {}^{233}U$$

A design to utilize the benefits of the thorium cycle has been explored. The design presented in this paper will describe how the thorium cycle can be utilized by the introduction of Thorium Channels into a CANDU-6. The purpose of this design is to enhance the lifetime of the fuel in the reactor. This can be visually represented with the following figure.



Figure 1. Reactivity of a 37-element fuel bundle in a CANDU-6 reactor.

During normal burn-up of a CANDU-6 reactor the average fuel is charge burn-up is given approximately by the burn-up value where the area under the curve represented by the blue shaded region is equal to the area above the curve represented by the red shaded region. This is due to the fact that the overall negative reactivity in the reactor must equal the overall positive reactivity to maintain criticality in the bulk reactor. Therefore an improvement to this curve would be to increase the irradiation of each region, thereby increasing the average discharge fuel burn-up. The measure of reactivity, masses of ²³²Th and ²³³U, and power contribution are used to determine the effects of the Thorium Channel on the system. The reactivity of the system is determined by the following equation;

$$\rho = 1 - \frac{1}{k} \tag{1}$$

This equation was used to determine the reactivity of the Thorium Channel on the system. ²³³U and ²³²Th follow two distinct mass transients over the time interval simulated in this project. ²³²Th shows an expected linear decline over time, as the absorption cross-section is not time-dependent. This relation can be written as;

232
Th $(t) = \int_{V} \left[^{232}$ Th $- \sigma_a \Phi(V, t)^{232}$ Th $(V, t) \right] dV$ (2)

²³³U is constantly being produced from neutron absorption of ²³²Th , and the subsequent decay of protactinium-233 (²³³Pa). As the ²³²Th is absorbing neutrons, the mass of ²³³U is increasing. However, not all of the ²³²Th is transmuted into ²³³U. There is a distinct probability that ²³²Th or ²³³Pa will absorb a neutron, or undergo a different event, before the decay occurs. This probability that ²³²Th or ²³³Pa will absorb an additional neutron is governed by the energy-dependent microscopic absorption cross sections for each respective nuclide and the total neutron flux of the region. The number of ²³³U atoms present at time *t* is then given by the equation;

²³³U(t) =
$$\int_{V} \left[{}^{233}\text{Pa}(t,V)e^{-\lambda t} - \left({}^{233}\text{U}(t,V)\sigma_{a}\Phi t + {}^{233}\text{U}(t,V)\sigma_{f}\Phi t \right) \right] dV$$
 (3)

Equation 3 assumes that the loss of ²³³U atoms to decay is negligible, since the half-life of ²³³U is 160 000 years. The relative power contribution of the Thorium Channel was used to determine impact of ²³³U produced on the total power. Power from the Channel would be created by ²³³U fission within. Since ²³³U is the only fissile isotope in the thorium channels, the relative power contribution from the Channel is given by the following formula;

$$P_{TC} = \frac{\left[\sum_{g=1}^{N} \left(\int_{V} \phi_{g} dV \times \sum_{f}^{g}\right)\right]_{TC}}{\left[\left[\sum_{g=1}^{N} \left(\int_{V} \phi_{g} dV \times \sum_{f}^{g}\right)\right]_{TC} + \left[\sum_{g=1}^{N} \left(\int_{V} \phi_{g} dV \times \sum_{f}^{g}\right)\right]_{CF}\right]},\tag{4}$$

Where TC and CF represent the Thorium channel and CANDU Channels respectively.

3.0 The Design

The design of this system involves modifying the CANDU-6 lattice to incorporate three additional clusters of channels placed vertically into the calandria and spanning the diameter of the circular face. The channels have an inner diameter of 3.5 cm and an outer diameter of 3.75 cm. The center channel is filled with thorium nitrate with a chemical composition of $(Th(NO_3)_4 \cdot 4H_2O)$. This solution is burned during operation of the reactor.

²³³U is a fissile isotope of uranium. Once the channel has produced a significant quantity of ²³³U, the solution will be extracted and purified using and external system of processes. The new solution will be placed back into the reactor within the channels at either end of the reactor. This solution, containing a large portion of ²³³U will be used to increase the neutron population of the reactor at the ends of the core to increase the discharge fuel burnup. A visual representation of this design is shown in Figure 2.



Figure 2. Visual representation of the addition of the Thorium Channel design to the CANDU-6 calandria vessel.

4.0 Methodology

To determine the effects of the design on a CANDU-6 reactor, the multigroup flux solving code, DRAGON was used to solve the neutron transport equation for the system. DRAGON uses an infinite lattice cell to determine the solution to the neutron transport equation. Therefore a three-dimensional cell was created to perform the simulation analysis on the system.

To keep the simulation as consistent as possible with the actual design, a thorium channel with a length of 28.575 cm was placed between two fuel channels. Each fuel channel contained four fuel bundles of length 50 cm. This geometry is presented in Figure 3 below and is the infinite lattice for which the results have been obtained so far. This supercell was reflected at the outer boundary surfaces in each direction for an infinite lattice.



Figure 3. Supercell geometry used as the input for DRAGON calculations (All dimensions are in mm).

The Thorium Channel was described earlier as a channel interstitial to the two fuel channels. The fuel channels are represented within the simulation as the fuel channel, the annulus gas, and the pressure tube. Within the pressure tube is a fuel bundle with a length of 50 cm. The bundle, for the purposes of this simulation was approximated by a fuel bundle consisting of one center pin and three annular rings. The pin is an exact replica of the center pin in a 37-element fuel bundle. Each ring contains the same mass of uranium that would be present in the pins within the centerline pin ring radius. The pin and rings are also cladded using the same mass of zircaloy as present in each respective ring. The dimensions of this approximation are presented in Figure 4.

There are some operational considerations that should be noted. For instance, the temperature of the Thorium Channel will operate at a nominal 60°C (333.15 K), the average temperature of the moderator in a CANDU-6. The moderator is also assumed to be the heat sink for the design.



Figure 4. Annular fuel representation used for DRAGON calculations.

5.0 Simulation of a Nominal Reactor with the Addition of Thorium Channels

A new set of simulations was required to increase the accuracy of the results and to provide data over a longer range of time. Changes to the initial simulation included adjusting the fuel contents of the CNADU bundles being burned. The simulation included bundles that have been burned in a lattice without the presence of the Thorium channels to a point where the infinite lattice returned a k-infinity of ~1.048. This would allow the Thorium channels to respond to the presence of burned fuel which included the effects of plutonium build-up and xenon poisoning.

5.1 Simulation without refuelling

Thorium channels were implemented vertically every 4 bundles of fuel, the first simulation were set to let the fuel burn without refuelling to illustrate how thorium and ²³³U would affect the reactivity in the fuel channels studied. The simulation was executed for a simulation time of 1200 days. The following graphs were tabulated from the results given in the output of the code.

5.1.1 Reactivity vs. time

The reactivity started declining immediately as expected due to uranium-235 (²³⁵U) burn up, and then plutonium peak was formed which made an increase in the reactivity, before it declines again to reach criticality after about 310 days. The simulation becomes subcritical after 310 days. Reactivity declines in almost a straight line behaviour after about 1000 days. However the values might be different but the reactivity curve has the same shape as if the thorium channels are not present.



Figure 5. reactivity vs. time for no refueling simulation

5.1.2 Thorium Mass vs. time:

Masses of 232 Th and 233 U formed are plotted against time to show the formation of 233 U as thorium declines in the channels.

Thorium mass vs. time:

As shown in the graph, thorium mass declines linearly with time. At time zero, thorium mass was about 363 grams, after 1200 days thorium mass is about 308 grams, which is more than 84% of the original mass. This implies that thorium mass contributed to breeding and decaying in a period of 1200 days is only about 15% of the total mass present.



Figure 6. thorium mass vs. time in non refueling simulation

5.1.3 Uranium-233 mass vs. time:

This graph demonstrates the mass build up for ²³³U in the thorium channels. The most amount of mass occurred after 570 days, only about 4.7 grams of ²³³U were bred. After the peak, ²³³U uranium-233 burn up increases and the burn up rate become greater than the breeding rate. The amount of ²³³U uranium-233 bred continue to decline with time, this indicates that the maximum amount of uranium can ever be achieved in this simulation is 4.7 grams per channel when 26 grams of thorium have been burned.

The first variable that was studied was the affect of the Thorium Channel on k-infinity of the infinite lattice. Results of the simulation are shown in the Figure 5 below.



Figure 7. uranium-233 mass vs. time with no refueling simulation

5.2 Simulation with Constant CANDU Fuel Burnup

This simulation is similar to the previous one except refuelling is simulated to maintain criticality under normal reactor conditions, negative reactivity inserted by xenon build up, leakage and reactivity devices are all counted for; negative reactivity insertion of about 80 mk is considered.

It is apparent from the graph that the thorium has a significant effect on the reactivity of the system. At first there is a substantial drop of reactivity from nominal. The reactivity then increases as ²³³U is bred in the Thorium Channels. Another important aspect to notice is that the reactivity of the system never reaches what a nominal reactor would. This is a trade-off effect of burning the thorium to produce uranium. To adjust for this and make the reactivity of the reactor critical, control rods will need to be removed slightly, thereby increasing the reactivity of the system back to a critical state.

The first variable that was studied was the affect of the Thorium Channel on k-infinity of the infinite lattice, the change of k-infinity caused by the thorium channels was calculated by subtracting the CANDU k-infinity from the total k-infinity of the simulation. Results are shown in the Figure 8 below;



From the graph, it is apparent that the reactivity changes due to the thorium channel reach peak reactivity around 600 days. This is where the solution would be removed from the system and the uranium-233 is extracted from solution to be utilized in the ends of the reactor. The exact changes in the mass of thorium and uranium can be found in the graphs below;



Figure 9. Change in thorium-232 mass over time.



Figure 10. Change in uranium-233 mass over time.

It is apparent from these graphs that the thorium is constantly being depleted via absorption of a neutron within the reactor. Once this transaction occurs, some of it eventually decays into 233 U. The breeding characteristics of the 233 U in the system are shown in Figure 10.

The power contribution of the Thorium Channel is presented in Figure 11 below. After about 1200 days the power contribution is about 5.5 %, the power contribution is almost a maximum at this value, as it appears in the graph. This graph shows a similar shape to both the reactivity and ²³³U mass graphs presented earlier. However it should be noted that the power contribution does not peak and drop off at the same simulation time as the previous graphs have done. This is due to a quasi-equilibrium attained between the ²³³U production and burnout rates. The power distribution remains around 5.5% during the quasi-equilibrium.



6.0 Conclusion

After the analysis of the results obtained during the simulations, it is apparent that the incorporation of the Thorium Channel adversely affects the initial behaviour within the infinite reactor. Through the variation of the amount of thorium nitrate in the system, it was shown that the TC can also be utilized as an additional neutron source after breeding uranium-233. The net effect of the thorium channels is insertion of negative reactivity due to the difference between the thorium mass and uranium-233 mass in the system at any present time. Based on the analysis of the simulations, the positive effects of uranium-233 on the reactor can be utilized and improved upon. Next steps taken for this project include finding the optimal fuelling method to maximize the net positive effects of uranium-233 analysing the effect of uranium nitrate solution on an infinite reactor lattice and to find a suitable method to extract uranium-233 from the system.

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