Assessments of design and operational parameter sensitivity towards plutonium production in heavy water reactors

Cecilia Gustavsson, Erik Branger, Stina Fredriksson, Axel Hallander, Isak Hedberg and Vaibhav Mishra

Department of Physics and Astronomy, Uppsala university

Abstract

A majority of the nuclear reactors in the world use low-enriched uranium as fuel, and will produce plutonium during operation. This will happen when neutrons undergo capture in ^{238}U instead of causing fission of ^{235}U, which is a likely reaction as low-enriched uranium is composed of > 95% ^{238}U. While the plutonium can be used as fuel in the reactor, it is also a material highly desired by states producing nuclear weapons. Not all reactors produce plutonium of the same grade, which significantly impacts its usability in a nuclear weapon. For this reason, certain reactor technologies have been favored for military plutonium production. Heavy water moderated reactors is one such family of reactors that has been used in current or now defunct nuclear weapons programmes by states such as India, Pakistan, Sweden, Switzerland and the United States.

A number of factors impact the rate and grade of plutonium production in a reactor. These include (but are not limited to) fuel design specifications (pellet radius, fuel density), operational temperature of the fuel as well as the coolant and moderator, and numerous other operational parameters such as specific power, cycle lengths and downtimes et cetera. The present study will look into investigating the relative sensitivity of plutonium production rate and grade towards these design and operational parameters. Based on the results from this evaluation, it is expected that we can better understand which parameters impact plutonium production quantity and quality the most. This will also help us understand the role and impact of uncertainties in these parameters and connect them to the plutonium content in the spent fuel produced by these reactors.

The simulation work in this paper has been carried out in the form of a supervised bachelor thesis work by students studying the Engineering Physics programme.

Introduction and background

Nuclear weapons require a critical mass of a fissile material; the most commonly used such being ^{235}U and ^{239}Pu. While uranium can be mined on Earth, plutonium does not exist in nature except as traces, and must hence be produced from uranium. This can be done in a nuclear reactor if uranium undergoes neutron capture and eventually builds up plutonium isotopes. The process is complex, and competes with uranium fission, which is necessary for reactor operation, since it is the process that provides the neutrons crucial for the capture itself.

Natural uranium consists of the isotopes ^{238}U (99.3 %), ^{235}U (0.7 %) and ^{234}U (0,005 %). Most power reactors use uranium enriched to 2-5 % in ^{235}U, and if a reactor is moderated by light water (as most commercial reactors are), enrichment is necessary for achieving criticality. Heavy water moderated reactors can run on natural uranium or a fuel that is slightly enriched; the latter making the time between fuel changes longer as well as reducing the fuel costs [1].

The objective of weapon-producing reactors is, however, not to save fuel or to provide electricity, but to produce plutonium of weapons-grade. There are several definitions of what exactly is meant by weapons-grade, but in principle, the plutonium vector should consist of at least 93% of the isotope ^{239}Pu and less than 7% of the isotope ^{240}Pu. For a LEU fuelled reactor, where ^{238}U is abundant, the fissile isotope ^{239}Pu is produced via the reaction:

$$^{238}U + n \rightarrow {}^{239}U \rightarrow {}^{239}Np \rightarrow {}^{239}Pu$$

Furthermore, the output of ^{239}Pu is not stable in time; if left in the neutron flux of the reactor, two things can happen to the ^{239}Pu. Firstly, it can capture more neutrons and build up heavier plutonium isotopes, (^{240}Pu, ^{241}Pu, ^{242}Pu, etc) which degrades the weapons quality of the plutonium. Secondly, the ^{239}Pu nuclei can fission in the reactor which consumes it. Consequently, military reactors operate on significantly shorter cycles as compared to civilian ones, as the plutonium quality is the limiting factor.

Independent assessments of a country's nuclear weapons stockpile is frequently made by analyzing the nuclear infrastructure available, as such large-scale infrastructure is typically difficult to keep hidden. However, details about the infrastructure obtained through estimates and analyses have large uncertainties, which will affect any estimates of material throughputs of the individual facilities. In the case of military reactors, optimized for plutonium production, design choices and operating parameters are unknown, creating difficulties in assessing the amounts and quality of plutonium produced.

Multiple factors such as reactor power, final burnup (i.e. time the fuel spends in the core) and enrichment will influence the resulting plutonium composition. The geometry of the reactor fuel also plays a role in plutonium production. Moderated neutrons produce fission and power, but during the moderating process, the probability for neutron capture is largest, due to the cross section resonances in ^{238}U. This leads to differences in the burnup at different radii of a fuel pin and an overall dependence on fuel geometry. Temperatures in the fuel and coolant as well as moderator will also play a role in the interaction between neutrons and target nuclei affecting the isotopic composition of the spent fuel.

Taking this background into account, it becomes evident that the plutonium output and quality are dependent on several factors and are the results of complex neutron physics and interactions. To investigate what impact these parameters have on the resulting plutonium composition and amounts, simulations using Monte Carlo methods are employed, using the reactor simulation code Serpent2 [2]. In this work, we are using Serpent2 to study heavy water reactors and the effect of operational as well as design parameters on the resulting plutonium output. The aim is to identify parameters that significantly affect the plutonium composition, adding uncertainty to production estimates, and also parameters that do not affect the production, hence knowledge about such parameters are less important for accurate estimates.

Earlier similar studies have been made for the civilian BR3 research reactor (a pressurized light water reactor or PWR) and a civilian sodium-cooled fast reactor (SFR) [3, 4]. In this work we are focusing on three different designs of pressurized heavy water reactors which are described in more detail in the sections below.

Heavy water reactors in this work

As mentioned in the introduction, heavy water reactors use heavy water as moderator. Due to the excellent moderation capabilities of heavy water, such reactors can use natural uranium as reactor fuel. In this work, we are focusing on three different heavy water reactor types which were chosen to represent a variety of fuel designs and operating characteristics, for both historic and current reactors. In the end of this section is a summary of factors where the reactor types differ, see Table 1.

The NRX reactor

NRX stands for National Research Experimental and was a Canadian research reactor and prototype to the later CANDU design. It was situated at Chalk River Laboratories in Canada, and moderated by heavy water but cooled by light water. It used natural metal uranium as fuel and was for a time the most powerful research reactor in the world. NRX was in operation between 1947 to 1993.

The NRX reactor was a research reactor intended for studying plutonium production but also for other types of reactor research [5]. Therefore, many different fuels were loaded in the core during its lifetime, including booster rods made of either enriched uranium or plutonium. From 1958, the metallic uranium rods were exchanged for UO_2 rods. In this paper, we are however only considering the NRX core with natural metallic uranium fuel.

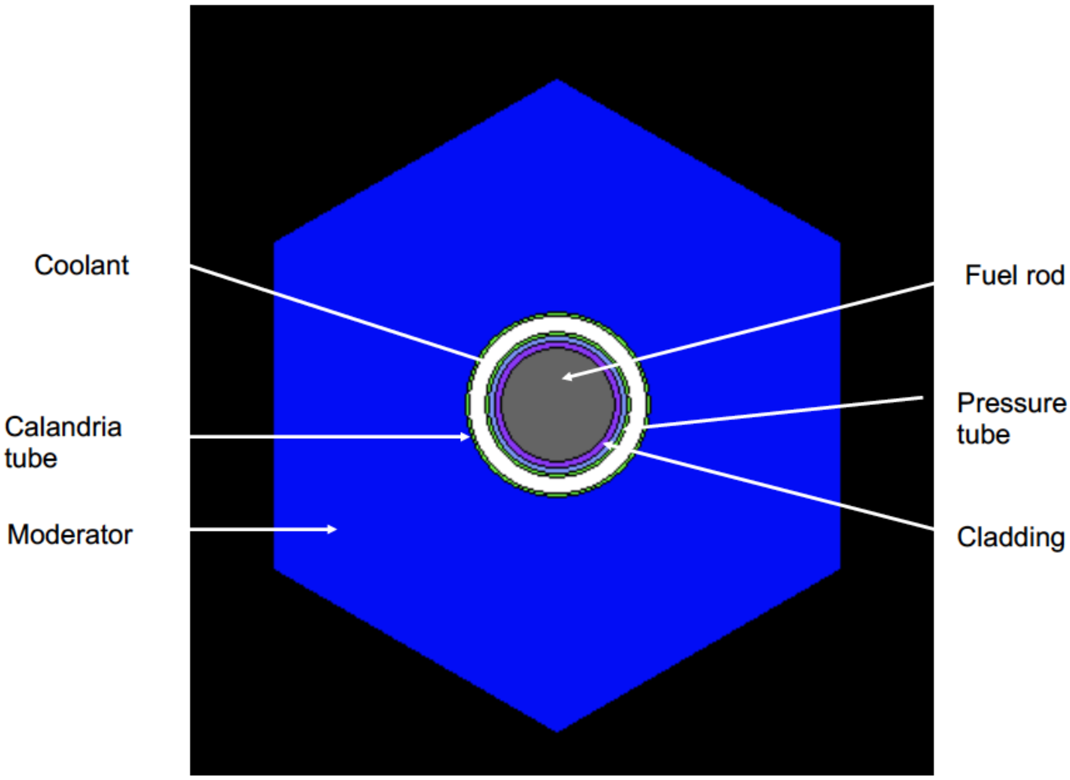

Figure 1: The NRX fuel element. Elements are placed in a hexagonal lattice.

The CANDU reactor

CANDU stands for Canada Deuterium Uranium and is a whole class of commercial heavy water moderated power producing reactors that have been developed by Atomic Energy of Canada Limited (AECL). Different designs exist and have existed, some CANDU types have been cooled by light water, others by heavy water, but the moderator has always been heavy water. Also the fuel design has varied greatly, from 7 pins in a 2-ringed circular arrangement up to 37 pins in a 4-ringed arrangement. With time, the assemblies have become both longer and of greater diameter, while the pins have seen a decrease in radius [6].

The first CANDU reactor in operation was the NPD - the nuclear power demonstration reactor in 1962 having an electric power of 25 MW. In this work, we are simulating a slightly later design; the Douglas Point reactor starting in 1966 with 210 MWe power. This particular reactor belongs to the class CANDU2 and has been the model for the Pakistani KANUPP reactor.

From the beginning, the CANDU fuel was made of natural UO_2 pellets, and the need for frequent fuel changes was solved by online refuelling. Still, many operating CANDU reactors run on natural uranium, but some are also using slightly enriched uranium fuel (SEU). The enrichment is usually between 0.9% and 1.2% and the benefits are lower fuel costs and the possibility of higher discharge burnup and less frequent fuel changes [1].

In this work we are considering a CANDU2-type of reactor with a 19-pin assembly and heavy water as both moderator and coolant. We have simulated the power to match that of KANUPP, i.e. 337 MW thermal.

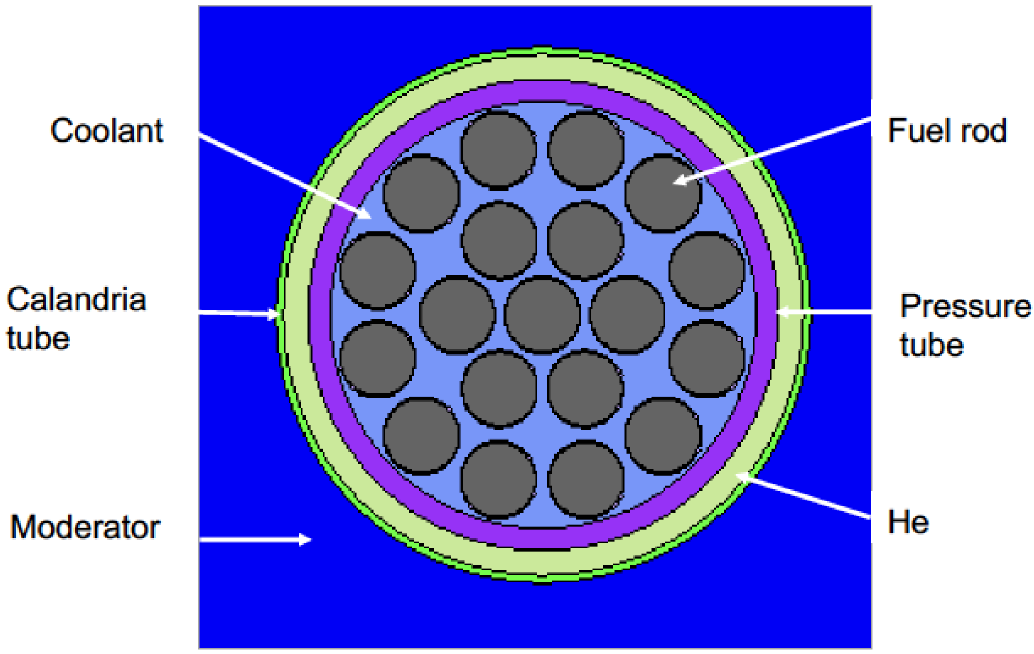

Figure 2: The CANDU assembly. The assemblies are placed in a square lattice.

The Ågesta reactor

The Ågesta reactor, a heavy water moderated and heavy water cooled power reactor, was the first commercial nuclear power plant in Sweden, in operation between 1963 and 1974 [7]. Its objective within "the Swedish Line" [8] was to generate district heating and power to the surrounding communities, but the reactor was also intended to play a role in the Swedish nuclear weapons programme, which was not canceled until 1972.

The Ågesta nuclear power plant operated with three different successive cores; core I, core II and core III [9]. The two first cores were very similar to early CANDU designs [6] with a circular arrangement of 19 fuel pins consisting of natural UO_2 pellets sheathed with zirconium. Ågesta had no pressure tubes for the coolant, but was of pressure vessel type. After severe fuel failures with the two first cores, a completely different fuel design was tried for core III - the core simulated in this work. Core III was a success, and during its years of operation (1970-1973), no serious incidents occurred, and the fuel had achieved the expected discharge burnup at the final shut-down. The fuel design developed for core III came from ASEA-ATOM who was at this time developing fuel for the coming Swedish light-water reactors. As basis for core III, the new square 8*8 assembly was chosen. These were however too large for the load openings in the Ågesta reactor, so a variant of the 8*8 fuel was developed; basically, the same fuel but with 5*5 pins in a square arrangement. In total, 96 such assemblies were loaded in core III. This meant in practice less uranium than cores I and II had, so to counter this, approximately half of the assemblies were enriched to 1.35%.

In this work we are simulating the 5*5 assembly developed for core III.

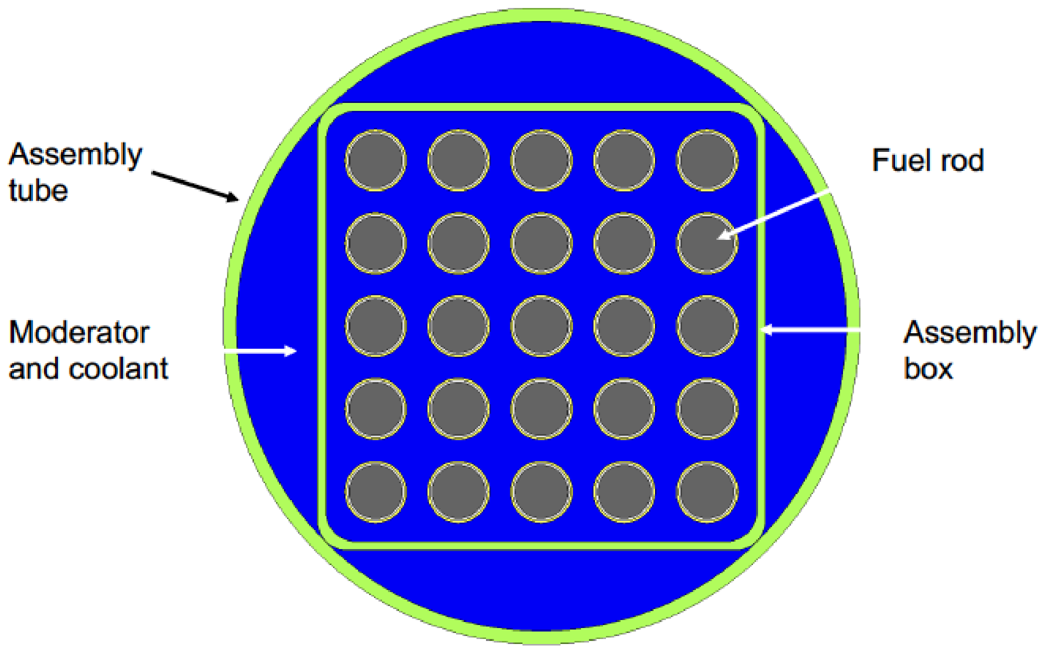

Figure 3: The geometry of the Ågesta III assembly. The assemblies are placed in a square lattice.

Table 1: Comparison of the three reactor types

	NRX	**CANDU2**	**Ågesta**
Start of operation	1947	1966	1970
Reactor type	pressure tubes	pressure tubes	pressure vessel
Moderator	D_2O	D_2O	D_2O
Coolant	H_2O	D_2O	D_2O
Fuel	metallic natural U	natural UO_2	natural UO_2
Assembly	1 pin	19 pins circular	5*5 pins square
Thermal reactor power (MW)	40	337	80

Simulations

The Monte Carlo particle transport code, Serpent2 [2] has been used to carry out the simulations in the present work. This code allows the user to specify reactor geometry and materials present before running the code. The set power history simulates the physical reactions that take place in a true reactor. The simulations were used to perform depletion calculation i.e. the code calculates the burnup of uranium which results in production of e.g plutonium. The output files will contain the isotopic composition of the irradiated fuel, and the various plutonium isotopes as well as their relative abundances. In this work we have set up an infinite 2D simulation meaning that only one assembly is modelled and subsequently mirrored as an infinite core by using periodic boundary conditions. In order to make sure the simulations are only run as long as the reactors can remain physically critical, criticality limits were calculated for the three cases. These limits take into account the simulated infinite cores and compensate for the inherent no-leakage of neutrons. The criticality limits were based on a reported such limit for CANDU reactors of $k = 1.045$ [10], and for the NRX and Ågesta criticality limits were calculated using the relative sizes of the cores as well as the known discharge burnup. Resulting criticality limits were 1.086 (NRX), 1.045 (CANDU2) and 1.084 (Ågesta).

Investigated parameters

As mentioned previously, the present study focuses on the impact of variation in reactor parameters (both design and operational parameters) on the quantity and grade of plutonium production in the spent fuel. The list of different parameters considered in the study are shown in Table 2, where the nominal values of each parameter is also shown.

Table 2: List of design and operational parameters and their nominal values in this study.

Parameters	**NRX**	**CANDU**	**Ågesta**
Fuel temperature (K)	473	683	873.5
Fuel density (g/cm^3)	19.1	10.47	10.47
Fuel rod thickness (cm)	1.73	0.71185	0.5235
Moderator temperature (K)	305	311	486
Moderator density (g/cm^3)	1.083	1.083	1.083
Coolant temperature (K)	340	537.5	486
Coolant density (g/cm^3)	1	0.863	1.083
Rod pitch (cm)	-	1.66	1.63
Initial enrichment (%)	0.7	0.7	0.7
Power density (MW/kg)	0.018	0.011	0.01653

Each of the parameters listed in Table 2 were subjected to a variation of ±10 % from its nominal value in steps of 2.5 % to assess its impact on the amount of plutonium and its vector composition at end of cycle (EOC). For some parameters though, this variation was not enough to see any change, so they were altered more, but within plausible limits. Notably, the initial enrichments studied were in the range of 0.5 % - 1.4 % which at the higher limit corresponds to a variation of 200%. Extra simulations runs with up to 200% of the nominal values were also studied for fuel temperature, coolant temperature and coolant density.

In the simulations, the Serpent input file deck representing the baseline or the nominal cases were prepared for each of the three reactor fuel types shown in Figures 1-3, and were suitably modified following the One At a Time (OAT) sensitivity analysis methodology [11].

The nominal and modified input decks were thereafter run using Serpent2 on a scientific linux-based computing cluster. Lastly, the sensitivity of the plutonium production and its grade was quantified for the three fuel types and documented using the Pearson's correlation coefficient [12].

Results

The sensitivity analysis for all three reactor types showed that the quantities of plutonium production and its quality were more sensitive to certain parameters than others. As described in the previous section, the relative sensitivity of the plutonium mass and quality are shown as Pearson's correlation coefficient in Figures 4-6 for the NRX reactor, CANDU reactor, and the Ågesta reactor respectively.

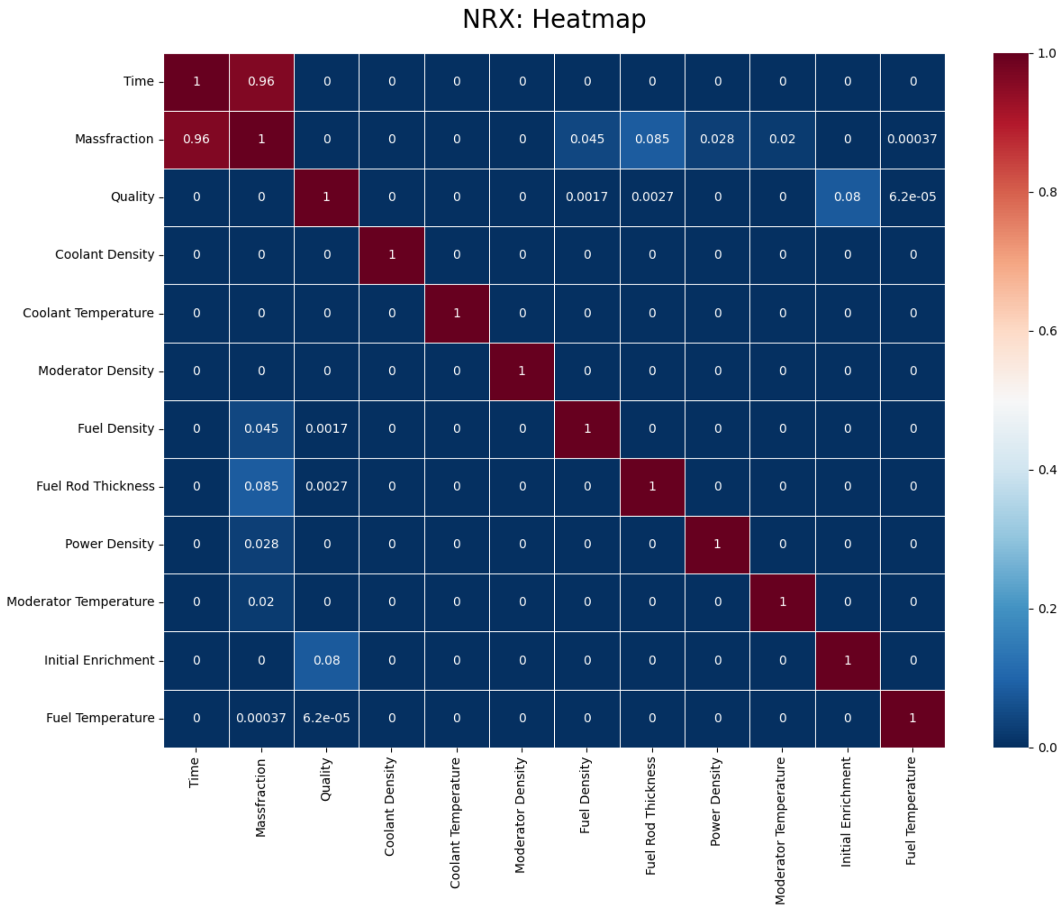

Figure 4: Heatmap between Pu mass and quality and reactor parameters for NRX reactor.

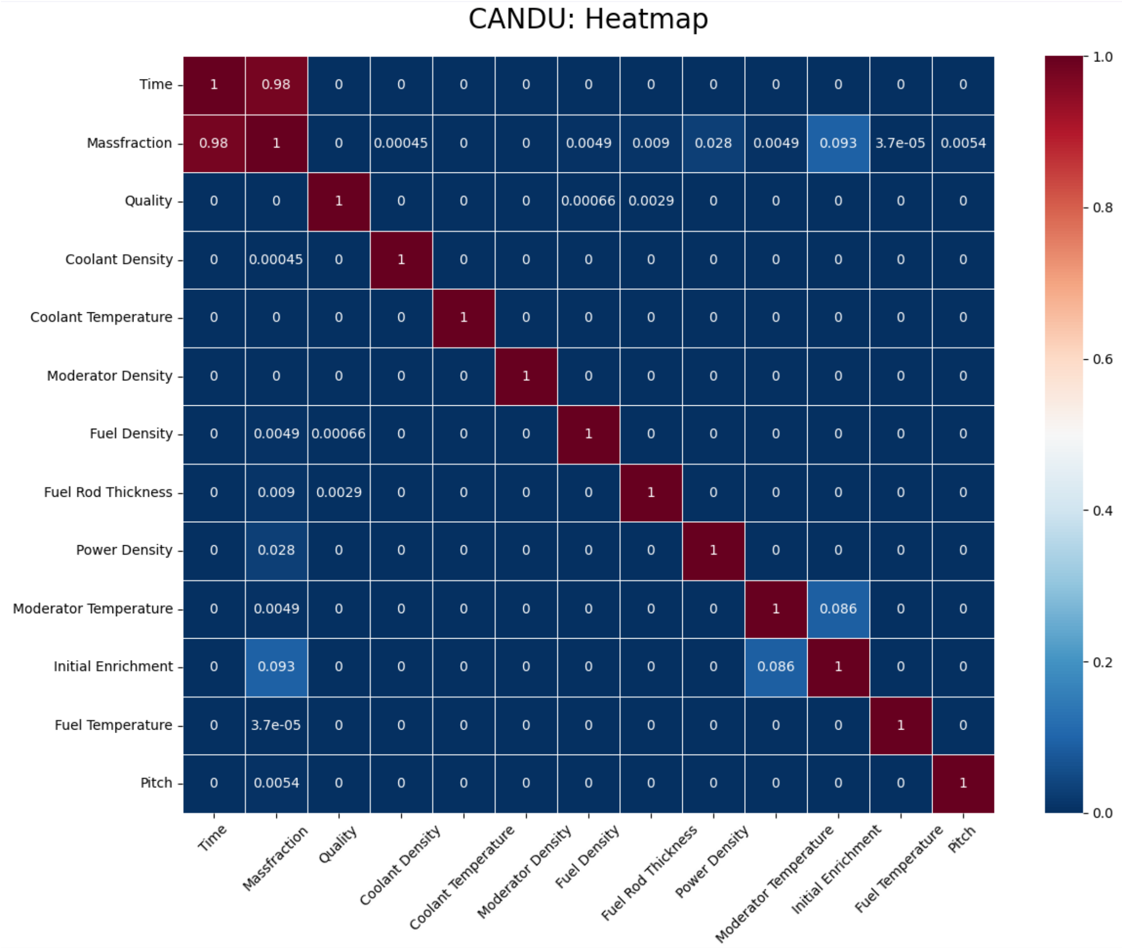

Figure 5: Heatmap between Pu mass and quality and reactor parameters for CANDU reactor.

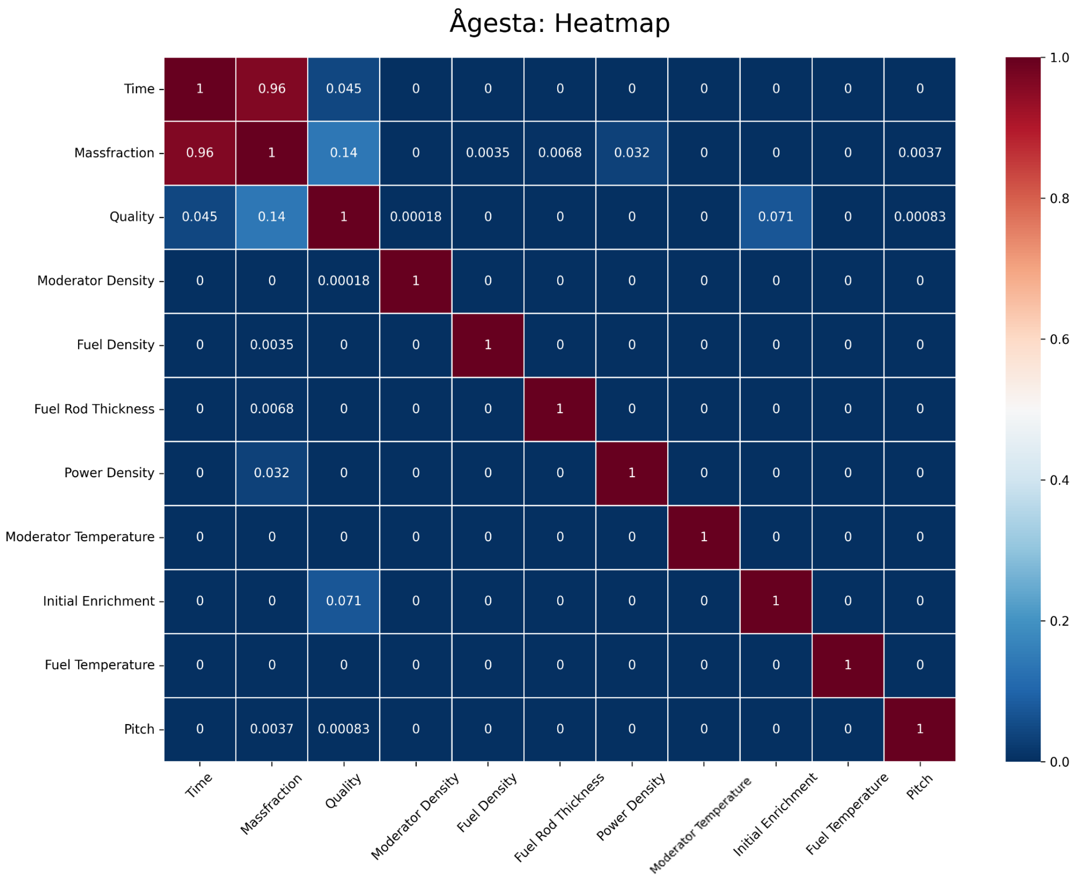

Figure 6: Heatmap between Pu mass and quality and reactor parameters for Ågesta reactor.

From the results in Figures 4-6, it may be deduced that for all reactor types, most of the parameters assessed have a minor impact on the quantity and quality of plutonium produced at the EOC. However, the total Pu mass produced had the strongest dependence on the 'time' parameter which denotes the time spent in the reactor core and is analogous to fuel burnup. The other design and operational parameters were found to be only weakly correlated to the plutonium production in the fuel. For instance, after time-spent-in-reactor, the correlation of plutonium mass at EOC showed minimal correlation to parameters such as fuel properties (such as enrichment, density, fuel rod thickness, fuel pitch), moderator properties (such as temperature and density) and coolant properties (such as temperature and density).

The absolute sensitivities (as a percentage difference from the nominal or baseline case) of mass of plutonium produced in each of the reactor cores towards each one of the design and operational parameters has also been depicted in Figures 7-9.

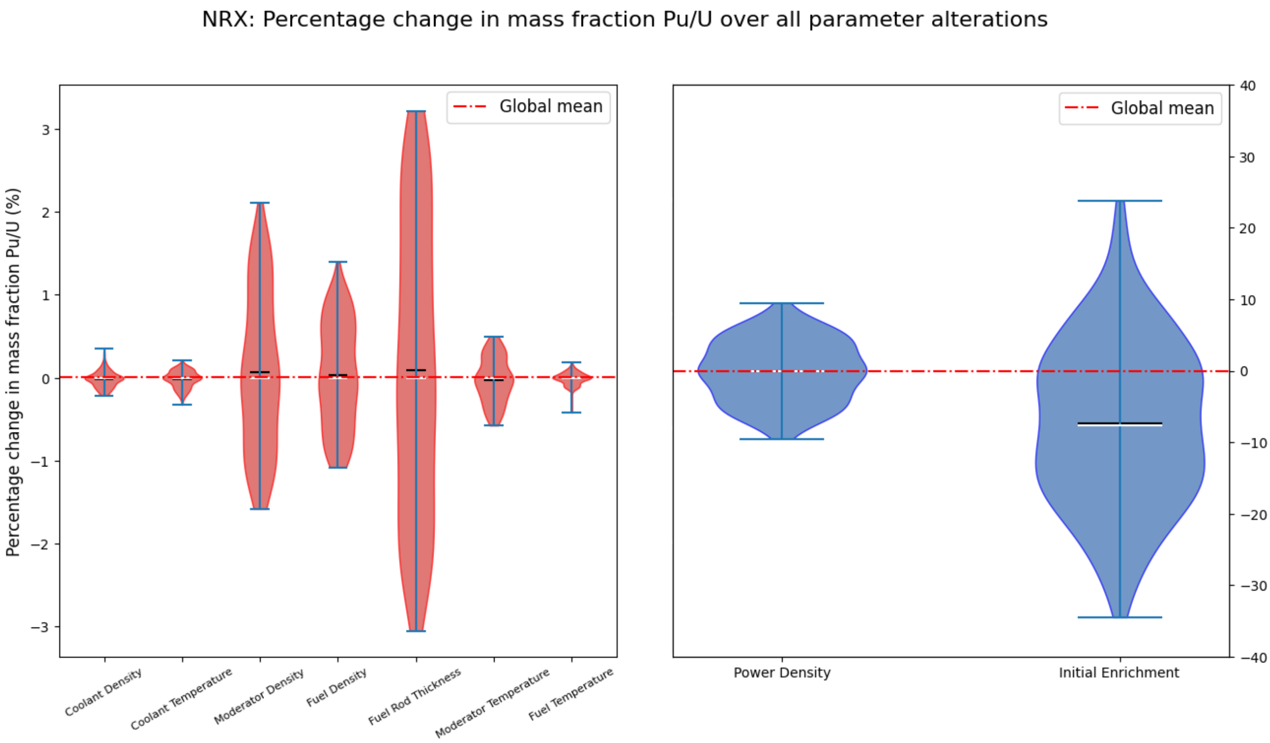

Figure 7: Violin plots showing Pu mass sensitivity towards reactor parameters for NRX reactor.

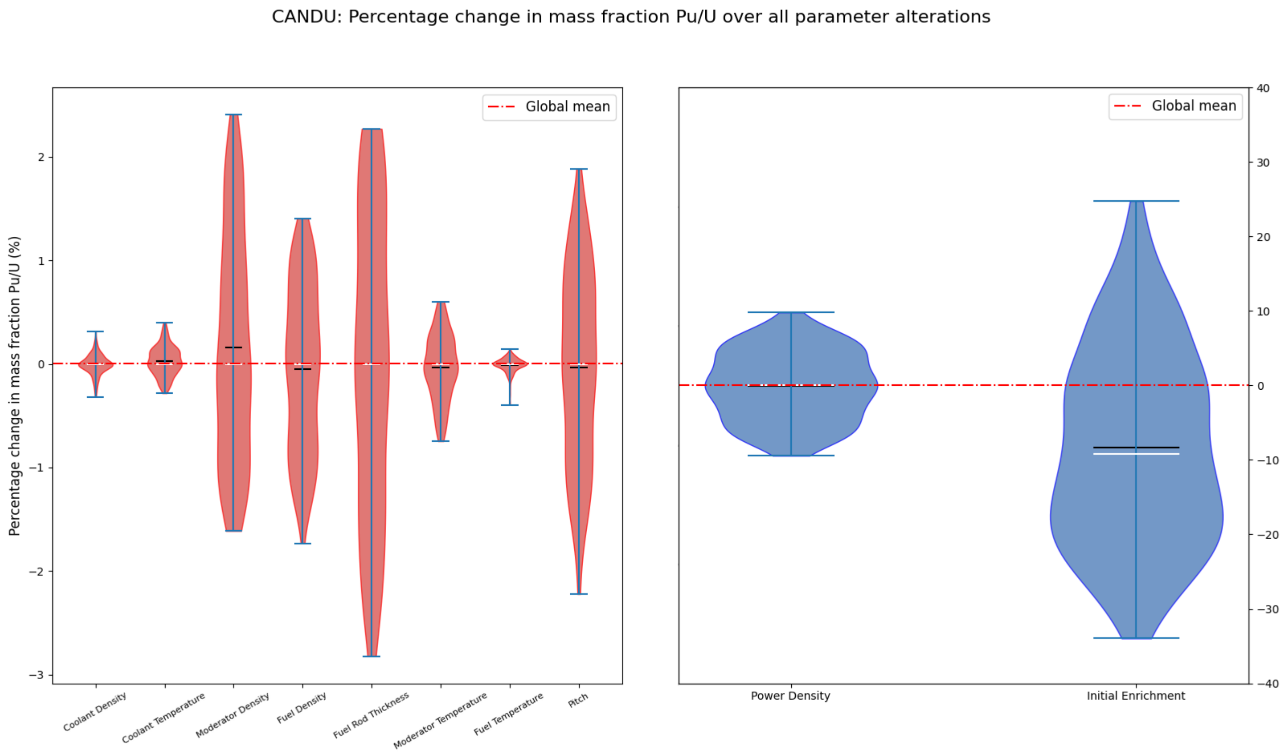

Figure 8: Violin plots showing Pu mass sensitivity towards reactor parameters for CANDU reactor.

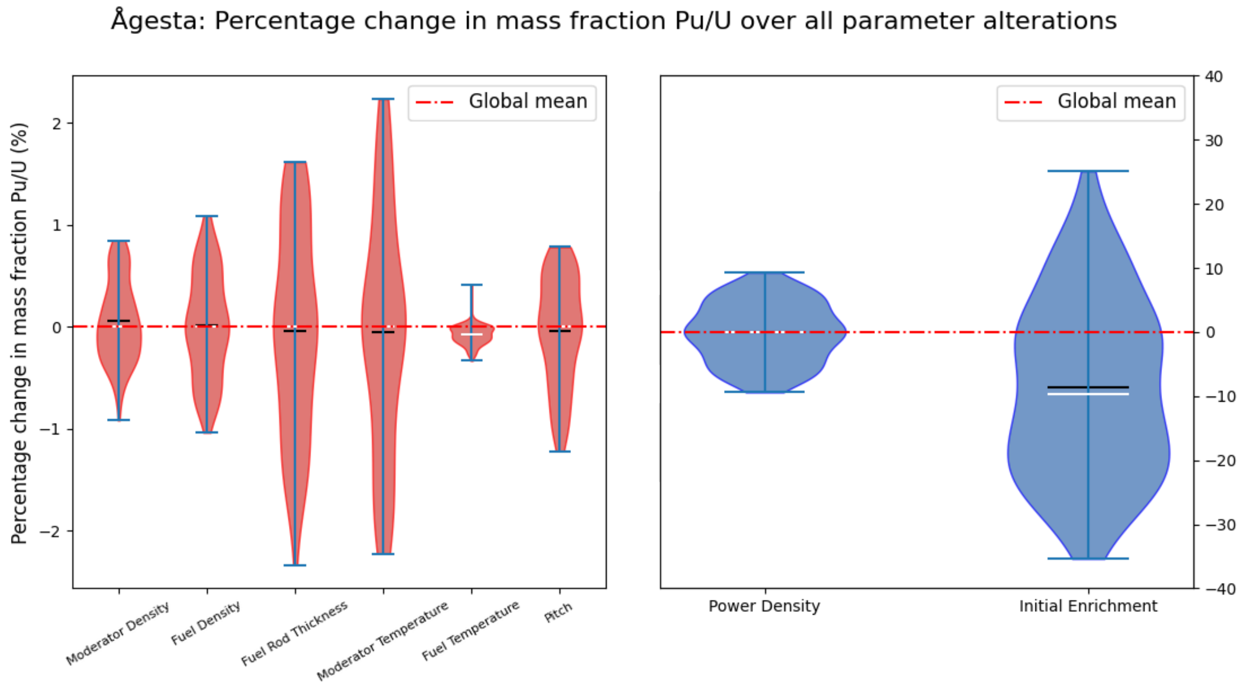

Figure 9: Violin plots showing Pu mass sensitivity towards reactor parameters for Ågesta reactor.

Figures 7-9 show that while overall trends in sensitivity of plutonium production towards reactor parameters are similar across reactor types, Ågesta and CANDU cores exhibit a marginally higher dependence on parameters like moderator density and fuel parameters such as rod thickness, density and pitch. All three cores show higher sensitivity towards power density and initial enrichment where the average global mean difference in plutonium production with respect to the nominal case was above 10% and up to 70% (in case of initial enrichment parameter).

Figures 10-12 show variation in plutonium production in the NRX, CANDU, and Ågesta cores respectively as a function of initial enrichment and time spent in core (or fuel burnup).

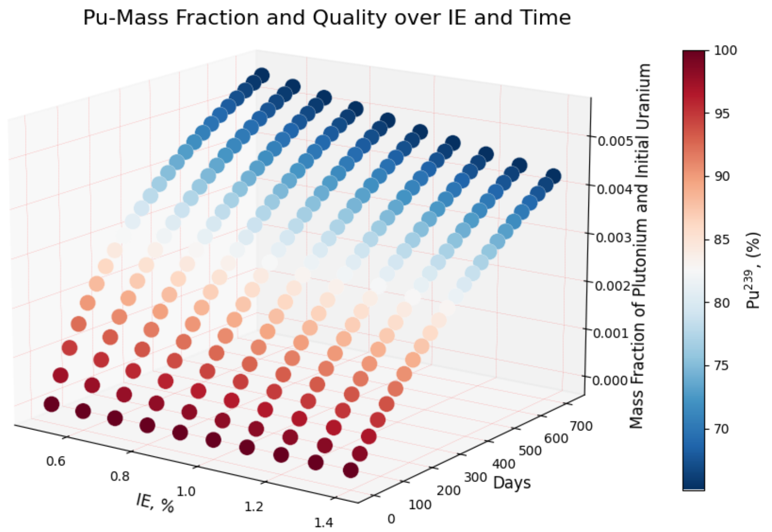

Figure 10: Plutonium build up and quality as a function of initial enrichment and time for NRX reactor.

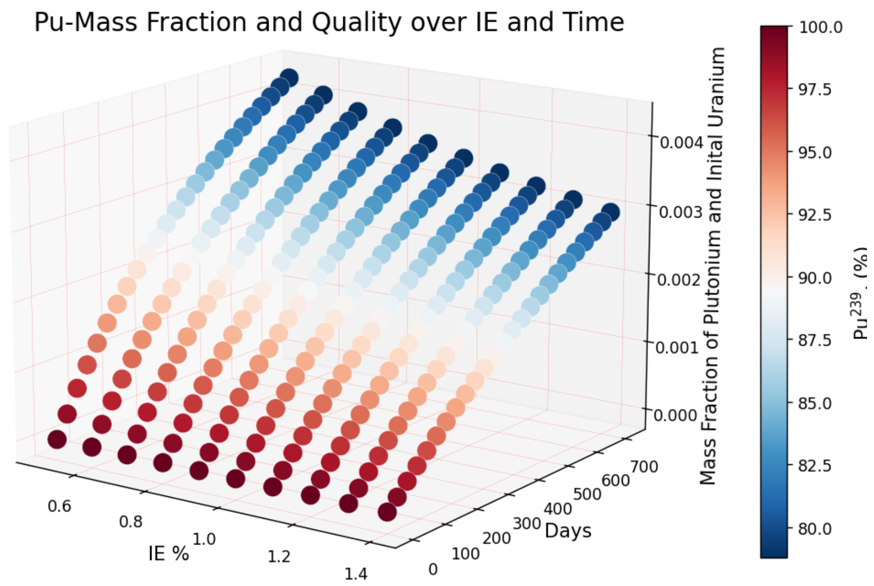

Figure 11: Plutonium build up and quality as a function of initial enrichment and time for CANDU reactor.

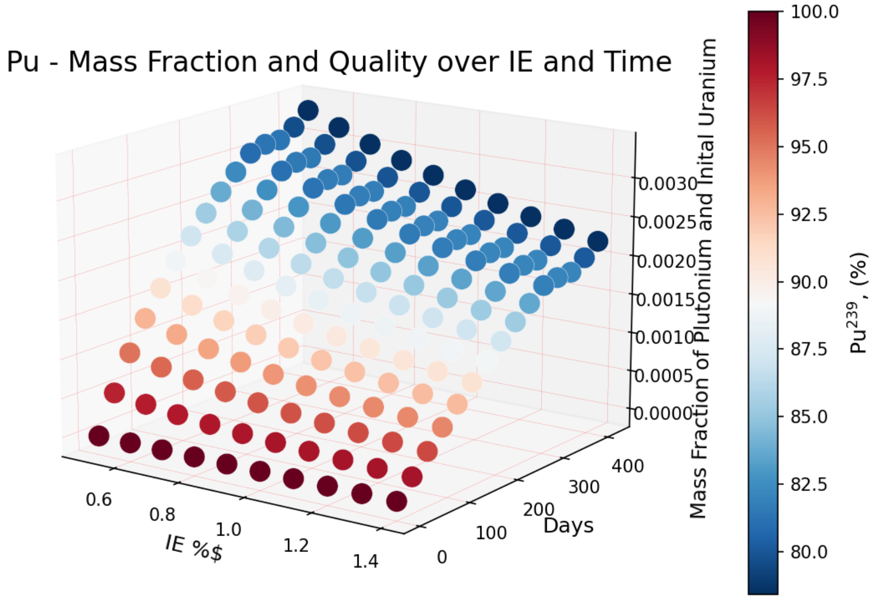

Figure 12: Plutonium build up and quality as a function of initial enrichment and time for Ågesta reactor.

One may infer from Figures 10-12 that lower initial enrichment levels favour production of more plutonium in all three reactor types. However, CANDU and Ågesta were found to be slightly more sensitive to the change (owing to the slope of the plot surface). Higher enrichment levels were found to favour the production of more ^{239}Pu wt. % but this effect was observed to be only marginal in nature.

Discussion and analysis

The present study has looked into quantifying the impact of variation in reactor design and operational parameters on its ability to produce plutonium. The results show that most parameters have very limited impact on the plutonium production, at least for the parameter ranges that were simulated here. The two most influential parameters are enrichment and power density, but for the CANDU reactor also the pitch (the distance between fuel rods) was influential. The more enriched the fuel is, the higher the burnup will be and the lower the plutonium content. Higher power density will result in more fission neutrons and more plutonium production. The pin pitch was changed independently while keeping the assembly pitch constant, meaning that when the distance between the fuel rods was increased, the distance between assemblies was decreased and the fuel more evenly distributed in the core. The net result for the CANDU reactor was that less plutonium was produced at larger pitches, i.e. when the rods were more evenly moderated.

Another parameter that was not explicitly studied, but does play a decisive role when it comes to plutonium quality is burnup or time in the reactor. The simulations showed that weapons-grade plutonium is achieved after 30-60 days of irradiation, but after that the plutonium will have too high content of heavier isotopes to be useful for nuclear weapons.

Even though this work has shown that plutonium production is rather insensitive to variations in the design and operational parameters investigated, it cannot be excluded that these parameters play a role. The chosen parameter ranges were quite small, some parameters clearly needed to be varied more to have an effect, and more importantly; when varying one parameter at the time, far from the whole possible parameter space is covered. We expect this work to be the foundation for further studies of plutonium production sensitivity.

Acknowledgement

We would like to acknowledge the Alva Myrdal Centre for Nuclear Disarmament at Uppsala University for supporting this work.

References

[1] Boczar P.G., *CANDU Fuel-Cycle Vision*, AECL Report, AECL-11937, 1998 May, https://www.osti.gov/etdeweb/servlets/purl/20047124

[2] J. Leppänen, M. Pusa, T. Viitanen, V. Valtavirta, and T. Kaltiaisenaho. *"The Serpent Monte Carlo code: Status, development and applications in 2013."* Ann. Nucl. Energy, **82** (2015) 142-150

[3] Conant, A., Erickson, A., Robel, M. et al. *Sensitivity and Uncertainty Analysis of Plutonium and Cesium Isotopes in Modeling of BR3 Reactor Spent Fuel*. United States, https://doi.org/10.13182/NT16-88

[4] Palfelt A., Thunberg W. & Winka A. *Determining the Sensitivity of Reactor Parameters in a Sodium Cooled Fast Reactor.* 2020. (MATVET-F). Available from: urn:nbn:se:uu:diva-413073

[5] Larson, E.A.G, A general description of the NRX reactor, AECL-1377, 1961, https://inis.iaea.org/collection/NCLCollectionStore/_Public/41/057/41057258.pdf

[6] R.D. Page, Canadian power reactor fuel, AECL-5609, 1976 https://canteach.candu.org/Content%20Library/19760101.pdf

[7] Ericsson, E., & Hasling, W. (1968). *Ågesta Reactor Plant Operational Experience from 1963 to 1967*. International Atomic Energy Agency (IAEA): IAEA, https://inis.iaea.org/collection/NCLCollectionStore/_Public/44/073/44073669.pdf?r=1

[8] Thomas Jonter (2010*) The Swedish Plans to Acquire Nuclear Weapons, 1945–1968: An Analysis of the Technical Preparations*, Science & Global Security, 18:2, 61-86, https://doi.org/10.1080/08929882.2010.486722

[9] Sandstedt, K.E., & Mogard, H. (1972). *Operating experience with the Agesta Nuclear Power Plant.* International Atomic Energy Agency (IAEA): IAEA.

[10] P. G. Boczar, et al., ”Recent advances in thorium fuel cycles for CANDU reactors”, Thorium fuel utilization: Options and trends (2002): 104, https://www.osti.gov/etdeweb/servlets/purl/20306969#page=110

[11] Gold, D., Reed, P., Hadjimichael, A., Malek, K., Karimi, T., Srikrishnan, V., Keller, K., Gupta, R., Vernon, C. and Rice, J., 2021, December. *Addressing uncertainty in MultiSector Dynamics research: an eBook guide for novice and experienced modelers*. In AGU Fall Meeting Abstracts (Vol. 2021, pp. GC15E-0740). Vancouver .

[12] Kent State University. SSPS: Pearson Correlation, https://libguides.library.kent.edu/spss/pearsoncorr