System Study of CANDU/LWR Synergy in Advanced Nuclear Fuel Cycles

Y. Friedlander McMaster University, Hamilton, Ontario, Canada

Abstract

This report proposes a study that will evaluate the effects of advanced nuclear fuel cycles on resource utilisation, repository capacity, waste streams, economics, and proliferation resistance. The proposed fuel cycles are designed to exploit the unique synergy that exists between light water and CANDU reactors. Also, several fuel cycle simulation codes have been proposed to be used.

1. Introduction

It is generally accepted in the scientific community that carbon dioxide emissions are a major contributor to global warming. While fossil fuels power plants have been identified as a major source for CO_2 emissions, their widespread use has not been abandoned. The nuclear industry represents a viable large scale alternative energy source with an order of magnitude reduction in CO_2 emissions [1]. However, several obstacles exist that retard the expansion of the nuclear industry. These include [2][3][4]:

- A negative public perception of the nuclear industry
- Limited (financially viable) global uranium reserves
- Economic competiveness with fossil fuels
- Long term disposal of nuclear waste
- Proliferation resistance

Advanced nuclear power plants, in particular Gen IV reactors are being considered as a part of the long terms solution [5]. However, by using current power plant designs and new reprocessing technologies, advanced fuel cycles can be developed that allow for the these solutions to be realized in a much shorter time frame.

Among the currently operated reactors, the **Can**adian **D**euterium Uranium (CANDU) reactor has unique characteristics that allow it an integral role in advanced fuel cycles. Because CANDU reactors use natural uranium, they have been designed to have an excellent neutron economy. This allows the reactors to use fuels with a fissile content that would be insufficient in light water reactors (LWR). Also, unlike LWRs, CANDUs are designed for on-line refueling; giving the operator the ability to flatten the power distribution in the reactor by using different fuel management schemes. This translates into flexibility to move between fuel types while still remaining within operating margins [6]. Lastly, the simplicity in design of the CANDU fuel bundles lends itself to the easy fabrication of more complex fuel compositions.

The purpose of this study is to evaluate the relative merit of different fuel cycles options that arise from introducing CANDUs into a LWR reactor park. This report outlines candidate fuel cycles for the study as well as possible evaluation criteria and fuel cycle analysis codes.

2. Fuel cycles

The following are fuel cycles are being considered as candidates for this study:

2.1 Reference case

As a reference case, the advanced fuel cycles can either be compared to a once through fuel cycle in either a LWR or a CANDU. Alternatively, they can be compared to conventional processing such as is practiced currently in France, Japan, UK, India, and Russia.

2.2 TANDEM

In the 1980s, AECL and KAERI researched the possibility of a reprocessing technology that avoids the proliferation risks of associated with conventional reprocessing schemes.. This resulted in the creation of the TANDEM cycle [7]. The TANDEM cycle co-precipitates Pu and U from spent PWR fuel using a chemical decontamination process, thus never creating a separate Pu stream. The Pu and U can then be diluted with natural uranium, depleted uranium, or enrichment tails to create a MOX fuel with a specifically tailored fissile content. The chemical decontamination process is based on a simplified version of PUREX so the process is expected to be economically comparable. Studies have shown the TANDEM cycle has the potential to greatly reduce the uranium consumption while limiting the additional proliferation risks [8]. However, work on the TANDEM cycle has been dormant for many years as the collaboration between AECL and KAERI shifted focus to the DUPIC cycle.



Figure 1 - The TANDEM fuel cycle

2.3 DUPIC

The Direct Use of PWR fuel in CANDU reactors (DUPIC) has been developed over the past two decades by AECL, KAERI, and the US. The fabrication of CANDU fuel bundles from spent PWR fuel has been successfully demonstrated by using the dry reprocessing technique: oxidation and reduction of oxide fuel (OREOX) [9]. The process first involves mechanical decladding of the spent fuel. Repetitive cycles of oxidation and reduction then reduce the fuel to a fine powder. This is followed by milling and fuel pellet fabrication, much as normal CANDU fuel would be made. It

is expected that the fuel will be carried in CANFLEX bundles. Because of the radioactivity of spent fuel, the entire process must be performed remotely. However, despite this added cost, it is expected to be cheaper than the significantly more complex aqueous processes currently used in the fabrication of MOX fuels [7]. A further advantage of the OREOX process is that the volatile and some of the semivolatile fission products are released [10]. This allows for special treatment of ¹³⁵Cs which is a major contributor to the radiotoxic risk of spent fuels in the long term and ¹³⁷Cs which is a major heat contributor in the short term [7].

Increased proliferation was the driving factor in the development of the DUPIC cycle and remains one of it's most advantageous features. There is no separation of plutonium during reprocessing and the high activity of the spent fuel provides an inherent defense against diversion [11]. Furthermore, it is expected that the DUPIC cycle will improve uranium utilisation and reduce the repository size relative to energy generation [12]. This should provide significant economic benefits in both the front and back end of the fuel cycle.



Figure 2 - The DUPIC fuel cycle

2.4 Fluoride volatility

As part of an effort to develop a flexible and proliferation resistant reprocessing technology for future reactor parks that contain both LWRs and FBRs, a process called FLUOREX has been developed by Hitachi in Japan [13]. The process includes the fluorination of spent LWR fuel followed by an aqueous solvent extraction similar to PUREX. It has been proposed that the fluorination process, without the aqueous extraction can be used to create novel advanced fuels that can be used in CANDU reactors [14].

Spent fuel is separated from the fuel assembly using mechanical decladding and an oxidation reduction method such as OREOX, releasing the volatile oxides. The SNF is then moved to a hot chamber and exposed to fluorine. Under these conditions, approximately 75-95% of the UO_2 forms a gaseous UF₆ by the following exothermic reaction [15]:

$$UO_2 + 3F_2 \rightarrow UF_6 \tag{1}$$

A small amount of fission products also form volatile fluorides and, together, with the UF_6 are separated from the solid residue. An optional uranium purification step can be added to achieve a decontamination factor as high as 10^7 . The remaining U, Non-volatile FPs, Pu, and actinides constitute the solid residue referred to as Pu ash. This stream contains too many neutron absorbers to be critical in a LWR. However, the fuel is still more reactive than natural uranium and does not require additional purification steps in order to be used in a CANDU reactor [14]. Both streams can be converted into oxides and fabricated into fuel bundles by well established technologies. However, the highly radioactive Pu ash would require expensive remote handling.



Figure 3 - An example of a fuel cycle using fluoride volatility

There are several options as to what to do with the U stream. As, enrichment plants use UF_6 , reenrichment and use in LWRs is a logical option. Alternatively, the uranium can be used without additional enrichment in a CANDU. Lastly, it has been proposed that if this process is used with spent *CANDU* fuel, the uranium may be sufficiently depleted to be categorized as low level waste which would greatly decrease disposal costs [16].

The uranium stream would provide a significantly larger burnup over natural uranium when used in a CANDU reactor [17]. This would translate into large savings in uranium utilization. Initial WIMS simulations show that the Pu ash can achieve burnup as high as 60GWd/t in a CANDU reactor. Furthermore, as well as removing ¹³⁷Cs during decladding, 38% of the ²³⁹Pu would be consumed [14]. This would dramatically reduce the heat load of the spent fuel, alleviating some of the burden on the backend of the fuel cycle.

2.5 Inert matrix fuel

The issue with using conventional fuels to reduce transuranic (TRU) inventories is that the presence of uranium serves as a source of TRU that counteracts the benefits of the transmutation. To overcome this obstacle, research is being conducted into the possibility of using inert matrix fuels (IMF). In IMF, a neutronically transparent material is used as the actinide carrier, rather than uranium. Different candidate materials have been researched in several countries. Each is investigated for appropriate thermal conductivity, neutron absorption cross sections, reactivity in water, etc. Canada has chosen to focus its efforts on SiC as a potential material, however, practical fabrication of such a fuel is at least 10 to 20 years away [18].

There are two proposed ways that IMF can be used to reduce TRU inventories in spent fuel. A CANDU can be full-core loaded with an IMF containing all the TRU from spent LWR fuel [18] Alternatively, 30 channels can be loaded with an IMF containing only Am and Cm, while the rest of the channels are loaded with unenriched reprocessed uranium (RU). The Pu, possibly mixed with Np, could then be used in MOX fuel in either a CANDU or LWR [6]. Although this would introduce a relatively higher proliferation risk, it reduces reliance on IMF. Both schemes can be achieved by using a UREX or UREX+ variant to reprocess the spent fuel.

The options have been demonstrated to achieve at least 70% destruction of the target TRUs which would result in a reduction in the long term heat load of 70%-80% [18][19]. Furthermore, the IMF would be an appropriate waste form so postprocessing would be unnecessary before disposal.



Figure 4 - a) A fuel cycle burning all the TRU in an IMF b) A fuel cycle burning only Am and Cm in an IMF

2.6 Reprocessed uranium

The uranium in spent LWR fuel has an enrichment of roughly 0.9% so it can be used in CANDU reactors without re-enrichment. Despite slightly larger amounts of ²³⁵U and ²³⁶U, RU is expected to behave similarly to slightly enriched uranium (SEU) in a CANDU reactor [20]. SEU can reduce uranium requirement by approximately 25% as well as provide up to 30% savings in disposal costs [6].

4. Evaluation criteria

The relative merit of different fuel cycles depends on what is desired of the specific region. For example, a fuel cycle implemented in India may be geared towards improving uranium utilisation as their native supplies are limited. Canada, on the other hand, has large amounts of deposits so other criteria may be the driving factors. At any rate, it is helpful to quantify and rank the fuel cycles' abilities to fulfil certain criteria so that, when a body decides on its priorities, the merits of the fuel cycles can be easily compared. The following are some commonly quantified properties of the fuel cycles that may be compared in this study.

4.1 **Resource utilisation**

It is difficult to accurately forecast uranium reserves far into the future as the extent of new nuclear power plant builds and uranium exploration have large margins of uncertainty. However, even conservative estimates predict a definite strain on global uranium reserves by the middle of the century [21]. A major advantage that the nuclear industry has over fossil fuels is the relative lack of volatility in the price of resources. However, to maintain this advantage, the dependency on uranium exploration must be minimized. Even if global reserves are maintained at moderate levels, regions that lack domestic reserves will look to secure its energy production by minimizing their reliance on uranium imports.

Also, there is pressure from environmental organisations to limit the amount of uranium exploration and mining. Limiting the uranium requirements can alleviate some of these pressures. Furthermore, consuming perceived wastes such as civilian plutonium reserves can demonstrate a fuel cycle's ability to satisfy energy demands with minimal environmental impact.

4.2 Decay heat

It is generally accepted that, no matter what path is chosen for the nuclear fuel cycle, a geological repository is required for the final disposal of high level waste. However, different fuel cycles will require different sizes of repository which would have both environmental and economic ramifications. The size if the required repository is relatively independent of the volume of the waste but, rather, is determined by the heat load [2] Therefore, decay heat of the HLW is an important property of the fuel cycle to calculate.

4.3 Radiotoxicity

The chief environmental concern with the nuclear industry is the possible release of radiotoxic elements. However, a properly constructed deep geological repository can be expected to not release any radionuclides until several thousand years. Furthermore, only the *mobile* fission products can be expected to contribute to the dose to the environment until upwards of a million years [2]. Therefore, one must be careful when using HLW radiotoxicity as a design criterion for

fuel cycles. A better measure would be the activity of certain nuclides at times that appropriately correspond to their relative mobility.

4.4 **Proliferation resistance**

A major deterrence for a nation's choice to pursue reprocessing options is the possible risk of diversion of weapons useable materials. A metric to value relative proliferation resistance is often qualitative. However, there are several attempts to develop quantitative measures that can be used [22]. These are based on the time and ease to separate a significant quantity (SQ) of weapons useable material as any step in the recycling process.

4.5 Waste streams

Every step in the fuel cycle creates different wastes. The wastes have varying degrees of environmental and radiotoxic risk. These wastes should be tracked in order to give a more complete assessment of the environmental effects of each fuel cycle.

4.6 Economics

Cost analysis of undeveloped technologies is hampered by large uncertainties. The costs can be estimated relative to existing technologies by taking into account factors such as relative complexity, radiotoxic hazards, and scale of deployment. However, as these cannot be estimated with any amount of certainty, it may be more useful to conduct a sensitivity analysis to determine at what prices each step in the fuel cycle must be in order for the fuel cycles to be financially equivalent.

Another economic factor that should be considered is the energy consumption for each step in the fuel cycle.

5. Method

Most fuel cycle simulation codes are comprised of two main parts: a depletion code and a mass flow code. The depletion code determines the isotropic composition of the fuel at discharge and the mass flow code tracks the flow of the nuclear material. For this study, isotropic composition data is being taken from previous simulations so the depletion code is unnecessary.

The mass flow calculations can be performed in several ways and depends on the code used. The mass flows can be limited by varying nuclear demands or capacities. They can also be dynamically influenced by economic models. The calculations may be done assuming a continuous flow of materials in a reactor park or they can be done by tracking the mass flows of an individual batches [23]. For this study, the code used will be chosen by its availability and its ability to evaluate the proposed fuel cycles for the aforementioned criteria.

5.1 Candidate codes

5.1.1 <u>Nuclear Fuel Cycle Simulation System (NFCSS)</u>

NFCSS is available for free through the IAEA website. It uses the depletion code CAIN but allows the use to manually select the input and discharge isotropic compositions. The code can track fuel

stockpiles and uranium requirements over many years for a changing reactor park. Various recycling strategies are supported [24].

5.1.2 Dynamics of Energy System of Atomic Energy (DESAE)

DESAE is another free simulation code provided by the IAEA. It has similar capabilities to NFCSS but includes an economic model and a decay heat calculation for a limited number of fission products [25].

5.1.3 Verifiable Fuel Cycle Simulation (VISION)

As a part of the Advanced Fuel Cycle Initiative (AFCI), the US Department of Energy developed a fuel cycle simulation code called VISION. As well as tracking mass flows, the code can perform a long term disposal analysis including heat load, radiotoxicity and dose. Furthermore, tools to analyze the fuel cycles for economics and proliferation metrics are included [26].

5.1.4 Dynamic Analysis of Nuclear Energy Systems Strategies (DANESS)

Developed by Argon National Laboratories (ANL), DANESS is designed to demonstrate the dynamic dependence between a reactor park's demands and economic capabilities. However, the code is customisable to adhere to the objectives of different studies. The code allows the fuel cycles to be studied in detail according to economic, environmental, and socio-political criteria [27].

5.1.5 <u>Commelini-Sicard (COSI)</u>

COSI was developed by the French Atomic Energy Commission. Unlike the previous codes, which use a continuous mass flow model, COSI tracks the fuel by batch. It conducts a very detailed analysis, especially in the back end, where it can track a near complete list of radionuclides in the spent fuel for upwards of a million years [28].

6. Conclusion

A study that will examine the ability that various nuclear fuel cycles have to exploit the synergy between light water and CANDU reactors has been proposed. Various fuel cycles have been presented as candidates for analysis. They are:

- DUPIC cycle
- TANDEM cycle
- Annihilation of TRUs using an IMF
- Burning spent fuel reprocessed by fluorination

Also, the criteria by which the fuel cycles can best be judged have been discussed. Broadly, they are:

- Resource utilisation
- Heat load of spent fuel
- Radiotoxicity of spent fuel
- Proliferation resistance
- Economics

Several computer codes have been examined for their capability to achieve the objective of this study. The next step in this study will involve the selection of a code and refining the proposed fuel cycles and judging criteria according to the capability of the code.

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