Nuclear Engineering and Technology 50 (2018) 794–800

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Technical Note

Comparison of proliferation resistance among natural uranium, thorium–uranium, and thorium–plutonium fuels used in CANada Deuterium Uranium in deep geological repository by combining multiattribute utility analysis with transport model

Shinya Nagasaki*, Xiaopan Wang, Adriaan Buijs

McMaster University, 1280 Main Street West, Hamilton, ON, L8S 4L7, Canada

ARTICLE INFO

Article history:
Received 17 November 2017
Received in revised form
5 February 2018
Accepted 12 February 2018
Available online 1 March 2018

Keywords:
CANada Deuterium Uranium Fuel Cycle
Deep Geological Repository
Multiattribute Utility Analysis
Proliferation Resistance
Transport Model of Radionuclides in Repository
Used Thorium Fuel Disposal

ABSTRACT

The proliferation resistance (PR) of Th/U and Th/Pu fuels used in CANada Deuterium Uranium for the deep geological repository was assessed by combining the multiattribute utility analysis proposed by Chirayath et al., 2015 [1] with the transport model of radionuclides in the repository and comparing with that of the used natural U fuel case. It was found that there was no significant advantage for Th/U and Th/Pu fuels from the viewpoint of the PR in the repository. It was also found that the PR values for used nuclear fuels in the repository of Th/U, Th/Pu, and natural U was comparable with those for enrichment and reprocessing facilities in the pressurized water reactor (PWR) nuclear fuel cycle. On the other hand, the PR values considering the transport of radionuclides in the repository were found to be slightly smaller than those without their transport after the used nuclear fuels started dissolving after 1,000 years.

1. Introduction

CANada Deuterium Uranium (CANDU) is a pressurized heavy water power reactor that has been in operation in many countries since the 1980s. There are 31 CANDU reactors in operation globally (Argentina, Canada, China, India, Pakistan, Romania, and South Korea). One key feature of CANDU is that heavy water functions both as moderator and coolant. The high moderation efficiency offers CANDU great flexibilities for using other types of fuel, including thorium (Th), mixed oxide (MOX) fuel, or even spent fuel from light water reactors. Especially, the CANDU system with Th fuel is considered to be one of the promising options for the future [2]. It has also been pointed out that the Th fuel cycle in CANDU is seen as being more proliferation resistant than the uranium fuel cycle [3–6] although the International Fuel Cycle Evaluation (1977–1980) concluded that the proliferation resistance (PR) of Th fuel cycle is generally similar to that of the Pu fuel cycle.

PR has become an important notion in characterizing a variety of nuclear fuel cycle options. Since the 1970s, a number of methodologies of PR assessment have been developed around the world [7–14]. Two popular projects are (i) the Generation IV International Forum's project of Proliferation Resistance and Physical Protection (PR&PP) of nuclear energy systems [7] and (ii) the International Atomic Energy Agency (IAEA's) Innovative Nuclear Reactors and Fuel Cycles (INPRO) project for developing a methodology for the holistic assessment of nuclear energy systems [8]. Other methodologies such as Technology Opportunities for Proliferation Resistance [9] and Japan Atomic Energy Agency's methods [10] are mainly dependent on expert judgment and came out with a qualitative assessment. The PR&PP, the multiattribute utility analysis (MAUA) [11], the fuzzy logic method [12], and the risk-informed probabilistic analysis [13] provide quantitative values relevant to PR. The MAUA incorporates subjective or objective inputs to calculate PR values. Recently, the INPRO collaborative project with Generation IV International Forum proliferation resistance and safeguardability assessment Tools [15] has been conducted. However, only a few studies have conducted the assessment of the PR for used nuclear fuel and high-level radioactive waste disposal [8,9,11,16]. Furthermore, to the best of the authors' knowledge, no article has taken into account the transport of radionuclides in the repository after the closure of the repository.

* *Corresponding author.*
E-mail address: nagasas@mcmaster.ca (S. Nagasaki).

https://doi.org/10.1016/j.net.2018.02.001

Postclosure safety assessments have demonstrated that the used nuclear fuels generated from the current CANDU nuclear fuel cycle (i.e., natural uranium fuel/once-through system) will be safely managed [17,18]. Safe management of used nuclear fuel is essential for public acceptance of the use of nuclear energy. However, the assessment of PR for used nuclear fuel disposal in the CANDU system has not been carried out. It may not have to be considered that a state would try to divert the nuclear materials from a closed repository. However, a postclosure monitoring may have to be implemented as a part of the postclosure safety case to detect radioactive material and/or other toxic material in the environment that could be attributable to the disposal facility [19,20]. The type of planned postclosure monitoring such as monitoring on the surface, through a ventilation line or an access tunnel, or near the underground facility varies between countries. The duration of postclosure monitoring at used nuclear fuel repository is dependent on a lot of factors. In the example of the postclosure monitoring for low-level radioactive waste repository in Japan (at least 300 years), it may be more than several hundred years. If an unexpected/undesirable incident occurs in the repository during the postmonitoring period, the implementer and/or regulatory authority may have to consider the retrievability or the reversibility of the disposal process to some extent [21]. These suggest there may be some access routes to the underground facility until the postclosure monitoring is officially completed. In this article, we considered a case where a state accesses to the used nuclear fuel matrix to divert during the operation and postclosure of repository.

The MAUA methodology was developed by Charlton et al. [11]. This method successfully allowed us to compare the PR trend for different nuclear fuel cycles and facilities. Their method used 14 attributes to assess a separate PR value for each process in a nuclear fuel cycle. Based on this MAUA methodology [11], recently, Chirayath et al. [1] have developed a computer code named PRAETOR (Proliferation Resistance Analysis and Evaluation Tool for Observed Risk) to aid in comparing the PR of nuclear installations. The PRAETOR code uses a metric, which considers both intrinsic and extrinsic measures at a nuclear installation, and the nonproliferation metric computed by the PRAETOR code is based on 63 attributes [1]. The details of MAUA and the PRAETOR code are described later in the article.

The proliferation aspect of used nuclear fuel disposal should be treated carefully because of its unique features. A significant amount of U and Pu will exist in the repository for thousands of years. Over such a long period, the total inventory of U and Pu in the repository changes, and their radioisotope compositions also change. In the reference case scenario [17,18,22], after the container failure, used nuclear fuel starts dissolving. The dissolved radionuclides will be transported by diffusion and advection with groundwater, leaving the precipitates of low solubility elements such as U and Pu in the repository. The PR in the repository must thus be expected to change with time.

The purpose of this article is to study whether Th fuel (Th/U and Th/Pu MOX fuels) in CANDU has an advantage of PR in the used nuclear fuel repository, compared with the PR of the natural uranium fuel case. In general, ThO_2–UO_2 MOX fuel has been considered for the Th fuel cycle. Furthermore, ThO_2–PuO_2 MOX fuel was also proposed as one idea to positively use surplus Pu [23].

In this article, the PRAETOR code with 63 attributes [1] was used to assess the PR values because Chirayath et al. showed that the PRAETOR code gave reasonable insights into the PR values of pressurized water reactor (PWR) spent fuel assemblies in two nuclear material diversion scenarios for a nuclear fuel cycle installation [1]. The transport calculation procedure of radionuclides in the engineered barrier was adopted to take into account the effects of dissolution, precipitation, diffusion, and migration of U, Pu, and other radionuclides in the repository [24]. The repository was assumed to be located at a depth of 500 m in the granite geological media [18]. The repository design concept developed by the Nuclear Waste Management Organization of Canada was adopted [18].

2. MAUA and the PRAETOR code [1]

According to IAEA [25], the PR is defined by the characteristic of a nuclear energy system that impedes (i) the diversion or undeclared production of nuclear material or (ii) the misuse of technology by states to acquire nuclear weapons or other nuclear explosive devices. The PR consists of two essential measures; the intrinsic features of the nuclear materials and the extrinsic barriers at the nuclear installation. The intrinsic features result from the technical design of the nuclear energy systems, and the extrinsic measures are those that result from the implementation of states' undertakings related to nuclear nonproliferation [1].

A research group of Texas A&M University has developed a methodology for the PR value assessment of nuclear fuel cycle technologies based on the MAUA theory [26–29]. They also have developed a computer code, named PRAETOR, to aid in comparing the PR values of nuclear installations [1].

2.1. Multiattribute utility analysis [1,11,26–29]

Generally speaking, the MAUA theory is based on compiling multiple factors into a single metric to facilitate easy decision-making. Furthermore, MAUA is considered to be able to incorporate complex and interrelated components in a decision.

In the MAUA theory, we first develop a set of individual attributes, (x_i), which can best describe the system under consideration. The values of the attributes we develop are mapped to a utility value (u_i) between 0 and 1 using appropriate objective functions, which are usually called utility functions. The higher the utility value is, the higher the PR of the system is. We also assign an adequate weight (w_i) to each utility value which reflects its relative significance. The MAUA theory provides a methodology to convert the multiple utility values and their weights into a single value of PR. Therefore, we can compare the PR values of different nuclear fuel cycle systems with each other [27,28].

It is crucial to assume preferential and utility independence of the chosen attributes, so that the MAUA theory relationships are valid [1]. The general form of the MAUA function is

$$U(x_1, x_2, \cdots, x_n) = \sum_{i=1}^{n} k_i u_i(x_i) + K \sum_{i=1,\, j>i}^{n} k_i k_j u_i(x_i) u_j(x_j) + K^2 \sum_{i=1\, j>i,l>j}^{n} k_i k_j k_l u_i(x_i) u_j(x_j) u_l(x_l) + \ldots + K^2 k_1 k_2 \ldots k_n u_1(x_1) u_2(x_2) \ldots u_n(x_n), \quad (1)$$

where the function u_i represents a utility function for the individual attributes which are normalized to a scale from 0 to 1; the constants k_i are weighting factor ($0 < k_i \leq 1$) for each attribute indicating the significance of one attribute relative to the others; U is the overall utility value (single metric); and the constant K is a scaling parameter which is a solution to the following equation (2) with the constraint that $K > -1$ [1]:

$$1 + K = \prod_{i=1}^{n} (1 + K k_i). \quad (2)$$

When the sum of all k_i is equal to unity, then $K = 0$, and the equation (2) can be rewritten as the equation (3) "additive utility function" given by [1]:

$$U(x_1, x_2, \ldots, x_n) = \sum_{i=1}^{n} k_i u_i(x_i). \quad (3)$$

On the other hand, when the sum of k_i is not equal to unity, then $K \neq 0$, and we can obtain the equation (4) "multiplicative utility function" given by [1]:

$$1 + KU(x_1, x_2, \ldots, x_n) = \prod_{i=1}^{n} (1 + Kk_i u_i(x_i)). \quad (4)$$

The additive utility function provides a weighted average of all the individual attributes. Hence, it is desirable to use the additive utility function when our goal is to find a system that performs well against as many measures of PR as possible [1]. However, this also limits the influence of any one attribute to the value of its weight [1]. On the other hand, while the result of the multiplicative utility function is still a PR value between 0 and 1, it now allows for extreme values to affect the result more heavily [1]. Furthermore, this serves adequately in comparing two technology options against one another [1]. Therefore, in this article, we used the PRAETOR code with the multiplicative utility function form.

2.2. PRAETOR code

In total, 63 attributes are taken into account in the PRAETOR code. The PR values from the PRAETOR code range between 0 and 1 with high values indicating higher PR [1]. Comparisons using PR values between various nuclear fuel cycle components, arrangements, and systems can indicate where to allocate nonproliferation assets and expertise [1]. The assessment results by the PRAETOR code inform the decision-makers where limited nonproliferation resources most effectively raise PR hurdles and best hinder potential proliferation efforts [1].

The PRAETOR code requires two input files. The first file contains numeric values or string text inputs as appropriate for all the 63 attributes. As the template of the first file is prepared by Chirayath et al. [1], what users need to do is to add the values of mass of U and Pu per significant quantity and the radiation level in terms of dose, for example, to the template file. The second input file needs to be supplied by the user. It contains the weight values required for the MAUA [1]. Chirayath et al. [1] also listed the expert weights for each of the 63 attributes. The output file of the PRAETOR code contains the utility value for each attribute value and the PR values.

According to Chirayath et al. [1], the overall PR value in the output file should be used only for comparison among the different nuclear fuel cycle scenarios. That is because the PR value for one scenario does not have much physical meaning unless it is used to compare with the PR value for another scenario [1]. Therefore, the assessment results by the PRAETOR code are most useful to compare the PR values between two or more scenarios and to identify the relative strengths against proliferation of nuclear materials within a single system or between systems under discussion [1]. More details of the PRAETOR code are described elsewhere [1].

3. Calculation methods

3.1. Fuels

In this article, three different CANDU fuels were considered to investigate the advantage of PR for Th fuel in the used nuclear fuel repository; (1) natural U fuel, (2) Th/U MOX fuel (30% Th+70% U (1.58% enriched U)) [30], and Th/Pu MOX fuel (96% Th+4% Pu). Plutonium used in Th/Pu fuel came from the reprocessing of natural uranium fuel used in CANDU. The thorium and plutonium mixing ratio was decided by optimization of CANDU reactor operation [31].

3.2. Burnup calculation

The burnup and decay calculations for three nuclear fuels in CANDU were carried out by using the TRITON [32] and ORIGEN-ARP [33] codes in SCALE code, version 6.1 [34].

In the CANDU system considered in this article, fuels were arranged in the channels located on a square lattice. The whole core was contained in a horizontal cylindrical calandria tank. Every fuel channel assembly was composed of a zirconium–niobium alloy pressure tube, a zirconium calandria tube, stainless steel end fittings at the end, and spacers to support the pressure tube and separate it from the calandria tube. The annulus between the pressure tube and the calandria tube was filled with CO_2 to provide thermal insulation for heat loss reduction. For the burnup calculations of CANDU fuel, a 37-element fuel bundle was modeled to represent a whole reactor core. The burnup history also took into account a fact that CANDU was refueled at full power at the rate of two channels per day. The average burnup of CANDU fuel was 7,927.5 MWd/t for natural U fuel and 7,927.5 MWd/MTHM for Th/U and Th/Pu fuels. Th fuel bundles were assumed to be loaded with a homogeneous MOX mixture of either Th and U or Th and Pu. The masses of U, Pu, ^{237}Np, and Am in the discharged used nuclear fuels (g/ton-fuel) are summarized in Table 1.

3.3. The PRAETOR code calculation

The PRAETOR code, the user's manual, the example of input files, and other necessary files for calculation were provided to us by Chirayath of Texas A&M University. By including the intrinsic and extrinsic features in the PRAETOR code, the PR values associated with different nuclear fuel cycles can be determined [1].

The PRAETOR code has two options; the additive utility function form and the multiplicative utility function form [1]. Because the latter serves adequately in comparing two or more nuclear fuel cycle scenarios against one another [1] as mentioned previously, we used the PRAETOR code with the multiplicative utility function form in this article. We used the expert weights which Chirayath et al. [1] listed for each of the 63 attributes in the calculation.

Table 1
Mass (g/ton-fuel) of U, Pu, ^{237}Np, and Am in the discharged used natural U fuel, Th/U mixed oxide fuel [30% Th+70% U (1.58% enriched U)], and Th/Pu mixed oxide fuel (96% Th+4% Pu).

Isotope	Natural U fuel	Th/U mixed oxide fuel	Th/Pu mixed oxide fuel
U-232	—	5.99×10^{-1}	1.58
U-233	2.17×10^{-5}	3.00×10^{3}	2.95×10^{3}
U-234	4.47×10^{-2}	1.70×10^{2}	9.54×10^{1}
U-235	2.05×10^{3}	4.58×10^{3}	1.97
U-236	7.68×10^{2}	9.91×10^{2}	7.95×10^{-1}
U-237	1.94	1.37	1.18×10^{-3}
U-238	9.85×10^{5}	6.85×10^{5}	1.05×10^{-3}
Total U	9.92×10^{5}	6.94×10^{5}	3.05×10^{3}
Pu-238	4.14	2.02	3.04×10^{1}
Pu-239	2.49×10^{3}	1.61×10^{3}	1.71×10^{4}
Pu-240	1.07×10^{3}	5.17×10^{2}	1.24×10^{4}
Pu-241	2.66×10^{3}	9.22×10^{1}	1.70×10^{3}
Pu-242	7.38×10^{1}	1.66×10^{1}	8.81×10^{2}
Total Pu	3.91×10^{3}	2.23×10^{3}	3.22×10^{4}
Np-237	2.78×10^{1}	1.90×10^{1}	2.34×10^{-2}
Am-241	1.98	6.94×10^{-1}	2.67×10^{1}
Am-243	3.37	5.15×10^{-1}	7.58×10^{1}

3.4. Transport calculation in repository

The current conceptual design of a deep geological repository in the Canadian Shield (crystalline rock; the main mineral composition is granite) in Canada consists of a repository constructed at a depth of approximately 500 m below surface. The used nuclear fuel bundles discharged from CANDU reactors will be encapsulated in containers. The containers will be surrounded by compacted bentonite. Bentonite is a type of clay that swells on contact with water, resulting in its natural self-sealing property. The details of the repository design concept are described elsewhere [18].

An outline of equations used in this article for the dissolution of used nuclear fuel, the diffusion of radionuclides in compacted bentonite, and environmental constraints around fuel matrix and in the compacted bentonite are described in the Appendix. The details of the equations are described elsewhere [18,22,24], and the values of parameters related to the repository design such as length of compacted bentonite are listed in the reference [18].

The equations describing the used nuclear fuel dissolution and dissolution rates for Th/U and Th/Pu fuels were assumed to be the same as those of natural U fuel [18] for the following reasons. First, the formula for dissolution of Th/U and Th/Pu CANDU fuels has not been reported. Second, the fuel dissolution rates of Th/U and Th/Pu fuels for CANDU may be dependent on the fuel composition and burnup history, but this dependence has not been reported either. Finally, the dissolution rate is not significantly sensitive to the transport calculation results in the repository [18]. The thermodynamic data compiled in the Japan Atomic Energy Agency database [35] and the sorption data summarized by Vilks [36] were used for the calculations.

The groundwater systems in the Canadian Shield are expected to have a relatively high ionic strength [18,36]. However, a repository site location has not been chosen in Canada yet, and the sorption data such as sorption distribution coefficients of U and Pu in saline solutions are not sufficiently compiled in any sorption databases in the world. Hence, in this article, the groundwater in the deep geological repository environment was assumed to have a low ionic strength.

In this article, we assumed that after being discharged from CANDU, used nuclear fuel bundles were stored in the spent fuel pool for 6 years and in dry cask storage for 44 years before they were moved into the deep geological repository and that the repository was closed 100 years after the discharge. After 1,000 years, the fuel matrix is assumed to start dissolving into the groundwater (this is the same scenario as references [18,22,24]). We assumed that the inventory in the repository was measured once a day during the operation period (50–100 years), and further inventory measurement was not conducted after the repository was closed. The trend of PR values in the repository with time was calculated for the used Th/U, Th/Pu, and natural U fuels. In this article, U, Pu, and other radionuclides existing only in the used nuclear fuels including the precipitate around the bundles were considered in assessing the PR. Namely, we assumed that U and Pu present in the buffer materials and natural barrier were no longer attractive for proliferation and radionuclides present there did not contribute to the heat and radiation barriers in the bundles.

For comparison, the trend of PR values without the transport calculation and the PR values for enrichment and reprocessing facilities in PWR fuel cycle (4.5% slightly enriched U fuel; 45,000 MWd/t) were also calculated. Burnup calculation results from the previous work [16] were used.

4. Results and discussion

The trend of PR values in the repository is illustrated in Fig. 1. The PR values for enrichment and reprocessing facilities in the PWR fuel cycle are also shown in Fig. 1. The PR values for all fuels used in CANDU in the repository gradually decreased with time due to the reduction of heat and radiation generation in the used fuel bundles. Because the frequency of inventory measurement changed from daily measurement to no measurement when the repository was closed, the PR values decreased by 0.09 after 100 years. After 1,000 years, Am-241 which was a contributor to the heat and radiation generation along with Pu-239 and Pu-240 started dissolving because of its relatively high solubility (2×10^{-7} mol/L [22]). Furthermore, the sorption distribution coefficient, K_d (see Appendix), of Am is much smaller than that of U and Pu [35,36]. Dissolved Am transported through the compacted bentonite due to a low K_d value and was removed from the used nuclear fuels matrix. On the other hand, U and Pu with very low solubility (10^{-9} mol/L [22]) did not dissolve so much and therefore remained in and around the bundles. In the case of Th/U fuel, Po-213 and other daughters of U-233 were minor contributors, and their solubility and K_d values are high and low, respectively, compared with U and Pu. Hence, the PR values further decreased.

The results of the PR value calculation by PRAETOR showed the following:

(1) the PR value for used Th/Pu fuel was slightly higher than that for used natural U and Th/U fuels because of the heat and spontaneous neutron generation from Pu, but the difference in the PR values among three fuels was practically negligible, and
(2) the PR values for all used nuclear fuels in the repository were comparable with those for enrichment and reprocessing facilities in PWR fuel cycle. When the inventory in the repository was stopped being measured after the repository closed, the PR values became smaller than the PR for the enrichment facility. The PR values became very small when the used nuclear fuel started dissolving and the actinides and fission products were removed from the used nuclear fuel matrix.

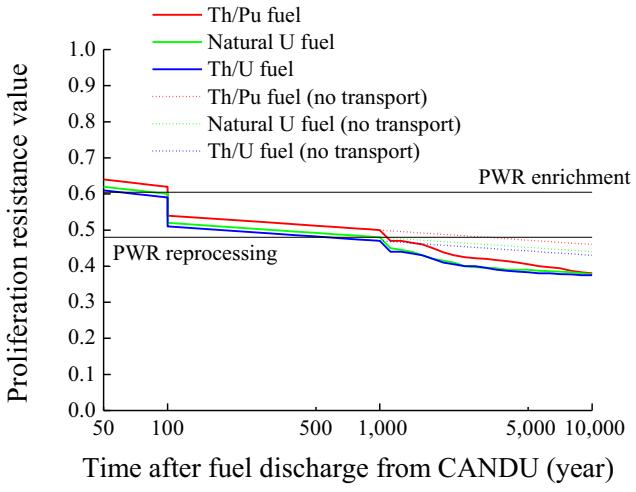

Fig. 1. Proliferation resistance of Th/Pu, Th/U, and natural U fuels used in CANDU in deep geological repository. Dotted lines represent the proliferation resistance without transport calculation of radionuclides in repository. Black solid lines represent the proliferation resistance of enrichment and reprocessing facilities in the PWR nuclear fuel cycle.
CANDU, CANada Deuterium Uranium.

Th fuel and the Th fuel cycle are considered to have excellent inherent features for PR [3–6,37]. However, there is no advantage for Th/U and Th/Pu fuels used in CANDU from the viewpoint of PR in the deep geological repository compared with natural U fuel.

Charlton et al. [11] showed the PR values in the geological repository for a once-through fuel cycle of a PWR, but the transport of radionuclides was not taken into account. Their PR values in the repository were about 0.55 between 100 and 500 years and 0.5 between 500 and 1,000 years. Their PR values from before the onset of dissolution of radionuclides from the used nuclear fuel are comparable with the PR values in Fig. 1, but their methodology and the PRAETOR code do not take into account the transport of radionuclides in the disposal system, and therefore, the PR values after the onset of dissolution of used nuclear fuel and after the start of the transport of radionuclides can perhaps not be evaluated.

The largest prompt variation of PR value in the repository appeared at 100 years only due to the change in the utility function value for the attribute “frequency of measurement”. Although the MAUA methodology was developed to assess the PR of diverse nuclear fuel cycles from mine and mill to geological repository [1,11], the features of deep geological disposal system might not be fully included. In the former MAUA methodology developed by Charlton et al. [11], the attribute “physical barriers” was taken into account. An “inaccessible” implied that the material could not be physically accessed, for example, material being irradiated in the reactor. A “canyon” referred to a completely enclosed, underground structure for which it was very difficult to gain access. However, in the PRAETOR code, representing the latest MAUA methodology, this attribute is not considered anymore [1]. In addition, to use the used nuclear fuels for proliferation after the closure of repository, they had to be retrieved from the repository, for example. However, such a process and time required to retrieve the fuels from the deep geological repository were not taken into account anymore. Therefore, to study the PR of various nuclear fuels in the deep geological repository using the MAUA methodology as implemented in the PRAETOR code, not only several new attributes which are relevant to the deep geological disposal system may need to be added but also some of the current attributes may need to be modified. The current attributes are adequate for the processes in which nuclear materials move relatively quickly. However, U, Pu, and other actinides and fission products in the repository do not move practically at all on the long term. The attributes which represent the time-integrated proliferation risk might be needed. Introduction of new attributes and modification of current attributes which are also realistic from the mining to the disposal will be investigated in future work. If the PR value for the repository is still comparable with that for enrichment and reprocessing facilities even after new attributes are introduced and the current attributes are modified, the adequate countermeasures against the proliferation possibility will be implemented in the repository for the safety and security of used nuclear fuel disposal.

5. Conclusions

Using the PRAETOR code [1], the PR values of Th fuels (Th/U and Th/Pu MOX fuels) used in CANDU in the deep geological repository were evaluated and compared with the PR value of the natural U fuel case. It was found that there was no significant difference in the PR values and the trend of PR values with time in the deep geological repository among the Th/U, Th/Pu, and the natural U fuel used in CANDU. This suggests that the advantage of Th fuel cycle in the CANDU system, compared with the natural U fuel cycle, is smaller than previously considered [3–6,29] from the viewpoint of PR in the deep geological disposal.

The PR values became very small when the used nuclear fuel started dissolving and the fission products were released from the used nuclear fuel. To evaluate the PR of various nuclear fuels in the deep geological repository using currently available MAUA methodology, not only several new attributes relevant to the deep geological disposal system may need to be added but also some of the current attributes may need to be modified.

Acknowledgments

This work was supported by the National Science and Engineering Research Council of Canada under Grant RGPIN-2014-05732. Authors greatly appreciate Dr Sunil S. Chirayath of Texas A&M University and Professor William S. Charlton of University of Nebraska for offering the PRAETOR code, user manual, and other files, for helping with the calculation, and for providing valuable comments and suggestions. Authors thank Dr Jorshan Choi of Berkeley Nuclear Research Center, Dr Yusuke Kuno of IAEA, Professor John Luxat of McMaster University, and Dr Jeremy Whitlock of Canadian Nuclear Laboratories for their helpful and constructive comments and ideas.

Appendix

Equations for transport of radionuclides in repository.

The dissolution of fuel matrix was considered to be expressed in terms of a long-term dissolution rate. The concentrations in the vicinity of the fuel are described by the mass balance equation (A-1), which includes the effects of congruent release with dissolution, solubility limits with precipitation/dissolution, diffusion into the compacted bentonite, and radioactive decay/ingrowth.

$$V^F\frac{\partial A_{ij}^F}{\partial t} = 2\pi r_{in} L\varepsilon^B D_{pi}\left.\frac{\partial C_{ij}^B}{\partial r}\right|_{r=r_{in}} + M_{ij}g_{si} - V^F\lambda_{ij}A_{ij}^F + V^F\lambda_{IJ}A_{IJ}^F \quad \text{(A-1)}$$

where

C_{ij}: concentration of dissolved nuclides [mol/m^3];

V^F: volume of the hypothetical water-filled region around used nuclear fuel matrix [m^3];

M_{ij}: inventory of isotope j of element i in the used nuclear fuel matrix [mol];

g_{si}: fractional rate of decrease of the used nuclear fuel matrix volume [y^{-1}];

D_{pi}: diffusion coefficient in pore water [m^2/y];

ε^B: porosity of compacted bentonite [-];

L: length of compacted bentonite [m];

r: distance from the center of the used nuclear fuel matrix [m];

r_{in}: inner radius of the compacted bentonite [m];

t: time after dissolution starts [y]; and

λ_{ij}: decay constant [y^{-1}].

F and B represent the used nuclear fuel matrix and compacted bentonite, respectively. ij represents isotope j of target element i. IJ represents isotope J of element I which is the parent isotope of ij. Total amount of nuclide in a unit volume of the hypothetical water-filled region around the used nuclear fuel matrix, A_{ij}^F, is given by equation (A-2)

$$A_{ij}^F = C_{ij}^F + P_{ij}^F, \quad \text{(A-2)}$$

where P_{ij}^F is the concentration of precipitate around the used nuclear fuel matrix.

The fractional rate of decrease of the used nuclear fuel matrix volume is given by

$$g_{si} = \frac{a^{fuel}}{\rho^{fuel} V^{fuel}} k, \tag{A-3}$$

where

a^{fuel}: surface area of the used nuclear fuel matrix [m^2];
ρ^{fuel}: density of the used nuclear fuel [kg/m^3];
V^{fuel}: volume of the used nuclear fuel matrix [m^3]; and
k: used nuclear fuel matrix dissolution rate [$kg/m^2 \cdot y$].

The time-dependent amounts of nuclides in the used nuclear fuel matrix are given by

$$\frac{dM_{ij}}{dt} = -M_{ij} g_{si} - \lambda_{ij} M_{ij} + \lambda_{IJ} M_{IJ}, \tag{A-4}$$

where $M_{ij}(0)$ is the nuclide inventory at the time when the dissolution starts.

The migration of nuclides in the compacted bentonite region is expressed by equation (A-5), which represents nuclide diffusion, sorption, precipitation/dissolution, and decay/ingrowth. Sorption is described by instantaneous/reversible/linear reactions (see equation (A-7)), and precipitation/dissolution is instantaneous/reversible reactions

$$\frac{\partial A_{ij}^B}{\partial t} = \varepsilon^B D_{pi} \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_{ij}^B}{\partial r} \right) - \lambda_{ij} A_{ij}^B + \lambda_{IJ} A_{IJ}^B, \tag{A-5}$$

where A_{ij}^B in equation (A-5) represents the total amount of nuclide in a unit volume of compacted bentonite (equation (A-6))

$$A_{ij}^B = \varepsilon^B C_{ij}^B + \left(1 - \varepsilon^B\right) S_{ij}^B + P_{ij}^B. \tag{A-6}$$

The concentrations of sorbed nuclide in a unit volume of the solid part of compacted bentonite, S_{ij}^B, are given by

$$S_{ij}^B = \rho^B K d_i C_{ij}^B, \tag{A-7}$$

where ρ^B is the density of bentonite excluding the pore [kg/m^3] and Kd_i the sorption distribution coefficient of element i [m^3/kg].

Precipitation occurs when

$$\varepsilon^B C_i^* + \left(1 - \varepsilon^B\right) \rho^B K d_i C_i^* < \sum_j A_{ij}^B, \tag{A-8}$$

where C_i^* is the element solubility [mol/m^3]. The concentration of precipitate in a unit volume of compacted bentonite, P_{ij}^B, is derived by equation (A-9)

$$P_{ij}^B = A_{ij}^B - \left(\varepsilon^B C_i^* + \left(1 - \varepsilon^B\right) \rho^B K d_i C_i^* \right) \frac{A_{ij}^B}{\sum_j A_{ij}^B}, \tag{A-9}$$

where the concentration of dissolved nuclides in a unit volume of pore water in the compacted bentonite, C_{ij}^B, is given by equation (A-10), which accounts for the limited elemental solubility (C_i^*) which is partitioned among stable and radioactive isotopes

$$C_{ij}^B = C_i^* \frac{A_{ij}^B}{\sum_j A_{ij}^B}. \tag{A-10}$$

These equations are solved for the following initial and boundary conditions.

$A_{ij}^B(r, 0) = 0$ at $r_{in} \le r \le r_{out}$
$A_{ij}^B(r_{out}, t) = 0$ at $t > 0$.
where r_{out} is the outer radius of the compacted bentonite [m].

Conflicts of interest

The authors declare no conflicts of interest.

References

[1] S.S. Chirayath, R. Elmore, G. Hollenbeck, N.G. Chandregowda, W.S. Charlton, R. Metcalf, J.C. Ragusa, Proliferation resistance analysis and evaluation tool for observed risk (PRAETOR) - methodology development, J. Nucl. Mater. Manage. 43 (2015) 22–47.
[2] CISION, Candu Energy Inc, Welcomes Positive Review of AFCR Technology in China, January 29, 2018. http://www.newswire.ca/news-releases/candu-energy-inc-welcomes-positive-review-of-afcr-technology-in-china-516331731.html.
[3] World Nuclear News, Thorium Use in Candu Units to Be Assessed July 15 2009, 2009.
[4] P.G. Boczar, Candu Fuel-cycle Vision, AECL Report, AECL-11937, 1998. http://www.iaea.org/inis/collection/NCLCollectionStore/_Public/31/010/31010434.pdf.
[5] M. Yamawaki, H. Yamana, H. Unezaki, Research, development and trend of thorium fuel cycle I, J. Atom. Ener. Sci. Jpn. 47 (2005) 802–821 (in Japanese).
[6] M. Yamawaki, H. Yamana, H. Unezaki, Research, Development and Trend of thorium fuel cycle II, J. Atom. Ener. Sci. Jpn. 48 (2006) 20–34 (in Japanese).
[7] I.V. Generation, International Forum, Evaluation Methodology for Proliferation Resistance and Physical Protection of Generation IV Nuclear Energy Systems, Rev. 6, 2011.
[8] International Atomic Energy Agency, in: Guidance for the Application of an Assessment Methodology for Innovative Nuclear Energy Systems. INRPO Manual - Proliferation Resistance, 2008. IAEA-TECDOC-1575, Rev. 1 November 2008.
[9] US DOE, Technological opportunities to increase the proliferation resistance of global civilian nuclear power systems, TOPS Task Force on the Nuclear Energy Research Advisory Committee, US DOE, 2001.
[10] N. Inoue, M. Hori, Methodologies of Nuclear Proliferation Resistance Assessment. Proceedings of 44th Annual Meeting of the Institute of Nuclear Materials Management, Phoenix Arizona, 2003. July 13–17.
[11] W.S. Charlton, R.L. LeBouf, C. Gariazzo, D.G. Ford, C. Beard, S. Landeberger, M. Whitaker, Proliferation resistance assessment methodology for nuclear fuel cycles, Nucl. Technol. 157 (2007) 143–156.
[12] J. Li, M.S. Yim, D. McNelis, Assessment the proliferation resistance of nuclear fuel cycle systems using a fuzzy logic-based barrier method, Nucl. Technol. 162 (2008) 293–307.
[13] D.S. Blair, P.E. Rexroth, G.E. Rochau, T.T. Sype, G.D. Wyss, A Risk-based Methodology for Nuclear Proliferation Decisions, Sandia National Laboratory, 2002. SAND2002–1579C.
[14] C.G. Bathke, B.B. Ebbinghaus, B.A. Collins, B.W. Sleaford, K.R. Hase, M. Robel, R.K. Wallace, K.S. Bradley, J.R. Ireland, G.D. Jarvinen, M.W. Johnso, A.W. Prichard, B.W. Smith, The attractiveness of materials in advanced nuclear fuel cycles for various proliferation and theft scenarios, Nucl. Technol. 179 (2012) 5–30.
[15] E. Hass, Proliferation resistance and safeguards-by-design, in: IAEA Symposium on International Safeguards, Vienna, Oct 20-24, 2014, 2014.
[16] S. Takagi, D. Kawasaki, S. Nagasaki, Y. Kuno, Proliferation resistance assessment for radioactive wastes, in: Proceedings of 16th Pacific Basin Nuclear Conference, Aomori, Japan, Oct. 13–18, 2008.
[17] Nuclear Waste Management Organization, Adaptive Phased Management. Postclosure Safety Assessment of a Used Fuel Repository in Sedimentary Rocks, NWMO TR-2013-07, 2013.
[18] Nuclear Waste Management Organization, Adaptive Phased Management. Used Fuel Repository Conceptual Design and Postclosure Safety Assessment in Crystalline Rocks, NWMO TR-2012-16, 2012.
[19] International Atomic Energy Agency, Monitoring and Surveillance of Radioactive Waste Disposal Facilities, IAEA Safety Standards for Protecting People and the Environment, Special Safety Guide, No. SSG-31, 2014.
[20] Nuclear Waste Management Organization, Project Phases, January 30, 2018. https://www.nwmo.ca/en/A-Safe-Approach/About-the-Project/Project-Phases.
[21] Nuclear Energy Agency, Reversibility of Decisions and Retrievability of Radioactive Waste, Considerations for National Geological Disposal Programmes, OECD, 2012.
[22] Japan Atomic Energy Agency, H12: Project to Establish the Scientific and Technical Basis for HLW Disposal in Japan, Supporting Report 3: Safety Assessment of the Geological Disposal System, the Japan Nuclear Cycle Development Institute Technical Note, JNC TN 1410 2000-004, Tokyo, Japan, 2000 (in Japanese).
[23] World Nuclear Association, Thorium, January 30, 2018. http://www.world-nuclear.org/information-library/current-and-future-generation/thorium.aspx.
[24] S. Nakayama, J. Ahn, T. Ikeda, T. Ohe, M. Kawanishi, M. Tsukamoto, H. Kimura, M. Munakata, Interlaboratory modelling for performance assessment of engineered barriers in high-level radioactive waste disposal, J. Atom. Ener. Soc. Jpn. 34 (1992) 342–364 (in Japanese).
[25] International Atomic Energy Agency, Proliferation Resistance Fundamentals for Future Nuclear Energy Systems, IAEA Department of Safeguards, STR-332, 2002.

[26] R.L. Keeney, H. Raiffa, Decisions with Multiple Objectives, New York, Wiley Press, 1976.
[27] R.L. Keeney, H. Raiffa, Decisions with Multiple Objectives, Preferences and Value Tradeoffs, Cambridge University Press, Cambridge, 1993.
[28] R.A. Krakowski, A Multi-attribute Utility Approach to Generating Proliferation Risk Metrics, LA UR-96-3620, Los Alamos National Laboratory, 1996.
[29] Krakowski, Review of Approaches for Quantitative Assessment of the Risks of and Resistance to Nuclear Proliferation from the Civilian Nuclear Fuel Cycle, LA-UR-01-0169, Los Alamos National Laboratory, 2001.
[30] M. Türkmen, O.H. Zabunoglu, Use of Th and U in CANDU-6 and ACR-700 on the once- through cycle: burnup analyses, natural U requirement/saving and nuclear resource utilization, J. Nucl. Mater. 429 (2012) 263–269.
[31] Private Communications with Dr. Jeremy Whitlock, Canadian Nuclear Laboratories, and Prof. John Luxat, McMaster University, 2015.
[32] M.D. DeHart, M.A. Jessee, Triton. A Multipurpose Transport, Depletion, and Sensitivity and Uncertainty Analysis Module, Version 6.1, 2011.
[33] J.E. Horwedel, L.C. Gauld, S.M. Bowman, ORIGEN-APR. Automatic Rapid Processing for Spent Fuel Depletion, Decay, and Source Term Analysis, Version 6.1, 2011.
[34] A. SCALE, Comprehensive Modeling and Simulation Suite for Nuclear Safety Analysis and Design, Version 6.1, 2011.
[35] Japan Atomic Energy Agency, Thermodynamic, Sorption and Diffusion Databases, January 30, 2018. https://www.jaea.go.jp/04/tisou/english/database/database.html.
[36] P. Vilks, Sorption of Selected Radionuclides on Sedimentary Rocks in Saline Conditions – Literature Review, NWMO Technical Report, NWMO TR-2011-12, Toronto, Canada, 2011.
[37] K. Kurosaki, Thorium Fuel Cycle. In Text Nuclear Fuel Cycle, Reprocessing and Recycle Technology Division, Atomic Energy Society of Japan, 2015.