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INHALATION RADIOTOXICITY OF IRRADIATED THORIUM AS A HEAVY WATER REACTOR FUEL

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Prepared by
Rédigé par

Edwards Geoffrey W R - Reactor Physicist

Reviewed by
Vérifié par

Hyland Bronwyn - Reactor Physicist

Approved by
Approuvé par

Radford Darren D. - Manager, Computational Reactor Physics

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286 Plant Road
Chalk River, Ontario
Canada K0J 1J0

286, rue Plant
Chalk River (Ontario)
Canada K0J 1J0

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INHALATION RADIOTOXICITY OF IRRADIATED THORIUM AS A HEAVY WATER REACTOR FUEL

G.W.R. Edwards, N.D. Priest, R.B. Richardson

Atomic Energy of Canada Ltd., Chalk River, Ontario, K0J 1J0, Canada
edwardsg@aecl.ca, priest@aecl.ca, richardsonr@aecl.ca

The online refueling capability of Heavy Water Reactors (HWRs), and their good neutron economy, allows a relatively high amount of neutron absorption in breeding materials to occur during normal fuel irradiation. This characteristic makes HWRs uniquely suited to the extraction of energy from thorium. In Canada, the toxicity and radiological protection methods dealing with personnel exposure to natural uranium (NU) spent fuel (SF) are well-established, but the corresponding methods for irradiated thorium fuel are not well known. This study uses software to compare the activity and toxicity of irradiated thorium fuel ('thorium SF') against those of NU.

Thorium elements, contained in the inner eight elements of a heterogeneous high-burnup bundle having LEU in the outer 35 elements, achieve a similar burnup to NU SF during its residence in a reactor, and the radiotoxicity due to fission products was found to be similar. However, due to the creation of such inhalation hazards as U-232 and Th-228, the radiotoxicity of thorium SF was almost double that of NU SF after sufficient time has passed for the decay of shorter-lived fission products. Current radio-protection methods for NU SF exposure are likely inadequate to estimate the internal dose to personnel to thorium SF, and an analysis of thorium in fecal samples is recommended to assess the internal dose from exposure to this fuel.

I. INTRODUCTION

Heavy Water Moderated Reactors (HWRs) have a good neutron economy, and, as a consequence, considerable flexibility in the range of potential fuel types in addition to the normal natural uranium (NU) [1]. Thorium, a fertile material which breeds fissile material (U-233) during irradiation, is an attractive option for reactor fuel in an era of decreasing natural uranium resources, but requires that fissile material be added to a fuel assembly to support the nuclear reaction initially. Such a fissile/fertile fuel assembly may be heterogeneous (different fuels in different elements) or homogeneous. In this paper we investigate the radiological hazard of irradiated thorium as part of a heterogeneous bundle having low enrichment uranium (LEU) in the outer elements and thorium in the inner elements. Examining the hazard of irradiated pure thorium in this way provides a more portable way of applying the results to homogeneous, thorium/fissile materials which will have an additional hazard created by the irradiation of whatever fissile nuclide (typically Pu-239) is mixed with the initial fuel.

A list of all the nuclides in spent fuel (SF), such as irradiated thorium or NU, along with their associated activities, is a 'Reference Hazard'. A Reference Hazard is useful for radiological protection because the whole extent of personnel exposure can be estimated by the presence in the body of a relatively small number of radionuclides chosen to be easily measurable.

The radiological protection problems associated with exposure to irradiated fuel following its discharge from the reactor are a function of the initial composition of the fuel, power, burnup (total energy produced by fission) and cooling time (time in a spent-fuel pool) after discharge. The types and amounts of radioactive fission products produced do not change significantly with a change of the nuclide undergoing fission (here mainly U-235 and Pu-239 in NU, and U-233 in thorium). The total fission product activity and associated radiotoxicity for short cooling times is roughly proportional to the assembly power in the reactor and, for long cooling times, to the total assembly burnup. The activity and toxicity of the actinide components in the spent fuel is dominated, for fuels with a low concentration of fissile isotopes, as here, by the nature of their fertile actinide constituent and by the total fluence (time integrated neutron flux) in the fuel.

For actinides, transuranic alpha emitters are the most important with respect to radiological protection [2]. Plutonium isotopes are a normal constituent of irradiated low enriched uranium fuels, including NU. Indeed, in an HWR, more than half of the energy output of a full burnup NU fuel bundle results from the fission of Pu-239 produced during irradiation by neutron capture (and subsequent beta decays) on initial U-238 present in the fuel. However, plutonium is normally present for insufficient time during a typical HWR fuel cycle to generate significant quantities of highly radiotoxic transplutonic actinides.

The principle constituent of thorium, in thorium fuels, is Th-232. The only other thorium isotope present is Th-230, which is a daughter of U-234 and hence present at a level depending on the initial uranium to thorium ratio of the mined ore. The irradiation of Th-230 and Th-232 produces a range of radionuclides that have high radiotoxicity and are unique to thorium fuels. Little

plutonium, and even smaller quantities of transplutonic actinides, will be created by the irradiation of thorium.

In order to work safely with irradiated fuels, it is important to know the radionuclide composition of the nuclear materials to ensure that appropriate bio-monitoring procedures are in place, and that doses can be evaluated following exposure. Such data are well established for normal-burnup conventional NU fuels containing isotopes of uranium and plutonium, but, as described above, thorium fuels may include isotopes that are not normally present. Bio-monitoring and dosimetric procedures either have not yet been established, or may need to be revised, for these isotopes. For this reason, the present study evaluates the radionuclide composition of irradiated thorium as a function of cooling period in order to identify potential bioassay and dosimetry problems. The present study determines which radionuclides will present the greatest health risk following intakes, and which radionuclides can be reliably assayed to estimate total internal dose from the Reference Hazard. In addition, the adequacy of the AECL computer code to estimate internal doses following an intake, GenmodPC [3], was investigated to determine whether all required radionuclides, their emissions data, and their elemental biokinetic models are present to calculate accurate dose estimations.

In this paper, fuel Reference Hazards have been generated for NU fuel and for irradiated thorium as part of a heterogeneous (LEU, Th)O_2 bundle. Fuel code data at discharge and after 100 and 2700 days (7.4 years) of post-discharge cooling, have been used to calculate the dosimetry metric $\tilde{E}(50)$, the committed effective dose (CED, in Sv) per kg of fuel inhaled as particulates. The nuclides contributing the most to the CED were studied to determine the status of current knowledge regarding bioavailability, biokinetics, the suitability of International Commission on Radiological Protection (ICRP) recommended radionuclide models, and whether appropriate bioassay and dosimetric tools are already in place.

II. METHODS

II.A. Fuels Analyzed

A standard 37-element HWR natural uranium fuel bundle is shown Fig. 1, and its fuel parameters and some typical irradiation data are given in Table 1. The second fuel bundle studied here was a 43-element heterogeneous (LEU, Th)O_2 bundle [1] containing 35 smaller outer elements in two rings and 8 larger inner elements in two rings (Fig. 2 and Table 2). In this work, a Reference Hazard for the irradiated thorium in the inner 8 elements of the bundle in Fig. 2 is presented and compared to a Reference Hazard for the NU in Fig. 1. A more complete analysis of this second bundle, including a Reference Hazard for the LEU in the outer 35 elements has already been published elsewhere [4] (which also analyzes low-void-reactivity fuel).

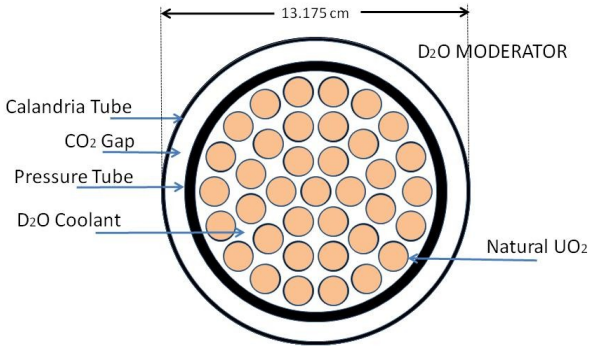


Fig.1. Natural Uranium HWR Fuel Bundle.

Table 1. Model Parameters Used for an NU Fuel Bundle

Fuel	0.71 U-235, 99.29 U-238, as 10.593 g cm^{-3} UO_2
Average Burnup	6.95 MWd kg^{-1} IHE
Irradiation Time	187.4 days
Average Power	37.1 W g^{-1}
Power at Discharge	30.7 W g^{-1}

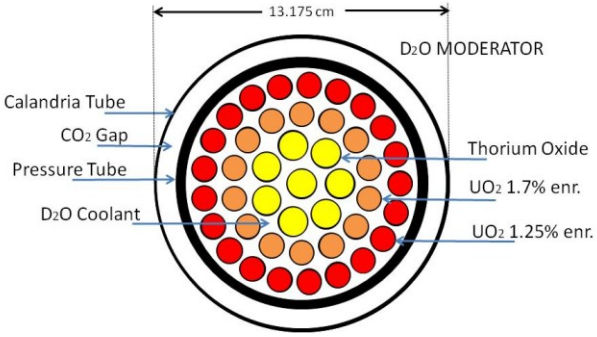


Fig. 2: High-burnup heterogeneous (LEU,Th)O_2 fuel bundle.

Table 2. Model Parameters for a high-burnup heterogenous (LEU,Th)O_2 fuel bundle

Fuel Inner	0.0025 Th-230. 99.9975 Th-232, as 9.46 g cm^{-3}, ThO_2
Fuel Intermediate	1.7 U-235, 99.29 U-238, as 10.593 g cm^{-3} UO_2
Fuel Outer	1.25 U-235, 99.29 U-238, as 10.593 g cm^{-3} UO_2
Thorium Burnup	9.11 MWd kg^{-1} IHE
LEU Burnup	19.85 MWd kg^{-1} IHE
Average Burnup	17.48 MWd kg^{-1} IHE
Irradiation Time	510 days
Average Power	24.6 W g^{-1}
Power at Discharge	25.2 W g^{-1}

Mined thorium contains some Th-230 created by U-234 decay because uranium and thorium are found in

the same ore deposits. ^{230}Th fractions of 5 ppm (0.0005%) or greater are seen where there are approximately equal amounts of uranium and thorium in the initial ore, and when the deposit is old enough for Th-230 decay to come into equilibrium with U-234 decay [5]. Ores with a significant amount of uranium may have Th-230 fractions of up to 50 ppm. The thorium in this study was assumed to have a relatively high Th-230 content of 25 ppm (0.0025% Th-230 in total thorium).

II.B. Fuel Composition Calculations

Fuel composition data were calculated using an AECL designed code 'WOBI' (WIMS-ORIGEN Burnup Integration) version 2.3.0 [6]. This code, which alternates neutron flux calculations from the 2D transport code WIMS-AECL 2.5d [7] and fuel depletion calculations in ORIGEN-S 4.4 [8] has been validated for NU assemblies in pressurized heavy water moderated reactors (PHWRs or HWRs) and for LEU assemblies in light water reactors (LWR) [6].

For each fuel bundle, an input data file describing the fuel bundle, pressure tube and calandria tube was constructed. Typical HWR values [9] for fuel temperature, coolant temperatures and coolant densities were used. The calandria tube assembly was modeled as being located inside a unit cell of heavy water with a moderator to fuel ratio equal to that of a typical HWR reactor. At the unit cell edges, partially reflective boundary conditions were imposed on the neutron flux to simulate the influence of the wider reactor environment. The neutron leakage from the unit cell was adjusted continuously as the simulated burnup progressed to keep the ratio of neutron loss and production nearly equal (as is the case globally when a reactor is running at steady power). This approach simulates the gradual shift of the bundle from being a source to a sink of excess neutrons in the reactor during aging, and slightly affects the actinide production per unit burnup. For this simulation, a steady irradiation without any downtime was assumed.

Following irradiation, the WOBI code was used to model the changes in fuel composition as a function of cooling time. For this analysis, the fuel composition at discharge (0 days), and for log-spaced cooling times afterwards (100 days, 300 days, 900 days and 2700 days), was calculated. The results for 100 and 2700 days give an approximate picture of the major processes changing the radiotoxicity in the first few years after discharge and will be reported here. Full results, including the 0, 300 and 900 day radiotoxicities, are reported elsewhere [4]. For each fuel and each cooling time, the WOBI code produced the activity of radionuclides present in the irradiated fuel. The in-reactor simulation was normalized to an axial slice of a fuel bundle containing 1 kg of initial heavy elements (any actinide, but excluding oxygen), but the final specific activities are normalized per kg of the fuel component being analyzed.

II.C. Radiotoxicity Calculations

For each fuel, and cooling time, the radioactive nuclides in the WOBI output were placed into one of three categories:

1. Fission products (and their progeny),
2. Fuel beta emitters (actinides only) and,
3. Fuel alpha emitters (actinides only).

The CED, also denoted E(50), is proportional to the excess cancer mortality risk over an assumed normal remaining lifetime of 50 years to an average healthy nuclear worker after exposure to a unit mass of fuel in the form of inhaled particulates. When normalized to exposure to a 1 kg fuel mass, it is denoted $\tilde{E}(50)$ (Sv kg^{-1}) and is calculated as:

$$\tilde{E}(50) = \sum_j \left(e_{j,inh}(50) \cdot \tilde{A}_{j,inh} \right) \qquad (4)$$

where $e_{j,inh}(50)$ (Sv Bq^{-1}) and $\tilde{A}_{j,inh}$ (Bq kg^{-1}) are respectively the dose coefficient per unit of activity intake by inhalation (ICRP, 2007) of a radionuclide j, and the activity per unit mass of fuel for the same radionuclide.

The dose coefficient of a radionuclide depends on, among other factors: its physical half life ($t_{1/2}$), the energy of the emitted radiation and the energy deposited per unit track length, the intake route, the lung and gut solubility, the residence time in radiosensitive tissues and organs, and the excretion characteristics. For nuclides retained in the body, long-lived radionuclides have more total decays over the 50 year dose integration time than short-lived radionuclides (for a given initial decay rate), increasing their relative dose coefficient.. The ICRP's unitless radiation weighting factor, proportional to the biological damage per unit energy deposited, is 20 for alpha particles and 1 for beta particles and gamma rays, and as a consequence, the inhalation dose coefficients for alpha emitters tend to be significantly higher than those of beta and gamma emitters of comparable half life. A typical alpha-emitter within an insoluble compound was observed to have an inhalation dose coefficient of the order of 10^{-6} Sv Bq^{-1}, while the longer-lived beta or gamma emitters had dose coefficients of the order of 10^{-8} Sv Bq^{-1}.

For the calculation of radiotoxicity, values of the inhalation dose coefficient were obtained principally from ICRP Publication 68 [10], under the assumption that the radionuclides were inhaled within an insoluble fuel matrix – ICRP absorption type S (slow) - as particles with an activity median aerodynamic diameter (AMAD) of 5 μm (geometric standard deviation = 2.5 μm). For radionuclides with more soluble elemental oxides (or an F (fast) or M (moderate) absorption type for all compounds

according to ICRP recommendations) this required a non-ICRP absorption type S value of the committed effective dose coefficient to be specified for the radionuclide because of the surrounding matrix material. These soluble contaminants are minor constituents of the fuel mass and it is reasonable to assume that they dissolve as the insoluble oxide fuel matrix dissolves. This reallocation of absorption type (e.g., for Am, Np, Cm, I, Cs which the ICRP classifies as absorption type F, M or vapors) is possible within the AECL GenmodPC v5 internal dosimetry code, but not within IMBA (an alternative code marketed by the Health Protection Agency, UK) and for this reason GenmodPC v5 generated inhalation dose coefficients were used for non-ICRP type S calculations. However, in these cases, only the assumed solubility of the radionuclide in the lung was changed to S type in GenmodPC internal dosimetry code and all other dosimetric parameters (including use of M or F type gut absorption factors) and values employed for the radionuclides are as recommended by the ICRP.

In this work, the Reference Hazard was first trimmed by eliminating all radionuclides not contributing at least 0.1% to the total activity within their category. $\tilde{E}(50)$ was then calculated for the remaining nuclides and the dominant contributors in each category are shown in pie graphs. Finally, the $\tilde{E}(50)$ data were tabulated, with only those radionuclides shown which were contributing at least 1% to the total $\tilde{E}(50)$ of the fuel. This last tabulation of the spectrum of radionuclides and their associated $\tilde{E}(50)s$ is termed the 'Radiotoxicity Reference Hazard', and is presented in this paper for NU and thorium SF, after 100 and 2700 days of cooling.

III. RESULTS – COMMITTED EFFECTIVE DOSE

III.A. NU Fuel

The radionuclides contributing to the committed effective dose for are shown in Fig. 3a-d and Table 3, and Fig. 4a-d and Table 4 for cooling times of 100 and 2700 days respectively. The tables show only those isotopes that were calculated to contribute ≥1% of the total $\tilde{E}(50)$.

At discharge (data not shown), the radiotoxicity from fission products is dominated by countless short-lived fission products. The radiotoxicity of fuel beta emitters is dominated by the isotope Np-239, and that the radiotoxicity of fuel alpha emitters is dominated by Cm-242. At this time, the radiotoxicity of fission products contributes more than 97% of the total CED, which is ~3.6×10^6 Sv kg^{-1}. After 100 days cooling, the shortest-lived fission products have decayed and fission products represent only 75% of the total $\tilde{E}(50)$ (Fig. 3d) at this point. The fuel beta emitter Np-239 ($t_{1/2}$ = 2.4 days), which is very active at discharge, is no longer a significant contributor. Of the important nuclides at discharge, only Cm-242 ($t_{1/2}$ = 162.8 days) continues to dominate its category. The longer half lives of the fuel beta emitter Pu-241, ($t_{1/2}$ = 14.4 years), and fuel alpha emitters (other than Cm-242) with respect to most fission products guarantee an increasingly larger share of the total $\tilde{E}(50)$ for these groups as cooling proceeds.

Between 100 (Table 3) and 2700 days (Table 4) after discharge, the total activity of the fuel drops by a factor of 40 while the total $\tilde{E}(50)$ drops by only a factor of 4. This is because $\tilde{E}(50)$ increases with the total number of expected decays occurring in the body, and this is smaller (per unit of initial activity) for short-lived isotopes.

After 2700 days (Table 4) the fission product radiotoxicity is dominated by Cs-137 ($t_{1/2}$ = 30.07 years) and ^{90}Sr-90 ($t_{1/2}$ = 28.8 years). At this time, the actinides from fuel activation now contribute 66% of the total (Fig. 4c), although in absolute terms their total dose is lower than at discharge, dropping from 2.3×10^5 Sv kg^{-1} to 2.0×10^5 Sv kg^{-1}.

The biokinetic models for all the important radionuclides of NU fuel are coded within the GenmodPC v5 internal dosimetry code. From a radiological protection point of view the monitoring of workers exposed to irradiated and aged (≥2700 days) NU fuel presents few challenges. Bioassay regimens are firmly established to monitor intakes of the hallmark radionuclides of long-term cooling, i.e., Cs-137, Sr-90, Pu-239, Pu-240, Am-241 and Cm-244 [11]. However, confirming the exposure of workers to fuel cooled for shorter periods of 100 days to 900 days could present difficulties since bioassay methods are not fully established for some significant radionuclides, including Ce-144, Pu-241 and Cm-242. Nevertheless, at these cooling times, sufficient 'indicator' radionuclides, such as Pu-239 and Pu-240, are present in the radionuclide mix to enable estimates of the total committed effective dose using bioassays for these Pu alpha emitters and knowing the full spectrum of radionuclides present (i.e., the Reference Hazard). Moreover, the detection limits for Pu-239 and Pu-240, by urine bioassay analysis using thermal ionisation mass spectrometry (TIMS) or accelerator mass spectrometry (AMS), are sufficiently low to allow the quantification of doses as low as 5% of the 20 mSv radiation worker dose limit [12] [13].

III.B. Thorium Fuel

In the (LEU,Th)O_2 assembly, the LEU and thorium oxides are separated into different fuel elements, with the inner 8 elements containing ThO_2 and the outer 35 elements containing UO_2. It is reasonable to assume that simultaneous exposures to radionuclides from both types of fuel is unlikely. Only the more novel of the two fuels, the irradiated thorium, is analyzed here.

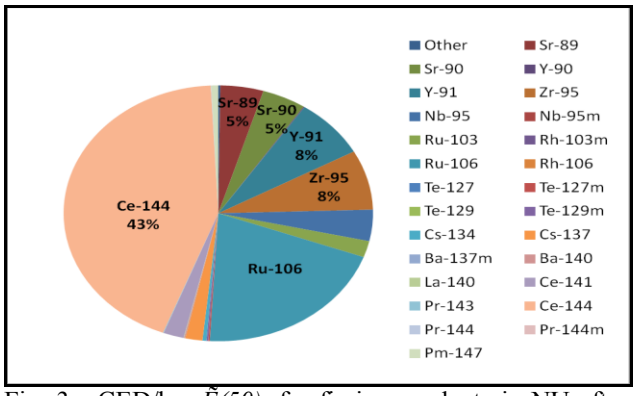


Fig. 3a. CED/kg, $\tilde{E}(50)$, for fission products in NU after 100 days cooling.

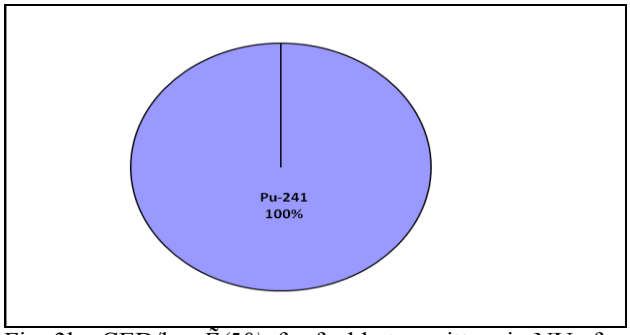


Fig. 3b. CED/kg, $\tilde{E}(50)$, for fuel beta emitters in NU after 100 days cooling.

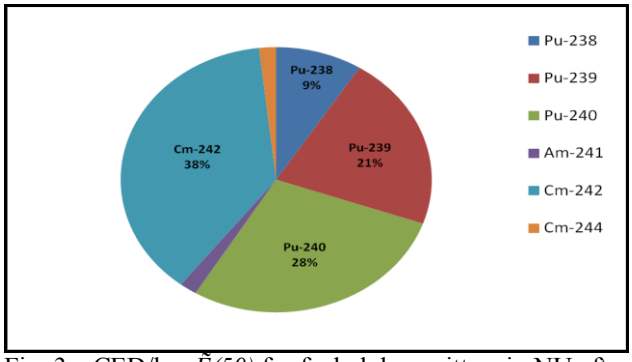


Fig. 3c. CED/kg, $\tilde{E}(50)$,for fuel alpha emitters in NU after 100 days cooling.

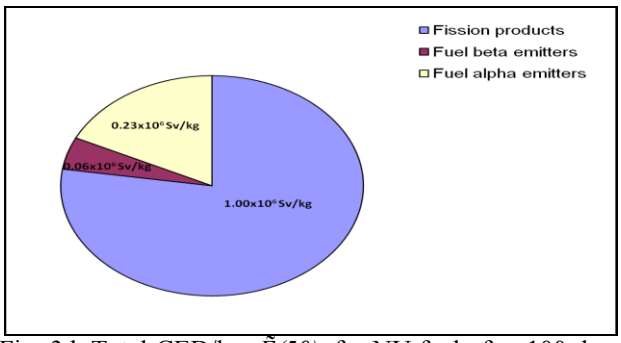


Fig. 3d. Total CED/kg, $\tilde{E}(50)$, for NU fuel after 100 days cooling.

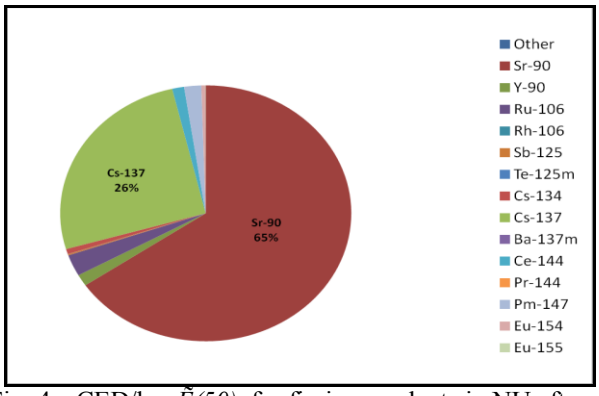


Fig. 4a. CED/kg, $\tilde{E}(50)$, for fission products in NU after 2700 days cooling.

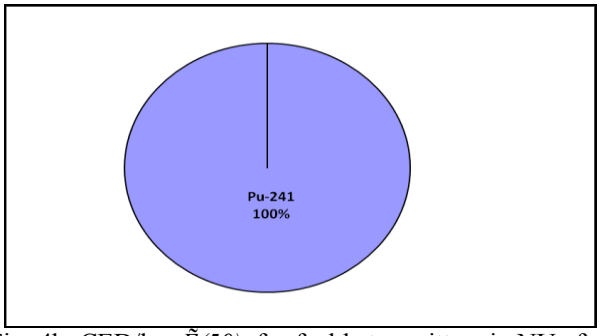


Fig. 4b. CED/kg, $\tilde{E}(50)$, for fuel beta emitters in NU after 2700 days cooling.

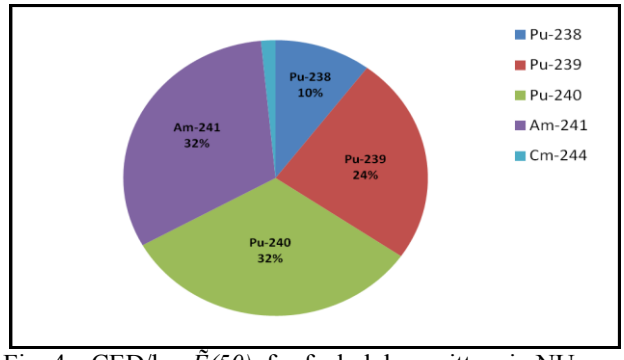


Fig. 4c. CED/kg, $\tilde{E}(50)$, for fuel alpha emitters in NU after 2700 days cooling.

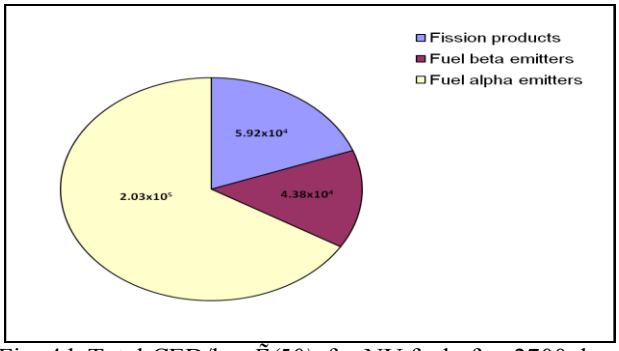


Fig. 4d. Total CED/kg, $\tilde{E}(50)$, for NU fuel after 2700 days cooling.

Table 3. Radiotoxicity Reference Hazard of 100-day-cooled NU

Nuclide	Activity (Bq kg^{-1})	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)/Total$	Category[1]
Cs-137	8.58x10^{11}	1.80x10^{4}	1.39%	f.p.
Ru-103	9.44x10^{12}	2.07x10^{4}	1.59%	f.p.
Pu-238	1.97x10^{09}	2.09x10^{4}	1.61%	fuel-α
Ce-141	6.91x10^{12}	2.15x10^{4}	1.66%	f.p.
Nb-95	3.05x10^{13}	3.98x10^{4}	3.07%	f.p.
Sr-89	8.16x10^{12}	4.54x10^{4}	3.49%	f.p.
Sr-90	6.01x10^{11}	4.62x10^{4}	3.56%	f.p.
Pu-239	5.91x10^{09}	4.94x10^{4}	3.80%	fuel-α
Pu-241	7.26x10^{11}	6.17x10^{4}	4.75%	fuel-β
Pu-240	7.83x10^{09}	6.56x10^{4}	5.05%	fuel-α
Zr-95	1.83x10^{13}	7.60x10^{4}	5.85%	f.p.
Y-91	1.25x10^{13}	7.62x10^{4}	5.86%	f.p.
Cm-242	2.17x10^{10}	8.73x10^{4}	6.72%	fuel-α
Ru-106	5.94x10^{12}	2.04x10^{5}	15.73%	f.p.
Ce-144	1.52x10^{13}	4.36x10^{5}	33.60%	f.p.
Other			2.42%	
Total	1.47x10^{14}	1.30 x10^{6}		

Table 4. Radiotoxicity Reference Hazard of 2700-day-cooled NU

Nuclide	Activity (Bq kg^{-1})	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)/Total$	Category
Cm-244	3.29x10^{08}	3.12x10^{3}	1.02%	fuel-α
Cs-137	7.28x10^{11}	1.53x10^{4}	4.99%	f.p.
Pu-238	1.96x10^{09}	2.08x10^{4}	6.81%	fuel-α
Sr-90	5.04x10^{11}	3.88x10^{4}	12.67%	f.p.
Pu-241	5.14x10^{11}	4.38x10^{4}	14.30%	fuel-α
Pu-239	5.91x10^{09}	4.94x10^{4}	16.15%	fuel-α
Am-241	7.44x10^{09}	6.41x10^{4}	20.94%	fuel-α
Pu-240	7.83x10^{09}	6.56x10^{4}	21.43%	fuel-α
Other			1.68%	
Total	3.58x10^{12}	3.06x10^{5}		

The radionuclides contributing to the committed effective dose for are shown in Fig. 5 and Table 5 for a cooling time of 100 days, and Fig. 6 and Table 6 for a cooling time of 2700 days. Table 5 and Table 6 show only those isotopes that were calculated to contribute ≥1% of the total $\tilde{E}(50)$.

The distribution of isotopes contributing to the radiotoxicity of fission products (Table 5) is broadly similar in irradiated thorium to that for NU fuel, except for the notable decline in the importance of Ru-106 which has both a very low yield from U-233 fission and a relatively high yield from Pu-239 fission. Other changes are due to differences in fission product yields (higher around mass 90 for U-233 fission in thorium fuels), fuel power (lower in the irradiated thorium than in the NU, reducing the short-lived fission product abundances and their corresponding radiotoxicity), and total fuel burnup (higher in the irradiated thorium than in the NU, increasing the long-lived fission product abundances).

Like NU fuel, fuel beta emitters in irradiated thorium, after 100 days cooling, are dominated by a single isotope. However, in irradiated thorium it is Pa-233, not Pu-241. At discharge (not shown), Pa-233 provides ~50% of both the total activity and $\tilde{E}(50)$, although its short half life guarantees that its contribution drops rapidly to 18% of $\tilde{E}(50)$ at 100 days post-irradiation (Fig. 5d and Table 5). The daughter of Pa-233 is U-233, an important contributor to the dose from fuel alpha emitters.

The landscape of fuel alpha emitters is completely different in irradiated thorium than in NU. Virtually no transuranic nuclides are present (because these would require many neutron captures, proceeding through isotopes with high fission probability such as U-233 and U-235) and $\tilde{E}(50)$ is dominated by U-232 and its daughter Th-228. The radiotoxicity of these isotopes is augmented by the presence of several short-lived gamma emitting isotopes in the decay chain under Th-228, so that every internal Th-228 decay is necessarily followed by several subsequent decays of daughter isotopes.

Overall, for a burnup of 9.11 MWd kg^{-1} IHE, and after 100 days cooling the total committed effective dose of thorium SF (1.36 Sv kg^{-1}) is comparable to that of NU SF (1.30 Sv kg^{-1}) (Table 3 and Table 5), but the fission product contribution is less, ~50% instead of ~75%, with both the fuel alpha and fuel beta components increasing to make up the difference (Fig. 5d). Between 100 and 2700 days the total activity drops by a factor of 50, slightly more than in NU SF, and the overall radiotoxicity by a factor of 2, somewhat less than in NU SF. The larger drop in activity is mostly due to the influence of the beta emitter Pa-233, formed by neutron capture on Th-232, and which has a longer half life than its counterpart Np-239 in NU fuel. This longer half life means that it still contributes significantly to fuel activity after 100 days cooling.

The slower decline of radiotoxicity (in thorium SF as opposed to NU SF) is due to the fact that, while the shorter-lived fission products are disappearing, an increasing amount of the highly radiotoxic Th-228 (and its daughters) is building in to the spent fuel during this period from the decay of U-232. After 2700 days, the radiotoxicity of the thorium SF has become twice as high of that of NU SF, mainly due to this effect (Table 4 and Table 6).

Production of U-232 in irradiated thorium can proceed in a number of ways but typically involves a fast neutron initiated (n,2n) reaction, as in Eqns. 5 and 6 here.

[1] See discussion at beginning of Section II.C.

Table 5. Radiotoxicity Reference Hazard of 100 days cooled thorium SF

Nuclide	Activity (Bq kg^{-1})	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)/Total$	Category
Ce-141	5.15x10^{12}	1.60x10^{4}	1.18%	f.p.
Cs-137	9.25x10^{11}	1.94x10^{4}	1.43%	f.p.
Ru-106	6.38x10^{11}	2.19x10^{4}	1.62%	f.p.
U-233	3.71x10^{09}	2.58x10^{4}	1.90%	fuel-α
Nb-95	2.22x10^{13}	2.90x10^{4}	2.14%	f.p.
Zr-95	1.32x10^{13}	5.48x10^{4}	4.04%	f.p.
Sr-89	1.04x10^{13}	5.76x10^{4}	4.24%	f.p.
Th-228	2.30x10^{09}	7.45x10^{4}	5.49%	fuel-α
Sr-90	9.93x10^{11}	7.63x10^{4}	5.63%	f.p.
Y-91	1.29x10^{13}	7.85x10^{4}	5.78%	f.p.
Pa-233	7.62x10^{13}	2.40x10^{5}	17.69%	fuel-β
U-232	1.01x10^{10}	2.62x10^{5}	19.29%	fuel-α
Ce-144	1.29x10^{13}	3.69x10^{5}	27.24%	f.p.
Other			2.27%	
Total	1.79x10^{14}	1.36x10^{6}		

Table 6. Radiotoxicity Reference Hazard of 2700 day cooled thorium SF

Nuclide	Activity (Bq kg^{-1})	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)/Total$	Category
Cs-137	7.85x10^{11}	1.65x10^{4}	2.44%	f.p.
Ra-224	9.07x10^{09}	2.49x10^{4}	3.69%	fuel-α
U-233	3.74x10^{09}	2.61x10^{4}	3.86%	fuel-α
Sr-90	8.34x10^{11}	6.41x10^{4}	9.50%	f.p.
U-232	9.38x10^{09}	2.44x10^{5}	36.15%	fuel-α
Th-228	9.03x10^{09}	2.92x10^{5}	43.33%	fuel-α
Other			1.04%	
Total	3.79x10^{12}	6.75x10^{5}		

Th-232 (n, 2n) Th-231 → β + Pa-231 (5a)
Pa-231 (n, γ) Pa-232 → β + U-232 (5b)

Th-232 (n, γ) Th-233 → β + Pa-233 (6a)
Pa-233 (n, 2n) Pa-232 → β + U-232 (6b1)
Pa-233 → β + U-233 (n, 2n)U-232 (6b2)

Another possibility is two thermal captures on Th-230, if it is present.

Th-230 (n, γ) Th-231 → β + Pa-231 (7a)
Pa-231(n, γ) Pa-232 → β + U-232 (7b)

Formation of Pa-231 via Eqn. 7a and 7b is normally quite small, relative to its overall formation rate, because Th-230 is present in the fuel in only trace quantities. Repeating the calculations of this work with the initial Th-230 reduced from 25 ppm to 0 ppm had the effect of reducing U-232 in the spent fuel by 10%. Since U-232 ($T_{½}$ = 69.8 y) and its daughter Th-228 ($T_{½}$ = 1.9 y), dominate the $\tilde{E}(50)$ for long cooling times (summing to ~80% of the $\tilde{E}(50)$ after 2700 days cooling, as can be seen in Table 6), a ±25 ppm uncertainty in Th-230 fraction in the initial fuel translates approximately into a ±10% uncertainty in fuel alpha emitter radiotoxicity.

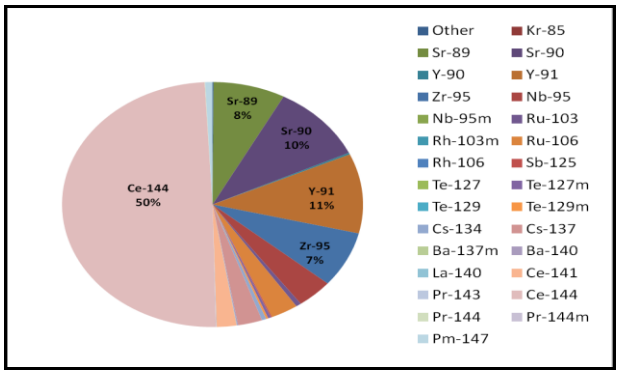
The presence of different radionuclides within thorium fuels, compared to NU fuels, presents significant difficulties to bioassay analysis for radiological protection purposes. A significant challenge is that both Th-228 and U-232 are released slowly from an insoluble thorium oxide matrix in the lungs. Therefore, the activity excreted in a 24-hour urine sample would be insufficient by current bioassay methods employed for NU fuels to determine an intake equivalent to a CED of 1 mSv. Instead, bioassay would have to proceed using fecal analyses, where the intake estimate is highly sensitive to the collection time allocated since exposure, which can only be partially mitigated by a series of frequent sample analyses.

IV. RESULTS – RADIOLOGICAL PROTECTION

IV.A. Overall radiotoxicity of fuels

At discharge, and up to 100 days of cooling, the overall radiotoxicity of both NU and irradiated thorium (although less so than NU) is dominated by short-lived fission products whose abundances are proportional to the assembly power. Thus NU, which exits the reactor at a power of 30.7 W g^{-1} (Table 1) has a higher fission product radiotoxicity, 1.00x10^{6} Sv kg^{-1}, than irradiated thorium, which exits the reactor at a power of 25.2 W g^{-1} and has a fission product radiotoxicity of 7.46x10^{5} Sv kg^{-1}. However, the high radiotoxicity of the beta emitter Pa-233, and the alpha emitters U-232 and Th-228, more than make up the difference at 100 days, making irradiated thorium slightly more radiotoxic than natural uranium at this point.

Both the radiotoxicity of NU and irradiated thorium gradually switch from being dominated by fission products to being dominated by fuel alpha emitters as cooling progresses to 2700 days. However, in irradiated thorium the radiotoxicity of fuel alpha emitters grows substantially because Th-228, a highly radiotoxic isotope because it stands at the top of a long decay chain containing many short-lived isotopes, is still coming into equilibrium with U-232 decay over this period. In contrast, the radiotoxicity in fuel alpha emitters in NU fuel declines somewhat as Cm-242 decays during this period. After 2700 days cooling, radiotoxicity in both fuels is dominated by the long-lived fission products Sr-90 and Cs-137, and by fuel alpha emitters. The higher burnup of the irradiated thorium and the high radiotoxicity of the nuclides in the U-232/Th-228 decay.



Fig, 5a. CED/kg, $\tilde{E}(50)$, for fission products in irradiated thorium after 100 days cooling.

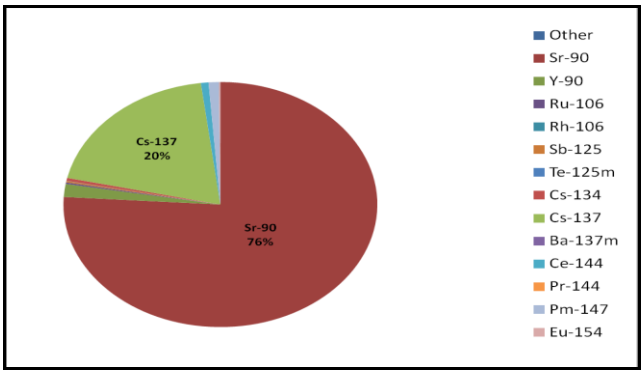


Fig. 6a: CED/kg, $\tilde{E}(50)$, for fission products in irradiated thorium after 2700 days cooling.

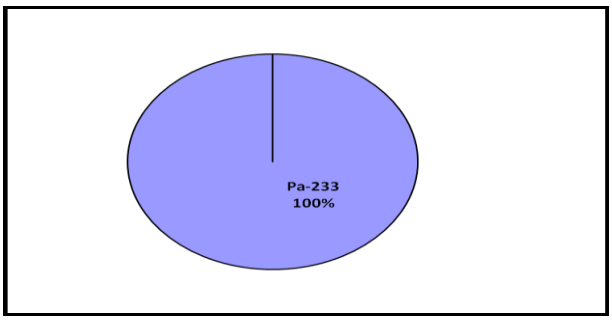


Fig. 5b. CED/kg, $\tilde{E}(50)$, for fuel beta emitters in irradiated thorium after 100 days cooling.

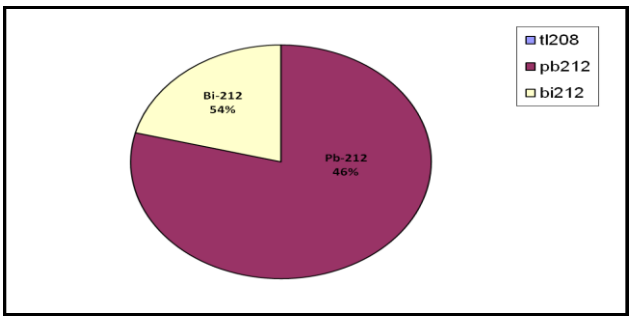
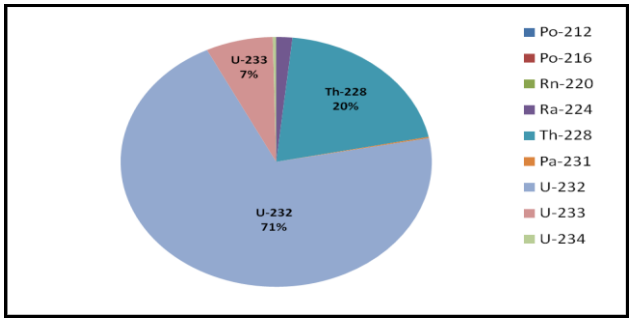


Fig. 6b. CED/kg, $\tilde{E}(50)$, for fuel beta emitters in irradiated thorium after 2700 days cooling.



Fig, 5c, CED/kg, $\tilde{E}(50)$, for fuel alpha emitters in irradiated thorium fuel after 100 days cooling.

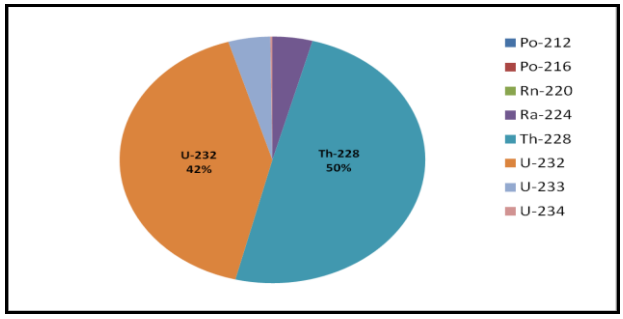


Fig. 6c. CED/kg, $\tilde{E}(50)$, for fuel alpha emitters in irradiated thorium after 2700 days cooling.

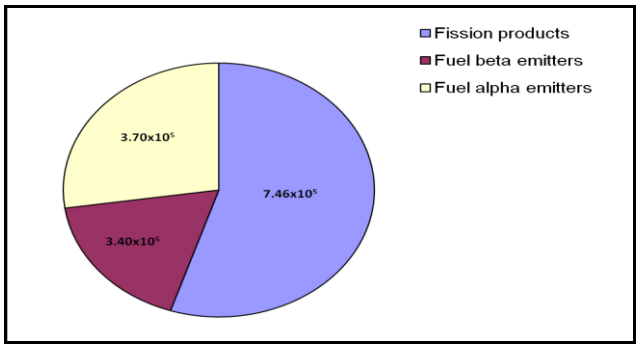


Fig. 5d. Total CED/kg, $\tilde{E}(50)$, of irradiated thorium fuel after 100 days cooling

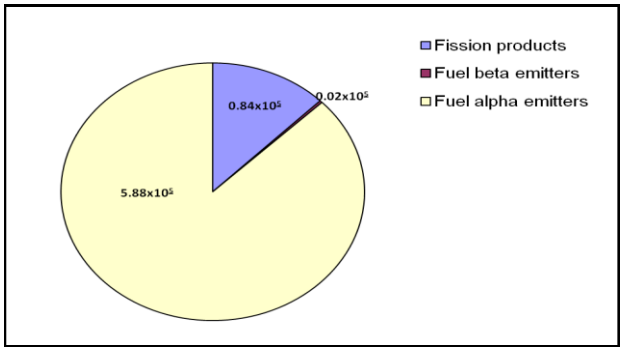


Fig. 6d. Total CED/kg, $\tilde{E}(50)$, of irradiated thorium fuel after 2700 days cooling.

chain, make the radiotoxicity of irradiated thorium more than double that of NU by this point

IV.B. Bioavailability and biokinetics of radionuclides

Type S solubility is a worst-case assumption for the inhalation dose coefficient of radionuclides such as: Ce-144, Th-228, U-232 and Pa-233, but not for Pu-238, or Pu-239, consistent with the ICRP type S classification for uranium dioxide and thorium dioxide. In practice, it is highly likely that the in-vivo solubility bioavailability of the fuel matrices concerned will vary not only as a function of chemical species (uranium dioxide, plutonium dioxide, thorium dioxide), but also with the fuel burnup. It is known that not only can irradiation of fuel affect its solubility [14] [15], but also that the porosity of the oxide particles may increase – giving them a larger effective specific surface. However, although physical and chemical data are available, to date no biological data are available on the bioavailability[2] of the irradiated fuels considered here. In contrast, the biokinetics/metabolism of many of the important radionuclides (e.g., Sr-90, Cs-137, Ra-224, U-isotopes, Pu-isotopes, and Am-241) is well established and has been incorporated into appropriate ICRP dosimetric models [16]. For some radionuclides, particularly those produced by thorium fuels (e.g., Pa-233 and Th-228) and for cerium and curium isotopes, much less information is available on their biological behaviour, and the ICRP dosimetric models recommended for these isotopes are less firmly based.

IV.C. Dosimetry Code

Comparison of the GenmodPC radionuclide library with the Reference Hazard produced for the NU fuel considered in this report indicated no important omissions for periods of cooling of ≥100 days. For the fuels at discharge, missing radionuclides include: fission products Kr-85, Nb-95m and Pr-144m (present in both NU and thorium) and thorium decay chain products: Rn-220, Po-216, Po-212 and Tl-208 (present mainly in irradiated thorium). These radionuclides are of minor dosimetric significance and in all cases were calculated to account for 0.2% or less of the total radionuclide activity in the fuel. Moreover, since the basic metabolic data for the elements of these missing radionuclides are presently coded within GenmodPC v5, it has been a relatively trivial task to modify files to include the emission data for the missing radionuclides.

IV.C. Implications for Bioassay

Currently within Canada the most commonly used method to identify and quantify CED following intakes of irradiated fuel is either a semi-annual or annual urine bioassay to quantify ^{239}Pu intake, followed by dose attribution for the remainder spectrum of radionuclides using a work-specific Reference Hazard. This is possible because the thermal imaging mass spectrometry (TIMS) detection limit for Pu-239 (~2 μBq) is low and because the fraction of Pu-239 in the Reference Hazard for normal NU fuels is high for all times of cooling and for all normal fuel burnups. Without TIMS (or accelerator mass spectrometry, which has similar detection limits) the minimum $\tilde{E}(50)$ that could be attributed to intakes of NU fuels using urine bioassay analysis would be greater than 5% of the annual worker 20 mSv dose limit [17] required by the Canadian regulator for the internal dosimetry of workers. An alternative method of analysis, alpha-spectrometry, has detection limits for Pu-239 that are 500 to 1000 times higher than those achieved with TIMS. While the use of urine bioassay for Pu-239 is adequate for NU fuel, the results of this study show that the methods currently employed need to be upgraded for thorium fuels which will contain inadequate quantities of Pu-239 to make this method tenable. In the absence of TIMS analytical methods for alpha emitters in fecal samples, alpha-spectrometry may be used for dose attribution, but the monitoring period should be monthly rather than semi-annually. Thorium oxide fuels that contain no higher actinides present a significant dosimetric challenge, as intakes that result in a committed effective dose of 1 mSv result in insufficient excretion of both Th-228 and U-232 to be adequately detected by current urinalysis methods. Exposures to thorium fuels require a bioassay regimen that includes the analysis of fecal samples that are collected monthly.

V. CONCLUSIONS

The radionuclide contents of a typical NU spent fuel assembly, and of the pure thorium elements of an advanced HWR fuel, were simulated and Radiotoxicity Reference Hazards for 100 days and 2700 days of cooling have been generated. After 100 days cooling, both fuels have similar levels of radiotoxicity, ~1.3×10^6 Sv kg^{-1}, but between 100 and 2700 days, the irradiated thorium radiotoxicity drops by only a factor of two, half as much as that of the NU. The overall drop in radiotoxicity in both fuels is due to the decay of fission products, and the lesser drop in irradiated thorium is mainly due to increase in the radiotoxicity of the U-232/Th-228 decay chain over this period. In contrast, in the NU, the initially important Cm-242 decays to insignificance, reducing the overall radiotoxicity of alpha emitters.

While the bioavailability and biokinetics of uranium fuels are well understood, less is known about thorium fuels. In particular, thorium oxide fuels may be much less

[2] Bioavailability references the uptake by particular tissues after exposure of the body to a radionuclide.

biosoluble than other HWR fuels, resulting in higher inhalation (and lower ingestion) radiation doses per unit activity of intake. More work is required to understand the biology of thorium and its compounds. Until this is done, the higher radiotoxicity in thorium fuels will present significant challenges to health physics personnel and will require an increase in physical barriers between fuel and workers as well as modifications to current bioassay practices to prevent unacceptably high exposures. In particular, standard biomonitoring for Pu-239, which is suitable to monitor worker intakes of NU fuel, will be completely unsuitable for irradiated thorium. This material may require a monthly fecal monitoring regimen for excreted U-232/Th-228.

The computer code GenmodPC v5 contains sufficient radionuclide data to calculate intake doses for both NU and irradiated thorium (with a few minor omissions relating to radionuclides that contribute <0.2% to the total activity).

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REFERENCES

1. Boczar P, Hyland B, Bradley K and Kuran S. "Achieving resource sustainability in China through the thorium fuel cycle in the CANDU reactor." *Proceedings of the 18th international conference on nuclear engineering*. Xi'an, China. 2010.
2. Lochard J, Richardson RB, Lange F, Sundararajan AR, Nomura Y. "Safety, health and environmental implications of the different fuel cycles." *Key Issue Paper No. 4 of Working Group 4 of the International Symposium on Nuclear Fuel Cycle and Reactor Strategy: Adjusting to New Realities.* International Atomic Energy Agency. Vienna, Austria. 2006.
3. Richardson RB, Dunford D. "Incorporation of current ICRP recommendations in the Genmod internal dosimetry code". Radiat Prot Dos **79**:375; 1998.
4. Priest ND, Richardson RB, Edwards GWR, "Radiotoxicity of irradiated advanced heavy water reactor fuels". Jour. Health Phys. **104/2**: 195-210, 2013.
5. Sims KWW, Gill JB, Dossetto A, Hoffmann DL, Lundstrom CC, Williams RW, Ball L, Tollstrup D, Turner S, Prytulak J, Glessner JJG, Standish JJ, Elliott T. "An inter-laboratory assessment of the thorium isotopic composition of synthetic and rock reference materials". Geostand Geoanal Res; **32**(1):65-91; 2008.
6. Edwards GWR, "Coupling of WIMS-AECL and ORIGEN-S for depletion calculations". *Proceedings of PHYSOR 2010 – advances in reactor physics to power the nuclear renaissance*. Pittsburgh, USA. 2010.
7. Irish JD and Douglas SR. "Validation of WIMS-IST". *Proceedings of the 23rd Annual CNS Conference*. Toronto, Canada. 2002.
8. Oak Ridge National Laboratory. "SCALE 4.4 modular code system for performing standardized computer analyses for licensing evaluation for workstations and personal computers". Oak Ridge National Laboratory; CC5-545; 1998.
9. IAEA, Heavy Water Reactors: Status and Projected Development, IAEA Technical Report Series No. 407, (2002).
10. International Commission on Radiological Protection. "Dose coefficients for intakes of radionuclides by workers". Oxford: Pergamon Press; ICRP Publication 68; Ann ICRP **24**:4; 1994
11. Gagné A, Surette J, Kramer-Tremblay S, Dai X, Didychuk C, Larivière D. "A bioassay method for americium and curium in feces". J Radioanal Nucl Chem; 2012.
12. Elliot NL, Bickel GA, Linauskas SH, Paterson LM. "Determination of femtogram quantities of ^{239}Pu and ^{240}Pu in bioassay samples by thermal ionization mass spectrometry". J Radioanal Nucl Chem 267:637-650, 2006.
13. Dai X, Christl M, Kramer-Tremblay S, Synal H-A. "Ultra-trace determination of plutonium in urine samples using a compact accelerator mass spectrometry system operating at 300 kV". J Anal Atom Spectrom, 27(1): 126-130, 2012.
14. Roth O, Jonsson M. "On the impact of reactive solutes on radiation induced oxidative dissolution of UO_2". J Nucl Mater **385**:595–600; 2009.
15. Nilson S, Jonsson M. "H_2O_2 and radiation induced dissolution of UO_2 and SIMFUEL pellets". J Nucl Mater. **410**:89-93; 2011.
16. International Commission on Radiological Protection. "Age-dependent doses to members of the public from intake of radionuclides: Part 4 inhalation dose coefficients". International Commission on Radiological Protection Publication 71; Ann. ICRP **25**:3-4; 1995.
17. Canadian Nuclear Safety Commission. "Design guide for nuclear substance laboratories and nuclear medicine rooms". Canadian Nuclear Safety Commission; GD-52; 2010.