Toxicity of irradiated advanced heavy water reactor fuels (2)

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ABSTRACT

The good neutron economy and online refuelling capability of heavy water moderated reactors enable them to use many different fuels types such as: low enriched uranium, plutonium, or thorium, in addition to their traditional natural uranium fuel. The toxicity and radiological protection methods for these proposed fuels, unlike those for natural uranium, are not well established. This study uses software to evaluate the composition and toxicity of three irradiated advanced heavy water reactor oxide fuels as a function of post-irradiation time. All three fuels use plutonium assumed to be recovered from light water reactor (LWR) fuel via reprocessing. The first heavy water reactor fuel investigated is a homogeneous thorium-plutonium fuel designed for a once-through fuel cycle. The second fuel is a heterogeneous thorium-plutonium-^{233}U bundle, with graded enrichments of ^{233}U in different parts of a single

fuel assembly, assumed to be part of a recycling fuel cycle in which ^{233}U from previous cycles is recovered. The third fuel is one in which plutonium and ^{241}Am is mixed with natural uranium. Each of these fuels turns out to be considerably more radiotoxic, for several years after reactor shutdown, than standard natural uranium reactor fuel would be, and it is shown that initial plutonium content is the most important factor affecting final radiotoxicity. For natural uranium, the isotope ^{239}Pu is a significant contributor to the internal dose from exposure, and evaluating its presence in urine using mass spectrometry is sufficient to estimate internal doses as low as 1 mSv – the level required by regulation. If this method is extended so that ^{240}Pu is also measured, then the combined amount of ^{239}Pu and ^{240}Pu is sufficiently high in the thorium-plutonium fuel that internal exposure to this fuel can be monitored using this method, but the fraction of these isotopes in the other two fuels is sufficiently low that they would remain below the detection limit. Thus, new techniques such as faecal measurements of ^{239}Pu (or other alpha emitters) will be required for these fuels.

1. INTRODUCTION

Heavy water reactors (HWR)s have a good neutron economy, and, as a consequence, considerable flexibility in the range of potential fuel types (Whitlock, 2000). In addition to the normal natural uranium (as UO_2) fuel, HWRs can potentially use many other oxide fuels*. In general, a reactor fuel will contain fertile constituents such as ^{238}U or ^{232}Th, together with a fissile component, which can be a combination of ^{235}U, ^{233}U or plutonium isotopes. Each of these fuels produces unique radiological protection problems if workers are exposed to the irradiated fuel following its discharge from the reactor. These problems are both a function of the radionuclide inventory of the fuel and of the cooling time (time in reactor spent-fuel pool) after discharge from the reactor. In general, for fuels of similar burnup†, the radioactive fission products produced during irradiation are broadly similar for all the fissile fuel components of potential use as HWR fuels (principally: ^{239}Pu, ^{241}Pu, ^{233}U, ^{235}U), and do not serve to greatly differentiate them. However the activity of individual fission products can increase significantly for higher burnup fuels because long-lived products build up throughout irradiation. In addition, the composition of irradiated fuels may be expected to vary depending on the nature of their fertile actinoid constituent.

Plutonium isotopes are a normal constituent of irradiated fuels containing the fertile component ^{238}U. Indeed, more than half of the energy output of a full burnup (7.5 MWd kg^{-1} of initial heavy elements) natural uranium fuel bundle results from the fission of ^{239}Pu that is produced during irradiation by neutron capture (and subsequent beta decays) on ^{238}U. However, plutonium is normally present for insufficient time during a typical HWR fuel cycle to generate significant quantities of transplutonic actinoids. If plutonium is either added to the initial fuel charge (creating a so-called Mixed Oxide or MOX fuel), or if NU or enriched uranium is subjected to a longer irradiation time, the production of other actinoids from neutron captures (possibly with later radioactive decay) on plutonium and uranium will increase. Of the radionuclides produced in fuels containing initial plutonium (or in high burnup uranium) fuels, alpha emitters are the most important with respect to radiological protection (Lochard et al., 1997).

In many countries, India and China being well known examples, supplies of thorium are more abundant than those of uranium. The principle constituent of thorium is ^{232}Th. The only other thorium isotope present is ^{230}Th, which is a daughter of ^{234}U and hence present at a level

depending on the initial uranium to thorium ratio of the ore. ^{232}Th is not fissile and cannot support a neutron chain reaction by itself, but it is fertile and under neutron irradiation produces the fissile nuclide ^{233}U. In this way, the presence of thorium in reactor fuels supports nuclear fission at a high rate even as the initial fissile constituents added to the fuel are depleted (IAEA 2002). However, in addition to ^{233}U, the neutron irradiation of ^{230}Th and ^{232}Th produces a range of highly radiotoxic isotopes unique to thorium-based fuels.

In order to work safely with irradiated fuels, it is important to know the radionuclide composition of the nuclear materials to ensure appropriate bio-monitoring procedures are in situ and to allow doses to be evaluated following exposure. Such data are well established for normal-burnup conventional nuclear fuels containing isotopes of uranium and plutonium. As described above, new fuels may include isotopes that are not normally present in conventional NU fuel. Consequently, bio-monitoring and dosimetric procedures have not yet been established for personnel working with some of the new or proposed fuels. In some cases bio-monitoring and dosimetry procedures may have to be developed. In other cases, e.g. for thorium, older procedures developed by AECL for experimental fuels may need to be revised. For this reason, the present study evaluates the radionuclide compositions of novel fuels, as a function of burnup and cooling period, in order to identify potential bioassay and dosimetry problems. Relevant questions are: Do we have sufficient information on the composition of novel fuels to estimate their bioavailability following exposure? Do we have sufficient information on the new fuels to generate a comprehensive 'Reference Hazard' list of the radionuclide activities depending on burnup and cooling? Which radionuclides will present the greatest health risk following intakes, and do we have the technologies to assay radionuclides for internal dosimetry? Do the computer codes used to estimate internal doses following an intake (e.g., GenmodPC developed by Richardson and Dunford, 1998) have all the radionuclides, their emissions data, and their elemental biokinetic models required to allow dose estimations?

Fuel code data at discharge and for different periods of post-discharge cooling have been used to list Reference Hazard radionuclides for two Th-Pu MOX oxide fuels (LWR plutonium mixed with thorium or plutonium + ^{233}U mixed with thorium) and one U-Pu MOX oxide fuel (LWR plutonium and ^{241}Am mixed with natural uranium), and to calculate the dosimetry metric 'committed effective dose' ($\tilde{E}(50)$) per kilogram[‡] of fuel inhaled as particulate material. The radionuclides present in each fuel component 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 if required.

This work follows on from that of a previous paper (Priest et al., 2013) in which similar calculations were done for: normal-burnup NU, dysprosium-poisoned uranium, pure thorium and low enrichment uranium (LEU) fuels.

2. METHODS

2.1 Fuels Analyzed

A description of the two Th-Pu MOX fuel bundles used in this analysis is supplied in Table 1 and Table 2. The first fuel described, in Table 1, is a Th-Pu MOX fuel designed for once-through operation. In such a fuel cycle, ^{233}U builds in to the fuel from neutron capture on ^{232}Th as the plutonium is depleted, increasing the total energy which can be extracted from the bundle. Such a fuel is one of the simpler types of fuel being considered, assuming a source of plutonium is available, as a way of extracting energy from thorium. Table 2 describes the second fuel material, Th-Pu-^{233}U in which some of the plutonium in the initial fuel composition is replaced by ^{233}U (which would have to be extracted from irradiated thorium). A fuel of this type would decrease the requirement for an external source of plutonium. Both Th-Pu bundles are heterogeneous, with poisoned central pins (to increase the reactivity safety margin on accidental coolant voiding). In addition, the Th-Pu-^{233}U is located in a fuel bundle having radially graded enrichments of ^{233}U and plutonium in its fuel elements. However, for this analysis, only exposure to an average of the irradiated fuel material in this bundle is considered.

The third fuel analysed here is a U-Pu-^{241}Am MOX one in which plutonium and ^{241}Am from an LWR is mixed with natural uranium, allowing more energy to be extracted from LWR fuel. ^{241}Am is left in because it often represents a problem for the ultimate disposal of nuclear fuel due to its high decay heat, and it is therefore convenient to remove it from the LWR repository waste stream when possible. The fuel for this bundle is described in Table 3.

2.2 Fuel code calculations

Fuel code calculations were undertaken as previously described (Priest et al., 2013). Fuel data were calculated using an AECL designed code ‘WOBI’ (WIMS-ORIGEN Burnup Integration) version 2.3.0 (Edwards, 2010). This code, which alternates neutron flux calculations from the 2D transport code WIMS-AECL 2.5d (Irish and Douglas, 2002) and fuel depletion calculations in ORIGEN-S 4.4 (ORNL, 1998) has been validated for NU assemblies in HWRs and LEU assemblies in LWRs (Edwards, 2010).

For each fuel bundle, an input data file describing the fuel bundle, pressure tube and calandria tube was constructed. Inside the calandria tube, fuel temperature, coolant temperatures and coolant densities were set at typical HWR reactor values. The calandria tube assembly was

modelled as being located inside a unit cell of heavy water with a moderator to fuel ratio equal to that of an HWR reactor fuelled with NU. 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.

The bundle configurations and power histories of the two Th-Pu MOX fuels and the U-Pu-Am MOX fuel are given in Table 1 to Table 3. For each bundle, the selected burnup and days of (continuous) irradiation given correspond to the upper end of their expected operating life. Final fuel compositions are affected by the bundle power at discharge (for cooling times short compared to the total irradiation time) and total burnup (for longer cooling times) and by details of the in-reactor history of the fuel (for intermediate cooling times similar to the total irradiation time). This analysis uses a continuous-irradiation power history to full burnup which assumes no reactor downtime and a high flux throughout. This approximation is conservative for the creation of most radionuclides in the fuel[§] and allows an assessment of the relative toxicity of the different fuels to which workers may be exposed.

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. For each fuel and each time of cooling, 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 actinoid, but excluding oxygen). For heterogeneous fuels, with different enrichment in different pin rings, the final results represent an average of all fuel in the bundle.

Calculations for NU, done as a reference case to which the current calculations are compared, are not repeated here from the earlier work (Priest et al., 2013).

2.3 Initial Compositions

The initial fuel composition of the uranium used in the NU (in the U-Pu-^{241}Am MOX fuel) had a nominal 0.005% ^{234}U, 0.711% ^{235}U, 99.284% ^{238}U composition, approximating the natural abundances of isotopes uranium in ore.

^{232}Th is nearly stable, with a half life of 1.4x10^{10} years. Because the half lives of all other thorium isotopes are short on geological scales, nearly all the thorium in the world is primordial ^{232}Th. However, mined thorium contains some shorter-lived ^{230}Th, created by ^{234}U decay, because uranium and thorium tend to be 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 ^{230}Th decay to come into equilibrium with ^{234}U decay (Sims, 2008). Ores with a significant amount of uranium may have ^{230}Th fractions of ten times this value (50 ppm). The thorium in this study was assumed to have a relatively high ^{230}Th content of 25 ppm (0.0025% ^{230}Th in total thorium).

2.4 Toxicity calculations

For each fuel, the radionuclide specific activity output by WOBI (in Ci kg^{-1}) was assessed for 0 days (discharge), 100, 300, 900 and 2700 days of cooling and ranked according to the radionuclide atomic number (Z). Subsequently, the radionuclide was allocated to one of three categories:

1. Fission products and their progeny,
2. Fuel beta-emitting activation products, and
3. Fuel alpha-emitting activation products

Radiotoxicity may be measured in terms of specific committed effective dose (CED), proportional to the increase in fatal cancer risk, over an assumed remaining normal lifetime of 50 years, for an individual exposed to a unit mass of irradiated fuel. This CED is denoted $\tilde{E}(50)$ (Sv kg^{-1}) and is calculated as:

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

where $e_{j,inh}(50)$ (Sv Bq^{-1}) is the committed effective dose coefficient for activity intake of insoluble Type S by inhalation (ICRP, 2007) of a radionuclide *j*, and $\tilde{A}_{j,inh}$ is the activity per unit mass for the same radionuclide $\left(\text{Bq kg}^{-1} = \text{Ci kg}^{-1} \times 3.7\text{x}10^{10}(\text{Bq Ci}^{-1})\right)$.

The dose coefficient of a radionuclide depends on, among other factors: its physical half life, 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. Radionuclides with half lives of the same magnitude as the 50 year integration time will have little change in decay rate and hence a large number of nuclear decays occur for each initial Becquerel, increasing the 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. As a consequence of this, and other effects, the inhalation dose coefficients for alpha emitters are 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 each fuel component and cooling time, the specific activity and *Ẽ(50)* for all fission products, fuel beta-emitting and fuel alpha-emitting radionuclides contributing ≥1% of the total *Ẽ(50)*) were tabulated as the 'Reference Hazard'. The Reference Hazard Tables also include values for the total specific fuel activity and committed effective dose of all nuclides in the fuel, irrespective of whether or not a radionuclide met the '≥1% of total *Ẽ(50)*' cut. Finally, for each fuel, graphs were prepared showing changes in the total specific activity and *Ẽ(50)* as a function of fuel cooling time to 2700 days.

2.4.1 Dosimetry Code

The most commonly used internal dosimetry code for radiological protection dosimetry is IMBA, marketed by Public Health England (formerly the Health Protection Agency) in the UK. This code was first produced in 1997 to meet the needs of the UK nuclear industry – particularly British Nuclear Fuels Ltd. IMBA Professional Plus is available as a series of modules that together comprise a comprehensive dosimetry tool for radiological protection dosimetry. IMBA implements standard ICRP internal dosimetry calculations. The extent to which IMBA can be modified to change dosimetric parameters for calculation is limited because the code, does not

solve the compartmental algorithms of the elemental and systemic biokinetic models, but displays retention, excretion and dose data to best-fit curves of results derived elsewhere from the implementation of the standard ICRP models. This restricts the usefulness of IMBA when non-standard radionuclide assumptions are made or if models need to be modified.

For the present paper, calculations were made using GenmodPC v5 (Richardson and Dunford, 1998). This code implements ICRP models, solving the compartmental algorithms of the elemental and systemic biokinetics models. The dosimetric data generated are verified against published ICRP dose coefficients. Because it runs the ICRP models, any parameters within the model may be changed to meet the need for non-standard dose calculations. Comparison of the GenmodPC radionuclide library with the Reference Hazard produced for the NU fuel in our previous paper (Priest et al., 2013) indicated no important omissions for periods of cooling of ≥100 days. For the fuels at discharge, missing radionuclides include: fission products ^{85}Kr, ^{95m}Nb and ^{144m}Pr (present in all fuels), and thorium decay chain products: ^{220}Rn, ^{216}Po, ^{212}Po and ^{208}Tl (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.

2.4.2 Solubility Type

For the calculation of $\tilde{E}(50)$, values of the inhalation dose coefficient were obtained principally from ICRP Publication 68 (ICRP, 1994), assuming 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) this required a non-ICRP absorption type S value of the committed effective dose coefficient to be specified for the radionuclide. These soluble contaminants are minor in parts of the fuels' 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 ^{241}Am, which the ICRP classifies as absorption type M) is possible within the AECL GenmodPC v5 internal dosimetry code, but not within IMBA and for this reason GenmodPC v5 generated inhalation dose coefficients were used for non-ICRP type S calculations.

Type S solubility is a worst-case assumption for the inhalation dose coefficient of radionuclides such as: ^{144}Ce, ^{228}Th, ^{232}U and ^{233}Pa, but not for ^{238}Pu, or ^{239}Pu, 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 (Roth and Jonsson, 2009; Nilsson and Jonsson, 2011), 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 of the irradiated fuels considered here. In contrast, the biokinetics/metabolism of many of the important radionuclides (e.g., ^{90}Sr, ^{137}Cs, ^{224}Ra, U-isotopes, Pu-isotopes, and ^{241}Am) is well established and has been incorporated into appropriate ICRP dosimetric models (e.g. ICRP, 1995). For some radionuclides, particularly those produced by thorium fuels (e.g., ^{233}Pa and ^{228}Th) 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.

3. RESULTS - FUEL ACTIVITY AND TOXICITY

3.1 Th-Pu MOX Fuels

3.1.1 Th-Pu MOX

The radionuclides calculated to be present in the Reference Hazard for this fuel at discharge (0 days) and after cooling times of 100, 300, 900 and 2700 days are shown in Table 4 to Table 8. These tables show only those isotopes that were calculated to contribute ≥1% of the total $\tilde{E}(50)$ after the inhalation of oxide fuel particles. Fig. 1a and Fig. 1b show the changing proportion of the total specific activity and $\tilde{E}(50)$, respectively, from fission products (dominated by beta-gamma emitters), fuel beta emitters and fuel alpha emitters, from discharge to 2700 days of cooling.

3.1.1.1 General Observations

The total specific activity of the irradiated fuel falls by more than two orders of magnitude between discharge and 2700 days post-irradiation (Fig. 1a), mostly as a consequence of the loss of short-lived fission products and the fuel beta emitter ^{233}Pa (produced by thorium irradiation and having a $T_{1/2}$ of ~27 days). In contrast, the $\tilde{E}(50)$ resulting from inhalation exposure (Fig. 1b) drops only by a factor of about three, being dominated from the beginning by fuel alpha emitters, most of which have medium to long decay times.

3.1.1.2 Fission Products

At discharge, the most important fission products are ^{144}Ce and ^{106}Ru, contributing ~5% of the total specific CED. A large number of other fission products (each too small a contribution to be listed here) contribute another 5% of the CED, but available dose coefficients for short-lived fission products are rather sparse (only about 1/3rd have such data) so this value is somewhat uncertain. The dominance of ^{144}Ce and ^{106}Ru is due to their medium length half lives of ~1 year – short enough for complete decay of these isotopes in the body, but long enough that they build up to large concentrations in discharged fuel[††]. After 2700 days (~8 years) these two isotopes have undergone 8 or more half lives of decay, reducing their activities to less than 1/250th of their initial value, and neither they nor any other fission product remains important to the CED.

3.1.1.3 Fuel Beta Emitters

At discharge, there are two fuel beta emitters: ^{233}Pa and ^{241}Pu, which are important, contributing ~8.4% of the total radiotoxicity. ^{233}Pa decays away fairly rapidly and after ~100 days only ^{241}Pu remains. The decay of this relatively short-lived actinoid over the next few years gradually produces the highly radiotoxic fuel alpha emitter ^{241}Am.

3.1.1.4 Fuel Alpha Emitters

Of the actinoids at discharge, ^{242}Cm ($T½$ = 162.8 days) initially contributes the most to the $\tilde{E}(50)$ of the Th-Pu MOX fuel, but its relatively short half life compared to most of the other alpha emitters renders it insignificant after a few hundred days. Most of the drop in radiotoxicity of the fuel alpha emitters is due to the decay of this isotope. After a few hundred days, the second and third most important actinoids, ^{238}Pu and ^{244}Cm with half lives of 87.7 and 18.1 years respectively, become the dominant contributors. The disappearance of ^{242}Cm (as well as the shorter-lived fission products and ^{233}Pa) substantially decreases the total radiotoxicity of the fuel and therefore increases the relative importance of the actinoid ^{232}U ($T_{1/2}$ = 69.8 years), which appears at the 1% level after 100 days and increases in proportion thereafter. In addition, the daughter of ^{232}U, ^{228}Th, grows into the fuel over a period of 6 to 8 years until a secular equilibrium in decay rates is reached with its parent. ^{228}Th is highly radiotoxic, in large part because it stands on top of a long decay chain of relatively short-lived isotopes, and by 2700 days its contribution to the CED is larger than that of its parent.

3.1.2 Th-Pu-^{233}U MOX

The radionuclides calculated to be present in the Reference Hazard for this fuel at discharge (0 days) and after cooling times of 100, 300, 900 and 2700 days are shown in Table 9 to Table 13. These tables show only those isotopes that were calculated to contribute ≥1% of the total $\tilde{E}(50)$ after the inhalation of oxide fuel particles. Fig. 2a and Fig. 2b show the changing proportion of the total specific activity and $\tilde{E}(50)$, respectively, from fission products (dominated by beta-gamma emitters), fuel beta emitters and fuel alpha emitters, from discharge to 2700 days of cooling.

3.1.2.1 General Observations

At discharge the activities of Th-Pu MOX and Th-Pu-^{233}U MOX are quite similar, since they are dominated by short-lived fission products and the fuel beta emitter ^{233}Pa. The concentrations of

these isotopes in the spent fuel is mainly fuel power dependent and this is similar in both cases. As in Th-Pu MOX, the total specific activity of Th-Pu-^{233}U fuel falls by more than two orders of magnitude between discharge and 2700 days post-irradiation (Fig. 2a) as a consequence of the decay of the short-lived fission products and ^{233}Pa. However, in Th-Pu-^{233}U the activity at 2700 days is somewhat lower than in Th-Pu-MOX because the initial concentration of plutonium is much less (about 28%), leading to a much lower production of ^{241}Pu, a dominant contributor to the fuel decay rate at this time.

Also similar to Th-Pu MOX fuel, the decline in $\tilde{E}(50)$ is much less then the decline in activity over this time period (Fig. 2b). The drop in $\tilde{E}(50)$, close to a factor of five, is about 25% greater than for Th-Pu MOX, because the contribution of fuel alpha emitters in Th-Pu-^{233}U fuel is reduced, leaving a larger share of the CED for the short-lived fission products (which are mainly dependent on fuel power). The fuel alpha emitters are reduced because the initial Pu is much less in Th-Pu-^{233}U fuel and the ^{233}U which replaces it does not undergo neutron capture to form radiotoxic actinoids.

3.1.2.2 Fission Products

The suppression of the overall fuel actinoid contribution in this fuel leads to a number of fission products contributing more than 1% to the radiotoxicity of the fuel at discharge. Of these, only ^{106}Ru is reduced in importance relative to the Th-Pu MOX fuel, since it has a relatively low yield in the fission of ^{233}U and has a high yield in ^{239}Pu fission. By 2700 days, the only important fission product contributors are the two classic medium-long-lived fission products ^{90}Sr and ^{137}Cs.

3.1.2.3 Fuel Beta Emitters

The fuel beta contribution to $\tilde{E}(50)$ is initially dominated by ^{233}Pa, but this decays away rapidly ($T_{1/2}$ = 27 days) and after 2700 days the only important contributor is ^{241}Pu.

3.1.2.4 Fuel Alpha Emitters

The spectrum of fuel alpha emitters is quite similar to that of Th-Pu MOX fuel and these isotopes dominate the $\tilde{E}(50)$ throughout this time period, albeit with those actinoids created by Pu captures reduced by an amount similar to the reduction of initial Pu in the fuel. ^{232}U and ^{228}Th are correspondingly more important, but the actinoids whose genesis was in the initial plutonium – in particular ^{238}Pu and ^{244}Cm, remain dominant overall.

3.2 U-Pu-^{241}Am MOX

The radionuclides calculated to be present in the Reference Hazard for this fuel at discharge (0 days) and after cooling times of 100, 300, 900 and 2700 days are shown in Table 14 to Table 18. Except for ^{241}Pu, which has been added for comparison with the other fuels, tables show only those isotopes that were calculated to contribute ≥1% of the total $\tilde{E}(50)$ after the inhalation of oxide fuel particles. Fig. 3a and Fig. 3b show the changing proportion of the total specific activity and $\tilde{E}(50)$, respectively, from fission products (dominated by beta-gamma emitters), fuel beta emitters and fuel alpha emitters, from discharge to 2700 days of cooling.

3.2.1 General Observations

At discharge, the activity of this fuel is similar to both the other fuels. Its overall activity is dominated by short-lived fission products, most of which do not contribute to $\tilde{E}(50)$, and to a lesser extent by fuel beta emitters. The most important of these beta emitters at discharge is ^{239}Np, a rather short-lived ($T_{1/2}$ = 2.356 days) actinoid isotope which does not appear in the thorium-based fuels as it is mainly produced by neutron capture on ^{238}U. The behaviour of the activity over the first 2700 days cooling (Fig. 3a) is also very similar to that of the other fuels, with a significant decline of almost three orders of magnitude due to the decay of the short-lived fission products and, to a lesser extent, ^{239}Np. (The decline in activity of ^{239}Np, which dominates the fuel beta emitters, over the first 100 days cooling, is not well captured by Fig. 3a due to a lack of intermediate points. The overall decline should be steeper over the first 10 days to match the short half life of ^{239}Np).

The overall $\tilde{E}(50)$ is dominated, to an even larger extent than the other fuels, by fuel alpha emitters. It declines somewhat less than the other fuels (just over a factor of three) during 2700 days cooling, but flattens less than the others between 900 and 2700 days during which period it continues to decline noticeably. Both these effects are due to the large ^{244}Cm contribution in this fuel. In the first 900 days, the presence of ^{244}Cm suppresses the relative effect on total CED of the decay of ^{242}Cm. However, between 900 and 2700 days, after the ^{242}Cm is gone, the decline in CED is larger in this fuel than in the others due to the decay of ^{244}Cm, which has a relatively short half life itself ($T_{1/2}$ = 18.1 years).

3.2.2 Fission Products

The medium long-lived fission products ^{144}Ce and ^{106}Ru contribute significantly to $\tilde{E}(50)$ in this fuel, as for the others. The absolute activity is slightly higher for ^{144}Ce because the longer irradiation time of this fuel (1500 days vs. 900 days for the other two) allows ^{144}Ce to approach secular equilibrium more closely. The ^{106}Ru activity is affected similarly, but this isotope also experiences a larger increase (of about 50%) because of the dominance of plutonium fissions in this fuel.

3.2.3 Fuel Beta Emitters

The activity (and toxicity) of ^{241}Pu remains important throughout the first 2700 days of decay in this fuel, as for the others. The other important beta emitter, ^{239}Np, is quite short-lived and contributes only to the discharge toxicity. In this fuel, unlike the two thorium-based ones, there are no beta emitters like ^{233}Pa with an intermediate-length half life ($T_{1/2}$ = 27 days) which contribute significantly to the toxicity over the first 100 to 300 days.

3.2.4 Fuel Alpha Emitters

The principle difference between the toxicity of this irradiated fuel and that of the thorium-based fuels is that this fuel has a large ^{244}Cm component. This nuclide, because it requires several captures on the initial plutonium, is produced non-linearly with time and its appearance here in large quantities is due to the significantly longer irradiation time of this fuel. Another apparent difference is the relatively small amount of ^{238}Pu in this fuel, although this isotope remains important to the overall toxicity. There is a strong flux dependence in the production of ^{238}Pu, since one of the principle production pathways is the creation of ^{242}Cm and its subsequent decay. In high fluxes, the probability that the ^{242}Cm will capture a neutron before decaying is increased. The constant-power history chosen for this fuel in this study results in relatively large fluxes as the fissile plutonium depletes, increasing the ^{244}Cm contribution and suppressing that of ^{238}Pu.

3.3 COMPARISON OF TOXICITY TO NATURAL URANIUM

Fig. 4a and Fig. 4b show the activity and radiotoxicity of natural uranium (Priest et al., 2013) over the first 2700 days, and the overall radiotoxicity from Fig. 4b is compared against that of the other fuels in Fig. 5. It is apparent that the advanced HWR fuels will present considerably greater radiotoxicity problems. Unlike the advanced fuels, few highly radiotoxic actinoids are produced in natural uranium fuel, explaining the relatively low $\tilde{E}(50)$. This is because the NU

irradiation time (of about 280 days) is insufficient to built up large amounts of ^{242}Cm, ^{244}Cm or ^{238}Pu, while the absence of thorium in the fuel means that there is no contribution from ^{232}U, ^{228}Th and its decay chain either. The radiotoxicity of irradiated natural uranium fuel at early times is therefore dominated by fission products and the short-lived beta emitter ^{239}Np, the two together making up 95% of the total $\tilde{E}(50)$ at discharge. The decay of these fission products results in the more dramatic fall-off in radiotoxicity seen in NU fuel than in the other fuels. In contrast, the radiotoxicity of the three advanced HWR fuels are all dominated by actinoids produced by neutron capture in the fuel. The decline of this radiotoxicity is less over this period, and dominated by the disappearance of ^{242}Cm in the 100 to 900 day decay period.

Of the two thorium-based advanced fuels, both of which have similar irradiation times, the production of curium from plutonium irradiation appears to be more important than the production of isotopes in the ^{232}U decay chain via thorium irradiation, so the difference in initial plutonium content appears to roughly explain the difference in final radiotoxicity. On the other hand, the U-Pu-^{241}Am ,while having a smaller amount of initial plutonium than the thorium-based fuels, more than makes up for the difference because of: its initial ^{241}Am content (which eliminates one capture on the way to creating curium isotopes), its long irradiation time, and the continuous production of new plutonium via neutron capture on ^{238}U.

4. RESULTS - RADIOLOGICAL PROTECTION

4.1 Natural Uranium

As previously reported (Priest et al., 2013) 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 in natural uranium i.e., ^{137}Cs, ^{90}Sr, ^{239}Pu, ^{240}Pu, ^{241}Am (Gagné et al., 2012). 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 ^{106}Ru ^{144}Ce, ^{241}Pu and ^{242}Cm. Nevertheless, at these cooling times, sufficient ‘indicator’ radionuclides, such as ^{239}Pu and ^{240}Pu, are present in the radionuclide mix to enable estimates of the total committed effective dose using Thermal Imaging Mass Spectrometry (TIMS) bioassays of urine samples for

these Pu alpha emitters and knowing the full spectrum of radionuclides present (i.e., the Reference Hazard).

4.2 Thorium-based Fuels

Unlike a pure thorium fuel irradiation (Priest et al., 2013), or thorium-^{233}U irradiation which would be qualitatively similar, the most radiotoxic irradiation products of thorium, ^{232}Th, ^{233}U, ^{228}Th are only a relatively small part (1-2% at discharge, 5-15% at 2700 days, for Th-Pu and Th-Pu-^{233}U respectively) of the Reference Hazards for both thorium-based fuels over this cooling range. As well, the most radiotoxic actinide known, ^{231}Pa, produced in the thorium-based fuels as an intermediary in the of production of ^{232}U, never achieves the 1% threshold of total dose to be included in the Reference Hazard. Thus for dose determination, methods to determine activity of thorium-derived isotopes in faeces, while desirable, may not be strictly necessary.

The overall importance of fission products is much reduced in these fuels, relative to natural uranium, because of the large increase in the radiotoxicity of actinoids created by neutron capture on plutonium. Although ^{137}Cs and ^{90}Sr remain the most important fission products at the longest decay times, ^{106}Ru and ^{144}Ce are virtually the only important fission products between 100 and 900 days. As mentioned in the previous section, bioassay methods for these nuclides are not fully established.

In the older fuels, the most important fuel alpha emitters are: ^{238}Pu, ^{241}Am, ^{242}Cm and ^{244}Cm. Of these, bioassay regimes to measure ^{241}Am and ^{244}Cm in faeces are available, but not for ^{238}Pu and ^{242}Cm. However, the biokinetics of curium are less well understood than those of plutonium and uranium and more work is required to determine the dose coefficients and pathways of curium through the body.

4.3 U-Pu-^{241}Am

This fuel had the highest $\tilde{E}(50)$ of all the fuels at discharge. Most of the conclusions in the previous discussion on thorium-base fuels remain applicable except that, unlike the thorium-based fuels, there is no contribution from the ^{232}U-^{228}Th decay series. The long irradiation time of this very high burnup fuel significantly increases both the relative and absolute ^{238}Pu and ^{244}Cm activities in the Reference Hazard, and, in particular ^{244}Cm dominates the radiotoxicity at long cooling times and is second only to ^{242}Cm at discharge. The uncertainty

of the biokinetics of curium, and the lack of bioassay techniques for ^{242}Cm, will make measuring exposure to this fuel problematic.

4.4 Internal Dose Monitoring via TIMS for ^{239}Pu and ^{240}Pu in Urine

Canadian regulation (CNSC, 2010) requires that detection techniques be available for internal doses as low as 5% of the 20 mSv worker dose limit (i.e. 1 mSv). Currently, the most commonly used method to achieve this, following intakes of irradiated fuel, is a TIMS analysis of the ^{239}Pu content in a semi-annual or annual urine sample followed b dose attribution for the remainder spectrum of radionuclides using a work-specific Reference Hazard.

The TIMS detection limit for ^{239}Pu is 0.4 fg/l (Elliot et al., 2006; Dai et al., 2012) (equivalent to ~1 μBq/l). This level of ^{239}Pu in urine corresponds to an $\tilde{E}(50)$ (from ^{239}Pu) of ~0.033 mSv (Lamont, 2005). Therefore doses as low as 1 mSv worker dose limit, from all sources, are measurable if the ^{239}Pu contribution is at least 3.3% to the total internal dose. This situation applies to natural uranium fuel cooled for 100 days or longer. Fig. 6 shows the fraction of ^{239}Pu contribution to $\tilde{E}(50)$ for the three fuels considered here and for natural uranium. The natural uranium fuel is above the 3.3% limit for 100 days cooling and longer. None of the other fuels have sufficient ^{239}Pu in their spent fuel that a 1 mSv dose could be estimated by measuring this isotope alone.

Extension of the TIMS analysis of ^{239}Pu in urine to measure ^{240}Pu in urine as well is practical and the TIMS detection limit for ^{240}Pu is similar to that for ^{239}Pu (Elliot et al., 2006; Dai et al., 2012). Fig. 7 shows the fraction of the dose contributed by the sum of these two isotopes vs. cooling time. This sum exceeds 3.3% of the $\tilde{E}(50)$, indicating that a 1 mSv dose from all nuclides could be measured by using the Reference Hazard, in NU fuel 100 days and older and also in Th-Pu fuel of any age. Th-Pu-^{233}U and U-Pu-^{241}Am fuels would still require an even more sensitive method.

An alternative method of analysis, alpha-spectrometry, has detection limits for ^{239}Pu that are 500 to 1000 times higher than those achieved with TIMS and are therefore not sensitive enough to determine CED following exposure to irradiated nuclear fuels.

5. CONCLUSIONS

The radionuclide content of three advanced HWR fuel bundles: plutonium+thorium, plutonium+thorium+^{233}U, and plutonium+uranium+^{241}Am, at typical operational burnups, were predicted at discharge after different periods of cooling. A Reference Hazard for each of these fuels at discharge and after 100 days, 300 days, 900 days and 2700 days of cooling has been generated and compared to that already generated for NU fuel (Priest et al., 2013) Examination of the results led to the following conclusions:

1. The radiotoxicity of the advanced HWR fuels investigated here was significantly higher, by approximately an order of magnitude, that that of NU. This will require more stringent radioprotection methods to limit worker exposure to these fuels.
2. The radiotoxicity of the advanced HWR fuels investigated here was dominated by actinoids derived from neutron capture on thorium or plutonium. The contribution of short-lived fission products to the radiotoxicity of the discharged fuel was ~10% for the Pu-Th and Pu-U-^{241}Am fuel and ~23% for the Th-Pu fuel. For comparison, short-lived fission products make up more than 75% of the radiotoxicity of NU fuel at discharge. The reduction in importance of fission products in the new fuels means that the radiotoxicity of the new fuels fell more slowly during the first 2700 days of cooling than NU fuel does.
3. The radiotoxicity of the thorium-based fuels was dominated by the activation products of the initial plutonium in these fuels. Actinoids from thorium capture (such as ^{232}U and ^{228}Th) were not a large component of the radiotoxicity. While detection of these nuclides (presumably in faecal samples since excretion of these nuclides in urine is negligible) would be desirable, detection methods for the products of neutron activation of plutonium (such as ^{241}Am and ^{244}Cm) should be sufficient, when paired with a Reference Hazard, to estimate $\tilde{E}(50)$.
4. The current method for tracking ^{239}Pu in urine, using TIMS, as an indication of exposure will not be sufficient for any of the new fuels, as ^{239}Pu represents too small a fraction of the overall radiotoxicity. However, extension of this method to ^{240}Pu would allow exposure to Th-Pu fuel to be monitored for fuels from discharge to 2700 days. In the other advanced fuels, the sum of ^{239}Pu and ^{240}Pu is still too small a fraction of the Reference Hazard for this method to be used at the current (~0.4 fg/l) detection limit.

Detection of these isotopes (or other alpha emitters) in the faeces will therefore be required.

5. Curium isotopes, in particular ^{242}Cm and ^{244}Cm, become much more important in fuels with an initial plutonium content – particularly high burnup fuels. There is no appropriate bioassay method available for ^{242}Cm, and the biokinetics of curium in the body is not as well understood as those of uranium and plutonium. This area will need more work.
6. GenmodPC v5 contains sufficient radionuclide data to calculate intake doses for all the fuels considered (with a few minor omissions relating to radionuclides that contribute <0.2% to the total activity). The ability to undertake the same tasks with IMBA will depend upon the IMBA Professional Plus version and modules available for use at any location.

6. ACKNOWLEDGEMENTS

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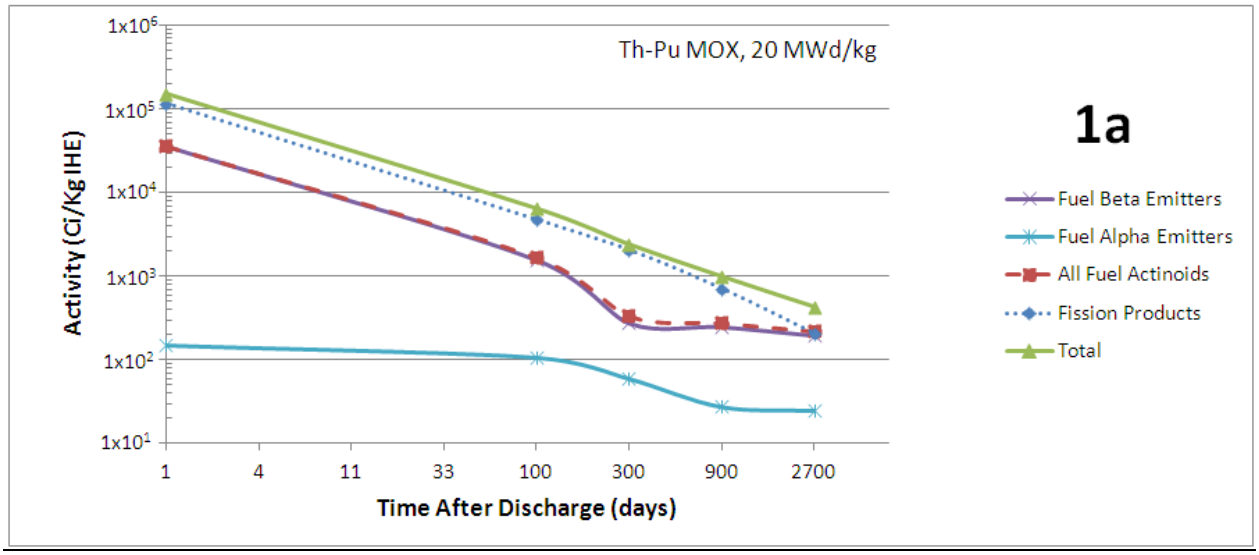
8. FIGURES



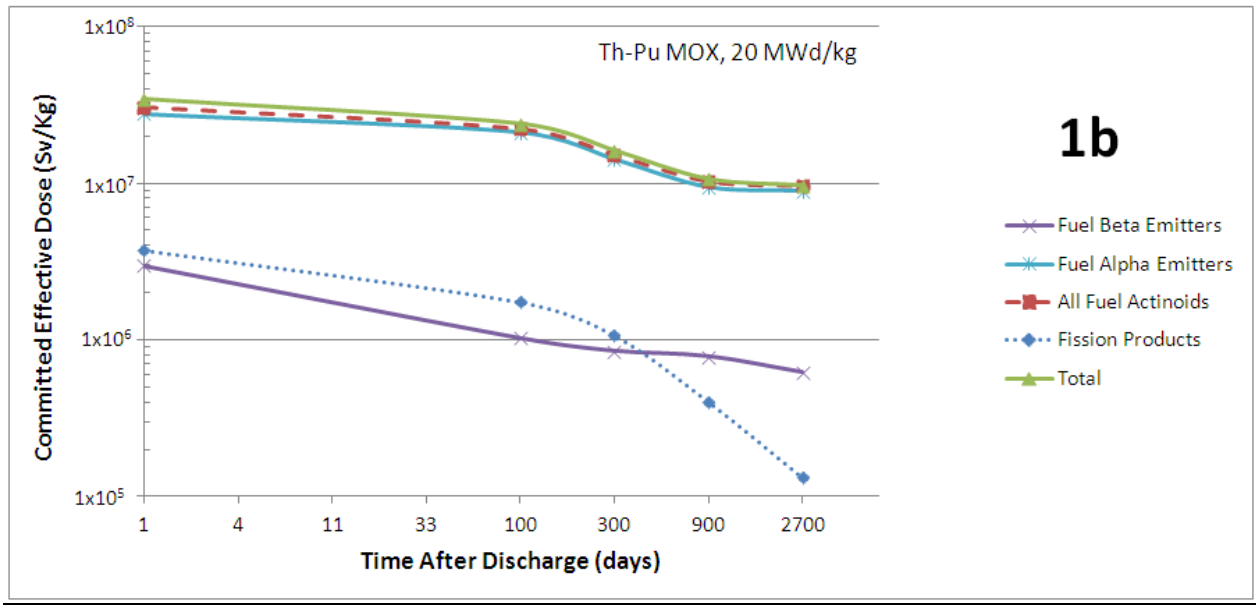
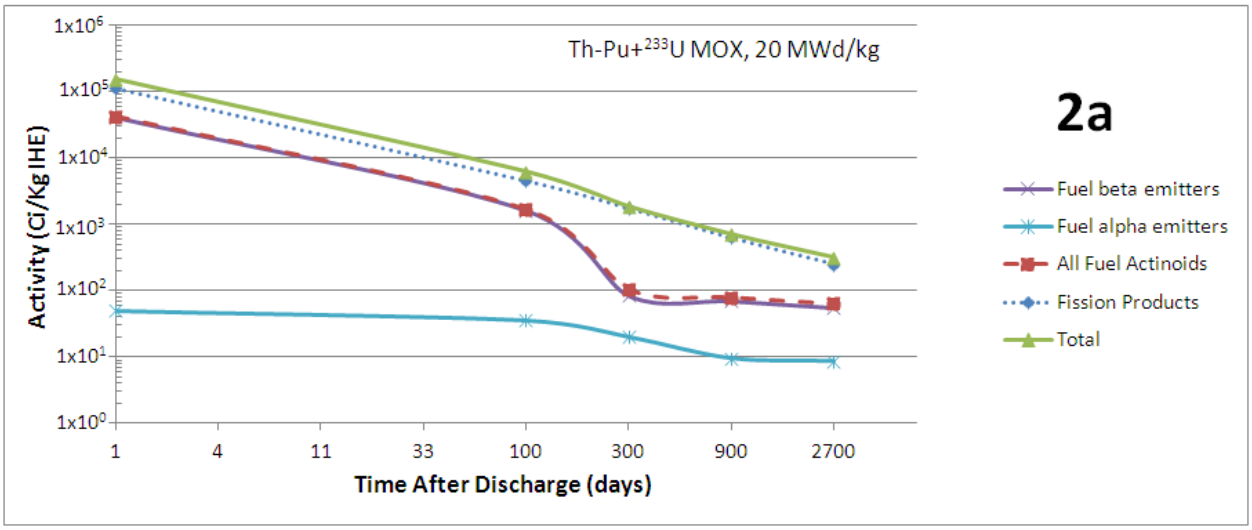


Figure 1

Average of all fuel elements from an irradiated thorium-plutonium fuel bundle. The specific activity (A) and committed effective dose (B), from the point of discharge from the reactor, to 2700 days of cooling in on-site storage.



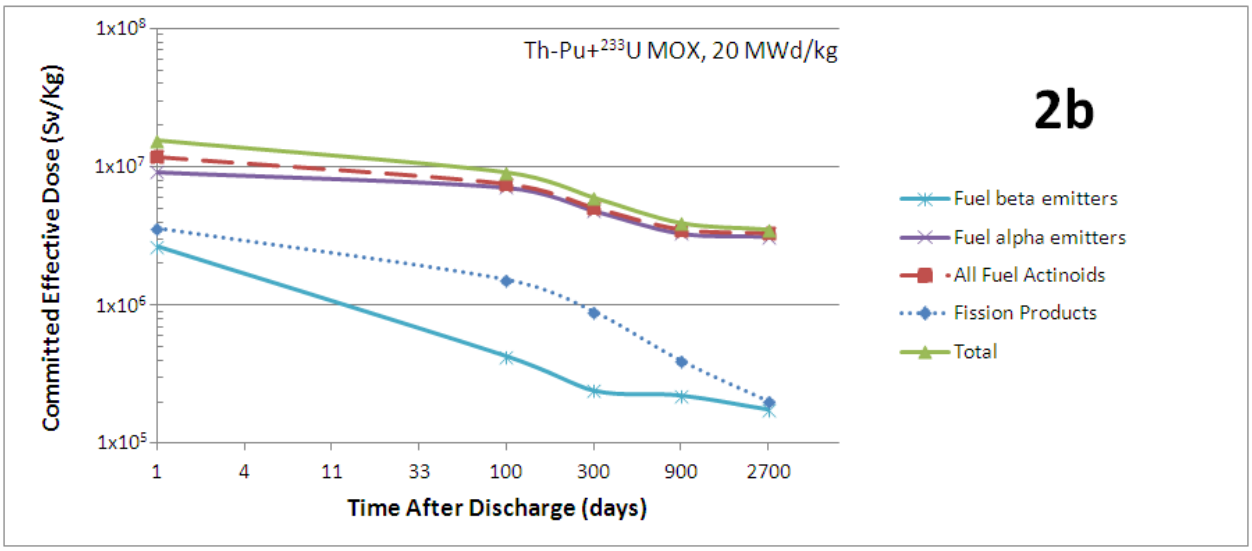


Figure 2

Average of all fuel elements from an irradiated thorium-plutonium-^{233}U fuel bundle. The specific activity (A) and committed effective dose (B), from the point of discharge from the reactor, to 2700 days of cooling in on-site storage.

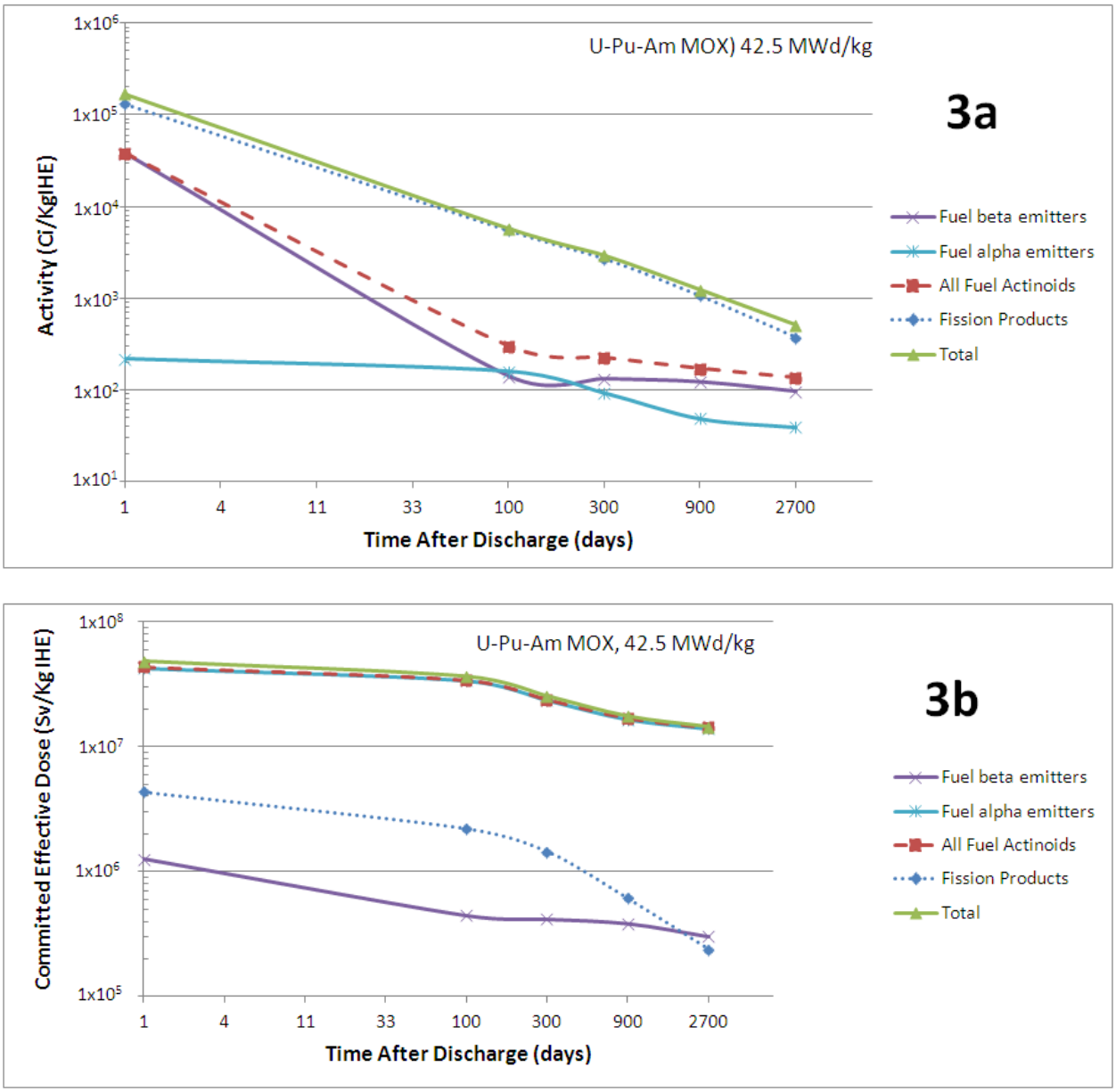


Figure 3

Average of all fuel elements from an irradiated uranium-plutonium-^{241}Am fuel bundle. The fuel specific activity (A) and committed effective dose (B), from the point of discharge from the reactor, to 2700 days of cooling in on-site storage.

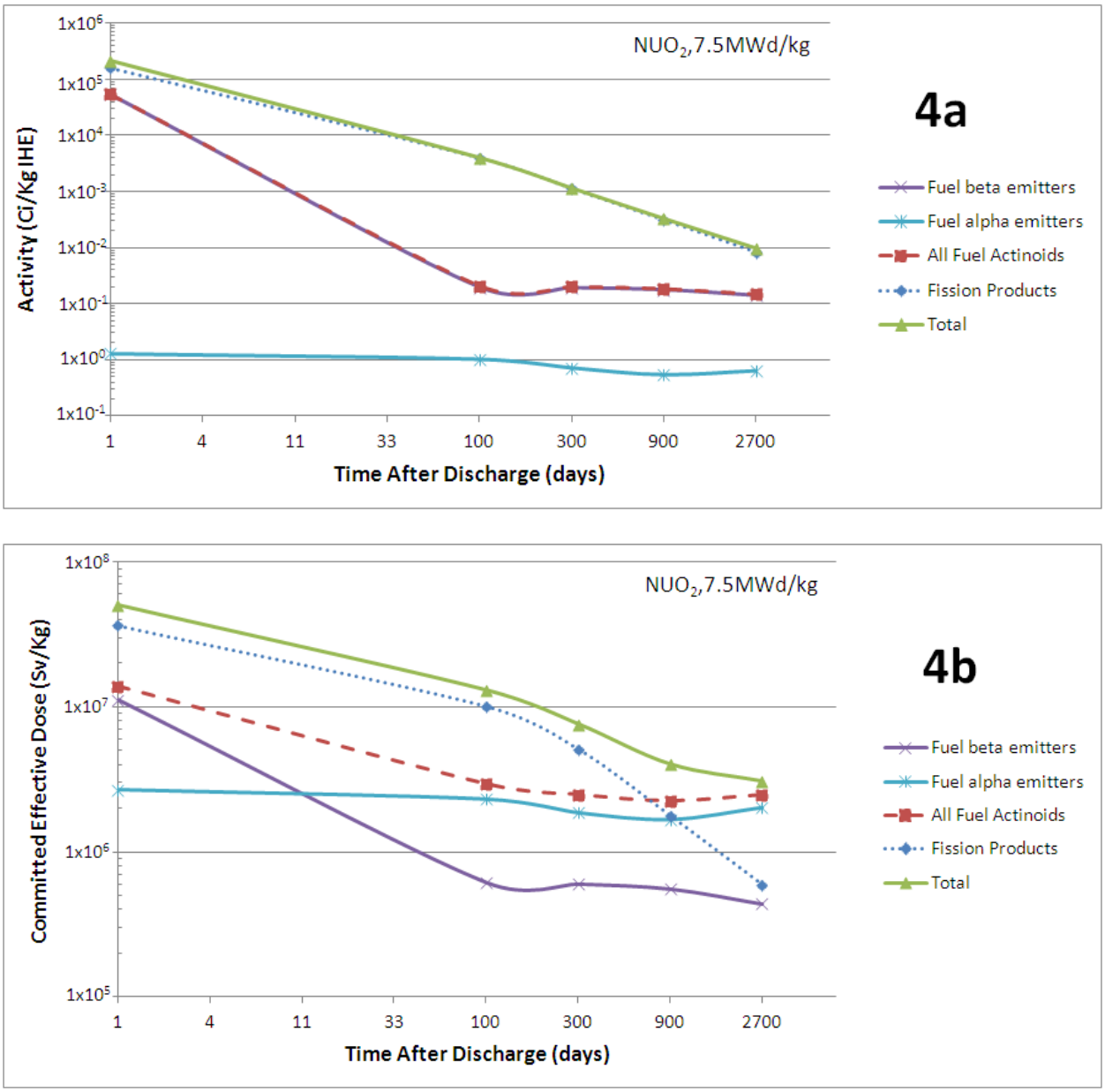


Figure 4

Average of all fuel elements of a natural uranium oxide fuel bundle. The fuel specific activity (A) and committed effective dose (B), from the point of discharge from the reactor, to 2700 days of cooling in on-site storage.

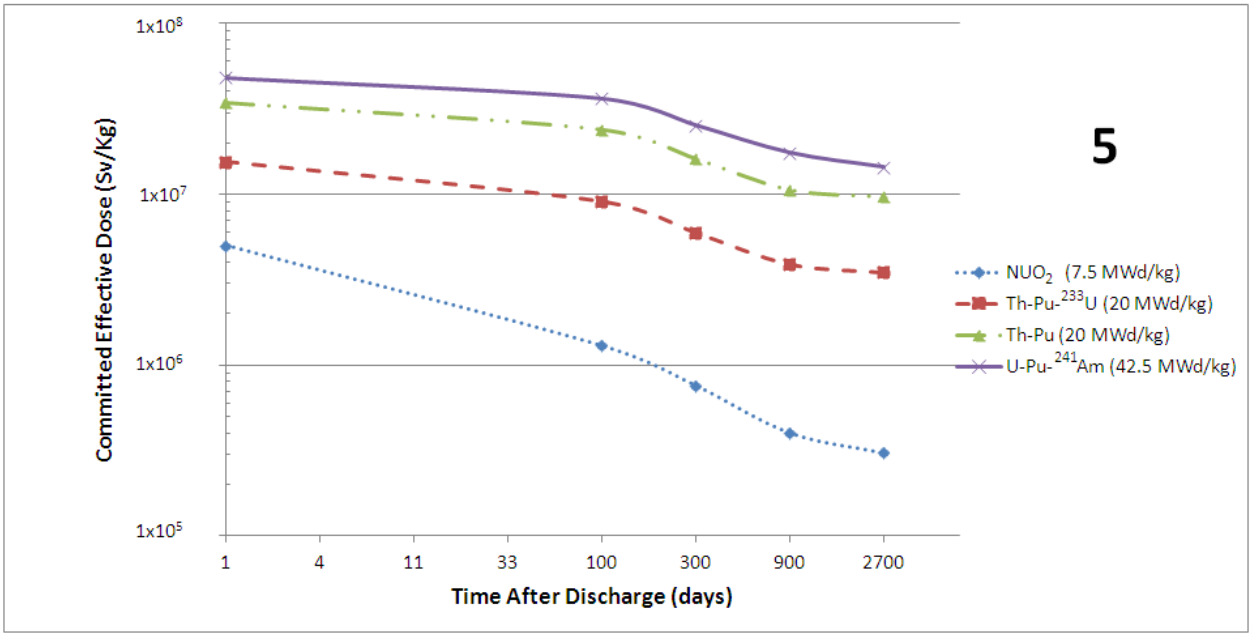


Figure 5

The calculated specific committed effective dose (Sv kg^{-1} inhalation, 5µm AMAD, ICRP Class S) i.e., $\tilde{E}(50)$ resulting from the inhalation of different advanced heavy water reactor fuels.

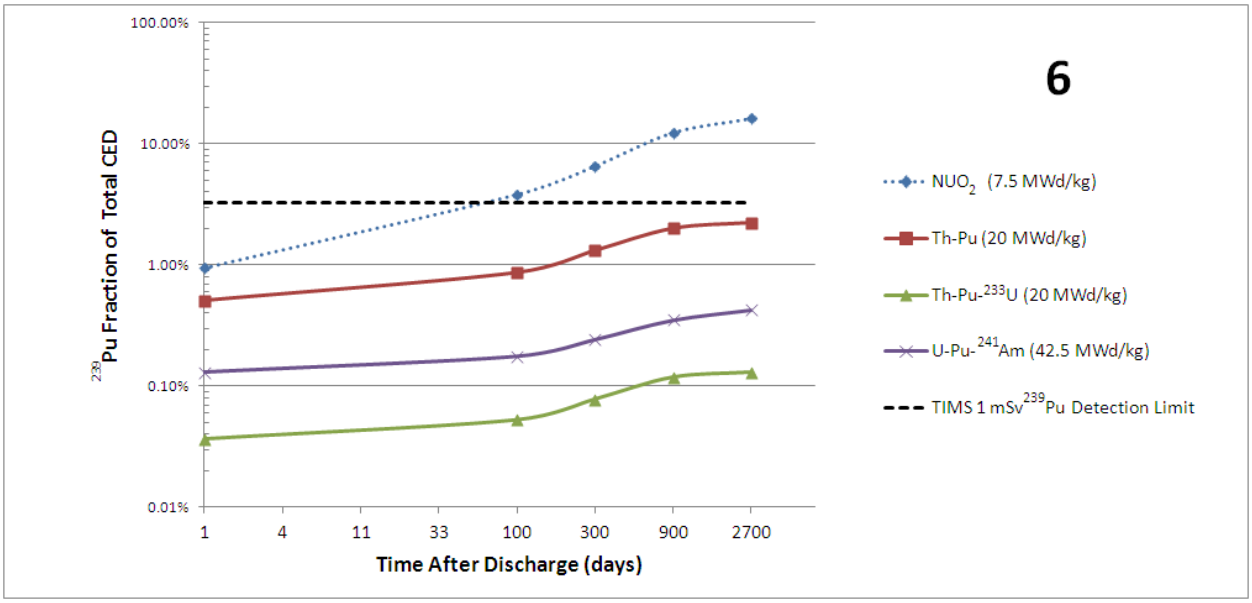


Figure 6

The fraction of the total $\tilde{E}(50)$ from ^{239}Pu in natural uranium and three other advanced heavy water reactor fuels. For an $\tilde{E}(50)$ of 1 mSv, a fraction of ^{239}Pu greater than ~3.3% (shown) will put an amount of ^{239}Pu in urine detectable via TIMS analysis

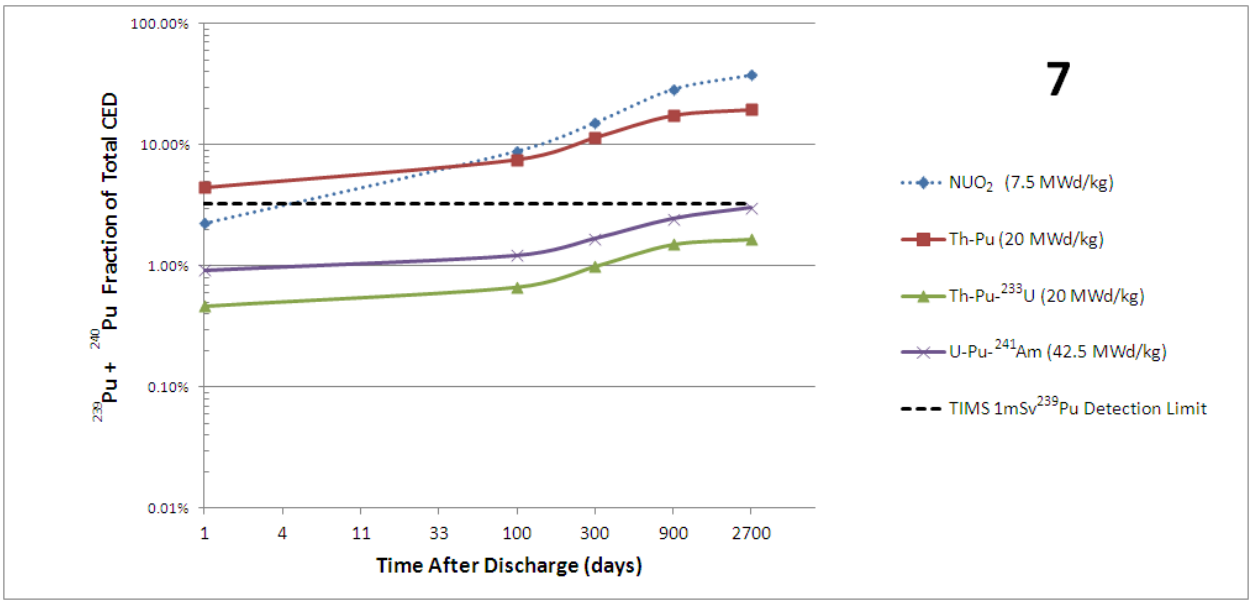


Figure 7

The fraction of the total $\tilde{E}(50)$ from $^{239}Pu+^{240}Pu$ in natural uranium and three other advanced heavy water reactor fuels. For an $\tilde{E}(50)$ of 1 mSv, a fraction of $^{239}Pu+^{240}Pu$ greater than ~3.3% (shown) will put an amount of $^{239}Pu+^{240}Pu$ in urine detectable via TIMS analysis

9. TABLES

Table 1. Element configuration and composition of unirradiated thorium-plutonium fuel.

Element Location	Number of elements	Fuel radius (cm)	Fuel material (% of mass)	Fuel density (g cm^{-3})	Fuel radionuclides (% of elemental mass)
Central	1		Zr-4 clad hafnium element		
Inner	7	0.53025	3.75% PuO_2 96.25% ThO_2	9.7	2.6% ^{238}Pu 54.2% ^{239}Pu
Intermediate	14	0.53025	3.75% PuO_2 96.25% ThO_2	9.7	23.8% ^{240}Pu 12.6% ^{241}Pu
Outer	21	0.53025	3.75% PuO_2 96.25% ThO_2	9.7	6.8% ^{242}Pu 99.9975% ^{232}Th 0.0025% ^{230}Th
Average Burnup			20.4 MWd kg^{-1} initial heavy elements		
Irradiation Time			650 days		
Average Power			31.3 W g^{-1}		
Power at Discharge			26.4 W g^{-1}		

Table 2. Element configuration and composition of unirradiated thorium-plutonium-U-233 fuel.

Element Location	Number of elements	Fuel radius (cm)	Fuel material (% of oxide mass)	Fuel density (g cm^{-3})	Fuel radionuclides (% of elemental mass)
Central	1	Zr-4 clad hafnium element			
Inner	7	0.53025	2.25% PuO_2 97.75% ThO_2	9.7	2.6% ^{238}Pu 54.2% ^{239}Pu 23.8% ^{240}Pu 12.6% ^{241}Pu 6.8% ^{242}Pu 100% ^{232}Th 100% ^{233}U
Intermediate	14	0.53025	0.8% PuO_2 2.1% UO2 97.1% ThO_2	9.7	
Outer	21	0.53025	0.8% PuO_2 1.7% UO2 97.5% ThO_2	9.7	
Average Burnup		19.9 MWd kg^{-1} initial heavy elements			
Irradiation Time		670 days			
Average Power		29.7 W g^{-1}			
Power at Discharge		26.3 W g^{-1}			

Table 3. Element configuration and composition of unirradiated transuranic mox fuel.

Element Location	Number of elements	Fuel radius (cm)	Fuel material (% of mass)	Fuel density (g cm^{-3})	Fuel radionuclides (% of elemental mass)
Central	1	0.6326	0.02%AmO_2 2.7% PuO2 97.28% UO_2	9.7	2.6% ^{238}Pu 54.2% ^{239}Pu 23.8% ^{240}Pu 12.6% ^{241}Pu 6.8% ^{242}Pu 99.284% ^{238}U 0.711% ^{235}U 0.005% ^{234}U 100% ^{241}Am
Inner	7	0.6326	0.02%AmO_2 2.7% PuO2 97.28% UO_2	9.7	
Intermediate	14	0.535	0.02%AmO_2 2.7% PuO2 97.28% UO_2	9.7	
Outer	21	0.535	0.02%AmO_2 2.7% PuO2 97.28% UO_2	9.7	
Average Burnup		42.4 MWd kg^{-1} Initial Heavy Elements			
Irradiation Time		1503 days			
Average Power		28.2 W g^{-1}			
Power at Discharge		28.2 W g^{-1}			

Table 4. Thorium-plutonium MOX Reference Hazard (at discharge). Only radionuclides shown are those with an $\tilde{E}(50)$ *of* ≥1% of the fuel total. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{106}Ru	6.54x10^{2}	0.56	8.3x10^{5}	2.4
	^{144}Ce	7.81x10^{2}	0.67	8.3x10^{5}	2.4
	All	1.16x10^{5}	100	3.7x10^{6}	10.9
Fuel beta emitters	^{233}Pa	1.69x10^{4}	46.97	2.0x10^{6}	5.8
	^{241}Pu	2.80x10^{2}	0.78	8.8x10^{5}	2.6
	All	3.60x10^{4}	100	3.0x10^{6}	8.6
Fuel alpha emitters	^{238}Pu	9.45x10^{0}	6.27	3.7x10^{6}	10.8
	^{240}Pu	1.96x10^{0}	1.30	6.1x10^{5}	1.8
	^{242}Cm	1.27x10^{2}	84.33	1.9x10^{7}	55.2
	^{244}Cm	1.06x10^{1}	7.02	3.7x10^{6}	10.8
	All	1.51x10^{2}	100	2.8x10^{7}	80.5
Total		1.52x10^{5}		3.4x10^{7}	

Table 5. Thorium-plutonium MOX Reference Hazard (100 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{106}Ru	5.42x10^{2}	11.44	6.9x10^{5}	2.9
	^{144}Ce	6.13x10^{2}	12.92	6.5x10^{5}	2.7
	All	4.74x10^{3}	100	1.7x10^{6}	7.3
Fuel beta emitters	^{241}Pu	2.76x10^{2}	17.50	8.7x10^{5}	3.6
	All	1.58x10^{3}	100	1.0x10^{6}	4.3
Fuel alpha emitters	^{232}U	2.58x10^{-1}	0.24	2.5x10^{5}	1.0
	^{238}Pu	9.65x10^{0}	8.99	3.8x10^{6}	15.9
	^{240}Pu	1.96x10^{0}	1.82	6.1x10^{5}	2.5
	^{242}Cm	8.35x10^{1}	77.73	1.2x10^{7}	52.1
	^{244}Cm	1.05x10^{1}	9.75	3.7x10^{6}	15.4
	All	1.07x10^{2}	100	2.1x10^{7}	88.4
Total		6.43x10^{3}		2.4x10^{7}	

Table 6. Thorium-plutonium MOX Reference Hazard (300 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{106}Ru	3.74x10^{2}	18.17	4.8x10^{5}	3.0
	^{144}Ce	3.77x10^{2}	18.32	4.0x10^{5}	2.5
	All	2.06x10^{3}	100	1.1x10^{6}	6.6
Fuel beta emitters	^{241}Pu	2.69x10^{2}	97.09	8.5x10^{5}	5.3
	All	2.77x10^{2}	100	8.5x10^{5}	5.3
Fuel alpha emitters	^{232}U	2.57x10^{-1}	0.43	2.5x10^{5}	1.5
	^{238}Pu	9.85x10^{0}	16.44	3.9x10^{6}	24.0
	^{240}Pu	1.96x10^{0}	3.26	6.1x10^{5}	3.8
	^{241}Am	9.66x10^{-1}	1.61	3.1x10^{5}	1.9
	^{242}Cm	3.57x10^{1}	59.48	5.3x10^{6}	33.0
	^{244}Cm	1.02x10^{1}	17.09	3.6x10^{6}	22.3
	All	5.99x10^{1}	100	1.4x10^{7}	88.1
Total		2.39x10^{3}		1.6x10^{7}	

Table 7. Thorium-plutonium MOX Reference Hazard (900 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{106}Ru	1.22x10^{2}	17.11	1.5x10^{5}	1.5
	All	7.13x10^{2}	100	4.1x10^{5}	3.8
Fuel beta emitters	^{241}Pu	2.48x10^{2}	99.80	4.7x10^{5}	7.4
	All	2.49x10^{2}	100	7.8x10^{5}	7.4
Fuel alpha emitters	^{228}Th	1.70x10^{-1}	0.62	2.0x10^{5}	1.9
	^{232}U	2.53x10^{-1}	0.92	2.4x10^{5}	2.3
	^{238}Pu	9.89x10^{0}	35.98	3.9x10^{6}	36.6
	^{240}Pu	1.96x10^{0}	7.12	6.1x10^{5}	5.7
	^{241}Am	1.64x10^{0}	5.97	5.2x10^{5}	4.9
	^{242}Cm	2.80x10^{0}	10.17	4.2x10^{5}	3.9
	^{244}Cm	9.62x10^{0}	35.00	3.4x10^{6}	31.9
	All	2.75x10^{5}	100	9.4x10^{6}	88.8
Total		9.89x10^{2}		1.1x10^{7}	

Table 8. Thorium-plutonium MOX Reference Hazard (2700 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	No individual fission products contribute more than 1% to total CED				
	All	2.06×10^{2}	100	1.3×10^{5}	1.4
Fuel beta emitters	^{241}Pu	1.96×10^{2}	99.41	6.2×10^{5}	6.4
	All	1.96×10^{2}	100	6.2×10^{5}	6.4
Fuel alpha emitters	^{228}Th	2.32×10^{-1}	0.94	2.8×10^{5}	2.9
	^{232}U	2.40×10^{-1}	0.98	2.3×10^{5}	2.4
	^{238}Pu	9.53×10^{0}	38.68	3.7×10^{6}	38.7
	^{240}Pu	1.96×10^{0}	7.96	6.1×10^{5}	6.3
	^{241}Am	3.37×10^{0}	13.67	1.1×10^{6}	11.1
	^{244}Cm	7.97×10^{0}	32.34	2.8×10^{6}	29.0
	All	2.47×10^{1}	100	8.9×10^{6}	92.2
Total		4.27×10^{2}		9.7×10^{6}	

Table 9. Thorium-plutonium-^{233}U Reference Hazard (at discharge). Only radionuclides shown are those with an $\tilde{E}(50)$ *of* ≥1% of the fuel total. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{89}Sr	1.22x10^{3}	1.09	2.5x10^{5}	1.6
	^{91}Y	1.29x10^{3}	1.15	2.9x10^{5}	1.9
	^{95}Zr	1.31x10^{3}	1.17	2.0x10^{5}	1.3
	^{106}Ru	2.41x10^{2}	0.22	3.1x10^{5}	2.0
	^{131}I	7.76x10^{2}	0.69	2.0x10^{5}	1.9
	^{140}Ba	1.40x10^{3}	1.25	2.4x10^{5}	1.6
	^{141}Ce	1.38x10^{3}	1.23	1.6x10^{5}	1.0
	^{144}Ce	8.55x10^{2}	0.76	9.1x10^{5}	5.9
	All	1.12x10^{6}	100	3.6x10^{6}	23.2
Fuel beta emitters	^{233}Pa	2.02x10^{4}	48.62	2.4x10^{6}	15.3
	^{241}Pu	7.83x10^{1}	0.19	2.5x10^{5}	1.6
	All	4.16x10^{4}	100	2.6x10^{6}	17.1
Fuel alpha emitters	^{232}U	2.78x10^{-1}	0.56	2.7x10^{5}	1.7
	^{238}Pu	2.43x10^{0}	4.86	9.5x10^{5}	6.2
	^{242}Cm	4.21x10^{1}	84.32	6.3x10^{6}	40.6
	^{244}Cm	3.99x10^{0}	8.00	1.4x10^{6}	9.1
	All	4.99x10^{1}	100	9.2x10^{6}	59.7
Total		1.55x10^{5}		1.6x10^{7}	

Table 10. Thorium-plutonium-^{233}U Reference Hazard (100 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{90}Sr	5.99x10^{1}	1.32	1.7x10^{5}	1.9
	^{106}Ru	2.00x10^{2}	4.42	2.5x10^{5}	2.8
	^{144}Ce	6.70x10^{2}	14.81	7.1x10^{5}	7.8
	All	4.53x10^{3}	100	1.5x10^{6}	16.8
Fuel beta emitters	^{233}Pa	1.55x10^{3}	95.23	1.8x10^{5}	2.0
	^{241}Pu	7.73x10^{1}	4.74	2.4x10^{5}	2.7
	All	1.63x10^{3}	100	4.2x10^{4}	4.7
Fuel alpha emitters	^{232}U	2.78x10^{-1}	0.78	2.7x10^{5}	3.0
	^{238}Pu	2.50x10^{0}	7.01	9.8x10^{5}	10.8
	^{240}Pu	4.74x10^{-1}	1.33	1.5x10^{5}	1.6
	^{242}Cm	2.76x10^{1}	77.57	4.1x10^{6}	45.5
	^{244}Cm	3.95x10^{0}	11.09	1.4x10^{6}	15.3
	All	3.56x10^{1}	100	7.1x10^{6}	78.6
Total		6.19x10^{3}		1.6x10^{7}	

Table 11. Thorium-plutonium-^{233}U Reference Hazard (300 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{90}Sr	5.91x10^{1}	3.39	1.7x10^{5}	2.8
	^{106}Ru	1.38x10^{2}	7.90	1.8x10^{5}	2.9
	^{144}Ce	4.12x10^{2}	23.59	4.4x10^{5}	7.3
	All	1.75x10^{3}	100	8.9x10^{5}	15.0
Fuel beta emitters	^{241}Pu	7.52x10^{1}	88.83	2.4x10^{5}	4.0
	All	8.47x10^{1}	100	2.4x10^{5}	4.0
Fuel alpha emitters	^{228}Th	1.11x10^{-1}	0.55	1.3x10^{5}	2.2
	^{232}U	2.77x10^{-1}	1.38	2.7x10^{5}	4.5
	^{238}Pu	2.57x10^{0}	12.81	1.0x10^{6}	16.9
	^{240}Pu	4.74x10^{-1}	2.37	1.5x10^{5}	2.5
	^{241}Am	2.57x10^{-1}	1.28	8.2x10^{4}	1.4
	^{242}Cm	1.18x10^{1}	58.97	1.8x10^{6}	29.6
	^{244}Cm	3.87x10^{0}	19.33	1.4x10^{6}	22.8
	All	2.00x10^{1}	100	4.8x10^{6}	81.0
Total		1.85x10^{3}		6.0x10^{6}	

Table 12. Thorium-plutonium-^{233}U Reference Hazard (900 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{90}Sr	5.68x10^{1}	9.05	1.6x10^{5}	4.1
	^{106}Ru	4.50x10^{1}	7.18	5.7x10^{4}	1.5
	^{137}Cs	6.59x10^{1}	10.50	5.1x10^{4}	1.3
	^{144}Ce	9.57x10^{1}	15.26	1.0x10^{5}	2.6
	All	6.27x10^{2}	100	3.9x10^{5}	10.1
Fuel beta emitters	^{241}Pu	6.95x10^{1}	99.30	2.2x10^{5}	5.6
	All	7.00x10^{1}	100	2.2x10^{5}	5.6
Fuel alpha emitters	^{228}Th	1.84x10^{-1}	1.95	2.2x10^{5}	5.7
	^{232}U	2.72x10^{-1}	2.88	2.6x10^{5}	6.7
	^{238}Pu	2.59x10^{0}	27.36	1.0x10^{6}	26.0
	^{240}Pu	4.75x10^{-1}	5.02	1.5x10^{5}	3.8
	^{241}Am	4.46x10^{-1}	4.71	1.4x10^{5}	3.6
	^{242}Cm	9.25x10^{-1}	9.78	1.4x10^{5}	3.5
	^{244}Cm	3.64x10^{0}	38.43	1.3x10^{6}	32.7
	All	9.46x10^{0}	100	3.3x10^{6}	84.3
Total		7.07x10^{2}		3.9x10^{6}	

Table 13. Thorium-plutonium-^{233}U Reference Hazard (2700 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{90}Sr	5.03x10^{1}	19.98	1.4x10^{5}	4.1
	^{137}Cs	5.88x10^{1}	23.34	4.6x10^{4}	1.3
	All	2.52x10^{2}	100	2.0x10^{5}	5.7
Fuel beta emitters	^{241}Pu	5.48x10^{1}	98.83	1.7x10^{5}	4.9
	All	5.54x10^{1}	100	1.7x10^{5}	5.0
Fuel alpha emitters	^{228}Th	2.51x10^{-1}	2.91	3.0x10^{5}	8.6
	^{232}U	2.59x10^{-1}	3.02	2.5x10^{5}	7.1
	^{233}U	1.40x10^{-1}	1.63	3.6x10^{4}	1.0
	^{238}Pu	2.49x10^{0}	29.00	9.8x10^{5}	28.0
	^{240}Pu	4.76x10^{-1}	5.54	1.5x10^{5}	4.2
	^{241}Am	9.29x10^{-1}	10.80	3.0x10^{5}	8.5
	^{244}Cm	3.01x10^{0}	35.01	1.1x10^{6}	30.2
	All	8.60x10^{0}	100	3.1x10^{6}	89.3
Total		3.16x10^{2}		3.5x10^{6}	

Table 14. U-Pu-Am MOX Reference Hazard (at discharge). Only radionuclides shown are those with an $\tilde{E}(50)$ *of* $\geq$1% of the fuel total. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{106}Ru	9.21x10^{2}	0.70	1.2x10^{6}	2.4
	^{144}Ce	8.95x10^{2}	0.68	9.5x10^{5}	2.0
	All	1.31x10^{5}	100	4.3x10^{6}	9.1
Fuel beta emitters	^{239}Np	1.76x10^{4}	46.53	6.7x10^{5}	1.4
	^{241}Pu	1.43x10^{2}	0.38	4.5x10^{5}	0.9
	All	3.78x10^{4}	100	1.3x10^{6}	2.6
Fuel alpha emitters	^{238}Pu	3.96x10^{0}	1.82	1.6x10^{6}	3.2
	^{242}Cm	1.69x10^{2}	77.60	2.5x10^{7}	52.5
	^{244}Cm	4.27x10^{1}	19.65	1.5x10^{7}	31.3
	All	2.18x10^{2}	100	4.2x10^{7}	88.3
Total		1.69x10^{5}		4.8x10^{7}	

Table 15. U-Pu-Am MOX Reference Hazard (100 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{106}Ru	7.46x10^{2}	13.63	9.5x10^{5}	2.6
	^{144}Ce	6.83x10^{2}	12.48	7.2x10^{5}	2.0
	All	5.48x10^{3}	100	2.2x10^{6}	6.1
Fuel beta emitters	^{241}Pu	1.41x10^{2}	99.86	4.4x10^{5}	1.2
	All	1.41x10^{2}	100	4.5x10^{5}	1.2
Fuel alpha emitters	^{238}Pu	4.26x10^{0}	2.67	1.7x10^{6}	4.6
	^{240}Pu	1.23x10^{0}	0.77	3.8x10^{5}	1.1
	^{242}Cm	1.11x10^{2}	69.48	1.7x10^{7}	45.5
	^{244}Cm	4.23x10^{1}	26.54	1.5x10^{7}	40.9
	All	1.59x10^{2}	100	3.4x10^{7}	92.7
Total		5.78x10^{3}		3.6x10^{7}	

Table 16. U-Pu-Am MOX Reference Hazard (300 days of cooling). Only radionuclides with an $\tilde{E}(50)$ more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	$\tilde{E}(50)$ (Sv kg^{-1})	$\tilde{E}(50)$ (% of total fuel)
Fission products	^{106}Ru	5.14x10^{2}	19.05	6.5x10^{5}	2.6
	^{144}Ce	4.20x10^{2}	15.57	4.5x10^{5}	1.8
	All	2.70x10^{3}	100	1.5x10^{6}	5.7
Fuel beta emitters	^{241}Pu	1.32x10^{2}	99.85	4.1x10^{5}	1.6
	All	1.32x10^{2}	100	4.2x10^{5}	1.6
Fuel alpha emitters	^{238}Pu	4.32x10^{0}	4.65	1.7x10^{6}	6.7
	^{240}Pu	1.18x10^{0}	1.27	3.7x10^{5}	1.4
	^{242}Cm	4.55x10^{1}	49.05	6.8x10^{6}	26.8
	^{244}Cm	4.08x10^{1}	44.02	1.4x10^{7}	56.6
	All	9.28x10^{1}	100	2.4x10^{7}	92.6
Total		2.92x10^{3}		2.5x10^{7}	

Table 17. U-Pu-Am MOX Reference Hazard (900 days of cooling). Only radionuclides with an *Ẽ(50)* more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	*Ẽ(50)* (Sv kg^{-1})	*Ẽ(50)* (% of total fuel)
Fission products	^{106}Ru	1.68x10^{2}	15.77	2.1x10^{5}	1.2
	All	1.06x10^{3}	100	6.1x10^{5}	3.5
Fuel beta emitters	^{241}Pu	1.22x10^{2}	99.84	3.8x10^{5}	2.2
	All	1.22 x10^{2}	100	3.8 x10^{5}	2.2
Fuel alpha emitters	^{238}Pu	4.47x10^{0}	9.17	1.8x10^{6}	10.0
	^{240}Pu	1.19x10^{0}	2.43	3.7x10^{5}	2.1
	^{241}Am	7.66x10^{-1}	1.57	2.4x10^{5}	1.4
	^{242}Cm	3.55x10^{0}	7.28	5.3x10^{5}	3.0
	^{244}Cm	3.83x10^{1}	78.55	1.3x10^{7}	76.9
	All	4.88x10^{1}	100	1.7x10^{7}	94.3
Total		1.24x10^{3}		1.8x10^{6}	

Table 18. U-Pu-Am MOX Reference Hazard (2700 days of cooling). Only radionuclides with an *Ẽ(50)* more than 1% of the fuel total are shown. 'Total' is a sum of the three 'All' rows.

Radionuclide category	Nuclide	Activity (Ci kg^{-1})	Activity (% of total category)	*Ẽ(50)* (Sv kg^{-1})	*Ẽ(50)* (% of total fuel)
Fission products	No individual fission products contribute more than 1% to *Ẽ(50)*				
	All	3.74x10^{2}	100	2.4x10^{5}	1.7
Fuel beta emitters	^{241}Pu	9.60x10^{1}	99.80	3.0x10^{5}	2.1
	All	9.62x10^{1}	100	3.0x10^{5}	2.1
Fuel alpha emitters	^{238}Pu	4.32x10^{0}	10.97	1.7x10^{6}	11.8
	^{240}Pu	1.21x10^{0}	3.06	3.7x10^{5}	2.6
	^{241}Am	1.61x10^{0}	4.09	5.1x10^{5}	3.6
	^{244}Cm	3.17x10^{1}	80.62	1.1x10^{7}	77.3
	All	3.94x10^{1}	100	1.4x10^{7}	96.2
Total		5.10x10^{2}		1.4x10^{7}	

10. ENDNOTES

[*] Possible CANDU fuels include reformed, but otherwise un-reprocessed, LWR or AGR oxide fuels.

[†] Burnup is a term for the total energy produced by a nuclear fuel per unit mass. A typical unit would be MWd kg^{-1}.

[‡] A kg is a convenient mass for calculations, not a realistic mass for inhalation exposure.

[§] Some radionuclides, like ^{233}U, which are created by the decay of a neutron absorbing intermediary (in this case ^{233}Pa) may be created more efficiently by a reactor history with regular downtime to allow the decay to occur, but the effect is usually small.

[**] The discharge compositions were plotted at 1 day in order to use a log scale for decay time.

[††] The mathematics of radioactive fission product production in irradiated fuel, assuming both a constant fission rate and that the fission product to be a negligible absorber of neutrons itself, is that the time dependence of the concentration approaches a limiting value, L, by means of the function $L \times (1 - 2^{-t/T_{1/2}})$, where $T_{1/2}$ is the half life of the isotope in question. After 3-4 half lives, 90% of the limiting value has been reached.