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Implications of alpha-decay for long term storage of advanced heavy water reactor fuels

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ABSTRACT

The decay of actinides such as ^{238}Pu, results in recoil damage and helium production in spent nuclear fuels. The extent of the damage depends on storage time and spent fuel composition and has implications for the integrity of the fuels. Some advanced nuclear fuels intended for use in pressurized heavy water pressure tube reactors have high initial plutonium content and are anticipated to exhibit swelling and embrittlement, and to accumulate helium bubbles over storage times as short as hundreds of years. Calculations are performed to provide estimates of helium production and fuel swelling associated with alpha decay as a function of storage time. Significant differences are observed between predicted aging characteristics of natural uranium and the advanced fuels, including increased helium concentrations and accelerated fuel swelling in the latter. Implications of these observations for long term storage of advanced fuels are discussed.

1. Introduction

The pressurized heavy water pressure tube reactor (HWR) is uniquely suited to accept a wide range of nuclear fuels due to its outstanding neutron economy, made possible through the use of neutron transparent materials. In conventional HWR, this neutron economy is exploited for the implementation of a natural uranium-based fuel cycle, which is inherently proliferation resistant as it does not require uranium enrichment capabilities. Furthermore, the capability to refuel on-power with small (0.5 m length) fuel bundles provides an enhanced degree of flexibility in refueling and fuel management. This fuel cycle flexibility of the HWR facilitates the use of alternatives to natural or slightly enriched uranium fuel that have the potential to extend the lifetime of natural uranium reserves while reducing the volume and decay heat load associated with spent nuclear fuel.

Decades of research have been conducted on the use of advanced fuels in Canada at Canadian Nuclear Laboratories (formerly Atomic Energy of Canada, Ltd.). A variety of alternative fuel cycles have been envisaged for the HWR, including those based on thorium (Edwards and Hyland, 2011) and those suited for actinide burning (Hyland and Gihm, 2011). These advanced fuel concepts bring with them challenges not only to the control and safety of operating reactors, but also to the safety of long term storage of the spent fuel.

In Canada, it is generally assumed that spent nuclear fuel will be stored in a deep geological repository (DGR) (NWMO, 2017). There is also provision in the DGR design for later fuel retrieval for reprocessing. Whether or not fuel is permanently or temporarily stored, its integrity is considered a key safety feature, serving as the first barrier to contain and isolate the fuel material from the environment. As such, there is interest in understanding how the characteristics of spent nuclear fuel evolve with time and how this may influence the effectiveness of barriers to radionuclide release (Ewing, 2015). Recent studies (Ahn et al., 2013; Wiss et al., 2014; Raynaud and Einziger, 2015) have shown that alpha-decay of actinides in spent nuclear fuel can result in the build-up of helium and swelling of the fuel matrix, leading to cladding stress, thus challenging fuel and fuel sheath integrity.

While there have been extensive studies on advanced fuel cycles in HWR (Edwards and Hyland, 2011; Hyland and Gihm, 2011; Bromley and Hyland, 2013; McDonald and Bromley, 2015), there have been few studies to date on long term storage of advanced HWR fuel (Edwards et al., 2013). The study of Edwards et al. (2013) focused on a scenario assuming failure of both fuel and fuel container and was used to determine potential differences in dose and radiotoxicity above a repository based on differences in fuel composition and associated release of spent fuel materials. In

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their study, no differences in the fuel structural properties were assumed.

In the work presented here, several conceptual advanced fuels have been investigated, with the goal to determine differences in their long term evolution, in particular the buildup of helium and fuel swelling associated with alpha decay. Conventional natural uranium dioxide (NU) fuel is modelled, along with mixed oxide (MOX), plutonium-thorium dioxide (Pu-Th), and low enriched uranium-thorium dioxide (LEU-Th) fuels.

2. Codes and methods

Lattice physics-based fuel depletion and decay calculations were performed using the CNL in-house codes, WIMS-AECL 3.1.2 (Altiparmakov, 2008) and WOBI 3.2.0 (Edwards, 2010). The accumulated number of alpha decays versus decay time was estimated based on the helium concentration obtained from WOBI decay calculations. In addition to helium production from alpha decay, there is also helium present in the fuel at discharge, which is produced from ternary fissions. For every 1 MWd/kg of fuel burnup, approximately 5×10^{15} helium atoms are produced per gram of heavy element, assuming approximately 200 MeV released per fission, and 1.7×10^{-3} fission yield of He (Serot et al., 2005). The displacement damage and fuel swelling associated with alpha decay were estimated using correlations determined by Raynaud and Einziger (2015). Additional details of the codes and methods are provided below.

2.1. WIMS-AECL

WIMS-AECL 3.1.2, is a two-dimensional multi-group deterministic lattice physics code that solves the integral neutron transport equation using collision probabilities (Altiparmakov, 2008). For this study, WIMS-AECL was used in conjunction with an 89-group nuclear data library derived from the ENDF/B VII.0 nuclear data evaluation (Altiparmakov, 2010). WIMS-AECL is used to determine the spatial and energy-dependent flux distribution in cluster geometry fuel (e.g. HWR fuel bundles) and surrounding materials (e.g. fuel channel and moderator). The code is also used to perform fuel depletion calculations.

2.2. WOBI

WOBI 3.2.0 (WIMS-Origen Burnup Integration) (Edwards, 2010), is a computer code developed at AECL that alternately calls the 2-D neutron transport code WIMS-AECL 3.1.2 (Altiparmakov, 2008), the SCALE (Hermann and Westfall, 2000) utility code COUPLE 5.1, and the SCALE depletion code ORIGEN-S 5.1. For each time step, WIMS-AECL generates a detailed model of the neutron fluxes throughout an assembly. These fluxes are used to generate reaction rates and effective cross sections in the fuel regions of the assembly for cross sections and processes in the WIMS-AECL library. Other cross sections from a more comprehensive SCALE AMPX-format cross section library are then added to these, and the SCALE utility COUPLE updates a one-group, problem-dependent library used by ORIGEN-S to advance the burnable material compositions to the end of the step.

The advantage of WOBI over a direct burnup calculation in WIMS-AECL is the inclusion of more nuclides and reaction types (coming from the SCALE AMPX-format library) than are currently available in any WIMS-AECL library. This is because the primary purpose of WIMS-AECL is the calculation of the neutron multiplication factor and nuclides and process that do not significantly affect the neutron flux may be neglected. The advantage of WOBI over ORIGEN-S by itself is the ability to pre-calculate reaction rates for ORIGEN-S for the geometry and materials of interest. Libraries that come with ORIGEN-S as part of the SCALE distribution are reactor and fuel assembly specific, and, as such, not applicable to the fuels investigated here.

2.3. Matrix swelling approximation

Fuel matrix swelling associated with alpha decay results primarily from displacement damage, with only about a 1% relative contribution to swelling expected from helium gas accumulation below $2 \times 10^{19} \alpha/g$ (Wiss et al., 2014). The displacement damage, in turn, can be estimated based on the number of alpha decays (Raynaud and Einziger, 2015) as

$$dpa = 2.5 \times 10^{-19} \alpha/g \quad (1)$$

where dpa is the displacement damage in displacements per atom, and α/g is the number of alpha decays per gram of fuel (heavy elements). The fuel lattice swelling associated with displacement damage can be estimated using

$$\frac{\Delta a}{a_0} = 3.528 \times 10^{-3} \left(1 - e^{-8.492 \times dpa}\right) \quad (2)$$

2.4. Fuel compositions

Several fuel types are considered in this study: natural uranium dioxide (NU), plutonium/uranium dioxide (MOX), plutonium/ thorium dioxide (Pu-Th) and low enriched uranium/ thorium dioxide (LEU-Th) fuels. The NU fuel is composed of natural uranium dioxide, the same as that used in conventional HWR in a 37-element bundle, with 0.71 wt% U-235 to give a burnup of approximately 7.5 MWd/kg (Page, 1976). The MOX fuel composition is homogeneous $(Pu,U)O_2$, using reactor grade plutonium with the following isotopic abundances (Pu-238/Pu-239/Pu-240/Pu-241/Pu-242: 2.75 wt%/51.96 wt%/22.96 wt%/15.23 wt%/7.1 wt%), expected to be obtained reprocessing high-burnup light water reactor fuel. The Pu is combined with depleted uranium (DU) to yield 0.91 wt % of Pu in total heavy elements (HE) to obtain approximately 13.5 MWd/kg using a 37-element bundle with the same geometry as that used for NU (Floyd et al., 1998).

The Pu-Th fuel composition is homogeneous $(Pu,Th)O_2$, using the same reactor grade plutonium as described above. Two different Pu-Th compositions were investigated: the first containing 1% PuO_2 (blanket fuel B02) and the second containing 5% PuO_2 (seed fuel S08). These were selected to be representative of high and low concentrations of plutonium in HWR fuel, and suitable for a seed-blanket core arrangement, as proposed in Bromley and Hyland (2013). The blanket fuel Pu-Th (B02) and the seed fuel Pu-Th (S10) have exit burnups of 41 MWd/kg and 31.1 MWd/kg, respectively.

The LEU-Th fuels were chosen similarly to the Pu-Th fuels: a low and high uranium concentration were chosen based on seed-blanket studies performed and documented in McDonald and Bromley (2015). The enrichment of the LEU is 5 wt% in total heavy element (HE), and the two compositions used 20% LEU (blanket fuel B04) and 40% LEU (seed fuel S08) respectively, with the balance being ThO2. The blanket fuel LEU-Th (B04) and the seed fuel LEU-Th (S08) have exit burnups of 41 MWd/kg and 14.7 MWd/kg, respectively.

The details of the various fuel bundle geometries can be found in Page (1976) for the NU and MOX fuels, (Bromley and Hyland, 2013) for the Pu-Th fuels, and (McDonald and Bromley, 2015) for the LEU-Th fuels.