A FRESH LOOK AT THORIUM FUEL CYCLES IN CANDU REACTORS

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ABSTRACT

AECL has long conducted research on thorium fuel cycles, and has recently increased its effort in this area. This paper discusses the status of and recent advances in the following areas of thorium fuel-cycle studies: fuel-cycle analysis, reactor physics measurements, fabrication, irradiation and post-irradiation examination, assessments of fuel performance, and waste management.

1. INTRODUCTION

The CANDU® reactor has an unsurpassed degree of fuel-cycle flexibility, as a consequence of its excellent neutron economy, on-power refuelling, channel design, and simple fuel bundle (Boczar et al. 1997). These features facilitate the introduction and full exploitation of thorium fuel cycles in an evolutionary fashion.

The thorium fuel cycle in CANDU reactors is of strategic interest for several reasons:

- The abundance of thorium in the earth's crust is about 3 times that of uranium. Hence the thorium fuel cycle ensures long-term nuclear fuel supply. For countries with abundant thorium reserves, the thorium fuel cycle in CANDU reactors would enhance both the sustainability of nuclear power and the degree of energy independence, using a single reactor type.
- In thorium fuel, ^{233}U is produced in-reactor through neutron capture in ^{232}Th, and subsequent beta-decay of ^{233}Th and ^{233}Pa. The concentration of ^{233}U in the spent fuel is about 5 times higher than that of ^{239}Pu in spent natural uranium (NU) UO_2 fuel. This isotope of uranium is a very valuable fissile material because of the high number of neutrons produced per neutron absorbed (η) in a thermal neutron spectrum.
- The thermal conductivity of ThO_2 (thoria) is about 50% higher than that of UO_2 over a large temperature range, and its melting temperature is 340ºC higher than that of UO_2 (Belle and Berman 1982, Fink et al. 1981). As a consequence, fuel operating temperatures will be lower for thoria than for UO_2, and all thermally activated processes, such as creep and diffusion of fission gas from the fuel, will be lower. Fission-gas release from the fuel should be lower for ThO_2 than for UO_2 operating under similar ratings.
- ThO_2 is chemically very stable and does not oxidize—a benefit for normal operation, postulated accidents, and waste management.
- Thorium-232 produces fewer minor actinides than ^{238}U does. The resultant lower radiotoxicity of spent thorium fuel is sometimes claimed to be a benefit in waste management. However, in the Canadian concept for engineered geological disposal, the actinides contained in used fuel are not a

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significant contributor to radiological risk (Goodwin et al. 1994), and this benefit is judged to be small.

- The amount of energy that can be extracted from mined uranium can be significantly extended using thorium fuel cycles, and a wide range of thorium cycles is feasible in CANDU reactors. In the limit, the self-sufficient
- equilibrium thorium (SSET) cycle is independent of NU, and of any external supply of fissile material (Critoph et al. 1975, Milgram 1984). The high neutron economy of CANDU reactors makes this fuel cycle theoretically achievable. Hence a single reactor technology can provide both short-term and long-term assurance of fuel supply. High conversion ratio CANDU reactors utilizing thorium would also be synergistic with more expensive, fast breeder reactors (FBRs), supplying fissile material.
- The once-through thorium (OTT) cycle in CANDU reactors provides an evolutionary approach to exploiting some of the energy potential of thorium without recycling. The optimal OTT cycle is economical today, both in terms of money and in terms of uranium resources. This cycle creates a mine of valuable ^{233}U, safeguarded in the spent fuel, for future recovery if desired.
- Since commercial thorium fuel recycling facilities have not been built, there is an opportunity to establish a new, proliferation-resistant technology for recycling.

While AECL has long maintained a thorium fuel cycle program, the level of effort has been increased recently. This paper discusses the status of research, and recent advances in the areas of fuel-cycle studies, reactor physics measurements, fabrication, irradiation and post-irradiation examination (PIE) of thorium fuels, and assessments of fuel performance and waste management.

2. THORIUM FUEL CYCLES IN CANDU REACTORS

2.1 General

Because thorium itself does not contain a fissile isotope, neutrons must be initially provided by adding a fissile material, either within or outside the ThO_2 itself. How the neutrons are initially provided defines a variety of thorium fuel-cycle options in CANDU reactors; there are, however, 2 broad classes: recycling options, in which the ^{233}U is recycled into fresh fuel; and the OTT cycles, where the rationale for the use of thorium does not rely on recycling the ^{233}U (but where recycling remains a future option). Recent fuel-cycle studies conducted by AECL have focused on the OTT cycles, as nearer term options.

2.2 The OTT Cycles

The OTT cycle produces a mine of valuable ^{233}U in the spent fuel, at little or no extra cost, available for future recovery, predicated by economic or resource considerations.

High neutron economy, on-power fuelling, the channel design, and simplicity of the fuel bundle provide a great deal of flexibility in approaches to the OTT cycle. In one concept, channels would be fuelled either with ThO_2 bundles or with "driver" fuel, typically slightly enriched uranium (SEU). The driver fuel would provide the neutrons required to convert ^{232}Th to ^{233}U in the thoria fuel. In such a system, the thoria would remain in the core much longer than would the driver fuel. In the optimal OTT cycle, a combination of feed rates, burnups, uranium enrichment, and neutron flux level would be chosen so that the cycle is economical compared with either NU or SEU (in terms of either resource utilization or money), without taking any credit for the ^{233}U produced. Simple scoping studies using a lattice code have shown that such OTT cycles do indeed exist (Milgram 1982), although their implementation would pose technical

challenges in fuel management. Other driver fuels could also be considered, such as "DUPIC" fuel from recycled spent pressurized-water reactor (PWR) fuel, or even NU (Dastur et al.1995).

An alternate strategy has recently been assessed for introducing the OTT in CANDU reactors. Compared to NU fuel, this option has better uranium utilization, comparable fuel-cycle costs, lower void reactivity, higher thermalhydraulic margins, a simpler fuel management scheme, and lower bundle and channel powers. However, the uranium utilization and fuel-cycle costs are not as low as for SEU, or for an "optimized" OTT cycle in which thoria and driver fuel are irradiated in separate channels, at different feed rates. This "mixed-bundle" option is a practical means of utilizing thorium in operating CANDU reactors, within the current safety and operating envelopes, and with no significant hardware changes.

The CANFLEX bundle (Inch et al. 1998) was used as the reference fuel for the mixed-bundle studies. Thoria is located in the centre of the bundle (in the central pin, and next ring of 7 elements, comprising ~25% of the fuel volume), while SEU driver fuel occupies the elements in the outer 2 rings of 14 and 21 elements. The SEU enrichment is chosen to give the desired bundle burnup. Lattice calculations using WIMS-AECL (Donnelly 1986) were done for bundle-average burnups of 20, 40, and 60 MW•d/kg HE (normalized to a NU burnup in CANFLEX of 7.6 MW•d/kg HE). Compared to NU, improvements in uranium utilization of 14%, 24%, and 21% respectively, were achieved, with comparable front-end fuelling costs. (Fuel-cycle costs for the mixed bundles would be lower if back-end disposal costs were included.)

Detailed fuel management studies were done using the RFSP code (Rouben 1996) for a CANFLEX mixed bundle having an enrichment in the outer 35 elements of 1.8%, with ThO_2 in the central 8 elements. Two fuel management strategies were considered. In the first, only one fuel type was used throughout the entire core, and the adjuster rods were removed. Adjuster rods provide flux and power flattening with NU fuel but are not needed for this purpose with enriched fuel. (In fact, their presence presents a fuel management challenge because they over-flatten the power in the centre of the channel, resulting in an asymmetric double hump in the axial bundle power distribution.) The bundle-average burnup is 22 MW•d/kg HE, with the average burnup of the SEU elements 25 MW•d/kg HE, and that of the ThO_2 elements 10.4 MW•d/kg HE. These are burnups that are readily accommodated in the CANFLEX bundle. A simple, 2-bundle shift fuelling scheme results in a refuelling rate of 5.5 bundles per day, with excellent axial power profiles—the bundle power peaks at bundle position 3 at the inlet end of the channel, then decreases along the length of the channel. This is good from the perspective of fuel performance (only fresh fuel sees an increase in bundle power during refuelling), and thermalhydraulic margins (higher critical channel power than for a cosine-shaped, or outlet-end-peaked axial bundle power distribution). Compared to NU fuel, maximum bundle and channel powers are lower, void reactivity is ~4 mk lower, and uranium utilization (quantity of NU required to produce a unit of power) is 21% better. Even though the thorium burnup is relatively low, the ^{233}U (including ^{233}Pa) content has built up to a level greater than 1.3%, close to its equilibrium value.

The second fuel management option accommodates the adjuster rods. To compensate for their over-flattening, 6% Gd was added to the central ThO_2 element in the same CANFLEX mixed bundle as the one used in the first option, in the inner 124 channels in the adjuster rod region of the core. The other channels employed the same mixed CANFLEX bundle, without Gd. The Gd burns out very quickly but suppresses the initial reactivity sufficiently that the resultant axial power distribution is acceptable (the fuel bundles experience a declining power history after their initial boost). The effect of the Gd on discharge burnup is acceptably small (0.12 MW•d/kg HE, i.e., 0.6% of the nominal discharge burnup). Another feature was the use of 60 dedicated high-burnup thoria channels at the periphery of the core (16% of the whole core), irradiated to a burnup >50 MW•d/kg HE. The elements in the outer 2 rings of these bundles contained 1.7% ^{235}U, using 20 wt % enriched uranium. These bundles experience essentially a constant reactivity and bundle power, to very high burnups, which greatly facilitates fuel management. In essence, they behave the same as do equilibrium ThO_2 bundles for their entire irradiation period. (It takes ~2000 d before a pure ThO_2 bundle reaches equilibrium, and a near-constant reactivity and power.) The fuel burnup penalty

attributed to the presence of these 60 high-burnup thorium fuel channels is also very small (~0.4% of the nominal core-average fuel burnup), partly because the fuel acts as a radial "blanket", reducing neutron leakage from the core. Compared to NU fuel, bundle and channel powers are lower, and the uranium utilization is improved by 14%. The presence of the Gd in the central bundles also reduces void reactivity, and full-core void reactivity is ~6 mk lower than for NU fuel.

These studies represent only a first look at practical fuel management strategies for the OTT cycle. They illustrate the flexibility of the CANDU reactor and CANDU fuel design in accommodating thorium fuel, and show that practical OTT cycles can be devised for existing CANDU reactors. Further analysis will be performed for other OTT cycle strategies. As well, reactor physics methods have just been developed by AECL, to allow explicit representation of the fuel flux and history in the fuel management simulations (Arsenault et al. 1997), which was not done in these preliminary calculations.

2.3 Thorium Cycles Involving Recycling

AECL performed extensive studies for thorium recycling options in the 1970s and 1980s. One useful reference is Veeder and Didsbury (1985), who used a special version of WIMS-AECL to analyze and compare the resource utilization of various CANDU reactor fuel cycles, including once-through NU and SEU fuels, and reprocessing cycles based both on both uranium and thorium. The thorium cycles considered only homogeneous mixtures of ThO_2 with admixed fissile material, with the initial fissile material being either ^{235}U or plutonium. The ^{233}U and any remaining fissile topping material was recycled from the spent fuel, and new fissile topping material was added to maintain burnup. The results show that for thorium cycles, the largest improvements in uranium utilization are realized in replacement generating units that inherit the ^{233}U produced in the units initially using thorium. For such systems in equilibrium, savings in NU requirements of up to 90% compared to once-through fuelling with NU are indicated.

The ultimate uranium-conserving fuel cycle would be the SSET cycle, in which no fissile topping (and hence, no NU) would be required in equilibrium. The ^{233}U concentration in the recycled fresh fuel matches the ^{233}U concentration in the spent fuel. Further improvements in neutron economy would be required to achieve this: reducing the fuel rating to lower the flux and hence neutron capture in ^{233}Pa, increasing the moderator purity, removing the adjuster rods from the core, and, most significantly, enriching the Zr used in the pressure and calandria tubes to remove most of the high cross-section ^{91}Zr. Nonetheless, these changes are feasible.

2.4 Thorium as a Carrier in Plutonium Annihilation

A special application for thoria that has recently received attention is its use as a matrix material for the annihilation of weapons-derived plutonium (Chan et al. 1997). Such Pu-ThO_2 cycles could achieve a very high efficiency in plutonium destruction. One bundle configuration considered employed ~2.6% weapons-derived plutonium in ThO_2 in a CANFLEX bundle that was modified to reduce void reactivity by replacing the central 8 elements by a large central graphite displacer; there were 35 fuel elements in the outer 2 fuel rings. A burnup of 30 MW•d/kg HE was achieved, and >93% of the fissile plutonium was destroyed. Good neutron economy is the key to high efficiency in plutonium destruction with ThO_2. Of course, ^{233}U is produced, through neutron capture in ^{232}Th, and is partially burned in situ. This material is safeguarded in the spent fuel with all the proliferation-resistant barriers afforded by spent fuel. The spent Pu-ThO_2 fuel would be stored until a decision was taken to recycle the contained ^{233}U (and thorium), based on economic and resource considerations (and if necessary, on the availability of a proliferation-resistant recycling technology). Of course, highly enriched uranium (HEU) could also be used as fissile topping for ThO_2, as an option for dispositioning weapons-derived HEU.

2.5 Proliferation-Resistant Thorium Fuel Cycles

To obtain the full energy potential from the thorium cycle in the long term requires recycling of the ^{233}U. The absence of a commercially established facility for recycling thorium fuel opens up the opportunity of incorporating the highest degree of proliferation resistance right from the start in the design of such a facility. One degree of proliferation resistance is provided in thorium fuel cycles by the presence of ^{232}U in the spent fuel, which renders the ^{233}U less attractive for diversion because of its copious emission of alpha particles, and the penetrating 2.6 MeV gamma ray associated with ^{208}Tl in the ^{232}U decay chain (although the ^{208}Tl daughter product builds in with a half-life of several years, and so would not be present immediately in chemically separated uranium).

The DUPIC cycle (Sullivan et al. 1997) is one example of proliferation-resistant recycling technology applied to the uranium fuel cycle. A number of features of this cycle result in a very high degree of proliferation resistance: a dry, thermal mechanical process is used for recycling the fuel; fissile material is not separated; high radiation fields are associated with the process and with the final fuel; and finally, because of the high neutron economy of CANDU reactors, the reconditioned spent PWR pellets can be recycled into CANDU fuel without the addition of further fissile material. Similar considerations can be incorporated into the design of a proliferation-resistant thorium recycle process.

2.6 An Evolutionary Approach to Thorium Fuel Cycles in CANDU Reactors

The OTT cycle allows an evolutionary approach to a CANDU thorium cycle, without irrevocably committing to thorium through the construction of a recycling facility. One strategy would be to start with NU fuel, facilitating localization of the fuel fabrication technology. SEU could then be introduced, lowering fuelling costs even further, improving uranium utilization, and reducing the quantity of spent fuel. (Alternatively, a country with a dual CANDU-PWR reactor mix could recycle the spent PWR fuel into CANDU utilizing the DUPIC cycle.) From there, the OTT cycle could be introduced, and the spent fuel stored until a decision would be taken to recycle the contained ^{233}U (and thorium), predicated by economic and resource considerations. Proliferation-resistant recycling technology could be developed, ideally through international collaboration. Once the decision to recycle is taken, the CANDU fuel-cycle flexibility allows a multitude of options. In the very long term, a CANDU-FBR synergism would allow optimal use of the fissile material produced in the FBR in a CANDU thorium fuel cycle.

3. REACTOR PHYSICS MEASUREMENTS FOR THORIUM FUELS

In the late 1980s and early 1990s, AECL completed 2 sets of measurements in the ZED-2 critical facility on 36-element bundles of CANDU-type fuel containing thorium as the fertile material. These measurements were designed to provide data for validating calculations for thorium-fuelled CANDU lattices performed with lattice-cell codes like WIMS-AECL. The first set of measurements was on fuel consisting of a uniform mixture of ThO_2 and PuO_2 in which Pu was about 2 wt % of the mixture. The second set was on similar-geometry oxide fuel in which the fissile material was ^{233}U at 1.23 wt% of the ThO_2-$^{233}UO_2$ mixture. Measurements were completed for both fuel types at 2 lattice pitches, 31 cm and 24.5 cm, with 3 different coolants in the fuel channels: heavy water, void (air), and light water. Detailed reaction rate distributions were measured in and around the fuel bundle at the centre of the core, for a variety of foils of different elements. The reaction rate measurements for the ^{233}U-Th fuel have been compared with WIMS-AECL calculations (Zeller et al. 1989, 1991).

Insufficient fuel was available to create uniform critical cores of the fuels, so the substitution technique was used to measure lattice buckling (a measure of the reactivity of the test fuel lattice). In this technique, a uniform critical reference core is first created, and its critical size is measured. For these measurements, the reference core consisted typically of 55 assemblies, each containing five 28-element natural UO_2

CANDU bundles, arranged in a hexagonal lattice. Into the central region of this lattice 1, 3, 5, and finally 7 assemblies, each containing 5 bundles of the thorium-containing fuel are successively substituted, the critical size of the reactor being measured for each geometry. An analysis involving a calculational model of the reactor is used to derive the buckling of the substituted fuel lattice for each substitution geometry, and an extrapolation gives the best estimate of the buckling (reactivity) of a critical core of the substituted fuel. In addition, the change in critical size of the reactor was measured as the water coolant and fuel in the 7 substituted rods were heated to 300°C. The substitution analysis methodology has been recently improved, and the analysis of the substitution measurements to yield lattice bucklings using these improved methods is underway.

4. FUEL FABRICATION AND IRRADIATION

AECL's past experience in fabricating ThO_2, $(Th,U)O_2$ and $(Th,Pu)O_2$ fuels includes the following fabrication routes: co-precipitation, dry-powder blending/milling, wet-powder blending/milling, sol-gel microspheres, and extrusion. The latter 2 processes are very experimental and are not discussed here.

There are several methods of co-precipitating UO_2 and ThO_2 (Belle and Berman 1984, Radford and Bratton 1975). AECL's experience in co-precipitation has focused on the addition of ammonia to nitrate solutions. In this process, uranium and thorium are dissolved into a nitrate solution to form $UO_2(NO_3)_2$ and $Th(NO_3)_4$. Ammonia is added to the solution to precipitate $(NH_4)U_2O_7$ (ammonium diuranate - ADU) and $Th(OH)_4$ (thorium hydroxide). The precipitate is calcined to form blended UO_2 and ThO_2 powder, which is subsequently processed into fuel pellets. The microstructure and quality of pellets made by this process are generally very good.

Dry blending can be done by several methods, including mixing in a V-blender, dry-ball milling, vibratory milling and jet milling. These tend to be dusty processes. There can be problems with uniformity on a microscopic scale, and intensive mixing is required. AECL investigated dry blending, using previously ground thoria powders that were mixed with highly enriched uranium powders and dry-ball milled before pressing and sintering. The microstructure of fuel pellets made by this route was quite uniform and devoid of cracks and large pores.

As is the case with dry blending, there are several wet mixing methods, including wet-ball milling and attrition milling. Wet processes have the advantage of not being dusty. In AECL's experience, it was found that product quality was poor and not reproducible with wet processes. The causes of the variations from batch to batch were not well understood. The fuel pellets used as reference fuels in the Whiteshell Reactor 1 (WR 1) irradiations were manufactured using a process of attrition milling, pan drying (forming cakes), granulation, pressing and sintering. This process yielded pellets with non-uniform microstructures and evidence of residual granules from the fabrication process.

AECL conducted an extensive program to develop thoria fuels for use in CANDU reactors, from the mid-1970s to the mid-1980s. Test irradiations were performed in the WR 1 reactor at AECL's Whiteshell Laboratories. Fission-gas release from standard reference $(Th,U)O_2$ pellets in these irradiations was significantly higher than from other thoria fuel irradiations and was generally comparable to UO_2 under similar conditions.

Thoria is expected to have fission-gas retention superior to the fission-gas retention of UO_2, especially at higher powers (see next section). Very recently, AECL investigated the fabrication of ThO_2 (with or without admixed enriched UO_2), and discovered that the primary cause of the non-uniform microstructure was the lubricant that was blended with the granulated fuel before final pressing. By removing the admix lubricant and using a wash to lubricate the pellet die, pellets with uniform microstructure free of residual granules and with high sintered density could be fabricated. Although the admix-lubricant technique for

lubricating dies has been very successful in UO_2 fuel fabrication, it does not appear to work well with ThO_2-based fuels.

As a result of AECL's investigation into factors controlling microstructure, a program is underway to fabricate and irradiate thoria fuels with controlled microstructures, to demonstrate the effect of microstructure on fuel performance and the superior performance of thoria fuels over UO_2. Pure ThO_2 and $(Th,U)O_2$ pellets have been fabricated and loaded onto a demountable-element bundle, which is currently being irradiated in the NRU reactor at the Chalk River Laboratories.

In addition to the WR 1 irradiations, AECL also irradiated four 19-element bundles in the NPD reactor between 1977 and 1987. The fuel in two of these bundles contained 2.6 wt % UO_2 (enriched to 93 wt % ^{235}U) and the other two contained 1.45 wt % UO_2. The irradiation was done at low powers (less than 30 kW/m) to discharge burnups of approximately 40 MW•d/kg HE. The fission-gas release from these fuels was typically less than 1%, demonstrating good performance at low powers.

5. WASTE MANAGEMENT ASPECTS OF THORIA FUELS

Thoria-based fuels are also appealing from a waste management perspective because ThO_2 is chemically stable and almost insoluble in groundwater. By far the most important chemical difference between ThO_2 and UO_2 is the fact that thorium is present in its maximum oxidation state, Th(IV), whereas uranium is not. Therefore, oxidative dissolution of the matrix is not an issue with thoria fuel. Redox conditions could affect the leachability of ^{233}U from irradiated thoria, but this would be limited to surface dissolution and is unlikely to be a major concern.

The inertness of thoria to oxidation is also relevant to interim dry storage of irradiated fuel before geological disposal. In contrast with UO_2, air oxidation of the fuel matrix in defective elements is not an issue with thoria-based fuels. Moreover, the thoria structure can easily accommodate oxidation of minor solid-solution components such as U and Pu. Thus fuel oxidation is unlikely to be a concern during dry storage of thoria-based fuels, and hence the maximum storage temperature would be limited by some other factor, probably cladding degradation (EPRI 1989).

The solubility of crystalline thoria in aqueous solution at 25°C and pH > 5, in the absence of complexing agents, is extremely low. The release of actinides and those fission products that are retained by the thoria matrix is expected to be limited by the solubility of ThO_2. Such release would be exceedingly slow in an engineered disposal vault of the type envisaged for CANDU UO_2 fuel. No credible aqueous or geochemical process has been identified that would greatly accelerate ThO_2 fuel-matrix dissolution under disposal conditions (Taylor et al. 1996).

Thoria crystallizes with the fluorite structure, as do all other actinide dioxides. Extensive solid-solution formation occurs between these oxides, and the fluorite structure can also accommodate substantial levels of actinides in other oxidation states, as well as many fission products. Thus no phase segregation of actinides is expected to occur within the fuel, either during operation or after disposal, and it is reasonable to assume that release of actinides will be controlled by the slow dissolution rate of the thoria matrix, provided that the fuel is initially homogeneous.

Calculated environmental releases and subsequent radiation doses arising from a CANDU UO_2 fuel disposal vault are dominated by the "instant" release of soluble and mobile fission products (in particular, ^{129}I) from the fuel-sheath gap of the fuel. Grain-boundary inventories may also be released rapidly, as compared with matrix dissolution. It is likely that similar findings would emerge from a detailed assessment of thoria fuel disposal, especially given our expectation of extremely slow matrix dissolution. Therefore, it is important to consider the irradiation history and microstructural behaviour of the fuel, and

to have reliable information on the segregation of mobile fission products to the gap and grain boundaries in thoria-based fuels.

Grain growth in the central region of fuel pellets is a major cause of fission-gas release to the gap, because the gases and other incompatible elements are swept from their original resting places in the fuel matrix and become concentrated at the grain boundaries. There, they form features such as fission-gas bubbles and noble-metal particles (Matzke 1980). Interlinkage of fission-gas bubbles on grain-boundary intersections eventually creates tunnels that permit venting of other fission products to the fuel-cladding gap. Thorium oxide is a better thermal conductor than UO_2 is; it also has a higher melting point and slower cation diffusion. Therefore, for a given power rating and fuel geometry, it would be expected to run cooler and undergo less grain growth.

Fission-gas release rates are expected to be somewhat lower for thoria-based fuels than for UO_2 fuels with comparable geometry, microstructure and power history (see also Section 4). This conclusion is based on the lower diffusion rate for xenon in ThO_2 than UO_2 (Matzke 1992, Shiba 1992) and the smaller burst release in ThO_2 (Kaimal et al. 1990). The expected low gas release rates from thoria-based fuels are supported by in-pile experiments on ThO_2 and $(Th/U)O_2$ fuel assemblies. Goldberg et al. (1977, 1978) measured fission-gas release in a set of 51 thoria-based fuel rods over a range of linear powers, burnups and compositions. The authors gave an expression for the rate of fission-gas release, which suggests that rates are significantly lower than for UO_2 under comparable operating conditions.

In many cases, the segregation and, hence, the leachability of volatile, non-gaseous fission products, such as cesium and iodine, is correlated with fission-gas release (Stroes-Gascoyne et al. 1987, Stroes-Gascoyne 1996), and thus we expect the release of these fission products to be lower for a thoria-based fuel than UO_2. Jones et al. (1977) reported low gas releases for $(Th/U)O_2$ fuels, and also noted that fission-product release from defected thoria elements was one to two orders of magnitude lower than for UO_2. Experimental data obtained by Matzke (1967) supports this notion; he found that the release of Br, Cs and Rb from thoria was generally slower than from UO_2.

Diffusion of fission products in UO_2 and ThO_2 remains poorly understood, but generally appears to involve U or Th ion vacancies. High-temperature, out-of-pile annealing experiments on lightly irradiated or ion-implanted samples appear to be consistent with modestly lower fission-product diffusion rates in ThO_2 than UO_2 —roughly paralleling the difference in cation lattice diffusion (Matzke 1967, 1980; Akabori and Fukuda 1991; Prussin et al. 1988; Naik 1992). Fission-product migration in-reactor involves further complexity; indeed, Matzke (1980) has suggested that 5 different diffusion coefficients are required to model fission-gas transport! Nonetheless, the overall trend is evidently maintained: under equivalent operating conditions, fission-product segregation and release tend to be lower for ThO_2 fuels than for UO_2 fuels.

To summarize, the high degree of chemical stability and the low solubility of thoria make irradiated thoria-based fuels attractive as waste forms for direct geological disposal. Moreover, there is good reason to expect lower fission-gas releases (and correspondingly lower gap and grain-boundary inventories of other fission products) in thoria fuels than in UO_2 fuels with comparable power history. In order to realize these beneficial qualities of thoria-based fuels, an appropriate fuel-fabrication process must be utilized to achieve an acceptable degree of microscopic homogeneity. Detailed PIE and leaching studies of thoria-based fuels, coupled with a thorough understanding of their physical and chemical properties, are needed to support these preliminary conclusions.

6. SUMMARY

The high neutron economy of the CANDU reactor, its ability to be refuelled while running at full power, its fuel channel design, and its simple fuel bundle provide an evolutionary path for allowing full exploitation of the energy potential of thorium fuel cycles in existing reactors.

AECL has done considerable work on many aspects of thorium fuel cycles, including fuel-cycle analysis, reactor physics measurements and analysis, fuel fabrication, irradiation and PIE, and waste management studies.

The thorium fuel cycle in CANDU reactors provides long-term assurance of nuclear fuel supplies, using a proven, reliable reactor technology. Those same CANDU features that provide fuel-cycle flexibility also make possible many thorium fuel-cycle options.

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