Imminent: Irradiation Testing of (Th,Pu)O_2 Fuel – 13560

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ABSTRACT

Commercial-prototype thorium-plutonium oxide (Th-MOX) fuel pellets have been loaded into the material test reactor in Halden, Norway. The fuel is being operated at full power – with instrumentation – in simulated LWR / PHWR conditions and its behaviour is measured 'on-line' as it operates to high burn-up. This is a vital test on the commercialization pathway for this robust new thoria-based fuel. The performance data that is collected will support a fuel modeling effort to support its safety qualification. Several different samples of Th-MOX fuel will be tested, thereby collecting information on ceramic behaviours and their microstructure dependency. The fuel-cycle reasoning underpinning the test campaign is that commercial Th-MOX fuels are an achievable intermediate / near-term SNF management strategy that integrates well with a fast reactor future.

INTRODUCTION / CONTEXT

Commercial motivations for developing any thorium-based fuel derive from the benefits offered to the in-core and post-discharge management of nuclear fuel, rather than from any uranium resource savings that may be achieved. This position statement realistically factors-in current market conditions and nuclear policy imperatives – though not all thorium advocates may agree.

The dominance of back-end fuel cycle factors in building a case for thorium fuels is due to: (i) the fact that thorium dioxide (ThO_2) has advantageous physical & chemical properties which make it well suited as a fertile ceramic matrix that can tolerate long periods in storage conditions, and, (ii) the fact that spent fuel inventories are an ongoing management issue for both governments and utilities, and that technology solutions could be applied to this spent nuclear fuel (SNF) ahead of the time when closed cycles with fast reactors are fully established.

A new nuclear fuel designated “thorium-MOX” is proposed, comprising mixed thorium and plutonium dioxides in ceramic pellet form. It combines the robust properties of ThO_2 (also called thoria) with fissile plutonium that has been separated from SNF and destined for recycling / destruction. A thorium-MOX fuel is an achievable prospect for vendors to offer operators of light water (LWR) or heavy water reactors (PHWR). Its achievability is among its key attributes. The use of thorium-MOX in existing LWRs or PHWRs can be postulated as an intermediate SNF management strategy in advance of a deployed advanced reactor fleet.

Indications to date suggest that thorium-MOX fuel is superior to uranium-MOX fuel in many respects, including its in-core operating characteristics - the measurement of which is the primary goal for the irradiation trial being run in the Halden reactor.

THORIA AS A FERTILE FUEL MATRIX

Thorium dioxide has been previously studied as a fertile ceramic matrix for plutonium and minor actinide-bearing fuels[1]. There are two broad dimensions to its feasibility: (i) neutronically, *ie*, that it can be optimized to achieve high levels and rates of plutonium consumption (destruction) [e.g., 1-15]. (ii) its material properties, *ie*, the suite of physical and chemical attributes that lead to strong/safe performance in terms of fission gas release, margin to melting, resistance to leaching, etc.

Advantageous Material Properties

Thorium dioxide has material properties that make it well suited for use as a fertile fuel matrix [*e.g.*, 16-30], especially compared with uranium dioxide as used in current MOX fuels.

- Thermal Conductivity: this is higher for pure ThO_2 than for pure UO_2, and while the admixture of plutonium reduces the conductivity somewhat, it remains higher than for urania-based ceramics [*e.g.*, 21-23]. This indicates that ThO_2 matrix fuel may operate at lower temperature and that in a shut-down scenario there will be less contained heat in the core.
- Melting Point: this is approximately 365^{o}C higher for ThO_2 than UO_2 [24], thereby providing extra safety margin for the fuel in a loss-of-coolant accident scenario, and potentially enabling higher operating power level in normal conditions (favorable for utility operators).
- Chemical Stability & extremely low solubility: ThO_2 is highly inert and cannot be oxidized (unlike UO_2) due the maximum +4 oxidation state for thorium. It is also much less soluble than UO_2 in aqueous media [*e.g.*, 25-27] which is of safety benefit in certain abnormal reactor operation scenarios and it provides added safety margins in waste management and spent-fuel storage contexts.
- Fission Gas Retention: The diffusion of fission gases (most notably; xenon, krypton, iodine) is considerably slower through the ThO_2 crystal lattice, meaning that less gas is released from an operating fuel ceramic, keeping fuel-rod pressures lower and minimizing cladding stress [30,31].
- Ceramic Compatibility: PuO_2 can be intimately blended with thorium dioxide to form a stable, high density sintered ceramic [*e.g.*, 32]. The homogeneity of plutonium distribution can be tailored, depending on the desired MOX-microstructure.

1 Much of the reference list concerns this topic, as do other papers in this Conference session [45-48].

- Neutronics: Thorium has a higher thermal neutron absorption cross section than U-238, thus, more fissile plutonium can be loaded into a thorium-MOX fuel (compared to regular MOX). The reactivity swing over the life of such a fuel is flatter due to the increasing fission contribution of U-233 as fuel ages in an LWR spectrum.

Collectively, these properties provide merit in both the reactor operation (higher power rating) and spent fuel management contexts. In the spent-fuel phase, the high stability and very low leachability of the thoria fuel ceramic would contribute extra safety margin to a SNF storage facility.

Effective Plutonium Consumption & Energy Extraction

Thorium itself produces no plutonium as it 'burns' in a power reactor, since successive neutron absorption results mainly in lower-mass uranium isotopes. This, along with the high thermal fission cross sections for Pu-241 and Pu-239, mean that mixed thorium-plutonium fuels can be designed with the aim to destroy 50-70% of the plutonium they contain, while extracting its fissile energy. Reactor operation coefficients remain well within regulatory limits. Indeed the fuel could be loaded into present-day reactors with essentially no hardware changes.

A large amount of work has been done to model and plan various plutonium minimization strategies using thorium-MOX LWR fuels. Theoretical and experimental irradiation programs have been performed in the US and Europe, as briefly summarized here:

- A number of US studies have made detailed, quantitative assessments of the use of thorium-plutonium fuel in LWRs [*e.g.*, 1-10]. Some investigations have specifically considered weapons-grade plutonium [*e.g.*, 8-10], and others have assessed the inclusion of other TRU components with a view to minimizing minor actinide inventories to achieve waste management benefits [*e.g.*, 7]. Together these have shown the broad feasibility of consuming ~900-1100 kg plutonium per GWe-year as thorium-MOX LWR fuel.
- Recent German analysis of a PWR operating with a full thorium-MOX core [11] confirms an effective plutonium consumption rate of ~57% (element) corresponding to over 1t per GWe-year. Importantly, this detailed assessment showed that temperature reactivity coefficients were always negative. The delayed neutron fraction was lower than for all urania-based fuels, but this is likely to be manageable through enhanced reactivity control measures such as the use of enriched boron.
- The IAEA has reported [12,13] on several country studies on the use of thorium fuels as a means to consume plutonium inventories. Of particular significance is work done in: (i) Korea, showing 49% & 60% (reactor/weapon) plutonium consumption in a non-optimized PWR, (ii) Russia, showing 59% weapons grade plutonium consumption in a non-optimized VVER PWR.
- Studies in Europe [13-16] have also established that thorium-plutonium fuels are effective

for consuming significant quantities of plutonium and greatly reducing fissile content. Plutonium consumption rates of ~47% (115 kg per TWhe) are reported for a full thorium-MOX PWR core.

- A Scandinavian study [17] showed the effectiveness of thorium-MOX fuel in consuming plutonium in BWRs (and PWRs), and that neutronic safety parameters were well within operating safety limits.

Th-MOX FUEL TESTING & QUALIFICATION

Any new nuclear fuel needs to be rigorously qualified to assure that it will function safely in a range of anticipated conditions. A key part of this process is physically testing the fuel's operational performance in order to determine where various safety limits lie. A fuel developer must quantify and understand key operating behaviours for the fuel material – including its: thermal conductivity, thermal expansion, radiation-induced swelling rate and fission gas retention. Only with knowledge of these phenomena is it possible to build models of how fuel properties evolve as burn-up proceeds.

The licensing of thorium MOX fuels is dependent on a reliable, predictive thermo-mechanical behaviour code/s being established for the specific type of $(Th,Pu)O_2$ ceramic that is slated for commercial LWR/PHWR use. Such code/s need to be benchmarked against measured irradiation performance data. Thus, the commercialization of thorium-MOX fuel is highly reliant on physical fuel testing.

Thorium-MOX Fuel Irradiation Experiment

Norwegian company *Thor Energy* has initiated an experimental thorium-MOX irradiation program in which all of the key properties and behaviours will be measured for prototypical thorium-MOX ceramic as it operates in simulated commercial reactor conditions. The project includes thorium-MOX fuel specimens manufactured in different facilities, including some pellets from an earlier European research project, thereby ensuring a strong base of representative types of thoria ceramic.

The irradiation itself is carried out by the *Institute for Energy Technology* - operators of the research reactor in Halden, Norway. Highly instrumented testing rigs are constructed to house the test fuel and 'on-line' data is obtained as it runs at the power rating appropriate for the experiment – in this experiment the starting linear heat generation rate (LHGR) is around 35 kW/m. Figure One shows an engineering drawing of the complex arrangement of instruments in a very small space above the experimental fuel column. Figure Two shows how the test rig is arranged in the reactor core.

The suite of instrumentation comprises: thermocouples that are inserted into the center of the fuel column, extensometers on the cladding and on the fuel column, internal rod-pressure measurement devices that do not require cladding tube penetration. The extensometers and pressure transducers are based on high precision linear voltage differential transformers (LVDT). Spatial (therefore pressure) resolution is very high.

The irradiation conditions experienced by test fuel pellets in the Halden heavy water reactor closely simulate those of an LWR. Measurements being made during the experiment are summarized in the data collection Tables where they appear according to the experimental objective they address. Data deriving from post-irradiation examination (PIE) will also be extremely important and this is also noted in the following five Tables.

Thermal Behaviour	**Data Collected to Quantify Behaviour**	
	On-line Experimental Measurables	*Data from Pre/PIE Measurements*
Thermal conductivity parameters	Center-line fuel temperature, Coolant flow and temperature, neutron flux	Thermal conductivity of fresh fuel, Thermal conductivity of irradiated fuel
Thermal conduction pathway changes	Center-line fuel temperature, Cladding elongation (ind gap closure)	Neutron radiography of fuel pin, microscopy, thermal conductivity of mini-segments of fuel ceramic

Table One Data collection outline relating to thermal property behaviours for Th-MOX

Fission Gas-Release Behaviour	**Data Collected to Quantify Behaviour**	
	On-line Experimental Measurables	*Data from Pre/PIE Measurements*
FGR onset	Rod pressure, Center-line fuel temperature	
FGR amount	Rod pressure	Rod-puncture, gas pressure measurement
Composition of released fission gases		Rod-puncture gas analysis by mass spectrometry
Amount & composition of retained fission gases		Fission gas profiling within the pellet ceramic structure by SIMS and other adv microscopy techniques.

Table Two Data collection outline relating to FGR property behaviours for Th-MOX

Mechanical Behaviour	**Data Collected to Quantify Behaviour**	
	On-line Experimental Measurables	*Data from Pre/PIE Measurements*
Cracking and relocation	Cladding elongation (indicating onset of PCMI)	Neutron radiography, gamma scanning
Densification / swelling	Fuel-column elongation, fuel temperature, Rod pressure	Fresh pellet density, geometry and resintering characteristics.
Pellet-Clad Mechanical Interaction (PCMI)	Fuel-column elongation Cladding elongation, Center-line fuel temperature	Microscopy (OM & SEM), fuel rod profilometry, neutron radiography, irradiated fuel density. Microscopic and element analysis of bond region.
Fuel ceramic microhardness		Destructive analysis on fuel ceramic, punch tests.
High burn-up structure	Rod pressure, fuel-column elongation, cladding elongation, fuel temperature	Grain size & porosity distribution. Microscopy of both fractured & polished ceramic with known burnup.
Fuel inhomogeneities		Neutron radiography, microscopy, both pre- and post- irradiation.

Table Three Data collection outline relating to mechanical property behaviours for Th-MOX

Chemical Behaviour	**Data Collected to Quantify Behaviour**	
	On-line Experimental Measurables	*Data from Pre/PIE Measurements*
General corrosion		Visual inspection (for crud, colour change), eddy current inspection
Stress corrosion cracking		Cladding metallography, microscopy & EPMA
Oxygen mobility & high-oxygen-affinity fission products		Microscopy and EPMA, rod-puncture gas analysis. mXAFS, mXRD if available.
Hydrogen in cladding		Microscopy, neutron radiography, melt extraction
Fuel-rod Moisture		Neutron radiography

Table Four Data collection outline relating to chemical property behaviours for Th-MOX fuel.

Nuclear Behaviour	**Data Collected to Quantify Behaviour**	
	On-line Experimental Measurables	*Data from Pre/PIE Measurements*
General burnup characteristics		Bulk fuel composition / FP and HM concentrations by mass spectrometry, SIMS Radial and axial burn-up distribution by neutron radiography, gamma scanning, EPMA, mass spectrometry, autoradiography, neutron flux and power data.
Actual power level	Initial calorimetry, coolant flow and temperature, neutron flux	

Table Five   Data collection outline relating to nuclear property behaviours for Th-MOX fuel.

The irradiation project is jointly steered and financed by a consortium of research partners which comprises Westinghouse (Sweden/USA, fuel manufacturer), Fortum (Finland, nuclear power utility), Rhodia (France, thorium supplier) and Thor Energy. Advisors and executing partners are NNL (UK, test pellet supply) and IFE (Norway, test reactor operators). The program is supported in part withNorwegian state funds through the national Norwegian Research Council which is providing about 40% of the entire project.. The consortium is still open to new partners.

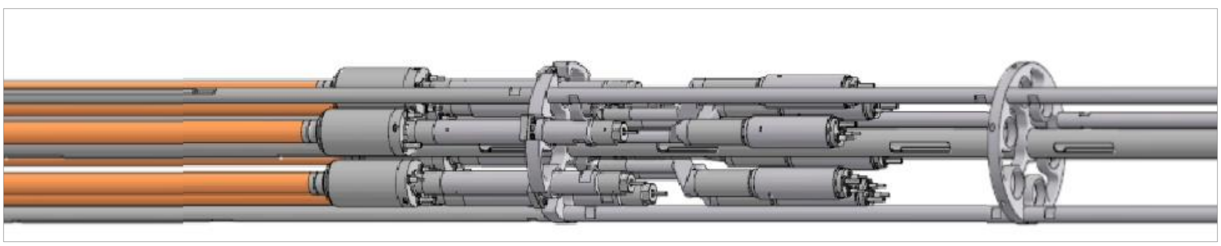

Figure One   Engineering drawing showing the arrangement of instrumentation above test fuel pins within the Th-MOX fuel testing rig. Included are LVDTs to measure internal rod pressure, cladding extension and fuel column contraction/elongation.

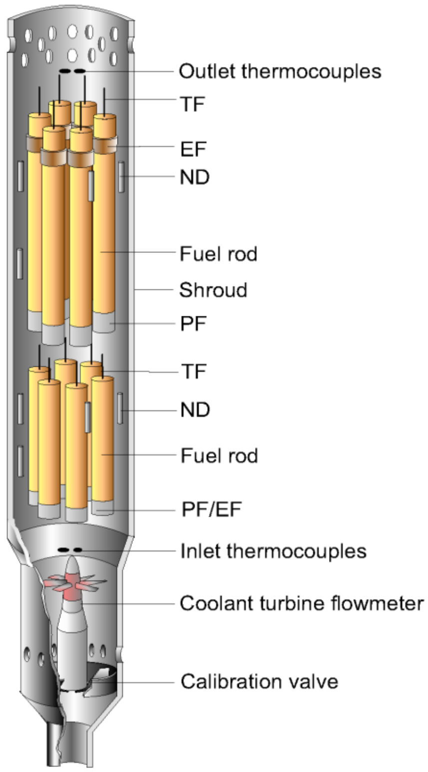

Figure Two A sketch of the second instrumented fuel testing rig that will be used in the thorium-MOX fuel testing campaign in the Halden HBW reactor in Norway.

Th-MOX FUEL – A TECHNOLOGY SPRINGBOARD

The use of thorium-MOX fuels in current/new-build LWR and PHWRs can be described as an interim fuel cycle strategy that can be implemented in the relative near term due to the licensablility of these reactor-type. Another benefit deriving from the use of thorium-MOX fuel relates to the technology development options that will open up once there is some operating experience for this fuel.

Compatibility with Advanced Claddings

Thoria is robust and can tolerate long residence in a power reactor core as fuels designed for high burn-up. These can offer cost savings and lead to high plutonium consumption per irradiation cycle, however the current limitation on fuel burnup comes from the zirconium cladding. Silicon carbide (SiC) offers greater strength, chemical resistance and thermal conductivity (at least during early fuel life) and tests to date [33] show that it should perform well as a cladding material for long irradiation periods.

With an initial Pu-loading of 19%, a batch average burnup on the order of 126 MWd/kg can be achieved for an SiC-clad Th-MOX fuel which is greater by a factor of 2.5 than that feasible with

Zr-clad 5% enriched UO_2 fuel. This leads to a major reduction in the amount of spent fuel produced per kWh. Attaining such burnup at standard power density requires three 32.7 month cycles (8.2 years total) which is well beyond the reach of Zircaloy-clad fuel [44].

Based on chemical compatibility indications it will be feasible to design high burn-up SiC-clad thorium-MOX fuels – and it will be possible to phase these in gradually with regular fuel. There are clearly more neutronic, material compatibility and manufacturing studies to be performed before such a fuel is commercialized. That work has started and the irradiation experiment described here also contributes to this end-goal.

Optional Recovery of Fissile Uranium-233

Thorium-MOX LWR fuels could be optimized to give higher U-233 conversion factors, if that became desired at some point. In any case, fissile U-233 generated in thorium-MOX fuels could be recovered some time the future – when economic feasibility, proliferation risk concerns, and chemical separation technology readiness issues combine to make this a viable strategy. It is noteworthy that U-233 is superior to plutonium as a fissile driver material for LWR fuel since it is much more amenable to multiple cycling in thermal reactors.

CONCLUSION

Thoria-based fuels for LWRs and PHWRs are a highly prospective technology for consuming / transmuting transuranic nuclides of concern *in currently licenced infrastructure*. Such fuels need to be qualified to assure their safe performance in the usual suite of normal/accident scenarios of prime concern to regularors. A pivotal step toward this goal is to test representative fuel under simulated operating conditions.

The trial operation of a commercial prototype thorium-MOX fuel in the Halden fuel test reactor is therefore a significant step toward the broader use of thoria fuel ceramics for achieving 'near-term' fuel cycle goals – most notably with respect TRU minimization.

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