Chapter 18
Overview of European Experience with Thorium Fuels

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Abstract Since the early 1970s, studies and experimental projects have been undertaken in Europe to examine the potential of thorium-based fuels in a variety of reactor types. The first trials were mainly devoted to the use of thorium in high-temperature reactors. These projects can be seen as scientific successes but were not pursued on a commercial basis because of the priority given in Europe to the development of light water reactors. Later on, thorium oxide was considered as a potential matrix for burning plutonium (possibly also minor actinides), and several core design studies, as well as experiments, were undertaken. The most recent such concern the BR2 and HFR Material Test Reactor (MTR) irradiations in Belgium and in the Netherlands, respectively, as well as the KWO PWR in Obrigheim in Germany, in which thorium-plutonium oxide fuel (Th-MOX) was successfully irradiated up to 38 GWd/tHM. The results of these experiments have shown that Th-MOX behaves in a comparable way as conventional uranium-plutonium oxide fuel (U-MOX). More work is still needed before Th-MOX will reach sufficient maturity to implement it on a large scale in power reactors, but all currently available results indicate that licensing Th-MOX for LWRs should be feasible. Finally, European research projects are still devoted to the study of thorium salts in molten salt reactors, a design that incorporates on-line reprocessing and needs no specific thorium fabrication, adding therefore the benefits of thorium without its main challenges.

Keywords Fuel • HTR • LWR • MSR • Plutonium • Thorium

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18.1 Introduction

Natural thorium (Th) has only one isotope, ^{232}Th, which is fertile. In a thermal reactor, Th can absorb neutrons and, following nuclear reactions, produces ^{233}U, which is fissile. Under optimized breeding conditions, a sustainable Th-^{233}U cycle can be reached, but the thorium cycle needs a seed or driver fuel, which can be based on ^{235}U or on Pu.

^{233}U as a fissile nuclide features high neutron production in a thermal and epithermal neutron spectrum. This ability offers improved neutron economy for reactors fueled with ^{233}U rather than ^{235}U or ^{239}Pu, particularly at thermal energies in light water reactors (LWRs). In theory, breeding (formation of fissile nuclides) is achievable at thermal energies with a Th/233U fuel, which is not the case with U-MOX fuel. However, even though breeding can be demonstrated at an experimental level, optimal breeding is not achieved in the current fleet of LWRs. In today's context, U-MOX fuels are not reprocessed, and here Th-MOX offers perhaps its best advantage over U-MOX. The excellent chemical stability of the thorium oxide matrix makes it an excellent candidate for direct disposal, and thus also for once-through fuels allowing burning excess Pu without production of higher actinides. Another alternative would be to use Th-MOX fuels in LWRs as a means to initiate the breeding of ^{233}U for future use in other reactor types, as an option to save natural U and to further improve the U-Pu fuel cycle.

In addition to the LWR/FR scenario, two reactor types have been considered for a breeding Th fuel cycle in the future: high-temperature reactors (HTRs) and molten salt reactors (MSRs). HTRs represent the fastest route to implement a closed breeding Th fuel cycle. The technology exists conceptually but needs to be developed before commercialization (which is pending). Also, supporting technologies associated with fuel manufacturing, reprocessing, transport, waste management, and final disposal need to be developed. MSRs represent a longer-term development option for Th fuel cycles. In MSRs loaded with Th-based fuels, breeding may be achieved over a wide range of neutron energies. On-line reprocessing is an important feature of MSRs, which enables continuous re-use of the nuclear fuel by extracting the fission products.

The potential development of a closed Th fuel cycle faces some obstacles. Reprocessing is one of these, as Th oxide is more stable than U oxide. In contrast to the Purex process, which has been industrially operational in the U-Pu fuel cycle for more than 30 years, the Thorex process, which has been investigated for many years in laboratories, faces some difficulties: it requires stronger acids (and therefore more advanced corrosion-free materials for process vessels) and longer dissolution times. Remote-controlled fuel manufacturing represents another challenge as Th-based fuels have high-energy gamma radiation from the presence of ^{232}U after irradiation, which requires remote fabrication and handling in heavily shielded facilities. Thus, this fuel fabrication, transport, and reprocessing are more complex than the present practice for U oxide fuel, for instance.

This presentation summarizes the history and status of the main European research programs (cordis.europa.eu) with Th use. These programs concerned HTRs, LWRs, and MSRs. Emphasis is given here on the latest two developments.

18.2 Thorium European Research Programme History

During the early years of nuclear energy R&D in Europe, between 1960 and 1980, the main experimental projects involving Th fuels were related to the HTRs (DRAGON OECD international project in the UK, ATR and THTR reactors in Germany) and also to an irradiation of Th-MOX fuel in the Lingen BWR in Germany. These projects can be seen as scientific successes, but they were not pursued on a commercial basis because of the priority given in Europe to the development of LWRs (except in the UK, where low-temperature gas-cooled reactors were developed), with UO_2 as reference fuel, and, for countries having selected the reprocessing cycle strategy, the recycling of the recovered Pu as MOX fuel.

Afterward, several studies were undertaken to examine worldwide interest in Th. In 1997, M. Lung wrote a report entitled “A present review of the thorium fuel cycle” [1] at the request of the European Commission. Then, in the 4th EURATOM Framework Programme, a review of the benefits of the Th cycle as a waste management option was carried out [2].

As a result of these studies, it was recognized that this option presented major advantages in term of actinides management through the “burning” of excess Pu in a non-U matrix (Th oxide), at least for those countries in Europe that considered Pu as a waste and not a source of energy for future utilization in fast reactors. These assessments opened the door to several European irradiation experiments during the 5th EURATOM Framework Programme using Th-MOX, namely in the KWO PWR in Obrigheim (Germany), in the HFR MTR in the Netherland (operated by NRG), and in the BR2 MTR in Mol (SCK•CEN) (“THORIUM CYCLE [3]” and “OMICO [4]” projects). These efforts were pursued and completed within the 6th EURATOM Framework Programme, with the demonstration at laboratory scale that this fuel would behave in a comparable way as current MOX fuel (see Sect. 18.3). In the 6th EURATOM Framework Programme, the fuels irradiated in the programs THORIUM CYCLE and OMICO were further investigated (postirradiation examination, radiochemical analysis, and leaching tests) in the “LWR-DEPUTY” project [5] and a strategy study on the “Impact of Partitioning, Transmutation and Waste Reduction Technologies on the Final Nuclear Waste Disposal” (“RED-IMPACT”) was performed [6].

In parallel, efforts at the European level started in early 2000 and are still under way concerning the development of the MSR, using a Th-^{233}U cycle in liquid Th fluoride fuel. Between the 5th and the 7th EURATOM Framework Programmes, several projects (MOST, ALISIA, EVOL) were funded (see Sect. 18.4).

Within the European nuclear research community, a Technology Platform named SNETP (Sustainable Nuclear Energy Technology Platform: www.snetp.eu) gathers most of the stakeholders involved in reactor research. SNETP issued a “Strategic Research Agenda” in May 2009 (revised in 2013 following the Fukushima accident) with an Annex (in January 2011) devoted to Th. In the annex, Th systems are noted as having significant long-term potentialities but also significant challenges before reaching industrial implementation. The two aspects (Pu management, molten salts) mentioned in this chapter were specifically recognized in the Th Annex to the Strategic Research Agenda.

18.3 Th-MOX Fuels Irradiated in LWR Conditions

Within the European Framework Programmes, the study of Th fuels behavior in LWRs was first aimed at comparing the behavior and the applicability of various matrices to be used for the transmutation of Pu and minor actinides (projects THORIUM CYCLE, LWR-DEPUTY, OMICO). Comparisons were made with standard fuels (UO_2, MOX), and also with so-called inert matrices fuels (using, for example, Mo or MgO as matrix in CERMET and CERCER fuel types, respectively). As explained earlier, irradiation experiments were performed in three facilities, namely, the KWO PWR, HFR, and BR2 Material Test Reactors.

The THORIUM CYCLE project was a 4-year project with the following participants: the coordinator NRG (NL), BNFL (UK), CEA (F), FZK and KWO (D), and JRC-IE and JRC-ITU (EU). The goals of this project, which started on 1 October 2000, were to supply key data for application of the Th cycle in LWRs. In particular, it included the study of

- The behavior of Th-based fuel at extended burn-up through an irradiation experiment of four short fuel pins [UO_2, $(U,Pu)O_2$, ThO_2, and $(Th,Pu)O_2$] up to 55 GWd/tHM in HFR, and an irradiation experiment of one short fuel pin [$(Th,Pu)O_2$] to 38 GWd/tHM in a PWR (KWO); it should be noted that a previous irradiation of $(Th,Pu)O_2$ in Germany (Lingen) achieved a burn-up of 20 GWd/tHM [7];
- The core calculations for Th-based fuel, including code-to-code validation, sensitivity check for significant isotopes ^{232}Th and ^{233}U, and the calculation up to 80–100 GWd/tHM for Th-MOX fuel.

The irradiation test in KWO enabled the investigation of the operational safety of Th-MOX rod behavior under realistic pressurized water reactor (PWR) conditions. The short test rod was inserted in a MOX assembly to provide the most realistic boundary conditions possible. The foreseen MOX carrier assembly had already been irradiated for one cycle. The cladding appeared in good condition after irradiation, and its creep-down, measured at the reactor site during the shut-down periods, as well as its general behavior, were well within the bounds of experience for UO_2 fuels. The fission gas (Xe and Kr) release was about 0.5 % [8], which is about half that for equivalent MOX fuels at the same burn-up, but the linear power was lower than in equivalent U-MOX studies. Taking into account experimental uncertainties, the fuel behavior seems to be at least as good as U-MOX.

The THORIUM CYCLE project was completed in 2006, but the postirradiation experiments were performed under a subsequent experiment called LWR-DEPUTY (coordinator, SCK.CEN). In this program, the main tests on Th-MOX consisted of additional fuels studies (microscopy, radial distributions of elements and isotopes) and radiochemical analyses. The objective of these analyses was to obtain a reliable experimental database for burn-up analysis and to evaluate changes in the heavy nuclide content:

- To optimize the dissolution and analysis strategies
- To establish the first dataset on heavy nuclide and fission product content in irradiated Th-MOX to assess the overall uncertainties
- To use this dataset in a benchmark analysis program

The OMICO Project [4] was conducted from 2001 to 2007. Its scope included the study and modeling of the influence of microstructure and matrix composition on Th-MOX fuel in-pile behavior in normal PWR conditions. The following tasks were undertaken:

- Fabrication of the Th-MOX fuels at the JRC-ITU
- Irradiation in the "CALLISTO" PWR loop in BR2, representing real PWR conditions; the burn-up achieved at the end of this project was about 13 GWd/tHM
- Nondestructive examinations (gamma-spectrometry, visual examinations) and microstructure studies

It should be noted that the pins were instrumented for pressure and fuel temperature determination. The test matrix was such that the Th-MOX could be compared with U-MOX and UO_2 fuels. Another test parameter consisted of the fabrication process (homogeneous versus heterogeneous powder mixtures). The results of the temperature/pressure readings were primarily used to benchmark computer code models for Th-MOX fuels behavior in the first stage of their life.

Besides the irradiation, fuel characterization was performed, including thermal diffusivity measurements, and the results were published [4, 9]. The results show a similar thermal conductivity for (nonirradiated) Th-MOX as compared to U-MOX.

In the LWR-DEPUTY [5] project, selected samples of the OMICO and THORIUM CYCLE programs were extensively studied to provide experimental datasets suitable for evaluating their in-pile performance. The experimental data were the basis of a benchmark exercise on the Th-MOX fuel pin irradiated at the NPP KWO to investigate the qualification of the numerical tools and software packages. A scoping study of the leaching behavior was also conducted. In addition to the experimental work, steady-state and transient analyses were performed for different PWR designs fueled completely or partially with Th-MOX fuel. An assessment of steady-state parameters (reactivity, shutdown margin, and reactivity feedback coefficients) has been performed in comparison with UO_2. All feedback coefficients are favorable for a safe operation under steady-state conditions. A comparative analysis of control rod ejection scenarios has also been performed, and it was found that the maximum values obtained for fuel and clad temperature and maximum fuel enthalpy are in line with the acceptance criteria for the current generation PWRs.

After 10 years of research sponsored through the EURATOM programs, the following conclusions can be drawn regarding the behavior of Th-MOX fuel in LWR conditions:

- Th-MOX has great potential and its fabrication as an oxide fuel is feasible
- Even at a laboratory-scale production route, Th-MOX shows a good in-pile performance
- Know-how on Th-MOX has increased, but
- Fuel performance obviously needs to be further improved before code calculations can predict specific Th-MOX behavior

As a general conclusion, the results of these experiments have shown that Th-MOX behaves in a comparable way (even better in some aspects) to MOX, and that licensing Th-MOX in a LWR should not be problematic, although more experimental data on fuels representative of the future commercial fuels would be needed. Experimental data also demonstrate that Th fuels will be more resistant to corrosion than U fuels in the case of spent fuel geological disposal.

18.4 The Molten Salt Reactor

The MSR, which incorporates the reprocessing on line and needs no specific Th fabrication, adds the benefits of Th without its main challenges. In particular, breeding may be achieved over a wide range of neutron energies, which is not the case for the U-Pu cycle.

Under the European Framework Programs, conceptual developments on fast neutron spectrum molten salt reactors (MSFRs) using fluoride salts open promising possibilities to exploit the ^{232}Th-^{233}U cycle. In addition, they can also contribute to significantly diminishing the radiotoxic inventory from present reactor spent fuels, in particular by lowering the masses of transuranic elements. Finally, if required because of expansion of nuclear electricity generation breeding beyond the iso-generation could be achieved. With the Th-U cycle, doubling times values are only slightly higher than those predicted for solid-fuel fast reactors working in the U/Pu cycle (in the range 40–60 years). The characteristics of different launching modes of the MSFR with a thorium fuel cycle have been studied, in terms of the safety, proliferation, breeding, and deployment capacities of these reactor configurations [10].

Between Framework Programmes 5 and 7, several projects (“MOST”, “ALISIA”, “EVOL”) were conducted, and promising developments and results were obtained in particular in the following areas:

- Conceptual design studies
- Safety developments, in particular, to study the residual heat extraction; tests with liquid salts have been undertaken to prove the ability of the cold plug system to act as a security valve on the loop circuit
- Fabrication of the salt mixture (LiF-NaF-KF) to be used in the French molten salt loop (FFFER project) has been achieved
- Experimental investigation of physicochemical properties of fluoride salts
- Experimental tests of the metallic-phase extraction process;
- Corrosion studies and experiments (this remains one of the main challenges for the development of the reactor system)

Finally, it should be noted that the MSR with its Th cycle is one of the six reference systems selected for R&D collaboration in the framework of the Generation IV International forum. The main contributors are the European partners, supported by Russia as observer.

18.5 Conclusions

Since the early 1970s, studies and experimental projects have been undertaken in Europe to examine the potential of Th-based fuels in a variety of reactor types. These projects have all been successful from a scientific point of view, but not all were followed up relative to the overall development of nuclear industry in Europe. High-temperature reactors (HTRs), although very well suited for Th use, have not been deployed to the benefit of LWRs. Results on the use of Th matrices in Th-MOX fuels in LWRs are encouraging, but still need demonstration at a larger scale in commercial conditions. Finally, the probably most efficient use of Th would be in a salt, to feed MSRs. Conceptual studies and related experimental programs are under way.

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