

Strategic Research Agenda - Annex

January 2011

**Thorium cycles
and Thorium as
a nuclear fuel
component**

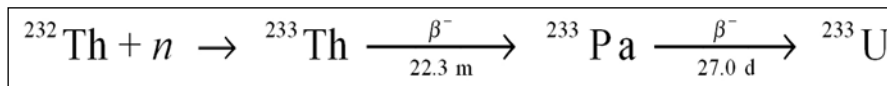
Thorium cycles and Thorium as a nuclear fuel component

Introduction

Thorium as a nuclear fuel component has attracted interest since the dawn of nuclear power. The main reason for the early interest was that breeding nuclear fuel cycles were expected to become a requirement for the expansion of nuclear power foreseen at the time. In this respect, the Th-²³³U-cycle has one major advantage over the U-Pu-cycle, as breeding may be achieved in thermal spectra. Given the difficulties to construct economically competitive fast spectrum reactors, thermal reactors operating in a breeding mode through the Th-²³³U-cycle were, and still may be, an attractive alternative.

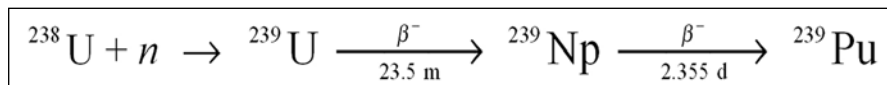
Fundamental physics of thorium fuels

Thorium in nature consists exclusively of the fertile nuclide ²³²Th. When irradiated by neutrons, it produces fissile ²³³U, as described by Equation 1 below:



Equation 1: ²³²Th capturing a neutron forms ²³³Th, rapidly decaying to ²³³Pa, which with a half-life of 27 days decays to fissile ²³³U.

This compares to the production of plutonium from ²³⁸U described by Equation 2:



Equation 2: ²³⁹Pu is produced from ²³⁸U in a process similar to that producing ²³³U from ²³²Th. An important difference though is the shorter half-life of the intermediate product, ²³⁹Np.

The production of fissile ²³³U is delayed due to the 27-day half-life of ²³³Pa. This has implications for the fuel cycle as the reactivity increases for quite some time after the reactor has been stopped, as well as in fuel that has been removed from the core. This needs to be considered in the safety analysis. Also, it provides a means of reactivity control, especially when on-line refuelling is adopted.

Neutron yield per absorption – η

An important reason for the historical interest in thorium as a nuclear fuel component is the high neutron yield per absorption in ²³³U. As depicted in Figure 1, fissioning of ²³³U provides a higher neutron yield than other nuclides for almost the entire thermal and epithermal energy range. This has led to a range of ambitious programmes, all aiming to achieve breeding in thermal reactors. The feasibility of breeding has been shown experimentally in the Shippingport PWR. This, however, required low burnup and a very unorthodox core design without control rods, where reactivity was controlled by moving the fuel assemblies.

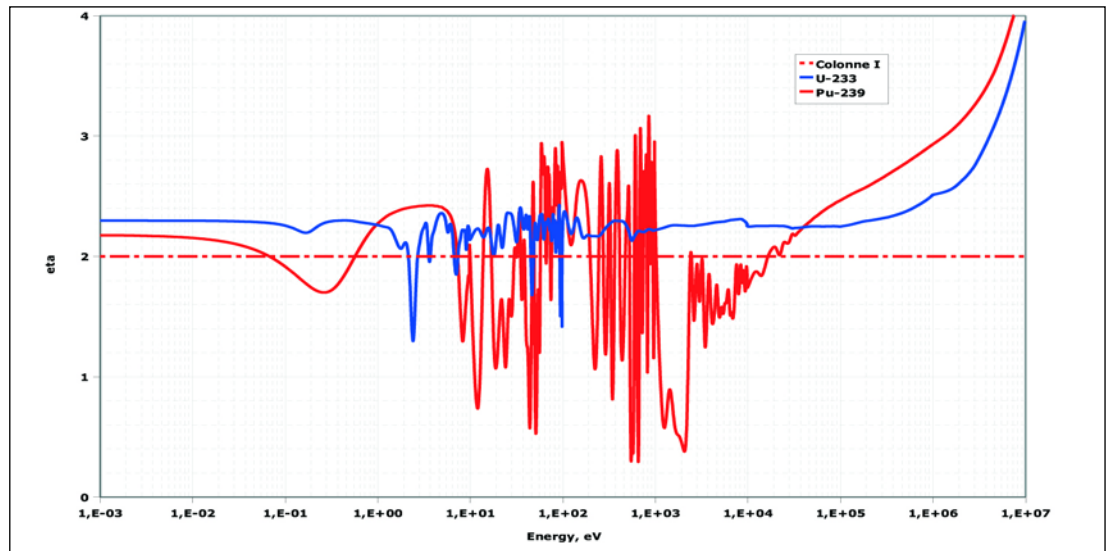


Figure 1: The neutron yield per absorption (η) for ^{233}U is higher than for the other fissile nuclides, e.g. ^{239}Pu , for almost the entire thermal and epithermal energy range.

Seed fuels

The fissile component of thorium fuel is generally referred to as seed fuel. The seed fuel options are ^{233}U , ^{235}U , and $^{239}\text{Pu}/^{241}\text{Pu}$. A closed thorium fuel cycle in equilibrium would only utilise ^{233}U as seed fuel. This, however, requires initial production of ^{233}U from ^{232}Th . This production requires another seed fuel. In the past thorium programmes, highly enriched uranium (HEU), i.e. almost pure ^{235}U , was foreseen. This is an obvious choice since it simplifies reprocessing while the build-up of transuranium elements is avoided. Today though, the use of HEU in commercial nuclear power reactors is excluded through international agreements. Low enriched uranium is rather inefficient as seed fuel, which leaves medium-enriched uranium and plutonium as the option available. Utilising plutonium as seed fuel would mean the fuel would be similar to MOX-fuel, the difference being that the fertile ^{238}U component would be replaced by ^{232}Th .

When HEU is used as seed fuel with a thorium matrix, the production of transuranium elements is minimised. This is a potential advantage for spent fuel disposal, as the minor actinides in particular account for an important part of both the heat load and the radiotoxicity of spent uranium fuel after caesium and strontium have ceased to be the dominating nuclides. With plutonium as seed fuel, this potential advantage almost entirely disappears. Thorium-plutonium fuel will hereafter be referred to as Th-MOX. In both Th-MOX and standard MOX, most of the minor actinide production, (typically more than 75 %), is due to neutron captures in plutonium. It should also be considered that, if Th-MOX is directly disposed, thorium daughter nuclides build in through decay over time, eventually making the thorium fuel more radiotoxic than its uranium counterpart.

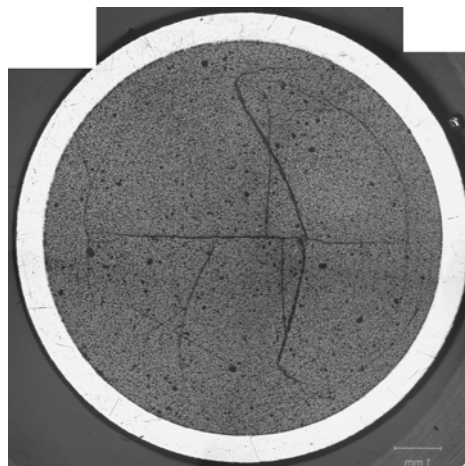


Figure 1: Th-MOX pellet irradiated in Obrigheim within the FP5 THORIUM CYCLE and LWR-DEPUTY projects



²³²U and its daughters

Thorium based fuels are associated with high-energy gamma radiation. During fuel irradiation, ²³²U is produced through (n, 2n)-reactions in ²³³U. In the ²³²U-decay chain, ²⁰⁸Tl and ²¹²Po are of particular interest. Both of these emit 2,6 MeV-gammas as they decay to ²⁰⁸Pb. This high-energy gamma is hard to shield, which has consequences for all handling of irradiated thorium bearing fuels and for reprocessed uranium from such fuels. ²³²U also occurs in reprocessed uranium fuel at parts per billion (ppb) concentrations, and even at these trace levels causes the gamma irradiation field to increase significantly, with consequential effects on radiological protection. Thorium fuels are characterised by much higher ²³²U concentrations that will require remote fabrication and handling in heavily shielded facilities. This makes fuel fabrication, transport and reprocessing more complex than the present practice for uranium oxide fuel, for instance.

The Indian programme

Presently, the most ambitious research on thorium fuel cycles in the world is being conducted in India. These efforts should be understood as part of a long-term strategy of energy independence in India. With insufficient fossil and limited uranium resources available domestically, the Indian approach is to make use of the abundant domestic supply of thorium. It should be acknowledged that the Indian programme has a strong focus on security of supply and assumes an eventual scarcity even of (domestic) fertile uranium. This is hardly the case in any other country, as fertile ²³⁸U is generally considered plentiful.

The Indian programme is divided into three stages. In the first, heavy water reactors (HWR) are used to optimise plutonium production from the scarce domestic natural uranium. In the second, sodium cooled fast reactors are to be utilised to produce plutonium from ²³⁸U and ²³³U from thorium in blankets surrounding the core. In the third stage, heavy water reactors optimised to feature a high conversion ratio are to be used with Th-²³³U-fuels. Breeding is not expected in the third stage. ²³³U will have to be supplied by fast reactors.

The Indian development of thorium fuels is driven not by the scarcity of ²³⁵U, which could have been dealt with by introducing fast reactors. Rather the programme is motivated by the scarcity of domestic fertile uranium, ²³⁸U, for which ²³²Th is a replacement.

At present, the first stage of the Indian programme, HWRs producing plutonium, is in place. The second stage is close, as the first full-scale fast breeder reactor is under construction, with expected completion in the autumn of 2011. The near breeder AHWRs for the third stage are under development. Several important decisions remain before the design is finalised.

Thorium in various reactors

The potential benefits of using thorium as a fuel component vary depending on the choice of reactor. In some, breeding is possible. In others, high conversion ratios are achievable, though breeding is not possible.

Light water reactors - LWR

Breeding has been proven experimentally in a light water reactor. In the Shippingport experimental PWR the production of ²³³U exceeded the consumption. The core had very particular arrangements for control though. Rather than using control rods, fuel assemblies were moved through the core to adjust reactivity. Further, the fuel had to be removed and reprocessed at very low burnup where the conversion ratio is optimal. Without these kinds of arrangement, breeding may not be achieved in LWRs, as the neutron economy is simply not sufficiently favourable.

It seems unrealistic that large power reactors would ever operate in the same manner as the Shippingport experiment, both from a licensing perspective and for economic reasons.

Thorium use in light water reactors would have to build on evolution of present practice rather than on revolutionary changes. The main advantage of LWR technology is the huge experience acquired over the years through operating reactors. If revolutionary changes were to occur, requiring conceptual changes in the core or fuel design, other reactors better suited for breeding in the Th-²³³U-cycle may just as well be developed.

Assuming an evolutionary development of thorium fuels for light water reactors, and the use of plutonium as seed fuel, one would end up with a thorium based mixed oxide fuel, where thorium took the role of ²³⁸U as fertile component. Breeding would not be achievable for this fuel-reactor combination. However, natural uranium savings would be possible. It is likely that the potential for saving natural uranium compares with the use of mixed oxide based on plutonium and ²³⁸U, but optimisation of the savings would require further studies. In this context, high conversion LWRs are interesting as they might achieve high conversion ratios with thorium-based fuels. It may also be interesting to compare the performance of Th-MOX in BWRs to that in PWRs.

The main incentive to use thorium-bearing fuels in light water reactors would be to gain experience of the technologies that would be required to go to a breeding Th-²³³U-cycle at a later stage. Though a breeding cycle would require a different reactor, some of the associated technology would be applicable.

Development of a thorium fuel cycle today would probably begin with mixed thorium-plutonium fuel assemblies in light water reactors. This is expected to be necessary to gain experience of thorium related technologies e.g. in fuel manufacturing and reprocessing. It is also a way of "phasing in" thorium in the commercial fuel cycle, as light water reactors could provide ²³³U during a transition period.

Directly introducing both a new reactor and a new fuel is associated with huge risks and is unlikely to be attractive to utilities. It should be noted though for the purpose of achieving breeding with thorium fuels, it is likely that the light water reactor would not prove adequate.

Heavy water reactors - HWR

Heavy water reactors feature a more favourable neutron economy than light water reactors. The low neutron absorption in the heavy water moderator and coolant allows for the use of natural uranium to be used as fuel directly, without enrichment, whereas in light water reactors the ²³⁵U content has to be raised artificially from 0,7 % to the 3-5 % range. Heavy water reactors are known to be excellent converters for producing plutonium and this also makes them suitable for breeding ²³³U from thorium.

With rapid on-line fuel shuffling and a low target burnup, breeding is indeed possible. But it is difficult to combine breeding with economical operation of the reactor. Achieving this is a key aspect of the Indian programme. It seems, though, that despite efforts to further improve the neutron economy, breeding may be out of reach for any economically realistic burnup. High conversion ratios will be achieved, however, which improves the fuel economy, especially when the fuel is recycled.

Economical breeding in heavy water reactors seems out of reach though breeding is in theory possible. With Th-²³³U fuel high conversion ratios will be achieved, improving the fuel economy.

High temperature reactors - HTR

Just like HWRs, graphite moderated helium gas-cooled high temperature reactors feature neutron economies superior, for example, to those of light water reactors. Consequently, the German HTR-development programme was clearly aimed at establishing a breeding thorium based commercial fuel cycle. HEU was foreseen as seed fuel for the reactors, which were expected to achieve breeding at sufficient core size. In the German programme, pebble bed reactors were chosen rather than the American approach of prismatic block type HTRs. The pebble bed strategy allows a further improved conversion ratio as burnup may be optimised for each fuel pebble.



Today, HTRs appear to be the fastest route to a breeding cycle in the thermal spectrum. Even though the German work was very extensive and solved many problems associated with thorium in HTRs, there are still hurdles to be overcome. An important one is the switching from HEU to plutonium seed fuel. Issues concerning the reprocessing of the fuel also remain unsolved. Most of all, a three-stream hydrometallurgical reprocessing method needs to be developed, in which thorium, uranium and the plutonium seed fuel can be separated and retrieved. There is also scope for design optimisation, especially since conditions have changed since the time of the German programme.

Generally, due to the minimisation of neutron leakage in large reactors, conversion ratios are higher. In large HTRs, a breeding thorium cycle is known to be possible. Current HTR development is towards small, inherently safe reactors. It remains to be examined whether breeding can be achieved in small reactors, e.g. through the use of fertile blankets. The development of better fuels and materials will eventually enable larger inherently safe HTRs.

HTRs represent the fastest route to implementing a closed breeding thorium fuel cycle. The technology exists conceptually, but needs to be developed before commercialisation. Also, a range of supporting technologies associated with fuel manufacturing, transport, waste management and final disposal needs to be developed.

Fast reactors - FR

Breeding can be achieved in fast reactors both in the uranium- and in the thorium cycle. At the high neutron energies typical of fast cores, fissioning of plutonium provides the best neutron economy and hence the highest conversion ratio. In fact sodium cooled cores loaded only with Th-²³³U fuel may just barely breed, implying very long doubling times.

As plutonium fuelled sodium cooled fast reactors grow large enough, they show positive reactivity feedback to coolant voiding. Introducing Th-²³³U fuel into the core generally leads to a less positive or even a negative void coefficient. The main reason for this is the more even neutron yield with varying neutron energies in ²³³U compared with the Pu-nuclides. The hardening of the neutron spectrum resulting from sodium voiding increases the relative neutron yield per absorption more for plutonium than for ²³³U.

Th-²³³U-fuels can be used to improve the void reactivity coefficient in sodium cooled fast reactors. However, they may not offer as high conversion ratios as uranium-plutonium fuels.

Molten salt reactors - MSR

For molten salt reactors, thorium based fuels have several advantages. Breeding may be achieved over a wide range of neutron energies, which is not the case for the uranium-plutonium cycle, for which breeding is unlikely to be achieved in a molten salt reactor.

Online reprocessing is an important aspect of a molten salt reactor system. The fluoride volatility process represents a straightforward reprocessing scheme for the liquid salt thorium fuel. For uranium-based fuels, the fluoride volatility process is less well suited since these would have to be reprocessed at far higher temperature, implying a more challenging process.

Molten salt reactors utilising thorium based fuels represent a long-term development option for thorium fuel cycles. Reactors operating in thermal, intermediate or fast neutron spectra can be employed.

Accelerator driven systems - ADS

Accelerator driven subcritical reactors are suggested for transmutation of minor actinides present in spent nuclear fuel. These are usually referred to as accelerator driven systems, ADS. A common view is that the most efficient transmutation is achieved for fuels consisting of approximately 50 % plutonium and 50 % minor actinides, where americium dominates the minor actinide part. The main purpose of the plutonium component is to maintain reactivity, thus allowing for longer operation and a higher burnup fraction. The disadvantage is the associated breeding of undesired minor actinides. If an inert matrix were used, no additional fissile material would be produced leading to a rapid drop in reactivity and hence to short periods of operation.

An alternative to diluting the minor actinides in a plutonium matrix would be the option of utilising thorium. The production of ^{233}U would act similarly to preserve the reactivity. To benefit fully from this option, the ^{233}U remaining at the end of the ADS irradiation cycle would have to be used in another reactor. As no such system has been envisaged, the option of using a thorium matrix has attracted little interest. The absence of a well-developed reprocessing scheme for thorium fuels is another key factor that affected the decision to propose plutonium matrices for ADS fuels.

A major disadvantage of utilising a thorium matrix in an ADS is the delay in production of ^{233}U following a neutron capture in ^{232}Th . This needs to be compensated for e.g. by increasing the accelerator current. With a plutonium matrix, this is not an issue.

Other incentives

The main reason for the historical interest in thorium as a nuclear fuel component has undoubtedly been the possibility of achieving breeding in thermal neutron spectra. There have, however, been proposals where the use of thorium was suggested for other reasons.

Natural uranium savings

In commercial light water reactors, a breeding thorium cycle is unlikely to materialise. However, it is possible to achieve savings of natural uranium. The most obvious example is recycling of plutonium diluted in a thorium matrix, forming Th-MOX. The savings achieved by adopting this strategy should be benchmarked against use of standard depleted uranium-based MOX.

So far, there has been no convincing evidence that Th-MOX would significantly outperform standard MOX. It is possible though that multi-recycling strategies in combination with high burnup would work to the advantage of Th-MOX.

Thorough investigations of the potential for saving natural uranium by adopting different thorium-fuel strategies for the existing LWR fleet are an obvious short term task in a strategy aimed at future breeding thorium cycles in HTRs or MSR. Even if Th-MOX does not show any improvement in performance compared with standard MOX, its use would provide experience of fuel manufacturing and reprocessing necessary for breeding cycles in other types of reactors.

Physical properties of Th

Thorium has some properties that are interesting from a nuclear fuel perspective. For example, it forms a very stable oxide. This may be interesting both for in-core applications and for waste management through direct disposal.

Pu-burning

Thorium matrices have been investigated for the purpose of incinerating separated plutonium in light water reactors. To maximise the plutonium depletion, a matrix that does not produce new plutonium should be used. Nevertheless, a fertile matrix is required to maintain reactivity. Thorium would be a suitable choice.

This may be an interesting option for some countries with large stockpiles of separated plutonium.

Reprocessing

Historically, a great deal of work has been invested in the development of the Thorex process for hydrometallurgical reprocessing of thorium-based fuels. Essentially, the process is a refinement of the Purex process developed for the uranium-plutonium cycle. The main difference compared to Purex related to thorium oxide being more stable than its uranium counterpart. It therefore requires stronger acids, larger volumes and longer dissolution times. The need for more advanced materials in the process vessels is a consequence of the use of strong acids.



The historical development of the Thorex process focused entirely on a two-stream process, where thorium and uranium were separated and the fission products passed to the waste. This was the obvious path, as highly enriched uranium was foreseen as the seed fuel choice. With HEU excluded, the need for a three-stream reprocessing scheme has arisen. Today, there is no three-stream version of Thorex, but, there are theoretical ideas about how such a scheme would look. The development of any new scheme would require fundamental research. This could probably be based on past experience, but there is also a significant need for innovation to adopt the old scheme to separate plutonium.

It would also be of interest to investigate different options for pyrometallurgical reprocessing of thorium bearing fuels. This field is poorly examined.

Non-Proliferation aspects of Thorium

The International Nuclear Fuel Cycle Evaluation (INFCE) study (1978-1980) under the auspices of IAEA concluded that the thorium fuel cycle, with fissile material contents up to 20%, would present comparable technical characteristics as the U/Pu-cycle and thus comparable in inhibiting proliferation. Despite the good fissile characteristics of ^{233}U , resulting into a small bare metal sphere critical mass of 16.5 kg compared to 10.2 kg for ^{239}Pu and 47.9 kg for ^{235}U , and the small spontaneous neutron emission rate, the presence of even small quantities of ^{232}U significantly complicates the handling of ^{233}U during a weapons fabrication process. Also the presence of ^{228}Th building up to a nearly constant level complicates in various aspects the use of ^{233}U given the high energy γ -emissions.

Quick fabrication of the weapon after separation of the ^{233}U may be considered to reduce the ^{232}U impact during fabrication. Other approaches would consist of producing ^{233}U in thermalised blanket regions of fast reactors where the higher energy neutron flux would be smaller resulting in less ^{232}U being created. Given the decay chain of ^{233}U , γ -emissions from daughters and associated U and Pa-isotopes would provide a 'marker' or 'tag' of the presence of ^{233}U and especially ^{232}U facilitating the detection and tracking of a nuclear weapon including it's the origin of the nuclear material used.

Various other options to deter from using ^{233}U for a nuclear weapon are considered with especially the denaturing of ^{233}U by addition of ^{238}U being mostly considered. The use of thorium fuel cycle would then go associated with a mixture of Th and ^{238}U (e.g. depleted uranium) resulting, after irradiation, into a much diverse set of isotopes (including Pu-isotopes) rendering the use of the ^{233}U as weapons material less attractive (despite presence of Pu). A comparable approach would consist of mixing the separated ^{233}U with ^{238}U after reprocessing.

Anyhow, the proliferation resistance of the thorium fuel cycle is comparable to the U/Pu-cycle especially when one considers that the path towards a thorium fuel cycle would initially demand synergy with the U/Pu-cycle, i.e. the use of medium enriched uranium (MEU) or Pu to initiate the breeding of ^{233}U from the ^{232}Th . For the ultimate longer-term $^{232}\text{Th}/^{233}\text{U}$ cycle, without any further need for MEU or Pu, proliferation concerns may be more important given the very good neutronic characteristics of ^{233}U though routes to denature such materials are envisaged and rather easily to implement.

Summary - The strategic path

Thorium is a fertile element that produces fissile ^{233}U when bombarded with neutrons. ^{233}U as a fissile nuclide features a high neutron production over a wide range of energies. This offers improved neutron economy for reactors fuelled with ^{233}U rather than ^{235}U or $^{239}\text{Pu}/^{241}\text{Pu}$, particularly at thermal energies. In theory, breeding is achievable in thermal spectra with ^{233}U as the fissile

component. Various attempts have been made to show this in practice, and these have succeeded. However, even though breeding can be shown in an experiment, it has proved challenging to achieve it in a commercial reactor.

There seem to be two alternative reactor options for a breeding thorium cycle in the longer run: high temperature reactors and molten salt reactors. It is doubtful whether breeding will be possible in commercial heavy water moderated reactors. In commercial light water reactors, breeding is not an option though thorium may here present a means in further improving the U/Pu-cycle and initiating a breeding of ^{233}U for future use in other reactor types. Breeding can be achieved in fast reactors operating in a thorium fuel cycle, even though it would be more efficient to use uranium-plutonium fuels in such systems.

Assuming HTR or MSR is considered a viable path for future commercial nuclear power, thorium fuels are of interest. In this case, HTR should probably be regarded as a medium-term option, while MSR would be the long-term option. In the short-term, development of thorium fuels and their associated fuel cycle facilities, e.g. fuel manufacturing and reprocessing, could be started by developing fuels for the operating LWR-park and especially fuels that allow for synergies with current U/Pu cycle. In LWRs, thorium fuels would represent an option to save natural uranium and/or to further improve U/Pu-cycle, which in many ways is comparable to the current practice of using MOX for the same purpose.

The potential development of a closed thorium fuel cycle faces some obstacles. Reprocessing is one; remote controlled fuel manufacturing is another. Provided these can be overcome and that new reactors are developed in parallel, nuclear power via the thorium route may become sustainable. The closed thorium fuel cycle thus represents an alternative option for long-term nuclear development.

The breeding closed thorium fuel cycle is an alternative option for the long-term nuclear development. It seems the closing of the thorium cycle requires development of either HTRs or MSRs. Also, a three-stream reprocessing route for thorium based fuels needs to be developed.

It is recommended that the development of HTRs and MSRs go hand in hand with the development of thorium-based fuels as well as with the development of the thorium fuel cycle, including reprocessing, fuel manufacturing and fuel handling.

In the short term, thorium may find limited use in LWRs to jump-start the development of fuel cycle technologies required for the closed fuel cycle envisaged at a later stage.

An R&D-programme on thorium-fuels and fuel cycle options, in synergy with U/Pu-cycle, is welcome and this rather independent of specific choices of reactor technology. R&D on Th-oxide fuels including the reprocessing of these fuels and the recycling of the U, Pu and Th-vector from the used fuels is to be envisaged allowing to assess the various development paths for such Th-bearing fuels. Th-fuel irradiation experiments both in furthering the understanding of such fuels with various U and Pu compositions as well as the reprocessability of these is to be envisaged as important and necessary R&D-steps in the nearby-future allowing to map the available options for a, whenever needed, use of such Th-bearing fuels in both LWRs as transition step towards use on future reactor types.

