Nuclear Science
2015

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Extended Summary

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NEA No. 7228

NUCLEAR ENERGY AGENCY
ORGANISATION FOR ECONOMIC CO-OPERATION AND DEVELOPMENT

Perspectives on the Use of Thorium in the Nuclear Fuel Cycle

Since the beginning of the nuclear era, thorium has been acknowledged as an interesting resource for its potential use as nuclear fuel. In the early period of nuclear energy, thorium had been considered as a possible supplement or even a replacement for uranium which was feared to be scarce at the time. Thorium is in all likelihood relatively abundant on earth and presents a number of intrinsic nuclear and chemical properties that would make its use as a potential nuclear fuel particularly interesting. However, in the early years of nuclear energy, it was soon discovered that the supply of natural uranium was not as limited as initially projected. Moreover, thorium lacks a fissionable isotope – a major drawback for thorium, as it is impossible to start a fission chain reaction purely on natural thorium – and consequently any nuclear system using thorium would initially be dependent on prior generation of fissile matter (extracted *from* uranium or bred *in* uranium systems). A uranium-plutonium fuel cycle was therefore not only an easier but a necessary first step, in line with the strategies of the main countries developing nuclear energy at the time.

Extensive reviews of thorium use in nuclear reactors have been published in the literature over the years. From 1950 to the late 1970s, the thorium fuel cycle was the subject of numerous studies and pilot experiments. Power reactors were operated with thorium-based fuels, demonstrating the feasibility as well as the complexities associated with their use.

In the late 1970s and 1980s, public support for nuclear power declined after the Three Mile Island accident in April 1979 and the Chernobyl accident seven years later in 1986. In conjunction with these events, the price of uranium fell to very low levels in the 1980s and thus the search for an alternative to uranium became economically of little interest.

Also, the cessation of efforts to deploy commercial spent fuel reprocessing in the United States in the late 1970s – motivated primarily by the perceived potential proliferation risks associated with the separation of fissile materials – ended interest in closed nuclear fuel cycles in the United States. This also had an impact on decision making in other countries.

In the case of thorium cycles, the decision to end reprocessing removed the possibility of recovering the ^{233}U from irradiated fuel in the United States. In other countries, such as France, the uranium-plutonium fuel cycle was under full development, with the implementation of partial recycling and no impetus to change from the uranium-plutonium fuel cycle.

The thorium option was, however, never abandoned and has continued to be studied with fluctuating intensity, particularly in academia. Today, the availability of fissile material (plutonium or enriched uranium) arising from the well-established uranium-plutonium fuel cycle makes the implementation of thorium fuels feasible in principle, although the necessary economic drivers for devoting significant industrial resources to that end have not yet been clearly established.

The 2015 Nuclear Energy Agency (NEA) report *Introduction of Thorium in the Nuclear Fuel Cycle* identified general conditions under which a transition to a thorium fuel cycle would become a practical option, providing details of the technical challenges associated with the various stages and options during that transition.

An in-depth, technical assessment of the potential of thorium-based fuels can only be made within the scope of a well-defined case study. In order to be complete, such an assessment must address many factors, including the type of reactor system hosting the fuel, the type of thorium-bearing fuel studied, the fuel management schemes chosen and the deployment scenarios considered. It should also address both the front and back end of the fuel cycle and consider the national context in which these analyses would be applied. Failure to look at the complete picture, and instead enumerating only latent advantages or disadvantages of the use of thorium as a general resource for nuclear energy, can be misleading. It is therefore important to bring the discussion of thorium into a scientific and technological context, covering the short- to longer-term realities of the nuclear energy industry.

Renewed interest in thorium

Today, many countries are developing or exploring the construction of new nuclear power plants. While several factors, including the financial crisis, the Fukushima Daiichi accident in March 2011 and the natural gas boom in North America, have impacted plans to build new plants in several countries, future energy scenarios continue to project "a significant development of nuclear energy to meet energy and environmental goals, albeit at a somewhat slower rate than previously projected" (NEA, 2012). The arguments behind these projections are based on the importance of issues including:

- low carbon energy production;
- security of energy supply;
- availability of competitive baseload electrical supply;
- other potential uses of nuclear energy, such as process heat generation.

On the other hand, issues related mainly to spent fuel management from the current uranium-plutonium cycle, still present challenges. These challenges include the:

- deployment of geological repositories for direct disposal of intact spent fuel and/or disposal of vitrified waste;
- development of uranium-plutonium fast neutron reactors (FRs);[1]
- transition to generation IV systems.

In some countries, concern over the eventual disposal of spent nuclear fuel has grown. Delays in repository programmes have meant that most countries will have to face an accumulation of spent fuel and high-level waste.

Drivers for thorium fuel development

Given thorium's potential for reduced minor actinide production, its introduction into the fuel cycle continues to be given consideration (IAEA, 2012). In the last fifteen years,

1. Technical and economic challenges are to be resolved before commercial deployment of fast reactors may be considered. At least for the foreseeable future, the development of fast neutron reactors will continue to be driven by governmental programmes with limited industrial prospects before 2050.

increased focus has been given to research on thorium-based fuels for medium-term utilisation in present or evolutionary (generation III+) reactors so as to provide additional options in the management of uranium and plutonium.

Thorium could potentially play a useful role as a complement to the uranium-plutonium fuel cycle as it would enhance the management of used fuels and radioactive waste while also providing a means of dealing with stockpiles of plutonium in the absence of fast reactors. Additionally, thorium could provide flexibility within the context of uncertainty around the long-term availability of relatively cheap uranium.

Studies are being carried out in several areas, aiming at quantifying the potential benefits related to thorium fuels. More specific areas of interest where thorium could play a favourable role in the future are:

- ***Improvements in the utilisation and management of fissile materials.*** Improvements in the current and in future fuel cycles will open up possibilities from the short term onwards. Of particular interest is the possibility of reaching higher conversion factors or even breeding conditions in thermal or epithermal neutron spectra evolutionary generation III+ systems that use thorium-based fuels, with the aim of recovering the fissile material from the used fuel.
- ***Fuel performance.*** Thorium has very promising physicochemical characteristics that are good candidates for improving fuel performance, in particular in the form of thorium dioxide, which has a high thermal conductivity, a low thermal expansion coefficient and a high melting temperature.
- ***Waste management.*** Thorium fuels may lead to less minor actinide production per unit of energy produced, although this depends on the fissile seed used. Thorium may be used as an inert matrix in view of burning plutonium (and possibly other actinide elements) which could provide an option for plutonium management. In the long term, $Th/^{233}U$ fuel cycles would lead to less minor actinides in the waste streams.

It should be recognised, however, that there remain considerable uncertainties associated with the practical application of thorium. With the lack of clear economic incentives to deploy thorium, the industrial development of thorium as a replacement for uranium in the fuel cycle is likely to remain limited.

Basic considerations

Fertile-fissile cycles

Today, water-moderated reactor technologies are dominant around the world. According to IAEA statistics (IAEA, 2013), of the 437 power reactors in operation in 2013, more than 90% were water moderated: 273 were pressurised water reactors (PWRs), 84 boiling water reactors (BWRs) and 48 pressurised heavy water reactors (PHWRs). These technologies rely on the extraction of energy released by fissions triggered by thermalised neutrons. Heavy nuclides that fission under thermal neutron interaction are called “fissile”. Others may be called “fertile” because they are more likely to absorb a thermal neutron instead of undergoing fission and thereby transmute into a heavier nuclide, which may be fissile.

When irradiated by thermal neutrons, ^{232}Th and ^{238}U follow similar processes. ^{232}Th *breeds* ^{233}U in a completely analogous way to that in which ^{238}U *breeds* ^{239}Pu. These two processes of neutron “radiative capture” (n,γ) reactions, followed by consecutive radioactive decays (β^-) represent the two practical “fertile/fissile fuel cycles”; the $^{238}U/^{239}Pu$ fuel cycle and the $^{232}Th/^{233}U$ fuel cycle (Figure 1).

Figure 1. The thorium and uranium fertile/fissile fuel cycles

$$^{232}\mathrm{Th} \xrightarrow{(n,\gamma)} {}^{233}\mathrm{Th} \xrightarrow[T_{1/2}=22\mathrm{min}]{\beta^-} {}^{233}\mathrm{Pa} \xrightarrow[T_{1/2}=27\mathrm{d}]{\beta^-} {}^{233}\mathrm{U}$$

$$^{238}\mathrm{U} \xrightarrow{(n,\gamma)} {}^{239}\mathrm{U} \xrightarrow[T_{1/2}=23.5\mathrm{min}]{\beta^-} {}^{239}\mathrm{Np} \xrightarrow[T_{1/2}=2.3\mathrm{d}]{\beta^-} {}^{239}\mathrm{Pu}$$

Conversion ratios and breeding

Although it requires an initial fissile "seed", a thorium fuel cycle may ultimately become autonomous in terms of fissile inventory as creation of the fissile nuclide ^{233}U occurs in fertile thorium, provided the retrieval of this ^{233}U is viable through reprocessing and separation. Under certain conditions, it is possible to create (or breed) more fissile material than is consumed by the reactor.

The conversion ratio (CR), also called the breeding ratio, is defined as the ratio of the rate of production to the rate of consumption of fissile nuclei in the fuel of a reactor in operation.[2] If CR is greater than or equal to 1, the system is called a "breeder". Reaching a conversion ratio of 1 allows a reactor in a closed fuel cycle to function independently[3] from any continuous external supply of fissile matter (assuming negligible losses at the reprocessing stage).

Achieving conversion ratios superior or equal to 1 is a key aspect of the sustainability of advanced nuclear reactor concepts (generation IV and beyond). The concept of breeding is by no means exclusive to the thorium cycle and can also be achieved with the uranium cycle in fast neutron reactors. However, thorium has an advantage in this respect over uranium cycles since it can achieve conversion ratios close to or slightly in excess of unity in thermal (slow) neutron spectra due to the favourable inherent neutronic properties of ^{233}U in this neutron energy range.[4]

2. The conversion ratio depends therefore on effective cross-sections, which vary during burn-up as the composition of the fuel evolves. A usual indicator of the conversion ratio used is the fissile inventory ratio, which can be defined as the ratio of final over initial fissile mass established at discharge of the fuel.
3. A functioning breeder reactor would, in principle, only require the initial fissile inventory to reach equilibrium. Since fissile matter is consumed and produced at the same average rate in the core of a breeder reactor, the actual consumption of matter in the reactor would then only rely on the input of fertile matter into the fuel cycle. This fertile matter – either uranium or thorium – is much more abundant than fissile matter (i.e. in the case of uranium, ~140 times more ^{238}U than ^{235}U) and more readily available.
4. It must, however, be noted that achieving breeding in thorium-fuelled thermal systems is an extremely complex task, in particular for solid fuels. The Shippingport 60 MWe Light Water Breeding Reactor (LWBR) used Th/^{233}U fuel and demonstrated in the late 1970s the possibility of reaching breeding ratios slightly greater than 1 (1.014). The LWBR was indisputably an engineering success made possible by having very low burn-ups, which favours high conversion ratios but sacrifices reactor performance, and by allowing for the complexity of a costly and sophisticated operation of the core, notably by removing all neutronic poisons and control rods. Such complexity is deemed unrealistic today for the safe or economical operation of reactors used for electricity generation.

Thorium resources

Thorium is a common natural element and fairly evenly spread in the crust of the earth. The main mineral host for thorium is monazite, a rare earth phosphate generally containing between 6 to 12% thorium-oxide. Countries possessing significant amounts of thorium resources include Australia, Brazil, Egypt, India, Norway, Russia, Turkey and the United States.

The latest estimates in the joint report by the NEA and the IAEA on *Uranium: Resources, Production and Demand* identified thorium resources amounting to about 6.2 million tonnes (NEA, 2014). Some studies, however, such as the one published by the United States Geological Survey (US Geological Survey, 2014), provide lower estimates of thorium resources of around 1-2 million tonnes.

There is currently no standard classification for thorium resources and these identified resources do not have the same meaning in terms of classification as identified uranium resources.[5]

On the basis of current incomplete knowledge of the available resources and the lack of an effective thorium market, it is difficult to compare available thorium and uranium resources. It is, however, possible to affirm that immediately exploitable thorium and uranium reserves in the crust of the earth are probably of the same order of magnitude, at several million tonnes.

Availability of thorium

To date, thorium production has mainly emerged as a by-product of mining activities for rare earth elements and/or uranium due to its association with these elements in nature. Under the present conditions, dedicated extraction and production of thorium is not considered economically viable due to the current lack of a thorium demand and market. However, such a dedicated extraction would probably not be needed in the short to medium term, because the increase of demand in the past decade for rare earth ores (mainly used in electronic equipment) has resulted in a by-product supply path of thorium ores that is currently independent of the demand for thorium from the nuclear market. Currently, demand for thorium is mainly accounted for by non-energy related companies, which use this material for applications such as high-temperature ceramics, melting tanks, catalysts, welding electrodes and some alloys.

Furthermore, many of the world's monazite deposits, such as the thorium-recovering sites of India, are co-located with titanium ore (ilmenite). Thorium by-product recovery from rare earth elements, as well as from currently active titanium mines, could potentially be greater than the estimated volumes of uranium consumed yearly by the world's entire nuclear reactor fleet.[6]

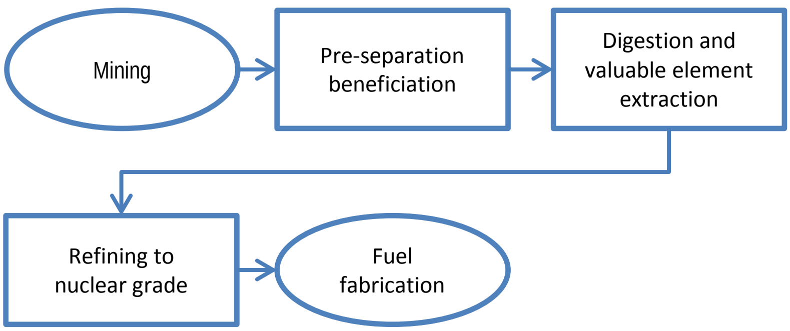
5. Uranium resources depend on the extraction price and are currently estimated in the order of 5 million tonnes recoverable at a cost of less than USD 130/kgU.

6. Around 7 000 metric tonnes of uranium are used yearly by the currently installed world nuclear capacity (376 GWe). The demand for rare earth elements was estimated to be 200 000 metric tonnes in 2014. For monazite alone, the *United States Geological Survey Minerals Yearbook* for thorium (2004) mentions 5 700 metric tonnes of annual monazite production (with around 0.1 Th/REE content), mostly as a by-product of ilmenite from the beach sands of India. Since about 7 million metric tonnes of titanium is recovered annually, the potential could exist for two or three orders of magnitude greater thorium production than currently observed.

Mining

Thorium is generally easier to mine than uranium. One of the advantages of thorium mining is that thorium can be extracted from open pit monazite deposits (presently, the main source of thorium), which are easier to mine than most uranium-bearing ores. Management of thorium mine tailings is also simpler because of the much shorter half-life of one of its daughter products, ^{220}Rn (55 sec), compared to the equivalent daughter product of uranium, ^{222}Rn (3.8 days). However, the radioactivity of the mined products is much higher for thorium than for uranium, because of the thorium decay chain product ^{208}Tl, which emits 2.6 MeV gamma rays.

Figure 2. Overview of thorium recovery process



The uranium mining industry produces large amounts of unused depleted uranium, which is mostly ^{238}U. If a thorium mining industry were to be developed, the volumes of thorium needed to be mined would in all likelihood be significantly lower than the volumes involved for mineral uranium today, and even lower in the case of higher conversion or breeding cycles that optimise the use of thorium.

The overall abundance of thorium is not an issue being considered in any nuclear energy development scenario for the foreseeable future. Even if thorium-based fuel cycles were to be aggressively pursued worldwide, the quantities of thorium that emerge today as a by-product of the extraction of other minerals (rare earth elements, titanium) would be able to provide more than ample quantities of thorium for its use in the nuclear industry for this century and likely beyond.

Fuel cycle aspects

Fuel fabrication and testing

The technical feasibility of using thorium as a fuel component in current power reactors has been shown in numerous studies. Thorium dioxide (thoria) has material properties (low-thermal expansion coefficient, high-thermal conductivity and high-melting temperature) that make it well-suited for use as a fertile fuel matrix in present reactors and for consuming plutonium or transmuting transuranic nuclides, especially as compared with the uranium dioxide currently used in MOX fuels. Thoria-based fuels for light water reactors (LWRs) and heavy water reactors (HWRs) show a potential for improved fuel performance in terms of reduced fission product release and reduced erosion, which lead to fewer fuel defects. Test irradiation programmes of thoria fuels have been carried out in the past,[7] are currently on-going[8] or are foreseen[9] to determine the key properties and behaviour of thorium dioxide fuels, such as thermal conductivity, swelling and fission gas release as a function of burn-up. The results constitute a significant step towards the broader use of thoria fuel ceramics in existing reactors.

Thorium-based fuels would need to be properly qualified to assure their safe performance in the usual suite of normal and accident scenarios of prime concern to regulators. Although no major obstacles have been found thus far, processes have to be further developed to manufacture thorium-based fuels at an industrial scale.

Performance of newly developed fuels would then need to be confirmed in further test programmes in agreement with licensing authorities, taking into consideration today's heightened safety requirements. The introduction of thorium in a reactor core would most likely follow a progressive approach – single fuel rod irradiation tests, followed by irradiation of test assemblies, leading to the introduction of thorium fuel in several assemblies or reload batches in the core.

A recent report by the United States Nuclear Regulatory Commission (NRC) on the *Safety and Regulatory Issues of the Thorium Fuel Cycle* (NRC, 2014), highlights that thorium's "…fundamental nuclear properties have impacts on a number of key areas related to reactor and safety analyses, including steady state and transient performance, fuel handling and management (fresh and irradiated), reactor operations and waste management. The uncertainties in relation to these data and the resulting impact on key safety parameters need to be fully evaluated" (Ade et al., 2014). Such evaluation processes, even for existing infrastructures, would be resource- and time-consuming.

Reprocessing and re-fabrication of thorium-based fuels

The reprocessing of thorium-based fuel cannot be achieved with the processes that are currently used for uranium-plutonium fuel. The envisaged solution is to use the THOREX (thorium recovery by extraction) liquid-liquid extraction process, developed from the PUREX (Pu and U recovery by extraction) process concepts used for uranium-plutonium fuel. The THOREX process (and its variants) has been successfully used at pilot scale, notably in the United States to reprocess about 900 tonnes of thorium fuel (~1.5 tonnes ^{233}U) and also in France. However, the dissolution of thorium metal or oxide has proven to be more complex than for uranium and requires the addition of hydrofluoric and nitric acids. The corrosiveness of these acids with respect to the process equipment must be buffered by adding aluminium nitrate. Such specific processes are yet to be developed at an industrial scale and would ultimately require further development of the dissolution, the liquid-liquid extraction and the conversion steps. The influence of the burn-up of the fuel and the impact of the conversion and fabrication processes, as well as the fuel microstructure, would also require further investigation.

7. In particular, at AECL for thoria fuels in HWRs. For LWRs, the (2000-2005) Obrigheim $(Th,Pu)O_2$ PWR test programme, or the German-Brazilian "Thorium Utilization in PWRs" programme in the 1980s.
8. The currently (2014) ongoing IFE/Thorium Energy thorium fuel rods irradiation campaign at the OECD/NEA Halden Reactor.
9. An Areva/Solvay R&D collaboration on thorium fuels was announced by both companies in early 2014.

THOREX has yet to reach the maturity of the commercial PUREX process. Currently, no alternative to THOREX exists for reprocessing thorium-based fuels, although other extractants have been investigated. Additional innovative reprocessing methods under study (i.e. fluoride volatility) could be developed, but full industrial reprocessing of thorium fuels can nevertheless only be envisaged, at the earliest, in the medium to long term.

A major challenge associated with thorium reprocessing from solid fuels is related to the unavoidable presence of ^{232}U, which accompanies ^{233}U. One of its decay products, ^{208}Tl, is a 2.6 MeV gamma emitter that requires substantial changes in the downstream fuel fabrication process, because it cannot be handled in normal glove boxes, as is the case for (U, Pu)O_2 MOX fuel.

A recycle fuel fabrication process, remotely operated within a fully shielded facility, must be developed and demonstrated for thorium fuels. This will present very large technological and commercial hurdles associated with significant costs.

Different options for thorium implementation

While the longer-term options of thorium use aim explicitly at the replacement of uranium-plutonium in a "pure" Th/^{233}U fuel cycle, such solutions would need to transit through a period where thorium, together with the ^{233}U produced, is gradually introduced into the fuel cycle.

A full transition to a breeder thorium fuel cycle is undoubtedly a long-term process. India, a country that for strategic reasons of energy independence has made great efforts at a national level to fully develop and deploy the thorium fuel cycle, does not envisage reaching a self-sustained Th/^{233}U cycle before 2070 (Vijayan, 2013; Bhardwaj, 2013). The three-stage development programme supported by India is an example of the long-term staging of thorium introduction, limited not only by fissile material availability but also by the level of readiness of thorium breeder technologies.

It is important to differentiate the different categories of potential thorium implementation in reactors, both in the short (before 2030), medium (2030-2050) and longer term (post-2050). These different uses of thorium share very different development challenges and advantages.

Short term

Thorium as an additive to the uranium-plutonium fuel cycle

In the short term (short by industrial timescales, i.e. before 2030), the introduction of thoria in small quantities as an "additive" in uranium-plutonium fuels can be considered as a practical option. A typical 5% to 10% mass fraction content of thoria in uranium-plutonium oxide fuels could be a means to improve the neutronic characteristics of PWR fuel assemblies, allowing for better core power flattening as well as a reduction in the use of burnable poisons such as gadolinium.

While this use of thorium surely does not address global objectives such as uranium savings, fissile material management or improvements in ultimate waste management, it may well be part of an initial step towards thorium use in generation-III or III+ reactors and could augment the irradiation experience towards higher burn-ups with thorium in LWRs or HWRs.

An important benefit that may emerge from thorium implementation in today's reactors is the accumulation of technological knowledge and operational experience, which could potentially be transposed to evolutionary reactor designs in the medium term.

Current industrial development activities for this option remain, however, somewhat limited, given the likely burden of licensing work and the lack of clear economic incentives for nuclear power plant operators to pursue this route.

Medium term

Thorium in generation III or III+ reactors as a complement to the uranium fuel cycle

Most likely over the next 20 years, new nuclear power plants under construction will nearly all be water-moderated technologies designed to last for about 60 years. Light water reactors will therefore continue to dominate nuclear energy production for most of the 21st century, with heavy water reactors also present in the market and showing promise as potential hosts for thorium fuels.

A 2011 NEA report on *Trends towards Sustainability in the Nuclear Fuel* Cycle (NEA, 2011) concludes that "the successful large-scale reactor technology demonstration efforts conducted in the past suggest that there should not be insurmountable technical obstacles preventing the use of thorium fuel and its fuel cycle in the existing and evolutionary LWRs. However, the industrial infrastructure, research, design and licensing data are not in place to allow a rapid deployment of thorium fuels in current reactors in the short term".

A medium-term (2030-2050) option would be to use thorium in existing systems – in either LWRs (i.e. PWRs, BWRs) or HWRs (i.e. CANDUs). Thorium use could be considered, for example, both in a homogeneous and a heterogeneous manner through the following means:

- mixing thorium with uranium and/or plutonium oxides as a homogeneous fuel;
- using fuel assemblies with a combination of uranium and/or plutonium oxide fuel rods and thorium oxide fuel rods;
- using a heterogeneous core approach, with the introduction of thorium fuel assemblies separated from the rest of the assemblies (UOX or MOX).

In the case of existing reactors, thorium would likely be used either as pure thoria in heterogeneous assemblies or cores, or with different combinations of thorium in homogeneously mixed oxide form with either low enriched uranium (LEU, enriched uranium containing less than 20% of the isotope ^{235}U), or plutonium (of adequate isotopic quality). Highly enriched uranium (HEU, enriched uranium containing 20% or more of the isotope ^{235}U) or pure ^{235}U would generally not be considered because of the worldwide consensus on the ban of their use for proliferation reasons.

Various studies have indicated that a workable approach for thorium use in LWRs could be via the heterogeneous option. Given the not-yet-proven industrial feasibility of mixed thorium-uranium-plutonium reprocessing schemes, the heterogeneity option would initially be at the level of fuel assembly, where the thorium and ^{233}U are part of the uranium-plutonium cycle, but without the development of a dedicated ^{233}U reprocessing/re-fabrication route.

Because of the current lack of reprocessing facilities and industrial processes for reprocessing thorium fuels, any near- to medium-term use of thorium fuels will likely be implemented in a once-through mode. In general, once-through fuel cycles using thorium would aim at achieving increased burn-up of the fuel, since the ^{233}U will be burnt *in situ* rather than recovered. In this respect, evolutionary generation III+ variants of PWRs, BWRs and HWRs could take advantage of such an improved utilisation of fissile resources.

Towards higher conversion

Hardening the neutron spectrum in the reactor is the principle behind high conversion in reduced-moderation generation III+ evolutionary variants of PWRs, BWRs and HWRs. These concepts have implemented modifications to their respective base designs that, while not allowing them to achieve breeding ratios of 1, could nevertheless lead to higher conversion ratios using thorium-based fuels, due to their inherently favourable neutron economy in thermal or epithermal neutron spectra. ^{233}U bred by thorium – which has the highest number of spare neutrons available for breeding at these neutron energies - is well-suited to these evolutionary designs, notably in the slightly modified, tighter fuel lattices of HWRs or BWRs such as modified CANDUs (Bromley, 2004; Bromley and Hyland, 2014) or potentially in resource-renewable BWR designs (RBWRs) (Ganda et al., 2012).

Even if high conversion does not reach breeding, achieving higher conversion ratios would still be desirable in view of the better utilisation of natural resources. A rapidly growing generation III or III+ nuclear reactor fleet could thus benefit from the use of thorium in parallel to uranium so as to improve their overall fissile material balance and the management of spent fuel through (multi-)recycling of uranium, and particulary plutonium. These types of strategies could become attractive options in cases where fast neutron reactor deployment, in synergy with generation III reactors, does not occur, is not sufficiently deployed or is compromised for any reason.

This potential can be further increased if multi-recycling of thorium fuel is implemented. Schemes with thorium facilitating the recycling of reprocessed uranium in CANDUs, or improving the recyclability of plutonium in LWRs, are under study and are deemed realisable in the medium term provided reprocessing challenges can be met.

Thorium-plutonium fuels in mixed oxide forms (Th-MOX) have long been considered a good option for plutonium disposal, as no significant quantities of new "second-generation" plutonium are produced through neutron irradiation of thorium. It should be noted, however, that the presence of MOX could, in some scenarios, result in an increase of the production of transuranic elements in the fuel, particularly compared to UOX systems. Nevertheless, Th-MOX provides a credible option for recycling of plutonium in the medium to longer term, provided the reprocessing challenges are met at an industrial level.

In general, the introduction of spent fuel recycling for thorium-based fuels would open up many variants of fuel cycle options and several scenarios can be envisaged in the medium to long term for a transition to the longer-term goal of establishing a full Th/^{233}U closed cycle in dedicated reactors.

It should be highlighted, however, that plutonium availability would be the principal limiting factor in the deployment of thorium-plutonium or thorium-uranium-plutonium cycles, along with long transition periods, which could typically be of the order of two reactor generations or over 100 years (Vallet, 2012).

Long term: Dedicated breeder systems using Th/^{233}U closed fuel cycles

Longer-term options (i.e. post-2050) for thorium implementation may investigate an increased use of thorium by using combinations of reactor types or dedicated breeder reactor systems to establish a full Th/^{233}U fuel cycle. All these longer-term options would require fissile material – either plutonium of adequate fissile quality from the existing uranium-plutonium fuel cycle or ^{233}U bred during a transitioning period – in order to be feasible.

All advanced applications of thorium as nuclear fuel would require significant research and development in thorium-based fuel technology, as well as proper qualification of the fuel.

An autonomous thorium fuel cycle is only possible if the ^{233}U is recycled. Advanced fuel cycle options in particular will depend on successful development of processes associated with spent thorium fuel reprocessing and re-fabrication of irradiated fuels with ^{233}U, including appropriate consideration of radiation protection and non-proliferation issues. These options would also depend on the successful implementation of the THOREX reprocessing method at industrial scales.

Generation-IV concepts with thorium

The Generation IV International Forum (GIF) has chosen molten salt reactors (MSRs) as one of its six concepts; the only one that specifically considers the use of thorium. MSR concepts implement very innovative fuel management approaches with the use of fuel (thorium- or uranium-based) in liquid form, which in principle allows continuous "online" reprocessing of the fuel in order to extract fission products and ^{233}Pa (the precursor of ^{233}U and a neutron absorber). Liquid fuel concepts, in principle, allow greater power densities and smaller initial fissile inventories than solid fuel concepts. The online liquid fuel management of MSRs allows for theoretical breeding ratios equal or greater than one.

In its latest *Technology Roadmap Update for Generation IV Nuclear Energy Systems* (GIF, 2014), GIF has extended the viability study phase of MSR concepts until 2025, reflecting the fact that MSR concepts are still in need of substantial development before they are deemed technologically feasible. Particularly challenging is the essential step of the online treatment of liquid fuel, which requires the implementation of pyro-chemical processes. Much, however, remains unknown about the actual feasibility of these processes or performances. In addition, safety analysis methods in their current form cannot be applied to liquid-fuelled MSRs, because of the innovative form of the fuel (i.e. absence of cladding, molten fuel conditions under normal operation, continuous circulation of fuel in and out of the active core). Development of methodologies for the design and safety evaluations of liquid-fuelled MSRs is also necessary (Serp et al., 2014).

In 2011, China announced the start of an ambitious R&D programme on molten salt reactors led by the Chinese Academy of Sciences (CAS) aiming, in particular, at the construction of a 2-MW pilot thorium molten salt reactor-liquid fuel (TMSR-LF). In 2014, the TMSR-LF prototype was in a pre-conceptual design phase with limited use of thorium in the foreseen candidate compositions for the molten salt fuel.

Large R&D and technology developments are still needed to exploit the potential benefits of thorium-fuelled molten salt reactors, whether fast or thermal.

Hybrid reactor concepts

Other, even more innovative hybrid reactor concepts that combine the characteristics of different future reactor concepts – namely, hybrids of accelerator-driven systems with molten salt reactor systems, hybrids of fission and fusion reactors – have been envisaged as potentially making use of thorium. Although these concepts may have interesting theoretical properties, they inevitably reflect the disadvantages, uncertainties and unknown factors related to the various base technologies that compose them. These unknown elements are often independent from the fact that the concepts may or may not use thorium and, as such, would first need to be further studied, developed and demonstrated. Consequently, such composite or "hybrid" concepts are very unlikely to provide any credible application for commercial electricity production in this century.

Waste management aspects

The use of thorium as nuclear fuel is often associated with advantages in the radiotoxicity of the resulting waste as compared to conventional uranium fuels. It must not be overlooked, however, that the implementation of thorium fuels with the view of developing a self-sustainable thorium fuel cycle will require the use of mixed fuel forms (thorium-LEU or thorium-plutonium fuels) during very long transition phases before a full Th/^{233}U cycle can be achieved. For these mixed fuel forms, a comparison in terms of advantages or disadvantages over current UO_2 fuels will strongly depend on the mixed fuel form considered and on fuel management and recycling strategies.

While a pure Th/^{233}U cycle will indeed produce considerably less plutonium and minor actinides than conventional UO_2 fuels, this is not the case for thorium-plutonium mixed fuel forms, and is less clear for thorium-LEU fuels. Furthermore, decay products from ^{233}U drive radiotoxicity to a higher level than that of LEU or U/Pu for the period between about ten thousand years and one million years, primarily due to the presence of ^{234}U (mainly produced through neutron capture on ^{233}U) and its decay product, ^{226}Ra. The relative differences between radiotoxicities resulting from the use of both cycles vary greatly depending on the recycling strategies and recycling efficiencies considered and must therefore be interpreted with care.

Irradiation of thorium fuel also gives rise to specific long-lived heavy radionuclides, some of which have a more important radiotoxicity than their counterparts in the uranium cycle. The most prominent are:

- ^{233}U with a half-life of about 160 000 years. It has a daughter product (^{229}Th), which contributes significantly to the radiotoxicity of the spent fuel. This difference becomes much less noticeable if ^{233}U is recycled.
- ^{231}Pa with a half-life of 33 000 years. It has a particularly high radiotoxicity, even higher than the principal isotopes of plutonium (^{239}Pu and ^{240}Pu).
- ^{232}U with a half-life of 70 years. The main burden comes from its decay products (^{212}Bi and ^{208}Tl), which are highly radiant (gamma radiation of 1.6 and 2.6 MeV) and which would impose heavy shielding requirements in recycling plants.

In terms of the potential reduction of the radiotoxic inventory of the waste compared to uranium-plutonium fuels, the benefits of a self-sustaining Th/^{233}U fuel cycle at equilibrium are more modest when considering the transition needed to establish that equilibrium cycle.

The long-term radiotoxicity of thorium-based spent nuclear fuels is more accurately described as being comparable to that of uranium-based spent nuclear fuels.

Non-proliferation aspects

The non-proliferation of nuclear weapons has always been an essential consideration to take into account in the deployment of any nuclear energy technology or process. This issue was addressed in particular in the framework of an extensive study conducted between 1978 and 1980 on the overall nuclear fuel cycle (IAEA, 1980). A general conclusion reached in this report was that the technical obstacles to military use of thorium cycles with uranium enriched to less than 20% are similar to those of the uranium-plutonium cycle. The fissile nuclide ^{233}U, produced in thorium fuel cycles, is categorised under the same basis as plutonium (see the IAEA Convention on the Physical Protection of Nuclear Materials).

There are four key physical characteristics to take into account when assessing the difficulty of using fissile materials for military purposes:

- The critical mass of a bare homogeneous sphere of fissile material, that is without a neutron reflector.
- The spontaneous neutron emission, which should be as low as possible to guard against the effect of pre-detonation. For highly neutron-emitting fissile materials, it becomes practically impossible to design a "reliable" weapon.
- The intrinsic radiation heat generated within the fissile material itself. Excessive heat can complicate the process of making a weapon or jeopardise its operation.
- The levels of external radiation associated with the fissile material must be considered because of the impact on radiological protection of the personnel handling these materials, as well as for reasons of potential radiation damage to electronic components.

The following remarks can be made based on these criteria:

- The mass of ^{233}U needed to make an atomic weapon is not altogether different from the mass required to make a plutonium weapon, as the critical masses of ^{233}U and plutonium are fairly close.
- The most important difference between uranium (either ^{233}U or ^{235}U) and plutonium originates from spontaneous neutron emission, which in plutonium is mainly due to the isotope ^{240}Pu – present in more or less significant proportions depending on the origin of the plutonium – and in particular the burn-up of the fuel from which plutonium was extracted.
- With ^{233}U, it is possible, due to its very low spontaneous emission of neutrons, to produce a simpler type of weapon than with plutonium, which requires the use of a much more complex implosion device. This simplification in the case of ^{233}U can, however, be mitigated by the high alpha activity of ^{232}U, which could trigger (alpha, n) reactions on light elements that may be present in trace amounts in the fissile material, causing an unwanted emission of neutrons. Nevertheless, this process would produce far fewer neutrons than the spontaneous emission of neutrons from ^{240}Pu, and its effects can be minimised by reducing the levels of contamination in light elements.
- Heat emission from ^{233}U is generally lower than plutonium and does not pose a major problem.

In summary, it is possible to make an atomic weapon with ^{233}U, and such devices have in the past been fabricated and detonated.

Self-protection from ^{232}U

The presence of ^{232}U (specifically in the case of thorium-based fuel cycles) is often cited as providing self-protection against proliferation. This is primarily due to the decay products of ^{232}U, which are strong gamma emitters. ^{208}Tl, for example, upon decay emits a 2.6 MeV gamma ray, leading to significant shielding and remote handling needs for any reprocessing activity.

However, the degree of proliferation resistance that this gamma field provides is in reality heavily dependent upon the threat scenario, in other words on the facilities available to the proliferators, their financial capacity to build or otherwise acquire shielded facilities, and conversely, their willingness to expose themselves or personnel handling these materials to radiation doses.

Economic aspects

Given that the thorium cycle has never been deployed at an industrial scale, there is no accurate data on the costs associated with the different stages of this cycle. There is no supply chain of thorium and consequently no indexed thorium market. It is therefore difficult to provide meaningful cost projections for any system using thorium, but it is nevertheless possible to draw some general conclusions by estimating costs as compared to conventional uranium-plutonium fuel cycles at different stages.

The case of a thorium fuel cycle can be qualitatively compared to the French reference case, where a closed uranium-plutonium fuel cycle is implemented on a large industrial scale (see table below).

Cost breakdown of the nuclear fuel cycle in France

Uranium	24.6%
Uranium conversion	3.3%
Uranium enrichment	21.3%
Fuel fabrication	16.4%
Interim storage and reprocessing of spent fuel + recycling	26.2%
Final disposal of ultimate waste	8.2%

Estimates based on data from the Ministère de l'économie et des finances (France), 1997.

Considering the different aspects of the above breakdown, the following remarks can be made.

In terms of the raw material, the comparison of uranium with thorium cannot be based on a market price, since such a market price does not currently exist for thorium.[10] It could be necessary to seek new resources once available stocks of thorium are exhausted. However, as thorium would be extracted together with other marketable materials (such as rare earth elements or titanium, for instance), the price would probably be much lower than that of uranium, especially as exploitable deposits are mostly open air, which facilitates the recovery of minerals.

The enrichment step is not applicable for a closed thorium cycle in equilibrium, but must still be considered for the transition phase if the fissile material used initially is in the form of LEU. In such cases, in particular for LEU at close to 20% enrichment, it would generally be necessary to feed the cycle with significantly larger amounts of natural uranium and separative work units (SWU) than for the standard uranium cycle for the same energy output, which would be a significant drawback in terms of the general optimisation of uranium consumption. The amount of this surplus largely depends on, for example, reactor type, fuel burn-up and fuel management schemes. For thorium-based cycles using recycled fissile material (plutonium or ^{233}U), the cost associated with these materials heavily depends on the back end of the chosen fuel cycle.

When considering the fabrication or re-fabrication of fresh or recycled thorium-based fuels, it is necessary to distinguish the type of fissile material chosen:

- If LEU is used as fissile material in once-through cycles, two different kinds of nuclear materials (thorium and LEU) would have to be managed, which leads to additional

10. In fact, there presently exists a stock of about 25 000 tonnes of thorium in the world, which would likely be initially traded at low prices, as they are mainly considered a storage burden for the industry or organisations owning them.

costs compared to the manufacture of standard enriched uranium fuels. This is particularly the case for the different types of heterogeneous fuel management options being considered in different reactor types, where thorium and uranium are physically separated. Specific studies related to homogeneous thorium-uranium fuel cycles in light water reactors have shown that these fuel cycles are not economically competitive over conventional uranium cycles with current fuel management strategies (Joo et al., 2003).

- If plutonium is used with thorium in once-through cycles, processes should not be very different from those that are used today in the manufacture of MOX fuel, and therefore the costs should be comparable.
- If the fissile material is ^{233}U, which implies closing the fuel cycle, the presence of the radioactive daughter products of ^{232}U would require operation behind radiological protection. Recycle fuel fabrication would certainly generate very significant additional costs, which are difficult to estimate since there are no technically and economically proven processes and equipment that have been developed and demonstrated for remotely-operated recycle fuel fabrication within a fully shielded and contained facility, especially at the large industrial scale that would be needed. Essentially, no development has been done at the pilot scale with uranium containing several thousand ppm of ^{232}U.

The final step in the back end of a closed nuclear cycle is the final disposal of medium- and high-level long-lived radioactive waste, for which the solution adopted almost universally is that of a deep geological repository. There is no reason to believe that there is a significant economic difference between uranium and thorium cycles in this final stage, especially as it is assumed here that only residual waste from reprocessing is disposed of after separation of all recyclable materials.

Conclusions

If the use of nuclear energy were to expand significantly in the world, some of the major challenges to be addressed would include improving the utilisation of mineral resources while reducing ultimate waste streams. The large-scale deployment of fast neutron reactors is still uncertain and the realisation of geological repositories has been delayed in some countries. In the absence of fast neutron reactors, the issue of plutonium management would have to be dealt with in the medium to long term.

The use of thorium in the nuclear fuel cycle as a complement to the uranium-plutonium cycle may have potential for improving the medium-term flexibility of nuclear energy and its long-term sustainability. More specifically, the following options for its introduction into the nuclear fuel cycle could continue to be investigated:

- the use of thorium as a means of burning plutonium (and possibly other higher actinides) as an option for plutonium management;
- the possibility of reaching higher conversion factors in thermal or epithermal neutron spectra in evolutionary generation III+ systems that use thorium-based fuels, with the aim of recovering the fissile material from the used fuel;
- the promising physicochemical characteristics of thorium-dioxide, which may allow improved performance of thoria-based fuels over current fuel designs.

It must be noted, however, that the development of new fuels or new reactor concepts is a time- and resource-consuming process likely to span several decades. Furthermore, any industrial application of thorium as a nuclear fuel would continue to require the input of fissile material from the existing uranium-plutonium cycle until the required amounts of ^{233}U could be produced and ultimately make the thorium cycle self-sustaining.

Until this point, an important factor governing the rate at which ^{233}U could be produced from the introduction of thorium-plutonium or thorium-uranium-plutonium cycles is plutonium availability. The limitations imposed by fissile plutonium availability result in rather long transition periods between thorium-plutonium and Th/^{233}U systems, which are likely to be of the order of many decades.

The development of a fully self-sustaining Th/^{233}U cycle would also require the development of industrial scale reprocessing capabilities to recover the ^{233}U from spent fuel, along with fuel fabrication facilities to prepare the material for reuse. In particular, impediments to closing the thorium fuel cycle may arise from the following issues:

- To date the THOREX process has been demonstrated in pilot-plant facilities but has yet to reach the maturity of the commercial PUREX process. Other extractants and alternative processes (e.g. fluoride volatility) are also being investigated but are still at a conceptual stage.
- A major challenge associated with the recycling of ^{233}U is the presence of radioactive ^{232}U. Remotely-operated and fully shielded recycled fuel fabrication processes would likely be needed, for which there are currently no specific equipment or proven processes at the industrial scale.

Ultimately, thorium technologies require significant further development. Given their cost and the lack of clear economic incentives for nuclear power plant operators to pursue this route, industrial development activities for thorium remain somewhat limited at this time. Even if these economic incentives were to appear, it is to be expected that short- to medium-term development of thorium fuels would be carried out in a step-wise fashion and in synergy with the existing uranium-plutonium fuel cycle.

In the longer term, the potential introduction of advanced reactor systems may present an opportunity to realise the full benefits of a closed Th/^{233}U fuel cycle in dedicated breeder reactors (generation IV or beyond) that are still in the design and study phase. Molten salt reactors in particular may offer the prospect of using thorium fuels with online recovery and re-use of the ^{233}U while recycling long-lived actinides and ensuring minimal losses to the final waste stream. It must, however, be recognised that the development, licensing and construction of such novel systems is a long-term undertaking.

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Perspectives on the Use of Thorium in the Nuclear Fuel Cycle

Since the beginning of the nuclear era, significant scientific attention has been given to thorium's potential as a nuclear fuel. Although the thorium fuel cycle has never been fully developed, the opportunities and challenges that might arise from the use of thorium in the nuclear fuel cycle are still being studied in many countries and in the context of diverse international programmes around the world. The full report, *Introduction of Thorium in the Nuclear Fuel Cycle: Short- to Long-term Considerations*, provides a scientific assessment of thorium's potential role in nuclear energy both in the short to longer term, addressing diverse options, potential drivers and current impediments to be considered if thorium fuel cycles are to be pursued.

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NEA No. 7228

The full publication NEA No. 7224
is available at: oe.cd/14K