Thorium for fission power

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Note

In this document the references are coded by Q-numbers (e.g. Q6). Each reference has a unique number in this coding system, which is consistently used throughout all base papers by the author. In the list at the back of the document the references are sorted by Q-number. The resulting sequence is not necessarily the same order in which the references appear in the text.

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Thorium

Thorium is a radioactive metal, 3-4 times more abundant in the Earth's crust than uranium. Thorium has a characteristic terrestrial isotopic composition, consisting for nearly 100% of Th-232, and traces of other isotopes, for example Th-228 and Th-230.
Thorium is a highly reactive metal. At standard temperature and pressure, thorium is slowly attacked by water, but does not readily dissolve in most common acids, the exception being hydrochloric acid. It dissolves in concentrated nitric acid containing a small amount of catalytic fluoride or fluorosilicate ions; if these are not present, passivation can occur. At high temperatures, it is easily attacked by oxygen, hydrogen, nitrogen, the halogens, and sulfur. It can also form binary compounds with carbon and phosphorus.
Finely divided thorium metal presents a fire hazard due to its pyrophoricity and must therefore be handled carefully. When heated in air, thorium turnings ignite and burn brilliantly with a white light to produce the dioxide. In bulk, the reaction of pure thorium with air is slow, although corrosion may eventually occur after several months; most thorium samples are however contaminated with varying degrees of the dioxide, which greatly accelerates corrosion. Such samples slowly tarnish in air, becoming gray and finally black.
(https://en.wikipedia.org/wiki/Thorium)

None of the thorium isotopes is fissile. The concept of thorium as nuclear fuel is based on the conversion by neutron capture of non-fissile thorium-232 into uranium-233, which is as fissile as plutonium-239. Consequently the application of thorium as fuel for nuclear power requires a special nuclear system. We will return to this issue below.

Promised advantages

According to its advocates thorium fuelled nuclear power would have significant plus points, such as:

- Thorium reactors are cheap and can be assembled in factory-made modules
- Thorium reactors can be designed as inherently safe power reactors.
- Thorium power does not poses proliferation risks, because a thorium reactor produces no transuranic actinides.
- Thorium power produces less radioactive waste than from uranium-fuelled NPPs, because no long-lived actinides are formed and only short-lived radioactive waste (fission products) are produced.
- Thorium is 4 times more abundant in the earth's crust than uranium.

During the 1960s, 1970s and 1980s the nuclear industry asserted that natural uranium could be 100% fissionable in fast breeders. In later years this claim has been reduced to 60% or 50%, still nearly a 100 times as much than in a conventional reactor. This promise turned out to be infeasible, because the breeder cycle as envisioned cannot be realized, as follows from the Second Law of thermodynamics.

Some people believe that thorium could be completely fissioned. In their view thorium reactors would open a limitless energy source for mankind, reviving the nuclear dreams from the early 1960s, which were based on the U-Pu breeder concept. These optimistic stories entirely pass over above mentioned inherent difficulties and particularly the Second Law of thermodynamics, see also reports m01 *Uranium-plutonium breeder systems* and m38 *Nuclear power and the Second Law*.

2 Uranium-233

Uranium-233 does not occur in nature, due to its relatively short half-life of 158000 years. Uranium-233 is a fissile nuclide that is prepared from non-fissile thorium -232 by neutron irradiation in a nuclear reactor. After irradiation the thorium target elements are to be reprocessed to separate the U-233 from the remaining Th-232.
Among a number of other countries, the USA conducted Th-232/U-233 research in the 1950s and 1960s (e.g. in the Shippingport reactor). In the USA, and probably also in the former Soviet Union, U-233 has been envisioned as fuel in very compact military reactors (to be transported in containers) for special applications, e.g. for electricity generation on remote locations, in large (military) spacecraft, in nuclear-powered rockets, in nuclear ramjets of an atomic bomber, but also for civil power reactors. These technical developments were halted in the 1970s, apparently due to various problems. One of these problems is the presence of uranium-232, a strong gamma-emitter, which makes U-233 difficult to handle. Methods to limit the content of U-232 are expensive.

Uranium-233 has a critical mass much less than U-235 and is comparable to plutonium in terms of weapons-usability. Between 1955 and 1968 several nuclear weapons test were conducted using uranium-233 [Alvarez 2012] Q594. In 1998 India detonated a very small device based on U-233 called Shakti V [WNA-Th 2015] Q302.
In the United States about 1550 kg of U-233 was separated. Of this amount about 123 kg may be unaccounted for, enough for some 13 nuclear explosive devices. The radiation level from contaminants is not considered to be an adequate barrier to prevent a terrorist from making an improvised nuclear device. Storage of the US stockpile of U-233 is a safeguard, security and safety risk. The production of the stockpile also has left a disposal burden [Alvarez 2012].

How is the situation concerning U-233 in elsewhere in the world? Several countries are still involved in the development of a thorium-232/uranium-233 nuclear breeder system.

Safeguards of uranium-233

The safeguard conditions of uranium-233 are not very clear. Regarding the situation in the USA [Alvarez 2012] Q594 states:

> Our nuclear facilities may have done a poor job of keeping track of this dangerous material. Now, the Department of Energy has indicated it plans to waive safety requirements to dispose of it. But if the U.S. government makes a mess, they should clean it up. All uranium-233 should be accounted for, stored safely, and disposed of safely.

Cost

With regard to the cost of U-233 [ORNL-6952 (1999] Q377 reports on page xvii:

> "It is estimated that the original production costs of high-quality U-233 were $2 to 4 million/kg. Low quality material is much less expensive since it can be produced in a light-water reactor (LWR).
> Irradiation service costs (excluding target fabrication and chemical separation costs) to produce U-233 today using the Advanced Test Reactor (ATR) in Idaho are estimated at about $30 million/kg. Because of the shutdown of facilities, the U.S. Department of Energy (DOE) production capability is limited. The ATR, which is the largest DOE reactor currently operating, could produce only about 0.3 kg/year. Only India has a current capability to produce significant quantities of high-quality U-233. Newer production techniques using heavy-water reactors may lower this cost."

3 Thorium breeding cycle

Thorium cycle

Thorium is a radioactive metal, slightly more abundant in the Earth's crust than uranium and with similar chemical properties as uranium. The concept of the thorium reactor is based on the conversion by neutron capture of non-fissile thorium-232 atoms into uranium-233 atoms, which are fissile. The use of thorium as nuclear fuel would imply a breeder cycle, similar to that of the uranium-plutonium breeding cycle.

A thorium reactor would consist of an active reactor core in which fissile nuclides are fissioned, surrounded by a blanket with thorium-232. The active core would generate heat for power generation and the neutrons for the transmutation of thorium-232 into uranium-233. After a given amount of absorbed neutron radiation the material would be removed from the blanket and transported to a reprocessing plant. There the material would be separated into several fractions: remaining thorium-232, newly formed uranium-233 and unwanted nuclides. The thorium would be replaced into the blanket of the reactor and the uranium-233 would be used to fabricate fuel elements for the core of the reactor.

Thorium reactors

Theoretically uranium-233 could be used in all types of nuclear reactors, based on thermal neutrons as well as on fast neutrons.
Several power reactor concepts using thorium have been proposed, including once-through and breeder recycle modes. In the once-through mode thorium would replace uranium-238 in the fuel. A small part of the thorium would be transformed into uranium-233, which would contribute to the fission process. In this way less uranium would be required to generate a set amount of energy. The conservation of the uranium consumption would be minor. If the fuel would be enriched with U-233 a reprocessing stage would be required. This mode resembles the use of plutonium in LWRs.

India is the only country in the world at this moment having an operable experimental U-233 reactor. India has been operating a low-power U-233 fuelled reactor Kamini at Kalpakkam since 1996 – this is a 30 kWth experimental facility using U-233 in aluminium plates (a typical fuel-form for research reactors). Kamini is water cooled with a beryllia neutron reflector. The total mass of U-233 in the core is around 600 grams. It is noteworthy for being the only U-233 fuelled reactor in the world, though it does not in itself directly support thorium fuel R&D. The reactor is adjacent to the 40 MWth Fast Breeder Test Reactor in which ThO_2 is irradiated, producing the U-233 for Kamini [WNA-Th 2015] Q302.

Apparently there exist good reasons not to use U-233 in military reactors or in weapons and to discontinue the research towards the thorium power reactor. The high radioactivity of uranium-233 and unavoidable contamination with unwanted radionuclides impeding the controllability of the fission process in uranium-233 may be an important factor.

Time frame of building up a thorium cycle

Only small quantities of U-233 exist in the world at this moment, the USA has 1710 kg of it in storage, 905 kg of which still contained in spent fuel. Of the U-233 stored in separated form 608 kg have the high isotopic quality necessary for reactors and weapons. Inother publications slighte different figures are mentioned. The U-233 stocks in other countries are unknown. The largest DOE reactor currently operating could produce

only about 0.3 kg/year.

To generate sufficiently pure U-233, special reactors are required, likely not appropiate for use as power reactors. It would take decades to construct these reactors and to generate sufficient U-233 to start up the first operating Th-232/U-233 breeder system. Then it would take 9 doubling times to attain a thorium breeder capacity equalling the current nuclear capacity (370 GWe). Even assuming an unrealistically short doubling time of some 20 years, 9 doubling times would mean a period of two centuries.

In this scenario a perfectly operating breeding cycle is assumed, including the separation processes of the spent fuel and the fuel element fabrication. The high radioactivity of U-233 would demand remote operations of the material throughout all steps of the fuel handling. The monetary costs, but also the energy requirements of the fuel cycle will be high.

Theoretically this period could be shortened, perhaps to some 100 years, if:
1 Today's world nuclear power reactor fleet would be replaced by a new generation of LWRs which are appropiate to breed uranium-233 from thorium; the currently operating power reactors are not.
2 The spent fuel from all power reactors of the world would be reprocessed.
Both assumptions are unrealistic.

Hybrid reactor

A major drawback of the thorium cycle is that a genuine thorium breeder reactor cannot sustain a fission process in itself and breed enough U-233 to achieve a steady state but always need an external accelerator-driven neutron source, or the addition of extra fissile material, such as plutonium ur uranium-235..
Even if 370 GW of thorium breeders would come on line in 2050, their contribution to the world electricity generation capacity would be less than 7% by that time, if we assume a growth of 2% a year of the world electricity demand. The contribution to the total world energy supply would be less than 1%.

Feasibility

Problems hampering the development of a true Th-232/U-233 system include, according to (ORNL-5388 1978 [Q376]):
- the high radioactivity of U-233, which is always contaminated with traces of U-232,
- similar problems in recycling thorium due to the highly radioactive Th-228, a decay product of both Th-232 and U-232
- technical problems, not yet satisfactorily solved, in reprocessing.
- the use of U-233 as fuel requires specially developed reactors.
- by recycling of U-233 its isotopic composition deteriorates, and so its usefulness as fissile material decreases, by the increasing generation of the unfavourable isotopes U-232, U-234 and U-236
- the separation processes needed to recycle fissionable material are inherently incomplete, so significant losses are unavoidable
- the recycling of Th-232/U-233 fuel has yet to be demonstrated
- assumed the recycling would be technically feasible, it is still unknown if the cycle would produce sufficient U-233 to expand the Th-232/U-233 capacity, or even to maintain itself
- due to the increasing radioactivity the separation processes deteriorate and the separation will get even more incomplete
- due to the increasing radioactivity the fuel handling and fresh fuel fabrication becomes increasingly difficult.

Similar problems arise with the uranium-plutonium breeder cycle. After dacades of research, in seven countries and investments of some €100bn the U-Pu breeder proved to be unfeasible.

The feasibility of the thorium breeding cycle is even more remote than that of the U-Pu breeder. This is caused by specific features of the thorium cycle on top of fundamental limitations. The realisation of the thorium-uranium cycle would require the availability of 100% perfect materials and 100% complete separation processes. None of these two prerequisites are possible, as follows from the Second Law of thermodynamics. It can be argued beforehand that the Th-U breeder cycle will not work as envisioned.

An overview of research projects in the past and of advanced thorium reactor concepts is given in [WNA-Th 2015] Q302.

Energy balance

Not mentioned in the numerous publications on the advantages of nuclear power based on thorium are the energy investments required to construct a given thorium system, to operate, maintain and refurbish it during its operational lifetime and to decommission the installations after their useful life ended. It would be questionable if the energy balance of any thorium fuelled nuclear power system could be positive.

4 Molten salt reactors

A molten salt reactor (MSR) is a class of nuclear fission reactor in which the primary nuclear reactor coolant, or even the fuel itself, is a molten salt mixture. MSRs run at higher temperatures than water-cooled reactors for higher thermodynamic efficiency, while staying at low vapor pressure. The nuclear fuel may be solid or dissolved in the coolant itself. In many designs the nuclear fuel is dissolved in the molten fluoride salt coolant, e.g. uranium tetrafluoride (UF_4). An MSR is not typical for thorium-based systems, for it can be fuelled by enriched uranium-235, plutonium and uranium-233. The fluid becomes critical in a graphite core which serves as the moderator. Solid fuel designs rely on ceramic fuel dispersed in a graphite matrix, with the molten salt providing low pressure, high temperature cooling.

The Aircraft Reactor Experiment ARE (1954, 2.5 MW(th)) was primarily motivated by the small size that the design could provide. The Molten-Salt Reactor Experiment MSRE (1965–1969, 7.4 MW(th)) was a prototype for a thorium fuel cycle breeder reactor nuclear power plant. The reactor was fuelled by a mixture of LiF-BeF_2-ZrF_4-UF_4; the breeding blanket of thorium salt was omitted.
As of 2011, the ARE and the MSRE remained the only molten-salt reactors ever operated, according to [MSRwiki 2016] Q693.

Although the concept is not new there is a renewed interest in thorium-fuelled MSR technology in China, Japan, Russia, France and USA, along with other Generation IV reactor technologies. One of the Generation IV reactor designs is a molten-salt-cooled, molten-salt-fuelled reactor; the initial reference design is 1000 MWe. This would mean an upscaling of 7 to 3000 MW(th) from the last operating MSR, 40 years ago.ß

5 Seaborg Wasteburner/ Copenhagen Atomics Wasteburner

The Seaborg Wasteburner [Seaborg 2015] Q695 and Copenhagen Atomics Wasteburner [Copenhagen Atomics 2014] Q694 are similar in design.
Without discussing both concepts in detail, some remarks may be appropiate.

Molten Salt Reactor MSR

As far as known only two small experimental molten salt reactors have ever been operated in the world, both in the USA and both based on enriched uranium-235 as fissile material:

- ARE, Aircarft Reactor Experiment, in 1954, power 2.5 MWth
- MSRE, Molten Salt Reactor experiment, during 1965-1969, power 7.4 MWth

The MSRE was intended to explore the possibilities of an MSR with thorium, but the experiments were finished without thorium in the reactor.
The MSR technology is far from proved, not even a pilot plant has been constructed, let alone a full-scale installation (1000-3000 MWth).

Integrated Fast Reactor IFR

The IFR concept comprises a fast reactor with an integrated reprocessing facility to remove fission products, so the the reactor could operate without removal of spent fuel and loading with fresh fuel.
This concept has never been tested. Some partial laboratory experiments have been conducted to explore some chemical implications of the separation processes.

Th-232/U233 breeding cycle

Thorium (consisting of nearly 100% Th-232) is not fissile and has to be converted into fissile uranium-233 by irradiation with neutrons. This would imply a breeding cycle (see text above). A crucial part of that cycle is the reprocessing of the irradiated fuel, to separate the newly formed U-233 from the remaining thorium-232 and to remove fission products. Reprocessing is a very complicated process. The separation processes comprise chemical and physical equilibria which are governed by the Second Law of thermodynamics. It is principally impossible to achieve 100% perfect separation and to produce 100% pure materials. In practice this means that all fractions after a separation process are impure and that a part of the wanted and unwanted materials will be lost into the waste streams.

Unavoidable contamination

The recovered U-233 would unavoidably be contaminated with highly radioactive non-fissionable isotopes of uranium (U-232, U-234 and U-236). Reprocessing of fuel from a thorium reactor would become more difficult after each cycle, because of the increasing radioactivity. The materials could only be handled by remote control, for reason of the high radioactivity.
Because all uranium isotopes have identical chemical properties, the unwanted isotopes cannot be removed from uranium-233 by chemical separation processes (reprocessing).

Thorium reactor

As far as known there exists only one experimental U-233 reactor in the world: the 0.03 MWth Kamini in India.

Containers

The concept of the Copenhagen Atomics Wasteburner with the reactor and associated installations packed in containers may be derived from the early concepts investigated in the USA during the 1950s of military power units to be used in remote locations. This container concept has been abandoned after the first experiments in the 1950s.

Prototype

The prototype of the Copenhagen Atomics Wasteburner would have a nominal power of 50 MWth. For comparison: the present world operating nuclear capacity is some 370 GWe, or roughly 1 000 000 MWth.

Hybrid reactor

A genuine Th-232/U-233 reactor cannot sustain the fission process: the rate of forming new U-233 nuclides from Th-232 would be lower than the rate of fissioning U-233 nuclides. This implies that a thorium reactor always needs an extra neutron source either by fissioning added U-235 or plutonium, or from an external accelerator-driven neutron source.

Burning actinides

Both Wasteburner concepts claim that transuranic actinides from conventional nuclear fuel, from the existing nuclear power plants, can be burned. This would imply that conventional nuclear fuel has to be reprocessed first, to remove the fission products and unused uranium. Reprocessing of spent nuclear fuel is an extremely contaminating and extremely expensive process.
The transuranic actinides are highly radioactive and very difficult to handle. Moreover spontaneous fission of these radionuclides may make the fission process in a reactor difficult to control.

Reprocessing waste

In both Wasteburner concepts handling of the removed fission products is unclear. Are the gaseous fission products discharged into the air? What happens with the other wastes? How are the removed from the sustem?

Timescale

It would take decades to generate sufficient U-233 in special (non-power) reactors to start up the first thorium reactor. Even if the Th-232/U233 cycle would work as advertised, it would take many doubling times of the breeder to come at the present nuclear capacity. The timescale would have to be measured in centuries.

Energy balance

The energy investments required to construct a given thorium system, to operate, maintain and refurbish it during its operational lifetime and to decommission the installations after their useful life ended might be prohibitive: most likely the system would work as an energy sink instead of an energy source.

Second Law

Examining the crucial components of the Wasteburner concepts, it can be concluded that these systems are inherently infeasible, because they are implictly based on the assumptions of 100% perfect separation processes, 100% pure materials and the absence of ageing processes. These assumptions are in conflict with the Second Law of thermodynamics.

Some other alleged advantages of thorium reactors are disproved by [PSR-IEER 2009] Q617 and [Lovins 2016] Q696.

Epilogue

A quote from Amory Lovins in The Ecologist [Q696] seems to apply to publications such as [Copenhagen Atomics] Q694, [Seaborg 2015] Q695 and [SAMOFAR 2015] Q697:
"The nuclear industry is forever reinventing itself with one brilliant 'new' idea after another, Amory Lovins wrote in this classic 2009 essay. But whether it's touting the wonders of future SMRs, IFRs or LFTRs, the reality never changes: the reactors they are building right now are over time, over budget and beset by serious, entirely unforeseen technical problems."

A quote from Admiral Rickover, quoted by Amory Lovins, seems also to apply to above optimistic publications, and many other, on this subject:
"No new kind of reactor is likely to be much, if at all, cheaper than today's LWRs, which remain grossly uncompetitive and are getting more so despite five decades of maturation. 'New reactors' are precisely the 'paper reactors' Admiral Rickover (mastermind of the US Navy's development of the Pressurized Water Reactor, the PWR) described in 1953:
"An academic reactor or reactor plant almost always has the following basic characteristics:

It is simple.
It is small.
It is cheap.
It is light.
It can be built very quickly.
It is very flexible in purpose.
Very little development will be required. It will use off-the-shelf components.
The reactor is in the study phase. It is not being built now.

On the other hand a practical reactor can be distinguished by the following characteristics:

It is being built now.
It is behind schedule.
It requires an immense amount of development on apparently trivial items.
It is very expensive.
It takes a long time to build because of its engineering development problems.
It is large.
It is heavy.
It is complicated."

References

Q302
WNA-Th 2015
Thorium
World Nuclear Association, updated September 2015
http://www.world-nuclear.org/information-library/current-and-future-generation/thorium/aspx

Q376
ORNL-5388 1978
Abbott L S, Bartime D E, Burns T J, ed.
Interim assessment of the denatured 233U fuel cycle: feasibility and nonproliferation characteristics,
ORNL-5388
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, December 1978.

Q377
ORNL-6952 1999
Forsberg C W & Lewis L C,
Uses for Uranium-233: What should be kept for future needs?
ORNL-6952
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6180, September 24, 1999.

Q594
Alvarez 2012
Alvarez R,
Managing the uranium-233 stockpile of the United States. Questionable conditions of taking inventory, storing, and disposing of a nuclear explosive material,
Institute for Policy Studies, Agust 2012.
‹Bob-Alvarez-Managing-Uranium.pdf›
www.ips-dc.org/reports/managing_uranium

Q617
PSR-IEER 2009
Makhijani A & Boyd M,
Thorium fuel: no panacea for nuclear power,
Factsheet by Physicians for Social Responsibility (PSR) and Institute for Energy and Environmental Research (IEER).
January 2009, updated July 2009.
‹thorium2009factsheet.pdf›
http://ieer.org/wp/wp-content/uploads/2012/04/thorium2009factsheet.pdf

Q693
MSRwiki 2016
Molten Salt Reactor
last modified 5 May 2016
https://en.wikipedia.org/wiki/Molten_salt_reactor

Q694
Copenhagen Atomics 2014
Copenhagen Atomics. Affordable Sustainable Clean Energy.
White Paper.
November 2014
‹ CA_Whitepaper2014.pdf ›
http://www.copenhagenatomics.com/?page_id=1141

Q695
Seaborg 2015
Seaborg Wasteburner. Molten Salt Reactor. Whitepaper
SEAB-WP-2015-001
Seaborg Technologies, March 2015
‹ ppp2015.pdf ›
http://seaborg.co/ppp2015.pdf

Q696
Lovins 2016
Lovins A,
'New' reactors? Same old story
The Ecologist, 12th April 2016
http://www.theecologist.org/essays/2987536/new_nuclear_reactors_same_old_story.html

Q697
SAMOFAR 2015
SAMOFAR project, kicked-off by the Delft Technical University, 24-25 August 2015
see:
http://samofar.eu/samofar-kick-off/
http://samofar.eu/project/
http://samofar.eu/concept/
retrieved 7 May 2016