

Chapter 2: Literature survey

“Creativity is not the finding of a thing, but the making of something out of it, after it is found.”

~James Russell Lowell ~

Overview

This chapter supplies background on thorium as an element, including past applications of thorium, thorium reserves and the mining of thorium. The different reactor types that can accommodate or use thorium-based fuel in their cores are introduced with their basic information. An overview of the past experience on several research and power reactors is given. The material properties, fertile and fissile isotope properties, as well as the decay chain of thorium are discussed for purposes of evaluating thorium as a fuel.

2.1 Background on thorium

This section provides background on thorium, including the applications of thorium in the past, a discussion on thorium reserves and the mining of thorium. The various reactors that can burn thorium-based fuels are introduced.

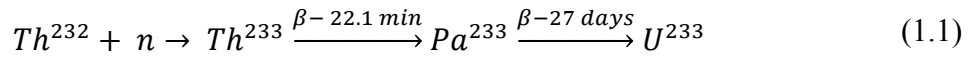
2.1.1 General

Thorium, with the chemical symbol Th, is a metallic element with an atomic number (amount of protons) of 90. The name Thorium originates from the name Thor, the Norse god of thunder or the mythical Scandinavian god of war. This element was discovered between 1828 and 1829 by the Swedish chemist, Jöns Jakob Berzelius (WNA, 2011a; MII, 2011). Thorium is a radioactive element and belongs to the actinide group (MII, 2011). Pure thorium has a shiny, silvery, white colour, but slowly blackens through oxidation in air. Thorium metal

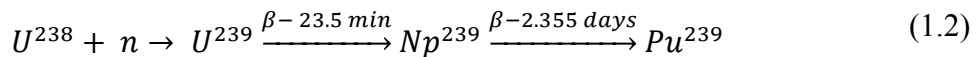
burns radiantly in air with a bright white light. Th^{232} decays slowly, with a half-life three times longer than the Earth's age (WNA, 2011a).

Thorium mainly occurs in the monazite (rare earth-thorium phosphate) mineral, thorite (ThSiO_4), and in thorianite (ThO_2). Thorium and its combinations, particularly thorianite, have abnormally high melting temperatures. Thorium-dioxide is known as one of the oxides with the highest melting point, namely 3300°C (MII, 2011; WNA, 2011a).

Thorium has only one natural occurring isotope, namely Th^{232} . This isotope is fertile and cannot fission. It should therefore capture one neutron to transmute into the fissile isotope U^{233} . The addition of a fissile isotope to Th^{232} is essential in a nuclear reactor, because power and neutrons need to be supplied until a sufficient amount of U^{233} is produced. The fissile candidates are enriched uranium (U^{235}) or plutonium (Trellue *et al.*, 2011). Equation 1.1 explains the conversion reaction from Th^{232} to U^{233} (Lamarsh & Baratta, 2001).



In comparing thorium to naturally occurring uranium, it needs to be said that uranium needs to be enriched to sustain a stable nuclear chain reaction. U^{238} can also capture a neutron(s) to produce fissile plutonium isotopes Pu^{239} and Pu^{241} (Trellue *et al.*, 2011). Equation 1.2 explains the conversion reaction from U^{238} to Pu^{239} (Lamarsh & Baratta, 2001).



Equations 1.1 and 1.2 are similar reactions.

2.1.2 Thorium applications

The commercial uses of thorium include: lantern-shrouds, welding electrodes, ceramics, glassware, and laboratory apparatus (Shultis & Faw, 2002; MII, 2011; WNA, 2011a). During the 19th century people used thorium in healing practices and thorium-dioxide as contrast

material for X-rays (Lamarsh & Baratta, 2001). Due to the high melting temperature, thorium-dioxide was used for coatings on tungsten threads in light bulbs.

Glass with added amounts of thorium-dioxide is used for the production of high quality lenses. Thorium and magnesium are alloyed to produce strong, creep resistant and light metals for aerospace purposes (MII, 2011). Thorium has an application in the petroleum industry and is used as a catalyst in the reaction of ammonia to nitric acid (Rageb, 2011).

Thorium is also investigated as a non-proliferation nuclear fuel source. Thorium fuel cycles were studied during the 1950s, but were not commonly employed due to the achievement of uranium fuel cycles (Trellue *et al.*, 2011). The geographical spread of thorium resources was not as prevalent as uranium reserves in the leading technology countries like USA, Canada and France (Unak, 2000). Another reason for uranium cycles developing faster than thorium cycles is due to the fact that uranium could produce plutonium for weapons.

2.1.3 Reserves

Thorium is assumed to be abundant, due to a half-life three times longer than U^{238} (see Table 2.1). Thorium is believed to be between three and four times more abundant than uranium (Trellue *et al.*, 2011; WNA, 2011a). In the lithosphere, thorium is 2,5 times more abundant than uranium, but unfortunately uranium deposits have been explored more extensively than thorium (Vapirev *et al.*, 1996).

The concentration of thorium in the crust of the earth is approximately three times more than uranium see Table 2.1. According to estimates on the abundance inside the earth's crust, thorium contains more energy than the sum of fossil fuels and uranium (MII, 2011).

Table 2.1 Comparison of abundance properties of naturally occurring thorium and uranium

	Th ²³²	U ²³⁸	Ratio Th/U
T _{1/2} (y)	1,4*10 ^{10c}	4,5*10 ^{9c}	3,14
Solar system abundance (%)	1,09*10 ^{-10b}	2,94*10 ^{-11b}	3,71
Crustal average abundance (mg/kg)	69,6 ^b	2,7 ^b	25,8
Earth's oceans abundance (mg/L)	1*10 ^{-6b}	0,0032 ^b	0,0003
Earth's crust abundance (ppm)	6 ^d , 7,2 ^c , 9,6 ^a	2,7 ^a	2,81

^{a.} (Greeneche, 2010)

^{b.} (Shultis & Faw, 2002)

^{c.} (Lamarsh & Baratta, 2001)

^{d.} (WNA, 2011a)

^{e.} (MII, 2011)

Another major supply of thorium comes from the phosphate mineral, monazite. Monazite can hold up to 12% thorium, but normally ranges between 6% and 7%. The global monazite resources are projected to be approximately 12 million tonnes, where India possesses two-thirds on their south- and east coasts (WNA, 2011a). Monazite would not have been mined for only the thorium content, but also for the need of the rare earths. Other minerals with higher thorium concentrations, such as thorite, (ThSiO₄) would be the main thorium sources when a thorium market has been established (Hedrick, 2007).

The prevalent stores of thorium are inside placer deposits, which are heavy mineral sands deposited by flowing water. Selected amounts of thorium have been extracted from igneous veins and carbonatites (igneous carbonate deposits), which are evaluated to half a million tonnes. Other igneous deposits, like alkaline igneous rocks, have smaller concentrations, but may hold resources of more than 2 million tonnes (MII, 2011; WNA, 2011a). Table 2.2 compares investigated and supposed uranium and thorium reserves at prices below 80\$/kg and between 80 and 130\$/kg.

Table 2.2 Investigated and supposed uranium and thorium deposits in tonnes (Vapirev *et al.*, 1996)

		Uranium	Thorium
Investigated	Below 80\$/kg	14 747 000	645 000
	80-130\$/kg	546 000	398 000
Supposed	Below 80\$/kg	1 604 000	2 256 000
	80-130\$/kg	1 114	489 000

The leading countries in terms of thorium reserves are Australia, Brazil, Canada, Egypt, Greenland, India, Norway, Russia, South Africa, Turkey, United States and Venezuela (MII, 2011; WNA, 2011a). It is also believed that thorium reserves are larger and more spread out over the world (Puill, 2002). Figure 2.1 compares different studies on reported thorium reserves. It should be noted that each study has unique assumptions, which explains the differences.

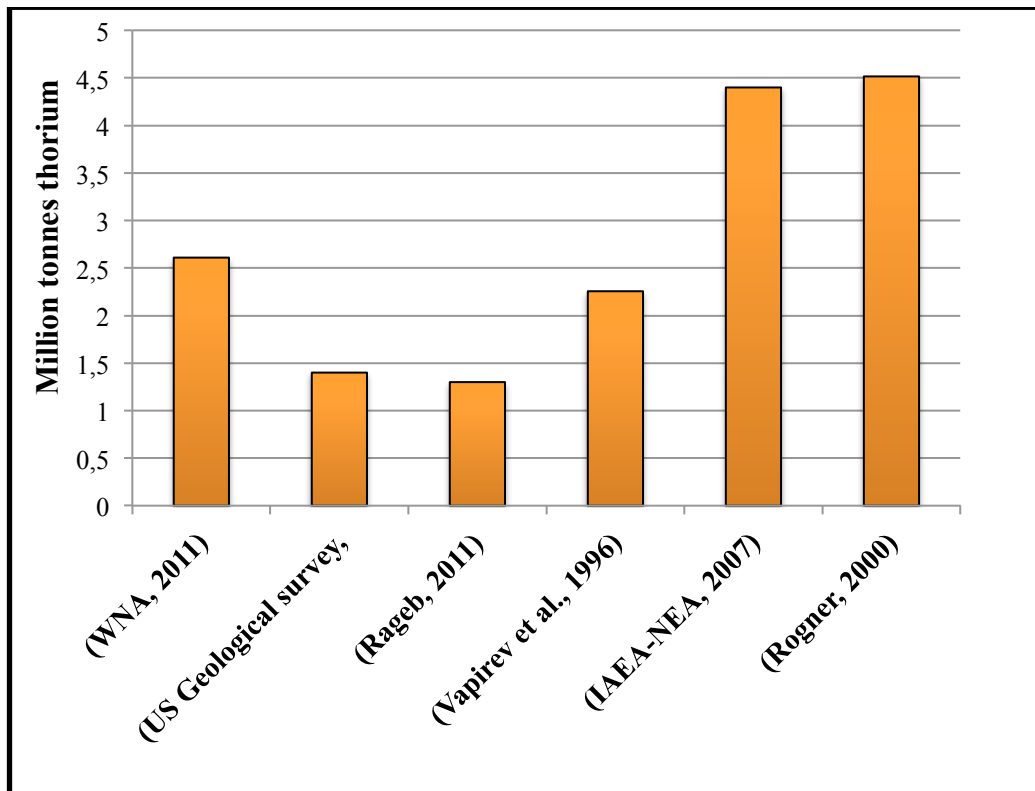


Figure 2.1 Reported thorium resources from different references

2.1.4 Fuel mining, production and reprocessing

Thorium can be mined from granite rocks, phosphate rock deposits, rare earths, and tin ores, coal and uranium mines tailings. Uranium mines based on brannerite ores produce millions of tons of tailings that contain thorium. It is also been proposed to extract thorium from the waste product of coal power stations, ash. A coal-fired power plant produces tons of ash per year, which contain thorium. Thorium is much more energy dense than coal and the concentration of thorium in coal have 31 times more energy than the remaining coal content (Rageb, 2011). The concentration of thorium will vary in different coal samples and the specific

concentration of thorium in South African coal needs to be determined. Due to the large proportion of coal-fired power stations in South Africa, extracting thorium from coal ash may be viable.

Thorium is created in the metallic form through Ca or Mg reduction of ThO_2 and through the electrolysis of anhydrous thorium chloride in a bonded mixture of Na and K chlorides. Thorium chloride is produced by calcium reduction of ThCl_4 mixed with anhydrous zinc chloride or by the reduction with an alkali metal of ThCl_4 . Water corrodes thorium slowly, but acid does not attack thorium, except for HCl (Rageb, 2011).

Retrieval of thorium from monazite normally implicates leaching with sodium hydroxide (NaOH) at 140°C and then an involved precipitation process to produce pure ThO_2 (WNA, 2011a). When Th^{232} is not supplemented by uranium, the environmental impact of thorium mining is much less compared to uranium mining. If Th^{232} is extracted, the activity of the tailings will decrease 20 times in 30 years (Vapirev *et al.*, 1996).

Plutonium-dioxide mixed with thorium-dioxide forms a solid solution and $(\text{Th/Pu})\text{O}_2$ can be manufactured by the powder route, by adding powdered thorium-dioxide and plutonium-dioxide together. $(\text{Th/Pu})\text{O}_2$ can be produced using the Micronized Master Blend (MIMAS) method with the same equipment as for conventional MOX without improving on radiological protection. $(\text{Th/U})\text{O}_2$ -fuel produced from recycled uranium; need more radiological protection, due to strong gamma rays. U^{232} has strong gamma radiating daughter products, primarily Tl^{208} (Schram & Klaasen, 2007).

2.1.5 Decay chain and daughter products

Th^{232} and U^{233} decay to a stable Pb^{206} and Bi^{209} respectively. Ra^{220} forms along the decay chain of Th^{232} and is a gas at ambient temperatures. This Ra^{220} has dangerous daughter products and appropriate ventilation is required, especially in mines (Rageb, 2011). The longest living daughters of Th^{232} products are Ra^{228} (6,7 years) and Th^{228} (1,9 years) (Vapirev *et al.*, 1996). Another intermediate product of Th^{232} named protactinium (Pa^{233}) is a strong absorber of neutrons, which in turn reduces the U^{233} production (WNA, 2011a).

U^{232} can be produced from Th^{232} via a $(n,2n)$ reaction and via a radiative capture (n,γ) or through both these reactions from the bred U^{233} . U^{232} can also be produced by two successive

neutron captures of Th^{230} . The existence of the isotope U^{232} (with a half-life of 72 hours) in thorium-based fuel cycles makes these thorium-based fuel cycles (at high burnups) a higher radiation danger than Pu^{239} .

U^{232} has strong gamma radiating daughter products, primarily thallium (Tl^{208}) and bismuth (Bi^{212}). These two isotopes release high-energy gamma rays, especially the 2,6MeV gamma rays from Tl^{208} , which can cause problems during manufacture, transport, recycling, and disposal (Rageb, 2011). Additional shielding needs to be developed for these handling operations (Puill, 2002).

It should be noted that these gamma-emitting daughters of thorium are also the reason why thorium-based fuels have a stronger proliferation resistance.

2.1.6 Thorium fuelled reactor options

This section depicts all the different reactor types that can utilize thorium-based fuels in their cores. Thorium is technically feasible in most current (LWRs & HWRs) and potential reactor designs (IAEA, 2012). The four main potential thorium-driven reactors are: high temperature gas cooled reactors (HTGRs), light water breeder reactors (LWBRs), molten salt breeder reactors (MSBRs) and accelerator driven systems (ADS). Thorium can also be used in PWRs as discussed in chapter 3. Each reactor is introduced with the basic information.

2.1.6.1 High temperature gas cooled reactor (HTGR)

General Atomics developed a high temperature gas cooled reactor HTGR that utilizes graphite as the moderator and helium as coolant (Galperin *et al.*, 1997). The HTGR reactor is fuelled with fertile thorium, mixed with HEU. As the reactor burns U^{235} , U^{233} is produced from the fertile thorium. Although this reactor is not a breeder, the fissile material needs to be maintained. The fuel elements are coated $(\text{Th,U})\text{C}_2$ particles (Lamarsh & Baratta, 2001).

Two different fuel possibilities exist, namely pebbles and prismatic fuel elements. General Atomics is developing the Gas Turbine-Modular Helium Reactor (GT-MHR), which utilizes prismatic fuel. This reactor is based on the US knowledge, particularly from the Fort St Vrain reactor. The GT-MHR core is capable of using different fuel options, including Th/HEU , $(\text{Th/U}^{233})\text{O}_2$ and $(\text{Th/Pu})\text{O}_2$. PBMR (Pebble bed module reactor) development is based on the

AVR and THTR of Germany while China and South Africa are involved in the developments (WNA, 2011a).

2.1.6.2 Breeder reactors

Reactors are called breeders when they produce more material than what they consume. The regeneration factor, η must be greater than 2 for breeding. This is due to the fact that one fission neutron must eventually be absorbed in fuel to keep the reactor critical and maintain the chain reaction. Thorium (U^{233}) is an attractive option as a breeder due to a η larger than 2 and displays great characteristics at thermal energy levels.

Breeding can be described in terms of the doubling time, which is the time it takes a breeder reactor to breed enough fissile material to fuel a second reactor. The breeding ratio is defined as the average number of fissile atoms produced per fissile fuel atom burned (Lamarsh & Baratta, 2001). A shorter doubling time and higher breeding ratio, results in more efficient breeding.

Other fissile isotopes need to be mixed with thorium, as described in section 2.1.1. The fissile isotopes serve as drivers, and provide all the neutrons initially, but are gradually complemented by U^{233} as it breeds from the thorium (WNA, 2011a). Examples of breeders include the molten salt breeder reactor (MSBR), with the liquid fluoride thorium reactor (LFTR) and the light water breeder reactor (LWBR) (Radkowsky reactor). These two types of breeders are described in the following section.

Molten Salt Breeder

MSBR is a breeder reactor that utilizes the $(Th/U^{233})O_2$ -fuel and operates in the thermal energy range. Here the U^{233} , thorium and coolant are mixed into a homogenous fluid by using fluoride salts. The reactor operates at elevated temperatures, but not high pressures, and the fluid stays a liquid at temperatures $\sim 1400^\circ C$ (WNA, 2011a).

The LFTR is an advanced case of the MSBR and it uses U^{233} coming from a liquid thorium salt blanket. U^{233} is continuously added to the core, after being extracted from the blanket. The core contains fissile U^{233} tetrafluoride ($U^{233}F_4$) in molten salts around $700^\circ C$ and at low pressure. A graphite construction surrounds the core to reflect neutrons and moderate neutrons to intermediate energies. The resultant fission products (FPs) dissolve into the salt

and are gradually separated. Xenon (gas) moves to the surface and the rest of the FPs are caught chemically (WNA, 2011a).

Actinides production is less for Thorium but more for compounds with an atomic mass larger than 235. The few actinides that form remain in the fuel to be transmuted and finally fissioned. The blanket contains fertile thorium tetrafluoride (ThF_4) in the same molten salt; due to the heat from the core. The U^{233} that is bred from the (ThF_4) forms soluble uranium tetrafluoride (UF_4). Fluorine gas is bubbled through this mixture to produce uranium hexafluoride (UF_6) gas that moves to the top of the mixture. It is then captured and reduced in a reduction column back to soluble UF_4 via hydrogen gas (WNA, 2011a).

This UF_4 is then sent to the core. The safety function of this design is achieved through a freeze plug. When a power outage occurs, the fuel drains into a catch basin where it becomes subcritical. MSBRs were studied extensively in the 1960s and were selected for further development in the Generation IV programme, due to the availability of advanced materials. China, Japan, Russia, France and the USA all show renewed interest in the MSBR (WNA, 2011a).

MSBRs do not use solid fuel rods with cladding or cooling water to transfer the heat. Fission energy is transported directly by the molten-salt fuel itself. The use of liquid fuel supports handling of the fuel. The reactor pressure of MSR is around 0,5MPa, which eliminates the need for a pressure vessel like in PWRs and eliminates steam explosions. Hydrogen will not be generated in MSR, due to the absence of both zirconium and water. The production of americium and curium is 83 times smaller for this reactor than in common UO_2 in PWRs (Kamei & Hakami, 2011).

Radkowsky Reactor

The Radkowsky Thorium Power (now Lightbridge) Corporation was involved in the development of the seed-blanket concept, especially for the Russian VVER and Western PWRs (WNA, 2011a; Fridman & Kliem, 2011). ThO_2 is used for the blanket and UO_2 for the seed assembly. The design is founded on the Shippingport LWBR (Fridman & Kliem, 2011). The function of the blanket is to breed and fission U^{233} , whilst the seed provides the neutrons for the blanket.

The seed can be 20% enriched U^{235} or plutonium. One can add some U^{238} inside the blanket to improve the proliferation resistance, because any uranium then separated from the blanket cannot be used for weapons. A by-product in the blanket, namely U^{232} , decays quickly and has strong gamma emitting daughter products. This complicates handling U^{233} and therefore presents proliferation resistance. The plutonium produced in the seed will have a high concentration of heat generating Pu^{238} , which also makes it unattractive for weapons (WNA, 2011a).

2.1.6.3 Accelerator driven system (ADS)

An accelerator produces high-energy protons, which hit a heavy material target, for example Pb, Pb-Bi, or Hg. A spallation reaction occurs and high-energy neutrons are produced, which are directed at a subcritical thorium reactor. Fast fission occurs in thorium and the neutrons multiply. The neutron economy can be controlled, by using neutron-multiplying material (Be) and external sources (IAEA, 2000).

The benefits of ADS are:

- a sufficiently smaller production of long-lived actinides;
- a minimal probability of runaway reaction;
- an efficient burning of minor actinides and a low-pressure system.

The disadvantages are that ADS are:

- more complex; less reliable power production due to accelerator downtime;
- the large production of volatile radioactive isotopes in the spallation target;
- the beam tube may break containment barriers (Anon., 2008).

2.2 Past experience

Thorium has been researched from the early stages of the nuclear era and countries have gained experience on thorium-based fuels on different research and power reactors. The most important cases are summarised Table 2.1, which provides historical background into thorium-based fuels. Most of the current and future designs are based on these reactors and are therefore relevant for this study.

Table 2.3 Summarised table of past thorium-based reactors

Name	Power	Burnup	Fuel	Time	Type	Country	Application
Angra 1	900MWe ^c	-	(Th/U)O ₂ , (Th/Pu)O ₂ ^c	1979 ^c	PWR ^c	Brazil ^c	Power ^c
AVR	15MWe ^b	15GWd/t ^b	(Th/HEU)C ₂ ^a 100000 pebbles ^b	1967-1988 ^b	HTGR ^b	Germany ^b	Test ^b
CIRUS	40MWth ^c	18GWd/t ^c	(Th/Pu)O ₂ ^c	Still in operation	PWL ^c	India ^c	Test ^c
Dragon	20MWth ^b	-	(Th/HEU)C ₂ 10:1 ^a	1964-1973 ^b	HTGR ^a	UK ^b	Test ^b
Elk River	22MWe ^a	8GWd/t ^c	(Th/HEU)O ₂ ^a	From 1964 ^a	BWR ^a	USA ^c	Power ^c
Fort St. Vrain	330MWe ^b	17GWd/t ^b	(Th/HEU)C ₂ ^a	1976-1989 ^b	HTGR ^b	USA ^b	Power ^b
Indian Point I	270MWe ^c	32MWd/kg ^c	(Th/HEU)O ₂ ^c	1962 ^a	PWR ^c	USA ^a	Power ^c
Kakrapar KAPS	200MWe ^a	-	(Th/U)O ₂ ^a	From 1993/1995 ^a	PHWR ^b	India ^b	Power ^b
Kamini	30kWth ^b	-	U ²³³ -Al alloy plate type fuel ^c	From 1996 ^b	Neutron source ^b	India ^b	Test ^b
Lingen	60MWe ^b	-	(Th/Pu)O ₂ ^b	1986 ^a	BWR ^b	Germany ^b	Power ^b
Peach Bottom	110MWth ^b 40MWe ^a	-	(Th/HEU)C ₂ ^a	1967-1974 ^b	HTGR ^b	USA ^b	Test ^b
Shippingport	60MWe ^a	60MWd/kg ^c	(Th/U ²³⁵ ,Pu)O ₂ ^b Seed-blanket ^b	1977-1982 ^b From 1957 ^a	LWBR ^b	USA ^b	Power ^b
THTR	300MWe ^b	15GWd/t ^a	(Th/HEU)C ₂ ^a 674000 pebbles ^b	1983-1989 ^b From 1985 ^a	HTGR ^b	Germany ^b	Power ^b

^a. (Greeneche, 2010)

^b. (WNA, 2011a)

^c. (IAEA, 2002)

2.3 Properties

The material properties, fertile and fissile isotope properties, as well as the decay chain of thorium are discussed. The strengths of thorium are noted and mainly compared to uranium to show that thorium is a viable alternative fuel option. The weaknesses are also shown to identify the potential problems that need to be addressed.

2.3.1 Material properties thorium fuel

ThO₂ is the form in which thorium is used as a fuel. The material properties for ThO₂ are discussed and compared to the reference materials such as UO₂ and MOX. The radiotoxicity of the ThO₂-fuel is also mentioned.

Strengths

ThO₂ is more stable and robust than LEU from a metallurgical (Caner & Dugan, 2000) and chemical point of view and can tolerate higher burnups. Metallic thorium's interaction with steam is less intense than for metallic uranium (Greeneche *et al.*, 2007).

ThO₂ is the highest oxide of thorium and does not vary considerably from this stoichiometric composition when subjected to air or water at temperatures up to 1727°C (Herring *et al.*, 2001). The specific power and burnup for reactors utilizing thorium-dioxide can be raised, due to the higher melting point shown in Table 2.4 (Trellue *et al.*, 2011). These characteristics permit higher safety limits and high thermal efficiencies (IAEA, 2012).

The probable radiotoxicity of depleted fuel is believed to be lower due to the lighter weight of thorium compared to uranium and plutonium and the lower production of minor actinides (Puill, 2002). Depleted and irradiated thorium-based fuel, presented a reduced radiotoxicity for the first 10000 years in the repository when compared to UO₂-fuel (Galperin *et al.*, 2002).

TOX (Th/Pu)O₂, UOX and MOX have comparable physical characteristics and can syncrystallize in the centered cubic form. This property is important for the manufacture and stability of hybrid oxide fuels and certainly allows the manufacture of very-high burnup fuels (Lung & Gremm 1998).

Weaknesses

ThO₂ has a ~10% lower density than UO₂ as can be seen in Table 2.4, which is a drawback, due to the resulting lower concentration of heavy nuclei. From 1000 years up to 1 million years thorium-based fuel presented an increased radiotoxicity, due to decay products like radium (Trellue *et al.*, 2011).

Table 2.4 Material properties of different oxide fuels

	ThO ₂	UO ₂	PuO ₂ ^b
Melting point (°C)	3300 ^a	2760 ^a	2400 ^b
Density (g/cm ³)	10 ^a	10,96 ^a	11,5 ^b
Thermal conductivity at 600 °C (W/cm/°C)	0,044 ^a	0,0452 ^a	-

^a. (Greeneche, 2010)

^b. (Puill, 2002)

2.3.2 Fertile isotope properties (Th²³² & U²³⁸)

Thorium is naturally available as Th²³² and the properties of this fertile isotope are discussed and compared to the reference fertile isotope, namely U²³⁸.

Strengths

Thorium-based fuel presents a good breeding ratio at thermal energies. High conversion rates from Th²³² to U²³³ can be achieved in the thermal spectrum due to the larger absorption cross-section of Th²³², compared to U²³⁸ (Kang-Mok & Myugn-Hyung, 2005). The thermal capture cross-section of Th²³³ is about three times more than that of U²³⁸ as shown in Table 2.5. A breeding cycle comparable to U²³⁸/Pu²³⁹ can be established with Th²³²/U²³³ only more effective (WNA, 2011a). Epithermal neutrons dominate the conversion of Th²³² to U²³³; harder neutron spectra in thermal reactors will increase the conversion ratio in thorium-based fuel (Si, 2009). The proportion of thorium neutron absorption to neutron loss in parasitic material is higher for Th²³² than for U²³⁸ (IAEA, 2012).

Weaknesses

The larger thermal capture cross-section of Th^{232} compared to U^{238} would demand much higher fissile enrichment requirements for Th^{232} fuelled thermal reactors (Kim & Downar, 2001). This would influence the conversion factor, which is the ratio of fissile nuclei resulting from capture to fissile nuclei eliminated by absorption. Uranium is advantageous in terms of conversion factor, compared to thorium (Puill, 2002).

Both Th^{232} and U^{238} can fission above threshold energies, but U^{238} has a lower fission threshold, as can be seen in Table 2.5, which is advantageous to U^{238} . The fission cross-section of U^{238} in the fast energy spectrum is between three and five times higher than Th^{232} . Fast fission of U^{238} contributes between seven and eight per cent of the total energy, compared to the two per cent of Th^{232} (Puill, 2002).

U^{232} is an undesirable by-product in thorium-based fuel cycles, due to the fact that it decays into thallium (Tl^{208}) and bismuth (Bi^{212}) as discussed in section 2.1.5. These two isotopes release high-energy gamma rays, which can cause problems during manufacture, transport, recycling, and disposal. Additional shielding needs to be developed for these handling operations (Puill, 2002). It should be noted that these gamma-emitting daughters of thorium are also the reason why thorium-based fuels have stronger proliferation resistance.

Table 2.5 Various properties of the fertile isotopes of thorium compared with uranium

	Th²³²	U²³⁸
Mass density (g/cm³)	11,72 ^c , 11,71 ^d , 11,5 ^a , 11,7 ^b	18,95 ^c , 19,1 ^d , 18,7 ^a , 18,9 ^b
Melting Point (°C)	1750 ^c , 1800 ^a	1135 ^c , 1133 ^a
Boiling Point (°C)	4788 ^c	4131 ^c
β_{eff} fast fission (pcm)	2030 ^a	1480 ^a
Fission Threshold (MeV)	1,5 ^b	0,8 ^b
σ_c thermal (barn)	7,4 ^a	2,7 ^a , 2,73 ^b
RI at Infinite Dilution (barn)	85 ^a	275 ^a , 272 ^b
Thermal Conductivity @ 600°C (W/cm/°C)	0,45 ^a	0,42 ^a
σ_a (barn)	7,40 ^d	σ _a = 7,59 (σ _f = 4,19) ^d
σ_s (barn)	12,67 ^d	8,90 ^d
Σ_a (cm⁻¹)	0,2249 ^d	0,3668 ^d
Σ_s (cm⁻¹)	0,3850 ^d	0,4301 ^d

^a. (Greneche, 2010)

^b. (Puill, 2002)

^c. (Shultis & Faw, 2002)

^d. (Lamarsh & Baratta, 2001)

2.3.3 Fissile isotope properties (U²³³ & U²³⁵)

Strengths

U²³³ has the highest neutron yield amongst all the fissile isotopes at thermal energies (Trellue *et al.*, 2011). This high η value of U²³³ results in much smaller swings of fissile content and reactivity than fuel cycles using U²³⁵. Over the core lifetime, power peaking is less compared to uranium cores, which makes thorium-fuelled reactors more controllable (Greneche *et al.*, 2007).

U²³³ has a high fission cross-section and low capture cross-section. This low capture cross-section of U²³³ limits unwanted transmutation. The ratio of production to absorption by fission of U²³³ is ~2.29, which is high and point to possibilities of a thermal breeding reactor (Puill, 2002).

The nuclear parameters of U²³³ have a significant weaker dependence on power and temperature, which eases reactor safety and operation, when changing from cold to hot conditions (Greneche *et al.*, 2007). At epithermal energies, η varies the least among fissile isotopes, which reduces the reactivity effects of changes in the neutron spectrum due to coolant transients. Th²³²/U²³³ fuel is less affected by spectrum hardening which reduces its

void and temperature coefficients (Kazimi *et al.*, 1999). U^{232} is an undesirable by-product in thorium-based fuel cycles, but also the reason why thorium-based fuels have stronger proliferation resistance.

The production of fission products (such as Xe, Sm, etc.) is considerably lower for U^{233} , compared to that of U^{235} and plutonium. This means that the average absorption cross-sections of the fission products of U^{233} decreases by about 25-30%, which results in reduced reactivity losses and increased core lifetime (Greeneche *et al.*, 2007). See Table 2.6 for the fission product yield values.

Weaknesses

U^{233} compared to U^{235} has a lower delayed neutron fraction, β , and behaves kinetically similar to Pu^{239} (Trellue *et al.*, 2011). A lower delayed neutron fraction can make the system very sensitive during reactivity changes. The delayed neutron fraction for U^{233} is around 0.22% (See Table 2.6). β plays a vital role in the time behaviour, reactor kinetics and essentially the control of nuclear reactors. Delayed neutrons significantly increases the reactor period, which makes it much more controllable (Lamarsh & Baratta, 2001).

One of the main weaknesses of thorium-based fuels is the high concentration presence of Pa^{233} . Pa^{233} has a long decay period (27 days), when compared with Np^{239} (2,3 days) produced from Pu^{239} in the uranium cycle. This results in an extended delayed reactivity after shutdown. The neutron capture in Pa^{233} results in the loss of a U^{233} nucleus, which would have been formed by the decay of Pa^{233} . This can lead to a significant reduction of the conversion factor more significant as the thermal neutron flux increases (Greeneche *et al.*, 2007).

Smaller recoverable energy per fission of U^{233} results in slightly more fuel to maintain the same power level as enriched uranium fuel (Waris *et al.*, 2010). The intermediate product of Th^{232} protactinium (Pa^{233}) is a strong absorber of neutrons, which in turn reduces the U^{233} production (WNA, 2011a).

Table 2.6 Comparison of nuclear properties for all the fissile isotopes

	U^{233}	U^{235}	Pu^{239}	Pu^{241}
η_{th}	2,29 ^a , 2,287 ^d , 2.3 ^b	2,07 ^a , 2,068 ^d , 2,077 ^b	2,11 ^a , 2,108 ^d , 2,109 ^b	2,145 ^d , 2,151 ^b
η_f	2,27 ^a	1,88 ^a	2,33 ^a	-
$\eta_{th\ ave}$	2,27 ^b	2,06 ^b	1,84 ^b	2,17 ^b
$\eta_{epi\ ave}$	2,16 ^b	1,67 ^b	1,88 ^b	2,49 ^b
Q	200,29 ^c	202,61 ^c	211,41 ^c	213,41 ^c
T_{1/2}(y)	59,2*10 ^{3c}	703,8*10 ^{6c}	24,11*10 ^{3c}	6,564*10 ^{3c}
σ_c at thermal energies (barn)	46 ^b , 47,7 ^d	101 ^b , 98,6 ^d	271 ^b , 268,8 ^d	368 ^b , 368 ^d
σ_f at thermal energies (barn)	525 ^b , 531,1 ^d	577 ^b , 582,2 ^d	742 ^b , 742,5 ^d	1007 ^b , 1009 ^d
α	0,088 ^b , 0,096 ^f	0,175 ^b , 0,171 ^f	0,365 ^b , 0,504 ^f	0,366 ^b , 0,331 ^f
β_{eff} (pcm)	270 ^a , 310 ^f	650 ^a , 690 ^f	210 ^a , 260 ^f	490 ^a , 500 ^f
I^{135} fission yield (atoms/fission)	0,0475 ^d	0,0639 ^d	0,0604 ^d	-
Xe^{135} fission yield (atoms/fission)	0,0107 ^d	0,00237 ^d	0,0105 ^d	-
Pm^{149} fission yield (atoms/fission)	0,00795 ^d	0,01071 ^d	0,0121 ^d	-
Total yield	0,066 ^d	0,077 ^d	0,083 ^d	-

^a. (Greeneche, 2010)

^b. (Puill, 2002)

^c. (Shultis & Faw, 2002)

^d. (Lamarsh & Baratta, 2001)

^e. (Trellue *et al.*, 2011)

^f. (Kazimi *et al.*, 1999)

2.4 Conclusion

This chapter supplies background on thorium as an element, including the applications for thorium in the past, thorium reserves and the mining of thorium. The different reactor types that can accommodate or use thorium-based fuel in their cores are introduced with their basic information. An overview on the past experience on different research- and power reactors are given. The material properties, fertile and fissile isotope properties, as well as the decay chain of thorium are discussed to evaluate thorium as a fuel.