On the proliferation resistance of thorium-uranium nuclear fuel

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On the Proliferation Resistance of Thorium-Uranium Nuclear Fuel

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1. Introduction

Civilian nuclear power reactors are either fuelled with low-enriched uranium (≤ 5% ^{235}U) or, to a much lesser extent, uranium-plutonium mixed-oxide (MOX) fuel. An alternative fuelling scheme is to use thorium, which naturally contains >99.99% of fertile ^{232}Th. On capturing a neutron, ^{233}Th undergoes subsequent β^- decays to ^{233}U. ^{233}U can undergo fission on capturing a neutron of any energy. An overview of the potential thorium-based fuel-cycles and its impact over the whole nuclear fuel cycle can be found in a recent IAEA Technical Document.[1] In terms of proliferation resistance, proponents of thorium-based fuel cycles mention that much less plutonium is produced and the plutonium generated has a larger component of heat-generating ^{238}Pu.[2,3] Furthermore, a small amount of short-lived ^{232}U is generated and from subsequent decays from this nuclide, a 2.6 MeV γ ray is emitted. For trace contaminations (~1 μg), the resultant whole-body dose is of the order of millisieverts at a distance of 1 metre which would make the spent fuel highly radiotoxic and difficult to reprocess.[4] The main counterpoint to this is that ^{233}U, produced in such a nuclear energy system, is a specific nuclear material with a bare critical mass of 15.5 kg, which is comparable to that of ^{239}Pu (10.2 kg).[5] Under IAEA Safeguards Agreements, pure ^{233}U is treated identically to plutonium (i.e. a significant quantity is defined as 8 kg and amounts over 2 kg are treated as Category I materials).[6-8] Also, for uranium-thorium fuels in existing nuclear energy systems, as thorium is fertile (and hence absorbs neutrons), higher concentrations of ^{235}U are needed to maintain criticality at the start of the fuel cycle.

This paper will highlight the historical and potential future uses of ^{232}Th and ^{233}U both in terms of civil and military applications. A brief section on the differences between ^{233}U and ^{239}Pu will be presented and how these could impact proliferation-resistance assessments. Finally, a set of open questions regarding ^{233}U will be presented, some of which will be answered as part of this research project.

2. Civilian Nuclear Fuel Applications of ^{232}Th and ^{233}U

Currently, thorium has little commercial application and as such is mainly treated as a waste by-product from rare-earth element mining.[9] Given its abundance, which due to its longer half-life is expected to be three times greater than that of uranium (> 57 MT compared with 19.2 MT), it has been touted as a potential future nuclear fuel. It should be noted that currently only 6.5–7.4 MT of thorium has been identified so far, compared with 5.4 MT of reasonably-assured uranium reserves.[10,11] However, data on thorium reserves has not been published for a number of countries, most notably China.

In terms of civil nuclear applications, thorium dioxide (ThO_2) has been used in a number of research, prototype and commercial reactors. For light water reactors (LWRs), the Shippingport Nuclear Reactor in the USA operated for 29,000 effective full power hours from 1977 to 1982 with a core consisting of ceramic thorium-uranium fuel containing 1–5% uranium (enriched to 98% ^{233}U).[12] Since this reactor's operation, modifications to the Rosatom-designed 1000 MW_e VVER-1000 have been proposed to incorporate low-enriched uranium (containing 20% ^{235}U) and thorium.[13] This core design is referred to as the VVER-T.[14] Similarly, researchers at MIT have investigated two different strategies of fuelling a Westinghouse PWR with low-enriched uranium (also containing 20% ^{235}U) and thorium.[15] Significant research into thorium-based fuels for LWRs is currently being undertaken by ThorEnergy and also LightBridge.[16,17]

High-temperature gas-cooled reactors (HTGRs) can also use thorium, either in prismatic blocks or as a bed of pebbles. A number of test reactors were built and operated with Th-LEU fuel between the 1960s and 1980s including Peach Bottom, USA (1967–1974), Dragon at Winfrith, UK (1964–1975) and AVR in Germany (1968–1988).[18,19,20] Two demonstration commercial reactors were built thereafter. First, a 330 MW_e prismatic HTGR was built at Fort St Vrain, Colorado, USA and a 300 MW_e pebble-bed modular reactor was built in Hamm-Uentrop, Germany.[21,20] Both reactors experienced numerous difficulties and were closed prematurely, although these closures were not due to failures or

shortcomings in the thorium-fuel components of the programmes. The Very High Temperature Reactor concept, an evolution of such high-temperature reactors, is one of six potential nuclear energy systems suggested as part of the Generation IV International Forum.[22]

In the near-term, India is hoping to use thorium in the Advanced Heavy Water Reactor (AHWR).[23] These reactors would form the third stage of India's three-stage nuclear energy program.[24] It is envisaged that these reactors would operate in a closed fuel cycle and use reprocessed ^{233}U and a small component of ^{239}Pu in the central seed region of the fuel cluster (near equivalent to a fuel assembly in a PWR), with fertile ^{232}Th forming the outer blanket region of the fuel cluster. A modification of the core layout is being investigated to allow such a reactor to operate with low-enriched uranium and thorium.[25] Also, it is envisaged that blankets of ^{232}Th will be placed in Indian-designed, sodium-cooled fast breeder reactors to breed extra ^{233}U required for AHWRs.[26] On a separate note, India has already utilised 600g of ^{233}U, alloyed with aluminium, to form the core of the 30 kW KAMINI research reactor at the Indira Gandhi Centre for Atomic Research, Kalpakkam, India.[27]

Another two potential nuclear energy systems that could make use of thorium are molten salt breeder reactors (MSBRs) and accelerator driven sub-critical reactors (ADSRs). Development of MSBRs is underway in the USA, China and Japan. These reactors would be based on developing and up-scaling the Molten Salt Reactor Experiment that operated at Oak Ridge National Laboratory from 1965 to 1969.[28] Although there are ambitious research programs under development for such reactors, various technical challenges still need to be overcome.[29] Research and development of ADSRs is ongoing in the UK, USA, Europe, China and India.[30] It should be noted that significant improvements in the reliability of current accelerator technology are needed.[31] For both technologies, it is expected that closed fuel cycles would pertain.[32]

Other countries that have shown an interest in thorium include Norway, which recently completed an appraisal of thorium and its potential to be indigenously adopted.[33] Norway has an estimated 170,000 tonnes mainly contained in hard-rock formations. The 2008 report gives no final assessment into the ability for thorium-based energy generation to be deployed but recommends that it should be maintained as an open option. In the UK, The Weinberg Foundation, a not-for-profit, non-governmental organisation, was launched in 2011 to promote thorium as a future nuclear fuel.[34] Although such a foundation is geared towards being technology-independent, particular support has initially been made towards molten salt reactor concepts. In 2012, an All-Party Parliamentary Group on thorium energy has been launched that will look at the potential benefits of the fuel and reactor technologies.[35] Separately, the UK National Nuclear Laboratory recently issued a position paper on thorium which states that 'the thorium fuel cycle does not currently have a role to play in the UK context, other than its potential application for plutonium management in the longer term', although it supports future research and development on this fuel cycle.[36] In recent years, the UK National Nuclear Laboratory has been involved in European-based research programmes on thorium-based fuels.[37]

3. Military Applications of ^{233}U

Although the majority of research relating to fission devices since the Second World War has focused on utilising plutonium, some research has also been performed on ^{233}U. Two particular references in open literature will be presented below, although in no sense should this be taken as an exhaustive representation of the subject.

First, from Hansen's book *U.S. Nuclear Weapons*, as part of its development of artillery-fired atomic projectiles, the US started development of an 8-inch shell (the MK 33) in 1952.[38] Hansen notes 'requirements for nuclear materials … included the request for construction of equipment and facilities for producing uranium-233 for use as a nuclear explosive in the new shell'. 'In late February 1953, results of tests at Los Alamos Scientific Laboratory (now Los Alamos

National Laboratory), indicated that U-233 would not be a satisfactory nuclear explosive and the Army changed its requirements to use uranium-235 as the nuclear fuel for the 8-in shell'.[39]

Second, from the Nuclear Weapon Archive website, it is mentioned that Shot MET ('Military Effects Tests'), part of Operation Teapot, comprised of a Pu/^{233}U core.[40] The purpose of this Shot was '(1) To study the effects on military equipment, and (2) To allow Department of Defense personnel to observe a nuclear detonation'.[41] The delivered payload from this Shot was 22 kT, though the predicted yield was suggested to be 33 kT.[40] Given that Ref. [40] does not reference the origin of the data, the original source material would need to be identified and reported to provide extra credibility. What appears surprising is that in comparing the material properties of ^{233}U to ^{235}U and ^{239}Pu, ^{233}U could potentially be more advantageous.

4. Material Properties of ^{233}U Compared to ^{235}U and ^{239}Pu

The material properties of ^{233}U, pertinent to weapons-usability, have been previous ascertained and compiled by C.W. Forsberg et al.[42] These centre on the mass, isotopic mix, heat and radiotoxicity of the material. As mentioned in the introduction, ^{233}U is a specific nuclear material of which a significant quantity, defined as the 'the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded', is defined to be 8 kg.[7] This is compared with 8 kg for mixed plutonium (with the exception of plutonium containing more than 80% ^{238}Pu which is exempt from safeguards). An estimate of the bare critical mass for ^{233}U is 15.5 kg.[5]

In terms of physical protection categorisation, as outlined in INFCIRC/225/Rev.3, the above compilation of weapons-usable material properties suggests that ^{233}U denatured with ^{238}U should have an equivalent categorisation to that of denatured ^{235}U.[43] The suggested categorisation and amendments (shown in italics) to INFCIRC/225/Rev.3 are presented in Table 1. The most recent guidance from the IAEA, INFCIRC/225/Rev.5 published in 2011, still maintains the same categorisation as the third revision.[8]

Table 1: Material Definitions for Safeguarding Unirradiated Pu, ^{235}U and ^{233}U

		Category		
Material	Form	I	II	III
Pu	Any material form, with exception of plutonium containing ≥ 80% ^{238}Pu	≥ 2 kg	< 2 kg, but > 0.5 kg	≤ 500 g, but > 15 g
^{235}U	U enriched to ≥ 20% ^{235}U	≥ 5 kg	< 5 kg, but > 1kg	≤ 1 kg, but > 15 g
	U enriched to ≥ 10% ^{235}U, but < 20%		≥ 10 kg	< 10 kg
	U enriched above natural, but < 10%			≥ 10 kg
^{233}U	Any material form	≥ 2 kg	< 2 kg, but > 0.5 kg	≤ 500g, but > 15 g
	U with ≥12% ^{233}U	*≥2 kg*	*<2 kg, but >0.5 kg*	*≤ 500g, but >15 g*
	U with <12% ^{233}U, but ≥6%		*≥4 kg*	*<4 kg*
	U with <6% ^{233}U, but ≥0.66%			*≥4 kg*

In terms of the isotopic mix of uranium, currently 'Weapons Grade' uranium is associated with having a ^{235}U content of greater than 94%[44] and non-weaponisable uranium has a ^{235}U content ≤ 20%.[13] However, with the introduction of

fissile ^{233}U and fertile/poisonous ^{232,234,236}U, an equivalent classification scheme to that of plutonium (as reported in Ref. [44]) would need to be formulated.

For plutonium isotopes, ^{239}Pu is the main fissile isotope. ^{240}Pu acts as a 'poison' (i.e. absorbs rather than generating neutrons) and has a sizable spontaneous fission component which can cause pre-detonation of a nuclear device.[45] This is the main reason why plutonium is used solely in implosion devices. ^{238}Pu has a relatively short half-life ($T_{½}$ = 87.7 years) and undergoes a series of α decays to ^{206}Pb which provides a large amount of decay heat. This decay heat can potentially cause degradation of the explosive charge.[44] ^{241}Pu, although fissile, can undergo β^- decay into ^{241}Am which can significantly poison a system.

For uranium isotopes, both ^{233}U and ^{235}U are fissile with differing bare critical masses. ^{236,238}U both act as neutron poisons. ^{234}U is also a poison but has a smaller spontaneous fission component compared to ^{238}Pu. ^{232}U has a comparably short half-life ($T_{½}$ = 68.9 years) and undergoes a similar series of α decays to ^{208}Pb. Compared to ^{238}Pu it provides a sizable decay heat, but from the β^- decay of ^{208}Tl into ^{208}Pb, a 2.6 MeV γ ray is emitted which is difficult to shield.

This radiotoxic ^{232}U component can be formed by the ^{233}U(n,2n) reaction and can potentially make the handling and reprocessing of ^{233}U a significant challenge. The work of Kang and von Hippel looks at the radiotoxicity of spent fuel from current nuclear energy technologies.[46] It is noted that for very high burn-up fuels (e.g. PWR fuel with a discharge burn-up of 70 GWd/t, compared to a typical average burn-up of 45–50 GWd/t), the resultant radiotoxicity of 5 kg of bare uranium metal, with an assumed ^{232}U component of 0.001% is 130 mSv/hr at 0.5 metre, after a decay time of 1 year. However, the build-up of ^{232}U can be mitigated by employing low burn-up fuel cycles, using segregated/targeted thorium channels and with swift reprocessing. The take-home message from the work of Kang and von Hippel is that 'the proliferation resistance of thorium fuel cycles depends very much upon how they are implemented'.

5. Open Questions Regarding the Proliferation-Resistance of Thorium-Uranium Civil Nuclear Fuel

1. What reasons led the US to stop pursuing ^{233}U for weapons purposes and would developments since 1955 make ^{233}U a concern?
2. What is the Definition Weapons Grade Uranium vector inclusive of ^{232}U and ^{233}U (compared with J. Carson Mark's report)?
3. Would thorium-uranium systems provide additional pathways to obtaining specific and alternative nuclear materials?
4. Is 20% low-enriched uranium (although not weaponisable) a sensible upper limit for uranium enrichment, assuming particular diversion routes? If it is not suitable, what impact would this have on thorium-uranium systems?
5. Similarly, what impact would alternative enrichment technologies (electromagnetic/atomic vapour laser isotope separation) have on the definition of low-enriched uranium?
6. Would thorium-uranium systems be more difficult to safeguard (particularly in terms of material accountability)?

6. Conclusion

Thorium has the potential to be implemented in existing and future novel nuclear energy systems. However, as ^{233}U is formed in such systems and is a specific nuclear material near equivalent to plutonium, thorium fuel is of legitimate concern in terms of its proliferation resistance. In this work, we consider thorium-uranium fuels in civil nuclear power plants. We conclude that it is erroneous to say that the thorium-uranium fuel cycle is benign.

7. Terminology

α decay: Transmutation of an unstable heavy nucleus into a lighter nucleus by emission of a ^{4}He nucleus (α particle).

β⁻ decay: Transmutation of an unstable nucleus with excessive number of neutrons, by conversion of one of its neutrons into a proton, emitting an electron (β⁻ particle) and anti-electron neutrino.

γ ray: Electromagnetic radiation emitted from a nucleus with excess internal energy. For nuclear energy systems, this is typically subsequent from β⁻ decay.

Bare critical mass: Mass of fissile material needed (without any reflectors or tampers, hence 'bare') for a sustained chain-reaction to take place.

Burn-up: Energy output from nuclear fission per unit mass of initial nuclear fuel.

Criticality: The ability for a chain-reaction to occur. A critical system has an output to input neutron ratio of 1. If ratio is greater than 1, it is super-critical. If ratio is less than 1, it is sub-critical.

Decay heat: The heat released from radioactive decay.

Fertile: Ability for a nucleus, on absorbing a neutron, to transmute into a fissile nucleus. Main fertile isotopes include ^{232}Th and ^{238}U.

(n,2n) reaction: Nuclear reaction involving energetic 'fast' neutrons, where an incident neutron impinges on a target nucleus and two neutrons are emitted. Converts fissile ^{233}U and ^{239}Pu into undesirable ^{232}U and ^{238}Pu.

Pebble-bed: Fuel form consisting of layers of thorium dioxide, uranium dioxide and graphite, moulded into spheres with a radius of ~5cm.

Prismatic: Fuel form consisting of thorium dioxide, uranium dioxide and graphite in microspheres, with a radius of ~0.15–0.3mm, and are embedded into hexagonal shaped compacts which fit into a large graphite block.

PWR: Pressurised Water Reactor. A light water reactor in which water is maintained at a high enough pressure to prevent boiling.

Spontaneous fission: Ability of a nucleus to undergo fission spontaneously without the need for a neutron.

VVER: Vodo-Vodyanoi Energetichesky Reactor, Russian-designed variant of the Pressurised Water Reactor.

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UK PONI was established in 2010 as a cross-generational forum dedicated to fostering dialogue and building expertise amongst emerging nuclear scholars.

The issues surrounding nuclear weapons and nuclear energy are complex and multifaceted, requiring a broad understanding of everything from technical intricacies to developments in International security. Changes to the shape, size and function of the world's nuclear arsenals, have the potential to profoundly affect global dynamics. Developments in civilian nuclear energy will similarly influence the form and direction of international non-proliferation efforts. These trends will ensure that nuclear issues continue to be at the top of the defence and security policy agenda. Yet despite the continuing importance of nuclear issues, there is little evidence that sufficient expertise is being grown to sustain those with expertise in the field.

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