THORIUM ENERGY FUTURES

S. Peggs (BNL/ESS), W. Horak, T. Roser (BNL), G. Parks (Cambridge U.), M. Lindroos (ESS),
R. Seviour (ESS/Huddersfield U.), S. Henderson (FNAL), R. Barlow, R. Cywinski (Huddersfield U.),
J.-L. Biarrotte (IPN), A. Norlin (IThEO), V. Ashley, R. Ashworth (Jacobs), A. Hutton (JLab),
H. Owen (Manchester U.), P. McIntyre (TAMU), J. Kelly (Thor Energy/WNA)

Abstract

The potential for thorium as an alternative or supplement to uranium in fission power generation has long been recognised, and several reactors, of various types, have already operated using thorium-based fuels. Accelerator Driven Subcritical (ADS) systems have benefits and drawbacks when compared to conventional critical thorium reactors, for both solid and molten salt fuels. None of the four options – liquid or solid, with or without an accelerator – can yet be rated as better or worse than the other three, given today's knowledge. We outline the research that will be necessary to lead to an informed choice.

INTRODUCTION

Fossil fuels account for over 80% of the world's energy production, with uncertain supply security at the same time that global demand is expected to increase by 60% by 2050. Secure, reliable and affordable energy sources are critical for geopolitical stability [1]. Nuclear fusion and other primary source technologies could make a major impact, but their development places their contributions beyond 2050 [1]. Renewable technologies are insufficient to reliably supply all the energy needs of the modern world [2].

Uranium-based nuclear fission technologies are an important energy source for many industrialised nations, although use has been limited by: high cost; perceived adverse safety; the potential for proliferation; and the management of nuclear waste. However, a wider future role for nuclear energy is inevitable if the growing demand for electrical energy is to be met, if these issues can be mitigated.

A thorium fuel cycle, combined with alternative technologies, presents numerous potential advantages [3]. Thorium fuel cycles are intrinsically more proliferationresistant, reduce plutonium production, and can consume legacy plutonium and waste actinides. The different characteristics of the thorium cycle options arise not only from the different system designs, but also from the physical properties of the fuels themselves [4, 5].

SAFETY, PROLIFERATION, AND WASTE

Safety: Molten salt reactor (MSR) and hightemperature gas-cooled reactor (HTGR) thorium reactors have safety advantages over conventional light water reactors (LWR) [4, 5, 6]. The liquid fuel in an MSR may provide improved reactivity feedback and can be removed from the reactor core [6]. HTGRs can withstand very high temperatures in excess of 2000 K without fuel failure [7]. As demonstrated at the German AVR, these designs may survive a complete loss of power event [4]. The use of thorium in LWR fuel can have advantages over solely U-Pu systems because of its better thermal, physical, and irradiation performance [7]. For example, thoria (thorium dioxide) has a melting point 500 C higher than UO₂, improving its accident tolerance. ADS systems have their own enhanced safety characteristics: they can be turned off "instantly", and have reduced fissile loadings.

Proliferation: Thorium occurs in nature as a single fertile isotope. In itself, it cannot be enriched to produce weapons grade material. Consequently it has been argued that thorium is "proliferation resistant" [8, 9]. Additionally, production of Pu-239 is extremely small in most proposed thorium cycles, due to the different actinide distribution, thereby posing a significantly lower proliferation risk.

However, thorium cycles do generate U-233 as the fissile isotope, rather than the Pu-239 bred from U-238 in the U-Pu cycle. U-233 is in principle useable in nuclear weapons, but the inherent co-generation of U-232 provides significant protection due to the steady growth of hard-gamma emitting Tl-208 in its decay chain. This makes non-state weapons manufacture difficult, and easily detectable. It also makes fuel reprocessing more difficult, but it is still required to remove long-term proliferation risk from spent fuel [9]. The International Atomic Energy Agency (IAEA) notes that no U-233 based weapons are under development and that the development of such a weapon would be difficult [9].

Waste: Thorium systems have the ability to burn actinides, and provide several routes to plutonium stockpile disposal [8]. Waste forms for thorium-bearing fuels have been studied and shown to have advantages based on their physical and chemical properties [5, 7, 10, 11].

SOLID FUEL REACTORS

Thorium can in principle be used as a solid fuel component in all the main reactor types that have operated to date: boiling and pressurised light water reactors (BWRs and PWRs), heavy water reactors (HWRs), HTGRs, and sodium-cooled fast reactors. Thoria based fuels offer higher operational safety margins and accident tolerance due to various favourable, robust, properties: a very high melting point; non-oxidizability; general chemical inertness; reasonable thermal conductivity; and a strong ability to retain fission products within its crystal lattice.

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Reactors without Accelerators

Thorium cycles have been investigated in a variety of reactors, including Gas-Cooled Reactors (GCRs), BWRs, and PWRs [12, 13, 14]. These studies demonstrated good performance of thorium in oxide form in LWRs and in carbide form in GCRs [15]. The Light Water Breeder Reactor programme in a PWR [16] demonstrated the feasibility of a closed Th-U-233 fuel cycle, confirming that U-233 breeding is achievable using a heterogeneous epi-thermal spectrum U-Th core. Near-complete transuranic waste incineration has been suggested in a thorium-fuelled PWR [17]. India has operated thorium-fuelled research reactors for many years: first the Purnima-II reactor (1984-6) and, since 1997, in the 30 kW Kamini research reactor.

Thor Energy is developing and testing a thorium fuelform comprised of intimately mixed thorium and plutonium oxides sintered into high-density ceramic pellets, and clad either in standard zirconium alloy, or with an advanced multi-layer silicon carbide composite. The $(Th,Pu)O_2$ fuel is a readily achievable variant of already-licensed mixedoxide fuels, well-matched to today's predominant LWR platform. Results are also applicable to an HWR, potentially an excellent reactor platform. Thor Energy is commencing a series of irradiation tests of prototype fuel in the Halden test reactor in Norway, to monitor changes in the fuel as it is operating: dimensional changes (in pellets and cladding); temperature; and fission gas release.

Accelerator-Driven Subcritical Cores

The accelerator provides a controlled external source of fast neutrons in the reactor core, to breed fissile material at the same rate or faster than it is consumed. Some neutrons maintain a chain reaction while others breed Th-232 to U-233. Eventually the U-233 becomes the fissile fuel, with fertile thorium being added to the mix as necessary.

Proton-driven spallation is the usual choice (especially for GW-scale reactors), although other particles including electrons have also been proposed. Heavier nuclei bring little advantage at considerable cost. The required beam current I is given by

$$I = e \frac{(1-k)P}{nfE_f} \tag{1}$$

where k < 1 is the "criticality" factor of the reactor, P is the thermal power, n is the number of neutrons produced per incident proton, of which a fraction f induce fission, and E_f is the mean energy release per fission. A typical 1 GWe reactor requires average currents of about 10 mA and a beam power of about 10 MW, beyond today's most powerful accelerators.

In 1996 the Indian Atomic Energy Commission initiated design studies for a 200 MWe PHWR ADS system fuelled by uranium and thorium [19]. Jacobs Engineering Group Inc. (Jacobs) have produced a conceptual design of an Accelerator-Driven Thorium Reactor (ADTRTM)



Figure 1: The ADTRTM power station. Top: site layout. Bottom: power reactor building.

600 MWe power station, based on the lead-cooled fast reactor illustrated in Figure 1 [20, 21]. The ADTRTM challenges previously established criticality margins, with a proposed k value of 0.995 selected to bring the accelerator requirements within the realm of current technology. Analysis of the system neutronics and safety led to a novel solution for the measurement and control of reactivity that is central to the commercial viability of the ADTRTM [22].

MOLTEN SALT REACTORS

The Molten-Salt Reactor Experiment (MSRE) shown in Figure 2 operated from 1965 until 1969 using a fuel/moderator mixture of lithium, beryllium, thorium and uranium fluorides [23]. It built on the success of the 1954 Aircraft Reactor Experiment [24], and on other early liquid-fuel schemes; the basic feasibility of such designs is well demonstrated. The graphite moderator ensures the correct neutron spectrum while the fuel salt circulates in a closed loop. Increased power (in a fluctuation) expands the liquid fuel, in turn reducing the power output. This safety feature was tested during MSRE operation, performing as expected. MSRs have further safety advantages and some disadvantages in comparison to solid-fuel reactors [25].

More modern Lithium Fluoride Thorium Reactor designs are being proposed as an alternative fuel cycle solution by several advocacy groups [26, 27]. A major problem

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Figure 2: The Molten Salt Reactor Experiment (MSRE).

with solid fuels is the accumulation of Xe-135, which acts as a short-lived neutron poison: in the MSR the Xe-135 is removed at the primary salt pump bowl. Introducing a small quantity of oxygen in the gas prevents corrosion of the piping.

Most MSR advantages are maintained in an ADS implementation. The target in the salt bath produces additional neutrons, making it easier to optimise performance. One promising target design, from BARC, uses an inert gas injected into a long vertical column to create a mass imbalance that circulates the lead-bismuth eutectic (LBE) and also removes gaseous poisons [28].

It is sometimes said that the accelerator trip requirements for an ADS MSR are considerably relaxed compared with a solid-fuel ADS reactor, because the timescales for stresses are longer since there is no Hastelloy cladding on the fuel rods. The comparison depends on the power rating of the fuel pins in the solid-fuel reactor. Recent studies have shown that in some cases the fuel cladding is the limiting factor [29] while in others it is the inner barrel [30]; an ADS MSR has no need for fuel cladding but does require an inner barrel.

The ADNA Corporation proposed the GEMSTAR design of a medium-power MSR facility [31]. The Department of Atomic Energy in India is aiming at a much larger facility. What is now required is a small molten salt research reactor that can be operated with or without an accelerator to obtain a quantitative comparison of the performance of the two alternatives, just as the MYRRHA project [32] will make the comparison for a solid fuel reactor.

ACCELERATOR IMPLICATIONS

Optimising spallation output in terms of proton energy cost per neutron gives a broad plateau beginning at about

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1 GeV, a value chosen by the Spallation Neutron Source (SNS) [33]. A lower value of 600 MeV reduces the capital costs of the MYRRHA proposal, but provides enough neutrons for a power of 57 MWth [32]. The European Spallation Source [34] energy of 2.5 GeV is rather higher.

At least 20 neutrons are produced for each incident 1 GeV proton in a typical spallation target -1 neutron per 50 MeV of beam energy. About 150 MeV of wall-plug energy is therefore required for each neutron, assuming a typical accelerator efficiency of 30%. Maximising this efficiency is an important accelerator design goal.

Beam losses must be kept to a low level to allow handson maintenance: a proton loss rate of 1 W/m is considered a maximum. This is also a major design challenge.

The d-t reaction of a 100 keV deuteron beam on a tritium target provides an alternative to spallation for neutron production [35]. Most of the beam energy is lost to interactions with electrons, requiring about 750 MeV for each 14 MeV neutron [36]. A plasma could be used to suppress these energy losses by a factor of ten. Although technically very difficult, this could make fusion competitive with spallation for neutron production.

Reliability and Availability

Extremely high accelerator reliability is required in a full-scale ADS power station. Repeated short trips lead to thermal stresses in reactor components, and long trips lead to economically very damaging losses of generating power [37]. Recent analyses of maximum allowable beam trip durations considering thermal stress and damage to reactor components suggest that the required accelerator reliability is much less severe than originally thought (but is still very challenging) [30]. Several uncertainties remain, especially concerning irradiated materials properties, erosion and corrosion related aspects in an LBE environment, and reactor restart sequences.

Erroneous initial optimistic predictions of the number of allowed emergency reactor stops due to beam trips could lead to premature replacements of components, reduction of reactor lifetime, and to a dramatic decrease of availability. Thus, the MYRRHA project requirements conservatively allow fewer than 5 beam trips (longer than 3 seconds) per 3-month operating period, following PHENIX reactor experience [38, 39]. In any case, major improvements in accelerator reliability are most certainly required for the ADS mission.

Industrial experience shows that reliability is achievable – at a cost – through redundancy, under-rating, graceful failure, and planned maintenance, using a holistic analysis of the complete system. It may be advantageous to use several independent accelerators, so that when one is off-line the current in the others can be increased.

Accelerator Technologies

Linear accelerators (linacs) can provide the necessary energy and current, and appear to offer sufficient reliability,

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availability, and rapid fault recovery. However, they are expensive relative to cyclotrons or synchrotrons. A 1.4 MW linac like the SNS would be \sim 300 m long, at a cost of \sim 0.7 B\$, so that it would nonetheless not dominate the total project cost, assuming that a thorium reactor costs about the same as a uranium reactor, \sim 5 B\$.

Cyclotrons are restricted to non-relativistic energies, and are limited by space-charge effects and by the need for orbit separation, but achieve MW-class beam powers at PSI and TRIUMF. A recent proposal for a 5 MW, 800 MeV, strong-focusing cyclotron would overcome both of these limitations [40]. There are also other proposals for ADS cyclotrons [41].

Rapid Cycling Synchrotrons (RCS) like ISIS operate at frequencies as fast as 50 Hz with beam powers well above 100 kW. Much faster repetition rates are attractive, but face the need to develop rapid frequency swing Radio Frequency (RF) systems, and also face technical issues with magnet and vacuum chamber eddy currents.

Fixed-Field Alternating Gradient (FFAG) accelerators currently exist only as low power prototypes, both proton [42] and electron [43]. An isochronous FFAG could be run with continuous beam like a cyclotron to achieve large currents. Non-isochronous designs face the same frequency swing challenges as RCS RF systems.

NECESSARY RESEARCH

Nuclear Energy Systems

Significant research is needed in the 3 main elements of all nuclear energy systems: front-end options, nuclear power alternatives, and disposal options. Several countries, including China, India, Russia and the United States all have active thorium nuclear energy programmes. The IAEA recently extended its long history of thorium projects with a new cooperative research programme.

Front-end options include the mining, milling, and conversion of thorium. Research in this area was mostly done in the 1950s and 1960s. Most current research on front-end options is focused on the separation of thorium from rare earth elements. Little or no techno-economic research has been performed to establish what the needed costs for thorium front end options are, as compared to uranium-based cycles.

Nuclear power alternatives. Extensive research has been performed on thorium nuclear power alternatives. Nearly every major reactor type has been computationally simulated. An almost equally impressive amount of research has been done on ADS systems. Although most of the research is simulation, a significant number of experimental tests have also been carried out, including test irradiations, materials testing, and thermal hydraulic tests. More materials research is needed, particularly for molten salt based systems, where existing data relies heavily on the extensive, but older, Oak Ridge National Laboratory programmes. **Disposal options** for thorium based systems are less well developed than front end systems or nuclear power systems. Several papers have been written specifically on thorium-based system disposal, with some experimentation. Much of this research overlaps with disposal options for uranium- and plutonium-based systems [4, 12].

Accelerator and Target

A recent study assessed the technological readiness of accelerator and target technology for ADS systems, and outlined the necessary R&D activities [44]: it concluded that accelerator and target technology had advanced significantly in the last two decades, and was now ready for a full scale demonstration of the coupling of a MW-class accelerator to a subcritical core. Recent experimental accelerator/reactor coupling demonstrations have been made at low power at GUINEVERE [45] and KURRI [42]. The key performance requirements are very high proton beam power ($\simeq 10$ MW), very low beam loss (< 1 W/m) and very high reliability. Key research areas include:

- 1. development and demonstration of highly reliable high-current proton injector systems;
- 2. advancing the state-of-the-art in accelerator systems, including linacs, FFAGs and RF systems;
- 3. improved understanding of beam loss mechanisms, emittance and halo growth;
- 4. development of highly reliable and fault tolerant accelerator systems and accelerator components.

Required spallation target R&D includes:

- 1. LBE handling, oxygen control and cleanup;
- 2. development of windowless target technologies;
- 3. materials irradiation studies;
- 4. full scale mock-up of cooling systems;
- 5. engineering the accelerator-to-target sub-critical blanket interfaces.

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REFERENCES

- [1] International Energy Agency: "World Energy Outlook", (2006); "Key World Energy Statistics", (2010).
- [2] F.E. Trainer, Energy Policy, V. 23, 1009-1026 (1995).
- [3] "Towards an Alternative Nuclear Future", Thorium Energy Association, ThorEA, (2010).

ISBN 978-3-95450-115-1

08 Applications of Accelerators, Technology Transfer and Industrial Relations

- [4] M. Lung & O. Gremm, "Perspectives of the Thorium Fuel Cycle, Nuclear Engineering & Design", V.180, 2, p.133-146, ISSN 0029-5493, (1998).
- [5] P. Rodriguez & C.V. Sundaram, "Nuclear and Materials Aspects of the Thorium Fuel Cycle", J. of Nucl. Mats., V.100, p.227-249, ISSN 0022-3115, (1981).
- [6] K. Furukawa et al., "A Road Map for the Realization of Global-Scale Thorium Breeding Fuel Cycle by Single Molten-Fluoride Flow", Energy Conversion and Management, V.49, 7, p.1832-1848, ISSN 0196-8904, (2008).
- [7] A. Talamo, "A Novel Concept of QUADRISO Particles Part III: Applications to the Plutonium-Thorium Fuel Cycle", Prog. in Nucl. Energy, V.51, p.274-280, ISSN 0149-1970, (2009).
- [8] A. Galperin, P. Reichert, A. Radkowsky, "Thorium Fuel for Light Water Reactors – Reducing Proliferation Potential of Nuclear Power Fuel Cycle", Sci. & Global Sec. V.6, 3, (1997).
- [9] R. Sollychin, "Exploring Fuel Alternatives", IAEA Bulletin 51-1, p.43-44, (2009).
- [10] P. Demkowicz et al., "Aqueous Dissolution of Urania-Thoria Nuclear Fuel", Nuclear Technology, V.147, p.157, (2004).
- [11] , D. Barrier et al., "Thoria, a Quasi-Inert Matrix for Actinides Dispositions", J. of Nucl. Matls., 352, p.357, (2006)
- [12] IAEA, "Thorium Fuel Cycle: Potential Benefits & Challenges", IAEA-TECDOC-1450, (2005).
- [13] M. Todosow at al., "Use of Thorium in Light Water Reactors", Nuclear Technology, 151, 168-176, (2005).
- [14] T.K. Kim & T. Downar, "Thorium Fuel Performance in a Tight-Pitch Light Water Reactor Lattice", Nuclear Technology, 138, 17-29, (2002).
- [15] J. Belle & R.M. Berman, Eds., "Thorium Dioxide: Properties & Nuclear Applications", US DOE/NE-0060, (1984).
- [16] R. Atherton, Ed., "Water Cooled Breeder Program Summary", WAPD-TM-1600, Bettis Atomic Power Lab, (1987).
- [17] B.A. Lindley & G.T. Parks "Near-complete Transuranic Waste Incineration in a Thorium Fuelled Pressurized Water Reactor", Annals of Nuclear Energy, 40, 106-115 (2012).
- [18] "(Indian) Nuclear Power Programme Thorium", http://www.dae.gov.in/publ/ar0203/chap3.pdf
- [19] S. Banerjee, "Towards a Sustainable Nuclear Energy Future", WNA Symposium (2010).
- [20] C. Rubbia et al., "Conceptual Design of a Fast Neutron High Power Energy Amplifier", CERN-AT-95-44(ET), (1995).
- [21] V. Ashley & R. Ashworth, "The Technically Viable ADTRTM Power Station", Nuclear Future V.7 3, (2011).
- [22] C. Rubbia, "Accelerator Driven Nuclear System with Control of Effective Neutron Multiplication Coefficient", PCT Patent, PCT/EP2010/054132, (2010).
- [23] R.C. Briant et al., Nucl. Sci. Eng., 2, 797 (1957).
- [24] P. N. Haubenreich & J.R. Engel, "The Molten-Salt Reactor Experiment", Nucl. Appl. Tech., 8, 118 (1970).
- [25] C.W. Forsberg, "Safety & Licensing Aspects of the Molten Salt Reactor", Trans. Am. Nucl. Soc. Ann. Meeting, (2004).

- [26] Thorium Energy Alliance, http://thoriumenergyalliance.com
- [27] The Weinberg Foundation, http://www.the-weinberg-foundation.org
- [28] P. Satyamurthy, L. M. Gantayet & A. K. Ray, PRAMANA 68, No 2, 343 (2007).
- [29] A. Ahmad, S.L. Sheehy & G.T. Parks, "The Effect of Beam Interruptions on the Integrity of ADSR Fuel Pin Cladding:", Annals of Nuclear Energy, 46, 97-105 (2012).
- [30] H. Takei et al., Journal of Nuclear Science & Technology, 49:4, p.384-397, (2012).
- [31] Ch.D. Bowman, ADNA Corporation Reports: ADNA97-12; ADNA/97-13; ADNA/97-14; ADNA/98-03; ADNA/98-04; Ann. Rev. Nucl. Part. Sci. 48, 505-56, (1998).
 Proc. 3rd Int. Conf. on Accelerator-driven Transmutation Technologies & Applications, ADTTA99, Prague, (1999).
- [32] D. De Bruyn et al., "Achievements and lessons learnt within the Domain 1 DESIGN of the Integrated Project EURO-TRANS". Technology & Components of Accelerator-driven Systems, Karlsruhe, OECD - NEA, p. 47-52, (2011).
- [33] "Spallation Neutron Source Project Completion Report", SNS 10000000-BL0005-R00 (2006).
- [34] S. Peggs, Ed., "ESS Conceptual Design Report", ESS report ESS-2012-1, (2012).
- [35] A. Hershcovitch et al., "Compact, Energy Efficient Neutron Source: Enabling Technology for Thorium Breeder & Accelerator Transmutation of Waste", APS Bulletin 54, (2009).
- [36] A. Hershcovitch & T. Roser, "Compact, Energy Efficient Neutron Source: Enabling Technology for Various Applications", BNL report C-A/AP/364, unpublished, (2009).
- [37] S.J. Steer et al., "Predicting the Contractual Cost of Unplanned Shutdowns of Power Stations: ADS Reactor Case Study", Elec. Power Systems Res., 81, 1662-1671 (2011).
- [38] J-L. Biarrotte et al., "Accelerator reference design for the MYRRHA European ADS demonstrator", LINAC 2010, (2010).
- [39] G. Rimpault et al., "Accelerator required characteristics for efficient ADS operation", submitted for publication to ANS Nuclear Technology, (2013).
- [40] P. McIntyre, "Strong-focusing cyclotron for high-current applications", these proceedings, (2012).
- [41] L. Calabretta, "A Multi Megawatt cyclotron complex to search for CP violation", Cyclotrons-10, (2010).
- [42] M. Tanigaki et al., "Construction of FFAG accelerators in KURRI for ADS study", APAC 2004, (2004).
- [43] S. Machida et al., "Acceleration in the linear non-scaling fixed-field alternating-gradient accelerator EMMA" Nature Physics 8, p 243 (2011).
- [44] H. Ait Abderrahim et al., "Accelerator & Target Technology for Accelerator Driven Transmutation & Energy Production," http://science.energy.gov/~media/hep/pdf/ files/pdfs/ADS_White_Paper_final.pdf, (2011).
- [45] A. Degtyarev et al., "Subcritical Molten Salt Reactor with Fast/Intermediate Spectrum for Minor Actinides Transmutation", Proc. of GLOBAL 2011, (2011).
- 08 Applications of Accelerators, Technology Transfer and Industrial Relations