PROSPECTS FOR ACCELERATOR-DRIVEN THORIUM SYSTEMS

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Abstract

To meet the tremendous world energy needs, systematic R&D has to be pursued to replace fossil fuels. Nuclear energy, which produces no green house gases and no air pollution, should be a leading candidate. How nuclear energy, based on thorium rather than uranium, could be an acceptable solution is discussed. Thorium can be used both to produce energy or to destroy nuclear waste. The thorium conference, organized by iThEC at CERN in October 2013, has shown that thorium is seriously considered by developing countries as a key element of their energy strategy. However, developed countries do not seem to move in that direction, while global cooperation is highly desirable in this domain. As thorium is not fissile, an elegant option is to use a proton accelerator to drive an "Accelerator Driven System (ADS)", as suggested by Nobel Prize laureate Carlo Rubbia. Therefore, the accelerator community has an important challenge to meet: provide the required proton beam for ADS.

BURNING FOSSIL FUEL TILL THE END?

If, by the end of the 21st century, people in developing countries are allowed to live as well as people do in Europe today, the world power consumption will have to increase by a factor 3 or more. Today, fossil fuels represent 87% of world primary energy consumption [1], and their consumption is still increasing, while resources are finite. Even in countries broadly developing renewable energies, fossil fuel consumption remains high, as fossil fuels are used as backup when there is no wind or no sun.

There are at least three good reasons to replace fossil fuels for energy production:

- Their impact on global warming – the atmospheric CO_2 level is higher than ever in the past 15 million years, and increasing faster than ever before. The IPCC reported in March 2014 that by 2100, a global temperature increase larger than 2˚C is more likely than one of less than 2˚C [2];
- Burning fossil fuel is having a severe impact on air pollution. Burning coal cost Europe alone 42.8 billion Euros in annual health care expenses [3]. The ambient air pollution caused the premature death of more than 400 000 Chinese in 2013. WHO reported that in 2012, around 7 million people died – 1 in 8 of total global deaths – as a result of air pollution exposure [4];
- At the present rate fossil fuel will run out relatively quickly on the human time scale, with present reserves of 53, 56 and 110 years [1], respectively for oil, gas and coal, while the current tendency is to increase fossil fuel consumption.

ENERGY R&D

Politicians will not invent the solution; it has to come from innovation. Innovation implies investment in both applied research and fundamental research. Without fundamental research there is no innovation.

Relying entirely on wind and solar energy would imply that, by the end of the century, their contribution to the world energy production would have to increase by a factor 130 or more. This is not realistic; at least not until the issue of the storage of electricity is resolved. In addition, the dispersed and fluctuating nature of these natural energy sources implies an important extension of the electric grid (50 000 km of new power lines in Europe).

Energy R&D has to be systematic, without prejudice. Nuclear fission energy, in particular, must not be left out. It produces no CO_2, no air pollution such as NOx, SOx, etc., and it has the potential to provide abundant, base load type of electric energy for many centuries. Furthermore, nuclear fission technology exists and is well understood.

The question that should be asked is: How should nuclear energy be exploited to be acceptable to Society? Where "acceptable" means that shortcomings of the present generation of critical reactors based on uranium, should be avoided: (a) accidents such as at Chernobyl, Three Mile Island, or Fukushima; (b) waste management (storage up to one million years is the only option developed so far); (c) proliferation of nuclear weapons (the uranium fuel cycle was developed for military purpose); (d) sustainability (uranium reserves for Pressurized Water Reactors (PWR) will last less than 100 years at the present rate).

THORIUM

Thorium is an abundant natural element: 1.2×10^{14} tons in the Earth's crust. It is as abundant as lead, and three to four times more abundant than uranium. Recovering only one part per million would provide the present world power consumption of 15 TW, for 18 000 years. "Thorium is a source of energy essentially sustainable on the human time scale", said Carlo Rubbia at ThEC13 [5].

Isotopically pure, natural thorium (^{232}Th) has an α-decay with a half-life of 14 billion years (almost stable). Thorium occurs in several minerals including thorite, thorianite and monazite and is often a by-product of mining for rare earths.

Known and estimated recoverable resources are between 6.6 and 7.4 million tons according to IAEA [6],

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which represents about 1000 years of present world energy consumption.

FISSION ENERGY FROM THORIUM

Thorium is fertile, not fissile, so it can only be used in breeding mode, by producing fissile ^{233}U (Fig. 1), in a neutron capture and decay chain analogous to that producing plutonium (^{239}Pu) from the isotope ^{238}U of uranium.

However, breeding uses almost all the thorium adding a factor 140 gain in reserves compared to ^{235}U in PWRs (in addition to the factor of 3 to 4 in abundance).

More importantly, the use of thorium minimizes long-lived nuclear waste production. For instance, it takes 7 successive neutron captures to produce ^{239}Pu from ^{232}Th. For similar reasons, the production of minor actinides is highly suppressed. As a consequence, thorium in a fast neutron flux may also be used to destroy nuclear waste produced by present nuclear power plants. There are other interesting properties of thorium: the high melting point of thorium dioxide, the highest of all oxides and one of the best refractory materials (3300°C compared to 2865°C for UO_2) and the high melting point of metallic thorium (1750°C compared to 1130°C for metallic uranium).

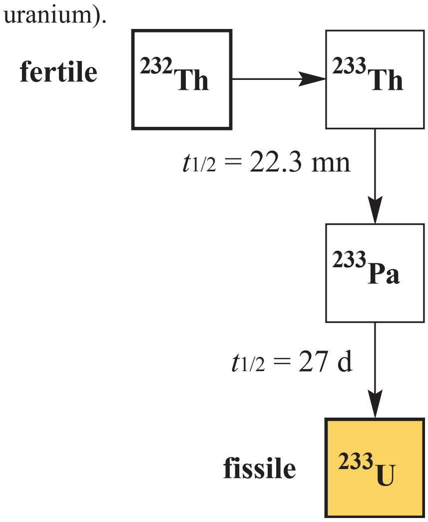

Figure 1: ^{233}U breeding chain from ^{232}Th. Horizontal and vertical arrows indicate neutron captures and β-decays, respectively.

The thorium fuel cycle has a major advantage over the uranium fuel cycle, in that it is very resistant to the proliferation of nuclear weapons [7].

WHY IS IT CHALLENGING TO USE THORIUM?

Firstly, it is necessary to produce (breed) ^{233}U from thorium in some way. Then, thorium mixed with a fissile element cannot be simply substituted to PWR fuel because of neutron inventory issues, mainly due to the larger neutron capture cross-section of thorium and to the long half-life of the intermediate element, protactinium (^{233}Pa, $t_{1/2} \approx 27$ days).

Even though ^{233}U is generally a better fissile element than ^{235}U and ^{239}Pu, it is precisely where one would want to use thorium to minimize nuclear waste production, namely in the fast neutron part of the energy spectrum, that ^{239}Pu is somewhat better than ^{233}U.

WHAT ARE THE OPTIONS?

There are three main methods for using thorium:

- Breeding ^{233}U in thorium blankets around critical reactors to introduce it in advanced thermal or fast critical reactors;
- Continuously recirculating or recycling the burnt fuel after removing accumulated neutron poisons and continuously refuelling the reactor, in order to always have fresh fuel, to guarantee a positive neutron inventory. This is the solution implemented in pebble bed and molten salt critical reactors;
- Using a proton accelerator to provide the excess neutrons needed to sustain the neutron capture and fission chain in a subcritical system, but also to burn unwanted nuclear waste. This is the so-called Accelerator Driven System (ADS), as proposed for instance at CERN in the 1990s by C. Rubbia [7] and promoted by iThEC [8].

The Indian Three-Stage Strategy

India, with little uranium but abundant thorium resources, has the most advanced working scheme for using thorium (including front-end and back-end of the fuel cycle) [9]:

- Use heavy water (CANDU) or light water (LWR) reactors to produce plutonium from India's small uranium supply;
- Use sodium-cooled uranium-plutonium fast reactors with a thorium blanket to breed ^{233}U;
- Reprocess blankets and manufacture ^{233}U-thorium fuel for advanced thermal reactors.

The Indian scheme certainly works from the technical point of view. However, issues remain concerning the complexity of developing and maintaining three nuclear technologies; the sustainability, as it requires uranium in the first stage; and the lack of a solution for the accumulated nuclear waste.

Pebble Bed Critical Reactors

Farrington Daniels at Oakridge National Laboratory (ONL), in the USA, first proposed pebble bed reactors, in the 1940s. Initial developments took place in Germany (AVR Jülich), followed by the THTR-300 project [10]. Further developments were made in South Africa, the USA, China and Turkey. These are generally graphite-moderated, gas-cooled, high-temperature critical reactors, in which pyrolytic carbon pebbles coated with fireproof silicon carbide containing the fuel circulate through the core. These systems have several drawbacks:

- Passive cooling by natural air convection, a desirable feature, implies no containment in case of accident;
- Water cannot be used for cooling in case of accident;

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- Graphite which is used as moderator is flammable;
- The burnup is limited and minor actinides have a small fission probability, as the neutron flux is thermalized by the graphite. Pebble bed reactors produce more high-level nuclear waste than current nuclear reactors;
- Fuel handling is delicate, as pebbles are cycled through the reactor. It was an accident in the pebble circulation and instability of the fuel temperature of the AVR that stopped the development in Germany;
- Reliance on highly resistant fuel pebbles makes nuclear waste partitioning and transmutation virtually impossible.

Molten Salt Critical Reactors (MSR)

This is a technology that is surprisingly concentrating industry's interest world-wide, in China, India, the UK, the USA, the Czech Republic, France, and even Switzerland, at least until recently. MSR were pioneered at ONL in the 1960s, with the 7.4 MWth Molten Salt Reactor Experiment, using UF_4.

In MSR, the fuel circulates in the core as a high-temperature molten salt mixture, and there is chemistry on-line, in the fuel loop outside the core, to extract fission fragments and eventually protactinium, in order to help the neutron inventory.

The main advantages are: liquid fuel allows the burnup to extend indefinitely, as a result of on-line reprocessing; the heat is produced directly in the heat transfer fluid; the high temperature (500˚C – 600˚C) is favourable to the conversion of heat into electrical energy; and passive cooling is possible for decay heat removal, after dumping the molten fuel by gravity into a reservoir underneath the core.

There are however important issues: delayed neutron emission occurring outside the core; possible failure of on-line chemistry, which would make it dangerous to re-inject cooler fuel into the core; corrosion with high temperature salts; the possibility to extract quasi pure highly proliferating ^{233}Pa; and the resulting delicate licencing issues.

Furthermore, unless salts other than lithium fluoride are developed, these systems will produce a thermal to epithermal neutron energy spectrum, which does not favour transmutation of minor actinides.

There is a particularly well-focussed and most ambitious effort in China [11]. At the end of March 2014, the Chinese Government decided that the first fully functioning thorium MSR reactor should be built within ten years, instead of the 25 years, originally considered.

Accelerator-Driven Systems (ADS)

The third and probably the most elegant way of using thorium consists of providing extra neutrons, with an external source, using a proton accelerator. The first use of an accelerator in this context goes back to G. Seaborg who produced the first μg of ^{239}Pu, in 1942, with the Berkeley 60 inch cyclotron. In the 1950s, Lawrence's (MTA) and Lewis' ADS projects were dropped or slowed down when rich uranium deposits were discovered in the USA, and it was realized that several hundred mA of beam intensity, hundreds of MW, would be needed to produce the required beam. No amplification was built into the system. In contrast, today's systems only need 1 to 10 MW of beam power.

There was renewed interest in ADS in the 1980s, when the USA decided to slow down the development of fast critical reactors. The Fast Flux Test Facility [12] at Argonne National Laboratory was shut down by DOE in 1993.

- H. Takahashi at Brookhaven National Laboratory proposed several ADS systems including the idea of burning minor actinides (PHOENIX [13]);
- Ch. D. Bowman at Los Alamos National Laboratory, proposed a thermal neutron ADS [14] for the transmutation of nuclear waste (ATW) using molten thorium fuel and chemistry on-line to extract fission products and ^{233}Pa;
- Japan launched Options for Making Extra Gains from Actinides (OMEGA [15], now JPARC) at JAERI (now JAEA).

In the 1990s, Carlo Rubbia gave a major push to the ADS technology [7], by launching a vigorous research programme at CERN, based on the development of innovative simulation of nuclear systems, specific experiments to test basic concepts (FEAT [16], TARC [17]), and construction of an advanced neutron Time of Flight facility (n_TOF [18]) to acquire neutron cross-section data, crucial to simulate reliably any configuration with new materials

ADVANTAGES OF THORIUM ADS

Safety

ADS eliminates the possibility of criticality accidents by allowing the system to be subcritical. Therefore, void coefficient, temperature coefficient, delayed neutron fraction β_{eff} are no longer "critical" parameters. However, subcriticality requires an external neutron source.

As any future generation nuclear system, ADS can be operated with passive safety features. This is especially true of cooling to avoid core melt down or limit its consequences, by borrowing from US advanced fast critical reactor designs (RVACS [19]).

ADS will not use dangerous coolants such as liquid sodium foreseen in some Generation IV options [20]. Lead or lead-bismuth eutectic mixture, are proposed as both target for protons and coolant, guaranteeing a fast neutron spectrum.

Waste Management

Compared to uranium once through fuel cycle, the combination of fast neutrons, thorium based fuel, and recycling of long-lived transuranic actinides (TRU) reduces long-lived waste production by several orders of magnitude. It also allows efficient destruction of the long-term component of present nuclear waste, the TRU.

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Military Proliferation

The use of thorium fuel implies insignificant production of neptunium, plutonium and other minor actinides. The uranium extracted from the spent fuel is a mixture of uranium isotopes, with a critical mass of about 28 kg [7], which implies a strong gamma emission coming from the decay chain of ^{232}U, lethal in a 10 mn exposure at less than one meter. Therefore, it would be extremely difficult to manufacture a bomb with such mixture.

In the re-processing of spent nuclear fuel in a fast neutron ADS, it is not necessary to separate out plutonium as is done in PUREX [21]. Therefore, the pyro-electrolysis method can be used to extract the entire TRU mixture to manufacture fresh thorium based fuel, unlike what is currently done with MOX fuel in France, where only plutonium is recycled.

BASIC PROPERTIES OF SUBCRITICAL SYSTEMS

The theory of subcritical systems is interesting in itself, to get insight into their physical properties, which are quite different from those of critical systems. In particular, their response to fast reactivity changes is spectacularly more moderate than for critical systems. The knowledge of the neutron flux geometry is important for calculating the generated power distribution and the uniformity of fuel burnup. Even though it is nowadays possible to simulate precisely extremely complex systems, with Monte Carlo methods, an analytical approach can be used by making some simplified assumptions to extract the general properties of such systems and compare them with critical systems [22].

A unique property of subcritical systems is that the neutron multiplication factor changes whether the accelerator is on ($k_s \equiv k_{source}$) or off (k_{eff}). When the accelerator is on, non-fission neutron multiplication due in part to the hard energy spectrum of spallation neutrons is important, through (n,Xn) reactions on lead. The change of geometrical flux distribution between the fundamental mode characterizing k_{eff} and the neutron flux geometry with the source characterizing k_s, also plays a role. As a consequence, k_s is always smaller than k_{eff}, which means that switching off the accelerator, hence the neutron source, not only stops the main power generation, but also moves the system further away from prompt criticality. This is a major safety asset for ADS.

Accelerators for ADS

The required accelerator power (P_{beam}) can be expressed as a function of the desired ADS fission power (P_{ADS}), the neutron multiplication coefficient k_s and G_0, a constant that depends on the beam energy, the target material and the detailed geometry of the system:

$$P_{beam} = \frac{\left(1-k_s\right)}{k_s G_0} P_{ADS}$$

Therefore, the accelerator power is a trade-off between accelerator power and criticality margin (Fig. 2). For given k_s and G_0 the fission power changes with the beam power, allowing the possibility of modulating the power output. This could be a useful feature if ADS systems are associated with fluctuating renewable energy sources.

Compared to uranium, neutronics with thorium is very favourable to power modulation because of the much longer half-life of ^{233}Pa (27 d) compared to ^{239}Np (2.3 d). What was a problem with the use of thorium in critical reactors becomes an advantage in the case of ADS.

The Paul Scherrer Institute (PSI) separate-turns cyclotron achieved a beam power of 1.4 MW (2.4 mA, with 0.59 GeV protons) [23]. This beam could already produce, a power P_{ADS} = 210 MW_{th} with $k_s = 0.98$, in an Energy Amplifier, as designed by C. Rubbia [7].

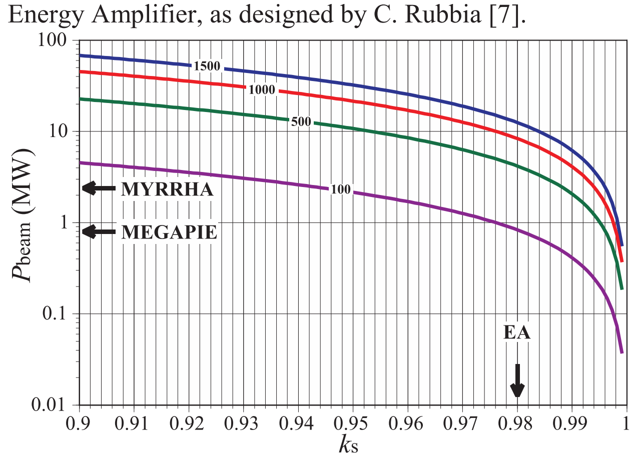

Figure 2: Beam power versus k_s. Curves of constant fission power are shown, labelled in MW_{th}. The k_s value for C. Rubbia's Energy Amplifier [7] is indicated, as well as the beam power achieved with MEGAPIE [24] and planned for MYRRHA [25].

Accelerator Requirements for ADS

In principle, it does not matter how the external neutron source is provided. In practice, for industrial applications, there are a number of well-defined requirements for the accelerator:

Beam particle: The choice is protons for their simplicity of production and because they are most efficient in producing neutrons by spallation.

Beam Energy: Optimum neutron production is obtained for $E_{beam} \geq 900$ MeV. At lower energy, protons tend to loose more energy by ionization, which does not produce neutrons. Above 900 GeV, there is an energy gain plateau, as shown by the FEAT experiment [16], slowly decreasing as pion production increases.

Beam power: Typically 1 to 10 MW depending on the choice of k_s value, and on the desired power output. A large operational range of beam intensities might be required to follow electrical power demand. The maximum range will depend on the core design. Beam power stability is obviously important, as a 1% fluctuation of beam intensity causes a 1% fluctuation of the thermal power.

Beam spot size (footprint): Large at impact on window separating the vacuum chamber from the target (studies at JAEA have shown that densities of up to 0.1-0.2 mA/cm^2 can be reached today). For MYRRHA [25] the design value is 0.07 mA/cm^2.

Beam losses: They have to be controlled such as to minimize irradiation of the accelerator elements and of the environment. The figure of merit is 1 W/m for LINACs, while beam losses are localized for cyclotrons. Beam losses are a main issue for any high power beam, not only for ADS. They have direct impact on maintenance and repair.

Reliability: Fatigue of mechanical structures, in particular of fuel elements, requires the minimization of beam trips. For instance, for MYRRHA, there are no constraints on the number of trips of duration of less than 0.1s. However, no more than 100 trips per day with 0.1 s $< T_{trip} <$ 3 s, and 10 trips in three months with $T_{trip} >$ 3 s are allowed [25].

Energy efficiency: One must maximize the accelerator electric power efficiency, $\eta \equiv P_{beam}/P_{grid}$. This is relevant to the overall energy efficiency of the system.

Size of accelerator: This might be a feature in favour of cyclotrons, since for nuclear waste elimination, one might want the accelerator to fit within the site of a nuclear power plant, to avoid transport of waste.

Cost: This is obviously a very important element to be taken into account.

In the end, the solution chosen among LINAC, Cyclotron or FFAG technologies, will be the one best fulfilling all of the above requirements.

To conclude, one main criticism of ADS has been that “the accelerator does not exist and will be too expensive”, this is obviously a challenge to take on by the accelerator community.

ADS Developments

Even though R&D on ADS is certainly not at the level required by the importance of the energy issue, a large amount of development is taking place, worldwide.

The PSI cyclotron beam has already reached the power range of industrial applications. MEGAPIE [24], a spallation target ran successfully for three months at SINQ, the Swiss Spallation Neutron Source, at a power of 0.8 MW. SNS, the US Spallation Neutron Source [26], is running at ONL, at 1.4 MW.

The MYRRHA project at SCK•CEN, Mol, Belgium, could be the first ADS prototype of significant power, if funded. It will use a LINAC (≤ 4 mA and ≤ 2.4 MW, with 0.6 GeV protons), but unfortunately thorium is not on the agenda, and the system will be transformed into a critical research reactor after only a few years of operation as an ADS.

At Troitsk [27] in Russia and at the Institute of Modern Physics in China (CADS) [28] ADS is considered for burning minor actinides, and a discussion in India is taking place to use ADS to simplify the Indian thorium utilization scheme.

Japan recently re-launched the Transmutation Experimental Facility program (TEF-T and TEF-P) at J-PARC, which is now part of the roadmap toward the ADS proposed by JAEA for nuclear waste transmutation and consists of two activities, under JPARC:

• Development of the ADS Target Test Facility (TEF-T) to verify the feasibility of the beam window, which is a challenge for ADS and to consider it as a material test facility;

• Further development of the Transmutation Physics Experimental Facility (TEF-P) to overcome difficulties in reactor physics issues such as subcritical core and a minor actinide loaded core.

There are other new ideas, which were presented at the ThEC13 conference: Molten Salt ADS by C. Rubbia, C. Pyeon (Japan) [29], and J-S Chai (Korea) [30]. There are also relevant studies of corrosion with high temperature lead or lead-bismuth eutectic mixture, material compatibility, which have resulted in the production of new material resistant to corrosion in lead up to about 550 ˚C.

DESTRUCTION OF NUCLEAR WASTE

ADS associated to thorium fuel is the only practical way of destroying minor actinides, a component of long-lived nuclear waste. Plutonium can also be destroyed efficiently as shown by C. Rubbia, for instance in the case of Spain [31]. According to the simulation validated by the FEAT [16] and TARC [17] experiments, an ADS, such as an Energy Amplifier, could destroy three times the amount of waste produced by a PWR running at the same thermal power [7]. The destruction of plutonium and of minor actinides by fission releases energy that can be used to produce electricity, thereby minimizing the cost of destroying nuclear waste.

CONCLUSION

The energy problem is too important not to explore systematically all options for the development of abundant, clean and safe energy sources. There is no reason to keep thorium out of the energy R&D effort, especially in developed countries, which already master the technological know-how, and should play the leading role [32].

The physics of Accelerator-Driven Systems is entirely understood. Conceptual designs exist. Now a prototype of significant power is needed to validate technological solutions and to learn how to operate such systems.

When taking into account the need for safety, proper waste management and non-proliferation, thorium in a fast neutron ADS is a most promising option for energy production and waste elimination.

ADS is a challenging innovation but there is no show stopper. The ball is clearly in the camp of the accelerator community.

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