Why Accelerator-Driven Transmutation of Wastes Enables Future Nuclear Power?

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Abstract.

Criticality concerns, decay heat management and radioactive waste handling are perceived as the primary, unsatisfactorily resolved technological problems of nuclear reactors. They all originate from very specific features of a fission phenomenon: self-sustained chain reaction in fissile materials, very strong radioactivity of fission products and very long half-life of some of the radioactive fission and activation products

Accelerator-driven transmutation systems, which operate in a subcritical mode and stay subcritical, regardless of the beam being on or off, can in principle address the safety issues associated with criticality, particularly for advanced fuel containing a high fraction of minor actinides. Subcriticality can also improve the controllability of this nuclear system through a simple electronic control of the accelerator. Subcriticality provides also substantial flexibility in fuel processing and managing. Accelerator-driven transmutation systems can accept such fuels that would be impossible or difficult to use in critical reactors, and can extend their cycle length ensuring good transmutation performance. Moreover, an advanced subcritical core design can also address some concerns of decay heat management.

However, a significant development of accelerator technology has to be achieved before a construction of the first industrial ATW facility can be realized. The high-intensity accelerator with a beam power in the range of 10-100 MW has to be available with the stability, efficiency, reliability, operability and maintainability features never demanded before from the accelerator technology.

1 INTRODUCTION

This review article due to the scope of the LINAC2000 conference does not cover all of the important aspects of Accelerator-driven Transmutation of Waste (omitting for example reprocessing chemistry) and has limited reference list. For more complete references look into [1]

Nuclear reactors based on self-sustained fission reactions, or so called “critical” reactors - after a spectacular development in fifties and sixties of the 20th century, that resulted in deployment of over 400 nuclear power reactors - are today wrestling more with public acceptance than with irresolvable technological problems. In a whole spectrum of reasons, which resulted in today’s opposition against nuclear power, some of them can be effectively addressed by a successful combination of nuclear and accelerator technologies. These hybrid systems, commonly called Accelerator-Driven Systems (ADS) or Accelerator-Driven Transmutation of Wastes (ATW), integrate a subcritical reactor core, i.e. a fissile material assembly unable to support a self-sustained chain reaction, with an intense spallation neutron source driven by a powerful particle accelerator. This intense neutron source supports the desired fission reaction rate in a fissile assembly taking advantage of the finite neutron multiplication capabilities of this assembly.

The basic goal of ATW is reduction of hazards related to handling and management of radioactive wastes through nuclear transmutation and improvement of operational safety of nuclear power facilities.

When coupled with the spallation process, high power accelerators can be used for an effective transmutation. Typically, several tens of neutrons will be produced from each proton colliding with the target. This means that a reasonable beam of protons (for example 5-10 mA at 1 GeV of proton energy) can produce a large number of neutrons per unit of time - see Fig. 1 [2]. The typical spectrum of neutrons emerged in spallation processes is presented on Fig. 2. This neutron spectrum is not very different from a typical fission neutron spectrum, having the most neutrons emerging with energies between 1 and 2 MeV. However, one can observe a very distinct difference – a tail of high-energy neutrons (with a yield of about 10 %) over 20 MeV reaching the maximum energy equal to energy of the incident protons.

2 TRANSMUTATION PROCESSES

Nuclear transmutation can be practically induced by any particles or quanta enabled to penetrate nuclei and to interact with nucleons. However, charged particles have to pass through a Coulomb barrier, which requires high energies and it is an energetically costly and ineffective process. $\gamma-$quanta on the other hand, have relatively small cross sections for transmutation reactions - like (γ,n) reactions – and moreover there are no monoenergetic γ-sources, making γ-transmutation energetically very ineffective. The most effective nuclear process that can be used for transmutation of radiotoxic isotopes is doubtless neutron absorption. Neutrons are not repelled by nuclei and interaction cross

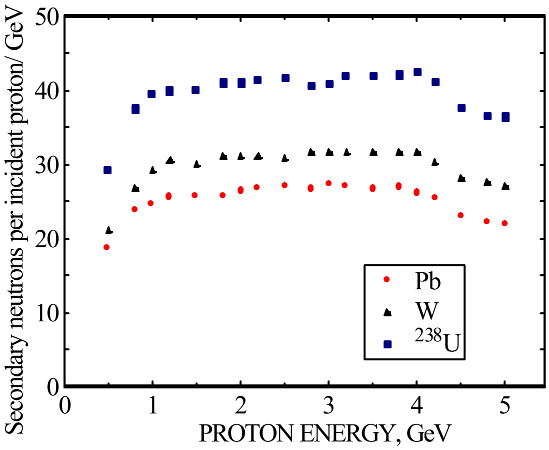

Figure 1. Number of neutrons per incident proton and its energy (GeV) produced in a spallation processes in different thick targets.

sections for many transmutation reactions are sufficiently large.

A final measure of transmutation efficiency, or its "figure of merit" is not a trivial issue as long as a final criterion for hazards related to radioactive waste is not well defined. Therefore, **reduction of source radiotoxicity of the nuclear wastes** seems to be the least controversial reference goal for transmutation of radioactive wastes.

In a short time perspective, like 100 years, fission products ^{90}Sr and ^{137}Cs, and Pu – isotopes, dominate radiotoxicity of spent fuel. ^{90}Sr and ^{137}Cs belong to short-lived fission products and are of big concern in a case of nuclear accident. They can however, be readily retained in storage facilities for reasonable periods to minimize their threat to the human environment. Neither ^{90}Sr nor ^{137}Cs can be effectively transmuted by neutron absorption.

In the long time, comparable with life-time of containers in geological repositories, the radiotoxicity is determined by transuranic elements: ^{239}Pu (up to 100 000 years), ^{242}Pu, ^{237}Np and long-lived fission products ^{129}I, ^{135}Cs and ^{99}Tc. Plutonium and other actinides have a very low mobility in geological environment, so they do not easily enter the biosphere. On the contrary Iodine, Cesium and Technetium, being much more mobile can leak the geological repository. Proliferation concern is another strong argument for transmutation of actinides, particularly Plutonium. Increasing a worldwide stockpile of Plutonium in spent reactor fuel must be of concern, above all in few hundred-year perspective when a "protective" barrier of radioactivity of short-lived fission product will decay out. Fortunately, most of these isotopes of concern can be effectively transmuted.

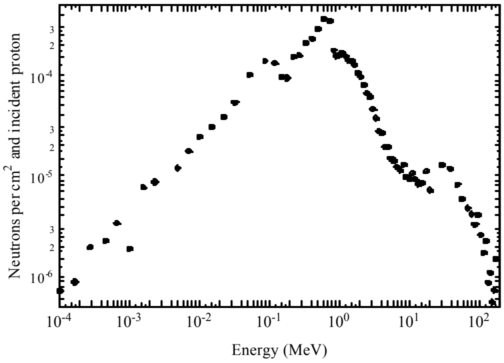

Figure 2. Energy spectrum of spallation neutrons produced by a 1 GeV proton beam.

Virtually every transuranic elements, Np, Pu, Am and Cm can be fissioned by one or few successive neutron absorptions with, in many cases, energy surplus, neutron gain and transmutation of transuranic elements into fission products. All transuranic isotopes are net neutron producers in fissions induced by fast neutrons (so called fast spectrum fissions). In thermal neutron spectrum corresponding to Light Water Reactors, only ^{239}Pu, ^{241}Pu and Cm-isotopes are "unconditional" neutron producers, other transuranic isotopes like ^{237}Np, ^{238}Pu and ^{241}Am may become neutron producers in a very high neutron flux. In these cases beta-decay of the intermediate neutron capture products competes with a fission probability.

Transmutation of fission products through neutron absorption is also possible for the long-lived radiotoxic isotopes like ^{99}Tc and ^{129}I, converting them into stable Ru and Xe, respectively. However, transmutation of fission products, in contrary to transmutation of transuranic isotopes, is a purely neutron consuming process and requires excess of neutrons. This surplus of neutrons can be obtained in different ways:

In critical reactors, which can be designed as "burners", in order to use all available neutrons for transmutation processes. Only reactors with an excellent neutron economy can be burners, which limits the choice to fast reactors with the hardest possible neutron energy spectrum, revival of heavy water moderated reactors or use of highly enriched fuel in standard LWRs. Neither of these choices is very probable today. Moreover, criticality conditions, dependence of safe reactor operation on a delayed neutron fraction and negative temperature feedbacks put severe constraints on the possible use of critical reactors;

In subcritical systems driven by an intense external source of neutrons – in ATW. An external neutron source and subcritical operation open new possibilities for transmutation.

3 ACCELERATOR-DRIVEN TRANSMUTATION

The main components of ADS are a high-intensity accelerator delivering a particle beam of 5 to 40 MW power, a transmuter - a sub-critical reactor with spallation source, and chemical reprocessing – see Fig. 3.

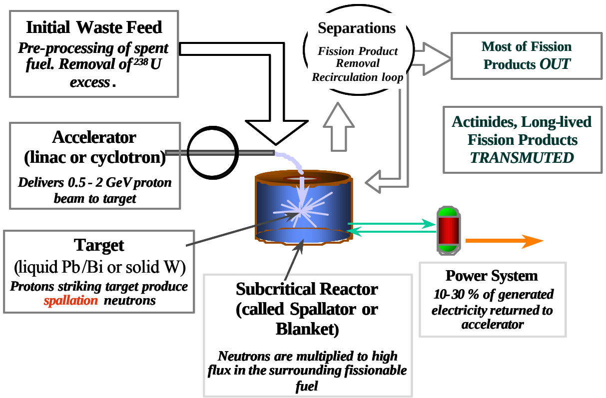

Figure 3. A schematic view of an Accelerator-Driven Transmutation System

When a particle beam (in most designs - protons) from accelerator hits a beam target of heavy elements, large quantities of neutrons and charged particles are obtained, largely through spallation of the atomic nuclei in the target. Most of the charged particles are slowed down and stopped inside the target or in its vicinity as an effect of Coulomb interaction, while the neutrons easily penetrate the target and surrounding subcritical core. If the spallation target is placed in the center of a subcritical core, the latter can act as a neutron multiplier even if it would not otherwise be self-sustaining. This is due to the fact that losses of neutrons can be compensated for through the supply of new neutrons from the spallation target. Through the fissions that occur in the core during neutron multiplication, more energy can be generated than is consumed to produce the accelerator beam. The external neutrons supplied by spallation target sustain a constant power of the system and play the same role as delayed neutrons in critical reactors. This results in another type of "self-sustaining" system, in which delayed neutrons are replaced by the spallation neutrons. Consequently, k_{eff} may have values much below 1. That is this system is called subcritical.

The neutrons emerging from both the target and the fuel in the subcritical core originally have high energies varying from a "usual" fission spectrum energies up to an energy of incident protons, see Fig. 2. By introducing a moderator, the neutron energy can be reduced (neutrons can be slowed down) in the same way as in a thermal reactor. The advantage of this is that most reaction cross-sections are greater at low neutron energies than at high energies. Thus, less fissile material is needed for a given reaction rate at low neutron energies than at high neutron energies. Conventional moderators - water and graphite - normally require encapsulated solid fuel and are therefore less suitable as moderators in ATW, mainly due to the large gradients in power density. In subcritical systems power density varies in space as exponential function not as cosine or Bessel functions like in critical systems. It results with high power densities around the spallation target and low power on peripheries. Consequently, nuclear fuel in a liquid form is required, e.g. molten salts, where actinides are dissolved in different types of fluoride salts. More sophisticated solutions can be also applied, like multiple target system, in which subcritical core surrounds 3-5 target modules fed by split proton beam or even fed by separate accelerators (in this case only use of cheap cyclotrons makes economical sense) [1]. Acceptable fission power distribution in the core can be obtained in this way but the technical complexity of this system increases considerably. Moreover, one has to cope with a very significant reactivity swings requiring either sophisticated fuel feeding procedures or very flexible accelerator working with current varying almost by a factor of 5 [1].

Using fast neutron spectrum it is easier to design a suitable subcrtical core for ATW than for a critical fast reactor, since the spallation source can deliver neutron flux of very high intensity. Also longer neutron free flow path in the fast systems makes the power peaking problem much less severe than in the thermal systems and consequently makes possible use of solid, reactor like fuel rods.

The conversion of heat from the core into electricity is more than sufficient to operate the accelerator.

4 COMPONENTS OF ACCELERATOR-DRIVEN SYSTEMS

4.1 Accelerator

Linear or cyclotron accelerators have been proposed for ATW. Historically, the proposed accelerator parameter used to be well ahead of accelerator technology state of the art – see Fig. 4. In spite of a vivid discussion in the accelerator community [3] the final choice of an accelerator type will depend on an intercomparison and complex optimisation of the whole ATW-system including economical constraints and also local traditions of research groups, which will succeed to build the first demonstration facility. The optimal parameters of the accelerator are relatively easy to estimate from a nuclear point of view: proton energy should be around 1 – 1.5 GeV (see Fig. 1 – there is no significant gain in number of neutrons per proton energy over 1 GeV) and proton current would depend on desired beam power, which for a demonstration facility would be in the limit of 4–10 MW, corresponding to 5 – 10 mA of the proton current.

However, taking into account development and constructional costs, it may appear that even

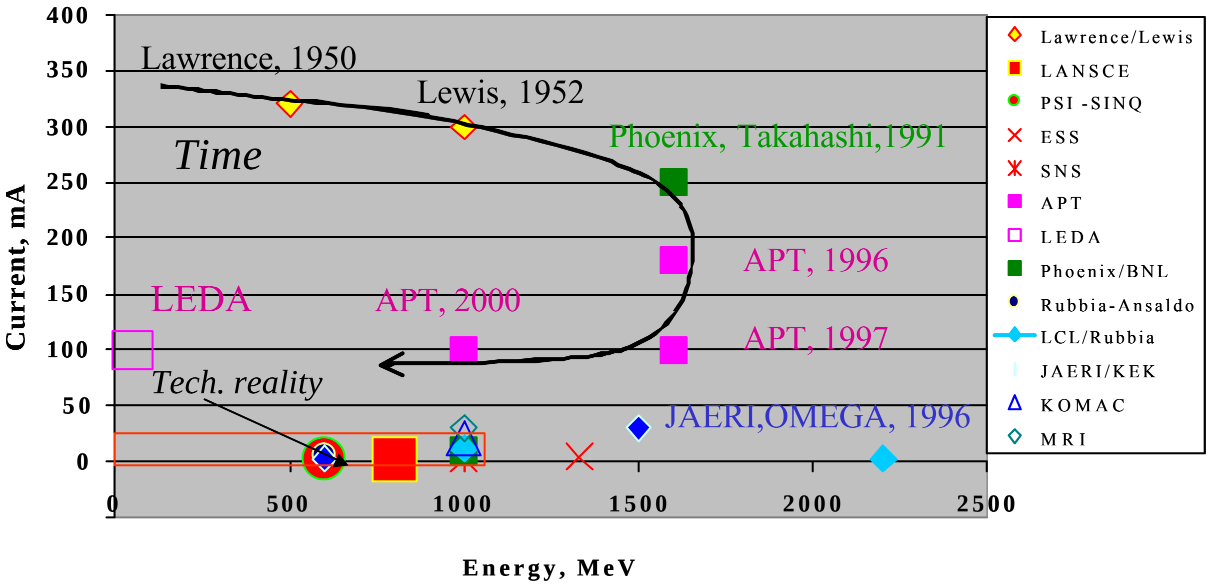

Figure 4. Accelerators proposed for ATW or other spallation neutron sources.

accelerators with proton energy around 500-800 MeV may be interesting for the first demonstration systems. Moreover, economical assessments for commercial accelerator-driven systems, taking into account also decommissioning costs may significantly differ from assessments for demonstration facilities.

Two most powerful accelerators today, represent both accelerators types: linac at Los Alamos National Laboratory, running (if required) at 800 MeV and 1 – 1.5 mA proton beam, and cyclotron at Paul Scherer Institute, having a 1.8 mA proton beam with 590 MeV energy. Both accelerator types require intensive development in order to match the requirements that are common for nuclear power systems. For example the number of accelerator beam trips must be reduced by few order of magnitudes, from several thousands trips per year which is a common number both for the PSI cyclotron and LANL linac, down to about several tens trips a year [1]. The reliability and availability of the accelerator in accelerator driven systems is one of the most important issues.

Even if there are no obvious showstoppers for improvement of accelerators, an intensive research work should focus on:

for linacs:

- Improved reliability and trip-free performance,
- Extensive use of superconductors (development of lower-beta superconducting cavities and cryomodules), high-gradient superconducting rf-cavities,
- Increase of electrical field gradients leading to reducing the size,
- Increase of current and possibly beam splitting/sharing to share accelerators in development stage;

for cyclotrons:

- Improved reliability and trip-free performance,
- Increase of beam current - novel concepts overcoming space charge difficulties.
- Cost reduction through compactness and robust constructions.

4.2 Target

Few different technical solutions have been envisaged for an ATW spallation target. A solid tungsten target clad in stainless steel and cooled by sodium and a liquid metal target, in which target fluid is also used as the primary cooling loop have been so far presented as the most promising options. The solid tungsten target design was developed in details in the APT project [4], however this solution for ATW system is less attractive than a liquid metal target; e.g. a liquid Lead-Bismuth eutectic (LBE). Other possible metals are liquid Lead-Magnesium eutectic and Mercury. Mercury target is a preferred option for spallation neutron scattering facilities, like Spallation Neutron Source (SNS) [5] project and European Spallation Source (ESS) [6].

The advantages of LBE are chemical inertia, high boiling temperature, relatively low melting temperature (123.5°C), good heat conductivity and no immediate volume expansion upon solidification (however slow volume expansion in a solid state due to recristallization requires some precautions). A significant disadvantage of the LBE spallation target is generation of ^{210}Po, a short-lived hazardous alpha emitter formed by neutron irradiation of Bismuth.

The key technological problem for the target design is a design of a target window which can withstand radiation damages of proton beam and backscattered neutrons, thermal stresses caused by accelerator trips and corrosion in LBE environment. Effects of spallation products on LBE corrosion control are one of the key problems to be investigated.

4.3 Subcrtical core: coolant/fuel system

Transmutation efficiency and the system performance depend very strongly on the choice of coolant and fuel types. Different conceptual designs have been proposed for the coolant/fuel systems in the last few years. The very common feature for most of them is the choice of a fast neutron spectrum in order to transmute efficiently minor actinides. As a consequence a liquid Lead or LBE as a coolant became a primary choice of many preconceptual designs. Several options of ATW fuel have been also proposed varying from a solid oxide fuel (e.g. Pu+MA) based on a well established fuel technology to unexploited metal and nitride fuels. In the longer time perspective Th-based fuel cycle is considered as particularly attractive for ATW. This fuel cycle, having worse neutron economy than U fuel cycle would additionally benefit from the external neuron source.

For a solid fuel system cooled by LBE there is an open question of spallation target integration into a subcritical core. Having the same coolant, the spallation target could be integrated into the core cooling system having the same first cooling circuit. It would definitely simplify the construction of the ATW, however the price would be a contamination of the whole primary liquid metal loop with the spallation products. So it will probably be preferable to have a spallation target with a separated cooling circle, to contain the spallation products into the minimal volume.

Another ATW option which is now considered is an advanced gas-cooled (He) ATW with LBE spallation target and MOX or even more advanced fuel, like particle fuel. Other very novel systems are also under considerations, like thermal neutron ADS cooled with liquid lead solution of Pu and PuMA, or liquid lead suspension of Pu and/or PuMA oxides. Conceptual design of this ATW, called Jülicher Transmuter, has been developed in the IABAT-project [1].

For an effective transmutation, ATW system requires advanced fuel like metallic, Uranium free-fuel, blend of actinides and Zirconium. This metallic fuel provides the high rates of heat transfer required. Use of a metal fuel makes pyrometallurgical processing attractive for recovery and recycle of the discharged ATW fuel. However, Uranium (or Thorium) free fuel implies a rapid drop of k_{eff} with burnup of the fuel. To compensate reactivity drop and to keep a constant power level, accelerator has to deliver particle current varying by a factor of 3-5 during a single irradiation period, moreover such a core may require several reloading per year. MA-fuel without ^{238}U can be only used in subcritical systems, it is unacceptable in critical reactor (as a full load) due to the small delayed neutron fraction and small Doppler effects.

Nitride fuel cores with enriched ^{15}N can also ensure good transmutation performance, however this technology is even less developed than metallic fuel and requires significant R&D efforts.

5 CONCLUSIONS

Accelerator-Driven Transmutation Systems open new possibilities to perform transmutation of nuclear waste addressing some of important concerns related to conventional nuclear power. It relaxes criticality concerns, has a potential for effective transmutation of nuclear wastes including incineration of a Pu-stockpile. Spent fuel from existing Light Water Reactors can be effectively transmuted in ATW, with radioactive waste streams with virtually no actinides and free of Tc and the long-lived Iodine isotope, i.e. without the most cumbersome isotopes. ATW opens also new possibilities to design subcritical nuclear power reactors combining transmutation with commercial nuclear energy generation. Development of these transmutation systems requires an extensive research program of interdisciplinary dimension covering nuclear physics, nuclear technology including high intensity, medium energy accelerators and spallation targets, reactor physics, material sciences, chemistry and nuclear chemistry, radioactive waste treatment technologies etc.

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