TRANSMUTATION OF ISOTOPES — ECOLOGICAL AND ENERGY PRODUCTION ASPECTS*

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This paper describes principles of Accelerator-Driven Transmutation of Nuclear Wastes (ATW) and gives some flavour of the most important topics which are today under investigations in many countries. An assessment of the potential impact of ATW on a future of nuclear energy is also given. Nuclear reactors based on self-sustained fission reactions — after spectacular development in fifties and sixties, that resulted in deployment of over 400 power reactors — are wrestling today more with public acceptance than with irresolvable technological problems. In a whole spectrum of reasons which resulted in today's opposition against nuclear power few of them are very relevant for the nuclear physics community and they arose from the fact that development of nuclear power had been handed over to the nuclear engineers and technicians with some generically unresolved problems, which should have been solved properly by nuclear scientists. In a certain degree of simplification one can say, that most of the problems originate from very specific features of a fission phenomenon: self-sustained chain reaction in fissile materials and very strong radioactivity of fission products and very long half-life of some of the fission and activation products. And just this enormous concentration of radioactive fission products in the reactor core is the main problem of managing nuclear reactors: it requires unconditional guarantee for the reactor core integrity in order to avoid radioactive contamination of the environment; it creates problems to handle decay heat in the reactor core and finally it makes handling and/or disposal of spent fuel almost a philosophical issue, due to unimaginable long time scales of radioactive decay of some isotopes. A lot can be done to improve the design of conventional nuclear reactors (like Light Water Reactors); new, better reactors can be designed but it seems today very improbable to expect any radical change in the public perception of conventional nuclear power. In this context a lot of hopes and expectations have been expressed for novel systems called Accelerator-Driven Systems, Accelerator-Driven Transmutation of Waste or just Hybrid Reactors. All these names are used for description of the same nuclear system combining a powerful particle accelerator with a subcritical reactor. A careful analysis of possible environmental impact of ATW together with limitation of this technology is presented also in this paper.

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1. Introduction

Nuclear reactors based on self-sustained fission reactions, or so called "critical" reactors — after a spectacular development in fifties and sixties of 20^{th} 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 ADS is reduction of hazards related to handling and management of radioactive wastes through nuclear transmutation and, possibly, improvement of operational safety of nuclear power facilities. Transmutation (or rather nuclear transmutation) is defined as the transformation of one isotope into another isotope or element by changing its nuclear structure. Nuclear transmutation was first demonstrated by Rutherford in 1919 [1], who transmuted ^{14}N to ^{17}O using energetic α-particles. I. Curie and F. Joliot produced the first artificial radioactivity in 1933 [2] using α-particles from naturally radioactive isotopes to transmute boron and aluminum into radioactive nitrogen and oxygen. It was not possible to extend this type of transmutation to heavier elements as long as the only available charged particles were the α-particles from natural radioactivity, since the Coulomb barriers surrounding heavy nuclei are too great to permit the entry of such particles into atomic nuclei. The development of electrostatic (van de Graaff [3]) and linear accelerators (Wideröe [4]) and the invention of cyclotron by Lawrence [5] removed this barrier and opened new possibilities for transmutation experiments. The first accelerator-driven transmutation was demonstrated by Cockroft and Walton in 1930 [6] on their linear accelerator, on which they bombarded Li-target with energetic protons (energy varying from 125 to 500 keV), "transmuting" Li into 2 α-particles in a simple reaction

$$^7_3\mathrm{Li} +^1_1 p \rightarrow^4_2 \alpha +^4_2 \alpha\,.$$

The accelerator-driven transmutation was successfully developed by G. Seaborg and his collaborators in experiments aiming to create transuranic elements and their numerous isotopes. In 1941 Seaborg, McMillan, Segré, Kennedy and Wahl [7] transmuted ^{238}U (in the form of U_3O_8) into ^{239}Np and consequently to ^{239}Pu by the bombardment with 6 MeV deuterons in 60-inch cyclotron.

However, only when coupled with the spallation process, high power accelerators can be used for an effective transmutation. Spallation refers to nuclear reactions that occur when energetic particles (*e.g.* protons, deuterons, neutrons, pions, muons, *etc.*) interact with an atomic nucleus — the target nucleus. In this context, "energetic" means kinetic energies larger than about 100 MeV per nucleon. At these energies it is no longer correct to think of the nuclear reaction as proceeding through the formation of a compound nucleus. The initial collision between the incident projectile and the target nucleus leads to a series of direct reactions (intranuclear cascade) whereby individual nucleons or small groups of nucleons are ejected from the nucleus. At energies above a few GeV per nucleon, fragmentation of the nucleus can also occur. After the intranuclear cascade phase of the reaction, the nucleus is left in an excited state. It subsequently relaxes its ground state by "evaporating" nucleons, mostly neutrons. Shortly, spallation can be described as a nuclear reaction in which the energy of each incident particle is so high that more than two or three particles are ejected from the target nucleus and both its mass number and atomic number is changed.

The spallation process is depicted in Fig. 1, showing two stages of the process (intranuclear cascade and evaporation). For thick targets, high-energy (> 20 MeV) secondary particles (plus their progeny) can undergo further spallation reactions. For some target materials, low energy (< 20 MeV) spallation neutrons (*i.e.* the cascade-evaporation neutrons) can enhance neutron production through low energy (n, xn) reactions. For heavier nuclei, high-energy fission can compete with evaporation in a highly excited nucleus. This process is illustrated in Fig. 1. Tantalum, tungsten, and lead are examples of materials that can undergo spallation/high-energy fission.

Spallation produces large numbers of neutrons per an incident proton. 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. 2 [8]. The typical spectrum of neutrons emerged in spallation processes is presented on Fig. 3. 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 incident protons.

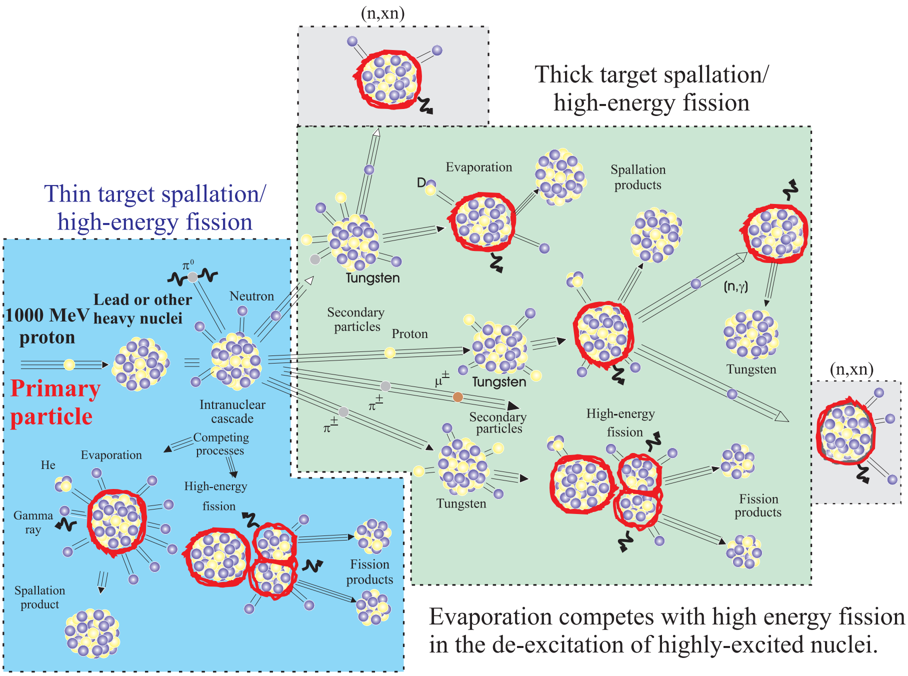

Fig. 1. A model of spallation processes in thin and thick targets.

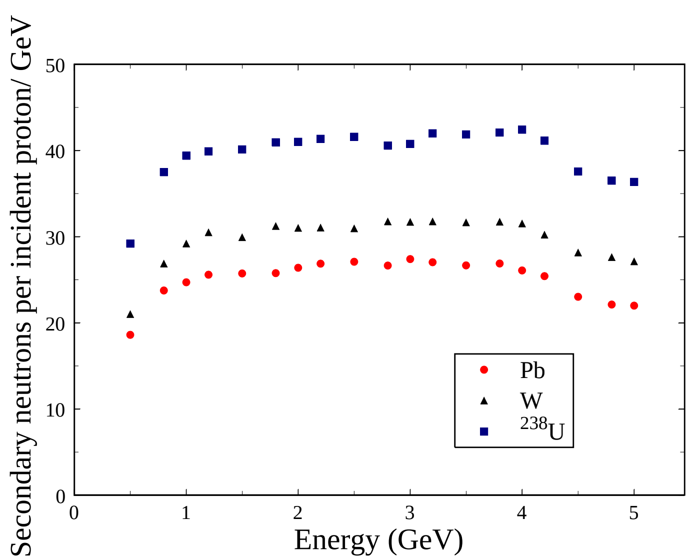

Fig. 2. Number of neutrons per incident proton and its energy (GeV) produced in a spallation processes in defferent thick targets.

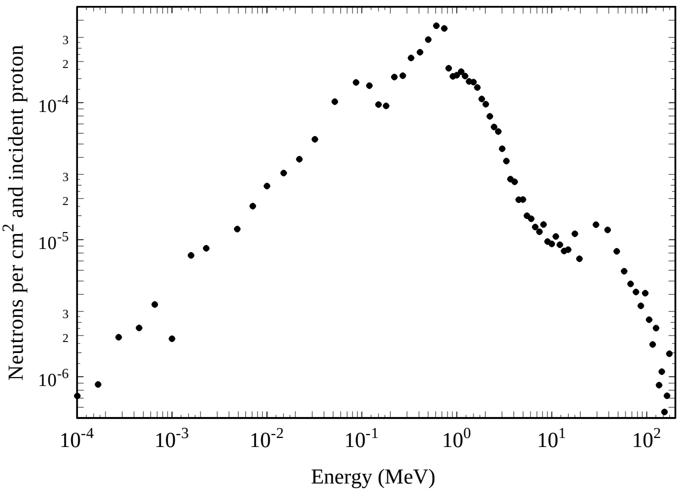

Fig. 3. Energy spectrum of spallation neutrons produced by a 1 GeV proton beam.

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. γ — 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, which make also these processes energetically very ineffective. This situation is exemplified on Fig. 4. So the most effective nuclear process that can be used for transmutation of radiotoxic isotopes is neutron absorption. Neutrons do are not repelled by nuclei and interaction cross 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. Radioactivity itself is not a good measure of hazards or noxiousness of radioactive wastes, therefore radiotoxicity, in most cases the ingestion radiotoxicity, is used as a measure of the biological consequences of radioactivity. However, in the case of geological (or underground) disposal of radioactive waste, which is a reference case, evaluation of ingestion radiotoxicity is a very complicated process of simulation of a long time transport of each isotope from a repository bed into the biosphere. Final results strongly depend on the assumptions, specific models and extrapolations used in the calculations. On the contrary, a nuclear part of these simulations, *i.e.* calculation of a source term of radiotoxicity can be

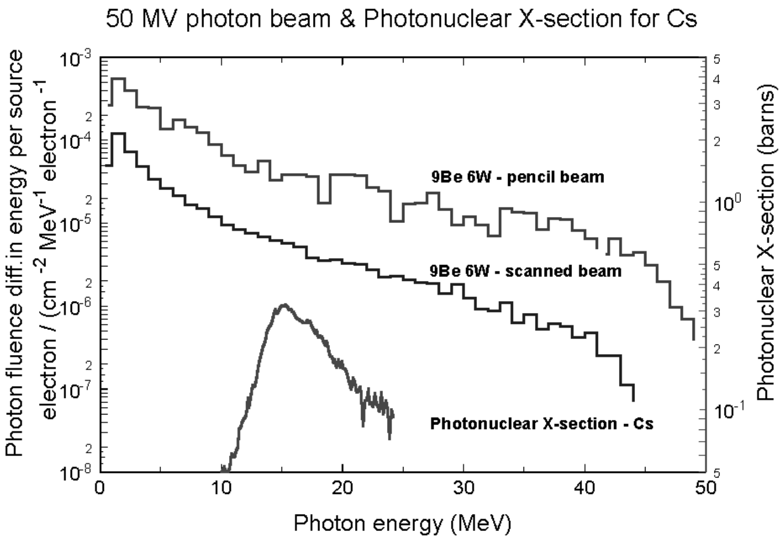

Fig. 4. Spectrum of 50 MV bremmstrahlung γ-source from W-target of an electron accelerator (left Y-axis) and a photonuclear reaction cross section for Cs (right Y-axis).

done very reliably with a high precision. 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 world-wide 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 can be effectively transmuted.

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 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. In the case of ^{237}Np and a thermal spectrum, the lowest neutron flux converting ^{237}Np from neutron consumer into neutron producer is expressed by

$$\Phi_{\mathrm{lim}} = \frac{\ln 2}{\sigma_f \, T^{\beta}_{1/2}} \, ,$$

where σ_f is a fission cross section for ^{238}Np, $\mathrm{T}^{\beta}_{1/2}$ is a half-life of beta-decay of ^{238}Np. For a thermal neutron spectrum, like in a molten-salt system, this limiting flux is about 2–3×10^{15} n/cm^{2}s.

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 plentifulness 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. This implies use of reactors with an excellent neutron economy, which limits the choice to fast reactors with the hardest possible spectrum, possibly 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 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 ADS. 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. 5.

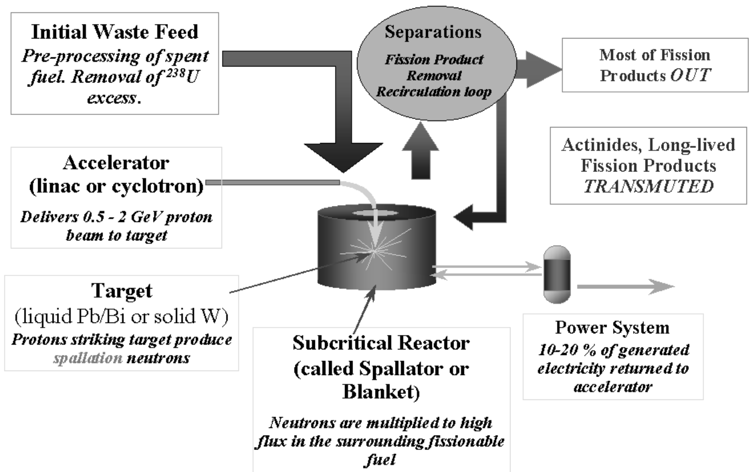

Fig. 5. A schematic view of an Accelerator-Driven Transmutation System.

When a particle beam (in most designs — protons) from accelerator hits a thick 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 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 proton 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, $k_{\rm eff}$ may have values much below 1.

The conversion of heat from the core into electricity in the conventional manner, via steam generators, turbines and generators produces electrical energy, which is more than sufficient to operate the accelerator.

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, as shown on spallation neutron spectrum in figure 3. By introducing a moderator, the neutron energy can be reduced (neutrons can be moderated) 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, that is for a given energy: In principle, considerably higher neutron fluxes can be achieved in this type of system than in a thermal self sustaining reactor.

Water and graphite normally require encapsulated solid fuel and are therefore less suitable as moderators in accelerator-driven systems 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, "thermal" molten salts, where actinides are dissolved in different types of fluoride salts have been considered to be a better combination of fuel and moderator. The homogenization of the fuel and subcriticality of the system mean that a substantial neutron flux is obtained close to the target, with a high transmutation rate, while most of the core has a considerably lower neutron flux. This can not be compensated for by increasing the supply from the accelerator-driven target, since material damage on the accelerator window, above all on the wall between the target and core, will be unacceptable at high proton and neutron fluxes [9]. More sophisticated solution can be applied, like multiple target system, in which subcritical core surround 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) [10]. 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 [10].

Using fast neutron spectrum it is easier to design a suitable neutron multiplying blanket/core for a subcritical system 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 heat generated by fission processes in ADS can of course be used to produce electrical energy. Some of this heat is used up to feed the accelerator.

In a fast accelerator-driven system, this share is typically on the order of 4–15%, and comparable to the energy which is used for secondary needs in a critical reactor.

4. Components of accelerator-driven systems

4.1. Accelerator

Linear or cyclotron accelerators have been proposed for ADS. In spite of a vivid discussion in the accelerator community [11] the final choice of an accelerator type will depend on an intercomparison and complex optimisation of the whole ADS including economical constraints and probably 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. 2 — 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 5–10 MW, corresponding to 5–10 mA of protons.

However, taking into account development and constructional costs, it may appear that even 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, are representing both accelerators types: linac at Los Alamos National Laboratory, running at 800 MeV and 1–1.5 mA proton beam, and cyclotron at Paul Scherer Institute, having a 1.5 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. The reliability and availability of the accelerator in accelerator driven systems is an important issue.

A linear accelerator is a preferred option for Los Alamos ATW group [12]. The accelerator design takes advantage of accelerator development work done in the frame of Accelerator-Production of Tritium (APT). Downsized APT accelerator for ADS would be a 45 MW (1 GeV energy and 45 mA current) superconducting linac driving several transmuters through a sophisticated system of beam splitters.

Most of the European groups are focused on advantages of cyclotron, proposing to overcome the technological limit of cyclotron current of about 10–15 mA, through use of multiple cyclotrons and more advanced designs like separated orbits cyclotrons [11]. Multiple cyclotrons, if cheap enough would be a good option to improve the reliability of ADS-facility.

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 highly desired, space charge challenges
- Cost reduction through compactness and robust constructions

4.2. Target

Two different technical solution can be envisaged for an ADS 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. The solid tungsten target design was developed in details in the APT project, however this solution is not very attractive for ADS. The preferable target for ADS would be a liquid metal target; for most cases liquid lead-bismuth eutectic (LBE) has been proposed. Other possible metals are liquid lead-magnesium eutectic and Mercury. Mercury target seems to be a preferred option for spallation neutron scattering facilities, like American Spallation Neutron Source (SNS) [13] project and European Spallation Source (ESS)[14].

The great 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). Moreover there are 70 reactor-year experiences from LBE-reactors developed in Russia. 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 groups. Several options of fuel have been proposed and the first choice for a planned demonstration facility in Europe, taking advantage of an existing reprocessing technology would be a solid oxide fuel *e.g.* (Pu+MA) oxide in titanium stabilised austenitic steel or 9% Cr martensitic steel cladding. A system with this fuel-coolant combination and 130 $\mathrm{MW_{th}}$ power at $k_{\mathrm{eff}} \sim 0.97$ would be capable to burn about 100kg Pu+MA, 30 kg of ^{99}Tc and 20 kg of ^{129}I per $\mathrm{TWh_{el}}$. In the longer time perspective Th-based fuel cycle is considered as an option of further reduction of ADS waste radiotoxicity. Thorium based 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 ADS, however the price would be a contamination of the whole primary liquid metal loop with the spallation products. So for the first demonstration solution it will probably be preferable to have a spallation target with a separated cooling circle, to contain the spallation products into the minimal volume.

As a backup ADS option it is now considered in Europe an advanced gas-cooled (He) ADS with LBE spallation target and MOX or more advanced 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 ADS, called Jülicher Transmuter, has been developed in IABAT-project [10].

In USA the ADS of a primary interest is based on LBE cooling and metallic, uranium free-fuel, array of metal fuel pins — 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 $k_{\rm eff}$ 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 3-4 during a single irradiation period, moreover such a core has to be reloaded about 3 times a year.

An alternative, thermal neutron accelerator-driven transmutation system — Tier — has been proposed in USA by Ch. Bowman [15]. Tier 1 ($k_{\rm eff} = 0.96$, neutron flux 2×10^{14} n/cm^{2}s) is a graphite assembly with circulating molten salt (NaF–ZrF_4) fuel and liquid lead spallation target. This ADS-system with 750 MWth power generated by fission of 300 kg/y of Pu and MA corresponding to annual PWR production of these elements would be a once-through transmuter with 80% efficiency. Tier 2 ($k_{\rm eff} = 0.95$, neutron flux 4×10^{14} n/cm^{2}s) system would be then a back-end option transmuting the spent fuel from 4 Tier 1 units. The choice of NaF-ZrF_4 salt in favour of LiF–BeF_2, which had been proposed in earlier molten salt system, is determined partially by economy the once-through performance of Tier system, in which no salt recovery is foreseen.

ADS system under consideration in Japan are to support 10 LWRs with MA transmutation [16]. Following options are investigated:

Solid fuel system:

- Nitride fuel core with enriched 15N

- Tungsten target and sodium cooling

  or

- Pb-Bi target-cooling system.

Molten-salt system:

- Chloride salt as target, fuel and coolant with on-line processing.

Taking advantage of subcriticality of ADS fuel for these systems can be purely MA-fuel without ^{238}U. Such type of fuel is unacceptable in critical reactor due to their small delayed neutron fraction-β and small Doppler effects so it is considered that ADS can play a significant role as "Transmuter" in the back-end of fuel cycle.

Table I summarizes some advantages and disadvantages of different coolants for ADS.

TABLE I

Summary of main features of ADS coolants

Coolant	Advantages	Disadvantages
Pb or Pb-Bi	Good neutronics Good thermal properties Easy integration with spallation target Good thermodynamical efficiency 70 reactor-year experiences from military submarines Not inflammable	Corrosion and erosion problems, material compatibility problems Opacity (problems with in-site inspection) Solid at room temperature Generation of radiotoxic ^{210}Po Not clear if "licenceable" for commercial systems
Gas (He)	Easy in-site inspection and access to core No-void effects Many reactor-years of experiences	High pressure operation Decay heat removal, requires advanced fuel Difficult integration with spallation target
Na	Well established technology Good compatibility with constructional materials Fair neutronics	Chemical reactivity with air and water Problems with physical separation between spallation medium and primary coolant Opacity (problems with in-site inspection) Voiding problems
Molten Salt	Good neutronics Deep burnup possible Possible very compact designs with small inventories Small waste streams in associated reprocessing chemistry	Advanced reprocessing chemistry Fuel and waste integrated with coolant Needs a lot of R&D Not clear if "licenceable" for commercial systems

4.4. Reprocessing chemistry

Accelerator driven transmutation implies, and depend very strongly on the chemical processes, which are commonly called reprocessing chemistry. This chemical processes are inevitably necessary to perform effective transmutation, and can be briefly divided into three different steps: chemistry which process LWR spent fuel and produce primary fuel for the first step of transmutation, *i.e.* first cycle in ADS; chemistry which makes possibly recirculation of the in ADS and finally, processes of conditioning the final outcome from ADS, *i.e.* final waste and tailings [17].

To produce the primary fuel for the first cycle of the waste in ADS, aqueous separations can be used, which is a well-developed technology in countries like France, UK and Belgium and can be relatively easily adopted for ADS. This step would primarily remove the excess of 238U and some fission products from LWR wastes, without a necessity of separation Pu or other transuranium elements. For the second stage of ADS fuel cycle — recirculation of the fuel in ADS, aqueous chemistry cannot be used, because one would like to recicurculate this fuel without too long cooling time. Chemical reprocessing technology not sensitive for high radioactivity must be applied. Pyrometallurgical separation provides these capabilities and is considered to be more proliferation resistant. Pyroprocesses withstand the high heat and radiation anticipated during the processing of fuel that has been irradiated in the ADS transmuter. All separations, either aqueous or pyro-based, should be modularized and constructed close to the transmuter; thereby limiting materials transport to either spent fuel from current nuclear power reactors or waste forms from ADS.

5. Conclusions

Accelerator-Driven Systems open new possibilities to perform transmutation of nuclear waste on a safe and effective way. Spent fuel from existing Light Water Reactors can be effectively transmuted in ADS, with radioactive waste streams with virtually no actinides (in many cases long lived α-emitters) and free of Tc and long-lived Iodine isotope, *i.e.* without the most cumbersome isotopes. Moreover ADS open new possibilities to design subcritical nuclear power reactors combining transmutation with commercial nuclear energy generation. To develop these transmutation systems an extensive research program of interdisciplinary dimension has to be started. This program covers nuclear physics, nuclear technology including high intensity, medium energy accelerators, reactor physics, material sciences, chemistry and nuclear chemistry, radioactive waste treatment technologies etc. In many countries the synergy between neutron science, accelerator technology, nuclear physics and transmutation research has been already recognized and common research and development programmes have been formulated and launched [17].

Nuclear and particle physics has an important role in development of ADS, particularly:

- Development of nuclear models, creating nuclear data bases, improving and designing new computer codes for particle interactions in the medium energy range (up to 300 MeV).

- Development and optimization of high current accelerators with exceptionally high reliability and low beam losses.

- Development of spallation neutron targets.
- Material irradiation studies and development of theoretical and computer models for irradiation induced material damages.
- New approaches to a nuclear fuel cycle.

This list of important topics is open and will cover more and more topics emerging from experimental work, which will hopefully start soon.

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