

# The Impact of Nuclear Science on Life Science

## Introduction to ADS For Waste Incineration and Energy Production

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### 1. Background

Accelerator based transmutation has as a concept been discussed since the 40-ties mostly for breeding fissile material from fertile thorium and uranium (ref. 1). The high power accelerator needed for the transmutation techniques was not within technical reach until fairly recently. The accelerator development over the last decades for high-energy physics has prompted proposals to develop accelerator driven systems for nuclear waste incineration and energy production.

One of the most pronounced problems with the use of nuclear power to-day is how to safely take care of the burned nuclear fuel commonly called the reactor waste which is both highly radiotoxic and has a very long radiological half-life. Different means to ensure that the waste will not interplay with biological life on earth have been proposed through the years. At present, the main option in most countries having nuclear power production plants in operation is deep geological disposal with or without reprocessing. An alternative option to first incinerate and transmute the high level radioactive waste to reduce the amount, the radio-toxicity and the half-life before disposing the remaining waste in a geological repository is now under consideration.

More than 99% of the amount of transuranium actinides can be separated from the spent fuels with realistic assumptions in relation to present and foreseen partitioning techniques.

A special problem is to take care of the strategic materials (highly enriched plutonium ( $^{239}\text{Pu}$ ) and uranium ( $^{235}\text{U}$ )) from dismantled nuclear weapons. Different means to minimise the risk for proliferation and at the same time make use of the energy content of the materials by incineration in reactors or accelerator-driven sub-critical systems have been proposed (ref. 2). The problem is most acute in Russia but also exists in the USA.

To overcome the safety and environmental issues with the present fission reactors, different concepts of accelerator-driven sub-critical systems have been proposed for safe and clean nuclear energy production using the thorium-uranium or uranium-plutonium fuel cycles. The environmental cleanness is given by the use of the thorium-uranium fuel cycle that produces fewer heavy transuranic elements than the uranium-plutonium cycle or by using the uranium-plutonium cycle with a simultaneous incineration of long-lived elements. Furthermore, the ADS techniques involve breeding of  $^{233}\text{U}$  or  $^{239}\text{Pu}$  from the most abundant isotopes in natural thorium ( $^{232}\text{Th}$ , 100%) and uranium ( $^{238}\text{U}$ , 99.3%), respectively. Thus, by using the total fission energy content of these abundant natural elements the resources are enough to meet the world energy need for thousands of years. Also the proliferation risk of strategic materials can be diminished with the use of the thorium-uranium cycle by adding a few percent of  $^{238}\text{U}$  to the initial feed, which will result in an infinite critical mass of the uranium.

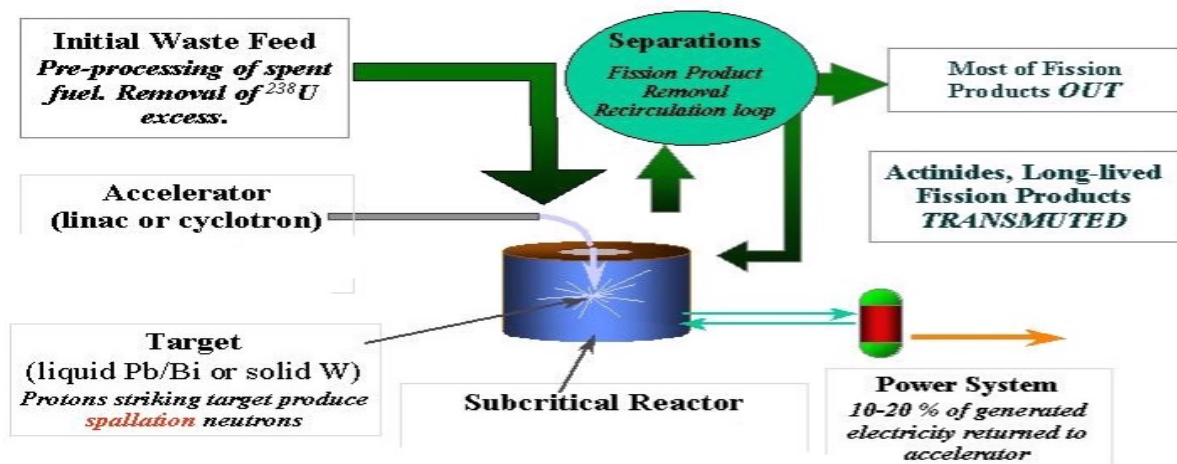
### 2. Main technology components of an ADS

An accelerator driven system consists of a high power proton accelerator, cyclotron or linac (ref. 3), delivering about 10 to 20 MW ( $E_p = 1$  GeV,  $I_p = 10$  -20 mA) beam-power to a heavy metal target, like Pb, W or U ADS systems. The beam power, of the order of 100 MW, has also been studied for special purposes as e.g. for accelerator-driven tritium production (APT) in the US. A proton of about 1 GeV energy generates about 30 neutrons by knocking out nucleons or cluster of nucleons in so called spallation reactions with the heavy target nuclei. The spallation neutron intensity will be  $10^{14}$ - $10^{15}$  n/s at the actual power level of the proton beam.

The spallation neutron energy spectrum is dominated by evaporation neutrons (about 90 %) with energies of a few MeV from the de-excitation of the reaction residues and has a tail of high energy neutrons up to the full energy of the proton beam from pre-compound reactions with the target nuclei.

The spallation target is surrounded by a sub-critical thermal or fast sub-critical assembly ( $k < 0.98$ ) (ref. 4) which contains the nuclear waste to be incinerated and/or nuclear fuel for energy production. The coolant is normally a liquid heavy metal or He-gas for a fast sub-critical reactor and a molten salt for a thermal reactor. The fuel is generally in solid form for the fast system and dissolved in the salt for the thermal.

Furthermore, processes are needed to separate the species to be transmuted in the spent reactor fuel as well as to clean the spent ADS fuel from fission products. The remaining transuranium elements (TRUs) and fission products from the separation processes are buried in a repository. The energy produced are fed to the electricity grid except 10-20 % which is used for running the ADS.



**Figure 1: Main components of an ADS transmutation facility**

### **3. Waste incineration**

#### **3.1 Introduction**

Different concepts to reduce the amount of transuranic actinides and long-lived fission products in the nuclear waste have been investigated. Transmutation by proton or heavier charged particle bombardment has been studied to eliminate certain radioactive fission products with very low neutron capture cross sections but is at present of minor practical interest. Also, the studies made on fusion-fission hybrid systems for nuclear waste incineration utilising the intense 14 MeV neutron field at the first wall of a magnetic confined fusion plasma Tokamak reactor are not further discussed in the present report as it is not likely to be realised within a foreseeable future.

In the following, the discussion is limited to incineration by neutron irradiation in a critical or sub-critical assembly. The basic principle of those concepts is to induce nuclear reactions that bring the actinides to fission and the fission products to transmute by neutron capture to more short-lived and stable species.

#### **3.2 The physics of nuclear incineration**

A number of physical constraints have to be taken into account when one is looking for an optimal transmutation system. The incineration efficiency is basically determined by the neutron economy and the neutron energy spectrum of the critical or sub-critical reactor.

The actinide nuclei with odd neutron numbers, like the uranium and plutonium isotopes  $^{233,235}\text{U}$  and  $^{239}\text{Pu}$  have large thermal fission cross sections in contrary to those with even neutron numbers which only fission by fast neutrons. Some of the latter transmute to thermally fissile nuclei by neutron capture, like  $^{232}\text{Th}$  and  $^{238}\text{U}$  which transmute to  $^{233}\text{U}$  and  $^{239}\text{Pu}$ , respectively, and are for that reason named fertile nuclei. Most of the fission products have large thermal capture cross sections and also large epithermal resonance capture cross sections in comparison with the cross sections for fast neutrons. This means that a thermal or a fast neutron transmutation system can only transmute a certain fraction of the nuclear waste in an optimal way.

A reactor having a fast neutron spectrum is needed to incinerate the non-thermally fissile transuranic isotopes. Among them are isotopes of the so called minor actinide elements (neptunium, americium and curium), which constitute a primary goal to incinerate. A fast critical reactor will have safety problems to burn fuel with a large content of minor actinides mainly for two reasons. The minor actinides have a low delayed neutron fraction resulting in a short power doubling time of the reactor and furthermore the Doppler feedback is low for those actinides. These safety problems of the fast critical reactor as a minor actinide burner are overcome by the sub-criticality of the reactor in an accelerator-driven system.

The accelerator driven sub-critical system with molten salt as coolant has a thermal or epithermal neutron spectrum. For this type of systems, the reactor waste is dissolved in salt consisting of light (beryllium and lithium) or medium (sodium and zirconium) weight fluorides. The thermal neutron energy spectrum of the molten salt concepts means that one obtains a very effective incineration of thermally fissile actinide isotopes like  $^{239}\text{Pu}$  and transmutation of fission products. On the other hand, the incineration will be less efficient of e.g. the minor actinides and small amounts of very heavy actinide isotopes will be build-up by

neutron capture in non-thermally fissile actinide isotopes. To counteract the reactivity loss by production of reactor poisons, an intense spallation source ( $10^{15-16}$  n/s) is required and/or an on-line separation system or by adding more reactivity (plutonium or highly enriched uranium) in the beginning of cycle. Experience of the molten salt techniques exists from the running of an experimental critical facility during about 4 years time in the 60-ties at the Oak Ridge National Laboratory (ref. 5).

An excess of neutrons has to be available for transmutation purposes in each step of the chain reaction for a critical reactor and in each neutron generation step for ADS. The ADS concepts have in general a larger excess of neutrons per fission for transmutation than the critical reactors, in particular, than the thermal critical reactor. The neutron excess per fission (G) can be expressed as (ref 6):

$$G = - \sum e_j D_j - (CM + L) \text{ for a critical thermal or fast reactor}$$

and

$$G = S_{\text{ext}} - \sum e_j D_j - (CM + L) \text{ for a subcritical accelerator driven system with an external source}$$

The (CM + L) is the number of neutrons per fission lost in parasitic captures, absorption in construction materials and leakage.  $D_j$  is the number of neutrons per fission needed to transmute isotope J down to fission products (can be positive or negative) and  $e_j$  is the fraction of isotope J in the fuel of the system.

The (CM + L) term is for all realistic power reactors about 0.3 neutrons per fission. A positive G means that transmutation can be achieved, while if negative, there is a lack of neutrons.

Physics analysis show that the transmutation of the minor actinides and long lived fission products can be made in standard critical reactors. However, for light water reactors one has to increase the enrichment to high values (about 10 %) to transmute the minor actinides while if one uses fast critical reactors one has to conceive a nuclear power park with a very large fraction of this type of reactors.

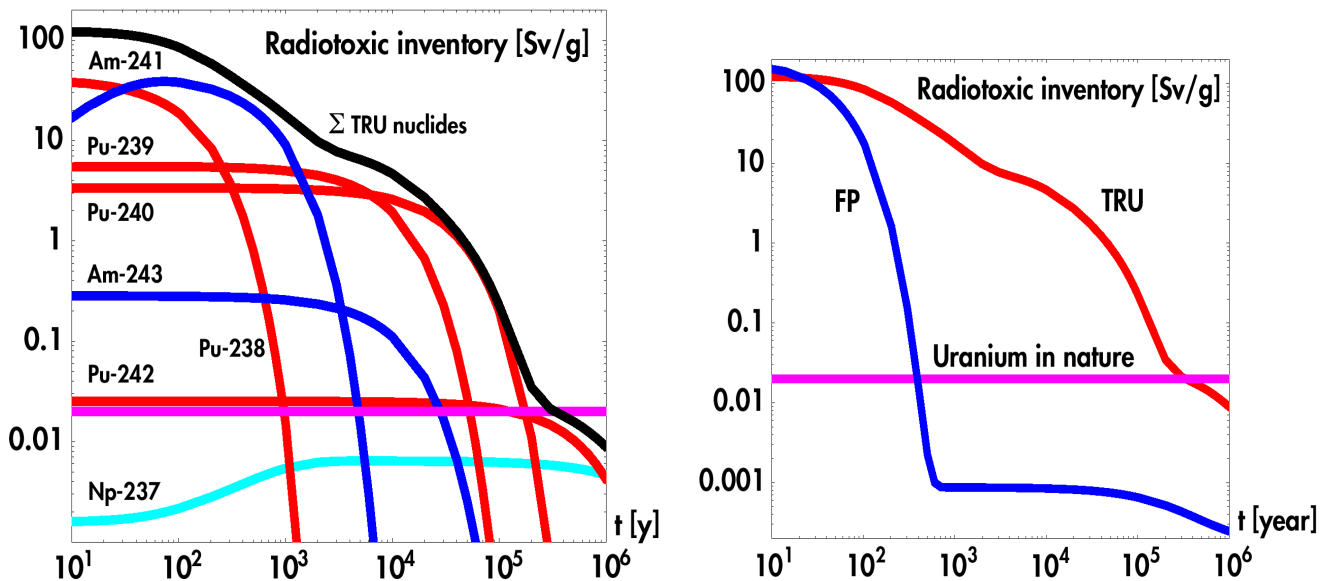
### **3.3 Light water reactor waste inventory**

The burned reactor fuel contains of two mayor element fractions, the actinides, mainly uranium but also in smaller amounts heavier elements, the transuranium elements (TRUs) like plutonium and the minor actinides (MA), neptunium, americium and curium and fission products which are medium weight elements from fission processes in the uranium fuel (Table 1). The TRUs are being formed by successive neutron captures starting with neutron capture in  $^{238}\text{U}$  and  $^{235}\text{U}$ . The actinide elements are highly radiotoxic and quite a few of their isotopes have half-lives longer than 10,000 years or more. Most of the fission products have very short half-lives but a few of them have long half-lives up to millions of years. Some of the long-lived products have also high solubility in water and there is a risk for migration of these elements if groundwater is entering the repository.

**Table 1, Annual production of plutonium, minor actinides and fission products from a 3000 MWth pressurized light water reactor with fuel burned to 33,000 MWD/ton (After 10 years decay)**

Isotope	Half-life (yrs)	Mass kg/yr
<u>Plutonium and Minor Actinides (MA):</u>		
<sup>237</sup> Np	2,100,000	14.5
<sup>238</sup> Pu	80	4.5
<sup>239</sup> Pu	24,000	166.0
<sup>240</sup> Pu	6,600	76.7
<sup>241</sup> Pu	14	25.4
<sup>242</sup> Pu	380,000	15.5
<sup>241</sup> Am	430	16.6
<sup>243</sup> Am	7,400	3.0
<sup>244</sup> Cm	18	0.6
<u>Long Lived Fission Products (LLFP):</u>		
<sup>79</sup> Se	65,000	0.2
<sup>90</sup> Sr	29	13.4
<sup>93</sup> Zr	1,500,000	23.2
<sup>99</sup> Tc	210,000	24.7
<sup>107</sup> Pd	6,500,000	7.3
<sup>126</sup> Sn	100,000	1.0
<sup>129</sup> I	17,000,000	5.8
<sup>135</sup> Cs	3,000,000	9.4
<sup>137</sup> Cs	30	31.8
<sup>153</sup> Sm	90	0.4

**Fig. 2 Time dependence of the specific radiotoxic inventory for fission products and transuranium elements.**



The time dependence of the specific radiotoxic inventory of spent light water reactor fuel is shown in fig. 2. While the fission products decay to the equilibrium radiotoxic inventory level of the natural corresponding uranium needed in less than 1000 years; it takes about 200,000 years for the transuranium elements to decay to the same radiotoxic level.

### 3.4 Partitioning

Two different types of processes can be applied to the separation of long-lived radionuclides: hydrochemical (“wet”) and pyrochemical (“dry”) processes (ref. 7).

The PUREX process is the most important hydrochemical reprocessing technique to separate uranium and plutonium from spent fuel and is based on the dissolution of the fuel in nitric acid. For the extraction of minor actinides, the process should be modified/extended for which extensive research is being currently performed.

An alternative to hydrochemical processes are pyrochemical processes in which refining is carried out in molten salt. In nuclear technology, they are often based on electrorefining or on distribution between non-miscible molten salt-metal phases. The mayor advantages of pyrochemical techniques to reprocess advanced fuels, in comparison to hydrochemical techniques, is a higher compactness of equipment and the possibility to form an integrated system between irradiation and reprocessing facility, thus reducing considerably transport of nuclear materials. In particular, for advanced oxide fuel (mixed transuranium, inert matrix or composite) and metal fuels, but also nitride or thorium based fuels, pyrochemistry is to be preferred. In addition, the radiation stability of the salt in the pyrochemical process compared to the organic solvent in the hydrochemical process offers an important advantage when dealing with highly active spent minor actinide fuel. Shorter cooling times reduce storage costs.

The separation techniques for the minor actinides and the long lived fission products are on a laboratory research level for both the wet and dry techniques. The goal is to obtain a separation efficiency of > 99,9% for the actinides and > 95 % for the fission products to minimize the amount of waste from the reprocessing (ref. 22).

### 3.5 Incineration strategies and concepts

Two different incineration routes can be followed the “double” or the “single strata”.

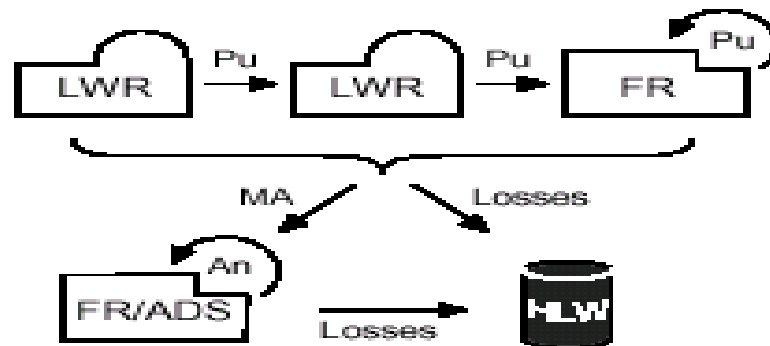
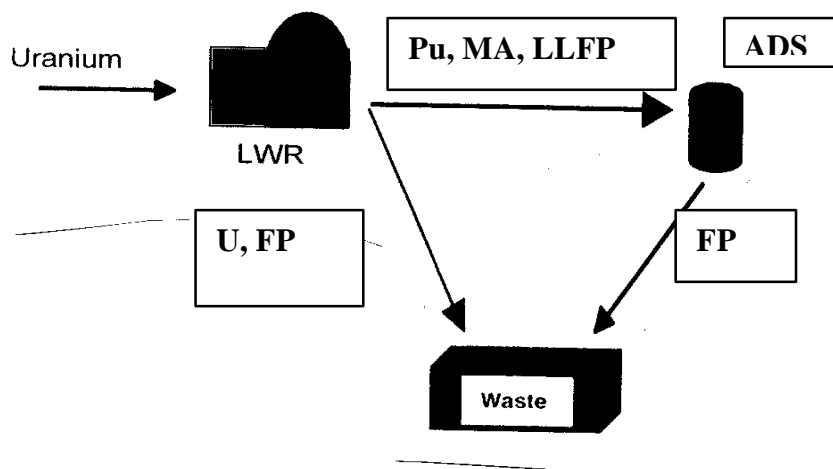


Figure 3A, Outline of the “double strata” option, (ref. 21)

In the so called “double strata” scenario the plutonium is separated from minor actinides and long lived fission products.. Uranium and plutonium are first extracted from the spent fuel and the remaining minor actinides and long lived fission products sent to an independent partitioning and transmutation facility (Fig. 3A). The plutonium is mixed with uranium, both as oxides, to form the so called MOX fuel which is burned in thermal and/or fast reactors. Several different sub-critical fast reactors for the “double strata” ADS are being studied with either liquid lead or eutectic liquid lead-bismuth, liquid sodium or helium gas as coolant and solid metallic, oxide, nitride or carbide fuel. Furthermore, most of the sub-critical fast reactors are designed to contain a special thermal/epithermal zone for the transmutation of the long lived fission products  $^{99}\text{Tc}$  and  $^{129}\text{I}$ .

All these concepts rely to some extent on experiences with power or prototype fast reactors like the Russian submarine reactors for eutectic liquid lead-bismuth, Phenix/Superphenix and JOYO/MONJU programmes in France and Japan for liquid sodium. There is also the project of the French (Framatome) and US (General Atomic) HTGR project for a critical gas-cooled reactor.

In particular, gas-cooled reactors with fuel pellets have also been investigated by General Atomics (ref. 8) and a Spanish-Israeli team (ref. 9) where the pellets are possible to be continuously loaded and unloaded.



**Figure 3B, Outline of the “single strata” option**

The other route is the “single strata” where the plutonium, minor actinides and long lived fission products are transmuted together (Fig. 3B). A number of studies focused on the “single strata” option are in progress. They include studies on ADS with the same types of fast sub-critical reactors as for the “double strata” concept but also thermal or epithermal accelerator-driven sub-critical systems with molten salt as fuel and coolant.

#### 4. ADS energy production concepts

Different accelerator-driven systems have also been investigated for energy production, in particular “The energy amplifier (EA)” proposed by C. Rubbia, CERN (ref. 10), “The accelerator-driven energy producer (AEP)” proposed by C. Bowman, LANL (ref. 11) and the TASSE concept proposed by I. Slessarev et al (ref. 12). All three concepts use the thorium/uranium fuel cycle. The EA was described in the 1994 NuPECC report (ref. 13) and has a liquid-lead cooled sub-critical fast reactor while the AEP and TASSE have molten salt sub-critical thermal reactors.

The accelerator-driven incineration systems are all producing energy when burning the nuclear waste. The energy content of the transuranic waste is about 25% of the energy produced by the reactor forming the waste. A simultaneous energy production and incineration system is also feasible using the thorium/uranium fuel cycle with plutonium as a primer and, at the same time, incinerating the minor actinides and the long lived fission products. Both sub-critical fast systems with liquid lead-bismuth or He-gas coolant as well as thermal systems with molten salt are being studied for multipurpose use.

## **5. Research on ADS**

The research on ADS for waste incineration/transmutation and energy production has considerably expanded during the last few years. The focus of the present ADS research world wide is on the incineration and transmutation of nuclear waste while the production of nuclear energy by ADS is on a longer time scale.

The funds available for partitioning and transmutation research have increased by about a factor of ten going from the 4th to the 5th Framework Programme of the European Commission. The support now covers 14 research projects involving studies on partitioning, fuel, spallation, sub-critical reactors, nuclear data etc.

The research ministries of France, Italy and Spain have agreed on collaboration on research of ADS. Several other European countries have joined this effort on a working-group level. This Technical Working Group (TWG), chaired by C. Rubbia, has presented in April 2001 a “road-map” report on the research and development leading to an European ADS demonstration facility (XADS) in year 2015 and a prototype (XADT) around 2030 with close synchronisation with the 6<sup>th</sup> and 7<sup>th</sup> Framework Programmes of the EU (ref. 14). Two different concepts have been proposed for XADS namely one liquid metal cooled ADS by ANSALDO and one gas cooled by FRAMATOME, both with MOX fuel.

A new project (AAA or 3A) has been initiated in the USA to merge and harmonise the researches related to high power accelerators. AAA stands for “Advanced Accelerator Applications” and includes the former “Accelerator Production of Tritium (APT)” and “Accelerator Transmutation of Waste (ATW)” projects. It has three components:

- Transformation of the APT project into an Accelerator Demonstration Facility project.
- Building of an Accelerator Demonstration Facility based on APT design
- Testing and demonstrating the technologies relevant for transmutation following a road-map study initiated by the US Department of Energy (ref. 15)

Several studies are also being carried out in Russia under the auspices of MINATOM. Among them is one project of sub-critical accelerator-driven systems for energy production using the thorium/uranium cycle with weapons grade plutonium as a primer which eliminates the proliferation risk and at the same time makes use of the energy content of the plutonium (ref. 16)

The OMEGA project in Japan (ref. 17) with the aim to study different means to optimise the use of the reactor waste has run since 1988. The aim of the first phase of the project, which was to evaluate different concepts and make R&D on key technologies, has been achieved. The second phase is under way, which includes engineering experiments on key technologies and demonstrations. Some key questions for the research are “Reductions of Minor Actinides and Long Lived Fission Products and the time requested”, “Generation of Secondary Waste”, “Increase in Radiation Dose”, and “Economy”.

The Korea Atomic Energy Research Institute is developing a Hybrid Power Extraction Reactor (HYPER) for the transmutation of nuclear waste and energy production. (ref. 18) The system is being designed to utilise fast neutrons (Pb/Bi eutectics as target and coolant) for the transmutation of TRUs and has 4 thermal target regions for the transmutation of fission products.

The problem of finding an acceptable place for a geological repository in the Czech Republic has speeded the research on accelerator-driven transmutation. A national research program (LA-0) has been initiated which focus on the problems related to the physics, chemistry and material questions in a sub-critical reactor with molten salt (ref. 19).

While design studies are being made of the accelerator part of the system (ref. 3), more basic studies are in progress on the spallation target and on the sub-critical reactor and the coupling between these two components of the system (ref. 4). At the same time, partitioning methods are developed for certain long-lived elements in the waste as requested for an efficient transmutation.

Furthermore, fundamental research related to ADS applications is in progress in many fields like nuclear data (both experiments and reaction model code developments) (ref. 20), material research (in particular corrosion and radiation resistance), fuel development, liquid metal and molten salt thermal hydraulics etc. Coordination and exchange informations of mostly basic research projects are also arranged on an international level by the International Atomic Energy Agency (IAEA) and the OECD/Nuclear Energy Agency.

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