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Accelerator transmutation of wastes (ATW) – Prospects and safety

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ABSTRACT (ENGLISH)

Accelerator transmutation of nuclear waste - **ATW** - has during last years gained interest as a technologically possible method to transform radioactive wastes into short-lived or stable isotopes. Different ATW-projects are described from the physical and technical point of view. The principal sketch of the safety analysis of the ATW-idea is given. Due to the very limited technical data for existing ATW-projects the safety analysis can be performed only qualitatively. Difficulties related to chemistry and material problems can cause some risks for the health and environmental safety for the closest environment. General public should not be effected.

ABSTRACT (SWEDISH)

Transmutation av kärnavfall, ATW, har under senare år aktualiserats som en tekniskt möjlig metod att omvandla kärnavfall till kortlivade eller stabila produkter. Olika aktuella varianter av ATW beskrivs från fysikalisk och teknisk synpunkt. En principiell säkerhetsanalys av ATW-konceptet genomförs. Här är det i nuvarande läge i stort sett endast kvalitativa aspekter som kan belysas; data föreligger i dag mycket begränsat. Ur risksynpunkt synes kemi- och materialfrågor, i första hand kan riskerna ge arbetsmiljöproblem. Allmänheten torde inte att berörs.

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LIST OF ABBREVIATIONS

ABC	Accelerator Based Conversion
ALMR	Advanced Liquid Metal Reactor
AOO	Anticipated Operational Occurrences
ATW	Accelerator Transmutation of (Nuclear) Wastes
BTA	Basic Technology Accelerator
CANDU	CANadian Deuterium-Uranium - canadian heavy water reactor design
CRIEPI	Central Research Institute of Electric Power Industry
CTR	here: lowest contemplate wall loadings (power per surface unit)
DBA	Design-Basis Accidents
dpa	displacement-per-atom
EBR	Experimental Breeder Reactor
ETA	Engineering Test Accelerator
FP	Fission Products
GeV	Giga-electron-Volt - energy unit
HEX	Heat Exchanger
HFIR	High Flux Irradiation Reactor
HT9	martensitic steel
IFR	Integral Fast Reactor
ING	Intense Neutron Generator
JAERI	Japan Atomic Energy Research Institute
k_{eff}	the effective multiplication factor
keV	kilo-electron-Volt - energy unit
LAMPF	Los Alamos Meson Production Facility
LOCA	Loss-Of-Coolant Accident
mA	milliampere
MA	Minor Actinides
MeV	Mega-electron-Volt - energy unit
MTA	Material Testing Accelerator
MW_T	MegaWatts-thermal
Linac	Linear accelerator
LWR	Light Water Reactor
n	neutron(s)
OMEGA	Options Making Extra Gains from Actinides and fission products - japanese R&D programme

ORNL	Oak Ridge National Laboratory
PNC	Power Reactor and Nuclear Fuel Development Corporation
P-T	Partitioning-Transmutation
R&D	Research and Development
RR	Residual Risks
s	second(s)
SA	Severe Accidents
STATS	Separation Technology and Transmutation Systems

SUMMARY

Accelerator transmutation of nuclear waste - **ATW** - has during last years gained interest as a technologically possible method. ATW - a high technology nuclear synergetic system - can convert the heat generated by transmutation into electricity. It could produce enough electricity to keep the accelerator running and still have as much surplus power left over as a midsize commercial reactor. However, ATW minimizes the criticality concern, which is quite serious for reactors, since ATW never achieves a self-sustaining chain-reaction.

Different ATW-concepts are under serious considerations in USA, Japan and in France. This report has the purpose to give some aspects on the prospects and safety of ATW. Since there is no direct empirical experience of ATW, mostly a qualitative evaluation of the ATW safety can be given here.

We concentrated our efforts mainly on the ATW-designs developed at the Los Alamos National Laboratory. Our choice was determined by the fact that Los Alamos projects are today the most complete and developed ones.

Conclusions of this report can be summarized as:

- ATW can enhance geological repository program
- ATW would not remove need for the first geological depository but it has the potential for significantly impacting future high level waste management strategy
- ATW reduces end-of-life wastes without leaving the burden to the future generations. The extremely long timescales of the toxicity of non-processed wastes is reduced to the "human dimension" of several centuries where engineered barrier systems could be used
- ATW can ease requirements of future geologic repositories by practically eliminating key radionuclides that are major contributors to long term risk.
- In is advanced design ATW offers a very an attractive undercritical nuclear energy system with very low system inventories of radioactive isotopes
- ATW as a thorium burner offers inexhaustible energy supply

ATW-concepts are still under development so this report could review only some important aspects of the safety and prospects of the current projects. ATW-ideas are in continuing process of evaluation .

1. INTRODUCTION

Accelerator transmutation of nuclear waste - **ATW** - has during last years gained interest as a technologically possible method. The main reason for this is the recent development of the accelerator technique. It is now possible to generate a beam of high energy particles (protons or deuterons) with high intensity currents of the order of hundreds milliamps. This beam hitting a suitable target creates a very intense neutron spallation source [1] . These neutrons can then be used to transmute some long-lived nuclear waste into stable or shortlived products. The waste might come from nuclear weapons or from nuclear reactors. This high technology nuclear synergetic system can also generate net energy [2] . Different ATW-concepts are under serious considerations in USA, Japan and in France.

No ATW-plant exists today but a first test one, might be constructed around the year 2000. It is then of interest to describe and analyze the technical feasibility of the different ATW-concepts. As it is the case with nuclear reactors and with different methods of spent fuel management, the safety of ATW should also be carefully evaluated.

This report then has the purpose to give some aspects on the prospects and safety of ATW. Since there is no direct empirical experience of ATW, mostly a qualitative evaluation of the ATW safety can be given here.

We shall first define what we shall mean with **safety**. **Safety** is the contrary to risk. Risk contains two components:

random or uncertain negative consequences for human life, health and environment,

and a measure of the likelihood of these consequences.

Unfortunately, there is no standard definition of the risk. In some context risk is a synonym for the *probability* of an undesired event, in another risk means the *consequence* of this undesired event. In nuclear safety, people often use "risk" as a synonym for the mathematical expectation of consequence, that is, the product of the *probability* and the *consequence* [3]. In reality, the perceived risk is more closely related to the consequence than to the probability, probably because people can visualize the consequence but not the probability. Our interest is risk in connection

with decisions. Considering nuclear waste the question is to provide the decision making body with sufficient information about the risk for deciding if the risk is acceptable or not and to find weak links where changes can substantially improve the safety. It will also be of interest to compare different waste handling methods with respect to risk.

There is one dominating difficulty when trying to estimate risk for the ATW-concept compared with the situation e.g. for nuclear reactors. ATW does not exist but as a technical well founded plan for a plant. That means that only a qualitative sketch of a risk analysis can be given. We shall anyhow try to give safety aspects to the different parts of ATW in the following sections. One important argument in favour of ATW compared with rock disposal is the relatively short dangerous time period for the resulting transmuted waste. (Here should be noted that even the ATW-output will contain some quantities of long-lived radioactive products.) To compare risks for the biosphere during 100 years with risks during 100 000 years gives rise to difficult methodological problems. The quantification of the risks for e.g. intrusion at a very distant future requires extrapolations which are extremely difficult. However, the risks for both systems has to be identified and estimated. The radiological exposition risks for the personnel at the sites and for nearby population should be carefully compared for those different systems.

There are two predominant principles concerning the handling of nuclear waste:

(i) The probability for negative consequences for the biosphere should be negligible.

(ii) The handling of the waste should be the responsibility of the now living generations.

It is of course also essential to consider the eventuality of proliferation of nuclear weapons.

This implies three waste handling strategies which also can be combined:

(i) Keep the waste from any contact with the biosphere.

(ii) Disperse the waste so that the risk will be of the same magnitude as the similar natural risks.

(iii) Transform the waste into a less dangerous form, i.e. transmutation of nuclear waste.

Up to now the concepts considered in most countries are examples of the first of these strategies - e.g. in Sweden the waste will without reprocessing be placed deep in the rock.

In order to evaluate in a fair way the risks for the third strategy - ATW, we must clearly define the system to be studied. We should consider all risks for human life and

health as well as for the environment from the construction of the ATW plant during its "production" phase up to the decommissioning of the plant and disposal of the residual radioactive material.

Also the nuclear weapon proliferation aspects of an ATW-plant should be considered.

In the following sections different ATW- concepts will be described, their prospects analyzed and corresponding risks evaluated if possible. First the ATW-concept is described in its historical context and then we concentrated our efforts mainly on the ATW-designs developed at the Los Alamos National Laboratory. Our choice was determined by the fact that Los Alamos projects are today the most complete and developed ones.

2. INTRODUCTION TO ATW CONCEPT

The basic mechanism underlying transmutation involves exposing radioactive elements to a stream of neutrons (or gamma quanta) and has been studied by scientists since the discovery of neutrons in 1932. Fast-moving as well as slow-moving neutrons can cause elements such as plutonium, which has a half-life time of about 24 000 years, to fission and releasing at this process a large amount of useful energy. Slower-moving neutrons can also be absorbed by the waste elements, transforming them into heavier isotopes which could have a shorter radioactive half-life or are stable.

Either a particle (eg. proton or deuteron) accelerator or a fission reactor can supply the neutrons for transmutation. The most advanced research on reactor-based transmutation involves the Integral Fast Reactor (IFR), which is now being developed at Argonne National Laboratory. The IFR is a modified breeder reactor with passive safety features which is designed to irradiate its own waste so that it can be recycled, thus eliminating the reactor's need for new fuel and reducing its volume of high-level waste. With minor modifications, an IFR-type reactor could also transmute, or "burn" high-level waste from other reactors [4].

The concept of using an accelerator to produce neutrons is not new; it was proposed at an early date of the nuclear age - 1950 - by E.O. Lawrence et al. at Berkeley [5]. To be independent of foreign sources of uranium used to make plutonium for weapons, a project was planned to produce plutonium from the low ^{235}U concentration tailings accumulating at the diffusion plant at Oak Ridge National

Laboratory. Thus, the so-called MTA Project (Material Testing Accelerator) [6] was initiated. The MTA project proceeded quite far into the problems of engineering, developing, and building the prototype of an accelerator-breeder facility; the project was terminated when substantial amounts of uranium ores were discovered in the western part of the United States.

Since 1952, W.B. Lewis working in Canada, has promoted the use of accelerator for producing ^{233}U fuel for the closed fuel cycle of the CANDU reactor. Since the CANDU reactor, burning ^{233}U, has near-breeding characteristics (conversion factor > 0.9), a single 0.3 - 1 GeV accelerator breeder can supply fuel for about 12 reactors. In the 1960s, the Canadian concept evolved into the idea of an Intense Neutron Generator (or ING), which would have supplied an intense neutron source for research. However, the expense of this project would have consumed a large part of the budget allocated to scientific research in Canada, and such a redefinition of priority was not accepted. Since then the use of neutrons for transmutation of the long-lived radioactive nuclei of nuclear waste into stable or short-lived species has been under many considerations. Thermal neutrons have not been considered useful for this purpose for several reasons. First, the typical nuclear waste has spent of the order of two-three years in a reactor thermal flux of approximately 10^{14} n/cm^2·s. Most of those waste nuclei with high cross sections for neutron capture have already been converted into nuclei with smaller cross sections by irradiation in this flux. Therefore most of the nuclei in fission product waste have low absorption cross sections which means they cannot be transmuted effectively in a thermal neutron flux of typical reactor intensity. Secondly, the higher actinide waste, which consists of neptunium and higher atomic number elements except plutonium, is thought not to transmute well in a thermal flux. Transmutation in this case implies destruction by fission since there are no stable higher actinide nuclei. Thermal fission cross sections for the primary constituents of the actinide waste ^{237}Np and ^{241}Am, are very small compared to their thermal capture cross sections so that thermal neutrons are thought to convert the material to a heavier mass isotope, easier to fission, rather than to directly induce fission.

Although neutron reaction cross sections in the higher energy range (keV-MeV range) are much smaller in general compared to those for thermal neutrons, the fission-to-capture ratio for the waste actinides is considered to be much more favorable this energy range. These fast neutrons have been therefore favored and fluxes at these energies at the 10^{15}n/cm^2·s level are practical. A prominent feature of the systems using fast neutrons is the large waste inventory (typically > 10 tonnes) which must be present in the system because of the small cross sections in the higher energy range. Moreover frequent fuel reprocessing and refabrication is required together with very

long time requirements for waste destruction. Such an accelerator-based system was proposed by the workers at Brookhaven National Laboratory [7] and is now carried out in Japan as a part of an extensive project called OMEGA. ("OMEGA" stands for Options Making Extra Gains from Actinides and fission products) [8]. However the extensive studies of both reactor- and accelerator based systems with fast neutrons have not been sufficiently encouraging to provide a viable alternative to storage of high level nuclear waste on a geologic time scale [9].

Recent advances in linear accelerator technology including new ion sources for producing the beam, the radio-frequency quadrupole for launching the beam into the linac accelerator structure, efficient klystrons to power the new accelerator structures, and improved beam focusing technique make possible constructions of high-current radio-frequency proton accelerators in the 0.8 - 1.6 GeV energy range, which could produce a thermal neutron flux at the 10^{16} n/cm^2·s range [2].

When 1.6 GeV protons strike a large radius target consisting of heavy nuclei such as lead, approximately 40-50 neutrons are generated per proton. Spallation is also possible on the very light nuclei, like lithium or lithium salts. Striking protons produce a cascade of high energy particles in lithium target, which surrounded with uranium multiplier can give almost as much neutrons per proton as a heavy nuclei target. The energy deposition for this process is about 30 MeV of proton energy per neutron compared with about 200 MeV of fission energy deposited per useful neutron from a self-sustained chain reaction in fissile material such as ^{235}U. The heat per unit volume which must be handled for a given neutron production rate is therefore considerably smaller from the spallation source than for a reactor.

The most promising and advanced ATW-concept based on the idea of using thermal neutrons has been developed at Los Alamos National Laboratory [10],[11]. This innovative technical approach is to use a high current proton/deuteron accelerator to create an intense source of **thermal** neutrons by moderating fast spallation neutrons. This technical approach is in fact substantially different - in spite of many similarities - than other transmutation concepts that use a **fast** neutron spectrum (such neutrons could be generated as well by a fission reactor as by accelerator) and which tend to focus primarily on destruction (incineration) of actinides. An important breakthrough at Los Alamos was the realization that intense fluxes of thermal energy neutrons could be used to great advantage in destroying actinides other than plutonium, which is readily burned by either fast or thermal neutrons. The first step in burning the minor actinide waste nuclide ^{237}Np is to capture a neutron leading to the key nucleus ^{238}Np. ^{238}Np is an unstable isotope with a decay half-life of 2.1 days and a very large thermal neutron fission cross section of 923 barns. ^{238}Np may either absorb a second neutron

leading to fission or it can decay to ^{238}Pu - Figure 1. In a low flux , decay is more likely, and successive neutron capture leads eventually to fissionable ^{239}Pu with the emission of 2.8 neutrons. However, three neutrons are absorbed and about 1/3 of the of the absorptions in ^{239}Pu do not lead to fission, so on average about 4 neutrons are required to fission each original ^{237}Np nucleus.

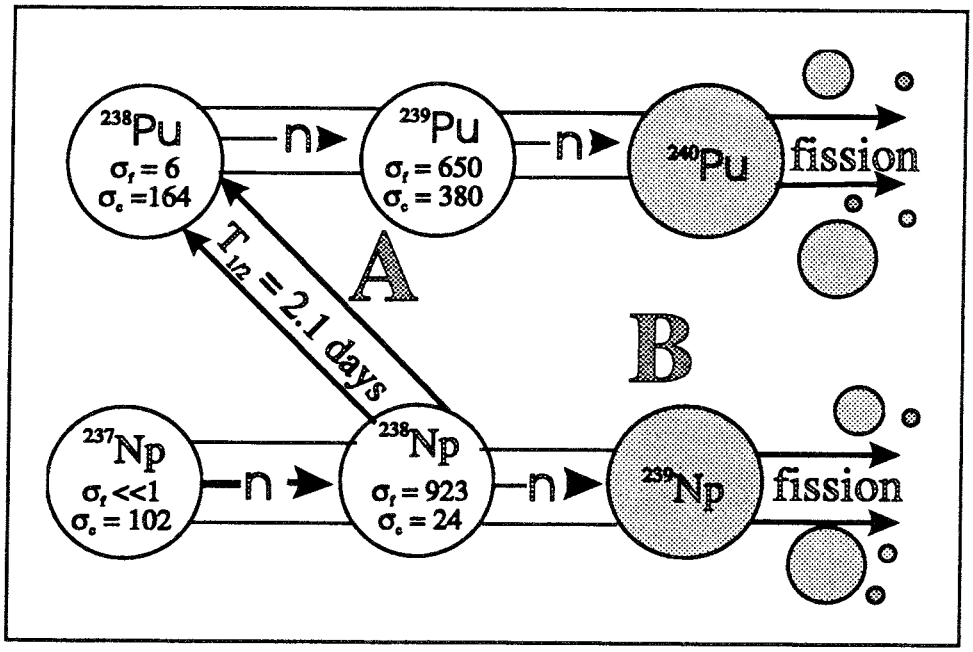

Figure 1 Two-step fission of ^{237}Np. At low fluxes ($< 2 \cdot 10^{15}$n $cm^{-2}s^{-1}$) ^{237}Np acts as poison, absorbing more neutrons than it produces (route A). At higher fluxes ^{237}Np becomes a fuel, generating more neutrons than it absorbs (route B).

In the large thermal flux ($> 2 \cdot 10^{15}$ n cm^{-2} s^{-1}) ^{238}Np nucleus can undergo fission most of the time, before it has a chance to decay. In that case more neutrons are liberated than are absorbed. Therefore, in a high thermal flux the ^{237}Np acts as a fuel, whereas in a low flux it is a poison. The other minor actinide nuclide ^{241}Am behaves similarly. Curium and plutonium isotopes present no special problems to maintaining the neutron economy of the system [12]. This two-step capture-fission process thus produces a favourable neutron economy. At transmutation rates equal to those of fast neutron systems, the actinide waste inventory in the thermal neutron system at one time can be up to 100 times smaller. This small inventory affords major safety and engineering advantages.

ATW minimizes the criticality concern, which is quite serious for reactors, since ATW **never achieves a self-sustaining chain-reaction**. The effective multiplication factor - k_{eff} - of the ATW system never reaches value of one, it is deigned to be at the

level of 0.9. If problems arise, such as an excess of heat in the transmuting waste, the accelerator can be simply turned off and the reactions immediately cease. The risk of a core-meltdown and explosion is thereby significantly reduced due to ATW's subcritical performance.

Like the reactor based approaches ATW can convert the heat generated by transmutation into electricity. It could produce enough electricity to keep the accelerator running and still have as much surplus power left over as a midsize commercial reactor. The very prominent feature of the ATW-system is the possibility of the effective burning of plutonium, which opens a new way of the destruction of weapons-return plutonium with the maximum societal benefit - converting plutonium into useful electrical energy. This version of the ATW-idea is called Accelerator-Based Conversion (**ABC**) and it is capable of rapidly burning plutonium in a low-inventory sub-critical system. The world's inventory of the reactor-produced plutonium is increasing at a rapid rate - around 80 tonnes per year. Figure 2 [13] shows the present inventory of plutonium and the rate at which it will accumulate if the number of power reactors continues unchanged. Currently, approximately 800 tonnes has been generated by nuclear power reactors and at current discharge burnups this amount is increasing by 80 tonnes per year. By the year 2015 there will be nearly 2000 tonnes of plutonium deposited at hundreds of sites around the globe. Swedish plutonium inventory by the year 2010 will exceed 60 tonnes. Figure 2 also shows a possible impact of the Accelerator-Based Conversion on the world's plutonium inventory. If ABC plants are introduced at app. one and one-half per year until 15% of the LWR generating capacity is replaced by ABC plants, the lower curve is obtained. In the long term, ABC plants could run on thorium cycle to allow even further reduction in plutonium.
By using higher burnup (and possible recycle) the plutonium accumulation can be decreased, however, in all cases the accumulation of plutonium increases and never gets below 2000 tonnes.

In its most advanced design the ATW-system coupled with reactor based plutonium burners offers the option for burning of 100 tonnes of weapons plutonium (about 1/3 of the current world inventory) in 30 years with a parallel transmuting of existing light water reactor wastes at the end of the LWR-life [14].

However, transmutation involves much more than simply bombarding the radioactive waste. Before it is placed in the ATW facility, the waste must undergo laborious chemical processing at various stages, primarily to separate elements requiring different treatment. This chemistry seems to be the most difficult challenge of these concepts [15].

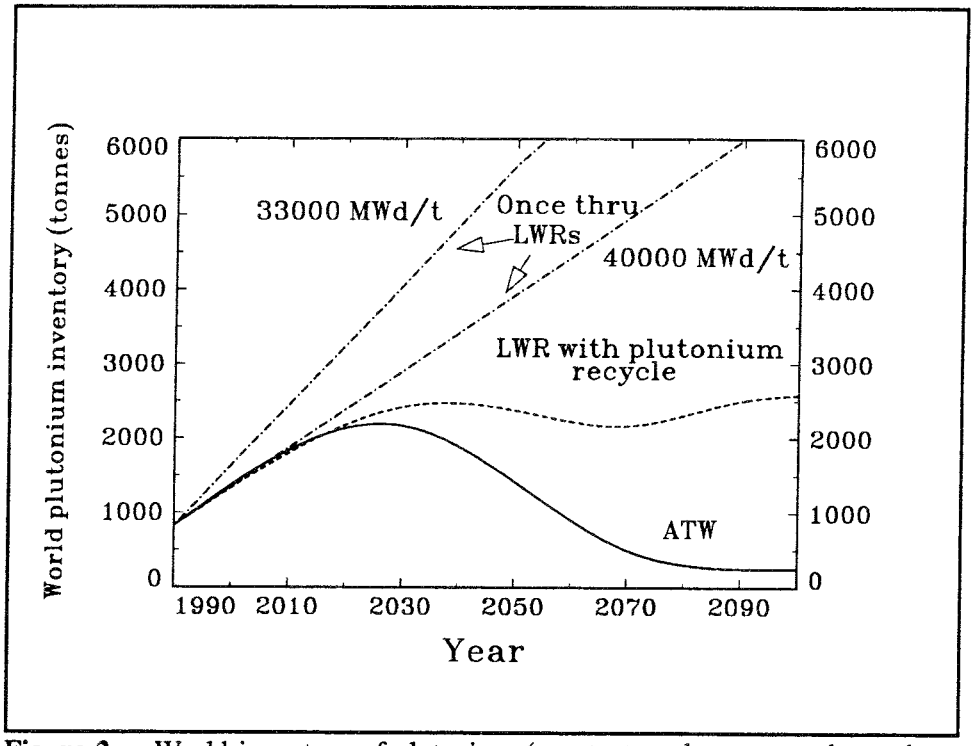

Figure 2 World inventory of plutonium (constant nuclear power demand - 1990 - assumed)

3. ATW-PROJECTS

Scientists at Los Alamos National Laboratory have under consideration two neutron-production-transmutation concepts to transmute key long-lived high level wastes (HLW) radionuclides: fission products and actinides. One concept is water-chemistry based and it is called the **aqueous system**, another one, **non aqueous system** is based on the molten salt chemistry. Corresponding two ABC-projects [16] based on the same principles are under serious considerations. The **ABC**-project, due to its specific goals, is beyond the scope of our report, however we **fully recognize and appreciate** its meaning in the context of the international safety and disarmament.

Another accelerator-driven transmutation system based on the fast neutron spectrum is being under development in Japan in the frame of OMEGA project.
In the following sections we describe in details those different ATW-concepts.

3.1 LOS ALAMOS BASE CASE AQUEOUS SYSTEM

So called base-case aqueous system is based on aqueous chemistry and aqueous neutron moderation. Heavy water is the carrier of the elements to be transmuted (in slurry-form), heat carrier and the neutron moderator. The target blanket module for this system consists of two thermally isolated systems; the target and the blanket. The target design is heavy-water-cooled solid tungsten and lead with a thermal power from beam deposition of 100 MW. The blanket surrounding the target is a heavy water moderated lattice of CANDU-like pressure tubes containing an actinide-oxide/heavy water slurry. Flow of the slurry through an external heat exchanger carries about 1540 MW of power out of blanket.

The target and blanket are intimately coupled neutronically. Efficient utilization of the spallation neutrons requires that most of the neutrons are absorbed productively in actinides (for large neutron multiplication) or fission products with minimal parasitic absorption in transmutation products, structure or target materials. It requires the efficient on-line removal of the transmutation products (to avoid further possible activation) and an efficient shielding of target from the thermal neutrons

The schematic diagram of the base-case (not neutron optimized) ATW aqueous system [17] is presented in Figure 3. On this figure - for the visibility's sake - only one target/blanket is shown.

The current processing concept incorporates individual processing loops for the fission products as well as two processing loops for the actinides. Two loops are used for the actinides to reduce the total system inventory. One loop is the primary feed loop for LWR actinides; the actinide blanket residence time in this loop is 30 days with a 10-day cooling/processing time outside the blanket. The processing removes the neptunium and plutonium from the remaining actinides and fission products for quick recycle in the blanket. The remaining actinides, primarily americium and curium, are cooled for 180 days, then separated from the fission products and recycled to the blanket to form a second loop. The actinides in this loop have a 180-day blanket residence time and 180-day cooling/processing time.

The main features of this system are:

1. 1600 MeV, 250 mA accelerator which drives 4 target/blankets
2. **Solid** W/Pb targets (see Figure 7)

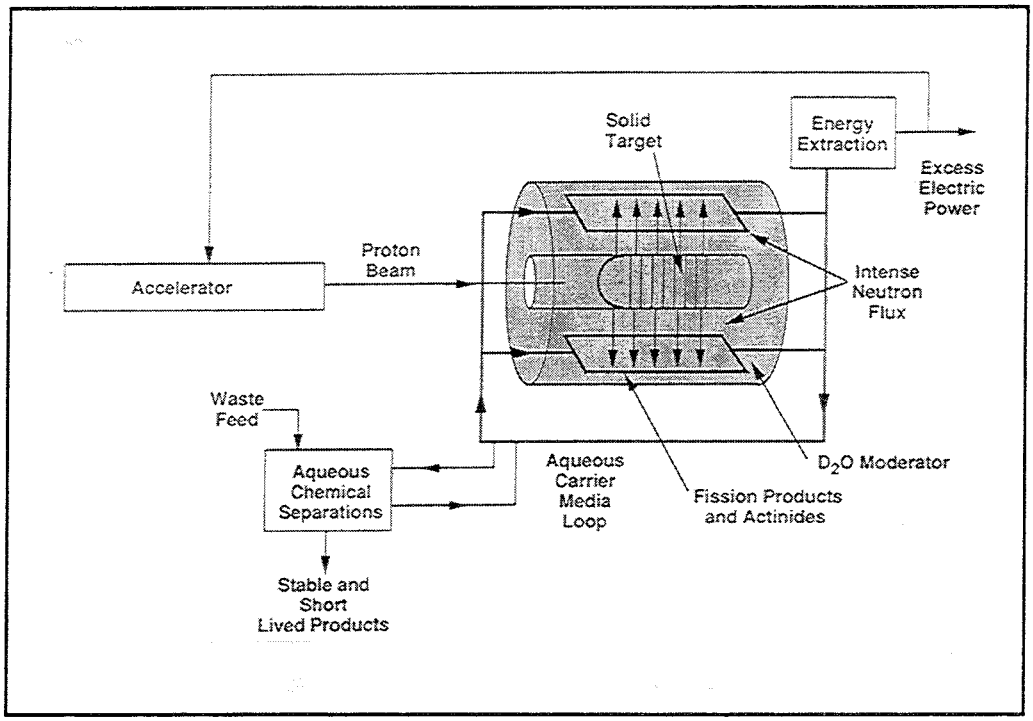

Figure 3 The base-case ATW aqueous system

3. D_2O CANDU-like blanket design:
   - 250 double wall pressure tubes
   - heavy water moderator and coolant
3. Oxide suspension for actinides (slurry) - slurry loading: 500 g/l
4. Supports actinide and technetium/iodine discharge from eight 3000 MW_T light water reactors
   - aqueous solution for technetium
5. Proven aqueous separations
6. Builds on demonstrated target and nuclear system technologies
7. Not optimized - numerous avenues for improvement
8. Plutonium burner operation mode possible with much better neutron economy than LWR feed.

These feature are schematically presented on Figure 4 [18].

Design goals for this system can be described as:

- *High transmutation efficiency*
  - high transmutation rates
  - rapid actinide/fission product processing
  - high light water reactor support ratio

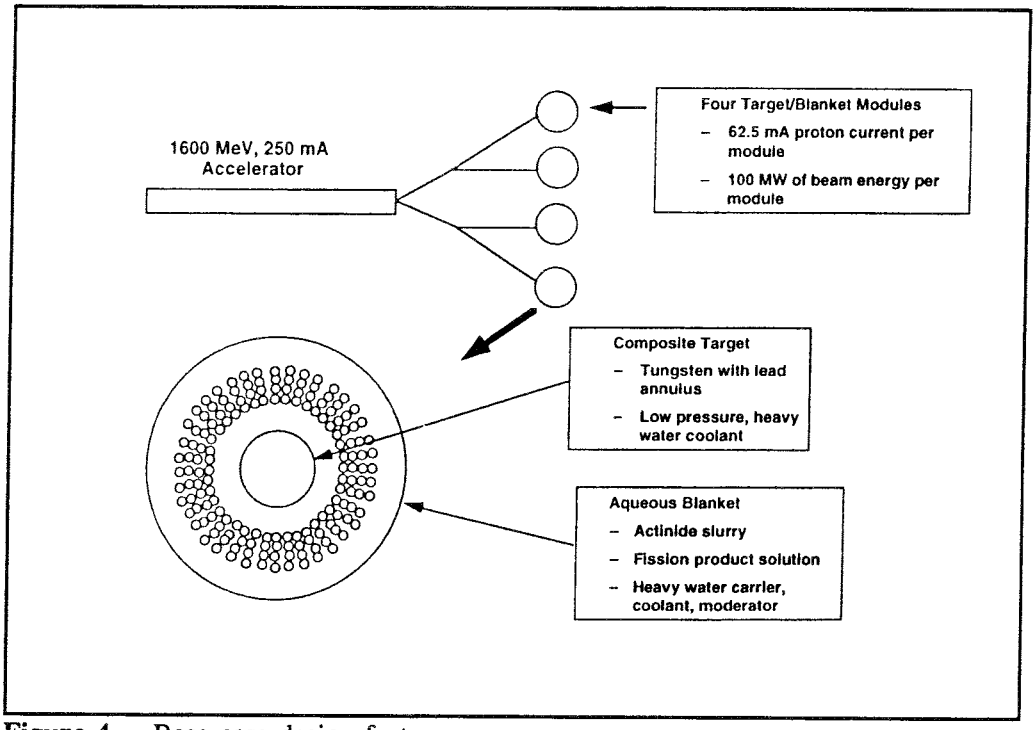

Figure 4 Base case design features

∵ *Environmental safety and health consideration*

- reduce end-of-life waste
- **minimal system inventories**

∵ ***Safety by design***

- passive decay heat cooling
- subcritical operation

∵ *Cost considerations*

- efficient neutron utilization
- reasonable thermal efficiency (but high pressure of the slurry is required)
- produce sufficient electricity to power accelerator and send excess to grid

∵ *Use existing technologies to extent possible*

The practical, detailed design of the aqueous concept is based on the technology used in heavy water CANDU reactors. The parameters of CANDU design adopted for the ATW aqueous system are presented on Figure 5.

This transmuter design :

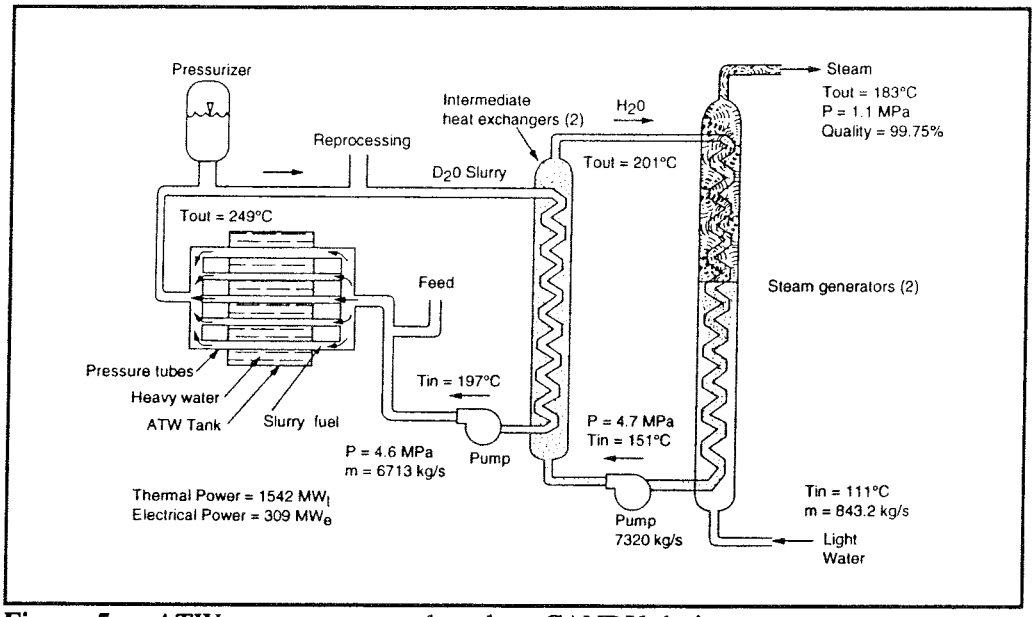

Figure 5 ATW-aqueous system based on CANDU design

- burns 2500 kg/yr actinides
- transmutes 250 kg/yr Tc

  203 kg/yr from LWR feed

  47 kg/yr internally generated from actinide burning
- transmutes 79 kg/yr iodine

  62 kg/yr from LWR feed

  17 kg/yr internally generated from actinide burning

3.1.1 Transmutation of actinides.

A lot of experience concerning actinide slurries was gained at Oak Ridge National Laboratory during the development of the homogeneous nuclear reactor:

- Thorium and uranium oxide slurries up to the concentration 1500 g/l and 300°C were tested
- Zirconium alloys showed good resistance to erosion for 500 g/l slurries in several tests with velocities up to 6 m/s

For the base case ATW, the slurry operates at a peak temperature of 250°C, a concentration of 70 g/l and a velocity of 3.7 m/s. To increase both the thermal and neutron efficiency one is considering slurries that operate up to 325°C, a concentration of 500 g/l and a velocity of 6 m/s (or even higher). These conditions are well within the experience at ORNL. Care must be taken in the design to keep the flow turbulent (to avoid settling), and to avoid "dead spots" where particle accumulation may occur.

Figure 6 presents the double actinide loop concept which significantly reduces inventory of actinides [19].

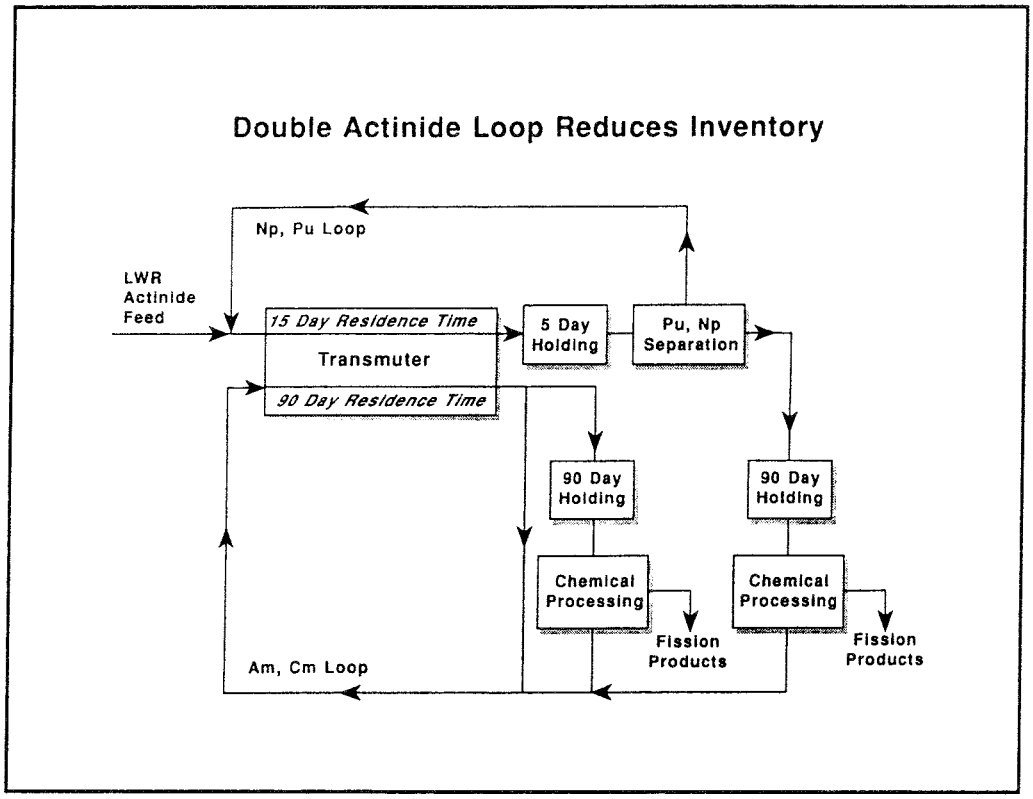

Figure 6 Double actinide loop for ATW-system

Annual discharge of fission products from actinide burning at ATW system:

- 90 kg long-lived fission products
  - ^{79}Se 0.3 kg
  - ^{93}Zr 34 kg
  - ^{107}Pd 47 kg
  - ^{126}Sn 2.0 kg
  - ^{135}Cs 6.0 kg

- 2410 kg stable and short lived (< 30 yr) fission products
  - ^{90}Sr 14 kg
  - ^{137}Cs 75 kg

3.1.2 Transmutation of fission products (^{99}Tc and ^{129}I)

Special attention in nuclear waste handling is given to two fission product isotopes, namely ^{99}Tc and ^{129}I. ^{99}Tc, a β-emitter, E_{max}= 0.292 MeV, is one of the most troublesome radioactive fission products due to its large fission yield - 6.1% for thermal neutron induced ^{235}U-fission, and to the long half-life, $T_{1/2}$=213 000 years. ^{129}I is one of the longest lived fission products - $T_{1/2}=15{,}7\cdot10^6$ years. The fission yield from the thermal fission of ^{235}U is 0.75%. It is also a β-emitter (E_{max}= 0.606 MeV), emits a weak γ at 39.6 keV, and is considered not to be very radiotoxic. However, technetium and iodine are highly mobile and dominate long-term radiological risk in geological repository of the spent fuel due to water intrusion and leaking. These risks are strongly dependent on the type of the geological repository bedrock. Due to the specific geological formation of the swedish bedrock the long-term radiological risk of the future swedish geological waste repository due to technetium and iodine mobility is considered to be very small. It is, however, important to realize that the total inventory of ^{99}Tc in swedish spent fuel will exceed 5 tonnes at year 2010. The respective number for ^{129}I is 1.2 tonnes.

In ATW-system ^{99}Tc is transported through the blanket in a heavy water solution. A separate heat exchanger is required to remove the small amount of sensible heat that it absorbs. Although transmutation via thermal neutron absorption is capable of significant reducing the long-term environmental or repository risk posed by ^{99}Tc an essential problem is the ability to separate a stable ruthenium (to which Tc transmutes) with as little as possible Tc contaminant. While the chemistry of the second-row transition elements is difficult and very complex [20], particularly so for Ru, several technologies are known which readily lend themselves to the scale proposed for transmutation. In addition to ion exchange, for which a considerable body of literature, including published separation factors is available, volatilization via ozonolysis is a competitive technology with substantial industrial experiences and some advantages over ion exchange in terms of waste stream management and processing. The recommended chemistry is:

- Form chosen for transmutation: $^{7}LiTcO_4$.
- Tc/Ru separation via ozonlysis, which demonstrates Tc/Ru separation factors higher than 10^5

The successful transmutation of ^{129}I depends on the rate of the transmutation. The thermal neutron capture cross sections for ^{129}I are 20 and 10 barns, to isomeric

and ground states of ^{130}I, respectively. Neutron capture consideration must be given for the stable ^{127}I that would also be present (σ_c= 6.2 b) in the transmutation system and would reduce the transmutation efficiency. The iodine - ^{129}I - fission products are currently designed to be placed in solid form, with the stable xenon transmutation product continually removed from the system [21]. The design is complicated by the low conductivity and low melting point of iodine, however, with the rods of approximately 0.5 to 1 cm radius and with the energy release from the formation and decay of ^{130}I the temperatures could be maintained at the level where elemental iodine would not vaporize appreciably. The neutronics of ^{129}I transmutation is very favourable - neutron capture leads to the stable isotopes of xenon (^{130}Xe, ^{131}Xe). The chemical form chosen for iodine transmutation is I_2 . The important points for this kind of chemistry are:

- *I_2 has multiple oxidation states*
- *Uncertain and complex radiolysis products exist*
- *Iodine and its compounds have corrosive properties*

3.1.3 Target/Blanket materials.

The solid tungsten target design is presented in details in Figure 7.
Total radioactivity in the target is small compared to that in the blanket for applications with fissionable material. The inventory of the long lived radioactive isotopes in Curies is less than 1% that of the blanket and it is dominated by long-lived Pb isotopes (^{205} Pb and ^{202}Pb, look Figure 19). Gas and volatile radioactivity is about 0.01 % that of the blanket. All the long-lived radioactive isotopes produced in the target can be finally transmuted - at the end of target life - in the blanket.

The target/blanket structural materials for ATW base case design were chosen based on experiences from the material studies for many different reactors and accelerators.

- *Target: Tungsten rods in high power density regions, lead rods clad in aluminum in annulus*
- *Structure: Aluminum in target, moderator tank and actinide external tubes*
- *Beam entry window: Inconel 718*
- *Actinide transport: Zircalloy-4*
- *Technetium transport: Zircalloy-4*
- *Iodine tubing: ceramic lined Zircalloy-4*

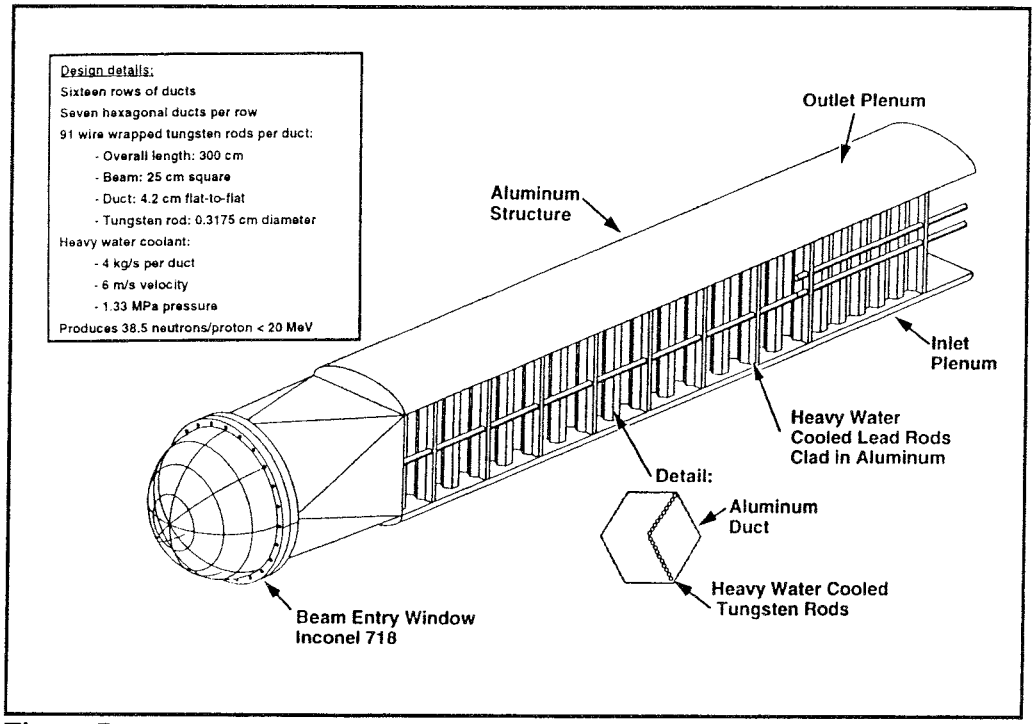

Figure 7 Tungsten target design

3.1.4 Inventory of radioactive materials

Tab. 1 presents some basic data concerning the radioactive inventory at equilibrium together with annual system feed requirements for the baseline ATW system with 4 target/blankets as presented on Figure 4.

Tab. 1 Inventory of some radioactive isotopes at equilibrium

Baseline ATW system
250 ma, 1.6 GeV Accelerator
4 Target/Blankets
Supports 10 LWRs @ 100% capacity

Isotope	Blanket/HEX Inventory (kg)	System inventory (kg)	System feed (kg)	Processing Loss (eff = 10^{-5}) (kg)
^{237}Np	97.6	130.	147.	$3.17 \cdot 10^{-2}$
^{238}Np	0.841	1.12		$2.73 \cdot 10^{-4}$
^{239}Np	$4.02 \cdot 10^{-3}$	$5.36 \cdot 10^{-3}$		$1.3 \cdot 10^{-6}$
subtot	98.4	131.0	147.	$3.19 \cdot 10^{-2}$
^{238}Pu	82.7	110.	45.7	$2.69 \cdot 10^{-2}$
^{239}Pu	222.	296.	1680.	$7.21 \cdot 10^{-2}$

Isotope	Blanket/HEX Inventory (kg)	System inventory (kg)	System feed (kg)	Processing Loss (eff = 10^{-5}) (kg)
^{240}Pu	596.	794.	775.	$1.93 \cdot 10^{-1}$
^{241}Pu	178.	237.	256.	$5.78 \cdot 10^{-2}$
^{242}Pu	2870.	3830.	157.	$9.33 \cdot 10^{-1}$
^{243}Pu	4.03	0.538		$1.31 \cdot 10^{-4}$
^{244}Pu	11.6	15.4		3.76· 10-3
subtotal	3960.	5290.	2910.	1.29
^{241}Am	36.7	73.4	168.	$298. \cdot 10^{-3}$
^{242}Am	0.182	0.364		$1.48 \cdot 10^{-5}$
^{242m}Am	0.568	1.14	0.22	$4.61 \cdot 10^{-5}$
^{243}Am	715.	1430.	3.02	$5.81 \cdot 10^{-2}$
^{244}Am	0.0284	0.0569		$2.31 \cdot 10^{-6}$
^{244m}Am	.0236	$2.68 \cdot 10^{-4}$		$1.91 \cdot 10^{-6}$
^{245}Am	$1.34 \cdot 10^{-4}$	$2.68 \cdot 10^{-4}$		$1.09 \cdot 10^{-8}$
subtotal	753.	1510.	198.	$6.11 \cdot 10^{-2}$
^{242}Cm	35.	70.		$2.84 \cdot 10^{-3}$
^{243}Cm	0.881	1.76	0.112	$7.15 \cdot 10^{-5}$
^{244}Cm	3130.	6260.	5.87	$2.54 \cdot 10^{-1}$
^{245}Cm	29.7	59.3	0.311	$2.41 \cdot 10^{-3}$
^{246}Cm	1140.	2280.		$9.24 \cdot 10^{-2}$
^{247}Cm	42.8	85.5		$3.47 \cdot 10^{-3}$
^{248}Cm	236.	473.		$1.92 \cdot 10^{-2}$
^{249}Cm	$2.44 \cdot 10^{-3}$	$4.88 \cdot 10^{-3}$		$1.98 \cdot 10^{-7}$
subtotal	4610.	9230.	6.29	$3.75 \cdot 10^{-1}$
Actinide total	**9430.**	**16200.**	**3270.**	**1.75**
^{99}Tc	639.	767.	268.	$3.11 \cdot 10^{-2}$
^{127}I	126.	252.	20.2	$1.02 \cdot 10^{-2}$
^{128}I	$1.750 \cdot 10^{-3}$	$3.49 \cdot 10^{-3}$		$1.42 \cdot 10^{-7}$
^{129}I	116.	232.	85.1	$9.41 \cdot 10^{-3}$
subtotal	242.	484.	105.3	$1.96 \cdot 10^{2}$
fission products total	**881.**	**1250.**	**373.**	**$5.07 \cdot 10^{-2}$**

3.2 LOS ALAMOS ATW NON-AQUEOUS (MOLTEN SALT) SYSTEM

ATW non-aqueous system called also an advanced system was designed to optimize the neutron economy. This system is still in the very conceptual stage, many details are under considerations but the main features seems to be decided: graphite moderation of the neutrons (possibly the HTGR-like or pebble-bed reactor-like design), helium or liquid lithium (or lithium salt) cooling, flowing lithium, lithium salt or flowing lead target, and molten salt chemistry (^{7}LiF and BeF_2). In difference to the aqueous chemistry the molten salt chemistry of actinides and fission products has never been really worked-out. Intensive research efforts together with a careful review of existing experimental data in molten salt chemistry are required [22]. The liquid lithium target gives the opportunity to decrease the accelerator beam energy to the level of few hundred MeV.

3.2.1 System outline

The most promising design option for an advanced ATW concept uses a lithium target with uranium multiplier and a multiplying blanket which is graphite moderated and lithium (helium) cooled. Molten salt (fluoride) is used and there is a thorium/uranium breeding region (Figures 8 and 9). Closed loop, non-aqueous separation would be used for both front and back-end fuel processing. The lithium-uranium target offers very efficient neutron production (estimated at 28 neutrons per 800MeV deuteron) [Figure 1] and low absorption of blanket neutrons because of its small size and presence of the fission product transmutation region surrounding it. This target produces a very small amount of heavy spallation products. The molten salt is contained in near-stationary conditions within graphite tubes, and cooled in the blanket by lithium metal or eutectic mixture of lithium and beryllium fluoride (alternatively helium gas). Isotopically pure lithium - ^{7}Li - must be used in the blanket, because of the large thermal absorption cross section of ^{6}Li. The multiplying blanket is completely submerged in a pool of the coolant fluid. The coolant is forced through the fuel elements by pumps placed outside the blanket. The fuel elements are designed to allow the free convection of the molten salt inside. As a result of this convective movement, high power densities can be reached in the blanket without boiling the fuel. For a thermal flux ^{7}Li has a negative void coefficient of reactivity and is probably the best coolant to use in high flux thermal systems.

Graphite-moderated, He- or lithium-cooled, molten salt design as presented on functional layouts - Figure 8 - lithium-cooled layout and Figure 9 - pebble-bed design.

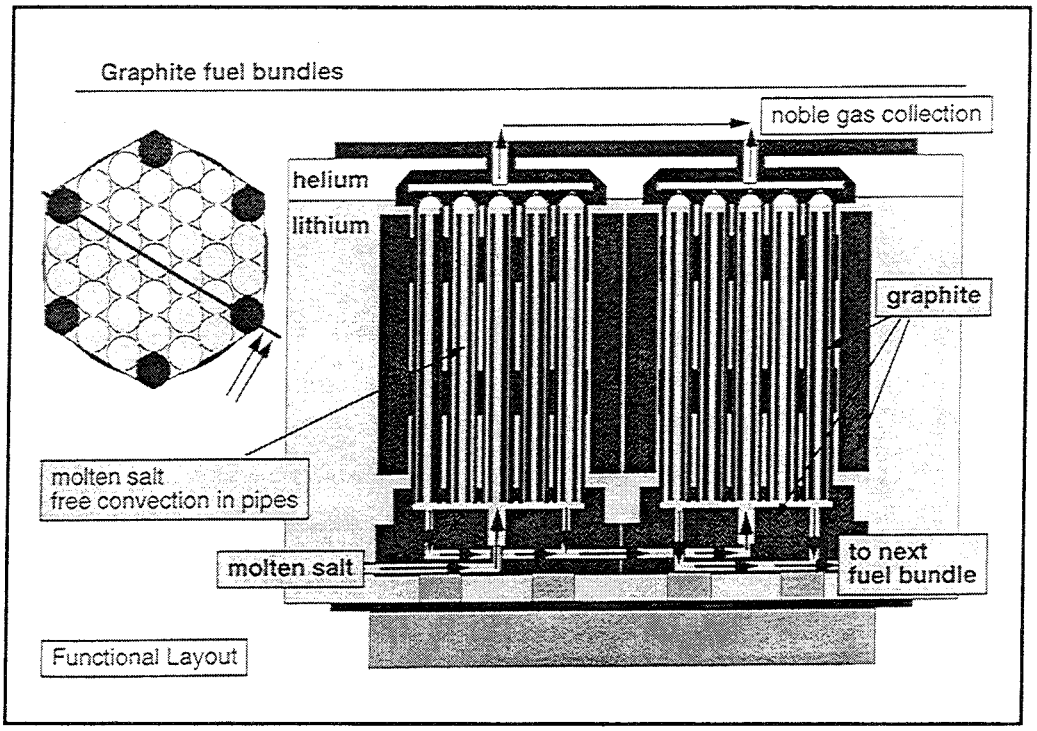

Figure 8 Graphite-moderated, molten salt, lithium-cooled advanced ATW system

Features of the ATW advanced system target-blanket:

- *Flowing liquid lead spallation source [23] with beryllium multiplier*
  24 neutrons per incident 800 MeV proton
  Allows high thermal neutron fluxes with existing materials and equipment

OR

Flowing liquid lithium-7 (or molten lithium salt) source with uranium multiplier

28 neutrons per incident 800 MeV deuteron (or proton)
Enhances moderation of neutrons, allows very high power density and flux

- *High fluxes allow maximum advantage from ^{238}Np and ^{242}Am fission and allow a minimum inventory*
- *Graphite moderator*

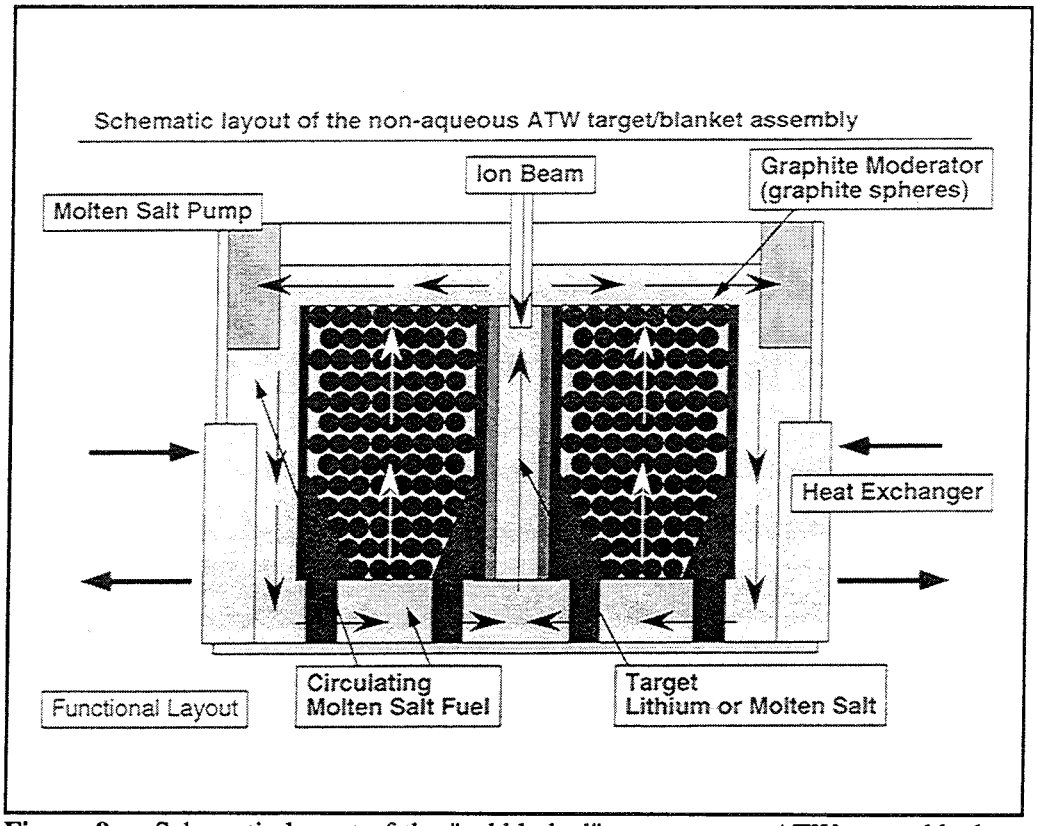

Figure 9 Schematic layout of the "pebble-bed" non-aqueous ATW target-blanket assembly

Low parasitic capture, excellent high temperature behaviour

- *Helium or liquid lithium (molten salt) cooled*

  Transparent to neutrons, inert in all condition

- *Molten salt fuel*

  Well suited to possible centrifuge separation (back-end chemistry) [24] and dry-halide partitioning (front-end chemistry)

- *Fertile thorium blanket, thorium breeding without reprocessing improves operating economy*

  Optimal utilization of leakage neutrons

- *Excellent neutron economy reduces accelerator size*
- *Construction materials allow high thermal-to-electric efficiency*

One design of non-aqueous system allows three distinct energy/transmutation

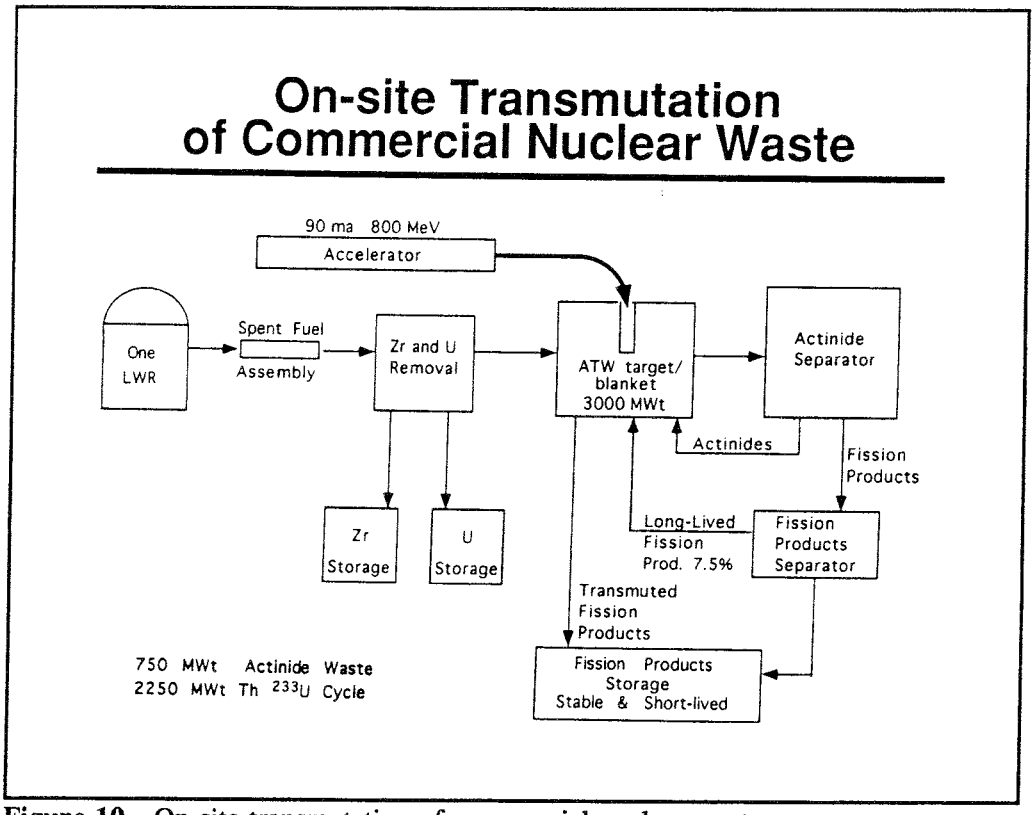

Figure 10 On-site transmutation of commercial nuclear wastes

functions:

On-site transmutation of commercial nuclear waste as shown in Figure 9

Energy park for commercial waste transmutation as presented in Figure 10

Power production with concurrent waste transmutation as demonstrated in Figure 11

The thorium-uranium cycle presented in the power production with concurrent waste transmutation concept - Figure 12 - opens practically unlimited resources of nuclear fuel without the necessity of using breeder reactors. Excellent neutron economy of these concepts reduces the accelerator size and the beam energy which makes this option technologically very attractive.

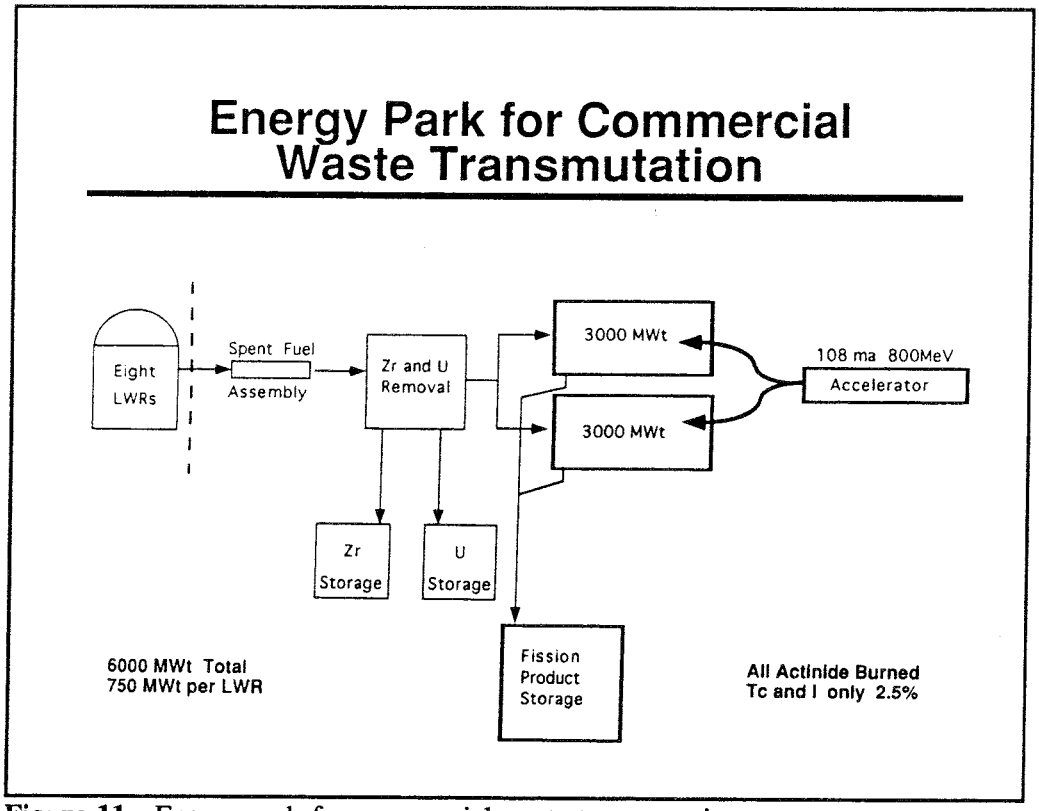

Figure 11 Energy park for commercial waste transmutation

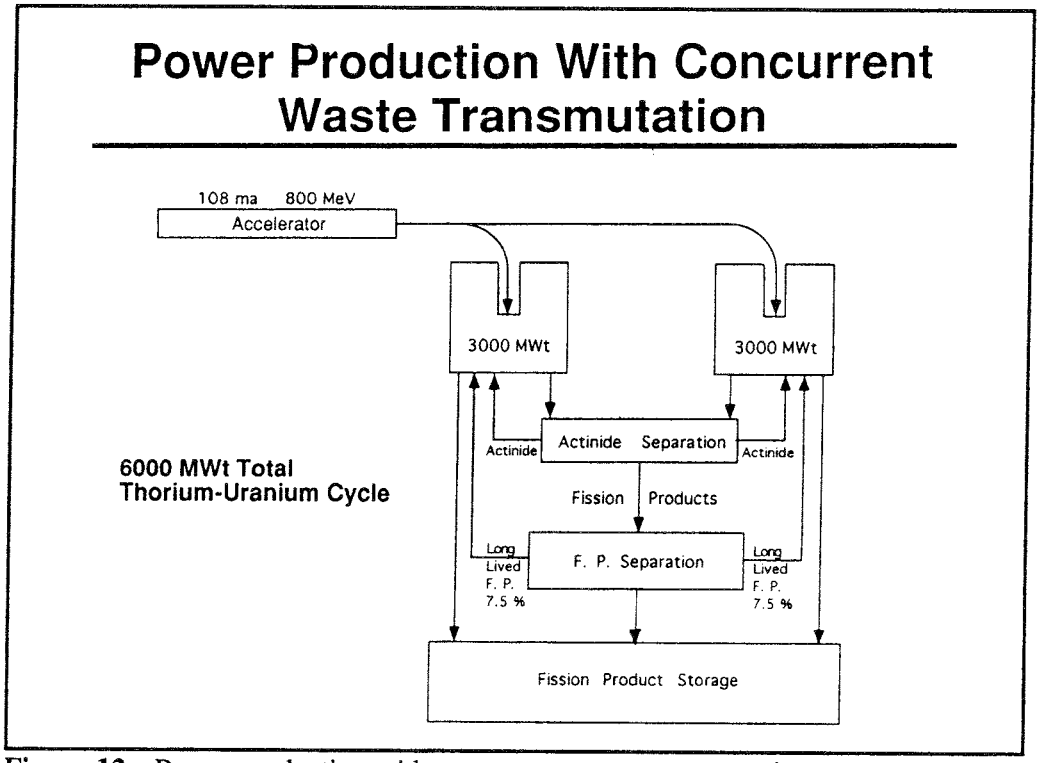

Figure 12 Power production with concurrent waste transmutation

3.2.2 Safety features

∵ Subcritical operation

∵ Low inventory of actinides and fission products

High thermal neutron flux with no dilution in external heat exchangers.

non radioactive coolant. No spallation products in target. Low pressure.

∵ Negative temperature and void reactivity coefficient.

∵ Passively safe in the event of loss of coolant accident.

Large thermal inertia and good heat conductivity.

Molten salt can be drained away without external intervention.

∵ Proliferation resistant fuel cycle

Uranium enrichment lower than 20%

Thorium breeding without reprocessing.

3.3 ATW DEVELOPMENT PLAN AT LOS ALAMOS NATIONAL LABORATORY

The time required for the development of first ATW-demonstration plant was estimated in Los Alamos for about 15-20 years. Figure 13 presents the ATW development plan divided into 3 phases and stretched out in time into 15 fiscal years. The plan for the first phase is roughly sketched as:

Phase 1 efforts:

Deliverables:

- Produce consistent point design, backed by experiment; determine credible material balance, performance, and cost data
- Provide credible solutions/data to major technical issues
- Basis for decision to proceeding to additional demonstration stages

Cost and duration:

- 3 years (after 1993)
- ~ 180 · 10^6 $ for 3 years [25].

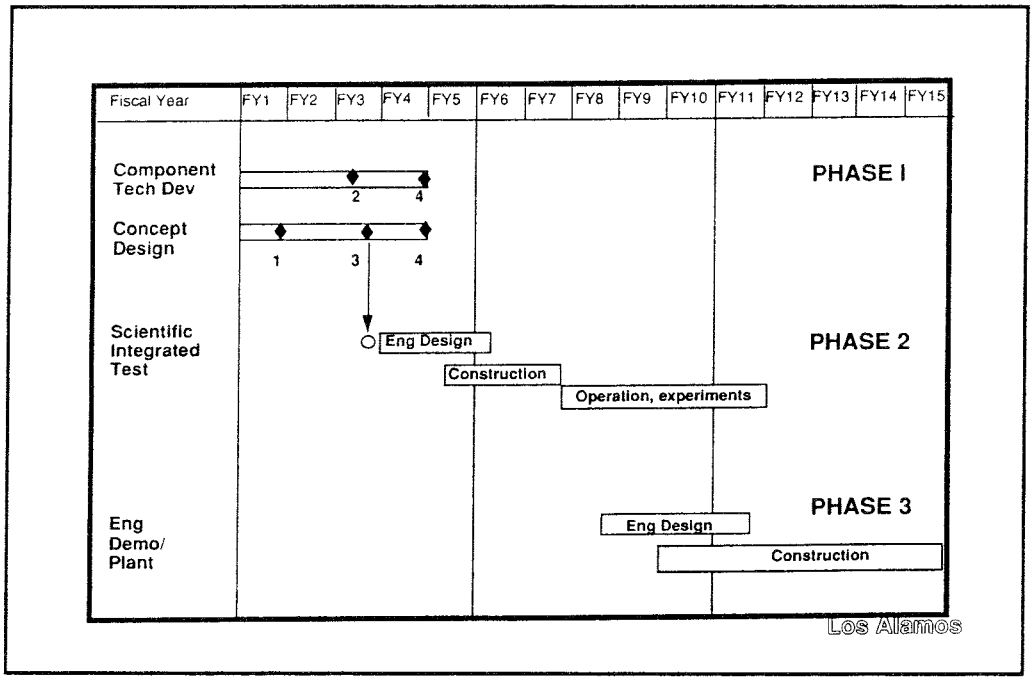

Figure 13 ATW development plan at Los Alamos National Laboratory

Phase 1 most important issues:

Fiscal year 1 - Complete self consistent point design that meets performance goals; initial material balance assessment

- Actinide and fission product chemistry processing components specified; performance and waste generation estimated
- Credible target reference design completed and used in preliminary radionuclide production calculations
- Blanket/processing system defined establishing base neutronics performance and design methodology
- Credible accelerator beginning reference design completed

Mid fiscal year 3 - Complete initial suite of key measurements needed for performance/material balance definition and assessment

- Target neutron performance/spallation product experiments
- Target mockup thermal hydraulic tests
- Beginning integrated component tests (LAMPF beamstop)
- Slurry definition, erosion fabrication tests
- Blanket component mockup flow tests

- Actinide flowsheet tests
- Fission product separation demonstration
- Initial waste stream results for process loops

3rd quarter fiscal year 3 - Complete improved concept design and material balance analysis using new experimental data and calculational results

- Decision - Initiate phase 2 start?

End of fiscal year 4 - Complete second stage of individual component test and demonstrations, incorporate results into preliminary system engineering design.

3.4 OMEGA PROGRAM IN JAPAN

The program is composed of two major R&D areas: the nuclide partitioning technology development and the transmutation technology development. The overall scope of the program is illustrated in Figure 14 [26].

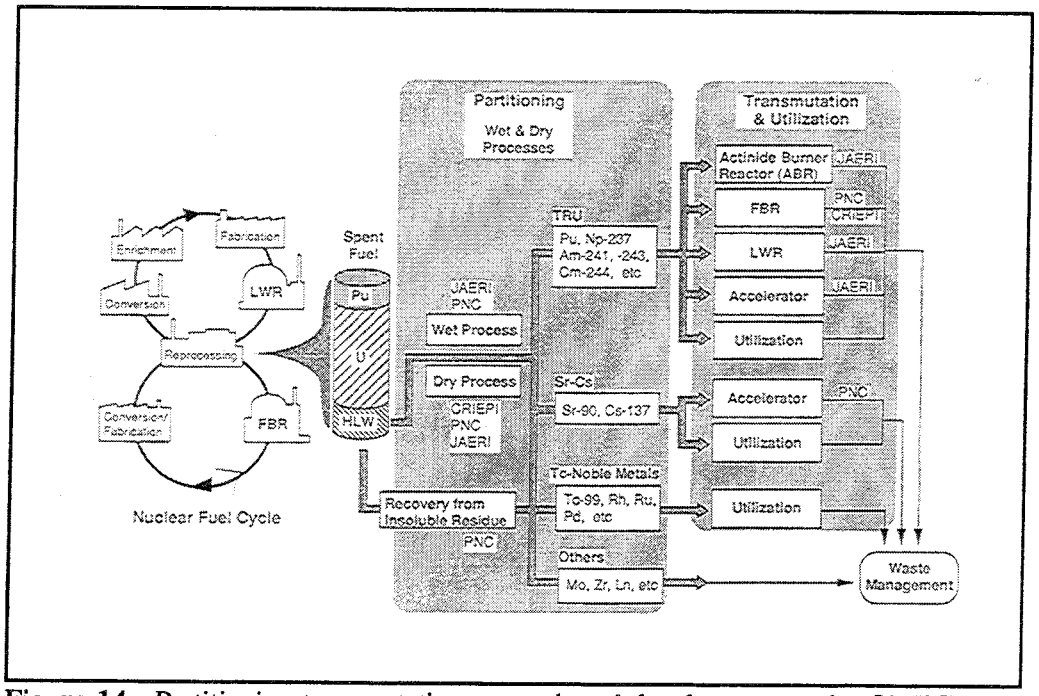

Figure 14 Partitioning-transmutation research and development under OMEGA program

The budget of OMEGA program is about $3 \cdot 10^9$ yen / year excluding salaries.

Several potential methods or concepts are being investigated in parallel in both areas by JAERI (Japan Atomic Research Institute), PNC (Power Reactor and Nuclear Fuel Development Corporation) and CRIEPI (Central Research Institute of Electric Power Industry).

Accelerator-driven transmutation system is under development mainly at JAERI.

Conceptual design study has been conducted on the accelerator-driven transmutation system, which is hybrid system composed of an intense proton accelerator with a proton energy of 1.5 GeV and a current around 40 mA, and a solid or molten salt target containing minor actinides and other long-lived nuclides - Figure 15. This system is capable to incinerate about 260 kg of minor actinides a year.

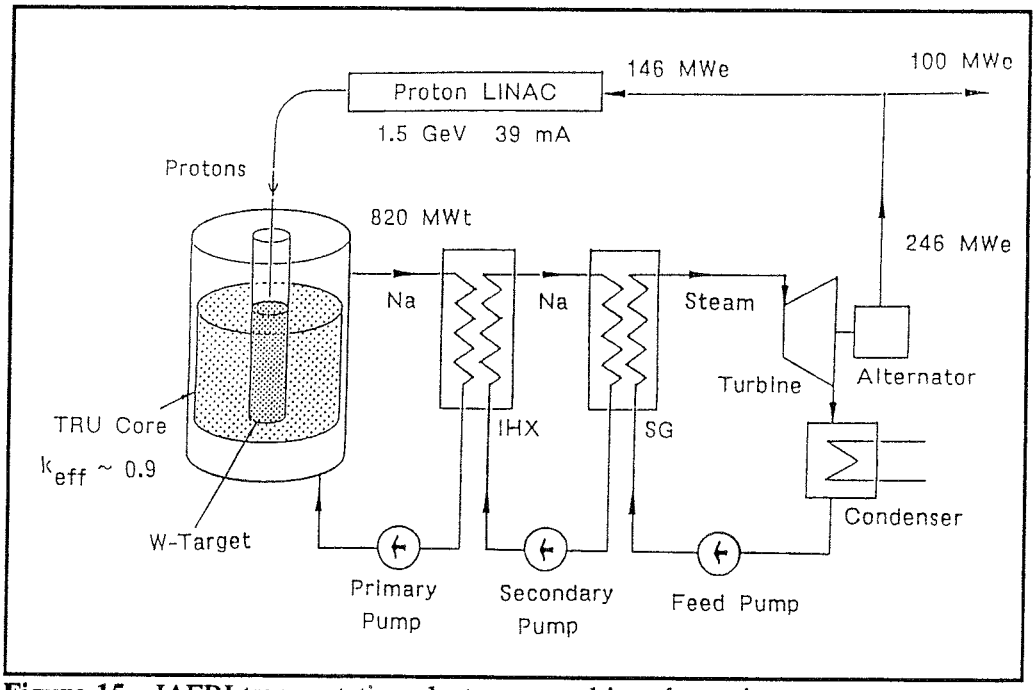

Figure 15 JAERI-transmutation plant concept driven by an intense proton accelerator

Core design of JAERI concept is based on solid-state pin-bundle type fuel elements and sodium coolant. It offers some advantages like high power density, and high temperature operation under normal pressure. Table 3 summarizes more important features of the transmutation system assisted by an intense proton linac which is under development at JAERI.

Study is also in progress on the system with a chloride molten salt assembly which is intended for transmuting not only minor actinides but also fission products.

In order to conduct various engineering test for the accelerator driven transmutation plant, a plan to construct the "Engineering Test Accelerator" (ETA) with a proton energy of 1.5 GeV and a current of 10 mA has been proposed. As the first step of developing the ETA, the "Basic Technology Accelerator" (BTA) with the proton energy of 10 MeV and the current of 10 mA is ready to be constructed to study a lower energy portion of the ETA.

An idea based on transmutation through photonuclear reaction (γ,n) by using bremstrahlung radiation from accelerated electrons is under development at PNC. The work is concentrated mainly on transmutation of ^{137}Cs using the giant resonance of the (γ,n) reaction. The main components of a high intensity electron linear accelerator with the current of 20 mA at the acceleration energy of 10 MeV are under construction. The first test operation of the accelerator is scheduled in 1995. However, the energy balance of this method is very poor and unless one can produce a high intensity monoenergetic γ-rays with an exact energy of the (γ,n) resonance, this method can not be considered as reasonable for transmutation purposes on a larger scale.

Japan is conducting a very impressive R&D program related to the partitioning and transmutation. The program is generally aiming at seeking the potential usefulness of the HLW and exploring the methods to reduce long-term burdens of the nuclear waste management in future. This program, very fortunately, is ".. not intended for seeking **short-term alternatives** for presently adopted back-end policies, but is conceived as **a research effort to pursue benefits for future generations through long term basic R&D's**. It will also contribute to revitalizing nuclear-related R&D's which brings the nuclear option into the 21 century" [8].

Tab. 3 Design parameters of minor actinides transmutation system assisted by an intense proton LINAC

Proton beam current			39 mA
Actinide inventory			3610 kg
k_{eff}			0.89
No. of neutrons per incident proton			40n/p
No. of fissions	(>15 MeV)		0.45f/p
	(<15 MeV)		100f/p
Neutron flux			$4x10^{15}n/cm^2 \cdot s$
Mean neutron energy			690 keV
Burnup			250 kg/y
MA thermal output		fuel	800 MW
		tungsten	20 MW
		total	820 MW
Power density		maximum	930 MW/m^3
		average	400 MW/m^3
Linear power ratio		maximum	61 kW/m
		average	27 kW/m
Coolant temperature			
	outlet	maximum	473° C
		average	430° C
Maximum temperature			
	fuel	center	890° C
		surface	548° C
	clad	inside	528° C
		outside	484° C

3.5 PERFORMANCE COMPARISON OF ATW-SYSTEMS

Significant improvements in the rate of actinide inventory reduction are possible with ATW. Defining the inventory reduction factor $\Psi(t)$ [27],[28] as

$$\Psi(t) = \frac{\textit{Actinide Inventory Without Transmutation}}{\textit{Actinide Inventory With Transmutation}}$$

one can calculate the time needed to achieve a 90% inventory reduction ($\Psi(t)=10$) for different systems:

- ∴ 184 years for an Advanced Liquid Metal Reactor system ($\beta=0.62$ - actinide burner mode)
- ∴ 16 years for the aqueous ATW system
- ∴ 2 years for an non-aqueous ATW system.

Figure 16 presents the performance comparison of different ATW concepts. Japanese OMEGA and Brookhaven's PHOENIX concepts are based on the fast neutron flux which increases very significantly the inventory of the system. As the consequence the performance measured as the ratio of mass burned/inventory drops down to the fast reactor level performance (IFR - Integral Fast Reactor, new generation of fast reactors developed at Argonne National Lab.). Superior performance of the advanced ATW depends on very good neutron economy. Tab. 2 presents the comparison of the different ATW concepts with the integral fast reactor performance.

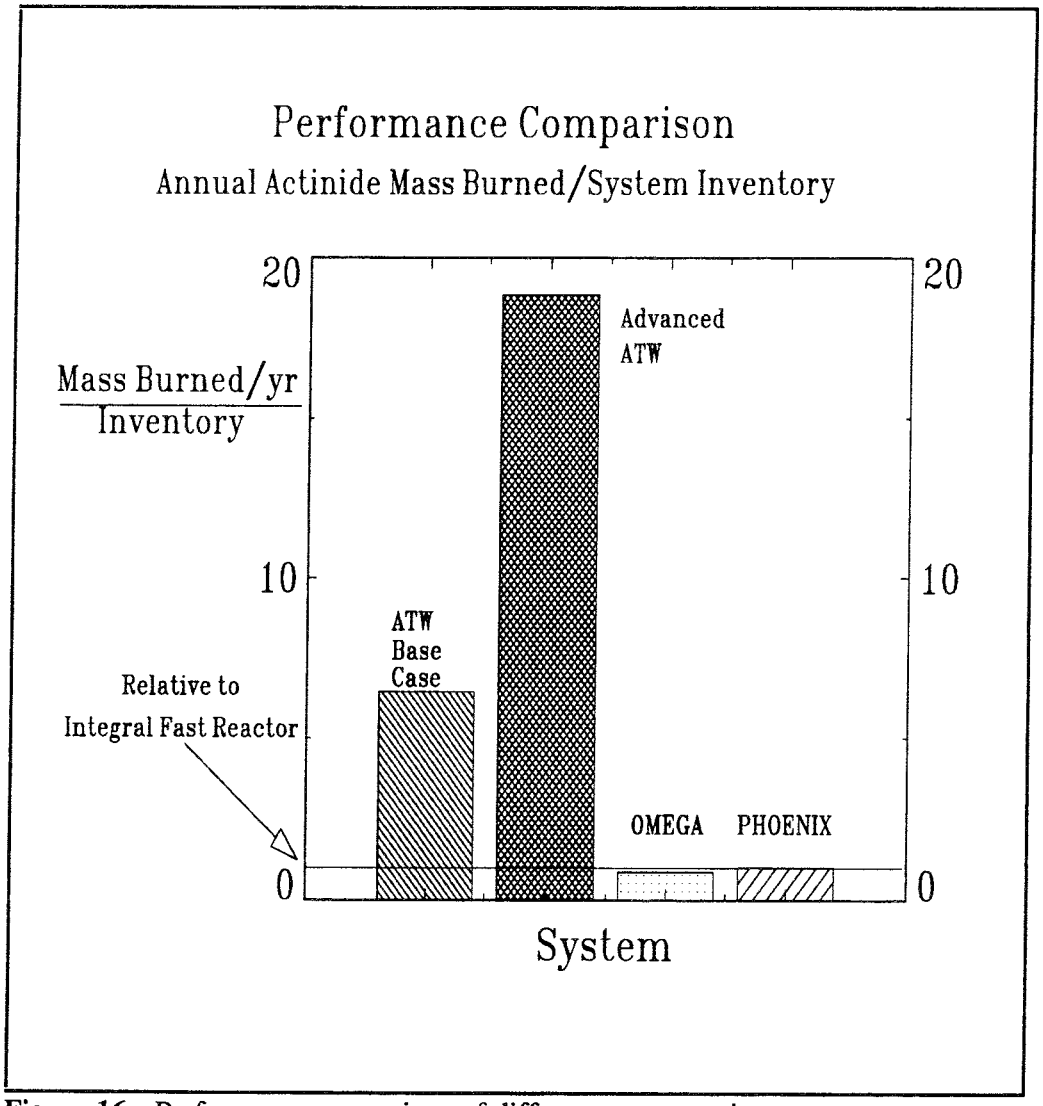

Figure 16 Performance comparison of different transmutation system

Tab. 2 COMPARISON WITH OTHER TRANSMUTATION SYSTEMS

Concept	Actinide (Pu,HA) (kg/yr)	FP (kg/yr)	Yearly Burn/Sys. Inventory	Blanket Inventory (kg)	Neutron Spectrum	Separations (Yearly Frequency)	Thermal Power (MW)	Electricity Net (MW)	Beam Power (MW)
ATW (BASE)	2600 (2320,280)	328	0.4	2000	T,E	aqueous (20)	6669	450	400
ATW (advanced)	326	70	1.2	210	T,E	physical (20)	3000	1000	90
IFR (Pu)	352 (308,44)	0.	0.07	4000	F	pyro (1/3)	1157	450	N/A
IFR (MA)	346 (26,320)	0	0.09	3000	F	pyro (1/3)	1157	450	N/A
OMEGA	202	0	0.05	2866	F	pyro, aqueous (1/3)	691	0	34
PHOENIX	2600	300	0.08	25000	F	aqueous (1/3)	3600	850	166

4. SAFETY CONCERNS OF ATW-SYSTEM

In order the perform a reasonable safety analysis of the ATW-system it is necessary to look carefully to every important component of this system. Figure 17 presents the main physical components of ATW concept.

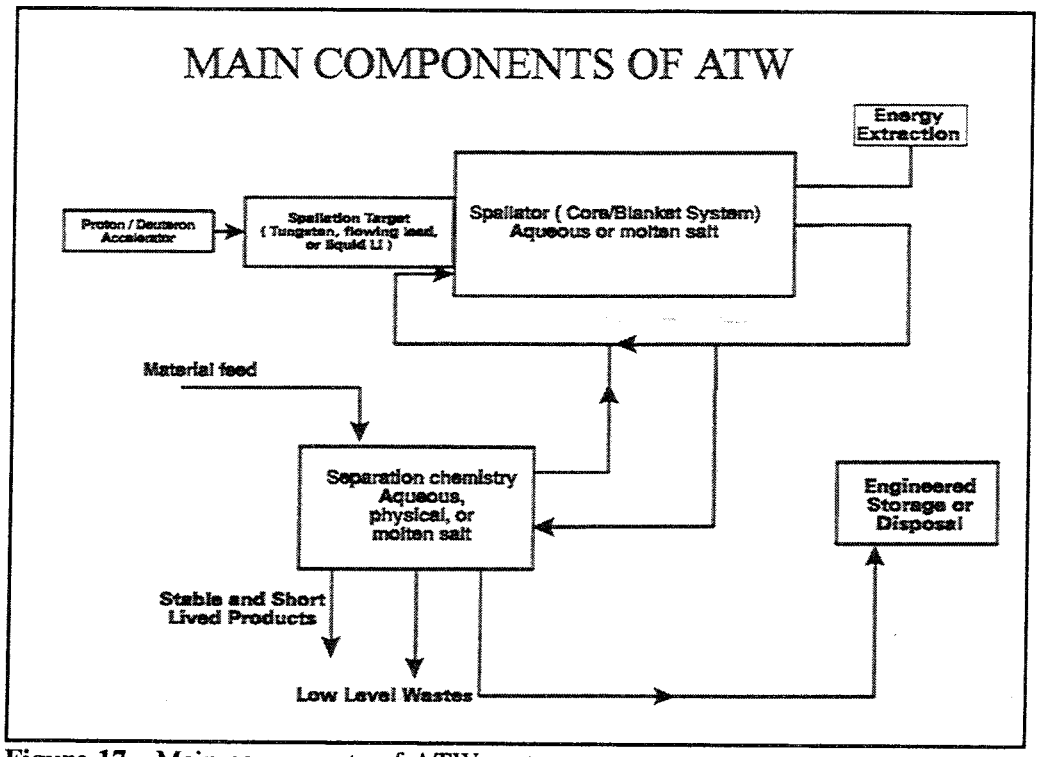

Figure 17 Main components of ATW system

We shall first point at some properties of the different components which might be important for the safety of the ATW system.

I. The construction phase

This phase will not offer any special safety problems but should be taken into account when comparing with other methods for taking care of the nuclear waste.

II. Transportation and prestorage of the nuclear waste from the reactors to the ATW plant.

Here some radiation risks might occur which should be evaluated. They are very similar to those associated with the presently assumed fuel cycle. However, the ATW system offers a very compact solution, with no needs for transportation between the reprocessing, refabrication and/or long term actinide waste storage. All steps could be performed on one site. With its full development the thorium based fuels cycle can offer the solution where nothing need leave the site except the generated electric power. Only truckload of thorium fuel and spent fuel from the existing LWRs need be delivered to the site.

III. Transformation of the solid nuclear waste into a form suitable for transmutation

This includes separation of short lived and stable products from long lived ones and thereafter getting the material in a slurry (for the aqueous system) or in a molten salt liquid (for the molten salt system). The possible risks here are partly the same as those associated with the separation in the transmutation loop. Due to the lack of experiences in this field in Sweden, an active international cooperation is needed to exchange the experiences with USA, France, Japan and Russia. Those countries have had very active partitioning-transmutation R&D programs. Special attention has to be paid to material flows in the system to minimize the generation of the low activity material streams. The chemistry has to be optimized with respect to high separation ratio and lowest possible material streams.

IV. Accelerator

The accelerator - hundreds of meters long - will be placed, as well as the whole spallator, under the ground. Under normal conditions no radiation can get out of the accelerator. An unstable proton beam might cause damage to the surrounding material which in turn could result in some radiation, primarily neutrons spallated by protons hitting the constructional materials. Furthermore, high vacuum in the accelerator may suck in some radioactive material from the spallator into the accelerator. In such a case the accelerator can be stopped at once and we should expect only minor induced

because of the possible thermal shocks in the target-blanket-core system.

The construction of the accelerator has to be reliably enough to provide the beam availability well over 70% including planned beam-off time.

V. Beam Entry Window

The beam entry window is the part of the system which is heavily exposed to the radiation, high energy proton/deuteron beam together with the flux of scattered neutrons will induce heavy radiation damages. An important function of the beam entry window, except for transmitting accelerator beam, is the separation between vacuum and high/normal pressure regions. Therefore the beam entry window is the weakest interface of the system. The window has to be exchanged every year, existing experimental data indicate the life-time hardly longer than 1 year, see Figure 18. Damages might occur between the replacements. Mainly risks for the repairment personnel are foreseen.

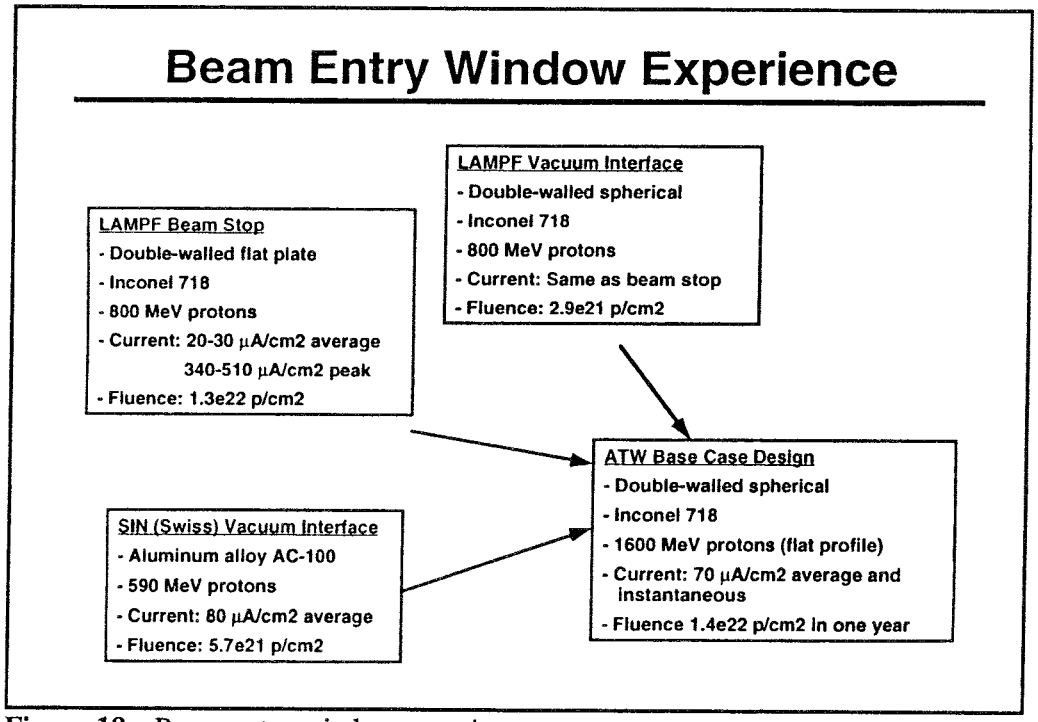

Figure 18 Beam entry window experience

VI Spallation Target

We shall separate into three main cases:

Solid target: Tungsten.
Here we have a horizontal geometry - straight proton beam. Then we don't need to bend the beam and thus generation of the extra radiation on the bender can be avoided. The distribution of the spallated neutrons can be kept relatively flat due to the duct design, see Figure 7. Because of the solid form of the target there is a possibility of severe radiation damages in the proton beam. Further we get cooling problems in the solid target and risk for water leakage and explosive vapours-D_2O buildup in contact with the hot tungsten.

Fluid target: Lead.

To provide the convection cooling in fluid target we must have a vertical geometry with accompanying problems for the bender. Further some activation problems i.e. buildup of long-lived isotopes (^{214}Po), see Figure 19. We have efficient heat transfer but complicated pumping system. There is some risk for explosive interaction between high temperature lead and the closest environment.

Fluid target: liquid lithium or lithiumfluorid.
Vertical geometry - see above.
Risk of fire in case of leakage.
Chemical problems - compatibility graphite and the target material, aggressive chemistry of liquid lithium.
Efficient heat transfer - the same medium is used st the target medium .
This target produces a very small amount of heavy spallation products (see Figure 19).
Radiation damage in target container material is one of the most challenging problems of ATW. Frequent replacement of the target container may be required during the plant life-time, but it appears to be difficult to design a replaceable container. Material problems of the target are discussed more detailed later in the Material Problems section 5.

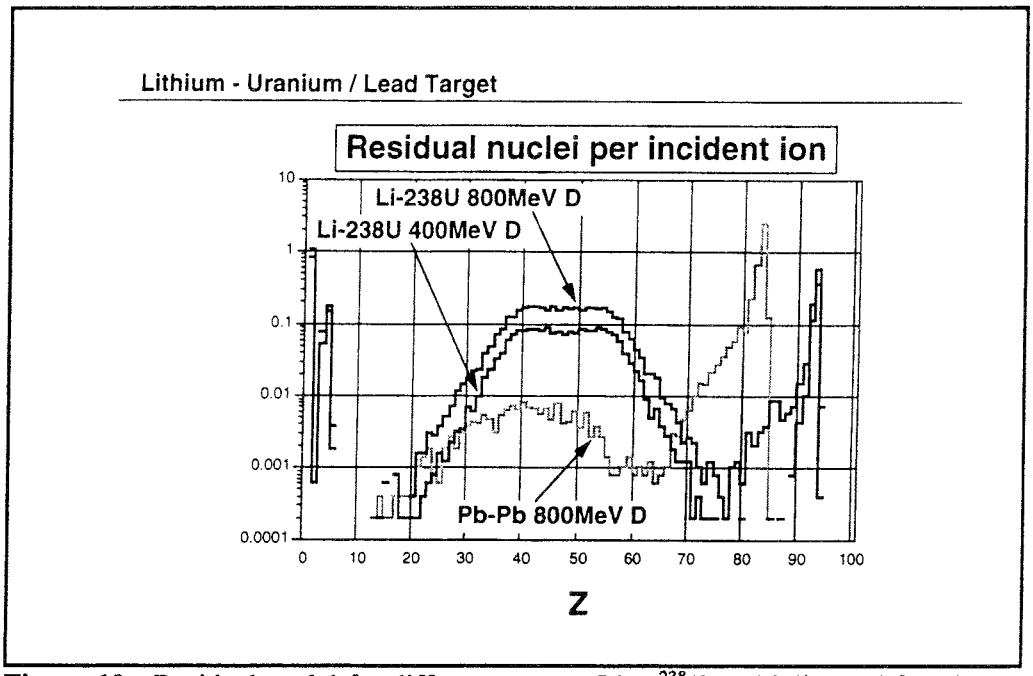

Figure 19 Residual nuclei for different targets: Li + ^{238}U multiplier and for Pb target. Two different energy of the deuteron beam: 400 and 800 MeV.

VII. Blanket/core system safety

The operation is sub-critical which eliminates the possibility of a critical accident. Furthermore, due to the very high transmutation rate, we have a low actinide inventory which diminish consequences of a leakage. The placement of the system under the ground also limits probable radioactive leakage to the environment.

The initiating event for most possible risks is material defects caused by stress under difficult conditions- high temperature, radiation, aggressive radioactive material in particular the molten salt case. See her next section - Material Problems.

5. MATERIAL PROBLEMS

5.1 THE NEUTRON ENVIRONMENT IN THE ATW TARGET FACILITY

A diagrammatic representation of the neutron environment in the ATW target facility

is shown in Figure 20. It should be noted that this figure is not a neutron spectrum in the conventional sense but rather a representation of the neutron flux above the value of the energy on the horizontal axis.

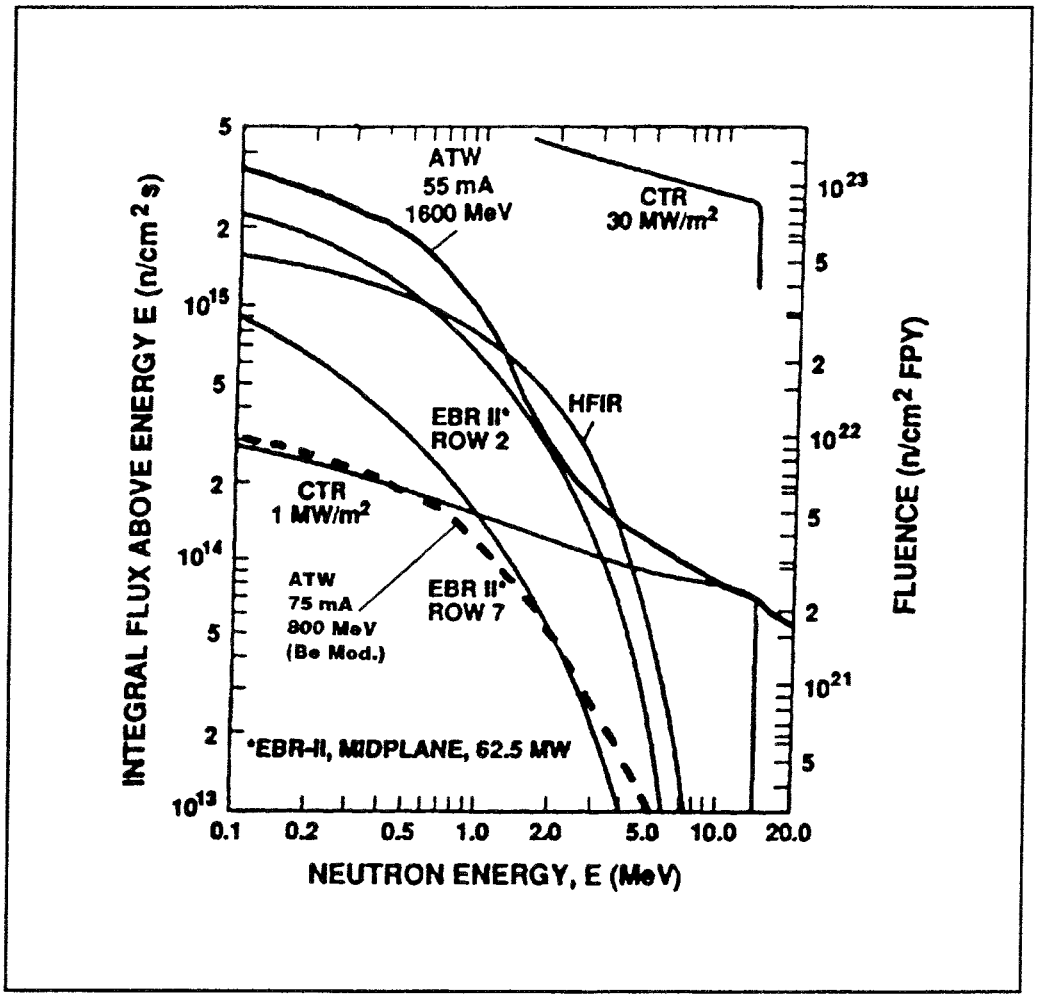

Figure 20 The neutron flux in a number of irradiation facilities given as the integrated flux above the given energy.

A comparison is made with EBR II, a sodium cooled fast reactor, and with HFIR, a water cooled high flux reactor. These are existing reactors in contrast to the fusion reactors labeled CTR (CTR label is identified with lowest contemplate wall loadings) in the Figure 20. With regard to the CTR:s it can be noted that a wall loading of about 5 MW/m^2 is a level which currently is considered as realistic for a fusion reactor [29]. Thus the ATW facility is subjected to a neutron flux which lies in the same range as existing fission reactors or projected fusion reactors.

However from a materials point of view there is a significant difference between the

fast reactor environment and the ATW environment and that is the large flux of high energy neutrons in the ATW facility ranging to the neutron energy up to 600-700 MeV - Figure 21.

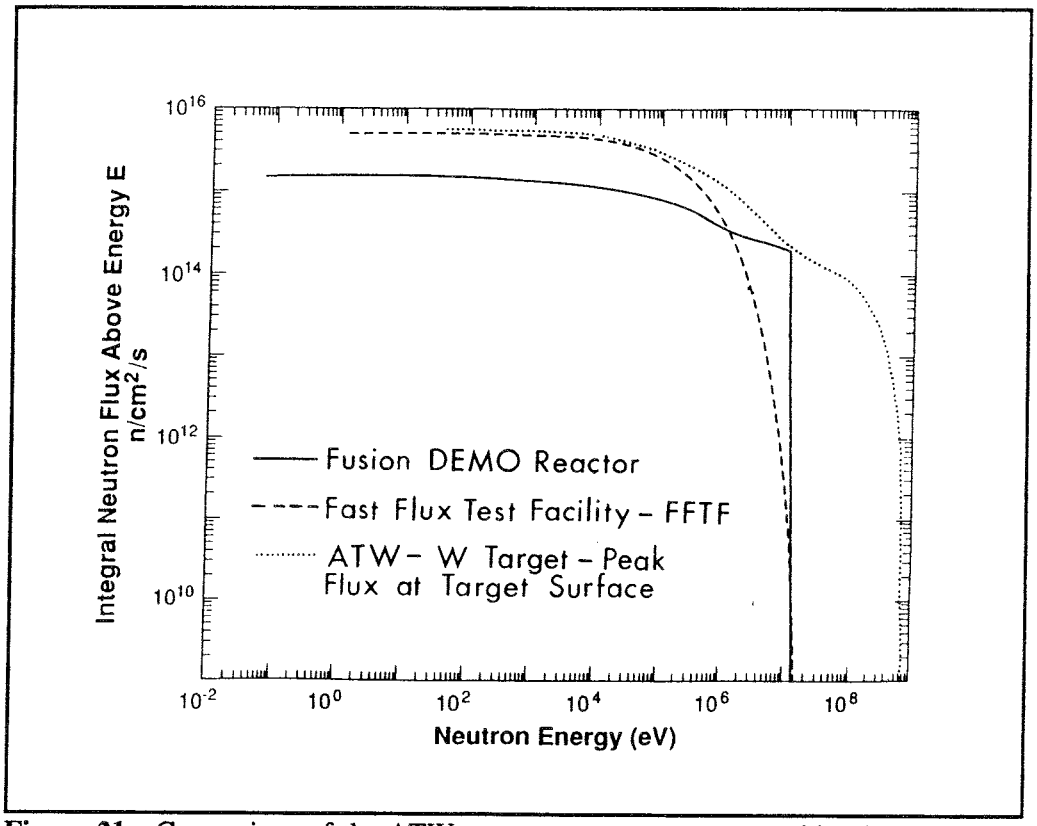

Figure 21 Comparison of the ATW neutron energy spectrum with other neutron facilities.

In addition to the larger production of displacements per neutron, the high energy neutrons will cause (n,p) and (p,α) reactions in the material, reactions which are almost absent in the fast reactor but which will produce significant concentrations of hydrogen and helium in the structural materials of an ATW facility. The effects of particularly the helium on the properties of irradiated structural material have been the subject of considerable attention in the development of fusion reactors and is still considered to be one of the major uncertainties in the development of fusion reactor technology. The reason for this uncertainty is quite simply that there still does not exist any irradiation facility where structural materials can be irradiated and tested with high energy neutrons in sufficient numbers to produce neutron doses of relevance to a fusion reactor. In fact it has been proposed to use a spallation source as a fusion materials irradiation facility, but the cost is so high that no single country or group of countries

are ready to spend the required amount of money on such a source.

5.2 NEUTRON IRRADIATION EFFECTS IN STRUCTURAL MATERIALS

The spectrum in Figure 20 can be used for obtaining an estimate of the neutron irradiation damage in the structural materials of the ATW target facility. The displacement damage rate can be estimated to be $2.6 \cdot 10^{-6}$ dpa/s in steels (dpa = displacements per atom) [30]. This will give 80 dpa in a year. For the other material proposed, Zircaloy 2, the displacement rate is expected to be about the same as that in the steel.

In a fusion reactor helium and hydrogen are produced in rates of about 14 and 40 appm per dpa respectively. In the ATW facility about 80% of the displacement damage is produced by neutrons with energies below 5 MeV which is the approximate threshold for significant gas production [1]. Thus the gas production should be of relatively smaller concern in the ATW than in the fusion reactor but a larger concern than it is in the fast reactor. It is certainly so high that high temperatures of the structural material must be avoided, otherwise low ductility creep failure will be a concern. A safe limit for steel would be about 450 C. This would also limit the possibility of swelling. For Zircaloy there is no information available on helium effects on low ductility creep failure. With regard to the hydrogen produced in the Zircaloy it will eventually form hydrides. However the amounts of hydrogen produced ($\approx$100 wtppm in 30 years) are far below levels which will be of any concern with regard to ductility. Any hydrogen produced in steel will escape by diffusion.

If swelling and creep failure is avoided by limiting the temperatures the remaining irradiation effect is the change in tensile properties and fracture properties. For the proposed martensitic steel HT9 data exist for irradiations up to about 200 dpa [31] in fast reactors. However the irradiation temperature for which HT9-steel data exist are higher than in the ATW facility. It can thus be expected that irradiation embrittlement effects will be higher in the latter facility.

It should be noted that for Zircaloy virtually no data exist for irradiations over about

30 dpa. It is thus largely unknown what changes will take place in the material if it is used in the ATW facility even for a one year exposure which will give about 80 dpa. It is known that mechanical properties tend to reach saturation values with increasing neutron dose. It is however quite uncertain whether true saturation has actually been reached in any of the values reported in the literature.

Another concern with Zircaloy is its dimensional stability. It is well known that zirconium components change shape in a neutron flux. Typically a tube or a plate will grow in the longitudinal direction and shrink in the thickness direction. For a fuel cladding tube the growth is of the order of a few tenths of a percent during a life time exposure, about 15 dpa. Thus significant shape changes should be expected in the ATW facility. It should be noted that the growth does not saturate with fluence. The long term behaviour is that it is directly proportional to fluence.

5.3 RADIATION DAMAGE OF TARGET STRUCTURAL MATERIALS FOR FLOWING LEAD TARGET [23]

The flowing lead is surrounded by a beryllium sleeve and probably by other structural materials outside the beryllium. These structural materials will be damaged to some degree by the neutron flux and it is useful to estimate lifetime of these components of the system. The beryllium performs the function of moderating the spallation neutron spectrum to reduce neutron damage to the other materials farther outside in addition to being a conduit for the lead and serving as a neutron multiplier. The radiation damage rate in the beryllium is expected to require frequent replacement of the beryllium and the target must be designed to allow fast replacement of this component to avoid long down-time for the system. The radiation damage to the beryllium from neutrons can come from displacement damage by neutrons with energies above about 100 eV and from gas production for neutrons above the (n,2n) threshold of about 1.7 MeV. The displacement damage is annealed away for the operating temperatures of about 500° and the primary damage is from the gas production mechanism. For the fluence of 10^{22} n/cm^2 with energies greater than 1 MeV beryllium samples were tested for compressive yield strength and failure strength without significant deterioration in performance. The beryllium should maintain its integrity for at least 10^8 seconds (3 years). During this period the radial swelling would be less than 1 mm. The axial

swelling would be about 1.2 cm. The radial swelling could be accommodated without significant influence on the target design. The axial growth would require more attention. However, since the beryllium is not a structural member of the target, this degree of material swelling should be readily accommodated.

The radiation damage of the graphite should be mostly due to displacement damage since the threshold for gas producing reaction is much higher than that for beryllium. Graphite is known to shrink first under radiation damage and than to grow. A thorough examination of the performance of graphite under radiation is necessary but from already existing data one can estimate that life-time of the graphite structure would be of the order of 20-30 years. It has been concluded that a major materials development program probably will not be required for the spallation target for ATW [23].

5.4 OTHER POTENTIAL MATERIALS PROBLEMS

In the proposed ATW facility it is proposed that the chambers with molten lead and molten salt, are designed with a double wall system such that the liquids are cooled at the walls, so that a layer of solid product form at the wall. In that way direct contact between the structural materials and liquid will be avoided and the possibility of corrosive attack minimized.

It is perhaps obvious to even the uninitiated reader that molten salt can be very reactive with a structural metal and it is thus imperative that this design solution actually will work. It is perhaps less well known that molten metal can embrittle structural metals. It is thus equally important that the layer of solid lead in the lead chamber can actually be formed. Both lead and mercury are potent embrittlers and lead can in fact have an embrittling effect even in the solid state on alloy steels [32]. Zircaloy is very brittle in the presence of mercury. Whether or not the small amounts of mercury present (0.1% ?) will have an effect is still unclear. It is well known that small amounts of Cd dissolved in liquid Cs have a strong embrittling effect on Zircaloy and that the effect is more pronounced in irradiated material [33]. It can be noted that the solid lead will have a mercury content of about half that of the liquid lead [34]. Whether or not such small contents have an effect will have to be tested before a commitment is made to the use of a zirconium alloy as a structural material for ATW facility.

5.5 REMARKS

The total neutron doses for the structural materials of the ATW target facility are such that no data exist for any of the proposed materials. It is thus possible that these materials will have to be changed several times during the life time of the facility. This will require special design solutions so that the work can be performed without to much radiation exposure to personnel. Another consequence is that frequent changes of material result in an increase in the amount of radioactive waste.

The major unknowns with regard to radiation effects in the materials are the effect of very high neutron doses on the mechanical properties and dimensional stability of Zircaloy. It will also be necessary to investigate the effects of helium on the creep failure of Zircaloy if the material is to be used at temperatures above about 250 C. For HT9 the effects of radiation are known up to higher doses than for Zircaloy. However the exposure temperature have in those cases been higher than temperatures to be recommended for an ATW with regard to the potential helium embrittlement problems.

It will be necessary to investigate any embrittling effects on both HT9 and Zircaloy from both liquid lead and solid lead with small amounts of mercury dissolved. It is especially important that such investigations are carried out on irradiation hardened materials since these are known to often be more susceptible to embrittlement than unirradiated materials. It has already been demonstrated that HT9 is less sensitive to liquid metals than austenitic stainless steel [31].

6. CONCLUSIONS AND REMARKS

We can summarize the safety aspects of the described systems in few points

6.1 SAFETY CONSIDERATIONS FOR ATW AQUEOUS SYSTEM

- Subcritical system
- Safety by design: Design the facility to minimize the impact of potential accidents (e.g. double wall piping to contain actinides) - must be

improved and thought through.

- Passive decay heat cooling [18]
- Must consider:
  - Operational transients that have a relatively high probability of occurring such as loss of power, and small pipe breaks
  - Low probability accidents such as pipe breaks, window failure, and beam spot reduction
  - Worst case scenarios such as beam trip failure during a loss of forced flow.

To be done:

- identify and characterize potential occurrences and accidents
- characterize consequences
- identify design requirements and mitigating conditions
- identify areas/issues which require further analysis

6.2 KEY SAFETY FEATURES OF ATW ADVANCED SYSTEM

1. Subcritical system

   High thermal neutron flux produced by accelerator driven source in multiplying blanket

2. Low inventory of actinides and fission products

   High thermal neutron flux target achieved through minimization of parasitic capture and leakage

3. Large negative temperature reactivity coefficient
4. Passively safe in the event of loss of coolant accident

   Large thermal inertia and low power density

   Molten salt can be drained away

5. Proliferation resistant fuel cycle

   Uranium enrichment lower than 20%

   Thorium breeding without reprocessing

6.3 SPECIFIC RISKS AND DISADVANTAGES

Aqueous system: energy production will require thermodynamical efficiency which is possible only with high pressure of aqueous slurry. Consequence: flowing fuel under high pressure which demands an extremely safe design of the blanket/target system. A small LOCA must not lead to any severe contamination problems. Difficult to design passive safety mechanisms.

Advanced system: risk of fire with graphite moderator, composition of the fuel/blanket must be designed to keep the negative temperature reactivity coefficient

The ATW concepts are still on the very conceptual phase of the development. Some immediate experimental tasks are necessary to **pursue the safety concerns**:

- Fundamental Materials Studies
  - Neutron flux and spectrum measurements, calculations and comparisons to predictions
  - Radiation damage, transmutation product generation
  - Defect production and spectral effects experiments

- Applied Studies
  - Nuclide production and decay heat in target
  - Mechanical properties of candidate materials
  - Qualification of target, structural and containment materials under prototypic conditions
  - Verification of thermal-hydraulic design
  - Radiolysis of coolant/moderator
  - Erosion/corrosion and material compatibility verification
  - Feed stream purity requirements
  - Transmutation product disposal

- Advanced Material Development
  - Composite such as Carbon-Carbon
  - Metal matrix such as Aluminum-Alumina

- Optional target materials

7. FINAL CONCLUSIONS

- ATW can enhance geological repository program
- ATW would not remove need for the first geological depository but it has the potential for significantly impacting future high level waste management strategy
- ATW reduces end-of-life wastes without leaving the burden to the future generations. Figure 22 shows the impact of the waste processing on the toxicity of the wastes. The extremely long timescales of the toxicity of non-processed wastes is reduced to the "human dimension" of several centuries where engineered barrier systems could be used
- ATW can ease requirements of future geologic repositories by practically eliminating key radionuclides that are major contributors to long term risk. It reduces uncertainty in long-term risk (what does not exist cannot cause a risk):
  - reduces possible actinide transport as colloid or complexes in failure scenarios with water
  - **reduces incentives for deliberate intrusion into repository**
- In its advanced design ATW offers a very attractive, undercritical nuclear energy system with very low system inventories of radioactive isotopes
- ATW as a thorium burner offers inexhaustible energy supply
- Using breeding technique ATW cuts down demands for uranium mining and milling which reduces health related risks per GW(e) -y of produced electrical energy. As an example of the quantification of these risks Table 4 shows the potential health effects for U.S: regulatory-basis fuel cycle (with uranium recycling), with 1000-year dose commitment. From this table can be read that just uranium mining and milling stands for 70% of the potential health risks related to electrical energy production.

As mentioned above ATW-concepts are still under development so this report could review only some important aspects of the safety and prospects of the current

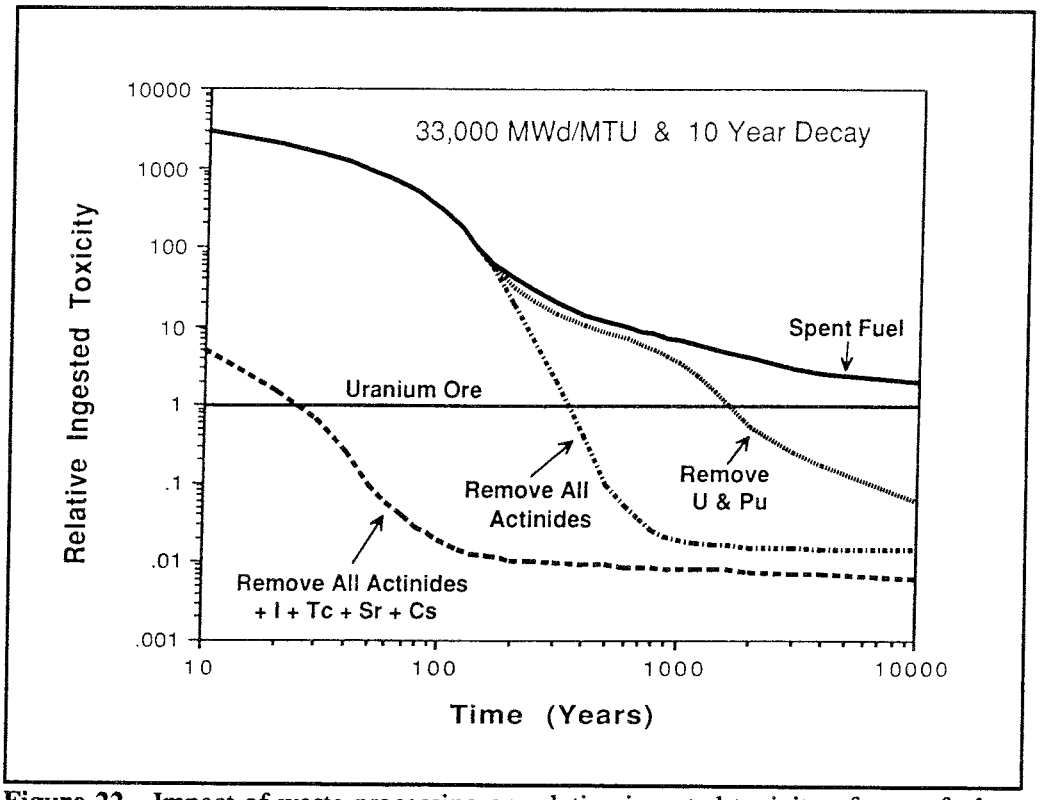

Figure 22 Impact of waste processing on relative ingested toxicity of spent fuel

projects. ATW-ideas are in continuing process of evaluation (e.g. STATS panel of the American National Research Council) and improvement. In the years to come we can expect more experimental data and new ideas which will make ATW more feasible and more acceptable way to deal with the nuclear wastes and to produce much cleaner energy. It is therefore of a great importance to follow the development of this research and to take an active part in the international cooperation to solve these difficult problems.

Table 4. POTENTIAL HEALTH EFFECTS FOR U.S. REGULATORY-BASIS FUEL CYCLE (U RECYCLE), 1000-YEAR DOSE COMMITMENT [35]

Source of risk	Total mortality per GW(e)-y	Total injury and disease per GW(e)-y
Uranium mining	0.516	13.3
Uranium milling	0.227	3.1
Conversion, enrichment	0.040	0.002
Fuel fabrication	0.027	0.21
Power generation	0.070	5.0
Reprocessing	0.057	0.21
Waste management, transportation	0.015	0.125
Catastrophic accident	0.04	0.15
TOTALS	**0.992**	**22.1**

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Ed Arthur
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Francesco Venneri
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Sekoshi Tani
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Hiroshi Hatta
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