HYBRID SYSTEMS STATE OF THE ART AND PROSPECTS

Introduction / The spallation process / Fissile material production concepts / Proposed transmutation concepts / Transmutation capability / Integral experiments / Safety analysis / Feasibility of Pu transmutation by hybrid systems / Conclusions

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1 INTRODUCTION

The present work was started in the framework of the ENEA contribution to the Nuclear Energy Agency (NEA) Task Force on Physics Aspects of Transmutation.

Taking into account the present interest in the hybrid systems technology. the paper was completed adding the initial developments concerning the systems devoted to fissile material production, together with the more recent developments regarding systems dedicated to electrical energy production.

In the last phase some parts of the original works describing the systems were included in the text.

The hybrid (accelerator-subcritical reactor) systems were thought to produce fissile material to avoid shortage before the discovery of the real amount of uranium in the world.

Later these systems were studied for transmutation of long lived waste.

At present. there are some proposals for their use as electrical energy production

Hybrid systems consist of a target (generally heavy metal) producing many neutrons from incident proton. The neutrons are introduced in a subcritical blanket.

In the present paper the main hybrid system concepts are described. The commercial introduction and the needs of experimental data to perform the projects in reasonably accurate conditions are discussed.

2. THE SPALLATION PROCESS

Nuclear spallation occurs when a heavy target nucleus is hit by a medium energy particle such as a 1 GeV proton (fig. I). Due to its high energy and correspondingly small de Broglie wavelength, this proton interacts with individual nuclei inside the nucleus, setting off a cascade of nucleon-nucleon collisions, i.e. the intranuclear cascade. In the course of this cascade, some energetic nucleons may escape from the nucleus and eventually hit another nucleus, thus giving rise to an internuclear cascade. The remaining nucleus is left behind in a highly excited state from which it returns to the ground state by "evaporating" off further nucleons. In the case of uranium, high energy fission may occur after evaporation of only a few nucleons, with further nucleons being evaporated from the fission fragments. Moreover, fast fission in ^{23}*U (and thermal fission in natural uranium) may further enhance the neutron yield, but also the heat per neutron. The neutron yield per proton as a function of neutron energy is shown in figure 2.

Spallation is thought to take place in two stages. In

RIASSUNTO I sistemi ibridi (acceleratore-reat tore sottocritico) furono pensati per produrre materiale fissile. Successivamente questi sistemi furono studiati per la trasmutazione delle scorie a lunga v ita. Attualmente vi sono delle proposte per i 1 loro uso nella produzione di energ ia elettrica Vengono presentati i principal concetti di sistemi ibridi. Viene discussa l'introduzione commerc iale. la capacità di trasmutazione (compresa quella per il plutonio) e la necessità di dati sperimentali per eseguire i progetti in condizioni ragionevolmente accurate. Sono presentate anche le principali aree da inv estigare m una analisi di sicurezza.

ABSTRACT *The hybrid systems (ucc,elerator-.subcriticul reactor) we - re thought to produce fissile material. Later these systems were studied for tran smutation of long lived waste. At present, there are some proposals for their use in electric power generation.*
The main hybrid system concepts are described. The commercial introduction, the transmutation capability (for plutonium too) and the needs of experimental data to perform the projects in reasonably acc urate conditions are discuss ed. The main areas to be investigated in a safety analysis are pre sented.

the first stage (the intranuclear cascade phase), the incident proton creates a high energy particle cascade inside the nucleus. During this intranuclear cascade, some high-energy (>20 MeV) "secondary"' particles and low energy (<20 MeV) "cascade" particles escape the nucleus: after the intranuclear cascade. the nucleus is typically left in a highly excited state. In the second stage (the evaporation phase), the excited nucleus relaxes, primarily by emitting low-energy (< 20 MeV) "'evaporation" neutrons. We define low-energy "spallMien"' neutrons as the sum of the low-energy cascade and evaporation neutrons.

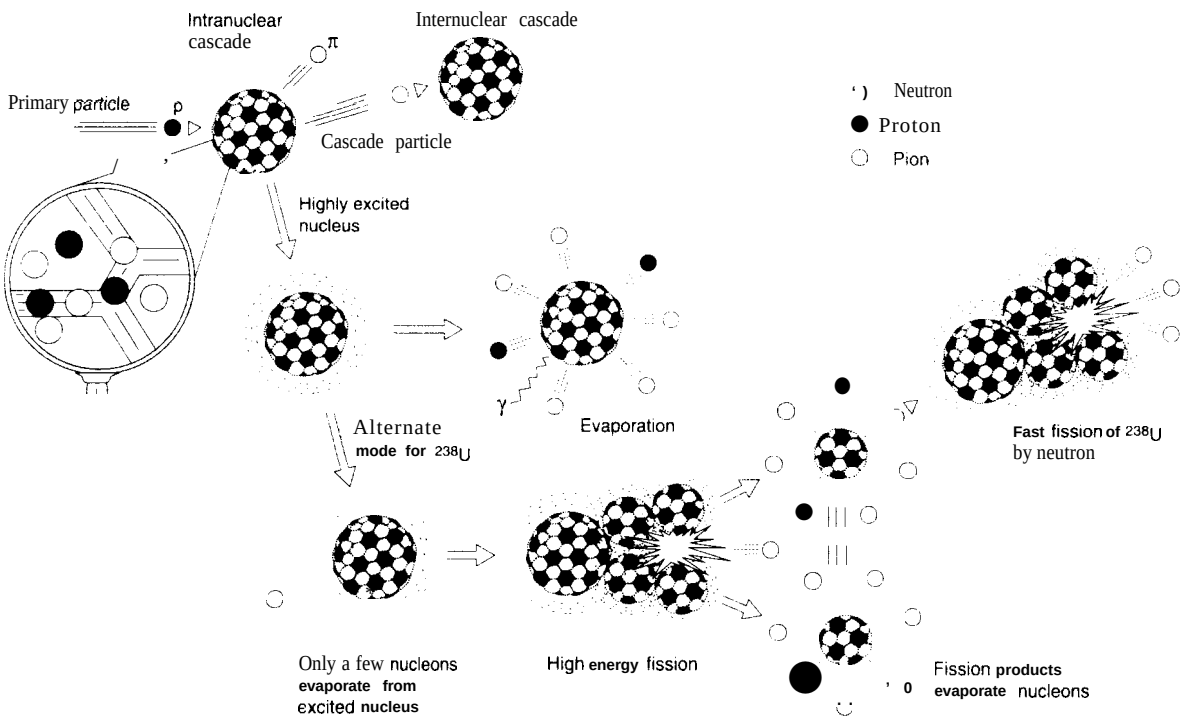
For thick targets, the high-energy secondary particles (Plus their progeny) can undergo further spallation reactions. For some target materials, low-energy-spallation neutrons can enhance neutron production through low-energy (n, xn) reactions. The total low-energy neutron production from a target is the sum of low-energy spallation neutron production plus the net production from low-energy (n, xn) reactions.

Using a code calculation as a model (which is at least approximately correct), it can be shown that more than 90% of the neutrons coming from a target of the cylindrical geometry shown, bombarded by 1 GeV protons, have energies below 20 MeV with an average of' only 4.8 MeV, and the remainder, less than 10% of the total, have energies below 400 MeV with an average of 105 MeV.

3. FISSILE MATERIAL PRODUCTION CONCEPTS

3.1. MTA project(1)

In 1947 E.O. Lawrence observed secondary neutrons from a uranium target bombarded by 90 MeV neutrons stripped from 190 MeV deuterons in the 184 inch cyclotron. B.B. Kinsey further investigated in 1948 the effect with a variety of target elements. Subsequently H. York refined the measurement techniques and measured the number of neutrons produced by direct bombardment of uranium targets with 190 MeV deuterons. York also derived an approximate analytical expression for the neutron yield (neutrons/deute-



About 5-6% of the neutrons released are cascade particles with energies up to that of the incident particle. The spectrum of the evaporation neutrons is similar to a fission spectrum

Fig. 1 – Schematic representation of the spallation process as set off by a medium energy proton striking a heavy nucleus

ron) as a function of deuteron energy for a thick uranium target.

From the beginning of these observations the programwasclassified because E. Lawrence was thinking about the possibility of using accelerator-produced neutrons for the production of ^{239}Pu and tritium using depleted uranium that was already piling up as a useless by-product of the nuclear-energy program. His purpose was to develop a process by which the USA could become independent of foreign sources of rich uranium ores in case those sources were cut off. At that time almost nothing was known about American uranium resources; it was then unknown whether uranium occurred in mineable concentrations in the USA.

Because he had a mind to a production plant as the outcome of the program, E. Lawrence. with AECL (Atomic Energy of Canada Limited) approval. sought an industrial partner to work with the Radiation Laboratory to participate in the research and development program and to take primary responsibility for the design of a production plant. the analysis of construction and operating costs and ultimately the construction and operation of the plant. Early in 1950 the AECL contracted with the California Research Corporation (now Chevron Research Corporation), a subsidiary of Standard Oil Company of California, to set up a subsidiary. the California Research and Development Company, which had to be the industrial partner in the MTA program.

Beginning in 1950 the MTA program consisted of the following elements:

1) measurements of pertinent cross-sections and the yield of neutrons from various target arrangements using primarily the 184 inch cyclotron 340 MeV proton, 190 MeV deuteron and 90 MeV neutron beams:

2) nuclear chemistry measurements of neutron capture and fission events by measurement of ^{237}Np and fission product activities in uranium foils distributed throughout uranium target assemblies;

3) theoretical investigation of nuclear excitation followed by neutron evaporation and nuclear fission, nucleon cascades by spallation nucleons and Monte Car-10 calculation of low energy neutrons produced by these processes;

4) high-current accelerator design and development: beam dynamics, Alvarez-type cavity resonant frequency and impedance calculations and measurements: intense ion-source injector development: high -frequency power amplifier tube development; construction and operation of three particles accelerators: A-12, A-54 and A - 48; electron model design and operation of a "clover leaf" cyclotron, and design studies of clover leaf and separated-beam cyclotrons for high-current acceleration of deuterons to 300 MeV:

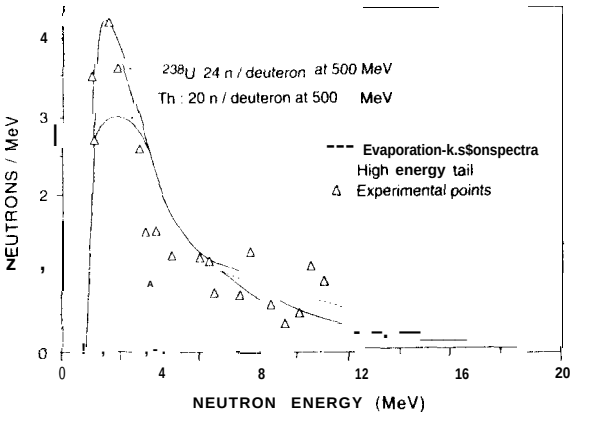


Fig. 2 – Neutron energy distribution, showing evaporati on-fission spectra for 90 MeV nuclear excitation of uranium and thorium. high-energy tail and points calculated from the measured spectrum

5) neutronics of subcritical multiplying lattice systems for ^{239}Pu and ^{233}U production and for excess power generation: subcritical, accelerator-driven electronuclear power breeder reactors;

6) detailed studies of plant design and production procedures. including chemical separation and power production, and parametric studies to optimise the economics of plant operation.

3.1.1. Accelerator design and development

Two significant conclusions were early reached in the measurement of neutron yields by bombardment of target assemblies with proton and deuteron beams from the 184 inch cyclotron. These were:

1) in direct bombardment of thick (- 30 cm) targets of uranium the yield in terms of neutrons per incident beam particle was 2570 to 30% greater for deuterons than for protons of the same energy;

2) in the case of deuteron bombardment, the insertion of a one-range thickness ("primary target") of lithium or beryllium in front of the uranium block ("secondary target") resulted in nearly the same neutron yield as bombarding the uranium directly. In this ar-Arrangement the deuterons were converted in the primary target to high-energy neutrons which were subsequently multiplied in the secondary target by a nucleon spallation cascade and nuclear excitation followed by neutron evaporation and fission of ^{238}U.

As a result of these two conclusions, accelerator design and development were directed toward the acceleration of high beam currents (≥ 0.25 A) of deuterons. Neutron yields measured for 190 MeV deuterons were projected to higher energies by a Monte Carlo type cascade calculation. From this calculation the deuteron energy for a production plant was initially set at 350 MeV and subsequently raised to 500 MeV.

In parallel with accelerator development, some alternative neutron-producing beam targets and surrounding assemblies for production of ^{239}Pu and ^{233}U were studied and evaluated economically by California Research and Development Company. These solutions ineluded subcritical, moderated lattice systems having a neutron multiplication factor (k_{eff}) ≤ 0.9 capable of both breeding plutonium and producing several times the power necessary to operate the accelerator, systems called accelerator-driven, subcritical power breeders. The final version of a production plant was designed to produce 564 kg/y of ^{239}Pu. The accelerator is a 50 MHz, 500 MeV, 320 mA Alvarez-type machine with a NaK-cooled beryllium-primary target, a NaK-cooled depleted uranium secondary target for neutron multiplication and a water cooled lattice with depleted uranium to absorb the neutrons.

The design of these systems may be such as to maximise ^{239}Pu production or to sacrifice some plutonium production in favour of greater power output. The C-50 design optimises ^{239}Pu production.

Table 2 of the original paper (1) includes estimates of the total capital investment, capital charge and operating costs from which the cost of the product is calculated as 124 \$/g of ^{239}Pu.

The MTA program was technically successful in the sense that the feasibility of converting fertile (depleted uranium or thorium) to fissile material (^{239}Pu or ^{233}U) was established, although considerable more work would have been required to arrive at an optimum design of a production facility.

3.2. *Canadian accelerator breeder system development (2)*

Assessments of a Fusion Breeder (FB) and an Accelerator Breeder (AB) indicated that both could produce fissile material at a cost, in 1981 Canadian dollars, that was three to four times the assumed 48\$ per fissile gram cost of ^{235}U. Fissile fuel costing included the revenues received from the sale of surplus electrical power generated by the breeder. Capital estimates for both methods were roughly 1500 M\$ per Mg/y fissile production rate. The fuel production rate of a FB was 2-3 Mg/y compared to an AB at 1 Mg/y. The FB produced about 1 GWe of electricity whereas the AB produced 0.1 GWe; power that would have to be fed to the electrical grid. Availability for a fuel breeder has not to be as good as that for nuclear power stations whose primary function is to generate electricity. Although the breeder generates electricity as a secondary function, its main purpose (producing fuel for power stations) is one stage removed from the generating process.

Advantages for an AB relative to a FB are that scientific feasibility has been proven and engineering problems appear to be easier. Technology associated with the blanket and target may be as complex as for a FB fertile -fissile blanket. No tritium breading blanket is required, however. The reference design for an AB is smaller and can be reached by well-defined steps in an organised and logically staged development program. Each stage ends with a unique research and development facility and presents an opportunity for program reassessment. For these reasons and because of the accelerator technological base at Chalk River Nuclear Laboratories (CRNL), we will pursue the first stage of an AB program while maintaining an active watch on developments related to a FB in the world.

3.2.1. Staged accelerator breeder development

An accelerator breeder producing 1 Mg/y as a reference size has a 500 m LINear ACcelerator (LINAC) that delivers 300 mA of 1 000 MeV protons to a suitable target/blanket. Higher production rates could be achieved by raising the output proton beam energy, with an associated increase in the length of the accelerator. For example. a 2000 MeV LINAC producing twice as much fissile fuel would be 1 km in length. The estimated 1500" M\$ cost for a 1000 MeV AB would represent a modest increase (2 m\$/kWh) to the 26 m\$/kWh charged today in the province of Ontario (an 8% increase in the price of electricity).

3.2.2. First stage

The first stage of the program is a 300 mA, 10 MeV proton LINAC, ZEBRA (Zero Energy BReeder Accelerator), that will test all aspects of launching the full current beam for an AB. The energy is high enough. without being overly expensive, to study beam transport, high beam loading, multiple tank operation, higher order mode excitation, frequency multiplying between different structures, high radio frequency (rf) power control and transmission, accelerator control, support systems. remote handling, accelerator turn-on, beam diagnostics, engineering techniques, emittance growth. shielding and reliability. Beam testing will be

important for the design of following stages and for checking computer codes used to predict beam dynamics. Performance of higher energy portions of an AB will depend on how well the bunched beam was formed and accelerated in the first part of the accelerator.

The 75 keV ZEBRA proton injector has complete current variability from zero to the full 375 mA needed for injection into a 4 m long Radio Frequency Quadrupole (RFQ) operating at 108 MHz with 1.5 MW of rf power. Lost beam in the RFQ represents the largest gas load for the vacuum pumps and must be considered in the design. The 2 MeV beam from the RFQ is further accelerated to 10 MeV in 4 m of post-coupler stabilised Draft-Tube Linac (DTL) operating at 216 MHz with 3.5 MW of rf power. The 3 MW proton beam from this 10 m LINAC will be transported to a carbon beam dump or to a beam analysing transport line. In addition, the beam, initially at reduced power levels, can be used to study characteristics of heavy metal targets in a liquid metal facility adjacent to the LINAC. The full 3 MW beam could deposit -40 kW/cm^2 on the surface of a liquid metal target.

Plans are now underway to build this 15 M$ LINAC, ZEBRA, in a new AECL laboratory to be located in Quebec.

3.2.3. Second stage

The remaining stages of the AB program would be built at a nuclear park using the team assembled at the new laboratory to oversee design and construction. Although divided into three stages the program has just one facility that is added to and upgraded following overall program reassessments. The second stage, EMTF (Electronuclear Materials Test Facility), should be completed by the mid 2000's at an estimated cost of roughly 75 M$ only for the 100 m LINAC and the related facilities. Beam from the 70 mA, 200 MeV LINAC will be delivered to a Pb-Bi target that will be used as an experimental facility providing neutron fluxes of 10^{15} n/cm's for materials and fundamental research. Reasons for the choice of this energy and current are given elsewhere. The output energy of 200 MeV represents the point at which a change would occur to a more efficient 432 MHz accelerating structure for the higher part of energy of an AB.

Beam from the LINAC could be shared to investigate accelerator characteristics of Coupled Cavity LINACS (CCL) to be used above 200 MeV. EMTF will be designed so that it could be upgraded to accelerate 300 mA of protons by the addition of extra rf stations to the installed 30 MW.

A third beam usage area for EMTF is a target/blanket development area with a 14 MW target, the design of which will be based on experiments with the 3 MW target on ZEBRA. Although the average neutron energy will be I/2 and the neutrons per proton (1.5 n/p) will be thirteen times less than that of a full AB, the power density in a fuel or blanket element in close proximity to the target will be similar to that of an AB (- 0.2 MW/L).

The reduced EMTF current, 70 mA, is adequate to study most aspects of the DTL portion of an AB. Comparison with full 300 mA operation of ZEBRA will provide the necessary information to design the last stage of the AB program.

3.2.4. Third stage

The third stage, to be completed in the mid 201 O's, is a pilot facility that will test all aspects of an AB at as low a power level as possible to still provide an acceptable engineering data base that can be used to design and construct a full power AB target/blanket. A 70 MW beam at 1000 MeV into a 150 MWe target/blanket required for this stage has fortuitously the same current as EMTF. The EMTF Pb-Bi target facility at the 200 MeV location will continue to operate on a shared beam basis with the pilot target/blanket or 70 MW beam dump. Upgrading EMTF to 1000 MeV is expected to cost approximately an additional 500 M$ including the target/blanket.

A large capital investment in the 140 MW of rf systems means that high efficiency, long lifetime, reliable, and economic rf tubes will be required. Development work on tubes and associated operating systems should be possible for this future need and that of the last stage.

3.2.5. Fourth stage

The fourth and final stage consists of adding more rf power stations to the LINAC and replacing the pilot target/blanket with one designed for 300 MW of beam power. Enough electricity could be generated by the turbine generators attached to the target/blanket to meet the needs of the accelerator and deliver 110 MWe to the electrical grid. Total cost of an AB is estimated at roughly 1500 M$ and completion is expected in the mid 2020's. The design of the LINAC and other components for this facility depends critically on ZEBRA and the experiments and developments that follow the first stage of the development program.

3.2.6. Target / blanket

"'Scoping" studies, of various blanket materials in a target/blanket arrangement that excludes engineering

complexities. were used to give simplified results for a 300 mA, 1000 MeV input proton beam. A Liquid Metal Fast Breeder Reactor (LMFBR) lattice, simulated by 50% fuel, 25% iron sheath and 25% sodium coolant, was used except for one set of molten salt cases and one set of cases that mocked a CANDU style lattice. The 100 cm radius cavity was necessary to power densities in the blanket to 0.3 MW/L when enrichment percentage in the blanket reached 2% levels. At 2% blanket enrichment, net fissile production rates for ^{233}U from thorium metal, thorium carbide. thorium oxide, thorium oxide CANDU lattice with either H_20 or D_20 coolant, and molten thorium salt were similar at - 2 kg per day.

Net production rates of ^{239}Pu at 2% blanket enrichment from uranium metal, uranium carbide, uranium oxide, uranium oxide CANDU lattice and molten uranium salt were 3.6, 3.1, 2.5, 2.2 and 1.8 kg per day, respectively. Fissile fuel production rates calculated for different percentage enrichments were used to determine fuel costs for different proton energies and currents. The calculations for production of ^{233}U and ^{239}Pu were made with the assumptions of an 11% capital charge rate, 17 $/g reprocessing charges, a facility with 80% availability, and a target/blanket that generates sufficient electricity to meet the needs of the accelerator.

Production rate increases as beam current increases and costs begin to reach an equilibrium level near 4 kg/d for ^{239}Pu and 7 kg/d for ^{233}U. These results suggest that an AB for ^{233}U production should have a higher beam power (factor of 2.5) than that for a ^{239}Pu production facility to minimise costs as much as possible.

3.3. *THORIum Molten-Salt Nuclear Energy Synergetics (THORIMS-NES) (3)*

In fission energy program, the following points should be considered for the radiowaste minimisation:

I – minimisation of nuclear chemical productions such as fission and spallation products, and trans-uranium elements: a higher thermal efficiency in electric generation will contribute to minimise Fission Product (FP) production. Thorium fuel cycle will have a big advantage to minimise or eliminate trans-uranium elements:

II – minimisation of spent fuel materials: all irradiated fuel materials should be fully used as far as possible and not be easily sent to graveyards. They should be fully burned out establishing a complete breeding fuel cycle;

111 – minimisation of fuel reprocessing steps and of reactor maintenance works.

3.3.1. Significant aspects of molten-salt reactor technology

A. Molten fluoride fuel concept

Molten-Salt Reactor (MSR, fig. 3) is classified to fast, epithermal and thermal reactors according to the kinds of neutron spectrum utilised. Fast or epithermal reactors not having some solid moderators might have more attractive due to the simple structure and high performance. However, it seems that no one has succeeded to present any practical designs yet. Fluoride salt fuelled thermal MSR would be the most promising reactor concept. The "Flibe ($LiF-BeF_2$) base molten-salt fuel" concept already established its sound technological basis by the brilliant long effort of Oak Ridge National Laboratory (ORNL), throughout 1947-1976. It depends on the successful operation of MSRE (Molten-Salt Reactor Experiment, 7.5 MWt) in 1965-1969, and on intensive R & D works on MSBR (Molten-Salt Breeder Reactor) (1963-1976). The excellent final summary on ORNL works could be found in ORNL/TM-7207 report. The short explanation will be given below.

However, the practical deployment of MSBR is not easy because of the following difficulties:

- necessity of core-graphite exchange in every 4 years,
- development of continuous chemical process in situ,
- necessity of improving doubling-time from the present estimation of 20 years,
- flexibility in power size.

Now, the principle 111 in previous paragraph should be accepted.

B. Flibe-base molten-salts

The molten $^{7}LiF-BeF_2-^{232}ThF-^{233}UF$ system will be one of the better and idealistic nuclear materials as a carrier of nuclear synergism due to the following characteristics:

1) a triple-functional medium working as:
   - nuclear reaction medium-fuel, blanket and target,
   - heat transfer medium-coolant.
   - chemical processing medium:

2) no radiation damage as an idealistic "ionic liquid":

3) high safety and economy assurance promised by chemically inert, low vapour pressure. moderate viscosity and thermal conductivity, and high heat capacity in fairly high working temperature (770- 1070 K).

Sodium technology has three severe disadvantages such as:

· high chemical reactivity,

– high thermal shock,

– oxidised vapour condensation in gas spaces, different from molten salt technology.

Molten-salt technology is much easier than water (high pressure, corrosive and reactive with zircaloy) and Na technologies.

C. Graphite

Graphite is the best material for neutron moderator and reflector in MSR and is really compatible with molten fluorides in bare state. This is a great benefit in designing MSR. The satisfactory materials were developed by the efforts of USA and France.

D. Containment materials

Fortunately, an easy manufacturable, weld able and high temperature resistant (till ca. 1100 K) alloy com-

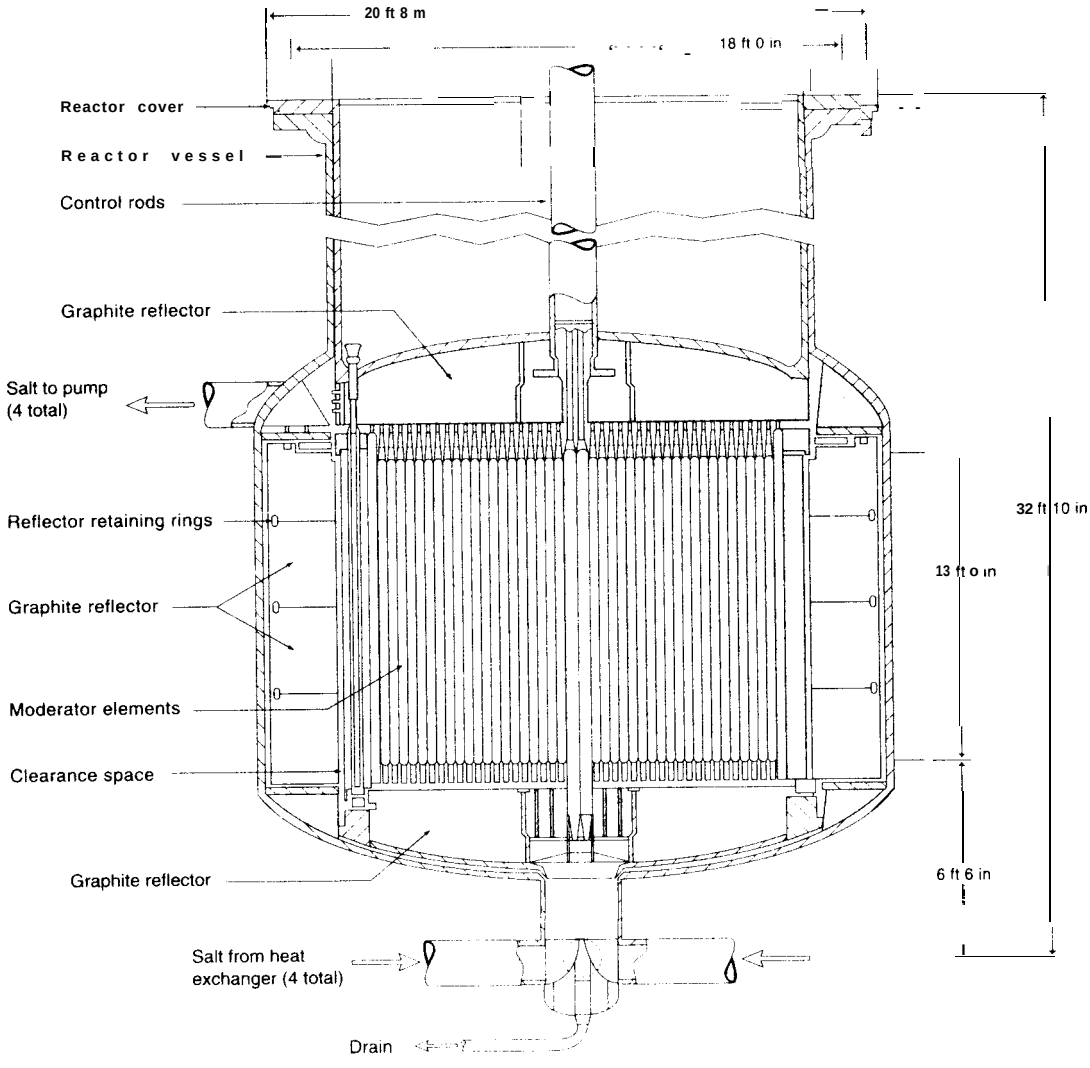


Fig. 3 – Sectional elevation of MSR vessel

patible with molten fluorides was developed basically according on the proposal of H. Inouye (ORNL). This alloy, Hastelloy N (Ni-(15-18)Mo-(6-8)Cr in w%), was modified by adding about 1 % Nb for the protection of Te attack on surface. This was experimentally supported by Kurchatov Institute.

This alloy needs not to be introduced in reactor-core region suffering severe irradiation and thermal shock. MSR-core is occupied by graphite and fuel salt only. The thinnest material will be 1/3 or 2/3 in diameter tube in the heat exchanger. Therefore, the reactor design would be very simple and easy. In practice, the corrosion of Hastelloy N by introduced contaminants such as air and moisture was surprisingly small and negligible even in the small experimental reactor MSRE. Commercial power stations would be much safer against corrosion.

E. Confinement of fuel salt and fission products

The triple confinement of fuel-salt is soundly established by:

- reactor vessel,

– high temperature containment, in which the atmospheric temperature is kept within about 500 "C,

- reactor container, in contrast with the solid-fuel reactors, which are also depending on the triple confinement.

However, in the latter the troublesome thin clad-tubes are placed in the high neutron flux, high flow velocity and high thermal shock region of core.

The fission product elements in fuel salt can be classified to the 4 groups concerning with their chemical behaviors:

Group (1): rare gas elements (practically no solubility in salt);

Group (2): stable salt elements such as rare earths, Zr, Ba, Sr, Cs, Br, I (no chemical problems):

Group (3): noble (insoluble) metal elements such as Mo, Nb;

Group (4): unstable salt element such as Te, O, H. D, T.

Group (1) elements are removed semi-automatically from salt. This is a great advantage for preventing their harmful neutron-absorption and their release to environment in accidents.

Group (2) elements are trapped definitely as ionic species in salt, and do not present any trouble.

Group (3) elements behave as:

- undissolved floating materials (shifting to cover gas system), and

– plate-out materials on graphite, or

– on metal surfaces.

Their mass ratios were 50- 10-40% in MSRE. Some part will be filtered out. The plate-out materials on graphite will be removed for reuse by grinding 0.5 mm in depth to diminish the quantity of radiowaste.

Group (4) elements, particularly Te, might induce a shallow surface brittleness on Hastelloy N. However, it is effectively protected by controlling electro-chemical redox-potential of fuel salt, and modifying the alloy composition by the addition of about 1 % Nb.

Tritium will be effectively transferred to coolant salt NaF-$NaBF_4$ (8-92 mol%) through the heat-exchanger tube-wall exchanging with H of water content (200 ppm) in coolant salt, and will be recovered in He cover-gas phase effectively minimizing T release to environment. MSR system only solved the problem of tritium management among all proven reactor systems.

3.3.2. Power producing reactors (MSR)

A. Design of simplified small molten-salt power station FUJI-11

Depending on the rational MSR technology above explained, the developmental program of ORNL was mostly concentrated on the large breeding reactors. Therefore, the design study of a small molten-salt power station, named as FUJI-series, was carried on Furukawa and his group.

The main design principle is:

- single fluid, multi-region type, graphite moderated core,

· no need of core-graphite exchange keeping low power density,

- no continuous chemical processing, except the simple removal of fission-gases (Kr, Xe) and tritium,

– high conversion ratio and low fissile inventory.

The purpose is the establishment, of nuclear power stations more simplified in the maintenance and operation modes and more flexible in size than MSBR.

Finally, as a standard design, the fuel self-sustaining compact MSR, 350 MWt FUJI-II was proposed. Its important parameters are shown in table 3 of ref. (3); about 90% of the volume in reactor vessels is occupied by moderator & reflector graphite.

This fuel self-sustaining character, meaning no need of fissile supply. Th-supply of 400 g/d is strictly effective for:

- minimisation of excess-reactivity,

– simplification of operation and maintenance,

– minimisation of fuel transportation and radiowaste, too.

This contributes to the improvement of nuclear-proliferation and terrorism resistance, and of coupling performance with fissile producing breeders (Molten-Salt fissile producing Breeders, MSBs), which is needed only for initial fissile supply.

B. Reactor chemistry

The behaviour of fission products accumulated in the fuel salt of FUJI-II at the final stage was analysed in detail; the final amounts are CSF = 0.06 mol%, SrF_2 = 0.05 mol%, BaF_2 = 0.005 mol%, CeF_3 = 0.17 mol% and ZrF_4 = 0.37 mol%, for example. The total amount of trivalent fluorides such as rare earth will be fairly large, but still soundly soluble in fuel salt. In conclusion, the fission products need not any separation except originally designed removals of fission gases and T, and partial filtration of solidified materials floating in fuel salt.

However, some precautions should be required in the full life to keep the sound reactor chemical conditions:

– pre-operational treatments such as gassing out of graphite, and surface-cleaning of piping and components by molten LiF-BeF_2 salt;

– redox potential control of fuel salt keeping its ratio UF_3/UF_4 in 0.02 ÷ O. 17;

– fuel salt composition control.

C. Safety

The FUJI-H power station excellent safety features are significant. Entire stop Of fuel salt flow in the reactor will not result in any damage, since the core-graphite temperature is only increased up to about 1300 K. One of the most severe accident will be a leakage of fuel salt from the primary system. However, spilled salt will be caught on the catch basin and automatitally guided to the emergency tank, which is dipped in a big water pool (fuel salt is chemically inert and insoluble in water).

3.3.3. Fissile producing breeders (MSB)

A. Proposal of several MSBS

For the next century, K. Furukawa proposed the following three types of Molten-Salt fissile producing Breeders (MSBs):

– Accelerator Molten-Salt Breeder (AMSB): by the neutrons generated from spallation reaction of Th nuclei in molten salt with 1 GeV protons;

– Impact Fusion Molten-Salt Breeder (IFMSB): by the application of new ideas of axially-symmetric mass-driver and shaped projectile accommodating DT-pellet;

– Inertial-confined fusion Hybrid Molten Salt Breeder (IHMSB): by the adoption of the first wall of molten-salt waterfall for the elimination of radiation damage.

These systems are designed to guarantee the straight supply of the fuel salt concentrated to 0.5-0.7 mol% in $^{233}UF_4$ content, which can be sent directly to the above molten-salt fission power stations fuelled in lower concentration (0.2-0.3 mol%). They should be developed within 2010-2030".

3.3.4. Accelerator Molten Salt Breeder (AMSB)

A. Technical problems in accelerator breeders (or spallators)

The physical principle of spallators was clarified by the successful material test accelerator project (1949-54) at Lawrence Radiation Laboratory, and by the intense basic researches in Chalk River Nuclear Laboratories (1952-84), Brookhaven National Laboratory, Kurchatov Institute of Atomic Energy, etc. and also in the development of molten-salt technology by ORNL.

The spallation reaction of heavy nuclei with 0.8-2 GeV proton will be one of the most effective methods for production of neutron, which is useful for fissile material production.

The technical problems in accelerator breeders are the developments of high current proton accelerator and target/blanket system. In the design there are problems such as:

– radiation damage

– heat removal,

– irradiation material shuffling and

– spallation chemistry of target/blanket.

Almost all would become simpler by the application of the single-fluid type molten-salt target/blanket concept. The reasons are the followings:

1) Th (or U) is chosen as a target nucleus, and single-phase molten fluoride system including ThF_4 (or $^{238}UF_4$) will serve as chemically inert target and blanket medium;

2) molten-salt target/blanket system promises simple and safe configuration, no radiation damage, easy heat removal, automatic material shuffling and easy the chemical processing;

3) the elimination of beam window will be possible due to the low vapour pressure of salts;

4) the degradation of neutron yield in the liquid metal target will be recovered by elimination of window and structural materials inside the target/blanket phase and by the inclusion of Be nuclei due to the BeF_2 component addition.

often liquid Bi, Pb or their alloys are chosen as target materials. However, they have several disadvantages as follows:

– container materials: applicable W, Mo graphite or carbon-steel are all not suitable for sound construction of nuclear facilities;

-- high density and high pumping power;

– complex chemical behaviour of spallation products such as Po vapour problem, formation of insoluble metallic or non-metallic compounds, etc. produces several troubles;

· the choice of "liquid" is not extended to fertile blanket zone, where no-w it is necessary again to consider cladding or barrier wall, and its radiation damage and neutron-loss by them, shuffling and exchange, heat removal, chemical processing, etc..

B. Design principle of AMSB

AMSB is composed of three parts: a 1 GeV-300 mA proton accelerator (LINAC etc.), a molten fluoride target/blanket system and a heat transfer and electric power recovery system. The schematic figure of its main parts is shown in figure 4. The size of target salt bath is 4.5-5 m in diameter and 7 m in depth. To keep the accelerator smaller in size, comparatively slow proton of 1 GeV was chosen and the current was specified mainly from the thermal output. The inside

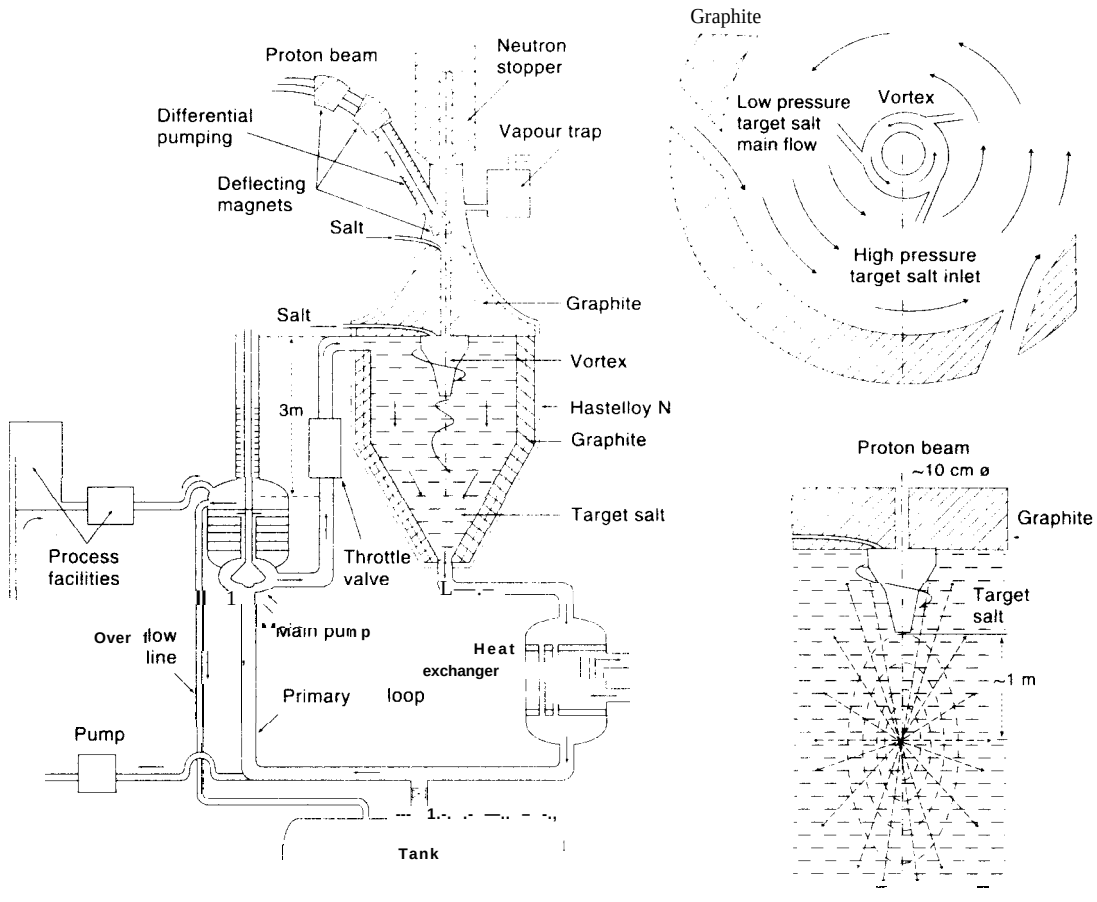


Fig. 4- Schematic figure of molten-salt target/blanket system of Accelerator- Molten-Salt Breeder (AMSB)

of the breeder vessel made of Hastelloy N (Ni-Mo-Cr alloy) is covered by thick graphite blocks immersed in salt. The target salt is introduced from the upper part forming a vortex of salt of about I m in depth. The proton beam is directly injected in off-centred position near the vortex bottom, reducing the neutron leakage and improving the heat dissipation.

This target/blanket system is subcritical, and not influenced by radiation damage. The heat removal could be managed by the dynamic salt, which is diluted in heavy isotopes. The shuffling is automatic. Except unknown engineering about a beam injection port, this simple configuration will be manageable in engineeting, depending on MSR technology in general.

C. Selection of molten-salt compositions

Considering the phase diagrams of tertiary salts ineluding U and Th fluorides, several candidate salt compositions are proposed. Now, the composition of

^{7}LiF-BeF_2-ThF_4 (64-18-18 mol%)

was chosen as a standard target/blanket salt.

The neutron yield will be improved by the addition of fissile nuclei to target/blanket system. By the calculation of Nakahara it was verified that the replacement of 3 a% of Th atoms in fluoride salts into fissile atoms may induce about an increase of 50% in neutron yield. Therefore, neutron yield of standard type AMSB will be increased by about 45% in fissile material production by the replacement of about 0.5 mol % of $^{233}UF_4$ into ThF_4 (corresponding to 2.8 a% in Th), that is:

^{7}LiF-BeF_2-ThF_4-$^{233}UF_4$ (64- 18-17 .5-0.5 mol%).

This type of AMSB using fissile added salts was named as the high-gain type AMSB, which is effective for the improvement of electric power recovery by an increased heat generation.

D. Nuclear and reactor-chemical aspects of AMSB

The absolute amount of spallation products is smaller than the fission products in reactors. The radiowaste concentration ratios in molten salts between AMSBS and MSBR of ORNL are only 1/14 for standard AMSB, and 1/6 even for high-gain AMSB.

The behaviour of produced elements was examined in the preliminary but considerable level in the similar manner as par. 3.3.1. We can understand that almost all the products would be manageable except oxygen, which may be gettered by Ti-metal hot trap.

3.3.5. Establishment of Th-^{233}U fuel cycle coupling with fissile producing breeders

K. Furukawa thinks that each "breeding & chemical processing (regional) centre" settled in 10-20 sites in the world will accommodate 4-10 Molten-Salt fissile producing Breeders (MSB), two chemical processing plants and one radiowaste managing plant. These regional centres should and might be heavily safeguarded. They are very simply connected with the MSR power stations by the molten-salt fuel cycle as mentioned in the above section. Other types of fission reactors such as the present U-PU cycle solid-fuel reactors could also be supported by the fissile materials (^{233}U or ^{239}Pu) produced by MSB, if necessary or in the transient stage. Spent solid fuel will be treated by the dry processing plant applying molten fluoride technology, which is now being developed in Dimitrovgrad, USSR.

The target/blanket salts of high ^{233}U concentration such as ().5 mol% prepared in MSBS, could be directly used as a fuel salt for MSR constituting the simplest molten-salt fuel cycle.

The excess or dirty fuel salts (not so much) from MSRS will be sent back to the chemical processing plant of regional centre, where the components will be easily isolated into:

a) fissile $^{233}UF_6$,

b) radiowaste (FP) and

c) fertile salt (^{7}LiF-BeF_2-ThF_4).

The component a) can be added to the supplying fuel salts after reduction, and the component c) should be used for the preservation of increasing $^{233}UF_4$ concentration in MSB target/blanket salts, if necessary, by adding some constituents.

3.3.6. Technological feasibility in THORIMS-NES

A. Advantages

In conclusion, integrating with the above design studies, excellent results in the THORIMS-NES will be obtained. The most significant ones are:

is – practical utilisation of the rich and popular resources of thorium;

- global (environmental) safety aspects: low thermal pollution by high thermal efficiency (43-46%), no production of trans-uranium (Pu, Am, Cm) elements, minimisation of radiowaste;

al- - social safety aspects (nuclear proliferation and terrorism): practically no Pu and easy incineration of trans-uranium elements, easy safeguard by high

reactivity of ^{233}U fuel, scarce fissile fuel transportation;

- basic (technological) safety aspects: chemical inertness. low pressure. negligible excess of nuclear reactivity:

– engineering safety aspects: simplicity in configuration, operation, maintenance and fuel processing, no core melt-down and recriticality;

– breeding fuel-cycle: easy completion of simple salt cycle, effective doubling time of 5-10 years in fissile production;

– power station: no restriction in siting, flexible power size, process heat supply to ca. 1070 K.

B. Technological handicaps

From the above explanations the rational features of molten fluoride fuel concept should be clear. However, in general, there are several doubts on it. Some conjectures on their reasons will be tried in the followings, although almost all of them seem to be "aberrations", that is, no essential reasons.

i) Non-popularity of fluid-fuel reactor concepts: till now all developed reactors except MSR were solid-fuel reactors. However, nuclear reactors are "nuclear chemical-reaction facilities", which should be "chemical plants". Solid-fuel reactors do not look like them. It seems not impossible that the solid-fuel concept would be "aberration" or "cul-de-sac (blind lane)", considering the difficulty of their fuel-cycle system establishment even spending 40 years or more.

ii) Failure of the other fluid-fuel reactors: this is another problem apart from MSR. There is a lot of unsuccessful solid-fuel reactor types. However, in analogy with the other fluid-fuel reactors, many people do think that MSR also might have a difficulty in its container materials. This opinion was exaggerated by the discovery of Te-attack phenomena on Hastelloy N after dismantling of MSRE on 1970. This was solved nicely during the final R & D stage (1972-76) in ORNL. USSR research group of Kurchatov Institute reconfirmed it by getting better results. However, nobody seems to refer on it beyond 1970. (Almost all textbooks of nuclear reactors do not think better to make a new part of fluid-fuel concept due to the essential difference of design philosophy from the solid-fuel concept.)

iii) No existence of fissile isotopes in Th resource: this is the reason why the first touch for fission-energy technology was from the U-PU cycle, clearly relating with nuclear armament technology. However, for the matured fission-energy era, this might not be essential or rational as explained in par. 3.3.1. After the choice of breeding fuel cycle concept, Th is equivalent with natural U and excellent in thermal reactor performance.

iv) Significant "simplicity" in scientific and technological bases, which mean few software and hardware (few components and instrumentations).

v) Historical non-opportunity in the Seventies: the success of MSRE operation and MSBR design study was significant in the years 1968-70.

In conclusion, non-popularity of MSR would be gradually solved by the understanding of the necessity of a new rational nuclear energy, which is already starting. However, its real demonstration by a pilot-plant such as mini FUJI-II is essential.

C. Technological problems

Reactor physics – MSR has a sufficiently thermalised neutron spectrum, in which neutronics is simple except the delayed neutron problem, and a small correction of critical mass is easy by the control of salt composition. The recent intense nuclear data study will be useful. The neutronics and beat generation of a spallator (AMSB) should be more improved theoretically. However, the essential check should be done experimentally.

Reactor chemistry – The technology of chemical impurity monitoring should be reconfirmed or developeal. The data base of modified Hastelloy N should be established in high temperature. However, the most important study is the tracing of long-term behaviour (par. 3.3.1 and 3.3.4) of fission and spallation products by the experimental reactors (mini FUJI-II and AMSBE).

Reactor engineering – Construction or operation of the first experimental facilities will not have any severe difficulties. However, among and after the long endurance test operation several engineering developmental efforts should be devoted to system simplification and technological maturing.

4. PROPOSED TRANSMUTATION CONCEPTS

4.1. PHOENIX concept (USA)

The PHOENIX concept (4) uses a large linear proton accelerator (LINAC) to drive and control one or more subcritical lattices of minor actinides (neptunium, americium and curium), to transmute the long-lived radioactive wastes from Light Water Reactors (LWRs), that are the most difficult to dispose of, and

to produce electric power in the process, One 3600" MWt machine would transmute the neptunium, americium, curium, and much of the iodine produced by about 75 LWRS and would generate a net of about 850 MWe for the electrical grid, as indicated in figure 5.

While not tied to a specific fuel reprocessing/recycle technology, much of the PHOENIX analysis performed thus far has been based on the proposed Clean Use of Reactor Energy (CURE) approach (5), which is a waste partitioning process based on the well-known Purex process and the newer Truex process. Within the CURE framework, certain elements are to be recycled, transmuted or simply separated from the major portion of the high-level wastes. The primary objective is to eliminate certain key nuclides from the bulk of the spent fuel so that the remainder can be packaged more easily (reduced heat load and shorter lifetime requirements) for disposal in the geologic repository currently planned by the US Department of Energy (DoE).

The PHOENIX concept assumes a large LINAC that can produce a 104 mA beam of 1.6 GeV protons.

A multiple (modular) concept was developed for the PHOENIX subcritical lattice. Each module resembles the core of the Fast Flux Test Facility (FFTF) (6), with the minor actinides formed into oxide fuel rods, replacing tbe uranium and plutonium in the FFTF fuel, The fuel rods are cooled using liquid sodium and are bundled into 217 pin assemblies, with 124 such assemblies making up a 450 MWt target module. From one to eight of these target modules are aligned in front of the proton beam, depending in part on the amount of fuel available at any given time.

The FFTF currently uses sodium coolant and oxide fuel and provides an existing data base as well as a potential site for experiments on minor actinides fuel. The prototype PHOENIX lattice was based on the FFTF lattice and scaled up to the required power level. As indicated in figure 6, the lattice parameters would be essentially identical to FFTF, with the simple replacement of uranium-plutonium oxide fuel with minor actinide oxide fuel.

Most of the neutrons in the PHOENIX lattice will be the result of fissions and material damage will be caused by those neutrons. There are strong reasons to believe that the PHOENIX target modules will experience material damage very similar to that in FFTF.

Coupled LAHET/MCNP calculation was performed

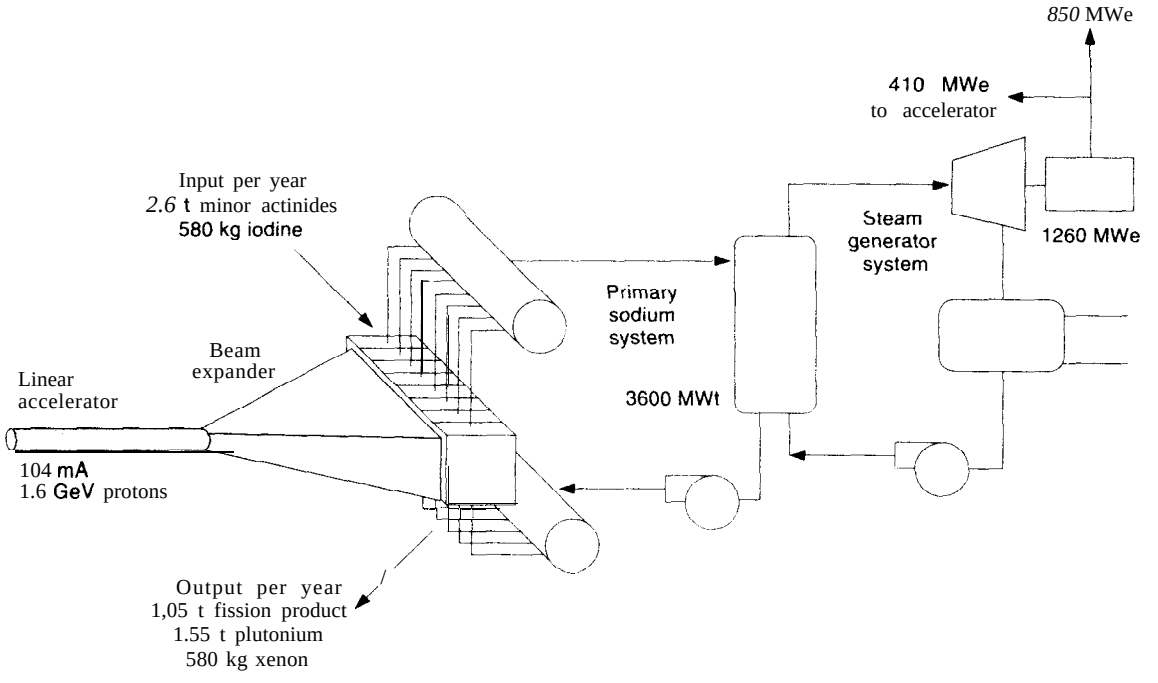
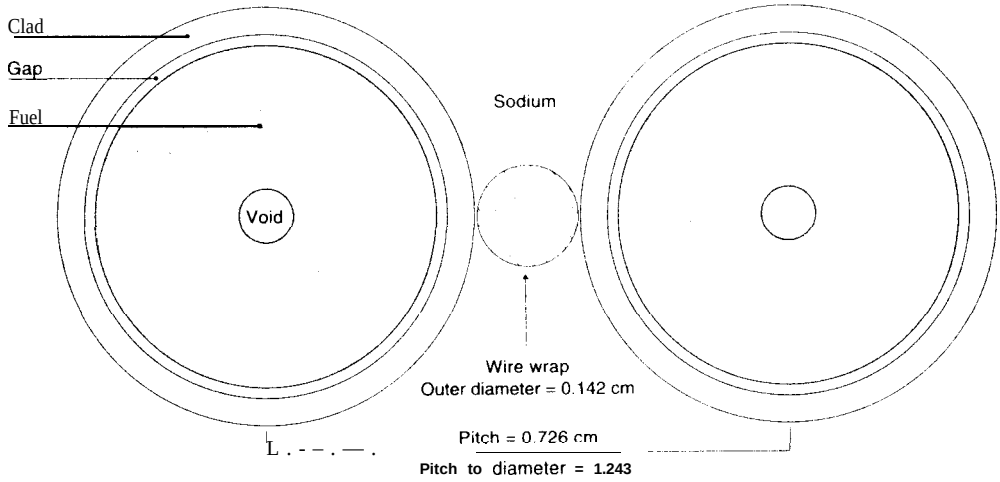


Fig. 5 – The PHOENIX concept (intermediate sodium system not shown)



Region	Material	Outer diameter (cm)	Fuel composition ($X\text{-}0_2$)
Void	Gas	O 076	0.418 ^{237}Np
Fuel	$X\text{-}0_2$	0.493	0.478 ^{241}Am
Gap	Gas	0.508	0.086 ^{243}Am
Clad	Stainless steel HT-9	0.584	0.017 ^{244}Cm
			0.001 ^{245}Cm

Assumed fuel properties are same as UO_2

Fig. 6 – PHOENIX lattice fuel pins, currently based on FFTF oxide fuel

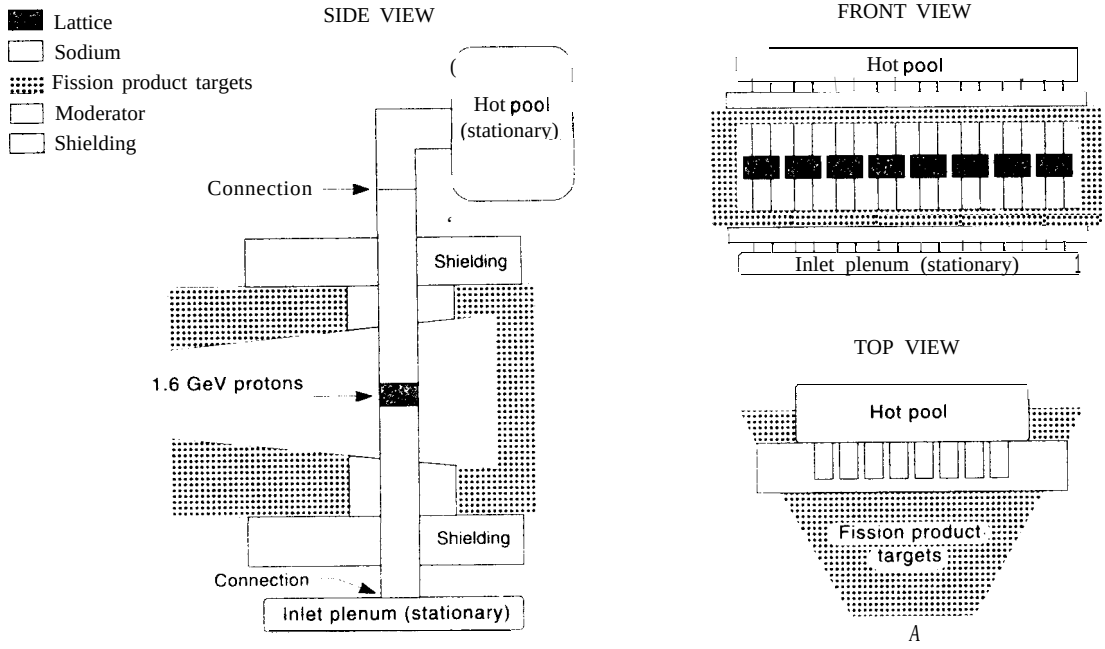


Fig. 7 – PHOENIX target chamber from three perspectives

or an essentially homogeneous three-dimensional rectangular representation of the target lattice, Subsequently, the lattice feature of the MCNP code was used to represent the lattice/target module geometry in its full heterogeneous detail to qualify the adequacy of the homogeneous modelling employed in the coupled calculation. The results of these calculations provided a Preliminary estimate for the target size: 188 cm wide, 72 cm deep and 75 cm high. This sizing gives a k_{eff} of about 0.8 for a new target, which is a good choice given the large reactivity increase expected during the first 2 years.

The use of modular targets would allow the removal of a troublesome target module without shutting down the machine for a prolonged period of time.

Lattice characteristics inside the hex-cans are very similar to FFTF. However, the shape of the module, 1.82 m x 1.88 m x 0.75 m is set to allow for considerable neutron leakage.

The arrangement of the eight modules within the vacuum chamber is illustrated in figure 7. Each module is designed to be removed from the vacuum chamber for reloading. PHOENIX could run effectively even with only one target chamber, although its throughput and efficiency are highest if all targets are loaded.

4.2. JAERI concepts (Japan)

4.2.1. Actinide transmutation plant with sodium-cooled solid target/core

The plant (7) is designed as a system that transmutes about 250 kg of Minor Actinides (MA) per year by fission, which corresponds to the annual actinide production of about 10 LWRS of 3000 MWt. The subcritical core is required to operate at k_{eff} of 0.9 or more to reduce the size of the proton accelerator and improve the energy balance of total system. Hard neutron spectrum is used.

Schematic diagram of the proposed concept is shown in figure 8. The accelerator injects 1.5 GeV proton beam into the tungsten target located at the centre of the core, which is loaded with actinide alloy fuel. A large number of neutrons are released by spallation reaction in the target and further fission reactions are induced in the actinide fuel region. The core generates a thermal power of about 820 MW and is cooled by forced upward flow of sodium primary coolant. In the energy conversion system, thermal energy is converted into electricity. A part of electric power is supplied to its own accelerator.

Metallic alloy of MA is used as fuel. In order to

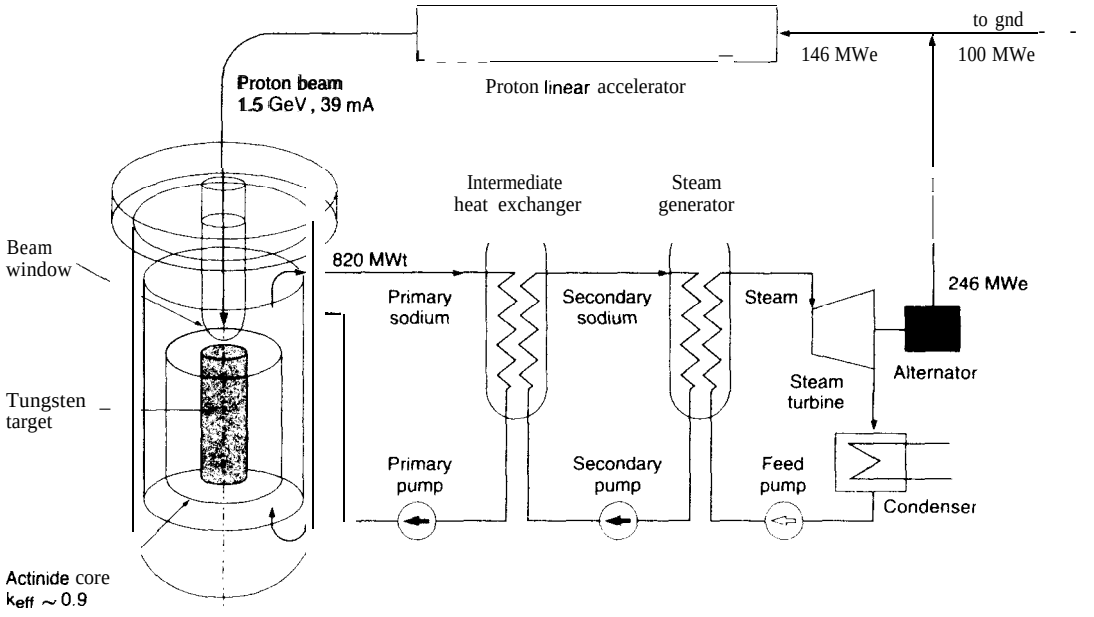


Fig. 8 – JAERI concept of accelerator-based actinide transmutation system with sodium-cooled solid target/core

TABLE 1 - JAERI ACTINIDE TRANSMUTATION SYSTEM WITH SODIUM-COOLED SOLID TARGET/CORE FUEL DESIGN PARAMETERS

Fuel composition	Np - (15) Pu - (30) Zr AmCm - (35) Pu (10) Y
Slug diameter, mm	4.0
Clad:	ODS steel
outer diameter, mm	5.22
thickness, mm	0.3
Active length, mm	1400
Pins assembly	55
Pin pitch, mm	8.7

keep sufficiently high phase stability of alloy, [be fuel is composed of two distinct alloy systems: Np-(15)Pu-(30)Zr and AmCm-(35)Pu-(10)Y. These alloy systems are proposed for a conceptual sodium-cooled acti nide reactor burner. Metallic fuel allows to implement a compact fuel cycle on pyrometallurgical processes and provides a hard neutron spectrum.

In the design, maximum fuel temperature is limited to 900 "C, because of low melting-point of MA alloy. Actinide fuel slug with a diameter of 4 mm is sodium-bonded to the cladding made of Oxygen Dispersion Strengthened (ODS) alloy. The outside diameter of the cladding is 5.22 mm, and the thickness is 0.3 mm. Fuel design parameters are summarised in table 1.

To ensure adequate fuel cooling during the out-of-core handling operation, the assembly design has several unique features; the structure members are six tie-rods near the comers rather than a hexagonal wrapper tube, the number of fuel pins per assembly is 55. the fuel pin pitch is 8.7 mm, fuel pins containing Am and Cm are arranged on the outermost row of the array while Np-containing pins are arranged on the inner rows.

Active length of the pins is 1.4 m. Each fuel pin contains a gas vent mechanism and an upper reflector .n its upper section. Because of relative wide fuel pin spacing, grid type spacers are employed rather than wire wrap spacers. Sodium coolant enters the assembly from the entrance nozzle at the bottom, flows upward through the space between fuel pins, and exits from the handling head at the top.

The core has two regions: the tungsten target and the actinide fuel (see fig. 9). The target, located in the centre of the core, consists of 61 target assemblies, in form of an approximate cylinder, of about 0.46 m diameter and of 1.4 m height. The target assembly has the same cross section as the actinide fuel assembly.

The target region is surrounded by the annular actinide fuel region, 1.4 m in outside diameter and 1.4 m in height. The fuel region is made of 378 fuel assemblies. Around the fuel region is the stainless steel reflector. Thickness of' upper and lower axial reflectors is 0.4 m and side reflector thickness is 0.6 m.

High energy protons are injected vertically downward through the beam window into the target, which is inactive, i.e. it does not contain fissile or fissionable materials. The volume fraction of tungsten is variable along the beam axis to produce an adequate shape of high energy neutron flux distribution from the target. This eliminates the power peaking at the core centre and thus considerably flattens the core power distribution.

The vessel diameter is about 4.6 m and the height is about 14 m. The reactor is of' loop type. Primary coolant sodium enters from the inlet nozzle, and then flows downward through annular space between the vessel and the core barrel into lower plenum.

Then, it turns upwards, removes the heat from fuel and target during its passage through the core. and exits from the vessel through the outlet nozzle.

A vertical tube for beam path is inserted into the reactor vessel down to just above the target region in the core. The bottom end of the tube is the beam window made of ODS alloy. The beam window has a sha-

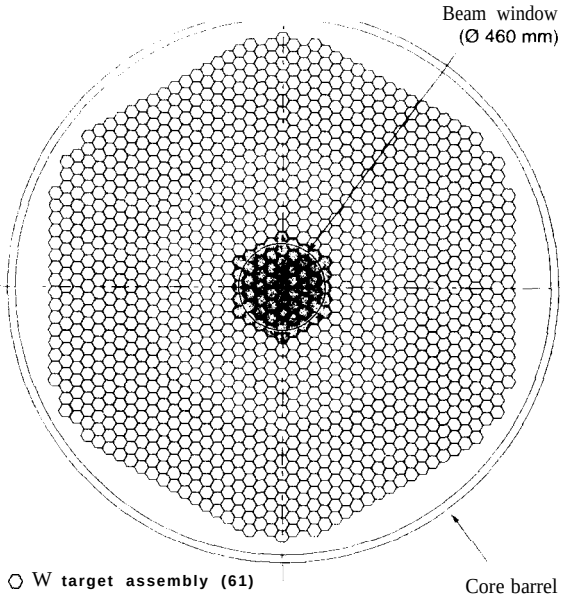


Fig. 9 – Core configuration of JAERI concept of accelerator-based actinide transmutation system with sodium-cooled solid target/core

pe hemispherical shell and is cooled by the upward impinging sodium flow from the target exit.

Core design parameters are shown in table 2. The volume of active core is about 2 m'. Maximum coolant flow velocity through the active core is limited to 8 m/s to avoid the possibility of flow-induced vibration of fuel pins. Core inlet sodium temperature is selected to be 330 "C. The coolant temperature lower than in standard sodium-cooled reactors helps to achieve a high core power density, or a high transmutation rate. at the expense of decreasing the power conversion efficiency.

The core generates a thermal power of 820 MW for the 1.5 GeV and 39 mA proton beam. 800 MWt are generated in the actinide fuel region and 20 MWt in the tungsten target region. In the fuel region, the maximum power density is about 920 MW/m^3 and the average power density about 400 MW/m^3. The maximum power density in the target regions is 360 MW/m^3.

The core operating parameters are given in table 3. The core inventory of minor actinides is about of 3160 kg. The k_{eff} is calculated to be 0.89. For an incident proton, approximately 40 neutrons are emitted by spa]-lation reactions in the target region. About 100 fissions are induced per proton in the fuel region. Assuming a load factor of 80%, the actinide burnup is approximately 250 kg, or 8%, after one year operation. A fairly hard neutron spectrum with an average neutron energy of 690 keV is achieved in the core.

Heat transport and power conversion systems in the plant design are based on the current state of technology for a sodium-cooled fast breeder reactor plant. The primary heat transport system consists of two sodium coolant loops. Each primary loop has an intermediate heat exchanger and a primary pump. The secondary system also consists of two sodium loops having a steam generator and a secondary pump.

In the power conversion system the steam drives a single turbine alternator to produce electricity. Due to the relative low coolant temperature, for a sodium-cooled system, the steam condition is similar to that of a LWR. The plant efficiency is roughly 30% and therefore the electric output is about 246 MW.

Assuming an efficiency of 40% for the 1.5 GeV-39 mA proton accelerator, electric power required to operate the accelerator is about 146 MW. This means that the proposed system is more than self-sufficient in terms of its own energy balance, having capability to supply 100 MW of surplus electricity to the external grid.

4.2.2. Accelerator molten-salt target system

The molten chloride salts have been studied as a solvent in the non aqueous reprocessing. Recently chloride systems have received serious research and development effort for Integral Fast Reactor (IFR) at the Argonne National Laboratory.

Concerning the TRans-Uranics (TRU) volubility in chloride salt systems. various phase diagrams composed of $PuCl_3$ are reported. Tbe system NaCl-$ThCl_4$-$PuCl_3$ is attractive considering the Pu volubility and the melting point respectively. The NaCl-$PuCl_3$ system is adopted in this work (8). The eutectic temperature is 453 'C for the composition of 64 (mol%) NaC1 -36 (mol%) $PuCl_3$, as the target salt in the accelerator transmutation system. The volubility of MA in this salt are not reported, though. Pu may be replaced by other actinides such as Np, Am and Cm.

TABLE 2- JAERI ACTINIDE TRANSMUTATION SYSTEM WITH SODIUM-COOLED SOLID TARGET/CORE: DESIGN PARAMETERS

Proton beam:	
energy, GeV	1.5
diameter. mm	400
Target	Solid tungsten
Fuel	Actinide alloy
Active Core:	
volume, m^3	2
length, m	1.4
Coolant:	Sodium
velocity, m/s	8
inlet temperature, °C	330

TABLE 3 - JAERI ACTINIDE TRANSMUTATION SYSTEM WITH SODIUM-COOLED SOLID TARGET/CORE: OPERATING CONDITION

Proton beam current. mA	39	
Actinide inventory, kg	3160	
Multiplication factor	0.89	
Neutrons/pro[on	40	
Fissions/proton:		
(>15 MeV)	0.45	
(<15 MeV)	I 00	
Neutron flux, n/cm^{2}s	4 10^{15}	
Mean neutron energy, keV	690	
Bumup, kg/y	250	(8%/y)
Thermal output, MW	82(I	
Power density. MW/m^3:		
maximum	930	
average	400	
Linear rating maximum, kW/m	61	
Maximum temperature, "C:		
coolant	473	
fuel	890	
clad	528	

To establish a harder neutron spectrum in the target system, chloride salt could be better than fluorine, since the mass number of chlorine is about twice that of fluorine, the parasitic neutron absorption cross section of chlorine is not so large. Therefore, the amount of sulphur produced, by the neutron absorption in the molten salt target would be small and its effects into the [at-get system will be neglected.

For the transmutation of the MA produced by ten LWRS of 3000 MWt, namely about 250 kg/y, the subcritical target system can operate at k_{eff} of ().9 or more. considering the reasonable size of the accelerator and the energy balance of the whole system.

The conceptual design of the molten-salt target system and its rough dimensions are shown in figures 1 () and l l respectively. The proton beam is injected in the salt target region I of figure l l through the beam window. MA are mainly transmuted in this region by the fast fission and spallation reactions. The fission chain reactions can be controlled by the proton beam. In region 111, the heat exchangers and centrifugal pumps are located inside the target core to decrease the total volume of the target,

The salt containing MA is circulated through region I to III by the pumps. The total capacity of the heat removal is about 800 MW, which corresponds to the

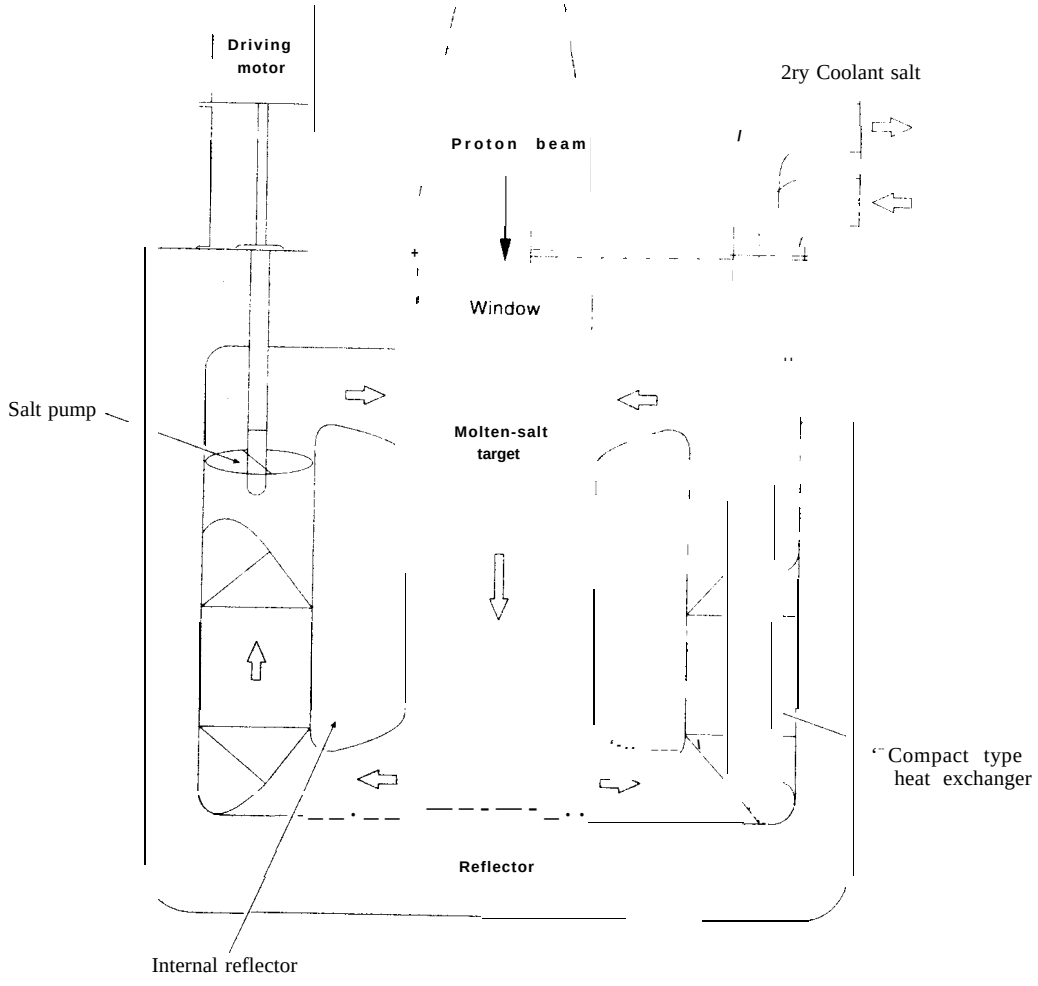


Fig. 10 – Concept of the JA ERI molten-salt target system

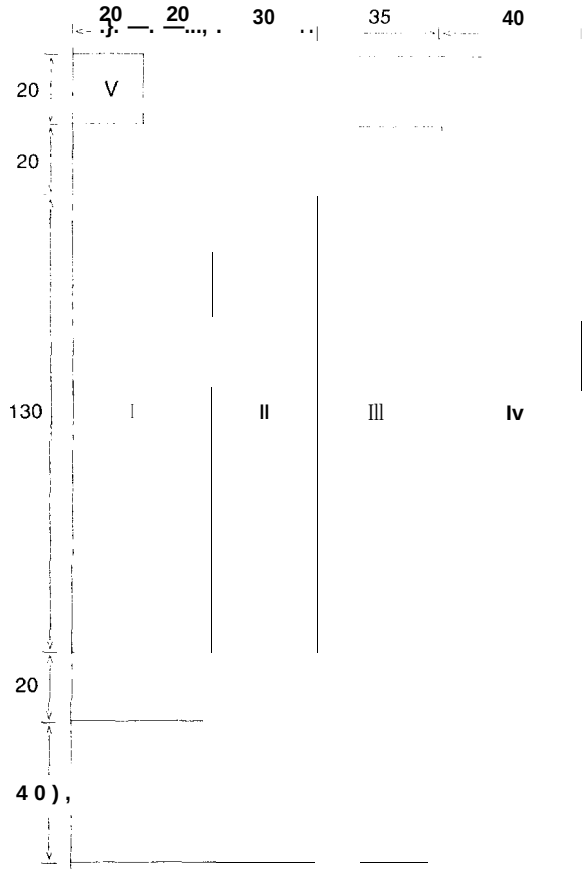


Fig. 1 I – Scales of the JAERI molten-salt target core (dimensions in cm)

heat generated by the incineration of 250 kg/y of MA.

The temperatures of the primary salt at the input and output of the heat exchangers are of 650 'C and 550 "C, respectively.

The region II is a cylindrical neutron reflector to protect the heat exchangers and pumps from the radiation damage.

The region IV is the neutron reflector; its external part is the core vessel.

The region V is the beam window including cooling gas flow space. High proton flux through the window causes severe radiation damage and excess heating. Preliminary analysis of HT-9 steel window was developed.

In the salt target the ratio of Pu to MA is assumed 15 to 85 mol%. The MA is composed of Np. Am and Cm, the ratios among its concentrations are considered the same that of LWR spent fuel, namely 56-39-S. The design parameters and the calculation results are shown in table 4. The calculated k_{eff} value is 0.92.

From the burning calculations it results that the amount of MA produced from 10 LWRS of 3000 MWt can be incinerated by a system of this type having a beam current of 25 mA and beam power of 1.5 GeV.

The neutron spectrum in region I is close to that of the fast reactors. The highest neutron flux is about $1.0\ 10^{14}$ n/cm^{2}s for the current of 25 mA.

The maximum neutron flux in the region 111 is about 2 orders of magnitude lower than in the region 1.

4.2.3. Transmutation of the long-lived fission products

High energy proton of 1.5 GeV is introduced to the Bi-Pb flowing target. The spallation neutron produced into the target passes through the wall of the container to the heavy water blanket. The neutron in the blanket transmutes the ^{99}Tc and the ^{129}I to stable nuelides. Tc is in the heavy water as a slurry of TcO_2 and 1 is solved as ion from solution of AlI_3. The transmutation products of Ru in the slurry can be separated from the TcO_2 by volatilisation of Tc_2O_7. On the other hand Xe from the ^{129}I can be removed easily from the heavy water by He-purge method.

The proton current needed to transmute the fission products coming from 10 LWRS of 3000 MWt is more than 150 mA (for 60% efficiency 250 mA is requested).

The radiation damage and corrosion to the wall of the Bi-Pb vessel are serious problems. The austenitic stainless steel, however. is thought to be applicable if the temperature of the wall can be controlled under 300 "c.

TABLE 4 -DESIGN PARAMETERS OF JAERI MOLTEN-SALT TARGET/SYSTEM AND RESULTS OF THE CALCULATION FOR MINOR ACTINIDE TRANSMUTATION

Target salt composition	63NaCl-36 (Pu, MA) Cl_3 (Pu : MA = 15: 85) (MA . Np, Am. Cm)
Volume. m'	2.6
Weight. t	9
Trans-uranic inventory, t	5
Effective multiplication factor, k_{eff}	0.92
Beam power:	
GeV	1.5
mA	25
Thermal output, MW	800
Transmutation rate of MA. kg/y	250

The transmutation reactions of these FPs are heat absorbing. To held the energy balance as a total sy - stem, the electric power for the FP transmutation shall be supplied by the margin of the power produced by the MA transmutation system.

4.3. Los Alamos concepts (USA)

4.3.1. ATW concept

Fission product waste arises from fission of either fuel or actinide waste materials. About 90% of fission products are non-radioactive or short-lived (less of 10 years), and can be left to decay naturally. Those having a longer half-life can be transmuted into non-radioactive or short-lived nuclei through neutron capture. At high neutron energies, this process is inefficient because capture cross-section is small. but at thermal neutron energies, capture cross-section increases. The most troublesome fission products are ^{99}Tc and ^{129}I because they readily form water-soluble compounds: this means that they could migrate out of a geological storage medium. These isotopes have high thermal neutron cross-sections and are easily transmuted to stable ^{100}Ru and ^{130}Xe respectively. The next most important long-lived nuclide is ^{135}Cs; this would need isotope separation before burning to make effective use of' the neutrons in the transmuter. The small quantities of ^{126}Sn and ^{79}Se can be readily transmuted. In contrast. ^{90}St and ^{137}Cs, among major components of' the waste. are more difficult to deal with. Their low thermal neutron cross-sections are so small that, even in the large flux levels present in the Accelerator Transmutation of Waste (ATW) blanket (9), only a halving in the effective half-life appears feasible. However, with a half-life of 30 years, these materials decay away naturally on a time scale reasonable in terms of human life span. We believe it is unnecessary to transmute ^{93}Zr and ^{107}Pd because their radioactivity is small. their decay products have low energies (non-damaging to tissue), and as noble metals they are not readily transported naturally through the environment.

in principle, the accelerator-driven neutron economy allows all of' the long-lived fission products to be transmuted, but tbe value of doing so varies between them and must be determined for each isotope by cost, risk and benefit analyses. The goal of ATW is to burn ^{99}Tc, ^{129}I, ^{135}Cs, ^{126}Sn and ^{79}Se.

I n the ATW concept the linear accelerator operates at 1600 MeV at a continuous-wave current of' 250 mA. The primary proton beam is then split into four beams, each having a current of 62.5 mA and an energy of 1600 MeV (see fig. 12). Each of the four beams is directed into four separated target/blanket modules. A modular design approach allows maintenance or replacement of components with relative ease and adds to the overall reliability of' the system. The modules have separate cooling loops for their respective targets. moderator. and blankets, and operate independently from each other.

The high-energy proton beam strikes a centrally located spallation target to produce an intense source of' neutrons. The base-case design is comprised of heavy water cooled tungsten rods. The power density is very high. and therefore requires sub-cooled boiling as the heat transfer mechanism. Because of the intense proton and neutron environment, the target assembly will require routine replacement.

Each target/blanket module is designed to burn the actinides and fission products from approximately two

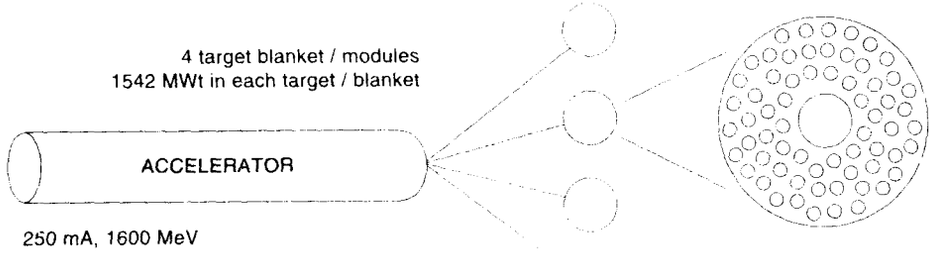


Fig. 12 – A *TW schematic*

light water reactors and produce approximately a 1542 MWt power. The blanket region and balance of plant design is based on existing heavy water reactor technology employed in the CANDU reactor system. Here, the fuel is contained in double-walled pressure tubes that pass through the blanket.

One main difference between the CANDU design and the ATW design is that an aqueous homogeneous continuous flow primary loop is employed in the ATW design, whereas solid heterogeneous fuel is used in CANDU. It is necessary to use the liquid fuel approach for actinide burning in ATW to take advantage of the high neutron flux and the rapid burn rates, as well as facilitate the continuous processing. This approach complicates the design somewhat because the fuel will be circulated both inside and outside the blanket region. To prevent the radioactive liquid fuel from coming into contact with the steam loop that drives the turbine-generator set, an intermediate light water heat transfer loop is added (see fig. 13). The additional intermediate loop adds another degree of safety margin in case of accidental radioactive release due to the rupture of the liquid fue" primary loop. The heat from the intermediate loop is transferred to a light water secondary loop for steam generation. In this concept, energy produced in the blanket from the fission of the actinides is recovered and used to produce electricity with about 30% thermal efficiency. This recovery of useful energy greatly reduces the overall cost of the facility.

A proposed layout for the target/blanket is shown in figure 14. The solid tungsten/lead target is surrounded by an annulus of technetium in D_2O, an actinide slurry region, and a D_2O reflector. The target/blanket design is evolving in an effort to enhance the safety, reliability, and efficiency (with respect to neutron utilisation). For example, it may be possible to keep the actinide slurry within the blanket region, and transfer heat to the D_2O moderator that is pumped outside the blanket. This retention will greatly reduce the overall volume of actinide slurry and reduce the risk of spills outside the blanket.

Another option that is under consideration is the use of a flowing lead target. The use of such a target adds complexity to the design but has the potential to increase the neutron utilisation efficiency. In addition, if greater accelerator currents are required, the liquid target can more easily remove the heat. The base-case target/blanket design and some of the design options are presented in more detail below.

As the target intercepts the proton beam, the heating rates are exceptionally high (9 MW/L peak) and the neutron flux levels produced are much higher than those for commercial nuclear reactors. The neutrons are very expensive to be produced when an accelerator proton beam is used. Therefore, the design of the target/blanket is crucial; it must use the neutrons as efficiently as possible while maximizing safety and reliability considerations. Several goals were specified in the design of the target/blanket (values given are for one LWR waste discharge):

- to transmute 33 kg of Tc and 7 kg of I per year;
- to transmute 300 kg of actinide mixture per year (Np, Am, Pu);
- to minimise parasitic capture in structures and target;
- to use materials that are compatible with fluids to minimise corrosion:
- to use material that avoids generation of long-lived radioactive nuclides;
- to use materials that can withstand intense irradiation;
- to provide a safe system that easily meets nuclear facility licensing and safety goals;
- to maintain subcriticality during normal and off-normal situations;
- to use passive modes of heat removal where possible;
- to ensure easy operation and maintenance;
- to minimise long-term research and development activities and costs.

There are several high atomic number materials that can be used for the spallation target including lead (Pb), lead-bismuth (Pb-Bi), tungsten (W), tantalum (Ta) and uranium (U). Through spallation on a lead target, approximately 50 neutrons are produced for each incident 1600 MeV proton. Neutron production is reduced to about 22 neutrons per proton for the 800 MeV application, and if a tungsten target is used, the neutron production decreases to approximately 18 neutrons per proton for the 800 MeV accelerator. For heat transfer and power density considerations, approximately 75% of the thermal energy in the beam is deposited directly into the target; the remainder of the power is removed by neutrons and gamma that leak into the blanket. For the 1600 MeV-62.5 mA proton beam, approximately 100 MWt power is generated, of which 75 MWt is deposited in the target.

For the ATW commercial waste application, two target design options were considered. The base-case design uses solid tungsten rods cooled with D_2O. As an option, a flowing liquid lead or lead-bismuth system is proposed, either of which act as the spallation target material and the heat transport medium.

The composition of the actinide waste was obtained from an ORIGEN code point depletion calculation

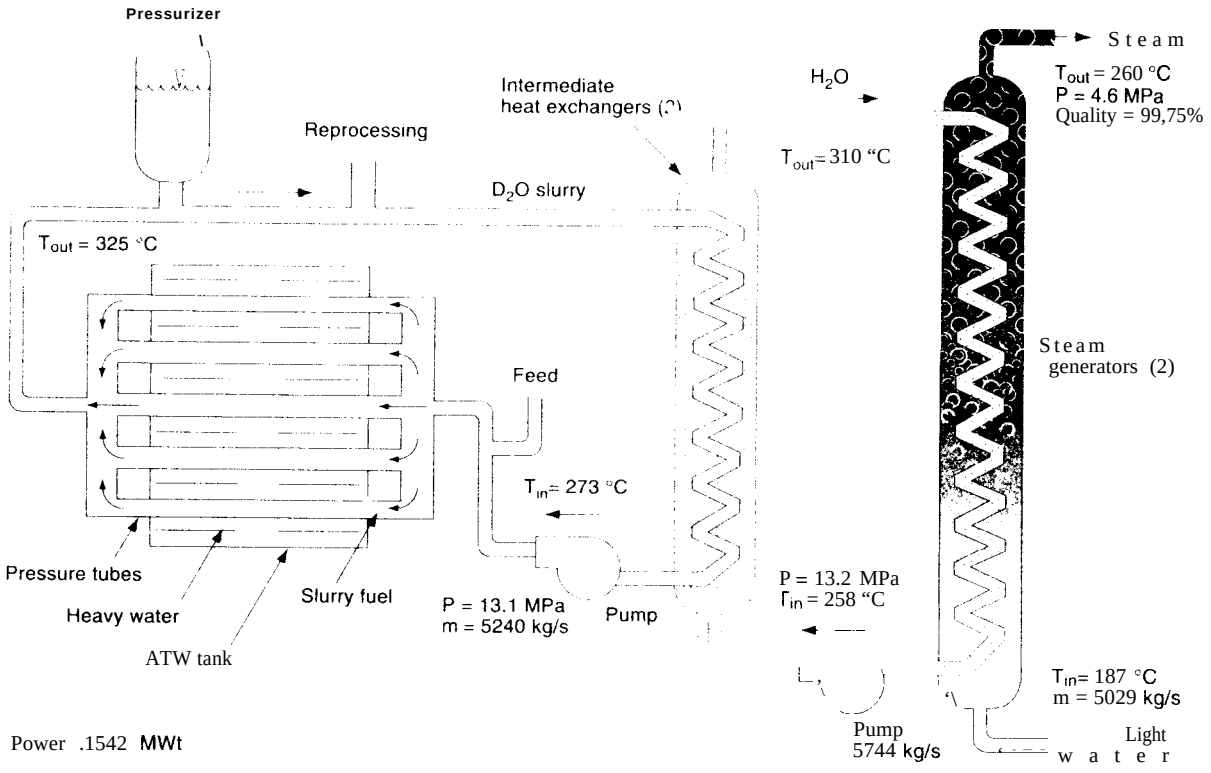


Fig. 13 – ATW balance of plant for the actinide slurry

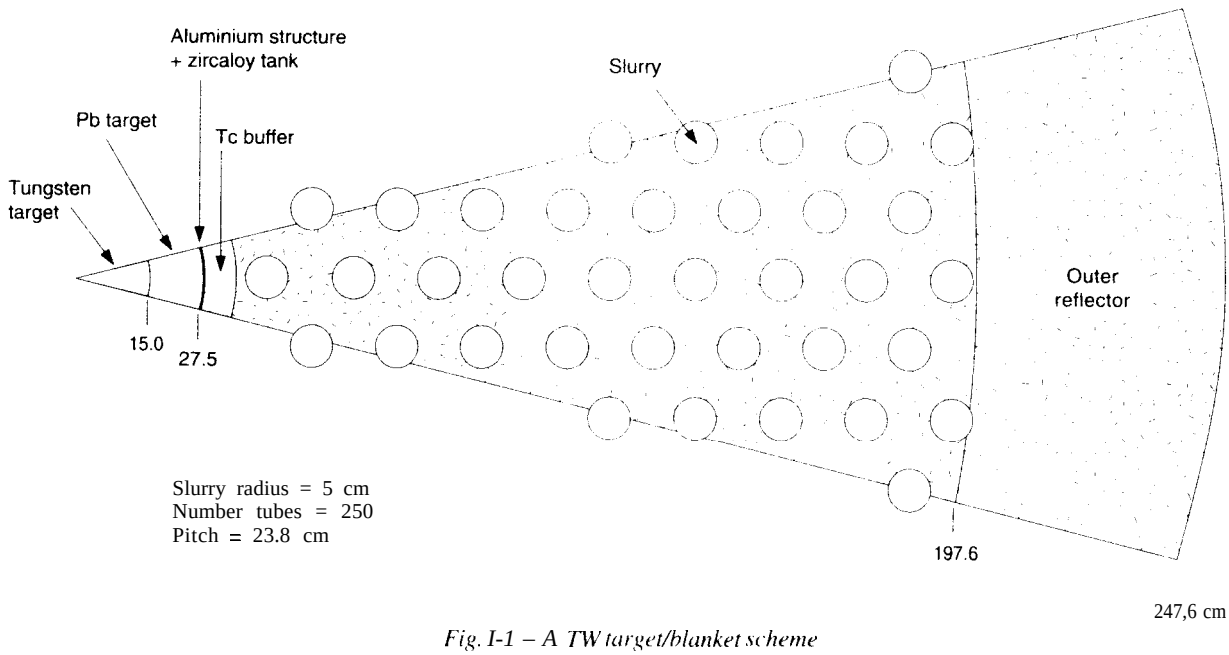


Fig. I-1 – A TW target/blanket scheme

using a CANDU neutron spectrum. Table 5 shows the relative isotopic composition of the PWR actinide wastes and the equilibrium composition after irradiation in a tlux of 1.0 10^{15} n/cm^{2}s that was used in the calculations. Plutonium isotopes constitutes almost 90% of the feed material mass but only 5070 of the equilibrium mass. The primary fissile isotopes are ^{239}Pu and ^{241}Pu.

TABLE 5- RELATIVE ISOTOPIC COMPOSITION OF THE PWR ACTINIDE WASTES AND EQUILIBRIUM COMPOSITION AFTER 1RRAD1ATION IN A FLUX OF 1.010" n/cm^{2}s

Isotope	PWR discharge	Equilibrium
^{237}Np	0.0449	0.0317
^{238}Np	0.0000	0.0003
^{239}Np	0.0000	0.0000
Total Np	0.0449	0.0321
^{238}Pu	0.0140	0.0230
^{239}Pu	0.5148	0.06 I ()
^{240}Pu	0.2372	(). [0()3
^{241}Pu	0.0785	0.0467
^{242}Pu	0.0481	().2630
^{243}Pu*	0.0000	0.0002
^{244}Pu*	0.0000	0.0026
Total Pu	0.8925	0.4968
^{241}Am	0.0513	().()()48
^{242}Am	0.0000	0.0001
^{242m}Am*	().0001	().000 1
^{243}Am	0.0092	().0874
^{244}Am*	0.0000	().()000
^{244m}Am*	0.0000	().0000
^{245}Am*	0.0000	0.0000
Total Am	().0607	0.0924
^{242}Cm	0.0000	().()213
^{243}Cm*	().0000	0.0006
^{244}Cm	().0018	().2794
^{245}Cm*	().0001	0.0051
^{246}Cm*	0.0000	().0004
^{247}Cm*	().0000	().(X)94
^{248}Cm*	0.0000	0.0625
^{250}Cm*	0.0000	0.()()0()
Total Cm	().00 19	().3787
Total	1.0000	I .0000

* Multigroup cross sections not currently available

Briefly, the present calculations indicate that a blanket combined with a 800" MeV- 140 mA (or equivalently a 1600 MeV-62.5 mA) accelerator may be able to transmute wastes from almost two pressurised water reactors.

The reference blanket has an actinide inventory of 325 kg in a 250 **cm** length. The corresponding Tc inventories are 82.0 and 99.2 kg in blankets operating at a k_{eff} of 0.95 and 0.93 respectively. The fluxes in the actinide blanket were calculated to be 1.38 10^{15} and 1.26 I 0^{15} n/cm^{2}s for k_{eff} of ().95 and 0.93 respectively.

4.3.2. Advanced ATW concept

Traditional liquid-fuel reactor systems pump the fuel from the core, where the heat is generated, to external **heat exchangers** for **heat removal.** The fact that the fuel is outside the core for some of the pumping cycle reduces the effective tlux and burnup rate and raises the actinide inventory. Thus it is proposed a target-blanket with slowly circulating higher-actinide liquid fuel and heat removal by a larger, thorium-bearing molten salt loop (1 O).

The most promising design makes use of a molten ^{7}LiF-^{9}BeF$_2$ salt target surrounded by a uranium layer and a graphite-moderated multiplying blanket (fig. 15). Most of the neutrons generated directly by the beam are produced in the uranium region. Molten salt ^{7}LiF-^{9}BeF$_2$ carries the LWR waste products and the thorium. The salt-uranium target allows efficient neutron production and low neutron parasitic absorption because of its small size and the presence of the surrounding fission product transmutation region. Since the proton or deuteron beam interacts mainly with the ^{7}LiF-^{9}BeF$_2$ salt, the target produces few waste products.

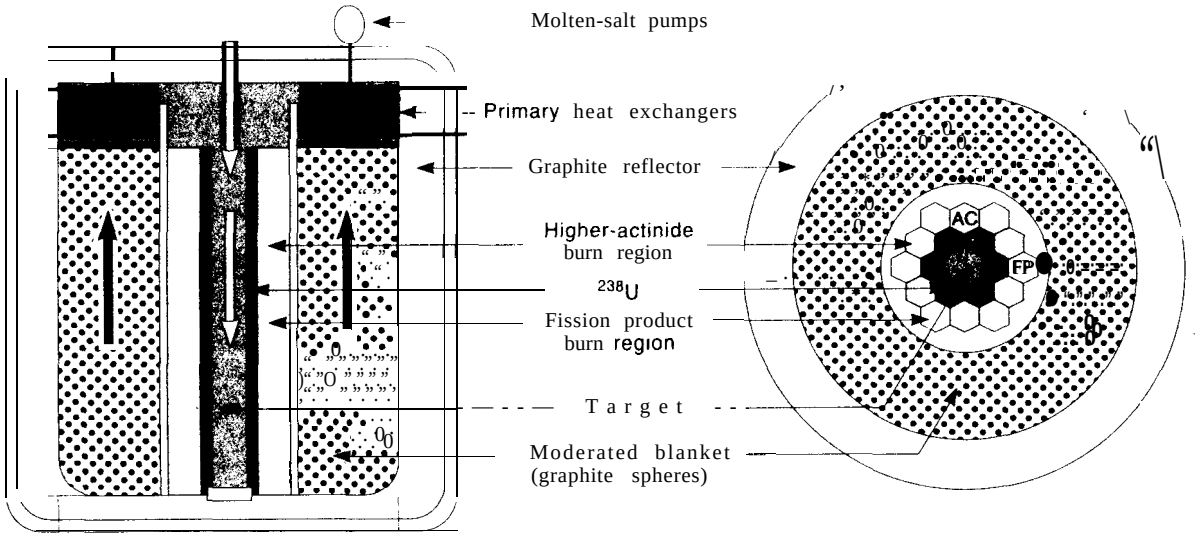
Np, Am and a fraction of the Pu coming from the spent fuel are fed slowly as fluorides in stationary containers (marked AC in fig. 15) near the centre of the blanket, in the highest flux region. There these isotopes take advantage of high flux burning, are fissioned in the most effective way and are then moved into the main molten salt loop. The heat generated in this central region is removed by the rapidly moving molten salt **in the main** loop. The molten salt in this loop circulates through a bed of graphite balls which supplies neutron moderation, and into intermediate (salt-salt) heat exchangers, where all the heat generated in the blanket is removed. Th breeding and the bulk of the power generation occur in the main loop, which is therefore, at the same time, target, fuel and heat-exchanging fluid, thus allowing a streamlined assembly design. The fission products isolated for transmutation are placed in special high-flux regions near the centre of the **blanket (marked** FP in fig. 15).

The on-site transmuter could be a single accelerator driving one or two target-blanket assemblies, burning the waste from a single LWR at the location of the commercial reactor, using as much as possible of the power-generating and support systems ah-eady existing at the site. No actinides would leave the site except uranium, which would eventually be re-used as fuel. If the transmutation and separation are done well enough, the transport and geological storage of long-lived actinides would not be needed.

Burning the actinide waste, including plutonium, from one 3000 MWt commercial reactor at the same rate at which the waste is produced would generate 750 MWt of power. The ATW plant would derive an additional 2250 MWt from the U-Th cycle while burning the waste originating in this cycle. Smaller ratios of power from thorium to power from LWR actinide waste are possible, using larger accelerators. Neutron economy arguments, based on the need of limiting neutron leakage as much as possible, demand, however, that at least half the power generated in a practical and economically viable ATW blanket comes from thorium.

The LWR waste is reduced linearly until it has all been put into the transmute. At this point, about 30 years later, the transmuter switches over to energy production exclusively from thorium. The remaining LWR actinide waste is reduced exponentially, and the system continually burns the thorium cycle waste. The equilibrium level of actinide waste during the energy production mode is only about 10 kg, compared with the 320 kg that accumulated every year in the LWR. At the end of the ATW plant life, this 10 kg would be burned in a new thorium energy production plant.

Processing of the LWR waste is needed before it can be fed as a fuel into the blanket. The spent LWR fuel assemblies could be converted to liquid fuel by using, for example, an eddy current or plasma torch process to melt and remove the zirconium cladding from the metal-oxide spent fuel. The oxide rubble would then be fluorinated to convert the uranium to highly volatile UF_6 which is removed readily. The remaining fluorinated actinides and fission products are dissolved in the molten salt and are then fed into the target-blanket.



The blanket is divided radially into four parts: 1) a central molten-salt column which is the target for the proton beam; 2) a uranium multiplier where most of the source neutrons are generated; 3) a fission product and actinide transmutation region, which also prevents thermal neutrons from the multiplying blanket from being parasitically absorbed in the target region; 4) the outer thorium-uranium multiplying breeding region of the blanket, which improves the neutron economy of the system and generates most of the electric power

Fig. 15 – ATW target/blanket schematic layout

4.4. *CERN concept*

4.4.1. Energy amplifier (11)

This illustrative case exploits the features of undermoderation with water already described previously. The typical thermal power which can be most readily produced in this way is of the order of 200 MW. In order to ensure sufficient cooling of the fuel, especially for larger power, the choice $V_m/V_f = 0.8$ is the most appropriate. Once the leakage of neutrons and the other losses in the fuel cladding etc. are taken into account, the system will be subcritical with $k = 0.92 \div 0.95$, corresponding to an energy gain $G = 33 \div 50$. A good design value is therefore $G = 40$. The beam has a energy of 800 MeV, an average current of 6.25 mA and a power of 5.0 MW. Both a LINAC and a cyclotron can easily satisfy such requirements.

The vast experience with Pressurised Water Reactors (PWR) can be used for the extraction of the heat. Of course other alternatives, like the boiling water mode. can be used as well, the choice being determined by the type of application.

The operating pressure of the device (PWR) is of the order of 154 bar corresponding to an inlet temperature of 291 "C and an outlet temperature of 322 'C. The coolant flow for the nominal power output of 200 MWt is 1.1 m^3/s. The required cooling transfer area inside the fuel core is approximately 300 m^2. Such a surface is provided even without cooling fins by cylindrical fuel rods of 2.5 cm diameter or smaller and a fuel mass of at least 21.4 t.

The general layout is shown in figure 16. It consists of two main separate parts, the final beam transport and the main energy amplifier assembly. The proton beam traveling in vacuum from the accelerator is focused by the magnetic quadruple lenses and reflected by 90 degrees with the help of the bending magnets. It enters the pressurised vessel through the long entrance collimator which has at its top end the pressure retaining window. Several beam observation devices are used to follow the beam trajectory. Heavy shielding floor is ensuring radiation safety. Neutrons from the fuel-moderator assembly can escape through the beam pipe. They are considerably reduced by the collimator. The narrow pencil beam passing through the collimator is collected in the beam dump, since contrary to the proton beam, they remain unreflected by the bending magnet.

Energy is extracted with the help of pressurised water contained in the main vessel. The cooling fluid enters through the inlet nozzle and exits through the outlet nozzle. It passes first between the inner walls of the main vessel and the core support barrel. Its flow is uniformed by the flow skirt and enters in the inner volume of the core support barrel and in the fuel assembly from below. It crosses the many channels of the fuel assembly, extracting efficiently the heat there produced and exits from the outlet.

The upper part of the main vessel houses the support structure, the fuel handling equipment and a number of control rods, mainly to be used to secure firmly in the non critical condition the fuel, after switching the beam off. The need for such device is primarily due to the fact that the fissile ^{233}U accumulates after shut-off because of the decay of ^{233}Pa. During operation these control rods can also be conveniently used to trim the neutron multiplication factor k_{eff} and hence the gain of the energy amplifier.

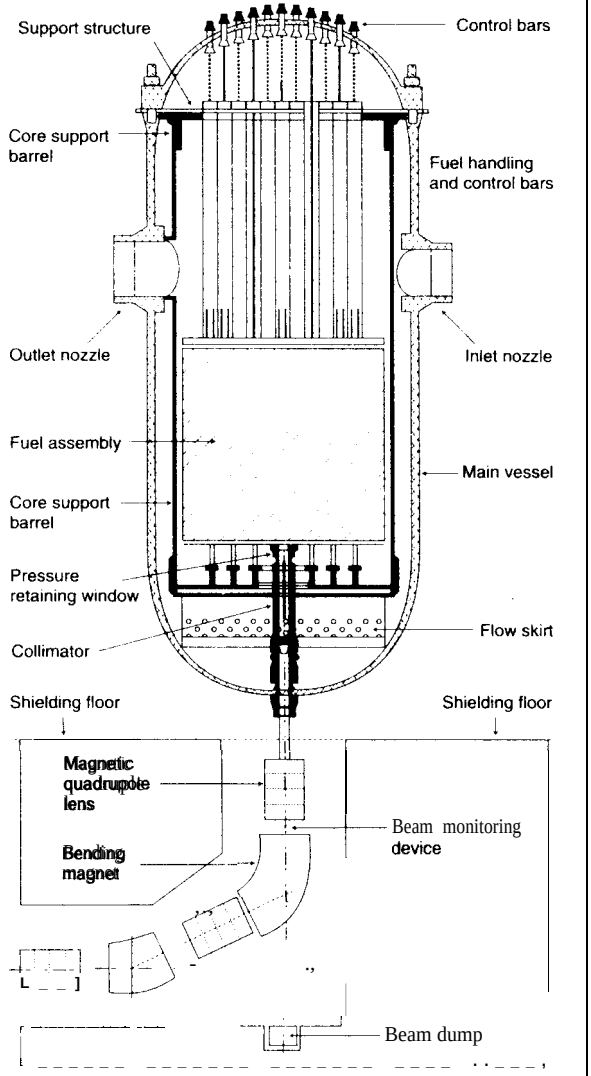


Fig.16 – Energy amplifier general layout

A number of different fuel assemblies can be used. Note that almost infinite variations are possible on fuel-moderator configurations. Largely inspired to reactor designs a fuel assembly is shown schematically in figure 17. The fuel consists in thorium metal rods clad with a thin zircaloy sheet to prevent corrosion. They are grouped in subassemblies which constitute a rigid unit for easy handling. Note that metal could be replaced by pellets of ThO_2, ThC_2 or other chemically stable thorium compounds.

5. TRANSMUTATION CAPABILITY

5.1. *Accelerator-driven system*

Table 6 summarises the data existing in literature concerning the transmutation capability of the various accelerator-driven concepts.

In the case of the PHOENIX concept, in ref. (4) the comparison was performed between the waste inventory and the release limits for 10000 years after disposal (see fig. 18), limits specified in the Code of Federal Regulations (CFR), particularly 40CFR191 and 109CFR60. The conclusion of the comparison is that the decontamination factor of the partitioning phase must be 10^{-5} or better to meet the containment requirements.

A preliminary analysis performed at Los Alamos (see ref. (12)) indicates for a homogeneous blanket having a thermal neutron flux of 1.3 10^{16} n/cm^{2}s transmutation rates of 500 kg/y for ^{237}Np (with an inventory of 9.9 kg of ^{237}Np), and 207 kg/y for ^{99}Tc (inventory of 26 kg of ^{99}Tc).

6. INTEGRAL EXPERIMENTS

6.1. *Accelerator-driven system*

6.1.1. Experiments planned in USA

The nuclear species and their amounts formed when a target is bombarded by protons in the 1 to 2 GeV energy range is the source term needed to evaluate some accident scenarios involving an accelerator-driven transmutation system.

The high-energy transport codes predict nuclide formation but are accurate only in an "average" sense; the predicted amount for some particular nuclide may be as much as a few hundred percent in error even if production as a function of A or Z may be approximately correct on average. Because some particular nuclides of the many formed might have some special environmental safety and health or other significance requiring more precise quantification, it is necessary to evaluate the situation experimentally.

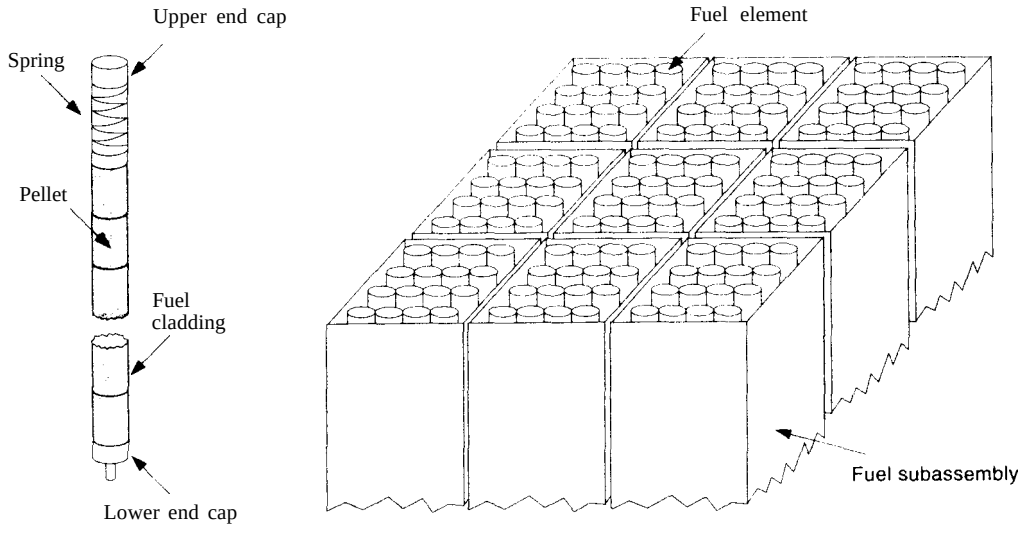


Fig. 17 – Energy amplifier fuel assembly

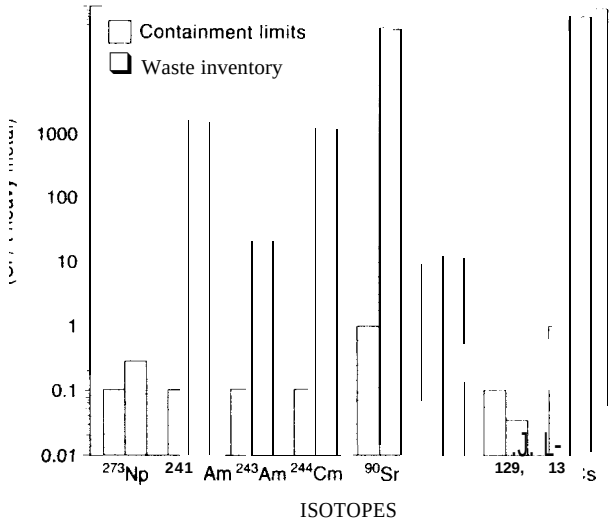


Fig. 18 – Waste inventory compared with release limits for 10000 y after disposal (limits specified in 40CFR191 and were in proposed 109CFR60)

The apparatus to generate the nuclides under conditions simulating a spallation neutron production target, used at one of beam lines of the Brookhaven National Laboratory's Alternate Gradient Synchrotrons, is shown in figure 19 (see ref. (13)). A range-thick cylindrical target of either of the two target materials favoured for the Acceleration Production of Tritium (APT) project, lead or tungsten. is divided into segments between which foils of the corresponding metal are placed. In this manner each foil is located at a unique depth into the target surrounded by metal of the same type. The overall dimensions of the target cm-respond to those for which there have been previous calculations and experiments of other parameters. An aluminium foil intercepts the proton beam of the desired energy impinging axially on the target to determine the proton fluence during the exposure by Al activation.

Each lead or tungsten foils is flanked by an aluminium foil that acts as a secondary dosimeter, a record of the size and position of the beam at the particular depth, and guards the sample foil from mechanically spilled debris from the target material.

Exposures to energies in the range of interest, about 1 GeV, are made for three successive time intervals differing by an order of magnitude.

The foils are assayed for their gamma spectra, using high-resolution calibrated (germanium diode) gamma detectors and standard gamma assay techniques at successive times, differing by several orders of magnitude so that the decay schemes can be deduced. The spectra are then analysed for the (quantitative) presence of the respective nuclides as a function of time after the end of irradiation.

Some interesting qualitative features of the spallation spectra can be demonstrated:

1 – an intense short-lived background underlying the gamma peaks of the spectra emitted by the target material within a short time after the end of the irradiation, which can be fitted quite accurately with a decreasing exponential in energy;

ABLE 6- TRANSMUTATION CAPABILITY: SURVEY OF DATA

oncept	Proton beam energy (MeV)	Proton beam current (mA)	Actinide burnup (kg/y)	Actinide composition	Actinide inventory (kg)	Number of LWR of 300()-36 600 MW served	Fission product burnup (kg/y)	Fission product inventory (kg)
HOENIX	1600	104		MA		75		
Iant with odium-cooled olid target/core	1500	39	250	MA	3160	10		
lolten-salt rget system	1500	25	250	I 5 mol% Pu, 85 mol% MA		10		
ission product ansmutation ystem	1500	250				10		
TW	1600	250	1200	90 mol% Pu, 10 mol% MA	I 300"	8	132 Tc, 28 I	Tc between 328-397

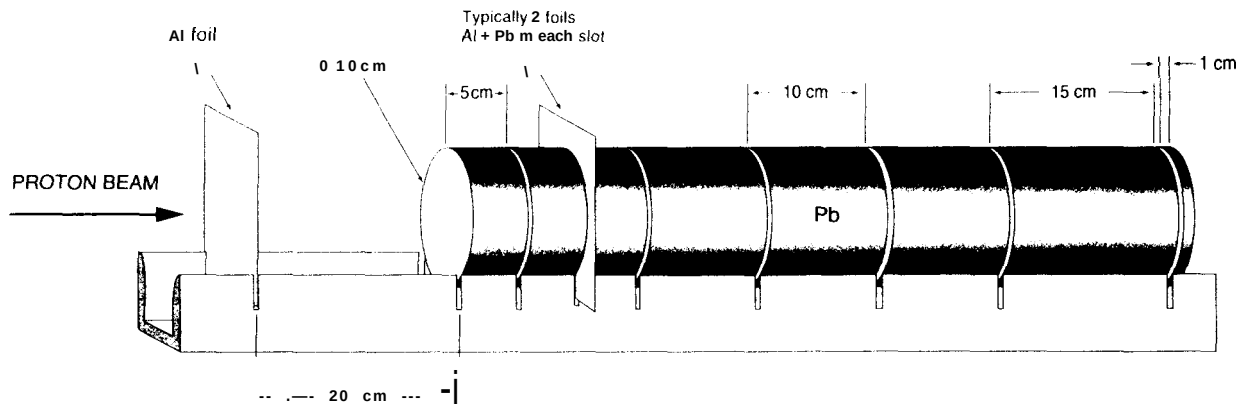


Fig. 19 – Experimental arrangement for activation of foil samples at the Brookhaven National Laboratory

2 – the **fact that the spectra measured at different times** for the same foil will be totally different in appearance due to the decay of some nuclides and to growth of others;

3 – the sharp fall-off. in intensity of the spectrum with energy and time compared to that for fission.

The afterheat that develops in target material for the APT due to the formation of radio nuclides during the proton-bombardment spallation process is expected to be less than that formed in a nuclear reactor with comparable neutron fluence by perhaps an order of magnitude if transport code calculations are to be believed and similarly for the effective decay time of that heat. Nevertheless, the question of afterheat in an APT target is still of interest from the thermal-hydraulics point of view during normal operation and as a safety issue in case of failure of the cooling system, even it' the accelerator beam is removed from the target immediately upon fault detection.

The present method of the experimental determination of afterheat, developed in connection with the APT program, bypasses the involved process of determining the principal nuclides by directly evaluating the heat produced by the spallation-induced radioactivity using a novel type of calorimeter originally developed at Argonne National Laboratory for assaying plutonium-bearing material. In this calorimeter, a constant-temperature environment is supplied by a servo-driven electrically warmed heat shield instead of constant-temperature water bath employed in the conventional instrument.

The action of the calorimeter is to supply heat to the sample to keep it at a constant elevated temperature even though the power output of the sample is decaying. The amount of heat supplied to the sample by **the calorimeter is the basic experimental data and is** essentially the complement of the decay heat.

The samples being assayed are first exposed to a proton beam and then retrieved by remote-handling devices. The first experiments (14) made use of the Brookhaven National Laboratory LINAC Isotope Producer with ().2 GeV proton energy impinging on lead target.

An experimental study was conducted at Texas A & M University (TAMU) (15) to compare measured and calculated spallation product yields from a lead target.

The experiment consisted in irradiating a thin, pure lead target (2 mm thick) in a 120 MeV deuteron beam. The lead target was exposed for 2 h at a nominal beam strength of I nA. After irradiation, the lead target was removed and counted on a well-calibrated, high-purity germanium detector at the TAMU Nuclear Science Centre. The characteristic gamma rays emitted from the lead target were recorded at various times for 1 month following the irradiation.

A special spallation product, decay gamma library was constructed and incorporated into the standard GENIE gamma-ray peak and nuclide identification soft ware. A total of 11 different nuclides in nine mass chains were identified from the multichannel counting spectra

The LAHET code was used to predict the generation rates for the product nuclides formed as a result of the deuteron-beam/lead-target interactions. The activity calculations were performed for nine mass chains from A= 198 to A=206 and contained isotopes of tellurium, lead and bismuth.

Results of the calculations and measurements at two decay times are presented in table 7. The ratios among the calculated and measured activities are generally less than 2 for all of the identifiable products.

Additional experiments employing copper, gold and thorium targets are planned.

Because the importance of the n/p parameter, it is desirable to resolve the difference between the code prediction and existing experimental data. The experiment (16) is intended to do that by creating a con fi - guration that is simple enough from a material and geometric standpoint that its performance according the codes (principally LAHET and MCNP) can be predieted unambiguously.

The experiment uses Brookhaven National Laboratory's Alternate Gradient Synchrotrons. Momentum-analysed protons in the gigs-electron-volt range will pass through two "paddle" detectors, further defining their energy by time of flight and arranged to produce a coincidence for each proton of the correct energy passing through to a lead or tungsten target located within a neutron detector. This detector is a cylindrical (CH_2) moderator pierced by ^3He proportional counters arranged concentrically with the cylinder axis.

6.1.2. Experiments planned in Japan

Spallation integral experiments (8) are underway in order to obtain data on nuclide production, to estimate the yields of neutrons and spallation products, and to investigate the validity of the spallation simulation code NMTC/JAERI. The 500 MeV booster proton synchrotron facility at the National Laboratory for High Energy Physics is used for the experiments. Proton beam was injected into a cylindrical assembly made of lead, with a diameter of 600 mm and a length of 1000 mm. A target was installed in the centre of the lead assembly. Target materials used so far are lead and tungsten. Experiments with a target of depleted uranium is also planned. The activation samples were Al, Fe, Ni and Cu cylinders with dimensions of 6 mm in diameter and 4-10 mm in length. These samples contained in 8 mm diameter mental capsules were inserted in the I () mm diameter holes drilled through the assembly along the beam axis at various radial positions. The number of induced reactions in the samples was obtained by measuring gamma-ray with a 100 cm^3 germanium detector.

Experimental results were compared with prediction by NMTC/JAERI code. The agreement was generally fairly good, but there were significant discrepanties on the centreline of the assembly. It is inferred that these discrepancies are attributed to the high energy nucleon streaming through the gap between samples and the assembly.

6.1.3. Experiments planned in Switzerland

Simple code comparisons for the irradiation of thin samples of actinides with high-energy protons have revealed considerable differences in the total yield and the shape of the mass distribution for both spallation and fission products. Since pure theoretical models are being compared with experimentally adjusted models, these differences are partly understood. However, in view of the more stringent requirements indicated above, experimental verification is desirable, especially for actinides for which the experimental data are scarce.

To check the respective modes, a basic validation experiment, ATHENA (17), in which thin samples of actinides are irradiated with 590 MeV protons from the Paul Schemer Institute ring accelerator, is underway. A uranium sample, consisting of 258 mg of UO_2, enca-

TABLE 7- COMPARISON OF MEASURED IN TAMU EXPERIMENT AND CALCULATED ACTIVITIES (μCi)

Isotope	Hal flife	Counting data set #2 2534 min after end of irradiation		Counting data set #5 7266 min after end of irradiation	
		Measured	Calculated	Measured	Calculated
^{199}Tl	7.42 h	().612	0.377	–	–
^{200}Pb	21.5 h	4.26	2.44	().411	().191
^{200}Tl	26.1 h	7.23	3.82	2.18	().972
^{201}Pb	9.33 h	2.00	1.25	–	–
^{201}Tl	3.04 d	1.79	3.31	0.955	1.65
^{202}Tl	12.2 d	0.131	().343	0.083	0.285
^{203}Bi	11.8 h	1.51	1.38		–
^{203}Pb	2.16 d	3.98	5.77	2.07	2.14
^{204}Bi	11.2 h	1.13	(),958		–
^{205}Bi	15.3 d	0.396	().445	0.389	().384
^{206}Bi	6.24 d	0.408	().645	0.299	0.533

psulated in a stainless steel tube with an inner diameter of 2.5 mm, was irradiated using a relatively low beam current, and y-spectroscopy was performed to confirm that the proton beam can properly be focused on the sample. First analyses gave promising results for ratios of calculated to measured activities. In the near future it is planned to irradiate a uranium and a thorium sample using a higher proton beam current. The utilised irradiation facility, Pirex, can handle currents up to *20* μA, which should make it possible to apply alternative measurements techniques such as inductively coupled plasma, mass spectrometry and total reflection X-ray fluorescence spectroscopy.

7. SAFETY ANALYSIS

At present stage of development of the hybrid systems, it is possible to see that the analysis must be focused at least at window, target cooling, interface target/blanket, dynamic behaviour.

7.1. Beam window

The beam window is probably the most critical component in the accelerator-based transmutation system. The major difficulties are caused by radiation damage and heat generation. Radiation damage by high energy particle is not fully understood.

Radiation damage can be alleviated by expanding the beam diameter at the window. Enlarging the window diameter needs a proportional increase in the window thickness to withstand the pressure difference across the window. As a result, the maximum temperature and the thermal stress in the window can not be reduced by increasing the diameter. In the JAERI concept, the design pressure difference is 0.25 MPa. The maximum allowable pressure difference is estimated to be about 2 MPa for the hemispherical window of ODS steel under uniform irradiation.

Window material should have high irradiation resistance, high creep rupture strength, high thermal conductivity, and good compatibility with coolant. Candidates are ferritic steels, vanadium alloys, SiC, C/C composites, etc..

The beam window should be designed to be replaceable. A multi-wall structure may be required to provide means for detection of window failure.

As noted above, the windowless design may be possible with lead, lead-bismuth, etc.. However, it seems a hard question to contain the radioactivity.

To reduce the window damage, it is necessary to expand the beam cross section area and to flatten the intensity profile. The undesirable beam tails also need to be removed.

Thermal power of a subcritical core is controlled by adjusting the beam current or energy. A proton accelerator can be electrically controlled much faster than the mechanical movement of the control rods. The time required for beam shutdown is probably within few ms.

Change in beam intensity causes a problem. Beam trip imposes a large thermal shock. The consequence of beam trip should be considered. If the multiple cores are driven by time-shared beam, they suffer from high frequency thermal shock. Small fluctuations of beam intensity may be inevitable due to the instability in ion source. It should be assured that beam fluctuations do not affect adversely.

The design of radiation shielding is more complicated than the case of regular reactors, because it involves much higher energies and there is a large diameter beam transport system.

Design measures should be provided to prevent the radioactivity from escaping into the accelerator in case of window failure.

Modern high intensity accelerators claim their high reliability. Accelerator availability can be well above 70% including planned beam-off time. The highest possible availability is desirable for overall system operation.

Results of research at the Los Alamos Meson Physics Facility (LAMPF) provided considerable data about window survivability. The LAMPF accelerator is 800 MeV- 1 mA. The double-walled flat window used on the current beam stop at LAMPF has undergone 17500 h at 20-30 $\mu A/cm^2$, for a total proton fluence of 1.3 10^{22} p/cm^2. This window is made of inconel 718. Another inconel window is used for the vacuum to air interface. It is replaced yearly and accumulates approximately 2.9 $10^{21} p/cm^2$. The proposed window for the Swiss Nuclear Institute Neutron Source is being tested at LAMPF. This material is a steel (Fe(10.5)Cr) and has also accumulated 2.9 $10^{21} p/cm^2$ of fluence proton.

For comparison to ATW, we assume that the 1600 MeV-62.5 mA proton beam is roughly equivalent to 800 MeV - 140 mA with respect to material damage. This provides an average flux of about 224 $\mu A/cm^2$ and in one full power year 3.52 10-2 p/cm^2 of proton fluence. This fluence is a factor of 2.7 times the current fluence on the LAMPF beam stop still in service. Based on this experience, it may be reasonable to assume that a window in ATW would require replacement about every 6 months.

7.2. *Target cooling*

In the case of loss of target coolant during the proton beam action on the target, the energy deposited in the target can produce a melting of a solid target and a vaporisation in a liquid target. In case of solid tar-

get the loss of beam produces a strong thermal shock on the target material and if it is repeated can produce rupture by fatigue.

7.3. *Interface target-blanket*

The target-blanket interface is a critical component in case of large accelerators. It is crossed by a fast neutron spectrum with a gaussian energy distribution centred at 4.8 MeV with a high energy tail reaching 400 MeV with an average of 105 MeV.

7.4. *Dynamic behaviour*

Usually three types of unprotected reactivity accidents are considered:

– slow reactivity ramp insertion,

– fast reactivity ramp insertion,

– fast reactivity ramp insertion due to sodium voiding caused by a loss of coolant accident (LOF driven TOP).

Slow reactivity ramp insertions without a scram are for example the inadvertent withdraw of a control rod(s) (few cents/s or 0.0001 k_{eff}/s).

A typical fast reactivity ramp insertion occurred in the EBR 1 accident which was caused by an inward bowing of the fuel pins. Other accidents of this category are earthquakes or diagrid failures without a scram (up to a few \$/s or 0.01 k_{eff}/s).

7..5. *The NEACRP benchmark problem (18)*

As a first example the KfK benchmark problem defined as a rod ejection accident and proposed by the Nuclear Energy Agency Committee on Reactor Physics (NEACRP) is chosen. It consists of a fast reactor made up of a core with a bank of annular control rods, radial, and axial blankets and sodium coolant. The essential features of the problem are: axis-symmetry, two neutron groups and six delayed neutron precursor families and thermal feedback through Doppler effects in capture and fission cross sections.

The transient is obtained through steady control rod bank withdrawal. The absolute reactivity insertion (p(\$)) starts at 1 ms and increases at a rate of 170 \$/s for the duration of 16 ms (the speed of the control withdrawn is adjusted to produce a ramp of 0.548 cm/ms). After this time the reactivity is kept constant.

The analysis of this problem gives a first indication of the mitigating effect of using a sub-critical accelerator-driven system.

These systems are driven by a spallation source dimensioned so that they generate in steady-state operation the same power as the critical reactor, which is assumed to be 1 GWt.

In a critical reactor, its power oscillates and has two peaks in a short time interval. Super-prompt criticality produces these peaks. The power rises rapidly during the period of super-prompt criticality and reaches its peak value at the time when the Doppler effect reduces the reactivity to values below the super-prompt limit.

In the case where the reactor is operated in a sub-critical mode the neutron source is determined such that the system generates 1 GW thermal power and this source strength is maintained during the whole time the reactivity is increased. When the time reaches 17 ms, or when the thermal power of the reactor reaches 50 times of the initial power (50 GW), the neutron source is reduced by the shut-off function $\exp(-t/\tau)$ (τ = 1 ins).

The case in which the initial sub-criticality is -3 and -2 \$ respectively, the power increases only to 2.2 GW and 6 GW respectively after 16 ms and after 17 ms, then the power decreases almost proportionally with the neutron source strength of $\exp(-t/\tau)$. If on the other hand the neutron source is maintained constant (the accelerator is not shut-off), also the power remains almost constant in this time range. For a sub-criticality of -1 \$ only a single peak power of 530 GW was calculated. Even though this value is similar to the peak of a critical system, the integrated power, i.e. the total energy release during the excursion is much less.

An interesting result of this analysis is the fact that the power decreases even between the delayed and prompt critical state. This is due to the long time constant of the delayed neutrons. When the reactor is in an under prompt-critical condition, the neutron flux is controlled by prompt neutrons which decrease for a sub-supercritical condition. For the case of a critical reactor two power peaks (curve no. 4 in fig. 20) of 700 GW and 500 GW respectively were calculated, which is in good agreement with the results of the NEACRP benchmark.

When the neutron life time and delayed neutron portion becomes half the value of the original case, then the power change is well as the reactivity change in units of dollars (as a function of time) becomes identical to the first case. This means that power and reactivity are time scale invariant in both the neutron life time and delayed neutron portion β.

When the power change is slow, the reactor can be controlled by a mechanical movement of control rods or by a hydraulic dispersion of fluid neutron absorbers which are dispersed by melting fuel elements like an electric fuse mechanism. In a sub-critical reac-

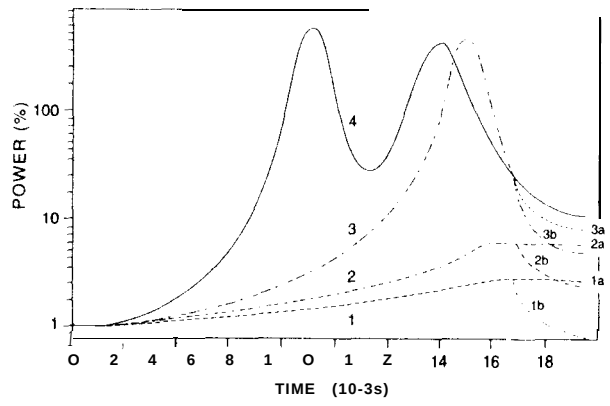


Fig. 20a – Power excursion after a reactivity insertion accident (170 $/s during 15 ins); curves I to 3: subcritical accelerator driven systems; curve 4: critical reactor

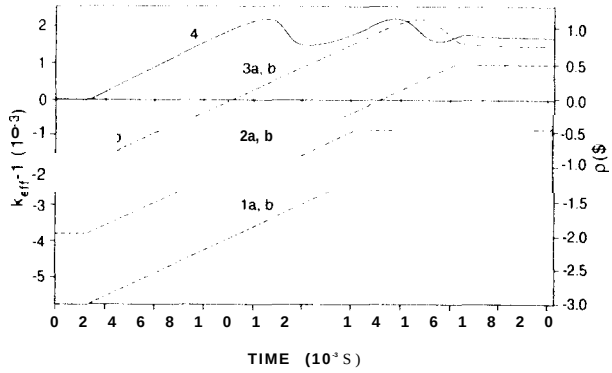


Fig. 20b – Reactivity behaviour during the accident scenario: curves 1 to 3: subcritical accelerator driven systems; curve 4: critical reactor

tor operated by spallation neutrons, the power change is much slower than in a critical reactor. This provides a great advantage from the point of view of reactor safety.

The few examples treated show that even slight] y sub-critical systems which require only a low accelerator current, respond much more benignly to a sudden reactivity insertion than critical systems. As can be seen from the figures *20* a) and b) already a sub-criticalit y of -3 $ reduces the energy release to less than a percent of that of a corresponding critical reactor.

It appears that a system with a k_{eff} of around 0.9-0.95 (= -30$- -15 $ for a fast reactor) would even look more attractive from the safety point of view. But these systems would require an expensive high current accelerator. In addition they are characterised by an inhomogeneous power distribution with a sharp peak around the target area. On the other hand, as we showed, accelerator-driven slightly sub-critical system behaviour is more benign than the "classical" reactors. They also have a relatively flat power distribution, and require only a low current and therefore much less expensive accelerators.

8. FEASIBILITY OF PU TRANSMUTATION BY HYBRID SYSTEMS

Reduction and recycling of wastes are one of the most recent trends in resource management and were pioneered by the nuclear community since the early days of nuclear power. Because of the small volumes of waste generated, nuclear industry has been a leader in pursuing "containment and disposal" rather than the more common "dilution and dispersal" strategy.

Nuclear industry has carefully considered the future management of-irradiated fuel discharged annually from some 400 commercial nuclear power plants now in operation world-wide including about 120 GWe nuclear electric power in operation in Western Europe and 45 GWe operational in the ex-USSR and East European countries (1 9).

Various means of management are considered for each main irradiated fuel constituents discharged from LWRS:

– uranium constitutes about 96% of the fuel unloaded from commercial power reactors. In the case of light water reactors, the most widespread type of reactor in the world, the spent fuel on discharge contains still 0.90% ^{235}U, whereas natural U contains only 0.7%;

– plutonium constitutes about 1% of the weight of the discharged fuel;

– minor actinides constitute about 0.1 % of the discharged fuel. They consists of about 50% of Np, 47% of Am and 3% of Cm;

– fission products constitute about 2.9% of the weight of the discharged fuel.

A typical 1000 MWe PWR unit operating at 75% load factor generates per year about 21 t of spent fuel at a burnup of 43 GWd/t and this contains about 20 t of enriched U, 230 kg of Pu, 23 kg of minor actinides, 750 kg of fission products.

Considering the papers presented at SAFEWASTE '93, it is clear that Belgium, France, Germany, Japan, United Kingdom and Russia reprocess their LWR spent fuel or have it reprocessed. This means that countries having more than one third of the total world installed nuclear power follow stockpiling plutonium and

producing High Level Waste (HLW). Then plutonium burning becomes an economical problem. while burning minor actinides and long lived fission products remains a need to increase the support of the public opinion. On the other hand the elimination of the minor actinides does not yield a radiological benefit unless the plutonium is also eliminated.

Plutonium problem is still more complicated because it is necessary to find a way for the weapons-grade plutonium final disposal.

The Secretary of Energy of the United States Government requested the National Academy of Sciences (NAS) Committee on International Security and Arms Control to evaluate disposal options for weapons-grade plutonium (20). The Idaho National Engineering Laboratory offered to assist the NAS in this evaluation by investigating the technical aspects of the disposal options and their capability for achieving high levels of plutonium annihilation.

Evaluations were performed for 13 options, involving the following reactors and accelerator-based systems:

1 - hybrid, Accelerator-Based Conversion (ABC) system,

2 – Advanced Light Water Reactor (ALWR),

3 – Advanced Liquid Metal Reactor (ALMR),

4 – Modular High-Temperature Gas-cooled Reactor (MHTGR),

5 – Molten-Salt Reactor (MSR),

6- Particle Bed Reactor (PBR).

Each option was evaluated in four technical areas: fuel status, reactor or accelerator-based systems status, waste-processing status, and waste disposal status. Each option was then rated on the basis of its deployment time.

Previous fuel development work for many of the options concentrated on uranium-based fuel. As a result, plutonium-based fuel development was limited, and demonstration of full-scale fuel fabrication was not completed. The development and fabrication of plutonium-based fuels will be on the critical path if annihilation of the plutonium is desired. All reactors and accelerator-based systems have technology development issues that must be resolved, including the examination of criticality and reactivity control during reactor operation.

Annihilation of plutonium is preferred to denaturing because it is possible to construct a nuclear explosive device from a wide range of plutonium isotopic concentrations. Calculations indicate that options with reprocessing can achieve plutonium annihilation in shorter time than non reprocessing options. Table 8 shows the maximum annihilation fraction of each option and the number of operating years required to achieve this fraction. Results from this table indicate that the options that can achieve 100% plutonium annihilation in the shortest operation time are the ABC and ALMR with plutonium fuel and reprocessing. These options are closely followed by MHTGR reprocessing option.

Waste characterisation and performance testing will be necessary for all plutonium-based fuels prior to their acceptance at a future geologic repository. Several waste disposal packages are possible, and each has disposal issues that must be considered in a comparative evaluation process. The likelihood of waste from the plutonium disposition options going to the first geologic repository is very low, and plans for a second repository have not been initiated. Monitored storage of plutonium, or its denatured form, could be required for several decades until a repository is available. Although it does not appear that any option has a notable advantage in the waste characterisation area, the ABC, ALMR and MSR options all have decreased repository requirements, such as radioactive lifetime of their final waste form and minimal criticality control issues.

Each of the plutonium disposal options is more than categorised by the deployment time. This categorisation shows the options falling into three distinct groups. The ALMR with uranium-plutonium fuel and the ALWR with mixed-oxide fuel use fuel types that are already developed. As such. these options are technitally most developed and could probably be deployed within 5 to 10 years. The ALMR with plutonium fuel, the ALWR with ternary fuel, and the MHTGR would probably require 10 to 20 years to deploy because each requires plutonium-based fuel development and testing. Because the ABC, MSR and PBR options will require significant development and design efforts, these options will probably require 20 to 30 years to deploy.

Therefore, if annihilation of the plutonium is desirable and reprocessing of the irradiated fuel is acceptable, the ALMR with plutonium fuel is believed to be the best option because of its rapid annihilation capabilities and its relatively early deployment time. If reprocessing of the irradiated fuel is unacceptable, either the ALWR with ternary fuel or the MHTGR are believed to be valid options because of their higher annihilation percentages than other non reprocessing options. If disposal is to start as soon as possible and only denaturing of the plutonium is desirable, both ALMR with uranium-plutonium fuel and the ALWR with mixed-oxide fuel are believed to be valid options because they are the most technically developed and can be deployed earlier.

This analysis confirms that the most suitable system for weapons-grade plutonium burning is the ALMR, the ALWR can help the annihilation. The hybrid system requires significant development and design efforts, but after their deployment they can solve the disposal problem in 41 years, while ALMR would need 160 years to reach the same goal.

C. Bowman claimed at SAFEWASTE '93 that the mass of the reactor discharged plutonium is now 5 times that of the weapons-grade one. This means that the most important problem to be solved would be the burning of the plutonium present in LWRS spent fuel.

In the existing nuclear power stations one third of the core can use uranium-plutonium mixed oxide fuel instead of enriched uranium. However, an increasing proportion of the isotopes ^{238}Pu, ^{240}Pu and ^{242}Pu limits the successive recycling to once or twice.

The recycling of the minor actinides in fast reactor-s and in PWRS is possible, but in the case of PWRS the unfavourable consequences for the fuel cycle (an increase of the neutron and gamma sources) are much greater.

The fast reactor technology required to control and adjust the plutonium inventory has already been developed. although the limited operational experience should be extended: fast breeder reactors exist in the USA (EBR II), in the UK (PFR), in France (Phénix, Superphénix), in Japan (Monju), in Kazakhstan (BN-350) and in Russia (BN-600).

Depending on the proportion of the ^{238}U isotope **in the core, a fast reactor can be a breeder, a regenerator or a plutonium burner.** Moreover the neutronic characteristics of these reactors allow them to use Pu containing a high proportion of even-numbered isotopes

TABLE 8 – ANNIHILATION LEVELS AND ANN1HILATION TIMES OF PLUTONIUM DISPOSITION OPTIONS ASSUMING A 75% OPERATING CAPACITY FACTOR

Concept	Number of reactors	Total thermal capacity (MWt)	Maximum annihilation level (%)	Operating time to achieve annihilation level (Y)
1. ABC	2	4260	100.0	41
2. ALMR uranium-plutonium fuel (with reprocessing)	9	4239	100.0	160
3. ALMR uranium-plutonium fuel (without reprocessing)	9	4239	10.5	17
4. ALMR plutonium fuel (with reprocessing)	9	4239	100.0	42
5. ALMR plutonium fuel (without reprocessing)	9	4239	23.6	10
6. ALWR mixed oxide fuel (with reprocessing)	2	3636	100.0	282
7. ALWR mixed oxide fuel (without reprocessing)	2	3636	17.0	48
8. ALWR ternary fuel (without reprocessing)	2	3636	65.4	32
9. MHTGR (with reprocessing)	9	4050	99.0	44
10. MHTGR (without reprocessing)	9	4050	70.2	31
11. MSR	1	3030	100.0	59
12. PBR (with reprocessing)	3	3600	100.0	50
13. PBR (without reprocessing)	3	3600	75.0	33

and therefore to burn plutonium of various origins, notably plutonium which has already been recycled one or more times in a LWR.

L.A. Baestlé (21) analysing different strategies of the deployment of LWRS and fast reactors reached the conclusion that the inventory of plutonium and Minor Actinides (MA) can be reduced 10 times, but the estimated number of the burner reactors should be comparable to that of the LWRS and these reactors must work for time comprised between 104 and 180 years.

Electricité de France presented at the Tel Aviv Meeting (22) a study calculating the quantities of Pu and MA considering a nuclear reactor installation strategy based on PWRS, European Fast Reactors (EFRs) and Liquid Metal Fast Breeder Reactors (LMFBRs) of the type without blanket.

To evaluate the long-term effects the simulation was followed up to the year 2100.

Beyond the year 2010 the total production of electricity by nuclear reactors is assumed to be stabilised at 450 TWh.

Until 2010 plutonium will be recycled in PWRS.

The strategy of intensive use of Pu will begin in 2010 with the substitution of the reactors started in the 1970s.

Until 2010 the reprocessed spent fuel will be in agreement with the PWR needs. Starting from this date the recycling capacity will be increased so that after 2030 all the spent fuel will be reprocessed.

The spent fuel cooling time considered was 3 years. Manufacturing time 2 years.

Recycling scenarios were simulated plus the no recycling one namely:

1 – no recycling:

2 – reprocessing, no recycling;

3 – reprocessing, recycling Np and Am in fast reactors;

4 – reprocessing, recycling Np, Am and Cm in fast reactors;

5 – reprocessing. recycling Pu in PWRs;

6- reprocessing, recycling Pu in PWRS and Np, Am in fast reactors;

7- reprocessing, recycling Pu in PWRS and Np, Am and Cm in fast reactors.

The weights of existing plutonium, and of stored and existing MA are presented in table 9. The analysed scenarios for multiple Pu and MAs recycling demonstrate that:

– Pu mass results approximately 540 t compared with 1350 t by 2100 for no Pu recycling,

– MAs. from 50 (scenarios 3 and 4) to 90 t (scenarios 6 and 7) compared with 200 to 300 t for strategies with no MA recycling (scenarios 1, 2 and 3).

If Cm is not incinerated, the cumulative amount of Cm will be in the order of 20 to 25 t by 2100.

This strategy, introducing massive amount of fast reactors, can reduce the Pu mass to one third and the minor actinide mass to one fourth; this means that at the end of this long interval of time in any case there will remain 510 t of Pu to be burned by hybrid systems.

On the other hand neither fast reactors nor thermal reactors can solve the problem of long-lived fission products. Hybrid systems driven by big accelerators seem to be the only solution for this task.

9. CONCLUSIONS

The most important problem in the nuclear fuel cycle is the disposal of long-lived waste. An alternative to the traditional deep burial solution is annihilation, and, in particular, plutonium destruction.

LWRS, the reactors of the present generation, can only be used for burning plutonium.

Fast reactors can destroy a large amount of plutonium and MA but it would be necessary to install them in numbers comparable to those of the LWRS. H ybnd systems can help the fast reactors in these tasks perhaps reducing the number needed.

Hybrid systems driven by large accelerators seem to be the only solution for the transmutation of long-lived fission products.

TABLE 9 – WEIGHTS OF EXISTING PLUTONIUM, MINOR ACTINIDES LOADED IN LMFBR AND MINOR ACTINIDES STORED AND EXISTING IN 2100 (t)

Scenario	1	2	3	4	5	6	7
Existing Pu	I 350	533	535	537	515	510	510
Loaded MA by year	()	o	3	4	0	5	6
Stored MA	300	205	16.5	()	'250	22	0
Existing MA	300	205	55	50	250	90	78

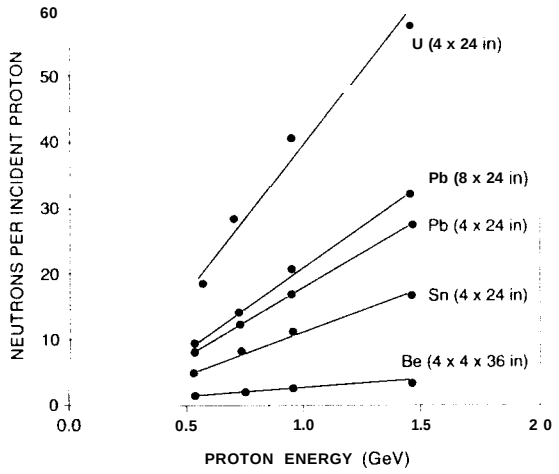


Fig. 21 – COSMOTRON experimental results: neutrons per incident proton from the bombardment of thick targets

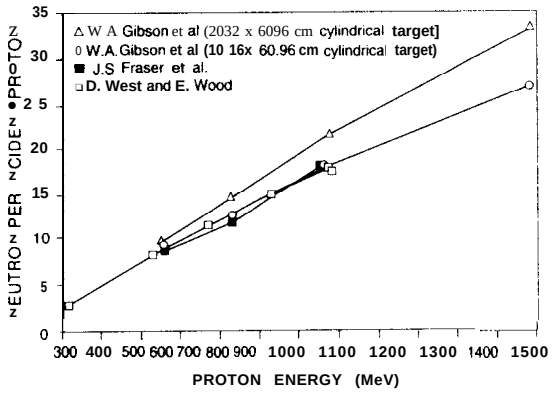


Fig. 22 – Neutron yield versus proton energy: published experimental data

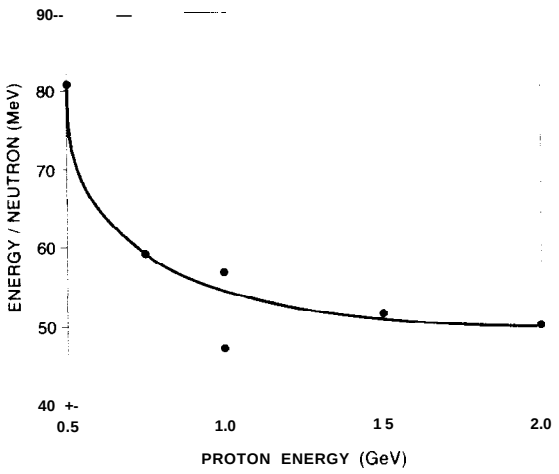


Fig. 23 – Energy required to liberate low-energy neutrons by high-energy bombardment of lead target

Experimental needs.

Figure 21 shows data from COSMOTRON experiment performed at Brookhaven National Laboratory (BNL) and figure 22 **shows** all the published data concerning neutron yield.

Table 7 presents the published data on spallation product distribution.

It is clear from this that the amount of experimental data is quite meagre. On the other hand, there are no data at all published on an area as important as the cooling of the target area. Figure 23 shows that the proton energy required for new experiments should be at values greater than 0.8 GeV.

In future, in order to improve the concept validated calculation codes, one of the most important activities in the research into intermediate energy fields could be the measurement of neutron yield, distribution of spallation products and afterheat.

As stated previously, some experiments are planned. In any case ENEA could promote or develop:

- a feasibility study for evaluating the CIRENE site (a shielded area with systems for fuel handling would seem to be ideal for realising intermediate energy physics experiments from which obtain values of fundamental parameters for the **design** of long-lived radioactive waste burning plants):

- the following experiments:

1 - target 4x4 cm, 0.03 cm thickness. 20 μA- 1500 MeV beam for measurement of the distribution of spallation products (ATHENA experiment):

2- cylindrical target 6x 18 cm (JAERI data), 1 mA- 1500 MeV beam for measuring the number of neutrons per proton;

3 - afterheat measurements, with beam characteristic and target similar to COSMOTRON experiment:

4- fuel assembly like CIRENE with ThO_2 for evaluating ^{233}U build-up.

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NOMENCLATURE

AB	Accelerator Breeder
ABC	Accelerator-Based Conversion system
AECL	Atomic Energy of Canada Limited
ALMR	Advanced Liquid Metal Reactor
ALWR	Advanced Light Water Reactor
AMSB	Accelerator Molten-Salt Breeder
APT	Acceleration Production of Tritium
ATW	Accelerator Transmutation of Waste
BNL	Brookhaven National Laboratory
CCL	Coupled Cavity Linac

CFR	Code of' FederalRegulation
CRNL	("balk River N uclear Laboratories
CURE	CleanUse of' Reactor Energy
DTL	Draft-Tube Linac
EFR	European Fast Reactor
EMTF	Electronuclear Materials Test Facility
FB	Fusion Breeder
FFTF	Fast Flux Test Facility
-fLW	High Level Waste
IFMSB	Impact Fusion Molten-Salt Breeder
IFR	Integral Fast Reactor
IHMSB	Inertial-confined fusion Hybrid Molten Salt Breeder
LAMPF	Los Alamos Meson Physics Facility
LINAC	LINear Accelerator
LMFBR	Liquid Metal Fast Breeder Reactor
LWR	Light Water Reactor
MA	Minor Actinide
MHTGR	Modular High-Temperature Gas-cooled Reactor
MSB	Molten-Salt fissile producing Breeder
MSBR	Molten-Salt Breeder Reactor
MSR	Molten-Salt Reactor
MSRE	Molten-Salt Reactor Experiment
NAS	National Academy of Sciences
NEACRP	Nuclear Energy Agency Committee on Reactor Physics
ODS	Oxygen Dispersion Strengthened alloy
ORNL	Oak Ridge National Laboratory
PBR	Particle Bed Reactor
PWR	Pressurized Water Reactor
rf	radio frequency
RFQ	Radio Frequency Quadrupole
TAMU	Texas A & M University
THORIMS-NES	THORIum Molten Salt Nuclear Energy Synergetics
TRU	TRans-Uranic
ZEBRA	Zero Energy BReeder Accelerator

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