Concepts of Accelerator Based Transmutation Systems

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Abstract

The search for an optimum strategy of rad-waste disposal is an extremely complex task depending on economic parameters and long term hazard estimates for which public consent still has to be achieved. Among the different options attention focuses on the possibilities of transmuting radio-nuclei such as minor actinides and long lived fission products by neutrons generated in spallation processes. At present research follows two main lines. One investigates the construction of fast subcritical actinide assemblies which are driven by spallation neutrons. As these subcritical systems rely mainly on neutron self-multiplication they require a relatively high actinide inventory, but only a low beam current. Studies show that in this case a multistage cyclotron arrangement could be used instead of a more expensive linear accelerator. The other line uses the spallation process to generate an intense thermal neutron field in which actinides are fissioned and long lived fission products converted. This project works with a relatively small radioactive inventory, but it requires a high current accelerator, based on technology at the edge of feasibility.

Introduction and Historical Review

More than ever before public opinion questions the underground disposal of long lived radioactive waste. In particular the α-emitting minor actinides (*Np*, Am, Cm etc.), the residues of plutonium after reprocessing and some β-emitting long lived fission products (^{135}Cs, ^{90}Sr, ^{99}Tc and ^{129}I) are considered to constitute an undue long-term risk.

Over the years national and international studies have been performed targeting on the reduction of the potential long-term hazard of radioactive waste generated by nuclear reactors. In a recent study based on the CURE approach [3], which is a waste partitioning process derived from the well established PUREX process and the newer TRUEX process, the long term radioactive ingestion toxicity for waste stream scenarios has been studied. The ingestion hazard, in terms of how much water is required in order to dilute the material to reach safe drinking water

standards, is compared to that of a typical natural uranium ore in Figure 1. The top curve shows the toxicity of the entire waste stream, assuming a once-through fuel cycle. Even after 10,000 years this waste remains two or three times more toxic than the reference uranium ore. If plutonium and uranium are separated, the toxicity reaches the reference value after approximately 1000 years as shown in the second curve. The additional removal of the minor actinides, i.e. neptunium, americium, and curium results in the third curve reaching the equal toxicity point after 3 to 4 centuries. In the remaining waste stream four isotopes are problematic: ^{90}Sr, ^{137}Cs, ^{99}Tc, and ^{129}I. The first two have half-lives of around 30 years and contribute significantly to the short term radio-activity and heat load, thus constituting a packaging problem. The latter two have very long half-lives, as well as high mobility in the case of leakage and water intrusion. If these four isotopes can be removed from the waste stream too, than the lowest toxicity curve is obtained reaching equal-toxicity within about 30 years. Similar hazard estimates have been worked out by several other authors (e.g.: [28, 14, 45]).

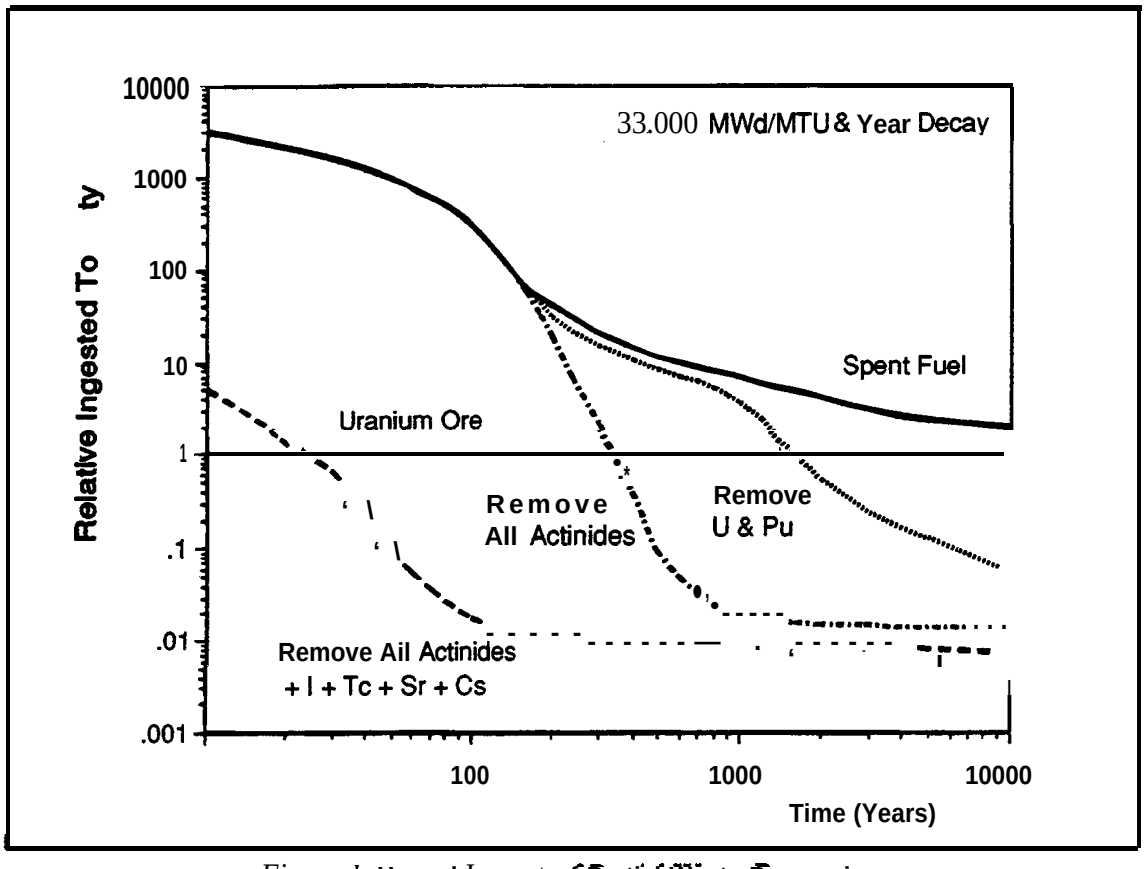


Figure 1: Hazard Impact of Partial Waste Processing

It was in the second half of the seventies that the OECD Nuclear Energy Agency assigned the Commission of the European Community a leading role in a chemical separation and transmutation exercise.

In this first internationally coordinated R&D programme on "Management and Storage of Radio-active Waste" R&D was carried out by the Joint Research Centre in collaboration with Community laboratories and industries financed by a shared cost action programme.

The main studies developed in this field were the following:

- Nuclear transmutation studies aimed at achieving an assessment of the by-product actinide (BPA) transmutation and its impact on the fuel cycle in terms of feasibility, benefits, costs and risks. LWR, FBR and HTR reactor physics calculations and conceptual design of fuel elements were part of these studies.

- Partitioning studies aimed at demonstrating the chemical feasibility of plutonium and BPA separation from HLW. Laboratory investigations and engineering assessment of HLW partitioning flow-sheets were prepared in this context.

Thus the Joint Research Centre organised, under the sponsorship of NEA, a First and Second Technical Meeting on Nuclear Transmutation of Actinides at the JRC Ispra in 1977 and in 1980.

The JRC has also contributed to the IAEA Coordinated Research Programme "Environmental Evaluation and Hazard Assessment of the Separation of Actinides from Nuclear Wastes Followed by Either Transmutation or Separate Disposal".

A report summarizing results and conclusions of the assessment studies on nuclear transmutation dealing with reactor physics, implications on fuel pin design, economic evaluations and risk assessment was finally issued in 1983 [28]. A parallel report on the chemical separation of BPAs from the HLW has also been published [17].

It is worthwhile highlighting some of the main conclusions of the Second Technical Meeting on Nuclear Transmutation of Actinides at the JRC Ispra [25]:

"On the basis of the various papers presented, of the findings of the workshop sessions and of the opinions prevailing during the final plenary session, the following conclusions and recommendations were reached:

- Partitioning and transmutation of by-product actinides is considered feasible in the sense that potential solutions of the technical problems involved exist, though a very large research development and demonstration effort over a few decades would be required before they could be applied under realistic conditions.

- The implementation on an industrial scale of partitioning and transmutation would *involve considerable modification of the fuel cycles and their associate reprocessing and fuel fabrication plants inter alia, on account of the high neutron emission and decay heat release of the by-product actinides (see conclusions of Workshop B and C).*

- The presence of by-product actinides in reactors would not raise particularly severe reactor physics problems (see conclusions of Workshop A), though there would be significant metallurgical and safety problems.

- The effort required appears appreciably less severe if partitioning and transmutation is applied in an advanced fuel cycle (e.g. the FBR fuel cycle) owing *to the expected introduction of new*

technologies such as high efficiency plutonium recovery from various waste streams and remote fuel fabrication.

- Of the various strategies which could be used, homogeneous recycling in FBR's appears the most suitable both with respect to transmutation efficiency and to compatibility with the FBR fuel cycle, which is likely to be the predominant fuel cycle during the period when partitioning and transmutation might be implemented.

- On the basis of the existing ICRP figures for ingestion of radionuclides, the reduction in the long-term potential hazard of radioactive waste by partitioning and transmutation appears to be marginal since the potential hazard of disposal in suitable selected geological formations is assessed to be small.

- If further reduction of the long-term potential hazard of radioactive waste disposal is envisaged, efforts should preferably be directed to improving techniques of alpha-waste reduction, conditioning and disposal. These objectives seem more readily achievable than the implementation of a transmutation strategy.

- Optimum schemes for reduction, conditioning and disposal of alpha-wastes should be derived from an analysis of fuel cycle models, risk models and fuel cycle strategies as outlined in the conclusions of the Workshop D. Preliminary cost/benefit calculations on the partitioning and transmutation of by-product actinited indicate that this is unlikely to be the optimum scheme. "

It is interesting to note that the conclusions drawn in 1980 reflecting the opinion of an international working group mentioned neither nuclear waste transmutation by accelerators nor even single purpose actinide burners. Even though these concepts were known at the time, they were considered to be too futuristic. Actually the whole exercise was carried out with the assumption that only established state of the art technologies should be used.

In the meantime a new fast reactor concept (the Integral Fast Reactor) in which actinides are recycled - has been developed by ANL and US industries. It is based on metallic fuel elements and a new on-site, non aqueous, reprocessing scheme. This reactor with enhanced inherent safety features is capable of recycling and transmuting its own actinides. As it was not designed as an actinide burner only a limited amount of actinides from LWR fuel element reprocessing can be added to its fuel cycle without hampering its inherent safety features. It also seems that in this reactor long lived fission products are not efficiently removed.

Specific actinide burners with improved transmutation performance have been envisaged in Japan. To take advantage of the more favorable fission to capture ratio in a hard neutron spectrum actinide fuelled fast reactors were the first choice. During the design studies it turned out that, because of the small delayed neutron fraction in actinide fuel and a short neutron life-time coupled with a small Doppler coefficient, such a reactor is more difficult to control than a normal FBR and therefore does not satisfy inherent safety requirements.

One way to operate such systems safely is to run them in a subcritical state and to feed them by "external" spallation neutrons. If the facility is sufficiently sub-critical small reactivity insertions do not lead to dangerous power excursions. For example, in a system only 3$ sub-

critical a sudden reactivity insertion of 1.1$ leads to a 44% power increase during the following second. A shut-down of the proton beam after this second would instantaneously decrease the power to 20%. This safety margin allows for more flexibility in the design since limitations by a non-negative sodium void coefficient and effects caused by poisoning and isotope changes due to burn-up are much less important. Otherwise the transmutation characteristic of an accelerator driven sub-critical facility is quite similar to a corresponding actinide burner reactor. It is therefor mainly the safety argument which moved accelerator based systems into the focus of interest.

In what follows we summarize firstly the present state of the art in accelerator design. Then we discuss the physics of accelerator driven systems. Finally, we give an account of prototypical fast and thermal transmutation systems presently under study.

Accelerators and Targets

As an alternative to the incineration of actinides and fission products in power reactors even in 1974 studies were made to employ charged particles (usually protons or deuterons) accelerated to energies above the spallation threshold. Without going into any details of spallation theory we just recall that charged particles exceeding an energy of several hundred MeV are able to penetrate the Coulomb barrier of heavy nuclei. In a fission-like evaporation process which follows such a reaction a shower of secondary particles, consisting mainly of neutrons and mesons is released. These high energy particles trigger similar reactions until the whole cascade loses its energy. The neutron yield increases with the charged particle energy and the mass number of the target material.

The spallation process has attended great interest as an intense neutron source. In targets of large mass numbers spallation cascades release per charged particle (usually a proton or a deuteron) several tens to hundreds of neutrons (depending on the target material) with an energy distribution similar to that of the fission spectrum.

Existing accelerators in the medium energy range (0.8 -2.0 GeV) are mainly dedicated to research and therefore limited to low currents as for example LAMPF at LASL with 1 mA beam power. For linear accelerators, based on new technologies realistic design goals worked out by LASL are specified around 250 mA (higher currents suffer from space charge problems). They are based on advances of ion sources for beam production, on the availability of radio-frequency quadruples injecting the beam into the linac, more efficient klystrons and improved beam focusing techniques.

Recent studies of tritium production by accelerators (APT) led to the development of a detailed design for a 1.6 GeV, 250-mA proton LINAC [5]. Its design was partly stimulated by the demanding requirements of SDI's neutral particle beam program. In this context a “classical” design proposal is based on LAMPF. Figure 2 shows the reference APT accelerator configuration. The lattice average acceleration gradient in the coupled cavity linac part of the design is 1 MV/m with a beam power of 250 mA.

A more recent proposal also drawn up by LASL applies the latest superconducting technology. It allows for a more efficient use of radio frequency and a three times higher

gradient than earlier designs. This results in substantial shortening of the accelerator as well as a substantial reduction in the cost estimate of construction. A 800 MeV linac based on this technology would be about 270 m long. The current assumed at 100% duty cycle is estimated at 93-mA for 1.6-GeV and 220 mA for 800 MeV.

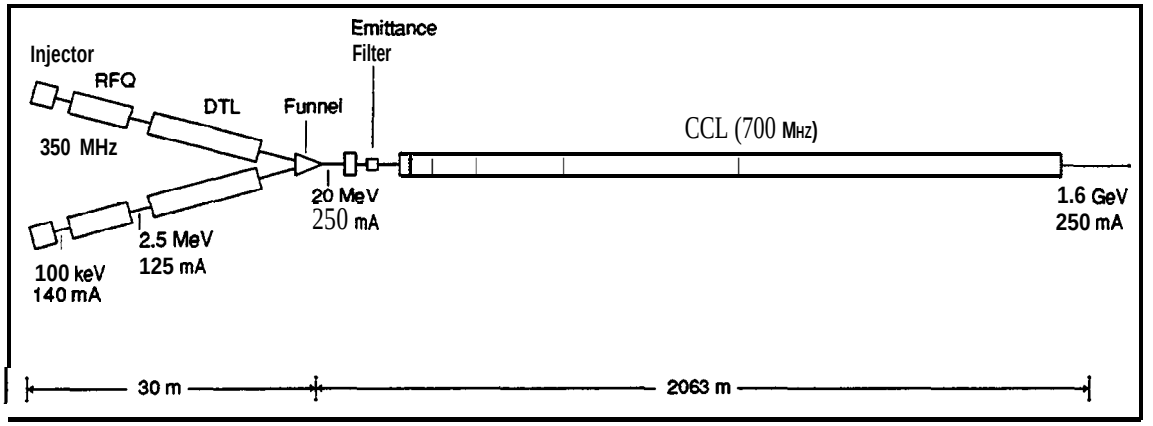


Figure 2: Reference APT accelerator configuration [5]

Besides linear accelerators which have been described in detail in many publications, the potential of less expensive cyclotron accelerators has also been investigated. Cyclotrons will surpass the mA threshold in the near future, as for example SINQ at PSI Villingen/Würenlingen. But their maximum current will be confined to about 10 mA.

At low energies the maximum achievable beam current of a cyclotron is rather limited. This consideration led to the concept of a so-called "multistage-parallel" cyclotron arrangement consisting of a number of low energy, low current cyclotrons feeding one high energy cyclotron. A preliminary study carried out in 1986 [3] led to a multistage cyclotron concept shown in Figure 3 providing 15 mA at an energy of at least 2-GeV. It would require the following assembly:

- three 60-70 keV ion sources
- a low energy 2 MeV, 5 mA injector stage consisting of three radiofrequency quadruple linacs (36 MHz) as developed by Müller from Darmstadt (Split Coaxial Resonator)
- an intermediate stage made of the same number of four sector cyclotrons with a phase width of 36° and frequency F_{Hf} = 36 MHz accelerating the 5 mA proton current to an upper limit around 200 MeV
- a final stage 15 sector cyclotron being fed by the three intermediate stages (3x5 mA at 200 MeV). This cyclotron works at 3x36= 108 MHz using 12 acceleration cavities at 600 keV. It can reach a proton energy of 2 to 3 GeV at a beam power of 30-45 MW.

In this assembly the following conditions must be satisfied:

- the time structure of the beam: the RFQ linac should work at the same radiofrequency as the injector (intermediate stage) cyclotron in order to avoid any beam loss. This implies the choice of a particular structure of the RFQ, namely that developed by Müller, called the Split Coaxial Resonator (SCR).
- Input-Output Conditions: the product *Br* where *B* and *rare the* average magnetic field and

radius respectively, must be conserved in the injection-ejection process of the different cyclotrons.

- Intensity Addition: raising the intensity to the required value in the first stage is achieved

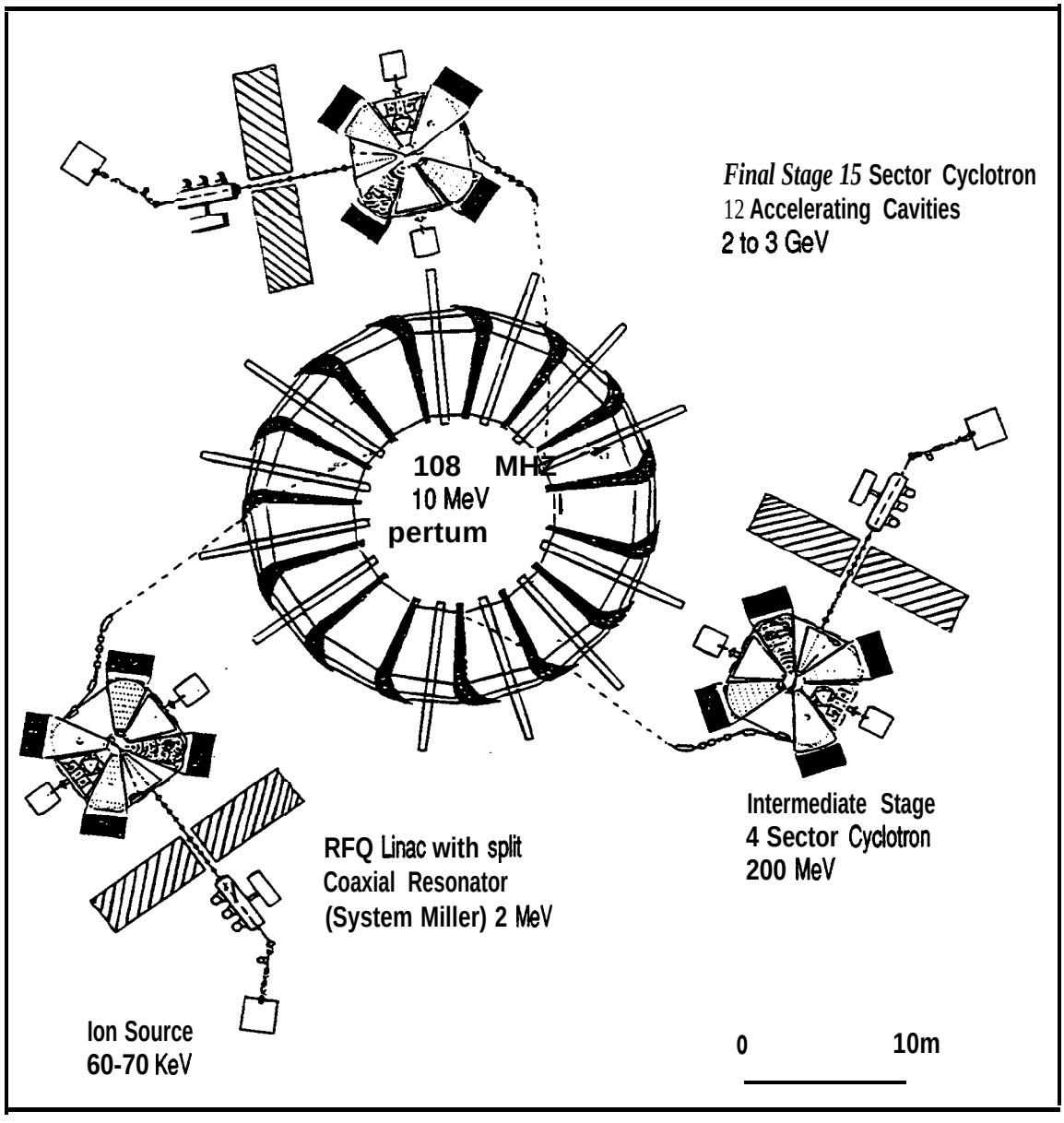


Figure 3: A schematic Multistage Cyclotron Arrangement

by adding the three beams of the intermediate stage. This requires the accelerating cavities of the final stage to work at a frequency three times larger than those of the injector cyclotrons.

Accelerator Driven Transmutes

Basic Physical Considerations

Three options of converting long lived radio-nuclei into short-lived isotopes are considered:

- the direct interaction of accelerated particles (protons) with nuclear waste materials; i.e. the direct **spallation** of **actinides** and/or fission products.
- the transmutation of long lived **rad-waste** into fast decaying isotopes using neutrons generated by a **spallation** process,
- the transmutation of long lived **rad-waste** into fast decaying isotopes in an accelerator **driven** "reactor-like" subcritical system.

At present **R&D** investigates the latter two options. One explores the possibility of moderating **spallation** neutrons **in** D_2O to generate an intense thermal neutron field in which actinides are fissioned and long lived fission product converted into fast deficaying isotopes. The other investigates the possibility of constructing fast subcritical **actinide** assemblies driven by **spallation neutrons.**

The Direct Spallation** of **Fission Products:

The original idea of exploiting the **spallation** process to transmute **actinides** and fission produets directly soon had to be given up. It turned out that the required particle currents were much larger than the most optimistic theoretical accelerator design goals which are around 300 mA. Indeed, it had been shown that the yearly destruction rate of a 300 **mA** proton accelerator would correspond only to a fraction of the waste generated by one LWR of 1 GW_e in the same period of time [12, 45].

***The Direct Use** of **Spallation** Neutrons*

To use only the **spallation** neutrons as they are generated in a proton target, the fission products would be placed around the target. For best efficiency, depending on the material to be transmuted, either the fast neutrons would be used as they are emitted from the target or they would be slowed down by a moderators to energy bands with higher transmutation cross sections as for example the resonance or the thermal region.

Assuming that it is possible to make all the **spallation** neutrons available for the transmutation process the following amount of energy is necessary to transmute the fraction q_{fp} of radio-nuclei per fission process in a nuclear energy system

$$E_{fp} = q_{fp} \frac{P_b}{n_{sp}} \frac{1}{\eta_b \eta_T} \; [MW] \qquad (1)$$

where q_{fp} = **fraction** of fission products to be transmuted
P_b = proton energy
n_{sp} = number of neutrons generated by one proton
η_b **efficiency** of **converting** electricity into proton **beam energy (= 0.5)**
η_T = efficiency of converting thermal energy into **electricity** (=0.33)

In the case of a 1.5 GeV proton beam emitting 50 neutrons **per spallation** in a lead target the transmutation of $^{99}Tc, ^{129}I, ^{135}Cs, ^{90}Sr, ^{85}Kr$ *and* ^{93}Zr (constituting 28 % of all fission products) would require 0.285 30/0.5/0.33 = 51.3 MeV to transmute the fission product fraction of one fission process. This is 51.3/200 = 26 % of the total power production of the energy system under consideration ! Because of the very optimistic assumptions made in this estimate the real percentage of energy required would even be higher. Together with the cost for reprocessing it would make this type of accelerator transmutation prohibitively expensive, at least in a commercial nuclear energy system.

Accelerator Driven Subcritical Assemblies

To improve neutron economy there **remains,** however, the possibility of multiplying the **spallation** neutrons in a subcritical assembly. In such a system the main part of transmutation is performed by fission neutrons in a reactor-like facility. Technically this is realized by surrounding a proton target region by fissionable material in a cooling system. **In** most designs a circulating liquid lead-bismuth alloy is proposed to remove the high specific heat released in the target. It must, however, be mentioned that the specific heat production per neutron is considerably lower than in a fission process (30 MeV against 80 MeV).

First the power production P_{fi} of a subcritical assembly fed by **spallation** neutrons is quantified:

$$P_{fi} = n_{sp} \frac{a \cdot k}{\nu(1-k)} \frac{i}{C} \; `f \qquad (2)$$

where: k = multiplication factor

a = importance of the target position and target neutron energy distribution (usually $a>1$ for a central target position

v = mean number of neutrons in a fission process

E_f = power release per fission (=3.1.10-10 W)

n_{sp} = neutron yield from one proton

i = proton current

C= proton charge (= 1.6 10^{19}A see)

In Figure 4 the power production of an accelerator driven facility is shown as a function of sub-criticality (1-k). It was assumed that a proton beam of 1 GeV and 1 mA impinges on a *Pb-Bi* target releasing 50 neutrons per **spallation** with an importance of $a=1$. It leads to

$$P_{fi}(1 \; *) \cong \frac{k}{4(1-k)} \; [MW] \qquad (3)$$

It can be seen that **near** criticality a 1 mA current already generates a relatively high fission power. For k = *0.97* more than 100 MW can be achieved.

The additional neutrons **from** the subcritical system as well as its fission power which can be transformed into electricity, are now exploited to run the transmutation process. Expression 4 **quantifies the energy** required to transmute a fraction q_{fp} of fission products in such a system. A positive sign of E_{fp} means that there is even a surplus of energy, while a negative sign indicates the need to add energy to the system from outside.

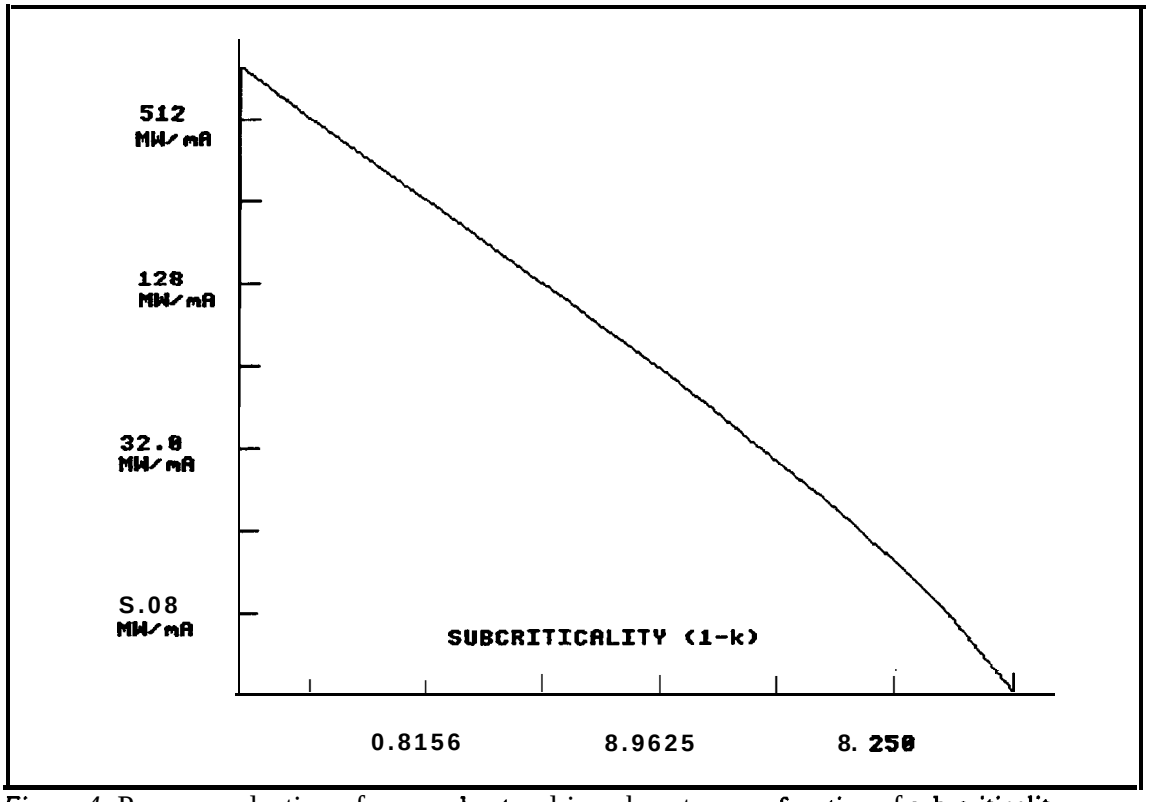


Figure 4: Power production of an accelarator driven booster as a function of sub-criticality (1-k@) assuming a proton beam of 1 mA at 1.5 GeV entering a lead target leading to a release of 50 neutrons per proton.

$$E_{fp} = \frac{n_{sp}\ \frac{k}{\nu(1-k)}\ E_f\ \eta_b \eta_T}{n_{sp}\left[\left(1 - \frac{k}{\nu(1-k)}\right)\eta_{fp} + \frac{k}{1-k}\left(\left(1 - J? - \frac{q_{fp}}{\nu}\right)\right)\right]} [MW] \qquad (4)$$

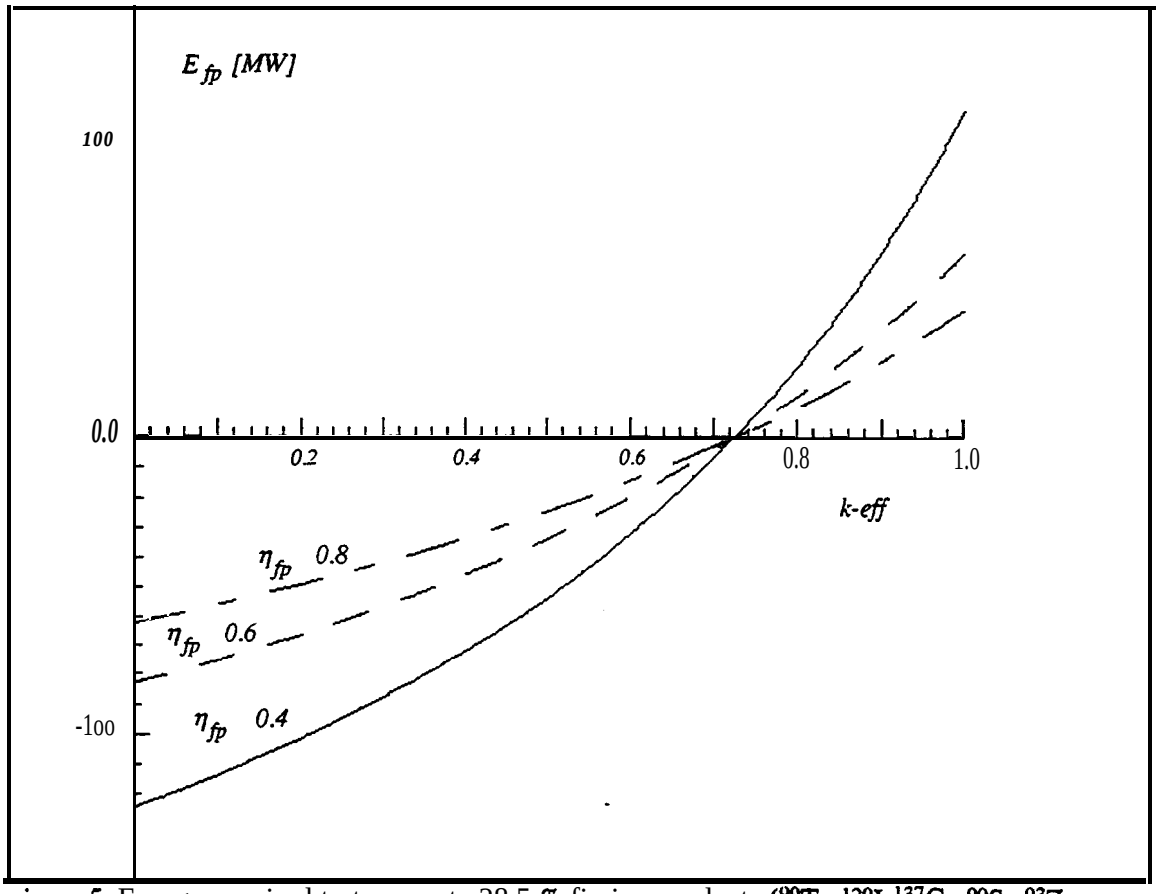
where

$$\eta_{fp} = \frac{\Sigma_a(FP)}{\Sigma_a(FP+Fuel+StructMat.)}$$

The condition for break-even or a positive energy balance is given by

$$k \geq \frac{1}{1 + \frac{n_{sp} E_f \eta_b \eta_T}{P_b \nu}}$$

Note that this expression is independent of the proton current and to a large extend also of the type of system considered. For a lead target and a proton beam of 1 to 2 GeV *break-even re*quires a *k* value near 0.75. But the amount of nuclei transmuted depends on the power of the system and therefore on the proton current as shown by Equation 2.



igure 5: Energy required to transmute 28.5 % fission products (^{99}Tc, ^{129}I, ^{137}Cs, ^{90}Sr, ^{93}Zr, ^{85}Kr) assuming $P_b=1000MeV, \nu=3.0, n_{sp}=34.3, \eta_b=0.5, \eta_t=0.33, q_{fp}=0.285$.

A more detailed analysis can be performed describing the transmutation facility by a set of time dependent differential equations as shown in [5]. Assuming equilibrium between the addition of waste and its burn-up the time derivatives can be set to zero transforming the system into a set of simultaneous equations. In the case of the LASL project 4 equations were used to describe the equilibrium conditions between the isotopes being considered.

As the most important nuclear waste materials which should be transmuted are the minor actinides, they are also the first candidates to "fuel" the subcritical facility.

Conceptual Designs of Fast Neutron Transmutation Systems

An Early Proposal by BNL - JRC

In 1985 the authors [4] proposed for the first time a hybrid system consisting of a sub-critical fast neutron minor actinide transmute, driven by proton generated spallation neutrons. In this proposal it was assumed that a multi-stage cyclotron, as described above, should serve as a proton accelerator. The possibility of reaching the GW range by relatively small proton currents

around 10 mA in slightly sub-critical systems justified the investigation of fast neutron transmutation facilities driven by a multistage cyclotron rather than a linac.

During steady-state operation the number of fissions depends mainly upon the degree of neutron self-multiplication in the subcritical system. The total number of fission is expressed by

$$N_{tot} = N_{(spallation + high\ energy\ fusion)} + N_{(low\ energy\ fission)} = N_{S+HF} + N_{LF}$$

$$= N_{(S+HF)} + S_h \nu \frac{k_{eff}}{(1 - k_{eff})} \qquad (5)$$

where:

N_{tot} = total number of fission per incident proton
N_{S+HF} = number of spallations and cascading high energy fissions per incident proton
S_h = number of neutrons generated by spallation + high energy fissions per incident proton
v = the number of neutrons emitted per fission
and
k_{eff} = *effective* multiplication factor for classical low energy fissions

In our considerations we assumed a V-shaped target region. The proton beam enters at the "bottom" of the V where it is de-focused by magnets so that the protons are spread out over the whole target volume. Figure 6 shows the subcritical minor actinide transmute driven by a proton beam which irradiates the surface of the subcritical core uniformly.

In order to make the power distribution in the subcritical system more or less flat (which is important from the heat removal point of view) the medium energy proton beam is shredded by a magnetic field at the entrance of the beam expansion cavity. This has the additional advantage that the effects of radiation damage due to the irradiation of the high intensity (medium energy) proton beam are reduced.

A disadvantage of this system is the large leakage of neutrons from the surface which is irradiated by protons. In order to use these leakage neutrons effectively a fertile material blanket region is installed around the beam expansion region. In the usual manner the fertile materials of this region are then converted to fissile materials by neutron capture.

If the surface on which the protons are injected is large, a long beam expansion region is required resulting in a large containment building housing the subcritical reactor. Because of the small cross sections in the higher energy range the assembly would require a relatively large actinide fuel inventory, most probably oxide or metal, and liquid metal or gas cooling.

Fission products like *Tc* and *I* could be placed in a moderator matrix (metal hydride or a metal deuteride) in the reflector. As was shown by [47] absorption in the epithermal resonance region may lead to effective transmutation. As there is enough space in the reflector *Tc can be* diluted so that the self-shielding effects can be mitigated.

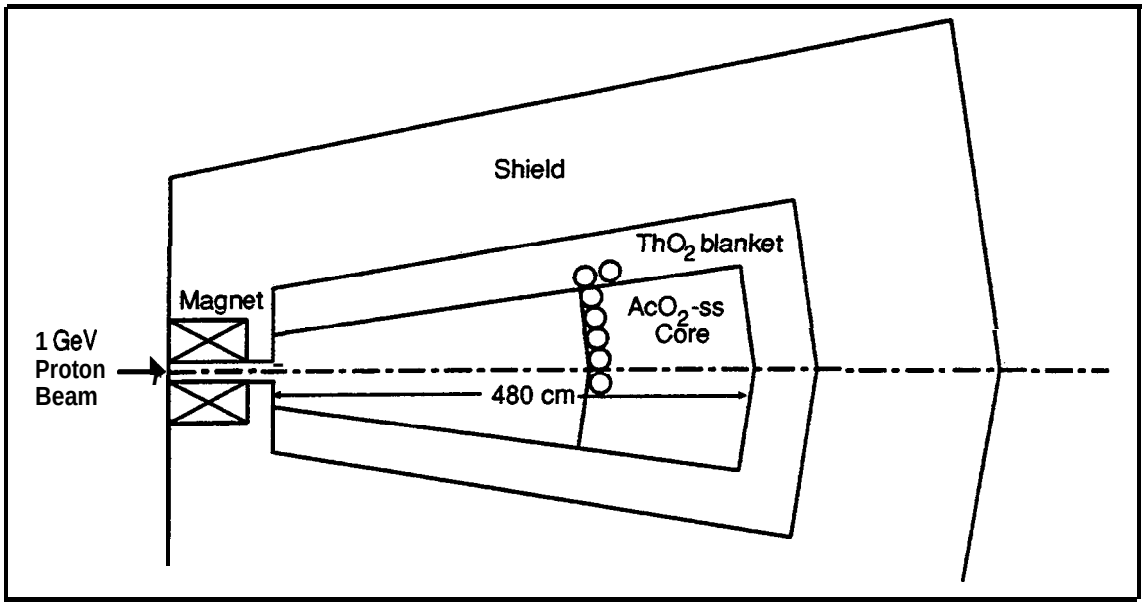


Figure 6: Schematic sub-critical target design

In the following we give an example of bum-up calculations carried out for this subcritical accelerator-driven fast actinide assembly. In the quantitative estimate of transmuting the actinides produced by ten light water reactors we assume that the subcritical target area exposed to the proton beam contains fuel rods consisting only of minor actinide oxides. This is, of course, a simplified assumption, since all reprocessing plants work with less than 1009'o separation efficiency. In reality the actinide waste will always contain some amount of uranium and plutonium, which would both have a negative effect on the transmutation rates presented here.

Our studies were carried out for systems cooled by sodium or helium. In both cases the fuel pellets were assumed to consist of actinide oxides cladded by steel canes. The following design parameters were chosen: SS Clad, outer diam.: 0.600 cm; Pellet, out diam.: 0.510 cm; Fuel pitch: 0.75-0.85 cm; Active length: 80 cm. The target zone was surrounded by a blanket region to utilize the large fraction of leakage neutrons which escape from this system having an unfavorable surface to volume ratio.

To calculate spallation effects and high energy fissions the nucleon-meson transport code NMCT - BNLF was applied. At low energies nuclear reactions were determined by conventional reactor codes.

The outcome of the study performed for a sodium or helium cooled system is summarized in *Table II. The* first surprising result is the low beam current required to incinerate the actinides produced by ten 1000 MW_e reactors. Depending on the beam power (1 to 3 GeV) and the self-multiplication of the target the required beam currents are between 5 and 15 mA. In the case of a sodium cooled system this requirement would rise by a factor of less than 2.

As a by-product this transmute could produce annually at least 100 kg of fissionable material in a uranium or thorium blanket and an excess electricity of 230-260 MW beyond the 40 to 70 MW required to operate the cyclotrons.

coolant	k_{eff}	Proton Beam Power *[MW]*	Beam Current *[mA]*			Reactor Power *[MW_t]*	^{233}U Production *[kg]*
			1 GeV	*2 GeV*	*3 GeV*		
Na	.90	27.9	27.9	14.0	9.3	900	85
He	.95	13.0	13.0	6.5	4 3	900	103

Table II: Characteristics of an accelerator-driven helium cooled sub-critical fast assembly transmuting the yearly actinides of 10 LWRS (1 GW_e)

The JAERI Projects

In the framework of the OMEGA project (Options Making Extra Gains from Actinides and fission products) JAERI launched a broad programme on the development of accelerators and target systems. In this context homogeneous and heterogeneous subcritical core structures are investigated.

JAERI heterogeneous Accelerator Driven System Studies:

First a project similar to the one mentioned above was studied by a JAERI group [37]. This design considered a horizontal proton target made of tungsten, 60 cm long in beam direction and with a vertical cross section of 10x1OO cm^2. Its fissile region is cooled either by sodium or a Pb-Bi alloy. To harden the neutron spectrum metallic fuel in the form of *Np-22Pu-20Zr* and *AmCm-35Pu-5Y* with high phase stability is envisaged (melting point at 900°C). The fuel assembly is similar to that of the LMFBR. Fuel pins with an active length of 100 cm are arrayed on a regular triangular pitch. The multiplication factor is assumed to be between 0.86 and 0.95. Several parametric studies referring to this system are summarized in Table III.

coolant	*Na*	*Pb-Bi*	*Na*	*Pb-Bi*
Proton Beam Current *[mA]*	22.6	*75*	182	*5.4*
k_{eff}	.92	*.86*	.94	*.95*
Actinide Loading *[kg]*	2866	*2013*	2682	*1584*
Bum-up rate *[%]*	7.0	*6.9*	4 3	*2.7*
weight *[kg]*	202	*139*	114	*42*
Waste Equivalent of 3 GW_t LWRS	7.6	*5.3*	4 3	*1.8*

Table III: Performance Characteristics of JAERI Fast Neutron Transmutation Proposal

JAERI's new proposal for a transmute employs a cylindrical target in *r-z* geometry bending the medium energy proton beam from the horizontal to the vertical direction. The conceptual scheme of this system is shown in Figure7. The project is backed by a R&D programme for a powerful proton accelerator in the 1.5 GeV and 10 mA range.

Injecting the protons into the target vertically requires only a small beam spreading, thus

facilitating neutron shielding and building lay-out. But the disadvantage of this Iodized neutron source design is a strong gradient in the radial power distribution in the case of a small multiplication factor ($k_{eff} \leq 0.9$) resulting in more difficult heat removal condition.

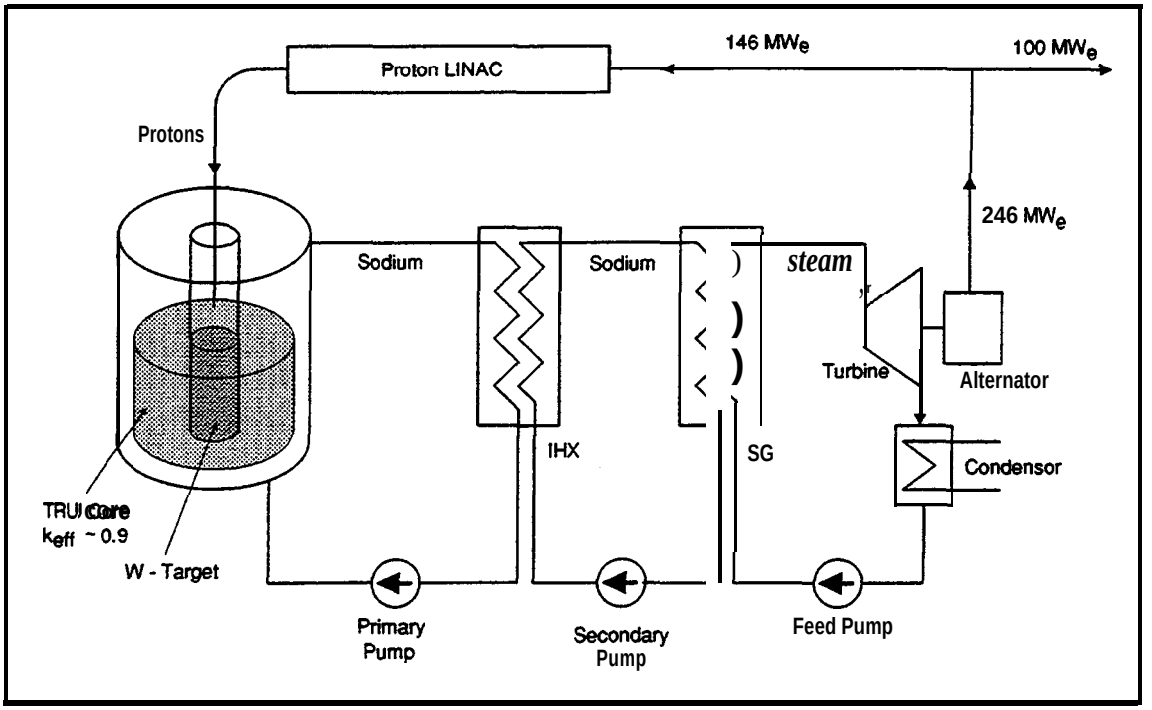


Figure 7: Conceptual flow diagram of JEARI's actinide transmutation plant

The reference parameters for this system are:

Proton Beam Current	*39 mA*	coolant	*Sodium*
k_{eff}	*0.89*	Power Density max.	930 $MW \cdot m^{-3}$
Actinide Loading	3160 *kg*	ave.	*400* $MW \cdot m^{-3}$
Bum-up	250 $kg \cdot y^{-1}$	Temperatures:	
Thermal output	820 MW	Coolant outlet	473 c
Neutron Flux	$4.10^{15}\ n \cdot cm^{-2} \cdot s^{-1}$	Fuel	890 °C
Mean Neutron Energy	690 *keV*	Clad	528 "C

JEARI's Accelerator-Driven Continuous Transmutation Molten Salt Facility

Conceptual design studies of a molten salt subcritical target system were carried out by JAERI [10]. The technical feasibility of these systems was demonstrated by the Molten Salt Reactor designed and operated at ORNL in the sixties. In the JAERI considerations the main requirements are a high volubility of TRU along with adequate nuclear characteristics and physico-chemical properties of the salt melt. Salts which satisfy these demands are either ^7LiF and BeF_2 or *NaCl in the* following composition: $64NaCl+31MACl_3+5PuCl_3$, (where MA=Np,

Am, Cm). From a reactor physics point of view *NaCl* might be the better choice. Because of its higher atomic weight the neutron spectrum will be harder than in the case of fluoride compounds, - a fact which favours actinide transmutation.

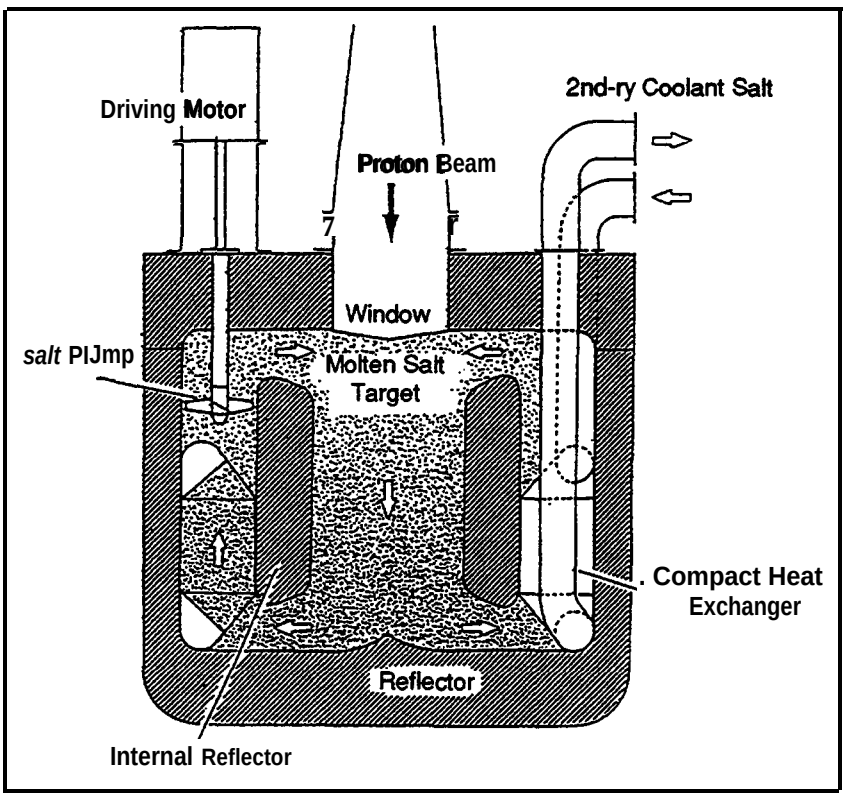


Figure 7: The JAERI Molten Salt Project

The actinides dissolved in the salt-melt serve as fuel for the sub-critical system and at the same time as target material for the proton beam. This leads to an important simplification of the whole system from the point of view of cooling as well as the chemical separation of spallation products and especially the target design.

The main advantage of a molten salt system is the possibility of re-processing the fuel continuously and of separating short-lived and stable fission products at the same time. The on-line separation of the fission products is envisaged to be made by a cold trap and an electro-chemical separation method. Furthermore, molten salt systems have the enormous advantage that the rather difficult and R&D intensive requirement of actinide fuel fabrication can be avoided. The rather steep power gradient around the entrance cone of the proton beam is in this type of system less of a problem than in a heterogeneous one. In a heterogeneous target driven reactor frequent shuffling of the solid fuel is necessary.

From the safety point of view one can assume that core meltdown accidents can be excluded. On the other hand corrosion together with radiolysis might pose major problems. In chlorine systems it is a main requirement to avoid any contamination with water.

Preliminary transmutation estimates result in a yearly total amount of TRU transmutation of 10 $kg \cdot mA^{-1} \cdot y^{-1}$ for a 1.5 GeV proton beam, a k_{eff} = 0.92 and a thermal output of the system of 32 MW_{th}/mA. The estimates show that the TRUS produced by 10 LWRS of 1 GW_e each can be transmuted by a proton beam current of 25 mA.

The Accelerator-Based Transmutation Proposal from BNL

The PHOENIX Concept developed at BNL [42] consists of modules of accelerator driven

sub-critical lattices containing minor actinide fuel. Classical fast reactor technology, such as oxide fuel elements and sodium cooling is assumed. Each module resembles the core of the Fast Flux Test facility (FFTF) with a k_{eff} = 0.9, however. From 1 to 8 target modules are aligned in front of a 104 mA beam of 1.6 GeV protons. The whole reactor-module serves as a target for a proton beam being expanded before entering the core region. The transmutation rates are listed in Figure 8. With these parameters the 8 modules would render 3600 MW_{th}.

Burn-up calculations were carried out for six two-year cycles. They reach equilibrium after a few years and produce little ^{239}Pu and only a modest amount of ^{242}Pu. The fraction of ^{238}Pu varies between 85% and 87% after each cycle. There is little variation of ^{237}Np but a noticeable increase of ^{243}Am and ^{241}Am.

Because of the number of modules which must be irradiated by the high current proton beam, a large spreading of the beam is required, resulting in a long beam spreading section. In order to avoid this shortcoming the high current proton beam should be split into small beams. A failure in the accelerator part or in one module would stop the whole system.

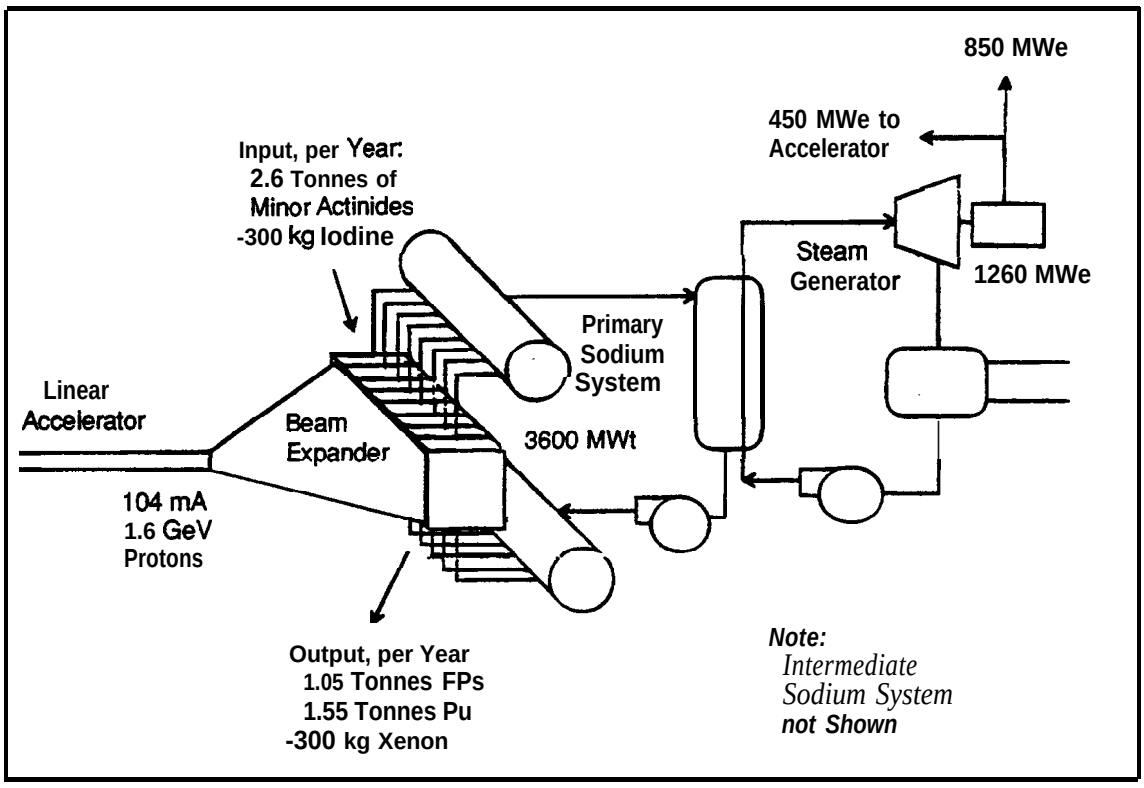


Figure 8: The PHOENIX Concept

High Flux Thermal Systems

Cross-section considerations show that fission products such as technetium and iodine can be more efficiently transmuted by thermal than by fast neutron systems. In Figure 10 the reduced (*effective*) half lives (τ_{eff}) of ^{99}Tc and ^{129}I are shown as a function of thermal and fast neutron transmutation. The cross sections of ^{99}Tc ($\tau = 2 \cdot 10^5$ [y]) are assumed to be 20 *b* for thermal and

0.2 *b* for fast neutrons. For ^{129}I (τ = 1.6" 107 [y]) 31 *b* and 0.2 *b*, respectively are taken. (In an epithermal neutron field there exists the possibility of enhancing neutron capture by resonance absorption.)

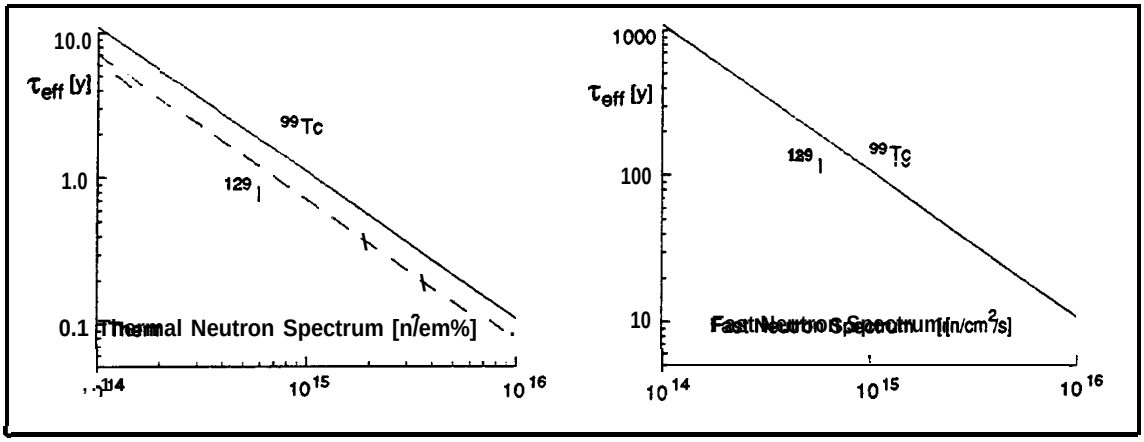


Figure 9: Effective half lives of ^{129}Tc and ^{129}I transmuted by different neutron spectra

In the case of the minor actinides the thermal cross sections are by orders of magnitude linger than the fast cross sections. But in this case the fission to capture ratio is less favorable for the thermal reactions. It means that the lower inventory of actinide fuel required to reach a certain multiplication factor is paid for by an increased build-up of higher order actinides.

But even in a thermal spectrum it is very difficult to transmute other fission products such as ^{137}Cs and ^{90}Sr (half live ~30y) because of their low absorption cross-section, 0.1 *b* and 1 *b* respectively. To transmute ^{137}Cs at ten times the natural decay rate a thermal neutron flux of 10^{17} *n-cm*$^{-2}$*-s*$^{-1}$ would be required.

Takahashi [31] studied the transmutation of fission products in a thermal flux field created by spallation neutrons in D_2O. The scheme was taken to be similar to LAFR. The 1.6 GeV protons are injected into a liquid Pb-target and the emitted spallation neutrons are thermalized in the surrounding assembly composed of (^{137}Cs + D_2O) and (^{90}Sr + D_2O). Figure 10 shows the geometrical parameters of the assembly. It turns out that even in this idealized configuration

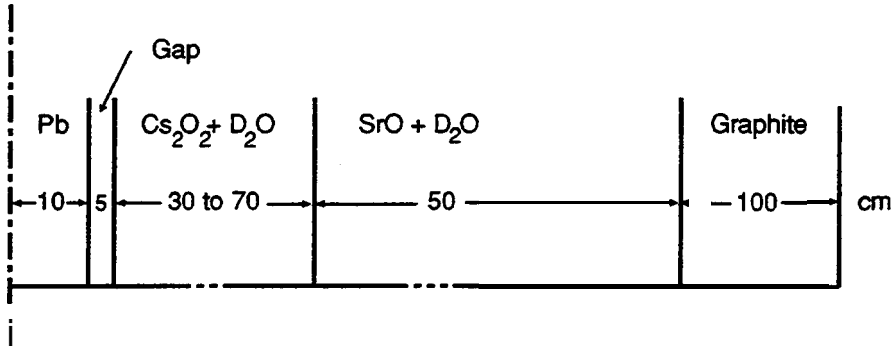


Figure 10: Configuration of thermal fission product transmute fed by spallation neutrons

because of the small cross-section of ^{137}Cs most neutrons escape unless a large amount of ^{137}Cs is put into the irradiation pool. However. a large concentration of ^{137}Cs lowers the transmutation rate for reasons of self-shielding as can be seen in Table IV. Also from the safety point of view it is not desirable to have a large fission product inventory in the assembly.

Table IV: Effect of Cs_2O_2 Volume Fraction on Neutron Losses in the Region surrounding a PbTarget

Volume Fraction of Cs_2O_2 in D_2O		10	50	80
Total loading *[kg]*	Cs_2O_2	450	2240	3580
	Sro	470	470	470
Fractional neutron loss in target region		0.49	0,34	0.24
Transmutation rate (see-l]	^{137}Cs	20.5 λ_{137}	11.0 λ_{137}	8.4 λ_{137}
	^{90}Sr	24.6 λ_{90}	14.8 λ_{90}	14.5 λ_{90}
Number of transmuted nuclei per proton	^{137}Cs	7.7	20.8	25.0
	^{90}Sr	15.0	10.1	8.5
Amount of transmutation per year [kg/y]	^{137}Cs	100	273	321
	^{90}Sr	134	80	75

Note: $\lambda_{137} = 7.33 \times 10^{-10}$, $\lambda_{90} = 7.52 \times 10^{10}$ *[see-l]*

The Los Alamos Intense Thermal Neutron Source

Recently LASL presented a well documented project on "Nuclear Energy Generation and Waste Transmutation Using an Accelerator-Driven Intense Thermal Neutron Source" [5]. It consists of an extensive study of incinerating fission products and minor Actinides, especially ^{99}Tc, ^{129}I and ^{237}Np.

The proposal deals with anew approach for a system either transmuting nuclear waste from civil and military origin or combining nuclear energy production (without a long term waste stream) with the transmutation of commercial nuclear waste composed of both minor actinides and fission products. The spallation process is used to generate an intense thermal neutron field in the 10^{16} n·cm^{-2}·s^{-1} range.

As shown in Figure 11 the transmutation system is composed of a proton accelerator, a heavy metal target surrounded by heavy water serving as a moderator for spallation and fission neutrons and a molten salt blanket containing actinides and fission products to be transmuted. The target is a flowing lead-bismuth alloy. Recently also a solid target made of tungsten and cooled by D_2O was considered.

The geometry of the system is composed of three sectors. The inner sector surrounding the

proton target contains heavy water. It is supposed to have a substantial epithermal flux component and is therefore particularly suited transmuting fission products such as ^{137}Cs and ^{90}Sr, which have a low thermal capture cross section, but epithermal absorption resonances. In this region a continuous flow system keeps the water temperature at 70°C.

For the transmutation of ^{137}Cs isotopic separation is necessary. This problem can also be solved in an elegant manner by decay fractionation. To this end the continuously extracted noble gas Xe is collected in chambers with increasing residence times to allow for isotope separation by natural decay processes. The same holds for the separation of ^{88}Sr from ^{89}Sr and ^{90}Sr where the decay products are chemically extracted.

The middle sector serves for the transmutation of minor actinides. It contains molten salt either in a cylindrical ring or in tubes passing through the D_2O moderator. The molten salt is $LiF\text{-}BeF_2$ as in the ORNL Molten Salt Experiment. It has an exit temperature of 720°C resulting from nuclear fissions of actinides and possibly additional fissionable material making the system sub-critical. The salt itself has a very low neutron capture cross section. The addition of actinides even in larger quantity does not alter the physical characteristics of the salt-melt.

The outer sector of the blanket is again filled with heavy water. It either serves as a

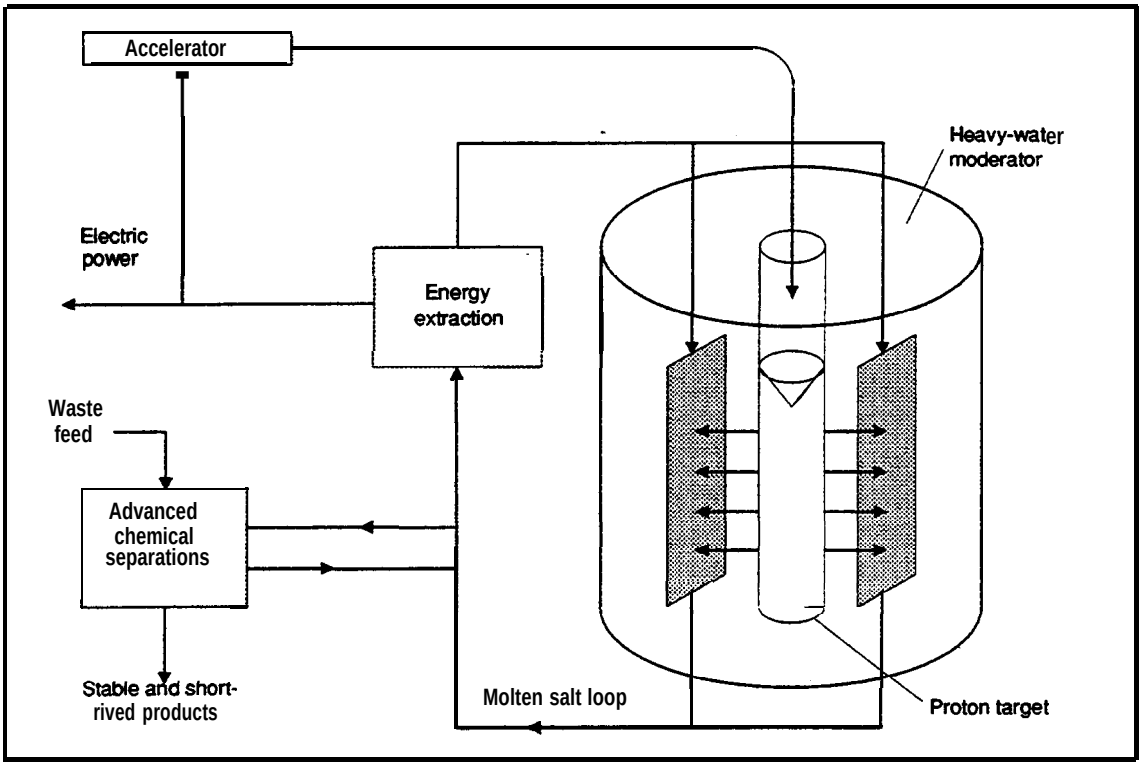


Figure 11: Schematic flow scheme of the LASL accelerator-driven nuclear waste converter.

reflector or as a region for the breeding of fissile material from natural thorium or uranium.

In an alternative proposal in addition to waste transmutation the system also produces

electricity to offset the reprocessing costs. In this case plutonium (or another material which can be fissioned by thermal neutrons) must be added to satisfy the neutron economy. In principle such an assembly could be operated as a reactor. But the high neutron flux and the rapid burn-up of plutonium and actinides might lead to fast reactivity swings. Hence for the same reasons as discussed above an accelerator driven sub-critical reactor is considered to offer the significant advantage of being an inherently safe system.

The composition of this system is very similar to the previous one. Again the inner D_2O ring would contain the "difficult" fission products and *Pu* would be added to the molten salt region. Driven by a 1.6 GeV - 13 mA proton beam, the facility could convert the waste of one 3 GW_{th} reactor and generate a power of 1060 MW_{th} in the molten salt. This implies that a 250 mA APT beam current could in principle process the waste from 19 LWRS operating at 3 GW_{th}. According to LASL studies the facility would run at a comfortable safety margin of $k_{eff} = 0.81$.

A similar system has been studied by LA-NIL. It differs from the LASL project by the replacement of the molten salt blanket by a heavy water slurry carrying the fissionable materials, such as actinides or plutonium.

All these systems look attractive because of their low actinide inventory and the possibility of a continuous reprocessing with the removal of short lived and stable fission products. On the other hand the high neutron flux may lead to serious difficulties, especially in the area of material damage, radiolysis and activation of structural materials. Also the ratio of the molten salt in and out of core residence time might lead toa less attractive ^{237}Np fission cycle.

One of the main reasons to use a 10^{16} n·cm^{-2}·s^{-1} flux is the transmutation of ^{237}Np. It constitutes the largest fraction of the minor actinides and cannot be fissioned by thermal neutrons. In a LWR ^{237}Np captures a neutron with σ_c = *1* 70 *b* forming ^{238}Np which decays with λ = 5.510-6 to ^{238}Pu. *This* isotope is again not fissionable and must become ^{239}Pu by neutron capture before it fissions. Because of the losses to capture in ^{239}Pu *this* chain to fission requires about 4 neutrons. If $\Phi\sigma_f > \lambda$ then the probability that ^{238}Np absorbs a neutron and fissions prior to its decay to ^{238}Pu *is* large.

The High Flux Particle Bed Reactor (BNL)

Encouraged by the advantages in control and safety features designers of *actinide burning reactors are* now pursuing the option of accelerator-driven systems, for example BNL with the High-Flux Particle-Bed Reactor System for rapid transmutation of actinides and long lived fission products. In this design a D_2O or beryllium carbide moderator/reflector contains zircalloy pressure tubes filled with coated gas-cooled fuel particles. The highly- effective heat transfer allows high power densities (- 5 MW/litre) and attendant high flux levels (10^{16} n·cm^{-2}·s^{-1}). Among the economic and safety benefits of this concept are the low inventory of radionuclides and the high-integrity, coated fuel particles that can withstand extremely high temperatures without releasing the fission products. On-line refuelling offers further safety and economical advantages as well as a low inventory of radio-nuclides (5% that in a commercial LWR).

A number of core configuration consisting of 92 to 85 Plutonium fuel drivers and 42

actinide loaded target PBR elements in a low temperature D_2O or beryllium carbide moderator/reflector were examined. In this concept fission products and actinides are always exposed to neutron irradiation. This is different from the LANL ATW scheme where a non-negligeable residence time outside the core reduces the incineration rate.

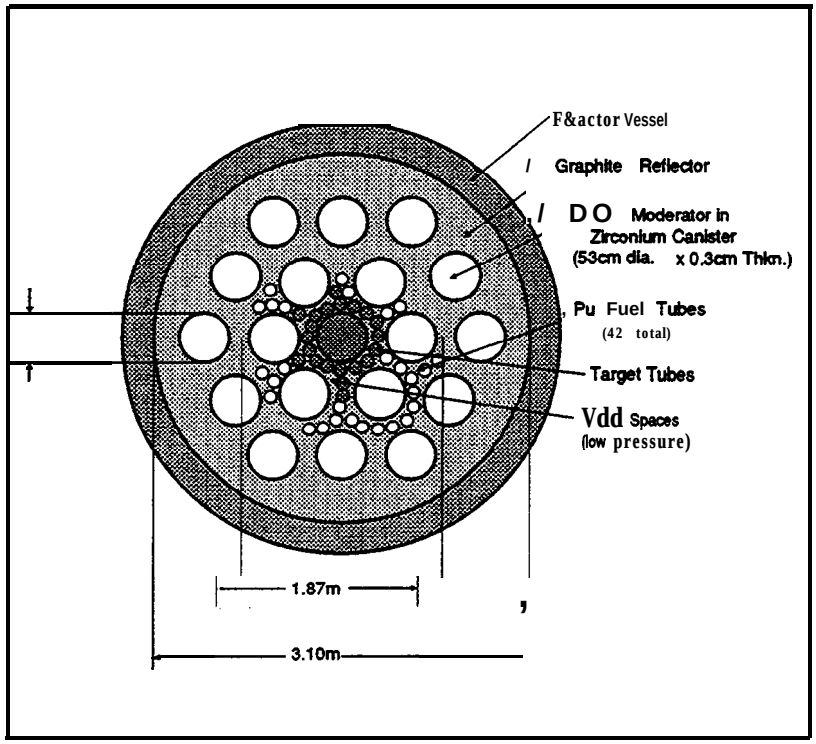


Figure 12: The BNL High-Flux Particle-Bed Reactor System

The reactor is operated in a subcritical condition by injecting a medium energy proton beam into a Pb target which is installed at the centre of the reactor serving as a neutron source. (see Figure 12.) Thus reactivity perturbations such as particle fuel movement due to fluctuations of the high pressure helium gas or the on-line fuelling can easily be controlled.

In the case of an accidental situation, the fuel particles can be ejected from the core region to a large volume container outside the core, where the decay heat can be removed by an emergency cooling system.

Conclusions

As we have seen, there exists a large variety of design options. However, they differ so much from each other that they present singular points in a multidimensional optimization space. It seems that so far only local optima, if any, have been found. The question still remains is there a global optimum? Which project is going to satisfy it? Still a synopsis of all the projects with pro and cons must be prepared. But there is certainly one very positive effect in all these studies: Politicians have recognized the importance of the question and it seems that R&D funds will become available. It is to be hoped that this is one of the reasons which will keep reactor physics alive for the years to come.

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