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TITLE: THE PHYSICS DESIGN OF ACCELERATOR-DRIVEN TRANSMUTATION SYSTEMS

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The Physics Design of Accelerator-Driven Transmutation Systems

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Abstract. Nuclear systems under study in the Los Alamos Accelerator-Driven Transmutation Technology program (ADTT) will allow the destruction of nuclear spent fuel and weapons-return plutonium, as well as the production of nuclear energy from the thorium cycle, without a long-lived radioactive waste stream. The subcritical systems proposed represent a radical departure from traditional nuclear concepts (reactors), yet the actual implementation of ADTT systems is based on modest extrapolations of existing technology. These systems strive to keep the best that the nuclear technology has developed over the years, within a sensible conservative design envelope and eventually manage to offer a safer, less expensive and more environmentally sound approach to nuclear power.

SUMMARY

When a medium-energy intense particle beam interacts with an appropriate target, neutrons are produced that are thermalized and can multiply in a surrounding moderating blanket containing fissile and fertile material. A neutron production of over 25 neutrons per incident particle per GeV is possible from such targets, with a multiplication of these neutrons by a factor of 20 to 30 in the subcritical blanket. Electricity is generated from the heat released by the fission processes, and a small part (between 5% and 15%) is used to drive the accelerator. A large excess of electric power is therefore available to the grid.

The basic elements, common to all the various ADTT systems, are a flowing lead spallation target driven by a GeV-class proton accelerator and a totally enclosed (pool-type) molten salt graphite moderated blanket.

The use of molten salt fuel allows the adoption of rather straightforward on-line fuel preparation (front-end) and cleanup (back-end) processes. Because no fuel extraction and refabrication is involved, these processes, under development at Los Alamos, do not introduce extraneous waste streams and have substantial proliferation and diversion barriers. Molten salt fuels also allow the attainment of substantially more powerful "inherently safe" units than possible with solid fuels, and owing to their high operating temperature and low pressure, they can achieve high thermal-to-electrical efficiencies, well exceeding those of current reactor designs.

Thermal-subcritical, accelerator-driven systems can achieve absolute safety from criticality accidents and have a neutron balance comparable to that of fast systems to enable them to perform the transmutation of the long-lived fission products. They also allow the complete burn of any nuclear fuel without reprocessing and, in the case of a thorium cycle, the full utilization of the fertile fuel without the use or production of enriched materials. Proliferation and diversion risks can be addressed effectively. End-of-Life inventories can be stored on-site as low level waste. As an example, the Accelerator Driven Energy Producer, based on the thorium cycle, is presented in some detail.

THE IMPACT OF ADTT

Nuclear systems under study in the Los Alamos Accelerator-Driven Transmutation Technology program (ADTT) will allow the destruction of nuclear spent fuel and weapons-return plutonium, as well as the production of nuclear energy from the thorium cycle, without a long-lived radioactive waste stream. In the area of Global Plutonium Inventory, ADTT can phaseout production and eliminate world inventory of commercial and excess-weapons plutonium in 30 years [1]. In the area of Nuclear Energy Production, ADTT can provide an alternative to conventional nuclear reactors and allow the complete utilization of fertile fuel (thorium) without excess breeding or the use of enriched material at any time in the cycle [2]. Finally in the area of Nuclear Waste Management, ADTT systems can drastically reduce the requirements for long-term storage of radioactive waste [3]. The same general ADTT nuclear design can be used in all three basic applications of the concept.

ADEP (accelerator-Driven Energy producer) is a thorium based energy producer, ATW (Accelerator Transmutation of waste) feeds on spent fuel, ABC (Accelerator Based Conversion) burns weapons-return plutonium. All these systems produce electricity and short-lived or stable fission product isotopes only (Fig.1).

The subcritical systems proposed by the ADTT program represent a radical departure from traditional nuclear concepts (reactors), yet they strive to keep the best that the technology developed over the years, within a sensible conservative design envelope.

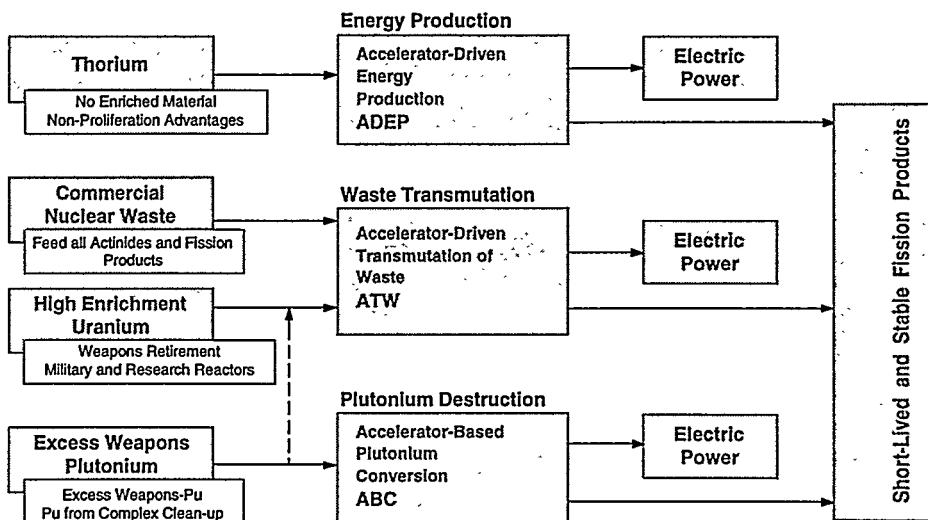


Figure 1. ADTT systems based on the same technology can use thorium, commercial nuclear waste, highly enriched uranium and excess weapons plutonium to produce energy.

OBJECTIVES OF ADTT

The Los Alamos ADTT concept is aimed at fulfilling the following 8 objectives:

- 1) Can burn 100% of its fuel, any fuel, to produce energy
- 2) Does not require fuel enrichment, extraction or refabrication
- 3) Does not build up plutonium as it produces power
- 4) Can destroy its waste as well as previously generated waste
- 5) Offers substantial enhancements to safety
- 6) Is diversion and proliferation resistant
- 7) Does not cost more than presently acceptable
- 8) Does not require technological leaps of faith

Traditional nuclear reactor technology, based on critical, solid-fuel systems, has clearly

never been able to meet all these objectives. However justified, the two assumptions of solid fuel and criticality have effectively negated the achievement of the 8 objectives. The simple removal of the two constraints of criticality and a solid fuel form widens the performance horizon of nuclear systems, to the point that all 8 objectives can be readily achieved.

The general features of the ADTT system proposed by the Los Alamos National Laboratory (LANL) group are shown in figure 2. When a medium-energy intense particle beam (protons or deuterons, 10-20 mA at 500-1000 MeV) interacts with an appropriate target, neutrons are produced that are thermalized and multiply in a surrounding moderating blanket containing fissile and fertile material in liquid (molten salt) form. A neutron production of over 30 neutrons per incident particle per GeV is possible from such targets, with a multiplication of these neutrons by a factor of 20 to 30 in the blanket. Electricity is generated from the heat released by the fission processes, and a part (between 5% and 15%) is used to drive the accelerator. The use of molten salt in the moderated blanket allows

high thermal-to-electrical efficiencies, exceeding those of current water-cooled reactors. A large excess of electric power is therefore available to the grid at rates that could be competitive with standard nuclear power costs.

In this system the molten salt fuel, a mixture of lithium, beryllium and thorium flu-

rideres, flows through a matrix of graphite, which forms the moderated, multiplying part of the blanket where the heat is generated, and into heat exchangers. Power generation occurs within the molten salt in the moderated region. The fission products isolated for transmutation are placed in the higher flux regions near the center of the blanket or in special heavy water high-flux, low-multiplication blankets, where they can absorb the extra neutrons necessary to convert them into stable species.

The system described in this article is driven by a 15 mA beam of 800-MeV protons and operates at 500 MWt with a k_{eff} of 0.95. The subcritical margin is sufficient to insure subcriticality in all conditions. The low power density of the system allows for an inherently safe design, where the after-heat cooling can be handled by purely passive means in case of emergencies. By completely consuming the nuclear fuel and the most troublesome fission products, this system would obviate the need for transport and geological storage of the long lived actinide species and fission product waste.

The operation in a driven subcritical mode eliminates the possibility of a criticality accident and provides quick and absolute shutdown by simply turning off the accelerator. On a more fundamental level the accelerator combines the traditional reactor control functions of control rods,

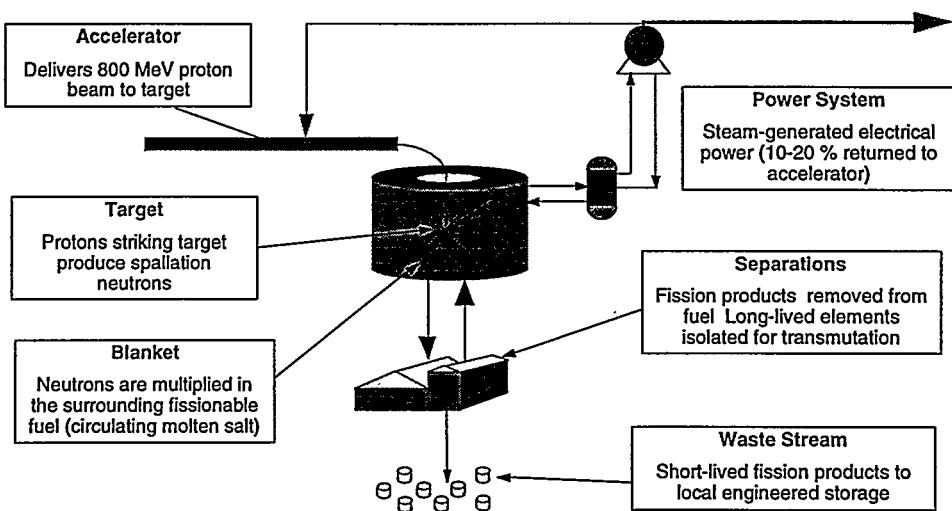


Figure 2. A top-level illustration of the Components of an ADTT system. ADTT is a closed nuclear system.

burnable poison and chemical shim in one streamlined mechanism which is flux- and burnup-independent. Coupled with the features of a liquid fuel system, this translates in the full utilization of the fuel without actinide extraction and excess breeding. Through the use of the accelerator drive and a fuel cleanup cycle, the fuel can be utilized completely. The low-level and short-lived radioactive waste remnants would be placed in near-surface managed storage at the power plant site itself.

BASIC DESIGN CHOICES

Graphite moderated thermal neutron spectrum

Fast systems have potentially a better neutron economy than thermal systems, because of the better ratio of fission to capture cross-sections in actinides. However the extra (fast) neutrons are not very useful for fission product transmutations (because of the very low cross section) unless they are thermalized. We maintain that it may not be safe to have thermalized neutrons in fast systems. Therefore, while fast systems can burn all actinides, they cannot straightforwardly destroy fission products. The accelerator drive introduces the extra neutrons missing from thermal-spectrum systems, effectively improving the thermal system neutron utilization to the point where there is very little difference between fast reactors and thermal subcritical systems in terms of neutron economy; that is, they can use the same nuclear fuel.

Thermal systems however have much larger fission cross sections, and therefore can work

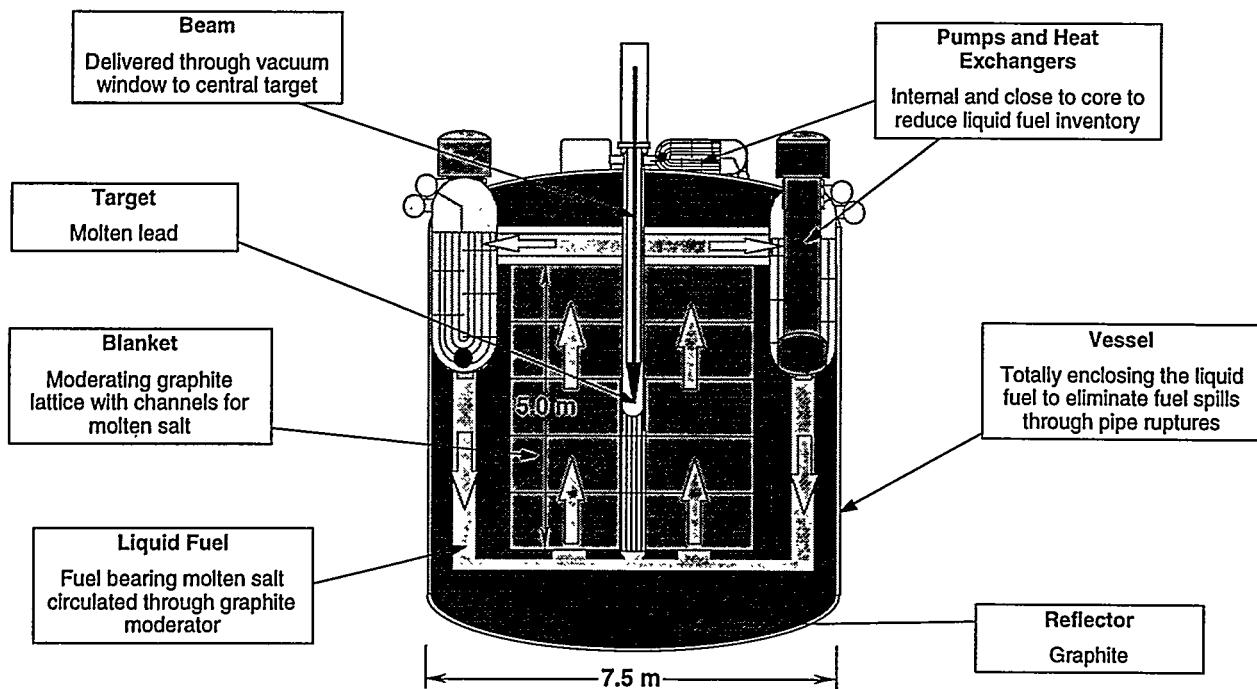


Figure 3. The prototype 500 MWt ADTT molten salt blanket. This blanket can be used for all the ADTT applications (ADEP, ATW, ABC). Graphite-moderated and fueled by molten salt, the ADTT blanket improves on the 1960's Oak Ridge molten salt reactor concept, by incorporating the many design improvements introduced by the newer liquid-metal reactors "pool" designs.

with much lower nuclear inventories and do not require enriched fuel to start. Looking at the thorium system, a thermal system might have an inventory of 100 kg of fissile uranium, whereas a fast system of the same size might require 10 tons of the same material. A thermal system driven by an accelerator can self-start in about 12 months, that is, drawing power from the grid a subcritical power system will produce enough uranium from thorium in one year to bring itself to full power, without having to use enriched start-up material. A fast system would require perhaps 100 years to do the same.

Fast systems operate in a region where absolute cross sections are very small. An accident that introduces a moderator into the fuel or fuel into a moderator will increase the reactivity. Water, for instance or even silicon dioxide (sand) will drastically increase the reactivity of fast systems fuels. There are very large numbers (literally thousands) of critical masses in fast systems. The consequences of accidents in these systems can be quite severe.

Thermal systems on the other hand operate in a optimum combination of geometry and moderation so that cross sections are highest. An accidental departure from the operational configuration will always lead to a less critical configuration. The rationale to develop critical fast systems (fast reactors) was the neutron economy, which allows these systems to breed autonomally. There is no compelling reason to have accelerator-driven fast systems, because the accelerator drive already provides all the advantages of the fast spectrum and also allows to keep all the good safety features of thermal systems.

Graphite is chosen as the moderator because of its high temperature compatibility with the liquid fuel choice, fluoride molten salt. In order for the graphite to last throughout the life of the plant, the power density in the blanket is limited to 5-10 w/cc in the moderated region. Because the fuel is kept clean, even at such low power densities the neutron flux is anticipated to be in the range 10^{14} to 10^{15} n/cm²s depending on whether a thorium bearing fuel is used or a pure actinide fuel. The main features of the graphite-moderated molten salt blanket are outlined in figure 3.

Liquid fuel (Molten Salt)

The choice of liquid fuels (and molten salt in particular) as the fuel in ADTT systems is based on the substantial advantages in the following 5 areas :

1)	Safety	Low-pressure operation
2)	Cleanup	On-line removal of fission products
3)	Complete Burn	No fuel fabrication and refabrication needed
4)	Efficiency	High temperature liquid
5)	Separations	Non-intrusive physical separations possible

Molten salts as reactor fuels and as coolants have been under study and development for over 40 years, and their chemical, physics, and irradiation properties are excellent. The Molten-Salt Reactor Experiment (MSRE) at ORNL, which was shut down in 1970 after about five years of very successful operation, contributed significantly to molten-salt reactor technology. The progress of the molten salt design study is covered in the entire February 1970 issue of Nuclear Applications and Technology, devoted to a review of molten salt reactor technology and to a description of a conceptual design for an molten salt breeder reactor [4]. The active work at Oak Ridge on the Molten Salt Breeder Reactor (MSBR) was phased out after the Liquid Metal Fast Breeder Reactor (LMFBR) was selected as the choice breeder. The last published results on molten salt technology

development at Oak Ridge date back to the late 70's. The successful operation of the Molten-Salt Reactor Experiment and the substantial amount of research and development accomplished on molten-salt reactor materials and processes at Oak Ridge indicate that with straightforward extensions in a few specific areas, a prototype Molten-Salt Accelerator-driven Transmutation Plant could be successfully constructed and operated.

The presence in the ADTT concept of the accelerator and separation subsystems, necessarily net drains on the energy balance, requires that the best possible efficiency be obtained in the power conversion process. Since the thermal to electrical efficiency is strictly related to the maximum temperature that is reached in the cycle, the liquid fuel should be able to operate at high temperatures without degradation of its physical and chemical properties. Safety concerns also require the use of liquid fuels that can reach high operating temperatures at low pressure. Molten fluoride salts can operate at temperatures well above 600 C at near-atmospheric pressure. This allows for the economical construction of rather large, low pressure vessels that can be all-enclosing, with integral pumps and heat exchangers, so that ducts and side-vessel penetrations can be totally eliminated, therefore reducing to extreme unlikelihood the event of a salt spill. In addition, since all the volatile fission products are continuously released and removed from the liquid fuel, and the others dissolved in the fuel itself are also removed, the source term important under severe accident conditions is substantially reduced vis-a-vis solid fuels.

The substantially more centrally peaked flux of accelerator driven systems in comparison to reactors also favors the use liquid fuels, where the fuel is uniform and all potentially unstable build ups of poisons or excess fissile material in special locations are completely eliminated.

The use of molten salt fuel, as opposed to solid fuels or liquid slurries, allows the adoption of rather straightforward on-line fuel preparation (front-end) and cleanup (back-end) processes. Because no fuel partitioning and refabrication is involved with molten salt use, these processes, under development at Los Alamos, do not introduce extraneous waste streams and have substantial proliferation and diversion barriers.

Molten salt fuels also allow the attainment of substantially more powerful "inherently safe" units than possible with solid fuels. The power level per unit is driven by the criterion that the unit should be passively safe in the event of a coolant failure, without need for salt drainage and removal. The 500 MW level per unit discussed later in the paper is a very conservative figure that might be the size of a first demonstration-type unit. Contrary to solid fuel systems, molten salt systems in fact have a heat source that is distributed throughout the large volume vessel. In ADTT molten salt systems, 2/3 of fuel is in heat exchangers and peripheral ducts and plenums at any given

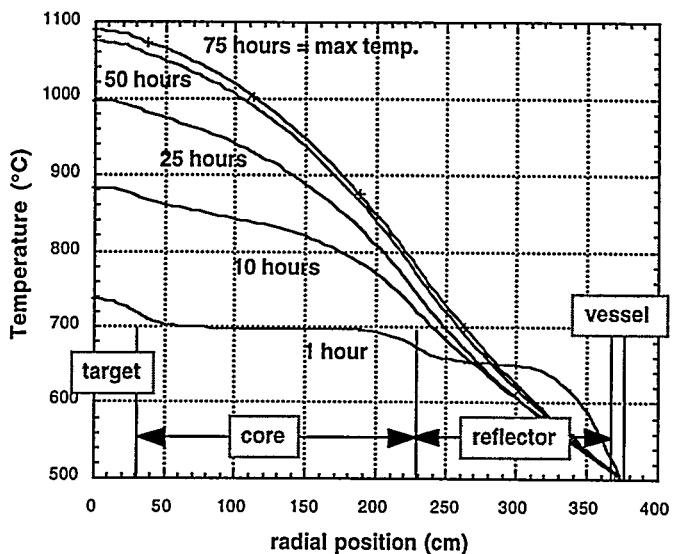


Figure 4. Temperature rise of a prototype 500 MWt ADTT blanket in a loss of coolant accident. Assuming only convective cooling and a passive heat removal mechanism, the maximum temperature of 1100 C is reached in 75 hours.

time and therefore any residual decay heat can be removed a lot faster than the case of solid fuel systems, where the heat source is concentrated at the center of the core, where it is most difficult to remove it when active cooling systems fail. The natural convection of the liquid fuel and the on-line removal of the fission products throughout operation also help considerably. A transient loss of coolant scenario is simulated in figure 4 for the 500 MW prototype module, which indicates a maximum temperature of 1100 °C reached 75 hours into the transient. We have performed calculations showing that the power per unit can be as high as 800 MWt and still satisfy the passive safety requirement that the salt not be drained in a loss of coolant accident. A much larger unit (3000 MWt) would require some means of draining the salt out of the blanket into a special passively cooled reservoir in the event of a coolant failure. Drainage in this case could still be obtained in a passive way through the use of melt plugs. Such a unit would have a power rating of 1200 MWe to the grid.

ADTT safety features

In spite of its non-conventional design choices, in fact precisely because of its non-conventional design choices, ADTT systems have very important inherent safety features [5]. Design characteristics, such as deep subcriticality, low power density, low operating pressure, no enriched fuel feed, no vessel penetrations and a distributed heat source, translate into inherent safety features, such as: no possibility for overheating and core damage due to decay heat in emergency situations, no possibility for criticality due to feeding error, no possibility for salt leaks. Subcritical systems are obviously slower to respond to reactivity insertions than critical systems, to the point that it is conceivable to maintain positive temperature coefficients in subcritical systems. The negative power feedback into the accelerator from the electricity extracted from the blanket should also insure that in the event of loss of coolant the accelerator/target system will not continue to pump neutrons into the blanket.

High-current Accelerators

The high-power accelerator technology required for ADTT has been under continuous development for the past three decades at Los Alamos. Accelerators up to five times larger than that required for the ADTT system described here have been proposed for the production of tritium for defense applications. These accelerators were reviewed by the Energy Research Advisory Board of the U.S. Department of Energy, subsequently by the JASONs, a scientific body which reviews proposed high technology programs of the U.S. Department of Defense, and were also evaluated by the General Accounting Office of the U.S. Congress. The result of these reviews was a general endorsement of the proposed accelerator technology with the provision that appropriate pilot and demonstration steps be made along the way towards the construction of a full scale facility [6].

The average power needed for the very largest of systems we propose (6×500 MWt), requires an accelerator average power of some tens of megawatts (100 mA beam). This can only be achieved with a linear accelerator. However, the highest power operational linear accelerator anywhere, (LAMPF), operates at around one megawatt (1 mA at 800 MeV). At first this seems like a very large extrapolation, but it is not nearly so bad as it appears. Firstly, in LAMPF, only every fourth bucket is filled with beam and so filling every bucket (if we use funneling) or every other bucket (if unfunneled) immediately gives a factor of four in average power respectively. Also LAMPF being a pulsed machine operating at 10% duty factor, going to 100% duty factor gives a

factor of 10. The charge in each microbunch can be increased by about a factor of four and still stay well within the stable space charge regime. Therefore an improvement by a factor of 160 ($4 \times 10 \times 4$) is possible by simple extension of proven technologies, so that up to 150 mA 1GeV LINACs can be built based on current technology [7]. Figure 5 shows a schematic view of a ADTT linac capable of driving 6 of the prototype 500 MW modules.

For circular machines the situation is a little different. The highest average power machine at PSI operates CW at about. PSI (the Paul Scheerer Institute in Switzerland) believes that its machine can go to about 1 to 2 mA without major changes, but to go to 10 mA would require a new design, which could be the Intermediate Separated Sector Cyclotron, presently proposed and studied by Rubbia [8]. Rubbia's design of a very compact circular machine in particular promises to become an efficient and inexpensive driver for the individual subcritical systems later described in this paper.

Primary issues for the ADTT accelerator are the efficiency, reliability and maintainability appropriate to an industrial setting. Experience with the use of an accelerator in an ADTT application can be readily gained using the existing accelerator at Los Alamos National Laboratory (LAMPF). As is, LAMPF is capable of reliably providing up to 1 mA of 800-MeV proton beam to a scaled down target-blanket similar to the one described here. LAMPF could thus drive an experimental deeply subcritical facility with a multiplication of 5 to a fission power level of about 5 MWt, a sufficient level for a thorough testing of the ADTT concept [9]. With modest improvements, leading to an increase in reliable current to about 2 mA, LAMPF would be able to drive a full size prototype module ($K_{eff}=0.95$, multiplication=20) to a fission power of 40 MW, sufficient for a full engineering testing of all the major components. With additional expenditures, small compared to the cost of a new accelerator, the LAMPF current could be increased to perhaps as much as 20 mA with a corresponding blanket power level easily exceeding 500 MWt, enough to power one of the prototype modules later described in the paper.

Liquid lead target technology

The function of a target in ADTT systems is the conversion of high energy (>500 MeV) protons to lower-energy (<20 MeV) neutrons and the delivery of these neutrons to the blanket in a useful manner. The conversion is achieved through the process of spallation, which is a simple way to describe the complex phenomena that take place when high energy protons hit a target. Through a series of internuclear cascades, neutrons and other charged particles are stripped off the target nuclei. Eventually neutrons will escape and propagate into the surrounding multiplying blanket. The ideal target should be simple in construction and operation, able to produce neutrons efficiently and to leak them to the surrounding blanket with minimal absorption losses.

At the high power densities generated by the beam currents proposed for ADTT systems,

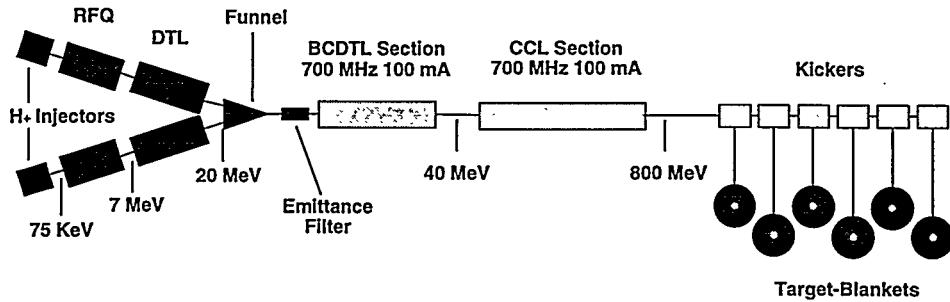


Figure 5. The ADTT linear accelerator design for multiple targets

liquid metal targets offer important advantages in practical implementations over solid targets. Besides the obvious ease of heat removal from liquid targets, there are no structural damage limit to the life of the target material, and the use of liquid targets would allow the primary target material to be reused beyond the structural life of the container material. This minimizes residual waste and maintenance time. High atomic weight, low melting point metals or alloys are needed. Among the low melting point heavy metals, lead and bismuth have very low neutron absorption cross-sections and therefore can be arranged in simple monolithic configurations. The lead-bismuth eutectic (LBE) has a lower melting point and operating temperature than either lead or bismuth and therefore should be favored in terms of structural consideration. The bismuth in the eutectic however creates greater materials compatibility problems with the structural container and produces radioactive polonium. Either lead or lead-bismuth eutectic would provide an adequate solution to ADTT spallation target [10].

The liquid lead (or LBE) target used for the ADTT design is inserted vertically into the blanket from the top. The reasons for this choice are simple. A horizontal target would require very heavy shielding against forward directed high energy neutrons. It would also introduce the risk of a radioactive molten salt spill from a side penetration into the blanket. Vertical insertion of the target from the bottom would also introduce penetrations in the bottom part of the blanket or the presence of permanent metal structures in the center of the blanket where neutron damage is most severe. In the vertical insertion from the top, a window is placed upstream of the target along the beam path and it is of traditional design. Past the window, the proton beam enters a hydrostatic helium plenum which keeps the lead surface down in the bottom of the target. The helium region is 300 cm long and takes the beam with negligible loss into the central region of the subcritical blanket. There the beam hits the lead free surface and slows down in a 90 cm long liquid lead (or LBE) volume. The beam stops in this length, and also (n,xn) neutron multiplication reactions happen in this region. There would be no window between the beam and the lead. This configuration avoids the most serious cooling problems of a hot window at the center of the blanket. The liquid lead flows down on one side of the split pipe, across the target area receiving the beam directly on its free surface and then flows up on the other side (see Fig. 6).

The target is a to-

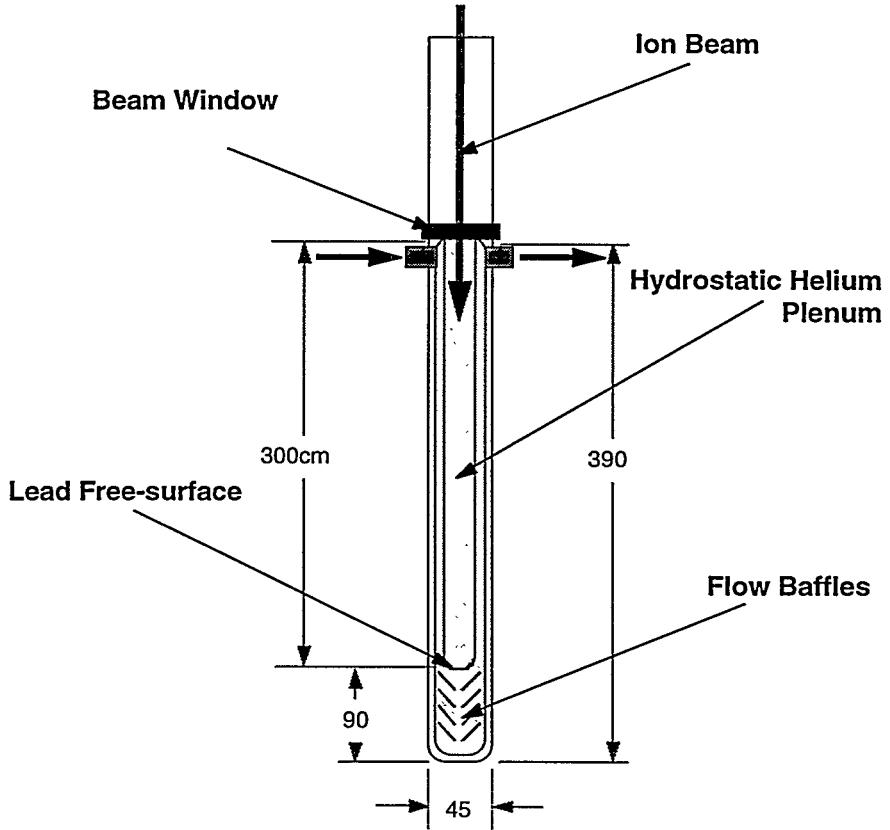


Figure 6. A schematic view of the prototype ADTT lead target

tally enclosed structure, bolted on the lid of the blanket and capable of being rapidly inserted and extracted from the blanket. The container structure has a double wall: the inside is lead compatible HT-9 alloy, the outside is made of molten salt compatible Hastelloy-N. Due to radiation damage to the steel, it is anticipated that the structure of the target most exposed to the neutron flux will have to be replaced as often as every 1-2 years. The same lead is however recycled through the life of the plant and beyond to the next ADTT system.

FUEL PREPARATION AND CLEANUP

It has long been the policy in the US not to promote the reprocessing of spent fuel. If by fuel reprocessing we intend the extraction and refabrication of the actinides in spent fuel to extract new power and close the nuclear fuel cycle, then ADTT provides an answer to that seemingly impossible task: a closed nuclear cycle that does not require reprocessing. What ADTT requires are much simpler processes: Fuel Preparation at the front-end and On-line Cleanup at the back-end.

Front-end Processes (ATW)

Front-end processes are only needed in the Accelerator Transmutation of Waste (ADTT-ATW) concept.

Uranium and zirconium are undesirable constituents present in large amounts of the spent fuel feed to ATW systems. Uranium in particular needs to be separated from the rest of the feed, as the continued production of plutonium in the course of the transmutation campaign would be entirely undesirable. The volatilization of fluorides is well tested way to separate the uranium and the zirconium from the spent fuel. The proliferation risks of this approach however might not be acceptable, in that there are no strong barriers to the extraction of plutonium and other actinides from the fuel in this process. Very strong proliferation barriers are instead present in a second process, presently under investigation at Los Alamos: uranium electrowinning [11].

The uranium electrowinning uses favorable differences in electrochemical potential to extract uranium and zirconium from spent fuel (see Fig. 7). If successfully implemented, it will allow the extraction of uranium from spent fuel without separation of plutonium and fission products, which is the limited scope of the fuel preparation processes for ADTT systems.

Briefly described the process consists of three main steps:

First, the spent fuel rod is fluorinated in a stream of HF/H_2 so that fluorides are produced. Fluorination in HF/H_2 prevents the formation of volatile hexafluorides.

Next, the fluorides are dissolved in molten Li_2BeF_4 .

Finally, a beryllium anode is inserted in the molten salt and all the fluoride species less stable

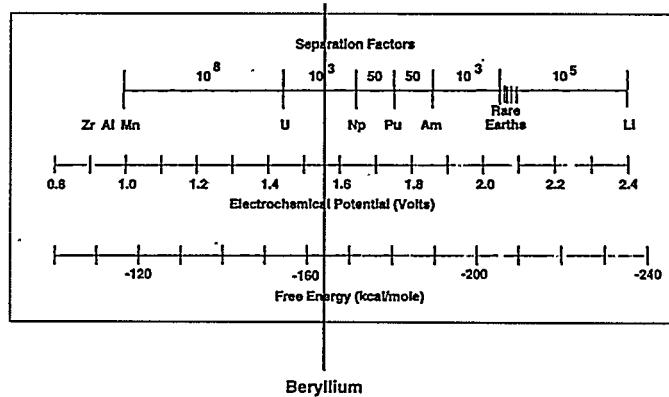


Figure 7. Electrochemical potential scale of the elements, showing the favorable position of beryllium, between uranium and the rest of the actinides

than BeF_2 will be reduced at the cathode. Since beryllium is in between uranium and all the other actinides in the electrochemical potential scale, after the deposition of the uranium, all of the beryllium would have to come out of solution before any of the actinides including plutonium, or the rare earths could be isolated. Since the salt mixture after the uranium has been extracted is typically 74:24:2 of Lithium, beryllium and actinide (plus fission product) fluorides, the presence of beryllium is a strong barrier to the extraction of actinides or rare-earths fission products during the spent fuel preparation step. The species that remain in the molten salt will be Np, Pu, Am, Cm, and lanthanide fluorides which can be fed directly into the molten salt blanket. Fission products (Rare Earths) stay in solution with the Actinides in the ATW feed (they are removed continuously at the back-end).

Back-end Processes (ATW, ABC, ADEP)

No actinide separation from the fuel is needed for ADTT operation and all actinide isotopes are fissioned and used for their energy content. The by-products of the fissions in the fuel (fission products), however, are continuously removed from the fuel. Short-life fission products can be left to decay naturally outside the blanket. Those with a longer half-life (>30 years) and which are of concern for long term storage, are transmuted into nonradioactive or short-lived nuclei through neutron capture.

The on-line cleanup processes envisioned for ADTT systems will not require introduction of external chemical agents.

Fission products in molten salt can be divided in three broadly different but roughly equivalent categories, and for each of these there is an appropriate physical means of cleanup. Volatile fission products and noble gases have low boiling points and tend to come out of the molten salt very easily. Removal of these volatile species and noble gases from the molten salt is obtained by helium flow or sparging. Because of the fast time scale of the sparging process, we call this "one-minute separation". Noble and semi-noble metals have low solubility in the salt and will plate out on internal surfaces. The process was observed at Oak Ridge in the molten salt experiment [4], and we plan to actively exploit it with the incorporation of passive extraction cells in the molten salt flow path. Again referring to the relevant time scale, these are the "one-hour separations". The remainder of the fission products, including the rare earths have good solubility in the salt and will tend to build up in concentration and stay in solution. Removal of lanthanides and other non volatile species will be done using liquid centrifugation and precipitation ("one-month separations"). Again the point should be emphasized that the ADTT back-end processing only requires fuel cleanup from fission products, not actinide isolation and extraction.

Helium Sparging of volatile fission products

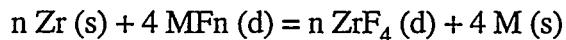
Volatile fission products will be carried away by a flowing stream of helium at the top (free) surface of the molten salt. The efficiency of the removal might be improved by "sparing", that is by injecting helium bubbles in the molten salt stream [4]. These bubbles act as collectors of the gaseous fission products. Again the fission products are removed by the flowing helium stream at the top plenum. After extraction from the blanket the helium stream carrying the fission products undergoes fractional distillation processes to extract the different fission products at different temperatures according to their boiling point. Special care is dedicated to xenon and argon, which are collected on cold zeolites. If extraction from the blanket is effected promptly, there is the possibil-

ity of achieving separation of cesium isotopes from the fractional decay of xenon.

Electrolytic separation of noble and semi-noble metals

Electrochemical separation techniques are proposed for removing the “noble metals” fission products: Mo, Nb, Ru, Rh, Ag, Cd, etc. [12].

The electrochemical method has been used extensively to produce pure metals from oxide or halide feed materials that have been dissolved in a molten salt. In this application of the method one is interested in purifying the molten salt and not in producing a pure metal. The electrochemical cell consists of a consumable anode, Zr or Be, which is placed in the molten salt and the “noble metals” are deposited on the cathode, a Ni rod. The fissile material, other actinides, and lanthanides remain in the molten salt. The reaction that describes the process is:



where M is a “noble metal”, n is the valence state of the “noble metal”, and d refers to a fluoride species dissolved in molten Li_2BeF_4 . The reaction is spontaneous because of the free energy difference between ZrF_4 and the “noble metal” fluorides. Therefore, in principle, the electrochemical cell can be operated in a passive fashion (i.e. no external source). Maintenance of this system involves the periodic replacement of the anode and cathode.

Molecular Centrifugation of soluble fission products

Molecular centrifugation uses the difference in molecular weight between carrier salt (light) fission product salt (medium) and actinide salt (heavy) to obtain effective separations of soluble fission products without recourse to chemical extractions [13]. The obvious consequence is the effective cleanup of lanthanides from fuel without acting on the chemically similar actinides.

Subjecting a liquid solution to large centrifugal fields will generate gradients of concentration increasing towards the outer edge of the spinning centrifuge rotor. These gradients and the speed at which they are attained are directly proportional to the square of the rotational velocity of the centrifuge and to the molecular weight of the solute. Other factors such as the solution viscosity and temperature also influence the process.

Centrifugal force can create solute concentration gradients in originally homogeneous solutions. The evolution of such processes is determined by the sedimentation coefficients and the mass diffusivities, which are predominantly dependent upon the molecular weights. A heavier solute will develop a steeper concentration gradient than a lighter solute. This leads to a relative enrichment

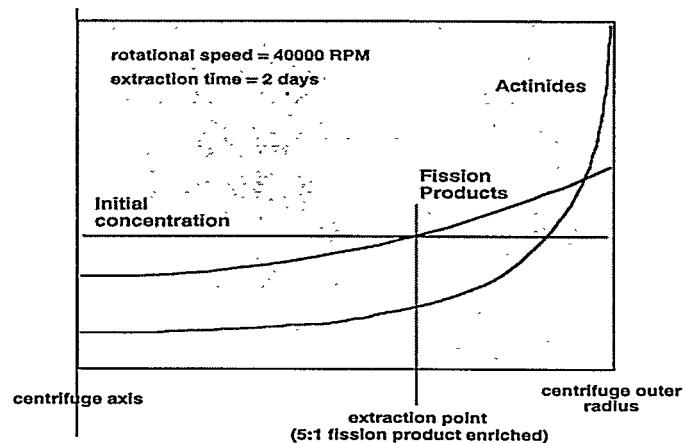


Figure 8. Working principle of centrifugation processes. Note that at the extraction point the fission product-to-actinide ratio is higher, but the overall concentration has not changed.

of one solute in one section of the centrifuge with respect to the other. It is always possible to identify an extraction point in the radial concentration profile where there will be an enrichment in the concentration of the lighter solute with respect to the heavier solute or vice-versa (see Fig. 8).

The process can be used in liquid solutions of molten salt fuel to isolate and separate the fission products from the rest of the actinides. Since the process relies on physical properties and always operates in dilute solutions, it is capable of removing only the fission products from the fuel, regardless of their chemical affinity to the actinides and it will not give rise to extraneous waste streams. Very little of the actinides are ever disturbed and the process is benign from a proliferation and diversion point of view. Through a series of centrifugation stages, very high enrichments of light-to-heavy or heavy-to-light solute can be reached ($>1000:1$) without appreciably increasing the overall density of the solute in solution and therefore without concerns for precipitation or for non-ideal behavior of the solution constituents. The separated fission products can be concentrated in a final centrifugation stage to reduce the waste volume.

AN EXAMPLE: THE ACCELERATOR DRIVEN ENERGY PRODUCER (ADEP)

As an example of the ADTT concepts I will briefly describe the Accelerator-Driven Energy Producer. The design features discussed are very preliminary and by no means optimal .

The Accelerator-Driven Energy Production (ADEP) concept employs an accelerator to provide neutrons to a moderated multiplying blanket. This allows the breeding of fissile ^{233}U from thorium, the fission of the ^{233}U with consequent production of electric power, the concurrent transmutation of long-lived fission products wastes to stable or short-lived species and the burning of all actinide waste generated. Thorium is sufficiently abundant in the earth crust to allow electricity production for thousands of years at any foreseeable power production level.

Natural thorium is used as fuel and is completely utilized without excess breeding. All actinides generated during power production are fissioned and never removed from fuel. Unlike fast breeder reactor systems, new energy sites can be started without production and transportation of excess enriched nuclear fuel. Long-lived fission products are transmuted and the need for geological storage of waste drastically reduced. ADEP can fulfill all the promises of Controlled Nuclear Fusion using current, proven technology.

When a thorium atom (^{232}Th) absorbs a neutron it is converted to an unstable thorium isotope (^{233}Th), which quickly decays into protactinium (^{233}Pa). In turn, the ^{233}Pa atom decays into ^{233}U with a half-life of 26 days. The nuclide ^{233}U has a large fission cross-section and a comparatively very small capture probability making it a very favorable fuel for nuclear energy production.

The thorium-uranium fuel cycle has two advantages over the traditional uranium plutonium cycle used in today's nuclear reactors:

1) The thorium-uranium cycle produces a very small amount of higher actinides (elements heavier than uranium), because of the small capture to fission ratio in ^{233}U and because of the presence of two other fissionable isotopes of uranium (^{235}U and ^{237}U) in the chain leading to plutonium and the other heavier actinides

2) The thorium-uranium cycle is regarded as safer than the uranium-plutonium cycle from a nuclear weapons proliferation standpoint, because of the presence of the hard-gamma emitter ^{232}U as a minor product of the cycle, and because of the possibility of straightforward isotopic dilution of ^{233}U with depleted or natural uranium in the feed or start-up fuel.

The thorium-uranium cycle has been extensively studied in the Oak Ridge Molten Salt Reactor Experiment (MSRE) and Breeder (MSBR) projects, and actually implemented in the origi-

nal High Temperature Gas Cooled Reactor proposed and built by General Atomic at Peach Bottom and Ft. Saint Vrain [14]. The conclusions of studies at Oak Ridge were that critical thermal breeders (reactor) could not breed fissile material in this cycle, unless the fuel could be kept painstakingly clean from fission product poisons and the protactinium precursor to ^{233}U could be promptly removed from the neutron flux and allowed to decay into ^{233}U . The introduction of the accelerator in ADEP largely eliminates these two problems and allows the thorium-uranium cycle to be exploited to its fullest in simple, practical systems.

The accelerator-driven spallation source supplements the number of available neutrons over that which can be achieved in a critical reactor by converting some of the electric power generated into neutrons. This neutron increase allows ADEP designs to produce energy using only natural thorium as feed, without highly enriched fuels and even without chemical extraction of the protactinium precursor to ^{233}U while at the same time transmuting the internally generated radioactive wastes. To do the equivalent functions, a critical reactor would require the use of highly enriched fuels.

What can ADEP systems transmute ?

ADEP systems can transmute all long-lived fission products for a price. One concern with the geologic storage of radioactive waste is the possibility of migration out of the storage medium. In this regard, ^{99}Tc and ^{129}I are the most bothersome of the fission products. Fortunately, these isotopes have high thermal neutron capture cross-sections and are easily transmuted to stable ^{100}Ru and ^{130}Xe respectively. The next most important long-lived nuclide is ^{135}Cs ; in this case, isotopic separation of the cesium nuclides is required before burning the ^{135}Cs , to make effective use of the neutrons in the transmuter. However, isotopic separation of ^{135}Cs and ^{137}Cs as well as ^{89}Sr and ^{90}Sr can be obtained naturally by radioactive decay in the ADEP system, exploiting the fast removal of their noble gas precursors from the liquid fuel. The small quantities of ^{126}Sn and ^{79}Se present among the fission products can also be readily transmuted. ^{90}Sr and ^{137}Cs are major components of the waste and are more difficult to transmute because of their very low thermal capture cross-sections.

The additional fraction of electricity needed to power the acceleration to accomplish the transmutation of key fission products is indicated in figure 9. In principle, the accelerator-driven neutron economy allows all of the long-lived fission products to be transmuted, but the value of performing the transmutation varies greatly among them and must be determined for each

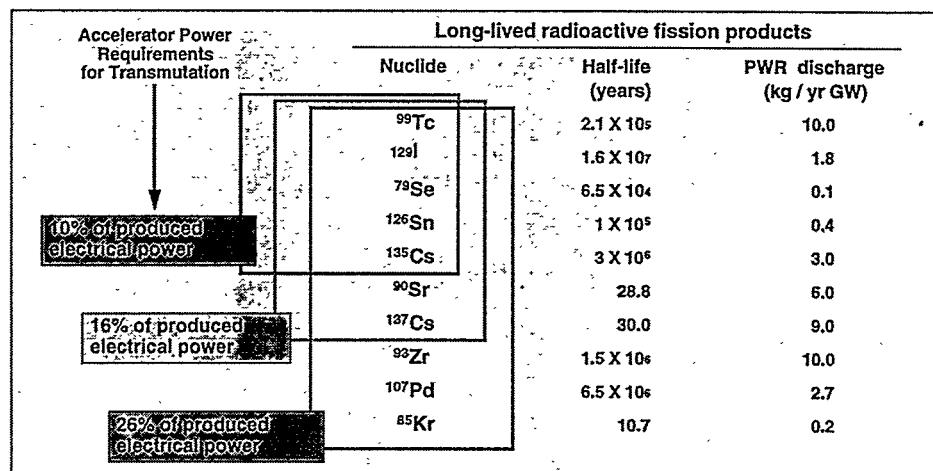


Figure 9. Fission product burning capabilities as a function of accelerator power . We think it is most appropriate to burn Technetium, Xenon, Selenium, Tin and $^{135}\text{Cesium}$.

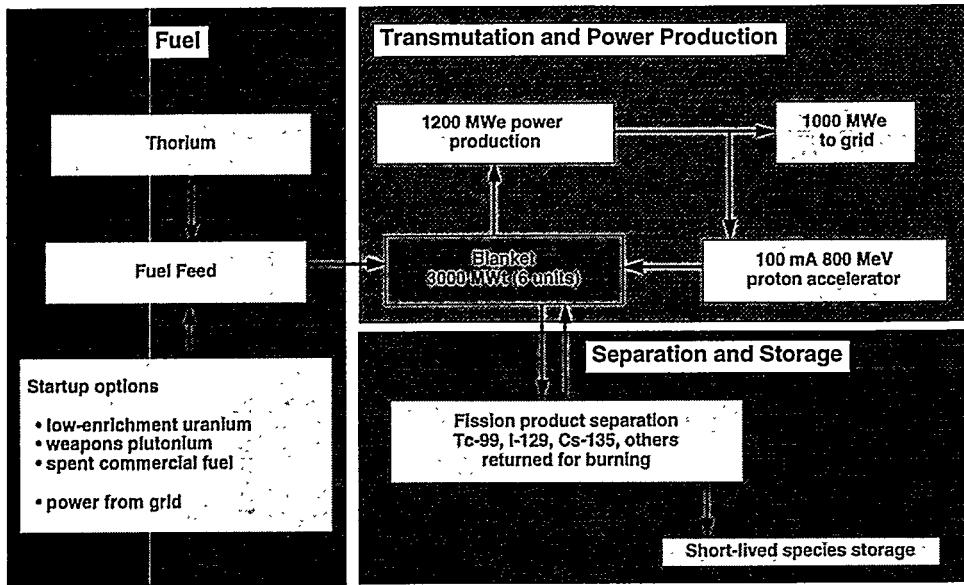


Figure 10. General features of the ADEP system. Electricity is generated from the heat released by the fission process, and a part is fed back to drive the accelerator. While the system can be started with a variety of enriched materials, particularly interesting would be the option of starting the system purely from power from the grid. This would effectively enable ADEP to start and sustain a thorium breeding cycle without isolating enriched materials of any kind. Make-up thorium is continuously fed. back-end separations and transmutation of long lived fission products are performed concurrently. The back-end processing removes the fission products from the fuel and isolates the isotopes requiring transmutation (^{99}Tc , ^{129}I , ^{126}Sn , ^{79}Se , ^{135}Cs). The residual radioactive waste will be suitable for on-site burial.

because their radioactivity is very small, their decay modes have very low energies, and as noble metals they are not readily transported by natural means through the environment. ^{137}Cs and ^{90}Sr cannot be effectively transmuted inside the low-flux ADEP blanket because of their small cross-sections. A large fraction of these isotopes will however decay naturally due to their relatively short 30-year half-lives during the life of the ADEP power plant, and if a more complete transmutation of these fission products is desired, special high-flux, low-multiplication blankets should be used. The 300-year containment of these isotopes (10 half-lives) is however deemed feasible, and may be all that will be required to deal with them. (Figure 10 shows the functional block diagram of the ADEP system).

Start-up

While the ADEP could start-up by beaming ions into its target and breeding its ^{233}U from scratch, this process would require some time (typically 1 year) to build up the blanket to full power. A convenient start-up mechanism could be provided where appropriate by feeding an initial charge of high-enrichment uranium, weapons plutonium or spent reactor fuel at the beginning of life for the ADEP plant (about 100 kg per 500 MWt module). The use of this initial feed allows the quick jump-start of the thorium uranium breeding cycle, and time to full ^{233}U power under beam is estimated to be about 100 days in simple start-up scenarios.

isotope through detailed cost, risk and benefit analyses.

Our fission product transmutation goal includes burning all of the ^{99}Tc , ^{129}I , ^{135}Cs , ^{126}Sn and ^{79}Se , which all together constitute 4.5% of the total fission products generated by the ADEP during power production. The remaining fission products are made up of stable nuclides, short-lived isotopes, and long-lived isotopes that do not present significant hazards. We believe, for instance, that it is unnecessary to transmute ^{93}Zr and ^{107}Pd

THE DESIGN OF ADEP

The design of the ADEP plant consists of various systems, or facilities, categorized as:

1. The target-blanket system, in which source neutrons generated by the interaction of a ion beam with the molten salt target are thermalized and subcritically multiplied; the fission heat generated in the fuel salt in its passage through a graphite moderated region is removed in primary heat exchangers
2. A coolant-salt circulating system, steam generators, and a turbine-generator plant for converting the thermal energy into electric power
3. An off-gas system for purging the fuel salt of fission product gases and gas-borne particulates and an associated electrolytic system for plating out noble and semi-noble metals.
4. A separation processing facility for fuel salt cleanup and recycle; this facility would continuously process a slip stream of molten salt.
5. Auxiliary salt handling equipment
6. General facilities and equipment, including controls and instrumentation, maintenance tools, auxiliary power equipment, waste management, storage and disposal systems, condensing water systems, electrical switchyard, stacks, and conventional buildings and services.

It is not the purpose of this paper to describe in any detail the ADEP system. Therefore I will only cursorily describe subsystems 1 through 4 and then just those parts that are unique to the ADEP system.

Target-blanket primary system

In the reference concept the molten salt fuel (a mixture of lithium, beryllium, thorium and uranium fluorides) flows through a bed of graphite moderator elements, which form the multiplying part of the blanket where the heat is generated, and into heat exchangers.

The (750-cm diameter, 700-cm height) blanket vessel contains graphite for neutron moderation and reflection, with the power producing region 80 cubic meters in volume (500-cm height, 450-cm diameter). All pumps and primary heat exchangers are also contained in the vessel. Shutdown and power level of the subcritical assembly are controlled by the accelerator beam intensity. Fuel concentration can be changed during operation without affecting the control properties of the accelerator. The target/blanket assembly, fuel processing facilities and power generating equipment are all located underground.

The center of the blanket is occupied by the liquid lead target, a self-contained unit which is inserted and extracted from the top of the vessel. The primary salt is circulated through six loops feeding from and into a common central plenum. Each circuit contains a single-stage centrifugal pump and a heat exchanger, all placed within the first containment vessel. Construction is modular, so that any heat exchanger or pump can be independently removed for maintenance.

After-heat removal system

As a design feature, the hastelloy-N vessel is totally enclosed and without openings, except for the lid at the top, so that any spillage of the salt is extremely unlikely, as it would require a major break of the vessel. Extended removal of the after-heat in the event of loss of circulation in either the primary or the secondary salt systems is designed into the vessel and surrounding containment structure, and does not require the transfer of the salt to a separate storage tank.

Secondary-salt circulation and steam-power

Heat is transferred from the primary salt to a secondary fluid, sodium fluoroborate, having a composition of $\text{NaBF}_4 : \text{NaF}$ (92:8 mole %) and a liquidus temperature of 390°C. The secondary salt goes to the steam generators to obtain 550 °C-temperature steam. Turbines and power generating systems are entirely conventional.

Off-gas collection system

It is important to provide quick removal of the volatiles in order to improve the overall reactivity of the systems, limit reactivity excursions due to xenon decay, reduce the heat load of the molten salt in the event of loss of circulation or cooling power, and limit the radioactivity release in the event of direct fuel exposure to the environment. The molten salt fuel offers simple means for the separation of these volatile species. Almost all noble gas fission products and 30% of the noble metals are removed by flowing helium at the upper plenum surface, within minutes from their formation, the rest of the noble metals plate out on special internal surfaces within a few hours.

Fuel-salt cleanup system

The cleaning cycle (Figure 11) must perform the following functions:

1. Remove all fission products from the molten salt.
2. Return all actinides to the blanket.
3. Isolate the long-lived fission products for return to the blanket for transmutation.

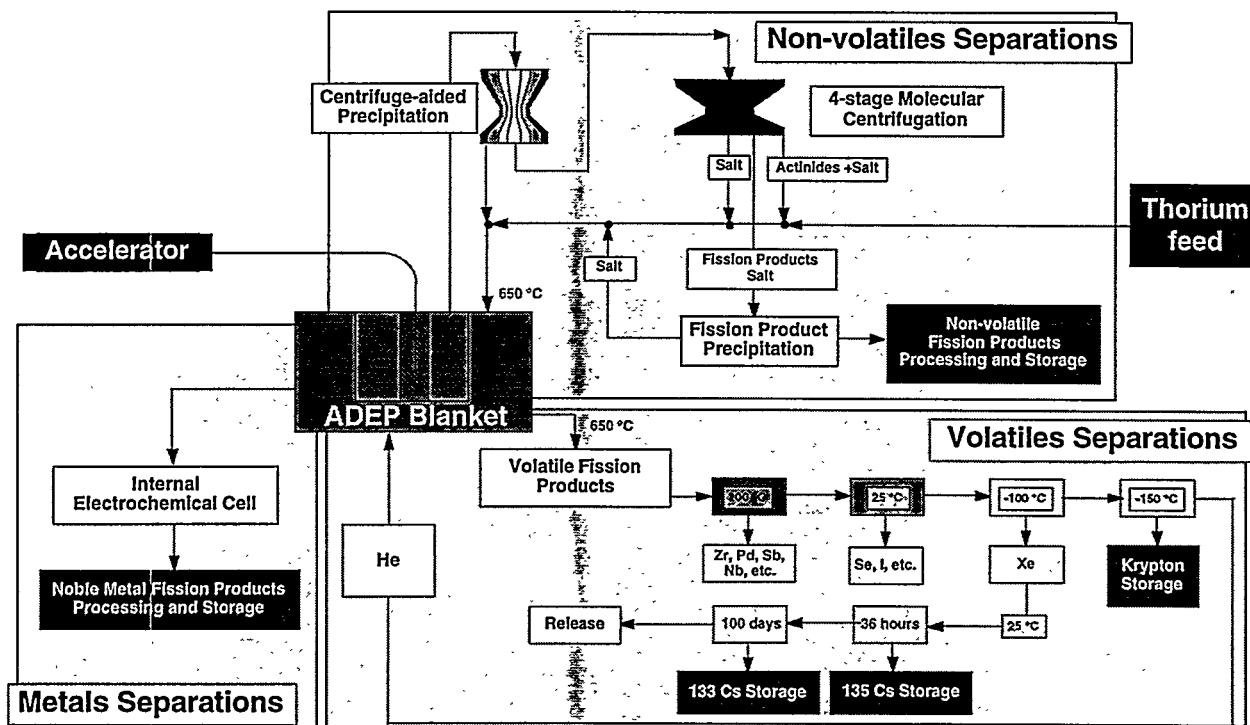


Figure 11. The molten salt ADEP flow-sheet for back-end cleanup processes. The three main groups of fission products receive different treatments.

The separation flowsheet proposed for ADEP is based on precipitation enhanced by centrifugation. It completely avoids the requirement for reagents, which might give rise to secondary waste streams. Starting with a nominal molten salt mixture of 12:1:69:18 ThF₄:UF₄:LiF:BeF₂ at an average temperature of 650 °C, the first process is a precipitation through cooled centrifugation. By the time the salt cools to about 400 °C, the salt composition will change to 2:54:44 of ThF₄:LiF:BeF₂. Nearly all of the ThF₄ precipitates, and the chemically similar UF₄ with it. These species can be returned to the transmuter blanket or be recycled through the centrifuge for further purification. The centrifugal fields required are modest and typical of those used in industrial centrifugation processes for removing precipitates from liquids.

Volatile fluoride fission products are removed throughout power production by the off-gas system. Low solubility fission product plate out in special places in the molten salt stream. Therefore only low volatility fission products and actinide fluorides are dissolved in the discharged salt. Because of their low concentration these fluoride salts will be carried with the LiF and the BeF₂ after the first precipitation.

A molecular centrifugation process follows this initial precipitation. During this centrifugation (4 parallel sets of 8 sequential stages) the residual fission products are extracted from the fuel. The end result is a stream of fission product bearing salt with less than 1 part per 1000 of actinides. This stream can be concentrated and fission products precipitated from it.

Waste Streams

Like the other accelerator-driven concepts originated at Los Alamos in the past few years, the ADEP concept is designed to limit its waste streams to a minimum. The ADEP transmutes the long-lived fission products generated during power production, and burns any higher actinide to a very low equilibrium level. It uses separation processes that do not involve external streams of reagents to be disposed and its high energy conversion efficiency limits the amount of waste heat released to the environment.

Four distinct radioactive waste streams have been identified from the ADEP operation at the end of its scheduled operational life: activated structural materials, short-lived solid fission products in fluoride form, noble metal fission products in metallic form, and noble gas fission products. The molten salt fuel is reusable in a new blanket and therefore is not considered a waste product.

All metallic components, including heat exchangers, vessel and all radioactively contaminated equipment will be cleaned using processes such as fluorine plasma etching that do not require the addition of solvents or reagents. The recovered actinides will be directly added to the molten salt fuel and burned in subsequent operations. The graphite moderator and reflector will be burned in a controlled environment, the residual fission products and actinides released by the burn will be scrubbed and recycled when appropriate in the blanket. Calcium carbonate will be the final form of the decommissioned graphite. No carbon will be released. Calcium carbonate waste will be mixed with the activated scrap metal and poured as low level radioactive concrete slabs for shallow burial on site.

All the troublesome fission products with long half-lives are transmuted to non radioactive materials in the ADEP blanket. Some shorter-lived isotopes, on the other hand, are too difficult to transmute, owing to their very small absorption cross section. No transmutation would probably be attempted on ⁹⁰Sr or ¹³⁷Cs. With a 30-year half-life, these isotopes will likely be lumped together with the other short lived and stable solid fission products, diluted with a salt of appropriate heat

capacity and then stored in supervised, engineered containers for up to 300 years before their radioactivity is down to safe levels. Alternatively, and at a high cost, both ^{90}Sr and ^{137}Cs , could be transmuted in special high-flux, low-multiplication blankets, where their effective half-life would decrease perhaps by a factor of 4. All noble metals recovered from the separation of fission products, including the weakly radioactive Zr and Pd, will be alloyed and stored in low level storage areas or recycled as seen appropriate. The noble gas Krypton, with a half-life of 10 years will have to be monitored for about 100 years in appropriate sealed containers.

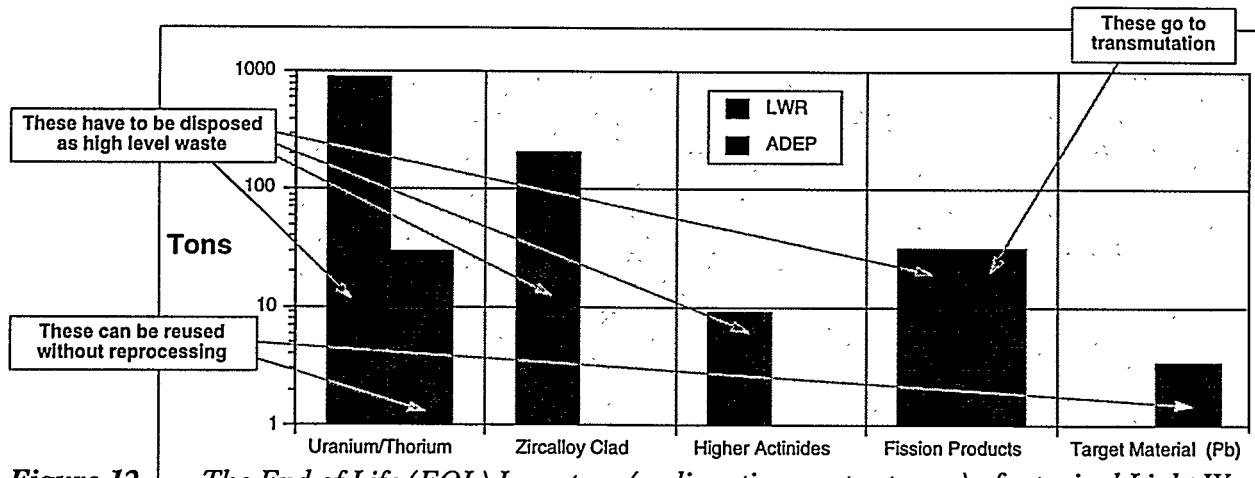


Figure 12. The End of Life (EOL) Inventory (radioactive waste stream) of a typical Light Water Reactor (LWR, 30 years at 3000 MWt) compared with the inventory of an Accelerator Driven Energy Producer (ADEP, 6x500 MWt modules)

Figure 12 compares the End of Life (EOL) radioactive waste stream of ADEP with that of a typical Light water reactor (LWR). LWR spent fuel must be reprocessed at plant EOL in order to be reused. The most optimistic estimates of fuel utilization for LWR systems range up to 40% following complex and repeated refabrication of fuel. On the other hand, ADEP fuel and target material are just as effective at End of Life as they were at the beginning of operations and ready to be placed in a new blanket without reprocessing. Full utilization of fuel and target material is readily achieved. Therefore the extent of the EOL radioactive inventory of ADEP and all other ADTT systems is driven by the cleanup efficiency, not inherently by the cycle.

A PROLIFERATION RESISTANT NUCLEAR SYSTEM

ADEP has very strong nonproliferation features:

- Complete fuel utilization without reprocessing
- No actinide waste stream
- ^{239}Pu production is very small and denatured with ^{238}Pu
- ^{233}U can be denatured with ^{238}U
- Power balance or thermal map can clearly identify diversion
- No fissile material is required for start-up or feed
- No fissile material needs to be extracted from the fuel
- On-line separations inhibit diversion
- System operation can be disrupted without radioactivity release

The ADEP concept could offer significant Safeguards and Non-Proliferation advantages over existing nuclear power production systems. The present system for nuclear power production involves different sites for fuel enrichment, fuel fabrication, commercial power production facilities, reprocessing plants and the disposal of high level wastes. The operations at each of these steps and the transfers between them offers opportunity for loss or diversion of nuclear material for clandestine purposes. The public is very concerned about the transport of spent fuel and high level waste in the back end of the fuel cycle and the safe storage of this high level waste for long periods. The ADEP system does away with the complex infrastructure of the current system and its transportation needs. Continued power generation at the site is possible through the use of the benign natural fuel thorium. Nuclear waste is destroyed at the same site without reprocessing, refabrication, or long-term actinide waste storage. Only one truckload of thorium fuel need be delivered to the site every thirty years. With the full development of the concept, nothing need leave the site except electric power.

The primary measures for identification of attempts to divert nuclear material within the existing nuclear power infrastructure involve physical security and nuclear material accounting supervised by the International Atomic Energy Agency. This material balance accounting requires taking differences between large numbers, which places severe demands on measurement accuracy to detect diversion of small amounts of material. In the ADEP system, only thorium and (possibly) a small initial charge of fissile fuel are brought into the power production site. The thorium is fed into the system and no actinide is removed. The thorium and all other actinides are confined to the molten salt loops in the blanket. Therefore by monitoring the thorium input rate, by knowledge of the system efficiencies, and by measurement of the electric power output of the system, it should be possible to detect the production of any fissile uranium which was not used for electric power production, without nuclear material measurements at any point in the system.

No ^{233}U or ^{233}Pa is ever isolated either during the operation or during the fuel cleaning process, and because of the subcritical nature of the ADEP system, the inventory of fissile material can in principle be burned down to completion.

The addition of 7-10% ^{238}U in weight to the initial thorium load ensures that the uranium present in an ADEP system at any time during its operating life will be unusable as weapons material without isotopic separation. Hardly any ^{239}Pu is produced (less than 3 kg is present in each 500 MWt modular blanket at any given time, dissolved in over 20 cubic meters of very hot salt and rendered unusable by the considerable fraction of the heat-laden ^{238}Pu isotope present). No buildup is possible because of the continuous cleaning of the fuel.

Even the surreptitious isolation of the protactinium inventory after a commandeered shutdown is not conducive to a high-risk proliferation scenario. Here again the dilution of the inventory (30 kg of ^{233}Pa at equilibrium 20 m³ of salt) and the 27-day half-life of the protactinium would ensure that a significant isolation of the potential ^{233}U inventory will not be possible by terrorist groups. The uranium that would be produced in this scenario is also not as desirable as ^{239}Pu in the manufacturing of weapons. Whereas the nuclear properties of ^{233}U and ^{239}Pu are roughly equivalent, the hard gamma radiation associated with the ^{232}U isotope and its daughters, always present with the bred ^{233}U , would preclude hands-on work and allow easy detection of smuggled material and of secret warheads. Clearly there are easier ways to procure better weapons-grade material.

The ADEP technology offers new opportunities for improving protection against production and diversion of nuclear material. It is important to attempt to build in as many of these features into the design from the beginning, instead of attempting to retrofit them into otherwise unsatisfactory designs, as it has been the case with current nuclear power technology.

ADEP Operating Parameters

fission power = 500 Mw power density = 6 w/cc
active core diameter = 450 cm
active core height = 500 cm
average core flux= $1.2 * 10^{14}$ n/cm²s average fuel flux= $5.0 * 10^{13}$ n/cm²s
molten salt composition = LiF-BeF₂-Th(U)F₄ 69-18-13 mole % (nominal)
Fuel salt temperature in-out heat exchangers = 1000 - 850 °K
salt volume = 20 m³ (total)
graphite volume = 250 m³ (total) 80 m³ (core)
graphite mass = 180000 kg
salt volume in core = 8 m³
salt volume in heat exchangers and plenums = 12 m³
salt flow in heat exchangers = 0.63 m/s
salt flow in core = 1800 kg/s - 0.96 m/s
secondary salt temperatures = 770 °K (in) 840°K (out)
pump power = 0.14 Mw (total for 6 primary pumps)
target material = molten lead
target size = 40cm diameter x 90 cm length
target lifetime = 2 years (exposed structure) indefinite (lead)
target neutron production = 22 n/p at 800 MeV

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