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ACCELERATOR-DRIVEN SYSTEMS FOR NUCLEAR WASTE TRANSMUTATION

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ABSTRACT

The renewed interest since 1990 in accelerator-driven subcritical systems for transmutation of commercial nuclear waste has evolved to focus on the issue of whether fast- or thermal-spectrum systems offer greater promise. This review addresses the issue by comparing the performance of the more completely developed thermal- and fast-spectrum designs. Substantial design information is included to allow an assessment of the viability of the systems compared. The performance criteria considered most important are (*a*) the rapidity of reduction of the current inventory of plutonium and minor actinide from commercial spent fuel, (*b*) the cost, and (*c*) the complexity. The liquid-fueled thermal spectrum appears to offer major advantages over the solid-fueled fast-spectrum system, making waste reduction possible with about half the capital requirement on a substantially shorter time scale and with smaller separations requirements.

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INTRODUCTION

Light-water reactors (LWRs) now deployed worldwide produce nuclear power from the fission of ^{235}U fed into the reactors and from the ^{239}Pu produced in the reactor. Other isotopes of plutonium are also produced, along with the minor actinide elements neptunium, americium, and curium. Much of the world has viewed the plutonium as an asset that could be recovered from the spent fuel. The plutonium then could be returned to LWRs for further burning as mixed oxide (MOX) fuel, or it could be sent to fast-spectrum plutonium breeder reactors to breed even more. Ultimately all of the plutonium and minor actinides (Np, Am, Cm) would be destroyed by fission, and then nuclear waste would consist mostly of fission products. Devising suitable storage canisters to permanently confine this waste is not possible, since some of the fission products and remnant actinides have half-lives in the range of thousands to millions of years. Geologic confinement was therefore adopted in order to rely on the natural geologic structure to confine the wastes after the canisters used to place the waste underground had deteriorated.

However, not all of the plutonium and minor actinides (PMA) can be destroyed by MOX burning. Therefore, fast breeder reactors, which had been invented for plutonium production, were reconsidered for plutonium destruction (1). Unfortunately, breeder-reactor technology has faltered owing to costs, safety concerns, a substantially longer than anticipated delay in the need to breed plutonium, and concern about the proliferation aspects of a worldwide plutonium-based nuclear energy economy. Accordingly, the United States adopted a policy of placing the reactor spent fuel in repository storage, without reprocessing and therefore without destroying either the plutonium or the minor actinide. This not only increased the repository-confinement performance requirements but also raised concerns about the possibility of recovering plutonium from the repository for weapons. Since no nation seems willing to store another nation's nuclear waste, this policy assures that every nation that has nuclear power, or aspires to have it, would have its own repository, which would have to be guarded indefinitely.

Insufficient attention was also paid to the possibility that criticality might lead to explosive release of nuclear energy in the repository. After the waste canisters have failed, the proper function of the repository geologic structure is to confine the waste if it wanders from its emplacement site. Rearrangement of the waste is also possible by accidental or purposeful intervention. Recent work (2) has shown that if the plutonium were to reconfigure into critical configurations, the neutron-moderating feature and the energy-confinement capability of the rock could lead to nuclear explosions with yields perhaps in the range of several tens of tons or higher. Although the likelihood of such events arising from natural causes might eventually be found to be acceptably small, it is possible to eliminate the risk completely by destroying actinide waste.

Application of scientific procedure to the establishment of geologic storage has unfortunate political consequences. Nuclear waste is viewed by much of society as the nation's most dangerous waste. Science responds that it can identify and characterize the one best site on any nation's territory for storage of this waste. Since there is a single best site, all of the nation's waste should permanently be stored at this best site. The consequence is that one community becomes host to all of the worst of the nation's waste. Such a situation does not exist for any other waste, and the evident unfairness of this consequence appears to be well recognized, since no government has yet selected the site for location of a waste repository. However, if by transmutation the waste toxicity were reduced by a factor of about 1000, then commercial waste, after an interim storage period of about 300 years, would approach the low toxicity necessary to eliminate the need for disposition by deep underground geologic storage. In that case, there would no longer be a need to identify one best site. Almost every region or state of a country could identify several sites to handle the region's nuclear waste, using siting studies conducted like those for any other waste. Such a situation would take the federal government out of the waste-siting issue. In the United States, siting would become a state issue; the only role of the federal government would be to establish regulations for site selection and qualification. Therefore, transmutation might either reduce the performance requirements for geologic storage of waste or eliminate altogether the need for central geologic storage for commercial nuclear waste.

The need for a new approach to destruction of wastes is perhaps best illustrated by considering the array of existing nuclear technology (Figure 1, *top*) that might be brought to bear on the destruction of plutonium and the minor actinides. The technology includes LWRs, fast breeder reactors, reprocessing, partitioning, fuel fabrication, fuel refabrication, interim storage, and repository storage and requires repeated transportation between these facilities. Figure 1 shows how this technology might be combined and implemented. Not included is an accelerator-driven system (ADS) for the destruction of Np, Am, and Cm,

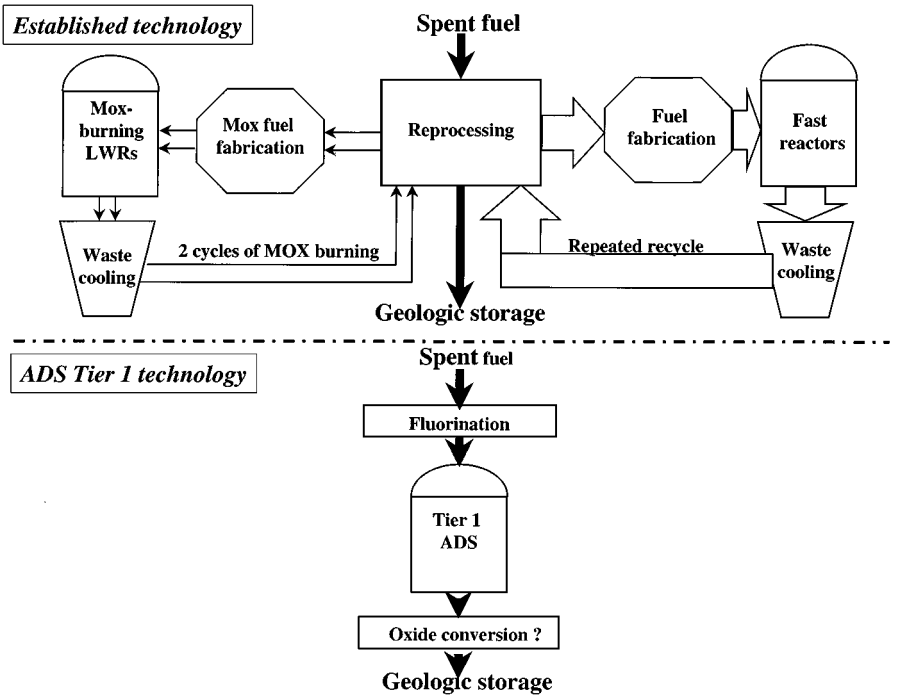


Figure 1 Comparison of established and new technology for waste destruction. *Top*, the deployment of current technology including light-water reactors for mixed oxide (MOX) burning, fast reactors, reprocessing, and fuel fabrication. MOX recycling presently appears to be limited to two cycles because of problems in reprocessing and fuel reactivity. The plutonium and minor actinide left after MOX burning is sent to fast reactors for repeated recycle. Fission product and a small amount of minor actinide are sent to geologic storage. *Bottom*, the requirement for the accelerator-driven system Tier 1 technology described in the text. The Tier 1 technology leaves somewhat more actinide for the repository than the established technology, but this probably is not important, since both systems require geologic storage of the remnant waste. The evident simplicity of the waste reduction with the accelerator is expected to compensate for the additional costs associated with the accelerator, although not all versions of the accelerator-based technology offer this simplicity.

which might be required. The lower portion of the figure shows the reduction in complexity that appears possible with an ADS described below.

Such a system might be deployable in France, where all of the component technologies exist, but many countries including the United States have not demonstrated the technology shown in Figure 1. Smaller countries such as Sweden or the Czech Republic have little hope of implementing such a complex system for disposition of their nuclear waste and might have serious difficulties

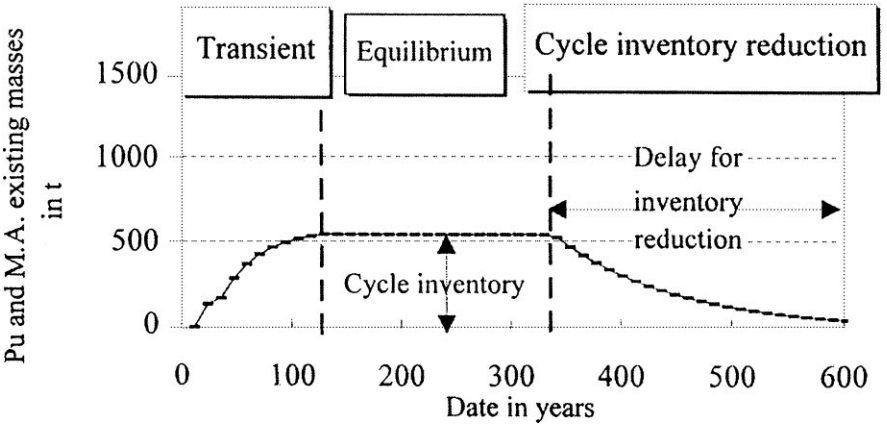


Figure 2 Plutonium and minor actinide inventory for France with existing technology. The figure shows the effect of implementation of the existing technology (Figure 1) on the plutonium and minor actinide inventory. The present inventory of about 200 tons stabilizes at about 600 tons in about 100 years. Part of the reason that the 600-ton inventory must be carried is the large inventory of fuel in storage, reprocessing, and the fast-spectrum reactors. If it is decided to close out nuclear power 200 years later, the existing technology would require about 270 years to reduce the inventory by a factor of ten.

in transport and other issues as they try to take advantage of the facilities of larger nations. Any new waste-disposition technology that is developed should lead to a great simplification of the infrastructure illustrated in the upper part of Figure 1.

The time scale for seeing benefits from waste destruction and the inventory requirements are also important considerations. Figure 2 shows the implementation of the infrastructure shown in Figure 1 (*top*) for destruction of PMA. It indicates the amount of PMA in France in three periods, assuming that the present nuclear power production rate remains stable for the next 300 years (3). A transient period is shown, during which the system comes to equilibrium. An equilibrium period of 200 years is assumed, followed by an inventory-reduction phase after a decision to close out nuclear power. Presently France has about 200 tons of PMA. The figure shows that this amount would grow to 600 tons before stabilization, partly because of the long time required for fast-spectrum systems to reach equilibrium and partly because of the inventories that would be carried inside and outside of the operating facilities of Figure 1. The French public would have to wait 100 years before the plutonium stops increasing. If this same system were deployed for the world's 400 existing reactors, the inventory would stabilize at about 4000 tons. The US National Academy of Sciences

estimates that this is sufficient plutonium for production of nearly one million nuclear weapons (4). Of course, it is important to note that if geologic storage after a single use, the option advocated by the United States, were implemented worldwide, the plutonium inventory would be five times 4000 tons by the year 2050.

The inadequacy of existing technology is also emphasized in the inventory-reduction phase of Figure 2. If phase-out of commercial nuclear power began at some point after equilibrium was established, 200 years would be required to reduce the inventory by a factor of five. This slow response time at the beginning and the end results mainly from the large inventory of PMA that the fast-spectrum reactors require for operation. The advent of a fast-spectrum ADS would not change this time scale significantly. One of this chapter's main points is the advantage of a thermal-spectrum ADS over a fast-spectrum ADS in terms of inventory reduction and time response at deployment of waste burning or at a shut-down of nuclear power.

THE CONCEPT OF AN ACCELERATOR-DRIVEN SYSTEM

Figures 3 and 4 show an ADS for transmutation of commercial nuclear waste (5). The system is driven by an accelerator in order to start many fission chains that run for a relatively short time, in contrast to a reactor, for which the chain runs continuously until the reactor is shut down. The effective multiplication factor k_{eff}, which is 1.00 for a reactor, can be reduced to the range of 0.98–0.95. One significant advantage of this mode is that the neutrons otherwise required to maintain the chain can be put to other uses, in particular, the destruction of nuclear waste. A second benefit is that constraints on reactor design required to keep $k_{\text{eff}} = 1$ can be relaxed and a broader design parameter space is practical. For example, an accidental injection of reactivity that would lead to a major accident for a reactor with $k_{\text{eff}} = 1$ would hardly be noticed with $k_{\text{eff}} = 0.96$.

The neutrons are produced by the accelerator beam via the spallation process. In most designs for an ADS, protons from the accelerator strike a liquid heavy-metal target, ejecting neutrons or protons in the forward direction with a lower energy than the incident particle has. These neutrons, and sometimes protons, then strike other nuclides, which in turn eject other forward-moving lower-energy particles. The cascade continues until the energy is spent. The total cascade length is about one meter for a 1-GeV beam energy. In any of these nuclear collisions, the struck nucleus is always excited to some degree and these hot nuclides boil off neutrons. About 90% of the neutrons are produced in boil-off reactions, while the remainder stem from the direct reactions already

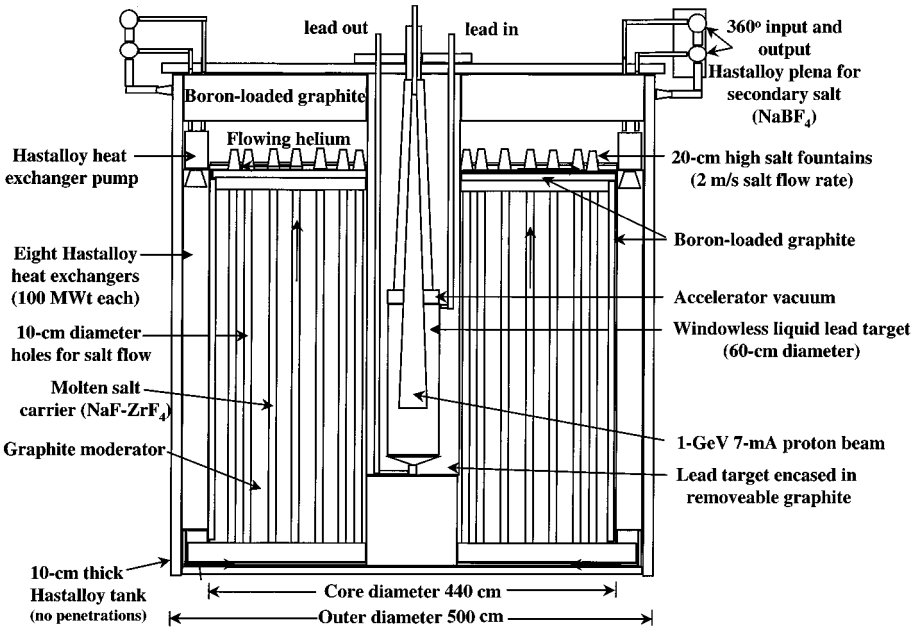


Figure 3 Design for once-through thermal-spectrum transmuter (750 MWt) for commercial reactor spent fuel. The system is a graphite-moderated liquid-fueled assembly with $k_{\text{eff}} = 0.96$; it operates at a fission power of 750 MWt. It is capable of transmuting the waste from one 3000-MWt LWR at the rate it is produced. The actinides and fission product continuously flow through the system via the carrier salt $NaF\text{-}ZrF_4$. The salt flows upward through holes in the graphite, then across the top to the outside of the system, down through internal heat exchangers, and through a plenum back into the graphite moderator. A salt-to-graphite volume ratio of 1/20 assures a well-thermalized spectrum.

discussed. The boil-off neutrons are emitted isotropically, in contrast to the forward-moving direct-reaction neutrons. Altogether about 30 neutrons are produced by each 1-GeV proton, mostly with energies between 1 and 10 MeV. They slow down by inelastic scattering until they reach about 600 keV, and by elastic scattering in the lead or surrounding material below 600 keV. These neutrons enter a blanket made up of hexagonal graphite rods with channels containing fissile material. Between 35% and 60% of the accelerator-produced neutrons start fission chains, which run for about 25 fissions before stopping. Therefore, if 50% of the accelerator-produced neutrons start a fission chain, one 1-GeV proton would generate $30 \times 25 \times 0.5 = 325$ fissions, which corresponds to an energy release of $325 \times 0.2\,\text{GeV} = 65\,\text{GeV}$. For a beam current of 12 mA, the fission power level would be 750 megawatts thermal (MWt).

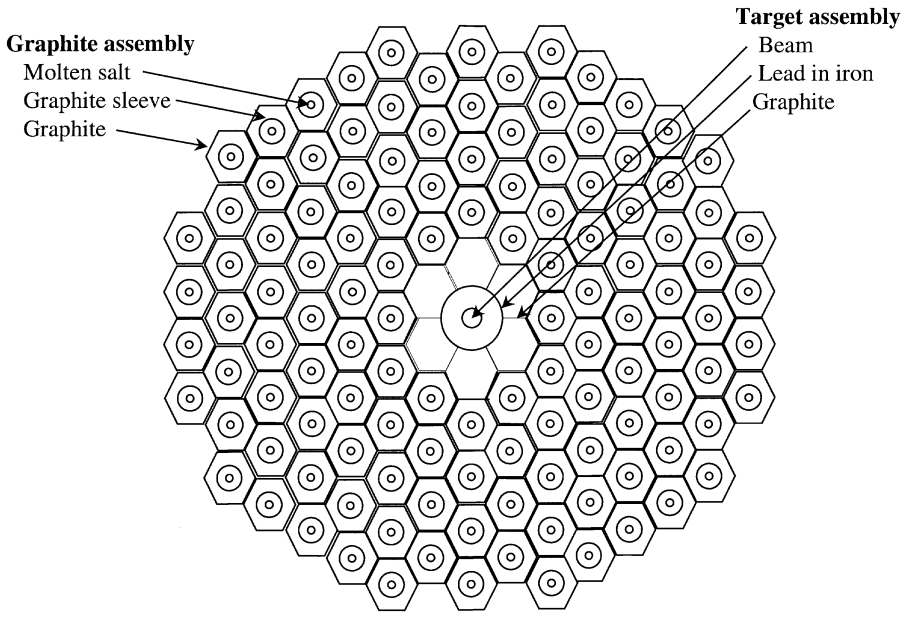


Figure 4 Plan view of Tier-1 proton beam target and core. Spallation neutrons are produced where the beam strikes in the center. The 10-cm beam diameter is considerably smaller than the 50-cm diameter of the lead target, allowing the higher-energy neutrons to multiply in the lead surrounding the beam column. The hexagonal 100-cm-diameter graphite moderates the neutrons into the thermal range before they enter the blanket region. The hexagonal graphite assemblies making up the blanket each have a 7-cm-diameter hole in the center for the molten salt flow and a 20-cm-diameter removable sleeve. The assemblies fit into a graphite or Hastalloy-N metal plenum at the bottom such that salt flow is only possible in the hole and not outside of the sleeve or between the assemblies. Solid hexagonal graphite assemblies providing a reflector thickness of about 50 cm around the outside are not shown.

The heat is deposited in the medium and is extracted and converted to electric power with a rather high thermal electric efficiency of about 42%, made possible by the high (up to 720 C) operating temperature of the molten salt or liquid metal design. Some of this power is fed back to the accelerator, which generally operates with a buss bar efficiency (6) of about 45%. For these numbers, the portion of the generated power required to drive the accelerator is $1/(65 \times 0.42 \times 0.45) = 8.5\%$. In most designs, only about 35% of accelerator-produced neutrons start fission chains. Clearly there is much to be gained in capital cost reduction and in power output by designing for the maximum efficiency in accelerator neutron utilization.

The target-blanket design in Figure 3 consists of a graphite moderator with actinide fuel and fission product carried by a molten salt of NaF-ZrF_4. The

spent-fuel assemblies are first fluorinated completely, so that everything is converted to fluoride except the noble gases. The resulting UF_6, some of the ZrF_4, and a few of the more volatile fission products are released as gases. NaF is added to the rest of the fluorinated actinides and fission products, and these waste products are fed into the blanket as solute in the NaF-ZrF_4 carrier. The salt is removed from the blanket periodically or continuously, depending on the design. There are several variants on the front-end and back-end chemistry for the various ADS concepts. Some systems feed no fission products to the blanket. Others require no back-end separation. Elimination of as much chemistry as possible is highly desirable, since more development is probably needed in chemistry than in the accelerator and reactor-like components of the system.

ADS Power Production Influence on Deployment

The fission power of the ADS of Figures 3 and 4 is 750 MWt. This is the power produced in the destruction of the 300 kg/year of PMA produced by a typical LWR operating at 3000 MWt and burning 1200 kg/year of fissile material. The United States would need 100 systems of this size (1/4 of an LWR), or 25 if they were scaled up in size to 3000 Mwt, just to keep up with the waste generation from its existing reactors. Clearly the introduction of ADS systems is a major undertaking that is impractical unless the system sells sufficient electric power to pay most of its capital and operating costs. The scale of the required deployment of the ADS fleet eliminates some options.

For example, some favor transport of the waste to one central site next to a repository where it could also be transmuted. This is logistically impractical for the United States, since it would require the introduction of more than 25 GWe into the grid from a single point. No national grid operates a single site that even approaches such massive power-generation capability. Another possibility for the United States would be to designate perhaps five sites for waste burning, reducing the power generation to 5 GWe per site. Although this is more nearly manageable, it is still substantially more power than any single US site produces. It is important to note that power production is broadly spread in the United States, with some variation in price (depending on the local market size and production capability), which is not completely flattened by long-distance transmission. The introduction of five GWe at any one site could disrupt the local market, with the probable result that the ADS-produced electricity would have to be sold at cut-rate prices.

A third alternative would be to destroy the waste on the site of the reactor. This arrangement has several potential advantages and disadvantages, but there is a major advantage for marketing power. A market already exists for power from the site or the reactor would not have been built there, and the addition of a transmuter to the site would increase the power output from the site by only 25%. Partly for the reasons given above, the size of the ADS discussed here

is 750 MWt, which seems to be the minimum size for practical deployment. Scaling the technology up to the size of commercial LWRs should perhaps await successful operation at this more modest size.

Criteria for ADS Design and Development

In order to effectively compare one system with another, it is important to summarize the important criteria. The ordering of the criteria in the following list should not be interpreted as a ranking of relative importance.

1. New nuclear technology should not increase the cost of nuclear power. The costs for nuclear power already are barely competitive for much of the world. In assessing total costs, it is necessary to recognize both government and private-sector costs, when both are present, and to include the development costs of the new technology.

2. As a new technology, transmutation should reduce the infrastructure complexity rather than add to it. Nuclear technology presently requires a highly complex technology infrastructure. Its primary competitor in much of the world is natural gas, for which the infrastructure requirement is gas wells, gas transport, and power plants.

3. The new technology need not use nuclear fuel optimally. Since the energy density in nuclear fuel is 10 million times higher than in chemical fuels, recovering the last 30–50% of energy possible does not seem especially important. It may be worth pursuing if the primary objective is to resolve other more important concerns such as the elimination of material useful for weapons in the fuel cycle. In this particular case, more complete energy recovery is a side benefit.

4. Overall operational safety in the nuclear infrastructure should be improved. Any risk addition with the new technology should be offset by risk reduction in the other elements of the infrastructure, or perhaps by elimination of some elements of the nuclear infrastructure.

5. Because the fundamentals of conventional nuclear weapons technology are well known and nuclear weapons are a significant international concern, the new technology must significantly reduce the likelihood of access to weapons-useful material. Both normal and off-normal evaluation of the operation of any new system must be performed from the proliferation perspective. Accelerator-driven technology offers the potential for reduction in the accessibility of weapons-useful material for many parts of the fuel cycle, including fuel preparation, fuel burning, and short-term and long-term waste storage.

6. The new technology should not allow a surprise downstream of new classes of nuclear weapons, which would halt the deployment of new technology after a long development period. Careful examination of the weapons-usefulness of new material produced in the course of transmutation is essential. In addition, the explosive potential (by spontaneous, accidental, or deliberate means) of material in permanent storage and at all other phases of the fuel cycle must be well understood.

7. The new technology and associated infrastructure should not require a single national site for permanent waste storage. Reduction of the toxicity makes benign engineered storage of remnant nuclear waste possible at many sites on a nation's territory.

8. The new technology should reduce the need for strong international oversight. The opportunities for cheating on international controls of nuclear technology are significant and are likely to remain so. In addition, the perceived benefits of abrogation of existing agreements are likely to remain strong. Therefore, any revision in the present nuclear infrastructure should reduce the amount of weapons-useful inventory in the fuel cycle as much as possible. A very large reduction in weapons material availability may be achieved by closing the fuel cycle. A further improvement by a factor of as much as 100 could come from the implementation of the accelerator-driven technology.

9. The introduction of accelerator-driven technology should provide major advances, rather than incremental improvements, in the resolution of current fuel-cycle concerns. Improvement over existing nuclear technology by an order of magnitude or more is possible using transmutation technology. The public, which watches nuclear matters closely, is likely to appreciate and support major nuclear advances much more than proposals for incremental improvement.

10. The transition from start-up to equilibrium should not be characterized by unsafe conditions nor generate other negative impacts.

DESIGN OPTIONS FOR THE ACCELERATOR-DRIVEN SYSTEM

Current ADS concepts usually include an accelerator; a liquid heavy-metal target for neutron production, surrounded by a subcritical blanket containing the actinide and fission products; associated front-end and back-end chemistry; and electric-power production facilities. A number of designs have been proposed and evaluated over the past decade as the understanding of the ADS

has evolved. Instead of describing these systems, this chapter focuses on describing and evaluating the primary issues for the ADS designs presently under consideration. The principal design choice to be made is between thermal- and fast-spectrum. The issue of liquid or solid fuel is also significant. A comparison of representative fast- and thermal-spectrum designs will address these issues (7, 8).

Fast-Spectrum Approach

The fast-spectrum ADS currently has many adherents. Most of the proposals made for transmutation employ a fast spectrum. These proposals usually feature either oxide-fueled and sodium-cooled systems with an average neutron energy of about 75 keV, or heavy-metal–cooled and solid metallic-fueled systems with an average spectral energy about three times higher. At least part of the interest in the fast-spectrum system derives from the many years of effort in nearly all nuclear countries to develop fast breeder-reactor technology. Although most of these national programs have been closed, a strong desire continues on both the political and technical sides to see benefit from the large investment made in this technology. These systems have a definite advantage in neutron economy, but as shown below, this advantage is not easy to exploit. Furthermore, existing designs do not productively use the extra neutrons from the accelerator. The fast-spectrum ADS has the disadvantages of requiring a long time to reach equilibrium and requiring a large waste-actinide inventory.

The principal design advantage of the fast spectrum is its capability to induce fission in all of the actinides, although the probability for fission in a fast spectrum of the even-even nuclides is only 30–50% of that for the even-odd isotopes. If the nucleus does not fission, then the neutron is captured and lost. If fission occurs, then about 2.8 more neutrons are produced after absorption of the neutron causing the fission and the nucleus is destroyed. One can calculate the balance between neutrons produced by fission and neutrons lost by absorption for each isotope present in the transmuter. This balance has been calculated for all of the actinides and is found to be positive for all actinide nuclides in LWR spent fuel (9). For a fast reactor with oxide fuel, the net neutron generation averaged over all of the nuclides except the uranium isotopes is about 1.2. This important result indicates that the neutron economy for a fast spectrum is excellent, so that even with parasitic capture and fission-product capture there would be a large neutron excess. It also shows that fast-spectrum systems do not require accelerator-produced neutrons to supplement the neutron economy for transmutation. This feature begs the question (answered below) of what useful role the accelerator plays in an accelerator-driven fast-spectrum transmuter.

Although the cross sections for actinides in a fast spectrum offer an advantage from the neutron-economy perspective, they carry the disadvantage of their

small size. Typically the fission cross section σ_f for the fast spectrum is about two barns. The burn-up rate is inversely proportional to the fission cross section and the effective flux ϕ_{eff}, which is typically about 2×10^{15} n/cm^2-s. (While the flux in the transmuter is usually about 7×10^{15} n/cm^2-s, the fuel typically spends about twice as much time outside the transmuter—in fuel cooling, destruction, separations, and refabrication—as inside, so that the actual flux must be reduced by a factor of three to find the effective flux.) The mean time for a reaction in this flux, given by $1/\phi\sigma$, is about 7 years. Because not all absorptions lead to fission of the fissile nuclides and most reactions do not lead to fission in the fertile nuclides, more than one absorption is required on average for destruction of a single nucleus. Therefore the mean time for destruction is about 15 years. If the fuel spends twice as much time outside the transmuter as inside, as in the example analyzed in Figure 2, the time for destruction is about 45 years, which is probably comparable to the lifetime of the transmuter. This long time might not create unresolvable problems, but a system that reaches equilibrium much sooner would be desirable.

The smaller the cross section, the larger the fuel concentration needed to achieve a particular reaction rate. The reaction rate R in units of absorption per cm^3-s is given by $R = \phi\sigma N$, where N is the number of actinide nuclei per cm^3. The power P of the transmuter is given by $P = VRE_f$, where E_f is the energy released per fission and V is the volume. Therefore the inventory $I = VN = P/\phi\sigma E_f$. For a power of 750 MWt, an effective flux (which takes account of fuel outside of the transmuter) of 2×10^{15} n/cm^2-s, and a cross section of two barns, the inventory is 2.3 tons. The power level of 750 MWt was used because this is the amount of energy released in the fission of the approximately 0.3 tons/year of actinide produced by an LWR operating at a thermal power level of 3000 MWt. Therefore, for the United States, with about 100 LWRs, the inventory of plutonium and minor actinide that must be maintained for a fast-spectrum system to burn waste actinide as fast as it is produced would be $2.3 \times 100 = 230$ tons. (This may be compared with the 1200 tons produced in these reactors over an assumed 40-year lifetime.) This burning rate does not reduce the US inventory but merely prevents it from growing. In order to double the destruction rate so as to reduce the inventory, twice as many transmuters and an inventory of 460 tons would be required. The inventory required for the thermal-spectrum system burning waste at the same rate is smaller by a factor of about ten.

Thermal-Spectrum Approach

The thermal-spectrum neutron economy is less favorable than that of the fast spectrum, so taking maximum advantage of the neutrons available is an important element in all thermal-spectrum designs. Parasitic capture of neutrons

by the system structure or by materials required to contain or transport the fuel must be minimized; accordingly, designs typically feature heavy water, enriched ^{7}Li, Be, and Zr. Parasitic neutron capture in fission products is also a concern. The reduction of fission-product absorption is one of the primary drivers toward liquid-fuel systems with capability for online removal of fission products by rapid recycling without fuel destruction and refabrication.

The disadvantages of rapid recycling are the potentially large amount of material that has to be processed and the high-performance separations required. If the purpose is to remove fission products but to keep all actinides in the system until they are destroyed by fission, the ability of the separation process to prevent leak-through of actinide into a fission-product stream is important. In a single cycle the leak-through of actinide might be small, but because there are many cycles the actinide has many opportunities to leak through, and the actinide content in the fission-product exit stream may be much greater than that achieved in a single separation (F Lelievre, private communication). Of course, rapid recycling, with liquid fuel and the large volume processed, could add considerably to the overall cost for transmutation.

The Molten-Salt Reactor Experiment (MSRE) (11) faced these problems in the late 1960s, attempting to demonstrate a Th-U–cycle thermal breeder. The system used a LiF: BeF_2 carrier for thorium, for the ^{233}U bred from the thorium, and for the fission products. While the performance of the molten salt reactor and the materials and separations components were satisfactory, the MSRE demonstrated that a breeding ratio of more than 1.03 probably would not be possible. Therefore, this approach was dropped, since by then it had been demonstrated that better breeding was achievable with a fast-spectrum reactor.

Nevertheless, the small-inventory feature of the thermal-spectrum system, made possible by the large fission and capture cross sections of the PMA, is highly attractive for waste destruction. Also, the neutron-economy issue is of less importance for destruction of LWR waste, since fissionable plutonium is fed into the system with other parts of the waste. Although the breeder must produce more fuel than it burns, the waste burner's neutron economics is helped along by the plutonium fed in. The addition of an accelerator to enable subcritical operation supplements the neutron economy further. Of course, the accelerator adds capital and operating costs to the system. If the amount of waste were small, cost would not be so important. However, the waste quantities are large, and destruction requires a large deployment of reactor-like systems, which must pay almost all of their costs by electricity sales. Therefore, if the benefits of small inventory and rapid approach to equilibrium are to be realized from the thermal spectrum, means must be devised to address the problems of neutron economy, separations efficiency, and cost.

At this point in the development of the ADS, the underlying principles are well understood for both thermal- and fast-spectrum systems. However, implementing this knowledge in a practical system requires compromises in design that affect performance in waste destruction. Therefore, a major focus of this paper is to describe the design and compare the performance of the fast- and thermal-spectrum systems, using one example of each. The chosen example of the fast-spectrum system is the design of the CERN (7) group led by Rubbia, which is the most thoroughly developed fast-spectrum design. Thermal-spectrum design has lagged in recent years; most recent is the Tier 1 and 2 system proposed (8) by the ADNA Corporation (Accelerator-Driven Neutron Applications). Significant differences between these approaches, both in technology and in performance, will become apparent. A later section briefly describes thermal- and fast-spectrum systems in the framework established for the ADNA Corporation (12) and CERN proposals. The input feed is that calculated using the Origin Code and reported in the 1990 document CURE (12).

THE THERMAL-SPECTRUM TIER 1 AND TIER 2 SYSTEM

Figure 5 shows a thermal-spectrum two-tiered system for dealing with the LWR waste stream. Tier 1 provides the option to destroy most of the actinide while recovering the fission energy for sale as electric power. Furthermore, it converts the plutonium to a form uninteresting for nuclear weapons, reducing the likelihood that the waste would be disturbed after placement in repository storage. It also converts the plutonium to an isotopic mixture for which criticality in a geologic repository would be virtually impossible even if the implaced waste were rearranged by natural or other means. The design is a continuous flow-through system with waste entering and leaving at the same rate. The actinide mixture is significantly transformed before it leaves. The system requires no back-end separations and only modest front-end separations compared to current reprocessing technology. This large reduction in chemistry requirements is a major factor in achieving practicality of the system and in reducing costs.

The output from this system could be sent to geologic storage or to the Tier 2 system, where actinide would essentially be completely eliminated. The Tier 2 system, however, would require full front-end and back-end separations and a somewhat larger accelerator, so its costs would be higher than the Tier 1 system's. On the other hand, the Tier 2 system must operate on only one fifth of the waste transmuted under Tier 1, so the higher costs might be tolerable. Of course Tier 2 would not be necessary if Tier 1 has already denatured the waste to such a degree that its geologic storage is acceptable to the public. The Tier 1

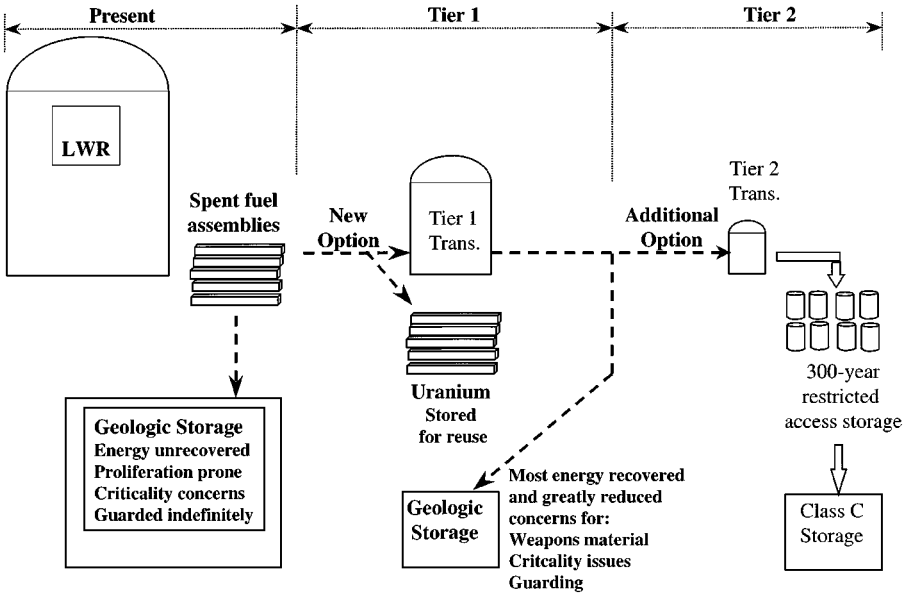


Figure 5 New options for nuclear waste disposition. To date, the United States has focused exclusively on the once-through process for disposition of commercial spent fuel, with the spent fuel assemblies going directly into geologic storage. The stored waste contains a large amount of potential nuclear energy and potential criticality is a concern. The plutonium and neptunium is weapons-useful material requiring perpetual guarding. The Tier 1 system would extract most of the fission energy and greatly reduce concerns for weapons-material criticality. It would require no back-end actinide and fission-product separations. However, it would not eliminate the need for geologic storage. The Tier 2 option would reduce the actinides from Tier 1 by a factor of about 100 for a total reduction by 500. The most bothersome fission products are reduced by a factor of 10–100, enabling eventual Class C storage. The size (area) of the three nuclear units is proportional to the deployment required to destroy the waste as fast as it is produced.

and Tier 2 designs are similar in most respects, except for the separations and the carrier salt.

The Tier 1 Accelerator-Driven System

The thermal-spectrum Tier 1 ADS described in detail next is a once-through system for commercial waste destruction. It requires only uranium and fission-product removal on the front end and no separations at all on the back end. The system is illustrated in Figure 6, which shows an ADS with liquid waste from commercial LWRs being fed into the system and transmuted product being removed for direct transfer to geologic reactors storage. The size of the two domed units is proportional to their fission-power generation. The fission heat is converted to electric power and sold in the commercial grid.

The only step required for preparing the feed of the LWR spent fuel into the transmuter is the fluorination of the waste, which allows the removal of the uranium and the separation of the fission products. The PMA and the zirconium cladding as fluoride salts are combined with NaF. A liquid eutectic of NaF-ZrF_4 is formed as a carrier for the PMA fed into the transmuter. Thus, this single relatively simple operation accomplishes the removal of the fertile uranium, the formation of an inexpensive eutectic salt, and the fluorination of the PMA.

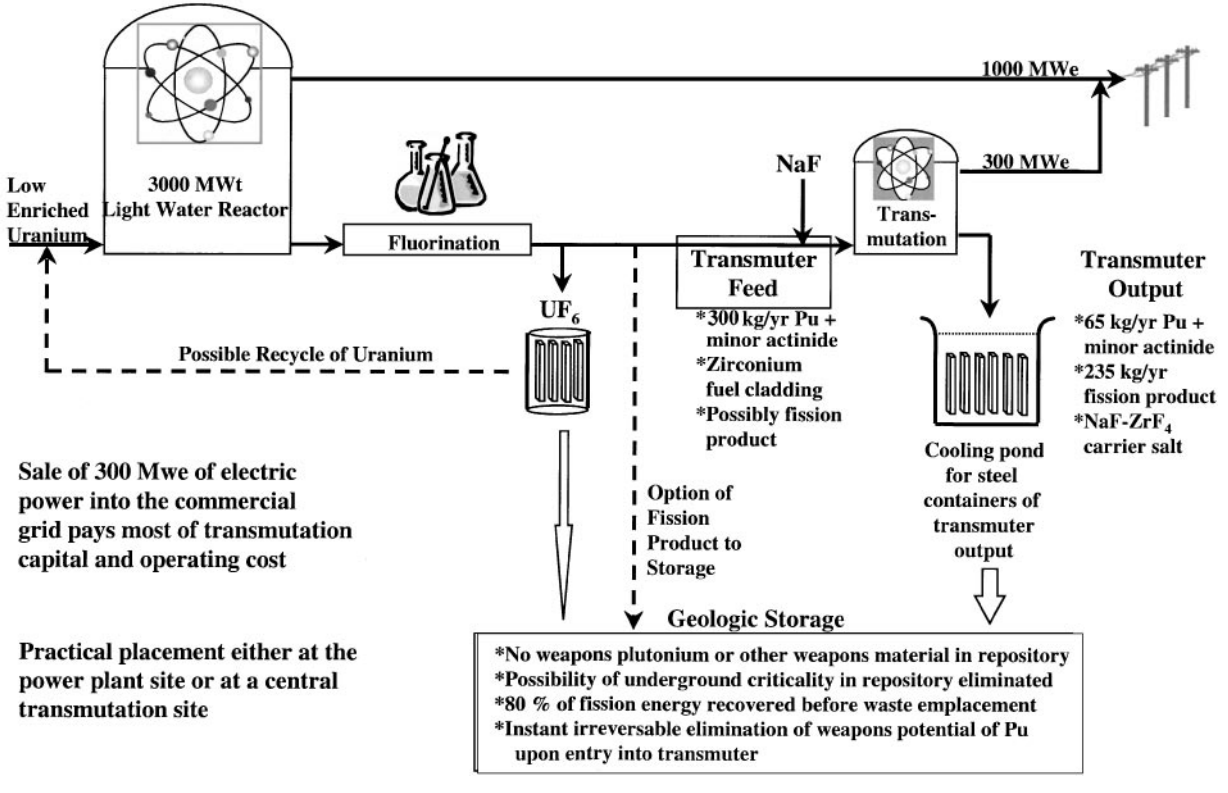
The performance of the transmuter is shown in Figure 7, which compares the input (13) and output isotopic compositions of the actinides. Only about 20% of the LWR actinide feed remains in the exit stream, so nearly all of the fission energy from the plutonium and minor actinide is recovered. Clearly, the exit isotopic composition is no longer dominated by the fissile species, and the weapons value and criticality potential are greatly reduced. If this material is sent to geologic storage, the actinide load is reduced overall by a factor of almost 5, the plutonium content is reduced by 7, and the neptunium is reduced by 10. The neptunium reduction is significant because it is the most mobile of the actinides in a geologic repository and because it is the only isotopically pure weapons-useful material in LWR spent fuel (14).

The system evaluated (Figure 3) consists of a graphite moderator penetrated with channels containing the molten salt that occupy 5% of the graphite volume. The NaF-ZrF_4 molten salt carries the fluorinated actinides and fission products through the graphite and the heat exchanger. Since the salt-to-graphite ratio is 1/20, it is a well-thermalized assembly. The average effective flux in the system is about 2×10^{14} n/cm^2-s.

The deployment of this technology is proposed in 750-MWt units, which are sized for placement on an existing LWR site and are capable of processing the waste from the typical 3000-MWt LWR as fast as it is produced. With $k_{\text{eff}} = 0.96$ and design allowing half of the accelerator-produced neutrons to start a fission chain, the ADS could be driven by a 12-mA 1-GeV accelerator, which probably can be built with modest extensions of existing technology. The beam-power capacity of the US accelerator proposed for tritium production is about 10 times larger.

By reducing the need for front-end separation and eliminating back-end reprocessing, eliminating fuel fabrication and refabrication, providing a greater thermal-to-electric efficiency than an LWR, and featuring a modest accelerator compared with other ATW systems, the thermal- spectrum Tier 1 ADS greatly enhances the prospect for economic viability of transmutation. The processed waste could be poured into steel containers as it is removed from the transmuter and could be stored in the LWR cooling pond before eventually being removed for geologic storage.

The usual process proposed for transmutation is a closed cycle in which the fission product is removed and the actinides returned for total burn-down.



Back-end separations usually involve the isolation of several fissile species if aqueous technology is used, although in the proposed nonaqueous separations the fission product is removed without isolation of the fissile species. Nevertheless, even for the nonaqueous technology there is concern about diversion of weapons-useful material in this back-end separations process. For the once-through process proposed here, there is no back-end separation.

In summary, the Tier 1 ADS allows the extraction of most of the useful energy from the actinides, reduces the amount of actinide going into geologic storage by a factor of five, eliminates concerns for accidental or induced criticality of the actinide waste stream, and makes the actinides unattractive as weapons material. It accomplishes these things by modest extensions of three well-studied technologies: the linac or cyclotron accelerator, fluorination, and the molten-salt reactor. It is expected that most of the cost of the transmutation would be paid for by sale of electric power into the commercial grid. The system design would allow placement on the site of existing reactors or as clustered facilities at government reservations such as the Savannah River Site in the United States.

TIER 1 DEPLOYMENT: NUCLEAR ENERGY GROWTH OPTION The impact of the deployment of the Tier 1 ADS on the US inventory of plutonium and minor actinide is shown in Figure 8, which presents the arrest in the growth of these materials and the reduction to a minimum equilibrium quantity. The figure presumes an indefinitely long deployment of 100 3000-MWt LWRs made possible partly by Tier 1 ADS deployment. Without transmutation, the inventory of plutonium and minor-actinide waste would grow to about 1800 tons from these LWRs by the year 2050. For simplicity it is assumed that the LWRs are deployed at the rate of 10 per year for 10 years. It is assumed also that the Tier 1 ADS technology would be ready for deployment by 2015. If one 750-MWt ADS were deployed for each LWR, it would only stop the growth in plutonium

Figure 6 Implementation of the once-through transmuter. Spent fuel assemblies from a light-water reactor are first converted to fluorides to remove the uranium as UF_6 and to prepare the rest of the waste for insertion into the transmuter as fluoride salt. The primary constituent of the input to the transmuter from the spent fuel is the cladding as ZrF_4. NaF is added to the mixture and the NaF-ZrF_4 becomes a carrier for the actinides and the fission products. There is no front-end separation producing a pure stream of plutonium. The waste flows continuously into the transmuter and out, spending about five years inside an effective flux of 2×10^{14} n/cm^2-s. Most of the actinide is burned away, and the remnant isotopic composition is transformed to material that is uninteresting as weapons material and incapable of supporting a thermal-spectrum chain reaction. The waste could be sent to a repository after cooling or it could be sent through a similar Tier 2 system (Figure 3) for complete burn-up of actinide and long-lived fission product.

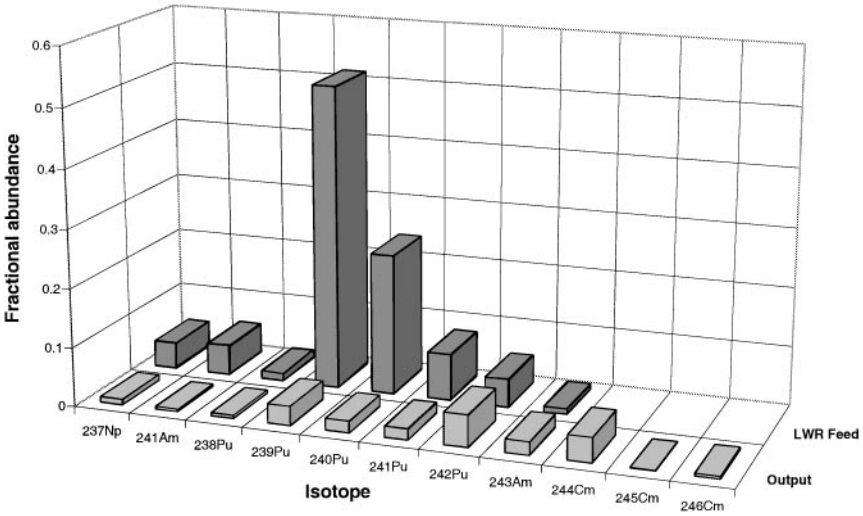


Figure 7 Transmuter actinide burn-up performance. *Back bars* show the isotopic abundance of the spent-fuel plutonium and minor actinide, which is the feed for the transmuter. *Front bars* show the major reduction in total actinide and the isotopic composition in the exit stream. Since the exit-stream isotopic composition and the equilibrium composition are the same, the input isotopic composition upon entry into the transmuters is immediately and irrevocably transformed to that shown as the *front bars*.

and minor-actinide waste or weapons-useful material with a national inventory of about 1000 tons. To eliminate the weapons potential from this material would require a system twice as large, using a 1500-MWt version of the Tier 1 technology (or two 750-MWt systems per LWR), as shown in Figure 8. The curves assume the deployment of the Tier 1 ADS at the rate of 10 per year between 2015 and 2025. By the year 2050, the weapons material is brought under control. The reduction of the weapons material is therefore achieved in about one human generation. The only weapons material left is the neptunium remaining in the transmuters and in the waste remnant.

The deployment of these transmutation systems would increase US nuclear power production by a factor of 1.5 to about 30% of total consumed electric power during the 35-year burn-down period. After that point the US ADS deployment, if it were maintained, would be twice as large as necessary. The ADS fleet could be maintained to accommodate the waste from an increase to 200 LWRs in the US fleet. In the event of such an increase, the nuclear-power market share in the United States would grow from about 20% to about 50%,

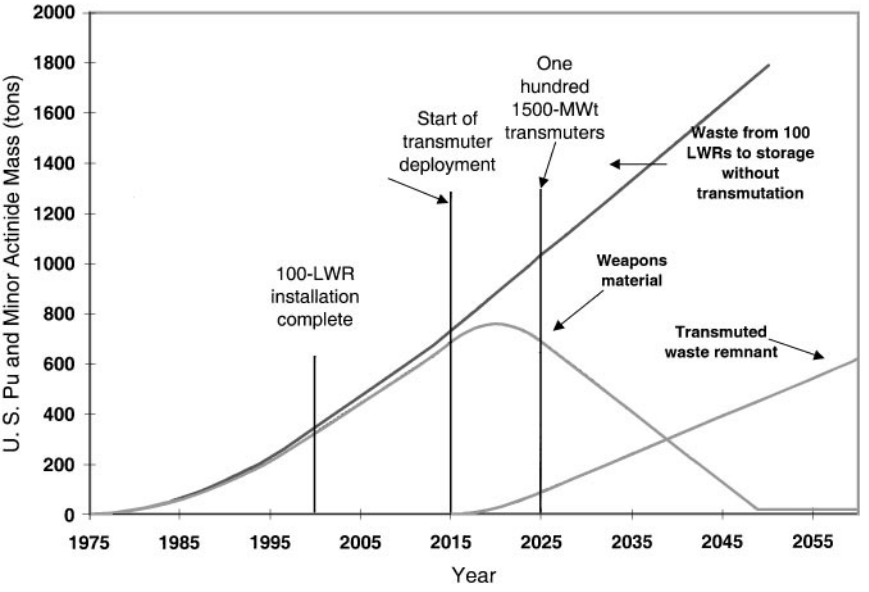


Figure 8 Tier-1 deployment; growth option. *Top* curve: Growth in plutonium and minor actinide waste without transmutation for the present US LWR fleet, continuing indefinitely. For simplicity it is assumed that the 100 LWRs were deployed at the rate of 5 per year for 20 years ending in 1995. If 100 LWRs continue in the United States, 100 Tier-1 ADS systems at 1500 MWt each will be required to reduce the weapons material (as shown) by a factor of about 100. It is assumed that these units would be deployed beginning in 2015 at the rate of 10 per year. All of the waste inventory would be transmuted by 2050. The actinide transmuted remnant, about one fifth of the LWR waste actinide, could be stored in a geologic repository or be reduced by an additional factor of 200 in the Tier 2 system.

assuming no growth in US demand for electricity. However, if half of the ADS systems were not replaced, the nuclear share would drop from the 30% during the waste-reduction phase to about 25% for the long term.

US TIER 1 DEPLOYMENT: NUCLEAR ENERGY CLOSE-OUT OPTION Figure 9 presents the nuclear close-out option for nuclear power in the United States with the Tier 1 technology. The deployment of 100 750-MWt transmuters between 2015 and 2025 allows about 16 years for development and demonstration of the transmutation technology. The upper curve in Figure 9 shows the accumulation of nuclear waste without transmutation, assuming that the LWR lifetime is 40 years. The curve immediately below it shows the build-up of weapons-useful material, which is mostly plutonium but also includes the

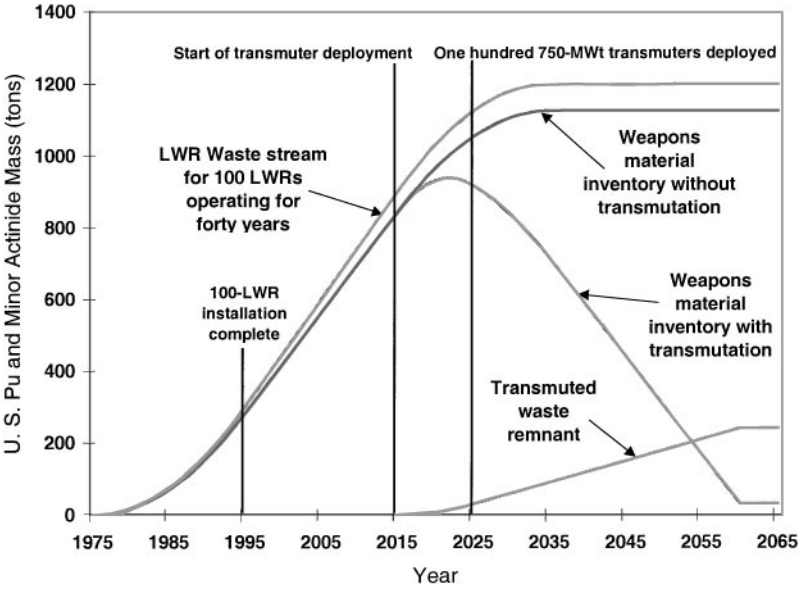


Figure 9 The nuclear power close-out option. *Top curve*: Generation of waste from 100 LWRs assumed for simplicity to have been deployed at the rate of 5 per year from 1975 until 1995, which are operable for 40 years. *Adjacent to top curve*: Weapons-useful material in the waste (plutonium and neptunium) without transmutation. *Two vertical lines* indicate the 10-year bracket for installation of 100 Tier-1 750-MWt transmuters. *Lowest curve*: Transmuted remnant waste, which is free of weapons material except for a small amount of neptunium, which could be sent to geologic storage or sent to a Tier-2 transmuter. *Curve above lowest*: Time dependence for weapons-material destruction. The weapons material is reduced by a factor of about 50.

neptunium. The next curve shows the time dependence for the reduction of the weapons material, which is reduced by a factor of about 50 in about 40 years. The lowest curve shows the accumulation of transmuted nuclear waste, which is free of weapons material and is reduced by a factor of about 5 below that of the untransmuted waste stream. This material could be sent either to geologic storage or to the Tier-2 transmuters, which would reduce the actinide content by an additional factor of 200 for a total actinide reduction factor of 1000.

US TIER 1 DEPLOYMENT: NO-GROWTH OPTION Transmuters would be deployed as the first LWRs are taken offline in about 2015, at the end of their 40-year lifetime. Four 750-MWt transmuters would be deployed for each LWR taken offline until the number of LWRs had dropped to 75 and the number of transmuters had reached 100. The burn-down period would last about 60 years. To reach equilibrium, the number of LWRs would eventually have to increase to

about 80 and the number of transmuters would decrease to about 80. This option is probably the least interesting for the United States, since nuclear power over the long term probably will either achieve a significantly greater share of the US market or be abandoned. The no-growth option is more interesting for countries with a major share of their power already coming from nuclear reactors, such as France and Sweden.

DESIGN PARAMETERS OF THE TIER 1 ADS The Tier 1 ADS would be impractical if the capture cross sections of the fission products fed from the LWR were too high. Simply weighting the capture cross sections of the fission products from LWR spent fuel with their abundance results in a large average fission-product capture cross section, and the burden on the neutron economy is impractically large for the system shown in Figure 3. However, if the fission product lifetime T in a neutron flux ϕ is much less than the residence time R_t, then the large fission-product capture cross section would not be operating all of the time that the nuclide is in the transmuter. The lifetime T is given by $T = 1/\phi\sigma$ where σ is the capture cross section and ϕ is the flux. The well-thermalized effective thermal flux under consideration here is 2×10^{14} n/cm^2-s. For a fission-product cross section of 1000 barns, $T = 5 \times 10^6$ s, or about two months. If the nuclide's residence time is five years, it is clear that the effective capture cross section of that nuclide would be reduced by about a factor of 30 (i.e. 5 years/2 months). The effective cross section associated with this 1000-barn cross section would be $1000/30 = 33$ barns. Therefore, the effective average fission-product capture cross section is substantially less than that of the mixture of fission products fed into the system. The average fission-product capture cross section, taking burn-up (including multiple daughters) into account, is 6.0 barns (5). This value is substantially lower than the common perception, and the resulting more favorable neutron economy is a key element in the design of a practical system of the type shown in Figure 3.

The Tier 1 ADS accepts PMA from the LWR and transmutes it to a different composition during its nominal 1.5×10^8-s stay in the transmuter. The system transmutes the fission products that have higher cross sections to some degree, and the effects of this are taken into account in calculation of the 6-barn average capture cross section. The actinide composition can be calculated using coupled differential equations that take fission and multiple capture into account for the input actinide distribution. The equilibrium composition of the actinides in the transmuter is of course the same as the exit composition.

Table 1 shows the results of this calculation for an effective flux of 2×10^{14} n/cm^2-s and a fluence of 3×10^{22} n/cm^2. The second column shows the normalized feed fraction. The fourth row for ^{239}Pu shows its feed fraction, which is 0.5152, followed by the equilibrium distribution for the ^{239}Pu that was not

Table 1 The transmutation of the feed to the equilibrium distribution[a]

Isotope	Fraction	^{237}Np	^{241}Am	^{238}Pu	^{239}P	^{240}Pu	^{241}Pu	^{242}Pu	^{243}Am	^{244}Cm	^{245}Cm	^{246}Cm	Total
^{237}Np	0.04	0.0093		0.00342	0.00167	0.0004	0.00031	0.00063	0.0002	0.0000	0.00001	0.00007	0.0164
^{241}Am	0.0515		0.004	0.0046	0.00224	0.00053	0.00041	0.00083	0.00032	0.0006	0.00002	0.00009	0.0136
^{238}Pu	0.014			0.00134	0.00065	0.00016	0.00012	0.00025	0.00009	0.00018	0.00001	0.00003	0.00282
^{239}Pu	0.5152				0.028	0.00659	0.00496	0.0104	0.00399	0.00747	0.00022	0.00108	0.0627
^{240}Pu	0.238					0.0119	0.00926	0.0188	0.00719	0.0135	0.00038	0.0002	0.0614
^{241}Pu	0.0788						0.00323	0.00656	0.00251	0.0047	0.00013	0.00068	0.0179
^{242}Pu	0.0481							0.0161	0.00616	0.0115	0.00033	0.00167	0.036
^{243}Am	0.0093								0.00179	0.00336	0.0001	0.00049	0.0058
Total	1.0	0.0093	0.004	0.00936	0.0325	0.01958	0.01829	0.05357	0.02229	0.04136	0.0011	0.00431	0.216

[a]The feed fraction is shown in the second column. The transmuted remnant in equilibrium is shown on the bottom row. The fraction of the input actinide remaining of 0.216 is shown in the lower right hand corner. The degree of destruction of any fed isotope is given by the ratio of the values in the last column to the values in the second column. For ^{239}Pu, the fraction of the ^{239}Pu fed that was not destroyed is .0627/.5152 = 0.122. The fraction of ^{239}Pu in the remnant waste is different, since ^{239}Pu is produced from lighter isotopes as well as being destroyed; this value is the total at the bottom of the sixth column divided by 0.216 or .0325/.216 = 0.15. The transmutation is for a fluence of 0.03 n/b.

Table 2 Results of Tier 1 transmutation

Isotope	Fraction fed[a]	Degree of Burn-up[b]	Final isotopic ratio normalized[c]	Final Pu ratio normalized
^{237}Np	.0450	0.207	.043	
^{241}Am	.0515	0.0777	.0185	
^{238}Pu	.0140	0.669	.043	.0698
^{239}Pu	.5152	0.063	.150	.243
^{240}Pu	.2380	0.0825	.0906	.146
^{241}Pu	.0788	0.232	.0847	.137
^{242}Pu	.0481	1.11	.2481	.402
				1.0
^{243}Am	.0093	2.40	.1032	
	1.0			
^{244}Cm			.1917	
^{245}Cm			.0056	
^{246}Cm			.0200	
			1.0	

[a]Isotopic feed normalized to unity.
[b]Degree of burnup = ratio of exit-stream feed concentration to inlet-stream feed concentration. This column = fraction of the isotope remaining from the feed taking into account the isotopic destruction and production. For ^{237}Np 20.7% of the feed of Np comes out of the transmuter; for ^{242}Pu more comes out than was sent in.
[c]Normalized isotopic abundance in the transmuter exit stream.

destroyed by fission but was transmuted to other nuclides. The sum of these amounts, given in the last column, is 0.0627. Therefore the fraction of the fed ^{239}Pu that was not destroyed by fission is $0.0627/0.5152 = 0.1217$. However, ^{239}Pu was also produced by neutron capture on other isotopes. The total, at the bottom of the sixth column, is 0.0325. Therefore the ratio of exit stream to inlet stream ^{239}Pu is $0.0325/.5152 = 0.063$. It is useful to note that the transformation from inlet to outlet distribution is immediate, since the outlet and the internal distribution in the transmuter are the same. Table 2 summarizes in several ways the results of the transmutation results in Table 1. Table 3 gives the equilibrium composition of the salt, and Table 4 shows the Tier 1 system parameters.

BEAM CURRENT REQUIREMENT The beam current requirement can be calculated starting with the fission power P of the system, which may be converted to the number of fissions per second. The average chain is $k/(1-k)$ fission events long (15); dividing the number of fissions by this factor gives the number of fission chains in the system, which must be started by the accelerator-produced neutrons. For a critical reactor with $k = 1$, the fraction of the fission neutrons that induce fission is $1/\nu_{\text{ave}}$, where ν_{ave} is the number of neutrons per fission averaged over the equilibrium composition of the fissioning species in the system.

Table 3 Equilibrium composition. The Tier-1 equilibrium composition of the salt and the total mass inside and outside of the flux is given for the carrier, the actinide and the fission product

Element	Mole Fraction	Mass (kg)	Element	Mole Fraction	Mass (kg)
ZrF_4	.4825	11,338	Pd	.00092	13.6
NaF	.4825	2,931	Ag	.00005	0.7
Np	.00052	17.3	Cd	.00007	1.1
Am	.00147	48.9	Sn	.00002	0.3
Pu	.00720	240.5	I	.00013	2.3
Cm	.00262	87.3	Xe	.00280	—
Se	.00005	0.5	Te	.00026	4.7
Br	.00002	0.2	Cs	.00124	22.8
Kr	.00028	—	Ba	.00089	16.9
Rb	.00029	3.5	La	.00062	11.9
Sr	.00060	7.2	Ce	.00125	22.3
Yt	.00035	4.3	Pr	.00051	9.9
Zr	.00060	33.6	Nd	.00197	39.3
Mo	.00236	30.2	Sm	.00042	8.7
Tc	.00054	7.4			
Ru	.00148	20.8			
Rh	.00031	4.5			

For a subcritical system, where the chains are started by accelerator neutrons, the fraction of the accelerator neutrons that induce a chain is therefore k/ν_{ave}. If each proton produces n neutrons, the current requirement is then given by

$$\begin{aligned} \mathrm{I\,(mA)} &= 5.2[(1-k)/k][\nu_{ave}/k][1/n]\mathrm{P(MWt)} \\ &= 5.2(1-k)[\nu_{ave}/nk^2]\mathrm{P(MWt)}. \end{aligned}$$

Therefore, for a power of 750 MWt, $k = 0.96$, $\nu_{ave} = 2.96$, $n = 30$ for 1-GeV protons on lead, $I = 16.7$ mA.

One can build the once-through Tier 1 system as a two-circuit system in which the LWR waste is fed near the center and is kept there for a period of time before it is allowed to mix with the rest of the salt. In this situation, the accelerator neutrons interact first with the more reactive actinide feed, so they are treated differently from the fission neutrons. The factor k/ν_{ave} (= 0.324 in the above case) must be replaced by the factor $\langle\sigma_f\rangle/(\langle\sigma_f\rangle + \langle\sigma_c\rangle) = 0.457$ for the Tier 1 system. Therefore the current required is $16.7 \times 0.324/0.457 = 11.7$ mA.

If the objective becomes to use one Tier 1 system on the site of one 3000-MWt LWR to destroy the waste from the one LWR, the Tier 1 unit would destroy 80% of the waste instead of 100% and the Tier 1 power would be 600 MWt instead

Table 4 ADS Tier 1 system parameters

Gross thermal power	750 MWt
Carrier salt	NaF:ZrF_4
Criticality factor k_{eff}	0.96
Thermal to electric conversion efficiency	0.42
Electric power	315 MWe
Beam energy	1.0 GeV
Beam current (one salt circuit)	16.7 mA
Beam current (two salt circuits)	13.4 mA
Accelerator efficiency	0.45
Power to grid (one salt circuit)	270 MWe
Power sales @ 35 mils/kwh; 80% uptime	$66.0 million/year
Debt retirement $500 million at 7% for 30 years	$40.3 million/year
Annual actinide burn rate	300 kg/year
Salt flow-through rate	2.2 liters/day
Front-end separations	Uranium removal only
Back-end separations	None
Location	Reactor site or central site
Vessel	
Diameter	425 cm
Height	500 cm
Thickness	10 cm
Weight	80 tons
Material	Steel
Liner	Hastalloy N modified
Liner thickness	2.5 cm
Liner mass	20 tons
Target	
Medium	Liquid lead
Window	None
Lead target diameter	60 cm
Lead target height	150 cm
Beam diameter	20 cm
Graphite outer diameter	100 cm
Container material	Steel
Cooling	External circuit
Internal piping	Steel in graphite
Weight of lead in target	5 tons
Weight of lead in target and circuit	10 tons

(*Continued*)

Table 4 (*Continued*)

Blanket	
Graphite diameter	425 cm
Flux diameter	375 cm
Flux height	375
Flux volume	38.5 m^3
Average flux	2×10^{14} n/cm^2-s
Fluence	3×10^{22} n/cm^2
Salt volume fraction	5%
Salt volume in flux	1.925 m^3
Average power density	19.5 watts/cm^3
Salt power density	390 watts/cm^3
Number of salt channels	122
Salt channel diameter	7 cm
Sleeve outer diameter	20 cm
Sleeve lifetime	3.3 years
Graphite matrix lifetime	100 years
Salt volume outside flux	1.925 m^3
Salt density	3.5
Total NaF mass	9.9 tons
Total ZrF_4 mass	16.9 tons
Graphite mass	110 tons
Number of internal heat exchangers	8
Salt velocity through graphite	4 m/s
Exit salt temperature	700 C
Entrance salt temperature	600 C

of 750 MWt. In that case the currents for the single-circuit and double-circuit systems are 13.4 and 9.4 mA, respectively. If it became practical to run the Tier 1 system at $k = 0.98$, as is assumed for the CERN system, then these single- and double-circuit systems at 600 MWt would require beam currents of 6.4 and 4.5 mA, respectively. Estimated capital costs for components are shown in Table 5.

Table 5 Estimated capital costs for Tier 1 components (in millions of dollars) with $k_{\text{eff}} = 0.96$

Accelerator	150
Fluorination	25
Target-blanket	150
Power production equipment	100
Balance-of-plant	75
Total	500

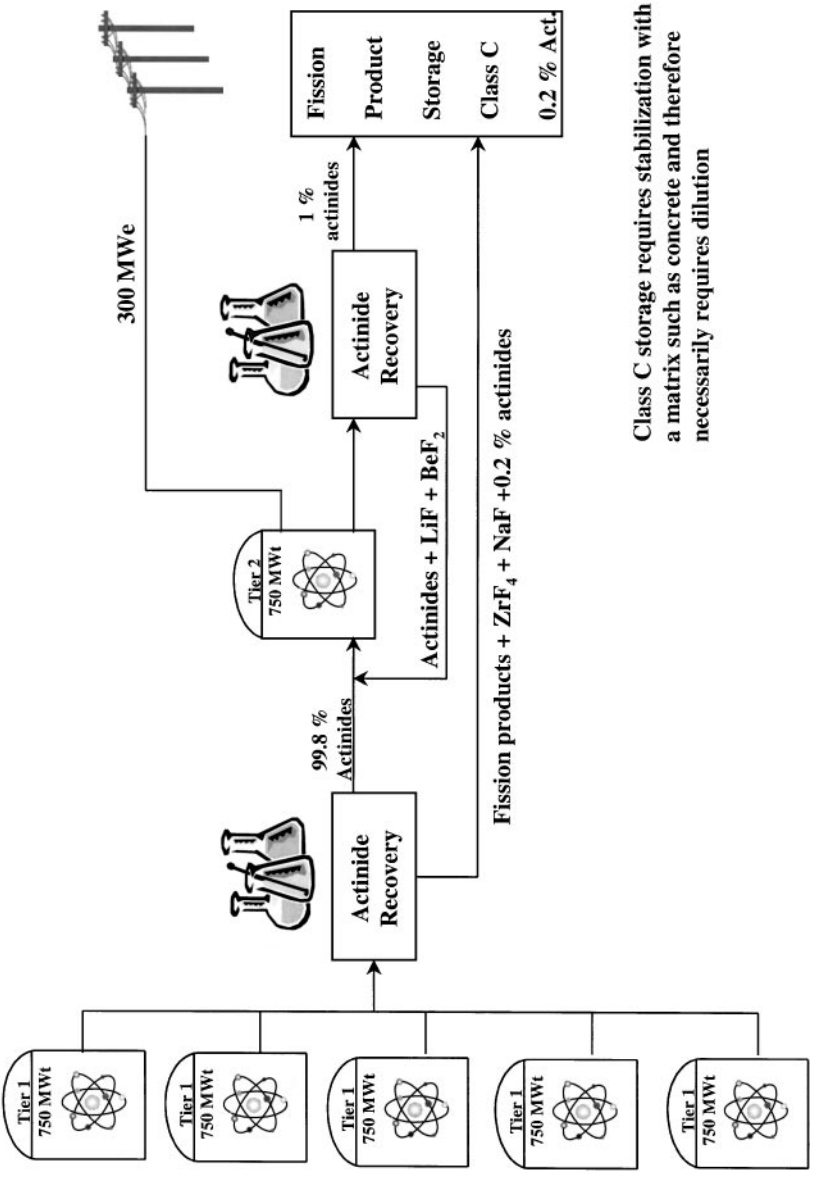
It should be noted that the costs in the table are not for a one-of-a-kind system but anticipate cost reductions from many identical system deployments per year. The annual expenditure for debt retirement of $500 million at 7% for 30 years would be $40 million per year. The receipts for power sales at 35 mils/kwh with 80% up-time would be $66 million per year, leaving $26 million per year for operating costs. Some fee should also be due the transmuter for having denatured the waste. Improved capital cost estimates would be available after detailed engineering design. However, the main factor in system economics is the price at which the power can be sold, and predicting that price 15 years in advance is highly uncertain. Nevertheless, the Tier 1 system will almost certainly have a clear cost advantage over waste-destruction systems requiring back-end separations and over transmutation concepts with solid fuel requiring fuel storage, destruction, separations, and refabrication.

The Tier 2 Accelerator-Driven System

The Tier 1 system enhances the viability of geologic storage by recovering most of the nuclear energy from PMA; by greatly reducing the possibility of explosive criticality in geologic storage from rearrangements of the fuel by intentional, accidental, or natural processes; and by destroying the plutonium and neptunium as weapons-useful material. The remnant of the waste still requires geologic storage. However, it is also possible to use nearly the same target-blanket design to destroy this remnant actinide using the Tier-2 system illustrated schematically in Figure 10.

The actinide from five Tier 1 ADS units is sent to a single Tier 2 ADS unit. The fission product and carrier salts of ZrF_4:NaF are removed on the front end. The mixture is fissioned at $k = 0.95$ and the remaining actinide destroyed by fission. Back-end removal of fission product is required for total burn-up of the actinide. The system would operate at a power level of 750 MWt and would send 270 MWe to the commercial power grid. If the back-end separations allow a 1% loss of actinide into the fission-product waste stream, the actual fraction of the LWR waste generated is only 0.2%, since five LWRs are being supported. However, to match this performance, the front-end separations must be performed with a 0.2% leak-through of actinides with the fission products. The fission products technetium and iodine also could be removed and burned in this system.

If both long-lived fission products and actinides are destroyed, the remnant fission product still will be strongly radioactive owing to the ^{137}Cs and ^{90}Sr isotopes with decay half-lives of about 30 years. However, after 300 years of interim storage, these nuclides would decay by a factor of 1000 and the remnant waste would reach Class C waste criteria. Geologic confinement is not required for the 300-year interim storage period, since canisters can be designed that



will easily outlive the radioactivity of the waste. Geologic confinement is not necessary for Class C waste either. Therefore, one major benefit of the Tier 2 system is the elimination of reliance on geologic storage. Since many sites in each state could meet Class C requirements, waste need not be sent to a single geologic storage site but could be stored near its production site. With the ADS Tier 1 and Tier 2 deployments, the waste storage would be transformed from a federal problem to a state problem, with each state handling its own waste. If the waste were reduced by a factor of 500 using transmutation and the 50 states shared equally in waste allocation, each state would be responsible for only 4×10^{-5} of the waste that would go to a single geologic storage facility such as Yucca Mountain. This great reduction in waste quantity should ameliorate problems of waste siting in the states. On the other hand, a reduction in actinide by 500 and the virtual elimination of underground criticality concerns and the weapons potential of the waste might be sufficient to persuade one community to accept all the nation's waste. The results of the calculation of the Tier 2 equilibrium isotopic distribution, when fed the distribution from the Tier 1 ADS, are shown in Table 6.

All numbers in the table have been multiplied by a factor of 1000. Therefore the sum of 216 is the total of the transmuted output of the Tier 1 system, 0.216. The equilibrium isotopic composition is shown in the bottom row and the sum is $311/1000 = 0.311$. Therefore the concentration of actinide in the feed salt is greater than that in the equilibrium mixture. This is because the actinide only disappears from the Tier 2 ADS by fission, whereas it is removed from the Tier 1 ADS by both fission and continuous drainage. The fission quantity $\Sigma_i N_i \sigma_{fi} = 6595$ and the average fission cross section is $\langle \sigma_f \rangle = \Sigma_i N_i \sigma_{fi} / \Sigma_i N_i = 21.2$ barns. The corresponding quantity for capture $\Sigma_i N_i \sigma_{ci} = 15{,}430$ gives $\langle \sigma_c \rangle = 49.6$ barns. Therefore the actinide capture is substantially more of a neutron-economy problem than in the Tier 1 system. This is partially compensated by the larger neutron multiplicity ν of 3.42 per fission. (This large value arises

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Figure 10 Deployment of Tier-2 system. If it is decided to destroy the remnant waste from the Tier 1 systems instead of placing it in geologic storage, the remnant waste from five Tier 1 systems would be transported to a central site. Front-end chemistry would remove fission products and NaF:ZrF_4 from the waste. The actinides would be sent to a 750-MWt transmuter, which would destroy the remainder of the actinide. Back-end separations would be required to remove the fission products and return the actinide to the transmuter. Because the Tier 1 system requires less front-end and no back-end separations, the Tier 2 cost is higher. However, since the Tier 2 system handles the waste from five Tier 1 systems, these additional separations costs are spread out over the output from five Tier 1 units and therefore have only incremental influence on the total transmutation costs. The single 750-MWt Tier-2 transmuter shown handles the remnant waste from about 6.25 LWRs.

Table 6 Calculation of equilibrium isotopic distribution for Tier 2 transmutation of Tier 1 mixture. The second column shows the isotopic feed distribution from the Tier 1 system with abundances multiplied by 1000. The Tier 2 transmutation results are shown in the bottom row again multiplied by 1000

Nucl.	Abund	^{237}Np	^{241}Am	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{243}Am	^{244}Cm	^{245}Cm	^{246}Cm	^{247}Cm	^{248}Cm
^{237}Np	9.3	2.42		0.986	0.512	0.127	0.103	0.316	0.151	0.446	0.0148	0.405	0.0203	0.075
^{241}Am	4.0		3.39	0.424	0.220	0.0548	0.0444	0.136	0.0648	0.218	0.0064	0.174	0.0087	0.0332
^{238}Pu	5.22			0.553	0.287	0.0715	0.0579	0.177	0.084	0.286	0.0083	0.240	0.114	0.0433
^{239}Pu	32.5				1.87	0.465	0.377	1.15	0.546	1.85	0.0536	1.47	0.0738	0.273
^{240}Pu	19.6					1.02	0.839	0.257	0.122	3.92	0.120	3.30	0.164	0.608
^{241}Pu	18.3						0.781	2.40	1.13	3.84	0.112	3.07	0.153	0.567
^{242}Pu	53.6							27.1	12.8	43.4	1.25	34.6	1.72	6.38
^{243}Am	22.3								5.33	18.1	0.526	14.5	0.723	2.68
^{244}Cm	41.4									33.2	0.977	26.8	1.34	4.97
^{245}Cm	1.2										0.0287	0.78	0.039	0.146
^{246}Cm	4.3											19.1	0.954	3.53
^{247}Cm	0.2												.0095	0.18
^{248}Cm	0.4													0.86
Totals	216	2.42	3.39	1.96	2.86	1.74	2.20	31.5	20.2	105.3	3.09	111.4	5.22	20.3

from the values of ν for ^{245}Cm and ^{247}Cm of 3.72 and 3.79 neutrons per fission, respectively.) However, the neutron economy of the Tier 2 system is tighter, and fewer neutrons can be lost in the carrier salt and in the fission product. For these reasons the Tier 2 systems operate with a LiF:BeF_2 salt, and fission product must be removed both at the front end and the back end.

THE CERN FAST-SPECTRUM SYSTEM PERFORMANCE

A CERN-based team (16) proposed an ADS called an Energy Amplifier for production of nuclear energy from thorium with a minimal waste stream. The team proposed a version of this system for the destruction of LWR waste and analyzed the performance of the system for the case of Spain. This system (Figure 11) is chosen for examination and comparison here because it appears to be the most fully developed fast-spectrum concept at this time. It consists of metallic fuel in an annular subcritical core surrounding an accelerator source of neutrons. The core is immersed in a liquid-lead coolant, which also serves as the neutron-production target. A long column of lead above the core enhances heat removal by thermal convection. This column also allows direct transfer of decay heat to air in case of a loss-of-coolant accident. The beam pipe in the lead transports the 20-mA 1-GeV proton beam to a window located near the middle of the core. Spallation neutrons produced in the lead drive the core, which is maintained at a nominal $k = 0.97$ at a thermal power of 1500 MW. This heat is converted to electricity and sold into the commercial grid to help pay the capital and operating costs of the facility. More specifications are given in Table 7, taken from the CERN reports (7). The TRU mentioned in the table is the plutonium and minor actinide produced in the LWR.

The LWR spent-fuel assemblies undergo decladding, and PMA are separated by nonaqueous means from the zirconium fuel cladding, the uranium, and the fission products. The PMA, along with thorium, form metallic fuel. This fuel is burned for two years; then the spent transmuter fuel is processed to remove fission product and is reconstituted, with additional plutonium and minor actinide to replace that fissioned. Therefore the system requires front-end processing and repeated back-end processing for all of the waste destroyed.

Reactivity Stability

As the fissile material in a reactor or solid-fueled ADS burns away, the reactivity decreases. In power- producing reactors, this effect is partly compensated by the production of ^{239}Pu as the ^{235}U in the initial fuel loading burns away. Pulling the control rods out as the burn-up progresses also compensates the reactivity

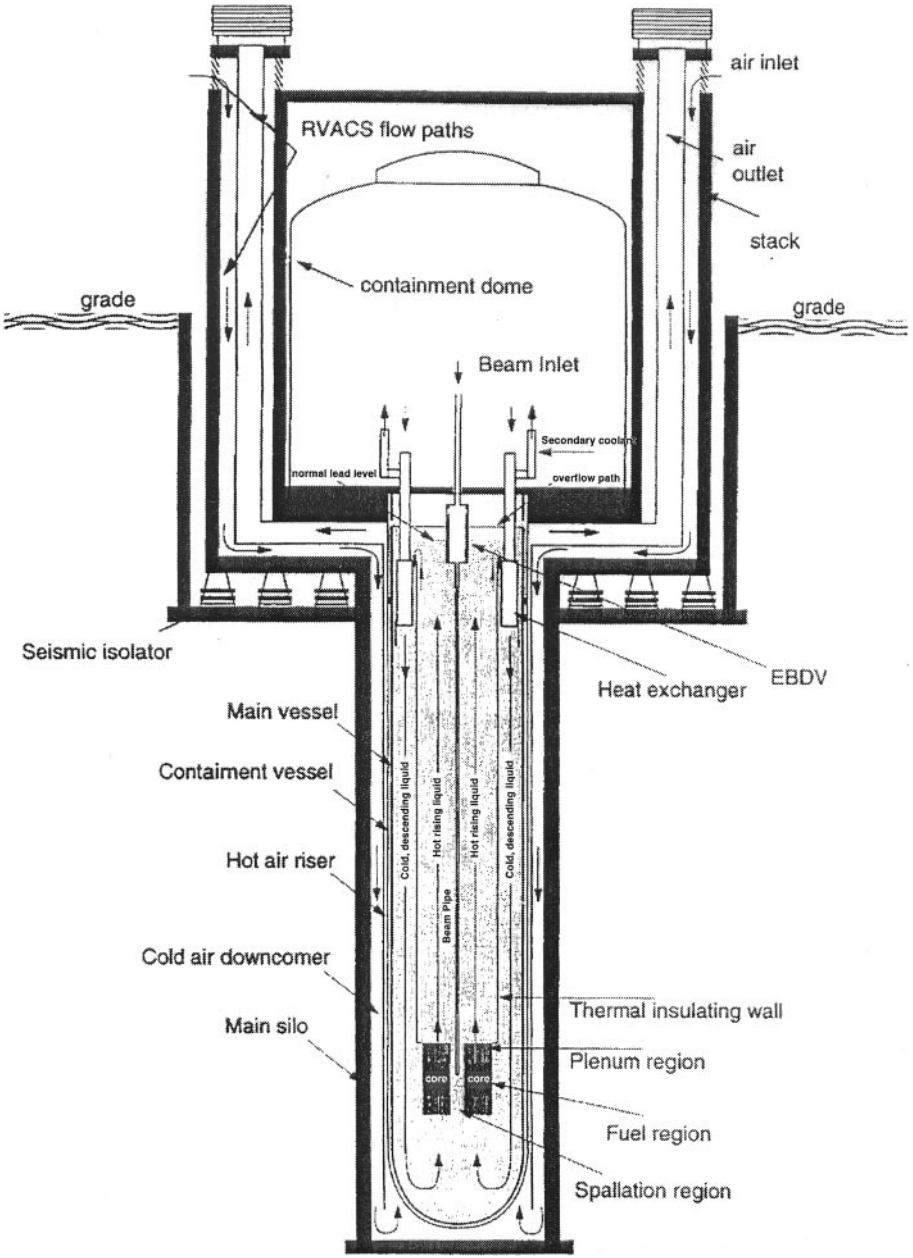

Figure 11 Energy Amplifier accelerator-driven system proposed by CERN (15).

Table 7 Main parameters of the Energy Amplifier operated as a waste incinerator

Gross thermal power per unit	1500 MW
Coolant	Liq. lead
Subcriticality factor k, (nominal)	0.97
Scram systems (CB_4 rods)	3
Main Vessel	
Gross height	30 m
Diameter	6 m
Material	Steel
Walls thickness	7.0 cm
Weight (excluding cover plug)	515 tons
Fuel Core (double structure, inner, outer core)	
Fuel mixture (asympt.)	^{232}Th + 30% TRU*
Fuel mass (asympt.)	9.2 tons
Number of bundles inner core	30
Number of bundles outer core	90
Specific power	160 W/g
Core power density, average	176 W/cm^3
Core power density, maximum	305 W/cm^3
Fuel Pins	
Outer diameter	8.2 mm
Cladding thickness	0.35 mm
Cladding material	HT-9
Active length	150 cm
Fuel-cladding gap thickness	0.1 mm
Inner void diameter	6.26 mm
Sub Assemblies (Bundles)	
Configuration	Hexagonal
No. hex rounds of pins (inner, outer core)	10, 11
No. pins in each bundle (inner, outer core)	331, 397
Flat to flat	234 mm
Pitch between pins (inner, outer core)	12.45, 11.38 mm
No. units	120

*TRU refers to Pu and minor actinides as used in this report.

reduction by several percent. In an ADS devoted purely to waste transmutation, there is no fertile ^{232}Th or ^{238}U, nor are control rods necessary in view of the typical operation at $k_{\text{eff}} = 0.96$. Reactivity stability is therefore a problem for a solid-fueled ADS. One way around this is to use a burnable poison in the solid fuel. As the fissile material is burned away, the poison is burned away at about the same rate, and the reactivity can be made fairly constant. However, this is a major blow to the neutron economy, since for every waste nuclide destroyed about one poison nucleus must be destroyed. The neutron excess of

1.2 neutrons per waste nuclide (9) therefore becomes 1.2–1 $\sim$ 0.2 and the fast reactor neutron economy becomes inferior to the thermal-spectrum value of 0.54 (9). Therefore the use of burnable poison is a questionable approach to maintaining reactivity stability.

The CERN system deals with this problem by including ^{232}Th in the solid fuel. Capture of a neutron on ^{232}Th produces the fissile material ^{233}U. As the waste burns out, the ^{233}U builds up and the reactivity is held more nearly constant. The analysis shows that k_{eff} varies between about 0.98 and 0.96 over one fuel cycle. The accelerator beam requirement is 18 mA at 1 GeV, but the current must vary from 60% to 130% of 18 mA. Variation of the accelerator current to keep the power constant is undesirable because the capital investment in the accelerator is not fully used at all times. In addition, the accelerator current must be changed up and down throughout the cycle, which adds complexity to the operational and safety issues.

The isotopic distribution in the waste is mostly stabilized after about five two-year fuel-burning cycles. If the fuel spends twice as much time outside of the flux as inside, then the time for reaching stability is about $5 \times 2 \times 3 =$ 30 years, which is most of the life of the transmuter. This may be a serious consideration if the objective is to reduce waste-actinide inventories significantly over a comparable period.

The asymptotic yields are summarized in Table 8, taken from the CERN report (16). The system burns ^{233}U but does not burn all that it produces. Table 8 shows a thorium feed rate of 0.74 tons for a two-year burn cycle. The amount of ^{233}U removed after the cycle is about 0.35 tons. The difference of about 0.39 tons, corresponds to the amount of ^{233}U produced and burned in two years. Therefore the ^{233}U extraction and ^{233}U burn rates are about equal at 0.175 tons per year. The ^{233}U produced would be separated, mixed with uranium separated from LWR spent fuel, and sold as new LWR fuel at an estimated value of $35 million per year to help offset the costs of waste destruction.

The ^{233}U is 94% enriched pure element and therefore it is weapons material. The plutonium also is weapons-useful material. Facilities exist in several countries to separate and burn this as mixed-oxide (MOX) fuel in LWRs. Transmutation in order to destroy commercial plutonium is a major justification for building and deploying the ADS. The amount of actinide waste material destroyed per year in the CERN system is about 400 kg (see Table 8) and the amount of ^{233}U extracted is almost half of that (175 kg), so the net actinide waste reduction is $400-175=225$ kg/year. Production of the ^{233}U therefore almost doubles the size of the facility necessary to destroy commercial waste actinide. Introducing ^{233}U production in order to operate with solid fuel carries a high price.

Table 8 Actinide consumption and production by the CERN ADS

Overall performance	
TRU* incineration rate	402 kg/year
^{233}U-rich uranium mix production rate	175 kg/year
Refill composition	
Fresh TRU	0.85 ton
Fresh thorium	0.74 ton
Residue of previous (actinides but not U)	
Thorium	5.35 ton
Rest	2.26 ton
Fuel metal total initial mass	9.2 ton
Discharge composition	
Recovered uranium mix	0.35 ton
Fraction of ^{233}U in Uranium mix	0.94 ton
Fission fragments	1.24 ton

*TRU refers to the plutonium and minor actinide from the LWR spent fuel.

Build-Up of Higher Isotopes

It is often claimed that the fast spectrum has a major advantage over the thermal-spectrum transmuter, since all of the isotopes can be fissioned in a fast spectrum to a significant degree. For the thermal spectrum, about half of the nuclides do not undergo fission upon neutron absorption but instead capture to the next heavier nucleus. Therefore the thermal spectrum is said to be less effective because it moves the actinide waste to heavier mass. In fact, the CERN report's opening page states that higher actinide cannot be eliminated in a thermal spectrum (7), and this is cited as a critical advantage of the fast spectrum over the thermal spectrum. Table 9 compares the distribution of isotopes for the Tier 2 system and the CERN system.

The thermal-spectrum figures are the Tier 2 inventories for a flux of 4×10^{14} n/cm^2-s in the transmuter, although the effective thermal flux is half as large because the actinide spends half its time in the heat exchangers. The fast flux in the transmuter is 7×10^{15} n/cm^2-s, while the effective flux is taken to be one third of this, since the actinide is estimated to spend twice as much time outside the flux as inside owing to the time required for fuel destruction, separations, and fuel refabrication. These figures, taken from the CERN report, are based on the average of the end-of-cycle-#8 inventories and the mid-cycle inventory of cycle #8 (7). Because for each cycle the waste spends two years inside the flux and an estimated four years outside, the equilibrium inventory is established

Table 9 Equilibrium inventory for thermal and fast spectrum[a]

Isotope	Thermal Flux (4×10^{14}) 2×10^{14}	Fast Flux (7×10^{15}) 2.3×10^{15}
^{237}Np	17.0 kg	118 kg
^{233}U	0	363
^{238}Pu	9.6	243
^{239}Pu	59.4	887
^{240}Pu	35.9	1388
^{241}Pu	33.5	257
^{242}Pu	98.0	410
^{241}Am	7.4	235
^{242}Am	0.17	30
^{243}Am	40.8	123
^{244}Cm	75.8	100
^{245}Cm	2.3	20
^{246}Cm	8.0	6.1
^{247}Cm	0.6	0.53
Sum	387.9 kg	4180.0 kg

[a]The isotopic inventories are given for thermal- and fast-spectrum systems operating at the same fission-power level of 750 MWt. For the thermal flux the effective flux in the transmuter is smaller by a factor of two than the flux in the transmuter, since the actinide spends as much time in the heat exchangers as in the flux. For the fast flux the effective flux is smaller by a factor of three since it is assumed that the actinide spends twice as much time outside of the transmuter in separations, etc as inside.

after about 50 years. The quantities are then multiplied by three because of the time spent outside and divided by two because the system power for comparison is 750 MWt instead of 1500 MWt as in the CERN report.

The build-up of higher actinides claimed for the thermal spectrum is not evidently a serious consequence, as shown in Figure 12, which exhibits the data of Table 9. It is true that the thermal-spectrum inventory contains relatively more higher actinides than the fast-spectrum inventory. However, the absolute amount of these higher actinides is smaller for the thermal spectrum because the overall inventory is much smaller. It is clear that total actinide inventory for the thermal spectrum is smaller by more than an order of magnitude because of the higher cross sections. Therefore, for a national LWR fleet of a given size operating in equilibrium with a national ADS fleet to burn LWR waste, the inventory of PMA that must be carried inside and outside the transmuter is much smaller for the thermal spectrum.

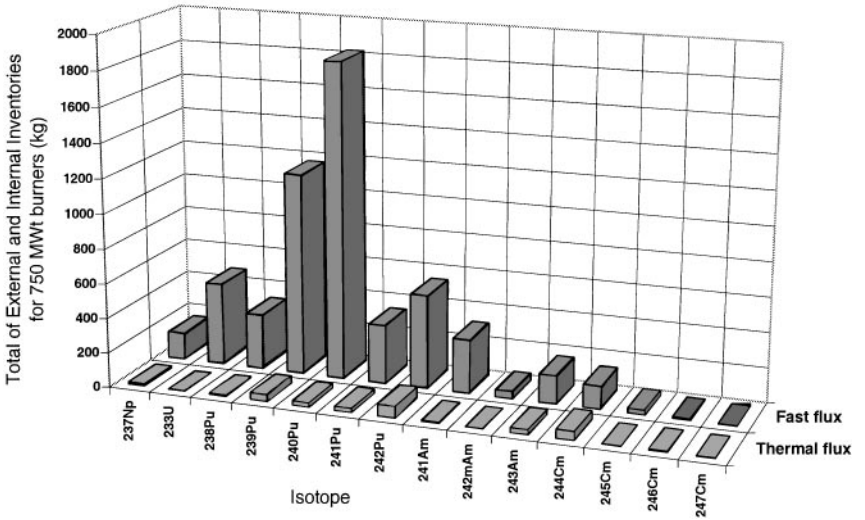

Figure 12 Comparison of the inventory of the CERN system with the Tier 2 ADS. Both are normalized to the same power level of 750 MWt. The build-up of heavier actinides is higher, relatively speaking, for the thermal spectrum than for the fast spectrum. However, this is unimportant in view of the much smaller inventory for the thermal spectrum.

Two isotopes in Table 9 and Figure 12 deserve special attention. The only actinide removed from the system is ^{233}U, which would be used to produce low-enriched fuel for LWRs. However, there will always be delays in getting this material into reactors for burning, as is the case for recycled plutonium, so this material also should be included as part of the total inventory necessary for transmutation. The ^{233}U quantity in Figure 12 and Table 9 associated with each CERN transmuter would be significantly larger.

Also, the quantity of ^{242m}Am is not the same as reported by CERN (7). The reason is that CERN used a thermal-spectrum branching ratio for ^{241}Am capture to estimate the amounts of ^{242}Am and ^{242m}Am (^{242}Am has a 16-hr half-life and is the ground state; ^{242m}Am is the metastable state with a half-life of 150 years). The ^{242}Am quickly decays to ^{242}Pu, but the ^{242m}Am remains, with a fission cross section higher than ^{239}Pu or ^{235}U by almost 10. For this reason, the isotopic mixture of americium in Table 9 is almost as reactive as pure ^{239}Pu. It is important that in destroying the LWR waste, one does not create new material such as ^{242m}Am that might have weapons usefulness (17). The LWR produces virtually no ^{242m}Am because the branching ratio is about 10% to ^{242m}Am and because any ^{242m}Am in an LWR is burned away in a few days. The same is true of a thermal-spectrum ADS. However, in a fast-spectrum ADS the material

is not burned away rapidly, and more of it is produced because the branching ratio to ^{242m}Am is larger by about a factor of three (17) than in a thermal-spectrum system. Therefore the CERN system produces significant amounts of this material, whereas a thermal-spectrum ADS produces virtually none.

Weapons Material Inventory

The amount of actinide in the ADS inventory is of concern because of environmental reasons, because of the complications that might arise in any required separations owing to criticality or decay heat (the thermal-spectrum Tier-1 system requires no back-end separations), and because of the extra expense of carrying a large inventory in the various recycling steps. However, another perhaps more important concern is the reduction of weapons-useful material from LWRs. It is useful to compare the amounts of weapons-useful material for both systems (see Table 9). Both systems yield plutonium with about the same ratio of even-mass to odd-mass isotopes. Because of decay heat and spontaneous-fission neutrons from the even-mass isotopes, neither isotopic composition is attractive for weapons use.

However, with regard to other weapons-useful material, two potential situations are of concern. The first is the recovery of weapons material from repository storage. If all of the weapons material is destroyed, as with the Tier 2 system and the CERN system, recovery from the repository is not a concern. For the Tier 1 system, the neptunium is the only useful weapons material (14) that would enter the repository. According to the US National Academy of Sciences estimates (4) of the amount of actinide required for a nuclear-fission weapon of about 5 kg, three weapons could be made from the 17-kg inventory of neptunium if it could be chemically recovered. This might not be easy, since the neptunium would be present in the salt at the mole fraction of about 1.6 parts per 10,000. Of course, if the waste went directly into the repository without transmutation, as is the current US policy, the amount of LWR waste per year from one LWR could be the source of about 50 plutonium and neptunium weapons.

A second concern regarding nuclear weapons is recovery from the transmuter itself in a nonproliferation-treaty abrogation scenario. In the event of widespread deployment of the transmutation systems, it is possible that a nation, after agreeing to all nonproliferation requirements of the International Atomic Energy Agency (IAEA) or other bodies, could abrogate and recover weapons material from one or more transmuters. It is important to note here that the key issue is whether more or better weapons material could be recovered from the transmuter than from the spent fuel that feeds into the transmuter. From Table 9, it is clear that the plutonium in both thermal- and fast-spectrum

equilibrated transmuters is much less desirable for nuclear weapons than the plutonium fed into them. Therefore, the primary nonproliferation issues of the ADS are the interruption of operation and removal of the inventory of the neptunium and ^{233}U for recovery of weapons material. We have already seen that about three neptunium weapons might be made from the interrupted operation of one thermal-spectrum ADS. However, nearly 100 could be made from the interruption of one fast-spectrum ADS. Taking 30 of the thermal-spectrum ADS units offline to obtain a 100-weapon arsenal would be time-consuming, expensive, and highly disruptive. Raiding one fast-spectrum ADS might be fast and cheap.

Back-end separations for the fast-spectrum solid-fuel ADS also provide the opportunity for separation of weapons material. Although the back-end separation facility built for the fast-spectrum ADS might not have the capability to separate out neptunium, it would naturally have the capability to separate ^{233}U. The Tier 1 thermal-spectrum ADS has no back-end separations system. Although the Tier 2 ADS requires back-end separations capability, it contains no weapons material. Therefore, in the thermal-spectrum ADS, there is no weapons material to extract except the ^{237}Np in the Tier 1 system. However, the neptunium could also be obtained directly from the LWR spent-fuel assemblies, so no new route to neptunium is opened by the thermal-spectrum ADS. In the fast-spectrum solid-fueled ADS, the necessity of back-end separations opens an alternative route to a nuclear arsenal.

This analysis ignores any ^{233}U that might have been accumulated from the fast-spectrum ADS and stored for eventual burning in LWRs. The ^{233}U problem for the fast-spectrum ADS could be eliminated by avoiding the use of solid fuel, which requires the production of ^{233}U for reactivity stability. However the 10-times-larger inventory of ^{237}Np would still be present.

Neutron Economy and the Role of the Accelerator

Because the neutron excess (9) per nucleus of waste actinide destroyed in a fast spectrum is about 1.2 and burnable poison is not used to stabilize k_{eff}, it is useful to examine the disposition of these excess neutrons. Table 10 offers such an examination, listing the sources and expenditures of neutrons. The sources include the waste actinide referred to as TRU make-up in Table 8 multiplied by the number of neutrons generated (1.2) in the destruction of an average nucleus of the waste. The ^{233}U also generates neutrons at the rate of $(\nu - 1 - \alpha) = (2.49 - 1 - 0.086) = 1.4$ per nuclide destroyed by fission. The 20-mA accelerator produces neutrons at the rate of 30 neutrons per 1-GeV proton. The loss of neutrons at the rate of one neutron per nucleus occurs for the thorium conversion to ^{233}U and for the destruction of the fission products ^{99}Tc and ^{129}I.

Table 10 Neutron production and beneficial use for fast spectrum[a]

Material	Mass kg/year	Neutron Source $\times 10^{27}$ n/year	Neutron Sink $\times 10^{27}$ n/year
Waste actinide feed	425	1.28	
^{233}U fission	195	0.67	
Beam 20 mA 1 GeV		0.11	
Thorium feed	370		0.96
^{99}Tc transmutation	50		0.30
^{129}I transmutation	15	2.06×10^{27}	0.070
Total			1.33×10^{27}

[a]The neutron source is 1.5 times as large as the neutron sink, leaving a large excess to be lost to parasitic capture and leakage. The neutron excess is 6.6 times larger than the accelerator production so that the accelerator is unnecessary for the neutron economy of a fast-spectrum ADS.

Dividing the production rate by the use rate gives 0.64 as the fraction of the neutrons produced that are used productively. Because only a small part of the excess neutrons can be taken up by parasitic capture, most of the excess 36% of the neutrons must simply leak away. If one leaves out the accelerator, the fraction 0.64 increases only to 0.68. Clearly the accelerator's role in the neutron economy is superfluous; a fast reactor's neutron economy is more than adequate.

Another possible role of the accelerator is to maintain constant power over the two-year lifetime of the fuel, during which k_{eff} changes between 0.98 and 0.96. However, this fluctuation is readily accommodated by reactor control rods, which routinely control reactivity changes at least twice as large. The only remaining role for the accelerator is to make possible the additional operational safety associated with subcritical operation, but arguing that fast reactors without accelerators are insufficiently safe might be difficult. This analysis demonstrates that the accelerator is essentially superfluous for a fast-spectrum transmuter. On the other hand, it has already been shown that the advantages of the thermal-spectrum transmuter are only accessible with an accelerator, since the accelerator-produced neutrons are essential to the neutron economy.

It is useful here to recapitulate the problems of the CERN-proposed fast-spectrum system arising from small reaction cross sections and the use of solid fuel. Owing to the small cross sections, the system takes a long time to equilibrate, and it requires large inventories. The consequences of solid fuel are (*a*) that the fuel spends more time out of the neutron flux than it otherwise would, owing to the need for fuel destruction, and fuel refabrication further increases the inventory; (*b*) the accelerator must be about a factor of two larger than is necessary for a liquid-fueled system; and (*c*) the production of ^{233}U needed to stabilize the reactivity decreases the facility effectiveness for waste destruction by a factor of two.

Fast-Spectrum Existing Technology Base

The technical base underlying this proposal is substantial. Lead-bismuth cooling has been used extensively in Russian Alpha-Class submarine reactors, and this technology has been largely declassified. Although the CERN proposal uses lead coolant, the difference in implementation should not be of great significance. The Argonne National Laboratory in the United States has extensively researched the metallic fuel. The pyroprocessing required with the metallic fuel also has been well researched, although most evaluators probably would not consider that development complete at this stage. The linear-accelerator technology is well developed at the Los Alamos National Laboratory and elsewhere, and the 20 mA of beam required is well below the 100 mA design objective of the Accelerator Production of Tritium project.

COMPARISON OF THERMAL- AND FAST-SPECTRUM ACCELERATOR-DRIVEN SYSTEMS

The differences in operational features of the thermal and fast spectrum are manifest in performance. The differences depend on the transmutation objective, but the thermal-spectrum ADS always performs substantially better. Figure 13 shows the situation for close-out of nuclear power for the US fleet of about 100 LWRs with actinide waste destruction. The top curve (same as the production curve in Figure 9) shows the waste generation for 100 LWRs which produce about 300 kg/year of PMA. The lowest curve shows the inventory for the installation of 50 1500-MWt ADNA thermal-spectrum transmuters (or 100 750-MWt systems) brought online between 2015 and 2025. The fleet would consist of 40 Tier 1 systems and 10 Tier 2 systems. The transmutation of the waste would be complete 35 years after completion of deployment.

The middle curve in Figure 13 shows the waste destruction using the same number of 1500-MWt CERN fast-spectrum transmuters. Almost half of the waste remains after the ADNA system has finished its waste destruction in 2060. The CERN systems also will have exhausted their 40-year lifetime in about 2060. To destroy the other half, a new fleet of CERN transmuters must be brought online. Continued reduction of the waste at the same rate is not possible for the close-out option because the CERN system is inventory-limited. This is evident in Figure 14, which shows the CERN system inventory and transmutation rates. The internal inventory for the CERN transmuter begins at about 2500 kg. This initial load stays in the system for two years. Assuming that after removal the fuel must spend twice as long outside as inside the transmuter for fuel destruction, separations, and refabrication, the fuel would not be returned to the transmuter until six years after it was first put in. Therefore,

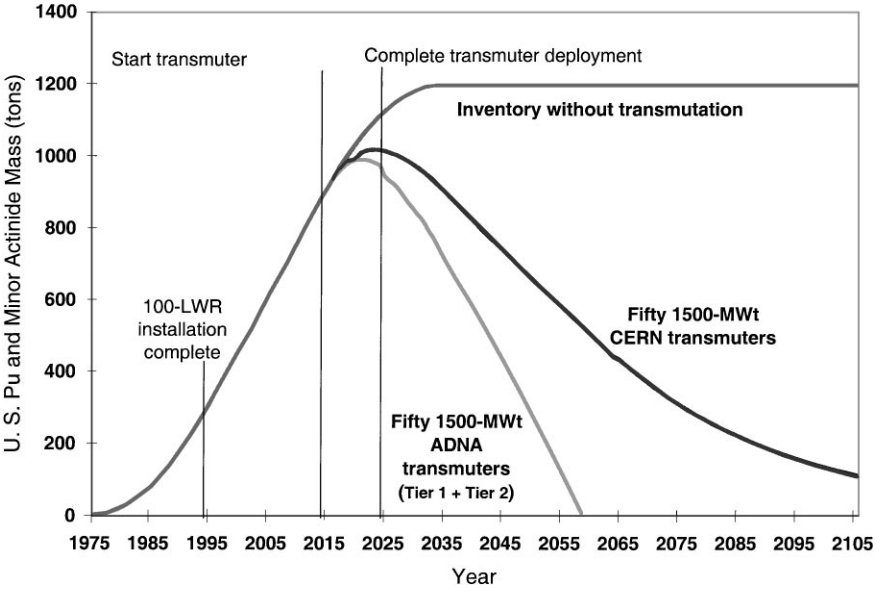

Figure 13 Comparison of CERN and ADNA systems in the close-out option. *Top curve*: Waste accumulation from 100 LWRs for a lifetime of 40 years. *Middle and lowest curve*: Time dependence of the waste for deployment of the power capacity of CERN or ADNA transmuters. The ADNA system has smaller inventory and does not require thorium and ^{233}U to maintain reactivity stability. If the objective is reduction of the 1200 tons by a factor of 100, the CERN system requires four times as long and about twice as many transmuters of the same power compared with the ADNA system.

the total fuel both inside and outside the transmuter would be three times as great as that shown in the middle curve of Figure 14. The inventory grows with time and reaches about nine tons in equilibrium, which is established after about 40 years.

The inventory requirement for 50 of these transmuters for the US waste is therefore $9 \times 50 = 450$ tons. When the inventory drops below that level, fewer transmuters can be fueled. The burn-up rate then decreases exponentially. The rate of waste destruction is $dW/dt = -Nb$, where W is the inventory of waste, N is the number of transmuters on line, and b is the rate of transmutation for one transmuter. However, N depends on the inventory I as $N = W/I$. Thus we find the expression $W = W_0 \exp(-bt/I)$. From Figure 14, the value of b near equilibrium is about 300 kg, whereas the inventory I is 9000 kg, so that $b/I = 0.0333$. The result of the calculation is shown in Figure 13, where the CERN inventory decays exponentially below the 450-ton inventory figure and reaches

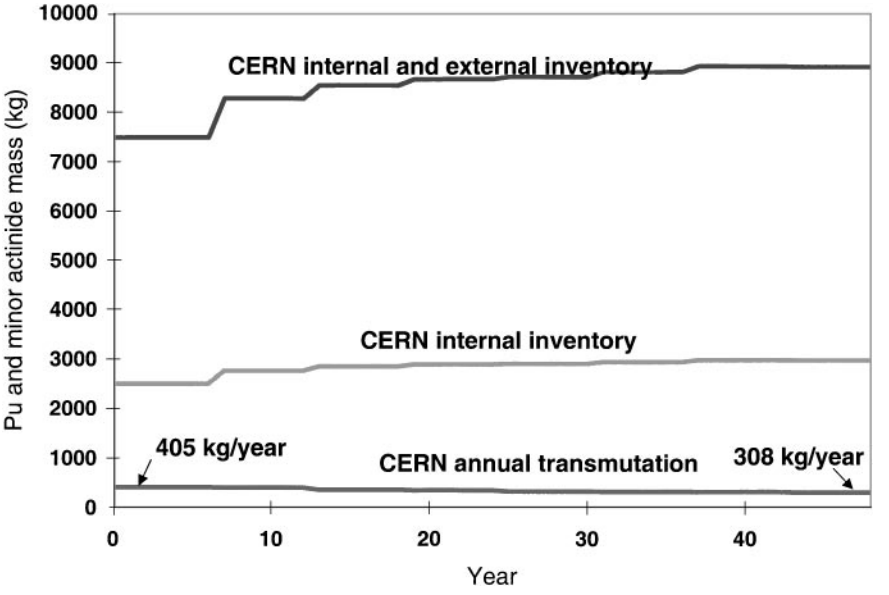

Figure 14 CERN system inventory and transmutation rates. The internal inventory for a CERN 1500-MWt system is shown as the *middle curve* (7). If the waste must spend twice as long outside for fuel destruction, chemical separations, fuel refabrication, and transport, the total of the internal and external inventory is three times larger (*upper curve*). Since the dwell time for the fuel inside the transmuter is two years and there must be two more loadings outside the transmuter, the inventory requirements change in six-year increments. The CERN system's annual transmutation rate decreases from an initial 405 kg/year to 308 kg/year as it approaches equilibrium in about 50 years.

10% of the waste produced by the year 2105. The CERN calculation of Figure 13 takes into account the time dependence for the transmutation rate to reach equilibrium.

To summarize the results presented in Figure 13 and 14 for close-out of nuclear power, the CERN system, because of its requirement for ^{233}U production from thorium and because of its large inventory, takes twice as long as the ADNA system for the same-sized fleet to complete the waste reduction. If the transmuters are assumed to have the same lifetime as present-day reactors (about 40 years), almost twice as many CERN transmuters must be built. Even by the year 2105, 10% or 120 tons of the waste remains and 15 CERN transmuters remain online. Another 70 years would be required to reduce the waste by a factor of 100 (from 1200 tons to 12 tons). By comparison, the external and internal inventory for the ADNA system's 1500-MWt transmuter is 0.516 tons

and the annual burn rate is 0.600 tons. Therefore inventory does not influence the ADNA system's transmutation rate until the inventory decreases to about 37 tons in about 2060. A goal of reduction by 100 after close-out of nuclear power would take four times longer with the CERN systems than with the ADNA design and would require about twice as many transmuters of the same power capacity.

The next comparison of ADNA and CERN systems is for continued LWR deployment. Figure 15 shows the result of deploying 100 1500-MWt CERN or ADNA systems for reduction of the waste inventory from a 100-LWR fleet maintained indefinitely. When deployment begins in 2015, the waste inventory would be about 725 tons and the CERN system would be unable to reduce it much over the next 40 years. However, the same-sized ADNA system deployment would eliminate the waste within 35 years. At that point the number of systems required to keep the inventory small would be 50 instead of 100 units.

With the ADNA system deployment, the only weapons material left after 2050 would be about 1 ton of neptunium dispersed in the 1250 tons of salt in the 50 ADNA transmuters and the plutonium and neptunium in the spent-fuel assemblies awaiting transmutation. The CERN ADS deployment of Figure 15 would not reduce significantly the weapons-useful material, although it would stabilize it at about 700 tons.

The large inventory of the CERN system also has implications not evident in Figure 15. The CERN inventory of PMA begins at about 7.5 tons and rises to 9.0 tons, as seen in Figure 14. Therefore, the inventory for 100 of these systems varies between 750 and 900 tons. However, the actual inventory from Figure 15 is never as large as 900 tons. This means that inventory in the spent-fuel assemblies and inside the transmuters limits the number of CERN systems that may be deployed and therefore the rate at which the waste can be destroyed. The actual CERN ADS performance is therefore not as good as Figure 15 indicates, since it is not possible to deploy all of the 100 CERN systems necessary to achieve the performance shown in Figure 15.

It is also important to note that the performance indicated in Figure 15 for the CERN system with continued deployment of LWRs cannot be improved by the deployment of more CERN systems. The deployment of twice as many CERN systems in order to double the transmutation rate would require 1800 tons of plutonium and minor actinide (9 tons $\times$ 200 transmuters) for fuel. These limitations can be stated analytically in the following equation:

$$\mathrm{d}W/\mathrm{dt} = N_{\mathrm{LWR}}p - N_{\mathrm{trans}}b = N_{\mathrm{LWR}}p - (W/i)b,$$

where N_{LWR} is the number of LWRs to be maintained on line, p is the annual production rate per LWR of PMA, N_{trans} is the number of transmuters and b is the

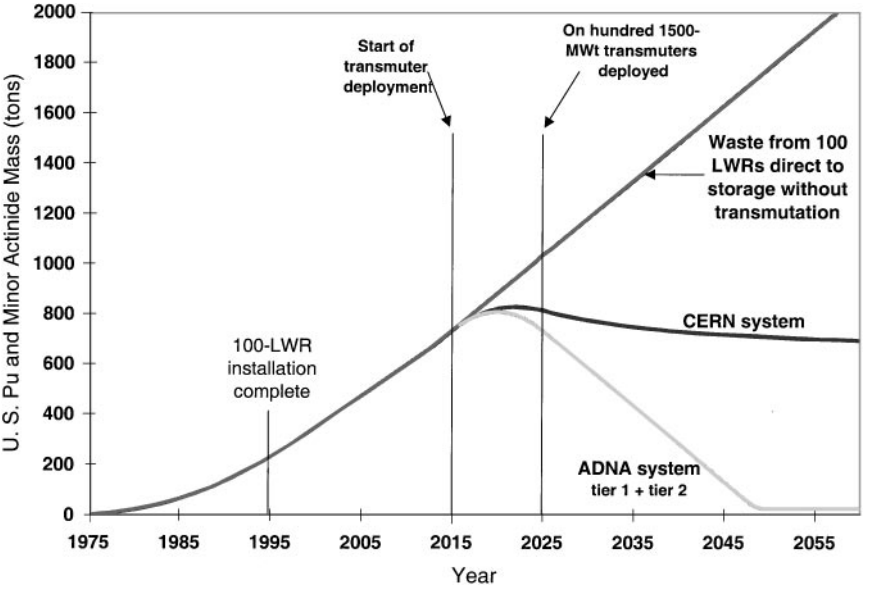

Figure 15 Comparison of ADNA and CERN system deployment for continued LWR deployment. The waste inventories are for once-through storage without transmutation, and with implementation of 100 1500-MWt CERN or ADNA systems. Because the 100 CERN systems destroy the waste only a little faster than the 100 LWRs generate it, the CERN system deployment only halts the growth of the waste pile; twice as many units would be needed to destroy the inventory. Because the ADNA system waste destruction rate is twice as large, the ADNA system is able to burn the inventory down to 25 tons. After the inventory has reached this level, only 50 ADNA systems are necessary to keep the waste inventory from growing. After 2050, the only weapons-useful material accessible is in the few spent-fuel assemblies awaiting transmutation. In the CERN system, there is as much weapons-useful material in spent-fuel assemblies in 2060 as there was when deployment began in 2015.

annual burn rate per transmuter. However, $N_{\text{trans}} = W/i$, where i is the inventory per transmuter if the goal is to deploy as many transmuters as possible to manage the waste as effectively as possible. In the burn-up of the waste, one eventually reaches equilibrium between waste generation and destruction with $dW/dt = 0$. For the CERN system, the minimum inventory is $W = 900$ tons, which may be compared with the 700 tons of Figure 15 when inventory constraints are not taken into account. The same calculation for the ADNA system gives 37 tons. Similar calculations for MOX burning of plutonium in LWRs show that MOX burning alone for control of plutonium, were it practical, would also require large inventories of plutonium.

Other Accelerator-Driven Systems for Commercial Waste Transmutation

The ADNA and CERN designs have been discussed in detail in this chapter because they represent the most thoroughly examined examples of thermal- and fast-spectrum systems. However, there are other designs, which are briefly described next.

OTHER THERMAL-SPECTRUM SYSTEMS Several thermal-spectrum designs have been examined over the past decade. The earliest design included D_2O moderation and coolant with actinide carried as oxide slurry (18). This system suffered from a very low thermal-to-electric efficiency, a high inventory owing to its low flux, and the likelihood of erosion problems in transporting solid material in a closed loop. A high-flux molten-salt system was also proposed (19) based on the Molten-Salt Reactor Experiment (MSRE) experience using the LiF-BeF_2 salt. Although this design carried no obvious technical problems that were not successfully resolved in the MSRE experiment, it required extensive front-end and back-end separations, which were likely to be expensive to apply in the transmutation of commercial waste. It was not possible to use this design in the once-through mode with throw-away of the carrier salt because the lithium salt must be isotopically pure ^{7}Li and because of the expense of the beryllium. Back-end separations would have been required for carrier-salt recovery, which would spoil one of the main advantages of the once-through Tier 1 system.

Perhaps the one thermal-spectrum system design deserving more study would be a liquid-fueled LiF-BeF_2 system without a graphite moderator but with a thick graphite reflector around the outside. The version studied was not nearly as well moderated as the Tier 1 system described above, and therefore the reaction cross sections are smaller and the inventories larger. The use of an expensive carrier salt means that back-end separations would be a requirement. However, this system might be successful in the Tier 2 application, since separations are required for that system and the carrier salt is recovered.

OTHER FAST-SPECTRUM SYSTEMS The primary focus of reactor development for the past 25 years has been fast-spectrum reactors, and the effort has strongly influenced fast-spectrum ADS design. Although problems with handling liquid sodium safely can probably be overcome, the positive void coefficient associated with this coolant is also a problem. Therefore, in most of the fast-spectrum systems, the interest is currently in lead or lead-bismuth metallic coolants, which are not pyrophoric and which exhibit a negative void coefficient. The neutron spectrum is also considerably harder when sodium is replaced by heavy metal,

and therefore the neutron economy is better for a lead-cooled system. The hardest spectrum and the best neutron economy are obtained through the use of metallic fuel instead of MOX fuel with heavy-metal coolant, although, as this chapter has shown, fast-spectrum designs thus far do not take significant advantage of this abundance of neutrons.

A somewhat different approach (20) to the fast-spectrum ADS is to destroy by fission as much as possible of the plutonium and neptunium in existing MOX burners and fast reactors and to destroy the americium and curium in an ADS. None of these nuclides are fissile, so a thermal spectrum is not so effective for destroying these materials by fission. Because these nuclides undergo fission with neutron energies in the 100 keV range or higher, the system has the potential to be an effective means of eliminating the higher actinides. Because the system does not burn plutonium, it would play only a minor role in the total LWR PMA destruction. The advantage to this is that not many would have to be built. The disadvantage is that the ADS system only adds complexity to an already complex system (shown in Figure 1), and it does not address the concern for a large inventory of plutonium illustrated in Figure 2.

SUMMARY

Thermal- and fast-spectrum accelerator-driven systems have been examined for use in reducing the inventory of plutonium and neptunium as weapons materials and as undesirable waste for long-term repository storage. Both approaches appear to be capable of destruction of the long-lived species in the waste to the point that a central repository is not required and the remnant waste can be stored in canisters. The lifetime of these canisters can be much longer than the activity of the remnant radioactivity (about 300 years). Both ADS approaches eliminate the need to rely on geologic containment of waste and the need to develop a single best place to store this waste, which many people consider the most dangerous waste known to man.

The thermal-spectrum system approach outlined here is a two-tier system. The objective of Tier 1 is not to eliminate the need for geologic storage but to eliminate some major concerns about geologic storage by extracting the energy available in the plutonium and minor actinide, denaturing the plutonium as weapons material, and virtually precluding underground criticality arising from rearrangements by natural, accidental, or malicious means. In this system, back-end separations are unnecessary and the front-end separations are greatly reduced. The cost-saving that results from the near elimination of the separations greatly enhances the economic practicality of the ADS; it is anticipated that electric power sales will pay for nearly all of the Tier 1 costs.

The elimination of the separations also greatly reduces the time and cost of development and demonstration of the technology. Therefore it is expected that the Tier 1 ADS could be deployed beginning in 2015. The well-documented MSRE program at ORNL, in which a molten-salt reactor was run for several years, demonstrated much of the required technology.

The Tier 1 systems are small and capable of destroying the actinide from one 3000-MWt LWR power plant on the site where the waste was produced. The near absence of separations is important in making this siting possible, along with the fact that the modest accelerator has a small footprint. There is no need to develop new nuclear sites for waste elimination when it can be destroyed where it is produced.

If it is decided that geologic storage is impractical, the Tier 2 thermal-spectrum technology can be deployed, eliminating reliance on geologic storage. The costs and time for development and deployment of the Tier 2 system are greater but the system must operate on only 20% of the waste, since 80% was already destroyed in the Tier 1 system. The incremental costs of the deployment of Tier 2 might still be acceptable when spread over the other 80% of the waste.

The fast-spectrum ADS technology as presently conceived is a single-step process for the destruction of all of the long-lived wastes. It therefore does not require geologic storage of its waste, so long as the ^{233}U is consumed. It is closely related to fast-reactor technology, which has been studied extensively for the past 30 years, and this is seen by many as an important factor in implementation of a fast-spectrum ADS. The fast spectrum also has the advantage of a better neutron economy than the thermal spectrum, although the thermal-spectrum neutron economy is apparently adequate, as illustrated in this paper. The fast spectrum has the disadvantage of small reaction cross sections, which increase the inventory and the time to equilibrium by more than an order of magnitude even though the fast-spectrum flux may be considerably higher than the thermal flux. These larger inventories and times to equilibrium are crucial in practical deployment, as shown above in the time displays for plutonium reduction. The fast spectrum builds up less of the higher actinide than a thermal spectrum does. However, this advantage is compensated by the smaller inventory of the thermal spectrum, as shown in several inventory comparison figures.

The most serious liability of current fast-spectrum designs is the use of solid fuel. Solid fuel and sub-critical systems are not well matched because, as the fuel burns away, it cannot be continually replenished, as can liquid fuel. Instead, the reactivity of the fuel decreases in practical situations by about 2–4%, which is about the same as the degree of subcriticality. In a normal reactor, much of this reactivity swing is controlled by breeding of more fissile material and

by control rods. The CERN system offsets some of the reactivity swing by producing ^{233}U, but still exhibits a swing from 0.96 to 0.98 over a fuel cycle. This reactivity swing shows up as a factor-of-two change of fission power, which is controlled by changing the accelerator current by a factor of two. This solution is not attractive because for much of the cycle half of the accelerator beam is unnecessary, and therefore the accelerator capital investment is not fully used. Because a significant fraction of the neutrons is allowed to leak away, the accelerator neutrons do not play an essential role in the neutron economy. The 2% reactivity swing could be readily controlled with control rods, since fast-reactor control rods typically control reactivity swings substantially higher. Therefore, it is not clear that the fast-spectrum sub-critical system offers a clear advantage over the fast reactor with solid fuel. The only role for the accelerator is in allowing the subcritical operation. The debate probably will continue for some time as to whether this perceived advantage is significant in improving heavy-metal–cooled fast-reactor safety.

The extensive separations requirements of the fast-spectrum systems eliminate the possibility of siting these systems on existing reactor sites. This restriction may reduce the political acceptance of transmutation and causes difficult problems in establishing a satisfactory price for the large amount of power that would be introduced at one or a few points into the commercial grid.

In the end, the selection of the ADS design for actual deployment will be more a business decision than a political decision. The technology used for tritium production is primarily a political decision in the United States because only one accelerator-driven or reactor unit is needed and the tritium is essential to maintain the nuclear stockpile. Therefore costs are secondary to security of procurement and other issues. However, waste transmutation, which could nearly pay its way, would be potentially big business no matter how it is done, since the plutonium and minor actinide produced amount to 25% of the fissile material burned in LWRs. If each of the approximately 400 LWRs in the world require one Tier 1 ADS or its equivalent, the worldwide market for 400 of these at an estimated cost of about $0.5 billion each would be $240 billion. This is comparable to the world airframe industry. The impact might be much larger if one takes into account the potential growth in nuclear-power deployment made possible by the resolution of the nuclear-waste disposition issue. In such a situation, performance and cost will always be vitally important in technology selection, and even a small advantage of one system over another can be a deciding factor.

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