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Optimization of Accelerator-Driven Technology for LWR Waste Transmutation

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Abstract

The role of accelerator-driven transmutation technology is examined in the context of the destruction of actinide waste from commercial light water reactors. It is pointed out that the commercial plutonium is much easier to use for entry-level nuclear weapons than weapons plutonium. Since commercial plutonium is easier to use, since there is very much more of it already, and since it is growing rapidly, the permanent disposition of commercial plutonium is an issue of greater importance than weapons plutonium. The minor actinides inventory, which may be influenced by transmutation, is compared in terms of nuclear properties with commercial and weapons plutonium and for possible utility as weapons material. Fast and thermal spectrum systems are compared as means for destruction of plutonium and the minor actinides. It is shown that the equilibrium fast spectrum actinide inventory is about 100 times larger than for thermal spectrum systems, and that there is about 100 times more weapons-usable material in the fast spectrum system inventory compared to the thermal spectrum system. Finally it is shown that the accelerator size for transmutation can be substantially reduced by design which uses the accelerator-produced neutrons only to initiate the unsustained fission chains characteristic of the subcritical system. The analysis argues for devoting primary attention to the development of thermal spectrum transmutation technology. A thermal spectrum transmuter operating at a fission power of 750-MWth fission power, which is sufficient to destroy the actinide waste from one 3000-MWth light water reactor, may be driven by a proton beam of 1 GeV energy and a current of 7 mA. This accelerator is within the range of realizable cyclotron technology and is also near the size contemplated for the next generation spallation neutron source under consideration by the U.S., Europe, and Japan.

Introduction

From the beginning of the development of commercial nuclear power, it was recognized that means for safe disposition of the remnant waste would have to be developed. The waste was to consist of fission products alone because it was expected that the actinide component of the waste would be destroyed by fission with enhancement of the total energy output from the fuel. Since the decay half-lives of the longest lived fission products are measured in the hundreds of thousands or millions of years, it was not possible to build storage canisters for which the integrity could be guaranteed for such a long period. Therefore geologic storage of the waste fission product was proposed for the purpose of confining by the stable geology of the site the movement of the waste after the emplacement canister had decayed away .

Nuclear proliferation concerns in the 1970s led to an additional challenge for geologic storage. The reprocessing of spent fuel from commercial power reactors to recover and burn the plutonium it contained was viewed as being a key element in an international market for plutonium. The reprocessing technology called PUREX produced a pure stream of plutonium so that new fuel assemblies containing the plutonium could be fabricated. Some of this pure plutonium output stream might be stolen for use in building nuclear weapons by terrorists or rogue states. Therefore in the late seventies President Carter issued a Presidential Order stopping the development of reprocessing and establishing the "once through" policy which required that commercial spent fuel assemblies be placed directly in geologic storage. The plutonium they contained and other higher actinide were

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therefore added to the burden of long lived radioactive species which must be contained by geologic storage. The U. S. policy since that time has been to discourage foreign countries from the development of reprocessing and to encourage the adoption of the once-through approach. Over the past twenty years few nations have adopted this policy partly because they wished to enhance the yield of electric energy from a given mass of fresh fuel by burning plutonium, and partly owing to proliferation concerns about the large amount of plutonium which would be stored in many repositories around the world in the once-through approach. (The world's inventory of plutonium in spent fuel would approach 6000 tons by the middle of the next century if nuclear energy continued to be deployed at the present capacity. Recent U. S. National Academy studies assert that only about 5000 grams are required for making a several-kiloton nuclear weapon.)

The concerns which President Carter was attempting to address about proliferation associated with reprocessing continues as long as the existing aqueous technology (PUREX and its extensions) remains as the separations technology of choice. The development of other separations and fuel fabrication technology which would allow the plutonium and higher actinide to be fissioned completely without separating a pure plutonium stream would eliminate the primary justification for the U. S. ban against reprocessing. The intent of other nations to burn the plutonium helps reduce the large plutonium inventory, but it does little good to burn half or two-thirds of the plutonium since the inventory would still be measured in thousands of tons. If plutonium is to be destroyed by fission to eliminate proliferation concerns, it must be destroyed completely so that the remnant is small, the isotopic content of the remnant is unfavorable for weapons material, and so that natural decay does not eventually convert the remnant into weapons material. All nations' concerns about plutonium would be met if plutonium and other higher actinide could be destroyed completely by a system which is efficient in generating electric power from the fission heat and which did not provide access to pure weapons material along the way. Accelerator-driven systems for spent commercial fuel promises to achieve these objectives.

With the end of the cold war the clean-up of the nuclear defense complex in the U. S. and Russia has also required both nations to address the disposition of excess weapons plutonium and an apparently large inventory of spent fuel from naval reactors containing highly enriched uranium. Attention to these materials also has drawn highly enriched spent research reactor fuel into the spotlight. A prominent option for the U. S. for these three highly enriched fissile materials was emplacement in geologic storage and a strong movement in this direction was initiated. Since these fissile materials and some of their fissile daughters have half lives of millions of years, dissolution of their emplacement canisters is assured allowing these fissile materials to reconfigure. This prospect provided the rationale behind Los Alamos work which showed that these materials could reconfigure into critical masses, that the reconfiguration could have positive feedback, and that the confinement provided by the surrounding rock taken together with positive feedback might give rise to large nuclear explosions in a repository[1]. Although it is difficult either to quantify the likelihood of such events or to show that the likelihood is too low as to be ignored, the possibility of the occurrence of such events has been confirmed[2]. Mitigation means to reduce the possibility of such events have been proposed and further study to evaluate such strategies has been proposed[2]. It also has been reported that commercial spent fuel also will exhibit criticality with positive feedback in emplacement configurations currently planned by the U. S. Department of Energy[3,4].

The viability of repository storage of fissile material also was dealt another blow by studies comparing the cost and speed of obtaining fissile material by removal from geologic storage with isotopic separation of ^{235}U or reactor production of ^{239}Pu. The results of the study

were that it was more than ten times faster and ten times cheaper to remove the fissile material from a repository than to produce it by either of the alternatives[5]. The proliferation concern about fissile material in repository storage was recognized by the IAEA which has concluded that the repositories must be guarded into perpetuity[6].

Finally the very nature of the scientific process for finding a best single site for storing the spent fuel assures difficulties in political acceptance. The purely technical challenge is to find the single best site in the nation for storage of nuclear high-level waste and to scientifically characterize it to the highest possible degree in order to understand the best emplacement procedure. The fact that there probably is in principle such a single best place imposes on the host community the obligation to accept this waste, which many believe to be the worst of the nation's wastes. Other types of society's waste can be stored locally, but the "worst" waste must be treated with the greatest care, stored in the single best place as verified by thorough scientific investigation, and must be monitored indefinitely since its long term toxicity never decreases on time scales relevant to human history or experience. The citizens of Nevada are unhappy with this and the rest of the nation understands that it's fundamentally unfair to force this waste on a single community which doesn't want it.

Current uncertainty on waste policy in the U. S.

The U. S. DOE and the Congress seem to be tabling for the time being Yucca Mountain or any other site for permanent waste storage. The Congress instead is considering a bill for interim storage of commercial spent fuel near Yucca Mountain. Of course this is not acceptable either to the Nevada residents because without means for ultimate disposition the interim site becomes a defacto permanent site. Furthermore if the waste were placed at a single site, there would be less motivation to search out means other than storage for dealing with the waste and there would be little possibility that the waste ever would be moved to another site. Apparently interim storage will not be implemented because President Clinton intends to veto the bill and there is not sufficient support in the Congress to override the veto. The commercial spent fuel therefore will stay for the foreseeable future on the reactor sites in dry storage. The situation will be viewed as unsatisfactory and other disposition strategies which do not have the single-best-site liability may finally get considered.

It is useful to note here that if the long lived species of the waste is destroyed sufficiently well such that storage canisters can be made which outlast the waste, then a single best geologic storage site is not required. The waste can be stored almost anywhere that it can be physically guarded since leakage from the cannisters is not an issue. Eventually it decays to low level Class C waste which can be stored without being guarded following established Nuclear Regulatory Commission and Environmental Protection Agency guidelines. It remains to be demonstrated that complete burn-up of the long-lived species is practical. However, no alternative to complete burnup of all long-lived species of the waste appears to be in prospect since, without geologic storage, neither the storage option or the partial burn-up option for weapons plutonium is viable. These same arguments also are relevant for the disposition of spent naval fuel and research reactor fuel.

The energy released in total burn-up of plutonium and minor actinide excludes the possibility that all of the commercial plutonium could be destroyed on a single site. The thermal power generated in burning the plutonium and minor actinides by fission using accelerator-driven systems (ADS) or by other means from a single commercial power reactor operating at 3000 MWt is smaller by about one quarter or about 750 MWt . This 4-to-1 ratio implies that it would take about 25 ATW systems operating at 3000 MWt for about 40 years to burn the waste from our roughly 100 reactors of 3000-MWt power. The cost to build and operate these twenty five 3000-MWt systems for 40 years is impossibly

large unless the heat generated from destruction of the waste by fission is converted efficiently to electric power and sold to pay these costs. All of this power from the equivalent of 25 reactors would have to be fed from this one site into the grid. This is far too much power to be absorbed from one site. Therefore except for nations with a very small committment to nuclear power, systems combining plutonium and minor actinide burning would have to be located on multiple sites. It may also be technically possible to burn the plutonium as MOX fuel in LWRs or in fast reactors and to burn the minor actinides separately in accelerator-driven systems. This approach has the disadvantage of requiring the carrying of large plutonium and minor actinide inventories and it requires the development of a challenging new technology with limited opportunity for the widespread deployment required to recover the development costs.

It becomes more practical to burn the plutonium and minor actinides if the waste is hauled to several regional sites. In the U. S., for example, if the commercial waste were equally distributed to Savannah River, Oak Ridge, Idaho Falls, the Nevada Test site, and Hanford, then only five ATW systems would be required on each site. The grid might well manage to accept this power of about 5 GWe from each of five sites and pay a competitive price for it. The remnant waste could then be transported to a geologic repository for final permanent disposition in this denatured form or stored locally in canisters which would reach Class C levels after 300 years. Canisters certainly can be made which can maintain their integrity until the waste remaining after transmutation has decayed to Class C levels. It might also become practical to destroy these materials on the reactor site where they are generated as described later in this report.

Weapons Usefulness of Commercial Plutonium

To express the need for total burn-up of commercial plutonium in the clearest terms, the following conversation might be of interest. This past February the author was sitting wearing a heavy ski jacket in the back of a cold conference hall in St. Petersburg, Russia beside a slightly bored Russian former nuclear weapons designer. The purpose of the meeting was to discuss technology affecting the future of the two kinds of plutonium...weapons plutonium taken from nuclear weapons in stockpile reductions and the commercial plutonium being produced worldwide in commercial nuclear reactors. The Russian was restless and presently he leaned over and whispered, "I don't understand the U. S. policy at all with regard to weapons plutonium? Why turn weapons plutonium into commercial plutonium when it's much easier to build nuclear weapons with commercial plutonium?" His further words are paraphrased below.

He said, "Three technologies must be mastered to make a nuclear weapon out of weapons plutonium. First, you have to master the compression technology...driving the plutonium into a highly compressed ball with conventional high explosive. Second you must produce a burst of neutrons to start a rapidly growing chain reaction and that's not so easy. And third, you have to time the burst of neutrons just right or the neutrons injected will come too late or to early. If you fail at any of these three requirements the bomb will be a dud." These requirements are well known and the role of neutron initiation had been stated in unclassified and published work[7] by Dr. Carson Mark, a leader in nuclear weapons design at Los Alamos in the 1960's. Mark's work also was referenced in the highly publicized study conducted by the U. S. National Academy of Sciences entitled, "Management and Disposition of Excess Plutonium[8]" and similar information has been communicated by Russian scientists[9].

He continued, "For nuclear weapons from commercial plutonium you need only the compression technology. Lots of neutrons are already present because the commercial plutonium contains isotopes which undergo spontaneous fission and produce neutrons

randomly. Because they are there already, one cannot control the timing of the injection of neutrons so the explosive power is quite uncertain. It might be anywhere in the range from 2000 tons of TNT to 18,000 tons. Who cares whether the explosion is 2,000 or 18,000 tons when the damage is proportional to the cube root of the yield and is therefore only about a factor of two different? Terrorists wouldn't and even a rogue nation's war planners wouldn't care much. So why does Washington keep pushing us to convert difficult-to-use weapons plutonium into easy-to-use commercial plutonium? Your policy is influenced too much by your weapons designers at Los Alamos and Livermore. With the advanced technology developed in the U. S. and Russia, sure, weapons plutonium is the best because the explosive power is highly dependable and therefore always the maximum and you also can make all kinds of fancy bombs such as nuclear artillery shells and so forth. But suppose you don't have nuclear weapons and you want to get them quickly and easily and you have the choice of commercial or weapons plutonium. Your U. S. weapons designers believe a terrorist organization or rogue state will choose the weapons plutonium. But the clever fellow who has to build a reliable bomb for the boss fast and cheaply will prefer commercial plutonium."

The Russians wish partly for this reason to burn the excess weapons and commercial plutonium as does most of the rest of the world. The Russians have developed new reactor technology to do this and the French, Japanese, and others also are working on the implementation of reactors. There is growing international interest in accelerator-driven systems with active programs in France, Japan, Russia, at CERN, and a more modest program in the U. S. at Los Alamos which promise to make complete destruction ultra-safe and affordable. From this array of technology studies could emerge practical means for total destruction of plutonium before the first plutonium anywhere in the world finds its way into geologic storage.

Input from the National Academy of Sciences

Aggressive development of this new accelerator-driven technology has not been endorsed by the National Academy of Sciences recommendations[10,11] and the U. S. policy has been rather neutral instead of supportive of such studies in other countries with one commendable exception[12]. The U. S. Department of Energy is proposing the adoption of the U. S. National Academy of Sciences recommendations which urge placement of plutonium of all types underground either with or without partial burn-up. It is now in the final stages of information gathering prior to a decision to embark on the implementation of these options[13]. It would be useful to examine the arguments which have steered thus far the selection of underground storage. Five positions underlying the National Academy of Science's recommendations are discussed below.

1. Reprocessing promotes an international market in plutonium.

Perhaps the weakest technical element in total plutonium destruction using existing technology is the PUREX process for separating plutonium from spent nuclear fuel. This technology was developed in the post war years and it or its derivatives are now widely deployed except in the U. S. It is presently not capable of dealing with the build-up of highly radioactive constituents of the waste produced in the course of complete plutonium destruction. One might develop the technique further to deal with its shortcomings, but it also has the problematic feature that it produces a pure stream of plutonium. The Swedes call this "naked" plutonium. The separation of this naked plutonium does not necessarily or perhaps ever match perfectly the feed into the plutonium-destroying systems. Therefore the excess must be stored, and perhaps to get a better balance between those who store and those who burn plutonium, it could be sold thereby creating a plutonium market. As with any commodity market it's not easy to prevent some of the commodity from being lost or

stolen. The U. S. is correct on the point of avoiding a market and should push on to prevent the development of a market in plutonium.

The solution to destruction of plutonium without producing a market in plutonium is not to ban reprocessing of any kind but to develop separations processes which do not produce pure plutonium. From the beginning the focus of U. S. accelerator-driven transmutation technology has been on separations which allow the destruction of plutonium without the production of naked plutonium[14]. Only the weakly radioactive zirconium fuel cladding and the uranium are removed so that the plutonium remains mixed with the most radioactive ingredients of the nuclear waste. The concentration of radioactivity of this product is about 100 times higher than in commercial spent fuel and this product can be fed directly into an accelerator-driven transmuter. From the viewpoint of the radioactivity of the material as a deterrent to diversion, it is therefore more feasible to divert the fuel assemblies themselves than to divert the product after preparation for transmutation. The U. S. approach also only makes accessible as much plutonium as the system burning it can use, so no excess is accumulated. With highly contaminated plutonium and no excess and with separation and burning integrated together on the same site, it is almost certainly feasible to develop means for destroying plutonium without promoting a market in plutonium. Therefore reprocessing need not promote an international market in plutonium.

2. Plutonium is safe in geologic storage.
The reasons why plutonium may not be safe in geologic storage already have been discussed in the introduction of the paper.

3. Accidental or purposeful repository intrusion is inconsequential.
Of course natural processes are not the only ones that could lead to critical configurations. The repository studies supported by the U. S. Department of Energy acknowledge the possibility of accidental intrusion into the repository as for example in drilling for water or minerals, although these studies had not recognized the possibility of criticality with positive feedback. The IAEA study and the Peterson study make the case that there are strong reasons to reenter and recover the plutonium from the repository for those wishing to obtain nuclear weapons capability. The repositories may be the richest or only source for other non-fissile materials of possible future interest. For example, all of the elements in the waste have isotopic concentrations different from those that occur naturally and have potential value for that reason. Mining a repository purposefully is therefore almost a certainty and if the mining is not done with great care, critical configurations could be created accidentally. Finally it is not out of the question that repository explosions might be deliberately induced for malevolent reasons. If the possibility for spontaneous criticality could be reduced to an acceptably low value (and how would that be decided?), the possibility of purposeful or accidental reconfiguration to criticality remains. Therefore accidental or purposeful intrusion into a repository can have serious consequences from a proliferation or criticality standpoint.

4. Conversion to the "spent fuel standard" is worthwhile.
For the many years while plutonium was stored in large inventories of nuclear weapons, the safety of weapons plutonium was not questioned. Since relative peace has brought major stockpile reductions, the disposition of the excess weapons plutonium particularly Russian plutonium has become an issue of major focus. There is good reason to want to get Russian weapons plutonium under control as quickly as possible. In response to this concern, the U. S. Academy of Sciences conducted a study entitled Management and Disposition of Excess Weapons Plutonium[8] to evaluate possible options. The recommendations included (1) declarations of weapons plutonium in the U. S. and Russia, (2) Safeguarded storage of this material, and (3) final disposition including storage

underground or partial burn-up in reactors before storage underground. After settling on underground storage, the issue of burning before storage underground was addressed by the NAS in a separate study with a report entitled, Management and Disposition of Excess Weapons Plutonium....Reactor-Related Options[11].

Both reports were strongly influenced by the concept of the "spent fuel standard." The plutonium in the commercial spent fuel as we have already seen is a mixture of isotopes which has some disadvantages for making sophisticated nuclear weapons. In addition the commercial spent fuel is in the form of spent fuel assemblies. The presence of the fission product radioactivity in the assemblies with the plutonium is felt to be another considerable deterrent to attempts to remove the plutonium for possible weapons use. Since there is so much more commercial plutonium than weapons plutonium, the transformation of weapons plutonium to commercial plutonium by burning in a reactor gets rid of the weapons plutonium but increases the amount of commercial plutonium by only about 10 %. Therefore conversion of weapons plutonium to commercial plutonium by partial burning is seen to be worthwhile.

The conversion of weapons plutonium to the spent fuel standard of commercial plutonium by partial burning would be an exercise of rather little value. As already shown, commercial plutonium is more useful to those we wish not to have plutonium than weapons plutonium. The value of the radioactivity as a deterrent decreases with time such that in about one to two hundred years the chemical separation of plutonium from the waste could be accomplished without the radioactivity being a significant barrier. Any advantage gained from the presently proposed policy of conversion to the spent fuel standard would be temporary and mainly would pass the resolution of the problem to future generations. They would have the responsibility for destruction following the probably dangerous task of recovery.

5. Weapons plutonium has only negative value.
Of course the main objective of U. S. plutonium policy might not be U. S. weapons plutonium but Russian weapons plutonium. The Russians understand that weapons plutonium has significant positive value and expect to receive come considerable societal benefit from the destruction of this material. The U. S. argues that weapons plutonium has negative value citing the energy value of the plutonium which is no different than for any fissile material. In the U. S. this case is valid because we currently have no technology available to extract the energy efficiently. However Russia has developed an advanced lead-cooled naval reactor[15] which its advocates would like to move into the commercial sector. It can burn the plutonium with significant advantage. In addition the Russians understand that the primary value of the weapons plutonium is not in the fission energy produced from burning but in the neutrons it produces. Much of the reason weapons plutonium is valued for sophisticated weapons is that it is an exceedingly rich source of neutrons. Since the key to nuclear energy is sustaining a chain reaction, the burning of weapons plutonium could enhance the reactor neutron economy allowing the chain to continue to operate while performing other useful functions such as destroying nuclear waste by transmutation using these neutrons. U. S. studies show that the economics of transmutation of commercial nuclear waste is very significantly enhanced by the burning of weapons plutonium and highly enriched uranium concurrently[16].

Fast vs. thermal spectrum destruction of waste actinides
Perhaps the most important technical issue for destruction of the plutonium and minor actinide waste from commercial nuclear power is whether to employ a fast spectrum or a thermal spectrum. Three arguments have been used in favor of the fast spectrum. The first is that the fast spectrum is more effective in destroying the nuclei exhibiting threshold

fission. This is a valid advantage because the cross section for fission of this class of nuclides is comparable to the capture cross section in the fast spectrum whereas in the thermal spectrum the fission cross section is negligible. The result is that nuclei which in the thermal spectrum only absorb neutrons to become heavier are often made to fission in the fast spectrum. Therefore the thermal spectrum is thought to have the undesirable feature of building up more of the heavier nuclides than the fast spectrum. The second advantage, which is a consequence of the first, is that the neutron economy of the fast spectrum system is superior to the thermal spectrum. The fast spectrum is able to induce fission in all of the nuclei with the accompanying fission neutrons boosting the neutron economy. For the thermal spectrum, virtually all of the absorption on the threshold fissioning nuclei result in neutron capture and a loss of a neutron. The third reason is that most nations with a major stake in nuclear technology have invested large sums in fast reactor research and development in the past twenty years displacing the thermal reactor studies of earlier times. Thermal reactor technology is therefore less familiar in the present research context and there also is a desire to see benefit from the enormous sums spent on the fast spectrum technology which has not been deployable up to the present. In summary the fast spectrum advantages of better neutron economy, less higher actinide build-up, and more familiar technology have drawn most of the attention to fast spectrum systems. It will be shown below that the thermal spectrum is markedly superior in spite of these advantages of the fast spectrum.

The advantage of the thermal spectrum system lies in the orders of magnitude higher reaction cross sections possible. Whereas the reaction cross sections for fast spectrum neutrons (~200 keV spectrum average) is only about 1 barn, the thermal reaction cross sections are typically several hundred barns. The number of neutron-induced reactions per unit volume is given by the product of the neutron flux, the cross section, and the actinide density. If the neutron flux for the fast and thermal spectrum systems were the same and the thermal fission cross sections were several hundred times higher for the thermal spectrum, then the actinide density would have to be several hundred times lower in the thermal spectrum to keep the same system power density. For the same power density the inventory of the thermal spectrum systems then could be several hundred times lower.

This advantage of achieving the same transmutation rate with a much lower actinide inventory is a compelling advantage. Even though the design characteristics of the thermal and fast spectrum system are quite different and probably not all of the advantage of the thermal spectrum can be preserved in a practical thermal spectrum system design, the advantage of the thermal spectrum system is commanding as will be shown next.

In comparing the effectiveness of reactors and accelerators, the flux level is an important factor. Fast spectrum reactors have been under study for many years in many countries and typically operate at a flux level of 2×10^{15} n/cm^2-s. Most thermal spectrum reactors use enriched fuel and therefore have not been designed for the excellent neutron economy which is demanded by a high flux thermal spectrum system. The exception is the widely deployed Canadian CANDU heavy-water-moderated reactor which uses unenriched uranium fuel. The thermal flux in these systems is 6×10^{14} and is limited by parasitic neutron capture at the operating temperature of about 250 degrees centigrade. If the temperature could be raised by a factor of three to about 750 degrees, the limitation from parasitic capture would allow the operation at a flux of about 1×10^{15}. While this temperature is not possible with an aqueous system, it may be possible with the graphite-moderated molten-salt reactor concept system studied at the Molten salt Reactor Experiment (MSRE) at the Oak Ridge National Laboratory. Therefore the flux level assumed for the fast spectrum and thermal spectrum system respectively is 2×10^{15} and 1×10^{15} n/cm^2-s.

The flux achieved in a reactor or accelerator-driven system is not usually the same as the time-averaged flux actually experienced by the material undergoing irradiation. In the fast spectrum reactors, the fuel is in solid form and must be removed about once per year. According to the Integral Fast Reactor (IFR) operational plan, the material would spend one year inside the reactor (i. e. exposed to the flux) and two years outside undergoing non-aqueous separations and refabrication into solid fuel. Therefore the effective flux to which the fuel is actually exposed is reduced by a factor of three to 0.66×10^{15} by this time spent outside of the reactor. The French Superphenix fuel would spend a substantially larger fraction of its time outside of the reactor owing to the aqueous processing employed there with a larger reduction in the average flux seen by the fuel.

One of the advantages of the accelerator-driven subcritical systems is the possibility to safely use liquid fuel. The fission power is deposited in the liquid and the fuel flows to an external or internal heat exchanger for heat removal. In the design study for practical deployment of the Oak Ridge MSRE technology, the liquid fuel in the form of molten salt flowed through an heat exchanger external to the tank containing the liquid fuel under irradiation. The fuel spent about 2/3 of its time outside of the neutron flux. If the heat were exchanged inside of the tank, the time spent outside of the flux could be reduced to the same as that spent inside of the flux. In this case a flux of 1×10^{15} is reduced to 0.5×10^{15}. An important advantage of a liquid fuel system is that no time (or fuel inventory) is required for solid fuel destruction or solid fuel refabrication. The fuel may be cleansed on-line promptly without the necessity for cool-down which also requires time and fuel volume. Therefore it appears to be practical to conduct on-line separations without significantly increasing the time spent by the fuel outside of the neutron flux. This is particularly true for the thermal spectrum system where the amount of fuel which must be processed may be much smaller than that required for a fast-spectrum liquid-fueled system.

Taking into account these considerations of time for fuel spent inside and outside of the neutron flux for either heat removal or fission product separations, the flux used in all of the following calculations is 0.5×10^{15} and 0.66×10^{15} ncm^2-s for the thermal and fast systems respectively. Anticipating the results to follow, it will be made clear that factors of a few either way for either system will not change the conclusion about the greater value of the thermal spectrum system for pluotnium and minor actinide destruction.

The amount of total actinide "waste" produced in a single 3000-MWth light water reactor (LWR) per year is given in Table 1. If this material is placed in a transmuter, a nucleus upon absorbing a neutron will either fission or will increase its mass by one unit to the next heavier nucleus of the element. Some fraction of a lighter nucleus can be transformed by many absorptions to a heavier nucleus before it is destroyed by fission. The differential equations for this process can be written for the beginning nucleus ^{239}Pu as follows:

$$dN_{239}/dt = F_{239} - N_{239}\phi\sigma_{c239} - N_{239}\phi\sigma_{f239}$$

$$dN_{240}/dt = N_{239}\phi\sigma_{c239} - N_{240}\phi\sigma_{c240} - N_{240}\phi\sigma_{f240} \qquad (1)$$

$$dN_{241}/dt = ...$$

where F_{239} is the feed rate of ^{239}Pu, ϕ is the neutron flux, and the subscripts f anc c refer to fission or capture following neutron absorption. The complete equations must include modest complications of alpha and beta decay.

The coupled differential equations can be solved either analytically or numerically to find the time dependence of the amount of any nuclide in the system. Various scenarios for dealing with the waste may be examined using these equations. Since the resolution of the commercial nuclear waste issue probably would reopen the door to indefinite use of nuclear energy, the most interesting question is the equilibrium amounts of the actinide isotopes in the system. This may be obtained by setting the derivatives in (1) to zero and solving the set of coupled algebraic equations.

The cross section parameters are of course substantially different for the fast and thermal spectrum systems. The thermal spectrum molten salt systems with graphite moderator operate at a temperature of about 0.1 eV where the reaction cross sections are about a factor of two smaller than at thermal energy. While spectrum averaging is necessary for the greatest accuracy for reaction cross sections, it is not vital since no actinide nuclei have resonances below 0.3 eV. The thermal spectrum system is expected to be well thermalized, but there is always a 1/v spectral component expressed as a resonance integral which modifies the rates somewhat. The cross section for these neutrons was taken into account by adding 5 % of the resonance integral cross section to the thermal cross section.

For fast spectrum analysis, spectrum averaging is very important because of the shelf in the cross section in the hundred keV range exhibited by the non-thermally fissile isotopes. The ratio of the capture and fission cross sections at 200 keV is taken from the Salvatores-Slessarev-Uematsu paper[20]. The absolute values for the thermally fissile isotopes is derived from the fission cross section at 200 keV since the cross section in the 200 keV range is typically nearly flat. For the non-thermally fissile isotopes, the absolute cross section is obtained from the capture cross section. This cross section does not exhibit a definite 1/v dependence, but the energy dependence is slow enough and close enough to 1/v that the value at 200 keV provides adequate accuracy. The cross sections used for both the thermal and fast spectrum system are given in Table 2.

The results from the calculation of the equilibrium inventory for the fast and thermal accelerator-driven systems, with no breeding of fissile material from fertile material such as ^{238}U or ^{232}Th spectrum systems ($\beta = 0$), is given in Fig. 1. The inventory is displayed for a system transmuting the waste from two light water reactors at the rate at which it is produced (about 300 kg per year per reactor). This waste production rate of 600 kg/year requires a transmuter fission power of about 1500 MWth. The isotopes are presented in order of increasing mass except for ^{241}Am which is shown at the right-hand edge of the figure. Several points are worth noting:

1. The thermal spectrum inventory is about two orders of magnitude smaller for all of the Np, Pu, and Am isotopes.

2. There is proportionately more of the higher actinides compared with plutonium isotopes in the thermal spectrum system indicating the advantages of the fast spectrum in reducing higher actinide burn-up.

3. Because of much smaller inventory of the lighter actinides in the thermal spectrum, the absolute amounts of curium isotopes in each system are similar and are small.

4. Inventory is proportional to $\phi\sigma$ and equilibrium time is proportional to $1/\phi\sigma$; the time to reach equilibrium is therefore very much longer for the fast than for the approximately four years required for the thermal spectrum system.

There is a general impression that transmutation increases the radioactivity of the feed material whether a fast or thermal spectrum is employed. This issue is examined in Fig. 2 where the alpha decay heat is compared for a 3000-MWth LWR. On the right-hand side of the figure is shown the alpha radioactivity decay heat at the end of 40 years of operation. On the left side is shown the equilibrium decay heat from the same system. Note that this decay power does not increase no matter how long the LWR or its follow-on system operates. In spite of the build up of the curium species, the alpha decay heat in the accelerator-driven system remains at about the level of the untransmuted waste. The equilibrium decay power for the fast spectrum is about one order of magnitude higher owing to the larger inventory. Therefore the fast spectrum does increase the equilibrium radioactivity while the thermal spectrum system does not.

We have seen already that commercial plutonium is easier to use than weapons plutonium in entry level weapons production. The possibility obviously exists for production in the transmutation process of other weapons-useful material. To gage this issue, weapons-relevant nuclear properties of weapons plutonium (w-Pu) and commercial plutonium (c-Pu) are compared in Table 3 with equilibrium isotopic distributions of Cm, Am, Pu and Np. The fissility parameter is the product of the fission cross section σ_f and the number of neutrons per fission ν. The amount of material for a nuclear explosion is expected to depend on this factor raised to some power because of the chain reaction nature of the energy generation. The National Academy of Sciences mentions that the mass requirement for nuclear weapon from ^{235}U is about three times higher compared to ^{239}Pu. Using the fissility parameter of 2.94 for ^{235}U and 4.85 for ^{239}Pu gives a mass requirement dependent on the square of the fissility parameter.

We have already discussed the problem of spontaneous fission neutrons introducing significant uncertainties in the weapon nuclear yield for commercial plutonium and reducing the yield very significantly for much larger spontaneous fission rates. The spontaneous fission rate for w-Pu, c-Pu, and the other isotropic mixtures are also compared in column four of the table. The alpha decay heat is another factor in the design of a nuclear weapon and the power in watts per kilogram is given in the last column. The amounts of the actinide isotopic mixtures carried inside of the transmuters after equilibrium is given in the second column. Parameters in the table which fall into a range significantly different from weapons-useful w-Pu and c-Pu are underlined. It is useful to examine each mixture individually.

Thermal spectrum Pu The spontaneous fission rate is five times higher than that for c-Pu. Since the rate for c-Pu is four times higher than for w-Pu and this higher rate significantly affects the weapon performance, it is reasonable to assume that a rate five times higher than c-Pu would preclude the use of this material in weapons. However the decay heat also is higher by a factor of three than c-Pu suggesting even more problems.

Fast spectrum Pu. This material exhibits the same disqualifications as potential weapons material as the thermal spectrum plutonium.

Thermal spectrum Am. The fissility parameter is essentially the same as for w-Pu, the spontaneous fission rate is lower by a factor of 100, and the decay heat is comparable to c-Pu. This material which is primarily 243Am appears to be useful in weapons. Fortunately there is little of it.

Fast spectrum Am. The decay heat appears to be a problem being about seven times higher than for c-Pu.

Thermal spectrum Np. The parameters indicate that the material is satisfactory for weapons use. Fortunately there is little of it.

Fast spectrum Np. This material appears to be satisfactory and there is a lot of it.

Curium. Both thermal and fast spectrum curium appears to be impossible to use because of both spontaneous fission rate and alpha decay heat.

The results of the analysis of Table 3 are given in Figure 3 where the amount of weapons-useful material in the transmuter per 3000-MWth LWR is compared for the thermal and fast spectrum systems in equilibrium. The thermal spectrum system contains about 38 kg compared with the fast spectrum inventory of 805 kg. If the LWR is operated to the end of life (40 years) and the fuel stored, the weapons-useful stored inventory is 12,000 kg. This stored inventory of course increases linearly indefinitely with time whereas the equilibrium inventories do not change. The advantage of transmutation therefore grows with time for either spectrum. The thermal system allows indefinite operation of an LWR with potential for nuclear weapons material sufficient for only a few nuclear weapons.

At the bottom of Fig. 3 we indicate the starting inventory of the thermal and fast spectrum systems. This starting inventory is all weapons-useful material. Between start-up and equilibrium, the thermal spectrum inventory of weapons-useful material decreases somewhat; the fast spectrum inventory increases by about a factor of four. While a complete dynamic calculation is possible and useful, the relatively small change between the initial and equilibrium inventories suggests that a dynamic calculation will not influence much the relative amounts of weapons-useful material in the thermal and fast spectrum systems at any time on the way to equilibrium.

To summarize, the fast spectrum system is inferior by about a factor of 100 to the thermal spectrum system in every important comparison. Even if the thermal spectrum operating parameters cannot be achieved and those of the fast spectrum can be exceeded, factors of ten would be required in both systems to make the fast spectrum system comparable in performance. Thus the apparent advantages of the fast spectrum system of better burn-up of higher actinides and better neutron economy evaporate in the face of the factor of 100 inventory advantage of the thermal spectrum system. It should also be noted that the neutron economy advantage of the fast spectrum is so large that casual examination indicates that the accelerator contribution to the neutron economy is not necessary; that is that a reactor could transmute the waste about equally well with however the same large inventory disadvantages. On the other hand, it is well known that the thermal spectrum system will not operate without the supplement of the accelerator-produced neutrons. The accelerator therefore *makes possible* the thermal spectrum transmutation, but the accelerator is *not essential* for fast spectrum transmutation. In view of this factor and the superior performance of the thermal spectrum system, there seems to be little justification for the study of fast spectrum accelerator-driven systems for pluonium and minor actinide burning until the thermal spectrum system have been thoroughly evaluated and found wanting.

Efficient use of accelerator-produced neutrons

While accelerators can provide supplemental neutrons to those normally present in a reactor, these neutrons are more expensive to produce than those from a reactor and so they must be used with the greatest effectiveness. We show below that the best use of these neutrons is to drive subcritical fission chains and or to use the neutrons to overcome situations where the short-fall in neutrons from reactors is small.

Supplementing the neutrons from fission.

Consider a system in which 100 fissions take place and in which the fission heat is converted to electric energy with a conversion efficiency of 40 %. Assume that all of this electric energy is fed back to the accelerator which operates with a bussbar-to-beam efficiency of 45 %. For 200 MeV of energy release per fission, the number of 1-GeV protons which can be accelerated is

$100 \text{ X } 200 \text{ X } 10^6 \text{ X } 0.4 \text{ X } 0.45 / 10^9 = 3.6$ protons of 1-GeV energy.

If each of these 1-GeV protons directed at a lead target produced 30 neutrons, the total neutrons produced from the fission heat of the 100 fissions would be $30 \text{ X } 3.6 = 108$. However each fission of ^{235}U which produced the electric energy to power the accelerator also produced 2.43 fission neutrons so that the 100 fission events produces $2.43 \text{ X } 100 =$ 243 neutrons. Therefore the neutron production using the accelerator provided an enhancement of 108 neutrons over the 243 which would otherwise be present from fission alone...an increase of 44 % or an increase in ν from 2.43 to 3.51. This is a remarkable increase of a full neutron per fission and it is now possible with existing accelerators.

Cost of the accelerator-produced and reactor-produced neutrons
Since accelerator produced neutrons and fission neutrons are produced by different means and at different locations in an accelerator-driven system, they can in principle be used for different functions. Some functions are more valuable or essential than others. It might make no difference how these neutrons were used if the cost of both accelerator and fission neutrons were the same. However if the production cost is significantly different, the more expensive neutrons should be used for the more highly valued function. We show next that the accelerator-produced neutrons are much more expensive than neutrons from fission.

The cost of accelerator-produced neutrons is fairly well known from the Accelerator-Production of Tritium (APT) program in the U. S. The capital cost of the 100-mA 1.3-GeV APT accelerator is estimated to be between \$2.5-3 billion. The cost of retiring this investment over a 30-year period is about \$200 million per year. The accelerator will produce 2 kilograms of tritium or 666 moles of atomic tritium operating at about 75% on-time. If superconducting technology is used, the bussbar-to-beam efficiency will be about 46 %. At a cost of \$0.05 per KWh, the annual power cost will be

$\pi \text{ X } 10^7 \text{ X } 1.3 \text{ X } 10^9 \text{ X } .05 \text{ X } 0.75 / (3.6 \text{ X } 10^6 \text{ X } 0.46) =$ \$92 million per year.

The operation cost of the facility is probably about half of that of a commercial power reactor or about \$40 million per year. The total costs are therefore approximately:

Principle and interest	\$200 million
Power	90
Operations	40
Total	\$330 million

There is little additional cost for converting the neutrons produced to tritium so taking the cost per mole of tritium and neutrons to be nearly the same, we arrive at a cost of the neutrons of (\$330 million per year) / (666 moles per year) = \$0.5 million/mole. While this cost is based on one-of-a-kind considerations and the cost could decrease considerably, the size of the accelerator is much larger than probably would be used for transmutation and so

some economy-of-scale influences would be lost for the accelerator used for transmutation. Therefore this cost for accelerator-produced neutrons might be realistic for the foreseeable future.

The cost of excess neutrons produced in a reactor burning HEU or plutonium can be roughly estimated by comparison with today's LWRs. If current reactors pay their way, the total cost of their operation is given by the buss-bar cost of the electric power sold. Dividing the number of excess neutrons which might be produced in such a system by the cost will give the cost per mole for the production of excess neutrons from fission. The parameters of a typical LWR are as follows:

Fission thermal power	3000 MWth
Electric power	1100 MWe
Energy per fission	200 MeV
Bussbar price of power sold	\$0.05 per KWh
Availability factor	0.75

These parameters give a fission rate $R_f = 9.4 \text{ X } 10^{19}$ fission per second. The number of useful neutrons U from such a system if 13 % of the neutrons are lost to the control rods, to leakage and to parasitic capture and if no fissile material is produced in the system is given by

$$U = \nu R_f[1 - (1 + \alpha)/\nu - 0.13] \qquad (2)$$

where ν is the number of neutrons per fission (2.43 for ^{235}U) and α is the capture-to-fission ratio (0.169). The number of useful neutrons is found to be 3500 moles per year. The value of the power sold is \$ 360 million per year. The cost per mole for the reactor neutrons therefore is \$360 million per year/ 3500 moles per year = \$100,000 per mole. The accelerator-produced neutrons are therefore about five times as expensive as fission neutrons.

Since the neutrons are so much more expensive from the accelerator than from fission, in an accelerator-driven subcritical system, *the accelerator-produced neutrons should be used only to initiate fission chains and the fission neutrons should be used for transmutation....not the accelerator-produced neutrons.* This holds unless the amount of material to be transmuted is small and is of such high value that the use of accelerator-produced neutrons is justified.

Calculating the number of useful neutrons for a subcritical system
If accelerator neutrons are only to be used to start fission chains in subcritical systems, then it is important to know how many useful neutrons are produced per accelerator neutron. We begin by calculating the number of neutrons in a subcritical system assuming initially that all of the neutrons are absorbed in fissile material. This assumption will be adjusted later. If the material is ^{233}U, then $\nu = 2.49$, $\alpha = 0.0861$, and the number of fissions we produce from 100 neutrons is $100/(1 + \alpha) = 92.1$ fissions. The number of neutrons produced is 92.1 X 2.49 = 229.3. Some of these neutrons leak away, some are parasitically captured, and some transmute waste. Some also produce fission and for $k_{eff} = 0.95$ the number of neutrons present is 95 % of those which were present in the previous generation. The number in this next generation is therefore 0.95 X 229.3 = 217.8 neutrons. These neutrons came from $217.8/\nu = 217.8/2.49 = 87.4$ fissions. The fissions consumed $87.4 \text{ X } (1 + \alpha) = 94.9$ neutrons.

We know that for a molten-salt graphite-moderated reactor, 13 % of the neutrons produced from fission are lost to leakage, parasitic capture, and to the control rods. If half of these are lost in control rods, then only 6.5 % are actually lost from a subcritical system without control rods. This number is 92.1 X 2.49 X 0.065 = 14.9 neutrons. The neutrons left over for useful work from the second fission generation is therefore 229.3 - 94.9 - 14.9 = 119.5 neutrons produced by the absorption on ^{233}U of the original 100 acelerator-produced neutrons. They must all be absorbed in useful work or by other non-fission means or the system k_{eff} will be higher than 0.95.

For the third fission generation, the number of neutrons at the beginning must be lower by 0.95 so that this number is found to be 0.95 X 217.8 = 206.9. Using the same analysis as for the second generation, we find the number of fissions is 83.1, the number of neutrons to induce these fissions is 90.25, and the number of neutrons lost is 14.2, and the number of useful neutrons remaining is 102.5.

These numbers are summarized below:

Fission generation	1	2	3
Neutrons for fission	100	94.9	90.25
Fissions	92.1	87.4	83.1
Fission neutrons	229.3	217.8	206.9
Neutrons lost		14.9	14.2
Useful neutrons		108	102.5

By inference one finds that the number of useful neutrons U from a source S may be written as

$$U = S[\nu(1-L) - 1]\,[k_{eff}/(1-k_{eff})]/\,(1 + \alpha) \qquad (3)$$

where L is the percent lost to parasitic capture and to leakage. However as the source neutrons also can be lost to parasitic capture etc., the number of available source neutrons must be reduced by the factor (1 - L) so that the final equation is

$$U = S\,(1-L)[\nu(\,1 - L) - 1][k_{eff}/(1-k_{eff})]/(1 + \alpha). \qquad (4)$$

Using a similar process we find that the number of fissions produced N_f is

$$N_f = [S/(1 + \alpha)][(1-L)/(1 - k_{eff})] \qquad (5)$$

If we add in breeding at the same rate as the fuel is burned in the system ($\beta = 1$), the equation becomes

$$U = S(1-L)[\nu(1-L) - 1 - (1 + \alpha)/k_{eff}]\,[k_{eff}/(1 - k_{eff})]/(1 + \alpha) \qquad (6)$$

Equations 4 and 5 can be used to calculate the fissions and useful neutrons per source neutron for $k_{eff} = 0.95$. The number of fissions calculated from Eq. 5 if all neutrons are absorbed on a thermally fissile nucleus is given in the first row for three fissile materials.

Using Eq. 4 the number of useful neutrons is shown in the second row for a non-breeding system and for special use of the accelerator neutrons to start a fission chain. We see that for $k_{eff} = 0.95$ (presumed multiplication of 20) that the number of useful neutrons is actually somewhat higher at 23-24 for ^{233}U and ^{239}Pu, but about 20 % lower for ^{235}U. If care is not taken to assure that the source neutrons are absorbed on fissile material, then those which are absorbed or lost elsewhere cannot initiate fission chains. For a system near $k_{eff} = 1$, no special use for the source neutrons means that only $1/\nu$ of these initiate fission chains. The number of useful neutrons is then reduced by this factor $1/\nu$ but increased by the accelerator neutrons spent otherwise $(1-1/\nu)$. The total number of useful neutrons per source neutron when the source neutrons are not treated specially is therefore $[(U/S)/\nu] + (1-1/\nu) = (U/S + \nu - 1)/\nu$. This number is given in the third row of the table above. The bottom row shows the advantage in terms of useful neutrons in reserving the accelerator-produced for intitating fission chains.

	^{233}U	^{235}U	^{239}Pu
N_f/S	17.21	15.99	13.75
U/S (source neutrons special)	23.23	19.3	23.65
U/S (source neutrons not special)\	9.88	8.53	8.86
Ratio (special)/(not special)	2.35	2.26	2.67

If all of the source neutrons are absorbed on fissile nuclides, the accelerator source (beam power) may be reduced by a factor of about ν or by a factor of about 2.4. It seems that this advantage has not been incorporated into any accelerator-driven transmutation system designs published in the literature thus far. The reduction in the accelerator size by a factor of two or more by this means, which is probably possible under practical engineering considerations, could have a significant impact on cost competitiveness of accelerator-driven transmutation technology.

One may define an accelerator effectiveness factor Γ for a target-blanket design as

$$\Gamma = \frac{\text{neutrons which start a fission chain}}{\begin{array}{l}\text{neutrons which start a fission chain +}\\ \text{neutrons which parasitically capture on structural materials +}\\ \text{neutrons which are captured in actinides without fission +}\\ \text{neutrons which transmute fission products +}\\ \text{neutrons which breed new fuel +}\\ \text{neutrons which escape}\end{array}}$$

The parameter Γ therefore varies between 0 and 1. Optimal designs for accelerator-driven systems will have Γ nearly equal to 1. Γ will be about 0.4 for reactors and for accelerator-driven systems for which the fission and accelerator-produced neutrons are used in the same way.

Possible sites for ADTT demonstration

There are a number of sites in the world where accelerators already exist or might soon exist where accelerator-driven transmutation might be demonstrated. Some of these sites are listed below along with the beam characteristics, the number of neutrons per proton on a lead target n/p, the number of neutrons per second n/s, and the fission power assuming a Γ factor of 1 and $k_{eff} = 0.95$.

Location		Current (mA)	Energy (MeV)	Status	n/p	n/s	Fission (MWt)
Europe							
	PSI	1.3	590	Exists	13	1.0×10^{17}	54
	Rubbia (injector)	10	120	Planned	0.75	4.5×10^{16}	24
Japan							
	KEK	5	1300	Planned	44	1.3×10^{18}	700
U. S.							
	LANSCE	1	800	Exists	22	1.3×10^{17}	70
	APT	100	1300	Planned	44	2.6×10^{18}	14,000
		100	1700	Planned	62	3.7×10^{18}	19,000
Russia							
	Troitsk	0.1	460	Exists	10	6.0×10^{15}	3
Unspecified site							
	IBA Corp.	2	160	--------	1.1	1.4×10^{16}	8

For Europe there are at least two prospects. The first at the PSI in Switzerland has been operating for almost two decades and could drive a system at 54 MWth. However the physical arrangement of the site may preclude an ADTT demonstration. The second is the injector cyclotron for the 10-mA 1-GeV facility proposed by Rubbia. The status of this proposal is currently undecided. The purpose in considering the injector is to indicate that the injector cyclotron alone is an interesting neutron source since it can drive a 24 MWth system. Japan is planning a general purpose spallation source and ADTT demonstration is part of the program. At the 700-MWth power level, it has been shown already in this paper that this system could transmute the waste for one 3000-MWth LWR.

There are two possible options in the U. S. The LANSCE accelerator at Los Alamos has operated for over 20 years at its design beam power. The original role of meson physics has been completed and the facility has been dedicated to neutron science. The new program leaves open the possibility for a major ADTT experiment at a power of 70 MWth. The LANSCE accelerator beam intensity probably can be upgraded by about a factor of two in current with an expenditure of about $20-30 million. The APT accelerator for accelerator production of tritium is an accelerator of enormous power. The lower power version has the capability to burn the waste from 23 reactors. In doing so it would feed into one point on the commercial grid a power of about 7 GWe which is a far larger feed at one point than anywhere now in the U. S.

The Russian version of LANSCE is the Moscow Meson Factory at Troitsk which also has been redirected to neutron science. In spite of its relatively modest parameters, the fission power is sufficient for interesting work. The last entry in the table refers to a cyclotron with 2 mA of beam at 160 MeV said to be available from the Ion Beam Accelerator Corporation in Belgium at a price of $22 million. This price is likely to be less than required to build a several megawatt target-blanket and therefore is attractive. Since the time for construction of the accelerator and for construction of a target-blanket are about the same, and since no target-blanket exists at present, it would be possible to purchase such an accelerator soon and to begin construction of a target-blanket and bring up an experiment as fast as it can be done anywhere.

There is a special niche for cyclotrons opened up by recognition of the importance of the G factor. At a recent meeting at Los Alamos on the prospects for building high power cyclotrons, it was concluded that 10 mA at 1 GeV represented a significant technical challenge but one that was likely achievable. To reach 15 mA at the same energy appeared to be unlikely. It seems likely therefore that a 7-mA 1-GeV accelerator can be built which is of sufficient size for the destruction of waste from a single 3000-MWth LWR. Such an accelerator is compact and a transmutation system of this kind could be accommodated on virtually every LWR site. Because of the length requirement of the linac, many sites could not accommodate a linac of this size nor would the linac be as economically practical as the cyclotron. Presently the likelyhood that this waste will not be moved from reactor sites in the U. S. raises the possibility that the waste might be destroyed on site by these cyclotron-driven systems

Summary
The role of accelerator-driven transmutation technology is examined in the context of the destruction of actinide waste from commercial light water reactors in fast and thermal spectrum systems. The reference fast (200 keV) spectrum used in the study is a 2×10^{15} n/cm^2-s system with fixed fuel in the system and with two extra loadings external to the system to accommodate the fuel destruction, chemical separations, and fuel fabrication. The average effective flux on the fuel is then a factor of three lower or 6.66×10^{14}. While advanced systems might be considered employing higher flux, shorter separations time, and no fuel fabrication, the present fast spectrum technology is the result of 20 years of work probably costing $20 billion or more. It seems reasonable to use for comparison the systems which have evolved from these large expenditures. The thermal spectrum system was a molten-salt liquid fueled system with a thermal (0.1 eV) flux of 1×10^{15} with the expectation that the fuel would be spending half of its time in the heat exchangers. The average thermal flux therefore is 5×10^{14}. This flux seems practical since the CANDU reactors have operated for many years at a flux of 6×10^{14} n/cm^2-s and at a lower neutron temperature. It is assumed that plutonium is burned in conjunction with the minor actinides.

The total inventory of plutonium and minor actinides carried in the fast flux is found to be 100 times larger than in the thermal system. Even though the thermal spectrum system inventory carries a larger *fraction* of curium than the fast spectrum, the *absolute amounts* in the two systems are about the same.

Arguments are presented indicating that commercial plutonium is preferable to weapons plutonium for development of a nuclear arsenal. The properties of the commercial plutonium is evaluated to determine what components of the Np, Am, and Cm inventories are of potential weapons use. The results of this are that the thermal spectrum system carries far less weapons-useful material....so little as to be uninteresting to one considering

removal of the inventory of these systems for malevolent purposes. The opposite conclusion is reached for the fast spectrum system whether a reactor or an accelerator is used for waste destruction.

The neutron economy is shown to be substantially better for the fast spectrum system than the thermal spectrum system but this advantage is not a determining factor. In a practical fast spectrum system approximately 0.4 neutrons per fission (14 %) are allowed to leak away[20]. This is substantially more than the neutrons provided by the accelerator for a system with $k_{eff} = 0.95$. From this perspective, the accelerator neutron enhancement is redundant; the neutrons are unnecessary except for enabling subcritical operation.

The thermal spectrum systems offer the advantage of much smaller inventory, but the neutron economy is insufficient for the thermal spectrum to be implemented as a practical thermal reactor. The accelerator-supplied neutrons are therefore vital for enabling functions in an accelerator-driven thermal system which are impossible to achieve in a thermal reactor (or in a fast reactor). Therefore the presence of the accelerator serves a vital function from the neutron economy perspective for the thermal spectrum system whereas for the fast spectrum it is unnecessary.

A thermal spectrum system therefore can be built at a fission power sufficient to destroy the waste from one 3000-MWth LWR using 5 to 7 mA of protons at 1 GeV. The thermal spectrum system therefore can be built to require about half as many neutrons from the accelerator as the fast spectrum systems for the same fission power. It is highly likely that a cyclotron can be built which can deliver at 1 GeV 10 mA of beam. But it is unlikely that a 1-GeV linac can be built to operate at 10 mA with capital cost and efficiency competitive with the cyclotron. Obviously the footprint of the cyclotron could fit on almost any commercial reactor site whereas the footprint of the linac would not fit on many sites. Therefore the destruction of the waste on the reactor site might be practical for the thermal spectrum using a cyclotron driver but impractical for the fast spectrum system which requires a linac driver.

There might be a niche for fast spectrum systems for minor actinide burning if fast and thermal reactors can successfully eliminate the plutonium. But the inventory of plutonium which must be carried by this array of reactors is too large not to cause concern. A 750-MWth fission system driven by a small accelerator and sited beside each LWR would reduce the weapons material inventory to a level of minimal concern and would allow present LWRs or advanced LWRs to operate indefinitely at the site without a build-up of weapons useful material.

Since commercial plutonium is easier to use, since there is very much more of it already, and since it is growing rapidly, the permanent disposition of commercial plutonium is an issue perhaps of greater importance than weapons plutonium.

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13. The receipt of public input on the report, "Storage and Disposition of Weapons-Usable Fissile Materials Draft Programmatic Environmental Impact Statement" was completed on May 7, 1996. The document may be obtained from U. S Department of Energy, Office of Fissile Materials Disposition, P. O. Box 23786, Washington, DC 20026-3786

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19. The first meeting in this series was entitled, "International Conference on Accelerator-Driven Transmutation Technologies and Applications," Las Vegas, NV, AIP Conference Proceedings 346 (1994)

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Plutonium and Minor Actinide From One Year of LWR Operation* at 3000 MWth

Isotope	Mass (Kg/yr)
^{238}Pu	4.5
^{239}Pu	166.
^{240}Pu	76.7
^{241}Pu	25.4
^{242}Pu	15.5
^{237}Np	14.5
^{241}Am	16.6
^{243}Am	3.0
^{244}Cm	0.6

* After 10-year decay

Thermal and Fast Spectrum Transmutation Cross Sections

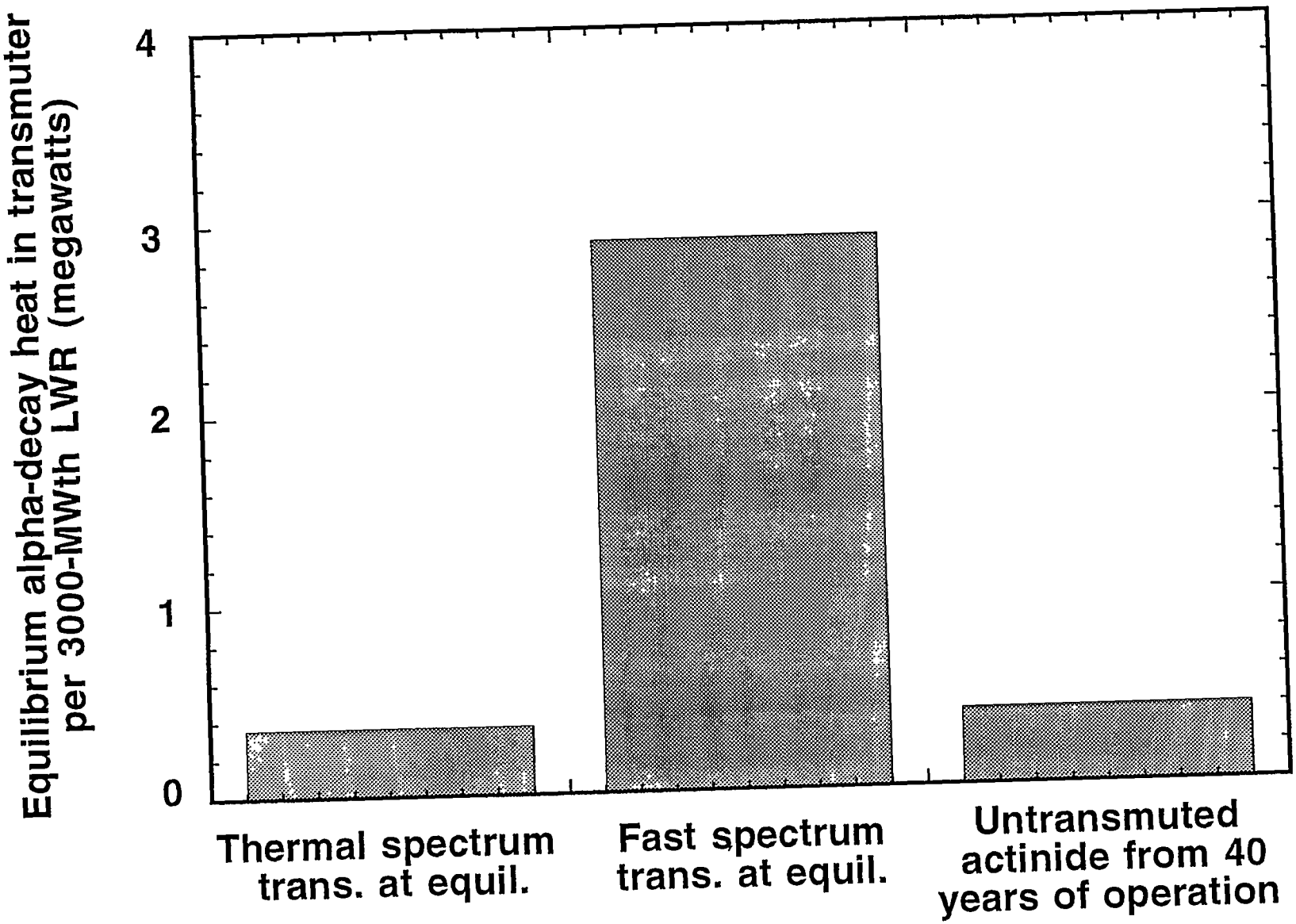
Isotope	Thermal*		Fast	
	Capture	Fission	Capture	Fission
^{237}Np	114	11.4	0.7	0.16
^{238}Pu	280	10.6	0.4	1.0
^{239}Pu	145	389	0.4	1.5
^{240}Pu	350	0.0	0.3	0.2
^{241}Pu	187	533	0.2	1.3
^{242}Pu	64.2	0.25	0.15	0.075
^{243}Am	130	0.1	0.6	0.07
^{244}Cm	40.1	1.15	0.5	0.35
^{245}Cm	189	1111	0.3	2.0
^{246}Cm	6.1	0.57	0.5	0.5
^{247}Cm	53	81	0.3	2.0
^{248}Cm	14.8	0.85	0.5	0.5
^{249}Cf	288	900	0.3	1.8
^{241}Am	375	2.3	1.0	0.13

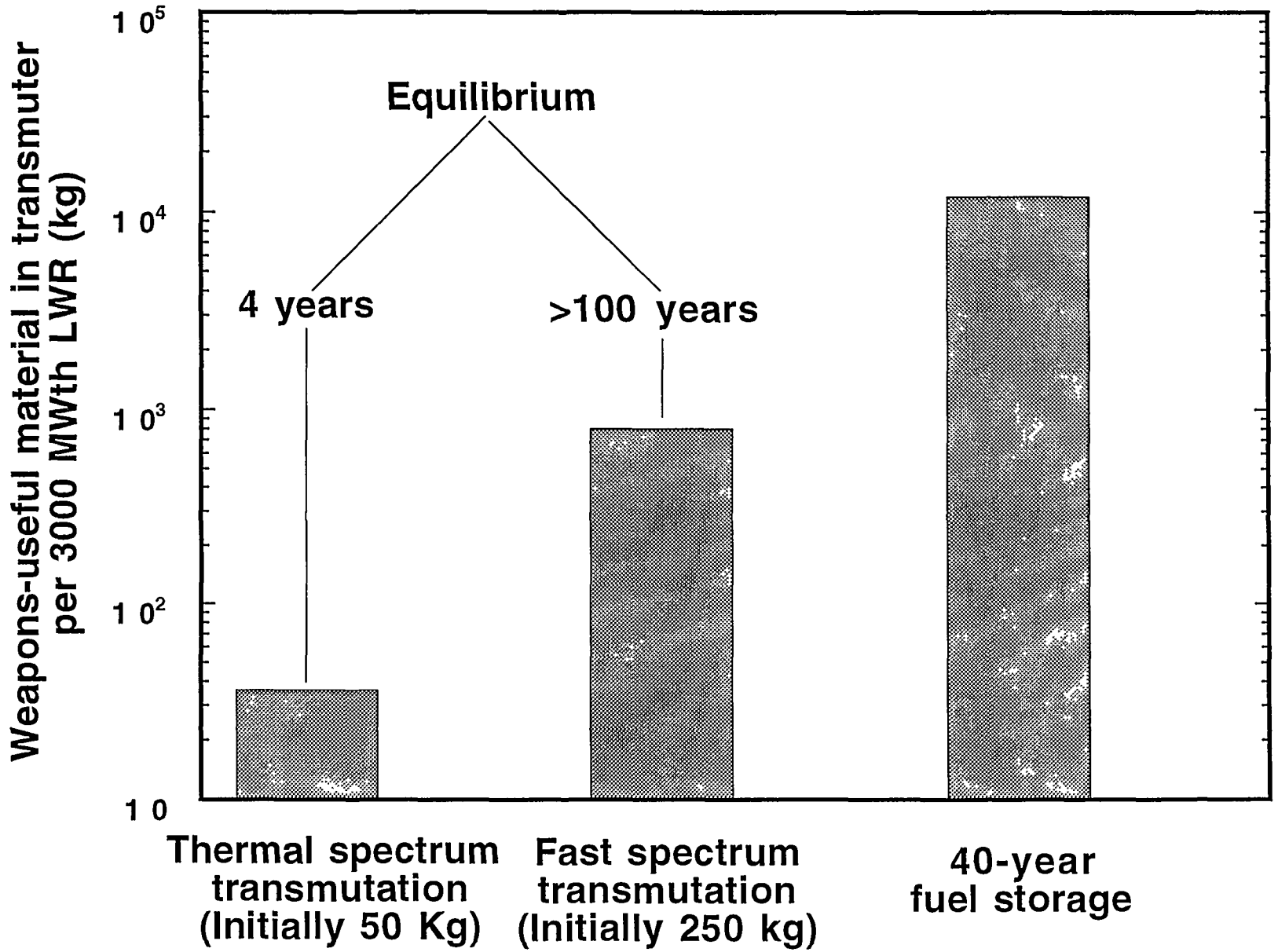
*These cross sections are the thermal cross sections reduced by a factor of two for 1000 Kelvin operating temperature and increased by 5 % of the resonance integral cross section.

Possible Weapons-Useful Material
(1500-MWth transmuter at equilibrium)

Material	Mass (kg/trans)	Fissility ($\nu\sigma_f$)	S. F. neutrons (fissions/s-kg)	Decay heat (watts/kg)
Weapons-Pu	--	4.85	2.8×10^4	1.8
Commercial-Pu	--	4.70	9.8×10^4	8.5
Pu (thermal)	242	4.53	5.0×10^5	26.9
Pu (fast)	46,400	5.42	6.3×10^5	103
Am(thermal)	63	4.81	2.9×10^2	10.2
Am (fast)	6627	4.81	4.2×10^2	57
Np (thermal)	14.6	4.10	<0.05	0.017
Np (fast)	1611	4.10	<0.05	0.017
Cm (thermal)	384	6.80	4.0×10^9	1900
Cm (fast)	628	6.80	4.0×10^9	1900

Equilibrium inventory (kg/1500 MWth)
10^5
10^4
10^3
10^2
10^1
10^0
Thermal (0.1 eV)
Fast (200 keV)
^{237}Np
^{238}Pu
^{239}Pu
^{240}Pu
^{241}Pu
^{242}Pu
^{243}Am
^{244}Cm
^{245}Cm
^{246}Cm
^{247}Cm
^{248}Cm
^{241}Am


Equilibrium alpha-decay heat in transmuter per 3000-MWth LWR (megawatts)
4
3
2
1
0
Thermal spectrum trans. at equil.
Fast spectrum trans. at equil.
Untransmuted actinide from 40 years of operation


Weapons-useful material in transmuter per 3000 MWth LWR (kg)
1 0^5
1 0^4
1 0^3
1 0^2
1 0
Equilibrium
4 years
>100 years
Thermal spectrum transmutation (Initially 50 Kg)
Fast spectrum transmutation (Initially 250 kg)
40-year fuel storage