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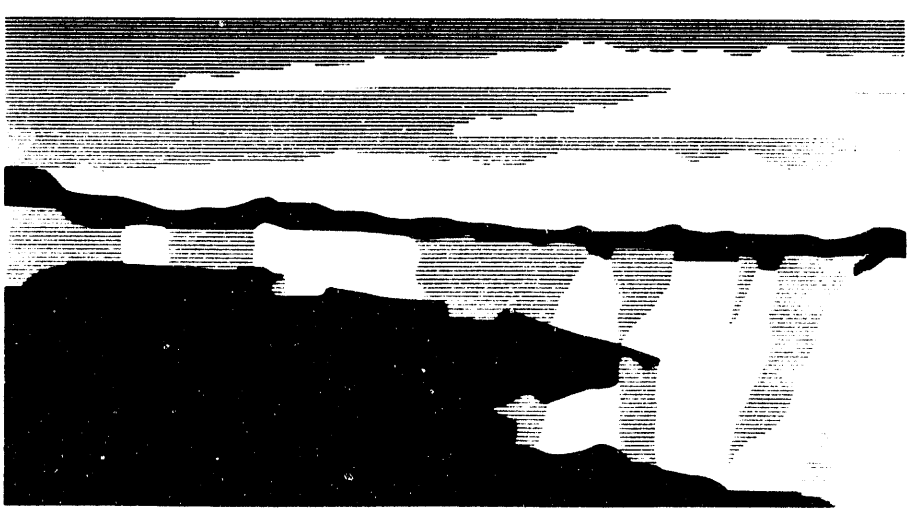

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COMPARISON OF ACCELERATOR-BASED WITH REACTOR-BASED WASTE TRANSMUTATION SCHEMES

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ABSTRACT

Accelerator-based transmutation of waste (ATW) systems for the destruction of commercial LWR spent fuel are compared with systems based on thermal reactors that accomplish the same objectives. When the same technology is assumed for the actinide-burning aspect of the two systems, it is seen that the size of the accelerator is determined only by the choice of how many of the long-lived fission products to burn. If none are transmuted, then the accelerator is not necessary. This result is independent of the choice of fluid carrier, and whether the actinides are destroyed in an ATW system or in a separate reactor.

Introduction

There is continuing interest in reducing risk from the disposal of commercial LWR waste, although it has been over 15 years since a major study concluded that there are "no insurmountable technological barriers to geological disposal" [1]. Accidental and intentional (to obtain weapons material) intrusion risks are associated with the transuranic elements, most notably plutonium. Although there is economic benefit from the sale of electricity generated by fissioning the plutonium in reactors, the cost of the fuel reprocessing may be greater than any economic gain. The justification for recycling and burning plutonium may lie in increasing the acceptability and decreasing the cost of a repository [2].

Accelerator-based transmutation of waste (ATW) systems would not only burn the plutonium, but all the higher actinides [3,4,5,6]. Reactors discussed here, which would also would be capable of doing this, would not be of conventional design, but would rather resemble the fluid-fueled concepts that were studied in the 1950's and 60's [7]. Thermal ATW systems offer the additional advantage of transmuting many of the long-lived fission products (LLFP's), such as ^{99}Tc [8]. A small repository would thus hold a large number of reactor-years of the remaining waste, without the legacy of leaving extremely hazardous waste unattended for millennia.

In this paper the differences between ATW and reactor based systems are discussed, with the aim of a fair comparison of the two systems assuming that similar technology is used in each. The results here are based on calculations performed with the established Monte Carlo neutron transport code MCNP [9].

Conceptual Target/Blanket Designs

Figure 1a shows one of many possible conceptual design for an ATW target/blanket assembly. The target will consist of a material that produces neutrons from either direct or spallation reactions. There will be in general an annulus of high-Z material, such as lead, for fuel

multiplication of the source through (n,xn) and (p,xn) reactions. The blanket contains a moderated heterogeneous lattice, using a flowing liquid which contains the actinides. Fission heat is carried away by convection and the liquid is cooled in an external heat exchanger. There are separate regions in the blanket where the LLFP's are transmuted.

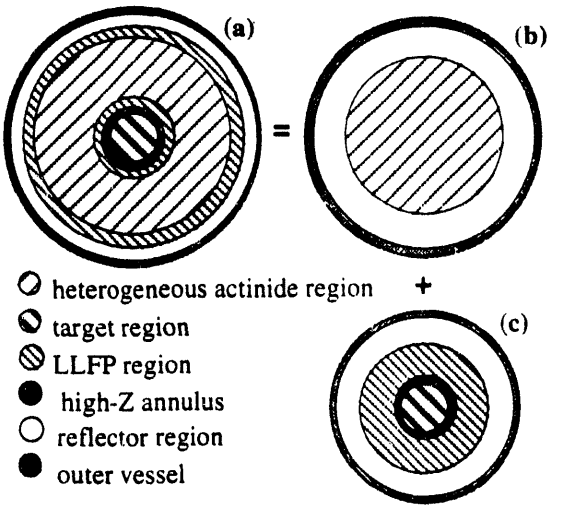


Figure 1. The concepts for target/blanket assemblies for ATW systems with and without the use of a reactor for actinide burning.
(a) the ATW system combining actinide transmutation and LLFP transmutation in the same blanket.
(b) actinide burning reactor that is based on the same heterogeneous lattice.
(c) the remaining LLFP-burning ATW blanket.

Fission products continually are produced in the slurry and must be removed for processing, or else they will become a very significant neutron poison for the system. The mean residence time of the constituents of the slurry in the target/blanket/heat exchanger before removal for chemical processing should be less than 15 days. The LLFP's produced by fission are removed by the chemical system and stored for later transmutation. Another chemistry system must be present to remove LLFP transmutation products (e. g. ^{100}Ru from transmutation of ^{99}Tc) .

The fluid-fueled actinide-burning reactor is shown in Figure 1b. Again, fission product concentrations must be kept to a minimum by continuous removal, requiring the use of liquid fuels. The prompt negative temperature coefficient of the liquid fuels eliminates the need for a resonance absorber. The same heterogeneous lattice is used, held in a critical configuration. The flowing liquid fuel is cooled in an external heat exchanger, and a chemical processing system is present to remove fission products. The outer wall serves as the pressure vessel.

The remaining ATW system, as shown in Figure 1c, then would burn the LLFP's only. The volumetric heat generation would be reduced by one or two orders of magnitude versus the combined actinide/LLFP ATW system. Chemistry systems to process and remove the transmutation products would still be required.

Neutron Balance

The neutron balance for either configuration is almost the same, provided that the reactor transmutes LLFP species with its reactivity margin. There may be some additional small effects due to changes in leakage. The actinide feed to the reactor or the ATW is half ^{239}Pu; therefore, initial criticality in the reactor is easily achieved. After ~10 years, the higher actinides build up in concentration to near the "equilibrium" values, resulting in a lowered reactivity. This process is illustrated in Figure 2, which shows the coupling between the species from neutron capture reactions and by α, β^-, EC decay.

To find the equilibrium concentrations for the actinide mixture, we solve a set of coupled linear algebraic equations implied by steady state conditions using a code TRANEQUI. The inputs

the code are the 1-group spectrum averaged cross sections for all the reactions, the decay constants and branching ratios, and the total flux.

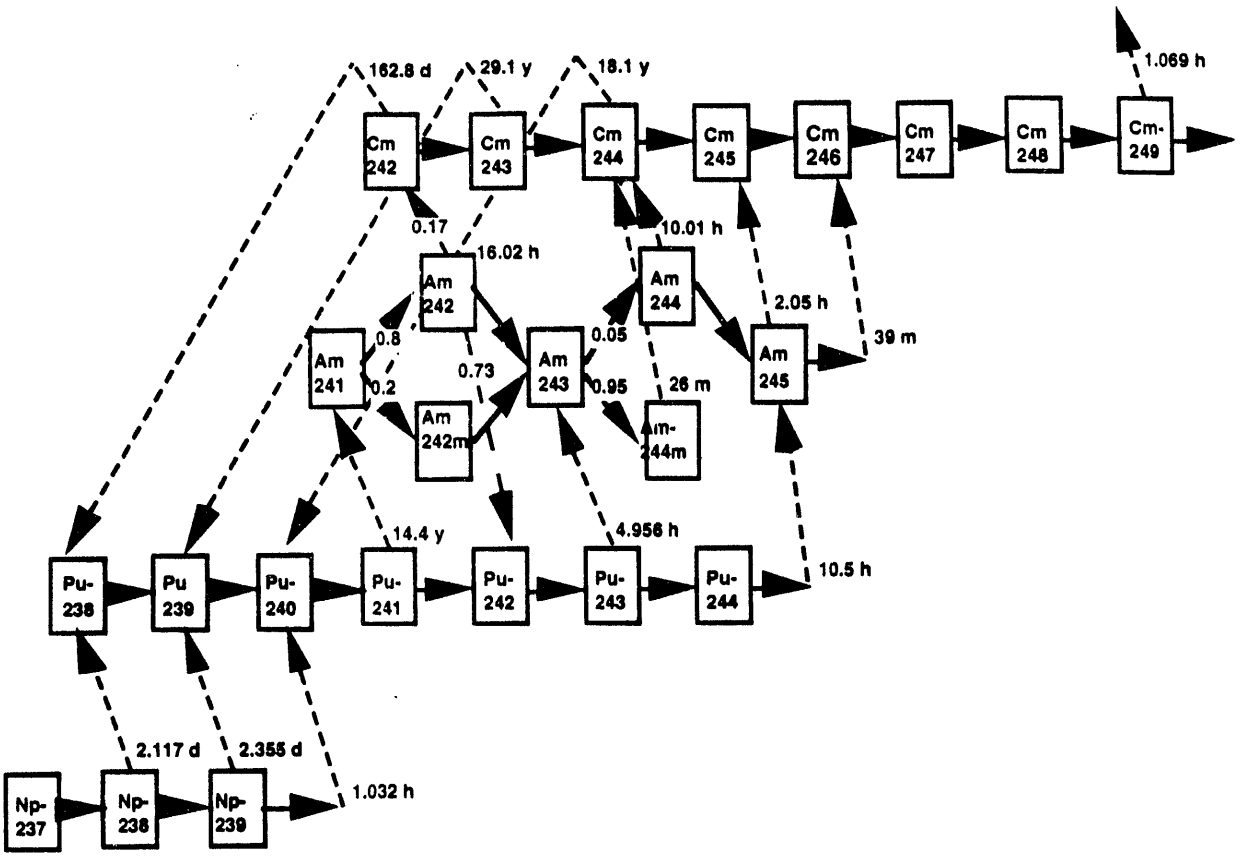


Figure 2. The set of 25 actinide species considered in the calculations for a commercial ATW system.

The unit cells in our MCNP calculations were those for a heavy-water moderated aqueous slurry system (Figure 3a) and a graphite moderated molten salt system (Figure 3b). The results presented here are the result of scoping studies, taking into account thermal/hydraulics limitations, chemistry requirements, etc.

In the aqueous system, the actinides are in the form of an oxide (500 g/l AcO_2) slurry in heavy water (1/4% H_2O impurity). The tubing that holds the slurry is made of high-temperature zirconium (97.5%) / niobium (2.5%) alloy, the same alloy used in CANDU systems. The inlet and outlet temperatures for the slurry as it travels along its 3 meter path in the tube are 275 C and 325 C, respectively. The slurry velocity is 12 m/s. The tubes are of a double-wall design, each wall thick enough to contain the 13.1 MPa operating pressure. The gap between the two walls acts as a thermal barrier. The simplest unit cell is a triangle as shown. The heavy water moderator outside the tubes is held at or near atmospheric pressure and 100 degrees C. Parasitic losses in this system are in the tubing, the H_2O impurity in the moderator and slurry, and in the entrained fission products in the slurry. The tubing losses increase with the operating pressure for the system. The system here operates at a pressure that gives a capability of 30% thermal/electrical conversion efficiency. A 50% holdup time in an external heat exchanger is assumed, and chemical processing residence times of 50% for Am/Cm, 25% for Np/Pu. These factors are important in determining the equivalent flux that the actinides see on average, in the input to the TRANEQUI code. The flux in the unit cell was determined to be 1.7×10^{15} n/cm^2/sec, based upon the slurry velocity, which in turn limits the power density.

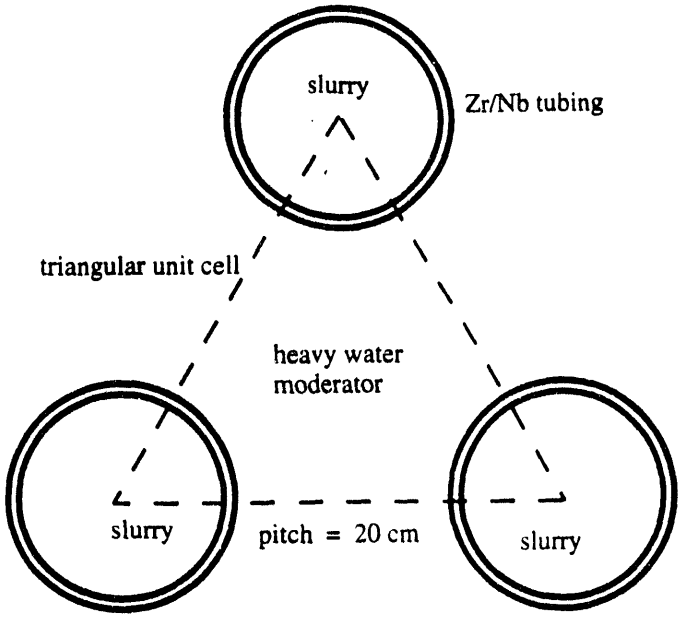
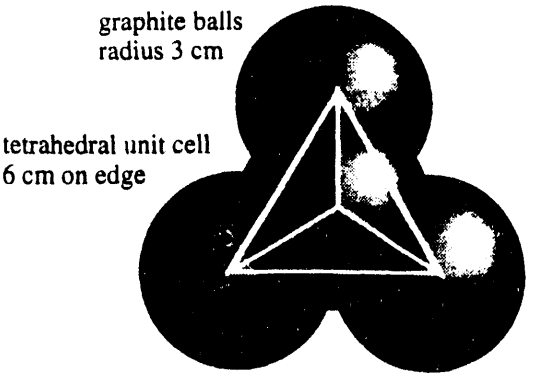


Figure 3 (a) Triangular unit cell for aqueous system.



(b) Tetrahedral unit cell for molten salt system.

In the molten salt system, the fuel is in the form of a fluoride salt (100 g/l AcF_4), dissolved in a molten salt carrier whose composition is 60% ^7LiF and 40% $^9BeF_2$. The carrier's melting point and operating temperature are 500 C and 650 C, respectively. The molten salt flows over the outside of a hexagonal close-packed set of high-purity graphite balls, of 3 cm radius. The simplest unit cell is a tetrahedron. In the selected design, the power density is about 200 MW/m^3 in the fuel, corresponding to a flux of $1.1x10^{15}$ $n/cm^2/sec$. No external holdup in a heat exchanger or in processing was assumed in the calculations. Xenon is removed continuously by a helium gas stripper. All other fission products are removed via centrifuge with a mean in-blanket residence time of 2 days.

The results of the calculations are summarized in Table 1. The eigenvalue is slightly higher in the molten salt system because reduced parasitic and fission product captures, even though α value is higher due to increased resonance capture in ^{239}Pu at the elevated temperature.

Table 1. Neutron Balance in unit cell calculations for aqueous and molten-salt concepts.

Quantity	symbol	value (aqueous)	value (molten salt)
fuel capture/fission	α	1.62	1.74
neutrons/fission	ν	3.05	3.06
neutrons/ fuel absorption	η	1.164	1.117
parasitic capture/fission	α_p	0.11	0.015
fission product capture/fission	α'	0.12	0.036
eigenvalue	k_{eff}	1.070	1.096

We have performed a simulation of an entire reactor similar to the system shown in Figure 1b, but because of space limitations, the details will not be presented here. In the aqueous case the tubing was made thinner because it is no longer a pressure boundary, and therefore the two systems obtained the same k_{eff} value. That value is 1.03. Either system can form a critical configuration throughout its lifetime, completely transmuting the LWR waste feed into fission products. There is a reactivity margin available to transmute LLFP's in these reactors, as well.

However, because of the presence of large quantities of higher actinides, the reactivity margin depends on the full utilization of the nominal in-core flux. In practical systems it may prove difficult to prevent flux dilution by holdup of the fuel in external components, e. g, heat exchangers or hold-up tanks.

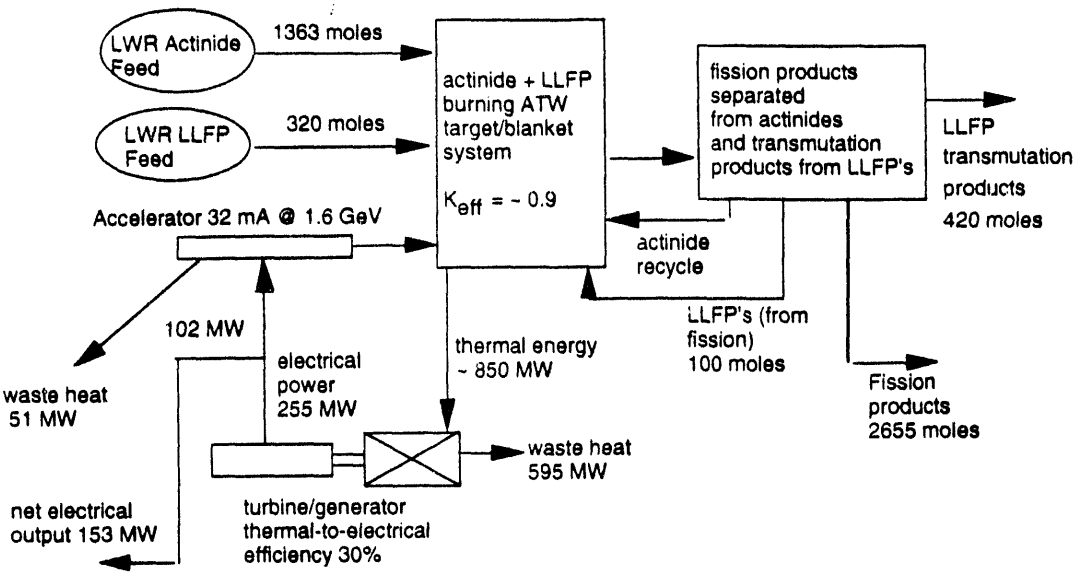
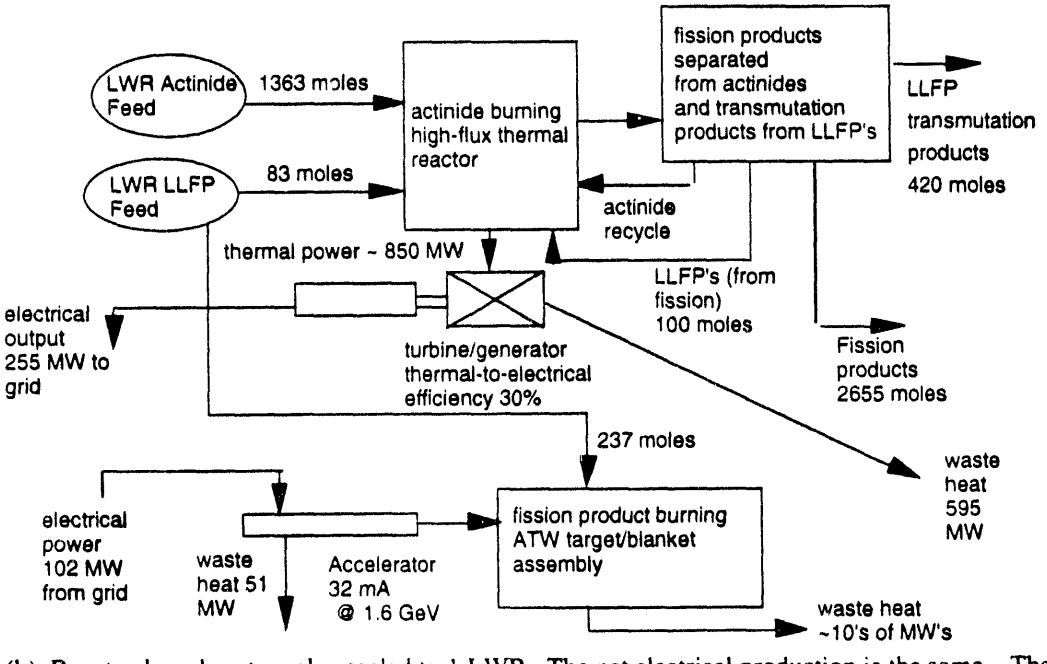


Figure 4. (a) Baseline aqueous ATW system scaled to 1 LWR worth of actinide and LLFP waste from commercial LWR spent fuel. The LLFP's burned are ^{99}Tc and ^{129}I only. No isotopic separation is assumed, therefore, the ^{127}I is burned with the ^{129}I.



(b) Reactor-based system also scaled to 1 LWR. The net electrical production is the same. The molten salt system is different in that the thermal/electric conversion efficiency is higher.

Mass and Energy Flows

The mass and power flows for an ATW system, normalized to a single 1 GWe LWR are shown in Figure 4a. In this specific example the flow rates correspond to the transmutation of the

actinides and ^{99}Tc and ^{129}I. There is no isotopic separation used for these balances, therefore ^{127}I is burned also. Heat produced is used to generate steam that runs turbine/generators. The accelerator uses less electricity than is made, and the surplus is sold to the grid. A small chemical facility removes LLFP's created by fission for recycle, and continuously recycles the actinides. Parasitic capture rates on target and structural materials have been taken from integrated conceptual ATW target/blanket designs [6].

A reactor-based system that accomplishes the same goals is shown in Figure 4b. The actinide-burning task is performed in a reactor that is fueled with the actinides. Electricity produced in this reactor is sold to the grid. Only a portion of the LLFP feed is transmuted with the actinides, and the rest is sent to an accelerator-based system for destruction by neutron capture. The LLFP's generated by the fission of the actinides are added to this stream. There is no electrical production in this portion of the system, and the heat generated is dumped. The accelerator buys alternating current (AC) electricity from the grid. The difference in the net energy streams between the two configurations is that the beam power is recycled to the thermal system in the first case. The beam power will be typically 5% of the actinide heating power, and so this loss is at most only a small fraction of the total power.

Power Requirements

The neutron balance of the ATW system is a function of the choices that are made for the burning of fission products. In both the aqueous and the non-aqueous case most of the neutrons used to transmute LLFP's are supplied by the accelerator, in either the configuration shown in Figure 1a or that shown in Figure 1b. The size of the accelerator is determined by the choice of how many of the LLFP's to burn. If none are transmuted, then the accelerator is not necessary.

Table 2. Long lived fission products (LLFP's) of interest in commercial LWR waste transmutation schemes.

species	isotopic moles per LWR-yr	elemental moles per LWR-yr	half life 1000's yrs	beta energy KeV	gamma energy KeV	elemental neutrons per atom	cross section barns
^{79}Se	2.5	23.8	65	160	0.0	1.2	1.5
^{93}Zr	257.5	1297.4	1500	60	0.0	2.1	2.1
^{99}Tc	259.5	259.5	210	293.0	0.0	1.0	17
^{107}Pd	67.9	427.3	6500	33.0	0.0	2.8	6.6
^{126}Sn	7.2	46.0	100	250.0	87.0	1.8	0.16
^{129}I	46.2	60.5	16000	150.0	39.0	1.0	15
^{135}Cs	74.1	588.1	3000	210.0	0.0	1.6	5.8

Table 2 lists the LLFP's produced in LWR's. All have low β^- decay energies. Some of the species have accompanying γ rays, also of low energy. Two different molar quantities are listed. One is just of the isotope itself and the second is for the element, including all isotopes. In the case of zirconium, the contribution from cladding is not included. Most species occur generally with several isotopes, as seen in Figure 5. Zirconium appears as 6 species, 90 through 96, not including the short lived 97. If isotope separation is not used, isotopes 90 through 94 must be transmuted to 95, which decays by two successive β^- emission to ^{95}Mo. The isotopes 90 through 93 thus require *more than* 1 neutron per transmutation. Isotopes 94 and 96 require each 1 neutron. The net average per atom is 2.1 neutrons per transmutation. These are the quantities listed in the last column in Table 2.

In Figures 6a and b the amount of electrical power needed to operate the accelerator is shown (on a per LWR basis) for each of the LLFP's, on an isotopic and elemental basis, respectively. In this

calculation, it is assumed that no LLFP transmutation is provided by a reactor. We assume a proton beam energy of 1 GeV. Based on computational experience, we select a reasonable number of number of neutrons liberated from the spallation target at 20 per proton. The efficiency for capturing the neutrons is 80%, and the capacity factor of the plant is 75%. The facility must buy the AC power from the electrical grid to operate the accelerator, and the conversion of that power into beam energy is taken to be 50%.[1]

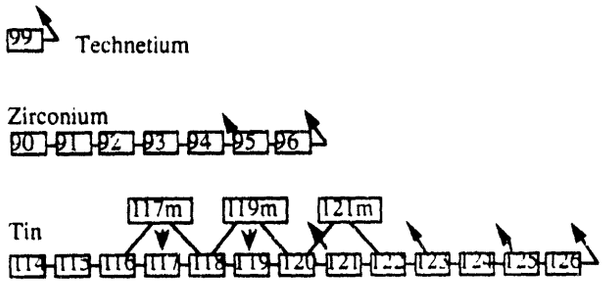


Figure 5. Transmutation schemes for some long lived fission product species. Upwards arrows represent β^- decay, which eliminates one atom of the element. Technetium is the only element of interest where a single neutron need be captured for transmutation. Others are more complicated, requiring more than 1 neutron per atom on average.

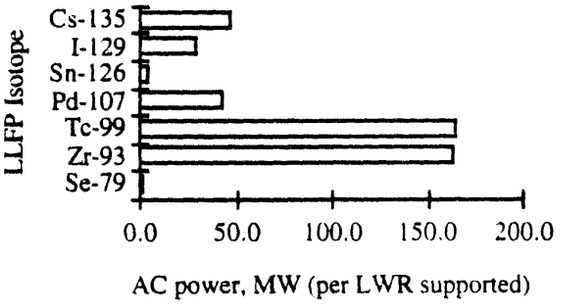
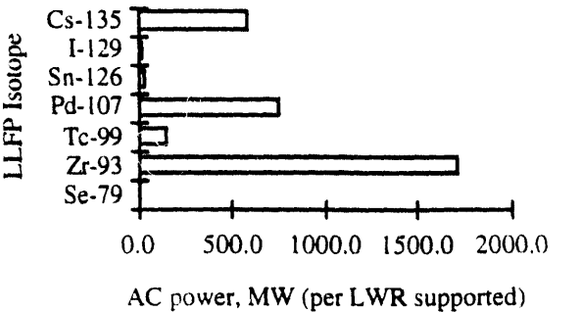


Figure 6 (a) The electrical power to operate the ATW accelerator for each LLFP isotope transmuted, when it is assumed that isotopic separation is used.



(b) The electrical power from the grid to operate the ATW accelerator for each LLFP isotope transmuted, when isotopic separation is *not* used.

Transmutation of the only species that appears as a single isotope (^{99}Tc) requires 160 MW of power, or 16% of the power of the LWR. When isotopic separation is allowed, the transmutation of all 7 isotopes would require 453 MW of power, or 45.3% of the electrical output of the LWR. The elemental ^{93}Zr would require more power than is produced in the LWR. Elemental transmutation is still expensive for ^{79}Se, ^{99}Tc, ^{126}Sn, and ^{129}I. The transmutation of all these combined would require 27% of the LWR's power.

The reactor in the system helps transmute LLFP's in two ways. Its reactivity margin is used to burn 0.13 moles of LLFP's per mole of actinides, saving the equivalent of 25 mA per LWR of beam current that would have been supplied by the accelerator facility. Thus, only 102 MW of AC power is needed for the accelerator to burn ^{99}Tc + I, rather than the larger quantities implied by Figure 6b. For additional species destroyed, the full amount of power shown in Figure 6 would be required. The actinide-burner reactor can supply 255 MW of AC power (for the 30% efficient aqueous system); the system would be a net producer of electricity unless all the species shown in Figure 3 are transmuted. The molten salt reactor, with its higher thermal/electric conversion efficiency, produces enough electricity to burn them all.

Conclusions

Complete elimination of all listed actinides and LLFP's can be achieved. In the absence of isotopic separation, however, the AC power required for most of the isotopes becomes much larger, and the transmutation of ^{93}Zr becomes prohibitively expensive. If isotopic separation is used and subsystem efficiencies are high, the actinide-burning portion of a transmutation system can supply a large fraction if not all of the electricity needed. If a liquid fuel thermal reactor is designed such that minimal dilution of the flux occurs in external regions, then using a target/blanket system to burn only fission products and a reactor to transmute actinides could be an attractive alternative to the single target-blanket solution.

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[1] The LAMPF facility at Los Alamos operates at an overall efficiency of about 4%, but recent advances in accelerator technology, such as the radiofrequency quadrupole, have given the accelerator community reason to believe that substantial improvements on this are possible.

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