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*ACCELERATOR - DRIVEN SUB-CRITICAL TARGET CONCEPT FOR TRANSMUTATION OF NUCLEAR WASTES

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ABSTRACT

A means of transmuting key long-lived nuclear wastes, primarily the minor actinides (Np, Am, Cm) and iodine, using a hybrid proton accelerator and sub-critical lattice, is proposed. By partitioning the components of the light water reactor (LWR) spent fuel and by transmuting key elements, such as the plutonium, the minor actinides, and a few of the long-lived fission products, some of the most significant challenges in building a waste repository can be substantially reduced. The proposed machine, based on the described PHOENIX Concept, would transmute the minor actinides and the iodine produced by 75 LWRs, and would generate usable electricity (beyond that required to run the large accelerator) of 850 MW_e.

1. INTRODUCTION

The PHOENIX Concept uses a large linear proton accelerator to drive and control one or more subcritical lattices of minor actinides (Np, Am, Cm), to transmute the long-lived radioactive wastes from light-water reactors that are the most difficult to dispose of, and to produce electric power in the process. One 3600 MW_t machine would transmute the neptunium, americium, curium, and much of the iodine produced by about 75 light water reactors (LWRs), and would generate a net of about 850 MW_e for the electrical grid, as indicated in Figure 1.

While not tied to a specific fuel reprocessing/recycling technology, much of the PHOENIX analysis performed thus far has been based on the proposed CURE approach (Ref. 1), which is a waste partitioning process based on the well known PUREX process and the newer TRUEX process. Within the CURE framework, certain elements are to be recycled, transmuted, or simply separated from the major portion of the high-level wastes. The primary objective is to eliminate certain problem components from the bulk of the spent fuel so that the remainder can be packaged more easily (reduced heat load and shorter life-time requirements) for disposal in the geologic repository currently planned by DOE.

The possible usage of hybrid accelerator - subcritical lattices for transmuting long-lived nuclear wastes has been discussed previously (Ref. 2 and 3). Because the minor actinides will fission quite efficiently in a very hard (fast) neutronic spectrum, there are various options to consider, including sodium, lead, or helium coolants and metal, oxide, or other fuel forms. For PHOENIX, we have made specific assumptions regarding the design of the accelerator and the composition of the subcritical lattices. These assumptions are quite modest regarding extensions of current technology, and the PHOENIX Concept described herein is considered credible.

The PHOENIX Concept assumes a large linear accelerator that can produce a 104 mA beam of 1.6 GeV protons. While such an accelerator is an extension of the present technology, an even larger machine producing 250 mA of 1.6 GeV protons was recently designed and evaluated for usage in a concept for producing tritium (Refs. 4 and 5).

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A multiple module (modular) concept was developed for the PHOENIX subcritical lattice. Each module resembles the core of the Fast Flux Test Facility (FFTF) (Ref. 6), with the minor actinides formed into oxide fuel rods, replacing the uranium and plutonium in the FFTF fuel. The fuel rods are cooled using liquid sodium, and are bundled into 217 pin assemblies, with 124 such assemblies making up a 450 MW_t target module. From 1 to 8 of these target modules are aligned in front of the proton beam, depending in part on how much of the "fuel" is available at any given time.

An alternative means of transmuting the minor actinides would be via reactors, most probably fast spectrum reactors since the minor actinides are strong absorbers of slow neutrons. In contrasting the two approaches, the reactor approach might appear at first to be simpler. However, the physics taking place within the PHOENIX proton-driven lattice is not much more complex than that in a reactor, as illustrated in Figure 2. The number of fissions induced is directly proportional to the number of neutrons released via the spallation and evaporation processes as the proton passes into the lattice. This number of neutrons released is roughly 50 for each 1.6 GeV proton in a minor actinide lattice of the size of the PHOENIX target modules. The number of fissions then depends on the k-effective for the lattice. Note that for a k-effective in the range of .9 to .95, there will be far more fissions triggered (167 to 352) by a proton than there will be direct proton induced spallation (destruction) of actinide atoms (5 or 6). Therefore, the neutronic spectrum and the resultant fission products would be quite similar to those in a comparable reactor, and PHOENIX target design would be based almost exclusively on current or planned advanced liquid-metal-cooled reactor technology.

2. RADIOACTIVE WASTES AND CURE

2.1 Toxicity Reduction Using CURE Processing (Ref. 1)

A major objective of the CURE process is to reduce the toxicity of the waste stream, as illustrated in Figure 3. The ingestion toxicity (how much water is required to dilute the material to reach safe drinking water standards) for different waste stream scenarios are compared against that of natural uranium. The top curve shows the toxicity of the entire waste stream, assuming a once-through fuel cycle. Even after 10,000 years, this waste stream remains two or three times more toxic than natural uranium. If the plutonium and uranium are separated, resulting in a second curve, the toxicity falls off significantly after 1000 years. Although it is not shown in Figure 3, the toxicity from the plutonium is 4 to 5 orders of magnitude greater than that of the uranium. The additional removal of the minor actinides, i.e., neptunium, americium, and curium from the waste stream results in a third toxicity curve. Note here that the waste stream reaches the equal-toxicity point after 3 or 4 centuries. Of the remaining waste stream, four isotopes stand out as problems: Sr-90, Cs-137, Tc-99, and I-129. The first two have half-lives of around 30 years and contribute significantly to the short-term radioactivity and heat load (a packaging problem). The latter two have very long half-lives, as well as high mobility and, thus, are problems when considering water intrusion and leaking. If these four key isotopes are removed from the waste stream (in addition to the actinides), the lowest toxicity curve results. The resulting waste stream would reach equal-toxicity within a few decades (i.e., ~30 years).

While relative toxicity is one measure of the hazards posed by the long-lived waste, another key measurement is risk - which factors in the likelihood of the material escaping to the environment. Because key fission products, especially the iodine and technetium, are far more likely to leach into the environment one must also address these fission products in order to significantly reduce the repository risks (Refs. 7, 8).

2.2 Key Elements and Isotopes

Of the uranium remaining in the spent fuel after 10 years of storage (the CURE assumption), only 0.8% is fissionable (U-235). While this uranium could be recycled into power reactors, some enrichment (or supplementing with plutonium) would be required before it could be used in light-water reactors (LWRs).

Fully two thirds of the plutonium is fissionable Pu-239 and Pu-241, and in contrast to uranium, plutonium is highly toxic. Because of the quantity of plutonium involved and the relatively high fissile content, the LWR waste plutonium may be better classified as a fuel, as opposed to a waste to be consumed (assuming that it is to be consumed at all).

The remaining actinides, often referred to as the "minor actinides", include neptunium, americium, and curium. Very few of the minor actinide isotopes fission thermally, and most of them absorb thermal neutrons effectively. After absorbing two neutrons, on average, they eventually evolve into fissionable isotopes. (Note: in a very high thermal neutron flux, it is possible to fission a short-lived fissile isotope before it decays, which could reduce the number of neutrons absorbed from two to one.) However, these minor actinides would initially be strong poisons if placed into LWRs.

Of the many fission products, only a few have sufficiently long half-lives to cause problems for burial. Seven such fission products are shown in Figure 4. The Sr-90 and Cs-137 are problems over the near term, i.e., a few hundred years, as they are highly active and contribute significantly to both toxicity and heat loading. The other five isotopes in Figure 4 are approximately equivalent when judged by half-lives and toxicity. However, the Tc-99 and I-129 are much more mobile and thus more likely to leach into the ground water. Therefore, it is these four fission products, Sr-90, Cs-137, Tc-99, and I-129 that are the highest priority for elimination.

2.3 Other Current Perspectives on Waste Processing

In previous studies (Refs. 9 and 10), researchers at the Oak Ridge National Laboratory (ORNL) compared the long-term repository risk reduction from actinide burning to increases in cost and contemporary risk. At that time (1977-80), they concluded that incentives for actinide burning did not exist. Recently (Ref. 11, published 11/90), the same group declared that "the now apparent challenges involved in characterizing and licensing a repository, combined with the specific repository regulations established in the last several years, appear to result in real incentives for actinide burning that will accrue during the next two decades." In particular, the authors cite a reduction in repository site characterization costs, an enhancement of repository site licensing, and an improvement in public acceptance.

However, the actinides are not the only significant hazards, as discussed in Reference 12. The author points out that "current analyses for repositories show that the doses from the fission products Tc-99, I-129, and Cs-135 are among the most significant long-term concerns."

The apparent contradictions between References 11 and 12 can be explained. If one focuses on current regulations and the hazards posed by all materials in the repository, in case of intrusion into the repository some time in the distant future, then the actinides are the major concern. However, if one focuses on the materials most likely to escape into the environment, driven only by natural forces, then some of the fission products become the major problem. Therefore, both references are correct to some degree. The CURE approach, and that adapted here, is to try to address all the potential problem materials under the assumption that the leaching/mobility questions are important.

3. THE PHOENIX CONCEPT

3.1 Characteristics of the Minor Actinides

The through-put and composition of minor actinides from a 3410 MW_t light-water reactor (LWR) are indicated in Figure 5. These figures are based on the assumption of 10 years of decay, without separation, before processing. The composition estimated for project Omega (Ref. 3) is somewhat different, as the minor actinides are separated from the uranium and plutonium after 3 years, and the reprocessing is performed after only 8 years. Either way, the minor actinide isotopes from LWR spent fuel do not fission well in a thermal spectrum.

The fission and capture cross sections with respect to fast neutrons are shown in Figure 6. Essentially all of the actinides will fast fission rather than capture fast neutrons. In addition, the number of neutrons per fission is in the range of 3 to 4 at high energies. Therefore, while the minor actinides are poisons in thermal reactors, they will fission in a fast spectrum reactor.

If one wishes to fission efficiently, and thereby eliminate the large inventory of LWR waste minor actinides, a fast-spectrum lattice is the best choice. In order to obtain the hardest spectrum, one would probably use a metallic fuel and a coolant that has little impact on the neutrons, e.g., helium or lead. However, metallic form minor actinide fuels would require considerable development and testing, and helium and lead have not been used in U.S. fast reactors. Therefore, although the LMR oxide fuel option may not have the highest neutronic efficiency, it is the best understood option, and would likely be efficient enough for the purpose of transmuting actinides. However, development of an advanced metallic form of minor actinide fuel would likely reduce the "burnup" reactivity increase and reduce the peak current required from the accelerator. Similarly, adaptation of a lead-based coolant would have significant advantages. Clearly, various cost-benefit trade-off studies should be performed once the basic concept has been established.

3.2 Spallation and Neutronics

With increased development of particle accelerators for applications ranging from research, to materials production and modification, and to the Strategic Defense Initiative, these machines have become larger, more powerful, and more efficient. The basic physics that results when high-energy charged particles are driven into targets of heavy elements has been known for several years. While empiricisms remain regarding the precise features of the intranuclear cascade process, there exist sufficient data and supporting theory to make reasonably accurate ($\sim \pm 15\%$) predictions for protons of a given energy level impacting on an actinide nucleus.

Figures 7 and 8 indicate current best estimates, based on data and calculations, of the number of TRU nuclides spalled and the number of neutrons ultimately released, as a function of the incident proton energy in large targets (smaller than the PHOENIX targets, however). For an incident proton at 1.6 GeV, 5 or 6 nuclides of Np, Am, or Cm (they will be very similar) will be spalled. More importantly, about 50 neutrons will be knocked free as the proton penetrates the lattice; most of these resulting from evaporation.

If the target were to be of materials that could not fission, most of the neutrons would be absorbed in the lattice, and there would be little TRU conversion. However, in a subcritical lattice, the neutrons would be multiplied, as was shown in Figure 2. An effective multiplication factor of 0.9 results in the 50 neutrons becoming 450 neutrons. Most of these neutrons result from the fission of about 167 nuclides, again as was shown in Figure 2. In combination with the spalled nuclides, the single proton

results in the destruction of 172 target nuclides. Should the multiplication factor be 0.95, the same proton could trigger destruction of 357 of the TRU nuclides.

3.3 Fission-Product Targets

In order to keep a lattice containing minor actinides sufficiently subcritical with a hard neutronic spectrum, a significant fraction of neutrons must be leaked from the lattice, particularly as the reactivity increases with "burn-up" (which is really build-up in terms of fissile isotopes). The availability of neutrons provides an opportunity to reduce some of the inventory of problem fission products.

The transmutation chains for iodine and technetium are shown in Figure 9. While there is no real advantage to converting the I-127, it will comprise 24% of the iodine, and will therefore be present to divert some of the neutrons that could be better used to convert the I-129. In the case of all three base isotopes, Tc-99, I-129, and I-127, the absorption of one neutron creates a stable isotope, and the absorption of subsequent neutrons has little impact, except for wasting neutrons.

In terms of the chemistry, the two candidate fission products are very different. Xenon is gaseous at all temperatures of interest, and elemental iodine also has relatively low melting and boiling temperatures. Iodine targets would have to be designed carefully to retain the gasses. As these targets are composed of neutron absorbers, the leakage of these poisons could result in a significant reactivity increase. On the other hand, both technetium and ruthenium melt above 2400K (3900F) and should be quite safe and stable, even in critical assemblies, i.e., reactors.

4. PHOENIX Design Studies

4.1 Lattice Types

In selecting a lattice type, the principal objectives are a hard neutronic spectrum and a high degree of safety, especially with respect to heat removal. Of the three most credible coolant choices, sodium, helium, and lead, sodium has the best heat removal capabilities. It can be used in a relatively high-power-density lattice, has a high boiling temperature and excellent natural circulation capabilities, and conducts heat very well. Sodium also tends to soften the neutronic spectrum somewhat, but that is not currently perceived as a major problem. In contrast, while helium would have no effect on the spectrum, it does not conduct heat well and tends to leak from systems that are not tightly sealed. Lead would soften the spectrum considerably less than sodium, but it does not conduct heat quite as well as sodium, and there has been little experience with lead coolant systems within the U.S. However, the use of lead-bismuth or lead-magnesium coolant systems would help the system efficiency by decreasing neutron captures and "burnup" reactivity swings, and is still being considered.

Regarding the fuel type, most liquid-metal reactors (LMRs) use oxide fuel, and it is believed that minor actinides could be substituted for the uranium and/or plutonium in the more common form of oxide fuel. While oxide fuels have high melting temperatures, the poor thermal conductivity and the neutron moderation impact of the oxygen atoms are notable disadvantages. If the metallic fuel developed as part of the Integral Fast Reactor (IFR) Program at ANL is utilized, the neutron spectrum would be somewhat hardened, which would decrease the "burnup" reactivity swing. However, substitution of americium, neptunium and curium for plutonium and uranium in the metal fuel would require some development effort. Although the melting point is lower for the metal fuel, the high thermal conductivity mitigates this problem somewhat. The "inherent shutdown" characteristics of the metallic fuel, in a reactor environment, would have little impact in PHOENIX, as the sub-critical lattice is driven by the accelerator. In addition, the inclusion of minor actinides significantly increases the sodium void worth

and decreases the Doppler feedback (Ref. 13). Finally, in contrast to oxide and metal fuels, there are little data available for carbide or nitride fuels (although the French are now studying nitride fuels).

The Fast Flux Test Facility (FFTF) (Ref. 6) currently uses sodium coolant and oxide fuel, and provides an existing data base as well as a potential site for experiments on minor actinide fuel. The prototype PHOENIX lattice was based on the FFTF lattice, and scaled up to the required power level. As indicated in Figure 10, the lattice parameters would be essentially identical to FFTF, with the simple replacement of uranium - plutonium oxide fuel with minor actinide oxide fuel.

The principal reason for basing the target design on FFTF is credibility. Because most neutrons in the PHOENIX lattice will be the result of fissions, and most materials damage will be caused by those neutrons, there are strong reasons to believe that the PHOENIX target modules will experience materials damage very similar to that in FFTF. Therefore, we are fairly sure that this design can stand close scrutiny. On the other hand, this target is not optimized, and some improvements can be expected.

4.2 Sizing and Design Calculations for Target Modules and Lattices

Sizing of the full-scale PHOENIX facility is constrained by economic factors associated with the large linear accelerator. As was discussed in Section 1, a current of 104 mA is sufficient to drive a $k=0.9$ subcritical lattice at 3600 MW_t. In principle, the 250 mA accelerator designed for tritium production (Refs. 4 and 5) could be utilized. A key feature of that accelerator design is the funneling of two 125 mA beams near the front of the machine. By limiting the PHOENIX accelerator to the 104 mA required to achieve 3600 MW_t in the lattice, we can eliminate the funneling aspect while retaining a significant margin below the current where funneling may be needed.

With respect to the lattice design, the extrapolation from the FFTF core is fairly modest. The number of protons and high energy neutrons (from direct collisions) is much smaller than the number of fission neutrons. In addition, the burnup level that is planned is lower than what is currently achievable, largely so the plutonium can be salvaged while it is mostly (86%) Pu-238. As the power peaking can be managed by beam shaping and lattice design, the use of a 3600 MW_t lattice (8 modules) is not considered to be a large extrapolation.

The contribution of the reference PHOENIX facility, regarding the CURE process, is illustrated in Figure 11. Note, that while PHOENIX is converting only about 3 MT/yr of the 2500 MT/yr waste stream, it disposes of the most difficult portion.

Initial design and calculations for obtaining a preliminary estimate of the over-all dimensions of the target, and some of the detailed neutronic characteristics of the target modules were performed with the LAHET and MCNP Monte Carlo codes. The LAHET portion of the analyses considered the interaction of the incident 1.6 GeV proton beam with the lattice and the subsequent generation of neutrons from high energy fission, spallation and evaporation events. The resulting neutrons are tracked until they leak from the lattice or scatter below some "low" energy cut-off ($\sim$20 MeV). The neutron source below $\sim$20 MeV is written out to a file as a function of energy and position within the lattice for the subsequent detailed slowing down calculation performed by MCNP. This analysis employs the latest ENDF/B-V cross sections in a detailed point representation. The highly heterogeneous nature of the target, coupled with the current limitations in the LAHET geometry lead to a two step approach. First a coupled LAHET/MCNP calculation was performed for an essentially homogenous 3-D rectangular representation of the target lattice. Subsequently, the lattice feature of the MCNP 384 code was used to represent the lattice/target module geometry in its full heterogenous detail to qualify the adequacy of the homogenous modelling employed in the coupled calculation. The results of these calculations provided a preliminary

estimate for the size of the target of 188 cm. wide, 82 cm. deep, and 75 cm. high. This sizing gives a k effective of approximately .8 for a new target, which is a good choice given the large reactivity increase expected during the first 2 years. Estimates of the leakage across the faces of the target and the neutron flux, spectra, and heat generation rates were obtained. The initial leakage is estimated to be roughly 40% (of total neutrons), although this may have to be increased during the burn-up cycle, e.g., by altering the geometry when necessary.

4.3 Burn-Up Calculations

Based on the lattice composition and neutronic spectrum of FFTF (the accelerator should harden the spectrum somewhat), burnup calculations were performed using the ORIGEN Code (Ref. 14). We assumed the entire lattice was initially composed of the minor actinides from the LWRs via the CURE processing facility. It was assumed that every 2 years the fuel was removed and reprocessed. A two-year decay while the "spent fuel" is cooled, processed, and reloaded was factored into the analysis. During this two-year period all fission products and the plutonium are assumed removed, and minor actinide makeup is assumed to be added, restoring the initial "fuel" loading.

The results of a 6-cycle, 12 year burnup calculation are shown in Figures 12 and 13. The principal reason for recycling every two years is to remove the plutonium while it is largely Pu-238. This has at least two advantages. First, the structural materials only have to survive 18 months of burnup (24 calendar months), which should be easily attainable (HT9 could likely survive longer exposures but some testing would be required). Second, the burn-up reactivity increases are reduced so they can be more easily compensated.

As shown in Figure 12, a near-equilibrium cycle is reached within a few years. This has several advantages, including the fact that an old target (fuel in for 7th cycle, for example) will behave much like a new target.

The increasing inventory of fissile isotopes causes the neutron multiplier to increase, as indicated in Figure 12. Because ORIGEN does not fully account for geometric factors, the increase in k plotted in Figure 12 is only approximately correct. If correct, it would imply that k, effective would increase from 0.8 to a little greater than 1. This increase can be overcome by varying the beam current, by changing the geometry (more leakage) from cycle-to-cycle, and by using burn-up compensating poisons.

The isotopic concentrations are shown in Figure 13. The two-year cycle produces little Pu-239 and only a modest amount of Pu-242. The fraction of Pu-238 at the end of each 2 year cycle varies between 85% and 87%. There is little variation in the Np-237 inventory from cycle-to-cycle. The gradual increase in Am-243 relative to Am-241 is notable, but it appears to be well stabilized by the 6th cycle.

Pu-238, because of its relatively "short" half-life of 87.7 years, is considered to be a good power source for use in the space program. For such usage, the Pu-238 must be fairly pure and contain only very small amounts of Pu-236 (3 year half-life). With the hard neutronic spectrum in PHOENIX, Pu-236 production will likely be too high in the Np-237 portion of the fuel. Therefore, in order to provide Pu-238 for space applications, the Am-241 and Np-237 would need to be separated physically. This has not been evaluated, but through careful engineering at least one-third of the Pu-238 production could probably be used for space applications.

Another interesting usage of the Pu-238 is suggested in a paragraph from p. 217 of Ref. 15. It seems that as little as 5% Pu-238 in a plutonium weapon would cause the explosives to melt prematurely.

Thus, there are some genuine possibilities for using the Pu-238 to render the other recycled plutonium (66% fissile) useless for weapons production, which suggests an entirely different approach to nuclear nonproliferation. Unfortunately, any attempt to further quantify that analysis at this time would almost certainly cross into a realm that is correctly limited by national security considerations, i.e., the details would be "classified".

4.4 Modular Design

While PHOENIX is designed to keep up with the waste stream from about 75 LWRs if fully loaded, it need not always run in such a mode. Initially, there may not be 24 metric tonnes of processed minor actinides available for loading. Also, during the first years of PHOENIX operation, a lower target loading and, therefore, a lower demand for beam current would make for an easier start-up and testing phase for the large linear accelerator. Finally, the use of modular targets would allow the removal of a troublesome target module without shutting down the machine for a prolonged period of time.

The preliminary size, shape, and important characteristics of the eight 450 MW_t PHOENIX Target Modules are indicated in Figure 14. Lattice characteristics inside the hex-cans are very similar to FFTF. However, the shape of the module, .75 M high by .82 M deep by 1.88 M across, is set to allow for considerable neutron leakage. This shape also maintains a negative sodium void reactivity worth and reduces the peaking into the target (calculated to be near the center, front-to-back). The fact that most neutron leakage is out the top and bottom or the front and back means that several modules could be placed side-by-side without significantly impacting on the neutron multiplication factor.

The arrangement of the eight PHOENIX target modules within the vacuum chamber is illustrated in Figure 15. Each module is designed to be removed from the vacuum chamber for reloading. PHOENIX could run effectively even with only one target chamber, although its throughput and efficiency are highest if all targets are loaded.

Based on the preliminary design, the fission product targets indicated in Figure 15 would be Iodine-129 (in a stable-compound form) possibly cooled by D_2O (Heavy Water). The D_2O would serve as a moderator, because the I-129 is most likely to absorb epi-thermal neutrons. Because water and sodium react violently if mixed, no water-cooled targets would be located close to the sodium piping. Instead, some other moderator, such as graphite or beryllium, would be located between the sodium pipes and water cooled fission product targets.

The region in and around the vacuum chamber will become highly radioactive after a few months of operation. Handling of target modules and fission product targets will be handled remotely, which is a major reason for using modular target units. Operationally, after the accelerator has been shut down for a few weeks, a module would be removed to a remote cooling area. A fresh module is then moved into the vacuum chamber and the accelerator is then restarted.

4.5 Accelerator Performance at Reduced Loadings

Regarding the operation of PHOENIX at reduced target loadings, one important factor is that the machine can be run at different duty factors in order to produce different currents. If the machine is running at a 60% duty factor, it is driving current for 600 milliseconds out of every 1 second, and is otherwise idle. For an assumed 60% duty factor, a current of 62 mA (average) would be delivered, which requires 265 MW of electricity. Such a current could drive the PHOENIX lattice at 3600 MW_t if the k-effective was slightly under 0.94. Even if the k-effective is significantly less than 0.9, the accelerator could drive the lattice at lower power levels, at least until the reactivity builds to 0.9.

The performance of PHOENIX with fewer than 8 target modules in place, assuming k-effective is 0.9, is illustrated in Figure 16. Note that there is net positive electricity generated in all cases, although the proportion available to the electrical grid improves as more modules are brought on line. With fewer than eight targets loaded, a higher current can be delivered per target module. Thus, a target with a first-cycle fuel loading could be driven along using a higher beam current until the k increases to 0.9.

5. POTENTIAL IMPACT ON HIGH LEVEL WASTE REPOSITORY

The partitioning and transmutation of long-lived radioactive waste isotopes should reduce the flow of problem isotopes to a high level waste repository. However, a modest reduction would be of questionable value, because one would still have to assure the integrity of the repository for a prolonged period. Further, the transmutation process would actually increase the volume of waste materials, although the added waste would be mostly short-lived isotopes. Thus, one needs some sort of quantitative objective in order to establish whether the proposed approach will achieve anything meaningful.

As was suggested in Figure 3, the real goal is to reduce the time period over which one must contain the wastes. If one can drop that period to a few decades, then the steel containers and the repository itself need only survive a credible number of years. But then one must ask "how low is low enough", i.e., how small does my residual waste inventory have to get before I can neglect it?

5.1 Repository Guidelines in the U.S.

In the U.S., guidance can be found in the Code of Federal Regulations, particularly 40 CFR 191 (Ref. 16) and 10 CFR 61 (Ref. 17). The former (which may be revised downward) specifies how many Curies per metric tonne of heavy metal of key isotopes can be allowed to escape the repository during the first 10,000 years after disposal. These limits, along with the activity in the spent LWR fuel (CURE assumptions), are shown in Figure 17. For several isotopes, one would have to assure on the order of 99.99% retention over the 10,000 years to meet 40 CFR 191. On the other hand, if one removed most of the problem isotopes, there could be far less waste inventory passing into the repository. For example, if one removed all but .1 Curie per metric tonne of the Am-241, then one could meet 40 CFR 191 a priori, i.e., containment could become unnecessary. In fact, if one could remove all but 1 part in 100,000 of the problem isotopes from the waste stream, then one could meet the current 40 CFR 191 requirements without any containment of the waste.

The other limit, from 10 CFR 61, is really for low level wastes, and because spent fuel is classified high level wastes and cannot (technically) be re-classified it does not apply to the repository. However, it provides an interesting indicator of how good is good enough. If we again go through the process of removing long-lived isotopes, we get another set of allowable partitioning losses, this time for the hypothetical conversion of high level wastes to "Class C" low level wastes. These are shown in Figure 18, which includes a per unit volume factor that comes from 10 CFR 61. Roughly speaking, it appears that partitioning losses of 10^{-5} or lower are required.

Thus for Figures 17 and 18, it appears that if the system partitioning losses are in the range of 10^{-5} or lower, the burden placed on the repository is fundamentally changed. This then would be the minimum objective of any partitioning and transmutation scheme in order to succeed within the U.S.

5.2 System Losses vs. Chemical Losses

An idealized waste partitioning and transmutation system is illustrated in Figure 19. However,

there would be partitioning losses, and these would allow some of the long-lived wastes to slip into the repository at both the initial partitioning phase and again at the reprocess partitioning phase. Further, the reprocess partitioning losses become multiplied as one reprocesses the transmutation targets. The net losses, therefore, depends on the amount transmuted per pass through the machine, as shown in Figure 20. Note that for a 90% transmutation machine, the net system partitioning losses are very close to the chemical partitioning losses. However, with a 10% transmutation machine, one needs chemical partitioning losses near 10^{-6} to get a net partitioning loss near 10^{-5}.

5.3 The Objective and Current Capabilities

From the materials shown in Figures 17 through 20, it appears that chemical partitioning losses in the range of 10^{-6} or lower will be needed to achieve the primary goal of fundamentally changing the repository. Unfortunately, this objective could even be more restrictive if the release limits are tightened.

Are such chemical separations possible? According to the Westinghouse Hanford Specialists who developed the CURE Process, chemical separations of 10^{-6} have been achieved in the laboratory, and even finer separations could be achieved (Ref. 18). As to how easily this can be achieved on a plant scale, only time will tell. This then is probably the weak link in the entire partitioning and transmutation chain.

6.0 SUMMARY AND CONCLUSIONS

Through a process of chemically partitioning the high level wastes and transmutating the key long-lived isotopes, one can significantly reduce the toxicity of the wastes and the risks (eliminating Tc and I, especially) due to placing that waste in a geologic repository. Much of the needed partitioning technology has been developed, although further work is required (Ref. 1). The PHOENIX Concept, discussed herein, is capable of transmuting large quantities of the problem isotopes, and of producing electricity in the process [Ref. 19]. We believe that the proposed means of partitioning the waste and of transmuting key long-lived portions of it are credible, and that these or similar approaches could be implemented when it becomes desirable.

At this time, we believe that the principle challenge is to reduce system partitioning losses to the degree where the advantages for the repository are clear. While the burden of proof now passes to the separation chemists, it appears that the required partitioning technology may become available within the next few years. When the partitioning technology has become sufficiently matured, the PHOENIX Concept and other transmutation devices should provide a viable capability for converting the long-lived waste isotopes to either stable or short-lived isotopes.

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Figure 1. The PHOENIX Concept

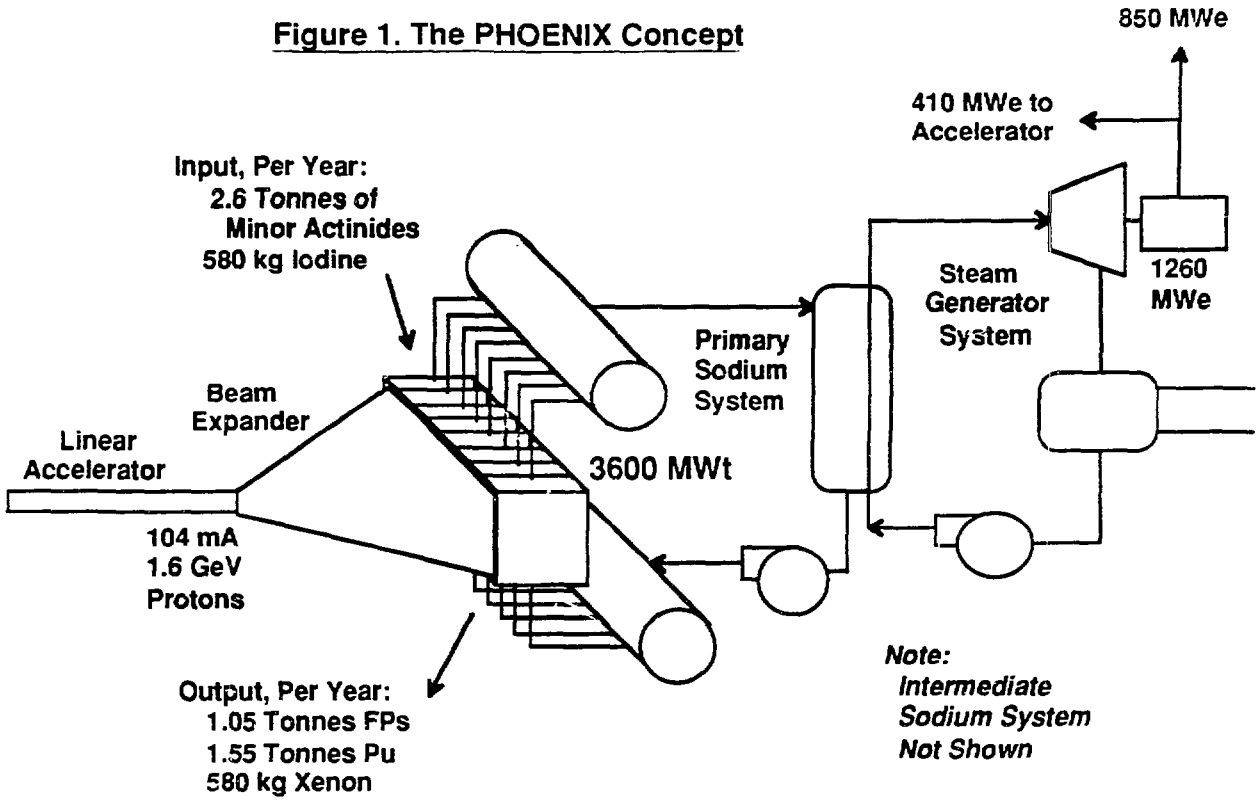


Figure 2. Conversion Ratio-Protons to Transmutations

Number of Transmutations = Number of Fissions + Number of Spallations (Proton Induced Fissions)

Number of Neutrons = Number of Source Neutrons x (1 + k + k x k + k x k x k + ...) ,

where: *Number of Source Neutrons = Number from Spallation + Number from Evaporation*

k=k, effective = Effective Neutron Multiplication Factor for the Lattice

Therefore: Number of Neutrons = Number of Source Neutrons x (1/(1-k)) *(Geometric Series)*

Number of Fission Neutrons = Number of Neutrons - Number of Source Neutrons

= Number of Source Neutrons x (k/(1-k))

Number of Fissions = Number of Fission Neutrons / v,

where: v = Number of Neutrons Released per Fission

Therefore: Number of Fissions = Number of Source Neutrons x (k/(1-k)) / v

Number of Transmutations = Number of Source Neutrons x (*k*/(1-*k*)) / v + *Number of Spallations*

Assuming: 5 Proton Induced Fissions, 50 Source Neutrons, 2.7 Neutrons per Fission:

$\Rightarrow$ If k = 0.95, Expect 357 Transmutations per Proton

If k = 0.9, Expect 172 Transmutations per Proton

Figure 3. Impact of Partial Waste Processing

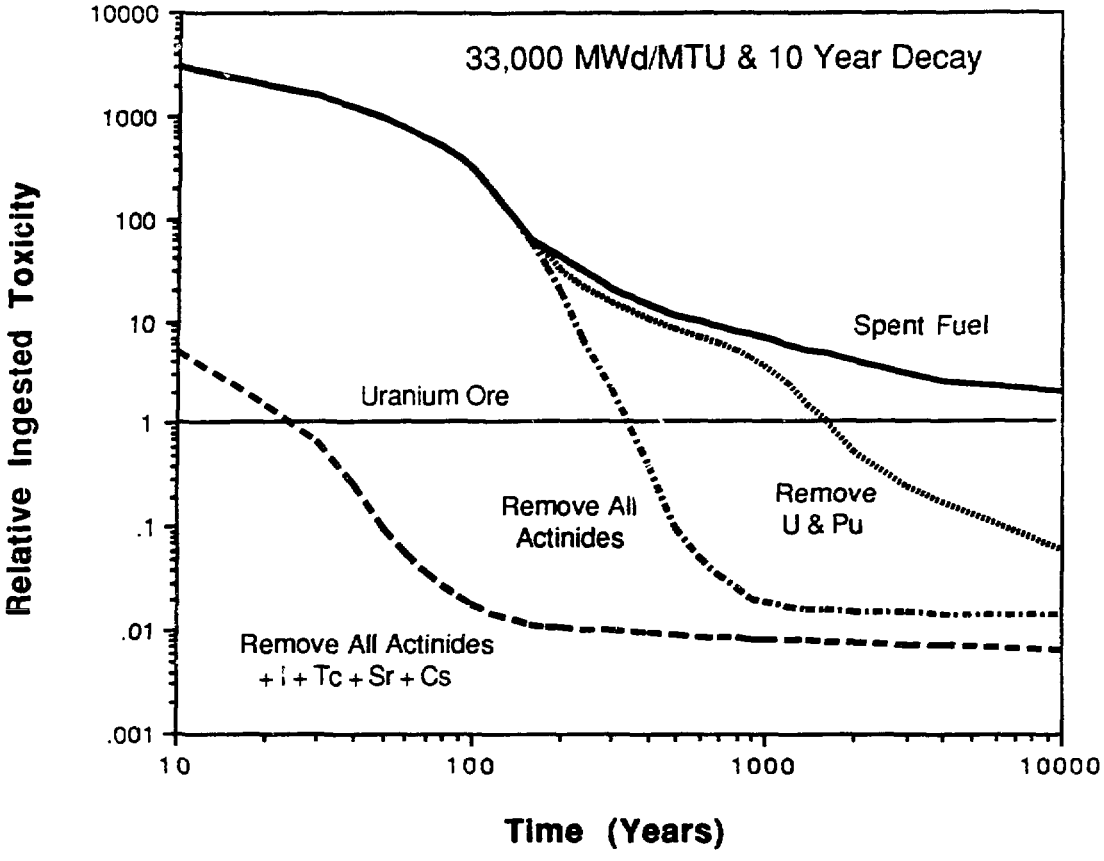
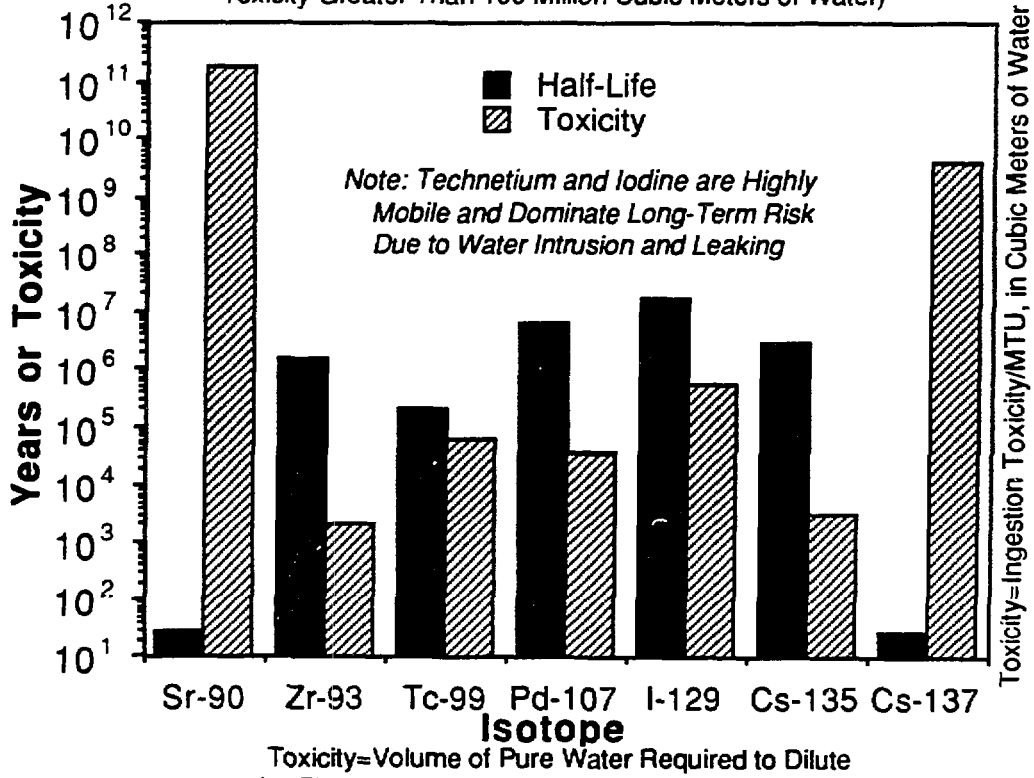


Figure 4. Characterisitics of Key LWR Fission Products

(At least 100g/MTU and Half-Life Longer than 100 yrs, or Toxicity Greater Than 100 Million Cubic Meters of Water)



Toxicity=Volume of Pure Water Required to Dilute the Element or Isotope to Drinking Water Standards

Figure 5. Minor Actinides Generated per year in a 3410 MWt LWR

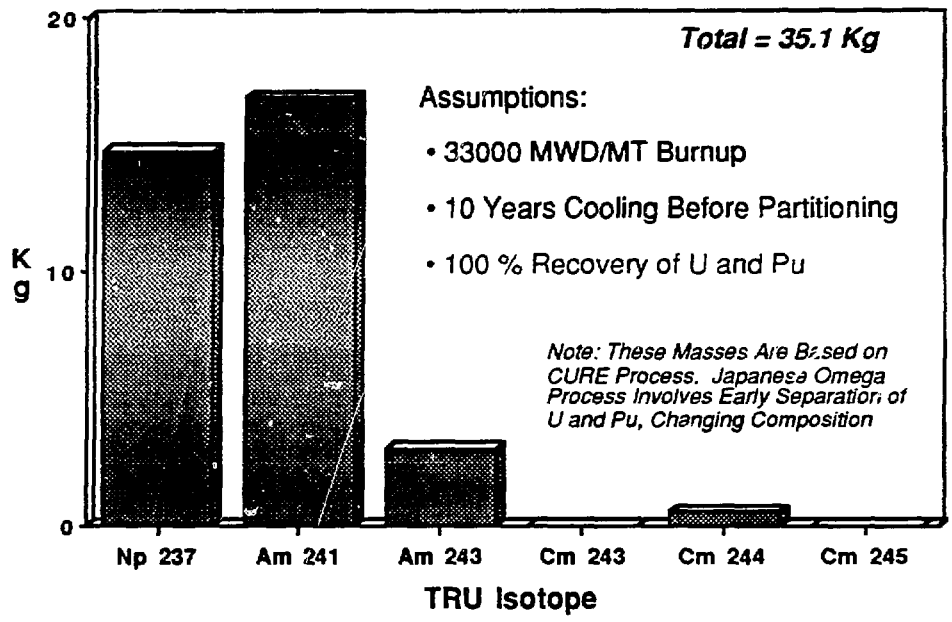


Figure 6. Fast Cross Sections for Key Actinides (1 to 20 MeV)

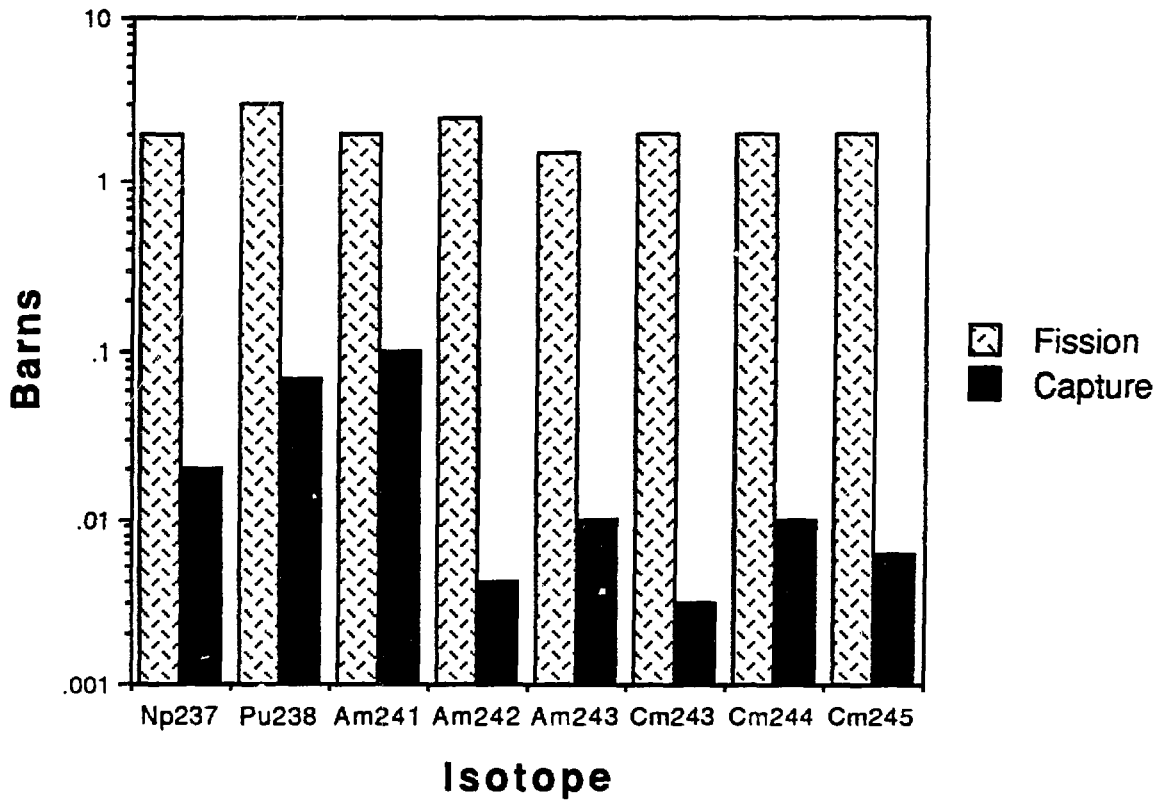


Figure 7. Number of Nuclides Spallated Per Incident Proton Energy

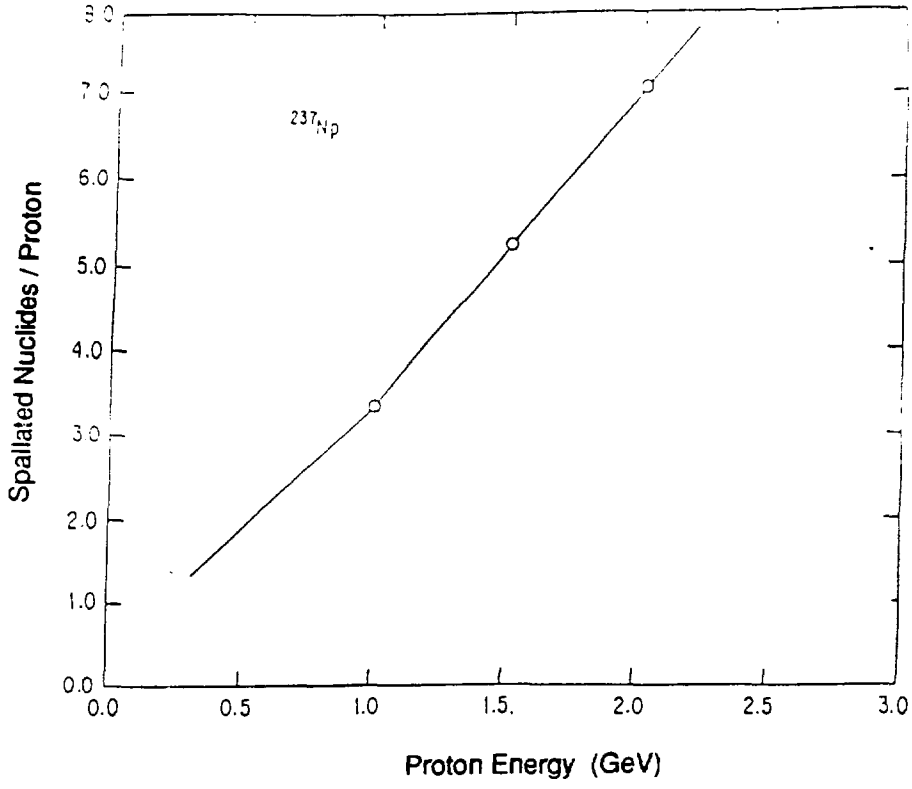


Figure 8. Number of Neutrons Produced Per Incident Proton Energy

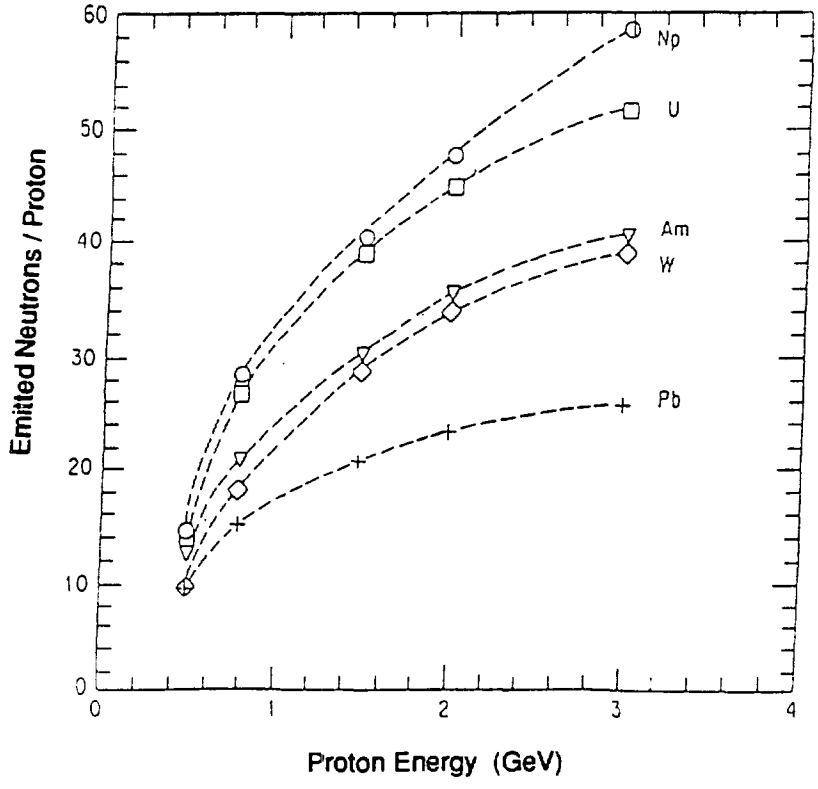
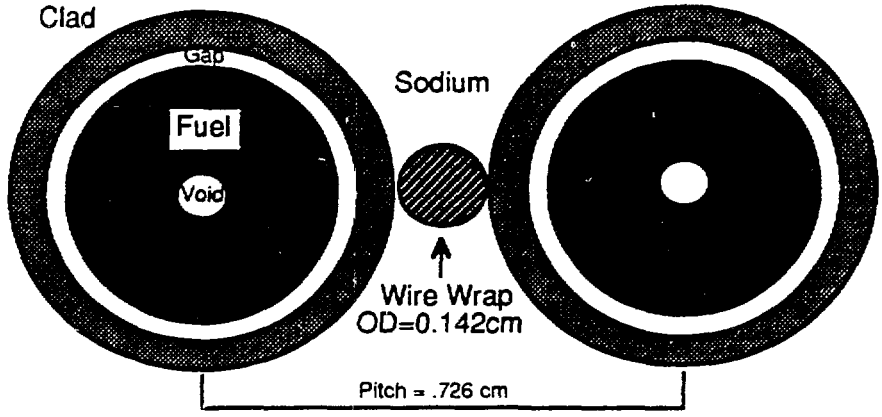


Figure 9. Transmutation Chains for Iodine and Technetium

	Initial Isotopes	Short-Lived	Stable Isotopes
76%	I-129 + n —>	I-130 $\xrightarrow{\beta-}$	Xe-130 + n —> Xe-131 + n —> Xe-132
24%	I-127 + n —>	I-128 $\xrightarrow{\beta-}$	Xe-128 + n —> Xe-129 + n —> Xe-130
			Xe is a Gas Above 166 K (-160 F)
100%	Tc-99 + n —>	Tc-100 $\xrightarrow{\beta-}$	Ru-100 + n —> Ru-101 + n —> Ru-102

Tc and Ru are Solids Below 2445 K (3942 F)

Figure 10. PHOENIX Lattice Fuel Pins Are Currently Based on FFTF Oxide Fuel



Region	Material	OD (cm)
Void	Gas	.076
Fuel	X-O2	.493
Gap	Gas	.508
Clad	SS (HT9)	.584

Fuel Properties:
Assume Same as UO2

Fuel Composition (X-O2)
.418 Np 237
.478 Am 241
.086 Am 243
.017 Cm 244
.001 Cm-245

Figure 11. PHOENIX Transmutes the WastesThat Can't Go Into Thermal Reactors

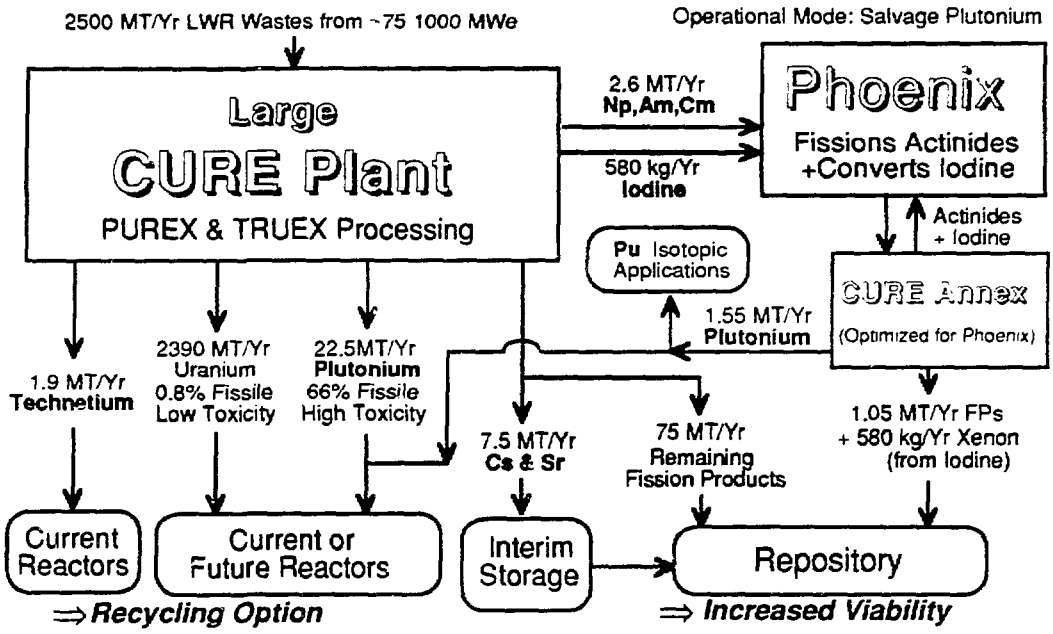


Figure 12. Reactivity Increase & Mass of Principal Actinides During First 12 Years of Burn-Up in 3600 MWt PHOENIX Lattice, Assuming Plutonium Removed During Reprocessing

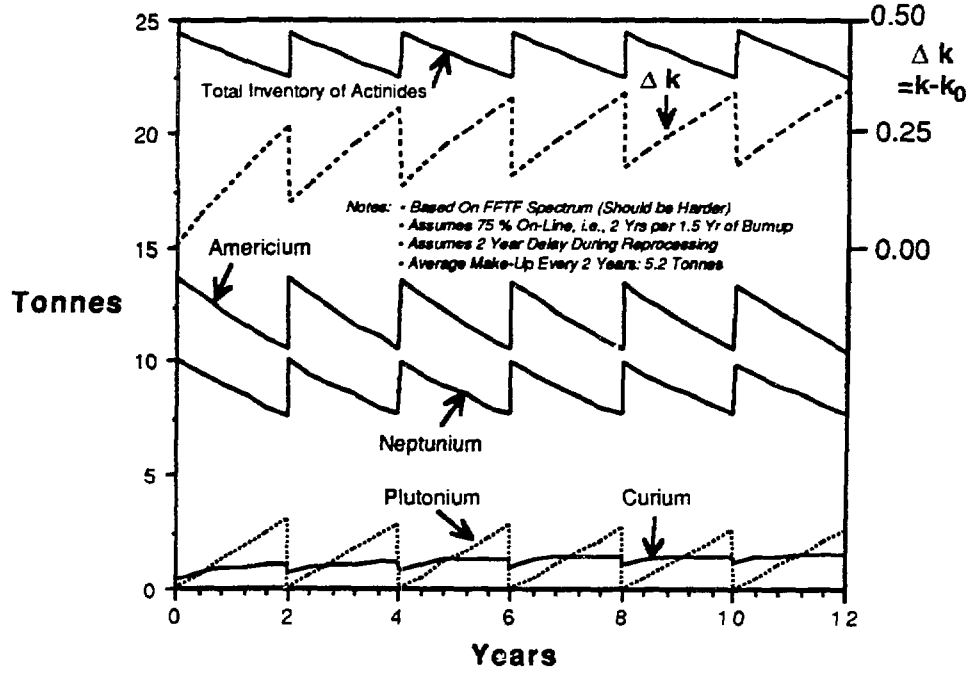


Figure 13. Mass of Key Isotopes During First 12 Years of Burn-Up in 3600 MWt PHOENIX Lattice, Assuming Pu and U Removed During Reprocessing

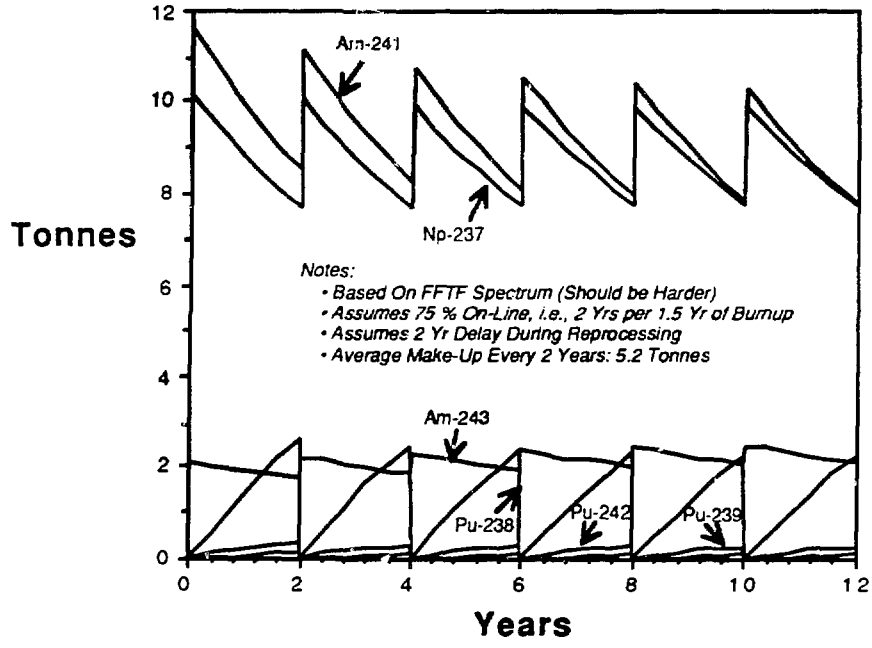
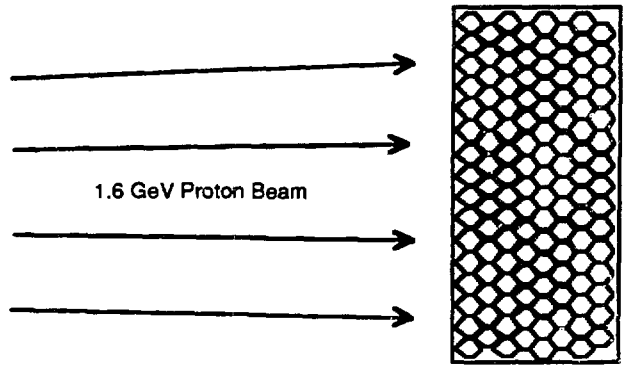


Figure 14. PHOENIX 450 MWt Sub-Critical Module

Approximate Size: 0.75 Meter High, 0.82 Meters Deep, and 1.88 Meters Across
Module Contains 124 Hex-Cans (4 * 16 + 4 * 15) (+ 1cm perimeter gap)
Inside Module/Outside Hex-Cans: Helium Gas + Sodium Leak Detection System



Each Hex-Can : .116 meters flat-to-flat, 4.7 meters high, and containing 217 fuel pins, flow area=.00432m2
Nominal lattice spacing=.120 meters, gap between assemblies: 0.0033 meters (collars)
Operating conditions: assembly flow=24 kg/s, inlet temp=560 K, outlet temp=760 K,pressure=0.5 MPa
Pressure drop = 0.806 MPa, average velocity=6.4 m/s, average sodium density = 850 kg/m3

Figure 15. PHOENIX Target Chamber from Three Perspectives

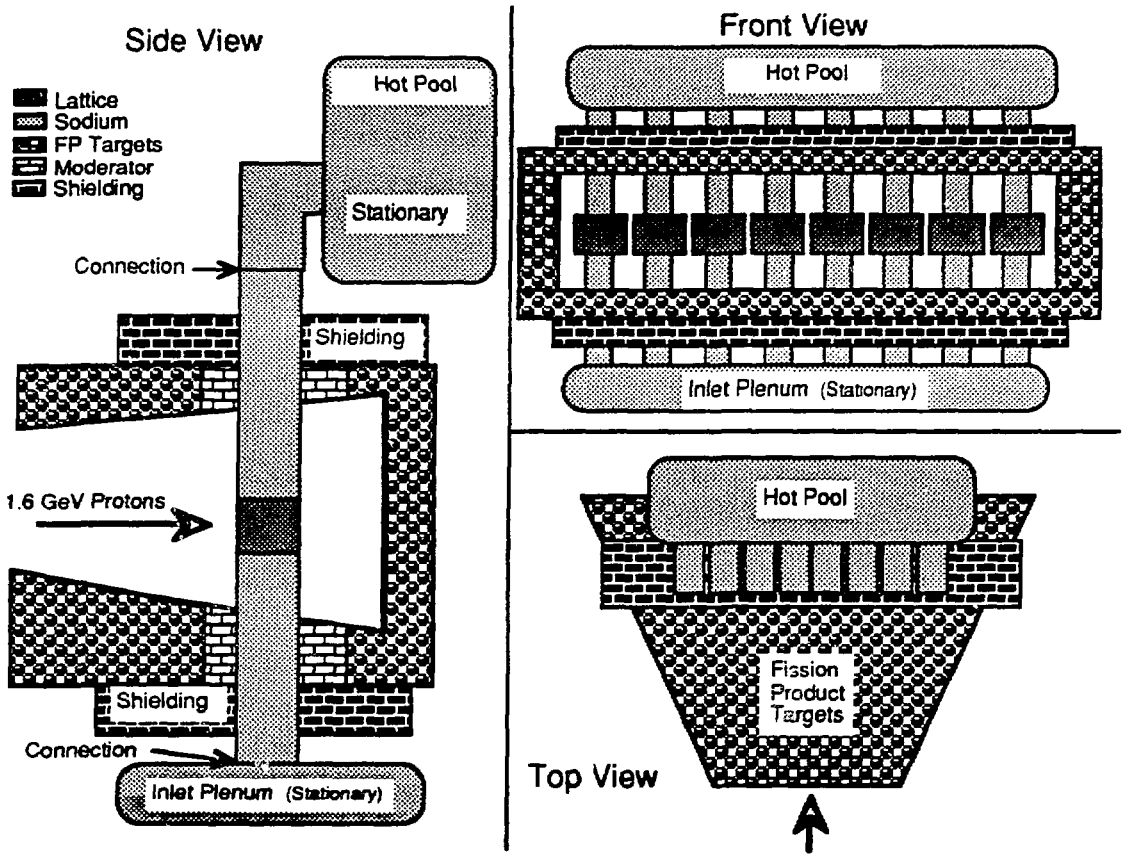


Figure 16. PHOENIX Power Production and Utilization (Accelerator & Grid) As a Function of Number of Target Modules On-Line

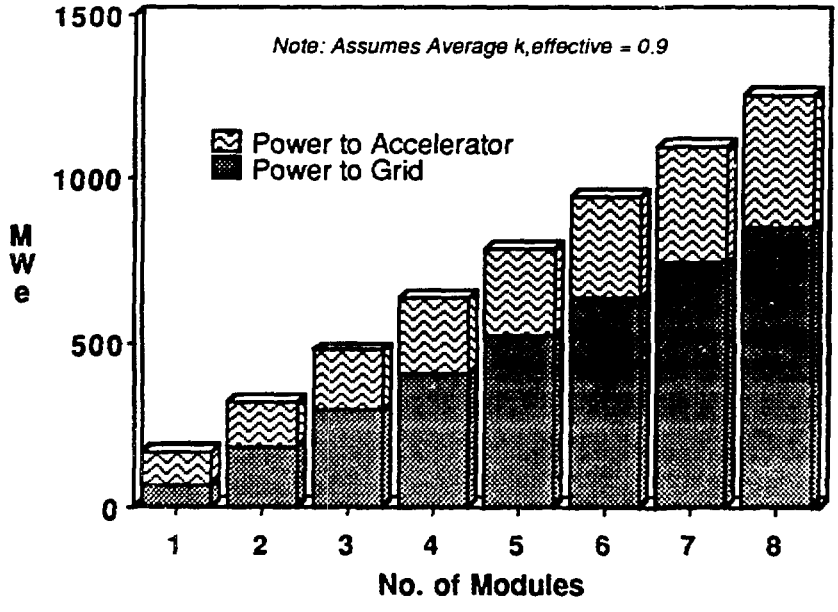
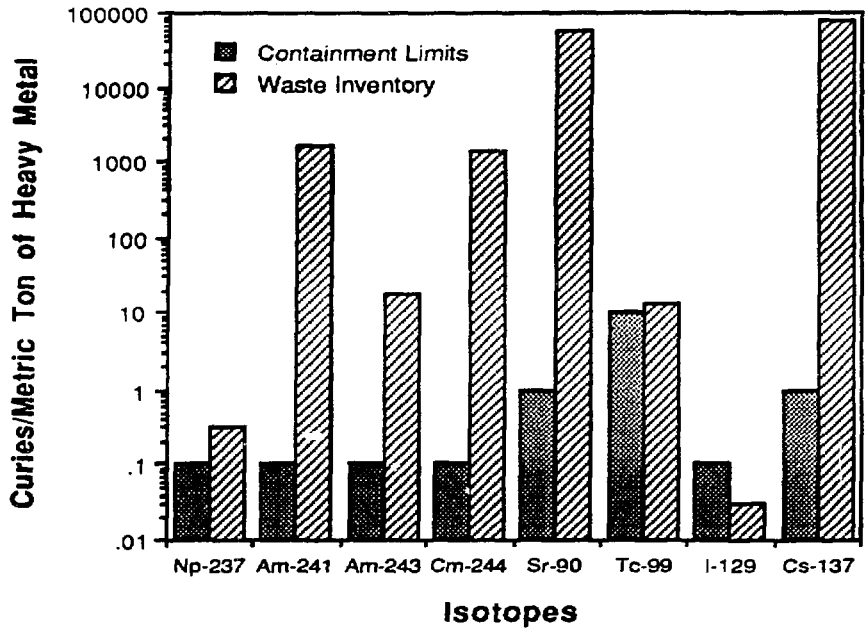
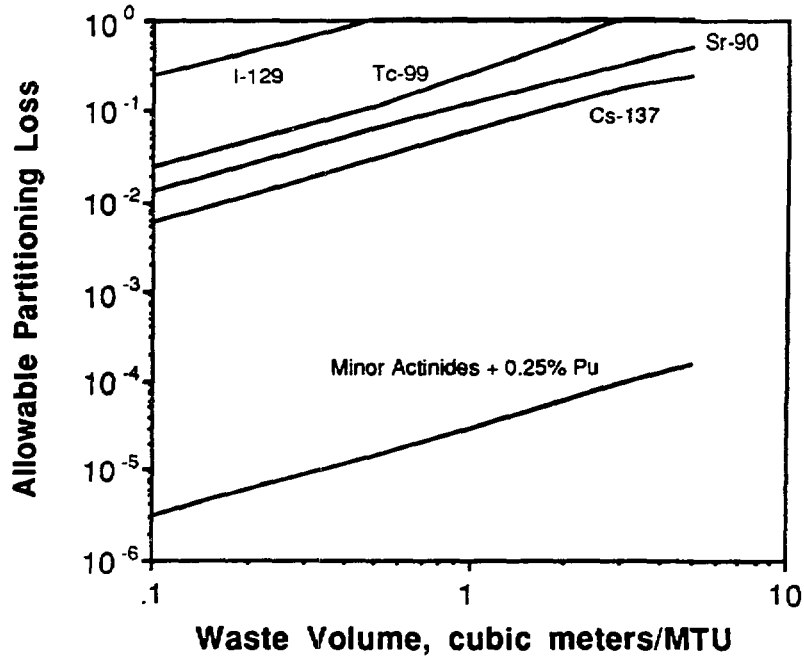


Figure 17. Waste Inventory vs. Release Limits for 10,000 Years After Disposal
[Limits Specified in 40 CFR 191 and Were In Proposed 10 CFR 60]



Conclusion: Partitioning At 10^{-5} or Better Should Meet These Containment Requirements

Figure 18. Allowable Partitioning Losses in Order to Produce Waste Meeting Radionuclide Specifications Shown in 10 CFR 61 for Class C Waste



Reference: "CURE: Clean Use of Reactor Energy", Westinghouse Hanford: WHC-EP-0268, May 1990

Figure 19. Waste Component Flows Through Partition & Transmutation System

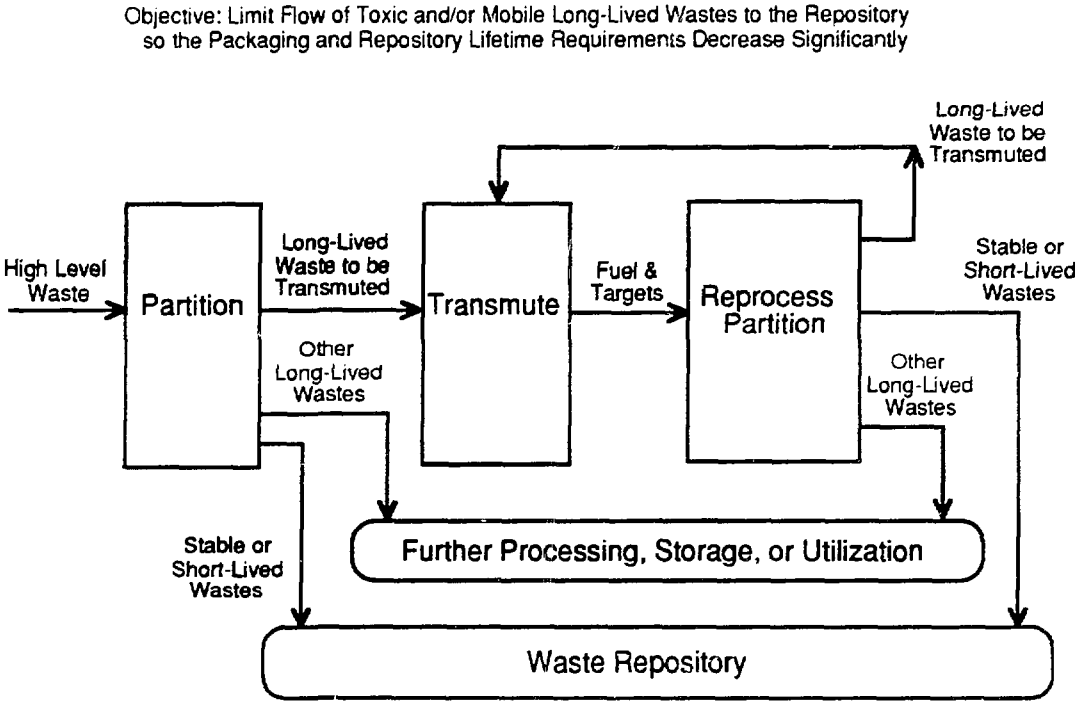


Figure 20. Net Partitioning Loss, Including Impact of Transmutation and Related Partitioning Steps, for Different Levels of Transmutation per Cycle