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*PROPOSED PARTITIONING AND TRANSMUTATION OF LONG-LIVED 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 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 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.

INTRODUCTION

As a result of difficulties in siting waste repositories and recent improvements in partitioning and transmutation technologies, there is an increasing interest in exploring alternate means of disposing of the long-lived nuclear wastes, i.e., by transmutation (References 1-3). Several transmutation options are under consideration, with each proposed approach appearing to have some advantages.

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 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 permanent disposal.

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.

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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 present technology, a 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).

A 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. The physics taking place within the PHOENIX proton-driven lattice is not much more complex than that in a reactor. 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. 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.

RADIOACTIVE WASTES AND CURE

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 2. 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 2, 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 become no more toxic than uranium ore within a few decades.

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 to significantly reduce the repository risks (Refs. 7, 8).

Other objectives of the CURE Program include 1) minimizing the waste stream of "secondary" wastes which are created during the partitioning and transmutation process, and 2) maximizing creation and utilization of isotopes useful for medical or other beneficial use. In particular, minimizing the creation of "mixed wastes" (nuclear and chemical hazards) is a key objective of any partitioning scheme.

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 supplementing with plutonium or enriched uranium may 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 high fissile content, the 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. As discussed earlier, it is the Sr-90, Cs-137, Tc-99, and I-129 isotopes that are the highest priority for elimination.

THE PHOENIX CONCEPT

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.

For an incident 1.6 GeV proton, test data indicates that 5 or 6 nuclides of Np, Am, or Cm will be spalled. The data also shows that about 50 neutrons will be knocked free as the proton penetrates the lattice; with most of these resulting from evaporation (Reference 4). 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, k-effective for the lattice of 0.9 results in the 50 neutrons becoming 450 neutrons. Most of these neutrons result from the fission of about 167 nuclei. In combination with the spalled nuclei, the single proton results in the destruction of 172 target nuclei. Should the multiplication factor be 0.95, the same proton could trigger destruction of 357 of the TRU nuclei.

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 3. While there is no real advantage to converting the I-127, it will comprise 24% of the iodine, and will therefore be present to absorb some of the neutrons that could be better used to convert the I-129. For 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 low probability and 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, leakage could result in significant reactivity increases. Both technetium and ruthenium melt above 2400K (3900F) and should be quite safe and stable, even in critical assemblies, i.e., reactors. We consequently elected to give high priority to converting iodine to xenon in the PHOENIX subcritical target assembly rather than technetium.

PHOENIX DESIGN STUDIES

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.

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. 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. The lattice parameters would be essentially identical to FFTF, with the simple replacement of uranium - plutonium oxide fuel with minor actinide (41.8% Np-237, 47.8 % Am-241, 8.6% Am-243, 1.7% Cm) 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.

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 burnup level that is planned is lower than what is currently achievable. 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 4. 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 (~20 MeV). The neutron source below ~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.

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. 9). 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 Figure 5 (the calculation of iodine transmutation is much more difficult but we estimate 500 to 600 kg/year). 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.

The increasing inventory of fissile isotopes causes the neutron multiplier to increase, as indicated in Figure 5. Because ORIGEN does not fully account for geometric factors, the increase in k plotted in Figure 5 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.

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. 10. It seems that as little as 5% Pu-238 in a conventional plutonium weapon would cause the explosives to melt. Thus, there may be 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".

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 6. 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 7. 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 7 would be Iodine-129 (in a stable-compound form). These targets would have to be cooled and the spectrum would have to be moderated, as I-129 is most likely to ahsorb epi-thermal neutrons

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 of each second, and is otherwise idle. For 60% duty

factor, a current of 62 mA (average) would be delivered, requiring 265 MW of electricity. Such a current could drive the PHOENIX lattice at 3600 MW_t if the k-effective was near 0.94. If the k-effective is 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, was also evaluated. There is net positive electricity generated in all cases, although the proportion available to the electrical grid improves as more modules are operated. For fewer than eight targets, a higher current can be delivered per target module. Thus, a target with a first-cycle fuel loading could be driven using a higher beam current until k reaches 0.9.

POTENTIAL IMPACT ON HIGH LEVEL WASTE MANAGEMENT

The partitioning and transmutation of long-lived radioactive waste isotopes would reduce the flow of problem isotopes requiring permanent isolation. However, a modest reduction may be of questionable value, because one might still have to assure the waste isolation for a prolonged period. Further, the transmutation process would increase the volume of waste materials, although the added waste would be much more dilute and contain mostly short-lived isotopes. 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, the containment need only survive a credible number of years. But how low is low enough, i.e., how small does the residual waste inventory have to get before it can be easily managed?

Repository Guidelines in the U.S.

In the U.S., some current guidance on waste isolation can be found in the Code of Federal Regulations, particularly 40 CFR 191 (Ref. 11) and 10 CFR 61 (Ref. 12). 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 from isolation during the first 10,000 years. These limits, along with the activity in the spent LWR fuel (Reference 1), are shown in Figure 8. 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 requiring isolation. 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 would be unnecessary. If one could remove all but 1 part in 100,000 of the problem isotopes, then one could meet the current 40 CFR 191 requirements without containment.

The other limit, from 10 CFR 61, is for low level wastes, and because spent fuel is classified high level waste and cannot be re-classified it does not strictly apply. However, it provides an indicator of how good is good enough. If we repeat 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. From this comparison, it appears that partitioning losses below 1 part in 100,000 may be required for the transuranic elements.

Thus, it appears that if the system partitioning losses are in the range of 10^{-5} or lower for TRU wastes, the burden of waste isolation placed on the repository is fundamentally changed. This then could be the objective of partitioning and transmutation scheme.

It should be noted that, although the regulatory guidelines are currently much less restrictive regarding the fission products, there are other considerations, such as heat load and "risk assessment" that will restrict the flow-through of certain fission products (thus, our intention to transmute iodine and technetium, as indicated in Figure 4). However, preliminary analysis indicates that partitioning losses higher than 10^{-5} would be acceptable for the fission products.

System Losses vs. Chemical Losses

An idealized waste partitioning and transmutation system is illustrated in Figure 9. However, there would be partitioning losses, and these would allow some of the long-lived wastes to slip through during 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, depend on the amount transmuted per pass through the machine, as shown in Figure 10. Note that for a 90% per pass transmutation machine, the net system partitioning losses are very close to the chemical partitioning losses. However, with a 10% per pass transmutation machine, one needs chemical partitioning losses near 10^{-6} to get a net partitioning loss near 10^{-5}.

The Objective and Current Capabilities

From the materials shown in Figures 8 through 10, it appears that chemical partitioning losses in the range of 10^{-6} may be needed to achieve the goal of fundamentally affecting waste management and disposal practices. Unfortunately, this objective could be made more restrictive if the release limits are tightened.

Are such chemical separations possible? According to the 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. 13).

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 disposal risks. 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. 14]. 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 desirable, provided the necessary research and development are pursued.

At this time, we believe that the principle challenge is to reduce system partitioning losses to the degree where the advantages for long-term waste isolation are clear. While the burden of proof now passes to the separation chemists and advanced reprocessing technologist, it appears that the required partitioning technology may become available within the next few years. When the partitioning technology has 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

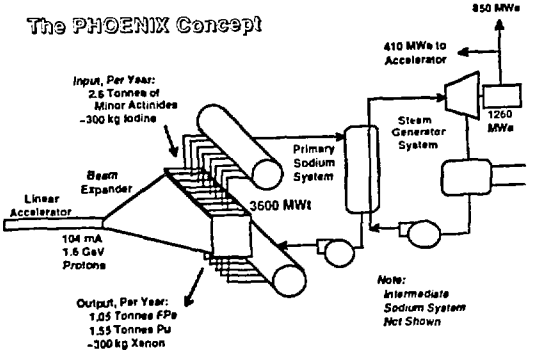


Figure 2. Impact of Partial Waste Processing

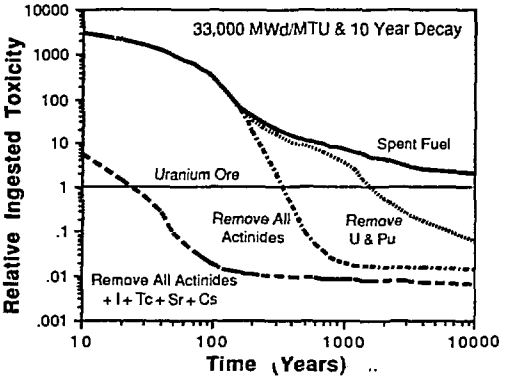


Figure 3. Transmutation Chains for Iodine and Technetium

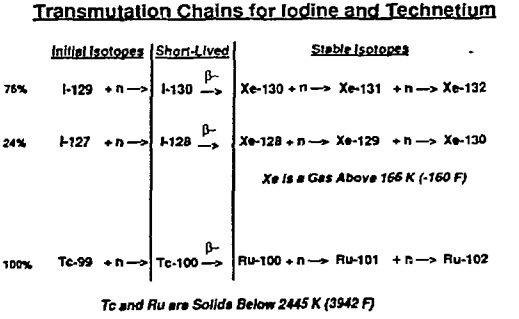


Figure 4. PHOENIX Transmutes Minor Actinides and Iodine

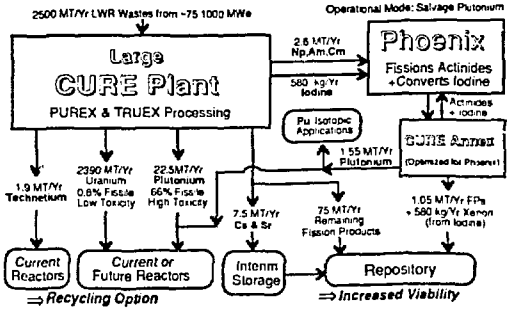


Figure 5. Burn-up Analysis for 3600 MW_t Target

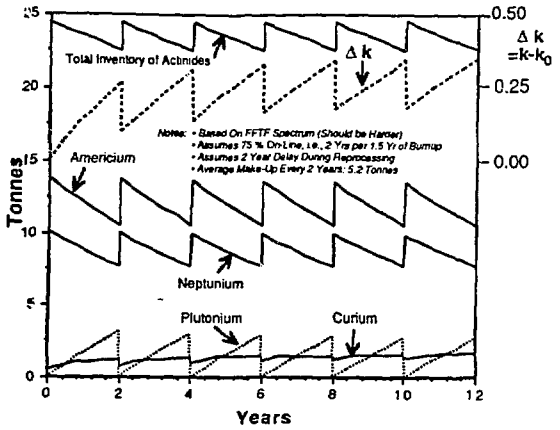


Figure 6. PHOENIX 450 MWt Sub-Critical Module

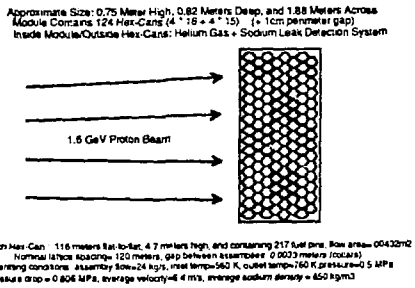


Figure 7. PHOENIX Target Chamber from Three Perspectives

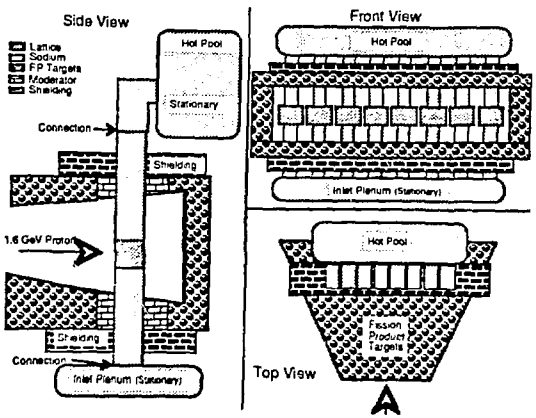


Figure 8. Waste Inventory vs. Release Limits Over the 10,000 Years After Disposal [Limits Specified in 40 CFR 191]

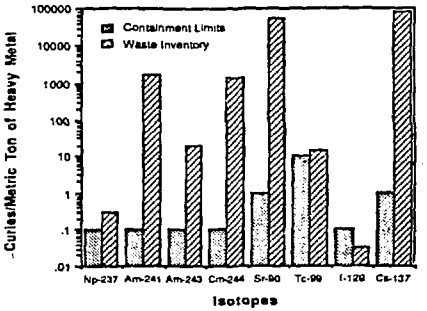


Figure 9. Waste Flows Through Partition and Transmutation System

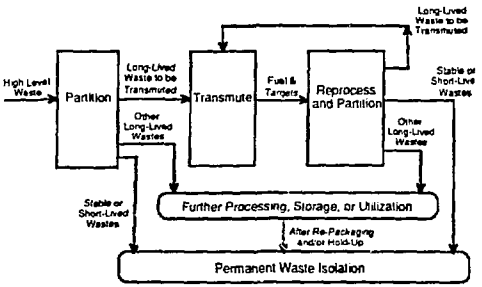
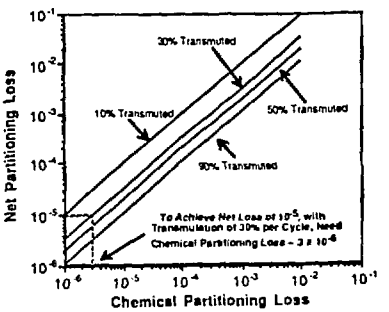


Figure 10. Net Partitioning Loss for Different Levels of Transmutation per Cycle



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