THE ONCE-THROUGH, HELIUM-COOLED CYCLE FOR
SECURE, CLEAN, SAFE, AND ECONOMICAL
TRANSMUTATION

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THE ONCE-THROUGH, HELIUM-COOLED CYCLE
FOR SECURE, CLEAN, SAFE, AND ECONOMICAL TRANSMUTATION

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ABSTRACT

Nuclear waste from commercial power plants contains plutonium, other actinides, and long-lived fission products. Based on current levels of global nuclear power generation, it has been estimated that by 2015 there will be more than 250,000 tons of spent fuel worldwide, containing over 2,000 tons of plutonium.

The main concerns related to the disposal of this waste are the potential for radiation release and exposure from the waste, and the possible diversion of fissionable material.

Transmutation of waste by fission and capture in nuclear reactors (under critical conditions) is a process that is well understood, and has long been considered a potentially beneficial element in the solution to the waste problem.

Recently, the development of high-power proton accelerators and targets raises the possibility of using a supply of spallation neutrons to extend fission and capture into subcritical operation, thus providing greater levels of transmutation than otherwise possible.

This paper describes the use of gas-cooled reactor and proton accelerator technologies for transmutation of nuclear waste in a once-through mode, without reprocessing after initial removal of fertile uranium and fission products from the waste. The proposed process uses a single type of transmuter that has the potential to eliminate essentially all weapons-useful material in the waste, and achieve a significant reduction in waste toxicity. It also has the potential to be economically attractive by generating significant amounts of electricity. Furthermore, the discharge from the process would be highly resistant to corrosion for long times.

A companion paper discusses the use of the same gas-cooled reactor and proton accelerator technologies in a “two strata” transmutation process that would use at least two different types of transmuters, possibly allowing even deeper levels of transmutation.

Transmutation of nuclear waste could have profound benefits for world political stability and the environment. It could drastically reduce the availability of weapons materials in the world, and reduce waste disposal requirements in terms of space and safeguarding time.

INTRODUCTION

Nuclear waste can be safely and securely stored in geologically stable repositories and allowed to decay for long times. However, technologies are becoming available that have the potential to add significant value to the disposal process. They provide for transmutation of nuclear waste into more stable materials that decay relatively fast and are not attractive for use in nuclear weapons, thus reducing long term toxicity and proliferation risks in the repositories.

THE PROBLEM

Nuclear fuel production begins with uranium ore, which is not without hazard as it contains some natural fission products and daughter elements that are created as uranium naturally decays to lighter elements. However, this naturally occurring material provides a useful benchmark for evaluating the nuclear waste that is ultimately produced. Uranium ore goes through several processing steps, including an enrichment step in which the fraction of the lighter uranium-235 is boosted from 0.7% to higher values (typically 3% for LWR reactor fuel) relative to the more naturally abundant uranium-238. As a fuel, the uranium is usually converted into oxide form (uranium oxide) and encased in a metal rod for use in LWRs.

In addition to the production of fission products, neutron capture in both uranium-235 and uranium-238 leads to the creation of plutonium, as well as minor actinides, (i.e., isotopes of elements with atomic number greater than 92), including neptunium, americium, and curium. The fuel is used until the uranium-235 content drops too low to sustain the chain reaction, around 0.8%. It is then moved to a spent-fuel water pool, where the hundreds of radioactive isotopes generated by the fission process begin to naturally decay away to stable, and generally harmless forms. After ten years of decay, spent nuclear reactor fuel is composed of the materials listed in Table 1.
Table 1. Spent Nuclear Reactor Fuel after 10 Years Decay

<table>
<thead>
<tr>
<th>Actinides</th>
<th>Fission Products</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium</td>
<td>95.6% Stable or Short-Lived 3%</td>
</tr>
<tr>
<td>Plutonium</td>
<td>0.9% Cesium &amp; Strontium 0.3%</td>
</tr>
<tr>
<td>Minor Actinides</td>
<td>0.1% Iodine &amp; Technetium 0.1%</td>
</tr>
</tbody>
</table>

An inspection of the table reveals that about 98.6% of the waste material is not of great concern: The uranium is no more hazardous than uranium ore, and can be separated from the other materials so that it meets Class C waste disposal standards. Unless the stable fission products happen to be chemically hazardous elements (e.g., mercury), they are also of little concern. That leaves the 1% that is composed of plutonium and minor actinides, as well as the 0.4% that is composed of cesium, strontium, iodine, and technetium, to be dealt with.

Plutonium is the unique waste component. It is fissionable, and capable of releasing significant amounts of energy. It is also a hazardous material, particularly if inhaled in particulate form. Because of its potential use in nuclear weapons, there is great sensitivity about isolating plutonium from other components of the nuclear waste stream. Minor actinides contain potential energy proportionate to plutonium, although their generally small thermal cross sections make them much more difficult to fission.

If the 1% of the reactor discharge that consists of plutonium and minor actinides is transmuted, the resultant waste stream would contain nearly 90% stable or short-lived fission products, and about 10% cesium, strontium, iodine, and technetium. This is a favorable trade-off, as a significant amount of energy would be produced, equal to about 18% of the energy that was produced in the reactor. Therefore, in transmuting the plutonium and minor actinides (i.e., Np, Am, and Cm) from the 100 nuclear power plants in operation in the U.S., one would be generating the equivalent to another 18 power plants worth of electric power.

But what of the 0.4% of problem fission products? The cesium and strontium problem is caused by a couple of isotopes having half-lives of about 30 years, which are hard to transmute. However, with 30-year half-lives, the inventory drops by a factor of nearly 10 every century, so four centuries of decay would drop the inventory by a factor of nearly 10,000. We can trust containers to provide isolation for that long, and the need for isolation thereafter is greatly diminished. In contrast, the technetium and iodine isotopes of concern are very long-lived and are primarily of concern well after the containers have ceased to be effective. Fortunately, the iodine and technetium isotopes of concern can be converted to stable isotopes if one has enough neutrons available. In that respect, we are fortunate that the transmutation of the plutonium and the minor actinides can provide many available neutrons.

The potential impact of removing and transmuting the plutonium and actinide wastes is illustrated in Figure 1. Note that after ten thousand years of storage, untreated nuclear reactor waste is still more than twice as hazardous as natural uranium ore, and continued isolation from the environment is still important. Furthermore, significant amounts of plutonium still remain in the untreated waste. In contrast, the step of transmuting the actinides offers the potential to make the waste stream less hazardous than uranium ore within three to four centuries, and reduce the need for isolation and safeguarding. Although this looks very attractive, it needs to be clarified that the real impact may be somewhat less dramatic than the figure shows since even some minor residual amounts of Plutonium and Minor Actinides will tend to make the treated waste toxicity curve cross the Natural Uranium Ore line farther to the right.

Figure 1. Impact of Removing & Transmuting Actinides.

GAS-COOLED SYSTEMS FOR TRANSMUTATION OF NUCLEAR WASTE

As stated above, if the 1% of the waste stream that is plutonium and minor actinides is transmuted, and the iodine and technetium isotopes of concern are also converted to stable isotopes, one has basic elements of a solution to the nuclear waste problem.

Let us consider some of these elements. In the transmutation process, the materials to be transmuted are irradiated in a neutron flux. Neutrons (1) can fission the atoms of the irradiated materials creating atoms of lower weight and generally more stable, or (2) are absorbed by the irradiated atoms. In the latter case, new heavier atoms are formed, which may in turn fission when hit by other neutrons. For the long-lived fission products, neutron absorption transmutes them to stable or short lived species.
Gas-cooled reactor technologies offer significant advantages in accomplishing this transmutation process. They are ideally suited for use with thermal neutron spectra since they allow operation at high temperatures and neutron energies that produce plutonium fission without the need for fertile material as a burnable control poison. In addition, they also are ideally suited for use with fast neutron spectra since they provide the hardest possible fast neutron environment for transmutation of higher actinides, which are more inclined to fission in the fast neutron energy spectra. This is due to the fact that the gas coolant is essentially transparent to neutrons and does not degrade the energy spectrum, as is the case with other coolants.

**Thermal Neutron Systems**

Generally both capture and fission cross-section for thermal neutrons are an order of magnitude larger than in a fast neutron spectrum. Thus, a suitably designed thermal system can destroy essentially all the proliferation-offensive Plutonium isotopes (Pu-239 and Pu-241). A helium-cooled, graphite moderated, thermal neutron energy spectrum assembly using ceramic-coated fuel, and operating as a critical system or as an accelerator-assisted subcritical system, is an attractive choice for this fission function for several reasons.

As shown in Figure 2, a Modular Helium Reactor (MHR) type assembly produces a relatively large flux in the thermal regime where fission cross sections are quite high. This promotes fission. In addition, the assembly operates in a temperature range (shown in Figure 2) in which the capture cross-section of erbium has a resonance at a neutron energy such that it can be used as a burnable poison to produce a strong negative temperature coefficient of reactivity. The lack of interaction of the helium with neutrons means that temperature feedback is the only significant contributor to the power coefficient. This provides for a stable operation of the reactor or subcritical assembly. In addition, it does not require U-238 as a burnable poison so that no additional plutonium is produced in the process.

Another feature of great importance in the thermal gas-cooled reactor or subcritical assembly is the use of ceramic-coated "TRISO" fuel particles. The ceramic materials are stable at high temperatures, and have very high melting points. This provides large thermal margins to ensure fuel integrity during loss of coolant events. Moreover, the coated particles are nearly spherical, and include large gas expansion volumes within the coated particles. The expansion volumes are able to accommodate the production of fission gas products within the coated particles with lower resultant internal pressures. In addition, the spherical shape is better able to withstand the mechanical stresses due to these pressures. The composite effect is that the particles can tolerate high levels of irradiation, and allow deeper levels of transmutation (burnup) without reprocessing. This capability has been demonstrated in multiple reactor irradiations.

![Figure 2. Neutron Flux Distribution and Cross-sections of Plutonium and Erbium in Gas-cooled Assembly](image)

An important advantage of the ceramic coatings is that they are much more durable than metallic coatings, and provide better reassurance of waste integrity in the repository. Extrapolated corrosion test results indicate that the incremental waste exposure in the repository due to corrosion of the ceramic coatings is expected to be negligible for hundreds of thousands of years (Figure 3).

![Figure 3. Particle Integrity](image)

Given these important features, the use of gas-cooled, graphite-moderated ceramic-coated-fuel thermal reactors, or accelerator-driven subcritical assemblies of the same type for destroying weapons grade plutonium has been studied in detail. The studies have led to the conclusion that an assembly operating as a critical system can transmute about 90% of Pu-
239, and 65% of a total load of Pu in a three-year pass. Then, if the 3-year irradiated load is further irradiated in an accelerator-driven subcritical assembly for one more year, the destruction of Pu-239 and total Pu increases to 99.9% and 87% respectively, with no intermediate reprocessing.

**Fast Neutron Systems**

Fast reactor systems are typically much smaller than thermal reactors since they need a high neutron flux and no moderator, and typically have higher fuel densities. This leads to higher power densities, more demanding cooling requirements, and more complex cooling and cooling control systems. The smaller delayed neutron fractions and complex reactivity feedback effects in fast neutron systems can potentially produce severe reactivity and heating effects.

The fission cross-sections in the fast-neutron region are smaller than in the thermal region; however, fission-to-absorption ratios are higher in the fast-neutron region than they are in the thermal region. So, even though many minor actinides are hard to fission at any energy level because of their low cross sections, their relative destruction rates are better than in thermal reactors, especially if a high neutron flux is provided.

Calculations show that a fast subcritical assembly cooled with helium gas allows a destruction rate of actinides of approximately 26% (in weight) per year. This is somewhat better than has been reported in other studies for liquid metal cooled systems, maybe because the gas coolant allows the production of a harder neutron energy spectrum, and consequently, a higher fission to absorption ratio. Given this rate of destruction, one could irradiate the actinides for several years (at 26% destruction per year) and then store it in a geological repository where further natural decay would take place. So, if one desires to destroy certain minor actinides, there is merit in using fast neutrons after plutonium is transmuted in a thermal spectrum and fast subcritical regime. And if one wishes to do so, it helps that the amount of actinides is a very small portion of the initial waste (0.1%) since the fast transmutation assemblies end up being smaller than they would have to be to transmute plutonium as well. These findings form the basis for a transmutation scheme utilizing both fast and thermal neutron spectra as discussed in the next section.

**THE THERMAL-FAST SOLUTION**

Based on the above considerations, we have explored the process of using (1) thermal neutrons (near the cross-section resonance peak) to do what they do best, i.e.: fission plutonium, and (2) fast neutrons to fission minor actinides. We call this the thermal-fast process. One advantage of using this process is that most of the transmutation of plutonium is done in the thermal regime where technologies are more mature, and development risks are lower. Since the amounts of minor actinides found in the waste material are significantly lower than the amounts of plutonium, it follows that the fast assemblies needed can be significantly smaller than the thermal assemblies.

The fuel cycle that we have studied for such scheme is as described in the previous section: Three years of transmutation of plutonium and minor actinides in a thermal neutron spectrum assembly operating in the critical mode, followed by one year of transmutation in the same thermal neutron spectrum assembly without reprocessing, operating as an accelerator-assisted subcritical system. At this point, essentially all fissile materials are burned up. What remains is mainly non-fissile minor actinides, which are moved to a fast neutron spectrum assembly operating as a subcritical system.

Burnup calculation results for this fuel cycle are shown in figure 4 for an initial 1,000-Kg charge of weapons-grade plutonium. As the figure indicates, most of the Pu transmutation is accomplished in a thermal critical regime. When this is followed by a one-year irradiation step in a thermal subcritical regime (accelerator-driven), essentially all Pu is gone. At this time, a three-year step of transmutation in a fast subcritical regime leads to Point C in the chart, when most of the initial charge is gone. If this remaining material is placed in a repository for 200 years, only 60 out of 1000 Kg of the initial charge are left.

![Figure 4. Incremental Transmutation in the Thermal-Fast Process](image-url)
239, and corresponds to the Point A-to-Point B trajectory in Figure 4. Two irradiated “TRISO” particles from this test are shown in Figure 5. The second (lighter) layer (from the outside in) is the Silicon Carbide coating. The two gray layers on each side of the Silicon Carbide layers are pyrocarbon layers that provide mechanical protection and pre-compression for very high structural strength margins in the silicon carbide. The central part of the particle contains the transmuted material. The dark areas within the Silicon Carbide layer are empty spaces occupied by fission gasses.

Figure 5. Irradiated TRISO Particles.

Pu Oxide
747,000 MW-days/tonne
>95% 
Pu Transmuted

Th-Pu Oxide
183,000 MW-days/tonne
>95% 
Pu Transmuted

The conclusions from the burnup levels shown in figure 4 and the durability of the silicon carbide “TRISO” particles illustrated in figure 3 are that (1) transmutation can reduce plutonium and minor actinides waste by about two orders of magnitude, and (2) the transmuted material will remain isolated from the repository environment for hundreds of thousands of years.

These are important potential environmental benefits of transmutation using gas-cooled reactor technologies.

Consider now how the manner in which the thermal and fast neutron energy spectra can be packaged together in an integrated assembly shown in figures 6 and 7.

Referring to these figures, the transmutation assembly consists of a steel vessel housing, inside of which there is an annular nuclear transmutation region that operates in the thermal neutron energy regime. In this annular region, plutonium and minor actinides from LWR waste are fissioned together in TRISO particles. Most of the mixture, about 90%, is plutonium. The remaining 10% is minor actinides. Fission neutrons in this annular region are thermalized in graphite blocks in which the TRISO particles are contained as shown in figure 8. Surrounding the annular thermal region there is an inner and an outer graphite neutron reflector. This thermal region operates in the critical mode for 75% of its cycle time, followed by operation in an accelerator-assisted subcritical mode for the remaining 25%.

Still referring to figures 6 and 7, in the center of the inner reflector there is a cylindrical region, approximately 15% of the size of the active thermal region, that operates in the fast energy neutron energy regime. This region consists of tungsten tubes that house TRISO particles already transmuted in the thermal region. Therefore, they contain mainly minor actinides. The main motivation for including this fast assembly inside of the thermal assembly is to take advantage in the fast fission process of the large heat storage and conduction heat removal capabilities of the thermal assembly.

The fast cylindrical region is designed so that, by itself, it is subcritical. However, neutrons reaching it by traveling from the thermal region through the reflector can cause fission and get amplified, thus creating subcritical transmutation.

As discussed above, the transmuter operates in the critical mode for approximately three years, which corresponds to 75% of its cycle time. In this mode, the fission process is driven by the critical reaction in the thermal region. After that, the
thermal region becomes subcritical, and is then driven for a fourth year to cause deep levels of transmutation by neutrons generated in a spallation target located in the center of the fast region. The target is driven by the proton beam illustrated in figures 6 and 7. Deep levels of transmutation can be achieved with no reprocessing thanks to the encapsulation in ceramic-coated microspheres of the materials to be transmuted, which accommodate the production of fission gas products within Helium then flows downward through cooling channels in the fission regions of the transmuter, and carries the heat at a temperature of 850 degrees C through the central part of the coaxial duct to a direct-cycle gas-turbine-generator system that generates electricity. The high operating temperatures and the characteristics of this direct (Brayton) power conversion system allow electric generation with a high net thermal efficiency of approximately 47%

Figure 8 TRISO Coatings and Graphite are Excellent Engineered Barriers for Normal Operation, Severe Accidents, and Permanent Disposal

internal expansion volumes.
A beneficial anti-proliferation effect of including the fast assembly within the thermal assembly is that the neutron economy in the integrated assembly cannot support breeding.

Going back to operating sequence, the fact that the transmuter needs the proton beam for only 25% of its operating time makes the entire process more economical because the accelerator can be time-shared by four transmuters in the plant configuration illustrated in Figure 9.

Still referring to figures 6 and 7, there is shown a coaxial duct in the lower part of the transmuter. The outer part of the duct brings in cold cooling helium to remove fission heat from the transmuter. It flows upward in an annular space between the inside of the vessel and the outside of a steel barrel that contains the thermal-fast assembly.

This high power conversion efficiency, the fact that 75% of the transmutation is done in a critical operating mode, and the fact that the proton accelerator is time-shared by four transmuters, leads to a favorable revenue-cost balance and the potential to attract private investment for the deployment of these units.

Roughly, the cost of the plant configured as shown in figure 9 (four 600 MWth transmuters, and one 15 MW beam accelerator) would be expected to be in the $1.5B to 2.0B range. This would translate into an annual cost of $190M assuming interest on and return of capital of 8.8%, plus typical nuclear plant operations and maintenance (O&M) costs increased by 50% to account for accelerator O&M. It also includes an allowance for decommissioning the plant, but assumes, however, that the fuel (to be transmuted) is government-supplied.
### Operating Sequence

<table>
<thead>
<tr>
<th>Time</th>
<th>Transmitter 1</th>
<th>Transmitter 2</th>
<th>Transmitter 3</th>
<th>Transmitter 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>$t_0$</td>
<td>Critical</td>
<td>Critical</td>
<td>Critical</td>
<td>Subcritical</td>
</tr>
<tr>
<td>$t_0 + 1$ yr</td>
<td>Critical</td>
<td>Critical</td>
<td>Subcritical</td>
<td>Critical</td>
</tr>
<tr>
<td>$t_0 + 2$ yrs</td>
<td>Critical</td>
<td>Critical</td>
<td>Critical</td>
<td>Critical</td>
</tr>
<tr>
<td>$t_0 + 3$ yrs</td>
<td>Subcritical</td>
<td>Critical</td>
<td>Critical</td>
<td>Critical</td>
</tr>
</tbody>
</table>

After the 1 year of subcritical operation:
- Fresh fuel goes into thermal sector
- Fuel compacts irradiated for 4 years in thermal sector are moved to fast sector

![Diagram of Plant Configuration](image)

**Figure 9. Plant Configuration**

The revenues, assuming 4 cents per kWe-hr, 75% plant availability, 47% transmuter thermal efficiency, and an accelerator efficiency (beam to electric power ratio) of 32% would be approximately $270M per year.

These estimates suggest that transmutation plants of this type have the potential to be economically viable under private financing.

From the safety standpoint, there are two important considerations. First, criticality. Safety in this respect is ensured by the use of erbium in the thermal assembly. Erbium has a neutron capture cross section that increases with temperature, and peaks a higher temperature than the fission cross section of plutonium 239. Thus, they are fissioned in a thermal fission region of the transmuter. Other actinides, particularly even-numbered isotopes, are more inclined to fission in a fast neutron energy spectrum. Thus, they are fissioned in a fast fission region of the transmuter.

The other important safety consideration is cooling. In this respect, the geometry of the assembly (tall and thin, and annular thermal configuration) has been shown to provide for passively safe conduction cooldown of a 600 MWh thermal spectrum-only assembly, even in a loss of coolant (LOCA) event. The effect on this feature by the inclusion of a fast fission region is currently being studied. However, preliminary conservative calculations suggest that including a fast region may still allow passive conduction cooling in a LOCA event if thermal region power is lowered so that total power of the transmuter is kept below 600 MWh.

Thus, it appears that levels of safety comparable to those that are encountered in gas-cooled reactors may also be achievable in the gas-cooled transmuters.

### CONCLUSIONS

Gas-cooled nuclear reactor technology offers the potential to eliminate essentially all weapons-useful material in nuclear waste, and achieve more than two orders of magnitude reduction in the amount of high-level waste. Repository heat loads and the toxicity of the waste are also significantly reduced. The process provides a transmuted waste form that is highly resistant to corrosion and durable, without generating mixed waste.

The process uses thermal and fast neutron energy spectra. Pu 239 and other fissionable materials have large fission cross-sections in the thermal spectrum. Thus, they are fissioned in a thermal fission region of the transmuter. Other actinides, particularly even-numbered isotopes, are more inclined to fission in a fast neutron energy spectrum. Thus, they are fissioned in a fast fission region of the transmuter.

The process transmutes about 75% of the waste in a nuclear critical mode. Then, use is made of a proton accelerator to generate spallation neutrons and drive the fission process in a subcritical mode to deep levels of burnup. Most importantly, these deep burnup levels are achieved with no plutonium reprocessing. This is made possible by encapsulating the waste to be transmuted in ceramic-coated microspheres that accommodate large amounts of fission products in spherical expansion volumes.

Deep burnup of plutonium 239 and fissionable materials with no plutonium reprocessing, and the use of the thermal region in the transmuter, which precludes breeding, are important proliferation resistance features of the proposed process.

Preliminary calculations suggest that the unique reactivity and cooling safety features offered by gas-cooled nuclear reactors can also be implemented in the proposed transmuter.

The use of a direct-cycle gas turbine-generator power conversion system with the proposed transmuter would lead to conversion efficiencies of approximately 47% when the transmuter is operating in the critical mode. This, along with the fact that the accelerator is only used for the 25% deep burnup phase of the cycle leads to a relatively high overall efficiency and low cost. Preliminary economic analyses suggest that the proposed transmutation process has the potential to be economically viable and attract private investment for deployment.