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NUCLEAR WASTE DISPOSAL UTILIZING A GASEOUS CORE REACTOR

by

Richard R. Paternoster

ABSTRACT

A feasibility study was undertaken of a gaseous core nuclear reactor designed to produce power to also reduce the national inventories of long-lived reactor waste products through nuclear transmutation. Neutron-induced transmutation of radioactive wastes can be an effective means of shortening the apparent half life.
NUCLEAR WASTE DISPOSAL UTILIZING A GASEOUS CORE REACTOR

Introduction

A major objection to the increased use of nuclear electric-power generation stems from envisioned difficulties related to the disposal and management of hazardous radioactive wastes. The vast majority of radioactive wastes are short-lived and easily managed with present waste disposal management techniques. However, several isotopes present in nuclear fission reactor wastes have half-lives of several thousands to several millions of years. These problem wastes are the main point of focus in the debate over disposal of nuclear wastes.

Ultimate disposal of the extremely long-lived wastes could reduce the waste disposal problem to humanly controllable time scales. Removal of long-lived wastes from the total accumulated waste would be the first step in this direction. Separation of iodine-129, the trans-uranium actinides, and other long-lived isotopes could be accomplished at several different stages in the waste disposal cycle. This could mean extraction at the fuel reprocessing site, after a five to ten year cool-down period, or after a 20 to 30 year decay interval. These separated long-lived wastes would be disposed of by methods which would completely remove them from the biosphere and hence any further consideration. Extraterrestrial transport of nuclear wastes (ETT) may become feasible in light of recent advances in the space program. Ultimate disposal into deep space or the sun is a remote possibility. The only other presently known method of ultimate disposal is through the process of neutron-induced nuclear transmutation. Induced nuclear transmutation of radioactive wastes converts long-lived problem wastes into relatively harmless short-lived or stable isotopes.
The concept of neutron-induced transmutation of radioactive wastes was first suggested by Steinberg et al. [1] as a means of effectively shortening the half-lives of waste Kr$^{85}$, Sr$^{90}$, and Cs$^{137}$. This would be accomplished in a neutron flux through the reactions

$$\text{Kr}^{85}(n,\gamma)\text{Kr}^{86} (\text{stable})$$

$$\text{Sr}^{90}(n,\gamma)\text{Sr}^{91} \rightarrow \text{Y}^{91} \rightarrow \text{Zr}^{91} (\text{stable})$$

$$\text{Cs}^{137}(n,\gamma)\text{Cs}^{138} \rightarrow \text{Ba}^{138} (\text{stable})$$

In examining the neutron capture cross sections for those reactions they concluded that neutron fluxes approaching $10^{16}$ n/cm$^2$sec would be necessary for shortening the half-lives appreciably. They further suggested that the most logical approach would be through the use of dual-purpose facilities for power production and waste transmutation. In such a scheme the major cost of waste burning, namely the cost of producing neutrons, is written off against the cost of electric power generation.

Wolkenhauer [2] has studied transmutation of high-yield fission products Sr$^{90}$ and Cs$^{137}$ in a controlled thermonuclear reactor (CTR) blanket. Gore and Leonard [3] have also studied transmutation of massive loadings of Cs$^{137}$ in CTR blankets with special flux trap designs. Cs$^{137}$ represents the most difficult case for this technique because it has the lowest thermal neutron capture cross section of the moderately long-lived fission products. These schemes appear attractive since the CTR power plant operating on a deuterium-tritium fuel cycle would produce an excess of neutrons. Within a few decades such systems may be found technically and economically feasible.
Claiborne [4] has considered disposal by transmutation of transuranium actinide wastes. Most of those long-lived alpha emitters have appreciable neutron capture and/or fission cross sections. Table 1 gives the capture and fission cross sections at thermal and resonance energies used by Claiborne. Actinide transmutation occurs by direct fission such as

\[ \text{Am}^{242m}(n,f) \]

and

\[ \text{Am}^{244}(n,f) \]

or by neutron capture followed by fission such as

\[ \text{Am}^{241}(n,\gamma)\text{Am}^{242}(n,f) \]

and

\[ \text{Am}^{243}(n,\gamma)\text{Am}^{244}(n,f) \]

In this scheme, those long-lived actinides are continuously recycled back into power reactor fuel elements and in this manner burned out. At fluxes presently available in power reactors \((10^{13} - 5 \times 10^{13} \text{ n/cm}^2\text{ sec})\) this would be a somewhat slow process spanning over several decades. Furthermore, serious problems arise in handling and safeguarding the recycled actinides during fabrication of fresh fuel elements.

**Requirements for Transmutation of Radioactive Waste**

Given a high enough neutron flux or a long enough period of irradiation any isotope with a non-zero neutron capture cross section can be transformed into other isotopes. However, the transmutation of radioactive waste imposes a number of constraints upon the process.
TABLE 1. THERMAL CROSS SECTIONS AND RESONANCE INTEGRALS FOR CAPTURE AND FISSION IN THE TRANSPLUTONIUM ACTINIDES

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Thermal Cross Sections (barns)</th>
<th>Resonance Integrals (barns)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Capture</td>
<td>Fission</td>
</tr>
<tr>
<td>95\textsuperscript{Am} \textsuperscript{241}</td>
<td>925</td>
<td>3.1</td>
</tr>
<tr>
<td>95\textsuperscript{Am} \textsuperscript{242m}</td>
<td>2000</td>
<td>6000</td>
</tr>
<tr>
<td>95\textsuperscript{Am} \textsuperscript{242}</td>
<td>0</td>
<td>2900</td>
</tr>
<tr>
<td>95\textsuperscript{Am} \textsuperscript{243}</td>
<td>105</td>
<td>.5</td>
</tr>
<tr>
<td>95\textsuperscript{Am} \textsuperscript{244}</td>
<td>0</td>
<td>2300</td>
</tr>
<tr>
<td>95\textsuperscript{Am} \textsuperscript{245}</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>96\textsuperscript{Am} \textsuperscript{242}</td>
<td>30</td>
<td>5</td>
</tr>
<tr>
<td>96\textsuperscript{Am} \textsuperscript{243}</td>
<td>200</td>
<td>600</td>
</tr>
<tr>
<td>96\textsuperscript{Cm} \textsuperscript{244}</td>
<td>10</td>
<td>1.2</td>
</tr>
<tr>
<td>96\textsuperscript{Cm} \textsuperscript{245}</td>
<td>343</td>
<td>1727</td>
</tr>
<tr>
<td>96\textsuperscript{Cm} \textsuperscript{246}</td>
<td>1.3</td>
<td>0</td>
</tr>
<tr>
<td>96\textsuperscript{Cm} \textsuperscript{247}</td>
<td>60</td>
<td>120</td>
</tr>
<tr>
<td>96\textsuperscript{Cm} \textsuperscript{248}</td>
<td>3.6</td>
<td>0</td>
</tr>
<tr>
<td>96\textsuperscript{Cm} \textsuperscript{249}</td>
<td>2.8</td>
<td>50</td>
</tr>
<tr>
<td>96\textsuperscript{Cm} \textsuperscript{250}</td>
<td>2</td>
<td>0</td>
</tr>
<tr>
<td>97\textsuperscript{Bk} \textsuperscript{249}</td>
<td>1450</td>
<td>0</td>
</tr>
<tr>
<td>97\textsuperscript{Bk} \textsuperscript{250}</td>
<td>350</td>
<td>3000</td>
</tr>
<tr>
<td>98\textsuperscript{Cf} \textsuperscript{249}</td>
<td>450</td>
<td>1690</td>
</tr>
<tr>
<td>98\textsuperscript{Cf} \textsuperscript{250}</td>
<td>1900</td>
<td>0</td>
</tr>
<tr>
<td>98\textsuperscript{Cf} \textsuperscript{251}</td>
<td>2850</td>
<td>3750</td>
</tr>
<tr>
<td>98\textsuperscript{Cf} \textsuperscript{252}</td>
<td>19.8</td>
<td>32</td>
</tr>
<tr>
<td>98\textsuperscript{Cf} \textsuperscript{253}</td>
<td>12.6</td>
<td>1300</td>
</tr>
<tr>
<td>98\textsuperscript{Cf} \textsuperscript{254}</td>
<td>50</td>
<td>0</td>
</tr>
</tbody>
</table>
The selection of prospective candidates for waste transmutation must begin with the radioactive wastes themselves. Reaction rates for neutron-induced transmutation processes can be specified in terms of neutron capture and/or fission cross sections. Unfortunately, high-yield fission product wastes have small neutron capture cross sections. An example is the capture cross section for the (n,γ) reaction in 10.7 year half-life Kr\(^{85}\). At the time of Steinberg's study [1], the thermal neutron capture cross section of Kr\(^{85}\) was believed to be 15 barns. Using this figure it was concluded that disposal by transmutation at Kr\(^{85}\) could be economically and technically feasible. A recent measurement [5] of this cross section found it to be 1.66 ± 0.20 b. Thus, an order of magnitude increase in the fluence is required to reach feasible transmutation levels.

Another criterion for neutron-induced transmutation of radioactive waste is isotropic purity. If maximum utilization of available neutrons is to be achieved isotopic impurities of a given waste isotope must be eliminated or reduced to lowest possible content. In the case of Kr\(^{85}\) transmutation, three stable krypton isotopes, Kr\(^{83}\), Kr\(^{84}\), and Kr\(^{86}\) are present with Kr\(^{85}\) in fission wastes. The low concentration of Kr\(^{85}\) in krypton wastes (7.2 percent) and its low capture cross section compared with Kr\(^{83}\) (\(\sigma_c = 2.5\) barns) makes enrichment of Kr\(^{85}\) necessary.

A final requirement for successful transmutation of radioactive waste is that the end product nuclide of the transmutation process be no more hazardous than the initial waste product. The ideal end product of a transmutation process would be a stable nuclide with a zero neutron capture cross section. A nearly ideal nuclide in this respect is waste I\(^{129}\) (\(t_{1/2} = 15.9 \times 10^6\) years). The transmutation reactions in this case are
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Thus even with several reaction paths the vast majority of \(^{129}\)I will be converted to stable isotopes of xenon. The situation is more complex in the case of actinide waste transmutation. In a sustained irradiation of waste actinides, such as curium, quantities of higher actinides, such as berkelium and californium will inevitably be formed. These higher actinides are generally alpha emitters with varying half-lives.

**Transmutation Systems and Waste Disposal**

The transmutation of \(^{129}\)I is first examined in the following. After a 160-day decay period, the short-lived iodine isotopes \(^{128}\)I, \(^{130}\)I, \(^{131}\)I, \(^{132}\)I and \(^{133}\)I decay away leaving stable \(^{127}\)I and the 15.9 \(\times\) 10\(^6\) year half-life, \(^{129}\)I. The spent fuel from present day LWRs contains about 40 grams/MTU of \(^{127}\)I and 231 grams/MTU of \(^{129}\)I at 160 days after discharge and 33,000 MWD/MTU burnup. These two isotopes constitute roughly 0.78 percent of the total fission product wastes with the \(^{129}\)I isotope being about 85 percent of this amount.

The proposed means for disposal of \(^{129}\)I by induced nuclear transmutation is by neutron capture to \(^{130}\)I which beta decays with a 12.4 hour half-life to stable Xe\(^{130}\). The thermal neutron capture cross section for the \((n,\gamma)\) reaction in \(^{129}\)I is 28 barns and the resonance integral is 50.4 barns. The remaining 15 percent of LWR iodine wastes is stable \(^{127}\)I with a thermal neutron absorption cross section of 6.2 barns and a resonance integral of 177 barns.
A schematic of the proposed system is shown in Figure 1. The 85 percent $^{129}$-15 percent $^{127}$ mixture available directly from the fuel reprocessing plant as liquid I$_2$ would be enriched to 95 percent $^{129}$ in an isotope enrichment facility. A laser isotope separation facility would be ideally suited for such a small scale operation. The enriched $^{129}$ would be inserted in the core of the power-burner reactor in a suitable concentration. The $^{129}$ upon capturing a neutron and decaying to stable xenon isotopes, primarily Xe$^{130}$ could be harmlessly vented to the atmosphere or sold commercially.

The specification of $^{129}$ enrichment is optional, but desirable from the standpoint of maximum neutron utilization for $^{129}$ burning. If 100 kg of unenriched iodine waste (85 percent $^{129}$-15 percent $^{127}$) is initially loaded for transmutation and exposed to a thermal neutron fluence such that 50 kg of $^{129}$ are transmuted, then about 2.6 kg of stable $^{127}$ are also transmuted to stable Xe$^{127}$. Irradiating 95 percent $^{129}$ enriched iodine in the same fluence results in the transmutation of 55 kg of $^{129}$ and only 0.9 kg of $^{127}$.

The actinide transmutation system would involve loading of several waste isotopes simultaneously for transmutation. The actinide wastes considered here are the isotopes of americium and curium. The amounts of the significant isotopes present in LWR fuel wastes following a ten year decay period are shown in Table 2. The two prevalent isotopes are Am$^{241}$ and Am$^{243}$. The most important isotope present in curium wastes is the 18.1 year half-life Cm$^{244}$. A decay interval of ten years after discharge from the reactor is reasonable to allow the wastes to cool down. Following this cool-down period the wastes would be loaded into the power-burner reactor for transmutation. After irradiation the transmuted waste would be removed from the reactor and stored to allow decay of short-lived actinides. Following this cool-down period the fission product wastes would be chemically removed and the remaining
Figure 1. Pathway for $^{129}$ Waste Transmutation in the Gaseous-Core Reactor.
TABLE 2. RELATIVE AND ABSOLUTE AMOUNTS OF AMERICIUM AND CURIUM ISOTOPES PRESENT IN AMERICIUM AND CURIUM WASTES FROM ATLANTIC 1 REFERENCE LWR FUEL AT TEN YEARS AFTER DISCHARGE AT 33,000 Mwd/MTU BURNUP

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Concentration in Spent Fuel Grams/MTU</th>
<th>Grams in Spent Fuel Load</th>
<th>Percent of Isotope Present in Element</th>
</tr>
</thead>
<tbody>
<tr>
<td>Am$^{241}$</td>
<td>48.3</td>
<td>4166.84</td>
<td>32.37</td>
</tr>
<tr>
<td>Am$^{242m}$</td>
<td>.92</td>
<td>79.37</td>
<td>.62</td>
</tr>
<tr>
<td>Am$^{242}$</td>
<td>$1.10 \times 10^{-5}$</td>
<td>.001</td>
<td>Insignificant***</td>
</tr>
<tr>
<td>Am$^{243}$</td>
<td>100</td>
<td>8627.0</td>
<td>67.01</td>
</tr>
<tr>
<td>Am$^{244}$</td>
<td>Insignificant*</td>
<td>Insignificant**</td>
<td>Insignificant***</td>
</tr>
<tr>
<td>Am$^{245}$</td>
<td>Insignificant*</td>
<td>Insignificant**</td>
<td>Insignificant***</td>
</tr>
<tr>
<td>Total Am</td>
<td>149.22</td>
<td>12873.21</td>
<td>100</td>
</tr>
<tr>
<td>Cm$^{242}$</td>
<td>$2.21 \times 10^{-3}$</td>
<td>.191</td>
<td>Insignificant***</td>
</tr>
<tr>
<td>Cm$^{243}$</td>
<td>.0679</td>
<td>5.86</td>
<td>.28</td>
</tr>
<tr>
<td>Cm$^{244}$</td>
<td>22.1</td>
<td>1906.57</td>
<td>89.98</td>
</tr>
<tr>
<td>Cm$^{245}$</td>
<td>2.14</td>
<td>184.62</td>
<td>8.72</td>
</tr>
<tr>
<td>Cm$^{246}$</td>
<td>.251</td>
<td>21.65</td>
<td>1.02</td>
</tr>
<tr>
<td>Cm$^{247}$</td>
<td>$3.31 \times 10^{-3}$</td>
<td>.286</td>
<td>Insignificant***</td>
</tr>
<tr>
<td>Cm$^{248}$</td>
<td>$2.29 \times 10^{-4}$</td>
<td>.0198</td>
<td>Insignificant***</td>
</tr>
<tr>
<td>Cm$^{249}$</td>
<td>Insignificant*</td>
<td>Insignificant**</td>
<td>Insignificant***</td>
</tr>
<tr>
<td>Cm$^{250}$</td>
<td>Insignificant*</td>
<td>Insignificant**</td>
<td>Insignificant***</td>
</tr>
<tr>
<td>Total Cm</td>
<td>24.56</td>
<td>2119.20</td>
<td>100</td>
</tr>
</tbody>
</table>

*Denotes $\leq 10^{-5}$ grams/MTU.
**Denotes $\leq 10^{-3}$ grams.
***Denotes $\leq 0.1\%$. 
actinides would be recycled into the next loading of waste. A schematic of the actinide transmutation system is shown in Figure 2.

THE GASEOUS-CORE NUCLEAR REACTOR CONCEPT

Gaseous-fueled, externally moderated cavity reactors have been under theoretical investigation in the United States since about 1954. The primary motivation for developing the gaseous-core reactor has been for high specific-impulse nuclear rocket propulsion. The two configurations that have received the most attention are: (1) the open cycle concept, in which the fissioning fuel is partially confined in the core by vortex-generated hydrodynamic forces, and (2) the closed cycle or nuclear light bulb concept, where the fuel is physically contained within a thin, radiation-transparent wall and recirculated for removal of fission product poisons. Uranium-235 fueled core regions ranging from 0.5 to four meters in diameter surrounded by reflector-moderator regions of practical thickness require minimum critical masses ranging from one to thirty kilograms. The critical particle densities of fissionable nuclei correspond to molecular densities of gases at less than atmospheric pressure.

Of the possible gaseous fuels, enriched uranium hexafluoride is the most obvious first choice. It sublimes at 54.6°C at atmospheric pressure and remains completely in the gaseous state at temperatures below 2000°K [7]. Three gaseous-core experimental facilities have been constructed to date. A 1.5 kW(t) U²³⁵F₆ fueled research reactor was constructed in the USSR in the late 1950s [8]. Two gaseous-core research reactors have been constructed in the United States. The most recent was a spherical gas-core critical experiment [9,10] performed in early 1970 at the National Reactor Testing Station.

The gaseous-core reactor concept has found little terrestrial application to date. A recent patent disclosure [11] proposes a UF₆ fueled reciprocating
Figure 2. Pathway for Actinide Waste Transmutation in the Gaseous-Core Reactor.
engine. This pulsed nuclear reactor concept is analogous to the internal combustion engine; only the working fluid is a UF$_6$-He mixture and the spark plug is an artificial neutron source. A detailed analysis [12] of the concept concludes that efficiencies greater than 40 percent are attainable. An 8400 MW(e) MHD power plant design has been proposed [13] which takes advantage of the high temperatures generated in the fissioning uranium plasma. This analysis concluded that large commercial power plants using a gaseous-core nuclear heat source coupled to the MHD generator could have thermal efficiencies in excess of 70 percent. Another design study by Gritton and Pinkel [14] indicates that attractive power levels in reactors of practical size can be obtained with gas pressures and wall temperatures within the potential capability of known structural materials. For a spherical gas core reactor with a radius of 152.4 cm to generated about 4000 MW(t) would require a uranium partial pressure of about 11 atmospheres. Practical heat removal considerations with water and lithium as coolants limit power output to 200 to 1500 MW(t), respectively, for a static fuel configuration.

The Dual Purpose Gaseous-Core Power-Production, Waste Transmutation Facility

The design of such a dual purpose facility would ideally combine both the economic production of electric power with attractive rates of transmutation for radioactive waste disposal operations. This means high efficiencies and high neutron fluxes are necessary.

Figure 3 shows a schematic cross section of the proposed gaseous-core reactor. The fuel is UF$_6$ enriched to 6 percent U$^{235}$. The interior core region is four meters in diameter surrounded by a reflector-moderator of D$_2$O with a thickness of 500 cm. The D$_2$O is encased in an outer reflector of 500 cm
Figure 3. Gaseous-Core Power-Burner Reactor.
of reactor grade graphite. The additional outer graphite reflector is provided to return as many of the neutrons to the core as possible.

Figure 4 shows a possible flow diagram for the UF₆ gaseous-core reactor. Fuel flows through the core, and is heated internally by fission energy. The gaseous UF₆ leaves the reactor at 1800°F and 800 psi and enters the regenerator where it gives it some of its sensible heat to an intermediate helium coolant loop. The UF₆ then goes through the primary heat exchanger giving up the remainder of its sensible heat and its latent heat to the helium working fluid. From there the UF₆ liquid at 600°F enters a sump tank. Here all the volatile fission products are removed. These include Kr, Xe, fluoride compounds of iodine, and several others. Nonvolatile fission products could be removed by an on-line reprocessing stream utilizing the fluoride volatility process [15]. From the sump tank the liquid UF₆ is pumped through an evaporator at 800°F and returned to the core.

Electric power could be produced if the helium working fluid was expanded in a turbine and recirculated back to the heat exchanger. Assuming the waste heat from the UF₆-He cycle was rejected to the surroundings at 100°F (43°C) and ideal Carnot efficiency of 82.4% is obtained. This simple calculation neglects all thermodynamic irreversibilities and heat losses. The point is, however, that this high efficiency is the result of the high temperatures produced in the gaseous-core nuclear reactor. If 75% of the ideal Carnot efficiency could be achieved in practice (typical in power plant design), a system efficiency of about 65% would result. Furthermore, if 15% of the reactor power is used to circulate the gaseous fuel and working fuel this leaves an overall plant efficiency of 50%. A power plant with this efficiency would reduce the amount of heat wasted to the environment by about a factor of two over present-day power plants.
Figure 4. Schematic Diagram of Gaseous-Core Circulating Fuel System
The two meter radius core reflected by 0.5 meters of D_2O requires a minimum of 14 kg of U^{235} to maintain criticality in the cold, clean condition. Due to the buildup of fission product poisons at hot, operating conditions, additional fuel must be added to compensate for this negative reactivity effect. Thus, a mass of 140 kg of U^{235}F_6 (in 6% enriched UF_6) is maintained in the core at any particular instant in time. Control of excess reactivity is by means of adjustable control rods located in the graphite reflector and by the addition of burnable poisons in the D_2O. Fine reactivity control would ideally be through the use of these burnable poisons; specifically radioactive waste products such as I^{129}, continually dissolved and burned in the D_2O. The control rods would then only be used for startup and shutdown control. Further reactivity control would be asserted through the addition of burnable radioactive wastes in the 16 target ports. These ports, located a distance of 12 cm into the D_2O reflector-moderator, would be used to load and transmute actinide wastes which are not suitable for dissolution in the moderator. The large negative temperature coefficient associated with the gaseous fuel density, and the Doppler coefficient of the 6 percent enriched fuel should make the reactor both very easy to control and inherently safe. Emergency shutdown control is provided by injection of high-pressure, gaseous B^{10}F_3 (thermal neutron absorption cross section, 3813 barns) directly into the core.

Let us now consider the waste transmutation aspects of the proposed facility. Mention has been made of loading radioactive waste iodine-129 in the D_2O reflector-moderator and actinide wastes in the target ports of the reactor. Waste I^{129} could be loaded continuously at the same rate at which it burned up and removed from the reactor. Actinide transmutation would involve batch loading and subsequent recycle after an appropriate burnout period.
The question remains of how much radioactive waste can be loaded into the reactor. Here one would like to load the largest amount possible without poisoning the reactor out of operation. Radioactive iodine-129 added to the D₂O will primarily affect the thermal utilization factor, \( f \). Using the known fuel composition and reactor configuration, values of \( f \) were computed for the feasible concentrations of \( ^{129}I \) added to the D₂O. A maximum concentration of 2600 ppm of \( ^{129}I \), or about 425 kg, can be added to the D₂O before the reactor becomes subcritical.

The insertion of actinides into the core of the reactor will have two complicating effects; the addition of fuel (and hence neutrons) in the form of fissile Am²⁴², Cm²⁴³, Cm²⁴⁵, etc., and the addition of neutron-absorbing fission products during the course of neutron exposure and actinide burnup. Claiborne [4] has concluded that the effect of recycling 99.5% of the waste actinides back into light water reactor fuels is minimal. A maximum average reactivity decrease of about 0.8 percent is attained in about five cycles through the reactor. This reactivity decrease can be counteracted by only about a 2% increase in the fissile material, which would amount to increasing the fuel enrichment about 0.1%.

Many of the actinides have substantial thermal fission cross sections, shown in Table 1 and hence, can achieve criticality if assembled in sufficient amounts. The only significant transplutonium actinide nuclides for which criticality in a thermal system could occur are Cm²⁴⁴ that contains fissile Cm²⁴⁵ and Cf²⁵² that contains sufficient target precursor, Cf²⁵¹. Because of the very low potential critical mass of Cm²⁴⁵ (942 g) and Cf²⁵¹ (940 g) the possibility of thermal criticality must be anticipated and included in the design. The published data of Clark [18] indicates that concentrations of 15 g/liter for Cm²⁴⁵ and 6 g/liter for Cf²⁵¹ may achieve criticality in a homogeneous mixture.
Advantages and Disadvantages of the Gaseous Core Power Reactor Concept

Compared with conventional solid-core nuclear reactors the gaseous core power reactor would have several advantages. These advantages and disadvantages derive mainly from the fact that the fuel is in the gaseous state. A basically simpler internal reactor core without the need for fuel pins, cladding and spacers is possible. Elimination of fuel element fabrication costs, UO2 to UF6 conversion charges would result in considerable savings in the monetary and energy cost of power production. Continuous replenishing of fissile fuel and continuous removal of fission product poisons would give rise to the possibility of continuous operation with increased fuel and neutron economy. Due to its inherent low inventory of U238 and its high neutron flux, less actinide waste is generated by the UF6 gas core reactor. Operating nuclear characteristics of the gas-core reactor were simulated with the ORIGEN computer code [19]. Figure 5 shows the buildup of fission product and actinide waste in the UF6 reactor and in a typical LWR of equal power output as a function of time. After three years of continuous operation the UF6 reactor contains about one order of magnitude less of long-lived actinide waste than the LWR. This behavior is due to the steady-state nature of refueling operations, as opposed to the "batch" fuel loading technique of present day LWRs. In addition, with continuous fuel (U215) addition and removal of xenon and other volatile fission product poisons, fuel burnups of 250,000 to 300,000 MWD/MTU were obtained in the computer results. The practical disadvantages of the UF6 gas-core reactor are primarily due to the inevitable dissociation of the fuel and its associated fluoride compounds. Specifically, they are: fuel dissociation, corrosion-limited lifetime of core and primary system components, and accountability of heavy-element by-product fluorides.
Figure 5. Comparison of Radioactive Waste Production of 3425 Mw(t) Fission Power Reactors.
In the circulating fuel system produced at high power operation. Here there is considerable experience in handling liquid fluoride salts from the Molten Salt Reactor (MSR) program. Also experience in handling UF₆ at the gaseous diffusion plants should prove extremely useful.

RESULTS AND CONCLUSIONS OF THE TRANSMUTATION STUDIES

Transmutation Rates and Reactions

The UF₆ gaseous-core reactor was chosen for this study as a neutron source because of its obvious potential for producing high neutron fluxes and generating useful power simultaneously. A reference reactor design was produced which could satisfy both of these requirements. The reactor power specified was 3425 MW(t) which resulted in an average thermal neutron flux across the core of 6.44 x 10¹⁴ n/cm²·sec. The ORIGEN computer code [19] was then used to study the transmutation rates of several waste isotopes present in the discharged fuel from reactors of the current LWR-type.

The waste isotopes chosen for the study were: I²⁹, Am²⁴¹, Am²⁴²m, Am²⁴³, Cm²⁴³, Cm²⁴⁴, Cm²⁴⁵, and Cm²⁴⁶. The amounts of the initial waste loading were determined from the composition of 20 discharged LWR fuel loads stored for a decay interval of ten years. The composition of the Am and Cm waste loading is the same as that given in Table 2. The quantities of I²⁹, Am and Cm used in the study are approximately equivalent to the waste product in 60 reactor years of operation.

In calculating rates of transmutation it is necessary to characterize the neutron energy spectrum of the reactor and the cross section dependence on spectrum for the particular target material. Thus the reaction rate depends not only on the magnitude of the neutron flux but also the shape of the neutron energy spectrum. The contribution of resonance energy neutrons to the reaction
rate is expressed by the ratio of resonance-to-thermal neutron fluxes, $\phi_{\text{res}}/\phi_{\text{th}}$.

The rates of waste transmutation were found to strongly depend on the value of $\phi_{\text{res}}/\phi_{\text{th}}$. The transmutation rate curves for $^{129}\text{I}$ are shown in Figure 6 for three values of $\phi_{\text{res}}/\phi_{\text{th}}$ in the range of interest. In this calculation the thermal neutron flux was $6.44 \times 10^{14} \text{ n/cm}^2\text{sec}$ and the initial loading is 400 kg of $^{129}\text{I}$ (the waste $^{129}\text{I}$ accumulated from 60 reactor-years of LWR operation). The effective neutron capture cross sections of $^{129}\text{I}$ for the values of $\phi_{\text{res}}/\phi_{\text{th}} = 0.058, 0.29$ and 0.58 were calculated to be 20.6, 32.3 and 46.9 barns, respectively.

For the specified neutron flux of $6.44 \times 10^{14} \text{ n/cm}^2\text{sec}$, transmutation rates of 68 kG/yr, and 77.6 kG/yr were calculated using the values of $\phi_{\text{res}}/\phi_{\text{th}} = 0.058, 0.29$, and 0.58, respectively.

The calculations for actinide transmutation were performed in a similar manner. The calculated transmutation rate curves of americium wastes for $\phi_{\text{res}}/\phi_{\text{th}} = 0.058, 0.29,$ and 0.58 are shown in Figures 7, 9, and 11, respectively. The same curves for curium wastes are shown in Figures 8, 10, and 12 respectively. It should be remembered in examining these curves that the Am and Cm wastes were irradiated at the same time in the neutron flux. Furthermore, these curves are the total waste composition of the initial waste loading plus the actinide by-product waste produced by fuel burnup in the UF$_6$ gaseous core reactor. It was felt that this would more realistically take into account the waste produced in affecting the transmutation process itself.

The curves presented in Figures 7 through 12 can be better understood with the aid of the cross section data presented in Table 3. The capture-to-fission ratio, $\alpha = \sigma_{\gamma}/\sigma_f$, is important in actinide transmutation. The relative probability that a compound nucleus decays by fission is $1/(1 + \alpha)$, and the relative probability that it decays by emission of capture $\gamma$-rays is $\alpha/(1 + \alpha)$. 
\( \phi_{th} = 6.44 \times 10^{14} \text{ n/cm}^2 \text{-sec} \)

Initial Loadings:
\( ^{129}I \) - 400 kg

Figure 6. \(^{129}I\) waste transmutation versus irradiation time for \( \phi_{res}/\phi_{th} = 0.058, 0.29, 0.58 \).
Figure 7. Americium waste transmutation versus irradiation time for $\phi_{res}/\phi_{th} = 0.058$. 

$\phi_{th} = 6.44 \times 10^{14} \text{n/cm}^2\cdot\text{sec}$

Initial loadings:
- Am$^{241}$ - 83.34 kg
- Am$^{242m}$ - 1.58 kg
- Am$^{243}$ - 172.54 kg
Figure 8: Curium waste transmutation versus irradiation time for \( \phi_{/\text{th}} = 0.058 \).

kg of waste remaining as original nuclide

\[
\phi = 6.44 \times 10^{14} \text{n/cm}^2\text{-sec}
\]

Initial loadings:
- \( \text{Cm}^{243} \): 0.111 kg
- \( \text{Cm}^{244} \): 0.361 kg
- \( \text{Cm}^{245} \): 3.69 kg
- \( \text{Cm}^{246} \): 0.435 kg
$\phi_{th} = 6.44 \times 10^{14} \text{ n/cm}^2\text{-sec}$

Initial Loadings:
- Am$^{241}$ - 81.34 kg
- Am$^{242m}$ - 1.58 kg
- Am$^{243}$ - 172.54 kg

Figure 9. Americium waste transmutation versus irradiation time for $\phi_{res}/\phi_{th} = 0.29$. 
Figure 10. Curium waste transmutation versus irradiation time for $\phi_{res}/\phi_{th} = 0.29$. 

$\phi_{th} = 6.44 \times 10^{14} \text{n/cm}^2 \cdot \text{sec}$

Initial Loadings:
- Cm$^{243}$ - 0.117 kg
- Cm$^{244}$ - 38.14 kg
- Cm$^{245}$ - 3.69 kg
- Cm$^{246}$ - 0.435 kg
Figure 11. Americium waste transmutation versus irradiation time for $\phi_{res}/\phi_{th} = 0.58$. 

$\phi_{th} = 6.44 \times 10^{14} \text{n/cm}^2\text{-sec}$

Initial Loadings:
- $^{241}\text{Am}$ - 83.34 kg
- $^{242m}\text{Am}$ - 1.58 kg
- $^{243}\text{Am}$ - 172.54 kg
Figure 12. Curium waste transmutation versus irradiation time for $\phi_{\text{res}}/\phi_{\text{th}} = 0.58$. 

Initial Loadings:
- $\text{Cm}^{243}$ - 0.117 kg
- $\text{Cm}^{244}$ - 38.14 kg
- $\text{Cm}^{245}$ - 3.69 kg
- $\text{Cm}^{246}$ - 0.435 kg 

$\phi_{\text{th}} = 6.44 \times 10^{14}$ n/cm$^2$-sec
<table>
<thead>
<tr>
<th>Nuclide</th>
<th>RES = 0.058</th>
<th>RES = 0.20</th>
<th>RES = 0.58</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\sigma_{\gamma}$</td>
<td>$\sigma_f$</td>
<td>$\alpha$</td>
</tr>
<tr>
<td>Am$^{241}$</td>
<td>707</td>
<td>4.17</td>
<td>169.5</td>
</tr>
<tr>
<td>Am$^{242m}$</td>
<td>1260</td>
<td>3780</td>
<td>0.33</td>
</tr>
<tr>
<td>Am$^{243}$</td>
<td>153</td>
<td>0.37</td>
<td>413.5</td>
</tr>
<tr>
<td>Cm$^{243}$</td>
<td>155</td>
<td>485</td>
<td>0.32</td>
</tr>
<tr>
<td>Cm$^{244}$</td>
<td>44</td>
<td>1.48</td>
<td>29.7</td>
</tr>
<tr>
<td>Cm$^{245}$</td>
<td>223</td>
<td>1150</td>
<td>0.19</td>
</tr>
<tr>
<td>Cm$^{246}$</td>
<td>7.81</td>
<td>0</td>
<td>--</td>
</tr>
</tbody>
</table>

TABLE 3. EFFECTIVE $(n,\gamma)$ AND $(n,f)$ CROSS SECTIONS (in barns) FOR Am AND Cm WASTE ISOTOPES FOR THREE VALUES OF RES = 0.058, 0.29, AND 0.58
Thus a high value of $\alpha$ means a low probability of fission following neutron absorption, and conversely, a low value of $\alpha$ means a high probability of fission.

From the data in Table 4 the major reaction paths can be identified. The major reactions for the americium isotopes are

$$\begin{align*}
&\text{Am}^{241}(n,\gamma) \rightarrow \text{Am}^{242}(n,f) \\
&\text{Am}^{242}(n,\gamma) \rightarrow \text{Am}^{243}
\end{align*}$$

and

$$\text{Am}^{243}(n,\gamma) \rightarrow \text{Am}^{244}(n,f).$$

The saturation behavior of $\text{Am}^{243}$ is due to by-product $\text{Am}^{243}$ produced by successive $(n,\gamma)$ reactions in the $\text{U}^{238}$ present in the fuel of the reactor. The major reaction paths at the curium nuclides is not so readily identifiable. It can be said, however, that successive $(n,\gamma)$ reactions on the curium isotopes are responsible for the production of significant amounts of nuclides with mass number greater than 244. The production curves for significant transmutation by-products with $A \geq 247$ is shown $\phi_{\text{res}}/\phi_{\text{th}} = 0.29$ and 0.58 in Figures 13 and 14, respectively. The by-product production curve for $\phi_{\text{res}}/\phi_{\text{th}} = 0.058$ has been omitted because the quantities of these isotopes produced in this case is at least two orders of magnitude lower than in the others.

The Effect of Transmutation on the Hazard Potential of Radioactive Wastes

The relative inhalation hazard (RIIH) and the relative ingestion hazard (RIGH), are quantitative measures of the potential danger which could result from release of radioactive wastes into the environment. The success of a particular transmutation scheme can be judged in terms of the overall reduction in the values of the RIGH and RIIH following transmutation.
Figure 13. Transmutation by-product production versus irradiation time for $\phi_{\text{res}}/\phi_{\text{th}} = 0.58$.

$\phi_{\text{th}} = 6.44 \times 10^{14}$ n/cm$^2$-sec
Figure 14. Transmutation by-product production versus irradiation time for $\phi_{res}/\phi_{th} = 0.29$. 

$\phi_{th} = 6.44 \times 10^{14}$ n/cm$^2$ -sec
The effect of transmutation on the nuclide RH, and RHG of $^{129}$I shown in Figure 15. The first section of the abscissa of this figure represents the reduction in the hazard measure due to the five year irradiation at $\Phi_{th} = 6.44 \times 10^{14}$ n/cm$^2$sec for the three values of $\Phi_{res}/\Phi_{th}$. No significant reduction in the hazard measure occurs after irradiation due to the 15.9 x $10^6$ year half-life of $^{129}$I. The top curve in this figure shows the hazard measure of $^{129}$I with no transmutation after fuel reprocessing. In the case of $^{129}$I the curves shown represent the total hazard measure since no by-products with significant radioactivity are made from the initial loading of $^{129}$I.

Figures 16 and 17 show the effect of transmutation on the total actinide RH, and RHG, respectively. The first section of the abscissa of these figures represents the reduction in the total actinide hazard measure resulting from transmutation of the quantities of americium and curium waste described in Figures 7 through 12. The three lower curves in Figures 16 and 17 are the hazard measure of the transmuted actinides (including by-products) plus the hazard measure of the burned nuclear fuel with 99.5 percent of the U and Pu removed at the end of the five year irradiation. The upper curve in both figures represents the hazard measure of all actinides (with 99.5 percent of the U and Pu removed) generated in 60 reactor-years of LWR operation. The differences in the shape of these curves represent the varying isotopic compositions of the irradiated and unirradiated wastes.

**Economic Considerations for Transmutation of Radioactive Wastes**

The cost of neutron-induced transmutation of radioactive waste is primarily the cost of producing neutrons. The major factors which govern the cost of neutrons are: reactor fuel and associated costs; plant depreciation including interest charges; and salaries, taxes, maintenance and general
Figure 15. Iodine-129 Hazard Reduction Following Transmutation.
Figure 16. Total Actinide RIHH Reduction Following Transmutation of Am and Cm Wastes.
Figure 17. Total Actinide RIGH Reduction Following Transmutation of Am and Cm Wastes.
operating costs. The cost of transmutation would also be a function of plant output because the sum of the plant costs listed above would remain fairly constant over a wide range of output, with fuel costs being the major variant. Plant output is determined by: reactor power, operating efficiency, and neutron utilization.

For large-scale transmutation of radioactive wastes in the 1980s and beyond, the price of target material will be determined primarily by costs associated with chemical separation from power reactor fuel residues. Estimates of unit recovery costs extrapolated to the mid-seventies for actinide targets and fission products are listed below [20]:

**Comparison of Chemical Recovery Costs**

<table>
<thead>
<tr>
<th>Actinide Targets</th>
<th>Fission Targets</th>
</tr>
</thead>
<tbody>
<tr>
<td>Am$^{241,243}$ $\sim$ $31/\text{g}$</td>
<td>Cs$^{137}$ $\sim$ $10/\text{g}$</td>
</tr>
<tr>
<td>Cm$^{242,244}$ $\sim$ $81/\text{g}$</td>
<td>Sr$^{90}$ $\sim$ $16/\text{g}$</td>
</tr>
<tr>
<td>Sr$^{129}$ $\sim$ $10/\text{g}$</td>
<td></td>
</tr>
</tbody>
</table>

As available quantities increase in the late seventies, and if a need for recovery of these materials is established, unit recovery costs could be reduced further.

Aside from developmental costs, several economic factors are in favor of the UF$_6$ gaseous-core reactor for use as a dual-purpose power reactor-transmutation facility. Fabrication of power reactor fuel elements accounts for about 40 percent of the total fuel cost [21] and chemical processing for about another 15 percent. Elimination of fuel fabrication costs and reduction in chemical processing costs could lower fuel costs by 45 to 50 percent of the UF$_6$ reactor. Increased operating efficiency resulting from continuous operation and increased neutron utilization resulting from continuous removal of
Fission product poisons could result in lower neutron costs than other reactor concepts. If salable power is generated in the reactor the assigned neutron cost could be lower still.

**Conclusions and Recommendations**

This study has attempted to ascertain the feasibility of nuclear transmutation in a UF₆ gaseous-core reactor. Some specific conclusions were obtained in the limited time available. Transmutation of ¹²⁹ by a five-year exposure to a thermal neutron flux of 6.44 x 10¹⁴ n/cm²/sec results in nearly order-of-magnitude reductions in the waste inventory of this nuclide. This reduction in inventory results in an order of magnitude decrease in the hazard potential of fission product waste ¹²⁹. A five-year transmutation of americium and curium wastes produces order-of-magnitude decreases in the overall hazard potential of actinide wastes generated in 60 reactor-years of LWR operation. The actinide results are in approximate agreement with those obtained by Claiborne [8]. It was found in this study that increased values of the resonance-to-thermal flux ratio, \( \phi_{res}/\phi_{th} \), resulted in increased rates of transmutation and increased reductions in hazard potential for both ¹²⁹ and the actinides. A rough breakdown of transmutation costs seems to indicate that the UF₆ reactor could be competitive with other transmutation reactor concepts.

Any study which attempts to determine the feasibility of a concept generally raises more questions than it answers. Several questions were raised during the course of this study.

Would recycling previously transmuted wastes accomplish further decreases in the hazard measure of actinide wastes?

What is the effect of varying the irradiation time on the inventory or hazard potential of actinide wastes, i.e., would three years irradiation accomplish the same effect as five years irradiation?
The technique of induced transmutation requires further study. Other long-lived isotopes should be examined for feasibility for the transmutation process. One such isotope which may prove sensible is $2.13 \times 10^5$ year Te$^{99}$. It is present in significant quantities in fission product wastes and has a 22 barn thermal neutron capture cross section and a 92 barn resonance integral. Further investigation into the process of neutron-induced transmutation may lead to realization of a practical method for disposal of long-lived radioactive wastes.
LIST OF REFERENCES


NUCLEAR FUEL CYCLE

WASTE MANAGEMENT AND THE EFFECT ON THE FUEL CYCLE

Sponsored by Nuclear Fuel Cycle Division

1. Storage Fee Analysis for Commercial High-Level Wastes, B. B. Field (ARCO), invited

During 1972 a study was completed for the AEC to develop and test a formula for determining storage fees to be charged commercial nuclear fuel processors for storing their high-level wastes in government repositories. The storage fee formula is incorporated in full-cost recovery to the government for the operating and capital costs directly related to its proposed waste storage operations. It is assumed that standardized canisters of wastes generated through the year 2000 will be placed into a Retrievable Burial Storage Facility for some interim period, possibly 100 yr, and then transferred to a more permanent repository. It is assumed that a one-time storage fee is charged at the time the canister is received for storage.

It was found that the fee must contain two components: (a) a component that covers the canister's share of constructing and operating the RBSF to transport where it is filled to expected capacity, and (b) a component creating an "endowment" fund whose earnings cover: (i) the costs of the 100-yr surveillance at the RBSF, (ii) the transition costs of constructing the more permanent repository for those canisters of transferring canisters to that repository, and of decommissioning the RBSF, and (iii) the perpetual surveillance costs at the new repository. It was found that the most important determinant of the storage fee is the cost of the interim facility. The next most important is the spread between cost escalation rate and endowment fund earnings (interest) rate. A simplified version of the formula is used to demonstrate these findings.


Public opinion insists on alternate options, other than the ones presently accepted as technically feasible, for disposal of radioactive waste, particularly for those isotopes with half-lives of several thousands of years. One of the recently proposed options in waste disposal by induced nuclear transmutation. Since many long-lived waste isotopes have appreciable neutron capture and/or fission cross sections, they can be made to decay into other faster decaying or stable isotopes by neutron irradiation. The key requirement for successful transmutation is the availability of a high neutron flux. Erotic reactors developed for space propulsion can fulfill this requirement. Of these, UF gaseous-core reactors are closest to technical feasibility.

This summary presents the results of a study in which the application of a UF gaseous-core reactor for radio-active waste disposal by nuclear transmutation was considered.

Several critical gaseous UF reactors have been built to date and an advanced prototype is presently under construction. Operating temperatures as high as 1800 K and neutron fluxes of $10^{16}$ n/cm² sec are considered feasible. Due to its high operating temperature, the gaseous-core reactor would be an effective power generating system, while at the same time radioactive wastes could be transmuted in the large irradiation volume available in the core of the UF reactor. Several options are available for loading waste into the reactor depending on the chemical form and composition of the waste. The results reported in this paper were obtained by modifying the ORIGEN code in a way that it could be applied to the UF gaseous-core reactor. Continuous fuel addition and removal of volatile fission products was simulated in these calculations.

Due to the inherent low fuel mass and high neutron flux, less total radioactive waste is generated by the UF, gaseous-core reactor. Figure 1 shows the buildup of fission product and actinide waste in the UF reactor and in a typical LWR of equal power output as a function of time. After three years of continuous operation, the UF reactor contains about one order of magnitude less of long-lived actinide waste than the LWR.

Figure 2 shows the case where the UF reactor has been initially loaded with several long-lived radioactive wastes (obtained from other reactors) and operated continuously for several years at a neutron flux of $6.44 \times 10^{16}$ n/cm² sec [3425 MWth], 50-atm fuel gas pressure, $6 \times 10^{-4}$ U fuel. In all cases the content of waste has been decreased by more than an order of magnitude in three years.

Thus, the UF gaseous-core reactor is attractive from the viewpoint that it has a significantly lower ultimate inventory than present-day light-water reactors. In addition, the disposal of radioactive wastes by induced transmutation in the UF, gaseous-core reactor can be a viable alternative to the problem of waste disposal.

Fig. 1. Comparison of radioactive waste production of 3425-mW(th) fission power reactors.

Fig. 2. Waste burnout vs irradiation time at $\phi = 6.44 \times 10^{20} \text{ n} / (\text{cm}^2 \text{ sec})$ in UF$_6$ gaseous-core power reactor.

3. Selection of Significant Elements and Radionuclides for Waste Management Assessment, S. E. Logan, Glenn A. Whan (U of NM)

Assessment of high-level radioactive waste management concept involves detailed examination of release and environmental transport mechanisms for radionuclides in a waste deposit. Calculations by the ORIGEN Code indicate approximately 260 radionuclides are present at the time of separation in fuel reprocessed at 160 days. A screening method is proposed to determine the most significant elements and nuclides present in each time interval, including buildup of actinide daughters, during long-term storage.

Several indexes for evaluating the relative hazard of radionuclides have been proposed, including the dilution volume hazard measure used extensively at the Oak Ridge National Laboratory. The release, in units of $\text{mCi/g}$, is the ratio of activity for each nuclide in a given quantity of waste to the corresponding ICRP value for air or water, for inhalation and ingestion hazard, respectively. A screening technique using this index is proposed as follows:

1. At each time in the ORIGEN output tables of ingestion hazard, select the three fission product elements and the three heavy metal elements comprising the largest hazard.

2. At each time, select all isotopes that collectively comprise over 99% of the hazard for each of the three elements from each category, which are significant at the time.

Selections made at each time assure representation for radiation hazard over the whole time range from short term to long term. Elements have significance as the first selection step because concentrations in waste options for varying these concentrations in response to