DISPOSAL OF HIGH-LEVEL NUCLEAR WASTE

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More than a half century after the beginning of the Nuclear Age, there is no satisfactory answer to the serious dilemma of how to dispose of the large quantities of radioactive wastes created by military and civilian uses of nuclear energy. This paper examines technological options for waste disposal, and concludes by favoring Midtibarrier Monitored Retrievable Storage (MMRS) The authors point out, however, that this form of storage (it is not really disposal) will require "continuous monitoring... essentially forever. " Thus, the best of the options will require something akin to a "nuclear priesthood" to pass along their skills at monitoring these wastes for thousands of generations — a sobering thought.

Our century's indulgence in nuclear technology has created radioactive wastes that are a problem not only in the present but will affect thousands of generations in the future. The problems are so long-term that they are beyond our capacity to plan for adequately. At a minimum, we should cease — with all due speed — to generate more nuclear wastes.

The Nuclear Age Peace Foundation s directors issued a policy statement on nuclear power in May 1996 calling for "a world adequately supplied by renewable, environmentally benign energy sources, and the worldwide elimination of nuclear power." A copy of the full statement is available from the Foundation.

—David Krieger

Introduction

Disposal of highly radioactive nuclear waste is a critical problem for our time and will remain so well into the future. There are two main waste sources: Nuclear power reactors and bomb-related nuclear material from the production facilities and from the decommissioned U.S. and (former) U.S.S.R. nuclear weapons.

This paper deals with disposal of (a) reactor spent fuel rods and (b) waste sludge from the bomb-grade plutonium separation process. Disposal of bomb-grade plutonium from decommissioned weapons and from existing stockpiles present somewhat different problems which are not treated here.* Nuclear waste disposal poses a number of different yet interconnected problems, all of which must eventually be resolved in an integrated fashion: technical, economic, health-related, environmental, political. The present paper addresses primarily technical issues, and does not attempt an analysis of the overall problem.

Management of radioactive waste is a complex, multi-faceted procedure. Spent commercial fuel rods present the most demanding challenge of all waste problems because of the high level of radioactivity. The fuel rods, relatively harmless before entering the reactor, emerge having become dangerously radioactive. They require storage tor at least ten years under circulated water in a pool inside the reactor containment structure.

By statute, the government, through the Department of Energy's Office of Civilian Radioactive Waste Management, has promised to provide disposal capacity for the waste generated by the nation's nuclear power plants. Some of the waste which has accumulated over 45 years of Cold War nuclear bomb production also falls into the high-level category.

The term "high-level" nuclear waste has had its meaning changed in the U.S. over the years. At the present time the Nuclear Regulatory Commission (NRC) has defined "high-level" very narrowly as mostly, but not entirely, spent fuel elements and reprocessed military wastes, such as sludges. They further define "spent fuel," concentrates of strontium-90 and cesium-137, and transuranics as something not necessarily included in their definition of "high-level" waste.

Because this NRC definition is contrary (if not actually contradictory) to standards of the rest of the world and makes no sense to the authors, "high-level" nuclear waste is defined here as all radioactive waste material coming from nuclear reactor fuel rods whether confined or not:

a) Spent nuclear fuel rods, clad or declad, from commercial electricity generating reactors; average radioactivity being more than 2.5 million curies per cubic meter.

b) Semi-liquid sludge from nuclear bomb fabrication waste processing residue — average radioactivity being about 3500 curies per cubic meter.

All this waste contains five shorter lived and longer lived radionuclides of main concern. The shorter lived are strontium-90 whose half life, $t_{1/2}$ is 28.5 years, and cesium-137 whose half life, $t_{1/2}$ is 30 years. See Ref. 1 for the half-life values used in this study. The radioactivity of these shorter lived nuclides is approximately 95% of the total radioactivity of the nuclides of concern. Total hazardous life for these shorter lived nuclides is considered to be between 600 years and 1000 years depending upon one's point of view.

The longer lived isotopes are plutonium-239 whose $t_{1/2}$ is 24,110 years, plutonium-240 whose $t_{1/2}$ is 6,540 years, and curium-245 whose $t_{1/2}$ is 8,500 years. Plutonium-238 whose $t_{1/2}$ is 88 years will have essentially disappeared after several thousand years, so in storage terms of the longer lived elements this isotope is not of concern as long as it will have been successfully contained for the next several thousand years. As for the life of these longer lived materials, the NRC considers 10,000 years as the storage time required; however, some people consider a lifetime as long as 100,000 years to 500,000 years as more appropriate.

A recent analysis, **Management and Disposition of Excess Weapons Plutonium, was published by the National Academy Press, 1994*

Table I
Radioactivity for 100 Tons of Spent Fuel *
Curies Remaining

Isotope	$t_{1/2}$ yrs	10 yrs	500 yrs	1000 yrs	10,000 yrs	100,000 yrs	200,000 yrs
Sr-90	28	2,000,000	15	trace			
Cs-137	30	3,000,000	40	trace			
Pu-239	24,110	22,000	27,000	29,000	56,000	8,000	240
Pu-240	6,540	49,000	175,000	170,000	69,000	7	trace
Cm-245	8,500	56,000	52,500	52,000	25,000	0.5	trace

* A typical 1000 megawatt reactor contains about 100 tons of enriched uranium, one-third of which is renewed each year.

Table I (above) extracted from Ref. 2 should be helpful. It must be noted that as some radioactive isotopes disintegrate, they create other radioactive isotopes in the process. Thus Pu-239 and Pu-240 increase at first and do not begin decreasing until many years later.

Table I illustrates, as does Figure 1 (below), rather spectacularly the fallacy of the NRC rationale for a 10,000 year waste storage lifetime, when the radioactivity for the plutonium isotopes are greater after that long period than at the outset. However, it must be noted that this Pu-239 is relatively confined and in general will not be disturbed, so the basic health hazards from such radioactive materials as radon and radium from uranium ores appear to be far more serious.

The general nuclear waste disposal approach is that the repositories should not be more dangerous than natural ores of uranium and thorium. In fact, they might be much less hazardous; after all, the natural ores have no barriers such as containers, and radium is leached from many of the ores so that traces get into the food chain. Spent fuel rods have to be stored between 13,000 and 14,000 years before their level of radioactivity decreases to that of natural uranium ore.

One of the most serious engineering problems is that of allowing for release of the prodigious heat emanating from stored nuclear power waste. Most of the heat comes from the strontium-90 and cesium-137 at the start, but the longer-lived actinides produce more in later years. As noted in

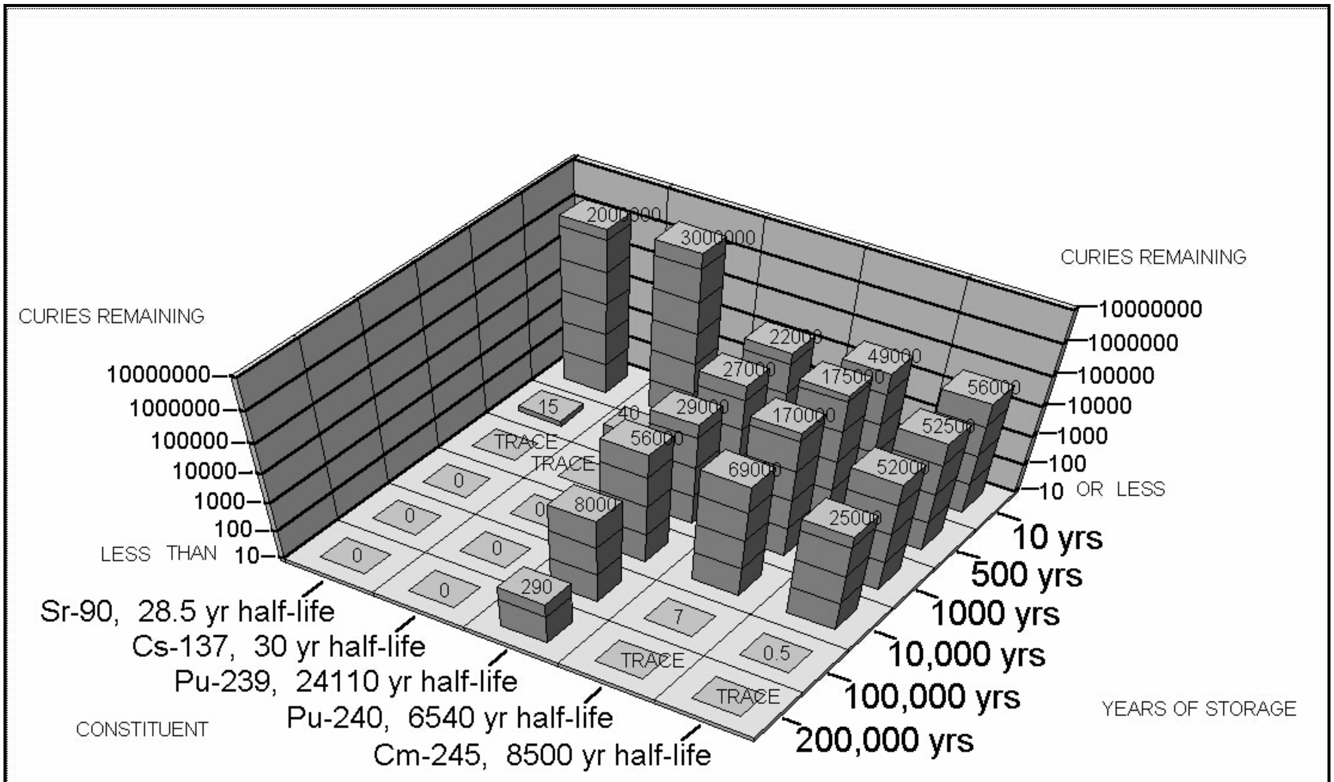

Figure 1. Radioactive Decay of 33 Tons of Spent Reactor Fuel. A typical (1000 MW) reactor contains about 100 tons of enriched uranium, about 33 tons of which becomes radioactive waste each year.

Table II (below), the heat liberated by spent nuclear reactor fuel decreases significantly as it ages.

From a practical engineering standpoint there is little difference between a 500 year lifetime and a 500,000 year lifetime. The 500 years is so long a time that no storage prototype system can ever be tested, thus the basic engineering considerations remain unchanged regardless of the waste lifetime. It is on this fact that any long-term storage conclusions are predicated. As is discussed below, any storage technique that utilizes permanent or nonretrievable ground burial is fundamentally a violation of basic engineering principles. This was pointed out to the nuclear industry over 25 years ago, but their response at that time was that they had "faith" that some satisfactory new technique would be developed, by the government of course and at taxpayers' expense, before it would be necessary to initiate long-term storage. Obviously, that has not happened and we are now faced with a nuclear waste disposal problem that has no fully satisfactory solution and probably never will have.

Multibarrier Monitored Retrievable Storage (MMRS)

This, unfortunately, is the final technique of choice for this particular waste disposal problem. It is unfortunate because there must be a continuous monitoring of the waste essentially forever. There are two fortunate aspects deserving mention: (1) the total volume of the waste involved is small by world standards, i.e., one football field for each type of waste each ten or twelve stories high, and (2) the number of people theoretically required to perform the monitoring task is also quite small, perhaps one hundred people or less worldwide. A ball park estimate of costs in present day dollars indicates that about $100 million is required over a 10,000 year time period for each 1000 megawatt nuclear power plant.

For the nuclear power plant waste, which consists of spent fuel rods, the most desirable inner barrier is the original cladding used for the nuclear fuel in the basic power plant configuration. This excellent cladding barrier is usually zirconium but sometimes stainless steel is used. The lifetime of this cladding has never been tested, so there is no telling exactly how long it can be depended upon. Safety engineering, however, dictates that because this barrier has already proved to be very reliable, it should be left in place and not removed. Further barriers have to be determined as a result of experimental development based upon both thermal characteristics and mechanical properties. Possibilities include glass, copper, ceramic, additional zirconium, stainless steel, nickel, or titanium. All this is for the power plant spent fuel rods only. Bomb waste having been processed requires another barrier or cladding before application of the "standard" multibarriers.

Because the bomb waste is initially in a semi-liquid sludge form, it has to be solidified at the outset. The quantities involved are approximately 105 million gallons for the U.S. as of 1994, so the total quantity worldwide would be about 200 million gallons. A ball park estimate of the solidified quantity results in roughly the same volume as the power plant waste with the identical radioactive nuclides. The major difference between this solidified nuclear bomb waste and the spent fuel rods will be that the former will probably be contained in vitrified or glassified cylinders as compared with the latter being in long slender cylindrical fuel rods with metallic cladding. Actually, if we fabricated the bomb waste's vitrified cylinders in long slender rods the same size as the spent fuel rods, the remainder of the waste disposal process could be identical for both waste components.

Of special note here is that the final configuration must be a solid container or cask whose outer surface is monitored. Engineering jargon usually refers to this approach as placing the canister in a "bath tub." Sensitive radioactive sensors in the "bath tub" must monitor this outer container surface continuously in an automated fashion. Such automation must incorporate Built-In-Self-Test, making use of many space exploration techniques.

While the waste canisters or containers are stored in shallow, underground but easily accessible facilities, all testing and monitoring should be performed by automated equipment. Such techniques preclude human errors caused by boredom, undetected equipment malfunctions, and misinterpretation of displayed information. Human intervention is necessary only for overall supervision and periodic testing of the automated equipment because of multiple error causation possibilities beyond the original design. We have to remember that there is nothing that is 100% safe; nuclear bombs for example only possessed six or seven safety

Table II
Thermal Power per Metric Tonne* of Spent Fuel

Age (years)	Rate of heat liberated (watts)	Percent of heat from strontium and cesium
1	12300	67
5	2260	69
10	1300	72
20	950	68
50	572	56
100	312	31
200	183	5

* 1 metric tonne = 1000 kilograms = 1 long ton = 2200 lbs

interlocks. Periodically, the nuclear waste monitoring equipment must be replaced and the waste canisters themselves will require retrieval and automatic repackaging every hundred years or more. It is noted that there are essentially two sets of automatic equipment, (1) the canister "bath tub" monitors and (2) the retrieval/repackaging mechanism. The latter might well be simply remote controlled equipment or a combination of semi-automatic components.

A summary of our viewpoint is that the best disposal method known to date consists of sealing the zirconium or stainless steel-clad spent fuel rods, without reprocessing, in copper or steel canisters and storing these in a geologic but easily accessible repository. This is the once-through fuel cycle. The spent fuel rods should be allowed to stand at least ten years under water so that most of the radioactive materials decay, and the rate of heat generation has fallen by about 86%. The repositories must have multiple barriers. The canisters must be arranged so that sufficient cooling air can circulate around them after disposal. The waste density must not exceed that required for adequate heat flow.

A major point to be made is that a very responsible and conscientious group of people is required to take care of our long-term nuclear garbage. This group must have substantial credentials for at least several centuries of resource concern and responsible treatment of their environment. Few groups in the world will qualify and it is worth considerable remuneration from the society at large to this select management group to perform the waste monitoring required. The compensation referred to, while quite large for the equipment and personnel involved in terms of the select group, will be minuscule compared with the monetary interest the U.S. presently pays on its debt or the amount societies throughout the world have been willing to spend on weapons of mass destruction.

Nonretrievable Geologic Storage

The major effort toward long-term high-level nuclear waste disposal has been in the area of depositing in the ground all the dangerous material in some sort of containers. This approach seeks to find a permanent disposal technique so the waste can be left for posterity without any possibility of future generations being at risk. While the motivation and results sought after are commendable, the reality of what is being attempted has not really been fully recognized.

Of prime importance here is the basic engineering principle alluded to above that any truly new system has to be tested for at least one life cycle in order for there to be reasonable confidence that there have been no design or fabrication errors. Given a new disposal system that has a life cycle of at least 300 years, the required engineering prototype test is nor possible. After twenty-five years, the faith of responsible nuclear power parties that government would figure out an acceptable solution eventually is as remote a possibility today as it was in the first place. Needless to say, that confidence in a permanent solution has now been thoroughly shaken, as basic engineering considerations dictated at the outset.

The geologic materials investigated throughout the world have included salt, granite, volcanic tuff, and basalt. Each particular site chosen, after much consideration of geologic and scientific aspects, has proven to have some flaw that makes such contemplated irretrievable burial unacceptable. In some instances fractures in the structure have occurred or been discovered whereby the nuclear waste could eventually get outside the confinement volume. Other problems include the buildup and then outflow of water. Earthquake susceptibility is always of concern and automatically precludes use of some sites.

In the end it does not look as though we can possibly have sufficient confidence in any one geologic site that would allow permanent disposal. One possibility, of course, is to treat the waste similarly to the way we instituted nuclear power in the first place, i.e., proceed with what seems satisfactory at the time and leave any serious long-term problems to be solved only after they have actually arisen. In other words, there is always the irresponsible option of letting our distant descendants be plagued with our 20th century errors.

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Burying of Casks Inside Underground Bomb Test Cavities

Given the already contaminated underground cavities made by bomb-testing in Nevada, a logical option would appear to be the use of these voids for permanent waste disposal. An important factor to be considered is the high level of radioactivity already present within those cavities. While leaks into the air occurred in some tests, in most cases all of the radioactivity from the explosions was confined. After all, this was the bomb-testing option of choice to prevent contamination of the atmosphere. A typical test was the Chesire experiment, conducted on February 14, 1976. It was a hydrogen bomb with a yield between 200 and 500 kilotons. It was detonated at a depth of 3830 feet, which was 1760 feet below the water table.

There is already considerable experience in drilling into bomb cavities. The purpose was to sample the radioactive materials for analysis, in order to estimate the yield and efficiency (which is the percentage of U-235 and/or Pu-239 which underwent fission). If the deeper cavities are chosen (to insure that they are well below the water table), it would be easiest to drill a shaft in the same place as the original one. By now, the fission products which are most dangerous, such as iodine-131, have all decayed. The only gaseous fission product left is krypton-85, with half-life 10.7 years. It is not nearly as dangerous as radon, and in any case only a small amount would diffuse out. Casks of waste would be lowered into the cavity using a cable suspended from a derrick, with the operator inside a shielded housing, if necessary. At the end, the cavity is filled with earth, and the shaft closed.

Although this burial technique looks promising and deserving of further study, it is by no means clear that this technique for disposing of hazardous waste is satisfactory. It could develop that creating new cavities for the express purpose of using them as repositories could become attractive. In that case, the site would be carefully chosen with the water table in mind, and the cavity blasted very deep. Hydrogen bombs might be best since most of the energy comes from deuterium fusion, thus minimizing the amount of radioactivity created.

So much for the positive aspects. Negative aspects include the idea that just because deep underground cavities are already contaminated with long lived radioactive nuclides from nuclear bomb explosions, we are not justified increasing the potential future health hazards by orders of magnitude. As with other geologic burials, there are possibilities of earthquakes, ground fractures, and unanticipated failures in the deep drilled shafts that would cause water leakage. However, of all the possible permanent disposal sites, these deep holes of hazardous remnants from past bomb development follies appear to be the most attractive, even though a time period of at least 10,000 years is too long to confidently conclude that there are no significant failure-modes.

Because permanent geologic disposal in nuclear bomb cavities violates fundamental engineering principles, it can be considered to be irresponsible for present generations to pursue that option. Perhaps considerations of our lack of knowledge today of what the worldwide land usage was many thousands of years ago will provide an understanding of our cautious conclusions here. We simply cannot be reasonably certain how the use of land throughout the world will evolve over the forthcoming thousands of years. Thus conscientious adherence to responsible behavior requires our not utilizing this bomb cavity technique at present. Further study might possibly result in something useful a hundred or more years hence.

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Burial Between Tectonic Plates

The interior of the Earth contains the elements potassium, uranium, and thorium, all slightly radioactive. This radioactive decay liberates heat, which keeps the Earth's core hot. The consequence of a hot, liquid core is movement of floating tectonic plates, and formation of mountain ranges and continents. Were this not the case, mountains and all land would erode down, and our planet would be covered with water. Without this radioactivity, we would not exist.

Geologists discovered many years ago that the continents are in constant motion relative to each other. Far below the ground tectonic plates are sliding very slowly over each other. The continents rest on these plates, so the oceans are changing size and shape while the surface continents are moving relative to one another. At the edge of a plate whose motion is toward the ocean, there will be a suhduction layer between that tectonic plate and the one below. Any material between the plates at that point will be pulled in between and remain there for at least several million years.

Concern over the years has been to consider just how one could perform the placement of high-level nuclear waste into a tectonic plate subduction layer. One major problem is digging down to that depth. But even more stringent than that is the problem of construction of shaft walls that will withstand the weight of all the earth above. The same problem is encountered when constructing a research module to descend to the ocean floor. While the ocean depth is a maximum of about 6 miles, the tectonic plate depth is as much as 50 miles. Finally, there are the construction strength problem differences between an enclosed submerged module in the ocean and the side wall problems in a shaft through which nuclear waste canisters are to be lowered.

There has not been, nor is there even a contemplated possibility of constructing a shaft that would be strong enough for this nuclear waste disposal option. Thus, another apparently attractive approach seems to be beyond our reach.

Transmutation

Soon after commercial generation of electricity via reactors started and their high-level waste began to accumulate, ways to simplify and manage the problem were sought. Among these was reprocessing to separate the waste into several fractions, and then, using neutrons, to transmute via fission the transuranium elements (neptunium, plutonium, americium, etc.) into nu-elides which have relatively short half-lives so that they lose their radioactive sting in a repository during an abbreviated storage time. The transuranium elements would require se-questering in a repository for many thousands of years.

If the nuclear waste is bombarded with neutrons, electrons, or other atomic particles so that it is changed from a long-lived to a short-lived radioactive material, the process has been termed "transmutation." About thirty years ago, people inquir-ing about the long-term nuclear waste disposal for commercial reactors were told that the military had the identical problem for its nuclear bomb waste. Because the military waste was already twenty years old, the word to one of the authors was that the military had not only decided that transmutation was the best solution to this problem but had already worked out all pertinent details. Many years and many nuclear reactors later, of course, we found out that the military had not developed any viable transmutation waste disposal system at all.

In fact, the basic problems with transmutation have been perennial. Each nuance has resulted in the same general result. Any process based on transmutation would require reprocess-ing to separate the waste into several fractions, and then, using neutrons, to transmute via fission the transuranium elements (neptunium, plutonium, americium, etc.) into nuclides which have relatively short half-lives. Considerable research has been carried out recently on these nuclear incineration techniques. Tests are being conducted at Hanford, Los Alamos, and Brookhaven National Laboratory on Long Island. Success of the proposed procedure depends on reprocessing spent fuel by either the PUREX process or a technique similar to the TRUMP-S process. The actinides would then be reintroduced into the reactor or bombarded with neutrons generated using an accelerator. Thus neutron sources might be either nuclear reactors, perhaps of the breeder type, or linear accelerators to produce high-energy protons, which collide with lead, bismuth, or tungsten targets. This produces abundant neutrons, which must be moderated using heavy water. The neutrons then cause fission of the actinides, and liberation of huge amounts of energy, as in a nuclear reactor.

Disposal of wastes by transmutation is intimately related to fast breeder reactors. While American reactors of this type were phased out by Congress in 1983, a new type, the Integral Fast Reactor, is now being studied. These breeder reactors use liquid sodium as coolant and have no moderator. They are being promoted as a way to cope with nuclear waste. The problem, of course, is that "we've heard that story before."

Even though the outlook for nuclear transmutation is most unpromising, a few details are perhaps in order. The accelerator procedure is highly unfavorable from the standpoint of energy consumption. The steel and other parts would be activated by neutrons, and become radioactive. It seems that about as much radioactive waste would be produced as is consumed, as stated above, if not more. Costs would be fantastic. The procedure could not easily be used with fission products. They absorb neutrons poorly; after all, they were in a neutron environment for years, and survived. Only two, iodine-129 and technetium-99, are easily transmuted to nonradioactive nuclides, and these are not particularly important. Technetium-99 (half-life nearly a quarter of a million years) is converted by neutrons into technetium-100 (half-life only 16 seconds) forming ruthenium. If this process is carried out while a stream of ozone is passed through the apparatus, volatile ruthenium tetroxide is con-stantly removed. Transmutation might be successful in this case, and perhaps that of iodine-129, but in general the tech-nique is not expected to be satisfactory.

In 1992 a group of nine qualified experts finished an exhaustive assessment of disposing of waste through transmuta-tion via fast breeder reactors, accelerators, and high tempera-ture electrolysis techniques (the Ramspott report, after the first author). These scientists are associated with the Lawrence Livermore National Laboratory, two universities, and a private firm. The study concluded that high-temperature electrolysis procedures for separating actinide metals in reprocessing high-level waste offers no economic incentives or safety advantages. Unfortunately, actinide separation and transmutation cannot be considered a satisfactory substitute for geological disposal.

Spacecraft Transport to the Sun

Of all the theoretically possible disposal techniques one can think of, this is one of the most preferable. Materials on the sun are already similar to our waste products, so our depositing high level nuclear materials on the sun would blend right in. Unfortunately, the numbers are such that we cannot do the job, either technologically or economically.

Given the liquid sludge nuclear bomb waste of about 10' gallons for the U.S. alone, the following ballpark numbers apply:

~0.1 = conversion factor for solidification.
~0.1 = conversion factor for gallons to cu ft.
~100 Ibs/cu ft density.
10,000 Ib effective spacecraft waste payload for an Apollo-type vehicle assuming the additional 7000 Ib payload will be required for containers and the retro-rockets.
10^8 x 0.1 x 0.1 x 100 x 10^{-4} = 10^4 spacecraft for only accumulated U.S. military waste.

Besides the fact that the U.S. does not have the economic resources to fund such a gigantic number of spacecraft, each vehicle would have to have perfect launch systems that would not blow up on the launch pad plus perfect guidance systems that would insure the vehicle not turning around back toward the Earth. Obviously, this is beyond any forseeable capability and must be abandoned as a possible option.

Conclusions

A major point emphasized in this study is that it is unethical to force a known potential environmental hazard on future generations when a reasonable alternative exists. This aspect was phrased above in engineering terms, i.e.

basic engineering principles; however, it could easily have been phrased in more socially oriented terms. This leads to the only responsible choice being the multibarrier monitored retrievable storage (MMRS) technique which will cost in present dollars between $100 million and $1 billion per 1000 megawatt power plant over a 10,000 to 100,000 year storage period.

It also needs to be pointed out that there are some important lessons to be learned from Mother Nature:

1) The natural nuclear reactors at Oklo in Gabon, West Africa, demonstrated that the plutonium and most metallic fission products did not leach out, even over thousands of centuries of leaching. Even the strontium-90 stayed in place until it decayed. The cesium-137 did migrate out, and the iodine fission products evaporated. Despite this favorable result, strictly speaking it applies to the particular geology of that area.

2) Another natural site teaches us more valuable lessons about the behavior of radioactive materials during long storage. There is a hill called Morro do Ferro in Brazil where there are 30,000 tons of thorium and 100,000 tons of rare earths. Much of the fission products are rare earths. Chemically, thorium resembles plutonium in some ways and the rare earths resemble curium and americium. Again, the evidence is that migration of the most dangerous materials from the surface over eons of weathering has been negligible.

3) Still another area whose study yields valuable lessons is the Koongarra ore body in Australia. This is a giant deposit of uranium ore in a common type of geological formation through which groundwater has been flowing for millions of years. Movement of uranium and its decay products has been investigated by drilling a series of holes through the ore body and surrounding layers. The results indicate that migration of only a few tens of meters has occurred on the weathered surface, and virtually no movement has taken place underground.

So with responsible behavior designing and implementing the MMRS long-term nuclear waste system, there is reasonable historical assurance that future disasters will probably be avoided even if some failures should occur in that system.

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