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This is a preliminary report. The information presented is of a preliminary nature and is not intended for use in open literature.

Preliminary Conceptual Designs for Advanced Packages for the Geologic Disposal of Spent Fuel

April 1979

Prepared for the U.S. Department of Energy
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Pacific Northwest Laboratory
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NOTICE

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PRELIMINARY CONCEPTUAL DESIGNS
FOR ADVANCED PACKAGES
FOR THE GEOLOGIC DISPOSAL OF SPENT FUEL

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ABSTRACT

Spent light water reactor fuel contains radioisotopes that present widely varying degrees of potential hazard to the biosphere. The fission products, notably ^{90}Sr and ^{137}Cs , are responsible for the major part of the radiation and decay heat evolution for time periods <1000 yr. The actinides present maintain their potential toxicity for as long as millions of years.

The present study assumes that the spent fuel will be disposed of in mined repositories in continental geologic formations, and that the post-emplacement control of the radioactive species will be accomplished independently by both the "natural barrier," i.e., the geosphere, and the "engineered barrier system," i.e., the package components consisting of the stabilizer, the canister, and the overpack; and the barrier components external to the package consisting of the hole sleeve and the backfill medium.

The present document provides an overview of the nature of the spent fuel waste; the general approach to waste containment, using the defense-in-depth philosophy; material options, both metallic and nonmetallic, for the components of the engineered barrier system; a set of "strawman" criteria to guide the development of package/engineered barrier systems; and four preliminary concepts representing differing approaches to the solution of the containment problem. These concepts use:

- a corrosion-resistant metal canister in a special backfill (2 barriers)
- a mild steel canister in a corrosion-resistant metallic or nonmetallic hole sleeve, surrounded by a special backfill (2 barriers)
- a corrosion-resistant canister and a corrosion-resistant overpack (or hole sleeve) in a special backfill (3 barriers)
- a mild steel canister in a massive corrosion-resistant bore sleeve surrounded by a polymer layer and a special backfill (3 barriers).

The lack of definitive performance requirements makes it impossible to evaluate these concepts on a functional basis at the present time.

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INTRODUCTION

A program to develop suitable packages^(a) for the safe handling, transportation, storage, and disposal of spent nuclear fuel was initiated in early 1977 by the U.S. Department of Energy (DOE) in response to the executive decision to forego plans for the reprocessing of spent commercial light water reactor (LWR) fuel. This program, originally called the Spent Unreprocessed Fuel (SURF) program, was designated the Spent Fuel Handling and Packaging (SFHP) program in early 1978. It is managed for DOE by the Rockwell Hanford Company.

Though the objectives of the program have been modified as emphasis has shifted from spent fuel storage to spent fuel disposal, Pacific Northwest Laboratory (PNL) has had the task since the program's inception of designing an "advanced package" for the spent fuel. An "advanced package" is a package that offers a greater post-emplacement (disposal) longevity in a given disposal medium than a "baseline package." In addition, it may offer a greater degree of resistance to breaching during predisposal handling, transportation, and storage. A baseline package in its simplest form can be thought of as a mild steel canister containing spent fuel and helium gas. The design of the baseline package was the responsibility of the Rockwell Hanford Company.

The dual advanced package/baseline package design approach has been followed up to the present time because of the lack of package performance criteria which define: (1) the required response of the package to design basis accidents during handling, transportation, and pre-emplacement storage; (2) the required package longevity under disposal conditions; and (3) the

(a) The "package" ordinarily comprises the spent fuel and those elements of the engineered barrier that are emplaced as a unit in the repository. It includes the spent fuel, the stabilizer material (the medium lying between the inner canister wall and the spent fuel), the canister (the primary container), and the overpack (a canister container attached to the canister). The barrier system consists of the package, the hole sleeve(s), and the backfill (embedment) medium.

acceptable failure rates during each phase of the package's life. Such performance criteria still do not exist, though a major effort has been mounted by the Office of Nuclear Waste Isolation (ONWI) to develop them.

This report **summarizes** only that part of the SFHP program done by PNL that was directed toward the development of candidate advanced packaging concepts for the disposal of spent nuclear fuel. This effort took place in the latter part of 1978 and the first quarter of 1979. The overall PNL **SFHP** program was recently terminated due to a reorientation of the basic spent fuel packaging program that placed less emphasis on advanced package designs.

It is not possible at the present time to present a spent fuel packaging concepts report that has adequately addressed all facets of the extremely complex problem, with all of the relevant data tied together in an orderly, coherent fashion. We have attempted in this report simply to bring together that information that existed at the time of the task termination, so that the technical data and packaging concepts that have been generated would be available to the sponsor. We present an overview of the nature of spent fuel waste and discuss repository and waste package concepts. The section on package requirements presents criteria to guide the development of **package/**engineered barrier systems. In addition, we discuss barrier material options, both **metallic** and nonmetallic. Next, we describe candidate barrier system concepts that represent differing approaches to the solution of the containment problem. The final section presents a summary and discussion of the previously presented material. Three separate supporting studies, dealing with: (1) the state-of-the-art of canister design; (2) the effect of stabilizer material on the **maximum** expected fuel cladding temperature; and (3) an assessment of polymers as long-lived barrier materials, all performed under the sponsorship of the PNL package design **subtask**, are included in their entirety as appendices to this report.

CHARACTERISTICS OF SPENT FUEL WASTE

The present task specifically addressed the problem of disposal of commercial LWR spent fuel. No other type of waste form was considered.

In the United States, spent fuel originates primarily from two basic types of nuclear reactors: boiling water reactors (BWRs) and pressurized water reactors (PWRs). In either case, the individual fuel rods consist of stacks of sintered UO_2 or $\text{UO}_2 + \text{PuO}_2$ pellets contained within a thin-walled metallic tubing, or "cladding". The great majority, perhaps 90%, of LWR fuel is clad in zirconium alloy, the remainder in stainless steel. The pellets possess the basic strength, hardness, and insolubility characteristics ordinarily associated with a hard, high-fired, refractory ceramic material.

In a typical, present-day BWR fuel assembly, the fuel rods are arranged in a 7 x 7 square array. Advanced fuels will utilize an 8 x 8 fuel rod array. A typical 8 x 8 assembly will have a length of 14.7 ft (4.47 m) and a 5.47 x 5.47 in. (13.9 x 13.9 cm) cross section, and will weigh approximately 615 lb (279 kg).

A typical PWR assembly contains 264 fuel rods arranged in a 17 x 17 square array, with an overall length of 12.6 ft (3.85 m), cross-section dimensions of 8.43 x 8.43 in. (21.4 x 21.4 cm), and an overall weight of approximately 1480 lb (669 kg).⁽¹⁾ At the present time, the spent fuel storage pools at nuclear reactor sites contain a total of approximately 20,000 assemblies. By the year 2000, it is estimated that more than 300,000 spent fuel assemblies will have been discharged from LWRs in the United States.^(2,3)

A typical UO_2 fuel consists initially of a mixture of two isotopes of uranium: ^{235}U , a fissionable isotope, usually present at a concentration of 3% to 4% and ^{238}U , the common, nonfissionable uranium isotope. The nuclear reactions that take place in an operating reactor are extremely complex; the fission of the ^{235}U atoms alone gives rise to hundreds of isotopic products. Neutron capture reactions produce an array of transuranic elements, which in turn participate in decay, capture, and, in some cases, fission reactions. Thus, the composition of a fuel pellet at any point in time depends on the

original fuel composition, the neutron energy spectrum, the time of exposure, the pellet temperature, the exposure duration, impurities present in the pellet, etc. Computer codes, such as ORIGEN,⁽⁴⁾ have been developed to assist in predicting the isotopic makeup of reactor fuel under various operating conditions. Tables of isotope inventories in spent fuel as a function of time after discharge have been prepared. (2,5)

The radioactive species present in spent fuel are generally grouped into two separate categories to simplify the approach to waste management concepts, viz., (1) short-lived fission products, and (2) long-lived fission products, heavy isotopes, and heavy isotope decay products. The short-lived fission products, such as ^{90}Kr , ^{60}Co , ^{137}Cs and ^{85}Kr , with a maximum half-life of about 30 yr, make by far the greatest contribution to the decay heat and radiation (primarily in the form of gamma rays) given off by the spent fuel in the first few hundred years after discharge from the reactor. The long-lived fission products and heavy isotopes, including ^{99}Tc , ^{29}I , ^{79}Se , ^{237}Np , ^{210}Po , ^{239}Pu , ^{241}Am , ^{213}Bi , and ^{238}U (which decays eventually to ^{226}Ra), constitute a radiation hazard to the biosphere that can persist for millions of years. The heavy isotopes formed by neutron absorption generally decay by alpha emission. For this reason, the isotopes present in nuclear waste can be roughly categorized as "beta-gamma emitters" (short-lived isotopes) and "alpha emitters" (long-lived isotopes).

An additional source of radiation from spent fuel waste arises from activation of the structural materials in the fuel cladding and assembly hardware. Some of the key isotopes produced are ^{60}Co , ^{63}Ni , ^{95}Zr , and ^{55}Fe . The time period over which these isotopes are hazardous is approximately the same as that of the short-lived fission products, though the intensity of the radiation produced is far less.⁽⁶⁾ On the other hand, the relatively long-lived isotope ^{14}C , with a half-life of 5,730 yr, is produced by the reaction of neutrons with nitrogen present as an impurity in the fuel pellets. This isotope merits special concern because of the potential for its entry into the biological carbon cycle.

Spent fuel contains approximately 96 wt. % of the uranium originally present, 3 wt. % of fission products, and 1 wt. % of transuranic elements and daughter products.⁽⁷⁾ Most of the fission products and essentially all of the heavy elements stay within the ceramic pellets during the entire period of reactor exposure. Because the pellet temperatures are much lower upon fuel discharge than during reactor exposure, there is little further tendency for migration of radioactive species from the pellet to the pellet/clad annulus.

However, some fission products, both solid and gaseous, do reside in or near the annular gap where they have migrated during reactor operation. The precise distribution of radioactive species and the nature of the chemical compounds that the species form are not well understood. However, it is obvious that the radioactive isotopes residing in or near the gap represent those species most apt to enter the fuel assembly environment if the fuel cladding is breached. Over very long time periods, of course, or in the presence of a very aggressive solubilizing agent, both the radioactive material in the pellets and the material in the annular (or plenum) region constitute hazards to the environment. In the case of relatively short time periods, such as those periods associated with transportation, handling, and packaging of assemblies from spent fuel storage pools, or in the presence of reagents relatively compatible with UO_2 , e.g., cold water, the precise position of the radioactive isotope of concern within the spent fuel pellets may be important in estimating the degree of hazard to the environment.

Fission gases collecting in the annulus and plenum regions of fuel rods obviously have the highest potential for release in event of a cladding breach. Of the fission product gases formed, only ^{85}Kr , with a 10.72-yr half-life, offers the potential for a radiation hazard from spent fuel that has been stored for a significant period of time (years). An appreciable quantity of Xe isotopes are formed as fission products, but their short half-lives make the hazard from this source negligible. The amount of fission product gases residing in the gap/plenum region of a fuel rod depends on the power history, hence pellet temperature history, of the given fuel element.

Fission gas release of up to 15% of the total formed has been observed in the case of PWR fuel that had attained a relatively high temperature.⁽⁸⁾

In addition to fission gases, it has been found that certain of the more volatile fission products collect at the pellet/cladding gap, especially in the region near pellet cracks. Cubicciotti et al.⁽⁹⁾ note that the elements Cs, I, Mo, Te, Pd, and Ba have been observed in the fuel/cladding reaction zone; and, because of their volatility, the minor yield fission elements Cd, Sb, Sn, and Ag can also be expected to deposit to some extent in that region.

As has been noted, the chemistry of the pellet/cladding reaction zone is complex and not well understood. The Cs constituent, for example, can be present in many different compounds, including CsUO_4 , CsI , CsTe , and Cs_2O , depending on the temperature and activity of the specific reactants. As in the case of the fission product gases, the amount of fission product deposit depends on the power history of the fuel rod in question.

The leaching of radioactive constituents from defected fuel rods has been the subject of only a few controlled, quantitative studies. On the whole, spent fuel storage pool experience has indicated that such leaching effects are not rapid in water at near-ambient temperatures, as there have been no unusual problems encountered in maintaining a satisfactorily low activity of pool water even in pools containing a significant number of fuel elements with ruptured cladding.⁽¹⁰⁾ Peehs, et al.,⁽¹¹⁾ describe, in quantitative terms, the evolution of Cs from a defected PWR fuel rod in ambient water as a function of time. It is not clear, however, that they have absolutely separated internal leaching effects from external (crud dissolution) effects. Katayama⁽¹²⁾ studied the leaching behavior of fragments of UO_2 fuel of 5.81 wt. % enrichment, irradiated to 54,450 MWD/MTU, in deionized water and typical Hanford area ground water at 77°F (25°C). He found that the relative leachability of the elements decreases in the order $\text{Cs} > \text{Sb} > \text{Sr} + \text{Y} > \text{Pu} > \text{Cm}$. (It is interesting to note that the fission products Cs, Sb, and Sr are removed at rates consistent with the fission product volatility ranking of Cubicciotti, et al.⁽⁹⁾) Lorenz, et al.,⁽¹³⁾ studied the release of fission products from "simulated" LWR fuel, i.e., specimens of clad UO_2 pellets with

the pellets precoated with radioactively traced CsOH , CsI , and TeO_2 , in atmospheres of flowing steam or air. The tests were performed in a relatively high temperature regime, 932° to 2372°F (500° to 1300°C) over short time periods (to 20 hr), to simulate transportation accidents and loss-of-coolant accident (LOCA) conditions. While the results are not directly applicable to disposal conditions, they are applicable to possible handling or transportation accidents that might occur pre-emplacment. It was found that the rate of release of Cs and I increased markedly with increasing temperature. In one test, for example, in which the specimen was held in flowing steam at 2012°F (1100°C), 36% of the Cs was lost from the specimen in the first hour. It was found that the rate of simulated fission product release was strongly dependent on the manner in which the cladding of the specimen was defected, i.e., rupture by internal pressurization (highest release rate) or defected by premachining (lowest release rate).

No attempt has been made in this discussion of fuel element characteristics to attach a degree of importance to the absolute rate of isotope emission from a fuel rod under various failure conditions. This type of radioisotope source term analysis and application is beyond the scope of this spent fuel package concepts document. This brief overview of spent fuel waste is intended only to give the reader with little background in reactor fuel technology some understanding of the origin of spent fuel waste, its chemical and physical makeup, and the complex nature of the radiation hazards associated with its great variety of isotopic constituents.

REPOSITORY CONCEPTS

The elimination of the hazards associated with nuclear waste can be accomplished most obviously, and perhaps simply, by disposing of the waste in a repository so remote from the biosphere that no postulated credible event could permit **it** to re-enter the biosphere in potentially harmful quantities. A variety of concepts have been proposed to effect such permanent isolation, including: (1) conventionally mined repositories in continental geologic formations; (2) disposal in very deep drill holes; (3) placement in rock formations in a manner such that the surrounding rock melts; (4) disposal in seabed sediments or deep underwater trenches; (5) ejection into outer space; and (6) disposal in the Antarctic ice cap. ^(7,14,15) Of these, the concept of a repository mined conventionally in continental geologic formations has received the most attention, and **it** is generally considered to be the repository concept that offers the greatest promise for near-term use. The PNL Geologic Package Development Program considered only this mined repository approach to spent fuel disposal in its conceptual design studies.

Different candidate geologic media are being considered for siting a mined repository for the disposal of high-level nuclear waste and spent fuel. The candidate media include: (7)

- rock salt, both bedded and dome
- anhydrite (calcium sulfate)
- granite, and other crystalline rocks
- shale, and related rock types
- flood basalt
- tuff
- "unsaturated rock", i.e., rock lying above the water table in arid regions of the U.S.

In any of these media, radioisotope entry to the biosphere is expected to be controlled by a combination of a long path length to the biosphere; a slow,

or nonexistent, rate of groundwater flow toward the biosphere; and absorption or precipitation of the radioisotopes along the water pathway.

Each of the candidate media has its own peculiar combination of favorable and unfavorable features for repository siting. Though **it** is not possible to generalize too extensively without knowing the characteristics of a specific site, the following qualitative observations may be made based on the average bulk characteristics of the media being considered: ⁽⁷⁾

- Rock salt has been studied more extensively than any of the other candidate media; its very existence denotes remoteness from aquifers; **it** has a high thermal conductivity; **it** tends to self-heal through plastic flow; and is easy to mine. On the other hand, salt has a high solubility; brines are potentially aggressive towards waste forms and canisters (or overpacks); pockets of brine and gas are associated with salt deposits; and salt does not provide the sorptive qualities of other rock types. Its tendency to flow under stress could pose an impediment to waste package retrieval.
- Anhydrite has a high thermal conductivity; **it** is chemically stable; **it** is much less soluble than salt; **it** has a much lower thermal expansion than salt; and **it** has a higher sorptive capacity than salt. However, anhydrite is brittle, and fractures that develop do not self-heal; **it** also must be considered, like salt, to be a soluble rock.
- Granite and similar crystalline igneous and metamorphic rocks are strong; they are chemically stable; they have a high temperature tolerance and a low porosity; the sorptive properties of such sites are considered good; the groundwaters associated with granitic deposits are noncorrosive; and the mining characteristics of such sites are predictable. Such deposits do not exhibit the high thermal conductivity and tendency to self-heal associated with salt deposits. In the event retrievability is desired over long time periods the resistance to self-healing must be considered a positive attribute.

- Shales exhibit a low permeability, plastic flow under lithostatic loads, and good sorptive qualities. The composition of groundwaters in shale varies widely--some would be aggressive toward canister materials. Difficulty is anticipated in keeping tunnels open in shale deposits because of its tendency to flow under stress. This could pose a problem in waste package retrieval. Shale as a medium has not been well characterized.
- Basalt is a very strong rock type; it has a high melting temperature and a moderate thermal conductivity; flow basalt formations are relatively impermeable; and groundwaters associated with basalt are generally of low ionic strength, hence not particularly aggressive toward waste forms and package materials. Groundwater flow and radionuclide transport through basalt formations are not well understood and are currently under study.
- Tuffs are products of volcanic ash depositions. They display a wide range of properties depending on the specific conditions existing during the original deposition. "Densely welded tuff" exhibits a high density; a low porosity; a low water content; a high thermal stability; and a compressive strength, thermal conductivity, and thermal expansion comparable to basalt. The welded tuffs have a significant fracture permeability and can function as aquifers. "Zeolitic tuff" has a relatively low density, a high porosity, a low permeability, a high water content, and high sorptive properties for radionuclides. Zeolitic tuff has a moderate compressive strength and moderate thermal conductivity. One repository design concept involves placing radioactive waste in welded tuff, which is relatively stable at high temperatures, and obtaining radioisotope sorption benefits from the layers of zeolitic tuff lying above and below the welded tuff layer.
- Unsaturated rocks, i.e., rock strata overlying deep water tables in some parts of the southwestern United States, have been proposed for repository siting because of the very low water flows through such formations and the high sorptive capacity of some of the common rock types. Such formations do not have a good thermal conductivity, and so the waste

would require either dilution or longer surface cooling prior to emplacement. Also, the shallower burial involved, from a few hundred to a thousand feet, increases the likelihood of exhumation by future man.

In all of the current concepts of conventionally mined geologic repositories, with the exception of the unsaturated rock candidate medium discussed above, a suitable rock formation is selected at a depth of 2000 to 4000 ft (600 to 1200 m), and one or more vertical shafts are sunk to that level. A grid of tunnels and drifts extends in a near-horizontal plane from the shaft bottoms. The waste packages are lowered from a surface cask to an underground transporter for final deposition in a prepared borehole located in a tunnel floor. After some prescribed period of time, i.e., the period of "easy" retrievability, the tunnels containing waste are backfilled; and, eventually, the entire facility is backfilled and sealed off from the biosphere. The waste is then considered to be disposed of, and no further effort on the part of man, except for routine monitoring activities, is expected to be required to ensure its permanent isolation.

The operation of a facility for disposal of high-level waste/spent fuel in the United States is still some years in the future, as a specific site for such a disposal facility has not yet been officially selected. Such a selection involves assurance that (1) the site is outside a region of high seismicity and volcanism; (2) the groundwater motion is slow and the path long; (3) the rock formation itself is sufficiently extensive and of the desired configuration; (4) the physical and chemical nature of the host rock are compatible with the waste form and waste packaging systems characteristics; and (5) that the site is not near commercially important mineral deposits. When such sites become officially selected, site-specific packaging system concepts can be addressed that take into account the environmental peculiarities that packaging systems must ultimately be compatible with, such as temperature, water chemistry, pressure, radiation stability, etc. Until that time the behavior of packaging systems must be roughly approximated by applying generic data, and package design and material selection must be approached conservatively.

WASTE PACKAGE CONCEPTS

The design of packages for the geologic disposal of high-level waste, whether it be calcine, vitrified waste, or spent fuel, has received very little attention. Almost no effort has been expended in either justifying the package as an integral element of the defense-in-depth radionuclide control philosophy, or in estimating the potential lifetimes under geologic disposal conditions of the package designs that have been proposed.

The major high-level nuclear waste package development efforts in the United States and abroad are summarized in Appendix A of this report.

Only in the case of the Swedish waste package development program has a systematic, detailed effort been made to design and justify package concepts for long-term, post-emplacement radionuclide control.⁽¹⁶⁾ The incentive for the Swedish program is contained in the Statement of Policy of the Swedish Government of October 8, 1976, viz., "...nuclear energy plants currently under construction may not be commissioned until the power company concerned can demonstrate...absolutely safe storage...of the high level waste..." The Swedish KBS (Karnbranslesakerhet) project was formed from four Swedish nuclear power utilities to demonstrate "how HW or spent fuel can be handled and finally stored"; "where a final storage...can be situated"; and "to describe the safety of the proposed arrangements..." The work has been directed by a group of approximately 20 people, and approximately 450 people have made contributions to the program. Because of the extent and timeliness of the KBS investigations, the work will be discussed here in some detail. The approaches used for vitrified waste and spent fuel will be described separately.

VITRIFIED WASTE DISPOSAL

In the proposed Swedish waste management scheme, the fuel will be reprocessed 2 to 10 yr after it has been taken from the reactor. The high-level reprocessing waste will be vitrified using the French AVM process.

The vitrified waste will be initially contained in cylindrical stainless steel vessels. These steel-clad waste packages will be stored at the reprocessing plant for at least 10 yr. After this initial storage period the packages will be shipped to an underground, air-cooled, dry storage facility where they will remain for about 30 yr. After this time period the packages will be prepared for disposal by encapsulation in 0.24-in. (6 mm) thick titanium casings. To reduce the intensity of radiation emanating from the packages, hence the radiolytic decomposition of the groundwater eventually expected to surround the package, a 3.9-in. (100 mm) thick layer of lead will be "inserted" between the steel cylinder and the titanium casing. The packages, now ready for disposal, will be placed in holes approximately 39 in. (1 m) in diameter and 15 ft (5 m) deep in the floors of tunnels approximately 1500 ft (500 m) below the surface of the ground. A "buffer mass" consisting of a mixture of quartz sand and bentonite will be packed around each package. After all holes are filled, the entire tunnel system will be filled with a mixture of sand and bentonite similar to that used in the storage holes.

A "reference group" made up of members of the Swedish Corrosion Research Institute has concluded that the stainless steel/lead/titanium composite canister could be expected to remain intact for 500 to 1000 yr.

SPENT FUEL DISPOSAL

The long-lived toxicity associated with the presence of actinides in spent fuel prompted the Swedish investigators to propose a more corrosion-resistant package for this service than the stainless steel/lead/titanium package concept offered for vitrified waste disposal. Copper and two ceramic materials, viz., aluminum oxide and glass ceramic of the betaspodumene type, have been selected for further study.

Copper will not react with pure, oxygen-free water. Since little oxygen is expected in groundwaters typical of the Swedish underground disposal sites, a copper package with walls 8 in. (20 cm) thick is expected to last "at least 5000 yr". The possibility of copper attack by sulfates or sulfides is currently under investigation.

A canister made of aluminum oxide would be expected to hydrate in the presence of groundwater, forming aluminum hydroxide. The rate of hydration is extremely slow at the temperatures and groundwater flow rates anticipated in a repository; a corrosion rate of less than 0.8 in. (20 mm) in 10,000 yr is expected. The Swedish corrosion reference group concluded that "...this alternative would appear to provide the necessary prerequisites for achieving a very long service life. Before a final evaluation can be made, however, more detailed investigations of corrosion in the environment would be desirable.. ."

There apparently has been no information published on the corrosion-resistance of the glass-ceramic material under repository-simulating test conditions.

A note in a recent news periodical⁽¹⁷⁾ offers further insights into the continually developing Swedish waste disposal program. It is reported that spent fuel would be encapsulated, after about 40 yr interim storage, in copper canisters 30 in. (77 cm) in diameter with walls 8 in. (20 cm) thick. Each canister will contain 500 fuel rods. The void space in the canister will be filled with lead. The packages will be placed in "depository holes" lined with blocks of highly compressed bentonite. The highly compacted bentonite will allow little water to pass through it and, because it swells when wet, will tend to make the depository holes self-sealing. It is reported that KBS is leaning towards the lead/copper/bentonite system rather than canisters made of alumina because of the relatively straightforward, currently commercial technology associated with the former.

WASTE PACKAGE LONGEVITY

The Swedish approach has as its objective the demonstration of "absolutely safe storage" of high-level waste. But the question remains as to how long a waste package, or engineered barrier system^(a) in a defense-in-depth isolation philosophy, should be expected to last. There are several approaches to answering this question, some of which are listed below:

(a) The package cannot be viewed as acting separately from the rest of the engineered barrier system in this analysis. The hole sleeve and backfill material must also be considered.

- (1) If the geologic formation is to perform all of the radionuclide control function, the barrier system need only last until the end of the retrievability period, perhaps 10 to 50 yr. This would ensure safe rehandling only.
- (2) If the barrier system is expected to perform as the primary radionuclide containment through the time period of high fission-product activity, it would have to last 300 to 1000 yr.
- (3) If the barrier system is to perform as a primary radionuclide containment for periods beyond the ability to predict significant or catastrophic geologic events, a barrier longevity of 5000 to 10,000 yr might be selected.
- (4) If the barrier system is to perform as the primary radionuclide containment during the time period of significant actinide activity, it would have to last for time periods beyond a million years.

Several factors should be taken into account before a barrier system longevity criterion is officially decided: (1) the results of systems studies based on the best available data; (2) the uncertainties inherent in the technical data; (3) the difficulty of predicting the nature and frequency of accidents which might completely nullify "protective" systems; and (4) the vagaries of public and political attitudes and opinions.

Two specific major benefits may accrue from barrier system longevities beyond the minimum retrievability period (10 to 20 yr):

- (1) A barrier system lasting for a substantial period of time (hundreds or thousands of years) offers the possibility of providing a positive barrier to radionuclides in the event that some unforeseen natural phenomenon disrupts the natural (geologic) barrier.
- (2) A long-lived barrier system could be expected to provide some degree of protection to the biosphere in the event of inadvertent violations of the repository by future man.

The benefits noted above provide the primary justification for the "defense-in-depth" disposal philosophy.

If a barrier system is to offer resistance to groundwater incursion for periods beyond that associated with "normal" retrievability (tens of years), then a single barrier concept is not likely to fulfill the requirements of every geology, nor is one system likely to be optimal for a very wide longevity time span. That is, a barrier system designed for a 5000-yr life in a basalt repository probably would not be the same one chosen for a 500-yr life in the same repository for economic reasons, nor would it be the system chosen for a 200,000-yr life for technical reasons. If the repository conditions are appreciably changed, e.g., basalt to salt, the barrier system concept would be expected to change also. Not all of the accommodation to the environment need be accomplished within the package itself, of course. The local environment of the package can be modified by the backfill material, e.g., by addition of relatively impermeable materials, such as cements or clays; the chemistry of the environment can be adjusted by adding deoxidizers, such as metal shot or other reducing agents, or by adding pH-adjusting compounds; and the emplacement holes can be sleeved with a wide variety of strong, corrosion-resistant materials. A well-thought-out combination of stabilizer material, canister/overpack materials, borehole liner material, and repository micro-environment adjustment should permit even barrier systems relying primarily on metallic components to function as primary radionuclide barriers for times up to, and perhaps beyond, 5,000 yr, at least under conditions expected in relatively benign hard-rock repositories. Such barriers could be designed to resist at least some degree of seismic activity and unanticipated groundwater flows during this period.

Swedish investigators claim that their thick-walled copper spent fuel package should last at least 5,000 yr, ⁽¹⁶⁾ because of the inertness of copper in oxygen-free water. It is interesting to note that copper tools made approximately 6,500 yr ago have been discovered in an excellent state of preservation in eastern Europe. ⁽¹⁸⁾ While this does not offer irrefutable evidence of the validity of the Swedish claim for longevity of fabricated copper objects, it is interesting to observe the longevity of such items when they are emplaced in (fortuitously) compatible environments.

If a barrier system is expected to last beyond 5,000 yr, reliance probably would have to be placed on noble metals, such as gold or platinum, or nonmetallic materials, such as glass ceramic, alumina, or graphitic materials. The latter options appear practical and technically reasonable, if special handling precautions are taken to ensure that only sound packages are ultimately emplaced. It must also be shown that post-emplacement shear and tensile stresses induced by deformations in the geologic medium will not exceed the maximum permissible loadings on each component of the barrier system. . .

PACKAGE REQUIREMENTS

For purpose of discussion, the package requirements can be divided into pre-emplacement and post-emplacement (disposal) considerations. The material requirements associated with each procedure, or unit process, are shown in Figure 1. The considerations presented in the figure are generic: The specific end use of the package and the specific set of package performance criteria that will be eventually defined will not substantially change the nature or the sequence of the required operations or the questions that must be asked regarding the performance of candidate package materials at each node of the flowchart. The flowchart includes vitrified waste, calcine, and spent fuel waste forms. Most of the differences in the processing and handling of these different forms, and the associated differences in package material requirements, occur early in pre-emplacement operations.

Defining the contribution of a package to control of radioactive wastes is relatively straightforward for the handling, transportation, storage, and short-time retrieval aspects of the geologic disposal scenario. Though the detailed criteria relevant to these operations have not yet been developed, no great difficulties are expected in meeting the required package performance requirements once they have been officially established.

The situation is considerably different in the case of the post-emplacement, post-retrieval package, for two reasons: (1) The technical requirements of waste disposal regarding defense-in-depth and engineered barrier concepts has not yet been officially promulgated; thus the ultimate role of the package under disposal conditions remains uncertain. (2) The expectable longevity of a package, hence its potential contribution to a barrier system under the environmental conditions expected in mined repositories, is open to technical questions. In the present study, of course, two assumptions have been made a priori: (1) A barrier system capable of acting as a long-lived containment for radionuclides is required. (2) The package is expected to perform a significant role in the barrier system. How good the barrier must be, i.e., how long it must last and at what integrity level, is an extremely important

FIGURE 1. Waste Package Flowsheet with Required Material Characteristics

question, however, as the answer will ultimately drive the development of the required technology. This answer can only be derived from a justified official position on the concepts of engineered barriers and defense-in-depth. For the purpose of the present study, a barrier system longevity sufficient to contain the radioisotopes during the period of high fission product activity was selected.

The technical questions alluded to are much more difficult to answer, since they are obviously not amenable to such a judgmental approach. The major question is comprised of two parts: Can realistic design concepts be generated that can contain radionuclides for times long compared to the lifetime of typical engineered structures? If such a design concept appears to be technically feasible, how can its longevity be proven prior to its being put into practice?

The present study addressed these questions. In order to make technical judgments on candidate package/barrier concepts, "strawman" performance criteria are required. The criteria used are discussed in the following section.

PERFORMANCE CRITERIA

A barrier system for disposal of spent fuel in geologic repositories must withstand a wide variety of adverse thermal, chemical, and mechanical conditions if it is to retain its integrity for time periods beyond those associated with ordinary retrieval (~50 yr). Both "normal" conditions and design basis accident conditions must ultimately be considered in predicting the longevity of the system. The longer the required system life and the more hostile the environment the greater are the design problems and the difficulty of assuring satisfactory performance.

A conceptual design effort requires some firm guiding principles to be efficacious. In the present study, these principles, i.e., performance criteria, were approached by first hypothesizing the information that will be eventually required to permit a satisfactory Title I barrier system/package design. These requirements are presented in Table 1.

TABLE 1. Required Information for Title I Design of an Engineered Barrier System for Disposal of Spent Fuel

1. Detailed description of barrier system end uses
 - a. Package **handling** and transportation requirements
 - b. Period of post-emplacement **package/barrier** system integrity
 - c. Period of required package retrievability
 - d. Acceptable failure rates
2. Description of geologic medium at specific disposal site
 - a. Thermal degradation mechanisms and kinetics
 - b. Response to radiation
 - c. Mechanical properties
 - d. Physical properties
 - e. Chemical composition of environment
 - f. Biological activity in environment
3. Definition of waste characteristics
 - a. Heat generation rate
 - b. **Maximum** permissible fuel temperature
 - c. Radiation characteristics
 - d. Criticality safety parameters
4. Description of credible accidents at specific disposal sites
 - a. Types of accidents
 - b. Frequency of accidents
 - c. Time of occurrence in "duty cycle"
 - d. **Loads/stress** imposed on barrier system components
 - e. Accident-caused changes in environment
5. Materials data corresponding to anticipated interaction of barrier system components with the environment associated with a specific disposal site

The general barrier end use requirements are set in Item 1, Table 1. The information provided under Item 2 will permit an assessment of the response of the geologic medium to the characteristic thermal and radiation emissions from the waste, obtained in turn from the data of Item 3. The result of this interaction, with the credible accident scenarios of Item 4 superimposed, will provide a description of the environment that the barrier system must be capable of resisting. The materials data of Item 5 can then be utilized to produce the Title I **package/barrier** system design to meet the **Item 1 requirements**.

It is anticipated that all of the information requirements outlined in **Table 1** will be considered in developing the licensable design of an engineered barrier system. Essentially none of the data currently exist, with the exception of parts of Item 3, and, in a generic sense, parts of Items 2 and 5.

The present study, involving only the conceptualization of packaging approaches with emphasis on post-emplacement requirements, obviously does not require all of the information of Table 1. A definition of the following items was considered sufficient for the task at hand:

1. Period of required post-emplacement barrier system integrity;
2. Period of required package retrievability;
3. Chemical composition of environment;
4. Maximum permissible fuel temperature;
5. Criticality safety parameters; and
6. Materials data corresponding to anticipated interaction of barriers with the environment.

The first five **items**^(a) in the foregoing list were addressed by assuming the following **strawman** performance criteria:

1. The barrier system is to last **1000 yr**.
2. The package is to be retrievable for 50 yr. Because of the **already-**stated barrier system longevity criterion of **1000 yr**, this requirement is primarily directed at the **borehole** preparation and general facility characteristics.

(a) The last item on the list, "materials data", cannot be arbitrarily selected or assumed.

3. The composition of the environment will be either saturated salt brine, corresponding to (wet) salt repository conditions, or water of low ionic strength, corresponding to benign hard rock repositories. Repository environments must be approximated by using generic data, as site-specific data are not yet available.
4. The temperature of the barrier system will not exceed 482°F (250°C) at any **environment/barrier** interface.
5. The maximum fuel cladding temperature will not exceed 715°F (380°C).⁽¹⁹⁾
6. The package will be designed to prevent criticality by maintaining the spent fuel in an appropriate geometrical configuration.

Additional constraints imposed on the conceptual design approach:

7. Spent fuel will be in the form of nondisassembled fuel bundles only.
8. The spent fuel cladding is not considered a part of the barrier system.
9. Additional degradation of the fuel cladding is not permissible during the life of the barrier system, **i.e., 1000 yr.**

The foregoing listing of **strawman** criteria and constraints is not intended to do more than provide a reasonable, but admittedly highly judgmental, set of guidelines for the generation of concepts for spent fuel disposal in a long-lived barrier system. No accident scenarios are considered; only normal, steady-state repository conditions.

The actual formulation of package end uses and **performance/design** criteria, in other than a **strawman** form, are beyond the scope of the present study; and, such criteria, once they are officially set, may bear little resemblance to those used here.

APPROACH TO PACKAGE/BARRIER SYSTEM CONCEPT DEVELOPMENT

The basic elements of a multiple barrier system approach to spent fuel disposal is shown in Figure 2. The barrier elements shown in the figure include those associated with the "package", **viz., the stabilizer(s), canister, and overpack(s)**; as well as those barriers considered external to the

FIGURE 2. Basic Conceptual Design Incorporating All Barrier Elements

package, viz. , the hole sleeve(s) and the backfill material (embedment medium). All of the barrier elements shown, with the exception of the geologic medium, are covered under the term "engineered barriers." The geologic medium is considered a "natural" barrier. A "defense-in-depth" approach means that the engineered barrier system is designed to provide, with a very low probability of failure, complete protection against radioisotope release for some prescribed time period, exclusive of any assistance from the natural barrier.

As noted previously, the barriers provided by the package cannot be functionally separated from those engineered barriers external to the package, when control of radionuclides after package emplacement is the issue. Thus, the package longevity criterion of 1000 yr assumed in the present study actually means "engineered barrier longevity", as the entire sequence of engineered barriers must be viewed as interactive elements of a total system. On the other hand, if retrievability is an issue, only the ease of extraction of the package itself from the repository would be considered, though the elements external to the package, such as the hole sleeve, could exert a profound effect on the ease of package removal, especially in a relatively plastic medium.

The relatively complex concept shown in Figure 2 can be compared to the most simplistic concept,^(a) with a minimum number of barriers, shown in Figure 3. All of the "optional" elements have been removed from the multiple-barrier concept, leaving only those absolutely required, in theory at least, for spent fuel handling, emplacement, and radionuclide control.

It is not possible at the present time to make a general recommendation for either a simple approach (Figure 3), a complex approach (Figure 2), or something in between. The final barrier element system design will have to be tailored to the geologic medium at the site in question, and data regarding

(a) It can be argued that even simpler engineered barrier systems can be conceived, e.g., a fuel assembly encased in a monolithic, homogeneous casting, without a canister element. It is not the purpose of the present discussion, however, to define precisely the boundaries of "simple" and "complex."

FIGURE 3. Simple Conceptual Design Incorporating Minimum Barrier Elements

materials performance in conditions found at the specific repository site must be obtained. It is possible, however, to discuss the role of each barrier element as it is currently perceived and make some comments on possible approaches to candidate package concepts.

Stabilizer

The stabilizer can play a number of roles in the concept of spent fuel packaging. If it is a solid material, such as metal (e.g., lead, aluminum, or zinc), a low-melting glass, or a ceramic (e.g., a preformed alumina shell), it has the potential to contribute substantially to the overall corrosion-resistance of the package and, in fact, could provide the major contribution to resistance to attack by the repository environment. In addition, a solid

stabilizer would help prevent the possibility of a criticality situation by controlling the geometry of the spent fuel. Processing difficulties, material costs, and the lack of ductility of the nonmetallic options are negative aspects of solid stabilizers.

Particulate stabilizers, notably sand, have received a great deal of attention by package designers. These materials circumvent most of the processing problems associated with solid stabilizers, as no complex high-temperature melting/pouring operations are required. However, a particulate stabilizer would provide minimal resistance to incursion of repository water, and so cannot be considered an absolute barrier. It is possible, however, that such stabilizer materials could provide a strong impediment to radionuclide movement by their sorption characteristics. Criticality control by geometrical constraint can still be accomplished by particulates, though not with the same degree of assurance that one would expect with a solid stabilizer.

Gaseous stabilizers would present by far the simplest processing situation, though no barrier is provided and no criticality control can be expected. An inert gas, such as helium, simply provides a nonaggressive medium for the fuel cladding and could perhaps offer a processing advantage, e.g., ease of leak checking.

Canister

The canister provides a first level of containment for the spent fuel and stabilizer. As the first level container, it would be expected to withstand any thermal, chemical, or mechanical traumas imposed by processing requirements, e.g., in acting as a mold for molten metal or glass. In addition, the canister might be expected to bear the brunt of some of the shock loadings, fatigue stresses, and extreme environmental conditions associated with all routine and accident handling and transportation conditions. The canister is ordinarily thought of as being a metallic vessel, though this is not an absolute requirement.

Overpack

The overpack is similar in principle to the canister. An overpack offers several options to the package designer: (1) It can be thought of as a second canister, applied (during pre-disposal handling or storage) if the first one fails, or is in danger of failing. (2) It can provide a level of uniformity to a variety of canister types, applied at the repository to accommodate acceptance criteria. (3) It can exhibit corrosion or mechanical properties superior to those of the original canister, thereby providing all, or the major part, of the resistance to the environment required by the package longevity criteria. (4) It provides primary containment redundancy, if required, for all stages of package handling, transportation, and emplacement. The overpack, like the canister, is also generally thought of as being a metallic vessel, though this need not be the case. The long-lived Swedish package concept,⁽¹⁶⁾ for example, employs an alumina container that in the present context would be considered an overpack.

Hole Sleeve

The hole sleeve's primary function, in current repository concepts, is to maintain an open borehole for easy package insertion and retrieval. This is especially important if the geologic medium is plastic, e.g., rock salt and certain shales. However, there is no reason that the borehole liner cannot assume a role in the multiple barrier concept, perhaps even a dominant role. It is conceivable, for example, that certain extremely effective barriers could more easily be formed or built in situ than transported and emplaced with the canister. Examples might include massive shells of special cements, cast in place, polymer barriers located some distance away from the package for thermal or radiation consideration, or impervious graphite vessels specially bedded in the surrounding rock. The final sealing of a borehole sleeve would obviously have to be done after package emplacement, and the appropriate technology would have to be developed to ensure a satisfactory seal.

Hole Sleeve Coating

The hole sleeve coating is included here only for the sake of completeness, i.e., to make the point that composite or laminar structures can be considered for all of the barriers discussed, so long as they have the requisite properties and are amenable to all processing operations, such as production of a final seal. An example of a hole sleeve coating might be a polymer layer on, or soaked into, a cement sleeve, a titanium sheet bonded to a cast iron sleeve, a zinc coating on a mild steel sleeve, etc.

Backfill Material

The backfill material, or embedment medium, generally thought of as a particulate material, a formed particulate (such as compressed bentonite), or a cementitious material, has several potential functions. It can be used to accomplish one or more of the following objectives: (1) to impede water access to the package; (2) to selectively absorb radioisotope species from the groundwater in the event of a package breach; (3) to adjust the micro-geochemical environment (pH, Eh,^(a) corrosion inhibitor addition) in the vicinity of the package to reduce the severity of package corrosion; (4) to adjust the Eh of the groundwater to maximize the adsorptivity of the isotopes considered to be the most hazardous; (5) to absorb the water present in the repository; and (6) to provide a mechanical strain relief buffer to the package in the event shear wave loading from seismic activity or large stresses induced by rock movements are anticipated.

Conclusion

An analysis of the foregoing list of potential barriers shows that a large number of barrier system design options are available. A final decision on the optimum system will not only require the site-specific technical data mentioned earlier in Table 1, but also a cost-benefit analysis and a material conservation analysis. Unfortunately, it is not possible to address any of these issues, in other than a qualitative sense, in the present document.

(a) The oxidation-reduction potential of the ionic species in the groundwater.

To develop preliminary packaging concepts, it is necessary to have some idea of the materials that are available that could possibly withstand the anticipated repository environments for adequately long time periods. This discussion is presented in the following section.

BARRIER MATERIAL OPTIONS

In order to evaluate the number of barriers needed, how substantial the barriers should be, how the barriers should be fabricated, the cost of a barrier system, and the impact of a barrier system on available material supplies, it is first necessary to determine what materials offer potentially useful barrier properties over time periods of hundreds of years in a possibly hostile repository environment. The backfill material is not specifically addressed here as a barrier; and the stabilizer, which can potentially be made of a wide variety of materials, would be considered included only if the decision were made to use a solid stabilizer material, either cast in place or made as an impermeable preformed shell. The canister, the overpack, the hole sleeve, and the hole sleeve coating are all considered potential barriers in this discussion.

A list of potentially useful barrier materials is given in Table 2. No claim is made for completeness, as, for example, hundreds of ceramic and polymer materials exist that could possibly offer a desirable property in some barrier system design. The purpose of the list is simply to provide an overview of the material types that should eventually be considered for application. Obviously, the longevity of the materials listed will be extremely dependent on the repository conditions. In the most benign case, e.g., cool, de-oxygenated water of low ionic strength in a low radiation field, all materials could initially be considered candidates. At the highest projected repository temperatures [perhaps 482°F (250°C)], and in the presence of strong radiation fields, only a relatively few of the materials listed, at least in the metal and polymer categories, remain contenders.

Each of the major material groups of Table 2 will be discussed in the following section.

METALS

More data exist on metals in anticipated repository environments than any of the other groups of Table 2, though this does not mean that they are

TABLE 2. Candidate Materials for Long-Lived
Spent Fuel Disposal Barrier Systems

•	METALS	Ti alloys Zr alloys Ni alloys Pb alloys Fe alloys Cu alloys
•	POLYMERS	polyimides polyamide-imides polyphenyl sulfone polyphenylene sulfide polyarylene ethers polyolefins polybenzyls (filled) polystyrene (modified) epoxies (filled)
•	CERAMICS	Al_2O_3 alumina $2\text{Al}_2\text{O}_3 \cdot \text{SiO}_2$ mullite $\text{Al}_2\text{O}_3\text{-ZrO}_2\text{-SiO}_2$ fused ZAS refractory CaTiO_3 perovskite $\text{CaO} \cdot \text{ZrO}_2 \cdot \text{TiO}_2$ zirconolite TiO_2 rutile ZrO_2 zirconia (CaO stabilized) ZrSiO_4 zircon carbides: TiC, SiC
•	CARBON	impervious graphite glassy carbon pyrolytic graphite
•	GLASSES	wide variety
	CEMENTS	high alumina cements specially tailored cements, grouting compounds, or chemical binders.

currently well characterized for this use. Metals are more susceptible to chemical attack than the nonmetallic groups of Table 2, and the specific nature of the environments involved, i.e., brines vs. bitterns vs. hard rock groundwater, can lead to markedly different material requirements. The feasibility of applying metals in salt (brine) environments and hard rock (low ion strength water) will be discussed here in some detail.

Braithwaite and Molecke⁽²⁰⁾ of Sandia, and Shannon^(21,22) of PNL have published data on the corrosion of metals in high-temperature brines. Braithwaite and Molecke used two brines (designated "Brine A" and "Brine B") and seawater in their tests. The compositions of the solutions are given in Table 3. The compositions of the alloys studied are presented in Table 4.

For the deoxygenated brine/seawater studies, the test coupons were sealed in pyrex or quartz ampoules, with an aliquot of solution, and exposed for 2 to 8 weeks at temperature in an autoclave. For the experiments with oxygen the coupons were held in the autoclave in open-ended quartz tubes. The results in deoxygenated brines are shown in Table 5; the results from oxygenated brine tests are presented in Table 6.

The Sandia results demonstrate the feasibility of long-lived metallic barriers, as even in the relatively aggressive oxygenated brines (Table 6) the corrosion rates of Ticode 12 and Hastelloy C-276 appear to be promisingly low.^(a) The data must of course be considered highly preliminary, as the specimens were not stressed; the tests were of short duration; no radiation effects were involved; the solutions were not refreshed; and perhaps not all corrosion attack processes were anticipated, e.g., attack by species not present in the test brines that will be present in a repository site groundwater.

(a) It is currently a matter of some conjecture whether oxygenated or deoxygenated corrosion environments yield data most relevant to eventual repository conditions. After mine closure the conditions could become anoxic; prior to closure oxygen would have relatively easy access to the environment. Design accident scenarios must also be considered.

TABLE 3. Composition of Solutions Used in Sandia Laboratory Studies (20)

<u>Ion</u>	<u>Seawater (ppm)</u>	<u>Brine A (ppm)</u>	<u>Brine B (ppm)</u>
Na ⁺	10,651	42,000	115,000
K ⁺	380	30,000	15
Mg ⁺²	1,272	35,000	10
Ca ⁺²	400	600	900
Sr ⁺²	13	5	15
Cl ⁻	18,980	190,000	175,000
SO ₄ ⁻²	884	3,500	3,500
	0.05	10	10
HCO ₃ ⁻	146	700	10
Br ⁻	65	400	400
BO ₃	--	1,200	10
pH	8.1	6.5	6.5
Total Dissolved Solids	35 g/l	306 g/l	297 g/l

TABLE 4. Nominal Compositions of Alloys Studied
by Sandia Laboratory Investigators(20)

<u>Alloy</u>	<u>C</u>	<u>Mn</u>	<u>Si</u>	<u>Cr</u>	<u>Ni</u>	<u>Mo</u>	<u>Fe</u>	<u>Cu</u>	<u>Others</u>
1018 Mild Steel	0.18	0.75	0.25	--	--	--	Bal	--	--
4130 Carbon Steel	0.3M*	0.5	0.3	1.0	--	0.2	Bal	--	--
Corten A Steel ^(a)	0.1	0.4	0.5	1.0	--	--	Bal	--	0.02 Ti
2 1/4 Cr-1 Mo	0.2	0.8	0.3	2.2	--	1	Bal	--	--
Naval Brass	--	--	--	--	--	--	--	60	39 Zn 1 Sn
90-10 Cupronickel	--	--	--	--	10	--	1.3	88.7	--
SS-304	0.08M	2.0M	1.0M	19	10	--	Bal	--	--
SS-316	0.08M	2.0M	1.0M	17	12	2.5	Bal	--	--
SS-Nitronic 50 ^(b)	0.06M	5	--	22	13	2.25	Bal	--	0.2 Nb 0.2 V
SS-20-Cb ₃ ^(c)	0.07M	2.0M	1.0M	20	34	2.5	Bal	3.5	--
SS-Ebrite 26-1 ^(d)	0.01M	0.4M	0.4M	26	0.5M	1	Bal	0.2M	--
Monel 400 ^(e)	0.2	1.0	0.2	--	66.5	--	1.2	31.5	--
Incoloy 825 ^(e)	0.03	0.5	0.2	21.5	42	3.0	30	2.2	0.9 Ti
Incone1 600 ^(e)	0.04	0.20	0.20	15.5	76.0	--	8.0	--	--
Incone1 625 ^(e)	0.05	0.2	0.2	21.5	63	9.0	2.5	--	4 Nb 0.2 Ti
Hastelloy C-276 ^(f)	--	--	--	15	59	0.7	5	--	4 W
Zircaloy 2	0.12	--	--	0.1	0.05	--	--	--	1.5 Sn 98.2 Zr
Titanium, C.P.	--	--	--	--	--	--	--	--	99.5 Ti
Ticode 12 ^(g)	0.012	--	--	--	0.84	0.34	0.09	--	98.9 Ti
MP35N	--	--	--	20	35	10	--	--	35 Co

* M denotes maximum

TRADEMARKS: a. U.S. Steel;
b. Armco Steel Corp.
c. Carpenter Technology Corp.
d. Allegheny Ludlum Industries
e. Huntington Alloys
f. Cabot Corp.
g. TIMET

TABLE 5. Corrosion Rates of Candidate Alloys in Deoxygenated Solutions 482°F (250°C), 5 MPa (725 psi) (20)

Alloy	Brine A		Brine B		Seawater	
	(in./yr)	(mm/yr)	(in/yr)	(mm/yr)	(in/yr)	(mm/yr)
1018 Mild Steel	0.067	1.7	0.0028	0.07	0.016	0.4
Corten A Steel	0.035	0.9	0.0020	0.05	0.0079	0.2
2-1/4 Cr-1 Mo Steel	0.039	1.0 ^(a)	0.0039	0.1 ^(a)	0.0079	0.2
Lead	0.016	0.4	0.012	0.3	0.012	0.3
Copper	0.0028	0.07	0.0020	0.05	0.0020	0.05
Naval Brass	0.039	1.0	--	--	0.0039	1.0
90-10 Cupronickel	0.0055	0.14	--	--	0.0028	0.07
SS-304L	0.00071	0.018	0.00039	0.01	0.00024	0.006
SS-316L	0.00059	0.015	--	--	0.00020	0.005
SS-Nitronic 50	0.00032	0.008	--	--	0.00012	0.003
SS-20Cb3	0.00028	0.007	--	--	0.00020	0.005
SS-Ebrite 26-1	0.0055	0.016	--	--	0.00020	0.005
Monel 400	0.0012	0.03	--	--	0.0039	0.1
Incoloy 825	0.00024	0.006	--	--	0.00016	0.004
Inconel 600	0.00035	0.009	0.00028	0.007	0.00020	0.005
Inconel 625	0.00020	0.005	0.00039	0.001	0.0047	0.012 ^(b)
Hastelloy C-276	0.00028	0.007	--	--	0.000059	0.0015
Zircaloy-2	0.00039	0.001	--	--	--	--
Titanium C.P.	0.00012	0.003	--	--	0.00024	0.006
Ti code 12	0.000024	0.0006	--	--	0.00020	0.005

(a) Crevice corrosion

(b) Pitting corrosion

TABLE 6. Corrosion Rates of Candidate Alloys in Oxygenated Solutions 482°F (250°C), 1020 psi (7 MPa) (20)(a)

<u>Alloy</u>	<u>Brine A</u>		<u>Seawater</u>	
	<u>(in./yr)</u>	<u>(mm/yr)</u>	<u>(in/yr)</u>	<u>(mm/yr)</u>
1018 Mild Steel	0.27	7.0	0.43	11.0
Copper	0.047	1.2	0.20	5.0
Lead	0.047	1.2	0.039	1.0
90-10 Cupronickel	0.016	0.4	0.028	0.7
SS-Ebrite 26-1	0.0094	0.24	--	--
SS-20Cb3	0.0039	0.1 ^(b)	--	--
Inconel 600	--	--	0.0039	0.1
Hastelloy C-276	0.0024	0.06 ^(c)	0.0079	0.2 ^(c)
Ti code 12	0.00016	0.004	--	--

(a) $O_2 = 600 \text{ ppm}$ in Brine A and 1750 ppm in Seawater

(b) Crevice corrosion

(c) Pitting and crevice corrosion

Shannon^(21,22) has performed a series of investigations of the corrosion resistance of metals in high-temperature synthetic geothermal brines. All of the tests were performed under deoxygenated conditions, and so represent relatively benign (nonconservative) test results. The materials studied are listed in Table 7.

The effect of increasing the NaCl concentration in the brine on corrosion rates exhibited by a number of alloys is shown in Table 8. Note that increasing the NaCl concentration increases the observed corrosion rate. In Table 9, titanium alloys and zirconium have been included, and H₂S has been introduced to the 1% NaCl solution. The 20% NaCl data of Table 9 do not agree with the corresponding data of Table 8, as the test durations were different. Not surprisingly, the lower rates (Table 9) are derived from longer-duration corrosion tests.

The agreement between Shannon's data and the Sandia deoxygenated brine data is good. In both cases, titanium alloys show outstanding performance, with certain nickel-base alloys not far behind. It must be re-emphasized that all of the tests were of extremely short duration, and extrapolation of these data to periods of desired canister longevity, i.e., hundreds of years, only shows feasibility of application of some of the alloys studied.

The foregoing discussion dealt with the application of metals in certain brine environments. Unfortunately, data relevant to material selection for specific hard rock repositories are no more available than those for brine service selection, as specific-site waters have not been used in high-temperature, environment-simulating studies. Inference from general corrosion studies regarding past applications of alloy groups must therefore be used. Also, prior to empirical verification, it is a reasonable general conclusion that metals withstanding the attack of oxygenated brine can withstand the attack of the low-ionic-strength waters expected in hard rock repositories. It would not be reasonable, however, to use the relatively exotic materials required for hot brine service if materials of lesser cost can be used for hard rock applications. The following materials can be considered candidates for benign **nonbrine** applications, for longevities of ~1000 yr in modestly thick sections:

TABLE 7. Nominal Composition of Alloys Tested in Simulated High Temperature Geothermal Brines (21)

Carbon Steels	Carbon	Mn	P	S	Si	Mi	Cr	Mo	N	Cu	Fe	H	O	Al	Other
A570 Sheet	0.13	0.42	0.008	0.017	--	--	--	--	--	--	--	--	--	--	
A538 Pipe	0.20	0.55	0.010	0.019	0.02	0.02	.007	.005	--	--	Rem.	--	--	--	
SAE 1010 Tube	0.10	0.30	0.007	0.020	0.0090	.04	.009	.010	--	0.02	Rem.	--	--	--	
A106 Pipe	0.20	0.55	0.10	0.012	0.15	0.04	.04	.010	--	0.03	Rem.	--	--	--	
SAE 1010 Tube [†]	0.03	0.27	0.017	0.006	0.0050	.03	.009	.006	--	0.06	Rem.	--	--	--	
A538 Tube [†]	0.23	0.71	0.010	0.019	0.15	0.06	.010	.007	--	0.008	Rem.	--	--	--	
API Grade Well Casing Steels															
C75	0.44	1.56	0.008	0.17	0.21	--	--	0.21	--	--	Rem.	--	--	--	
C95	--	--	0.014	0.020	--	--	--	--	--	--	Rem.	--	--	--	
J55	0.41	1.08	0.011	0.013	0.08	0.03	0.03	.004	--	--	Rem.	--	--	--	
K55	0.32	1.03	0.010	0.020	0.09	0.09	0.01	.009	--	0.07	Rem.	--	--	--	
L80	0.29	1.22	0.021	0.033	0.07	--	--	--	--	--	Rem.	--	--	--	
N80	0.28	1.20	0.010	0.012	0.13	0.08	0.04	0.0	--	0.12	Rem.	--	--	--	
P110	--	--	0.019	0.027	--	--	--	--	--	--	Rem.	--	--	--	
Chromium Steels															
4130 Tube [†]	0.23	0.48	0.09	0.011	0.25	0.035	0.82	.06	--	0.007	Rem.	--	--	--	
E-Brite 26-1	<0.001	0.020	0.10	0.011	0.21	0.114	26.23	.01	0.10	0.020	Rem.	--	--	--	
405	0.020	0.25	0.10	0.009	0.44	0.228	13.05	.002	--	0.051	Rem.	--	--	0.27	
410	0.12	0.20	0.09	0.014	0.27	0.224	12.23	.05	--	0.08	Rem.	--	--	<0.01	
430	0.051	0.24	0.11	0.009	0.29	0.304	15.89	.09	--	0.07	Rem.	--	--	0.05	
446	0.067	0.45	0.14	0.011	0.22	0.229	23.20	.06	--	0.06	Rem.	--	--	0.02	
406	0.11	0.36	0.11	0.005	0.26	0.23	12.45	.00	--	0.04	Rem.	--	--	1.75	
439	0.052	0.41	0.31	0.010	0.43	0.222	16.89	.08	--	0.09	Rem.	--	--	0.11	
29-4*	0.01 max	--	--	--	--	0.115 max	2	4	0.02 max	--	Rem.	--	--	--	
29-4-2*	0.01 max	--	--	--	--	2	2	4	0.02 max	--	Rem.	--	--	--	
26-15*	0.06 max	--	--	--	--	0.05 max	2	1	0.04 max	--	Rem.	--	--	--	
2-1/2 Cr, 1 Mo	0.12	0.50	0.02	0.014	0.2	0.11	2.14	105	--	0.18	Rem.	--	--	--	
6X*	0.03 max	1.5	--	--	--	2	22	65	--	--	Rem.	--	--	--	
E-Brite 26-1 Tube [†]	0.01	<0.10	0.105	0.0111	0.1	0.09	22.2	106	--	0.005	Rem.	--	--	0.05	
410 Tube [†]	0.10	0.39	0.10	0.009	0.5	0.33	11.89	.003	--	0.03	Rem.	--	--	0.17	
Titanium															
35-A	0.18	--	--	--	--	--	--	--	0.011	--	0.04	0.006	0.09	--	
15 Ni.	0.01	--	--	--	--	1.5	--	--	0.009	--	0.04	67 ppm	0.08	--	
0.2 Pd	0.03	--	--	--	--	--	--	--	0.01	--	0.16	<100 ppm	0.122	--	0.10 Od
6Al-4V*	--	--	--	--	--	--	--	--	--	--	--	--	--	6	4 V
Special Materials															
Hastelloy C-276*	0.02	1	0.03	0.03	0.05	Rem.	14.5	16.5	15.17	--	--	4-7	--	--	W > 4.5 C 2.5 V 0.5
Inconel 625*	0.05	0.15	--	0.008	0.25	61	22.5	9	--	--	2.5	--	--	0.2	0.2 Ti, 4 Cu
Inconel 600*	0.08	0.5	--	--	0.25	76	15.5	--	--	0.5	8	--	--	--	
Incoloy 825*	0.03	0.5	--	0.015	0.25	42	22.5	3	--	2.5	30	--	--	0.1	Ti 0.9
Zirconium															
Pure ytal Bar															

*Nominal

†Heats used in field test

TABLE 8. Effect of Alloy Composition on Uniform Corrosion⁽²¹⁾

Alloy	Temperature Pressure Oxygen pH		482°F 1000 psi (6.89 MPa) <0.01 ppm 4.6 to 4.8		Corrosion Rate, in./yr (mm/yr)	
	1% NaCl		5% NaCl		10% NaCl	
A570	0.012	(0.03)	0.035	(0.9)	0.043	(1.1)
A538 Heat 1	0.011	(0.3)	0.024	(0.6)	0.036	(0.9)
A538 Heat 2	-		0.06	(0.4)	0.023	(0.6)
C75	0.009	(0.2)	0.016	(0.4)	0.007	(0.2)
1010	0.014	(0.4)	0.038	(1.0)	0.041	(1.0)
4130	-				0.007	(0.2)
2-1/4 - 1Mo	0.003	(0.08)	0.023	(0.6)	0.012	(0.3)
410 Heat 1	0.003	(0.08)	0.004	(0.1)	0.030	(0.8)
410 Heat 2	-		0.009	(0.2)	0.020	(0.5)
E-Brite 26-1 Heat 1	0.002	(0.05)	0.0003	(0.01)	0.0007	(0.02)
E-Brite 26-1 Heat 2	-		0.0007	(0.02)	0.0008	(0.02)
Hastelloy C-276	0.0009	(0.02)	0.0012	(0.03)	0.0017	(0.04)
Inconel 625	0.0003	(0.01)	0.0001	(0.01)	0.0002	(0.01)
Inconel 600	0.0006	(0.02)	0.0009	(0.02)	0.0018	(0.05)
Incoloy 825	0.0004	(0.01)	0.0002	(0.01)	0.0003	(0.01)
29Cr-4-2	0.003	(0.08)	0.0002	(0.01)	0.0006	(0.02)
6X	0.0004	(0.01)	0.0003	(0.01)	0.0003	(0.01)

TABLE 9. Comparison of Alternate Alloys with Carbon Steels
in Oxygen-Free Brines - 1000 psig (6.89 MPa)(21)

(Based on 11-Day Tests)

	Corrosion Rate			
	250°C - 1% NaCl		250°C - 20% NaCl	
	pH 4.8 (in./yr)	H ₂ S 10 mg/l (mm/yr)	pH 4.6 (in./yr)	(mm/yr)
Average of ten carbon steels	0.007	0.18	0.087	2.2
<u>Chromium Alloys</u>				
2-1/4 Cr, 1 Mo	0.003	0.08	0.019	0.48
E-Brite 26-1	0.0006	0.02	0.0006	0.02
405	0.0025	0.06	0.0098	2.5
410	0.0014	0.04	0.120	3.0
430	0.0017	0.04	0.0062	1.6
446	0.0007	0.02	0.0008	0.02
406	0.0022	0.06	0.017	0.43
439	0.0014	0.04	0.015	0.38
29 Cr-4 Mo	0.0006	0.02	0.0008	0.02
29 Cr-4 Mo - 2 Ni	0.0004	0.02	0.0008	0.02
26 Cr-1 Mo - 1 Ti	0.0006	0.02	0.0008	0.02
6X	0.0001	0.003	0.0005	0.01
<u>Nickel Alloys</u>				
Hastelloy C-276	0.0015	0.04	0.0005	0.01
Inconel 625	0.0002	0.005	0.0001	0.003
Inconel 600	0.0092	0.23	0.0004	0.01
Incoloy 825	0.0002	0.005	0.0003	0.008
<u>Titanium Alloys</u>				
Ti 35A	<0.0002	<0.005	<0.0001	<0.005
Ti 1.5 Ni	<0.0002	<0.005	<0.0001	<0.005
Ti 0.2 Pd	<0.0002	<0.005	<0.0001	<0.005
Ti 6 Al-4V	<0.0002	<0.005	<0.0001	<0.005
<u>Zirconium</u>	<0.0001	<0.005	<0.0001	<0.005

- Iron-base alloys
 - Cast irons
 - Fe-Cr alloys
 - Fe-Cr-Ni alloys
- Lead
- Copper-base alloys
- Nickel-base alloys
- Zirconium-base alloys
- Titanium-base alloys

Obviously, as in the brine case, a final selection must be based on the conditions of the final repository site; the cost-effectiveness of each material type; material availability; the characteristics of the barrier system selected, e.g., the number, order, and position of the barriers; the required longevity of each barrier; possible environmental modifications that could promote the longevity of a relatively inexpensive barrier; and ease of barrier fabrication, processing, and final sealing.

POLYMERS

Polymers have not been given much attention in past waste packaging studies, probably because of their recognized sensitivity to heat and radiation relative to metals and ceramics. A combination of proper polymer selection and a barrier system design that reduces the temperature and radiation intensity at the polymer layer appears to offer promise in barrier system design, and would permit taking advantage of the great inherent resistance of polymers to attack by groundwaters and the potential economics associated with polymer materials. The polymer options are treated in more detail in Appendix B of this report.

To qualify for long-lived barrier system service, a given polymer's degradation mechanisms would have to be thoroughly understood, and experimental verification of stability would be required. A major difficulty in accepting polymers for long-lived service is the simple fact that polymer operating experience spans only a few decades, so that empirical evidence of longevity is not available.

CERAMICS

In general, ceramics offer the greatest resistance to chemical degradation. Ceramics may readily be selected that are extremely insoluble in expected groundwaters and that show strong resistance to phase changes and hydration reactions. However, ceramic bodies are far more susceptible to shock-induced fractures than their metallic (or polymer) counterparts, and so a great deal of attention must be paid to the design of the embedment medium or support devices surrounding the ceramic barrier. Also, it is generally true that use of a ceramic vessel leads to more difficulty in processing than one made of metal or polymer, if, for example, sintering reactions are required to effect the final seal.

If a ceramic canister, overpack, or hole sleeve is expected to provide a reliable long-lived barrier to radionuclide migration, it is imperative that methods be available to verify the integrity of the barrier prior to borehole filling or tunnel sealing, as barrier fracture could occur during the emplacement or sealing operation.

CARBON

Carbon in its various forms offers an excellent combination of properties for a long-lived radionuclide barrier. It is extremely inert to essentially all reactive species at the temperatures anticipated in geologic repositories. In the form of "impervious graphite" it exhibits strengths to 10,000 psi (compression). It is relatively inexpensive and easy to fabricate. As in the case of ceramic materials, carbon forms do not exhibit the fracture toughness of metals, and the final seal would require a special process.

GLASSES

A wide variety of glasses and glass-ceramic materials can be considered as candidates for waste barriers, in the form of canisters, overpacks, or hole sleeves. In general, glasses offer easier fabricability than ceramics, with perhaps slightly inferior chemical/physical properties when compared to

the stablest ceramic forms. They exhibit similar problems to ceramics in the need for careful embedment, special final sealing, and final quality assurance.

CEMENTS

A cement or grout that can be cast in place to form a massive barrier appears to offer promise in hole sleeve, or hole-sleeve embedment, applications. Radiolysis of water in ordinary hydration-type cements may preclude their application; also, the temperature levels must be kept below the maximum tolerated by a given cement type. If, however, these technical objections can be circumvented (polymers face a similar problem), then use might be made of these potentially cheap, strong materials. A monolithic cement hole sleeve surrounded by a flexible polymer, for example, would be potentially inexpensive and extremely resistant to water ingress. The final sealing and quality assurance of such a composite system would require special attention.

CANDIDATE BARRIER SYSTEM CONCEPTS

The development of **package/barrier** system concepts for spent fuel disposal, given the **multiplicity** of **design/material** options available, and without having firm knowledge of the **environment(s)** that the system must operate in or a given material's performance in the eventually chosen environment, represents an extremely challenging task. Simplification of the task was attempted by breaking the barrier system into three parts: the stabilizer, the **canister/overpack**, and the borehole. Each of these system components was conceptualized independently, with a "maximum number of barriers" assigned to each concept. ^(a) These were then examined for strong or weak characteristics, then assembled building-block style into the final composite "candidate barrier system concepts." The four concepts resulting from this process are those that appear to offer the strongest incentives for further study.

The basic stabilizer design options considered are shown in Figure 4. Whatever the specific design objectives are for a stabilizer, e.g., mechanical support for criticality preclusion, **it** must not seriously inhibit the flow of decay heat from the package. This area is specifically addressed in the work presented in Appendix C of this report. **If**, for example, reference is made to Figures C-1 and C-5 of Appendix C, **it** can be seen that the stabilizer concept of Figure 4a, even when filled with helium, has an inferior heat transfer to the concepts that have a particulate material **fill** (Figure 4b) or a solid ceramic shape with particulate **fill** (Figure 4c) **if** the particulate is assumed to be alumina, Al_2O_3 , and the solid shapes mullite, $2Al_2O_3 \cdot SiO_2$. The heat transfer performance of the concept of Figure 4d can be compared with the others **if** the thermal conductivity of the solid stabilizer is known, simply by referring to the curve of Figure C-2, Appendix C. Of course, the

(a) It is important to recognize that a "zero barrier" concept (Figures 3-5) does not mean that no barrier exists; but only that the engineered barrier concept described does not ~~add~~ a long-lived barrier to the pre-existing constraints to radionuclide dispersion, i.e., the fuel pellet, the fuel cladding, and the geologic medium.

a. Gas stabilizer
Number of barriers* 0

b. Particulate fill throughout
Number of barriers? 0

c. Solid ceramic shapes outside assembly; gas or particulate material inside assembly
Number of barriers? 0

d. Solid metal, glass, or ceramic material surrounding, or throughout, assembly
Number of barriers? 1

* Refers to the number of barriers added to the pre-existing constraints to radionuclide dispersion (the fuel pellet, the fuel cladding, and the geologic medium).

FIGURE 4. Basic Stabilizer Design Options

fuel assembly and canister characteristics cannot differ appreciably from those assumed in Appendix C if the results are to retain quantitative significance.

The concepts of Figures 4b, 4c, and 4d offer, in general, increasing degrees of support for the fuel and increasing processing complexity. Each of these three stabilizer concepts would preclude **criticality**^(a) even in the event of fuel rod disintegration with optimum mixing of fuel particles with water, if they held their positions relative to the fuel; i.e., if they constrained the fuel-water mixture to a region no larger, or only slightly larger, than the original cross-sectional dimensions of the fuel assembly.⁽²³⁾ Obviously, Concepts 4c and 4d offer potentially superior performance in this regard. Because of the processing complexity of 4d and because it is felt that effective radionuclide barriers can be more easily obtained outside the stabilizer component, Concept 4c was chosen as the most promising stabilizer concept.

The basic **canister/overpack** options considered are shown in Figure 5. The concepts range from a zero-barrier, thin, mild steel can (Figure 5a) to a double-barrier, **canister/overpack** design (Figure 5c). Concepts 5b and 5c differ only marginally, in that a "no-credit" steel can is associated with 5b. Such a can may provide processing ease, if it is on the inside; or handling and emplacement protection for a relatively fragile nonmetallic vessel, if on the outside.

It appears reasonable to incorporate at least one barrier into the basic **canister/overpack** design to insure some degree of package integrity during interim **handling** and storage; so Concepts 5b, 5c, and 5d are considered to be viable options for inclusion in the overall system. Note that Concepts 5b and 5c differ only by the equivalent of a mild steel vessel. Concept 5d is equivalent in concept to the Swedish lead-lined titanium can.⁽¹⁶⁾

(a) This statement is based on a criticality analysis of a single spent fuel package containing a single fuel assembly. Multiple-package arrays were not included in the analysis.

- a. Thin-walled. iron-base alloy canister *

Number of barriers: 0

- b. Thin-walled. iron-base alloy canister (Figure 5a) surrounded by overpack of corrosion-resistant alloy, e.g., cast iron, lead-, titanium, or copper-base alloy for hard rock application; titanium- or nickel-base alloy for salt application; or nonmetallic overpack, e.g., ceramic, graphite, glass, or cement tailored to specific application.

Note: Position of canister and overpack materials may be reversed.

Number of barriers*: 1

- c. Corrosion-resistant alloy or nonmetallic canister only

Number of barriers*: 1

- d. Concept of Figure 5c with corrosion-resistant metallic or nonmetallic overpack

Number of barriers? 2

* Refers to the number of barriers added to the pre-existing constraints to radionuclide dispersion (the fuel pellet, the fuel cladding; and the geologic medium).

FIGURE 5. Basic Canister/Overpack Design Options

The basic borehole barrier design options are shown in Figure 6. There are as many ways to prepare a borehole as there are to prepare a package; the concepts shown represent only a few of the many possibilities. It can also be immediately observed that whether a barrier goes on the package or is included in the borehole makes no difference to the number of barriers or the basic system configuration. Where the barrier goes depends on relatively subtle processing, handling, and emplacement considerations, which are beyond the scope of this effort. There may be sound reasons for preventing deformation of the geologic medium surrounding the borehole, perhaps to control displacement of the prepared backfill or perhaps to prevent overall rock shifting, which might prejudice the integrity of the repository strata. If these considerations were to be considered important, a stout sleeve inside the borehole, rather than an overpack at the canister, may be the preferred choice.

It appears highly desirable to have the borehole sleeved with a material that will last at least as long as the desired period of retrievability, e.g., ~50 yr. It may not be necessary to have a true barrier associated with the borehole, however, especially if a multiple-barrier canister/overpack concept is used. Thus, all of the borehole concepts shown in Figure 6 are considered viable options.

It should be noted that, despite first appearances, none of the concepts shown in Figure 6 are extremely complex. Attention would have to be given to assurance of final barrier integrity, especially in the case of required barriers made of brittle materials, and to effecting the final seal.

The four candidate barrier systems are shown in Figure 7. The maximum number of barriers ranges from two to four. The identities of the overpack and hole sleeves become inconsequential in these concepts, as previously mentioned. The derivation of the final four concepts is summarized in Table 10, along with the number of barriers ascribed to each component.

Each of the four final concepts is applicable to either salt or hard rock repository conditions. Suitable choices of materials for each component, including the backfill material, would have to be made depending on the anticipated environment.

- a Mild steel liner, Porous backfill or no backfill, or simple hole in rock *

Number of barriers: 0

- b. Mild steel liner or no liner with specially tailored backfill

Number of barriers? 1

- c. Specially tailored backfill, thick nonmetallic liner (e.g., ceramic, glass, graphite, special cement)

Number of barriers: 2

- d Concept of Figure 4c, but with polymer layer surrounding liner *

Number of barriers: 3

- e. Specially tailored backfill, corrosion-resistant alloy liner (e.g., iron- or copper-base alloy for hard rock nickel- or titanium-base alloy for salt)

Number of barriers? 2

- f. Concept of Figure 4e, but with polymer layer surrounding liner

Number of barriers: 3

* Refers to the number of barriers added to the pre-existing constraints to radionuclide dispersion (the fuel pellet, the fuel cladding, and the geologic medium).

FIGURE 6. Basic Borehole Barrier Design Options

a Corrosion-resistant metal canister in specially tailored backfill

Maximum number of barriers? 2

b Mild-steel canister with corrosion-resistant overpack or corrosion-resistant hole sleeve, in specially tailored backfill

Maximum number of barriers* 2

c Corrosion-resistant canister with corrosion-resistant overpack or corrosion-resistant hole sleeve, in specially tailored backfill

Maximum number of barriers* 3

d Mild-steel canister in thick, corrosion resistant bore sleeve with surrounding polymer layer, in specially tailored backfill

Maximum number of barriers? 3

With corrosion resistant can: 4

* Refers to the number of barriers added to the pre-existing constraints to radionuclide dispersion (the fuel pellet, the fuel cladding, and the geologic medium).

FIGURE 7. Candidate Barrier System Conceptual Designs

TABLE 10. Source of Components for Final Four Concepts

	<u>Stabilizer</u>	<u>Canister/Overpack</u>	<u>Bore Hole</u>
concept 7a (2) ^(a)	4c (0)	5c (1)	6b (1)
Concept 7b (2)	4c (0) or 4c (0)	5b (1) 5a (0)	6b (1) 6c (2) or 6e (2) ^(b)
Concept 7c (3)	4c (0) or 4c (0)	5d (2) 5c (1)	6b (1) 6e (2)
Concept 7d (3,4)	4c (0) or 4c (0)	5a (0) 5c (1)	6d (3) ^(b) 6d (3)

(a) Number in parentheses denotes number of barriers.

(b) Considered a less desirable approach.

The possible contributions of a backfill material to radionuclide control were discussed briefly in the PACKAGE REQUIREMENTS-CONCEPT DEVELOPMENT section of of this report. Backfill options are not treated here in detail because of the tremendous diversity of potential backfill end use objectives and the variety of materials, both natural and man-made, used singly or as mixtures, that can be thought of as candidates for backfill applications. In each of the four barrier concepts presented here the assumption is made that the backfill material can qualify as a true barrier. This can, in theory at least, be accomplished by virtue of water absorption, water permeability, or removal of radionuclide species from the groundwater.

One concept, 7d, employs a polymer layer; it is positioned at distance from the spent fuel to minimize thermal and radiation effects.

The distinction between stabilizer Concepts 4b and 4c, Figure 4, is not considered critical at our present state of knowledge. Stabilizer 4c could be replaced by 4b with no serious loss of credibility. Only a better understanding of credible accident scenarios will permit such a choice to be made with confidence.

SUMMARY AND DISCUSSION

Four preliminary engineered barrier concepts for spent fuel disposal are presented for consideration. The concepts are based on what are considered to be reasonable intended barrier end uses and a reasonable interpretation of existing materials performance data. They are, therefore, considered to be feasible approaches to long-lived barrier disposal philosophy, with sufficient inherent variety in the group and flexibility in the individual approaches to ensure against the overlooking of a potentially desirable material or configurational option. The "building block" approach to engineered barrier design used here is recommended to future studies, as it tends to simplify a complex task while allowing a methodical assessment of each feature. Obviously, such an approach must not be allowed to preclude the eventual overview and optimization of the system as a whole.

The concepts presented here obviously do not encompass all of the possible barrier options, and the concept selection process used is necessarily judgmental. An analysis of the overall approach taken should, if nothing else, help focus attention on the key issues currently inhibiting the technological development of barrier systems in the disposal of spent nuclear fuel:

- How long should the barrier system last?
- What degree of barrier redundancy is required?
- What are the repository steady-state conditions?
- What are the credible accident scenarios?

Once these questions have been answered, the relevant materials data may be obtained and the package/barrier system processing approach and emplacement concepts can be confidently addressed, with the ultimate objective being the simplest, most cost-effective barrier system for a given repository site.

Some of the strawman criteria adopted in the present study may be overly severe: e.g., the 1000-yr barrier longevity and the 482°F (250°C) maximum package temperature. It may be sufficient that a barrier system last only through the "thermal period," i.e., the period during which the temperature of the repository medium peaks and begins to fall off. This period,

wherein the "natural" barrier is considered to be most vulnerable to thermally-induced breaching, would easily lie within a barrier longevity of 300 yr. Or, it could be decided that a time period for barrier longevity sufficient for the spent fuel radiotoxicity to decay below that toxicity level found in the original uranium ore would be sufficient; this time period is less than 1000 yr. ⁽²⁴⁾ The maximum package temperature was arbitrarily assumed to be high, i.e., 482°F (250°C). This temperature is obviously strongly dependent on the age of the fuel, the near-field thermal conductivity, and the designated areal power loading on the repository surface. It is not difficult to imagine maximum temperatures <300°F (150°C) at the canister surface by suitable adjustment of these parameters. Also, it must be recognized that the temperature permitted in the barrier system will eventually not only be dictated by repository considerations, it will also be dictated by barrier longevity criteria.

Even with the rigorous strawman criteria conditions assumed, it appears that application of any of the four concepts is feasible in either salt or hard rock repositories. For salt (brine) application, titanium-, zirconium-, or nickel-base alloys, in thicknesses of <1 in. (2.5 cm); or ceramic or graphite materials, in thicknesses of perhaps 2 to 6 in. (5 to 15 cm); or polymers (assuming adequately low temperatures and radiation fields) of <1 in. (2.5 cm) thickness; appear to be candidates for long-lived barriers. Cements, to exclude water, and special backfill materials, for water sorption, ion sorption, or ion retardation, may be required in much thicker sections, e.g., from several inches to several feet (tens of centimeters).

For application in waters of low ionic strength, alloys of iron, e.g., cast iron, in thicknesses of perhaps 2 to 6 in. (5 to 15 cm), and lead- and copper-base alloys, in thicknesses of 1 to 4 in. (2.5 to 10 cm), can be added to the above list. (It must be recognized that some barrier materials, such as lead and copper, are potentially toxic in their own right, and so must be used with caution.) A final material selection can be made only when the given repository site conditions are known, and when a complete understanding of the candidate material's response to the specific environmental conditions has been obtained.

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APPENDIX A

LITERATURE REVIEW OF CANISTER TECHNOLOGY FOR HIGH-LEVEL WASTE DISPOSAL

R F. Geisendorfer

LITERATURE REVIEW OF CANISTER TECHNOLOGY FOR HIGH-LEVEL WASTE DISPOSAL

The technical status of canisters for the disposal of high-level nuclear waste has been reviewed by screening the available unclassified literature published through August 1978. The review has been accomplished as a part of Task II, Package Design, of the PNL Geologic Package Development Project.

The informational data base was established primarily from the following sources:

- Engineering Index
- Chemical Abstracts
- U.S. Patents
- NTIS (National Technical Information Service)
- NASA (National Aeronautics and Space Administration)
- STAR (Scientific and Technical Aerospace Reports)
- IAA (International Aerospace Abstracts)
- Nuclear Science Abstracts
- DOE Energy Database (June 1976 on)
- Nuclear Safety Information Center
- CIRC II (DOE-LLL Terminal - unclassified information only)
- METADEX (Metal Abstracts/Alloys Index)
- ENVIROLINE (Environment Information Center)
- Pollution Abstracts
- Oceanic Abstracts

Canisters, per se, were only infrequently the subject of a given study, but were usually considered as one integrated aspect of waste management. Accordingly, it was necessary to review the literature on waste management, and, in particular, that of high-level waste solidification, in order to extract canister information. From the informational data base, several hundred documents were reviewed and screened for pertinence. The results of the review and screening process are reported here as (1) a general bibliography; (2) an annotated bibliography; and, (3) a table summarizing canister development. The reported results should be considered as representative of the state-of-the-art of canisters rather than as all-inclusive. The intent has been to present a balanced view of recent, current, and conceptual canister designs relative not only to geologic disposal but also to other options, such as space disposal. In addition, concepts such as "self burial" are included in the annotated bibliography for the same reason.

Several obvious difficulties and/or problem areas hindering canister development for high-level waste disposal were detected during this literature review:

- The specific waste form(s) and permanent disposal method(s) have not yet been sorted out sufficiently to permit firm canister design criteria to

be established with confidence, or to allow for the realistic selection of materials of construction based on sound engineering and scientific judgment.

- Most worldwide R&D efforts have centered on the solidification aspects (processing) of the waste and the resulting waste form. Although such effort is of course crucial, inadequate attention has been given to the canister. Past canister efforts, with few exceptions, have been neither intensive nor extensive relative to canister design, materials, fabrication, or testing. All too frequently the canister is described as "cylindrical and of stainless steel" with little evidence cited supporting the choice of design, material(s) selected, or fabrication steps.
- Past emphasis has been on canister designs for temporary or interim storage with inadequate consideration given to permanent or long-term disposal requirements. For example, a single-wall, Type 304L stainless steel canister may well be an adequate choice for interim storage--it is certainly well characterized from the standpoint of fabrication and corrosion--and such a vessel may serve adequately as the "first wall containment barrier" for permanent disposal. However, it appears likely from the literature review that a multiple barrier approach will be needed for permanent isolation or disposal. Consequently, this multiple barrier aspect should be considered in the initial container design (for temporary and/or interim storage) to insure compatibility with outer barrier designs, materials and fabrication techniques. Sweden appears to be the world leader in the multiple-barrier approach to canister development and recognizes the advantages of "pure materials" relative to long-time metallurgical stability.
- Little evidence of "cross-fertilization" with other national (and international) technological or scientific resources relative to canister development has been shown. For example, in the U.S., perhaps the automotive industry could be tapped to aid in canister fabrication techniques and cost-effective design, while the aerospace industry's approach to damage tolerant design presently applied to airframes may be of benefit to canister designs. Additionally, university corrosion and elevated temperature materials research expertise could be used more fully.

In summary, worldwide canister development for permanent disposal of high-level waste is presently at a rather low state of technological development with apparently an inadequate use of available scientific and technological resources. Most innovative approaches to canister design for permanent disposal exist as concepts only--particularly noteworthy are those of Sweden. However, canisters for temporary or interim storage are more highly developed, appear adequate, and have been demonstrated full-scale in some cases. Some innovation is also evident. The major impediment to further timely development of canisters for permanent disposal appears to be the lack of definitive design criteria.

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ANNOTATED BIBLIOGRAPHY

GEOLOGIC STORAGE

UNITED STATES

1. An Analysis of the Technical Status of High Level Radioactive Waste and Spent Fuel Management Systems, JPL Publication 77-69, December 1977. Jet Propulsion Laboratory, Pasadena, CA, prepared for California Energy Resources Conservation and Development Commission.

Examines the technical status of principal U.S. programs for the management and disposal of high-level waste. Among the major unresolved technical questions listed is that of container design which is necessarily predicated on the as yet unsettled issue of the containment criteria the canister must meet.

2. Status of Commercial Nuclear High Level Waste Disposal, Special Report EPRI NP-44-SR, Electric Power Research Institute, Palo Alto, CA, September 1976.

Reviews available technology and concludes that it is adequate for the safe disposal of HLW by several different processes. Points out that a decision to implement and demonstrate a production scale system is needed. Considers the most important variables in canister design as: waste heat generation rate, canister diameter, and number of internal cooling fins (if any)--assumes the boundary conditions used is a maximum centerline temperature of 800°C to minimize devitrification. Although noting that canister sizes vary from 6 to 24 inches in diameter and 2 to 10 feet in length, a canister of 12 inches in diameter by 10 feet in length made from 304L stainless steel with simple drop-in internal fins (presumably of 304L) is recommended.

3. An Appraisal of Underground Radioactive Waste Disposal in Argillaceous and Crystalline Rocks: Some Geothermal, Geochemical, and Hydrogeological Questions, J. A. Apps, N. G. W. Cook, and P. A. Witherspoon, Lawrence Berkeley Laboratory Report LBL-7047, June 1978, pp. 18-20.

Contains a discussion on container integrity. Considers materials either extremely resistant to chemical attack, such as fused alumina, corundum or other ceramic, or those which may be in chemical equilibrium with the subsurface environment, such as copper. Points out that specialized expertise and protracted research will be needed before there will be adequate assurance

that barrier containers will indeed contain radioactive waste for up to 10^6 years--a difficult task in that there are no historical records to provide guidance for the long range effects, other than the geological record.

4. Preliminary Canister Size Criteria for Commercial Solidified High-Level Waste, J. R. La Riviere and E. L. Moore, ARH-ST-151, April 1977, Atlantic Richfield Hanford Company, Richland, WA.

Reviews canister size criteria (length, weight, diameter, shape) with respect to the several methods of waste solidification being considered in the U.S., such as calcination, vitrification (in-can melter, continuous ceramic melter), supercalcine, sintered glass-ceramics and metal matrices. Preliminary canister criteria are a cylindrical shape, 6.626 inches to 24 inches outside diameter, up to 17 feet in length and weighing a maximum of 12,000 lbs.

5. Thermal Analysis of a Fuel Cladding Repository Pilot Plant in Salt, H. C. Claiborne, ORNL-TM-5221, Oak Ridge National Laboratory, Oak Ridge, Tennessee, April 1976.

Considers canisters 15 feet in length of "standard 12-inch steel pipe with the fuel hulls compacted to 70% of theoretical density. No further canister design specifics are given.

6. Heat Transfer Considerations in the Canister Storage of High-Level Solidified Wastes, A. P. Hoskins and J. R. Berreth, Report ICP-1090, Allied Chemical Corp., Idaho National Engineering Laboratory, April 1976.

Considers heat transfer as a function of waste form (4 types), permissible heat generation rate, cooling by natural convection in air or water), and canister configurations (annular cylinders and solid cylinders with and without fins). Canister wall temperature during storage was limited to 300°C.

7. Deep Self-Burial of Radioactive Wastes by Rock-Melting Capsules, S. E. Logan, Nuclear Technology, Vol. 21, February 1974.

Examines the rock-melting canister concepts utilizing waste heat from HLW. Descent by gravity achieves deep disposal. Author claims that spherical capsules, 1 to 2 meters in diameter, could reach depths of greater than 10 km. Canister surface temperatures in the range 1150-2400°C are anticipated in basalt. Container materials considered as potentially suitable include the super-alloys up to 1200°C, several refractory metals, Nb-1% Zr up to 1330°C, TZM alloys up to 1430°C, and Ta-10%W up to 1830°C. Recommends investigations of high-temperature corrosion and erosion of container materials in contact with waste oxide melts and molten rock for refractory metals, ceramics, and composites.

9. An Approximate Solution for Self-Burial Rates of Radioactive Waste Containers, H. C. Hardee and W. N. Sullivan, SLA-73-0931; Sandia Labs, Albuquerque, NM, January 1974.

Predicts the migration velocity of a spherical container (heat source) melting through a medium such as rock.

10. Durability of Containers for Storing Solidified Radioactive Wastes, C. L. Angerman and W. N. Rankin, DP-MS-76-66, Savannah River Laboratory, Aiken, SC, 1976.

Based upon 10,000-hour heating tests, the authors conclude that for 100-year storage in unconditioned air, AISI 1020 carbon steel appears to be a better material than other alloys considered such as Cor-Ten A, Type 304L stainless steel, or Inconel 600, from waste-form computability, cost, strength, impact, and oxidation resistance standpoints.

11. Nuclear Wastes as a Heat Source, W. F. Witzig and M. E. Foster, Nuclear Technology, Vol. 38, No. 2, April, 1978, p. 258-263.

Proposes concepts of using solidified fission product wastes as a heat source to produce low-pressure steam in a nuclear waste boiler in addition to providing interim storage. Analysis uses a cylindrical Type 304 stainless steel canister 8.65 inches by 12 ft with a wall thickness of 0.325 inches; however, the desirability of canisters of higher power densities (greater than 303 W/l) is not discounted.

12. A Review of Metal-Matrix Encapsulation of Solidified Radioactive High-Level Waste, L. J. Jardine and M. J. Steindler, ANL-78-19, May 1978, Argonne National Laboratory, Argonne, IL.

Contains an excellent review of metal matrix encapsulation techniques such as casting and powder metallurgy including a survey of fabrication techniques. Although canister design was beyond the scope of the study, a thermal analysis of HLW canisters is presented based upon a cylindrical 304L stainless steel container with internal diameters of 6, 12, and 24 inches and a fill height of 10 feet.

13. Conceptual Design Considerations for the Storage of Solidified High-Level Waste in Canisters at a Commercial Fuel Reprocessing Plant, J. R. LaRiviere and E. L. Moore, ARH-SA-221, November 1975, Atlantic Richfield Hanford Company, Richland, WA

Discusses corrosion aspects of 304L stainless steel.

14. Feasibility of Applying Cathodic Protection to Double-Wall Waste Storage Tanks, E. L. Moore, Atlantic Richfield Hanford Co., Richland, WA, Report ARH-CD-975, June 1977.

Concludes that impressed current cathodic protection to the tanks considered (mild steel) may promote stress corrosion cracking under some conditions.

15. Summary of National and International Radioactive Waste Management Programs (excluding U.S.), K. M. Harmon, PNL-2598, March 1978, Pacific Northwest Laboratory, Richland, WA.

Emphasis is on waste treatment, particularly solidification of HLW. Some information on canisters and/or waste encapsulation methods is presented.

16. Alternatives for Long-Term Management of Defense High-Level Radioactive Waste, ERDA Report 77-43, Idaho Chemical Processing Plant, Idaho Falls, Idaho, September 1977.

Notes that canisters will serve as the primary containment vessel and that further effort is required to determine the design criteria and the conceptual design for post-treatment options being evaluated. Provides a schematic of a typical HLW canister 0.15 to 0.61 m in diameter by 0.6 to 4.5 m in length constructed of "303 series stainless steel" of unspecified thickness.

17. Geological Criteria for Radioactive Waste Repositories, G. D. Brunton, W. C. McClain, Union Carbide Corp., Oak Ridge, TN, Office of Waste Isolation Report Y/OWI TM-47, November 28, 1977.

Recognizes that repository rock must not react to any significant extent with the waste form container.

18. National Waste Terminal Storage Program Management and Technical Program Plan, FY-76 - FY-78, Union Carbide Corp., Oak Ridge, TN, Office of Waste Isolation, Report No. Y/OWI-1, April 21, 1976.

Does not address canisters, per se, but does have provisions for heat transfer/thermal analysis as one part of several long-term generic support studies.

19. Retrievable Surface Storage Facility Alternative Concepts Engineering Studies, ARH-2888 REV, Atlantic Richfield Hanford Company and Kaiser Engineers, July, 1974.

Contains a metallurgical evaluation by a consultant (W. K. Boyd, Battelle Columbus Labs) of five alternative interim (100 yrs minimum) storage concepts. Two of the concepts involve

water as the coolant while three involve air storage. Depending on the storage concept, the waste canister, typically a 12-3/4" O.D. by 10 ft long cylinder of 304L stainless steel 1/4" plate with plain carbon steel cooling fins, may be stored as received or enclosed in an overpack, typically AISI 1010 steel, or 304L stainless steel 3/8" to 14" in thickness.

Such important aspects as canister construction, materials selection, fabrication techniques and subsequent stress relief, external and internal corrosion, and radiation effects are analyzed.

20. A Materials Compatibility Information Data Bank, K. E. Mead, SAND-77-1237C, Sandia Laboratories, Albuquerque, NM, 1977.

Assists in the selection of materials to insure their compatibility with each other and with the environment. Appears to be a useful and versatile source of annotated information to aid the designer and materials engineer.

21. High Level Waste Canister Emplacement and Retrieval Concepts Study, Office of Waste Isolation, Report No. Y/OWI/SUB-76/16509, September, 1976, Boeing Engineering and Construction, Seattle, WA.

Considers several emplacement and retrieval concepts for waste canisters in bedded salt. The canister configuration chosen for the study was a cylindrical 304L stainless steel vessel with a 12-inch I.D. and 12.75-inch O.D. and 10 feet in overall length. The canister ends were hemispherical with the lower end inverted. The maximum allowable surface temperature of the canister was 700°F for purposes of the study.

22. Alternative Solidified Forms for Nuclear Wastes, J. L. McElroy and W. A. Ross, BNWL-SA-5824, Pacific Northwest Laboratory, Richland, WA.

Discusses solidification methods in a multiple barrier context of primary waste form, applicable coatings or films, matrix encapsulation and the canister. Presents a description of the inducto-slag melting process to reduce the volume of cladding hull waste.

23. High-Level Waste Ceramics: Materials Considerations, Process Simulation, and Product Characterization, G. J. McCarthy, Nuclear Technology, Vol. 32, January 1977, pp. 92-104.

Discusses tailor-made ceramics (supercalcine) for use in conjunction with the multiple-barrier HW concept. Four barriers are envisioned, namely, 5 mm diameter ceramic core pellets providing atomic-level fixation of nuclides, an impervious coating for protection against leaching, a metal matrix to encase the coated pellets in monolithic form, and finally, the sealed stainless steel canister, 1 to 2 ft in diameter by 8 to 10 ft in length.

24. Maximum Acceptable Temperatures of Wastes and Containers during Retrievable Geologic Storage, G. H. Jenks, Report No. Y/OWI/TM-42, Office of Waste Isolation, Oak Ridge, TN, August 1977.

Assumes a canister of Type 304L stainless steel for high-level waste and indicates that the maximum acceptable canister temperature is 375°C to limit the increased susceptibility to stress corrosion cracking (sensitization) upon subsequent exposure to water at atmospheric temperatures.

25. Determination of Performance Criteria for High-Level Solidified Nuclear Waste, J. J. Cohen, et al., Report No. NUREG-0279 (PB-269 857), prepared for the Nuclear Regulatory Commission, Washington, DC, by Lawrence Livermore Laboratory, Livermore, CA, July 1977.

Contains a section on canister breaching mechanisms including overheating leading to rapid corrosion by waste, external oxidation in air, creep to rupture, impact leading to rupture, corrosion by water, thermal shock rupture, bursting from internal pressurization, and melting. Considers three examples of construction materials: 1020 carbon steel, 304L stainless steel, and 316L stainless steel. Points out that high-nickel alloys are also conceivable choices.

BRITAIN

26. Problems in the Design and Specification of Containers for Vitrified High-Level Liquid Waste, A. D. W. Corbet, G. G. Hall, and G. T. Spiller, Report IAEA-SM-207/3, British Nuclear Fuels Ltd., Risley, Warrington, U. K.

Examines several important aspects affecting the design and specifications of a container for vitrified HLW. Materials availability, cost, fabricability, and properties such as oxidation resistance, high temperature strength and creep stability, compatibility with glass compositions, and corrosion resistance in fresh and sea water have been addressed relative to materials selection. Authors conclude that KK-40 (25Cr-20Ni-0.4C) has been a satisfactory material until now, but future work will investigate the use of higher nickel content alloys such as INCOLOY 800L to improve container reliability.

SWEDEN

27. Suitability of Titanium as a Corrosion Resistant Canister for Nuclear Waste, S. Henriksson, and K. Pettersson, Karnbranslesakerhet, Stockholm, Sweden, August, 1977.

Assessment of literature pertaining to corrosion of unalloyed titanium in contact with Baltic Sea water. Concludes that a 5-mm thick canister of titanium should have an extrapolated life of 1000 to 10,000 years.

28. **Handling of Spent Nuclear Fuel and Final Storage of Vitrified High Level Reprocessing Waste, Part I - General**, Karnbranslesakerhet, Stockholm, Sweden, 1977.

Contains detailed descriptions of the three major canister design concepts presently considered--steel, lead, titanium, pure copper, and alumina. The rationale for the design of each is also presented.

29. **HIPed Alumina Containers for Nuclear Waste**, Powder Metallurgy, August 1978, p. 372. (See also, Nuclear Engineering International, Vol. 22, Aug. 1977, p. 38.)

Reports the first full-scale fabrication of alumina containers using hot isostatic pressing. The ceramic containers were 2.5 m long with an outside diameter of 0.5 m and a wall thickness of 10 cm. The alumina starting powder was pressed at 1350°C and 15,000 psi in a Quintus (ASEA) press. The next step is to demonstrate that the technique can be used with radioactive material.

30. **Gold and Selective Storage Solves Nuclear Waste Problem**, Gosta Wranglén, *Annals of Nuclear Energy*, Vol. 4, pp. 527-528, Pergamon Press, 1977. (Also, *Environment*, Vol. 20, No. 2, p. 4, March 1978.)

Describes a novel method of disposing of high level transuranic waste that has been separated from the high level waste by reprocessing. According to the proposed method, the transuranic waste is compressed and sintered as an oxide body and enclosed in a relatively small, short cylinder of stainless steel alloyed with boron for neutron absorption and lined with cadmium and lead sheets to eliminate penetrating radiation. The cylinders are then coated with 1 mm of pure gold by electrodeposition (jointless coating) and followed by a thick layer of copper to mechanically protect the gold. The waste canisters are finally enclosed in a thick layer of bitumen and permanently disposed of deep in a suitable geologic formation.

31. **Evaluation Regarding the Feasibility of Titanium as Cladding Material for Nuclear Waste**, Sture Hendriksson and Kjell Pettersson, Report AE-MS-132, AB Atomenergi, August 1977.

Presents the results of a literature study on the properties of titanium such as corrosion and oxidation resistance.

CANADA

32. The Canadian Spent Fuel Storage Canister: Some Materials Aspects, D. G. Boase and T. T. Vandergraaf, Nuclear Technology, Vol. 56, January 1977, pp. 60-71.

Discusses design and materials aspects of concrete canisters for interim (50 to 100 years) storage of spent fuel. The canisters are cylindrical about 5 m high and 2.5 m in diameter. The cavity (0.75 m dia x 3 m high) is lined with steel plate 9.5 mm in thickness and lead 25.4 mm thick. The fuel bundles are stored within the canister in carbon steel baskets of welded construction. Preliminary experimental studies has revealed no serious difficulties with corrosion or deterioration of materials; minor cracking has been observed, however.

33. The CANDU Reactor System: An Appropriate Technology, J. A. L. Robertson, Science, Vol. 199, 10 February 1978, pp. 657-664.

Presents a good overview of the Canadian reactor program. Includes a summary of their waste management program and the major storage options.

BELGIUM (EUROCHEMIC)

34. Operation "Hot Mole", Jean Donea, Euro Spectra, Vol. 8, No. 11, 1969-72.

Contains a preliminary theoretical study, confirmed by lab experiments of the self-burial concept for the disposal of HLW. Considers both spherical and cylindrical containers. A major question is the suitability of container materials at temperatures up to 1500°C. Burial depth of 300 m for a cylindrical container is calculated as attainable for salt media.

USSR

35. Method for Choosing Safe Disposal Conditions for Solidified Wastes, Soviet Economic Mutual Assistance Committee on Peaceful Uses of Atomic Energy, Moscow, 1973, Translation Report ORNL-tr-2811.

Presents nomograms to aid in the selection of storage conditions for glass-like solidified waste with specific heat release of up to 700 W/m³ in finned and unfinned cylindrical containers with radii up to 1.4 meters. Container materials of construction unidentified. Containers are presumably hermetically sealed.

EXTRATERRESTRIAL (SPACE) DISPOSAL

UNITED STATES

36. A Review of NASA-Sponsored Technology Assessment Projects, A. C. Mascy, A. D. Alexander III, and R. D. Wood, NASA Technical Memorandum 78472, May 1978.

Contains a section on technology assessment of the space disposal of radioactive nuclear waste, the first phase of which has been completed. Concludes that final judgment of the appropriateness of space disposal should be made after the second and third phases now underway are completed and then compared with other technical alternatives on an equal basis. Appears more concerned with the techniques of technology assessments than the actual assessment.

37. Analysis of Temperature and Pressure Distribution of Containers for Nuclear Waste Material Disposal in Space, L. E. VanBibber and W. G. Parker, NASA-CR-121186, Astronuclear Laboratory, Westinghouse Electric Corp., prepared for NASA Lewis Research Center, April 1973.

This study considers the safety of a container package after an aborted flight resulting in an impact with the earth. The container studied was composed of spherical layers of alternate materials. The innermost material was the solid nuclear waste, followed by a thin layer of tungsten shielding. LiH shielding was placed next to the tungsten followed by enclosure in Type 316 stainless steel. Re-entry shielding materials such as graphite and teflon initially protecting the canister were presumed to be separated from the container for the post-impact analysis. Results indicated that container integrity could be maintained with partial burial with and without canister deformation but complete burial resulted in creep stress rupture failure in 4 to 12 days at power levels of 5 KW and above.

ICE SHEET DISPOSAL

FEDERAL REPUBLIC OF GERMANY

38. The Disposal of Radioactive Waste in Ice Sheets, K. Philberth, Journal of Glaciology, Vol. 19, No. 81, 1977.

A method of disposal of HLW in spherical containers (material unspecified) with a shielding layer of lead 10 cm thick and an outer radius of 0.2 m is proposed in which the containers self-bury to eventual depths of 100 to 150 meters in the ice sheets of

Antarctica or Greenland. Suggests advantages are retrievability and safety from contamination even with catastrophic dislocations of the ice sheet.

OCEAN (SEA BED) DISPOSAL

UNITED STATES

39. Seabed Disposal Program, D. M. Talbert, editor, SAND-77-1270, Sandia Labs, Albuquerque, NM, August 1977. Appendix I, Behavior of Candidate Canister Materials in Deep Ocean Environments, W. H. Smyrl, L. L. Stephenson, and J. W. Braithwaite, Sandia Laboratories Report SAND 77-0243, April 1977.

Based upon documented marine corrosion data, steels (including stainless steels), aluminum alloys, and copper alloys were eliminated from consideration since corrosion rates were too high by a factor of 10 to 100 to give the proposed 1000-year life for a 1-cm thick wall. Some other materials were selected for testing--pure titanium and zirconium, Ti-6Al-4V, Zircaloy 2 and 4, Hastelloy C-276, and Udmet 700. The results of 9-month tests under simulated deep ocean conditions showed that all alloys selected could give long life and are candidates for further studies.

PATENTS

UNITED STATES

40. U.S. Patent 4,115,311, Sept. 19, 1978, Nuclear Waste Storage Container with Metal Matrix, Kenneth R. Sump, Kennewick, WA, inventor, assigned to U.S. DOE, Washington, DC.

Describes a storage container for HLW having a gravity-sintered metal matrix providing impact strength and thermal conductivity. Considers copper and iron and their alloys, stainless steels and heat- and corrosion-resistant materials as appropriate. Suggests that AISI 304 or mild steel with a protective coating are suitable container materials.

41. U.S. Patent 4,021,676, May 3, 1977, Waste Canister for Storage of Nuclear Wastes, James B. Duffy, Fullerton, CA, inventor, assigned to U.S. ERDA, Washington, DC.

A cylindrical waste canister with internal fins spaced away from the wall to conduct heat away from the center without producing

unacceptable hot spots in the canister wall is described. Very similar to design currently being evaluated at PNL.

42. U.S. Patent 3,983,050, Sept. 28, 1976, Method for Storage of Solid Waste, William J. Meacham, LaGrange, IL, inventor, assigned to U.S. ERDA, Washington, DC.

Describes a metal canister for long-term storage of calcined high-level radioactive waste that is made "self-sealing" against a breach in the canister wall by the addition of powdered cement (which mixes with water to form a concrete patch) to the canister with the calcine before sealing for storage.

43. U.S. Patent 4,040,480, August 9, 1977, Lawrence M. Richards, Monterey Park, CA, inventor, assigned to Atlantic Richfield Co., Los Angeles, CA.

Describes a novel method of radioactive waste heat absorption and dissipation by the use of the heat pipe principle.

TABLE A-1. SUMMARY TABLE OF CANISTER DEVELOPMENT FOR HIGH-LEVEL WASTE DISPOSAL

Country	Major Stated Containment Objective(s)	Waste Form	Design Philosophy	Design		Technical Status			Actively Worked in 1978?
				Fabrication	Shape/Dimensions (outside)	Conceptual Only	Laboratory Demonstration	Scaleup (Prototype, Pilot, etc.)	
Sweden	"Completely Safe" terminal storage in crystalline primary rock - $\approx$ 3000 meters. Flexibility with respect to future options and technical developments.	Vitrified High-Level Waste	Interim supervised storage for 100 years or longer in stainless steel container subsequently encapsulated in more permanent materials.	Successive layers as follows: 3-4 mm stainless steel 100 mm lead 6 mm titanium. Multilayered canister surrounded by a buffer of quartz and bentonite.	Cylindrical/ 0.4 m diameter 1.5 m length	X			Yes, corrosion studies of candidate materials
Sweden	"	Spent Fuel	Direct disposal of fuel rods by placement in a material with very low reaction rate with host rock. First barrier is the relatively insoluble fuel cladding.	Pure copper 20 cm thick ness made from forging a cast block and boring out of internal cavity The three-part lid to be EB welded each part separately	Cylindrical/ 0.91 m in diameter, 4.9 m in length with hemispherical end caps	X			Yes, corrosion studies
Sweden	"	Spent Fuel	Alternative way of direct disposal of fuel rods by coiling and subsequent placement in a ceramic body.	Al ₂ O ₃ body of 10 cm thickness encased in steel for safe handling. Fabricated by hot isostatically pressing the Al ₂ O ₃ powder to full density at 1350°C and 15,000 psi. Top cap or cover is diffusion bonded to main container during hip cycles.	Cylindrical/ 0.5 m in diameter 2.5 m in length with hemispherical end caps		X 1/3 scale container	X Full scale without radioactive waste	Yes, corrosion, delay failure, fabrication techniques studies.

SUMMARY TABLE (Contd)

Country	Major Stated Containment Objective(s)	Waste Form	Design Philosophy	Design		Technical Status			Actively Worked in 1978?
				Fabrication	Shape/Dimensions (outside)	Conceptual Only	Laboratory Demonstration	Scaleup (Prototype, Pilot, etc.)	
Sweden		Transuranic waste separated by reprocessing and compressed and sintered as an oxide body	Multi-barrier approach using a noble metal in conjunction with radiation absorbers.	Boron containing stainless steel inner vessel lined with cadmium and lead sheet and coated with gold by electro-deposition to 1 mm thickness. The gold coating is followed by a "thick" outer layer of copper and finally a "thick" layer of bitumen (presumably in-situ)	Cylindrical1 "small (2 liters) and short"	X			
Britain	Permanent geologic disposal following interim storage. Also considering emplacement on or under the ocean bed.	Borosilicate glass; Finial /Harvest process.	Container suitable for use during vitrification at 950°C and subsequent interim storage in cooling ponds and/or air cooled vaults with potential for permanent disposal.	Stainless steel (HK-40) single wall container; annular shape also considered is basically two concentric cans. (Also evaluated wrought Incoloy 800). Plan to test lead, copper, gold, and titanium.	Cylindrical1 45-61 cm in diameter. 2.7 m in length, annular shape 122 cm O.D. X 76.2 cm I.D. X 2.76 m High. with wall thickness of 2.5 cm.		X	X Full-scale prototype (est. 1980)	Yes. corrosion studies of candidate materials.
Canada	To demonstrate safe disposal of nuclear waste by 1990s . To isolate NV from man's environment for much longer than hazardous lives by burial in deep seated (1000 m) cavities in igneous rock. "Plutons" - pilot scale repository planned ~ late 1980s.	Spent Fuel bundles	Interim storage (CANDU) container for spent fuel using passive air cooling approach and 3 separate barriers (50-100 yr duration)	Thick low carbon steel container or basket of welded construction contained in a weld-sealed outer steel can. Concrete outer pack is lined with lead 2.54 cm thick.	Cylindrical/ cavity is 0.75 m X 3 m outer dimensions of concrete canister is 2.5 m in dia. X 5 m in length. (square cross-section design with a 0.76 m cavity has also been proposed)		X		Yes

SUMMARY TABLE (Contd)

Country	Major Stated Containment Objective(s)	Waste Form	Design Philosophy	Design		Technical Status			Actively Worked in 1978?
				Material	Shape/Dimensions (outside)	Conceptual Only	Laboratory Demonstration	Scaleup (Prototype, Pilot, etc.)	
France	To provide interim storage in engineered facilities for up to 40 years followed by isolation in a suitable geologic formation.	Vitrified (Borosilicate glass) HLW from Pever process (pot vitrification) and, more recently, the two-stage calcination-vitrification AVM (Marcoule) process.	Process canister must be capable of withstanding temperatures up to 1150°C. It must serve also as the storage container	*Radioactive Stainless Steel (NS30 with C ≤ 0.2w/o)	Cylindrical/50 cm in diameter X 1 meter in length.			X	Yes (AVM process)
Eurochemic	Safe on-site storage 50-100 yrs. Considering waste isolation in claybeds.	Granular phosphate product from Lotes or Pamela process embedded in metal matrix (at 350°C).	Information lacking	Information lacking	Cylindrical/15 cm diameter, 1 m in length.				Yes
United States	Interim air/water storage and subsequent safe isolation of nuclear wastes in geologic media. Isolation of HLW for 1000 years minimum.	Phosphate glass or calcined product	Prototype (WSEP) canisters for simulated storage in water/air up to 10 years.	Mild steel, Types 304L and 310 stainless steel. Welded closure using press-in caps.	Cylindrical bottle/20-30 cm in diameter by length.			X (Radioactive Waste) 196-1970	Yes, canisters in storage being evaluated.
		Spray calcined in-can melted vitrified waste (borosilicate glass)	Canister is a part of multiple barrier system and must be capable of sustaining process melting temperatures of 1050-1150°C as well as surface storage.	304L stainless steel 1/4-inch wall vessel with 8 internal fins of same material to dissipate heat. A gap at the fin-wall interfaces is provided to prevent container wall hot spots.	Cylindrical/20-30 cm in diameter by 1.2 m 2.1 m in length.			X	Yes
		Ceramic-melter produced vitrified waste	Canister must withstand process temps (350°C) and interim storage	Mild steel container, 1/4-inch wall, finned & unfinned, welded closure.	Cylindrical/30 cm in diameter X 23 m in length				Yes

SUMMARY TABLE (Contd)

Country	Major Stated Containment Objective(s)	Waste Form	Design Philosophy	Design		Technical Status			Active/Worked in 1978?
				Fabrication	Shape/Dimensions (outside)	Conceptual Only	Laboratory Demonstration	Scale (prototype, pilot, etc.)	
USSR	Surface storage (above water table) emphasized. Considering deep geologic isolation as well.	Phosphate glass	Information lacking	Information lacking	"200 liter container"		X		?
Federal Republic of Germany	Permanent disposal in salt and/or iron mines.	Borosilicate glass (VERA process) or phosphate glass (Pamela process)	Container suitable for annealing cast glass (VERA) or the incorporation of glass beads in a metallic matrix, i.e., molten lead (Pamela) as well as interim storage and permanent disposal.	"Stainless steel" specific information lacking	Cylindrical bottle/ 21 cm in dia. X 80 cm in length; filling neck is 8 cm X 2.5 cm in diameter.		X	X Plns for pilot plant 192- 1983 with Euro-chemic (Pmela process)	Yes

APPENDIX B

APPLICATION OF POLYMERIC MATERIALS TO ENGINEERED BARRIER SYSTEMS FOR DISPOSAL OF SPENT NUCLEAR FUEL

W. E. Skiens

APPLICATION OF POLYMERIC MATERIALS TO ENGINEERED BARRIER SYSTEMS FOR DISPOSAL OF SPENT NUCLEAR FUEL

INTRODUCTION

This study is an assessment of the feasibility of using organic polymeric^(a) materials in engineered barrier systems for the disposal of spent nuclear fuel packages. Spent fuel barrier systems are expected to retain their integrity for prolonged periods in underground disposal facilities containing potentially hostile (corrosive) environments. Because of these corrosive environments, it was suggested that polymers might provide relatively inexpensive, stable, noncorroding elements in engineered barrier systems for spent fuel disposal.

The conditions assumed at the outset of this study for the stored packages are as follows:

- a. Storage time: ~1000 years
- b. Maximum radiation dose at canister surface: $\sim 1.3 \times 10^{10}$ rads
(Figure B-1)^(b)
- c. Storage environment: salt brine (primarily Na and Mg chloride) or solid salt surroundings. (Salt brines may be expected to be somewhat altered by radiolysis products.)
- d. Temperature: maximum of 250°C

It is also considered possible that some of these conditions might eventually be modified (i.e., made less extreme) in order to permit use of acceptable polymer materials. For example, there are few polymers that can be expected to withstand prolonged exposure at temperatures of 250°C. Also, since nearly 50% of the total radiation dose of the packages would be delivered in the first 6 to 7 years after removal from the reactor, it may be feasible to store the removed assemblies for 5 to 10 years in less hostile conditions before applying a polymeric encapsulant. This would mean that the

-
- (a) Polymers are defined to include all macromolecular materials, i.e., thermoplastic, thermo-set and elastomeric materials.
 - (b) The figure is developed from ORIGEN calculations which assume typical condition of PWR -3.3% enrichment, 30 MW power and burnup of 33,000 MWD/MTHM. (ORIGEN—The ORNL Isotope Generation and Depletion Code, M. J. Bell, ORNL-4628, May 1973.)

expected dose to the polymer would be considerably less than 10^{10} rads (6×10^9 rads after 10 yr) and a number of polymers can be expected to retain good mechanical and chemical properties in this dose range.

It should be pointed out at the outset that none of the polymers being considered for the proposed packaging application have been available for more than about 40 years and most for less than 20. It is therefore only possible to make assessments of the lives of these materials based on relatively short-term observations [1-3]. It must also be emphasized that none of the polymers have been tested under the hypothetical repository conditions outlined above and, as a result, any assessment made from isolated test data cannot reflect the possibility of synergistic effects on the polymers as a result of the total environment.

As an illustration of the type of effects that may occur it should be pointed out that radiation interacts primarily by one of two major mechanisms in polymers--either crosslinking or chain scission [4-8]. Although both of these mechanisms are occurring as any polymer is irradiated, usually one mechanism predominates and the resultant polymer either degrades to lower molecular weights by chain scission or increases in molecular weight as crosslinking occurs. Both mechanisms cause changes in the properties of the irradiated polymer and in many cases these changes are undesirable. Some types of polymers, however, are much more resistant to radiation (that is, their "G values" [9,10] are much lower) than others. In general, polymeric structures containing a high degree of aromatic character are much more resistant to radiation (and frequently also to thermal) exposure than aliphatic structures. However, radiation and thermal resistance or the inverse do not always imply chemical resistance or instability.

Another factor which must be considered in this application is the chemical products (species) produced by the radiolysis of the brine which is expected to surround the buried package. The influence of these species on a polymeric barrier element must be considered in conjunction with the radiation, thermal and normal chemical environment encountered in the salt mine repository.

In addition to the water or water vapor that surrounds the fuel package, the major ion concentrations expected in salt-mine repositories are [11,14]:

<u>Ion</u>	<u>Molarity</u>
Mg	2.3-3
K	0.4
Na	1-2
Cl	7-7.5
B	0.5
pH	Slightly basic at room temperature (may become more acidic at high temperatures)

FIGURE B-1. Dose Rates for Spent Fuel Packages
After Removal from Reactor

The effects of radiation and temperature on this solution will be to produce species such as H_2 , OH^- , Cl_2 or Cl_2^- , Br_2 , O_2 , O_3 , H_2O_2 , ClO^- , ClO_3^- , BrO_3^- , and HCl . Although this certainly does not detail all the possible chemical species that may be generated, many of these will appear only momentarily before decomposing at the relatively high temperatures in the repository. Such species as H_2O_2 , Cl_2 , ClO_3^- , O_3 , and BrO_3^- will decompose rapidly into O_2 , Cl^- (or Br^-) and H_2 . Such compounds as HNO_3 may also be present initially due to the irradiation of entrapped atmospheric N_2 and O_2 in the repository. Of the major chemical species stable under the conditions surrounding the package, perhaps the most hostile to many barrier materials will be HCl . HCl is not a product of radiolysis but rather of the hydrolysis of $MgCl_2$ at the high temperature surrounding the spent fuel package. It would be expected to attack many candidate container materials.

POLYMERIC ENCAPSULATION OF SPENT FUEL PACKAGES

Consideration of polymeric materials in spent fuel package design suggests possible utilization of a number of fabrication methods commonly used in polymer technology. These techniques include coating of a canister, an overpack, or a hole sleeve by painting, dipping, spraying, etc., or slipping the canister into a pre-molded container, or placing the canister (or hole sleeve) inside an extruded bag or sheet which is then subsequently sealed. Each candidate polymer system should be individually assessed for the best method of encapsulant formation. Other techniques such as using polymer/polymer or polymer/metal laminates may effectively combine the best properties of two or more materials. These and other engineering design considerations must be followed up as the study of this concept progresses.

POLYMERS

The materials that have been considered for this application are generally commercially available; however, no economic restrictions have been put on materials in the preliminary study. Thus, materials costing from a few cents to many dollars per pound have been considered.

All of the generic families of plastics and elastomers have been considered with an attempt to understand what their response will be to the environment expected--radiation, thermal and chemical--in salt mine repositories. The assessments detailed on the following polymeric materials are based on data from reports, journal articles, books, manufacturers' literature, and verbal information from manufacturers' technical representatives. Literature searches have been carried out using normal library procedures as well as computer techniques.

The polymers that appear to be potentially useful in this application are listed below with more detailed information on their properties on the following pages. This list makes no attempt to prioritize the materials, but specific recommendations will be made at the end of this report.

Polysiloxanes (silicones)
 Polyolefins
 Polyamides (aromatic nylons)
 Polyamide-imides
 Polyimides
 Polyurethanes
 EPDM rubber
 Polysulfone
 Polyethersulfone
 Polyphenyl sulfone
 Polyphenylene sulfide
 Polyphenylene oxide
 Polyarylene ethers
 Copolyesters
 Polybenzyls
 Epoxy resins

This use of polymers requires little structural strength in application and it appears that considerable radiation damage to the material can occur in many cases without loss of function of the polymer in its role as part of the fuel containment package.

In considering these materials and their properties one often comes away with the feeling that almost all of them might easily fulfill two of the three requirements (radiation, thermal and chemical constraints), but the third factor (and not always the same one) frequently makes the application in this use questionable. For example, the polyolefins (high-density polyethylene or polymethylpentene) can be expected to withstand both the radiation and chemical environment, but the thermal stability considerations (150°C maximum useful temperature) make the use of these materials questionable. In a different vein, the polyimides can be expected to stand up well to the indicated radiation dose and have outstanding thermal stability, but their structure suggests that the chemical environment may cause problems. Nearly all polymers have better long-term stability in an inert gas atmosphere or vacuum than in oxygen or air, thus modifying the atmosphere in the repository before sealing it might be desirable.

The individual polymers are briefly described here, as well as potential suppliers. Their potential usefulness and properties are indicated, along with some recommendations concerning their use.

(1) Polysiloxanes [10,13] (Dow Corning-Silastic, General Electric-Silicones)

The silicones (polysiloxanes), especially those containing phenyl groups and filled with SiO_2 have quite good radiation resistance, although at dose levels of 1×10^{10} rads expected for the entire 1000-year package life, siloxane stability would appear to be marginal. Withholding silicone encapsulation until the package has aged 6 to 10 years (thereby cutting the dose to the encapsulant to about one-half that expected from

a freshly removed fuel package) would enhance this polymer's chance of withstanding the radiation dose significantly. Silicones have excellent thermal resistance in the 200°-250°C range and the chemical resistance appears good, although testing in hot brines should be performed to evaluate this point since no data are available under these conditions.

(2) Polyolefins [10] (DuPont-Alathon, Phillips-Marlex, Mitsui-TPX)

Polyolefins such as high-density polyethylene and polymethylpentene have quite good radiation resistance, generally crosslinking (and toughening). Again, doses of 1×10^{10} rads may be enough to cause significant embrittlement of these polymers and loss of properties beyond that desired. Cutting dose by one-half or more should help. Chemical resistance is excellent in hot brine at least to the temperatures tested (~100°C). The greatest drawback to these polymers is their inability to take temperatures above the 100°-150°C range. If the temperature can be moderated to this range, stability should be good.

(3) Polyamides [14] (aromatic) (DuPont-Nomex or Kevlar)

The completely aromatic polyamides (i.e., Nomex, Kevlar) show very good radiation resistance compared to the aliphatic polyamides (nylon 6, 6/6, etc.). Their (aromatic polyamides) thermal resistance is also excellent. Thus, these materials may be expected to withstand prolonged temperatures in the 200°-250°C range and radiation doses up to 1×10^{10} rads without substantial losses in properties. The chemical resistance is reported to be very good, but testing will be necessary to ascertain long-term stability of the amide linkage in a hot-brine environment.

(4) Polyimides [14-16] (DuPont-Kapton or Vespel, Rhodia-Kinel, Upjohn-2080)

The polyimides are one of the more recently developed high-temperature, radiation-resistant types of polymers. There is a group of polymers in the polyimide family, the best of which are highly stable under high radiation doses (1×10^{10} rads) and thermally stable in the 250°-300°C range. Some of these materials are thermoplastics, although those possessing the highest stabilities are thermosets. The chemical stability of this family is very good, but again, there is the question of the long-term hydrolytic stability in brine at the high temperatures being considered. Testing will be necessary to determine the stability of the imide linkage in the hot-brine environment.

(5) Polyamide-imides [14,17] (Amoco-Torlon)

The polyamide-imide family is a hybrid of the two previously discussed families of polymers and has some of the best properties of both. In general, these are thermoplastic materials. They have good radiation resistance (less than 5% loss in tensile strength at 10^9 rads), and good

thermal stability at 250°C. The heat-distortion temperature of this polymer is reduced somewhat by moisture absorption at higher temperatures; however, under the low stress performance necessary in this application it can be expected to still perform well. Again, because of the lack of data, the question of long-term stability of the amide and imide bonds in the high-temperature brine environment must remain of some concern until some further information is available.

(6) Polyphenylene Oxide [14,18] (General Electric-PPO or Noryl)

This polyphenylether shows good radiation resistance together with excellent hydrolytic stability at elevated temperatures. It can probably stand prolonged exposure to aqueous or brine systems in the 200°C range.

(7) Polyaryl Ethers [14,19-21] (Raychem-Stilan, Uniroyal Arylon-T)

These are highly aromatized ethers which are purported to have excellent radiation stability (no change in tensile strength after 1×10^{10} rads at room temperature). They are also highly resistant to hydrolysis at high temperatures and should perform well in the 200"-250°C range in salt mine environments. Again, however, data is lacking to reveal expected long-term stability in the hot-brine environment--testing is necessary.

(8) Polyesters [22] (Carborundum-Ekkcel)

These aromatic copolyesters appear to have excellent high-temperature (200°-250°C) and radiation (10^{10} rads) stability. The question of hydrolytic stability in the hot-brine environment again has no immediate answer from any available information. It can be expected that the ester functionally will be somewhat sensitive to attack in basic systems, especially at high temperatures.

(9) Polybenzyls [23] (Ciba-Geigy-Xylok)

These polybenzylic compounds contain only C-C bonds in the main chain linkages and thus are reported to possess excellent hydrolytic and general chemical stability. They show only 3% decrease in flexural strength after exposure to 10^{10} rads. These materials are thermosets generally used in conjunction with glass or asbestos reinforcing as fillers, which usually helps to increase thermal and radiation stability.

(10) Polysulfone [14,24] (Union Carbide-Udel)

Polysulfone is a relatively new polymer with some excellent properties. It is considered a high-temperature, hydrolytically stable thermoplastic with good stability to radiation. It is reported to retain good physical properties even after radiation doses of up to 5×10^9 rads. A combined thermal-chemical environment of hot brine plus some radiolytic products from the brine leave an open question concerning stability which can be answered only with test evaluation.

(11) Polyethersulfone [25,26] (ICI-Polyethersulfone)

Polyethersulfone is characterized by the presence of aromatic ether linkages. This sulfur-containing polymer has good thermal stability to about 200°C. It purportedly has good radiation resistance and shows good chemical stability in high temperature aqueous systems.

(12) Polyphenyl sulfone [27,28] (Union Carbide-Radel, Uniroyal-Astrel)

This is another of the aromatic polysulfone family of high-temperature, radiation-resistant polymers. This material has a somewhat higher heat deflection temperature than polysulfone or polyethersulfone, and a continuous service temperature approaching 250°C. It is highly resistant to hydrolysis and chemical attack and can be expected to have good resistance to the hot-brine environment. Among these three sulfone polymers (polysulfone, polyethersulfone, and polyphenylsulfone) there are many similar properties; however, in some cases significantly different characteristics as well as costs exist, and these will need to be considered in detail before selection of materials for experimental studies.

(13) Polyphenylene Sulfide [29] (Phillips-Ryton)

Polyphenylene sulfide is another of the generation of high-temperature thermoplastic resins recently developed. Technical representatives of the manufacturer report little change in flexural strength or modulus of the material after exposure to 5×10^9 rads of γ or neutrons. Initial studies show the chemical resistance to be excellent and the polymer appears to retain its properties after aging in the 200"-250°C range. Although its overall properties would appear to make this polymer a prime candidate for use in the protection of spent-fuel packages in a salt-mine environment, testing of this polymer (especially in the hot-brine environment) must be carried out before its applicability can be firmly evaluated.

(14) Polyurethanes [1-3,30,31]

The polyurethane family of polymers is a broad one and consequently a wide variation in properties is available--from hard, rigid structures to those of an elastomer. However, it appears that selection of monomers with aromatic character and utilizing polymers with ether rather than ester functionality in the chain to provide hydrolytic stability might provide a material with both good radiation and chemical stability. The high-temperature limits for the use of polyurethanes are probably somewhat lower than some of the materials already discussed and these materials probably cannot be employed in systems above 150°C. However, there are many advantages in terms of costs and processing parameters associated with these materials.

(15) Epoxies [1-3,32] (Shell-Epons, Dow-D.E. R., Ciba-Geigy-Arocast, Etc.)

The epoxies represent an extremely large class of polymeric materials and some appear to have properties of interest in the proposed application. Those epoxies containing a high degree of aromatic character in the base resin, as well as in the curing agents, may be formulated to have good radiation and thermal resistance. Chemical resistance is generally good also in many of these formulations, but data is lacking to substantiate their stability for prolonged periods in hot-brine systems. Again, careful study of formulation and some preliminary experimental work should be carried out before including epoxies in an extensive test program.

(16) EPDM [33-37] (DuPont-Norde1, Goodrich-Epcar, Exxon-Vistalon)

This material, together with the silicones and certain polyurethanes, are the only elastomers of the large number reviewed that appear to have good potential for use in the present application. (The newer thermoplastic rubbers as well as the broad group of vulcanizable synthetic and natural rubbers were all evaluated for applicability.) The EPDM synthetic rubbers, prepared as terpolymers of ethylene, propylene and a nonconjugated diene, have properties such as high-temperature stability in aqueous systems that are needed in this application. Study of the available literature on the radiation stability of elastomers indicates that EPDM elastomers can be expected to retain reasonable properties even after exposure to a radiation in the 10^9 rads range. The primary mechanism of radiation degradation of this elastomer is by crosslinking and ultimately brittle failure. Many factors influence an elastomer's radiation resistance: The polymer's molecular structure, the compounding ingredients, the volume and thickness of the sample, the type of radiation, the dose rate and the temperature and environment of the sample during irradiation. The ability of EPDM rubber to withstand high-temperature hot water and/or steam is exceptional as well as its resistance to the attack of ozone and other oxidizing species. Under the expected conditions, EPDM rubbers appear to host the best total property package of the elastomers evaluated, but their use will probably be restricted to temperatures in the 150"-200°C range.

CONCLUSIONS

Among the thousands of polymers that have been synthesized in laboratories, there are perhaps experimental or highly complex polymers that might potentially be more stable as a containment material for spent fuel assemblies in salt-mine repositories than those treated in this review; however, we have dealt only with those materials which are commercially available. Certainly, as indicated earlier, both the total radiation dose levels and the temperatures expected in this application severely restrict the potential polymer candidates available. The total radiation dose expected over a thousand years

($\sim 1-2 \times 10^{10}$ rads for fuel fresh from the reactor), together with the fact that about half this dose is delivered in the first 10 years after discharge from the reactor, indicates that to provide maximum life to a polymer barrier element it would be highly desirable to delay this encapsulation and final storage of the assembly for up to 10 years after discharge. The chemical environment alone does not appear to be a particular problem for many polymers, but the effect of the high temperature together with the brine solution and radiolysis products leave a large area of unknown with nearly all of the materials reviewed. Undoubtedly, whatever can be done to moderate the package temperature would be desirable. Most of the polymers mentioned above can be expected to perform very satisfactorily in this repository environment to about 150°-200°C range, but if temperatures approach 250°C most polymers will not be suitable for long-term use.

Of the sixteen polymer systems described in this study, a few have been selected as the most likely candidates for spent fuel packaging materials. These are selected on the basis of their ability to withstand the following conditions of storage: $\sim 10^{10}$ rads of γ and n radiation, $\sim 250^\circ\text{C}$ maximum temperature and salt-mine brine solutions. If the radiation dose and/or the severe temperature can be further moderated, some other polymers in addition to those mentioned could also become prime candidates.

In consideration of their ability to withstand radiation and in recognition of their excellent thermal stability, the polyimides or polyamide-imides have to be one of the first choices of polymers for this application. The major question is their ability to withstand the chemical environment (although their chemical stability is good) at the elevated temperatures.

Another choice of materials must be the polysulfone family with the polymer of choice being the highly stable polyphenylsulfone. This selection would be followed closely by the related polyphenylene sulfide material which has very good radiation resistance and has been used in systems utilizing hot, flowing geothermal brines [38].

The polyarylene ethers hold a good deal of promise for this application also, being high-temperature, stable polymers with good radiation resistance as well as being very hydrolytically stable.

It must be emphasized, however, that these polymers have never been evaluated under the conditions assumed for this application and, consequently, it has been possible to characterize them only in terms of the individual properties of interest. Although these assumptions may be fairly valid in the case of radiation resistance, evaluation of the effects of high-temperature, salt-mine brine solutions are very difficult to assess. The effects of brine radiolysis products are assumed to be minimal; however, it cannot be stressed too strongly that experimental studies based on conditions similar to those anticipated in the repositories should be initiated in this program as it progresses.

It appears quite possible to accelerate radiation testing of these polymer systems by going to fairly rapid, high-dose tests. Chemical stability testing can be accelerated somewhat in many instances by increasing concentrations of specific ionic species. Thermal testing, however, cannot be carried out at much higher temperatures than those already suggested, since these are already near the temperature limits of acceptable use of organic polymers.

Design of the polymeric covering or packaging for the fuel assemblies also must be a consideration dependent on the methods of handling and burial of the assemblies. Molded containers might be desirable if possible rupture of containment vessels is a concern in the repositories. The design and development of specific enclosures will depend upon further definition of repository conditions.

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APPENDIX C

THE EFFECT OF VARIOUS GASEOUS, PARTICULATE, AND SOLID
STABILIZER MATERIALS ON THE MAXIMUM CLADDING TEMPERATURE
OF A FUEL ASSEMBLY WITHIN A CLOSED CANISTER

R. A. McCann

THE EFFECT OF VARIOUS GASEOUS, PARTICULATE, AND SOLID
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OF A FUEL ASSEMBLY WITHIN A CLOSED CANISTER

All of the concepts for spent nuclear fuel packaging addressed in the PNL Geologic Package Development Project assume that the spent fuel elements do not lose their basic integrity prior to failure of any of the engineered barriers. So long as the fuel rods retain a close approximation to their original configuration, heat loads along the longitudinal axis will be predictable; criticality will not be achievable; and retrieval of an assembly from a package, if necessary, will be a relatively uncomplicated operation. Cladding rupture and concomitant loss of fuel integrity would result if the temperature of the fuel elements were allowed to exceed a critical value, through buildup of decay heat.

The effect of various gaseous, particulate, and solid stabilizer materials on maximum cladding temperature has been examined to provide some insight into the relative merits of several different approaches to spent fuel stabilizer design. The assumed canister is fabricated from a pipe with an internal diameter of 13 in. (33 cm) and a length of 15 ft (457 cm). A single intact PWR fuel assembly is centered radially within the canister and has the following characteristics:

Number of fuel rods	289 (17 x 17 array)
Fuel rod diameter	0.37 in (0.94 cm)
Cladding thickness	0.0225 in (0.0572 cm)
Pitch-to-diameter ratio	1.3
Active length	144 in (366 cm)
Overall length (including tie plates)	160 in (406 cm)

The configuration of tie plates, placement of the active length within the overall length, and other constructional details are generally modeled to reflect a typical PWR fuel assembly. Emittances of 0.4 and 0.3 are assumed for the fuel rods and canister, respectively.

Most of the calculations were performed using a finite difference computer code developed especially for three-dimensional transient and steady-state hydrothermal analysis of fuel assemblies. The code takes into account the coupled heat transfer modes of conduction, convection, and radiation. Thermal radiation among rods and between rods and the canister is modeled in the fashion of Reference (1) and elaborated upon in Reference (2). The velocity and pressure fields due to natural circulation are determined consistent with the conservation laws of mass and momentum. Local (i.e., computational cell) flow resistances along principal directions within the fuel assembly are formulated as suggested in Reference (3) for multidimensional laminar and turbulent flow. The hydrodynamic equations are coupled to the energy equation and solved iteratively. The basic algorithm employed in the solution of the energy equation is an implicit alternating direction scheme described in Reference (4). Donor cell differencing using the cell Peclet number is used as in Reference (5).

The code permits temporally and spatially varying boundary conditions, thermo-physical properties, and heat generation rates. It is also adaptable to other fuel assembly configurations (e.g., three BWR assemblies) enclosed in a single canister. Certain of the results to be described were obtained using a two-dimensional version, for economy, where the three-dimensional version had previously indicated that axial effects were not significant.

The effects of some stabilizers were calculated at more than one assembly power level. The power levels used are considered maximum for their respective ages (time after reactor discharge) and are as follows:

<u>Spent Fuel Age, Year</u>	<u>PWR Fuel Assembly Thermal Power, Kilowatts</u>
3	1.786
5	0.964
10	0.554

All thermophysical properties used are based upon the recommended values found in Reference (6). The effective conductivities of granular materials and composites (i.e., fuel rods) were calculated according to the approach outlined in Reference (7). The uncertainty of effective conductivities based upon analytical models can be quite large, and this uncertainty increases as the disparity between the conductivities of the various phases increases. It is for this reason that experimental determinations are recommended if granular materials are found ultimately to warrant serious consideration as stabilizer materials.

Figure C-1 presents a pictorial representation of the cases that have been examined. It will be noted that the various cases (e.g., IA, IIA, etc.) generally fall into two categories: (1) a single stabilizer medium fills all void space within the canister; and (2) the stabilizer medium contained within the fuel assembly region is different from that found exterior to the fuel assembly. Each case is discussed in following paragraphs.

Figure C-2 shows the temperature difference between the cladding of the hottest (central) fuel rod and the canister temperature as a function of stabilizer conductivity (case ID of Figure C-1). This curve, for five-year old fuel, applies strictly to only those situations where the effects of radiation and convection can be shown to be negligible, i.e., the case of solid stabilizers. The consequences of solid stabilizers such as metallic lead or an aluminum-12.5 wt% silicon alloy may be predicted with confidence. Even the effect of some granular materials with helium--but not necessarily other gases--can be indicated with some degree of confidence, but caution is advised.

Steady-state results for a helium stabilizer (case IA) are shown in Figure C-3. Maximum cladding temperature is indicated as a function of canister temperature with thermal power level as a parameter. The effects at other power levels may be obtained by interpolation. Natural convection, while present, does not have an especially significant impact on the maximum cladding temperature. The shape of the curves generally reflect the increasing heat transfer rate due to radiation and conduction as the temperature increases.

Results for argon and nitrogen gas stabilizers (in addition to helium) are shown for five-year old fuel in Figure C-4. The temperature penalty associated with argon and nitrogen relative to helium is much smaller than might be predicted based solely on their thermal conductivities. The primary reason for this is that convection is now playing a much more important role in determining maximum cladding temperature. This illustrates just one of many situations where, in addition to thermal conductivity, other thermophysical properties are important (in this case, density, viscosity, and specific heat).

A comparison of results for the other cases illustrated in Figure C-1 are presented in Figure C-5. The maximum temperature difference between the cladding and canister versus canister temperature is predicted for five-year old fuel. The results will be discussed in order of increasing heat transfer effectiveness, i.e., efficacy in minimizing cladding temperature. The top curve in the figure, for reference, shows the previously discussed situation of helium gas alone, case IA.

Next in effectiveness are cases IIA and IIIA. The coarse alumina particles surrounding the fuel assembly are sufficiently large (~ 0.3 m diameter) so as not to infiltrate into the fuel assembly. The thermal conductivities of both alumina and mullite reflect 20-30% micro-porosity. The void fraction selected in the region of the alumina particles accounts for the near presence of boundaries and thus is somewhat greater than what would be predicted by theoretical maximum packing. In any event, the potential uncertainty in the analytically predicted thermal conductivity of granular materials can be relatively large, and therefore, the slight difference indicated between cases IIA and IIIA is probably not significant.

Case IC represents fine (approximately 100 micron) mullite particles and helium gas filling the entire canister. The effects of convection and radiation are largely suppressed. Since this is a conduction-dominated situation - at least for helium - this curve is in agreement with that which could be obtained from Figure C-2.

The most effective cases are IB and IIIB. Fine (approximately 100 micron) alumina particles are in the fuel assembly region and either alumina particles or mullite blocks fill the space between the fuel assembly and canister. Helium gas is also present. For reasons mentioned previously, little significance should be attached to the slight difference between these two cases.

The solid stabilizers used as examples for potential package designs are, of course, just two of many candidates. Alumina particles were selected primarily because of superior leach resistance and thermal conductivity. Mullite is a trade-off when fabrication of large blocks is involved.

Other **gaseous/solid** stabilizer combinations could give quite different results. It is understandably tempting to infer the characteristics of **alternate** package designs from other apparently similar designs but this should be done cautiously, especially when natural convection is present. And, it should be emphasized that the use of experimental rather than analytically derived values of granular material effective conductivity is advisable.

The heat transfer results displayed are not necessarily intended as an endorsement of any particular gas or package design since other factors **must** also be considered. Nuclear activation products, chemical reactivity with the fuel assembly, ease of leak detection, structural need for solid stabilizers, leach rate under geothermal conditions, cost, etc. will **all** be influential elements of any package design.

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- IA. GAS (He, Ar, N₂) THROUGHOUT SYSTEM
- IB. HELIUM WITH FINE PARTICLES OF Al₂O₃ THROUGHOUT SYSTEM
- IC. HELIUM WITH FINE PARTICLES OF MULLITE THROUGHOUT SYSTEM
- ID. SOLID STABILIZER THROUGHOUT SYSTEM

- IIA. HELIUM WITH COARSE PARTICLES OF Al₂O₃ OUTSIDE FUEL ASSEMBLY

- IIIA. HELIUM IN ASSEMBLY; MULLITE BLOCKS WITH COARSE PARTICLES OF Al₂O₃ OUTSIDE ASSEMBLY
- IIIB. HELIUM WITH FINE PARTICLES OF Al₂O₃ IN ASSEMBLY; MULLITE BLOCKS WITH COARSE PARTICLES OF Al₂O₃ OUTSIDE ASSEMBLY

FIGURE C-1. Pictorial Representation of Stabilizer Arrangements

FIGURE C-2. Cladding Temperature of Hottest Rod Above Canister Temperatures Versus Thermal Conductivity of Stabilizer for a Thermal Power of 0.964 kW

FIGURE C-3. Maximum Fuel Cladding Temperature Versus Canister Temperature for Helium Stabilizer

FIGURE C-4. Maximum Fuel Cladding Temperature Versus Canister Temperature for He, N₂ and Ar Stabilizers (5-yr-old fuel)

FIGURE C-5. Maximum Temperature Difference Between Cladding and Canister Versus Canister Temperature of 5-year-old Fuel. Refer to Figure C-1 for curve identification.

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