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**STATUS OF SOLIDIFICATION AND DISPOSAL OF
HIGHLY RADIOACTIVE LIQUID WASTES FROM NUCLEAR POWER
IN THE USA**

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ABSTRACT

Solidification of highly radioactive liquid wastes from reprocessing of spent nuclear fuels will soon be required in the USA for additional improvement in safety during interim storage, during transportation to final disposal, and during final disposal at a Federal repository. During the past 15 years, major research and development has been carried out to develop four basic processes for converting the high level liquid wastes to encapsulated solids. These processes are: spray solidification, pot calcination, phosphate glass solidification, and fluidized bed calcination. All processes are being demonstrated on an engineering-scale with radioactive wastes. In all four processes, the waste is heated to 400 °C to 1200 °C to drive off essentially all of the volatile constituents, leaving a solid or a melt that will cool to a solid.

Disposal of these radioactive wastes requires isolation from the biological environment for hundreds of thousands of years. The longevity of the hazard implies that the disposal system should be capable of providing this isolation without surveillance. After considerable study, natural salt deposits were concluded to be the most promising disposal locations in the USA for meeting these criteria. A concept for a facility to dispose of containers of solidified highly radioactive waste in a salt mine has been investigated in an experiment in a salt mine. The overall disposal concept, as well as equipment requirements and handling techniques, were demonstrated.

The US-AEC has proposed that all solidified highly radioactive waste from nuclear power fuels be shipped to Federally owned repositories. Studies are currently in progress on the detailed design and cost estimates for such a first demonstration disposal facility to be located in a salt mine in the central area of the USA.

The cost of high level waste management involving solidification in combinations with interim liquid storage, interim solid storage, transportation to final disposal, and disposal in a salt mine has been estimated to be about 35 to 50 mils/MW hr_e, or only slightly higher than the cost range for perpetual liquid storage.

INTRODUCTION

The conversion of high-level liquid wastes to solids as a pretreatment for storage is being developed in nearly all countries with significant near-future nuclear energy. Solidification is being developed so widely because it is the only reasonably attainable technique for achieving a substantial increase in the safety associated with the storing and disposal of the high-level waste from the nuclear power industry.

Storage of high-level wastes as solids will provide significantly more safety than storage as liquids because solid wastes are:

- Immobile,
- Less soluble in water,
- Considerably smaller in volume, and
- More rugged physically than those stored in liquids.

After solidification, the wastes must be stored virtually forever under stable, controlled conditions. Guidelines for such storage in the USA have been proposed in the Federal Register.^[1] These guidelines propose that all high-level wastes must be solidified within 5 years after the source fuel has been reprocessed, and the solidified waste must be shipped to a Federal repository within 10 years after fuel reprocessing.^[2]

ORIGIN OF HIGH-LEVEL WASTE

The origin and disposition of high-level wastes are shown in Figure 1. Here, one tonne of nuclear fuel is used in a nuclear reactor to generate electricity. The partially spent fuel is then removed from the reactor, aged for a few months, then processed for recovery of usable materials which are refabricated into new fuel elements. About 99.9% of the non-volatile fission products are contained in the aqueous high-level radioactive waste effluent from the reprocessing step. Currently, these aqueous wastes are stored in underground tanks which are susceptible to failure and must be replaced every 15 to 40 years. With solidification, the wastes would then be deposited in locations such as in salt caverns.

Most spent fuel reprocessing is done by the Purex solvent extraction process. The resultant high-level wastes are primarily aqueous solutions of nitrate salts, depending upon the reagents added to the solutions during reprocessing.

SOLIDIFICATION PROCESSING

As a result of research and development conducted in the past 15 years, four processes for solidification of high-level wastes have been developed in the USA to the point of radioactive demonstrations on an engineering scale. The processes are pot calcination, spray solidification, phosphate glass solidification, and fluidized bed calcination.

In all four processes, heat is applied to raise the temperature of the waste to 400 to 1200 °C. At these temperatures, essentially all of the volatile constituents (primarily water and nitrates) are driven off, leaving a calcined solid or a melt that will cool to a solid.

Solidification processes which form melts require significant chemical modification of the chemical composition of the waste. Generally, at least 70 mole % of inert chemicals are required to incorporate the fission products into materials that are meltable at reasonably low temperatures, or less than about 1000 °C. Melts have been developed in which the major meltmaking fluxes are phosphates, [3,4,5] silicates, [6,7] or borates, [8] or some combination of these. [9,10,11] In most cases, potentially workable chemical composition ranges have been defined. [12]

POT CALCINATION

Pot calcination, which was initially developed at the Oak Ridge National Laboratory, [12,13] is a batch process that has been developed to a state of readiness for commercial use. It is presently being demonstrated with full-level radioactivity by Battelle-Northwest in the AEC's Waste Solidification Engineering Prototype, or WSEP. [14,15] The process is simple and is adaptable to a wide variety of feed compositions.

In pot calcination, shown in Figure 2a, liquid waste is added to a pot that is heated in a multiple-zone heating and cooling furnace. The principal processing vessel, the stainless steel pot, is also the final container for the solidified waste. The waste is concentrated at a constant volume until scale (salt cake) forms on the walls of the pot. As calcination continues, the scale grows in thickness and reduces the capacity for boiling the remaining sludge, and therefore the feed rate must be reduced proportionately. When the feed rate is reduced to an "unprofitable" rate (about 5 liters/hr) the feed is shut off. At this point, the scale has grown inward from the pot wall and upward from the bottom to fill the pot, except for a thin-cone-shaped liquid-containing void in the upper 2 to 3 feet of the salt cake. Heating is continued until the remaining liquid is boiled to dryness and all of the waste in the pot has been calcined and has reached the temperature of 850 to 900 °C. The pot is then cooled in the furnace, removed, sealed, and taken to storage.

SPRAY SOLIDIFICATION

Spray solidification, which was developed by Battelle-Northwest,^[8] is a continuous process that is approaching readiness for commercial use. It is currently being demonstrated by Battelle-Northwest on a pilot plant scale with full level radioactivity in the WSEP.^[16] It is a continuous process with low hold-up volumes, it is adaptable to a moderately wide variety of feed compositions, and it produces a variety of good quality solids.

In the spray calciner, shown in Figure 2b, liquid waste (which contains some or all of the melt-making additives) is fed through a pneumatic atomizing nozzle into the top of a heated cylindrical tower. The atomized waste is sequentially evaporated, dried, and calcined to a powder as it falls into a continuous melter below the calciner where it is melted at temperatures of 800 to 1200 °C. Process gases from calcination flow into the adjacent filter chamber, carrying much of the calciner powder. The powder collects on the porous metal filters as the gas passes through. The powder is periodically blown off the filters and into the melter by sudden pulses of high-pressure steam or air directed backward through the filters by small nozzles. The molten calcine flows through an overflow weir or a freeze valve into the receiver-storage pot below. After the pot is filled, it is cooled in the furnace, sealed, and taken to storage. The pot for receiving the molten waste may be made of mild steel if the pot is kept below about 700 °C.

The flowsheets most used for spray solidification at Battelle-Northwest produce ceramic, orthophosphate solids of alkali metals, and iron. Calcium is added to retain sulfates when present.^[15] Alternative flowsheets which produce borosilicate, semi-glassy solids have been demonstrated recently. Some of these flowsheets have been demonstrated with a scheme which eliminates the melter and performs final melting in a stainless steel container.^[17]

PHOSPHATE GLASS SOLIDIFICATION

Phosphate glass solidification is a continuous process which has been extensively developed at Brookhaven National Laboratory and is approaching readiness for commercial use.^[18] It has been demonstrated on a pilot plant scale with full-level radioactivity in the WSEP.^[19] It is a continuous process that produces a good quality glass product.

In phosphate glass solidification, shown in Figure 2c, liquid waste which contains all of the melt-making additives is first fed to the evaporator where it is concentrated and denitrated by factors of 2 to 10 to a thick, syrupy, aqueous phosphate slurry. The slurry is then fed to the continuous melter where final volatilization of the water, nitrates, and other volatile constituents is accomplished, and the material is

heated to 1000 to 1200 °C to form a molten glass. The molten glass flows through an overflow weir or freeze valve into the receiver-storage pot below. After the pot is filled, it is cooled in the furnace, sealed, and taken to storage.

The pot for receiving the molten glass is similar to that for the spray solidification process.

The chemical adjustments required for the phosphate glass process consist mainly of adding phosphoric acid to the feed to obtain a metaphosphate melt, with the oxides of the alkali metals at least half of those of the total metal oxides in the melt.

FLUIDIZED BED CALCINATION

The fluidized bed solidification process is a continuous process that has been extensively developed for use with aluminum nitrate and zirconium-fluoride-aluminum nitrate wastes. Development of fluidized bed solidification was initiated at Argonne National Laboratory,^[20] and has been extensively demonstrated by the Idaho Nuclear Corporation.^[21] The process has been fully demonstrated with moderate radioactivity levels in production-scale equipment since 1963 and is ready for commercial application for these wastes.^[21] Development with the more unfriendly Purex wastes has been limited. The process provides a relatively high capacity for a given equipment size.

In fluidized bed solidification, shown in Figure 2d, the liquid waste is injected through pneumatic atomizing nozzles into the side of a heated (400 to 600 °C) bed of granular solids which is continuously fluidized by gas sparging upward through the fluidized bed reactor. Contact of the waste with the hot, granular bed results in evaporation and calcination of the feed as coatings of the bed particles. The calcine that is entrained with the calciner off-gases is removed from the gas stream by cyclone separators or filters, then returned to the mainstream of particles. The mainstream of particles is continuously removed from the reactor and transported to storage tanks.

CHARACTERISTICS OF SOLIDIFIED WASTE

The main incentive for converting high-level radioactive liquid wastes to solids is for improved isolation and safety during the millenia of time that the material must be contained. The most important concern is during the first 10 years after solidification when the heat removal problems, temperature, radioactivity, and rate of temperature change are the greatest, and during transportation to the long-term disposal site.

To provide this increased safety, the desired characteristics of solidified wastes with primary importance are:

- Good thermal conductivity,
- Low leachability,
- Good chemical stability and radiation resistance,
- Mechanical ruggedness,
- Noncorrosiveness to container,
- Minimum volume,
- Minimum cost.

The net effect of a high thermal conductivity is to increase the amount of allowable heat stored in a pot of a given size. This characteristic permits reduction of the amount of time that liquid waste must be stored before solidification and permits decreases in the volume of solidified wastes. Low leachability is desired to minimize the amount of radioactivity in water that would reach animal life, in the event that such water contacts a breached container of solidified wastes. Solidified wastes provide significantly improved leachability compared to liquid wastes. The reasons for the desirability of the other listed characteristics are obvious.

Table 1 shows the typical characteristics of solidified high level wastes from Purex wastes.

The solidified waste from pot calcination is a mixture of oxide scale of the metallic constituents in the original liquid waste. The solid is a porous, friable calcine with a low thermal conductivity and a relatively high solubility in aqueous solutions.

The spray solidified waste is a monolithic solid formed after cooling the melt. The solid is a tough, microcrystalline, rock-like material with good thermal conductivity and a moderately low solubility.

The phosphate glass solid is a monolithic, moderately brittle glass formed after cooling the melt. The glass has a fairly good thermal conductivity and a low solubility in aqueous solutions.

The fluidized bed calcine is granular with a mean particle diameter of about 500 microns. The granules may be composed of crystals or amorphous solids. The granules are generally spherical in shape and are moderately soft and friable. Their thermal conductivity is relatively low.

TABLE 1. Characteristics of Solidified High-Level Purex Wastes

	<u>Pot Calcine</u>	<u>Spray Melt</u>	<u>Phosphate Glass</u>	<u>Fluid Bed</u>
Form	Calcine cake	Monolithic	Monolithic	Granular
Description	Scale	Microcrystalline	Glass	Amorphous
Mole % Fission Product Oxides	Up to 80	Up to 20	Up to 25	Up to 50
Bulk Density, g/ml	1.1 to 1.5	2.7 to 3.0	2.7 to 3.0	1.0 to 1.7
Thermal Conductivity, Btu/(hr)(ft)(°F)	0.15 to 0.25	0.5 to 1.0	0.4 to 0.8	0.10 to 0.25
Maximum Heat, W/liter solid ^(a)	85	205	190	70
Leachability in Cold Water, g/cm ² -day	1.0 to 1 ⁻¹	10 ⁻³ to 10 ⁻⁶	10 ⁻⁴ to 10 ⁻⁷	1.0 to 10 ⁻¹
Hardness	Soft	Hard	Very Hard	Moderate
Friability	Crumbly	Tough	Brittle	Moderate
Residual Nitrate, wt%	≤0.05	≤0.005	≤0.0005	≤4.0
Volume, liters/1000 MW _{th}	1 to 2.5	1.2 to 3	1.5 to 5	1.5 to 5
Maximum Stable Temperature, °C	~900	Phase Separation at ~900	Devitrifies at ~500	~600
Container Material	SS	MS or SS	MS or SS	MS or SS

(a) Approximate values for storage in air in 8-inch diameter cylindrical pots to maintain pot centerline temperatures less than 900 °C and pot wall temperatures less than 425 °C. Average k values were used.

A photograph of samples of the four types of solidified wastes is presented in Figure 3.

OVERALL STATUS OF SOLIDIFICATION

The pot, spray, and phosphate glass processes have been demonstrated since 1966 for the Atomic Energy Commission at processing rates of 10 to 20 liters/hr of high level liquid waste, or waste from about 1 tonne/day of nuclear fuel. Demonstrations will be completed later in 1970. The fluidized bed process has been demonstrated since 1963 for the Atomic Energy Commission at high rates of 300 liters/hr of intermediate level wastes. Scale-up of all processes for Purex wastes beyond about 1 tonne/day equivalent fuel will require some additional nonradioactive development.

In 1971, the one tonne/day Midwest Fuel Recovery Plant near Morris, Illinois will be started up, and will convert its high level waste to granular solids by fluidized bed calcination. Within the next two years, the Atomic Energy Commission's waste solidification development program of currently-known concepts will be completed and reported. Effects of severe temperature and radiation on the properties of the solidified waste products will have been measured and evaluated. Evaluations will provide reliable bases for decisions on the management of high-level wastes, as well as for the design, operation, and safety of waste solidification plants.

FINAL DISPOSAL IN SALT

An ideal scheme for disposal of highly radioactive wastes is considered to be one which provides for complete isolation and absence of interaction of the waste from man's biosphere for time periods on the order of several hundred thousand years with no operation, maintenance, or surveillance required after disposal, and at an economical cost. [2]

Storage of wastes as liquids in near-surface tanks has been a successful means for containing the radioactive wastes to date. Such storage is considered to be temporary only, because the waste is contained in a mobile form within our biosphere, the tanks must be periodically replaced due to corrosion failure, and continual operation and surveillance are required.

Other disposal alternatives which have been investigated in the Research and Development programs sponsored by the US Atomic Energy Commission are: (1) disposal as solids in surface or near-surface vaults; [22] (2) disposal as liquids or slurries in caverns in deep geologic formations; [23] (3) disposal as solids in caverns in deep geologic formations. [24]

Disposal of wastes as solids in surface or near-surface vaults on commercial reprocessing plant sites appears acceptable for interim storage. However, for the very long term, surface disposal of solids is considered better than disposal in selected deep geologic formations because of the requirement of continued surveillance.

Safety considerations militate against the bulk shipment of high-level waste liquids or slurries. Consequently, storage of these wastes in deep geologic formations would require suitable geohydrologic conditions at the fuel reprocessing site, thus limiting the number and location of suitable reprocessing plant sites. Disposal of high level waste liquids or slurries in deep underground formations has not been evaluated economically or technically for power reactor fuel reprocessing waste. However, the need to expend millions of dollars to determine whether any selected geologic formation is positively suitable for such an application would appear to limit this concept. While this concept cannot, at the present time, be conclusively eliminated either on the basis of safety or economics, it is not expected to be an important concept in the nuclear power economy.

Disposal of high-level waste as solids in selected deep geologic formations appears acceptable from both the safety and economic standpoints. The initial stimulus for this part of the AEC effluent control R&D program commenced in September 1955, when, at the request of the Atomic Energy Commission, a committee of geologists and geophysicists was established by the National Academy of Sciences-National Research Council to study the possibilities of disposing of high-level radioactive waste materials on land, and to indicate what research was needed to determine the feasibility. [25]

The results of this Committee's study and their recommendations can be summarized as follows:

"Disposal in salt is the most promising method for the near future. Research should be pushed immediately on the structural problem of stability vs size of cavities at a given depth; on the thermal problem - getting rid of the heat or keeping it down to acceptable levels---."

In response to the NAS suggestion, a major part of the AEC ground disposal R&D program in recent years has been directed toward determining the suitability of using underground salt formations for the disposal of high-level solidified radioactive waste. The results of these studies, performed largely by the Oak Ridge National Laboratory, support the premise that bedded salt formations are good locations for disposal of these wastes.

Bedded salt formations are unique in that they are practically always dry, they have not been associated with ground water for millions of years, and they should retain their integrity for millions of additional years. Because of the plasticity of salt, fractures seal or close rapidly, thereby forming an impervious container for entrapped materials. Salt beds are generally located in areas of low seismic activity and widely separated from flowing aquifers as evidenced by the very existence of such deposits. Salt has a considerable compressive strength which is similar to that of concrete, and good thermal conductivity and heat capacity. Large spaces may be mined out, and, even at depths of 1000 feet, more than half of the salt area may be removed with only slight deformation of the support pillars.

Salt formations which are desirable for disposal of radioactive wastes should be at least 200 ft thick, and should be within 2000 ft of the surface. The locations of major salt deposits in the United States which meet these criteria are shown in Figure 4 as four cross-hatched areas (two in Michigan, one in New York, and the one in Kansas). Other areas are not necessarily ruled out for future waste repositories; but those shown are more than adequate. For example, the area in central Kansas alone is about 10,000 square miles.

Salt mine space required for all the high level waste from reprocessing of power reactor fuels in the US by the year 2000, aged 10 years to the year 2010 before disposal, is approximately 2500 acres, based on recent nuclear power predictions by the Atomic Energy Commission.^[26] By the year 2070 this need will have increased to 50 to 100 square miles, which is still a small portion of the thousands of square miles available which are suitable for waste disposal.

CONCEPT FOR DISPOSAL IN A SALT MINE

The high-activity, fuel reprocessing wastes which are solidified by one of the several processes discussed previously would be sealed in containers of about 6 to 24 in. in diameter, and up to about 10 ft long.

After possible interim storage at the fuel reprocessing site, the containers would be shipped to the salt mine which had been previously developed in nearly conventional fashion. At the mine, the waste containers would be unloaded in a hot cell located at the top of a special shaft extending to the mine level. After inspection and any necessary decontamination or recanning, the containers would be lowered, one at a time, into a shielded underground transporter positioned under the waste shaft.

Once a waste can is in the underground transporter, it would be moved to the current disposal room and deposited into a hole drilled in the floor. The hole would then be backfilled with 6 to 8 feet of crushed salt.

Figure 5 shows the layout of one sector of a typical mine designed especially for waste disposal. The mine space would be developed so that ventilation air serving concurrent excavation and disposal operations are completely isolated and that personnel never enter a ventilation air stream after it has passed a filled disposal room.

The holes in the floor would be spaced to dissipate the fission product decay heat without exceeding either the allowable waste temperature or the specified salt temperature of 200 °C. After all the holes in a given room have been filled, the room itself would be backfilled with crushed salt obtained from the excavation operations. The design of the entire operation, including the sizes of the disposal rooms and the support pillars, would be such that plastic flow, accelerated by the elevated temperatures, will cause a consolidation and recrystallization of the salt, thus returning the salt bed to very nearly its undisturbed state after a few decades.

PROJECT SALT VAULT

A field-scale test of this disposal concept was undertaken in an inactive salt mine at Lyons, Kansas. [27,28,29,30] The principal objectives were to show the feasibility and safety of both the concept and the waste-handling equipment, and to determine the effects of heat and radiation on in-situ salt which would permit detailed design of an actual disposal facility. In this experiment, which was carried out from November 1965 to January 1968, irradiated fuel assemblies from the Engineering Test Reactor at Idaho Falls were used to provide sources of intense radioactivity, while electrical heaters supplemented the decay heat generated by the fuel. The demonstration duplicated proposed actual disposal operations as nearly as practical.

The objectives of the experiment were met. There were no detrimental effects of heat and radiation on the salt bed, and enough information was obtained to permit design of a disposal facility. More than 4 million curies of radioactivity in 42 irradiated Engineering Test Reactor fuel assemblies were handled in 21 separate containers without the aid of surface hot cells (although hot cells would be required in an actual facility).

As anticipated from prior laboratory experiments, the structural and chemical properties of salt were not significantly altered by the intense radiation fields. Radiolytic production of chlorine gas did not occur at detectable rates. The structural behavior of the salt was affected by the elevated temperatures and thermal stresses surrounding the heat sources, and it was demonstrated that the effects (primarily due to increased creep rates) can be predicted and handled by standard mining techniques.

Small, brine-filled cavities in the salt, which constituted about 1/2 volume % of the salt were found to migrate toward the heat sources and into the holes containing the sources. In an actual disposal facility, this influx of moisture would amount to a maximum of 0.2 to 1 liter per year per hole, and could lead to stress-corrosion-cracking of portions of stainless steel waste containers, and failure of the containers in as short a time as a few months. Even if stainless steel waste containers do fail due to stress corrosion cracking, no problems are anticipated in the disposal operation because the holes and rooms would be backfilled before the containers failed and ventilation of the various rooms would be controlled. If the containers are of mild steel, only generalized rusting is expected, and container integrity should be maintained for a period of years.

CURRENT STATUS OF SALT DISPOSAL

Following the experimental studies, a preliminary study was made of a conceptual salt mine facility which would handle the entire output of high level waste from US power reactor fuels until about the end of the century.^[31] The overall conclusions from this study were that disposal in salt appears to be a practical, relatively low-cost scheme, and adequate bedded salt deposits exist in the United States for application of the disposal scheme. Currently, a more detailed design and cost study is being made at ORNL for a combined demonstration facility to be operable in the mid-seventies to handle both the solidified power reactor fuel wastes, as well as the essentially non-heat-generating wastes which contain small quantities of long-lived alpha emitters.

ESTIMATED COSTS

A number of cost estimate studies have been made by Blomeke and co-workers at ORNL. These studies have investigated the total costs of managing high level wastes, which include interim liquid storage, conversion to solids by pot calcination, interim solids storage at the fuel reprocessing plant, transportation of solids to a salt mine, and handling and burial of the solid waste in a salt mine. A summary of these cost estimates is presented in Table 2. For the cases examined, costs for total waste management are estimated to be in the range of 35 to 50 mills/MW hr_e, or \$9000 to \$12,000 per metric ton of fuel processed. These costs represent approximately 1% of the total cost of nuclear power, and approximately 10% of the total cost of reprocessing spent nuclear fuels. Comparable costs for perpetual storage as liquids vary from 30 to 35 mills/MW hr_e, or near the lower end of the range for storage as solids.

TABLE 2

COST RANGE FOR MANAGEMENT
OF HIGH-LEVEL WASTES FROM LWR FUELS ^(a)

<u>Operation</u>	<u>Cost Range,</u> <u>mills/MW hr.</u>
Interim Liquid Storage from 1 to 5 Years	14 to 22
Pot Calcination in 6 to 12-in. Diameter Pots ^(b)	4 to 18
Interim Solids Storage from 1 to 10 Years	2 to 10
Shipment of Solids 1000 Miles	1 to 3
Burial in Salt after Aging 1 to 10 Years	3 to 11
Totals for Specific Schemes	37 to 45

(a) 33,000 MWd/MT exposure

(b) 1 cubic foot of solids/10,000 MW_{th}

SUMMARY

Solidification represents the only practical and reasonably attainable technique for achieving a substantial increase in the safety associated with the storing and disposal of the high level waste from the nuclear power industry. The use of this technique is expected to soon become a policy of the US Atomic Energy Commission, wherein the solidification of highly radioactive liquid wastes will be required before shipment to a long-term federal disposal site.

Major AEC-sponsored research and development on processes for converting the high-level liquid wastes to encapsulated solids is nearly completed. From these studies, four basic solidification processes which are essentially ready for commercial application have evolved.

After considerable engineering and experimental study, mined caverns in natural salt deposits were concluded to be the most promising locations to provide the isolation required for hundred of thousands of years. Studies are currently being made on the details of a Federal demonstration disposal facility in a salt mine in central USA.

The cost of high-level waste management involving solidification in combinations with interim liquid storage, interim solid storage, and transportation to final disposal at a salt mine has been estimated to be in the range of 37 to 50 mills/MW hr_e, or only slightly higher than the cost range for perpetual liquid storage.

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Figure 1
High Level Wastes in the Nuclear Fuel Cycle

Figure 2a
Pot Calcination Process

Figure 2b
Spray Solidification Process

Figure 2c
Phosphate Glass Solidification Process

Figure 2d
Fluidized Bed Calcination Process

Spray Solidification

Phosphate Glass

Pot Calcination

**Fluidized Bed
Calcination**

Figure 3
Samples of Solidified High Level Waste

ROCK SALT DEPOSITS IN THE UNITED STATES (AFTER PIERCE AND RICH, U.S.G.S. BULL. 1148)
SALT DEPOSITS HAVING THICKNESS OF AT LEAST 200 ft AND LYING WITHIN 2000 ft OF LAND SURFACE.

Figure 4
Rock Salt Deposits in the United States

Figure 5
One Sector of Conceptual Waste Disposal Mine

INTERNATIONAL ATOMIC ENERGY AGENCY
in co-operation with
THE UNITED STATES ATOMIC ENERGY COMMISSION

**SYMPOSIUM ON ENVIRONMENTAL ASPECTS
OF NUCLEAR POWER STATIONS**

New York, 10-14 August 1970

ERRATA TO SM-146/23

STATUS OF SOLIDIFICATION AND DISPOSAL OF
HIGHLY RADIOACTIVE LIQUID WASTES FROM NUCLEAR POWER
IN THE USA

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Based on Work Performed for the
United States Atomic Energy Commission

ABSTRACT

Solidification of highly radioactive liquid wastes from reprocessing of spent nuclear fuels will soon be required in the USA for additional improvement in safety during interim storage, during transportation to final disposal, and during final disposal at a Federal repository. During the past 15 years, major research and development has been carried out to develop four basic processes for converting the high level liquid wastes to encapsulated solids. These processes are: spray solidification, pot calcination, phosphate glass solidification, and fluidized bed calcination. All processes are being demonstrated on an engineering-scale with radioactive wastes. In all four processes, the waste is heated to 400 °C to 1200 °C to drive off essentially all of the volatile constituents, leaving a solid or a melt that will cool to a solid.

Disposal of these radioactive wastes requires isolation from the biological environment for hundreds of thousands of years. The longevity of the hazard implies that the disposal system should be capable of providing this isolation without surveillance. After considerable study, natural salt deposits were concluded to be the most promising disposal locations in the USA for meeting these criteria. A concept for a facility to dispose of containers of solidified highly radioactive waste in a salt mine has been investigated in an experiment in a salt mine. The overall disposal concept, as well as equipment requirements and handling techniques, were demonstrated.

The US-AEC has proposed that all solidified highly radioactive waste from nuclear power fuels be shipped to Federally owned repositories. Studies are currently in progress on the detailed design and cost estimates for such a first demonstration disposal facility to be located in a salt mine in the central area of the USA.

The cost of high level waste management involving solidification in combinations with interim liquid storage, interim solid storage, transportation to final disposal, and disposal in a salt mine has been estimated to be about 37 to 45 mills/MW hr_e, or only slightly higher than the cost range for perpetual liquid storage.

FINAL DISPOSAL IN SALT

An ideal scheme for disposal of highly radioactive wastes is considered to be one which provides for complete isolation and absence of interaction of the waste from man's biosphere for time periods on the order of several hundred thousand years with no operation, maintenance, or surveillance required after disposal, and at an economical cost. [2]

Storage of wastes as liquids in near-surface tanks has been a successful means for containing the radioactive wastes to date. Such storage is considered to be temporary only, because the waste is contained in a mobile form within our biosphere, the tanks must be periodically replaced due to corrosion failure, and continual operation and surveillance are required.

Other disposal alternatives which have been investigated in the Research and Development programs sponsored by the US Atomic Energy Commission are: (1) disposal as solids in surface or near-surface vaults; [22] (2) disposal as liquids or slurries in caverns in deep geologic formations; [23] (3) disposal as solids in caverns in deep geologic formations. [24]

Disposal of wastes as solids in surface or near-surface vaults on commercial reprocessing plant sites appears acceptable for interim storage. However, for the very long term, surface disposal of solids is considered to be less desirable than disposal in selected deep geologic formations because of the requirement of continued surveillance.

Safety considerations militate against the bulk shipment of high-level waste liquids or slurries. Consequently, storage of these wastes in deep geologic formations would require suitable geohydrologic conditions at the fuel reprocessing site, thus limiting the number and location of suitable reprocessing plant sites. Disposal of high level waste liquids or slurries in deep underground formations has not been evaluated economically or technically for power reactor fuel reprocessing waste. However, the need to expend millions of dollars to determine whether any selected geologic formation is positively suitable for such an application would appear to limit this concept. While this concept cannot, at the present time, be conclusively eliminated either on the basis of safety or economics, it is not expected to be an important concept in the nuclear power economy.

Disposal of high-level waste as solids in selected deep geologic formations appears acceptable from both the safety and economic standpoints. The initial stimulus for this part of the AEC effluent control R&D program commenced in September 1955, when, at the request of the Atomic Energy Commission, a committee of geologists and geophysicists was established by the National Academy of Sciences-National Research Council to study the possibilities of disposing of high-level radioactive waste materials on land, and to indicate what research was needed to determine the feasibility. [25]

ESTIMATED COSTS

A number of cost estimate studies have been made by Blomeke and co-workers at ORNL. These studies have investigated the total costs of managing high level wastes, which include interim liquid storage, conversion to solids by pot calcination, interim solids storage at the fuel reprocessing plant, transportation of solids to a salt mine, and handling and burial of the solid waste in a salt mine. A summary of these cost estimates is presented in Table 2. For the cases examined, costs for total waste management are estimated to be in the range of 37 to 45 mills/MW hr_e, or \$9500 to \$12,000 per metric ton of fuel processed. These costs represent approximately 1% of the total cost of nuclear power and approximately 10% of the total cost of reprocessing spent nuclear fuels. Comparable costs for perpetual storage as liquids vary from 30 to 35 mills/MW hr_e, or near the lower end of the range for storage as solids.

SUMMARY

Solidification represents the only practical and reasonably attainable technique for achieving a substantial increase in the safety associated with the storing and disposal of the high level waste from the nuclear power industry. The use of this technique is expected to soon become a policy of the US Atomic Energy Commission, wherein the solidification of highly radioactive liquid wastes will be required before shipment to a long-term federal disposal site.

Major AEC-sponsored research and development on processes for converting the high-level liquid wastes to encapsulated solids is nearly completed. From these studies, four basic solidification processes which are essentially ready for commercial application have evolved.

After considerable engineering and experimental study, mined caverns in natural salt deposits were concluded to be the most promising locations to provide the isolation required for hundreds of thousands of years. Studies are currently being made on the details of a Federal demonstration disposal facility in a salt mine in central USA.

The cost of high-level waste management involving solidification in combinations with interim liquid storage, interim solid storage, and transportation to final disposal at a salt mine has been estimated to be in the range of 37 to 45 mills/MW hr_e, or only slightly higher than the cost range for perpetual liquid storage.