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FUEL REPROCESSING WASTES IN DEEP UNDERGROUND CAVITIES

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PLACEMENT AND SELF-PROCESSING OF NUCLEAR FUEL REPROCESSING WASTES IN DEEP UNDERGROUND CAVITIES*

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ABSTRACT

A method is presented for disposing of nuclear fuel reprocessing wastes by *in situ* incorporation in molten silicate rock. In this method, liquid wastes are injected into a deep underground cavity and allowed to self-boil. The resulting steam is processed at the ground surface and recycled in a closed system. When waste addition is terminated, the chimney is allowed to boil dry, thereby solidifying the waste. The heat generated by the radioactive waste then melts the surrounding rock, which dissolves the waste. Finally, the rock refreezes, trapping the radioactivity in an insoluble rock matrix deep underground. This method has significant environmental and economic advantages over previously proposed methods.

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INTRODUCTION

The increasing use of nuclear power reactors poses serious radioactive waste management problems for the future. Because of their high hazard and long decay times, reactor wastes must be isolated from the biosphere for many centuries. Although the United States Atomic Energy Commission has announced [1] that it will begin to store these wastes in engineered storage facilities above ground, a longer term solution to the disposal problem is needed.

Over the past 20 years, considerable money and effort have been devoted to finding a solution to the problem. Much of this effort has been directed toward a disposal method that would require high level wastes to be solidified and shipped to centralized repositories for permanent emplacement. Salt mines have been the most prominently considered sites for the purpose. Each operation in the waste solidification process entails a certain accident potential; transport of the solidified waste is also hazardous. Despite the extensive precautions planned for avoiding such hazards, the possibility of accident increases in proportion to the amount of handling, movement, and interim storage.

To reduce these hazardous and expensive operations, we propose the use of deep, underground cavities for the *in situ* incorporation of fuel reprocessing waste in molten silicate rock. The concept for an integrated fuel reprocessing and waste disposal facility discussed here meets the objective of safe, timely, and permanent removal of radioactive material from the biosphere at relatively low cost.

THE LLL CONCEPT

The LLL concept [2] for disposal of fuel reprocessing wastes involves the placement and self-processing of the wastes in deep, underground cavities. These cavities either may be excavated by conventional mining or they may be created as rubble-filled chimneys using nuclear or high explosives. The concept is depicted in Fig. 1. The capacities and properties indicated here are given as reasonable examples, not as requirements. To eliminate the need for waste transportation, the disposal cavity would be situated in the immediate vicinity of the nuclear fuel reprocessing plant, where almost all nuclear wastes are isolated. The cavity would be formed in silicate rock having a low permeability.

Liquid wastes from the plant operations would be injected into the cavity with either minimal treatment or none at all. Initially, only fresh, high-level wastes would be injected and allowed to self-boil. The resulting vapor would be conducted to the surface through an outlet hole. As soon as enough radioactive material is accumulated underground to assure continuous boiling, intermediate- and low-level waste would be added. The accumulation of radioactive material in the cavity would require additional cooling water to prevent the rock from melting.

At the surface, steam issuing from the outlet would be either condensed and recirculated to cool the cavity or processed for reuse in the plant. This closed system would ensure no release of contamination to the environment.

When a substantial fraction of the cavity volume is filled or when the cavity is no longer needed, waste and water addition would be terminated and the inlet hole would be permanently sealed. When steam and vapor are no longer released, the outlet hole would also be sealed.

In the cavity, the events depicted in Fig. 2 would then occur. For purposes of this description, a 25-yr waste-addition period is assumed. The indicated time spans are based on the plant operating conditions given in Fig. 1 and on heat-flow calculations that are discussed later.

When waste and water additions stop, the cavity temperature would rapidly rise to the melting point of the rock. The radioactive material would dissolve in the rock melt. As the molten rock mass increases in size, its surface area increases, resulting in a higher rate of conductive heat loss to the surrounding rock. Concurrently, the heat-production rate of the radioactivity diminishes because of decay. When the heat loss rate exceeds that of input, the molten rock will begin to cool and solidify. During this period, no environmental hazard could result, since the molten rock mass would be essentially immobile (movement to any cooler environment would cause rapid solidification). The heat surrounding the radioactivity would prevent the intrusion of groundwater. The ultimate result would be the permanent incorporation of the waste in an insoluble silicate rock matrix deep underground.

HEAT FLOW CALCULATIONS

Consideration of heat generation and transport within the cavity and surrounding rock is essential to a quantitative treatment of the concept. A computer code [2] has been developed to calculate the heat flow and the thermal effects in and around the cavity. The heat source term for these calculations is taken from recent work of Oak Ridge National Laboratory [3] for LWR and LMFBR fuel having a burnup of 3.3×10^4 MWd/ton (megawatt day/metric ton). Figure 3 shows the total thermal power generated in the waste from 1 ton of spent fuel as a function of postirradiation times between 0.5 and 100 yr.

Heat flow calculations were performed for various conditions. The results of a typical calculation are given in Figs. 4 through 7. This calculation assumed the following conditions:

Cavity properties

Cavity radius = 10.8 m
Cavity volume = 5.3×10^3 m³

Rock properties

Initial temperature = 50 deg C
In situ density = 2.7 g/cm³
Melting point = 1050 deg C
Heat of fusion [4] = 80 cal/g
Specific heat [5] = $0.216 + (9.60 \times 10^{-5} T) - (2.91 \times 10^{-3} T^2)$ (T in deg K) cal/(g deg K)
Thermal conductivity [6] = $(6.30 \times 10^{-3}) - (6.7 \times 10^{-6} T) + (3.41 \times 10^{-9} T^2)$ (T in deg C) cal/(cm sec deg K)

Waste disposal conditions

Age of waste	= 0.5 yr postirradiation time
Rate of waste disposal	= 1500 ton/yr
Duration of waste disposal	= 25 yr

In this example, the waste from each ton of fuel is assumed to be contained in 1.5 m³ of solution at 99 deg C. The cavity temperature is maintained at 200 deg C during waste disposal by regulating the rate of cooling water addition. The cavity dimensions listed above are nominal values for a 5-kt nuclear chimney at a depth of 2000 m.

Figure 4 shows the rate of heat production within the cavity peaking at about 67 MW. Figure 5 shows the required rate of cooling water addition reaching a maximum of about 1.8 m³/min at the 25-yr cutoff. Figure 6 shows the radius of the molten rock mass reaching a maximum of 96 m at 90 yr. Figure 7 shows the temperature profile as a function of distance from the original cavity center at various times after the start of waste disposal.

Table 1 shows the effects of additional cooling-water beyond the 25-yr duration of waste disposal. Although the volume of melted rock can be significantly decreased by such additional cooling, the time at which the maximum molten rock is attained is considerably lengthened.

Further calculational results indicate that neither the required cooling-water rate nor the rock-melting rate is significantly affected by varying the cavity size in the 5,000 to 30,000 m³ range.

SITE CONSIDERATIONS

Because transportation of liquid high-level wastes is unacceptable, cavities would have to be located at the sites of fuel-reprocessing plants. The required cavities could be produced prior to plant construction to avoid ground shock problems, if nuclear explosives are to be used. If conventional mining methods are to be used, the cavities could be emplaced at any convenient time. It should be noted, however, that it would be entirely possible to build a reprocessing plant to withstand the shock of a nearby 5-kt explosive. If the plant were so constructed, cavities could be created as needed after plant construction and connected to the plant by short pipelines.

Two phases of the operation must be considered in selecting a site with suitable geological and hydrological conditions. These are the waste addition phase and the rock melting phase. The requirement for the first phase is that the cavity be placed in a rock having a low permeability. A rock layer 100 m or more thick is required to contain the cavity and the associated fractures produced by the explosion. If the cavity is constructed by mining, a smaller layer may be sufficient. This ensures that a negligible amount of water enters the cavity prior to and during the early portion of the first phase and that no radioactivity migrates from the cavity.

A shale at the depth considered here (>500 m) will have a low permeability, and local fractures will either heal by plastic flow or will not extend far into the medium. Transmission of fluids will therefore be very low. The direction of flow will depend upon the pressure gradient. The

pressure in the cavity will be low compared to the formation pressure, so any leakage will tend to be into the cavity. For this reason, there is more concern that water might flow into the cavity than that solutions will migrate away, although a small flow into the cavity would not be harmful.

The requirement for the second phase is a silicate rock of sufficient dimensions to contain the molten rock at its maximum dimension. For the example used in this paper, the maximum radius is 96 m, which would require a material more than 200 m in thickness, and preferably 300 or 400 m. This material should contain a negligible amount of carbonate rock in order to avoid the generation of CO_2 during the melting phase. Once the rock starts to melt, the radioactive materials are dissolved in the melt and are soon surrounded by molten rock. The peripheral melt zone will contain little or no radioactivity because of the high viscosity of silicate rocks near their melting point. Water in the surrounding rock would be converted to steam ahead of the advancing melt and would be driven away, except for a small amount dissolved in the melt.

Another consideration is the expansion of rock as it melts. A rock with low porosity will increase in volume by 10 to 30%, depending on its composition. If the rock is porous, however, the volume increase will be balanced by the pore space. The fluid content of the pores would be driven away as the temperature increased. The permeability of the rock should be large enough to allow the fluid to migrate away without raising the pore pressure enough to cause failure of the rock under the stresses that may be present.

To provide further assurance that the waste is permanently isolated from the biosphere or from any water that might find its way to the surface, the cavity would be constructed at a depth greater than 500 m (and up to 2 or 3 km deep) and below a considerable thickness of impermeable rock.

During the first phase, when the drill holes are transporting water and steam between the cavity and the surface, care must be taken to avoid the introduction of radioactivity into the rock zones containing mobile water. The system would be operated at a pressure less than that in any water-bearing zones, except perhaps those within a few hundred feet of the surface. Any leak in the well casings below this depth would, therefore, be into the system, rather than out of it. To avoid contamination in the shallow zone, a site should be selected with no important aquifers within several hundred feet of the surface, and preferably none at all. Multiple casing might be provided in this shallow zone, with instruments located between the inner and outer casings to detect any leak so that corrective action could be taken. All holes would be cased, and tubing would be used within the casing to transport the waste.

Disruption of the flow of cooling water to the cavity during the first phase must be avoided to prevent loss of the cavity. Flow could be stopped because of a plant breakdown or earthquake damage to the drill holes. Should one of these occur, the small amount of water in the underground system at any one time would soon be evaporated from the cavity. Rock melting would commence and the cavity would be lost for future use. Another cavity would then have to be built unless a spare had been provided. Although this mishap entails an economic loss it would not be a hazard to safety.

CAVITY FORMATION

Whether the disposal cavity should be formed by means of an explosive blast (nuclear or high explosive) or by conventional mining and excavation will be determined by a more extensive comparison of the respective economic, environmental, and technical advantages of each method. The chief advantage of a nuclear chimney compared with an excavated cavity is lower cost. This is especially true at depths of 2000 m or more, if environmental considerations should dictate such depths. Furthermore, there are locations where conventional mining techniques may be prohibitively expensive because of temperature gradients or the likelihood of rock collapse.

An excavated cavity, on the other hand, might have some technical advantages:

- More clearly defined geometry
- Inspection before waste is committed to it
- More uniformly controlled boiling conditions
- Easier instrumentation
- Possible addition of other waste (e.g., contaminated fuel cladding, clothing, and other solid wastes) since the cavity is open
- Possible linings of refractory brick, metal, or other material

However, the advantages of an excavated cavity would be lost if the cavity collapsed during the boiling cycle. Collapse of large cavities may be difficult to prevent and studies are needed to determine whether cavity stability can be maintained. Since rock collapse is a possibility, some studies should be made to determine whether stable boiling in a rock-filled cavity can be achieved.

OPERATIONAL CONSIDERATIONS

A generalized plan for an integrated nuclear fuel-reprocessing and waste-disposal facility is shown in Fig. 8. As soon as the cavity becomes self-boiling, it will accept intermediate- and low-level waste from the plant as well as high-level waste. This waste plus the water from the condenser will be used to supply cooling water to the cavity. The principal function of the condenser will be to circulate enough water into and steam out of the cavity to keep the cavity well below the melting point of the rock during the operation of the facility. Sufficient water for use in the plant will be treated and circulated back through the plant.

ECONOMICS

Using an economic model based on a discounted cash-flow method [3], the cost estimates for high-level waste management by solidification and disposal in salt mines range from 0.03 to 0.05 mill/kWh(e). This does not include the cost of intermediate- and low-level waste disposal, which would amount to an additional 0.005 to 0.01 mill/kWh(e) [7,8]. Thus, the total cost of disposing of fuel-reprocessing liquid waste using state-of-the-art methods ranges from 0.035 to 0.06 mill/kWh(e) (or US \$9000 to US \$15,000 per ton of fuel, assuming a burnup of 33,000 MWd/ton and 32% efficiency).

To estimate the economics of liquid waste disposal using the LLL method, an analysis of costs for a typical reprocessing plant is made. For this analysis, a 1500 ton/yr plant is considered and a plant life of 25 yr is assumed. Only waste management costs are considered. Any economic advantage derived from integrating waste management with the fuel reprocessing operation or any possible economic benefits resulting from energy recovery from the waste are neglected.

For a waste disposal facility using a 5-kt nuclear chimney at a depth of 2000 m, the total capital expenditure has been estimated to be US \$10 million [2]. For a facility using a conventionally mined cavity at a comparable depth, the total capital expenditure, based on experience in mining underground chambers for the nuclear test program, is estimated to be US \$20 million. Based on a discounted cash-flow model and an operating expense of US \$0.5 million/yr, the unit cost ranges from 0.008 mill/kWh(e) for the former facility of 0.015 mill/kWh(e) for the latter facility (or US \$2000 to US \$3600 per ton of fuel, respectively).

The LLL method of waste management, using either a nuclear chimney or a mined cavity, would thus effect substantial saving over present state-of-the-art methods. By the year 2000, when an estimated 15,000 tons of fuel per year will be processed in the United States, use of the LLL method could result in an annual savings of over US \$100 million.

ENVIRONMENTAL CONSIDERATIONS

The major concern with respect to nuclear waste disposal is the avoidance of environmental pollution. Serious consequences could result from the release of only a small fraction of the radioactivity.

In the LLL concept, storage and processing are minimized and the need for transport of the wastes is eliminated, significantly reducing the potential for accidental releases. In addition, this concept offers several other environmental advantages:

- Elimination of radioactive waste from the biosphere at the earliest possible time. No surface storage of wastes required.
- Permanent binding of the radioactive waste in a rock matrix deep underground, giving assurance of its permanent elimination from the environment.
- A safe method of permanently disposing of intermediate- and low-level wastes. (This would be true even for very-low-level wastes that normally are diluted and dispersed to the environment in concentrations presently considered safe.)
- A method of disposal safe from the effects of such disasters as floods, earthquakes, or sabotage.

FURTHER RESEARCH AND DEVELOPMENT

Before this waste disposal concept is put into commercial practice, experimental work is needed to assure the safety of the process and to determine the optimum operating procedures. While it would be informative, useful, and necessary to do both experimental and calculational work

in several areas, we believe that the concept cannot be fully developed without a field test.

A field test would require the actual placement of a sufficient quantity of nuclear wastes into a cavity to undergo a complete melting and freezing cycle. By using fresh, high-level, power reactor wastes in a small cavity, the cycle could be accomplished in a few years. This procedure would minimize the time required for proper evaluation.

Calculations indicate a wide range in the acceptable scale of operation for a field test. As an example of a suitable scale of operation, a quantity of power reactor waste generating 0.4 MW thermal power could be injected into a small cavity during a period of 30 d. If the rock thermal properties listed previously in this text are assumed, a maximum rock melting radius of 5 m would occur at about 1.3 yr. The melting and freezing process would be monitored by measuring thermal gradients; ground water would be monitored for pressure changes and contamination. The frozen melt zone and its surroundings would be explored by drilling.

Although this scale of operation seems appropriate for an underground field test of the concept, it may be desirable because of technical, safety or economic considerations to experiment on a smaller scale. Calculations indicate that much smaller quantities of waste in cavities as small as 1 m in diameter can be used. This makes it possible to simulate an underground test using a few tons of rock in a hot cell before committing a larger quantity of radioactive waste underground.

Studies leading to and including the field test need to be undertaken both to simulate the complex processes that will take place underground and to better understand the individual processes that occur. This program would include:

- (1) Construction and operation of a pilot plant to determine the critical problems to be faced in underground tests. The pilot plant would be designed to simulate an underground test in a hot cell, utilizing small quantities of radioactivity. It would permit one to vary parameters more readily. A rock-filled chimney as well as an excavated cavity could be simulated. The effect of simultaneous boiling and localized rock melting on boiling stability and on vaporization-deposition could be determined. This pilot plant represents a simulation of complex processes. To more clearly understand the individual processes, these additional studies are needed.
- (2) Boiling and entrainment experiments to determine the optimum mode of boiling (e.g., whether to keep a level of boiling liquid in the cavity or to permit the liquid to flash to steam at the rock surfaces). Also, the amount of activity entrained into the vapor exhaust pipe should be known in order to design equipment to process such material and return it to the cavity.
- (3) Volatility and deposition studies to determine which of the fission products or rock components are likely to vaporize and deposit in the steam exhaust pipe and what conditions may be selected to prevent plugging of the inlet and exhaust pipes.

- (4) Solubility and leaching studies to determine the composition of the boiling waste solution in contact with rock. Whether the radioactive fission products remain in solution or precipitate out affects temperature distribution, localized melting, and stability of boiling in the cavity.
- (5) Phase relationships in the rock-water-radionuclide system and other studies related to rock properties and inter-relationships between the rocks and the boiling liquid waste.

SUMMARY

The method for nuclear waste management presented has the following basic features:

1. Nuclear fuel reprocessing and waste management would be integrated at a common facility, thereby eliminating the need for transportation of wastes.
2. Low-level and intermediate-level, as well as high-level wastes would be injected into a deep cavity in silicate rock and allowed to self-boil. The resulting steam would be processed and re-circulated in a closed system.
3. Following a suitable period of waste addition and continuous evaporation, the cavity contents and surrounding rock would be allowed to melt.
4. Finally, the melt would freeze, permanently trapping the radioactive waste in an insoluble rock matrix deep underground.

Further work will be required to develop this method of waste disposal. At this time, all aspects of the concept appear reasonable and, because of its potential economic and environmental advantages, we believe that the concept demands serious consideration.

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TABLE I. EFFECT OF ADDITIONAL COOLING WATER ON MAXIMUM MOLTEN ROCK

Duration of waste disposal, yr	Total cooling time, ^a yr	Maximum molten rock		
		Time, yr	Radius, m	Volume, 10 ⁶ m ³
25	25	90	96	3.7
25	30	103	88	2.9
25	40	125	80	2.1
25	50	138	74	1.7

^aTime during which cavity temperature is maintained at 200°C.

FIGURE CAPTIONS

- Fig. 1. Artist's concept of waste disposal in a nuclear chimney.
- Fig. 2. Artist's concept of the sequence of events that occur in the chimney when liquid waste is added.
- Fig. 3. Total thermal power in the wastes generated by the processing of spent LWR and LMFBR fuel as a function of postirradiation time.
- Fig. 4. Power output of waste in chimney as a function of time.
- Fig. 5. Required cooling-water rate as a function of time.
- Fig. 6. Melt radius as a function of time.
- Fig. 7. Temperature profile as a function of distance from the original cavity center at various times after the start of waste disposal.
- Fig. 8. Generalized flow diagram for an integrated nuclear fuel-reprocessing and waste-disposal facility.

RADIOACTIVE WASTE DISPOSAL

DEPTH - 2000m

CHIMNEY RADIUS - 1m

MAX MELT RADIUS - 10m

14 million gallons

LAWRENCE LIVERMORE NATIONAL LABORATORY
UNIVERSITY OF CALIFORNIA, LIVERMORE

100m

Fig. 1. Artist's concept of waste disposal in a nuclear chimney.

Time Sequence

Fig. 2. Artist's concept of the sequence of events that occur in the chimney when liquid waste is added.

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Fig. 4. Power output of waste in chimney as a function of time.

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