Lawrence Livermore Laboratory

HIGH LEVEL RADIOACTIVE WASTE ISOLATION BY INCORPORATION IN SILICATE ROCK


November, 1976

This paper was prepared for presentation at the 5th IAEA PNE Technical Meeting to be held in Vienna, Austria, November, 1976.

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately-owned rights. The views contained herein are not necessarily those of the U.S. Government.

"Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Energy Research & Development Administration to the exclusion of others that may be suitable."
HIGH LEVEL RADIOACTIVE WASTE ISOLATION BY
INCORPORATION IN SILICATE ROCK*

University of California
Lawrence Livermore Laboratory
Livermore, California

ABSTRACT

A number of technical possibilities for isolating high level radioactive materials have been theoretically investigated at various times and places. Isolating such wastes deep underground to insure long term removal from the biosphere is one such possibility which has been investigated. The present concept involves as a first step creating the necessary void space at considerable depth, say 2-5 km, in a very low permeability silicate medium such as shale. Waste in dry, calcined or vitrified form is then lowered into the void space, and the access hole or shaft sealed. Energy released by the radioactive decay raises the temperature to a point where the surrounding rock begins to melt. The waste is then dissolved in it. The extent of this melt region grows until the heat generated is balanced by conduction away from the molten zone. Resolidification then begins, and ends when the radioactive decay has progressed to the point that the temperature falls below the melting point of the rock-waste solution. Calculations are presented showing the growth and resolidification process. The use of a nuclear explosion presents one alternative way of creating the void space.

INTRODUCTION

A problem still facing development of nuclear power is that of the high level radioactivity generated in the fission process. Because of their extremely high hazard potential and long decay times, these by-products must be isolated from the environment for centuries. Any disposal

*This work was performed under the auspices of the U.S. Energy Research and Development Administration under contract no. W-7405-Eng-48.
or isolation method selected must be demonstrably capable of containment of the radioactivity under all circumstances prior to commitment of large quantities of these radioactive materials.

Over the past several decades a number of alternative methods of long-term storage have been suggested. Currently, geologic isolation is the most generally favored option for all forms of by-product radioactivity. As an example, present United States regulations require high level wastes from fuel reprocessing to be solidified within five years and shipped to a government repository within ten years after their generation. Current plans are to emplace the canisters of high level waste in a mined-out region of a geologic formation. Historically, bedded salt formations have been favored for these geologic repositories, although there is now a large effort to find suitable sites in other media such as shales and granites.

A common element in conventional methods of handling and isolating high level radioactivity is that provision must be made to dissipate the heat generated in radioactive decay. For conventional geologic disposal, this means that the canisters must be emplaced with sufficient spacing to prevent excessive heat build-up which could compromise the local integrity of the formation, or in the case of salt, cause melting and canister migration.

The concept presented in this paper is unique in that the decay heat is utilized to self-process the radioactivity and immobilize it in situ in a deep geologic formation. Basically, the concept involves creating a cavity volume in a suitable silicate formation and subsequently emplacing the radioactive material. The decay heat melts the surrounding rock which eventually resolidifies, encapsulating the radioactivity in the silicate matrix. This concept was first introduced several years ago. In that proposal, all radioactive wastes (including low level) over a period of about 25 years would be directly introduced into the void space. During this time, cooling water would be added to maintain the region at low temperature to prevent melting and loss of access. When the void space
was full (or earlier if desired), cooling would be stopped and melting allowed to commence. The concept is consistent with the management of almost any form and age of high level waste.

Void volume requirements for receiving the high-level wastes are modest. For shallower depths, conventional mining is certainly a viable way to construct the cavity. As the depth increases, the creation of void volume by use of a nuclear explosive may become a more economically attractive alternative. The costs of the nuclear explosive operations and additional surface facilities, if any are required, would have to be compared to those associated with a conventional alternative such as simply drilling out the void space as needed.

**A PNC DISPOSAL SCHEME**

The concept is depicted in Figure 1. A low yield nuclear chimney is formed in a deep silicate rock formation having low permeability. After sufficient time for decay of gaseous radioactivities, the chimney would be reentered and casing emplaced for access to the chimney for waste injection. Provision for interim steam circulation to the surface for cooling of the waste in the chimney must be provided during the waste injection phase.

Surface facilities would be constructed to provide hot cell handling capabilities for receiving, preparation, and slurry injection of the waste into the chimney. A heat exchanger would be constructed to condense the steam from the chimney and reinject it with additional cooling water into the chimney.

The waste, which we have assumed here to be in a solid form required for its transportation to the disposal site, would be removed from its container and ground or chopped to a suitably small size for slurry-injection into the chimney using sufficient water to prevent plugging of the injection line. During the injection phase sufficient cooling water would be added to maintain the chimney at a low temperature. Waste addition would continue
until the void capacity of the chimney was filled with solid waste, at which time waste and cooling water addition would be terminated, and the injection hole permanently sealed. When steam and vapor are no longer released from the chimney, the steam line would be sealed.

An artist's concept of the sequence of events is shown in Figure 2. After cooling stops, the chimney temperature would rapidly rise to the melting point of the rock and melting would begin. The chimney rubble would melt first, followed by the surrounding rock. The radioactive material would dissolve in the molten rock, and convective processes would mix newly melted rock with that previously melted. As the molten rock mass increases in size, its surface area increases, resulting in proportionately greater conductive heat loss to the surrounding rock. At the same time, radioactive decay diminishes the rate of heat production. When the rate of heat loss exceeds the rate of heat generation, the molten rock will begin to cool and solidify, proceeding from the outer bounds of the melt region toward the core. During this period, no significant migration of the molten mass can take place since any movement into a cooler zone would result in rapid cooling and solidification. The final result would be permanent incorporation of the radioactivity into an insoluble silicate matrix deep underground.

THERMAL CONSIDERATIONS

Consideration of heat production and transport within the chimney and surrounding rock is an essential aspect of this concept. Figure 3 is a log-log plot of the decrease in the thermal power level of radioactive waste with time which is valid over the time interval of interest. A computer code has been used to calculate the heat balance within the chimney-molten rock system and the thermal transport to the surrounding rock for a variety of waste disposal schemes. Typical rock properties assumed for these calculations are shown in Table 1.
Chimney Properties -- Volumes of high level waste are not large, and a single low yield nuclear chimney provides sufficient void volume to dispose of high-level wastes from a sizable nuclear economy. A 5-kt chimney at a depth of burial of 2000 m is used for this example. Chimney properties shown in Table 2 have been calculated using the method of Butkovitch and Lewis. For these rock properties, cavity volume for a given yield scales roughly inversely with depth. Therefore, if a greater depth for disposal were desired, a greater yield proportional to the depth would produce about the same void volume.*

Disposal Scheme -- We have assumed for this presentation a waste management system with interim storage of the high-level waste. The waste would be in solid form, either as calcine or one of several vitrified forms, and contained in steel canisters suitable for shipment. The waste would be accumulated at an interim storage facility until the PNE disposal site was ready to accept it. Specifically, we have assumed ten years accumulation of solidified high-level waste from two 1500 metric ton/yr reprocessing plants. [For perspective, this is roughly estimated to be equivalent to the waste from 10 years operation of one hundred 1000-MW(e) power plants or one million megawatt (electric) years of nuclear power generation.] This waste is further assumed to have been stored for ten years prior to disposal in the nuclear chimney over a one-year period. The dry, solid waste would fill approximately one-third of the total void of the 5-kt chimney at 2000 m depth.

Calculational Results -- Heat flow calculations were performed for continuous addition of the waste over a one-year period starting with the 10 year old and ending with the 20 year old waste. Thus, the last waste injected would be 21 years old. It was assumed that the slurry addition

*It is unknown whether or not cavity collapse to form a chimney will occur at very great depths. Non-occurrence of collapse, however, should not affect the viability of this concept, but a standing cavity that later collapsed during waste injection might result in loss of communication with the cavity and preclude further injection of waste.
Table 1

Rock Properties Assumed:

Initial Temperature = 50°C

In Situ Density = 2.7 g/cm³

Melting Point = 1050°C

Heat of Fusion = 80 cal/g  (Ref. 5)

Specific Heat = 0.216 + (9.60 x 10⁻⁵T) - (2.91 x 10³T⁻²) cal/g - deg
(T in °K)  (Ref. 6)

Thermal Conductivity = (6.30 x 10⁻³) - (6.7 x 10⁻⁶T) + (3.41 x 10⁻⁹T²)
cal/cm - sec - deg (T in °C)  (Inferred from Ref. 7)

Table 2

<table>
<thead>
<tr>
<th>CHIMNEY DATA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Yield</td>
</tr>
<tr>
<td>Depth of Burial</td>
</tr>
<tr>
<td>Rock Density</td>
</tr>
<tr>
<td>Water Content</td>
</tr>
<tr>
<td>Cavity Radius</td>
</tr>
<tr>
<td>Cavity Volume</td>
</tr>
<tr>
<td>Bulking Porosity</td>
</tr>
<tr>
<td>Chimney Rubble Mass</td>
</tr>
</tbody>
</table>
would be made up of a three-to-one ratio of water to waste by volume. Cooling water requirements to maintain the chimney at 200°C during the one year injection phase were calculated as that required in addition to the slurry water. The properties of the system were calculated at 0.01 year increments using one meter spacial zoning beyond the chimney.

The calculated power output of the waste in the chimney in megawatts is shown in Figure 4. The left plot shows in detail the rise in power output during the injection phase. Figure 5 shows the required rate of cooling water addition, which reaches a maximum of 0.6 m³/min to dissipate the 23 MW power output at the one-year cutoff. Figure 6 shows the radius of molten rock growing to a maximum of 80 meters at 70 years with a very slow rate of resolidification thereafter.

SITE CONSIDERATIONS

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.

In the first phase, the chimney is connected to the surface through drill holes; waste and water are introduced into the chimney; and steam is transported to the surface. In the second phase, after the volatiles are driven out of the chimney, the rock melts and the melt volume increases to some maximum size and then decreases as the melt freezes. In the example used here, the first phase lasts for only one year and the second phase lasts for a few hundred years.

The chimney itself should be located in rock having a low permeability. A rock layer 100 m or more thick is required to contain the chimney and the associated fractures produced by the explosion. This ensures that a negligible amount of water will enter the chimney prior to and during the early portion of the first phase and that, later in the first phase, no radioactivity will migrate away from the chimney.
A shale at the depth considered here will have low permeability, and local fractures will either heal by plastic flow or will not extend far into the medium. The pressure in the chimney will be low compared to the formation pressure; therefore any leakage will tend to be into the chimney. For this reason, there is more concern that some water might flow into the chimney than that any dissolved radioactivity will migrate away. A small flow into the chimney would not be harmful.

The requirement for the second phase is a silicate rock of sufficient thickness to contain the molten rock at its maximum dimension. For the example used in this paper, the maximum radius is 80 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 carbonat rock to avoid the generation of CO₂ during the melting phase.

An additional consideration is the expansion of rock as it melts. A low-porosity rock will increase in volume by 10 to 30%, depending on its composition. In a porous rock, however, the expansion can be compensated for by the pore space. Fluid in the pores would be driven away by the temperature increase prior to melting. The permeability of the rock should be large enough to allow the fluid to migrate away without increasing the pore pressure enough to cause failure of the rock.

To provide further assurance that the waste is permanently isolated from the biosphere or from any mobile water that might find its way to the surface, the chimney would be constructed at a depth of 2 km or greater below a considerable thickness of impermeable rock.

During the first phase when the drill holes are transporting water and steam between the chimney and the surface, care must be taken to avoid the introduction of radioactivity into rock zones containing mobile water. The system (chimney and holes) 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 leakage in the well casings below this depth or in the chimney would therefore be into the system rather than out of it.
To avoid contamination in a 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 would 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 chimney during the first phase must be avoided to prevent loss of the chimney. Flow could be stopped because of a facility breakdown or earthquake damage to the drill holes. Should one of these events occur, the small amount of water or steam in the underground system at any one time would soon be evaporated from the chimney and condensed in the hole or in surrounding rocks. The chimney would then melt and would be lost for future use. Such a mishap would result in an economic loss, but would not create a safety or environmental hazard.

ENVIRONMENTAL CONSIDERATIONS

Seismic -- The explosive yield required to make a chimney of sufficient size is sufficiently small that damage to structures would be limited to a relatively small area. A site 10 to 20 km from any small towns or population centers would minimize the number of minor damage claims.

Explosive Radioactivity -- The quantity of radioactivity from the detonation would be trivial in comparison to that of the waste to be injected into the chimney. Care would have to be taken to avoid gaseous radioactivity release upon reentry into the chimney, however.

Steam Transport of Radioactivity -- If the waste is glass encapsulated, it would be of such low solubility that significant water dissolution of the radioactivity followed by steam transport during the injection phase seems unlikely. Calcined waste, on the other hand, is quite soluble, and a few radionuclides could be in a chemical form that is slightly volatile and
small quantities may be transported to the surface with the steam. A closed-loop steam-condenser-reinjection system with adequate monitoring will assure containment of these materials.

**Radioactivity Migration During Melting** -- Once the rock starts to melt the radioactive materials would be dissolved in the melt and 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 points. 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 that would be dissolved in the melt. Thus, it is unlikely that radioactivity could migrate ahead of the melt front.

**Long-Term Migration of Radioactivity** -- The disposal site would be carefully selected to assure its isolation from mobile water. In the unlikely event of future natural phenomena or human activity that would compromise this isolation, there are additional barriers to migration of the radioactive material into the biosphere.

The resolidified rock matrix serves as a permanent, very low solubility barrier to leaching by any water that may migrate into the waste region after solidification. The large volume of rock melted serves to dilute the radioactivity (by up to a factor of a thousand in this case) and serves to further reduce the absolute leachability of the radioactivity dispersed in the silicate matrix.

The movement of any radioactivity that is dissolved will be significantly retarded by ion sorption in the transmitting medium. The magnitude of sorption depends on the species, its chemical state, and the chemical properties of the water and rock medium. With reasonable site isolation and assuming nominal flow rate and sorption properties, radioactive decay would render the waste harmless before it could intersect water supplies utilized by man.
Significant water intrusion cannot take place, however, so long as the resolidified melt and surrounding rock remain at temperatures sufficiently high to vaporize and disperse any in-flowing water before it encounters the waste laden region. Figure 7 shows temperature profiles outside the melted region at times of 20, 100, and 200 years. It is obvious that a thermal barrier to water intrusion will persist for many hundreds of years.

In summary, this concept presents four major barriers to transport of the radioactivity to the biosphere: 1) a deep site carefully selected to be isolated from mobile water; 2) encapsulation of the radioactivity in an extremely low-solubility silicate matrix; 3) a thermal barrier to water intrusion persisting for many hundreds of years; and 4) ion sorption and flow retardation of dissolved radionuclides in the surrounding geologic media.

TECHNICAL ISSUES TO BE ADDRESSED

Both experimental and engineering design work will be required to develop this concept to utilization. Although we believe a field test will be required for full development, a number of technical issues can be addressed in laboratory experiments or calculational studies. Among these are:

- Boiling and entrainment experiments to determine the optimum mode of boiling; e.g., whether to keep boiling liquid in the chimney or to keep the chimney dry with flash evaporation.
- Measurement of the solubilities of the various solid waste forms under waste injection conditions.
- Modeling studies of the dynamic behavior of the molten rock-waste system.
• Measurement of the steam volatility of soluble radionuclides and other chemical species so that estimates of their transport and deposition in the steam processing system can be made and equipment designed to handle these materials.

• Geochemical studies of rock-waste interactions both in the melting phase and after resolidification, particularly with respect to the question of possible segregation of waste species during the melting phase.

A small-scale field test could be executed using a small conventionally constructed cavity. The time-scale to go through complete melting and the beginning of the solidification process could be compressed to a few years using a small amount of fresh high-level waste.

In summary, we have presented a scheme for disposal of high level radioactivity. Preliminary calculations and engineering evaluation show the method to warrant further study; however, very little experimental work has as yet been done toward developing the concept beyond the calculational stage.
REFERENCES


FIG. 1 - ARTIST’S CONCEPT OF WASTE DISPOSAL IN A NUCLEAR CHIMNEY.
Fig. 2 - Time Sequence of Events.
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 THE CHIMNEY
FIG. 5 - COOLING WATER REQUIREMENTS
FIG. 6 - MELT RADIUS VS TIME

MELT RADIUS (m)

TIME (YEARS)

Fig. 6 - Melt Radius vs Time
Fig. 7 - Temperature Profiles at Various Times