DISPOSAL OF PARTITIONING-TRANSMUTATION WASTES WITH SEPARATE MANAGEMENT OF HIGH-HEAT RADIONUCLIDES

Charles W. Forsberg
Oak Ridge National Laboratory*
P.O. Box 2008
Oak Ridge, Tennessee 37831-6180
Tel: (865) 574-6783; Fax: (865) 574-9512
Email: forsbergcw@ornl.gov

Manuscript Date: October 25, 2000
File Name: ATW.Madrid00.article

Article For
6th Information Exchange Mtg. on Actinide and Fission Product Partitioning and Transmutation
Session: Nuclear Waste Transmutation
Organization for Economic Cooperation and Development Nuclear Energy Agency
Madrid, Spain
December 11–13, 2000

---

*Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U.S. Dept. of Energy under contract DE-AC05-00OR22725.

DISPOSAL OF PARTITIONING-TRANSMUTATION WASTES WITH SEPARATE MANAGEMENT OF HIGH-HEAT RADIONUCLIDES

Charles W. Forsberg
Oak Ridge National Laboratory
P.O. Box 2008; Oak Ridge, Tennessee 37831-6180
Tel: 865-574-6783, Email: forsbergcw@ornl.gov
United States of America

Abstract

An alternative approach is proposed for disposing of partitioning-transmutation (P-T) wastes to (1) reduce repository costs and (2) improve repository performance. Radioactive decay heat controls the size and cost of the repository. It is proposed that P-T wastes be separated into a high-heat radionuclide (HHR) fraction and a very-low-heat-radionuclide (VLHR) fraction to bypass this repository design constraint. There are five repository HHRs in spent nuclear fuel: cesium, strontium, plutonium, americium, and curium. P-T, by destroying the long-lived HHRs (plutonium, americium, and curium), is an enabling technology for separate low-cost disposal of the remaining HHRs (^{137}Cs and ^{90}Sr), which have limited half-lives ($T_{1/2}$ = ~30 year), small volumes, and high heat-generation rates. These characteristics allow the use of lower-cost disposal methods for these HHR wastes. Eight HHR disposal options are identified and described. With the removal of the HHRs, there are lower-cost, higher performance methods for disposal of the remaining VLHRs.

Introduction

Repository design and performance are primarily controlled by radioactive decay heat. Consider the proposed Yucca Mountain (YM) repository [1] in the United States. It is designed for ~70,000 t of spent nuclear fuel (SNF) and high-level waste (HLW). If there were no radioactive decay heat, the entire volume could be placed in a cube, which would be ~30 m on each side. The cost of such a repository would be very low. However, radioactive waste generates heat. To ensure repository performance, the repository temperatures are limited. The temperature is limited by packaging the wastes in ~11,000 waste packages (WPs) and dispersing the WPs over 100 km of tunnels. The repository program will cost several tens of billions of dollars.

From a distance, a schematic of the proposed YM repository (Fig. 1) appears as a large planar structure—like a horizontal underground car radiator. This typical characteristic of geological repositories is a consequence of the need to limit repository temperatures and dissipate decay heat. If waste partitioning and transmutation (P-T) is to have a major impact on the repository cost, it must be by changing how decay heat is managed in a repository.

Radioactive decay heat: sources and impacts

There are several temperature limits [2] on the repository: (1) waste-form limit, (2) package limit, (3) near-field rock limit, and (4) various far-field limits. Each limit is imposed to prevent damage to one or more barriers to radionuclide migration from the waste form to the accessible environment. Almost all repository decay heat from SNF (Fig. 2) is produced from five elements: cesium (^{137}Cs), strontium (^{90}Sr), plutonium (Pu: multiple isotopes), americium (Am: multiple isotopes), and curium (Cm: multiple isotopes). While there are other heat-generating radionuclides, these decay away quickly. These high-heat radionuclides (HHRs) can be divided into two categories: shorter-lived HHRs (^{137}Cs and ^{90}Sr) and long-lived HHRs (Pu, Am, and Cm).

The temperature limits in and near the WP are controlled by decay heat from the shorter-lived HHRs— ^{90}Sr and ^{137}Cs. The long-term temperature limits far from the WP are usually controlled by the longer-lived actinides. It takes a significant amount of decay heat over a long time to heat large quantities of rock to unacceptable temperatures. The removal of either the shorter-lived or longer-lived HHRs radionuclides from the waste provides some benefits to the repository, but the benefits are limited because both sets of radionuclides impose temperature limits on the repository—one set in the near term and the second set in the longer term.

If the HHRs are removed from the waste, alternative repository design options [3] exist that may significantly reduce the size and cost of a repository. A large repository is replaced with a mini-repository, and the size of the mini-repository is controlled by the fraction of the HHRs that are not removed. For this to occur, alternative methods for management of the HHRs are required.

- *Long-lived HHRs*. This conference is examining P-T of actinides, including Pu, Am, and Cm. If the P-T technology is successful and economically viable, this approach can be used to destroy these troublesome HHRs.

ORNL DWG 2000-272

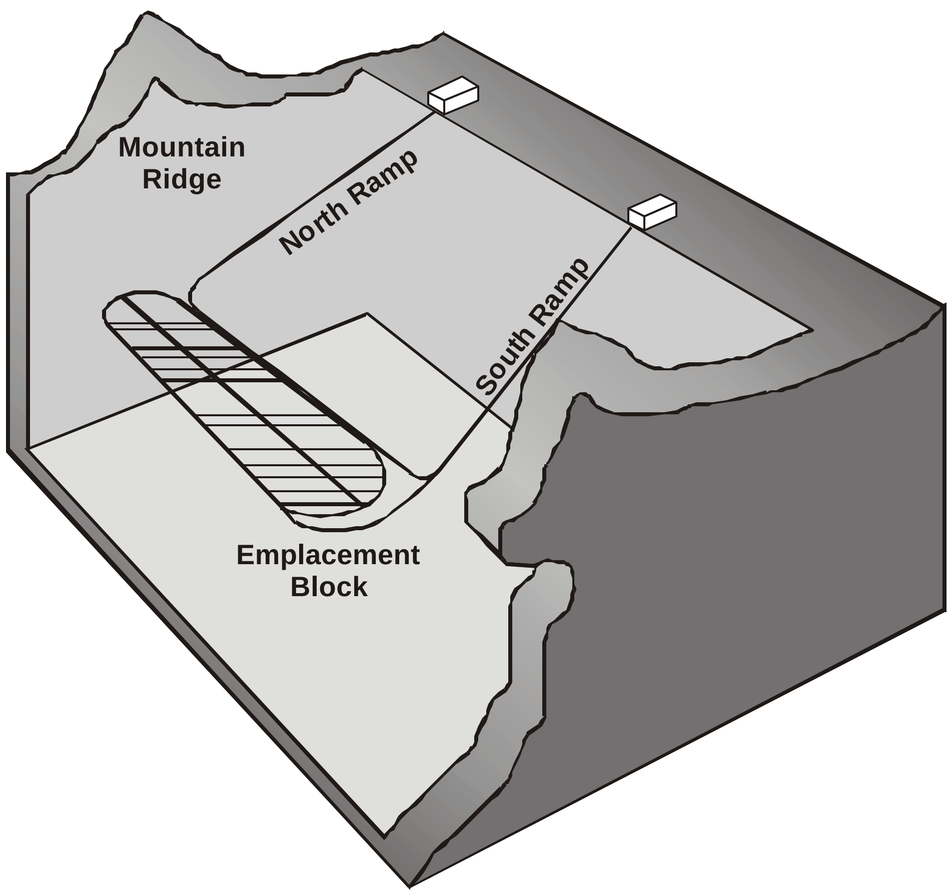

Fig. 1 **Schematic of the proposed YM repository**

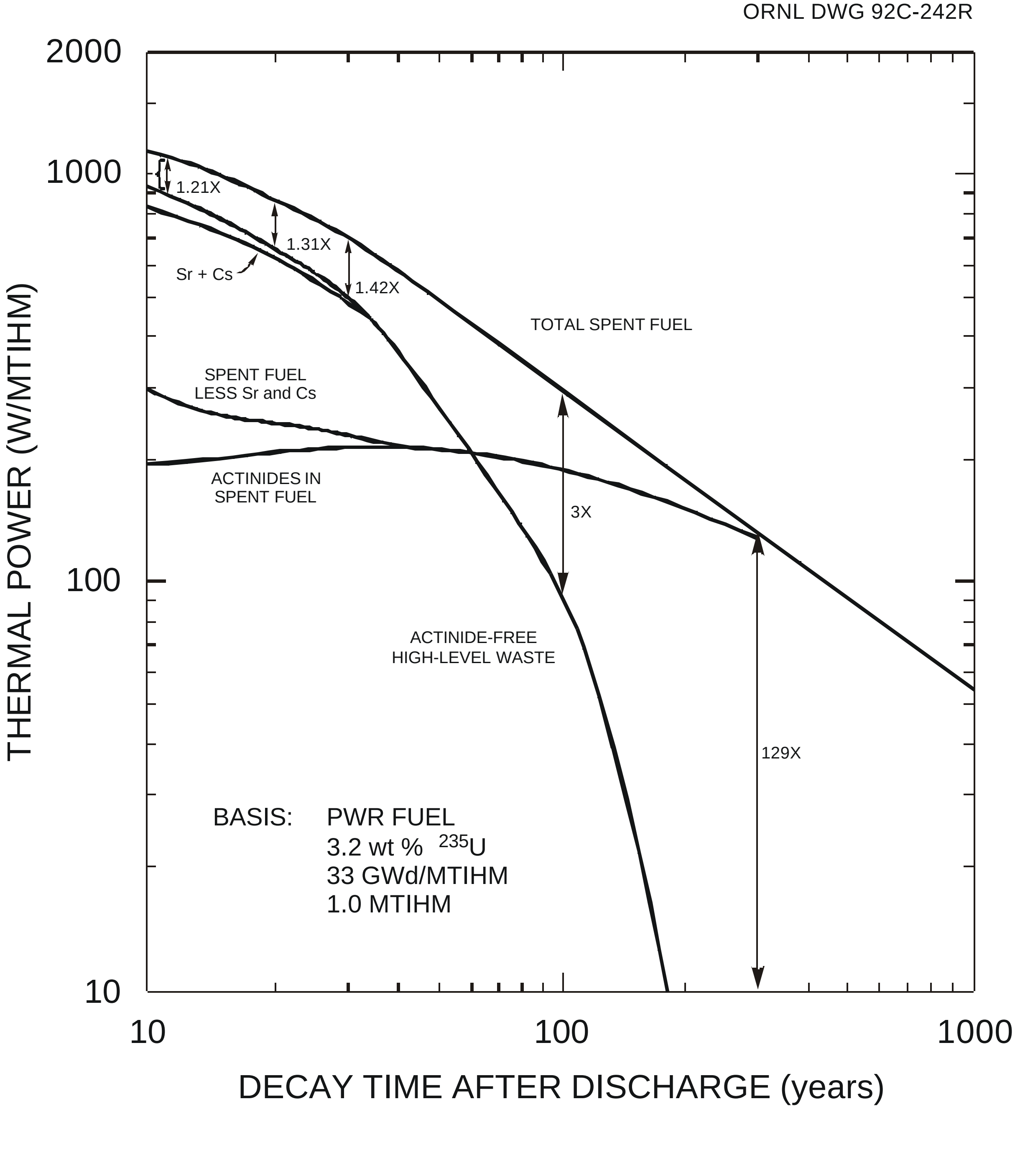

Fig. 2 **Decay heat from SNF**

- *Shorter-lived HHRs*. The ^{90}Sr and ^{137}Cs must be separately managed. Because the characteristics of shorter-lived HHR wastes are different than those of HLW and SNF, low-cost disposal methods may be available. These HHRs differ from SNF and HLW in four ways (characteristics):

  < *Half-life*. The short half-life ($T_{1/2}$. 30 years) allows the use of options that are safe for disposal of these materials but that would be difficult to demonstrate as safe over geological times if disposing of SNF or HLW with their large inventories of long-lived radionuclides.

  < *Waste volume*. The quantities of HHRs (cesium and strontium) are small. One tonne of 40,000-MWd light-water reactor (LWR) SNF contains 4.1 kg of cesium and strontium.

  < *Heat-generation rate*. High heat-generation rates create options that require decay heat to function.

  < *Fissile content.* These wastes include no fissile materials, and thus there are no safeguards or criticality concerns.

There is experience [4] in separating and packaging shorter-lived HHRs from HLW. Over 10^8 Ci of these HHRs were separated from defense HLW at Hanford, Washington, to minimize the cost of storing HLW in tanks. Tank capacity was limited by the decay heat, not the tank volume. The HHRs were packaged in 6.67-cm-diam capsules.

Management of shorter-lived HHRs

There are many methods to manage ^{90}Sr and ^{137}Cs. The method selected by any nation will depend upon institutional factors and the geology available to each nation to manage such wastes. *Near term and more speculative options* are described herein to emphasize that when the characteristics of the waste change, the disposal options change. This is an area of waste management where very few investigations have been conducted; thus, many of these options are not well understood. In parenthesis are the characteristics of the short-lived HHRs that are important for the disposal option.

Long-term storage (half-life)

The HHR wastes can be placed in long-term, dry-storage facilities, which are similar to those used for HLW and SNF. After the decay of most of the HHRs, the wastes can be disposed of in the repository.

Extended dry repository (waste volume, heat-generation rate, half-life)

The HHR capsules could be disposed of in a separate section of a dry repository above the water table [5]. The proposed YM repository in the United States is of this type, and thus this is a potential option for the United States. Long boreholes would be drilled into the rock from a central tunnel and then filled with small-diameter HHR capsules. The heat load would be controlled by placing low-volume HHR capsules in small-diameter horizontal boreholes (<15 cm in diameter) rather than placing large, HLW or SNF WPs in 5.5-m-diam disposal tunnels. The holes could be drilled in a horizontal plane (Fig. 3) or in a vertical array. Boreholes are less expensive than tunnels.

The HHR section of the repository would be designed as an "extended-dry" repository in unsaturated rock. By placing the boreholes closely together to obtain higher local heat loads and higher local temperatures (but sufficiently apart to avoid capsule damage), the local rock temperature would be above the boiling point of water for thousands of years. If the rock temperature is above the boiling point of water, there can be no groundwater flow near the capsules and no migration of radionuclides in groundwater. The shorter-lived HHRs decay before the high-heat section of the repository cools below the boiling point of water. The need for high temperatures requires closer spacing of the HHRs than is used for SNF and HLW; and thus, a correspondingly smaller repository section for these wastes.

The YM repository project investigated SNF extended-dry repository concepts [6] because of economic advantages. Such concepts have not been adopted for SNF or HLW because of the uncertainties in predicting long-term, extended-dry repository behavior after the repository cools. These uncertainties do not exist for HHRs that decay before the high-heat section of the repository cools down.

Conventional repository (half-life, waste volume)

The HHR wastes could be disposed of in a conventional repository. However, the repository size and cost may be significantly reduced. Boreholes, not tunnels, are needed for HHR placement. Elimination of long-term heat-decay loads and most long-term performance requirements (>1000 years) allows the use of simpler WPs and other simplifications.

Saltdiver (heat-generation rate, half-life)

Natural salt domes contain relatively pure salt in the shape of a mushroom with diameters measured in kilometers. The vertical dimension may be as large as 10,000 m. The saltdiver repository [3] uses the high-heat generation rates of HHR capsules to allow disposal at depths up to 10,000 m underground in salt domes. The HHRs are packaged into moderately large containers (saltdivers) that are placed in a salt dome. The high-density, high-temperature WPs sink by heating the salt under the WP until the salt becomes plastic or melts (at 800EC).

ORNL DWG 99C-391R

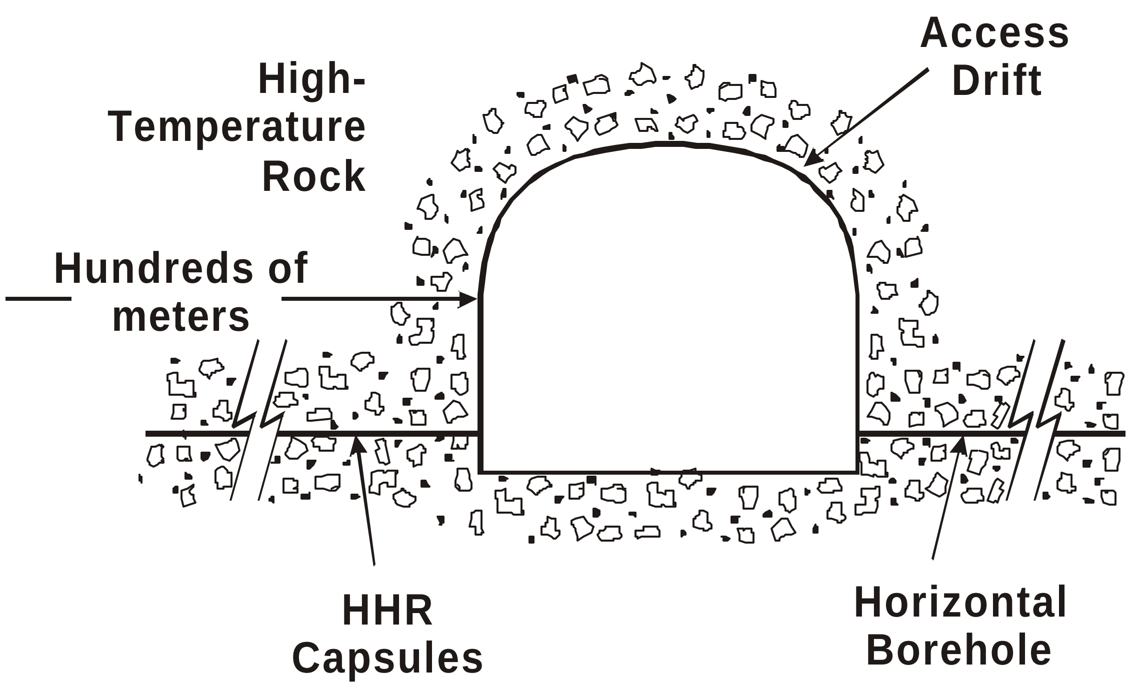

Fig. 3 **Shorter-lived HHR repository with boreholes (rather than tunnels) used to distribute decay heat load radionuclide**

Rock melt (heat-generation rate, half-life)

In the melt-rock repository [7], a large, spherical, underground cavity would be constructed several hundred to several thousand meters underground. Large quantities of HHRs would be placed in the cavity. During loading operations, active cooling systems control temperatures. After the cavity is loaded, the cavity would be sealed, and the cooling systems would be shut off. The HHRs would melt and then melt the surrounding rock. The radionuclides would then be incorporated into the molten rock. It is large-scale vitrification of waste. Ultimately, as the decay-heat levels decrease, the molten rock would solidify into solid rock.

During periods of high-temperature operations, the high temperatures cause plastic deformation of the rock beyond the melt zone, which seals all cracks. Several uncertainties [8] have been identified with this disposal option. However, the identified uncertainties apply only to HLW and SNF that contain long-lived radionuclides, not disposal of shorter-lived HHRs. Further analysis would be required to determine if there are unidentified failure modes when disposing of HHRs.

Borehole (waste volume)

The use of deep vertical boreholes (>5 km) has been considered for the disposal of various radioactive wastes. However, a major drawback is that a borehole has very limited volume. Drilling deep, wide boreholes is expensive. For short-lived HHRs, the volumes are very small; thus, this may be a viable low-cost option for these specific wastes.

Seabed (waste volume, half-life)

International programs [9] have investigated seabed disposal of SNF and HLW. Seabed disposal involves placing WPs into the clay layer, which covers most of the ocean's seabed. The clay layer has potentially excellent waste-isolation properties, and the ocean provides an independent backup mechanism (ocean dilution) if there were failures. There are major institutional problems and some technical problems associated with this option.

Demonstration of disposal viability of short-lived HHRs would be simpler than for other wastes because the shorter-lived HHRs remain hazardous for a much shorter period of time. Furthermore, there are no fissile materials associated with HHRs. Recent analysis has raised questions about the viability of disposal of wastes with fissile materials using this technology. New off-shore oil recovery technologies are making it increasingly easier to recover objects from the ocean seabed; thus, there is a concern about the recovery of any fissile materials by unknown parties if the disposal site is the ocean seabed.

Shallow-land disposal, half-life

The limited half-life may allow shallow-land disposal of the shorter-lived HHRs under some circumstances [3].

Management of low-heat, long-lived radionuclides

With most of the HHRs removed, the repository for the remaining wastes becomes a small facility [3,5]. The required repository would contain two sections: a section for wastes with significant decay heat and a section for the very-low-heat radionuclides (VLHR). Existing vitrified HLW and some P-T target wastes (deep-burn, once-through targets; certain target-processing wastes) would be disposed of in a repository section similar to existing repository designs. Because of the small quantities of these wastes, this repository section would be relatively small.

The wastes from processing light-water reactor SNF, after removal of the shorter-lived (^{90}Sr and ^{137}Cs) and long-lived (Pu, Am, and Cm) HHRs would be a VLHR waste. These wastes may be disposed of in a few lower-cost, high-performance silos without exceeding temperature limits. Depending upon the geology and efficiency of removal of HHRs, such a silo might accept the wastes from up to 10,000 tonnes of SNF.

There is experience with waste silos [10]. Sweden (Fig. 4) and Finland have constructed and are operating underground silos for the disposal of intermediate-activity wastes. The heat-generating characteristics of these wastes are somewhat similar to VLHR wastes. The Swedish waste silos are about 50 m high and 25 m in diameter. The costs per unit volume are a fraction of the cost of traditional WPs.

VLHR silos would be located in the middle of the repository at full repository depth to take advantage of the waste-isolation capabilities of the repository. The repository provides a major barrier against human intrusion, and the geology provides several barriers against radionuclide releases to the accessible environment. Silos are an alternative WP, not a replacement for the repository.

The replacement of WPs with large silos may result in significant improvements in the performance of the engineered barriers to radionuclide releases. The release of radionuclides from a failed WP is proportional to (1) the groundwater flow through the WP and the (2) solubility limits of the radionuclides in groundwater. By concentrating the VLHR wastes from up to 10,000 t of SNF in 1 silo rather than spreading it over ~1,000 WPs, the groundwater flow through the wastes per unit volume is reduced by a factor of 100 to 1,000. With the reduction of groundwater flow per unit quantity of waste, radionuclide releases are proportionally reduced. The large waste silo has a smaller surface-to-volume ratio than does each WPs.

Other considerations

Scaling factors

No detailed economic analysis of these repository benefits has been conducted. However, some comparisons [3] between conventional repositories and these alternative designs can be made. Consider the case where (1) P-T destroys the long-lived HHRs, (2) the shorter-lived HHRs are disposed of in an extended-dry repository such as YM, and (3) the long-lived, low-heat wastes are disposed of in a set of silos.

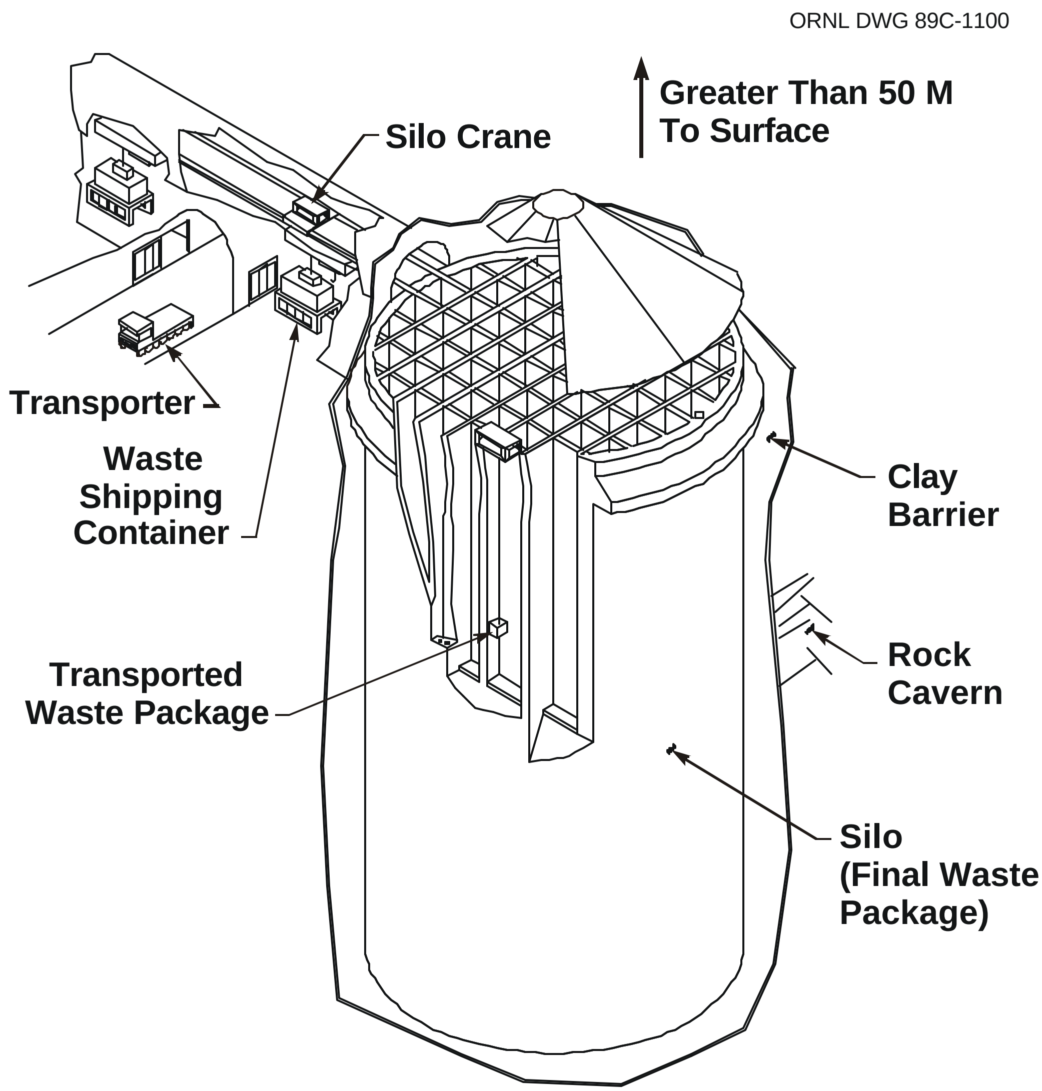

Fig. 4 **Swedish SFR silo for intermediate wastes**

In the conventional YM repository design, the SNF is disposed of in large WPs in 5.5-m-diam tunnels. For every 100 m of tunnel required for disposal of SNF, about 71 m of boreholes would be required to dispose of the shorter-lived HHR wastes from the SNF. For every 100 SNF WPs, 71 HHR capsules of similar length and a small fraction of a silo would be required for disposal of the HHR-VLHR wastes from that SNF. In effect, there are three major changes: (1) substitution of 5.5-m disposal tunnels with 15-cm boreholes for the HHRs, (2) substitution of thousands of WPs with a few silos, and (3) reduced heat load from destruction of the longer-lived HHRs.

The impact of these changes would be to significantly reduce the operational costs for the repository. Operational costs include the mining of disposal drifts for the WPs. It may not impact siting or licensing costs—an important fraction of the total costs. The economic incentives are strongly dependent upon the size of the repository. As the repository capacity increases and the cost per unit of waste decreases, operational costs become a larger fraction of disposal costs. Siting and licencing costs are essentially fixed costs.

The economic cost for the repository gains in an actinide P-T fuel cycle is the necessity to separate the cesium and strontium from the other waste streams. This cost is dependent upon the specific separations processes.

Cesium-135

The short-lived HHRs contain one long-lived radionuclide, ^{135}Cs. It has a half-life of 3×10^6 years. Performance assessments of proposed repositories [11] indicate that this long-lived radionuclide is not usually a significant risk to man nor a significant factor in terms of repository performance. There are several reasons for this:

- *Geochemistry*. Radionuclides, such as ^{129}I, ^{237}Np, and ^{99}Tc, which dominate the long-term risks from a repository are those most easily transported by groundwater with little retention by the geology. There is significant retention of cesium in most types of rock and ion-exchange of radioactive cesium isotopes with non-radioactive cesium in the rock.

- *Biological effects*. Differences in the accumulation rate of different radionuclides in specific human organs determines their relative hazards. The hazard from ^{135}Cs is low compared to many other radionuclides [12] because of its low rate of bioaccumulation.

For any HHR disposal option, a performance assessment of the risks from this radionuclide will be required. There are major engineering questions about the feasibility of isotopically separating this isotope from other cesium isotopes; thus, disposal with the other cesium isotopes is likely to be the most practical route. Such an assessment would be significantly simpler to make than for HLW or SNF because there is only a single radionuclide.

Conclusions

Repository designs and costs are controlled by radioactive decay heat. Any P-T option that destroys long-lived HHRs (Pu, Am, and Cm) is an enabling technology that may allow for lower-cost, higher performance repositories by separate management of (1) the shorter-lived HHRs and (2) the VLHR wastes. These repository benefits may exceed the other waste management benefits of actinide P-T fuel cycles such as reductions in radiotoxicity. The cost for these benefits is the requirement to separate cesium and strontium from the other P-T wastes.

An understanding of the costs and benefits of separate management of shorter-lived HHRs should be a high priority within any investigation of actinide P-T fuel cycles. This is an appropriate area for international cooperation. Most of the issues (selection of radionuclides to be destroyed by P-T, transmutation efficiencies, solidification of short-lived HHRs, repository design for low-heat, long-lived radionuclides, etc.) are common issues for all. It is an area of waste management where only very limited studies have been undertaken.

REFERENCES

[1] U.S. Department of Energy, *Draft Environmental Impact Statement for a Geological Repository for the Disposal of Spent Nuclear Fuel and High-Level Radioactive Waste at Yucca Mountain, Nye County, Nevada*, DOE/EIS-0250D, July 1999, U.S. Department of Energy, Washington, D.C.

[2] A. G. Croff, *A Concept for Increasing the Effective Capacity of a Unit Area of A Geologic Repository,* Radioactive Waste Management and Environmental Protection, 1994, **18**, 155–180.

[3] C. W. Forsberg, *Rethinking High-Level Waste Disposal: Separate Disposal of High-Heat Radionuclides (^{90}Sr and ^{137}Cs)*, Nuclear Technology, August 2000, **131**, 252–268.

[4] R. R. Jackson, *Hanford Waste Encapsulation: Strontium and Cesium*, Nuclear Technology., January 1977, **32**(1), 10.

[5] C. W. Forsberg, *Disposal of Partitioning-Transmutation Wastes in a Yucca-Mountain-Type Repository With Separate Management of High-Heat Radionuclides (^{90}Sr and ^{137}Cs)*, 4th Topical Mtg on Nuclear Applications of Accelerator Technology, November 2000 (American Nuclear Society).

[6] T. A. Buscheck and J. J. Nitao, *Repository-Heat Driven Hydrothermal Flow at Yucca Mountain, Part I: Modeling and Analysis*, Nuclear Technology,. December 1993, **104**(3), 418.

[7] J. J. Cohen, A. E. Lewis, and R. L. Braun, *Insitu Incorporation of Nuclear Waste In Deep Molten Silicate Rock*, Nuclear Technology., April 1972, **13**, 76.

[8] A. S. Kubo, *Technical Assessment of High-Level Nuclear Waste Management*, Ph.D. degree thesis, May 1973, Department of Nuclear Engineering., Massachusetts Institute of Technology, Cambridge.

[9] Nuclear Energy Agency, *Feasibility of Disposal of High-Level Radioactive Wastes Into The Seabed*., 1988, Organization for Economic Cooperation and Development, Paris

[10] J. Carlsson, *Nuclear Waste Management in Sweden,* Radwaste Magazine, 1998, **5**(6), 25.

[11] U.S. Department of Energy, *Total System Performance Assessment–Viability Assessment (TSPA-VA) Analysis Technical Basis Document*, November 13, 1998, Las Vegas, Nevada.

[12] U.S. Nuclear Regulatory Commission, *10 Code of Federal Register, Part 20,* Washington, D.C.