

《Review》

A Review of Nuclear Waste Management:

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방사선 폐기물 처리 처분에 대한 최근 동향

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Abstract

A brief review is presented for the state-of-art waste treatment technologies, siting issues, and regulatory requirements in geologic disposal of nuclear wastes. Key technical issues that need to be solved by waste management research program are also presented.

Introduction

The potential risks associated with management and disposal of radioactive wastes are *estimated* to be much smaller than those due to other routine activities involving risk.^(1,2) In spite of this estimation, the issue of management and ultimate disposal of radioactive wastes continues to be one of the major concerns regarding nuclear related activities. This concern suggests that the emphasis given in public debate of radioactive waste disposal is more to the level of uncertainty than to the level of risk. Due to the complexity of phenomena and processes involved in radionuclide migration, the degree of spacial and temporal extrapolation needed in the risk assessment, and lack of understanding overall system interaction, it is

generally believed that estimation of risks associated with radioactive waste disposal is accompanied by relatively large uncertainties. In order to reduce uncertainties and improve public confidence, more focused research is needed to improve our understanding of relevant phenomena and processes regarding waste isolation and migration through various engineered barriers and geologic systems.

This paper addresses various types of radioactive wastes with respect to origin, nuclear and chemical characteristics, waste treatment technology and disposal alternatives. The pertinent regulatory requirements for disposal of radioactive wastes are presented followed by a discussion of technical issues that need to be resolved to demonstrate implementation of the regulations. Although there is no complete identification and quantification of all uncertainties associated

with risk assessment, there are some well recognized sources of uncertainties. These sources of uncertainties will be discussed together with current research efforts to reduce them.

Waste Type and Characteristics

Radioactive wastes are traditionally categorized as high-level wastes (HLW), low-level wastes (LLW), or uranium recovery wastes (UR) in the United States. Europeans and Japanese use the additional category of "intermediate-level wastes (IMLW)" to characterize LLW with relatively high radionuclide concentrations. Transuranic wastes (TRU) are those wastes containing more than 10 to 100 nano curies of transuranic elements per gram of wastes. The waste categorized above is primarily based on origin of wastes rather than level

Table 1. Chemical Composition of HLW

| Facility | Waste Type | Composition* |
|----------------------|------------------------------------|---|
| US Reference | PW-4b(clean)** | Re ₂ O ₃ (32%) ZrO ₂ (12%) MoO ₃ (13%) |
| | PW-7a(dirty) @ | Re ₂ O ₃ (31%) U ₃ O ₈ (15%) |
| INEL(USA) | Aluminum Calcine (Non-fluoride) | Al ₂ O ₃ (89%) |
| | Zirconium Calcine (Fluoride) | ZrO (21%) Al ₂ O ₃ (21%) CaF ₂ (54%) |
| SRP(USA) | Sludge | Fe ₂ O ₃ (5~47%) Al ₂ O ₃ (3~36%) MnO ₂ (3~17%) U ₃ O ₈ (0~19%) |
| Marcoule (France) | Gas Grafite | Al(8~12g/l) Fe(6~10g/l) Na(4~6 g/l) U(2.5~3.5g/l) |

Note: * Fission products not included. weight % in ()

** PW stands for purex process waste

@ PW-7a is typical of HLW expected from Allied General Nuclear Service (AGNS), South Carolina

Table 2. Average Activity of HLW
(Savannah River Plant Sludge)

| Time After Irradiation, yrs | Radionuclide Activity, Ci/gal | | |
|--------------------------------|-------------------------------|----------------------|----------------------|
| | 1 | 5 | 10 |
| Ce-144-Pr-144 | 4.5×10^2 | 1.3×10^1 | 1.5×10^{-1} |
| Zr-95 | 9.6×10^1 | 1.8×10^{-5} | a |
| Y-91 | 7.6×10^1 | 2.5×10^{-6} | a |
| Sr-89 | 4.4×10^1 | a | a |
| Nb-98 | 6.0×10^0 | a | a |
| Ce-141 | 3.0×10^0 | a | a |
| Pm-147 | 1.0×10^2 | 3.6×10^1 | 9.7×10^0 |
| Ru-103 | 5.2×10^0 | a | a |
| Ru-106-Rh-106 | 2.4×10^1 | 1.6×10^0 | 5×10^{-2} |
| Sr-90 | 3.0×10^1 | 2.8×10^1 | 2.4×10^1 |
| Cs-137 | 1.6×10^0 | 1.5×10^0 | 1.3×10^0 |
| Te-129 | 9.4×10^{-1} | a | a |
| Te-127 | 6.4×10^0 | 5.9×10^{-4} | a |
| Cs-134 | 8.7×10^0 | 2.3×10^0 | 4.2×10^{-1} |
| Sm-151 | 7.5×10^{-6} | 7.3×10^{-1} | 7.0×10^{-1} |
| Pu-238 | 1.1×10^{-1} | 1.1×10^{-1} | 1.1×10^{-1} |
| Pu-241 | 2.4×10^{-2} | 2.0×10^{-2} | 1.6×10^{-2} |
| Cm-244 | 1.3×10^{-2} | 1.1×10^{-2} | 9.5×10^{-3} |
| Am-241 | 1.1×10^{-2} | 1.1×10^{-2} | 1.1×10^{-2} |
| TC-99 | 4.3×10^{-3} | 4.3×10^{-3} | 4.3×10^{-2} |
| Pu-239 | 3.5×10^{-3} | 3.5×10^{-3} | 3.5×10^{-3} |
| Eu-154 | 1.1×10^{-3} | 8.3×10^{-4} | 5.5×10^{-4} |
| Zr-93 | 8.6×10^{-4} | 8.6×10^{-4} | 8.6×10^{-4} |
| Pu-240 | 6.4×10^{-4} | 6.4×10^{-4} | 6.4×10^{-4} |
| Cs-135 | 2.2×10^{-5} | 2.2×10^{-5} | 2.2×10^{-5} |
| Sn-126-Sb-126 | 1.1×10^{-4} | 1.1×10^{-4} | 1.1×10^{-4} |
| Se-79 | 1.0×10^{-4} | 1.0×10^{-4} | 1.0×10^{-4} |
| U-233 | 2.1×10^{-5} | 2.1×10^{-5} | 2.1×10^{-5} |
| I-129 | 9.4×10^{-6} | 9.4×10^{-6} | 9.4×10^{-6} |
| U-238 | 6.4×10^{-6} | 6.4×10^{-6} | 6.4×10^{-6} |
| Pd-107 | 4.4×10^{-6} | 4.4×10^{-6} | 4.4×10^{-6} |
| Np-237 | 3.9×10^{-6} | 3.9×10^{-6} | 3.9×10^{-6} |
| Eu-152 | 1.7×10^{-3} | 1.3×10^{-6} | 1.0×10^{-6} |
| Pu-242 | 6.2×10^{-7} | 6.2×10^{-7} | 6.2×10^{-7} |
| Tb-158 | 6.0×10^{-7} | 6.0×10^{-7} | 6.0×10^{-7} |
| U-235 | 2.7×10^{-7} | 2.7×10^{-7} | 2.7×10^{-7} |

*Values $< 1 \times 10^{-7}$

of radioactivity concentration. Nevertheless, the level of radioactivity concentration in HLW is generally higher than that of LLW.

HLW in the United States is defined as spent

nuclear fuel*, liquid solution generated from the first stage solvent extraction cycle of nuclear fuel reprocessing, or equivalent⁽³⁾. HLW is produced exclusively from the nuclear fuel cycle. Chemical (and nuclear characteristics of the HLW are strongly influenced by type of fuel claddings, method of reprocessing, and burn-up (irradiation) level of fuels. The HLW from reprocessing is currently stored in double-walled stainless steel tanks as liquid or calcine form and kept at reprocessing plants. A typical chemical composition of HLW is shown in Table 1. It should be noted that chemical composition and concentration vary considerably from tank to tank

and batch to batch. This variation will have a significant impact on the choice of host matrix to immobilize HLW. Typical radioactivity concentration of HLW is shown in Table 2.

LLW in the United States is defined as those wastes other than HLW or UR⁽⁴⁾. This broad definition of LLW covers all of power plant generated wastes, institutional wastes and most industrial and fuel fabrication wastes. LLW is further classified as fuel cycle wastes and non-fuel cycle wastes. Characteristics of LLW are summarized in Tables 3 and 4. LLW is generally stored temporarily within the facilities where it is generated. When the storage capa-

Table 3. Fuel-Cycle LLW

| Source | Original waste form | Major radioisotopes | Activity (Ci/MTHM ^a) | Final waste form |
|--|---------------------|--|----------------------------------|--|
| UF ₆ conversion (fresh) | Liquid | U, Ro-226, Th-230 | 1.07 × 10 ⁻⁶ | 1.3 × 10 ⁴ liters of liquid waste per MTU |
| | Solid | U, Ro-226, Th-230 | 9.40 × 10 ⁻³ | 1.1m ³ of solid waste (including CaF ₂) per MTU |
| UF ₆ conversion (recycle) | Solid | Fission products | 0.57 | ~1.3m ³ of solid waste (including CaF ₂ and carbonate-leached ash) per MTU |
| | | Pu-238, Pu-241 | 4.12 × 10 ⁻⁵ | |
| | | U | 1.35 × 10 ⁻³ | |
| Enrichment (fresh) | Liquid | U | 1.11 × 10 ⁻⁶ | 675 liters of liquid waste per MTU |
| Enrichment (fresh+10% recycle) | Liquid | U | 1.16 × 10 ⁻⁶ | 675 liters of liquid waste per MTU |
| | | Np-237 | 1.67 × 10 ⁻⁹ | |
| | | Pu-239 | 3.33 × 10 ⁻¹² | |
| | | Fission products | 3.40 × 10 ⁻⁴ | |
| Fuel fabrication (fresh+10% recycle) | Liquid | U, Th-234, Pa-234 | 1.20 × 10 ⁻³ | 5.7 × 10 ⁵ liters of liquid waste per MTU |
| | Solid | U | 7.34 × 10 ⁻³ | 4.6m ³ of solid waste (including CaF ₂) per MTU |
| Fuel fabrication (mixed oxide) | Solid | U, Pu, Am | 3.47 | ~2265 (98.2)m ³ of solid waste per MT of plutonium (uranium) processed |
| LWR Operation ⁸ PWR(1000MWe) BWR(1000MWe) | Liquid | Fission products and | 19.3 | 184m ³ of resins, liquids and sludge typically solidified |
| | Solid | Activation products | 1.9 | |
| | Liquid | Fission products and activation products | 100 | |
| Reprocessing | Solid | | 12.2 | 326m ³ of general trash |
| | Solid | Pu, Am | 0.53 | ~1586(15.0)m ³ of solid waste per MT of plutonium (uranium) processed |
| | | Fission products | | |

^a Metric ton of heavy metal

* Although spent fuel is categorized as HLW, the discussion of HLW in this paper will be limited to reprocessed HLW.

Table 4. Non-Fuel Cycle Wastes

| Radionuclide* | Use/Purpose | Amounts Employed |
|---------------------|---------------------------------|--|
| Americium-241 | Smoke detectors | • up to 5 μ Ci in domestic models 15+ μ Ci in commercial/ industrial units |
| (With Beryllium) | Moisture Content-Density gauges | • in the order of 50mCi |
| Gold-198** | Oil & Gas Well Logging | • up to 15 Ci |
| | Research/tracer | • up to Ci's, depending on whether lab work or field studies |
| Carbon-14** | Research/tracer | • in the order of μ Ci |
| Cobalt-57, 58, 60** | Medical/diagnostic | • in the order of 1 μ Ci per does |
| Chromium-51** | Medical/diagnostic | • in the order of 50 μ Ci per dose |
| Caesium-137 | Moisture Content-Density gauges | • up to tens of mCi |
| | Medical/therapeutic | • up to 200mCi or so per treatment |
| | Oil & Gas Well Logging | • up to 1~2 Ci |
| | Density & Level gauges | • up to 5 Ci |
| Gallium-67** | Medical/diagnostic | • in the order of 1.5 mCi per dose |
| Tritium | Luminescent devices | • up to 200mCi in watches, • up to 25 Ci in exit & similar signs |
| Tritium** | Research/tracer | • in the ordel of μ Ci |
| | Oil & Gas well/tracer studies | • up to tens of Ci |
| Iodine-125** | Medical <i>in vitro</i> tests | • in the order of μ Ci |
| | Research/tracer | • in the order of μ Ci |
| Iodine-131** | Medical/dignostic | • up to 50 μ Ci or so per dose |
| | Medical/therapeutic | • up to 100 mCi per dose |
| Iridium-192** | Oil & Gas Well Logging | • in the order of μ Ci |
| Krypton-85 | Thickness gauges | • up to 1 Ci |
| Neodymium-147** | Research/tracer | • up to Ci's, depending on whether lab work of field studies |
| Polonium-210 | Static eliminators | • up to 100 mCi or so in industrial units |
| Scandium-46** | Research/tracer | • up to Ci's, depending on whether lab work or field studies |
| Strontium-90 | Thickness gaugas | • up to 25mCi |
| Technetium-m** | Medical/diagnostic | • up to 20mCi per dose |
| Thallium-201 | Medical/diagnostic | • in the order of 1.5 mCi per dose |

* Sealed sources excepted where marked**

cities are filled, the wastes are treated or processed as needed and finally shipped off site for disposal.

UR is composed of tailings from uranium milling operations, which extract "yellow cake" (U_3O_8) from uranium ore. Chemicals added during the processing of uranium ore become part of the tailings. Most mills use acid solvents to extract uranium from the ore. These mills discharge a mixture of solid tailings and liquid to an impoundment area, referred to as a tail-

ings pond or tailings pile. Part of the liquid is recycled to the mill but more of it evaporates or seeps into ground. The principal radiological health hazard due to UR comes from Ra^{222} , a decay product from U^{238} . Non-radioactive toxic chemicals such as arsenic, lead, selenim, sulphate, and nitrate may also cause health hazards.

Waste Management Alternatives

The general approach for waste management

consists of (1) waste treatment to make hazardous radionuclides less mobil or dispersible, (2) packaging of treated wastes for easy handling and transportation, and (3) disposal of waste packages in a location where radionuclides released from the waste packages will take long tortuous pathways before reaching to biosphere.

(1) Waste Treatment and Packaging
(Engineered Barriers)

Various waste treatment and packaging technologies have been developed during the past 10 years to immobilize and stabilize radioactive waste so that it becomes less mobile and dispersible. Containing radioactivity at its source is considered by some people to be the most effective method to minimize risk due to radioactive waste disposal. Others argue that natural barriers (e.g., geology) will provide more effective protection. In either case, containment of wastes within a well defined boundary (engineered barrier) will be a very convincing way of assuring public that their health and safety is adequately protected from radioactive wastes. The followings are some examples of waste treatment and immobilization methods currently used or proposed.

HLW Containment Materials

Containerization: Containment of radioactive waste within a durable canister is an obvious first step of immobilization. Canisters have been developed historically for the purpose of meeting handling and transportation safety requirements. Various metallic canister materials such as stainless steel, titanium alloys (Ticode 12) and nickel alloys are considered for HLW containment. A waste package design using low carbon cast steel overpack is currently considered by the U.S. DOE for HLW. Regular 55-gallon drums (carbon steel) are used for LLW, but U.S. industries are currently developing new container materials such as high-density polyethylene (Marlex by Chem Nuclear) to improve

Table 5. Typical HLW Borosilicate Glass

| Chemical Component | Frit (wt%) | Solid Waste (wt%) |
|--------------------------------|------------|-------------------|
| SiO ₂ | 57.5 | 48.8 |
| B ₂ O ₃ | 25 | 14.2 |
| Na ₂ O | 17.5 | 15.0 |
| Al ₂ O ₃ | | 8.4 |
| Fe ₂ O ₃ | | 2.6 |
| MgO | | 6.3 |
| Ni/Cr | | 0.2 |
| Fission products | | 4.5 |

corrosion resistance.

Vitrification: This is a well developed technology and applied to immobilize HLW in France (AVM), Germany (PAMELA), Japan, and United States.⁽⁵⁾ The composition of glass most widely used is based on the borosilicate system with various additives to improve the chemical durability of glasses. An example of glass composition is shown in Table 5. The glassy state is thermodynamically metastable and will be vulnerable to devitrification, phase separation, or hydro-thermal attack. However kinetic processes governing the above alterations are extremely slow under normal conditions. Since glass has non-crystalline amorphous structure, it can accommodate the range of chemical compositions and concentrations without disrupting the basic glass structure. This is one of very important features for solidification of HLW in light of wide compositional and concentration range of HLW shown in Table 1.

SYNROC: SYNROC is an assemblage of titanium based synthetic materials such as Hollandite, Perovskite, and Zirconolite. The concept of using SYNROC to immobilize HLW was introduced by Professor Ringwood of the Australian National University.⁽⁶⁾ Geochemical durability and persistence of these titanium based minerals have been proven by studies of various natural analog materials. Moreover, these crystalline materials exist in thermodynamically stable states of the chemical composition. The-

Table 6. Typical Synroc Composition

| | "Hollandite" 40% | Zirconolite 35% | Perovskite 25% | Bulk SYNROC Composition |
|--------------------------------|------------------|-----------------|----------------|-------------------------|
| TiO ₂ | 71.0 | 50.3 | 57.8 | 60.3 |
| ZrO ₂ | 0.2 | 30.5 | 0.2 | 10.8 |
| Al ₂ O ₃ | 12.9 | 2.5 | 1.2 | 6.3 |
| CaO | 0.4 | 16.8 | 40.6 | 16.2 |
| BaO | 16.0 | — | — | 6.4 |
| Total | 100.5 | 100.1 | 99.8 | 100.1 |

refore, in theory, any radioactive materials locked within these crystalline structural "cages" are very effectively immobilized. The base composition of SYNROC is shown in Table 6. Hot isostatic pressing (HIP) is required to produce a dense SYNROC monolith, and further research and development is needed to demonstrate full scale manufacturability. Some researchers argue that it is questionable whether one can ever synthesize mineral phases identical to naturally occurring one which took millions of years to reach equilibria with their surroundings. There are also some doubts with respect to SYNROC's capability to accommodate various waste compositions and concentration of waste streams. This possible inadequacy may lead to some limitation of waste loading factors in SYNROC.

Supercalcine: The concept of Supercalcine for HLW immobilization is similar to that of SYNROC, except that Supercalcine is composed of

primarily silica based ceramic phases such as Apatite, Pollucite, and Monazite.⁽⁷⁾ Primary ceramic phases of Supercalcine are shown in Table 7. Since Supercalcine is unconsolidated as produced, it may need a secondary binder such as metal matrix to reduce the potential surface area available for aqueous leaching of radioactivity.

FUETAP: This material is concrete formed under elevated temperature and pressure (≤ 70 kg/cm², $\leq 250^\circ\text{C}$.)⁽⁸⁾ FUETAP concept is considered for immobilization of defense HLW and TRU wastes. FUETAP is easy to make and relatively cheap. However, its chemical durability is not as good as that of glass or crystalline matrix under severe conditions.

Other containment materials such as coated particle (based on HTGR fuel technology), metal matrix, and cermet were proposed for immobilization of HLW.

Table 7. Supercalcine Phase

| Constituent | Fixation Phase | Structure Type |
|--------------------------|---|----------------|
| Cs,Rb | (Cs,Rb)AlSi ₂ O ₆ | Pollucite |
| Sr,Na,Mo | (Ca,Sr) ₂ [NaAlSiO ₄] ₆ (MoO ₄) ₂ | Sodalite |
| Sr,Ba,Mo | (Ca,Sr,Ba)MoO ₄ | Scheelite |
| Sr,RE*[PO ₄] | PEPO ₄ | Monazite |
| | (Ca,Sr) ₂ RE ₃ [SiO ₄] ₆ O ₂ | Apatite |
| Ce,U,Zr | (Ce,U,Zr)O ₂ + x, (Zr,Ce)O ₂ | Fluorite |
| Fe,Ni,Cr | (Fe,Ni)(Fe,Cr) ₂ O ₄ | Spinel |
| | (Fe,Cr) ₂ O ₃ | Corundum |
| Ru | RuO ₂ | Rutile |

* RE=rare earths, particularly La, Pr, Nd, Sm, Gd (and probably Am, Cm).

LLW Containment Materials

Hydraulic Cement: This material is most widely used to immobilize LLW in the United States. Hydraulic cement is a combination of silicates and aluminates. Base compositions for the Portland type cement are shown in Table 8. The technology is well developed and the cement can accommodate variety of waste streams with some chemical adjustment. Hydraulic cement is mechanically stable under burial conditions and does not produce any free liquid after proper setting. Leachability of mobile ions such as cesium is high, but can be reduced by using some

Table 8. Properties Portland Cement
Principal Compounds Present in Portland Cements

| Compound | Formula | Abbreviation |
|-----------------------------|---|-----------------------|
| Tricalcium silicate | $3\text{CaO}\cdot\text{SiO}_2$ | C_3S |
| Dicalcium silicate | $2\text{CaO}\cdot\text{SiO}_2$ | C_2S |
| Tricalcium aluminate | $3\text{CaO}\cdot\text{Al}_2\text{O}_3$ | C_3A |
| Tetracalcium aluminoferrite | $4\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot\text{Fe}_2\text{O}_3$ | C_4AF |

Compound Composition of Portland Cements

| Type of cement | Compound composition, % | | | |
|--------------------------|-------------------------|----------------------|----------------------|-----------------------|
| | C_3S | C_2S | C_3A | C_4AF |
| I. Normal | 45 | 27 | 11 | 8 |
| II. Modified | 44 | 31 | 7 | 13 |
| III. High early strength | 53 | 19 | 10 | 7 |
| IV. Low heat | 20 | 52 | 6 | 14 |
| V. Sulfate resistant | 38 | 43 | 4 | 8 |

Behavior of Principal Compounds Present in Portland Cement

| Property | C_3S | C_2S | C_3A | C_4AF |
|---|----------------------|----------------------|----------------------|-----------------------|
| Rate of reaction | medium | slow | fast | slow |
| Heat liberated per unit of compound | medium | small | large | small |
| Cementing value per unit of compound: Early | good | poor | good | poor |
| Ultimate | good | good | poor | poor |

additives such as polymers and Metso Beads*.

Urea-Formaldehyde (UF): UF was introduced to immobilize LLW from power reactors. UF is a polymerization product of urea (H_2NCONH_2) and formaldehyde (HCHO). Radionuclides are "mechanically trapped" in the UF matrix during polymerization. However, liquid can be "squeezed out" due to condensation reactions, which produce "free liquid". Due to this free liquid problem, the application of UF for LLW is phasing out.

Bitumen: Bitumen is widely used in Europe and Japan to immobilize LLW but has limited useage in the United States. Its technology has been well developed by the roofing and road covering industries. The basic chemical constituents of bitumen are carbon disulfide and vari-

ous hydrocarbons. Due to evaporation of water during the bituminization process, significant volume reduction can be achieved. Chemical durability of bitumen with wastes containing Na_2SO_4 is very poor and needs to be improved for application to LLW from boiling water reactors.

Dow Binder: Dow Industrial Services is marketing a proprietary vinyl esterstyrene polymer system for solidification of LLW. The binding process uses a combination of binder (vinyl ester resin) with small amounts of a catalyst and a promoter. The Dow binder has very good leach resistance property and is an effective binder for solidifying decontamination solution containing chelating agents.

Absorbent materials such as vermiculite, diat-

* Proprietary additives by Hittman Nuclear and Development Corporation.

omaceous earth, and absorbent clays are often used to immobilize liquid LLW containing oil, solvent, and other liquid wastes which are difficult to solidify.

UR Containment Materials

Chemical coatings, asphalt, soil cover, and rock cover (rip-rap) are used to reduce Ra²²² emission from uranium mill tailings, and to prevent wind erosion and water infiltration of UR tailing piles.

Various volume reduction methods such as (HLW), evaporation (LLW), incineration (LLW, TRU), and acid digestion (HLW, TRU) will improve the stability of wastes as well as reduce volume of wastes to be disposed. Volume reduction may be a very important consideration as costs of waste transport and disposal escalate.

(2) Siting of Waste Disposal Facilities

Subsurface land disposal is the U.S. choice for disposal of HLW and LLW. Ocean disposal is practiced by OECD countries for disposal of LLW and IMLW. Discussion of waste disposal siting in this paper is limited to the underground disposal methods. The most probable pathway for radionuclide migration and human exposure is through the groundwater system. Therefore, the basic principle of siting is to locate the site in a dry region with low hydraulic conductivity and to take advantage of long tortuous groundwater pathways. Other features that should be considered are geologic characteristics to insure long-term stability of site and geochemical conditions benign to waste packages as well as retarding the migration of radionuclides in groundwater transport (e.g., sorption, precipitation).

HLW Disposal Sites: There are no HLW disposal sites currently operating in the Free World. The existing HLW is temporarily stored in surface facilities. Most of the countries producing HLW such as United States, United Kingdom, France, Germany (FRG), Japan, and

Table 9 Candidate Host Rock for HLW Disposal

| Nation | Host Rock | Candidate Site |
|---------|---------------|------------------------|
| USA | Salt | Carlsbad Avery Island* |
| | Basalt | Hanford |
| | Tuff | Nevada Test Site |
| FRG | Salt | Gorleben Asse* |
| Canada | Granite | |
| UK | Granite Clay | Troon |
| France | Granite Clay | |
| | Salt | |
| Sweden | Granite | Strippa* |
| Belgium | Clay/Shale | Mol* |
| Japan | Granite/Shale | Akenobe* |

Note : * Underground laboratory only

Canada decided to dispose the HLW in deep geologic repositories constructed at depth ranging from few hundred meters to one thousand meters. The choice of repository host rock is influenced by the availability of stable rock masses in respective countries. Candidate host rocks for each country are listed in Table 9.

Formation of large size rock salt is due to absence of flowing groundwater during past millions of years. Therefore, it can be reasonably assumed that salt sites will be free from flowing groundwater for long times in the future. However, any significant amount of entrained water may be very corrosive with usually high ionic strength. The mean water content of salt is low (generally less than 1%), but there are wide localized variations. Additionally, experiments have shown that water tends to migrate up a thermal gradient toward a heat source (waste package). The chemical reactions that would take place under elevated temperatures and pressures, and in the presence of radiation, are complex and not completely understood. The brine solutions could be deleterious to the canister/overpack and the waste form.

Granite also contains water in very small amounts, largely in fractures. Investigation to date indicates granite to be a potentially more

benign environment than salt. For example, its water is generally of low ionic strength, thus reducing its corrosion potential. Shales are considered attractive because of their plasticity at expected lithostatic loading and their high sorptive capability. On the other hand, it is known that some shales undergo mineralogical changes at temperatures above 100°C. The effect of these changes on interactions with the waste form is unknown.

Basalt formations are typically developed from layered sequences of lava flows. Although many of the basalts are highly jointed, some flows may be relatively impervious to water. Additionally, secondary minerals, predominantly clays, may fill the joints and cracks, effectively sealing them. The geochemistry of water in the interbeds appears to be controlled by the nature of the sedimentary interbeds and not the basalts themselves. Their composition appears to show an equilibrium with feldspars.

Other proposed geologic host candidates are tuff, alluvium, and anhydrite. Research on these has recently been undertaken.

LLW Disposal Sites: The LLW has been disposed in near surface shallow trenches similar to those used in typical sanitary landfills. Due to increased public concerns on general environmental pollution, most countries are instituting more stringent environmental protection policies, which will require more careful evaluations of site hydrology, geochemistry and general geologic stabilities of the region. Better design of trenches including trench caps, lining materials, and drain systems will also be required.

Ideal LLW sites should be in the areas where population density is low and far removed from population centers. However, this condition cannot always be met by every country. Alternative disposal method such as totally engineered facility (e.g., Le Centre de la Manche-France)

of island disposal (Taiwan) is practiced.

The siting of UR piles are very limited to the location of uranium mines.

It should be emphasized that the site suitability consideration is strongly influenced by the availability of sites. The U.S. is very generously endowed with respect to available land space for waste disposal, which allows to pursue a policy of "defense-in-depth" by requiring durable engineered barriers and stable and impermeable natural barriers for waste disposal and isolation. Other countries may have to rely more on the engineered barrier due to limitations of available sites.

Regulation

A brief discussion of regulation governing the disposal of HLW, LLW, and UR in the U.S. is presented below:

(1) HLW

The U.S. Environmental Protection Agency (EPA) is responsible for establishing general radiation protection criteria and is expected to publish these criteria for the HLW in the form of regulation, Part 191 of Title 40 of the Code of Federal Regulations (40 CFR 191). The NRC is responsible for implementing the EPA's criteria. Therefore, the logical sequence of rulemaking should be publication of 40 CFR 191 followed by 10 CFR 60, which is the NRC's regulation for HLW management. Due to delay in EPA's rulemaking, NRC went ahead and finalized its regulation with the assumption that the final EPA's regulation will not be drastically different from the proposed one. The NRC's criteria include performance objectives of containment (300~1,000 years), controlled release of radionuclides (10^{-5} per year), and groundwater travel time (1,000 years). Additional criteria for retrievability, site selection, repository engineering, waste package fabrication, and

engineered components of repository are established in CFR 60.

(2) **LLW**

The NRC has recently finalized 10 CFR Part 61, which will be effective in December 1983. The performance objectives of 10 CFR 61 include dose limits to the general population and occupational personnel, and protection from inadvertent intruders. Other criteria for site selection, trench design, and monitoring are established in 10 CFR 61.

(3) **UR**

The regulation governing disposal of uranium mill tailings is established in Appendix A to 10 CFR 40. The highlights of the regulation are a radon emission limit (2 picocuries per square meter per second) and various tailings pile stabilization methods.

Technical Issues and Research Needs

The regulations require isolation or slow and gradual release of radionuclides to the biosphere during a period when radioactivity in wastes presents significant hazards. The hazard period can be a few hundred to several thousand years depending upon waste characteristics. Therefore, the main technical issue is related to confirmation of long-term characteristics projected (or assumed) in the assessment of waste isolation or waste migration. The key characteristics and parameters pertinent to waste isolation are long-term containment effectiveness of various waste treatment and immobilization methods, groundwater flow through various geologic media, and geochemical interaction between the radionuclides and surrounding rocks.

The difficulty of confirmation is primarily due to the uncertainties associated with the long-term prediction of realistic behavior of various waste isolation parameters based on short term and small scale experimental data. The rate of

radionuclide release from engineered barriers and migration through geologic media are extremely slow under realistic conditions. Degradation processes of engineered materials are gradual under disposal conditions. Therefore, experiments are generally conducted under various accelerated conditions to yield any measurable results. Short-term experimental data are extrapolated to predict long-term performance of various engineered and natural barriers. Extrapolation is also needed in space, since the experimental data are generally based on small scale tests, while a real disposal facility and its surrounding environment which have to be assessed extend over many tens or even hundreds of kilometers.

Additional uncertainty arises from the fact that our understanding of basic phenomena and processes related to waste migration is incomplete. For example, the mechanisms and process of radionuclide leaching from a waste form matrix such as glass or cement is not well understood, hence any long-term assessment of leaching behavior includes unquantifiable uncertainties. Failure mechanisms and the rate of degradation for various container/canister materials used to contain HLW (e.g., stainless steel, carbon steel, titanium alloys) are not well quantified. Consequently, no credit is given to containers or radionuclide transport calculations, thus resulting in an overestimation of risks. Geochemical interactions and resultant retardation of radionuclide migration is quantified by an empirical method based on gross effect measurements (so called K_d -measurements). Due to a lack of understanding of each contribution to retardation, accuracy and reproducibility of K_d values are very poor. The phenomena related to groundwater flow through various different geologic media need to be better characterized to have a realistic assessment of radionuclide transport far from the waste. Various existing

ground transport models have not been able to describe accurately what was observed at LLW burial sites such as the one at Maxey Flats, Kentucky.

In order to resolve these technical issues, current efforts in waste management research are focused on *mechanistic research*. For example, in order to understand and quantify radionuclide leaching behavior, emphasis is given to leaching mechanisms research using various advanced spectroscopic methods to characterize kinetics of leaching surfaces rather than to conventional solution chemistry measurements⁽⁹⁾. There are also research programs which attempt to interpret geochemical retardation phenomena on the basis of solubility limits, valence states of radionuclide species, and sorption-desorption mechanisms.

Natural analog research can also be a very effective tool to generate data and information that can be used to resolve technical issues and reduce uncertainties in understanding phenomena related to radioactive waste disposal. The hydrologic and geochemical research on the ore body at Oklo in the African republic of Gabon provided a valuable record of long-term radionuclide migration through geologic system. To the extent that radionuclide migration from such ore bodies in analogues to the long-term migration of radionuclides from HLW in deep geologic repositories, similar research will be very useful to resolve various technical issues regarding long-term radionuclide migration through geologic media if suitable natural analog can be identified.

For UR, research is needed to understand the effectiveness of various tailings pile stabilization techniques and identify potential contamination problems associated with alternative uranium extraction methods such as in-situ solution mining.

In conclusion, the major technical issue rela-

ted to radioactive waste disposal appears to be the level of uncertainties with respect to risk assessment rather than the level of absolute risk itself. To resolve the issue, waste management research should be focused on developing an understanding of basic phenomena and processes relevant to radionuclide transport and migration through various engineered and natural barriers.

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