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**Geologic Disposal of High-Level Radioactive Wastes—
Earth-Science Perspectives**

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By J. D. Bredehoeft, A. W. England, D. B. Stewart,
N. J. Trask, and I. J. Winograd

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*A summary of factors and processes
that must be understood for the safe containment
of high-level radioactive waste*

DEPARTMENT OF THE INTERIOR
DONALD PAUL HODEL, Secretary

U.S. GEOLOGICAL SURVEY
Dallas L. Peck, Director



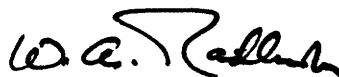
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FOREWORD

Earth scientists have been concerned with the problem of disposal of radioactive wastes since 1955 when the National Academy of Sciences gathered 65 scientists in Princeton, N.J., to consider the geological, biological, physical, and chemical aspects of waste containment. Investigations during the 1950's and early 1960's examined the feasibility of disposal of liquid high-level wastes in deep geologic basins and in salt mines. Scientists since then, sponsored largely by the U.S. Atomic Energy Commission and later by the U.S. Energy Research and Development Administration, carefully considered salt deposits as a repository for solidified high-level wastes. Within the past few years, in response to growing pressures for a resolution of the problem of disposing of the wastes, earth scientists at various universities and government laboratories, as well as at the U.S. Geological Survey (USGS), also began an intensive examination of the problem. As a result of this expanded examination, modified concepts of geologic disposal have evolved, and aspects of some older concepts have been questioned. Some of these changes in outlook and philosophy as perceived by USGS scientists are described in this Circular. Because the authors are confident that acceptable geologic repositories can be constructed, this paper should not be construed as an attempt to discredit the concept of geologic containment or the work done in the 1960's and early 1970's. However, the earth-science problems associated with disposal of radioactive wastes are not simple, nor are they completely understood. The many weaknesses in geologic knowledge noted in this report warrant a conservative approach to the development of geologic repositories in any medium. Increased participation in this problem by earth scientists of various disciplines appears necessary before final decisions are made to use repositories. Basic philosophical, as well as technological, issues remain to be resolved.



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Geologic Disposal of High-Level Radioactive Wastes— Earth-Science Perspectives

By J. D. Bredehoeft, A. W. England, D. B. Stewart, N. J. Trask, and I. J. Winograd*

INTRODUCTION

An effective solution to the problem of long-term storage of radioactive waste is essential to the expanded use of nuclear power. Radioactive waste is the inevitable byproduct of the generation of electricity by nuclear reactors or of the production of plutonium for weapons. Fuel assemblies must be removed periodically from nuclear reactors as fission products accumulate. These spent and contaminated fuel assemblies contain large concentrations of fission products such as ^{90}Sr and ^{137}Cs , which decay to innocuous levels during several hundred years, and of longer lived transuranic elements and their daughters, elements such as plutonium, americium, neptunium, and radium. These elements persist for as long as several million years.

The spent fuel from a nuclear reactor must be isolated either permanently or temporarily from the biosphere. Spent fuel can be processed chemically to recover most of the plutonium and unfissioned uranium for possible reuse. The residuum from this process is a highly radioactive byproduct which must also be isolated from the biosphere. This byproduct is what is usually referred to as "high-level waste." The United States does not plan to reprocess spent fuel chemically; thus, the immediate problem of commercial radioactive waste is one of handling, storing, and ultimately disposing of spent fuel assemblies.

Radioactive wastes from military activities are somewhat different. These wastes are mainly the residue after plutonium has been extracted from radioactive materials that have

been irradiated in a reactor designed specifically to produce plutonium. Although military wastes contain many of the same fission products and transuranic elements as commercial waste and occupy significant volumes, their total radioactivity is relatively low—at present about the same as that stored in spent-fuel assemblies from commercial reactors (Nuclear Energy Policy Study Group, 1977, p. 244).

The volume of commercial radioactive waste that will accumulate during the next few decades will depend upon the growth of the nuclear-power industry and on the form of the waste. King and Baker (1976, p. 206) estimated that by the year 2000, as many as 476,000 spent fuel assemblies will be on hand if none has been reprocessed. If these spent fuel elements were to be reprocessed, the resulting solid high-level waste would occupy approximately 3,000 cubic meters (m^3) (Blomeke and Kee, 1976, p. 99), and accompanying intermediate-level wastes contaminated by transuranic elements would occupy a volume one order of magnitude larger than the volume of high-level wastes (Blomeke and Kee, 1976, p. 108).

Wastes generated by military activities, 81 million gallons in liquid and solidified form, are held in temporary storage by the U.S. Department of Energy (DOE) at Hanford, Wash., Savannah River, Ga., and Idaho Falls, Idaho (Nuclear Energy Policy Study Group, 1977, p. 250). Because of their chemistry, most of these wastes present uniquely difficult problems in

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handling (Nuclear Energy Policy Study Group, 1977, p. 249).¹

It is generally accepted that repositories in geologic media can provide the most certain safe containment of radioactive waste. DOE is developing, and intends to have licensed sometime in the 1980's, a Waste Isolation Pilot Plant (WIPP) in bedded salt near Carlsbad, N. Mex. The Nevada Test Site and the Hanford, Wash., facilities are being explored for suitable geologic repositories. Evaluations of sites in bedded salt of the Salina and Permian basins, salt domes of the gulf coast, and salt anticlines of the Paradox basin are also continuing. Reconnaissance work to assess the potential of a repository in shale or crystalline rock is underway. In the first repositories used, wastes would be retrievable for 10 to perhaps as many as 50 years.

Radioactive wastes are highly toxic. A commonly cited measure of this toxicity is afforded by a computation of the quantity of water needed to dilute high-level wastes to levels specified in the Radiation Concentration Guides (RCG, see headnote to table 1). These volumes were tabulated by Blomeke and Bond (1976); they are reproduced in table 1 and are compared with the volume of the oceans and of fresh water in lakes, rivers, and glaciers in table 2. A volume of 5.2×10^{16} m³ of water, which is almost 4 percent of the oceanic volume

¹ An additional 600,000 gallons of liquid high-level waste and sludge derived from both commercial and military spent fuel are in temporary storage at West Valley, N.Y. (Nuclear Energy Policy Study Group, 1977, p. 252).

TABLE 2.—World supply and volume of annually cycled water (abbreviated from Nace, 1969)

[Nace's (1969) data were reported in km³ × 10⁻³. We converted his data to cubic meters to permit a direct comparison with the data of table 1 (1 km³ = 10⁹ m³)]

| Source of water | Global volume (in m ³) | Percentage of total water |
|-----------------------------------|------------------------------------|---------------------------|
| Oceans | 1.4×10^{18} | 97.6 |
| Rivers (average channel storage) | 1.7×10^{12} | 0.0001 |
| Freshwater lakes | 1.2×10^{14} | 0.0094 |
| Saline lakes; inland seas | 1.0×10^{14} | 0.0076 |
| Soil moisture; vadose water | 1.5×10^{14} | 0.0108 |
| Ground water (upper 1,000 m only) | 7.0×10^{15} | 0.5060 |
| Ice caps and glaciers | 2.6×10^{20} | 1.9250 |
| Annual river runoff | 3.0×10^{13} | 0.0021 |

(1.4×10^{18} m³), would be needed to dilute the wastes on hand at the year 2000 to levels specified in the RCG; this volume is almost double that of fresh water in global storage in lakes, rivers, ground water, and glaciers. Even after a million years, the volume of water needed to dilute these wastes to the levels specified in the RCG is significant in terms of water stored in individual major lakes and aquifers. Another crude measure of the potential waste hazard was provided by Cohen (1977, p. 28). He showed that the potential for millions to billions of fatal cancer doses is contained in the wastes provided by just 1 year of United States nuclear-power production if all the waste were to be ingested, an admittedly unlikely scenario. The above-cited tabulations are simplistic because they ignore the environmental pathways to man after production and burial of the wastes, yet such computations are a sobering reminder of the seriousness of the waste-disposal endeavor. Clearly, radioactive wastes con-

TABLE 1.—Properties of high-level wastes to be accumulated by the year 2000 (from Blomeke and Bond, 1976, p. 87)

[Toxicity is defined as the cubic meters of air or water that would be required to dilute the radioactive constituents to levels specified in the Radiation Concentration Guides (RCG; see column 2 of tables I and II, U.S. Code of Federal Regulations, Title 10, Pt. 20) as the maximum allowable for unrestricted use. MCi, megacurie; kW, kilowatt]

| | Accumulated by year 2000 | Time elapsed following year 2000, in years | | | |
|--|--------------------------|--|----------------------|----------------------|----------------------|
| | | 1,000 | 10,000 | 100,000 | 1,000,000 |
| Radioactivity, MCi | 158,000 | 63.1 | 24.5 | 4.7 | 1.6 |
| Fission products | 155,000 | 4.7 | 4.5 | 3.4 | 0.7 |
| Actinides | 3,400 | 58.4 | 20.0 | 1.3 | 0.9 |
| Thermal power, kW | 770,000 | 1,500 | 470 | 36 | 21 |
| Fission products | 657,000 | 5.3 | 4.9 | 3.1 | 0.2 |
| Actinides | 113,000 | 1,500 | 470 | 33 | 21 |
| Inhalation toxicity, m ³ air | 1.2×10^{22} | 3.4×10^{20} | 1.4×10^{20} | 1.0×10^{19} | 4.4×10^{18} |
| Fission products | 7.0×10^{20} | 1.8×10^{15} | 1.8×10^{15} | 1.3×10^{15} | 2.1×10^{14} |
| Actinides | 1.1×10^{22} | 3.4×10^{20} | 1.4×10^{20} | 1.0×10^{19} | 4.4×10^{18} |
| Ingestion toxicity, m ³ water | 5.2×10^{16} | 1.1×10^{13} | 3.3×10^{12} | 1.7×10^{12} | 6.4×10^{11} |
| Fission products | 5.2×10^{16} | 1.8×10^{10} | 1.8×10^{10} | 1.4×10^{10} | 2.4×10^9 |
| Actinides | 4.4×10^{14} | 1.1×10^{13} | 3.3×10^{12} | 1.7×10^{12} | 6.4×10^{11} |

stitute a major hazard. The magnitude of the associated risk depends upon the environmental pathways to man. An assessment of that risk depends upon our understanding of those pathways now and for thousands of years.

Wastes from a deep repository could be exposed to the biosphere either (1) by some geologic process such as tectonism, diapirism, or erosion that directly exposes the waste or (2) by some process that transports the wastes to the biosphere. Transport by ground water is acknowledged to be the most probable mechanism for moving the radioactive wastes the necessary distances from repository to the biosphere.

Development of acceptable waste repositories involves in the most general terms: (1) identification of sites that meet broad criteria for tectonic stability, slow ground-water movement, and long flow paths to the surface; (2) intensive subsurface exploration of such sites to determine the hydrologic and geologic conditions in and around the potential repository; (3) predictions of repository behavior based on the initial conditions and on various assumptions about the future; (4) evaluation of the risk associated with these predictions; and (5) a judgment whether these risks are acceptable. This Circular is not intended to address each of these steps in detail; we do discuss some of the key earth-science questions involved in the first four steps. Step 5, of course, requires a political decision and is beyond a technical discussion.

The authors of this Circular are confident that the steps outlined above can be carried out in such a way that the ultimate decision on the acceptability of a given site and waste-handling procedure will have a strong scientific and technical foundation. However, some key geologic questions are unanswered, and answers are needed before the risk associated with geologic containment can be confidently evaluated. In this Circular, we discuss briefly the various media that have been considered for geologic disposal of radioactive waste and point out the need for several relatively new techniques for characterizing the rocks in the immediate vicinity of a repository. We consider a variety of possible interactions among the mined opening of the repository, the waste, the host rock, and any water that the rock may contain. Many

of these interactions are not well understood, and this lack of understanding contributes considerable uncertainty to evaluations of the risk of geologic disposal of high-level waste. Because salt is being considered as a potential repository medium, we point out some geochemical uncertainties connected with its use. Knowledge of the ground-water flow path from a repository is important in providing a backup containment system if waste should leak from the repository, and we devote a section to considerations of describing and modeling such systems. In the next section, modeling and geologic prediction are discussed from the point of view of some philosophical aspects of prediction in the geosciences. A concluding section suggests research areas that we feel need emphasis. The discussion is qualitative throughout; quantitative treatment of several of the topics considered is underway in the Geological Survey and elsewhere.

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THE REPOSITORY HOST ROCKS AND SITE CHARACTERIZATION

Different rock types have been considered as the host rocks for radioactive-waste repositories. Rock salt received early attention because of its low porosity and permeability, its

plasticity, and its high thermal conductivity (Natl. Acad. Sci., 1957). In recent years, shale, limestone, granitic rocks, and basalt have been considered (Schneider and Platt, 1974). These rock types provide low primary permeability and, in many areas, a high degree of structural stability. Parts of the Precambrian shield are known to be extremely stable (Yardley and Goldich, 1975). The clay minerals in shales have a potential for significant radionuclide sorption (Brindley, 1977), although measurements of this property are still incomplete. Another rock type having a high sorptive potential is zeolitized tuff, which is widespread in the Basin and Range province of the Western United States. The overall desirable properties of these rock units, such as thickness, depth, homogeneity, and tectonic stability, have been discussed at length previously (ERDA, U.S. Energy Research and Devel. Adm., 1976) and are not reviewed here.

The identification of potential host rocks for radioactive-waste repositories and of broad geographical areas having generally acceptable properties is followed by more detailed studies of smaller specific areas of interest. The volume of rock that will be needed to accommodate an actual waste repository will probably be relatively small—approximately 10 km³ (ERDA, 1976). However, that volume must be intensively explored by all appropriate methods.

Planning a waste repository, like planning subsurface mining and tunneling, requires a full understanding of the subsurface geology, but the consequences of an undetected error in planning the waste repository are more severe. Finding unexpected ground-water conduits, shear zones, fault-displaced or pinched-out beds, abandoned excavations, or variable lithology could be hazardous and costly. The discovery of such anomalies during the excavation of a nuclear-waste repository could force abandonment of the site.

Confidence that a prospective site is likely to provide adequate containment derives from an understanding of structural and lithologic features apparent at scales from a few meters to tens of kilometers. Many critical features are subtle. For example, small faults, particularly strike-slip faults, can be extremely difficult to detect and to date. Fracture systems, which

could act as short-circuiting conduits for ground water, might be revealed from regional structure features clearly evident only at the scale of a Landsat image (Pickering and Jones, 1974; Drahovzal and others, 1974) or perhaps from buried structural features indicated by the distribution of soil moisture (Johnson and England, 1977). Furthermore, the necessary structural integrity and lithologic homogeneity cannot be assured simply through detailed surface geologic mapping and coring, particularly if the number of core holes is limited by the risk to the potential repository caused by extensive local drilling.

Techniques for remotely exploring a volume of rock are in various stages of development and should be applied to potential waste-repository sites as soon as practical. These methods are used from the surface, between boreholes, and at the head of an excavation. They include high-resolution seismic and acoustic techniques, which can detect fine-scale structural and lithologic variations (Laine and others, 1977), and electric and electromagnetic methods, which are primarily sensitive to the distribution of water (Scott and others, 1975; Daniels, 1977). Some of the most recently devised methods use short-pulse radar (Stewart and Unterberger, 1976; Watts and England, 1976) and continuous-wave interferometry (Rossiter, 1977); their use is limited to high-resistivity materials such as salt and perhaps dry crystalline rock.

The techniques cited provide a nondestructive way of characterizing the site in detail. Some of the borehole-to-borehole electrical techniques need to be further refined, but the critical need to map the original subsurface structure, lithology, and ground-water regime warrants the aggressive utilization of such techniques where appropriate.

PERTURBATIONS CAUSED BY THE REPOSITORY AND THE WASTE

The original host rock and any included water will be perturbed for substantial periods of time by the waste and by excavation of the repository. Three kinds of perturbations can be caused: (1) mechanical disturbances initiated by mining of the repository; (2) chemical disturbances caused by the introduction into the

subsurface of wastes and waste-generated fluids that are not in chemical equilibrium with the rock mass; and (3) thermal disturbances of some sort, very large if hot wastes are introduced; this stress compounds the mechanical and chemical perturbations.

A major engineering task in any repository will be to prevent water from entering the workings through fractures caused by the mining process. All excavations produce a disturbance in the in situ stress field. This disturbance can produce fracturing in the vicinity of the underground working. Most estimates suggest that this mechanical disturbance extends for distances that are about the same as the size of the underground opening (Cook, 1977). The mining disturbance can only increase the permeability of the rock in the vicinity of the repository. Moreover, current estimates (Union Carbide Corp., Nuclear Div., Office of Waste Isolation, 1977) suggest that the backfilling process will fill the underground working to a density of perhaps 80 percent of what it was before mining. Plastic deformation in shale and salt may, however, ultimately return these rocks to approximately their initial densities. In more brittle rocks, permeability may continue to increase even after backfilling of the repository as stresses are relieved. Shafts to the workings and exploratory holes in the vicinity of the workings will, of course, be carefully plugged but will still provide potential conduits for the entry and exit of water during substantial time periods.

Chemical effects brought on by the introduction of heat from the waste are discussed below. Chemical reactions that may take place at low temperatures are related to the incursion of fluids into the repository volume. These fluids will react with the waste components during long timespans, either dissolving them or producing a new waste form having changed solubility. For repositories in bedded or domed salt, the fluids at low temperatures will be saturated brines. The effects of these brines on the containment properties of salt are being studied at several laboratories. Interstitial brine is known to reduce the mechanical strength of salt. Although scientists usually assume that water will never enter the workings of a repository in salt in amounts of more than 1 percent of the mass

of solid salt and waste, the potentially serious consequences of greater amounts of water need to be explored. In addition to possibly entering through shafts and boreholes, water may enter a salt repository from brine pockets such as those that were found (unexpectedly) in one salt deposit (Boffey, 1975). The consequences of appreciable weakening of salt could be serious. One consequence could be an accelerated collapse of workings, which might facilitate sealing of a closed repository but would, at the same time, severely jeopardize retrievability and integrity in an active repository that was accidentally flooded. Another possibility is that the canisters or fuel assemblies could be moved in a direction determined by density contrasts and unrelieved stresses near them.

The presence of brine in a salt repository adjacent to waste canisters or fuel assemblies would create additional problems concerning the ultimate fate of the hazardous nuclides. The protective metal canisters and sleeves will certainly degrade quickly in the strong brine environment, and leaching of the wastes will be enhanced. Ultimate containment will therefore depend on the enclosing salt mass itself. Elements from the waste will be partitioned into the brines depending on such factors as complex formation, ion exchange, particle suspension in the dense brine, and colloid dispersion. The meager data available on this problem suggest that many elements will be partitioned into the liquid phase and that few elements will be partitioned into the solids. In other words, the capacity of salt to "fix" or adsorb the nuclides from the waste in insoluble form is apparently low. This is a widely recognized serious drawback of salt as the repository medium; the mechanical integrity of the salt medium thus becomes of extreme importance. A possible way around this difficulty is to wrap the wastes in materials that do have a strong tendency to fix the nuclides from the wastes; research on possible materials is beginning.

The amount of heat introduced into a host rock by the waste depends on the form of the waste. Canisters of high-level waste may produce as much as 5 kW 10 years after reprocessing (ERDA, 1976); this output declines to one-tenth of this value within 100 years. Spent fuel assemblies have lower heat output, approx-

imately 0.5 kW 10 years after removal from a reactor (Jenks, 1977), although the decay is less rapid.

Thermal energy introduced into the rocks surrounding the repository may cause complex mechanical and chemical changes. Increased temperatures in salt would further decrease mechanical strength of the salt-brine mixtures discussed above and would increase the creep rate of dry salt. Thermal expansion and later contraction will cause compressional and tensional stresses in confined media and may lead to widespread fracturing in brittle rocks. Preliminary calculations of thermal effects in brittle media have been carried out (for examples, see Ratigan, 1976). Usually such calculations use finite-element or finite-difference methods and deal with relatively close-in effects. A difficulty in such calculations for shale is lack of knowledge of the thermal properties of shale in three dimensions.

One of the more significant chemical changes caused by the introduction of thermal energy to a rock system will be the breakdown of hydrated minerals and consequent release of water. For example, mixed-layer montmorillonite-illite, a common constituent of shale, undergoes a phase transition involving increase in illite layers and the liberation of free water starting at temperatures as low as 100°C (Perry and Hower, 1972). At elevated temperatures in shale, this water will probably be in the form of steam. Oxygen, carbon dioxide, sulfur, and halogen gases may also be released. Interaction among preexisting minerals, volatile components, and waste will be promoted by high temperatures. Experience from the Geological Survey's geothermal studies indicates that hot moving fluids may alter existing minerals and form new ones, thus causing significant changes in permeability. Some of these changes could reduce permeability and fluid flow (Summers and others, 1978). These mineralogical changes may be accompanied by volume changes and attendant stress on the confining medium.

Thermal energy can also drastically modify hydrologic flow regimes by: (1) creating and driving convection cells; (2) inducing pressure gradients in fluids by their expansion, including the formation of systems containing both liquid

and gaseous phases; (3) decreasing viscosity of fluids; (4) changing the bulk permeability; (5) changing the rock-stress field in a manner to strongly affect the fluid-flow field; and (6) changing solution chemical characteristics. Participants in a symposium on the movement of fluids in largely impermeable rocks (Union Carbide Corp., Nuclear Div., Office of Waste Isolation, 1977) concluded that no adequate model now exists to describe fluid flow, energy or chemical species transport, or source kinetics during the time interval of the thermal pulse, namely, the first several hundred years after burial.

Conservative limits may need to be placed initially on the thermal load to be borne by a radioactive-waste repository until many of the problems outlined above have been resolved. We are not saying that it is not feasible to store hot wastes in a deep geologic repository. We are saying, however, that given the current state of our knowledge, the uncertainties associated with hot wastes that interact chemically and mechanically with the rock and fluid system appear very high.

A general rule might be to restrain the permissive maximum temperature to a level lower than that which will cause a gas (steam) phase to appear. The gaseous phase is to be avoided, if possible, because, among other reasons, its lower viscosity and larger volume than those of solid or liquid phases permit more rapid movement through porous media. The stress field and the presence of steam may interact to concentrate solution processes or chemical reactions in mechanically stressed regions near repositories; such interactions are still poorly evaluated, though they are potentially sources of breaches in containment.

Allowing the wastes to dissipate heat before burial also widens the spectrum of usable media. Montmorillonitic shales, for example, undergo a phase transition already mentioned involving reordering of mineral structure and release of free water at temperatures as low as 100°C. The fact that such mineralogical transitions yield large volumes of fluids is a possible deterrent for placement of hot wastes in such shale. Massive zeolitized ash-fall tuffs, which are common over large areas in the Basin and

Range province of the United States and which locally are as much as hundreds of meters thick, are also attractive as potential media for waste disposal. They have most of the same desirable properties as the montmorillonitic shales; they are low in permeability and very high in sorptive capacity (Ramspott and Borg, 1977). Unfortunately, phase changes starting at about 150°–200°C cause the release of free water and may preclude use of zeolitic tuffs for the disposal of hot wastes. Crystalline rocks have few of the hydrous phases present in shales and tuffs; their thermomechanical response to hot waste is unknown, however.

THE GROUND-WATER SYSTEM

The acknowledged most likely process for moving radioactive waste from repository to biosphere is transport by ground water. Model predictions of the fate of the emplaced waste must therefore take into account factors that affect transport by ground water. These include: (1) convective transport by the flow, (2) dispersive and diffusive transport, (3) chemical interactions with the rocks along the flow path, and (4) rates at which the wastes come into solution.

In the simplest one-dimensional case in which ground water flows through a porous medium, the velocity of flow is given by Darcy's law, which states:

$$v = \frac{-k}{e} \frac{\delta h}{\delta l}$$

where v is the average velocity of the fluid, k is the permeability, e is the porosity, and $\delta h/\delta l$ is the gradient of the hydraulic head. The average distance moved by water during an interval of time is a crude measure of how far long-lived radionuclides might be transported, assuming that the velocity field is uniform both in time and space, and that, as a worst case, the radionuclides are not sorbed by the porous media traversed. If, for example, we wished to restrict transport to 1 km in 10^5 years, the flow velocity should be no more than about 3×10^{-8} cm/s. Further, assuming a head gradient of 1/100 (1 percent) and a medium having a porosity of 10 percent, we compute a permeability along the flow path on the order of 3×10^{-7} cm/s (about 0.3 millidarcies). This permeability is not par-

ticularly low for natural rock materials. Rocks that have "intergranular" permeabilities at least this low include "tight" sandstones, shaly siltstones, a variety of shales, and many crystalline rocks, including most granites. The current DOE program to establish waste repositories is directed toward salt, shale, and crystalline rock in part because these media have low permeabilities. As the porosities of such media decrease, or as the head gradient increases, the permeability must be lower in order to maintain the same velocity of water flow. Several other factors in ground-water transport act to speed up or slow down the flow of contaminants.

Contaminants disperse and diffuse at the front of an advancing wave of ground water bearing such contaminants and tend to spread out their concentrations. These effects cause some contaminants to arrive faster than would be predicted by the average velocity of water movement (streamline convection) alone.

Adsorption and chemical reactions, including adsorption, ion exchange, and precipitation, retard or restrain the movement of some contaminants. Adsorption and ion exchange are strongly dependent on the chemical form of the contaminants, including their oxidation state and extent of complexing. Numerical measures of the sorptive properties of various chemical species vary through many orders of magnitude. They have yet to be determined for many of the actinide elements under repository conditions. However, the basic principles of these reactions are well known.

In order to construct a complete model of contaminant flow, the availability of the waste to percolating ground water must be known. This availability will depend in part on the form of the wastes. High-level wastes from reprocessing may be cast in a glass form that would have very low leachability (Marsily and others, 1977; ERDA, 1976); the leach rate of spent-fuel material appears to be comparable (Katayama, 1976). Even if the wastes have very low leachability, some contaminants will be released, and the rate of leaching may change significantly with time as the waste undergoes radioactive decay, interacts with the enclosing medium, or comes in contact with ground water of changing composition.

In summary, combinations of hydrologic factors can slow transport between the repository and the biosphere. Factors tending to lengthen the transport time include: (1) low permeability of the media, (2) high effective porosity of the media, (3) low gradient of hydraulic head, (4) high sorption capacity of the media, (5) long flow path, and (6) low rate of release of solutes by the wastes to the transporting fluids.

From the preceding general outline of factors influencing ground-water transport of radionuclides, preparing a predictive model of contaminant transport might appear relatively simple. Unfortunately, several large gaps exist in knowledge of the details of natural transport systems.²

Media in which ground-water flow is largely through fractures are apt to be present in any flow system and pose problems in predicting radionuclide transport. The presence of fractures increases the velocity of flow under the same gradient. Fracture flow velocity may be a few orders of magnitude faster than intergranular velocity. In addition, the sorptive capacity of fractured media is likely to be lower than that of a porous medium because the fluid is in contact with much less of the medium for shorter times.

In theory, fluid flow through fractured media can be predicted in two ways:

1. The flow through a network of fractures can be analyzed. The geometry of the network of fractures must be known, and thus the orientation of fractures and the size of fracture apertures must be known. Although the geometry of the fracture system may be known in the vicinity of an underground working, it seems difficult, if not unfeasible to know this in sufficient detail at any distance from the few boreholes or workings likely to be permitted at or near a repository.

2. The fracture system can be analyzed stochastically. This analysis uses variables such as permeability and porosity that are analogous to those for a porous medium. Unfortunately, this method of analysis is relatively recent and, as yet, is untested on a field scale.

In addition to natural fractures, manmade boreholes in the vicinity of the repository as

well as in the repository itself present problems. They must, even during hundreds of years, be considered as potential short-circuit pathways that could permit water flow from the repository horizon upward to shallow aquifers that may be utilized by man.

In addition to the problem of ground-water flow through fractures, a second major hydrologic problem involves measurements of fluid head and permeability in porous media of low permeability. These values are needed to characterize the hydrology around a repository, but meaningful in situ measurements of ground-water flow in low-permeability porous media have not been obtained routinely in the past. Although laboratory measurements of relevant variables are possible, experience in hydrology suggests that such measurements commonly vary by an order of magnitude or more from those obtained through in situ tests. The problem is compounded by the large number of variables that must be measured to describe the transport process through fractured porous media of low permeability.

The carbon-14 dating method provides a quasi-independent check on hydraulic computations of ground-water velocity, but only for waters that have been removed from the biosphere for less than 40,000 years, the practical limit of ¹⁴C dating of ground water. In all likelihood, the waters in the vicinity of most potential repositories will be at least tens of thousands of years old. Radioactive disequilibrium studies using the systems ²³⁴U/²³⁸U and ²³⁰Th/²³⁴U offer some potential for dating ground water 10,000 to 1 million years old; these methods need considerable refinement before they can be routinely applied, however. Another potential technique for determining rates of ground-water flow in some rocks involves measuring the amount of dissolved helium in the water and comparing this measure to the amounts of uranium and thorium that yield helium through radioactive decay in the rock traversed (Marine, 1977).

In order to predict contaminant movement, the physical and chemical properties of the media that control transport must be known for a length of flow path sufficient to describe the movement for the requisite radionuclide containment time (Apps, 1977). Although measur-

² These uncertainties were the subject of a recent symposium (Union Carbide Corp., Nuclear Div., Office of Waste Isolation, 1977).

ing these properties seems feasible, the manpower and observation time needed are not trivial. We need, as a minimum, the permeability and porosity of the media and the hydraulic head gradients all in three dimensions. In addition, we need to know the sorptive characteristics of the media along all paths, and we need to estimate the variable rates at which the solidified wastes will enter the transporting fluids. Needed, in particular, is information on the distribution and extent of major heterogeneities. The need for such data severely taxes both the available data base and the technology for generating it. Most of the requisite data are presently unavailable; most of the available data have such large error limits that their usefulness in predictive models is limited (Philip, 1975). Indeed, in a recent review of ground-water models in general, Bugliarello and Gunther (1974, p. 136) stated that "the complete description of the quality and quantity aspects of groundwater flow under complex * * * conditions * * * is still in the future." Some basic problems of geologic models in general are discussed below.

CONTAINMENT TIME FRAMES AND GEOLOGIC PREDICTION

The objective of geologic disposal of radioactive wastes in the ground is to preclude their reaching the surface or near-surface hydrosphere, and thereby the biosphere, until they have decayed to a point at which their concentrations no longer constitute a health hazard.³ Common geologic processes capable of transporting wastes to the surface or near surface after deep burial include ground-water movement, tectonism, diapirism, erosion, and volcanism. Equally important, tectonism, erosion, seismicity, glaciation, and climatic change may alter the initial conditions of ground-water flow described in the preceding section. Fracture permeability, hydraulic gradients, and flow-path length are all subject to change, and predictive models must take into account the probabilities of such changes (Burkholder and others, 1977).

³ We recognize that definition of what might constitute a health hazard upon release of selected radionuclides to the hydrosphere involves consideration of many biologic and surface-hydrologic processes capable of reconcentrating the wastes. In this report, we are concerned exclusively with pathways in the lithosphere.

HOW LONG IS NECESSARY?

How long must the wastes be sequestered and the individual and interactive effects of the processes discussed above be predicted? Different radionuclides require different containment periods ranging from several hundred to several million years. Six hundred years is the period commonly cited as mandatory for containment of ⁹⁰Sr and ¹³⁷Cs, which are expected to constitute 99 percent of the projected curie (Ci) accumulation in reprocessed waste by the year 2020 (Gera and Jacobs, 1972, table 2.5). These fission-product nuclides have half-lives of 29 and 30 years, respectively. Every 10 half-lives of a nuclide correspond roughly to a 1,000-fold reduction in radioactivity; hence, after 600 years, or roughly 20 half-lives, the activity of these nuclides is reduced to $10^{-3} \times 10^{-3}$ or 10^{-6} (one millionth) of its initial value.

Other longer lived fission products will also be present in the waste, depending in part on its form. Spent fuel assemblies, for example, contain significant quantities of ¹²⁹I that has a very long half-life of 1.6×10^7 years and that poses a potential hazard at least this long.

The toxicity during periods as long as 10^7 years computed for radium and selected actinide elements and actinide daughters in high-level waste from reprocessing are shown in figure 1; the toxicity computation is based on the hazard from ingestion. Such a measure of toxicity takes no account of pathways to the biosphere and to man (Pigford and Choi, 1976) but does help to pose the question of how long a time must be considered in geologic predictions to assure safe containment of waste. The critical feature of figure 1 is the very slow drop-off in total potential toxicity during the time indicated. As Gera (1975, p. 14) stated, "The variation in potential hazard from 100,000 to 10 million years is so limited and slow that it is virtually impossible to make a rational case for any specific length of required containment falling within that time interval. Either containment failure can be considered acceptable after a period on the order of 100,000 years, or containment must be assured for periods of time exceeding at least 5 million years." Gera (1975, p. 14) considered that assuring containment for longer than 5 million years is "clearly impracticable since totally reliable geologic

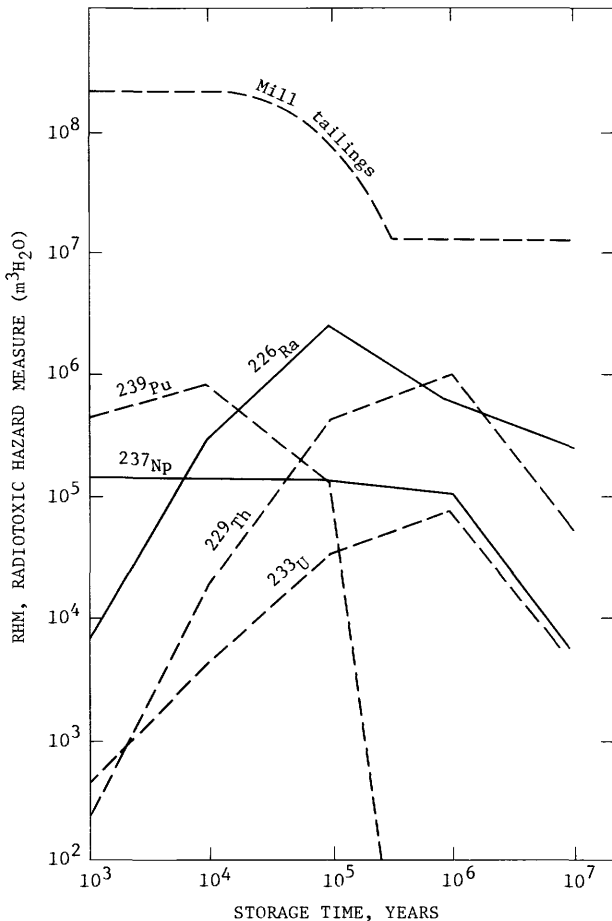


FIGURE 1.—Ingestion hazard of selected radionuclides in high-level waste during 10 million years. The radiotoxic hazard measure is obtained by dividing the number of curies present for a given nuclide by the number of curies produced by the maximum permissible concentration of that nuclide in a cubic meter of drinking water. Data are normalized for 1 metric ton of light-water reactor fuel. The nuclide curves are plotted from data in table 2 of Hamstra (1975); the curve for uranium mill tailings was derived from figure 2 of Hamstra (1975) and figure 7 of Pigford and Choi (1976).

predictions of the detail required over such long time frames are beyond present capability.”

Figure 1 also calls attention to the hazard from uranium mill tailings, which, although more than an order of magnitude more toxic than high-level waste, have customarily been treated in much more cursory fashion. Whether the hazards from high-level waste and uranium ore bodies and their derivative tailings are

comparable is not clear. The high-level wastes are in highly concentrated forms. This means that a breach of a repository and a direct short circuit to the biosphere could have serious consequences. On the other hand, the relatively low volumes of high-level waste should help to insure that it can be more easily handled and placed in a more secure geologic environment than the more voluminous and dispersed ore and mill tailings (Pigford and Choi, 1976).

GEOLOGIC PREDICTION

Although the hazards from geologically isolated radioactive wastes during long periods of time are difficult to specify exactly, it is pertinent to ask, “How credible are geologic predictions ranging from 1,000 to 10 million years into the future?”

We have not had, of course, any working experience with geologic containment for such long periods; thus, predictions must be based on conceptual models of what will happen to the waste after emplacement. Such models exist, and their complexity and sophistication have been increasing steadily in recent years (for example, see Logan and Berbaro, 1977; Burkholder and others, 1977; Tierney, 1977). In theory, the models provide a compendium of all possible events and processes likely to influence release of radioactivity from a repository to the biosphere and eventually to man. Connected with each event or process is an estimate of its future probability or rate. Potential interactions among the events and processes are also identified, and estimated functional relationships are incorporated into the models. The starting point for the models is a set of initial conditions, including emplaced radioactivity, rock geometry, and rock properties around the repository and along potential flow paths. The end point for the models is a set of risks usually expressed in radiation exposures to humans for a series of times after final disposal.

Application of these models to policy making in the field of radioactive-waste management requires that their limitations be clearly understood. Models of natural systems that have come into use in recent years fall into four categories (Holcomb Research Inst., 1976): (1) simple and predictable, such as agricultural

crop patterns; (2) complex and predictable, such as river hydrology or short-term weather patterns; (3) simple and unpredictable, such as ecosystem response to natural disasters; and (4) highly complex and unpredictable, such as interrelations among the species of an ecosystem or, we believe, the fate of radioactive wastes in geologic repositories. The unpredictability of radioactive-waste models stems from the lack of a method for determining the future rates of many events and processes, such as tectonism, and from the current lack of adequate data needed to allow the model to function from start to finish—for example, the data needed to characterize ground-water-flow systems.

Past geologic events such as faulting, seismicity, or climate change probably have not been random, but deterministic explanations for their frequency, place of occurrence, magnitude, and rate of change are difficult to establish. Regardless of whether deterministic or probabilistic models are favored to explain particular past geologic events, use of the geologic record to predict future events is a formidable task. Certainly, an assumption of the constancy of rates of geologic processes, namely substantive uniformitarianism,⁴ is open to question. The past rates of occurrence of geologic events and processes have varied widely over time and there appears to be no clear philosophical basis for determining rates for these events or processes in the future. Geology is basically a retrodictive rather than a predictive science (Kitts, 1976). A related problem is that the observational record of the past, on which estimates of the rates of occurrence of geologic events are made, is invariably incomplete (Ager, 1973). The seismic record for most of the United States, for example, extends back only about 200 years. For some parts of the stable interior, clusters of relatively few low-level events suggest that larger events have a finite probability of occurrence, but estimates of this probability must be assigned relatively high errors (S. T. Algermissen, oral commun., 1977). These considerations are not meant to suggest that the geologic record of the past can be ignored in

⁴ Substantive uniformitarianism contrasts with methodologic uniformitarianism that asserts merely the spatial and temporal invariance of the natural laws such as gravity. These concepts were discussed extensively by Gould (1965a, b), Hubbert (1967), Albritton (1967), Hooykaas (1963), and Simpson (1963, 1970).

siting radioactive-waste repositories, but only that estimates of probabilities of future events be recognized for what they are—estimates only. Although many of the current gaps in needed data and theory will be filled eventually, Burkholder and others (1977) noted that many processes probably can never be modeled precisely. For example, the exact form the waste will have thousands of years after disposal probably cannot be determined with certainty from either laboratory experiments or theory.

Of particular concern in assessing the uncertainty of geologic prediction is the effect of complex interactions among events and processes whose individual effects may be simple. Natural events such as earthquakes may affect a backfilled repository containing fluids very differently from the way in which they affect undisturbed rock; seismicity may itself be induced by fluid pressures. Potential interactions such as these must be analyzed in any systems analysis approach to waste isolation. However, unanticipated interactions have taken place in many engineering efforts whose components were presumably well characterized; the Apollo 13 near-tragic explosion is a conspicuous example (Cooper, 1973). Simpson (1963, 1970), Scriven (1959), and Mayr (1961) argued that long-term prediction in the biological and earth sciences is unreliable and impossible to perform with high confidence limits because of the great complexity of possible interactions among processes, both identified and unidentified.

Although validating a waste-management model for the timespans of concern will never be possible, we can ask the pragmatic question, "How good has the track record been in predicting the geologic response to man's disturbances for a shorter time, say 100 years?" Briefly, predictions in "routine" engineering geologic endeavors ranging from soil consolidation to tunnel geology have varied from good, in particularly favorable circumstances, to poor in some others where data seemed adequate and reliable (Lambe, 1973; Dowding and Miller, 1975). Two major difficulties revealed in studies of incorrect predictions have been the geologic definition of the medium and the choice of boundary conditions, rather than the mathematical modeling. The record may be even worse than it appears, for as Lambe (1973)

pointed out, many published and apparently successful predictions were made after the fact; he cautioned against overreliance on such predictions as opposed to those documented before the events took place. The interested reader is referred to the highly pertinent papers by Lambe (1973), Dowding and Miller (1975), Rowe (1975), and Meigh (1976) for details.

In summary, predictive models are an essential step in the selection and implementation of a radioactive-waste repository and a radioactive-waste management system. They are invaluable tools for analyzing the problem and for identifying factors that are likely to have the greatest effect on radionuclide migration. However, some components of the models are inherently unpredictable at present and are likely to change at different times. In no sense, therefore, will these models give a single answer to the question of the fate of radioactive waste in geologic repositories; rather they will provide a spectrum of alternative outcomes, each based on a set of uncertain assumptions about the future. Decision makers outside the earth sciences will have to evaluate these uncertain predictions in the light of pressing social and economic concerns.

RESEARCH NEEDS AND CONCLUDING STATEMENT

This Circular has dealt largely with the difficulties and uncertainties connected with the geologic disposal of high-level radioactive waste because, from our viewpoint, these are significant potential stumbling blocks that need critical attention. In emphasizing these problems, we do not intend to slight the extensive effort currently going forward to find safe repositories. Significant progress is being made. We offer the following suggestions for research efforts and emphasis in the hope that they will prove constructive.

First, the many questions concerning the behavior of rock salt must be resolved—questions centering on its high solubility. If relatively small amounts of brine can cause substantial decrease of mechanical strength and possible movement of waste during a relatively short time, special efforts will surely be necessary to insure retrievability from a salt repository

for periods as short as 10–25 years. The question of whether the workings of a mine in salt can be predicted to stay dry will have to be faced.

Second, systematic examination of media other than salt should continue. Some of the advantages of shale, crystalline rocks, and zeolitic tuff were mentioned earlier. The suggestion of Winograd (1974) that waste be placed at relatively shallow depths (30 to several hundred meters) in the thick (as thick as 600 m) unsaturated zones of the arid Western United States deserves consideration. The uncertainties connected with all these media are greatly reduced if the media are used, at least initially, only for relatively cool waste (surface temperatures $<100^{\circ}\text{C}$).

Third, the complex task of characterizing ground-water transport systems around potential repositories should continue, and this task should be recognized as costly and time consuming. Empirical data on flow through fractured media and on the detailed chemistry of sorption-desorption phenomena for the actinide elements need to be collected and used in conceptual models.

Fourth, more tools to evaluate potential repositories should be aggressively developed. Although it may seem unduly cautious to call for still more technology and more types of data, we see the need to devise methods of dating old (greater than 40,000 years) ground waters and of exploring the volume of rocks around the repository as amply justified by the seriousness of an inadvertent breach of a repository.

Fifth, more research emphasis is needed on the relatively short and long term effects of the repository structure and the waste on the environment around the repository. Such research should address the question of the extent to which the repository itself can localize escape of radionuclides to the environment.

A concluding statement concerns the uncertainties involved in geologic prediction for long timespans, discussed earlier. These uncertainties need to be faced candidly in public discussions of radioactive-waste disposal. Earth scientists can indicate which sites have been relatively stable in the geologic past, but they cannot guarantee future stability. Construction

of a repository and emplacement of waste will initiate complex processes that cannot, at present, be predicted with certainty. The inability to predict can be offset in part by adoption of a multiple-barrier or "defense-in-depth" philosophy for radionuclide containment. Such a philosophy provides a succession of independent barriers to nuclide migration. The waste form, the host rock, and the ground-water flow path all provide potential barriers. Continuing research is needed to measure the efficacy of these barriers and to obtain a better understanding of the processes involved.

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