## Geologic Disposal of Nuclear Wastes: Salt's Lead Is Challenged

## From salt beds to the seabed, most proposed geologic media are still very much in the running

Highly radioactive, long-lived nuclear wastes have been piling up at storage sites around the world since the dawn of the Atomic Age. In 1957, a committee of the National Academy of Sciences and the National Research Council concluded that the most promising means of protecting the world from these wastes is to bury them in natural beds of salt.

The promise of salt as a disposal medium has yet to be fulfilled. More than 20 years after the NAS-NRC report, a technical subgroup of the President's Interagency Review Group on Nuclear Waste Management concluded that "Although most is known about the engineering aspects of a repository in salt, on purely technical grounds no particular geologic environment is an obvious preferred choice at this time." \* Among the other geologic environments still being seriously considered are granites, basalts, shales, tuffs, and the sediments beneath the deep ocean floor.

Although wastes already exist that must be dealt with, public acceptance of any future nuclear power program is also at stake. Scientists are acutely aware that an extremely strong case must be made for any proposed disposal method if waste management is not to be a stumbling block. A false step of any sort, through haste, poor judgment, or simply bad luck, would have extremely serious consequences.

Some options have fallen by the wayside. Proposals to let the wastes' own heat melt them into the Antarctic or Greenland ice caps have been dropped. The political problems appear to be considerable and the stability of the ice caps over the 100,000 years or more required for the radioactive decay of the wastes is highly uncertain. Shooting them into the sun or into orbit around the sun has been put on the back burner because of high costs and the less-than-perfect reliability of launch vehicles.

Researchers are looking for geologic media that can prevent the wastes from

wandering away from where they are placed and that will behave in the distant future pretty much as they have in the past. Beds of salt seemed promising because they have a number of unusual but desirable characterisitics that could help contain wastes. One is that salt has no cracks in it that would allow the flow of water or brine, the most likely means of escape for wastes from a disposal site. Salt, which is actually an off-white translucent rock, flows under pressure. Thus, fractures should seal themselves. Unfractured salt has been shown in the laboratory to be among the most impermeable rocks in nature. In addition, the NAS-NRC report argued, the persistence of salt deposits for 200 million years or more demonstrates their isolation from circulating groundwater. Within the last few years, however, investigators have shown that another unusual characteristic of salt, its high solubility, has allowed groundwater to penetrate and alter salt beds in ways that are difficult to detect from the surface and are not yet fully understood. Also, pockets of brine can be left behind as evaporation of seawater forms the salt beds.

Some of this complex salt geology has become obvious since the selection of a site for the Waste Isolation Pilot Project (WIPP). WIPP is intended to demonstrate the practical disposal of solid wastes contaminated at low levels by the long-lived transuranic elements, such as plutonium. It may also be used for the disposal of spent fuel.

The initial search for the WIPP site ended in 1975 with the selection of a site for intensive evaluation in the Delaware Basin in southeastern New Mexico. This site was near an ancient reef that encircles the salt beds, which are themselves sandwiched between two water-bearing rock formations. Unexpectedly, drillers encountered complex distortion of the geologic structure, and they also struck a pocket of brine pressurized by hydrogen sulfide, a highly toxic gas, and by methane, which is explosive.

Abandoning this initial site as geologically unsuitable, researchers from Sandia Laboratories, who picked up the project during the drilling phase, selected the present site, which is about 10 kilometers farther from the reef. Evaluation of this site is based on extensive drilling and seismic reflection profiling, the acoustic technique used by oil companies to remotely detect deep geologic structures. Even so, if a brine pocket similar to the one found earlier were located at the new WIPP site, it might or might not be detectable, according to Leslie Hill of Sandia. The problem, common to studies of most geologic media, is that remote sensing techniques are still not sensitive enough to detect smallscale but significant changes in rock properties below the surface. On the other hand, attempts at direct detection by extensive drilling could threaten the integrity of any repository. The presence of a brine pocket not connected to any system of flowing water would not eliminate an area from consideration, but it would complicate construction of the repository.

Strong evidence that large portions of the salt beds in the Delaware Basin have been dissolved and carried away also limited the area suitable for a site. Because the salt beds are tipped downward to the east, water has entered the exposed ends of the beds in the west. Dissolution has also been caused by water entering on the east from the reef. Roger Anderson of the University of New Mexico estimates that 50 percent of the salt originally deposited by evaporation of the reef-enclosed sea has been removed by this dissolution. It is advancing in a wedge-shaped front that should not reach the site for some millions of years, according to Wendell Weart of Sandia. but Anderson has cautioned that the rate and mechanism of deep dissolution are not vet firmly established.

Another limitation on site location is the presence of boreholes drilled since the early part of the century in search of mineral deposits. A plethora of such holes around a study site near Lyons, Kansas, contributed to its abandonment in 1971. In the Delaware Basin, drilling was less intensive and more accurately recorded than at the Kansas site, so that

0036-8075/79/0511-0603\$00.50/0 Copyright © 1979 AAAS

<sup>\*</sup>Subgroup Report on Alternative Technology Strategies for the Isolation of Nuclear Wastes, TID-28818 (draft), October 1978; available from NTIS, U.S. Department of Commerce, Springfield, Virginia 22161 (\$9.50 printed copy, \$3.00 microfiche).



Salt

a 1-mile buffer zone around each hole is considered sufficient protection from possible infiltration of water.

A potential problem involving salt dissolution that is generally recognized as unresolved is the presence of breccia pipes or collapse chimneys in the Delaware Basin. These are columns of rubble 100 to 500 meters in diameter that extend vertically thousands of meters through the salt beds, sometimes even breaking the surface and forming a depression. They seem to result from the collapse of rock strata above a chamber that has been eaten out of the salt by flowing water. How the water entered the supposedly impermeable salt is not clear.

As a possible explanation, Anderson and Douglas Kirkland of Mobil Research and Development Corporation in Dallas have proposed that fresh water in the underlying Bell Canyon aquifer first penetrates cracks in a normally impermeable layer of anhydrite separating the aquifer and salt. This water forms a small amount of dense brine. The difference in density between the brine and the fresh water then drives a convective circulation in which the lighter fresh water rises through the crack, dissolves the salt, and sinks back through the same crack as the heavier brine. Anderson and Kirkland have demonstrated this brine flow in a laboratory model. Even if this is the

cause of collapse chimneys, Anderson notes, it is still not known whether the process continues today or where it might occur in the future. It is also not certain whether collapse chimneys will allow water to pass through them and possibly breach a repository.

Cross section of a

Water already known to be dispersed throughout the salt at the WIPP site has recently raised an additional concern. Microscopic pockets of concentrated brine, remnants of the seawater that evaporated to form the salt beds, bring the water content of the salt up to 0.5 percent. It has already been shown that the heat from the radioactive decay of the wastes could burst the brine inclusions and cause the released brine to migrate toward the waste containers. The heat would also promote reactions of magnesium chloride in the brine that would sharply increase the brine's acidity. Exactly how much brine might eventually collect around the waste is uncertain. Estimates range from a few liters to several tens of liters.

If brine migration could actually affect containment of wastes, through corrosion of the waste container or weakening of the salt formation, a number of technical solutions have been suggested. Simply reducing the maximum temperature in the repository would prevent the release of the brine in the first place. This could be done by letting the wastes cool more before emplacement, putting less waste in each canister, or spacing them farther apart. The canisters, glassy cylinders of waste sealed in a stainless steel jacket, could also be given greater protection from the brine. A likely protective shield is an alloy of 99 percent titanium. Jeffery Braithwaite of Sandia believes, on the basis of laboratory tests and reported alloy properties, that about 0.6 centimeter of titanium alloy would probably protect the wastes throughout the several hundred years of high heat output even if the initial temperature were as high as 250°C.

The Department of Energy is seeking other possible disposal sites in bedded salt, but salt in domes along the Gulf Coast is also being evaluated as a possible disposal medium. This salt was laid down in horizontal beds too, but its lower density caused it to form numerous ascending fingers of salt that pierced the denser overlying sedimentary rock. The domes immediately inland of the coast have obviously continued to rise during geologically recent time, but several of the hundreds of domes farther inland that may have stabilized have been chosen for further study. Joseph Martinez of Louisiana State University believes that nothing in his work or that of others on the Gulf Coast domes would rule them out as disposal sites, but a great deal remains to be learned, he says. If anything, he points out, the geology of salt domes is more complicated than that of salt beds because dome salt was modified both when it was in bedded form and when it was shaped into domes.

Expectations of extensive, undisturbed beds of dry salt may not have been realized but many researchers believe that the technical questions concerning salt will be resolved at least as promptly as those concerning other media, if not sooner. With its edge in engineering, salt may still be the first geologic environment selected for a repository. But other geologic environments have been under study all along, and their accelerated evaluation recently received a boost from the report of President Carter's Interagency Review Group. Nonsalt rocks under consideration include granites and basalts, which cooled in place from molten rock; shales, which are muds turned into rock by high temperature and pressure; and tuffs, which are volcanic ash solidified by its own heat

Although none of these types of rocks can be considered a typical nonsalt medium, they do have some general properties that can be contrasted with those of

SCIENCE, VOL. 204



Areas now under study by the seabed disposal program. Desirable characteristics for a repository beneath the sea floor include a water depth of 4000 meters or more, the presence of about 60 meters of red clay, and a continuous geologic record of a stable sediment environment. [Source: Charles Hollister, Woods Hole Oceanographic Institution]

salt. Unlike salt, all of these rocks are usually fractured to some degree because of contraction during cooling. If the cracks are connected to one another, water can pass through the rock. But these rocks also have the ability to chemically adsorb most waste elements. Thus, if groundwater leached wastes from a repository and then moved toward the surface, adsorption by the rock could delay the arrival of the wastes where they might do harm. Salt's adsorptivity is negligible. Slowing of waste dispersion by this means would occur only if and when the contaminated brine entered a nonsalt aquifer.

This ability to retard the dispersion of radioactive elements in spite of water movement has been demonstrated in an actual geologic setting by a group at Los Alamos Scientific Laboratory under the direction of William Daniels. The radioactivity used in the experiment resulted from a 0.75-kiloton nuclear test in 1965 294 meters below the surface at the Nevada Test Site. The rock there is a loose alluvium washed down from the surrounding tuffs. Many of the radioactive elements were trapped in glassy particles formed during the test, but some radioactivity dissolved in the water that filled the cavity following the test. In 1975 water began to be pumped from a borehole 91 meters from the cavity at a rate much higher than natural groundwater flows in the area.

Radioactive hydrogen-3 (tritium) in water molecules arrived at the pump after about 1.4 million cubic meters had been pumped. About 2.6 million cubic meters have now been pumped, but only tritium and krypton-85, a chemically inert gas, have been detected. The remainder of the radioactivity is apparently being held up by the tuff. According to Daniels, laboratory measurements suggest that more than 1500 years would be required for most elements to travel the distance already covered by the tritium.

The adsorptivity of a rock such as tuff can be measured in the laboratory by suspending particles of it in a solution containing the element. The ratio of the element adsorbed by the rock to that remaining in solution is called the distribution coefficient. The higher an element's distribution coefficient, the more effectively it is retained. Generally speaking, an element with a coefficient of 100 will take 100 times longer to travel a given distance through rock than the water that it is dissolved in.

Darleane Hoffman, Kurt Wolfsberg, and their colleagues at Los Alamos have determined distribution coefficients for tuff. They range from near zero to 100,000 depending on the element, the initial solution concentration, and the condition of the particular sample used. For example, strontium's distribution coefficient ranged from 50 to 14,000 and cesium's from 150 to 33,000. These are generally higher than those for granite or basalt and about equal to those for shales. Prediction of containment times for nuclear waste on the basis of such coefficients can be complicated further by other factors. Natural organic compounds may interfere by inhibiting adsorption. Also, an element may take on another form, such as insoluble colloidal particles, which are not adsorbed readily.

Prediction of containment times in nonsalt media is further hindered by the variability of the structure of rock formations. A case in point is the tuffs of southern Nevada, which are the subject of a nuclear waste disposal feasibility study being conducted by Allen Lappin of Sandia and Bruce Crowe of Los Alamos. They note that both the rocks beneath the tuffs and the tuffs themselves have faulted and been displaced, confusing efforts to trace the likely path of water once it has left a hypothetical disposal site. In addition, the hydrologic properties of tuffs may vary with depth; whether a particular layer of tuff is resistant to water flow or freely conducts it depends on how extensively the original ash particles became welded together and how much fracturing occurred.

All of these factors, plus the amount of rainfall, determine how soon groundwater passing through a repository might reach the surface. How soon contamination would reach the surface depends on the mineral composition of the tuffs, which can also vary considerably along a single flow path. Fresh tuff is often altered when fluids percolate through the rock. Altered tuff has a particularly strong attraction for transuranic elements, while fresher tuffs readily adsorb many of the other elements in wastes. Lappin notes that all of this geologic complexity and mineralogical variability would make the characterization of a prospective site more difficult, but it

could strengthen containment by providing a number of different physical and chemical barriers. These difficulties of site characterization are shared to some degree by all types of nonsalt rocks.

Another group of researchers is looking below the bottom of the ocean as a possibly less complicated, more predictable place to dispose of nuclear wastes. Innumerable drums of low-level wastes have been dropped onto the sea bottom, but since 1974 a group managed by Richard Anderson of Sandia is evaluating the feasibility of inserting high-level wastes beneath the seabed. In that way, the sediment would be the geologic barrier, not the ocean above it.

The vastness of the ocean has often been considered to be enough of a barrier, but workers in the seabed disposal program decided early that a location thousands of kilometers from land and beneath 5 kilometers of water was not isolated enough. For example, their first study area was about 1000 kilometers north of Hawaii. No known natural resources such as economically recoverable manganese nodules or fisheries exist there, so disturbance by humans would be unlikely, but natural routes of escape from the area may exist. Small, intermittent currents were measured near the bottom that could conceivably carry any released wastes across the Pacific in 1000 years or less. Another possible route is through the animals of the deep sea. Oceanographers have discovered over the last few years that the animals on and near the deep sea bottom can be quite mobile. Instead of being at the dead end of a one-way flow of food and debris from the surface, bottom animals may be linked to the surface through overlapping habitats of predators and prey. In spite of the possible dilution involved in these pathways, researchers turned to the geological formations beneath the seabed for a more reliable barrier.

In contrast to the geologic formations on land, the red clay deposits found in the center of large ocean basins appear to be relatively tranquil and uniform, according to Charles Hollister of Woods Hole Oceanographic Institution and Ross Heath of Oregon State University. They examined core samples of sediment about 10 meters long that were collected from the study area north of Hawaii. They found that the clay particles making up this sediment had been settling to the bottom evenly and without interruption or erosion for at least 10 million years. One particularly long core of sediment from the center of the surveyed area had recorded a similarly quiet environment during the past 70 million years.

The few characteristics, such as thin layers of airborne volcanic ash or subtle color changes, that can be used to distinguish one part of a core from another are very predictable, too. Cores taken 100 kilometers or more apart show the same patterns of these markings. Such uniformity, Hollister and Heath point out, would allow considerable geologic characterization of a particular site without disturbing it.

Red clay, which is actually various shades of chocolate brown, has a number of physical and chemical properties that would help to contain nuclear wastes for long periods. Like salt, it flows under pressure, only much more readily. Although there is no reason for natural cracks to form, clay's plasticity would tend to seal in waste canisters after emplacement. One means of emplacement being considered is the precisely navigated free fall of a long, bulletshaped waste canister that would penetrate about 30 meters into the bottom. Laboratory experiments by Armand Silva of the University of Rhode Island indicate that the clay might flow in behind the canister as it penetrated the bottom and immediately close up the hole. In any event, Silva says, natural closure would occur relatively quickly.

Although red clay has measurable permeability, water does not naturally flow through these sediments. Silva has found that the permeability of a number of red clays approaches that of moderately fractured granite (about 0.01 millidarcy), whether it was measured on an intact sample or one that had been severely distorted during collection. But there are no significant forces in undisturbed sediment to drive the water through the clay; diffusion would be the fastest type of migration. Heath has calculated that a waste element would require about 100,000 years to move by diffusion alone through 30 meters of sediment to the surface.

This travel time would be greatly increased by the high adsorptivity of red clays. Egberg Duursma of the International Atomic Energy Agency in Monaco, Kenneth Erickson of Sandia, and Heath have found that most distribution coefficients for red clay have high values at low waste concentrations; for example, 100 to 6000 for strontium-90 and 1400 for cesium. These are similar to the highest values for terrestrial materials. If an element's distribution coefficient were only 10, it would not escape from the sediment for a million years.

A major question that remains to be answered is the possible effects of the waste's heat and radiation on sediment properties. Calculations by Paul Dawson of Sandia suggest that if the only effect of the heat is to start convective circulation, the canister would stay pretty much where it initially came to rest. James Krumhansl and Dave McVey of Sandia have calculated that such convection would carry any wastes leached from the canister only about 2 meters before most of the radiogenic heat died away. But the material surrounding the canister might be changed into a viscous fluid by the high temperature, invalidating these predictions. The effects of heat and radiation on other properties of red clays, such as adsorptivity, are also as yet unknown. Corrosion problems in the seawater-clay medium would be similar to those anticipated in a salt repository.

The engineering problems involved in emplacing and, if necessary, retrieving nuclear wastes from the deep seabed will not be studied until 1983, when environmental aspects will have been evaluated, but researchers are cautiously optimistic. They point to the exploits of the Glomar Explorer, which apparently lifted a sunken Soviet submarine off the deepsea floor, and the Glomar Challenger, which has employed increasingly sophisticated technology to retrieve long cores from the deep sea.

Political obstacles to disposal of nuclear wastes in international territory are more uncertain. At the moment, no provisions of the Law of the Sea documents would bar such activity, and deep-sea disposal is considered to be a serious option for Japan and Great Britain, which have limited numbers of possible sites. Some observers argue that the regions being considered are of no real economic value to anyone and thus may escape the tortuous consideration being given such proposals as manganese nodule mining. In any case, they say, the difficulties could hardly be much greater than locating a disposal site in a U.S. congressman's district.

Researchers are feeling considerable pressure to develop data to support strong or even overwhelming arguments in favor of geologic disposal of nuclear wastes. Most believe that safe disposal is doable, probably in several different media. But significant questions remain concerning each geologic environment being considered.-RICHARD A. KERR

## Additional Reading

- J. D. Bredehoeft, A. W. England, D. B. Stewart, N. J. Trask, I. J. Winograd, Geological Disposal of High-Level Radioactive Wastes-Earth-Science Perspectives (U.S. Geological Survey Circular 779; available at no charge from Branch of Distribution, U.S. Geological Survey, 1200 South Eads Street, Arlington, Va. 22202).
  "High-level nuclear wastes in the seabed?" Oceanus 20 (No. 1), (winter 1977).