

HAZARDS OF MANAGING AND DISPOSING OF NUCLEAR WASTE

Although it is arguably for the benefit of society, the translation of basic nuclear processes into technological achievements produces nuclear wastes that can become environmental hazards if they are not properly cared for. Light-element fusion, heavy-element fission and radioactive decay have provided us with nuclear weapons, nuclear power plants and nuclear medicine. But they all produce radioactive materials that are unusable, no longer needed, or unwanted. Ultimately, these materials require long-term isolation from the biosphere.

Of particular interest, with regard to hazards in the US, are the 30 000 tons of spent fuel rods from commercial nuclear power plants and the 400 000 cubic meters of "high-level" radioactive wastes left over from the production of plutonium and highly enriched uranium (HEU) for nuclear weapons and naval propulsion reactors. The fuel rods from the power reactors have remained at the plants in "spent fuel pools" or, more recently, "dry cask storage." The latter are large concrete and steel containers on concrete pads. The wastes from plutonium and HEU production are, for the most part, stored in large tanks at government sites throughout the Department of Energy's weapons complex.

All these storage arrangements are temporary. For the most part they await decisions regarding "final" disposition in repositories such as Yucca Mountain in Nevada and the Waste Isolation Pilot Plant (WIPP) in New Mexico. But even after these wastes come to be in a state of final disposition, there will still be hazards and risks.

Hazards and risks

The Federal government distinguishes three forms of wastes.¹ (Also see the glossary on page 23.):

High-level waste. Most of the high-level waste in the US has come either from spent nuclear fuel removed from commercial power and weapons-production reactors or from the reprocessing of such spent fuel to extract additional fissile weapons material.

Transuranic waste. This kind of waste comes primarily from the processing and reprocessing of plutonium and highly enriched uranium. Transuranic wastes generate

When we bury long-lived nuclear wastes in geologic repositories, we have to worry about what may happen ten thousand—or even a million—years in the future.

William E. Kastenberg and Luca J. Gratton

less heat than do fission products. Furthermore, because they emit mostly alpha particles, they require little or no shielding. Such material is referred to as contact-handled waste. But about 3% of the transuranic waste does emit penetrating gamma radiation and therefore requires shielding.

That small fraction is called remote-handled waste. Transuranic waste includes contaminated materials such as protective clothing and glove boxes.

Low-level waste. The Nuclear Regulatory Commission classifies low-level waste into four groups, depending on the degree of hazard it poses, and hence the type and form of disposal it requires: Waste that can be disposed of by shallow land burial is classified (from least to most hazardous) as A, B or C. This classification scheme determines the required type of packaging and form of burial. Low-level waste too hazardous to be buried in a near-surface facility (called "greater than class C") requires disposal in a geologic repository.

The classification of radioactive waste listed above is based on hazard—that is to say, in terms of the quantity and type of radioactivity it emits. But the risk posed by any particular waste must also take account of the potential for exposure. For radioactive waste, there are two broad classes of risk assessments. For the first class, we might ask the questions: What can go wrong? How likely is it to go wrong? What are the consequences? These questions are typical of assessments that quantify the risk of system failure. The second class considers the potential exposure of humans and other "ecological receptors" if one assumes that some event, for instance waste-tank leakage, has already occurred or is bound to occur over long periods of time as a result of natural degradation and geological transport.

Both assessment classes involve uncertainties in the data and models used to quantify potential exposure and risk. There are, for example, two different kinds of performance assessment for a geologic repository. The first is so-called undisturbed performance, which evaluates past and present geological processes to predict future conditions that may lead to degradation of the waste package and transport of radionuclides into the groundwater. Such predictions are inherently uncertain over the time scales under consideration.

The second kind of assessment deals with so-called disturbed performance—that is to say, it considers the possibility of human intrusion and rare natural events such as earthquakes and volcanic eruptions. Predictions

WILLIAM KASTENBERG is a professor of nuclear engineering at the University of California, Berkeley. LUCA GRATTON is a graduate student in the university's nuclear engineering department.

of societal behavior thousands of years in the future may be of some help in repository design, but it is, perforce, highly speculative. Such uncertainties reappear throughout this article.

From a risk perspective, we focus on high-level and transuranic wastes in their current states of storage and their proposed future disposal in geologic repositories. Low-level waste, with its own sociopolitical set of constraints, is discussed in the article by Warner North on page 48. With regard to hazard and risk, the environmental and health impacts of low-level waste disposal are expected to be comparatively small. By contrast, the hazards and risks posed by the high-level waste stored at the various DOE weapons-complex sites can be regarded as very large. The waste tanks at the Hanford facility will serve us as a good example.

Storage of high-level waste

To produce weapons-grade plutonium, one irradiates uranium metal in so-called production reactors. In the US, from 1943 to 1988, this process was carried out primarily at the Hanford site in Richland, Washington. The irradiated uranium metal was cooled and treated in a chemical separation plant. The chemical separations generated several hundred thousand tons of wastes—transuranic, high- and low-level and “mixed,” a designation for waste that is both radioactively and chemically hazardous.

Over the past 50 years or so, a number of strategies have been employed to store the waste safely, pending a long-term or permanent solution. Such strategies included storage in single-shell tanks and then in double-shell tanks, adding chemicals such as sodium hydroxide or calcium carbonate to make the acidic waste alkaline, and removal of cesium and strontium to lower the heat load. In spite of these efforts, several major problems persist: Among other difficulties, some tanks are leaking and some have the potential to produce a chemical explosion.

The 149 single-shell tanks at Hanford, with capacities ranging from 2×10^5 to 4×10^6 liters, had design lives of about 20 years. Some leakage from these tanks was already suspected in 1956. By the late 1980s, 67 of them were known or suspected leakers, and an estimated 4 million liters of high-level waste had been released to the soil.

Chemicals had been added to the tanks to settle radionuclides from the liquid waste to the bottom. The upper liquid waste was then siphoned off and sent to shallow surface drainfields, where it percolated into the soil. No leaks have been known to occur from the double-shell tanks. At least 12 different contaminants have been identified in the groundwater beneath the tank storage site, including arsenic, chromium, cyanide, carbon tetrachloride, cobalt-60, strontium-90, technetium-99, iodine-129, cesium-137, tritium, and plutonium-239 and 240. The groundwater occurs at depths of 70 to 90 meters below the surface. It has been estimated that about 1 million curies have been released or leaked to the ground, and an additional 5 million curies have been disposed of in solid-waste burial grounds. These losses are from a total inventory of 210 million curies in the existing tanks.

In dealing with the waste tanks, one has to consider potential short- and long-term impacts. Among the short-term health issues are occupational radiological and chemical accidents, occupational radiological exposure during routine operation, and radiological transportation accidents on- and off-site. The kind of accident with the most severe potential health impact would be a sudden, energetic hydrogen gas combustion in a waste tank. At least 25 tanks at Hanford are currently estimated to be generating hydrogen gas (from ongoing chemical and radiolytic reactions) in sufficient quantities to cause an

energetic fire if it were somehow ignited. DOE is monitoring these tanks and ventilating them to allow the hydrogen to escape. In the tank generating the most hydrogen, a mixer pump has been installed.

The *Final Environmental Impact Statement for the Tank Waste Remediation System*² contains calculations indicating that, in the event of a large combustion at Hanford, one might expect as many as 22 latent cancer fatalities (20 site workers and 2 off-site members of the public) from direct radiation and inhalation of radioactive contaminants. This conclusion is based on a conservative set of assumptions, including worst meteorological conditions, maximally exposed individuals, and no evacuation or other interdiction measures.

The longer the wastes remain in the tanks, the higher is the probability that an energetic hydrogen gas fire will occur. The estimated probability is 72% if the wastes remain in the tanks for 100 years—the duration assumed for both the no-action and long-term-management alternatives in the environmental impact statement. This calculation yields an expectation value of about 0.2 cancer fatalities per year. That's quite high in comparison with the risk estimated for commercial nuclear power plants. It should be noted that the so-called long-term-management scenario, which requires only that the waste be transferred twice per century into new double-shell tanks, does not fare significantly better in these calculations than its no-action alternative over the course of 100 years.

The primary long-term impacts (from 100 years to 100 centuries) are considered to be groundwater contamination and land use restriction, and the potential health effects associated with contaminated groundwater, accidents and intruders. Movement of contaminants into groundwater is a long-term process. It depends on the nature of each contaminant, its processing into a more stable form, and the type of engineering barriers that have been put in place—for example, caps to prevent or limit intrusion of rainwater.

Long-term groundwater contamination (in this case, from releases during retrieval of tank waste, releases from what's left in the tanks after remediation, and releases from waste vaults) depends on the remediation process one chooses. In the absence of any action beyond transfer to new double-shell tanks twice per century, the fastest-moving contaminants at Hanford are expected to reach the groundwater in about 130 years and rise to maximum concentration at 210 years. It would then take another 20 to 50 years for these contaminants to reach the Columbia River. More ambitious remediation alternatives, such as vitrification *in situ*, yield transport times ranging from one to five millennia.

The Hanford final environmental impact statement lays out the risk to various potential users of the land at different times, up to 10 000 years, in the future. The long term risk of contracting cancer would be high for the no-action and long-term-management scenarios, ranging from 1% for recreational users to almost 100% for hypothetical “Native American” users living off hunting, fishing, gathering and subsistence farming.

Vitrification *in situ* reduces these estimated risks at least a thousandfold. The integrated risk, which depends on population distribution, was calculated to be in the range of 60 to 3000 excess cancer deaths over 10 000 years.

Spent fuel

A considerable accumulation of high-level waste exists in the form of spent commercial fuel temporarily stored in pools of water at nuclear power plants. To the extent that such pools meet all Nuclear Regulatory Commission requirements, they pose relatively little risk when compared



CORRODED PLUTONIUM-BEARING FUEL TARGETS from a Savannah River reactor formerly used to make plutonium-239 for weapons. These targets, 4 inches in diameter and 14 inches long, have since been converted to a stable metal form for long-term storage.

to the risk posed by DOE's high-level-waste tanks. Considerable efforts have gone into assessing the safety of the power-plant pools or dry-cask storage facilities. These assessments consider what can happen in the event of loss of cooling or other mishaps due to human error or external events like earthquakes.

The major concern with regard to spent fuel is, once again, focused on the DOE weapons complex. Good examples are the so-called K Basins at Hanford, in which almost 7500 canisters containing more than 2000 tons of spent uranium fuel are stored. A first priority is to expeditiously move the fuel away from the Columbia River, because some of the spent-fuel cladding was damaged during reactor discharge and handling. Moreover, the fuel was not intended for long-term wet storage; so it continues to degrade slowly. Continued storage in the K Basins presents obvious risks, but so does removal and transport to a new storage facility. Even the new facility would be a temporary measure. Final disposition would therefore involve additional risk some time in the future.

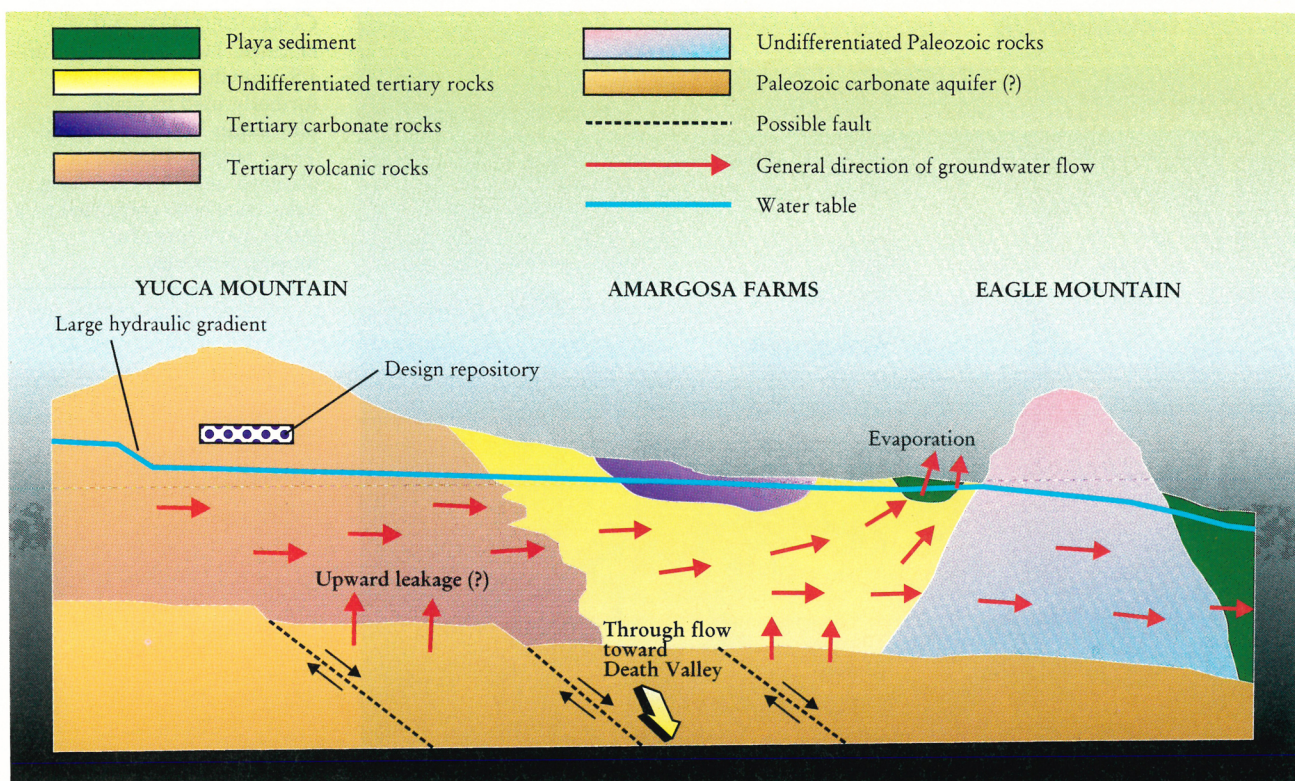
At present, geologic disposal appears to be the most attractive option for the ultimate disposition of spent fuel and high-level waste from both commercial power reactors and weapons-production reactors, as well as for the transuranic wastes from weapons activities over the last half-century. Geologic disposal for these waste categories is preferred for economic, political and technical reasons. The economic aspects of nuclear waste disposal are beyond the scope of this article. Some of the political issues are discussed in the companion article by Warner North, starting on page 48. The technical issues raised by the geologic disposal of transuranic and high-level wastes and

spent nuclear fuel have been more thoroughly researched, in the US and internationally, than any alternative disposal option. These studies give us an extensive, albeit untested, technical basis for the design and assessment of repositories. The progress already realized for both WIPP, which is designed for transuranic wastes, and the proposed Yucca Mountain high-level-waste repository, attests to the seriousness of the US in its pursuit of a geologic disposal option. This pursuit may provide internationally useful benchmarks against which future repository designs and performance criteria could be compared.

Risks of geologic deposition

Performance assessment provides figure-of-merit estimates for various geologic repository designs. The wastes must, of course, be adequately contained in the proposed geologic setting for a very long time. Conceptual repository models serve to yield the requisite long-term performance predictions. The assessment process employs extensive mathematical modeling that incorporates our most up-to-date theoretical and empirical understanding of the physical and chemical behavior of the waste constituents whose escape from the repository is to be prevented.

The performance of a repository depends on numerous variables describing both the waste form and the geologic environment over long time periods. The already daunting assessment process is further complicated by the need to consider infrequent or unknowable future events. Because these uncertainties grow with time and space, the modeling generally requires a stochastic treatment. The potential hazards of a high-level or transuranic waste



IDEALIZED GEOHYDROLOGIC CROSS SECTION of the vicinity of the proposed high-level-waste repository at Yucca Mountain, in a semiarid region in southwestern Nevada. The repository would be excavated out of volcanic tuff about 300 meters below the Yucca crest. (Adapted from ref. 6)

repository are typically described in terms of scenarios that might allow passage of degraded waste materials into the environment. The most pertinent scenarios for WIPP and Yucca Mountain differ from each other, primarily because of the sites' differences in geological conditions and waste classifications.

Waste Isolation Pilot Plant

The WIPP site is 650 meters underground within a salt formation in the Pecos Valley of New Mexico, 27 miles from the city of Carlsbad. Preliminary stages for transuranic-repository licensing by the Environmental Protection Agency are currently being sought. The presence of the large salt formation indicates that the region has been geologically stable and somewhat isolated from circulating groundwater for relatively long geologic periods. Furthermore, salt has desirable mechanical characteristics, such as its tendency to heal itself after fracture. By the time it's filled and closed, WIPP is projected to have accepted a combined inventory of 6 million cubic feet of contact- and remote-handled transuranic waste. The total activity of the remote-handled waste component will not exceed 5.1 million curies.

"Disturbed performance" scenarios dominate the potential-hazard analyses for the WIPP site. Of the so-called disturbed scenarios, human intrusion is the only significant potential contributor to waste releases identified in the WIPP Compliance Certification Application³ submitted to EPA last October. Tectonic, magmatic and criticality events were eliminated from further consideration in the assessment process.

The application for certification indicates that the WIPP site should satisfy EPA requirements even when breached by multiple boreholes in the distant future. It

was assumed for these purposes that future generations would drill roughly one borehole per square mile per century. To demonstrate safety margins, it was even assumed that a borehole with a degraded plug would intersect groundwater upstream of a well. Someone drinking two liters a day from that well, it was calculated, would still absorb less than 3% of the allowed EPA standard of 15 millirem per year.

A report⁴ compiled for the State of New Mexico suggests that criticality cannot occur at WIPP, provided the plutonium content of individual waste drums does not exceed a prescribed limit. Two scenarios that might undermine this conclusion involve very severe compression of the waste drums or long-term dissolution, transport and reconcentration of the fissile materials. But both scenarios are considered extremely unlikely, given the design and anticipated emplacement of the waste canisters and our understanding of the local hydrogeology.

A review⁵ of an early draft of the certification application states that the certification should expand its consideration of plausible repository-breach scenarios to show compliance with EPA requirements intended to give assurance—beyond the usual numerical predictions—of the repository's ability to provide long-term waste isolation. The review points out that the region is known to have oil, natural gas and potash reserves that might entice problematic mining activities in the distant future.

Yucca Mountain scenarios

The proposed Yucca Mountain high-level-waste site is a semiarid region in southwestern Nevada. Current plans call for the repository facility to be excavated from volcanic tuff at a depth of about 300 meters beneath Yucca Crest. That would still be 300 meters above the local water table.

The repository zone is unsaturated, which means that liquid water and gases can coexist naturally in the fractures and pore spaces of the volcanic tuff. When filled, the repository is projected to house 70 000 tons of spent nuclear fuel and 8000 tons of high-level military waste.

A recent assessment⁶ of the Yucca Mountain design provided undisturbed-performance estimates for times ranging from ten thousand to a million years after the repository is sealed. The scenario took account of the various natural processes that are expected to degrade the waste packages over these very long periods. The study also included consideration of a number of engineered barrier systems that could serve as isolation regions between the waste canisters and outlying tuff, and thus significantly extend the duration of waste containment.

The modeling results for the Yucca Mountain repository's undisturbed performance indicate that it easily satisfies EPA's 10 000-year dose limits under various difficult circumstances. Radiation doses were calculated for individuals drinking two liters a day of groundwater from the peak concentration in a contaminant plume, at a well 5 km downstream of the repository boundary. It should be pointed out, however, that the vast majority of the Yucca Mountain simulations resulted in no radionuclide releases at all to the environment.

The study found that performance assessment a full million years into the future depends sensitively on details of the repository design and the modeling parameters. On that time scale, high water infiltration rates lead primarily to neptunium-237 releases, while lower rainwater percolation leads to technetium-99 and iodine-137 releases. These are all highly water-soluble species that do not sorb significantly to rock. Increasing the infiltration rate a hundredfold increases the mean dose in local drinking water almost a millionfold. The study found similar sensitivity to changes in the flow speed of the aquifer.

Incorporating gravel backfill, a capillary barrier and extra "sacrificial" corrosion layers on the waste canisters into the design was shown to be capable of reducing the peak dose at the accessible environment by eight orders of magnitude. Risk also tended to decrease rapidly with increasing distance of the well from the repository.

An earlier performance assessment study⁷ assessing the feasibility of waste disposal at Yucca Mountain incorporated disturbed-performance scenarios arising from human intrusion and criticality events. The intrusion events were assumed to result from exploratory drilling at the rate of a few boreholes per square kilometer in ten thousand years. The results tentatively met EPA standards, though there were some calculated excursions above the long-term regulatory limits.

Concern over the possibility of a criticality event if minimally treated weapons materials are deposited at Yucca Mountain has stimulated an ongoing and vigorous debate in the scientific community. The issue received widespread attention following a controversial report⁸ that an "autocatalytic criticality event" might occur in a repository and do significant harm. Subsequent research by us and colleagues at Berkeley⁹ and others at Sandia⁷ led us to conclude that the potential for criticality at a repository like Yucca Mountain, containing minimally treated weapons wastes, is unlikely for plutonium but cannot be definitively ruled out for uranium. Plutonium-239 has a half-life of 24 900 years—moderate by actinide standards. It is relatively insoluble in water and therefore is not apt to travel large distances. It might be transported most efficiently as a colloid. Uranium-235, on the other hand, has a half-life of 713 million years, and it is more soluble in water. But it is not expected to readily precipitate out of solution in the geochemical environment of Yucca Mountain.

If a criticality event involving repository wastes should occur, its actual consequences and the resulting hazards are also a subject of debate. Estimates range from a "no consequences" scenario with negligible energy generation¹⁰ and marginal increases in temperature and fission products,⁷ to significant enhancement⁸ of the potential for radionuclide releases. Other investigators view the criticality controversy as somewhat irrelevant.¹¹ They point out that robust repository design and adequate preconditioning of waste can render the probability of a criticality event negligible. This view is credibly supported by various studies,^{7,9} but it also warrants more detailed investigation.

Adequacy of the safety regulations

Preliminary performance assessments for the proposed Yucca Mountain^{6,7,12} and WIPP^{3,13} sites support the conclusion that both repositories would perform adequately over the 10 000-year period under consideration. That is to say, they would satisfy the existing containment dose requirements. There is, however, some uncertainty about the regulatory criteria to which Yucca Mountain would be held accountable. In fact, current law requires the promulgation of new regulatory standards specific to Yucca Mountain. But it is unclear what specific criteria the new standards will embody. Will they be based solely on release probabilities, on risk to humans¹⁴ or on some combination of the two? It is also uncertain whether the regulatory period will be made longer than the current EPA requirement of 10 000 years. Two assessments for Yucca Mountain have found that doses to individuals drinking groundwater between 100 000 and a million years in the future will exceed the doses at 10 000 years by orders of magnitude.^{6,7} The potential for environmental and human exposures to radionuclides beyond the current 10 000-year regulatory period has led the National Academy of Sciences to endorse the adoption of much longer time horizons.¹⁴

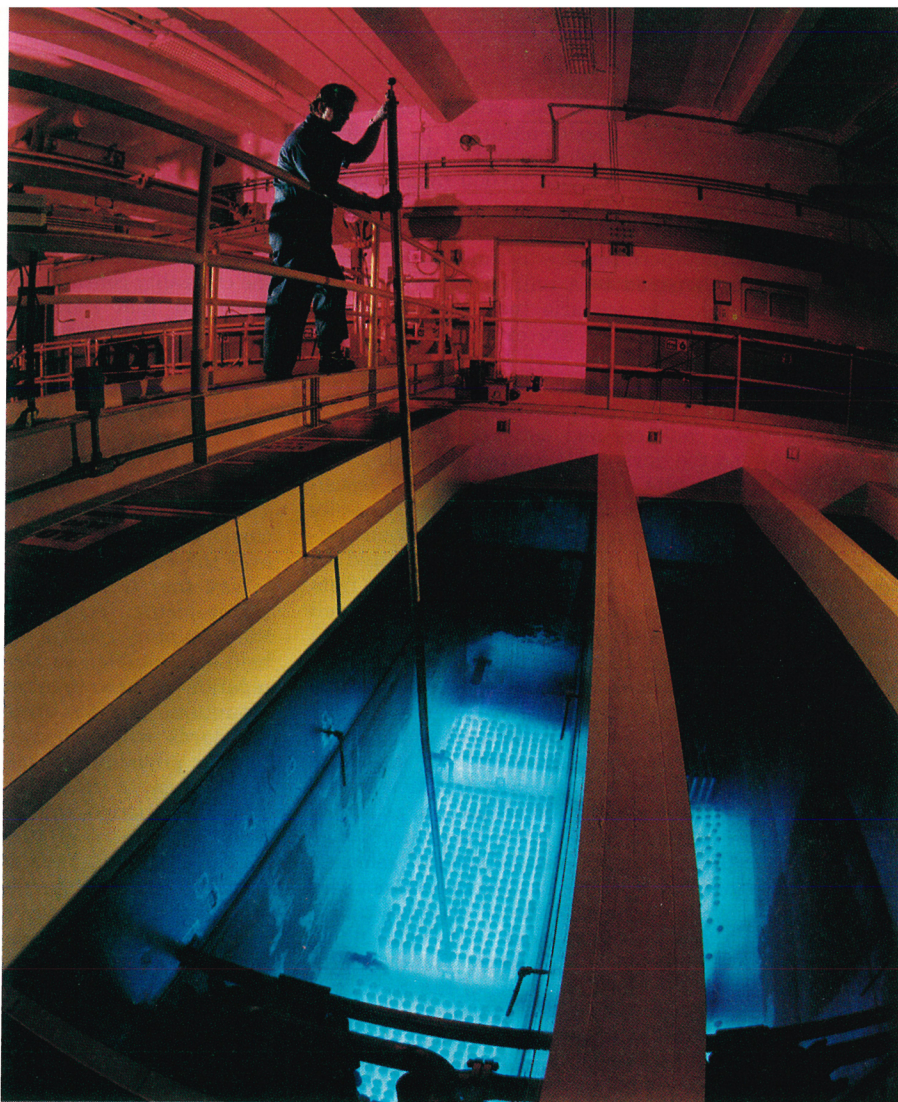
The preliminary performance assessments for high-level and transuranic waste disposal at Yucca Mountain and WIPP indicate that the proposed sites are likely to comply with existing regulatory requirements and that they pose few hazards. Nevertheless, the uncertainties in the quantitative models and in our understanding of the underlying physical, chemical, geologic and societal issues warrant a degree of cautious prudence as we commit wastes to any long-term geologic repository. We should exercise appropriate caution until the scientific and technical community can further reduce the uncertainties, or until it is adequately demonstrated that the repository design is robust enough to accommodate the present level of uncertainty. The fact that we don't yet know what standards Yucca Mountain will ultimately be required to satisfy adds to the uncertainty.

Some of these quantitative uncertainties may ultimately resist further reduction. But we believe they can be accommodated by robust repository design, albeit at greater financial cost. Engineered design features may, for example, drastically offset our uncertainties as to the criticality issue or the danger of exploratory drilling in the distant future.

This brief survey of issues associated with the disposal of high-level and transuranic wastes has touched on a spectrum of hazards and risks. It seems likely that the highest risks, those associated with the high-level waste tanks at the DOE weapons complex sites, will be reduced, albeit with great difficulty and at considerable cost. Provisional approaches to the DOE tanks and the accumulation of spent fuel at commercial nuclear power plants leave a legacy of waste awaiting ultimate disposal. Some of

A WATER POOL at the Hanford weapons complex site's Waste Encapsulation and Storage Facility (WESF) stores and cools capsules containing cesium-137 and strontium-90, two high-heat isotopes left over from plutonium production.

With the imminent decommissioning of the original Manhattan Project spent fuel processing plant adjacent to WESF, the facility is to be upgraded to stand alone for another 10–15 years while long-term storage of the hot cesium and strontium capsules is being planned.



these wastes have radioactive lifetimes measured in millions of years. The uncertainties inherent in predicting risks over such time spans challenge our ability to protect future generations.

The disposal of transuranic waste at WIPP, if it remains undisturbed, is predicted to pose no risks to neighbors for at least 10 000 years. But perhaps we need to examine the disturbance scenarios with greater scrutiny and, in either case, to look well beyond 10 000 years.

For a high-level-waste repository of the type proposed for Yucca Mountain, it is clear that natural processes will eventually redistribute the waste materials. Present design efforts are directed toward ensuring that, at worst, the degraded waste configurations will eventually resemble stable, natural ore deposits, preferably for periods exceeding the lifetimes of the more hazardous radionuclides. Perhaps that's the best we can hope for.

References

1. League of Women Voters Education Fund, *The Nuclear Waste Primer*, Lyons & Burford, New York (1993).
2. US Department of Energy and Washington State Department of Ecology, *Final Environmental Impact Statement for the Tank Waste Remediation System*, Washington DC (1996).
3. US Department of Energy, "The Waste Isolation Pilot Plant: Compliance Certification Application," submitted to EPA, October 1996.
4. S. C. Cohen, Supplement 2 in *Review of the WIPP Draft Application to Show Compliance with EPA Transuranic Waste Disposal Standards*, Environmental Evaluation Group of the State of New Mexico, DOE/AL/58309-61, Albuquerque, NM (1996).
5. R. Neill, L. Chaturvedi, W. Lee, T. Clemo, M. Silva, J. Kenney, W. Bartlett, B. Walker, *Review of the WIPP Draft Application to Show Compliance with EPA Transuranic Waste Disposal Standards*, Environmental Evaluation Group of the State of New Mexico, DOE/AL/58309-61, Albuquerque, NM (1996).
6. TRW, *Total System Performance Assessment—1995: An Evaluation of the Potential Yucca Mountain Repository*, Civilian Radioactive Waste Management System, B000000000-01717-2200-00136, Revision 01 (1995).
7. R. Rechard, ed., Sandia National Laboratories report SAND94-2563/1 (1995).
8. C. Bowman, F. Venneri, Los Alamos National Laboratory report LA-UR-94-4022A (1995).
9. W. Kastenberger, P. Peterson, J. Ahn, J. Burch, G. Casher, P. Chambre, E. Greenspan, D. Olander, J. Vujic, B. Bessinger, N. Cook, F. Doyle, L. Hilbert, Nucl. Technology **115**, 298 (1996).
10. W. Myers, W. Stratton, R. Kimpland, R. Sanchez, R. Anderson, Transact. Am. Nucl. Soc. and Eur. Nucl. Soc. **75**, 214 (1996).
11. C. G. Whipple, Scientific American, June 1996, p. 72.
12. M. L. Wilson *et al.*, Sandia National Laboratories report SAND93-2675 (1994).
13. R. P. Rechard, Sandia National Laboratories report SAND93-1378 (1995).
14. National Research Council, *Technical Bases for Yucca Mountain Standards*, National Academy Press, Washington, DC (1995). ■