From a potpourri of political and scientific options emerges the first step likely to be taken to package high-level radioactive waste for disposal

THE BOX WITHIN A BOX WITHIN A BOX

BY LINDA GARMON

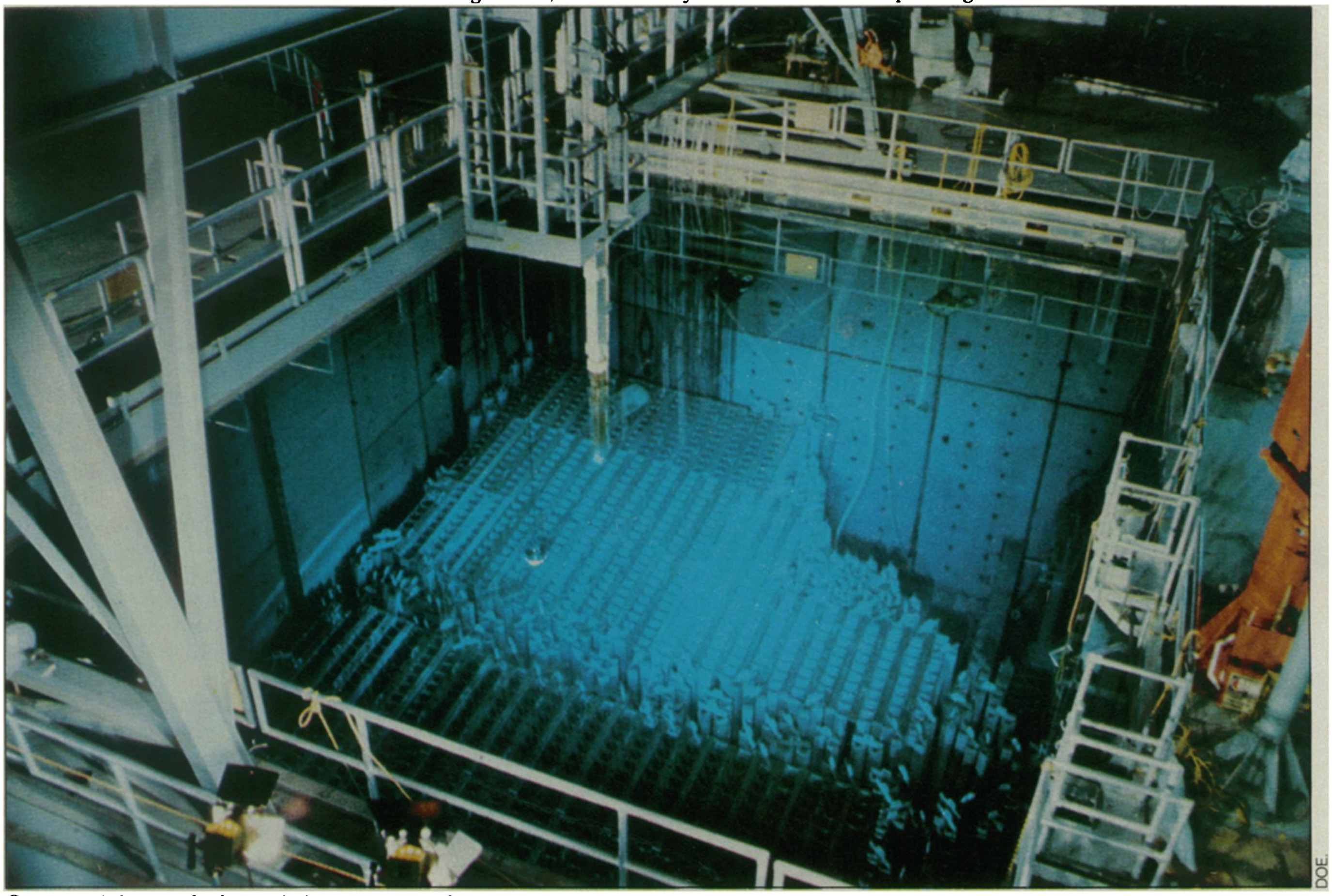
First of a three-part series on high-level nuclear waste management.

The magician taps the box with a mystical wand and assures that the volunteer's missing watch will be inside. But upon opening the first good-sized box, the volunteer discovers a second, smaller box. Likewise, opening box number two reveals only a third box, and so on until the missing article is retrieved. The nest of boxes leaves the impression that the trickster somehow overcame multiple barriers.

On stage, the multiple barrier effect is, of course, an illusion. In the scientific arena, however, a diverse group of researchers and policy makers are enlisting the aid of the real-world correlate of the nest of boxes to tackle one of the toughest problems of the nuclear age — the "disposal," or isolation from the biosphere, of high-level radioactive wastes. Under the multiple barrier plan, such wastes would be immobilized in a solid form, then would be placed in a canister, which in turn would be encompassed by a series of additional barriers — the ultimate one being a mined repository.

While Congress mulls various pieces of legislation regarding these repositories and the U.S. Department of Energy (DOE) plans exploratory shafts at selected geologic sites, much activity remains focused on the smallest in the nest of boxes — the initial barrier, or the form used to immobilize the high-level waste. For instance, DOE recently capped years of intensely competitive research when it decided to consider solidifying certain liquid military wastes into either a glass or a ceramic. At the same time, a long-standing controversy over what to do with civilian (commercial) radwaste has been rekindled. The initial waste form may be only one part of the package deal, but it now is a hot item on the agenda for the disposal of the high-level waste that has been accumulating in the United States for nearly 35 years.

That waste results from commercial power generation and from the extraction



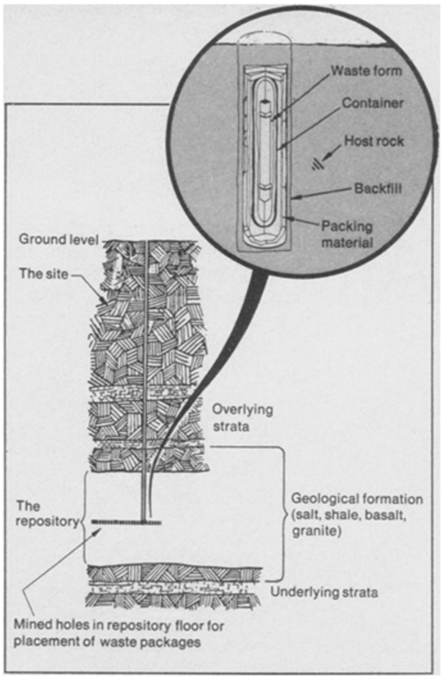
Commercial spent fuel now is in storage pools at reactor sites such as the Dresden Nuclear Power Station west of Chicago.

of uranium and plutonium isotopes for military weapons manufacture. In both instances, the process begins with reactor fuel composed of uranium oxide pellets sealed in metal tubes. During a fission-type nuclear reaction, the uranium atoms in this fuel assembly release heat as they split into the fission products — new and lighter atoms such as krypton, strontium, ruthenium, iodine and cesium. In addition to the fission products — which constitute only about 5 percent of the used fuel — the waste contains residual uranium and heavier elements, such as plutonium, produced in the fuel when uranium atoms capture neutrons. Some of the waste is stable (nonradioactive), but other components — dubbed "radionuclides" — emit hazardous bursts of energy in the form of alpha (positively charged particles), beta (high-speed electrons) and gamma (high-energy electromagnetic energy waves) radiation. Certain of these radionuclides continue to decay for thousands of years. Plutonium 239, for example, has a 24,000-year half-life — the time it takes for one-half of the atoms to decay. And therein lies the reason why nuclear waste is no ordinary trash: It radiates, and it radiates for a long time.

The brunt of the responsibility for isolating that radiation from the biosphere falls on DOE. In the past year or so, the department has come to favor the multi-barrier mined-repository strategy over the more exotic proposed radwaste disposal schemes — burial in polar ice caps and storage on remote islands or in outer space, for example. Mostly by default, aging radwaste by temporary storage has become the first step in this strategy. Used commercial fuel assemblies now lie in water-filled pools adjacent to power plants, and the mostly liquid defense wastes sit in double-walled steel tanks at three government facilities and at one commercial site in West Valley, N.Y.

But time is running out on temporary storage. Not only have commercial pools of used fuel assemblies been plagued with overcrowding, but also "some defense high-level wastes have been stored for 35 years, approaching and sometimes exceeding the life expectancy of their storage tanks," according to a recent General Accounting Office (GAO) report. "More importantly, some tanks have cracked and leaked significant amounts of radioactive materials into the ground," the report goes on to state. As a result, DOE has stepped up its efforts in dealing with the 8,000 tons of used fuel assemblies and the 75 million gallons of defense wastes it now has on its hands.

Twenty-two million of those defense waste gallons are from a land of sandy soil and pine trees. These wastes — generated by the E.I. du Pont de Nemours & Company-operated Savannah River Plant in Aiken, S.C. — are first in line to be treated. They are DOE's radioactive guinea pigs, and precisely how to treat them has been



IEEE Spectrum

The proposed radwaste packaging system involves several barriers: the waste form, a canister (container), a mineral filling (backfill) and the host rock.

the subject of much recent discussion.

There are a lot of different ways to treat a waste. You can make it into a glass. You can incorporate it into concrete. You can even make ceramic pellets out of it and then cast those pellets in metal. In each case, the objective is the same: to immobilize the waste with a barrier by making it a part of that barrier. In 1979, DOE recognized about 11 *potential* ways of achieving this effect (refer to the chart). Recently, with the Savannah River Plant wastes in mind, the department decided to take a closer look at what it believes are the two most effective forms: borosilicate glass and a crystalline ceramic called SYNROC.

Borosilicate glass is formed by melting together nuclear waste components and glass-forming additives such as boron oxide and silicon oxide. SYNROC, made famous by A. E. Ringwood and colleagues of the Australian National University in Canberra, is a ceramic material whose three minerals — zirconolite ($CaZrTi_2O_7$), hollandite ($BaAl_2Ti_6O_{16}$) and pervoskite ($CaTiO_3$) — accept waste components into their crystal lattices. DOE's recent decision to eventually convert Savannah River Plant waste into one of these forms did not take the radwaste community by surprise; inklings of such a resolution were evident after a meeting in Atlanta, Ga., last spring of both the developers of various waste forms and a peer review panel.

Researchers in the field have come to refer to that meeting as the "Atlanta shoot-out," a term they borrowed from the headline of a local newspaper article about the event. At the shoot-out, waste form researchers presented their latest data before the eight independent scientists of the peer review panel, chaired by materials science professor Larry L. Hench of the University of Florida at Gainesville. When the confrontation was over, the panel chose borosilicate glass and SYNROC as the most promising waste forms.

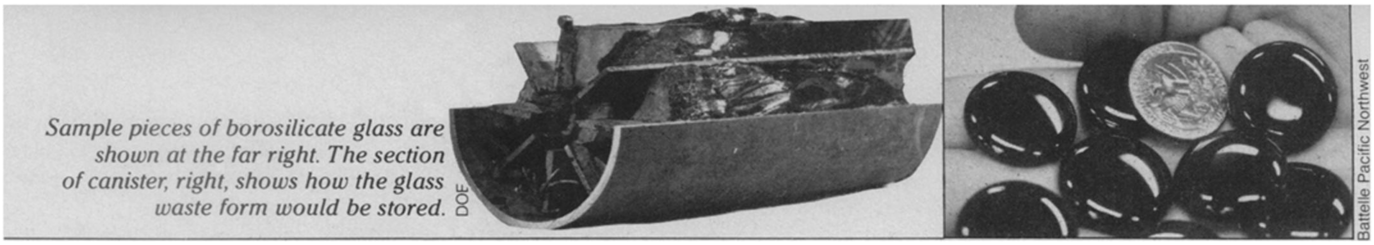
And thus began the show-down between borosilicate glass and SYNROC. Or did it? Most waste-form researchers already feel borosilicate glass is the shoo-in; some go so far as to say that SYNROC was listed merely for political reasons. "I think they [the panel] believe SYNROC — the idea of a synthetic rock — has a big PR value," said one waste-form developer. "My own personal opinion," he continued, "is that glass is quite acceptable. . . I think the thing that we need to worry about now — that we have not put enough thought into — is the actual [waste-treatment] plant. We want to put in a process whereby the people who have to work there eight hours a day get the minimum exposure. If we engineer a complicated plant — and I think SYNROC would require one — we're going to endanger these people."

Glass also earns extra points for being tolerant of a wide variety of waste forms. "Defense wastes are not uniform," explains Stephen V. Topp of the Savannah River Laboratory. "A lot of different fuel has gone into those reactors over the years," he says, and an immobilizing form that is insensitive to the resulting variations in defense wastes is desired. This should be a factor when deciding between glass and SYNROC, says Topp, who chaired the International Symposium on the Scientific Basis For Nuclear Waste Management at the Materials Research Society meeting last month in Boston. "Ceramics must be precisely tailored to the precise waste form, and that would vary from tank to tank [of wastes in temporary storage]," says Topp. "You'd like to make a waste form that would take a wide variety of junk," he says. "That's why glass shines."

Nonetheless, the show-down continues — at least officially — with the final DOE choice between borosilicate glass and SYNROC due by October 1983. DOE plans then call for the first high-level waste immobilization facility to be in operation at the Savannah River Plant by 1988. Treatment of the high-level waste at the Hanford (Richland, Wash.) and Idaho Fall, Idaho, defense sites will be dealt with thereafter.

But what's to become of the nuclear waste from the private sector? At least for now, neither glass nor SYNROC can solve this problem, because most commercial waste is not in a state that can be converted into these waste forms. Most commercial waste is in the form of spent-fuel rods — the used uranium fuel that has been removed from a nuclear reactor. But the fuel isn't really completely "spent," and technically, it is not even considered a waste: 95 percent of it is re-cyclable uranium.

The high-level defense wastes, on the other hand, result from reprocessing military spent fuel. At reprocessing plants, the



Sample pieces of borosilicate glass are shown at the far right. The section of canister, right, shows how the glass waste form would be stored. DOE

Battelle Pacific Northwest

THE CANDIDATES

For the past several years, DOE has funded research on these major alternative waste forms. Recently, the department decided to eventually convert Savannah River Plant waste into either borosilicate glass or SYNROC.

WASTE FORM	DESCRIPTION	DEVELOPER/ CONTRACTOR	POTENTIAL ADVANTAGE	POTENTIAL DISADVANTAGE
Borosilicate Glass	Waste and glass formers (frit) are melted together.	Savannah River Laboratory, Battelle Pacific Northwest	Best developed Relatively simple process	Hydrothermal reactions possible Glass melter required
High-Silica Glass	The extra silica (SiO_2) is added to increase the durability of glass.	Catholic University of America	Very low leachability	Hydrothermal reactions possible
FUETAP Concrete	Concrete monoliths, composed of waste and additives, are Formed Under Elevated Temperatures (150°C) And Pressure (100 psi).	Oak Ridge National Laboratory	Relatively simple process	Relatively high leachability Poor long-term stability Low waste loading
Hot-Pressed Concrete	Dense concrete monoliths are formed by hot-pressing waste and additives at 150°C to 250°C and 25,000 psi to 50,000 psi.	Pennsylvania State University	Good long-term stability	Relatively high leachability Large hot-press required
Supercalcine Ceramic/ Tailored Ceramics	Waste components are incorporated into various types of crystals. Supercalcine crystals are silica (SiO_2)-based; tailored ones are alumina (Al_2O_3)-based.	Pennsylvania State University, Rockwell International	Low leachability Excellent long-term stability High waste loading	Complex process Hot isostatic press may be required
SYNROC Ceramic	Waste components are incorporated into various types of crystals that all contain TiO_2.	Australian National University, Lawrence Livermore National Laboratory, N.C. State University, Argonne National Laboratory	Low leachability Excellent long-term stability High waste loading	Complex process Hot isostatic press may be required
Cermet (Urea Process)	Small waste-plus-additive crystallites are mixed with metal and then pressed.	Oak Ridge National Laboratory	Ease of quality assurance	Long-term metal corrosion Very complex process Large amounts of urea and off-gas
Glass Marbles in Metal Matrix	Waste-plus-frit marbles are loaded into a canister, and the void space is filled with metal.	Battelle Pacific Northwest Laboratory	Well developed Two barriers	Low-melting metal required Large surface exposed if matrix fails Glass melter required
Ceramic Pellets in Metal Matrix	Waste-plus-additive crystalline ceramic pellets are surrounded by a metal matrix.	Battelle Pacific Northwest Laboratory	Two barriers	Complex process Large surface exposed if matrix fails
Coated Ceramic	The waste-plus-additive ceramic is coated with materials such as pyrolitic carbon, which is basically graphite.	Battelle Pacific Northwest Laboratory	Multiple barriers Very low leachability Excellent long-term stability	Very complex process Coating technology required
Coated Ceramic via Sol-Gel	Spherical waste-plus-additive ceramic particles are coated with materials such as pyrolitic carbon.	Oak Ridge National Laboratory	Multiple barriers Very low leachability Excellent long-term stability No dry powders in process	Conceptual product only Complex process Sol-Gel and coating technology required

After "Features of Alternative Waste Forms" from "Preliminary Evaluation of Alternative Forms For Immobilization Of Savannah River Plant High-Level Waste" by J. A. Stone, et al.

fuel is separated into streams of uranium, plutonium and high-level waste. The uranium and plutonium are solidified and converted into either fresh reactor fuel or nuclear weapon material; the high-level waste — which contains fission products and chemical solvents used to dissolve the spent fuel — remains in a liquid state, awaiting immobilization into a waste form.

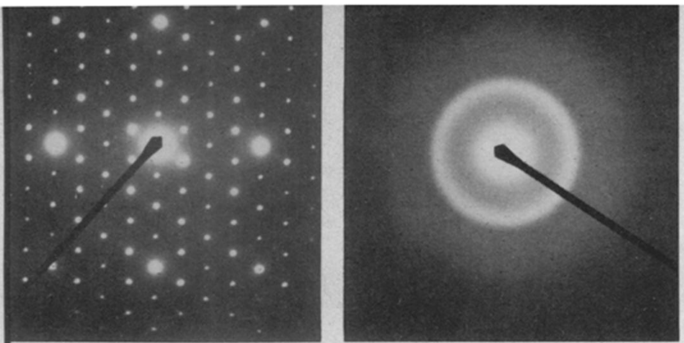
Reprocessing commercial spent fuel in this fashion was banned by Presidents Gerald Ford and Jimmy Carter, who feared the plutonium produced could be stolen and used to make nuclear weapons. President Ronald Reagan lifted that ban Oct. 8.

Still, "The private sector is saying, 'You've got to be kidding — the risks are too high,'" says Colin A. Heath, former director of DOE's Office of Waste Isolation. Because the future of U.S. nuclear power in the United States is uncertain, and because succeeding administrations could easily reverse current policy, investors fear pouring money into commercial reprocessing plants, Heath explains. Indeed, despite DOE's current search for methods to encourage commercial reprocessing, it now is viewed as the long-shot in civilian nuclear waste disposal.

The treatment processes that *are* likely for commercial waste range from simply packaging the intact spent fuel assembly to chopping it, dissolving the resulting exposed fuel in acid and converting the solution to a glass. DOE now is testing how stainless-steel encapsulated fuel assemblies fare in granite formations at the Nevada Test Site in southern Nevada and in a basalt out-cropping at Hanford.

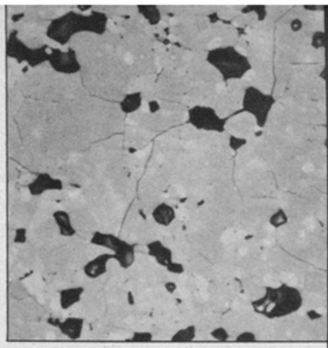
The GAO sees several drawbacks to this method. In its recent report — "Is Spent Fuel Or Waste From Reprocessed Spent Fuel Simpler to Dispose Of?" — GAO notes that spent fuel disposal only defers the nuclear proliferation threat to future generations who may exhume and reprocess the fuel. In addition, the disposal method would require several times the repository space of high-level waste forms.

Finally, GAO reports, "DOE's contractors and scientists believe that a waste package [all of the multiple barriers, save the repository] can be designed that will completely contain either spent fuel or



T. J. Headley, et al./Sandia National Laboratories, Univ. of N.M.

Self-damage caused ceramic waste forms to crack, right, and to go from a structured, far left, to an amorphous state, left. Photos at left are electron diffraction patterns.



Clinard, et al./Los Alamos National Laboratory

WASTE-FORM RADIATION: SELF-INFLICTED WOUNDS

If nuclear waste forms such as borosilicate glass and SYNROC are to be places where radioactive components check in but don't check out, then those forms must resist leaching in ground water. Because the damage these waste forms suffer from their own radioactive components can affect leach resistance, this self-injury is the subject of several current research endeavors. Thus far, while various effects are turning up in all of those studies, none paint a clear picture of radiation self-damage.

Clyde M. Northrup Jr. is shooting lead ions into simulated waste forms to study that phenomenon. Northrup and colleagues of Sandia National Laboratories in Albuquerque, N.M., use the implanted ions to mimic the effects of alpha-recoil nuclei. The heavy atoms — thorium, for example — in nuclear waste forms emit alpha particles. At the same time, due to conservation of momentum, the atoms' remaining nuclei recoil. The phenomenon can be likened to firing a shotgun: The bullet (alpha particle) speeds off in one direction, while the shotgun (recoil nucleus) kicks back in the other direction. Ninety percent of the radiation damage in waste forms, says Northrup, is associated not with emitted particles, but rather with the alpha-recoil nuclei. Northrup is studying such damage in borosilicate glass and ceramics similar to SYNROC. He presented preliminary results of the ongoing research at the meeting in Boston last month of the Materials Research Society (MRS).

Although the data are not yet quantified, they do suggest that lead ion implantation enhances leaching of glass and ceramics, Northrup reported. In addition, he said, results suggest that subtle changes in the borosilicate glass recipe cause "considerable" changes in that form's ability to resist leaching. "That's interesting," says Northrup, "because most of the people making these glasses are saying all borosilicate glasses are alike."

While Northrup's research technique may flag the superior formulation of a given waste form, whether it accurately simulates a waste form's self-damage is the subject of debate. One possible strike against it, Northrup explains, is that the implanted ions always travel in the same direction, causing a "preferential mechanical strain" that does not accurately reflect the havoc that alpha-recoils wreak.

In related research also presented at the MRS meeting, Frank W. Clinard and colleagues of Los Alamos National Laboratory in New Mexico doped samples of the zirconolite ($CaZrTi_2O_7$) component of SYNROC with plutonium 238. "We zeroed in on this particular phase because we think it is the most susceptible [of the three SYNROC crystallites] to radiation damage," Clinard says. He and co-workers stored the sample phase until alpha decay self-damage accumulated to a dose of 6×10^{24} particles per cubic meter, equivalent to a 1,000-year-old SYNROC. Extensive microcracking was observed. Because cracking exposes more waste form surface, it could enhance leachability; again, however, the results are preliminary and only serve to "point out an area that needs to be further investigated."

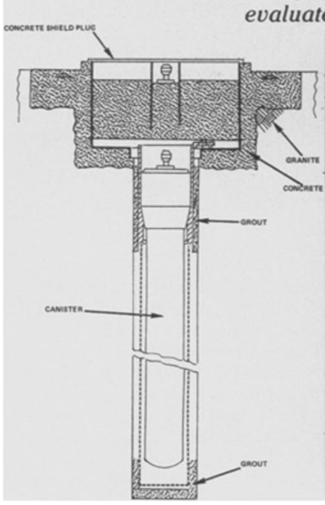
Finally, other researchers are turning to nature for answers. T. J. Headley of Sandia National Laboratories and Rodney C. Ewing and Richard F. Haaker of the University of New Mexico at Albuquerque are studying metamict minerals—a class of naturally occurring minerals that contain radioactive species such as thorium and uranium. "Metamict minerals provide a natural example of the potential long-term effects of radiation damage in proposed crystalline radioactive waste forms such as ... SYNROC," Headley and colleagues reported in the Oct. 8 NATURE.

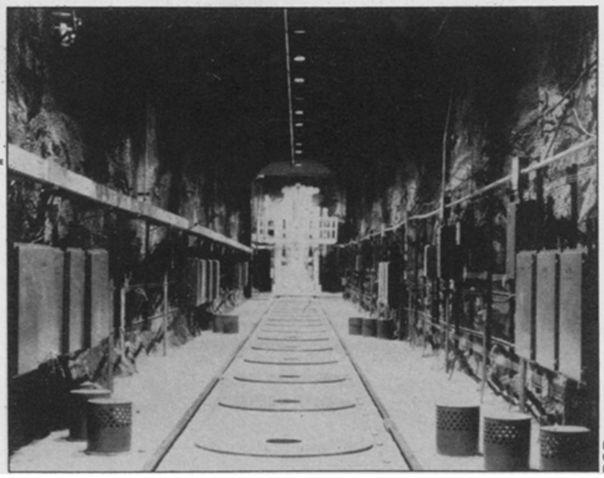
Headley used the electron beam of transmission electron microscopy to scrutinize metamict minerals. When the electron beam passes through a highly ordered crystalline structure, a highly ordered electron diffraction pattern results. But when the researchers beamed their metamict minerals, they observed only halo-like patterns characteristic of unordered structures. "The conclusion is that long-term radioactive decay of the internal uranium and thorium has damaged the crystalline lattice so much that it has been transformed to an amorphous state," says Headley. "However, it remains to be determined how much alpha damage is required to change the structure of the different crystalline phases in [SYNROC] and to quantify any changes in leach rates as the alpha damage progresses."

high-level waste for 1,000 years." This thousand-year guarantee appears adequate for fuel that has been reprocessed: The resulting high-level waste consists mainly of fission products that can decay to a relatively nontoxic level of radioactivity in that time period. Spent fuel, however, takes more time to detoxify, because it still contains plutonium and uranium, which decay to a nonradioactive state only after hundreds of thousands of years. Consequently, reports GAO, spent fuel eventually would "knock down" the multiple barriers in its waste package. In that case, the only box left in the nest isolating it from the biosphere would be the geologic repository. □

Next week: The geologic repository.

The diagram (below left) illustrates the method used to place spent fuel in granite at the Nevada Test Site. DOE has placed actual spent fuel assemblies in the Nevada granite to evaluate that rock's response to heat and radiation (below right).





DOE