A new approach to entombing nuclear waste

Ever since the beginning of the nuclear age more than 40 years ago, intensely radioactive and thermally hot reactor waste has been steadily accumulating. Even the by-products from the Manhattan Project, which led to the bombs that obliterated Hiroshima and Nagasaki, remain active at the Hanford Reservation within the State of Washington, U.S.A.

And at many other sites around the world thousands of tonnes of high-level radioactive waste — the leftovers from both nuclear power-generation and nuclear weapon-building — also await disposal.

The wastes, which will retain dangerously high levels of radioactivity for millennia, are currently stored in large steel tanks, usually above ground and in the open. Yet virtually all nuclear engineers consider such storage — in potential close contact with human life and the environment — generally unacceptable except as a temporary expedient.

Most people concerned with the management of this 'radwaste' agree that entombing the material hundreds of metres below the surface appears to be the most feasible method of safely and permanently isolating it from future civilisation(s).

Scientists and engineers at the CSIRO Division of Geomechanics believe they have devised a particularly attractive way of doing this that offers clear benefits over existing proposals.

Dr Barry Brady, Chief of the Division, and his colleagues suggest that high-level nuclear waste should be buried as part of a thermosyphon loop, within which circulating groundwater would keep it cool and confined. Initial experiments and computer simulations have produced encouraging results, and plans are being developed to test the scheme in a simulated repository in granite rock.

Omnipresent groundwater

Deep burial in stable, impermeable rock creates a natural barrier between the waste and the biosphere, and the degree of isolation can be strengthened by adding engineered barriers. For example, the waste can be immobilised in borosilicate glass, or SYNROC (see the box on page 5), and the whole mass encased in corrosion-resistant canisters.

The Oskarshamn nuclear complex in Sweden. Three power plants are in the background; in the foreground are the surface buildings of the Central Facility for Intermediate Storage of Spent Fuel (CLAB).

All these barriers are called for to counter contact with one particular entity — groundwater. Circulation of groundwater, aided by the convection that the heat from the waste generates, is seen as the most likely route for transporting radioactivity from its deep underground resting place back towards the surface.

Geologists are aware of it, but not many people realise that, below the relatively shallow level of the water table, groundwater is present everywhere — it even insinuates its way into what you might consider to be the most impervious rock. And so granite rock masses contain a small percentage of water and have groundwater flowing through them (although it may amount to only millimetres per year).

Any cavity you excavate underground will invariably fill with water (to water-table level) within a short time. If the rock suffers additional fracturing, water will flow even faster through the fissures. The difficulty when placing radioactive waste underground is that mining operations (drilling, use of explosives, and so on) inevitably cause fracturing and, more importantly, the stresses created by heat released from the waste do the same.

So the risk is that groundwater may corrode the radwaste canister, and leach radioactive material from the solid matrix in which it is embedded. These processes are greatly accelerated by elevated temperatures.

Although not finally agreed upon, performance goals set by the United States Nuclear Regulatory Commission call for the canister to last 1000 years, and for the annual release after that to be less than one-hundred-thousandth of the contained radioactivity. Further, the travel time to the surface should be at least 1000 years.

Obviously, efforts can be made to make the canister corrosion-resistant, and the matrix leach-resistant, but faced with a very exacting requirement that has never been attempted before, most engineers hesitate to guarantee that their handiwork will survive intact over geological time scales. Man-made structures are usually lucky to survive centuries, let alone millennia; yet, as the graph on page 6 shows, we would like to lock radwaste away for many, many times longer — perhaps the proverbial million years (easier to say than imagine).

What repository architect dares to convince the public that his design will survive all the possible changes that may come upon the earth within that vast time span?

Sweden has plans for a repository to replace the country's interim spent-fuel storage facility, and Swedish engineers expect that the copper-clad canisters to be used will have a life of a million years (based on a canister temperature of 200–300°C). The United States hopes to have a repository in place by the year 2000, but the temptation is to defer the day, believing that a better, assuredly safer, scheme will arise in the interim (and hope that an accident doesn't befall the existing temporary storages).

Another reason for delaying is the decay in the waste's activity with time. Only one-quarter as much radioactivity will issue from a canister after 10 years as comes from a 1-year-old package. The difference is critical in terms of the resulting rise in temperature of the mass. Whereas after 1 year buried reprocessed waste may rise in temperature by 1900°C, after 10 years it would be much less hot — about 250°C above its surroundings.

Since glass devitrifies (crystallises and becomes brittle) at temperatures higher than 700°C, it is desirable to wait some years before burial. Another consideration is that basalt fractures above 500°C (for granite, the temperature is 300°C), again favouring delay.

A final point on the side of procrastination is the boiling point of water, which at 1000 m below the surface (10 MPa pressure) rises to 180°C. Keeping the temperature below boiling point prevents hydrocracking — the cracking of rock by water expanding into steam.

When groundwater boils, it also makes the flow of groundwater through the rock's fractures and pores hard to simulate in a computer model, and hence practically unpredictable. Yet without satisfactory modelling it is impossible to say with reliability that a repository will behave safely.

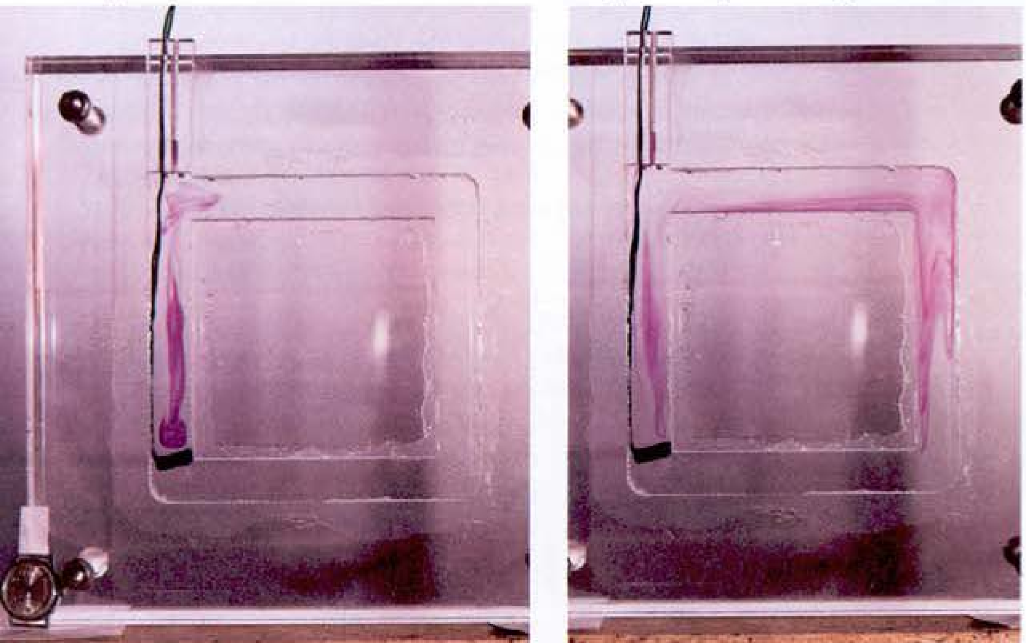
That's a big drawback, because every repository designed to date will give rise to boiling — unless the waste is stored another 100 years beforehand, or the effective concentration of the waste is diluted (which makes the capacity of a given repository that much less).

So we can see that one of the biggest bugbears of nuclear-waste disposal is its elevated temperature. Not only does this give rise to the specific problems outlined above, but higher temperatures greatly accelerate the processes of leaching and transport of radionuclides.

At Lucas Heights, spent fuel elements are held in stainless-steel-lined holes 16 m deep. Inspectors from the International Atomic Energy Agency are checking that all is well.

Thermosyphon loop

Here is where Dr Brady's scheme comes into its own. It is a way of cooling down the waste (in whatever form it may take) by circulating water over it using a thermosyphon — the system of water circulation that naturally occurs in solar water-heaters (see the diagram). Instead of treating groundwater as an unavoidable nuisance, it puts it to work in a controlled and predictable fashion. With an ample flow of water in the thermosyphon loop, temperatures will be much less than they otherwise would be, and boiling can be avoided.



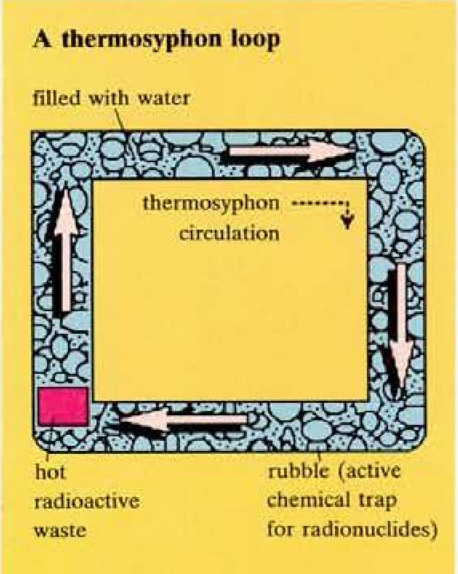
Dye traces out the path of liquid circulating in a transparent model of a thermosyphon loop.

As it happens, water surrounding a repository would, of its own accord, go into a similar circulating pattern — but on a slower and longer scale, and in a more uncontrolled fashion. Excavating a thermosyphon loop effectively creates a local hydraulic short-circuit, which confines that groundwater in the near vicinity of the waste. The water, bearing any radioactive burden, has lost much of its incentive to seek out fracture channels that may lead away from the repository.

Dr Brady envisages that the thermosyphon loop would not be an open channel but would be filled with rubble of predetermined size. This would relieve the loop of stress from surrounding rock, making it indefinitely stable. Moreover, the composition of the back-fill can be deliberately chosen so as to chemically capture, and precipitate, the radioactive species leached from the canisters.

It appears that this precipitation process may happen preferentially within fine cracks, giving an intrinsic self-sealing of the thermosyphon loop. Dr Bruce Hobbs of the CSIRO team is carrying out further

The thermosyphon would continuously cool the radioactive waste.



A thermosyphon loop

studies to confirm and elaborate the workings of this mechanism.

Another team member, Dr Lincoln Paterson, has constructed small-scale models of thermosyphons out of transparent perspex. He has placed electrical resistors in the model to provide a heat source, and has examined flow patterns by putting dye or fine particles in the loop.

Using this technique, he has looked at the effect of different sizes of back-fill (employing glass beads of various diameters), and has examined the influence of a large leak, such as a massive fault line may provide. All these experiments indicated that the thermosyphon loop is very robust.

Another colleague, Mr Harry Schlanger, has duplicated the experiments using computer models, and obtained effectively the same results. The computer runs verified that the system's performance remains

SYNROC imitates Nature too

The philosophy behind the thermosyphon loop proposal is one of imitating Nature. If we can see natural systems that have preserved themselves for relentless ages, that gives us strong assurance that a man-made copy may last as long.

We can apply the thermosyphon loop idea to any existing repository scheme, adding an extra margin of safety to it. Depending on the final safety margin desired, the repository may have its waste cased in various thicknesses of copper, stainless steel, or titanium; and it may have the waste immobilised in a matrix of glass (the first-nominated candidate), to which several European countries are committed, or in SYNROC (the Australian contender that offers superior performance to glass).

SYNROC, a synthetic form of very inert rock, was proposed in 1978 by Professor Ted Ringwood of the Australian National University in Canberra. Now the research effort on SYNROC greatly exceeds that on any other waste form except borosilicate glass.

The properties and performances of SYNROC have been studied in laboratories in Australia, Britain, the United States, Canada, Germany, and Japan. Formal agreements for collaborative research and development programs have been enacted between this country and Japan, Italy, and the United Kingdom.

Since 1979, the Australian Atomic Energy Commission (AAEC) and the ANU have received some millions of dollars in Australian government funding to investigate SYNROC. Recently, a full-scale plant has been built at Lucas Heights to demonstrate that (non-radioactive) SYNROC samples can be made on a commercial scale (about 10 kg per hour).

The recipe for SYNROC-C, the formulation designed to immobilise high-level waste from water-cooled reactors, calls for five simple chemical powders, the oxides of titanium, zirconium, calcium, barium, and aluminium. The radwaste is added as a nitrate solution (10–20% by weight) to the prepared powder (which is then a soft crystalline dark-grey material). Pressure and heat (up to 1300°C) create a black, dense, crystalline ceramic — SYNROC.

It then comprises about one-third hollandite ($BaAl_2Ti_6O_{16}$), one-quarter zirconolite ($CaZrTi_2O_7$), one-fifth perovskite ($CaTiO_3$), one-sixth rutile (TiO_2), plus one-twentieth metal alloys.

Almost all of the elements in high-level nuclear reactor wastes can be made to form an integral part of the crystal lattices of these very stable SYNROC phases, in the same way as close natural analogues of these minerals can be found to contain long-bound radioactive elements.

Its inertness makes SYNROC very resistant to leaching by groundwater. Tests have shown that glass will flake and crack badly after a few days' exposure to high-pressure water at 300°C, whereas SYNROC remains virtually unaffected, and is at least 1000 times more leach-resistant than typical borosilicate glass (such as pyrex).

The AAEC scientists have spiked SYNROC with small quantities of radioactive elements found in real radwaste. When they added neptunium-237, plutonium-239, americium-241, and curium-244, they found the leach rate at 70°C to be extremely low — less than 40 µg per day per sq. m. Further tests with strontium-90, caesium-134, and other fission products, which require handling in a 'hot cell', have shown leach rates almost exactly as predicted from non-radioactive tests.

It appears that the radioactivity of the waste causes no harm to SYNROC's crystal lattices. When SYNROC was irradiated for up to 6 months with fast neutrons from the AAEC's HIFAR reactor, it withstood the equivalent of 100 000 years of high-level waste containment with very little physical damage and no significant change in leach rate.

Under the collaborative agreement mentioned, the British Atomic Energy Authority has made small samples of SYNROC fully loaded with highly active wastes from a commercial power reactor. Tests with these have recently begun.

SYNROC: second generation immobilisation of radwaste. D. Coleby. *Nuclear Spectrum*, 1986, **2**, 22–6.

A 'hot cell' used for handling experimental SYNROC samples that have been spiked with fission products.

Hot-pressing SYNROC within stainless-steel bellows.

The final product — a 'pancake' of compacted SYNROC.

What is 'radwaste'?

High-level radioactive waste — 'radwaste' — is the unwanted fission material remaining from the 'burning' of uranium.

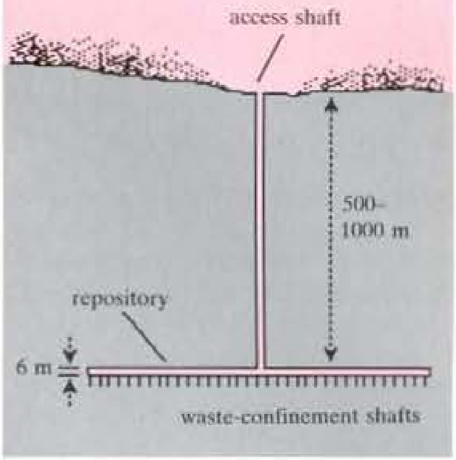
After about 3–4 years in a light water-cooled reactor, by-products from the reaction of the uranium accumulate within the fuel, and begin to dampen the reaction cycle. The spent reaction rods are removed and replaced with new ones.

A typical 1000-MW nuclear power station produces 30 tonnes of spent fuel a year. Uranium and plutonium can be recovered for recycling (by 'reprocessing'), leaving about 1 tonne of high-level waste — assorted fission fragments that contain almost all the radioactivity. It is usually formed into a solid, which is then embedded in glass (or, potentially, SYNROC).

'Reprocessing' is not a straightforward operation, and only the United Kingdom and France practise it on a commercial scale. In the United States, the operators of commercial reactors are stockpiling their spent fuel rods until more satisfactory arrangements can be made. This also raises the possibility that waste repositories may need to accommodate old fuel rods.

The level of radiation from high-level waste is intense: a person standing 10 m from an unshielded new waste canister can receive a 500-rem dose (which has a 50% chance of being fatal) in 10 minutes. The graph shows how the radioactivity decays with time. Over the first 1000 years, the radioactivity from fission fragments (strontium-90 and caesium-137 are major ones) has largely decayed, and activity from the longer-lived transuranic elements (neptunium, plutonium, americium, and curium) then predominates. These elements take about 100 000 to a million years to decay to acceptably low levels.

A plan for a repository



Thermosyphon loops would provide an extra degree of protection for any waste repository.

largely unaltered by the properties of the surrounding rock, allowing accurate prediction of the characteristics of any repository — in particular, temperature and diffusion of waste products.

One of the next things the scientists want to do is monitor the performance of a simulated repository. They plan to operate a non-radioactive version by excavating a thermosyphon loop in granite rock, and installing powerful electric heaters within it.

Natural analogues

Dr Brady is confident of the success of his thermosyphon system. With no moving parts, it should operate continuously for eons. After all, it mimics the same system as can be found in Nature. Apparently, many ore bodies throughout the world were formed by the thermosyphoning of minerals over millions of years.

If every other part of a nuclear-waste repository also has natural analogues known to last over geological time scales, then we should feel safe that our generation's radioactive waste can be buried and forgotten.

For example, ancient deposits of native copper are well known, suggesting that copper cladding should make a good shield against corrosion of the waste. Embedding the radioactive elements in SYNROC, a synthetic form of a rock known to have locked up radioactive atoms within its crystal structure over geological ages, provides similar assurance.

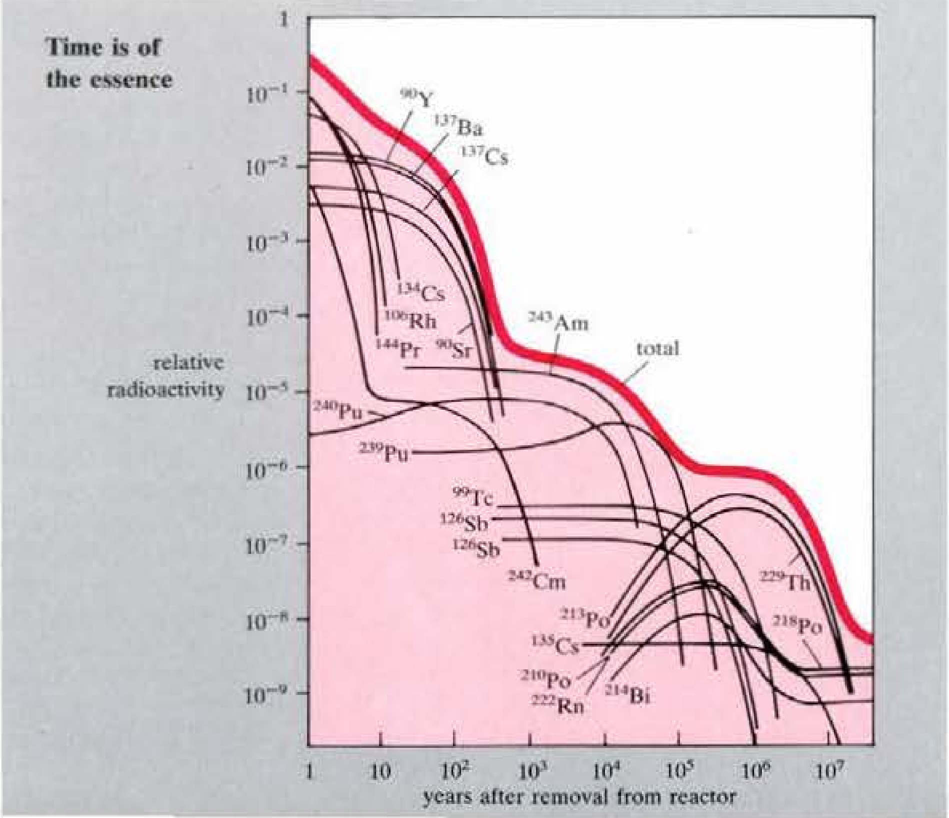
And, of course, the only reason we find uranium ore bodies today is because that material has, over many millions of years, fended off dispersion. Dr Brady likes to cite the case of the Oklo uranium deposit in the Gabon Republic. Two billion years ago, a rich uranium deposit went critical and became a natural fission reactor. The radioactive by-products, identical to those from man-made reactors, are still there today, apparently unmoved from where they were created.

By taking care and mimicking Nature, Dr Brady believes, we can design a perfectly safe scheme for burying nuclear waste. If we don't set to and build repositories, the danger of above-ground accidents, or careless dumping at sea, may be realised.

The new proposal is not an alternative to current schemes. Rather, it is a mode of operating them that provides an additional wide margin of safety. The Swedes are currently satisfied that their planned repository will remain intact for a million years. If they configure their waste into a thermosyphon system, they can expect the radioactivity to remain isolated for at least ten times as long.

Andrew Bell

Time is of the essence



The radioactivity of reactor waste diminishes with time. We would like to keep the waste isolated from the biosphere for many thousands of years.

More about the topic

A novel operating principle for a nuclear waste repository in crystalline rock. B.H.G. Brady, B.E. Hobbs, L. Paterson, and H.P. Schlanger. *Proceedings, 27th Symposium on Rock Mechanics, Tuscaloosa, Alabama, June 1986*, 910–16.