MINERALOGICAL MAGAZINE, APRIL 1985, VOL. 49, PP. 153-158

Radioactive wastes—an overview

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IN considering how best to deal with radioactive wastes it is necessary to: (*a*) identify the different types of waste which exist or which will be produced in the foreseeable future; (*b*) examine what techniques already exist for the treatment, storage, and disposal of the wastes and where further research and development is needed; (*c*) identify suitable storage and disposal sites.

The overriding factor is to be able to ensure that no harm will be caused to man or the environment, either now or in the future. In particular no one must be exposed to radiation levels in excess of the limits recommended by the International Commission on Radiological Protection (ICRP), 5 m Sieverts for the public, 50 m Sv for radiation workers.

Wastes containing only low concentrations of radioactivity can sometimes be disposed of directly to the air, land, or sea if suitable precautions are taken. Other wastes need to be isolated until the radioactivity has decayed to a low enough level, so that any possible future transfer of radioactivity to man will not be significant, and the radiation levels will be as low as reasonably achievable and within the ICRP limits.

Man-made isolation barriers may be satisfactory for wastes containing short-lived radionuclides only, i.e. those which decay away in hundreds of years, but it is advantageous to locate the waste in a suitable geological formation. Wastes which require isolation for thousands of years must be disposed of where permanent natural geological barriers supplement the artificial barriers.

There is everywhere a significant natural radioactive background and it is envisaged that projected waste disposal practices will increase this by much less than 1%, indeed, far less than the wide variations in the geographical distribution of this background.

Since it is not possible to demonstrate directly the effectiveness of any long-term isolation barrier, it is necessary to use mathematical models which predict the sequence of events both under the expected conditions and also under all foreseeable accident conditions. Laboratory and 'field' experiments are needed both to provide data for use in model calculation and to test, whenever possible, the validity of the assumptions made.

When potential disposal sites have been identified extensive on-site tests are needed to obtain the necessary data specific to each site.

Direct disposal of low-level wastes

Before low-level wastes can be disposed of directly, the potential impact of the disposal must be evaluated and monitoring processes set up. The critical group of people most likely to be at risk must be identified and the possibility of reconcentration processes, such as food chains, must be examined. For example, when liquids are discharged to rivers, the critical group will be those using the river, directly or indirectly, as a source of potable water. On the other hand the critical group for sea discharge will be those who eat fish, shell fish, or edible sea weed caught or collected near the discharge site. Pre-disposal studies and post-disposal monitoring are both needed.

The pre-disposal studies will enable the Authorizing Bodies to set limits on the levels of radioactivity to be disposed of, e.g. liquid waste streams are usually treated by chemical precipitation or ion exchangers which are selective to specific elements, e.g. Cs or Sr.

Low-level solid wastes are often buried in shallow, land-fill type trenches; others are enclosed in suitable containers which are deposited on the floor of the deep ocean at depths of 4000 m. Although the wastes themselves remain *in situ*, radioactive constituents can be leached out from the wastes by ground water or ocean currents. The likely movement of leachate through a trench site and the fate of the dissolved radionuclides, usually immobilization by adsorption in the adjacent soil, can be predicted from hydrological measurements and laboratory experiments. Bore holes opened up alongside such trenches are used to monitor the radioactivity of the ground water. Tritium, as HTO, is not adsorbed and some radionuclides form mobile non-sorbed chemical complexes with organic compounds present in the waste or soil. Complexing can sometimes be prevented by pre-treating the waste, but if tritium is present then a dry area remote from any aquifer may have to be chosen for the site, or else sea disposal selected.

Extensive oceanographic and marine biological studies were carried out to select a deep ocean site in the North Atlantic suitable for the disposal of solid wastes sealed inside concreted drums. In order to be able to predict how leachate mixes with the ocean it was necessary to make extensive measurements of the deep ocean currents, which are quite different from those at the surface. Again, checks were carried out to ascertain whether the sparse fish life at the bottom plays any role in the foot chain of surface fish. No bottom fishing is carried out at this depth, but some surface fishing may take place. Routine surveillance of the site has shown, as expected, that the radioactivity in deep-sea fish and in the sea water is not measurably different from the natural level.

Types of waste requiring isolation

These wastes can be divided into three categories: (*a*) high-level wastes (HLW) and unreprocessed spent fuel; (*b*) intermediate-level wastes (ILW) containing short lived radioactivity only; (*c*) intermediate-level wastes containing some long-lived radionuclides.

In countries, like France and the UK, where spent fuel is reprocessed to recover uranium and plutonium, nearly all of the fission products are removed at the first solvent extraction stage in nitric acid solution. This solution, which contains 95% or so of the total radioactivity present in the fuel, is concentrated and stored in high-integrity stainless steel tanks. The waste emits considerable amounts of heat due to radioactive decay and the tanks need to be cooled. Several independent cooling circuits are available. In due course these liquid wastes will be converted into solid blocks which are easier to store and supervise. Vitrification is the only solidification process which has been taken to the manufacturing stage and this is likely to be the process generally used in the near future. As indicated below, alternative processes, such as the production of synthetic rocks, are in the development stage. The blocks of waste will continue to emit substantial amounts of heat for decades so that, if they are disposed of during this period, the disposal site strata must be capable of withstanding a significant temperature rise. The actual temperature will depend on the density of the blocks in the repository. Again, heavy shielding and some cooling facility will be needed during transportation. Storage for periods of 50 years or more will reduce the thermal output by a factor of ten or more, enabling a wider range of rock types to be used, more blocks per repository and less shielding. There is thus every incentive to store these wastes and to defer decisions on their final disposal. By the year 2000, the total volume of solidified UK HLW will be about 1000 m^3.

Unreprocessed spent fuel contains all the uranium and plutonium produced in the fuel, in addition to the fission products. Similar amounts of heat are given off as from high-level wastes. Proposals have been made to encapsulate the fuel rods in one or other of a number of canister types, store to allow the radioactive decay heat to decay to a suitable level, and then to dispose of these in a similar fashion to solidified high-level waste. If, and when, spent fuel is exposed to ground water, the leachate will contain a large amount of uranium. and so be chemically different from the leachate derived from high-level wastes. Again, the uranium will gradually reach equilibrium with its radioactive daughters, notably radium. There will also be significant amounts of plutonium present.

Intermediate-level wastes which are free from long-lived radionuclides arise mainly at nuclear power station sites and are largely due to the production of neutron-activation products. The wastes consist of metallic components from the reactor core, including miscellaneous hardware associated with fuel element assemblies, and ion exchangers and sludges from water reactor circuits, and the reactor fuel element ponds.

A number of intermediate-level waste streams are produced at fuel reprocessing and fuel fabrication plants. Most of these contain long-lived radionuclides, especially alpha-emitting transuranic elements such as plutonium and neptunium. These wastes include fuel element cladding, residues from fuel dissolvers, ion exchangers and sludges from long term fuel storage ponds, evaporator concentrates and plutonium contaminated materials from the fabrication of fast reactor fuel elements.

Isolation options

As indicated, wastes can be isolated both by man-made barriers and by natural geological features. It is convenient to refer to the former collectively, as the 'near field' and the latter as the 'far field'.

The near field comprises (*a*) the 'waste form' itself, which will be chemically inert and designed to have a very low leach rate in ground water; (*b*) the waste container, e.g. a corrosion resistant canister; and (*c*) a backfill, e.g. an impervious clay lining.

The far field will be the surrounding undisturbed rock. Various rock strata, especially evaporites, argillites, and crystalline rocks (see below), have been proposed in various countries for repository sites. In addition, some deep ocean sediments are being investigated as possible alternatives to underground strata for the comparatively small volume of high-level wastes. The wastes would be buried in the sediment, the sea itself playing, at most, a subsidiary role acting to dilute and disperse any traces of low residual radioactivity that might migrate upwards from the burial site.

The isolation can be conveniently divided into three stages. The first is the pre-disposal 'cooling off' period of up to 100 years which applies only to high-level wastes. The second stage of 500 to 1000 years, applies to all wastes. During it, little or no groundwater (or sea water) will contact the waste because the back-fill retains its low permeability and the canister its integrity. Short-lived fission products will decay away so that intermediate-level wastes free from long-lived radionuclides will be rendered harmless. At the end of this period high-level wastes will resemble those long-lived intermediate-level wastes which contain transuranic elements.

The final period will need to last many thousands of years to allow long-lived alpha emitters such as ^{237}Np and fission products like ^{99}Tc to decay. These have half lives of 2.1×10^6 and $2{\cdot}1 \times 10^5$ years respectively. It is unreasonable to expect the waste-form, the canister, and the back-fill, to last undamaged for this time, and access of water to the waste is assumed to take place. The residual radionuclides will leach out slowly, with glass over some thousands of years, and migrate with the groundwater (or sea water) through the near field and the surrounding rock (or ocean sediment). There will be interaction between the dissolved species, the near field and the rock, and it is necessary to be able to predict convincingly that if any residual radioactivity does return to man then the levels will not be significant.

Mathematical modelling is necessary to enable 'sensitivity analysis' to be carried out on any projected disposal scheme. Thereby, the relative importance of such factors as low leachability, corrosion resistant canisters, underground water movement, and sorption of radionuclides, both on the near field and on the geological media can be established. The factors controlling the leaching of the radioactivity from the waste-form into groundwater are found, in general, to be of secondary importance to those that control the rates of migration with the groundwater through the near field and the surrounding rock. From an analysis carried out by the National Radiological Protection Board in the UK, for example, it can be deduced that, for high-level wastes, neither the development of waste-forms that leach out very much more slowly than from glass, nor the use of canister materials, which would last for as long as 100 000 years, would be justified if a suitable combination of near field and surrounding rock is chosen.

Not as much information is available for intermediate level waste disposal, but the general conclusions are likely to be similar.

Near-field barriers

The 'waste form' for high-level wastes. Although several different types of waste forms have been examined for their suitability for disposing of high-level wastes, most workers have concentrated on glass, especially borosilicate glasses. These can accommodate a wide range of waste compositions and have been found to be stable under radiation doses well in excess of those expected. They exhibit a very low leach rate even if broken up. Leach tests are comparative, not absolute, since they are normally measured in the laboratory by a modified 'Soxhlet' test which is very unrealistic in comparison with the likely leaching mechanism expected underground. The laboratory tests expose the glass to a continuous flow of water, whereas it is much more likely that only a very slow trickle of water will be available and a quasi-equilibrium condition will be more realistic. Attention is now being drawn to the consequences of this. In any event, the modelling studies referred to indicate that under reasonable geological conditions there is no great advantage in reducing the leach rate to an extremely low level. Of course a virtually unleachable waste form, possibly a synthetic rock, could be disposed of in geological formations which might otherwise be regarded as unsuitable.

Many glass compositions have been developed, as well as several types of processing plants. The French have built and operated a full scale active plant and demonstrated that vitrification is a realistic process. It is likely that other types of vitrification plants will eventually be built.

Although vitrification is generally regarded as perfectly acceptable, research is still continuing on alternative waste forms for high-level wastes. Most of this is concentrated on the manufacture of a range of synthetic rocks, e.g. SYNROC. These rocks are designed to incorporate the most important radioactive elements entrained within suitable mineral lattices. The composition of the synthetic minerals is based on natural mineral analogues which have remained stable over geological time. So far these have only been made on a small scale in the laboratory, usually with inactive simulates. Whether the properties of large specimens of

TABLE I. *Summary of intermediate- and low-level solid waste arisings in the UK to year 2000**†

	Conditioned volume to AD 2000 m^3	Total activity Ci		Average specific activity‡	
		α	$\beta\gamma$	$\alpha Ci/m^3$	$\beta\gamma Ci/m^3$
Intermediate-level wastes with significant alpha activity					
Cladding wastes	13000	5×10^5	6×10^7	40	5000
Sludges, resins, concentrates	23000	2.3×10^5	7×10^6	10	300
Pu contaminated wastes	16000	1.6×10^5	6×10^6	10	350
Other α-containing solids	200	2×10^3	10^5	10	500
Intermediate-level wastes with insignificant alpha activity					
Sludges, resins	5000	500	5×10^5	0.1§	100
Activated and contaminated items	19000	0	10^6	0	50
Misc. tritiated etc. Amersham International	9000¶	0	6×10^5	0	70
Low-level wastes	490000‖	Low	Low	Low	Low

* The small amounts of defence wastes are not included.
† Power programme as assumed for Sizewell inquiry evidence, i.e. all magnox reprocessing completed by 1995, 8 GW of AGR and 12 GW of PWR at AD 2000, 1850 tonnes of AGR fuel reprocessed, no PWR fuel reprocessed, by AD 2000. No arisings from reprocessing overseas fuel are included.
‡, § Specific activity covers a wide range, in particular ‡ where only a small part of this item contains α activity.
¶ Volume after packaging for sea disposal. Only approximately 10% of this is waste.
‖ LLW wastes buried at Drigg or similar site in unconditioned state.

synthetic rock waste forms, composed of several synthetic minerals, will match those of small specimens remains to be proven. Synthetic rocks require more rigorous manufacturing procedures in terms of temperature and pressure than glass. Much technological development will be required before fully radioactive full scale synthetic waste forms can be made. Unreprocessed fuel elements provide their own waste form. These consist essentially of ceramic-grade uranium oxide which has low leachability.

The 'waste form' for intermediate-level wastes. The volumes of intermediate level wastes are much greater than those of high-level wastes (Table I). Intermediate-level wastes, unlike high-level wastes, are very heterogeneous. Encapsulation in glass or synthetic minerals could be difficult and extremely expensive. Fortunately, less rigorous criteria apply, little heat is evolved, radiation damage is much less, and calculations indicate that leachability is not necessarily the most important factor. Indeed it was concluded at a recent meeting of the International Atomic Energy Agency (Utrecht 1982) that for short-lived radionuclides, packaging into suitable drums with no immobilization might be quite sufficient.

More attention, however, is needed for wastes containing long-lived radionuclides. For these a variety of immobilizing agents have been used or are under development, e.g. cement, epoxy resins, polyester resins, water-compatible polymers and bitumen. The leach rates from portland cement are high but can be reduced both by the use of mineral additives such as clinoptilolite and by impregnation with a suitable polymer. The immobilizing matrix selected is based to a large extent on the mutual compatibility of the waste and the matrix. For example cement can incorporate a higher proportion of inorganic than organic ion-exchange material. Glass and metal matrices have also been studied for use with plutonium-contaminated items such as fuel element cladding, but are not prime candidates.

Canister materials. Several metals and alloys have been examined as potential canister materials. Some, such as titanium alloys, are thermodynamically unstable but due to their protective oxide coating are very corrosion resistant. If the coating is damaged, rapid deterioration can take place. Others, such as copper, are thermodynamically stable under some, but not all groundwater oxidation conditions, and yet others such as cast iron are always unstable and corrode but at a predictable rate. Thus, thick canisters of say 100 mm cast iron, or copper under oxidizing conditions, will provide protection for sufficiently long periods. It is arguable which type of canister to use for short-lived radionuclides. One advantage of cast iron is that its corrosion product will help to keep the oxidation potential in the vicinity of the waste at a low enough value to prevent the formation of mobile species of long-lived neptunium and technetium. The oxidation potential is also controlled by the rate at which ground water brings dissolved oxygen to the site. Thus the choice of canister material is dependent on the hydrology and geochemistry of the geological stratum selected.

Mild steel drums and concrete containers alone have been proposed for intermediate-level wastes. Mild steel may last some hundreds of years in the right environment, i.e. high pH and low oxidation potential. Thus an excellent combination would be a concrete waste form within a mild steel drum buried in clay, or a clay back fill. Even if the waste form is bitumen or an organic resin it is likely that concrete will form an external barrier. The only geological environment likely to pose a problem are brine inclusions in acid salt mines but even here the choice of the correct back fill may overcome this. A concrete canister may be part of a concrete waste form or a protective barrier to another waste form.

Backfill. The backfill is the medium filling the voids in the excavation and has at least two functions. As in conventional mining practice it seals the repository from surrounding ground water, provides strength and helps to dissipate heat. In addition, it can chemically condition any water that does penetrate into the repository so that attack on the canister and waste form is minimized. Again it can chemically condition and react with any leachate so that dissolved species are more readily immobilized in the backfill and surrounding rock.

It is convenient to use 'mining practice' material for the bulk of the backfill and special compositions in the 'buffer zone' immediately surrounding the canister. Clay and concrete are the two materials most widely used for the backfill with clay/cement, clay/sand, and clay/mineral mixtures for the 'buffer zone'. Concrete barriers may be used to segregate canisters and also act as radiation shields.

Far field

The far field depends essentially on the hydrological, engineering, and geochemical properties of the surrounding rock. Three different types of rock are currently being examined as potential hosts; evaporites, argillites, and crystalline rock.

Rock salt, as beds or salt domes, is the most widely studied of the evaporites, anhydrite less so. Evaporites are impervious to water if the deposits are free of dislocations or discontinuities, and isolation essentially depends on this because there is little sorption of radionuclides from ground water. Some evaporite strata, however, contain layers of other rock types which may allow some water to enter; the existence and properties of these must be well characterized. Temperature gradients in salt arising from buried heat-emitting wastes can cause the movement of canisters and also any water present. Brine inclusions may cause canister corrosion. The properties of the enclosing strata must also be studied to be able to predict the likely consequence of any release of radioactive waste.

Both soft and hard argillites, e.g. clays and mudstones, have been studied. Soft clays are virtually impervious although they may be water logged. Such strata are, however, unsuitable for heat emitting wastes since elevated temperatures may cause the clay to dry out and crack. Clay trenches are proposed for those intermediate-level wastes of short half-life. Such trenches could be, say 6–9 metres deep, possibly with a concrete base to support the weight of canisters, and concrete walls and roof. Mudstones, are usually fissured but isolate leachate well by combination of their hydrological and sorptive characteristics.

Crystalline rocks, such as granite, have been extensively studied as repositories for heat-emitting wastes because of their good engineering properties. Such rocks are often extensively fissured with large water-carrying fractures and an extensive micro-fissure structure. Most radionuclides present in a leachate will be chemically sorbed by the rock and thus be removed from the groundwater. Other species, that may be mobile, will diffuse into the microfissures and be physically retarded in movement. Calculations based on laboratory and field trials suggest that a combination of chemical and physical retardation delay the return of radioactivity to the surface or the nearest aquifer for at least many thousands of years.

Calcareous and argillaceous sediments on the floor of the deep ocean are being studied as potential host strata into which high-level radioactive wastes may be placed. Such disposal would be too expensive for the much more voluminous intermediate-level wastes. At locations away from seismically active regions there is only a very slow movement of water upwards from the basement rocks to the ocean floor. The sorptive properties of these sediments are more than adequate to ensure that the long-lived radionuclides in the leachate will remain immobilized within a few metres of the canister. Experiments are needed to examine whether the technique of emplacing canisters several metres deep in the sediments will produce a short-circuit path along which leachate could migrate. There is also the possibility that heat emission might produce a convective plume of water. *In situ* experiments are planned.

Conclusion

Many low-level radioactive wastes can be discharged directly to the environment, gases to the atmosphere, liquids to rivers or the sea, solids to simple trenches or the bottom of the deep ocean.

Intermediate-level wastes of short half-life can be isolated by a combination of waste form, canister and backfill. High-level wastes after suitable storage, and intermediate-level wastes of long half-life, must depend for their isolation on a combination of the geochemical and physical properties of the near field and host rocks. Experimental results and mathematical models indicate that the return of radionuclides back to man can be delayed long enough for the radioactivity to decay to harmless levels if suitable geological strata are selected.

KEYWORDS: radioactive waste, environment.