AUSTRALIAN ATOMIC ENERGY COMMISSION

CURRENT STATE OF THE ART IN HIGH-LEVEL RADIOACTIVE WASTE DISPOSAL

J.M. COSTELLO

Reprinted from "Atomic Energy in Australia", 27, [1–4], January, 1984.
Editorial Office: Private Mail Bag, Sutherland, N.S.W. 2232

CURRENT STATE OF THE ART IN HIGH-LEVEL RADIOACTIVE WASTE DISPOSAL

J.M. COSTELLO

Australian Atomic Energy Commission,
Lucas Heights Research Laboratories

This paper was presented originally at the Australian Mineral Foundation Seminar "Major Issues in Nuclear Energy" which was held from 13 – 14 June 1984, in Adelaide, South Australia. The paper is reprinted here with minor amendments.

Introduction

The need for safe disposal of wastes containing radioactivity is an inevitable consequence of the harnessing of nuclear energy. These wastes may contain radioactive materials occurring in nature – as in the case of mining and milling of radioactive ores, or alternatively may be produced by nuclear fission reactions in nuclear power generation, or in the manufacture and use of radioisotopes for medical and industrial purposes.

Radioactive wastes are produced in all physical forms, that is, as gases, liquids and solids with concentration of radioactivity ranging from near background levels to several tens of hundreds of thousands of terabecquerels* (TB_q) per cubic metres of waste – a range of concentration in excess of 10^{15}. The radioactivity is continually reduced as the radioactive nuclei decay into stable isotopes, the rate of reduction being governed by the half-lives† of the particular isotopes contained in the waste.

Large quantities of radioactivity are contained in the spent fuel discharged from nuclear power reactors. The treatment and disposal of this spent fuel has attracted public attention.

This paper briefly describes the objectives and principles of radioactive waste disposal. Its main theme is a review of the status of developments in treatment and geologic disposal of the highly radioactive wastes contained in spent nuclear fuel. It has been based on a recent paper presented at the joint Australian Geoscience Council: Australian Academy of Science Symposium "Radioactive Waste Management: A Geoscientific Assessment" (Costello, 1983).

Objectives and Principles of Disposal of Radioactive Wastes

The basic objective in the disposal of radioactive wastes is protection of man and the environment from unacceptable harm. In effect this means achieving a sufficient degree of isolation or dilution of the wastes so that any return of radionuclides to the biosphere is at a rate and/or concentration sufficiently low as not to present an unacceptable biological hazard. A further objective is to achieve the degree of isolation required for long-lived wastes without reliance on long-term surveillance.

Three basic principles are employed in radioactive waste disposal. They are:

(i) Dilution and dispersion of short-lived or very dilute radioactive wastes. The radio-

* Becquerel (Bq): the amount of radioisotope which decays at the rate of 1 disintegration/second; 3.7 x 10^{10} Bq is equal to 1 curie (Ci).

† Half-life: the time needed for a number of atoms of a nuclide to be reduced by half through radioactive decay.

activity may be reduced to acceptable levels by dilution in the environment. Quantitative physical, chemical and biological data and knowledge of dispersion phenomena and reconcentration factors at the specific disposal site are essential.

(ii) Concentration and containment of long-lived radioactivity. These wastes are confined in specially engineered structures; thus uranium mill tailings are disposed of into above ground or subsurface retention systems, and it is intended to dispose of solidified highly radioactive wastes from spent reactor fuel by deep geologic burial.

(iii) Delay and decay storage of very short-lived radioactive wastes to permit their decay into non-radioactive species.

These principles are applied separately or in combination, depending on the nature and concentration of the radioactivity in the wastes, their toxicity, mobility, radioactive half-life, and type of radioactive emission (alpha (α)-particle, beta (β)-particle, gamma (γ)-ray or neutron). These characteristics govern the choice of management procedures to be adopted.

Spent Fuel from Nuclear Power Generation

A modern 1250 megawatt electrical (MWe) pressurised light water reactor (PWR), operating at 80% capacity, and capable of supplying the electrical needs of more than 1 million people, discharges 73 spent fuel assemblies each year. The spent fuel consists of about 32 tonnes (t) of uranium, 1.15 t of fission products, about 285 kg of plutonium, and about 50 kg of other transuranium elements. Spent fuel is intensely radioactive, hence it is temporarily stored under water at the reactor sites, the water providing radiation shielding and removing heat from fission product decay in the spent fuel. The spontaneous heat generation rate in freshly discharged fuel is about 2 MW t^{-1} of contained uranium, falling to 10 kW t^{-1} after one year's storage, and to 1 kW t^{-1} after 10 years.

Spent fuel may be managed by reprocessing, a technique which extracts residual uranium and plutonium for re-use in nuclear fuel cycles, and generates a high-level waste solution containing the fission products and other transuranic elements for conversion into a solid form for burial in deep, stable geologic formations. Alternatively, the unreprocessed spent fuel may be placed in long-term retrievable storage, or it too may be regarded as high-level waste, encapsulated and consigned to geologic disposal.

Status of Spent Fuel Reprocessing

In the USA, reprocessing of spent fuel from commercial nuclear power generation was deferred indefinitely in 1977 as an initiative to reduce risks from the misuse of plutonium; however, it received qualified political approval in 1981. Reprocessing is taking place in Western Europe, Japan and the USSR, where plutonium-fuelled fast breeder reactor (FBR) programs are well advanced, and also in India where a 50 MW experimental FBR is under construction. Reprocessing and plutonium recycle are vital parts of the breeder fuel cycle.

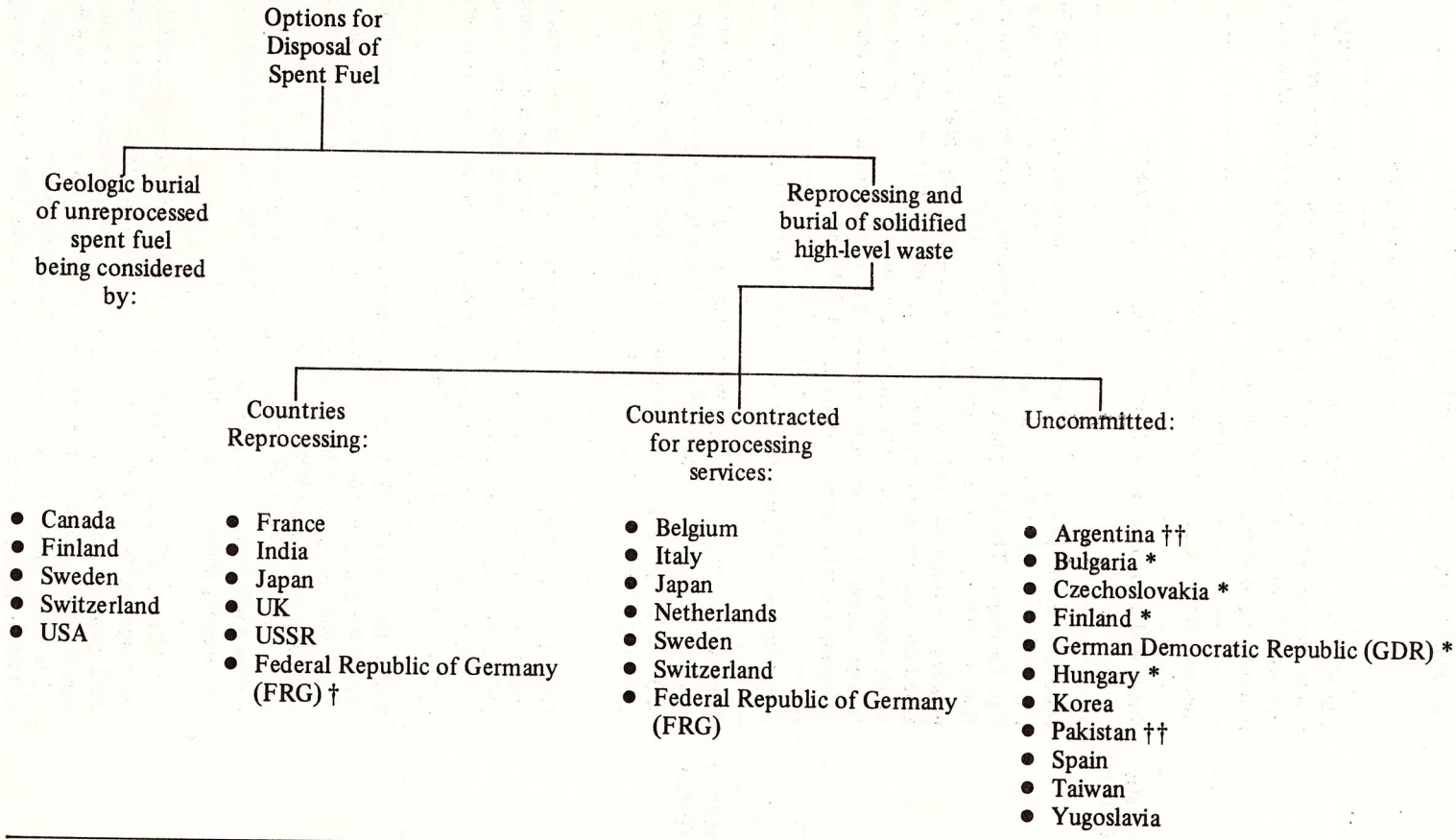
The Australian Atomic Energy Commission forecasts of nuclear power growth suggest that, by the year 2000, about 60 000 t of uranium in spent fuel may have been reprocessed in Europe and Japan, with a further 140 000 t awaiting reprocessing, and that a further 55 000 t in the USA will require reprocessing, long-term storage or geologic disposal (AAEC, 1981).

National attitudes towards spent fuel disposal are listed in Table 1. Of 24 countries with operational nuclear power stations above 30 MW, six are currently reprocessing domestic fuel, and seven have contracts with international reprocessing services. Five countries are considering geologic burial of spent fuel. The status of national programs for fuel reprocessing is summarised in Table 2.

Nature of High-Level Waste

Reprocessing of the fuel discharged yearly from a 1250 MWe LWR gives rise to about 20 m^3 of high-level waste concentrate. This is a solution of nitric acid which contains about 0.5% of the uranium and plutonium, together with about 99% of other actinide elements and fission products, other than rare gases and iodine originally present in the spent fuel. The fission products consist of isotopes of 42 elements representing every group of the Periodic Table, from lithium (Atomic No. = 3) to ytterbium (Atomic No. = 70); 33 of these elements are present as radioactive isotopes. The actinide

TABLE 1 – ATTITUDES TOWARDS REPROCESSING IN COUNTRIES WITH NUCLEAR POWER STATIONS ABOVE 30 MWe



* Spent fuel of Soviet origin ultimately to be returned to USSR.
† Plant understood to be under maintenance.
†† Pilot-scale reprocessing plants reported under construction.

TABLE 2 – NATIONAL PROGRAMS FOR REPROCESSING SPENT FUEL FROM COMMERCIAL NUCLEAR POWER GENERATION

Country	Reprocessing Capacity and Plans
Belgium	The 60 t U a^{-1} Eurochemic plant at Mol was operated between 1966 and 1974; it was then closed as uneconomic. A decision on recommencement of operations and possible increase in capacity is awaited. Belgium has contracted for French reprocessing of about 54 t U of fuel.
Canada	Research supporting vitrification development.
German Democratic Republic (GDR)	Spent fuel is to be returned to the USSR for reprocessing.
France	Natural uranium gas-graphite fuel has been reprocessed since 1958 in the 1 200 t U a^{-1} UP1 plant at Marcoule, and also since 1967 in the 900 t U a^{-1} plant at Cap La Hague. The UP2 plant, after adaptation, commenced reprocessing LWR fuel in 1976 at a nominal capacity of 100 t U a^{-1}. This capacity is being progressively expanded, and new plant, UP2-800, with 800 t U a^{-1} capacity is scheduled for operation by 1989. A third plant, UP3A, of capacity 800 t Ua^{-1} is under construction and expected to operate in 1987. A duplicate plant, UP3B, is also under consideration. France has international reprocessing contracts involving a total of about 6 000 t U of LWR fuel.
Finland	Spent fuel of USSR origin is to be returned to that country for reprocessing.
Federal Republic of Germany (FRG)	The experimental 35 t U a^{-1} WAK plant at Karlsruhe, operational since 1971, was reported closed in May 1980 for repairs to equipment. Construction of a 350 t U a^{-1} plant has been announced for Wackersdorf in Bavaria. A total of 1 700 t U of spent LWR fuel is contracted for reprocessing in France.
India	The 60 t U a^{-1} Trombay plant, reprocessing natural uranium metal fuel, became operational in 1965. The 100 t U a^{-1} Tarapur plant for reprocessing HWR and LWR fuel became operational in 1977. It is believed that a third plant of capacity 100 t U a^{-1} for spent oxide fuel from heavy water reactors is intended to be operational in the late 1980s at Kalpakkam.
UK	The 1 000 t U a^{-1} B204 plant reprocessed natural uranium gas-graphite fuel in the 1950s and early 1960s. The 2000 t U a^{-1} B205 plant has reprocessed this fuel since 1964 and is scheduled for a major renovation. The B204 plant, after modification, reprocessed LWR fuels between 1968 and 1973. A 1 200 t U a^{-1} thermal oxide reprocessing plant (THORP) is under construction and expected to commence LWR fuel reprocessing by 1990. The UK has international contracts for reprocessing about 3 100 t U of spent fuel.
USA	The 300 t U a^{-1} plant at West Valley, N.Y., operated intermittently from 1966 to closure in 1972. A novel 300 t U a^{-1} plant at Morris, Illinois was never operated owing to maintenance design problems. Reprocessing of commercial nuclear power fuel was deferred indefinitely in 1977. Construction was halted on the 1 500 t U a^{-1} plant at Barnwell, South Carolina, which could require an additional $800 million to complete. Its operation in the 1990s has been suggested.

TABLE 2 (cont.) – NATIONAL PROGRAMS FOR REPROCESSING SPENT FUEL FROM COMMERCIAL NUCLEAR POWER GENERATION

Country	Reprocessing Capacity and Plans
USSR	Spent fuel reprocessing is being carried out on a pilot scale; no data are available on the capacities or locations of Soviet reprocessing plants. However, it is understood that spent fuel of USSR origin arising in COMECON countries, e.g. Bulgaria, Czechoslovakia, GDR, is scheduled for return to the Soviet Union. The USSR has also negotiated for return of similar spent fuel from Finland.
Italy	20 t of LWR fuel has been contracted for reprocessing in the UK. The Eurex pilot plant at Saluggia has capacity equivalent to about 10 to 20 t U a^{-1} of LWR fuel and is used for reprocessing research and development.
Japan	A small 100 to 140 t U a^{-1} demonstration reprocessing plant at Tokai Mura has been in operation intermittently since 1977 on LWR fuel. A commercial 1 200 t U a^{-1} plant for reprocessing LWR fuel is planned for operation in 1990. Japan has contracted for reprocessing 1 600 t U LWR fuel in France and 1 600 t U in the UK. Japan has also renewed a contract for reprocessing 500 t U of gas-graphite fuel in the UK.
Netherlands	120 t U has been contracted for reprocessing in France.
Sweden	727 and 140 t U of LWR fuel have been contracted for reprocessing in France and the UK respectively. The majority of Swedish spent fuel (6 000 t) is to be stored in Sweden for up to 20 years in a central store (CLAB) pending a decision on its reprocessing or disposal.
Switzerland	470 t U has been contracted for reprocessing in France.

(continued from page 18)

and transuranic elements present range from actinium (Atomic No. = 89) to californium (Atomic No. = 98). The solution spontaneously generates heat at a rate between 1 and 10 kW m^{-3}, depending on the storage time of the fuel before reprocessing.

In Belgium, France, India, Japan, the UK and the USA, high-level waste solutions from commercial nuclear fuel reprocessing have been stored in stainless steel tanks for periods up to 25 years. The tanks require cooling to remove fission product heat, secondary containment to restrain any leakages from entering the environment, redundant spare tanks for transfer of solution in the event of tank failure, and constant supervision. Storage of high-level waste solution is an interim measure before solidification of the waste into a stable, practically insoluble glass or ceramic form suitable for disposal, e.g. by burial in deep geologic formations.

Solidification of High-Level Waste

Solidification of high-level waste solution involves evaporation of nitric acid and water, calcination of residual nitrates of fission products and actinides into oxides, and conversion of the oxides into a stable waste form. Candidate waste forms include variants of glass, ceramics, and also multi-barrier matrix waste forms in which the high-level glass or ceramic waste forms are incorporated into metal matrices. Several engineering alternatives have been developed for the evaporation and calcination operations, and for fusing, casting or pressing the waste forms. These operations must be carried out and maintained remotely owing to the high level of radioactivity.

The optimum solid high-level waste form should include the following characteristics:

- the ability to incorporate more than 52 fission products and actinide elements at a high proportion in the solidified product;
- radiation stability;
- thermal stability;
- low leachability and high resistance to groundwater attack;
- production at low temperatures to retain volatile elements; and
- production technology compatible with remote operation under highly radioactive conditions.

The ideal waste form with all the desirable characteristics has yet to be demonstrated. However, many solid forms are likely to be satisfactory in an appropriately engineered disposal system. Conversion of wastes into borosilicate glasses (vitrification) is in operation on an industrial scale for wastes from low burn-up fuel. The technology has been demonstrated on an engineering scale for wastes from high burn-up fuel (Bonner et al., 1980). Solidification of high-level waste from the annual reprocessing of fuel from a 1 250 MWe LWR could give rise to about 29 cylinders of borosilicate glass, each cylinder being 30 cm diameter x 3 m height (INFCE, 1980). Glass is considered to be adequate for a first demonstration system of solidification, transportation and ultimate disposal (USNRC, 1979), but second generation ceramic waste forms are under development, e.g. SYNROC, which may have superior overall characteristics.

Status of High-Level Vitrification

Table 3 summarises some national achievements, developments, and plans for the solidification of high-level waste. At present most nations with nuclear power programs have selected borosilicate glass as the principal form for immobilising their high-level waste. In terms of established, engineering scale processes, borosilicate glass is the only choice available.

France is the world leader in applied vitrification technology. Following small-scale research between 1958 and 1963, a continuous industrial scale vitrification plant, Atelier Vitrification Marcoule (AVM), commenced operation in 1978 on high-level waste from reprocessing of gas-graphite reactor (GGR) fuel. The AVM plant has demonstrated an annual capacity for the vitrification of 132 m^3 of fission product solution into 60 t of glass (Chotin et al., 1982). By May 1982, 546 m^3 of high-level liquid containing 3.3 EB_q (88 MCi) of radioactive waste had been vitrified in 734 canisters, each holding about 340 kg of glass. This high-level waste was produced by the reprocessing of 9 657 tonnes uranium (tU) of GGR fuel (Celeri et al. 1983). Two additional vitrification plants are planned to start up in 1986 and 1989 at Cap La Hague for solidification of waste from reprocessing oxide fuels. These plants together will be capable of solidifying wastes from about 1 600 t of spent LWR fuel annually, equivalent to the fuel discharged annually from about fifty 1 GWe reactors (Thiry et al., 1983).

French vitrification technology is being marketed internationally. Although the UK developed its own vitrification technology in the 1960s and 1970s, it was decided in 1981 to use French technology under licence. A vitrification plant is being built at Sellafield to commence operation in the late 1980s. Belgium and the Federal Republic of Germany (FRG) are reported to have negotiated contracts for access to AVM technology for possible application at the former Eurochemic reprocessing site at Mol and at the Karlsruhe reprocessing plant, respectively (CEA-Cogema, 1981). French technology was also considered for solidification of high-level wastes in the US. However, US vitrification technology has been chosen for immobilising US defence wastes at the Savannah River plant on grounds of process simplicity and cost (USDOE, 1983a). The Defence Waste Processing Facility at Savannah River is under construction and is scheduled to be in operation in 1988. The plant has a budget of US$900 million (Oertel et al., 1983). US technology has been chosen for vitrification of liquid HLW stored at the closed West Valley reprocessing plant (Knabenschuh, 1982), which was the only commercial nuclear fuel reprocessing plant to have operated in the US. It is expected that this solidification operation will commence in 1987.

India has a vitrification plant at Tarapur with a production capacity of 4 kg/hour and another is under construction at Trombay. A vitrification process developed by the FRG is under construction at Mol in Belgium for demonstration on a

TABLE 3 – STATUS OF SOLIDIFICATION AND GEOLOGIC BURIAL OF HIGH-LEVEL WASTES FROM COMMERCIAL THERMAL NUCLEAR POWER GENERATION

Country	High-level waste solidification		Geologic burial	
	Preferred waste forms	Developments	Preferred formations	Developments
Australia (AAEC)	SYNROC	Non-radioactive large-scale production development, together with leaching trials at high temperatures and pressure on inactive and irradiated specimens is being conducted. Experimental facilities for the fabrication of laboratory-scale specimens containing fission products and actinides have been constructed.	–	–
Belgium	Borosilicate glass beads in lead matrix (PAMELA Vitro-met), borosilicate glass blocks	A vitrification plant (PAMELA) using FRG technology is scheduled for operation at Mol by 1986, and a second plant using French technology is scheduled for 1987.	Clay	An underground laboratory, in clay, at Mol was completed in October 1984. An experimental program has commenced to study corrosion behaviour and waste package components. An operational underground demonstrational facility in the Mol clay is planned for use by 1995.
Canada	Borosilicate and aluminosilicate glasses, glass beads/lead alloy, matrix, crystalline ceramics, glass-ceramics and unreprocessed fuel	Nepheline syenite glass blocks containing aged fission products have been subjected to a long-term leaching trial since the early 1960s. The Waste Immobilization Process Experiment (WIPE) currently under construction, is a demonstration vitrification unit with a production capacity of 10 kg h^{-1} of glass.	Granite	Construction of an underground laboratory at Whiteshell is planned to study geology, mining techniques, heat transfer and back filling performance. Generic research in the 1980s is planned to be followed by a demonstration repository in the 1990s, and by a commercial repository after the year 2000 for disposal of either reprocessed fuel or HLW.
Denmark	–	–	Salt	Salt domes in Jutland are under examination. The program includes field studies, design and safety assessments for a repository supporting a 5 000 MWe nuclear program for 30 years, with burial assumed from 2010.

TABLE 3 (cont.) – STATUS OF SOLIDIFICATION AND GEOLOGIC BURIAL OF HIGH-LEVEL WASTES FROM COMMERCIAL THERMAL NUCLEAR POWER GENERATION

Country	High-level waste solidification		Geologic burial	
	Preferred waste forms	Developments	Preferred formations	Developments
German Democratic Republic (GDR)	–	–	Salt	A waste repository is being developed at Bartensleben.
Federal Republic of Germany (FRG)	Borosilicate glass, glass ceramics, phosphate glasses	A vitrification plant, PAMELA, using FRG technology is under construction at Mol. Pilot scale vitrification development is being carried out at Karlsruhe.	Salt	The disused Asse salt mine, used as a repository for low-level radioactive wastes since 1967 is being used for *in-situ* testing of thermal dissipation in salt. The salt dome at Gorleben is currently being investigated for possible siting of a high-level waste repository; a decision on site suitability is expected in the early 1990s.
Finland	–	–	Crystalline rock	Risk assessment studies, economic evaluations.
France	Borosilicate glass	The industrial scale AVM plant at Marcoule commenced operation in 1978. About 540 m^3 of HLW corresponding to reprocessing of over 9 000 t of natural uranium gas-graphite fuel were vitrified into 250 t of glass by early 1981. Two larger plants of similar design are scheduled to be operational at Cap La Hague in 1986 to solidify waste from up to 1 600 t U a^{-1} LWR fuel, corresponding to a nuclear generation capacity of about 40 000 MWe.	Granite	It is intended to store glass blocks for 30 to 50 years in air cooled vaults. A repository for disposal of low and intermediate level waste is expected to be operational by 1990. Sites for HLW disposal are under investigation. It is planned to select a site in 1988 for construction of an underground research laboratory with burial planned from 1991.
India	Borosilicate glass	Commissioning of a vitrification plant at Tarapur commenced in 1982. The plant has a capacity of about 120 kg d^{-1} of glass. Similar plants are planned at Kalpakkam and at Bhabha Atomic Research Centre.	Granite	Burial after 30 years of storage in an air-cooled underground vault is planned.

Table 3 – Continued

Italy	Borosilicate glass	Small scale demonstration vitrification plants are under consideration.	Clay, salt	The sediments near the Trisaia Centre in South Italy are under study.
Japan	Borosilicate glass, zeolite ceramics	Vitrification technology has been developed since 1976. The Vitrification Pilot Plant is planned for construction in the late 1980s.	Granite, diabase, shale, zeolitic tuff, limestone, schist	Storage of glass for up to 50 years. Underground disposal in Japan on an experimental basis may commence in 2015.
Netherlands	–	–	Salt	Inland salt domes are under study; former consideration of salt domes under the North Sea has been discontinued on grounds of cost.
Sweden	Borosilicate glass, unreprocessed fuel	–	Granite	A 3-year international study of geologic disposal commenced in 1981 at Stripa and is an *in-situ* experiment scheduled for completion in 1986.
Switzerland	Borosilicate glass	–	Granite	Feasibility studies for burial at depths of up to 1 500 m are in progress.
Spain	Borosilicate glass	Use of the West German PAMELA process under licence is under consideration.	Salt, shale, ceramite	A pilot plant repository is under consideration for the late 1980s.
UK	Borosilicate glass	The FINGAL vitrification process was developed between 1962 and 1966. This technology was the basis for the HARVEST engineering scale work commenced in the early 1970s. This national program was abandoned in 1981. French technology is being used for construction of an industrial scale vitrification plant scheduled to be operational in the late 1980s.	Granite	Some experimental drilling tests have been conducted in North Scotland but were discontinued in December 1981. It is proposed to store the vitrified waste at the surface for at least 50 years.

Table 3 – Continued on page 26

TABLE 3 – STATUS OF SOLIDIFICATION AND GEOLOGIC BURIAL OF HIGH-LEVEL WASTES FROM COMMERCIAL THERMAL NUCLEAR POWER GENERATION (Cont.)

Country	High-level waste solidification		Geologic burial	
	Preferred waste forms	Developments	Preferred formations	Developments
USA	Unreprocessed fuel; borosilicate glass, titanate ceramics, SYNROC, cermets, cement, coated particles, super calcine, metal matrix materials	The waste solidification engineering prototype plant (WSEP) had a capacity solidifying wastes from 1 t U d^{-1} of fuel. About 50 MCi of radioactive waste was encapsulated between 1966-1970. The Nuclear Waste Vitrification Plant (NWVP) in 1979 vitrified high-level waste containing about 0.4 MCi from reprocessing 1.5 t U of LWR fuel. 260 kg of glass was produced in spray calciner in-can melters, 20 cm diameter x 2.4 m height, which served as encapsulating canisters for the waste.	Salt, granite, basalt	A waste isolation pilot plant (WIPP) in New Mexico is under construction in salt for operation by late 1980s for disposal of transuranic waste of military origin. The Nevada terminal waste isolation project in granite is in use as test site for temporary (5 year) geologic emplacement of commercial spent fuel. The first National Terminal Waste Storage (NWTS) site is scheduled for operation by 1998.
USSR	Alumino-phosphate glasses	No details available	Salt	Studies have been reported on near-surface storage facilities for vitrified high-level wastes. Over 10^8 Ci has been reported disposed of by injection of liquid waste containing about 1 Ci l^{-1} into deep sandstone formations.

(Continued from page 22)

technical-industrial scale. This process, named PAMELA, is intended to incorporate waste from former reprocessing operations into borosilicate glass beads embedded in a lead matrix, or into blocks of borosilicate glass. Other countries with plans for vitrification plants include the FRG, Italy and Japan.

Industrial commitment to the French vitrification technology is illustrated in Figure 1. Reprocessing commitments by France and the UK to the mid-1990s include contracts for reprocessing 9 150 t U from spent LWR fuel from other countries, and 10 850 t U from domestic fuel (CEA-Cogema, 1981; Jones and Pearce, 1981). High-level wastes from reprocessing 20 000 tU, equivalent to about 660 GWe a year of installed nuclear power, will therefore be immobilised in borosilicate glass; this is equivalent to vitrifying wastes from more than three years of the present installed nuclear power program for the whole world (199 GWe in 1984).

Development of Advanced Waste Forms – SYNROC

Mineral-like crystalline ceramics have been under development at Pennsylvania State University for 10 years. These are refractory waste forms in which the fission products and actinide elements are immobilised in the crystal lattices (McCarthy and Roy, 1981). Unlike glasses, crystalline waste forms are thermodynamically stable, although they are more susceptible to radiation damage. The work is aimed at the development of a form with outstanding chemical durability which alone could minimise releases of radionuclides, even under the worst conceivable case of repository failure.

The SYNROC formulation, proposed by Professor A. E. Ringwood of the Australian National University (ANU), is the leading alternative waste form which is being developed in a collaborative program between the ANU and the AAEC. Ringwood (1980a) identified three refractory and leach-resistant minerals occurring in nature – zirconolite ($CaZrTi_2O_7$), perovskite ($CaTiO_3$) and hollandite ($BaAl_2Ti_6O_{16}$) – which have demonstrated stability for millions of years in a wide range of geologic and geochemical environments. When combined, these minerals are capable of accepting most of the high-level radioactive waste constituents into the crystal lattices.

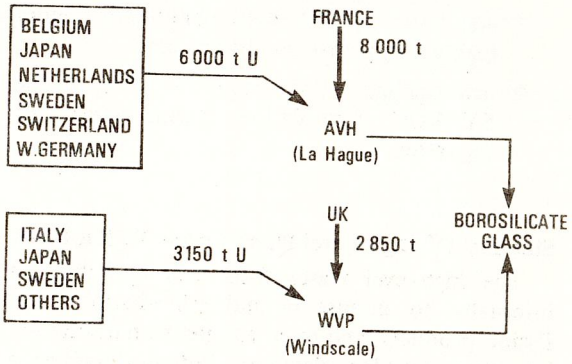


Figure 1. Reprocessing commitments by France and the United Kingdom in 1995.

Leach rates of alkali caesium (Cs) and alkaline-earth strontium (Sr) elements from non-radioactive SYNROC at 100°C into water which was replaced daily have been up to 1 000 times lower than those from borosilicate glass; under static conditions, where water was saturated with leach material at 100°C, leach rates from SYNROC were typically one-tenth of those from glass. The leach rate of SYNROC does not appear to increase greatly with temperature (Reeve et al., 1981). Specimens of non-radioactive SYNROC have been irradiated in a nuclear reactor to simulate radiation damage effects in waste stored up to 8×10^5 years. All specimens were intact after irradiation.

The relative merits and disadvantages of the alternative forms for immobilising commercial high-level wastes have been extensively reviewed (Johnston and Palmer, 1982; Mendel et al., 1981; CEC, 1981; IAEA, 1980; USDOE, 1981; Wald et al., 1980; USNRC, 1979). SYNROC has been ranked high in research potential. Japan and UK are collaborating with Australia on SYNROC research and development programs.

The program at the Lucas Heights Research Laboratories involves the fabrication of non-radioactive SYNROC on a multi-kilogram scale and continuation of radiation damage and leach tests (Reeve et al.,1983). This includes:

- preparation of, and leach tests on SYNROC samples containing radioactive fission products and actinides – laboratory scale equipment for manufacture of these samples has been constructed;
- assessments of the engineering practicability

and costs of SYNROC manufacture under highly radioactive conditions; and

- investigation of the reproducibility of the SYNROC formulation during large-scale operation.

Stability Of High-Level Waste Forms To Radiation

The high-level waste form will be subjected internally to intense β- and γ-irradiation from fission products, and to α- and neutron irradiation by some actinide elements. Potential radiation damage effects include displacement of constituent atoms, rupture of chemical bonds, valency changes, transmutation of radioactive nuclei into different elements, build-up of internal energy, and deposition of helium atoms in the waste form.

While the majority of radiation damage studies have been made on glasses containing high-level waste, comparatively little work has been done on the radiation stability of ceramics. Radiation studies on different waste forms have been reviewed by Mendel et al. (1981).

The effects of radiation are being studied in accelerated tests. High concentrations of short-lived β- and α-emitters will deliver in a few years doses to samples equivalent to those to be received by a real waste form over periods exceeding hundreds of thousands of years. Some tests in France and the FRG have delivered doses of β-radiation to glasses equivalent to doses from the storage of LWR wastes over several millions of years, without a detectable change in the glass structure (Amaury, 1979). In British tests, doses up to 5×10^{18} α decays/gram, equivalent to more than 10^5 years of storage, have been delivered to glasses; the resultant increase in leach rate was less than a factor of two (Marples, 1982; Roberts, 1980). Tests in the US have shown that changes to leachability or mechanical strength detected in glasses after α-doses equivalent to over 10^5 years of storage (Platt and McElroy, 1978) were not significant in an appropriately engineered disposal system.

Status Of Geologic Disposal Of High-Level Waste

No country disposes of high-level radioactive wastes at present. Many countries have active research programs involving *in situ* experiments and geological investigations for the establishment of demonstration and commercial high-level waste repositories (see Table 3). Geologic media under study include bedded and domed salt deposits, crystalline rocks, basalt, clay and tuff (OECD/NEA, 1982a).

The advantages and disadvantages of the various rock types being considered for waste repositories are set out in Table 4. No formation is without some disadvantages; the IAEA (1982a) recently concluded that a site with ideal characteristics is not essential for siting an appropriately engineered waste repository.

Major design efforts have been concentrated on the placement of high-level waste in land-based mined repositories. Disposal of waste in deep drilled holes, which has been advocated by Ringwood (1980b), could be regarded as a variant of the mined repository concept. The deep-hole proposal has to some extent been neglected, although countries with small nuclear power programs have shown interest (Elsam and Elkraft, 1981).

Geologic disposal of high-level waste (HLW) awaits identification of technically suitable and publicly acceptable sites. Local opposition to the siting of repositories has been a significant impediment to the demonstration of HLW disposal. Investigations of potential geologic sites have been deferred in a number of countries and discontinued in the UK. Sweden, the FRG, Canada and the USA are leading the search with *in-situ* studies of their geologic formations (OECD-NEA, 1983; USDOE, 1983b; Shemilt, 1982; Salander et al., 1980). The Dutch authorities have recently proposed that the OECD/NEA initiate a study of the concept of an international waste repository.

There has been a recent press report that the Republic of China has concluded an agreement with a West German consortium to act as agents for storing radioactive waste from Western European countries in China. It is reported that some 4 000 t could be involved up to the year 2000 and stored in the Gobi Desert at a charge of $US1500/kg. No country has announced interest in the offer by China.

The USA has the most advanced plans for waste disposal. The Nuclear Waste Policy Act of 1982 specifies a clear approach for the siting, licensing and development of repositories. Candid-

TABLE 4 – GEOLOGIC FORMATIONS FOR ULTIMATE DISPOSAL OF HIGH-LEVEL WASTES

Type of formation	Advantages	Disadvantages	Investigating countries
Rock salt comprising bedded salt and dome salt (from diapirs)	1. High thermal conductivity 2. Plastic condition at low temperature and pressure ensures fractures are self-sealing 3. The existence of rock salt indicates its isolation from circulating waters throughout geologic times past 4. Very low permeability	1. A natural resource commonly associated with other resources such as oil and gas 2. Subject to creep under load that accelerates with increase in pressure associated with deep burial, and increase in temperature associated with heat-generating radioactive wastes 3. May contain up to 3% bitterns in which the salt is highly soluble under the influence of heat 4. Bitterns that migrate to the waste canister walls in response to canister heat form pockets of highly corrosive fluid 5. Many radioactive elements in waste will dissolve if they come in contact with bitterns 6. Poor ion-exchange properties	German Democratic Republic (GDR) Netherlands USA Federal Republic of Germany (FRG)
Crystalline rock (granite, gneiss, granodiorite, gabbro etc.)	1. Underground openings are self-supporting or require minimum support 2. Fractured-rock permeability decreases with depth, and many such rocks have very low permeability 3. Weathered rock adjacent to fractures has ion-exchange properties 4. Large diameter holes can be drilled to great depths where temperatures exceed 300°C	1. Could be a major source of energy in the future, i.e. rock with a thermal gradient of 20°C/1 000 m 2. Fractures are not self-sealing 3. Low permeability backfill material required to seal repository 4. The larger the underground opening, the more fractures intersected	Austria Canada Denmark France India Japan Sweden Switzerland

TABLE 4 (cont.) – GEOLOGIC FORMATIONS FOR ULTIMATE DISPOSAL OF HIGH-LEVEL WASTES

Type of formation	Advantages	Disadvantages	Investigating countries
	5. Low thermal gradients are present in some areas (10°C per 1 000 m) 6. The regional groundwater system can be accurately modelled to predict flowlines and travel times		UK* USA
Basalt	As for crystalline rock above except for (2) and (6): 2. Zones with very low permeability do exist, but permeability does not necessarily decrease with depth 6. The regional groundwater system can be modelled, but it is more complex owing to variations in numbers of fractures between individual basalt flows and interbedded tuffs	As for crystalline rock above except for (1), and the addition of another point: 5. Because some basalts are highly fractured and permeable, selection of a low permeability thickness of basalt will require considerably greater effort in proving its suitability than would be required for crystalline rock	USA
Argillaceous formations (1) Clay	1. Good plastic characteristics and unsupported openings would be self-sealing 2. No current value as a resource 3. Very low permeability within the clay itself 4. Good ion-exchange properties	1. Subject to creep and all openings will require full support and full lining 2. The construction of a fully lined repository at depths greater than 250 m may not be possible or the cost may be prohibitive 3. High permeable sands are commonly interbedded with clays 4. Chemical reactions take place within the clay with increase in temperature, and a formation temperature above 100°C is not recommended 5. Little information is available on the migration of fluids within the clay in response to a thermal gradient	Belgium Italy

* The UK recently decided [UK Hansard, 1981] to cease current geological exploration activities in view of its intention to prolong temporary storage of high-level waste.

TABLE 4 (cont.) – GEOLOGIC FORMATIONS FOR ULTIMATE DISPOSAL OF HIGH-LEVEL WASTES

Type of formation	Advantages	Disadvantages	Investigating countries
(2) Shale and tuff	1. Less plastic than clay and unsupported openings would readily deform 2. No value as a resource unless containing hydrocarbons 3. Very low permeability expected, but some variation between individual beds 4. Good ion-exchange 5. Large diameter holes can be drilled to considerable depths and kept open by the use of dense circulating muds	1. All openings require support and full lining 2. The construction of a repository would be possible at depths greater than 250 m, but deformation of the installation would increase with depth 3. Some highly permeable beds may be part of the sequence 4. Reaction to a thermal gradient expected to produce changes in the clay minerals and their properties	Italy USA

(Continued from page 28)

ate rock types for the first repository are basalt, tuff and salt; a decision on the site is due in 1987. The first repository is scheduled to begin operation in 1998 to be followed by a second in 2002 although several years' delay in recommending a repository site has been reported (Nuclear Fuel, 1984). The repositories are required to accept both spent fuel and vitrified HLW.

Confidence in the disposal of high-level radioactive waste on or in the deep ocean floor as a technically acceptable alternative to land-based repositories has been expressed by France, Japan and the US (Anderson, 1982), and is the subject of considerable research effort from members of the OECD's Nuclear Energy Agency (OECD-NEA). Active participants in the OECD-NEA Seabed Working Group Program include Canada, the Commission of European Communities (CEC), France, the FRG, Japan, the Netherlands, Switzerland, the UK and the USA, and countries having observer status are Belgium and Italy. At present, dumping of HLW into the oceans is expressly prohibited under an international convention. However, there is current uncertainty on whether subsealed emplacement of waste into deep ocean sediments would constitute dumping. Differing views have been expressed on this question both within the US and in other nations. Major political questions on the use of the seabed are expected to occupy international forums for some years before the proposal could become a realistic option.

Isolation of Buried Waste

Driving forces which might return some of the buried radionuclides to the biosphere have been reviewed by Burkholder (1979) and are illustrated in Figure 2. Contact of buried waste with groundwater and the leaching of radioactive constituents from the waste form is generally considered to be the most probable pathway. The release of radionuclides from waste forms by leaching involves complex mechanisms of selective leaching and matrix dissolution. These mechanisms are not completely understood.

No waste form yet developed is totally insoluble in water; the rate of leaching depends on the type and composition of the waste form (glass or ceramic), the temperature, flowrate and composition of the water, and the exposed surface area of the waste. The various radioactive elements within the waste also have widely differing leach rates. Actinide elements leach 100 times more slowly than alkali metals from borosilicate glass. In addition, leach rates in water at 100°C are 35-50 times greater than at 20°C (Mendel et al., Amaury, 1979).

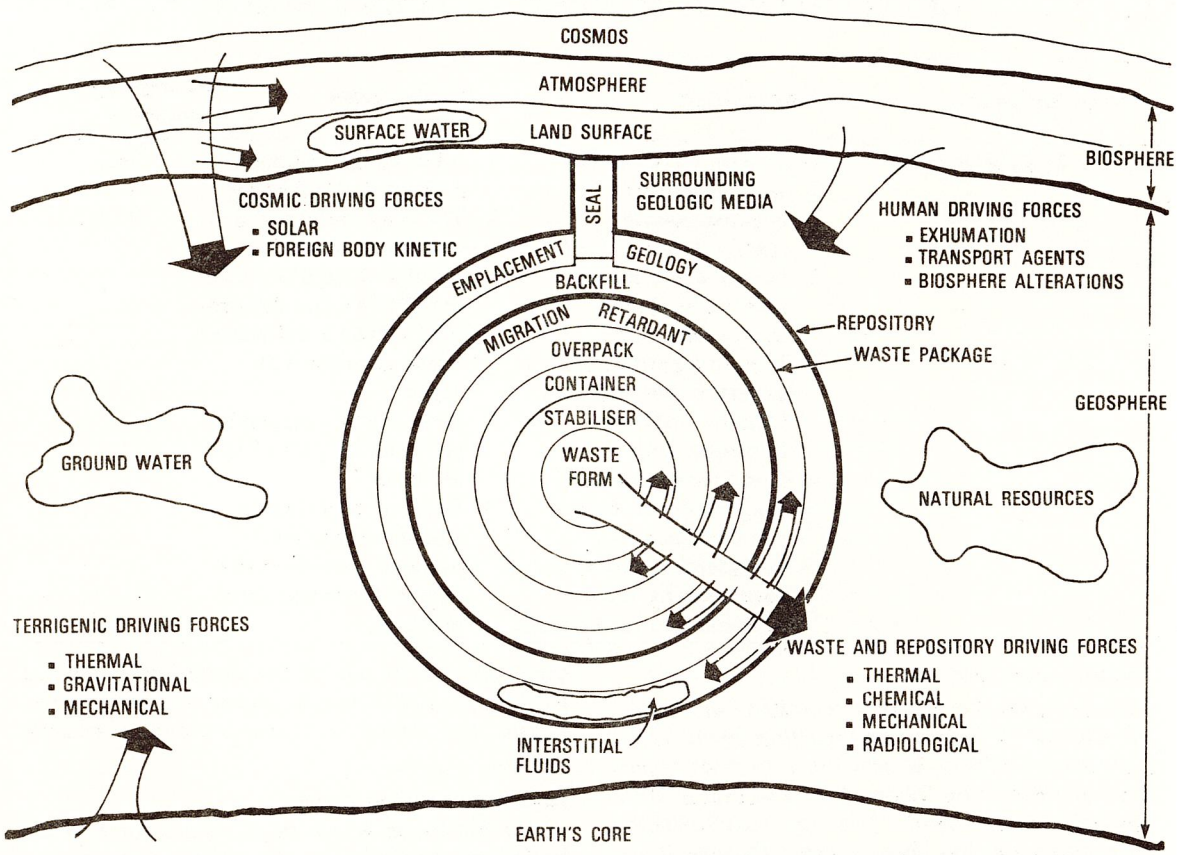


Figure 2. Waste isolation system and driving forces (after Burkholder, 1979).

Leach rates as low as 5 x 10^{-7} g cm^{-2} d^{-1} have been measured for Cs/Sr from some French glasses at 20°C. A conservative value of 10^{-5} g cm^{-2} d^{-1} has been assumed in environmental impact studies in the UK (Hill, 1979) and the USA (USDOE, 1980). This corresponds to a bulk dissolution rate of 0.014 mm a^{-1}, or a dissolution time of about 3 500 years for waste fragmented into pieces 10 cm in diameter.

Potentially, the greatest leach rates are during the initial few hundred years after burial, owing to local increase in temperature of the geologic medium and contained groundwater through fission product decay heat. Glasses in contact with water at high temperature (300°C) and high pressure (30 MPa) undergo rapid hydration, leading to fragmentation of the sample and leaching of about 30% of the Cs from the glass in 2 weeks (McCarthy et al., 1978).

High temperature leach tests under reflux in the laboratory have been criticised as being unrepresentative of waste disposal conditions which could reasonably be expected in geologic burial (Savage and Chapman, 1981). At groundwater velocities of 1 to 2 m a^{-1}, which are expected for crystalline rocks selected for waste disposal, chemical saturation effects could theoretically limit leach rates to less than 10^{-9} g cm^{-2} d^{-1} (Chapman et al., 1980). In an 88-day test under near-stagnant conditions at 200°C and 50 MPa, leach rates of borosilicate glass fell from 3 x 10^{-4} to 2 x 10^{-5} g cm^{-2} d^{-1} (Savage, 1981).

Temperatures can be lowered by reducing the concentration of fission products in the waste, storing the waste for some tens of years before burial to reduce the heat from fission product decay, and allocating adequate space for waste canisters in the geologic medium. Sweden has expressed an intention to restrict temperatures of the buried high-level waste and unreprocessed spent fuel to below 100°C by these methods (KBS-1, 1978; KBS-3, 1983). Maximum temperatures above 350°C are apparently still under consideration for vitrified commercial high-level waste in the USA (Westinghouse, 1983). More measurements of hydrothermal leaching data are required under realistic repository conditions using actual radioactive high-level wastes and a range of geologic media (Johnston and Palmer, 1982).

However, the possibility of entry of an aquifer into a repository after major ground movement, though remote, cannot be totally excluded, and geologic burial strategy employs sequential independent barriers to retain the radioactive elements underground. Waste packages have been designed to prevent access of water to the primary waste form for up to 1 000 years. In these packages, the radioactive glass or fuel is sheathed in a corrosion resistant metal, surrounded by an absorbent overpack, e.g. bentonite clay. The overpack has two functions: to hydrate, swell and seal the inner package against major water ingress, and to provide an environment which can absorb radioactive materials which might eventually be leached from the waste form.

Isolation from the biosphere is further assured if escaping radionuclides or heavy metals are trapped by chemical interaction or adsorption processes in the repository (Roy, 1981). This could occur either on the host rock or in adjacent geologic strata. Groundwater systems are determined by many complex factors. Hydrogeologic conditions at each prospective disposal site must be sufficiently understood that a reliable prediction of the possible interaction between the groundwater, the geologic medium and the emplaced waste can be made.

The complex geochemical mechanisms influencing the sorption of labile radionuclides (OECD-NEA, 1982b) indicate that engineered clay barriers may be particularly important in retention of long-lived actinide elements. Minimum performance criteria have been published for HLW packages and underground facilities in the US (USNRC, 1983). These are to be both designed and located so that, assuming anticipated processes and events, and including full or partial saturation of the geologic medium by water,

- the waste packages will contain substantially all radionuclides for at least 300 years, and up to 1 000 years after permanent closure;
- at any subsequent time, the annual release rate of radionuclides into the geologic medium will not exceed 10^{-5} a^{-1} of the quantity remaining after 1 000 years after closure, excluding those radiouclides contributing less than 0.1% of the total release rate limit;
- geologic sites will be selected in areas where groundwater will take at least 1 000 years to travel from the repository to the accessible environment.

Potential Radiation Doses from a High-Level Waste Repository

Assessments of radiation dose rates resulting from ingress of water into a high-level burial site have involved hypothetical models for the release of radionuclides and their rates of transport to and uptake in the human environment. The models have ranged from simple consequence analyses, which assumed worst case or realistic values for parameters used in the models, to sophisticated probabilistic analyses, which recognised the statistical variations in parameter values and their interactions on radiological dose (IAEA, 1981). The results of several dose assessment studies were reviewed and compared by Koplik et al. (1982).

Consequence analyses have considered geologic disposal of both vitrified high-level waste and unreprocessed spent fuel. The waste or fuel was assumed to be stored for up to 40 years to reduce heat from fission product decay, followed by

encapsulation of vitrified waste in corrosion resistant canisters of titanium and lead (KBS-1, 1978), and encapsulation of unreprocessed fuel in copper canisters filled with compressed copper powder (KBS-2, 1978; KBS-3, 1983).

The canisters were packed in bentonite and buried at depths of at least 500 m in geologic formations including granite, basalt, volcanic tuff and salt deposits. The consequences of repository failure in the first year after filling and decommissioning have been assessed (USDOE, 1979). However, this case was discounted as a credible situation and it was considered that detection and remedial action would protect individuals from the high estimated doses (USDOE, 1980).

The waste packages were assumed to be breached by groundwater at a minimum of 1 000 years after burial, when temperatures have fallen to about 25°C. Leaching of long-lived radionuclides occurred, and the contaminated groundwater percolated slowly upwards through the geologic medium and entered aquifers, surface waters or oceans. Retardation and decay of the radionuclides during transport to the biosphere were calculated from adsorption coefficients measured on the particular geologic media. Radiological doses were estimated for hypothetical populations exposed by drinking water from deep wells near the repository, using water from rivers or lakes to irrigate food crops, and by ingestion of fish and exposure to sea water and coastal sediments.

Estimates of dose from unit ingestion of certain α radionuclides have been revised on the basis of recent metabolic and radiological data (ICRP, 1979). These revisions reduced the dose estimates for radium 226 by a factor of 25, increased estimates for plutonium 239, americium 241 and curium 244 by factors of 7, 30 and 25, respectively, whereas the dose estimates for neptunium 237 were increased 250-fold. Use of this revised data has resulted in significant increases in doses estimated in earlier studies (IAEA, 1982b).

Neptunium 237 has become significant in dose estimation largely as a result of a 100-fold increase by the International Commission on Radiological Protection (ICRP) of the fractional absorption value for all neptunium compounds from the gastro-intestinal tract. The recommended value is 0.01 but there is considerable uncertainty about the data, which were based on a small number of experiments on rats. The ICRP noted that absorption of trace quantities of neptunium may be a factor of ten lower, as also may be the absorption of neptunium incorporated in food (ICRP, 1979). The high fractional absorption value may have been due to the presence of neptunium as the NpO_2^+ ion (IAEA, 1982B), and this may not be the principal chemical form of neptunium released to the biosphere from high-level waste. A recent review in the UK indicated that a fractional absorption value of 0.001 was more appropriate for adults ingesting neptunium in inorganic forms and forms likely to be encountered in food and water. However, values up to 0.01 were considered applicable to infants during the first year of life (NRPB, 1984). Evaluation of factors influencing the transfer fraction for neptunium is part of the AAEC research program at the Lucas Heights Research Laboratories.

The level of radiological dose estimates in the consequence studies depends on the validity of the mathematical model and the values of parameters used for calculation. Some studies on disposal of vitrified HLW have used combinations of parameter values which may be regarded as conservative (Hill, 1979; Hill and Lawson, 1980; Burton and Griffin, 1981). Other studies have used parameter values considered more representative of disposal of vitrified waste and unreprocessed fuel at carefully chosen repository sites (Pigford et al., 1983; Pigford, 1983), while some other studies have examined the effect of using both 'pessimistic' and 'expected' parameter values for disposal of high-level waste (KBS-1, 1978) and unreprocessed spent fuel (KBS-2, 1978; KBS-3, 1983).

Estimates, using combinations of parameters regarded as conservative or pessimistic, resulted in maximum radiation dose rates in the distant future several times that received from an 'average' natural radiation background, with the majority of the dose rate being derived from neptunium 237. These studies generally assumed a major influx of groundwater into the repository at water velocities up to 100 m a^{-1}, representative of highly fractured rock or sandy soils (Open University, 1975). Corrosion of the waste canisters was followed by dissolution of the waste form and all its radioactive constituents over periods of 3 000 – 30 000 years. Rapid transport of contaminated groundwater to the biosphere was assumed during periods

TABLE 5 – ESTIMATES OF THE MAXIMUM ANNUAL INDIVIDUAL RADIOLOGICAL DOSE FROM A HIGH-LEVEL WASTE REPOSITORY USING CONSERVATIVE OR UNFAVOURABLE PARAMETERS

Reference	KBS–1 modified*	Hill (1979)	Hill & Lawson (1980)	Burton & Griffin (1981)	Time of maximum dose years (a)
Radionuclide	mSv a^{-1}	mSv a^{-1}	mSv a^{-1}	mSv a^{-1}	
Technetium 99^m	6×10^{-2}	7×10^{-2}	5×10^{-2}	10^{-2}	$200 - 6 \times 10^3$
Iodine 129	10^{-3}	8×10^{-2}	5×10^{-2}	–	$200 - 6 \times 10^3$
Caesium 135	6×10^{-3}	3×10^{-4}	2×10^{-5}	3×10^{-4}	$9 \times 10^4 - 10^6$
Radium 226	2×10^{-3}	3×10^{-2}	10^{-3}	2×10^{-4}	$5 \times 10^4 - 10^6$
Uranium 233	10^{-1}	6×10^{-7}	4×10^{-7}	4×10^{-5}	$2 \times 10^4 - 5 \times 10^4$
Uranium 234	10^{-2}	10^{-5}	4×10^{-7}	10^{-6}	$3 \times 10^4 - 10^6$
Neptunium 237	25	60	5	10	$10^4 - 2 \times 10^5$
Plutonium 239	2×10^{-6}	2×10^{-9}	2×10^{-11}	3×10^{-9}	$4 \times 10^5 - 3 \times 10^6$
Maximum	25	60	5	10	

* Data adjusted to revise dose factors in ICRP (1979).

between 100 and 400 years, and low retention factors† chosen for neptunium corresponded to the location of the repository in a geological medium with oxidising characteristics. Table 5 lists the maximum radiological doses to individuals and the major radionuclides responsible for the dose from the waste repository from some consequence studies using conservative parameters. At the time corresponding to the maximum individual dose (10^4 to 2×10^5 a), neptunium 237 was predominant, contributing more than 99% of the maximum dose rate in the range 5 to 30 times that of an 'average' natural radiation background. It is of interest that the contribution from plutonium 239, a radionuclide of major public concern, was estimated to be less than 10^{-6}% of the maximum dose.

Studies in the UK using conservative data indicated some advantages for siting a repository on the coast. The effect of different food chains and the dilution mechanism resulted in annual dose rates of about one twenty-fifth of an 'average' natural background (Hill & Lawson, 1980).

By contrast, estimates using optimistic data measured at prospective geologic sites, and considered more realistic of expected repository conditions, resulted in calculation of maximum dose rates corresponding to a fraction of an 'average' natural radiation background, derived from neptunium 237, technetium 99, iodine 129, radium 226, and isotopes of thorium and uranium.

Examples of these estimates, given in Table 6, assumed low rates of water flow through the repository, increased dissolution time for radioactive waste constituents, and increased retention factors for neptunium corresponding to a rock with reducing properties. Groundwater velocities measured in unfractured rock strata were normally less than 1 m a^{-1} (KBS-3, 1983; Pigford, 1983). This velocity corresponded to the access of a few litres of water to each waste canister/a. Under these conditions, the rate at which waste constituents could dissolve in the groundwater was limited by solubility constraints. This resulted in expected dissolution times of 3 million years for vitrified waste (KBS-1, 1978) and 1 to 7 million years for unreprocessed spent fuel (KBS-2, 1978; KBS-3, 1983). Water transport times to the biosphere were in the range 100 years (KBS-3, 1983) to $10^4 - 10^5$ years (Pigford, 1983), and neptunium retention factors up to 23 000 in the Swedish studies. Implicit in the low doses predicted in Table 6 is the assumption that the repository is sited in a geologically and seismically stable area,

† Radionuclide transport time = groundwater transit time x retention factor

TABLE 6 – ESTIMATES OF THE MAXIMUM ANNUAL INDIVIDUAL RADIOLOGICAL DOSE FROM A HIGH-LEVEL WASTE REPOSITORY USING PROBABLE OR EXPECTED PARAMETERS

Reference	KBS–1* (1978)	KBS–2* (1978)	KBS–3* (1983)	PIGFORD† (1983)	Time of maximum dose (years)
Radionuclide	**mSv a^{-1}**	**mSv a^{-1}**	**mSv a^{-1}**	**mSv a^{-1}**	
Carbon 14	–	–	–	10^{-3}	10^{4}
Technetium 99^{m}	8×10^{-4}	2×10^{-2}	4×10^{-8}	2×10^{-5}	$2 \times 10^{4} - 10^{6}$
Iodine 129	2×10^{-5}	10^{-2}	3×10^{-4}	3×10^{-6}	$2 \times 10^{4} - 5 \times 10^{6}$
Radium 226	2×10^{-5}	9×10^{-3}	2×10^{-4}	5×10^{-7}	$6 \times 10^{5} - 5 \times 10^{9}$
Thorium 229-230	3×10^{-4}	10^{-2}	10^{-6}	–	$10^{5} - 10^{7}$
Uranium 223-2398	10^{-3}	3×10^{-2}	6×10^{-7}	2×10^{-6}	$5 \times 10^{5} - 5 \times 10^{9}$
Neptunium 237	9×10^{-3}	10^{-9}	6×10^{-6}	10^{-2}	$10^{4} - 10^{7}$
Maximum	10^{-2}	5×10^{-2}	3×10^{-4}	10^{-2}	

* Data adjusted to revised dose factors in ICRP (1979)

† Waste inventory 10 x that in KBS studies. Data refer to non-solubility limited waste dissolution in a basalt repository.

and that no major influx of groundwater will occur over extremely long periods of time.

Low maximum individual radiological doses are also predicted by probabilistic scenario analyses. The Canadian Systems Variability Analysis Code (SYVAC) has been developed to model the processes occurring in the repository, the geosphere and the biosphere, and to take into account uncertainties in the data associated with those processes (Dixon and Rosinger 1981).

In SYVAC, the data obtained from laboratory and field studies are used in the form of parameter distributions rather than 'best estimate' or 'conservative' single values. A value for each parameter is sampled in turn from its distribution to form a set. This set of values defines a 'scenario'. The SYVAC then determines the transport of radionuclides from the vault to the biosphere for this scenario, and estimates an individual dose. Repeated sampling produces different scenarios for which consequences are determined. Of 1 730 estimates of radiation dose to the most exposed individual, the great majority were reported to be less than 10 μSv a^{-1}, and in no case did the dose exceed 1 mSv a^{-1}, i.e. less than 50% of 'average' natural background.

Cost of High-Level Waste Disposal

A recent estimate of costs of conditioning and disposal of both spent LWR fuel and reprocessing wastes into a granite repository have been reported by the Swedish Nuclear Fuel Supply Company (SKBF, 1982) for the entire waste management operations supporting a power program of 300 GWe a^{-1}. These estimates include long-term storage facilities for spent fuel and vitrified waste, and also of repositories for reactor operational and decommissioning wastes. The levelised cost for the Swedish waste management program was equivalent to about 0.26 cents/kWh (undiscounted), or 0.14 cents/kWh at a discount rate of 10% a^{-1}. This latter value is equivalent to about 3% of a power generation cost of 5c/kWh.

Radiation Doses and Risks from a High-Level Waste Repository in Perspective

The annual doses and risks from living near a high-level waste repository in the far distant future may be placed in perspective by comparison with doses received today from nuclear power and its fuel cycle and from the natural radiation background.

An average global annual radiation dose of 0.0006 mSv a^{-1} has been estimated from nuclear

power production. In the UK, the estimated average annual dose from nuclear energy is 0.003 mSv; maximum individual annual doses to members of the public during the late 1970s were 1.5 mSv from reprocessing, 0.3 mSv from nuclear reactor operation and 0.06 mSv from upgrading and fuel manufacture (NRPB, 1981). British Nuclear Fuels Ltd estimated a lower maximum annual dose of 0.9 mSv for reprocessing in 1981 (BNFL, 1982).

Estimates of 'average' annual radiological doses from natural background have recently been increased to 2 mSv a^{-1} (UNSCEAR, 1981). The wide range of potential annual radiation doses from natural background sources is illustrated in Table 7. Thorium in beach sands at Kerala, India, contribute to an abnormally high background of up to 20 mSv a^{-1}. In Australia, sunbathing for 2 hours/day on Kingscliffe Beach in New South Wales can result in an external dose of 0.3 mSv a^{-1}, which is equivalent to a continual exposure rate of 3.8 mSv a^{-1}.

The presence of uranium in igneous rocks results in increased natural background doses from the accumulation of radon gas and its decay products in dwellings. A recent survey in granite areas of the south-west and northern UK has indicated doses of 5 mSv a^{-1} in 100 000 houses, 25 mSv a^{-1} in about 1 000 houses, with maximum doses of 100 mSv a^{-1} (NRPB, 1983). Regulatory proposals include a design level of 5 mSv a^{-1} for new buildings and a rehabilitation action level of 25 mSv a^{-1} for existing houses.

By comparison, the maximum individual dose rate estimated in consequence analyses using optimistic data ranged from 3×10^{-4} to 5×10^{-2} mSv a^{-1} (KBS-3, 1983; KBS-2, 1978). Analyses using a combination of conservative or unfavourable data indicated maximum dose rates in the range of 5 to 60 mSv a^{-1} for an inland repository (Hill & Lawson, 1980; Hill, 1979), and 0.08 mSv a^{-1} for a repository sited in a coastal geologic formation. While the upper limit of this range would be unacceptable for repository design, it may be noted that dose rates within the range are being delivered from natural background sources in India and the UK. Probabilistic analyses suggested doses less than 1 mSv a^{-1} (Dixon and Rosinger, 1981).

The risk of death from radiation-induced cancer averaged over age and sex (NHMRC, 1981) is about 1.25×10^{-2} Sv^{-1}. Exposure to a natural radiation background of 2 mSv a^{-1} carries, therefore, a proportionate death risk of 2.5 in 100 000 /a^{-1} of exposure.

TABLE 7 – ANNUAL INDIVIDUAL RADIATION DOSE TO HUMANS FROM NATURAL BACKGROUND SOURCES

Source	'Average' annual dose mSv a^{-1}*	Range mSv a^{-1}
Natural		
External		
Cosmic rays (sea level)	0.3	0.3 – 1.3 (2 500 m)
Ground and building materials	0.3	0.2 – 100**
Internal		
Potassium 40 in body	0.3	
Water and food ingested	0.1	
Air	1.2	
Total – natural	2.1	2.0 – 100
Man-Made Radiation		
1 Chest X-ray	0.4	0.2 – 0.7†
1 Dental X-ray	0.2	
Nuclear explosion tests	0.003	0.02 – 0.05 ‡
Luminous watches	0.02	0.002 – 0.03†
Jet air travel		
Sydney – Perth	0.03 ††	
Sydney – London	0.14	

* 1 milli sievert/year (mSv a^{-1}) = 100 m rem /year.

** Dwellings in granite areas, UK.

† per X-ray

‡ per year

†† per round trip

A similar statistical level of risk applies to the following societal activities in the UK (Flowers 1976):–

- smoking 40 cigarettes,
- travelling 2 000 km by car,

- travelling 10 000 km by plane,
- rock climbing for 40 minutes,
- canoeing for 2.5 hours,
- engaging in ordinary factory work for about 40 weeks, and
- simply being a human aged 60 for 8 hours.

Maximum risks for persons exposed to probable repository conditions corresponded to less than one-fortieth of the risk from these activities. Use of pessimistic data suggested up to thirty times this level of risk. The SYVAC analysis suggested one-half of these risks.

Technological Trends

An alternative form of high-level waste disposal which has been studied intensively by the CEC involves separation of the long-lived actinides from the fission products. The actinides would be recycled through nuclear reactors and transmuted into nuclides with shorter half-life and reduced radiotoxicity. Fission products and transmuted actinides would be buried in deep geologic formations. While partition and transmutation of actinides seems technically feasible, it has been considered unlikely to be cost effective in dose reduction (IAEA, 1982b).

However, actinide separation was strongly supported by the Working Group on Spent Fuel Management, chaired by Professor Castaing, in a recent report to the French Supreme Council for Nuclear Safety (CSSN, 1982). The Castaing Report recommended the development of processes for the removal of the majority of neptunium 237 from high-level waste, together with americium 241 and curium 245, which form neptunium 237 by radioactive decay. The separated actinides would undergo special treatment, including immobilisation in ceramics such as SYNROC, or nuclear transmutation. Industrial implementation of these processes in France was envisaged by the end of the century. The Group recommended against geologic disposal of unreprocessed spent fuel or vitrified high-level waste at the present time.

The case for actinide separation has not been accepted internationally. Initial reactions are that it could increase capital and unit costs of reprocessing by 20 to 25% (Nuclear Fuel, 1983). Future trends seem likely to be concentrated on resolving the uncertainties in the metabolic and radiobiological data for neptunium, and on improving mathematical modelling techniques using realistic hydrologic and geochemical input data, to give confidence that the radiation doses from carefully chosen sites shall be as low as reasonably achieveable.

Conclusions

(a) Many solid waste forms appear suitable for disposal in appropriately engineered repositories without presenting a significant biological hazard.

(b) Borosilicate glass continues to be the principal radioactive waste form selected by many nations and has been used on an industrial scale since 1978.

(c) SYNROC is generally accepted as an improved radioactive waste form offering great development potential.

(d) The technology for the disposal of radioactive wastes in some geologic formations is available.

(e) The procedures for geologic disposal require, among other things, input from advanced hydrogeologic organisations for the development of regional hydrogeologic models for the prediction of groundwater movement.

(f) More assessment is required of the consequences of repository failure during the first few hundred years after waste burial.

(g) Further study is necessary to improve confidence in the validity of and sensitivity to parameter values in mathematical models used to estimate radiological dose resulting from failure conditions, particularly with reference to neptunium 237.

References

Amaury, P. Solidification and Storage of High Activity Nuclear Waste from Reprocessing: The AVM Process. 4th Annual Symposium, Uranium Institute, London (1979).

Anderson, D.R. (ed.) (1981). Proceedings of the Seventh Annual NEA Seabed Working Group Meeting, Paris, France, SAND 82-0460 (1981).

Australian Atomic Energy Commission Twenty-ninth Annual Report 1980-81 (1981).

BNFL Annual Report on Radioactive Discharges and Monitoring of the Environment 1981. British Nuclear Fuels Ltd, Health & Safety Directorate (1982).

Bonner, W.F., Bjorklund, W.J., Hanson, M.S., Knowlton, D.E. Engineering-scale Vitrification of Commercial High-level Waste. Waste Management '80, Proc. Symp. on Waste Management, Tucson, Arizona, 2, 339 (1980).

Burkholder, H.C. Waste Isolation Performance Assessment – A Status Report. Office of Nuclear Waste Isolation, Battelle Memorial Institute, Columbus. ONWI-60 (1979).

Burton, W.R., Griffin, J.R. A Design Study of Long-term Storage and Underground Disposal Systems for Highly Active Waste. United Kingdom Atomic Energy Authority, Nuclear Power Development Establishment, Risley. ND-R-514 (1981).

CEC. Marples, J.A.C., Malow, G., Jacquet-Francillon, N., Heimerl, W., et al. Testing and Evaluation of the Properties of Various Potential Materials for Immobilising High Activity Waste. Commission of European Communities, Ispra. EUR-7138 (1981).

Celeri, J.J., Lavie, J.M., Lefebvre, J., Sousselier, Y. Les Aspects Industriels de la Gestion des Dechets Radioactifs. *Annales des Mines,* No. 2-3, Fevrier-Mars, 119-126 (1983).

Chapman, N.A., McKinley, I.G., Savage, D. The Effect of Groundwater Availability on the Release Source Term in a Low Hydraulic Conductivity Environment. Proc. Workshop on Radionuclide Release Scenarios for Geologic Repositories, OECD Nuclear Energy Agency, Paris (1980).

Chotin, M.M. Hugony, P., Pieraggi, A. (1982). Industrial Operating Experience at the Marcoule Vitrification Facility. ANS Topical Meeting: The Treatment and Handling of Radioactive Wastes, Richland, Washington (1982).

CEA-Cogema. Le Retraitment des Combustibles Irradies en France et a l'Etranger. Commissariat a l'Energie-Cogema, Paris (1981).

Costello, J.M. High Level Radioactive Waste Disposal: The International Scene. Symposium on Radioactive Waste Management: A Geoscientific Assessment. Australian Geoscience Council and Australian Academy of Science, Canberra (1983).

CSSN. Rapport du Groupe de Travail Sur la Gestion des Combustibles Irradies. Conseil Superieur de la Surete Nucleaire, Ministere de la Recherche et del 'Industrie, Dec. 1981 – Nov. 1982, Paris (1982).

Dixon, R.S., Rosinger, E.L.J. (ed.) Third Annual Report of the Canadian Nuclear Fuel Waste Management Program. Atomic Energy of Canada Ltd. AECL-6821 (1981).

Elsam and Elkraft. Disposal of High-level Waste from Nuclear Power Plants in Denmark. Salt Dome Investigations. Vol. 1. Summary. Jysk-Fynske Elsamarbejde (ELSAM), Fredericia, Denmark; ELKRAFT A.mb.A, Hellerup, Denmark. INIS-mf-6784 (1981).

Flowers, B.H. *Royal Commission on Environmental Pollution.* Sixth Report. Her Majesty's Stationery Office, London (1976).

Hill, M.D. Analysis of the Effect of Variations in Parameter Values on the Predicted Radiological Consequences of Geologic Disposal of High-level Waste. National Radiological Protection Board, Harwell. NRPB-R86 (1979).

Hill, M.D., Lawson, G. An Assessment of the Radiological Consequence of Disposal of High-level Wastes in Coastal Geologic Formations. National Radiological Protection Board, Harwell, NRPB-R108 (1980).

IAEA. Evaluation of Solidified High-level Waste Products. International Atomic Energy Agency, Vienna (1980).

IAEA. *Safety Assessment for the Underground Disposal of Radioactive Wastes.* International Atomic Energy Agency, Vienna, Safety Series No. 56 (1981).

IAEA. *Site Investigations for Repositories for Solid Radioactive Wastes in Deep Continental Geologic Formations.* IAEA Technical Report Series No. 215 (1982(a)).

IAEA. *Evaluation of Actinide Partitioning and Transmutation.* IAEA Technical Report Series No. 214 (1982 (b)).

ICRP. *Limits for Intakes of Radionuclides by Workers.* International Commission on Radiological Protection, Publication 30. Annals of ICRP, 3, 1-4, Pergamon Press, New York (1979).

INFCE. Waste Management and Disposal Report of INFCE Working Group 7. Vol. 7. International Nuclear Fuel Cycle Evaluation, IAEA, Vienna. ST1/PUB/534 (1980).

Johnston, R.J., Palmer, R.A. Stability of Ceramic Waste Forms in Potential Repository Environments: A Review. Prepared for USDOE by Rockwell International Energy Systems Group. DOE/ET/41900-13 (1982).

Jones, P., Pearce, D. The Economics of Nuclear Fuel Reprocessing: A Case Study of the Windscale THORP Plant. *Energy Economics,* October, p. 202 (1981).

KBS-1. Handling of spent Nuclear Fuel and Final Storage of Vitrified High-level Reprocessing Waste. The Karn-Bransle-Saakerhet (Nuclear Fuel Safety) Study, Stockholm (1978).

KBS-2. Handling and Final Storage of Unreprocessed Spent Nuclear Fuel. The Karn-Bransle-Saakerhet (Nuclear Fuel Safety) Study, Stockholm (1978).

KBS-3. Final Storage of Spent Nuclear Fuel. The Karn-Bransle-Saakerhet (Nuclear Fuel Safety) Study, Stockholm (1983).

Knabenschuh, J.L. Present Status of the West Valley Demonstration Project. *The Treatment and Handling of Radioactive Wastes,* Battelle Press, Columbo, Richland, 1983, p. 534 (1982).

Koplik, C.M., Kaplan, M.F., Ross, B. The Safety of Repositories for High Radioactive Wastes. *Rev. Mod. Phys.,* 54, 1:269-310 (1982).

Marples, J.A.C. The Effect of Ionizing Radiation on the Release of Radionuclides from Conditioned Waste. Workshop on Geochemistry and Radioactive Waste Disposal, OECD-NEA, Paris/(1982).

McCarthy, G.J., White, W., Roy, R., Scheetz, B., Komarneni, S., Smith, D., Roy, D. Interations between Nuclear Wastes and Surrounding Rock. *Nature,* 273, 216-217 (1978).

McCarthy, G.J., Roy, R. A Review of Ceramic Nuclear Waste Forms through 1978. Workshop on Alternate Nuclear Waste Forms and Interactions in Geologic Media, Gatlinburg, Tennessee, CONF-8005-107 (1981).

Mendel, J.E., Nelson, R.D., Turcotte, R.P., Gray, W.J., Merz, M.D., Roberts, R.P., Weber, W.J., Westsik, J.H. Jr., Clark, D.E. A State-of-the-art Review of Material Properties of Nuclear Waste Forms. Materials Characterization Center, Pacific Northwest Laboratory. PNL-3802 (1981).

NHMRC. Recommended Radiation Standards for Indivuals Exposed to Ionizing Radiation (1981). National Health and Medical Research Council, Commonwealth Department of Health, Canberra (1981).

NRPB. *Living with Radiation,* UK National Radiological Protection Board, 2nd edition; H.M.S.O. (1981).

NRPB. Human Exposure to Radon Decay Products Inside Dwellings in the UK. National Radiological Protection Board, Harwell (1983).

NRPB. Metabolic and Dosimetric Models for Application to Members of the Public. Recommended Models for the Metabolism of Iodine and Values for the Gut Transfer Fraction of Plutonium, Curium and Neptunium. NRPB-GS3, National Radiological Protection Board, Harwell (1984).

Nuclear Fuel. Panel Recommends Slowdown on Reprocessing while France Reconsiders Waste Management. *Nucl. Fuel,* 8. 1 (Jan. 1983).

Nuclear Fuel, DOE keeps 1998 date in Draft Waste Plan, but moves back Site Recommendation Date. *Nucl. Fuel,* 9, 1:8 (Jan. 1984).

OECD-NEA. Geologic Disposal of Radioactive Waste. Research in the OECD Area. OECD-Nuclear Energy Agency (1982(a)).

OECD-NEA. Geological Disposal of Radioactive Waste. Geochemical Processes. OECD-Nuclear Energy Agency Paris (1982(b)).

OECD-NEA. The International Stripa Project. Experimental Research on the Underground Disposal of Radioactive Waste. OECD-Nuclear Energy Agency, Paris (1983).

Oertel, G., Crandall, J.L., McElroy, J.L. Treatment and Conditioning of Wastes from US Nuclear Fuel Reprocessing Plants. International Conference on Radioactive Waste Management, Seattle, USA, IAEA-CN-43/187 (1983).

Open University. Water: origins and demand – conservation and abstraction. Milton Keynes, Open University Press, *Environmental Control and Public Health Course* PT 272:104-105 (1975).

Pigford, T.H. Long-term Environmental Impacts of Geologic Repositories. IAEA-CN-43/185, International Conference on Radioactive Waste Management, Seattle, USA, IAEA-CN-43/187 (1983).

Pigford, T.H., Blomeke, J.O., Brekke, T.L., Cowan, G.A., Falconer, W.E., Grant, N.J., Johnson, J.R., Matusek, J.M., Parizek, R.R., Pigford, R.L., White, D.E. *A Study of the Isolation System for Geologic Disposal of Radioactive Wastes.* US National Research Council, National Academy Press, Washington, D.C. (1983).

Platt, A.M., McElroy, J.L. Management of High-level Nuclear Wastes. Pacific Northwest Laboratory. PNL-SA-7072 (1978).

Reeve, K.D., Levins, D.M., Ramm, D.J., Woolfrey, J.L., Buykx, W.J., Ryan, R.K., Chapman, J.F. The Development and Testing of SYNROC for High-level Radioactive Waste Fixation. Waste Management '81 Symposium, Tucson, Arizona (1981).

Reeve, K.D., Levins, D.M., Ramm, D.J., Woolfrey, J.L. Third Report to the IAEA's Coordinated Research Program on the Evaluation of Solidified Waste Forms. IAEA/AAEC Research Agreement No. 2700/Cf (1983).

Ringwood, A.E. Safe Disposal of High-level Radioactive Wastes, *Search,* 11, 10:323-330 (1980(a)).

Ringwood, A.E. Safety in Depth for Nuclear Waste Disposal. *New Sci.* (London), 88, 1229:574-575 (1980(b)).

Roberts, F.P. Irradiation Effects on Borosilicate Glasses. Paper presented at the 10th ASTM Int. Symp. on Effects of Radiation on Materials, Savannah, Georgia (1980).

Roy, R. The Technology of Nuclear Waste Management. *Technol. Rev.,* 83, 5: 39-50 (1981).

Salander, C., Proske, R., Albrecht, E. The Asse Salt Mine – The World's only Test Facility for the Disposal of Radioactive Waste. *Interdiscip. Sci. Rev.,* 5, 4:292-303 (1980).

Savage, D. Private communication to J.M. Costello (1981).

Savage, D., Chapman, N.A. Radioactive Waste Containment (letter). *New Sci.* (London), 90, 1253:452 (1981).

Shemilt, L.W. Third Annual Report, Technical Advisory Committee on the (Canadian) Nuclear Fuel Waste Management Program. TAC-3 (1982).

SKBF. Radioactive waste management plan. PLAN 82. SKBF/KBS 82-09:2, Svensk Karnbransleforsorjning AB/Audelning KBS, Stockholm, Sweden (1982).

Thiry, H.B., Laurent, J.P., Ricaud, J.L. French Experience and Projects for the Treatment and Packaging of Radioactive Wastes from Reprocessing Facilities. IAEA-CN-43/114, 2:219. Proc. Int. Conf. on Radioactive Waste Management, IAEA, Seattle, USA (1983).

UNSCEAR. Report to the General Assembly. United Nations Scientific Committee on the Effects of Atomic Radiation (1981).

USDOE. Draft Environmental Impact Statement. Management of Commercially Generated Radioactive Waste. US Department of Energy. DOE/EIS-0046-D (1979).

USDOE. Final Environmental Impact Statement. Management of Commercially Generated Radioactive Waste. US Department of Energy. DOE/EIS-0046-F (1980).

USDOE. The Evaluation and Review of Alternative Waste Forms for Immobilization of High-level Radioactive Wastes. US Department of Energy. DOE/TIC-11472 (1981).

USDOE. Long-term Technology Program for Defense High-level Waste. Savannah River Operations Office, Aiken, South Carolina, USA, DOE/SR-WM-79-3 Rev. 2-83 (1983 (a)).

USDOE. Bibliography of the published reports, papers and articles on the Nevada Nuclear Waste Storage Investigations, US Department of Energy. NVO-196-24 (Rev. 3) (1983 (b)).

USNRC. Solidification of High-level Radioactive Wastes, Final Report (Roy, R., Panel Chairman). National Academy of Engineering, National Academy of Sciences. NUREG/CR-0895 (1979).

USNRC. Disposal of High-level Wastes in Geologic Repositories. *Fed. Register,* 48, 120:28194-28228 (1983).

Wald, J.W., Lokken, R.O., Shade, J.W., Rusin, J.M. Comparative Waste Forms Study. Pacific Northwest Laboratory. PNL-3516 (1980).

Westinghouse. Conceptual Waste Package Designs for Disposal of Nuclear Waste in Tuff, Westinghouse Electric Corporation. ONWI-439 (1983).