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LUCAS HEIGHTS RESEARCH LABORATORIES

AN ASSESSMENT OF OVERSEAS DEVELOPMENTS IN METHODS FOR TREATMENT AND DISPOSAL OF HIGH-LEVEL RADIOACTIVE WASTES

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Geology and Geophysics

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ABSTRACT

The principles of management and disposal of highly radioactive wastes contained in spent fuel from nuclear power generation are described. The status of developments in spent fuel reprocessing, high-level waste solidification and geologic isolation is reviewed. Some generic studies on the possible range of annual radiological doses to individuals from waste repositories are discussed and compared with doses from some existing nuclear power and fuel cycle operations, and with the dose received annually from an average background of naturally occurring radiation.

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BACKGROUND TO THE JOINT STUDY

Policies adopted by other countries on the ultimate disposal of high-level radioactive wastes and spent nuclear fuel are important to Australia because they are relevant to Australia's uranium marketing policy, and to the possible future introduction of nuclear power to Australia. From the viewpoint of public reassurance, it is also important to be able to demonstrate that the disposal of radioactive wastes overseas is being undertaken in a responsible and effective manner. Burial in deep geologic formations is considered to be the most effective means of ensuring long-term isolation of these materials from the environment.

A major source of up-to-date information on progress being made overseas in the ultimate disposal of radioactive wastes is the Radioactive Waste Management Committee of the Organization for Economic Cooperation and Development Nuclear Energy Agency (OECD-NEA). The Committee set up in 1975 a Coordinating Group for Geologic Disposal which initiated collaborative programs to identify deep, stable geologic formations considered to be technically satisfactory for the burial of highly radioactive wastes arising from the operation of nuclear power stations. In addition, there is collaborative work in individual countries between bodies with expert knowledge of radioactive wastes, geology and hydrogeology. For example, the US Department of Energy (USDOE) is collaborating with the US Geological Survey and the UK Atomic Energy Authority (UKAEA) is working with the UK Institute of Geological Sciences.

Informal contacts between officers of the Australian Atomic Energy Commission (AAEC) and the Bureau of Mineral Resources, Geology and Geophysics (BMR) had shown that BMR has an interest in the geological aspects of radioactive waste disposal and has appropriate engineering and hydrogeological expertise. In 1979, the AAEC proposed a collaborative technical assessment with BMR of developments overseas of various methods for the treatment and disposal of high-level wastes and spent fuel from generation of nuclear power. The AAEC agreed to assess developments in radioactive waste treatment and disposal by member countries of OECD-NEA, as well as developments in Australian research into such advanced waste forms as SYNROC. The Bureau, in collaboration with the AAEC, agreed to assess the relative merits of a variety of types of

geologic formations being considered for the immobilisation of the various waste forms over a range of repository conditions of temperature, pressure, presence of water, etc. These assessments would take into account the data being acquired by OECD-NEA.

A timescale of about two years, ending in April 1992, was considered realistic for the study.

1. INTRODUCTION

A modern 1250 MWe pressurised light water reactor (LWR), capable of supplying the electrical needs of over 1 million people, discharges each year 73 spent fuel assemblies. This fuel contains about 32 t of uranium, 1.15 t of fission products, about 285 kg of plutonium, and about 50 kg of other transuranium elements. A boiling water reactor (BWR) of similar size discharges 220 fuel assemblies containing about 20 per cent more uranium and 10 per cent more plutonium [Croff and Alexander, 1980]. 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 ten years.

Spent fuel may be managed by reprocessing, a technique in which residual uranium and plutonium are extracted for re-use in nuclear fuel cycles, and fission products and other transuranic elements constituting the high-level waste are converted 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 may be encapsulated and consigned to geologic disposal. The AAEC forecasts of nuclear power growth suggest that, by the year 2000, about 60 000 tonnes of uranium in spent fuel may have been committed for reprocessing in Europe and Japan, and that a further 55 000 tonnes produced in the USA will have required reprocessing, long-term storage or geologic disposal.

This report reviews the status of overseas developments in management and disposal of the high-level radioactive waste arising from the generation of nuclear power.

2. 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. Rapin [1980] has reviewed reprocessing operations in France, Japan, the UK and the Federal Republic of Germany (FRG). Reprocessing policy and development in the FRG have

been discussed by Scheuten and Salander [1981].

The nationally preferred options under consideration for the reprocessing and disposal of spent fuel are listed in Table 1. Of 22 countries with operational nuclear power stations above 30 MW, six are currently reprocessing domestic fuel, nine have contracts for international reprocessing services, and three are considering geologic burial of unreprocessed spent fuel [AAEC, 1980]. The status of national programs for fuel reprocessing is summarised in Table 2.

For some reprocessing contracts negotiated before 1980, it has been reported that the French and UK reprocessors have agreed to dispose of the resultant high-level waste; for contracts signed from 1980 onwards, the reprocessors have the option of returning waste to the countries of origin [Paige et al., 1980]. Some COMECON countries, which have their spent fuel reprocessed in the USSR, are apparently not expecting high-level waste to be returned [Dlouby et al., 1980; Mitzinger, 1979].

3. HIGH-LEVEL WASTE SOLUTION

Light water reactor fuel may generally be reprocessed after a storage period varying from 1 year [Hill and Lawson, 1980] up to 10 years [KBS, 1978a] to permit some decay of fission product radioactivity. Some spent fuel in the US has been stored for about 20 years. Reprocessing of the fuel discharged yearly from a 1250 MWe LWR gives rise to about 40 m^3 of high-level waste concentrate. This is a solution of nitric acid which contains about 0.5 per cent original uranium and plutonium, together with about 99 per cent of other actinide elements and fission products other than rare gases and iodine 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 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.

Concentrated high-level waste solutions from commercial nuclear fuel reprocessing have been stored in stainless steel tanks in Belgium, France, India, Japan, UK, USA, and USSR for periods up to 25 years, and no leakages have been reported from these storage tanks. The tanks require cooling to remove fission product heat and secondary containment to prevent leakages. Spare tanks are needed to enable the transfer of solution in the event of tank failure, and the plant needs 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 deep burial in geologic formations.

4. SOLIDIFICATION OF HIGH-LEVEL WASTE

The solidification of high-level waste solutions 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 wastes are encapsulated in metal, glass or ceramic matrixes (Table 3). 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 of the wastes. Solidification of high-level waste from the annual reprocessing of fuel from a 1250 MWe LWR could give rise to about 27 cylinders of borosilicate glass, each cylinder being 30 cm diameter x 3 m height.

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 of the desirable characteristics has yet to be demonstrated; however, many solid forms are likely to be satisfactory in an appropriately engineered disposal system. The conversion of wastes into borosilicate glasses (vitrification) has received most developmental effort, and is in operation on an industrial scale for wastes from low burn-up fuel from gas-graphite nuclear power reactors. The technology has been demonstrated on an engineering scale for wastes from high burn-up fuel from light water reactors [INFCE, 1980]. Glass is considered to be adequate for a first demonstration system of

solidification, transportation and ultimate disposal [USNRC, 1979], but second generation waste forms now under development have overall characteristics which may prove to be superior.

5. STATUS OF HIGH-LEVEL WASTE VITRIFICATION

Table 4 summarises some national achievements, developments, and plans for the solidification of high-level waste. Most nations with nuclear power programs have selected borosilicate glass as the principal form for immobilising their high-level waste.

France is the world leader in applied vitrification technology. Following small-scale research between 1958 and 1963, 480 PBq (13 MCi) of radioactive waste from reprocessed gas-graphite (GGR) fuel was solidified in the PIVER pilot plant at Marcoule into 12 t of borosilicate glass between 1969 and 1973. A continuous industrial scale vitrification plant, Atelier Vitrification Marcoule (AVM), commenced operation in 1978. The AVM plant technology and performance have been described by Bonniaud et al. [1980]; it has demonstrated an annual capacity for the vitrification of 132 m^3 of fission product solution into 60 t of glass. This capacity is equivalent to vitrification of wastes from about 3400 t U y^{-1} of GGR fuel which is greater than the rate of spent fuel arising from the French GGR program. The AVM plant is expected to vitrify the backlog of high-level wastes stored at Marcoule since 1958, as well as handle the routine production of the Marcoule reprocessing plant. By April 1982, 440 m^3 of high level liquid containing 3.7 EBq (72 MCi) of radioactive waste had been vitrified into 650 canisters [Chotin et al., 1982], each holding about 340 kg of glass.

The waste canisters are stored at Marcoule in concrete cells. Fission product decay heat is removed by forced air cooling; it is intended to cool the cells by natural convection after several years of storage. Maximum temperatures of the glass product are maintained below 600°C to avoid spontaneous crystallisation of the glass, a process known as devitrification.

A similar plant, Atelier Vitrification Hague (AVH), is being designed to vitrify high-level wastes from 800 t U y^{-1} of reprocessed LWR fuel at the Cap La Hague site. This capacity is equivalent to the generation of about 26 GWe of nuclear power.

The plant will require two processing lines similar to that of AVM, each consisting of a liquid waste calciner and melting furnace. It has a planned total capability for the production of 300 t y^{-1} of glass from 600 m^3 y^{-1} of waste solution at an operating level of 250 days per year

[Bonniaud et al., 1980].

By comparison, developments in waste solidification during the past 20 years in Canada, India, the FRG, the UK, the USA and the USSR (Table 4) have been on a laboratory or pilot plant scale. Engineering scale vitrification of commercial high-level waste in the US has been reported by Bonner et al. [1980]. French vitrification technology is being marketed internationally; the UK has abandoned its FINGAL/HARVEST process and announced that AVM technology under licence is to be used in a vitrification plant to be constructed at Windscale. Belgium and the FRG are reported to have contracted 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].

The 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 9150 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 t U, equivalent to about 660 GWe y of installed nuclear power, will therefore be immobilised in borosilicate glass; this is equivalent to vitrification of wastes from four years of the present installed nuclear power program for the whole world (155 GWe in 1982).

An alternative vitrification process, developed by the FRG, is under construction at Mol for demonstration on a 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.

6. DEVELOPMENT OF ADVANCED WASTE FORMS

Mineral-like crystalline ceramics have been under development at Pennsylvania State University for almost 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 relative merits and disadvantages of the alternative forms for immobilising commercial high-level wastes have been extensively reviewed [Mendel et al., 1981; CEC, 1981; IAEA, 1980; USDOE, 1980b; Wald et al., 1980; USNRC, 1979]. SYNROC is an alternative ceramic waste form which has been ranked high in research potential, particularly for salt-bearing US defence wastes [USDOE, 1980b]. However, it has recently been reported that defence wastes at the Savannah River Plant are to be immobilised in borosilicate glass [Nucleonics Week, 1982] on grounds of process simplicity and cost. SYNROC could be an alternative technology for immobilising wastes from commercial reprocessing in the US if the latter operation is recommenced; it has not attracted major interest in Europe owing to the industrial lead established by vitrification technology.

7. SYNROC

The SYNROC formulation, proposed by Professor A.E. Ringwood of the Australian National University (ANU), is being developed in a collaborative program between ANU and the AAEC. Ringwood [1978, 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 their crystal lattices.

Leach rates of alkali (Cs) and alkaline-earth (Sr) elements from non-radioactive SYNROC at 300°C into water which was replaced daily have been up to 1000 times lower than those from borosilicate glass; under static conditions, where water was saturated with leached 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, 1982]. 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 program at the AAEC's Research Establishment at Lucas Heights includes the fabrication of non-radioactive SYNROC on a multi-kilogram scale [Ramm and Reeve, 1982] and continuation of radiation damage and leach tests. Further investigations required include:

- studies of the engineering practicability and costs of SYNROC manufacture under highly radioactive conditions;

- the reproducibility of the SYNROC formulation during large scale operation; and
- preparation of, and leach tests on SYNROC samples containing radioactive fission products and actinides. Laboratory scale equipment for manufacture of these samples is in course of construction.

8. 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. The primary source for radiation damage in nuclear waste solids is the displacement of constituent atoms, mainly from contained α-emitters. Other effects include internal ionisation resulting in 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. The relative importance of these effects depends on the nature of the waste form, i.e. whether it is a supercooled liquid (glass) or a solid with a crystal lattice (ceramic).

The majority of the radiation 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 the actual 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, α-doses up to 5×10^{18} alpha decays per g, equivalent to over 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 which had been detected in glasses after simulated α-doses equivalent to over 10^5 years of storage [Platt and McElroy, 1978] were not significant in the context of storage in an appropriately engineered disposal system.

9. POTENTIAL LEACHING OF RADIOACTIVITY FROM VITRIFIED HIGH-LEVEL WASTE

Contact of groundwater with high-level waste results in the leaching of radioactive constituents into solution, and ultimately in total dissolution of the waste form. 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.

The release of radionuclides from waste forms by leaching involves complex mechanisms of selective leaching and matrix dissolution. These mechanisms are not completely understood. Actinide elements have been observed to leach 100 times more slowly than alkali metals from borosilicate glass; also, leaching rates in water at 100°C are 35-50 times greater than at 20°C [Amaury, 1979; Mendel et al., 1981].

A field test on radioactive glass in contact with a cool flowing aquifer has been conducted in Canada for over 20 years. Fifty blocks of nepheline syenite containing 50 TBq (1400 Ci) of fission products were produced between 1958 and 1960. Nepheline syenite is a naturally occurring alumino-silicate material. The glass blocks were buried below the water table in an alluvial soil. Rates of leaching based on ^{90}Sr release fell during 10 years to 5×10^{-11} g cm^{-2} d^{-1}, corresponding to a dissolution time of more than 10^8 years [Tomlinson et al., 1977].

Leaching rates as low as 5×10^{-7} g cm^{-2} d^{-1} have been measured for caesium and strontium 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 USA [USDOE, 1980a]. This corresponds to a bulk dissolution rate of 0.014 mm y^{-1}, or a dissolution time of about 3500 years for waste fragmented into pieces 10 cm in diameter.

By contrast, glasses in contact with water at high temperature (300°C) and high pressure (30 MPa) have been observed to undergo rapid hydration, leading to fragmentation of the sample and leaching of about 30% of the caesium from the glass in two weeks [McCarthy et al., 1978]. This situation has been compared with that of water entry to a repository containing high concentrations of radionuclides during the initial few hundred years when internal temperatures will be near their maximum.

High temperature leaching tests under reflux in the laboratory have been criticised as being more representative of waste disposal under a 'boiling waterfall' than conditions which could reasonably be expected in geologic burial [Savage and Chapman, 1981]. At groundwater flowrates

of 1 to 2 x 10^{-3} m y^{-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]. However, the possibility of entry of an aquifer into a repository after major ground movement, though remote, cannot be totally excluded. Accordingly, some countries have expressed intentions to limit temperatures in waste repositories to below 100°C, corresponding to leach rates which would be acceptable. 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.

10. STATUS OF GEOLOGIC DISPOSAL OF HIGH-LEVEL WASTE

10.1 Isolation from the Biosphere

The main alternatives for the isolation of radioactive waste in geological formations are as follows:

(a) Placement in a mined repository or a deep drilled hole.

(b) Placement on or below the deep ocean floor or within deep oceanic sediments.

10.1.1 Land-based repository

The mined repository concept is now thought to be feasible, and awaits identification of technically suitable and publicly acceptable sites; nevertheless, public opposition to the disposal of high-level waste in mined repositories has been strong. Investigations of potential geologic sites have been deferred in a number of countries and discontinued in the UK.

Groundwater is generally considered to be the most likely agent for the dispersal of radionuclides from a high-level radioactive waste repository, with radionuclides being transported to the biosphere in solution [Burkholder, 1979]. The strategy of prolonged isolation in the geologic environment is aimed at keeping water from coming into contact with the waste, and ensuring that if water should come into contact with the waste it is not returned to the biosphere until its acquired radioactivity no longer constitutes a hazard to either man or the environment. This suggests burial, within a container of high integrity (see Section 11), in a geologic medium that is either strong and of extremely low permeability or is known to be isolated from ground-

water. Isolation from the biosphere is further assured if escaping radionuclides or heavy metals are trapped by ion exchange processes in the repository. This could occur either on the host rock itself, on added clay or on other minerals, or in adjacent geological strata.

In a soundly selected and engineered repository only cataclysmic geological events - earthquake and volcanic activity - should pose a risk of catastrophically breaching a repository over a few thousand years. Drastic climatic change, resulting in the melting of the polar icecaps or encroachment of ice sheets, would be needed to have a possible impact of significance on a repository in the same time-scale. Such changes would also have vastly greater effects on the biosphere than the breaching of a repository.

With an increasing time-scale, prediction of geological events and of the long-term effects of slow, ongoing processes becomes more difficult. However sound site selection, including the selection of the host rock, and repository design can minimise even the long-term risk of the integrity of the repository being breached. Table 5 sets out the processes which pose the greatest theoretical threat to a repository and suggests the order of time in which each may be significant and the measures available to minimise risk. Nevertheless, the effects of these processes are not necessarily deleterious; for example, immersion of a site in water, or coverage by an ice sheet or lava could enhance the integrity of the repository rather than jeopardise it.

Conditions for emplacement in a geologic environment are as follows:

(a) It can be penetrated by engineered structures or large diameter drillholes, kept open until emplacement is complete, then completely sealed.

(b) It can resist perturbations around the waste, produced by radiation-generated heat or by radiation, which might lead to the entry of water. This suggests a strong, stable, virtually impermeable host rock, e.g. granite (permeability generally less than 10^{-9} m s^{-1}) or plastic self-healing material such as salt or clay.

(c) It has good sorption properties for the radioactive constituents of the waste.

(d) It preferably has high thermal conductivity which will disperse the heat energy from the waste.

The mined repository concept has received the major design effort and feasibility studies of crystalline rock and salt are well advanced.

The deep-hole disposal method could be regarded as a variant of the mined repository, since much of the research and experimental work on mined repositories in crystalline rock, salt and clay would be applicable to the development of a deep-hole repository. The deep-hole concept has to some extent been neglected, although countries with small nuclear power programs have shown interest in pursuing this line of investigation, which has been advocated by Ringwood [1980b].

10.1.2 Seabed Disposal

At this stage, no country considers placement of high-level radioactive waste on or in the deep ocean floor to be a suitable option, although confidence in seabed or sub-seabed disposal as a technically acceptable alternative to land-based repositories has been expressed by France, Japan and the US [Anderson, 1981]. At present, such disposal is expressly prohibited under an international convention, and major political questions on the use of the seabed are expected to occupy international forums for some years before such a proposal could become a realistic option. Procedures for placement of the waste, proving the methods of canister retrievability from malplacement (a stipulated requirement for ocean floor disposal) and monitoring the performance of ocean floor disposal are the subject of considerable effort from members of the OECD-NEA. Countries actively participating in the OECD-NEA Seabed Working Group Program include Canada, the Commission of European Communities (CEC), France, Japan, the Netherlands, UK and USA, while countries with observer status are Belgium, the FRG, and Switzerland [Talbert, 1982].

10.2 Current Status of Host Rock Types

Table 4 summarises national investigations of geologic formations and programs for the establishment of demonstration and commercial high-level waste repositories. Sweden, the FRG, Canada and the USA are leading with in-situ studies of their geologic formations [Thoregren et al., 1980; Salander et al., 1980; USDOE, 1980c; Boulton, 1980; Shemilt, 1981].

The advantages and disadvantages of the various rock types being considered for repositories are set out in Table 6. It may be seen that no formation is without some disadvantage. Borehole and shaft sealing is a requirement common to all repositories. The IAEA have recently concluded [IAEA, 1982] that a site with ideal characteristics is not essential for siting an appropriately engineered waste repository.

10.2.1 Salt

In the late Fifties and early Sixties, it was believed by most earth scientists concerned with waste disposal that salt deposits would be an acceptable geologic environment for the storage of all forms and all levels of radioactive waste. Pierce and Rich [1962] summarised data on all the known salt deposits in the United States. Their summary showed that salt, either as bedded salt or in salt domes, occurs in 23 States. These original regional studies of salt deposits were used in the selection of the bedded salt deposits in Kansas and later in New Mexico as potential locations for high-level waste repositories. The Kansas site was abandoned primarily for hydrogeologic reasons [De Buchanane, 1976], but in New Mexico construction of the Waste Isolation Pilot Plant (WIPP) is in progress; it will be the first US facility designed and constructed to gather data and demonstrate on a large-scale the feasibility of disposal of radioactive waste in bedded salt [Langley, 1979].

The Federal Republic of Germany carried out a number of research and development projects at the Asse salt mine, including the construction of a pilot plant for the handling and disposal of low- and intermediate-level radioactive waste. It is now proceeding with the construction of a high-level repository in the Gorleben salt dome [Kuhn et al., 1980]. Another project, under investigation in Denmark, is the feasibility of disposing of high-level waste in a deep hole in one of the salt domes in Jutland.

The US National Academy of Sciences advised the US Atomic Energy Commission in 1957 that salt was the formation of first choice for the disposal of radioactive waste. By 1970, considerably more information was available on the reaction of salt to increases in temperature and pressure and to radiation, together with studies on the physical chemistry of fluids in salt which had cast some light on the formation of breccias and solution cavities in salt. An early demonstration of the engineering techniques for the handling and placement of short-cooled spent fuel assemblies in salt were encouraging [Bradshaw and McClain, 1971]. However, the major disadvantages in the use of salt as a disposal medium are its potential for creep with concomitant difficulties if waste form retrieval is desired, and the corrosive nature of bittern inclusions (saturated solutions of chlorides). The determination of site-specific conditions such as hydrogeology is a requirement for all geological types. These factors may present long-term containment

difficulties for some sites and are discussed in more detail below.

Creep within salt

The creep of salt around openings in salt rock is governed by the naturally low limit of elastic behaviour of salt rock which is of the order of only 70 kPa (100 psi). The rate of creep becomes constant when the supported load remains unchanged [Baar, 1973]. Mine pillar deformation can cause vertical closure of openings, also at a constant rate, and is commonly accompanied by subsidence at the surface. An increase in temperature will cause the rate of creep to increase; however, design principles based on in-situ measurements in salt mines for more than 20 years provide a firm basis for the design of openings and for support systems within a salt formation [Baar, 1975]. Lower temperatures may be attained by reducing the density of canister placement by ageing the waste before burial, or by removing heat generated by the waste by ventilation until the repository is abandoned and sealed and the control of creep no longer required.

Because of the susceptibility of glass to leaching at higher temperatures, some countries intend to limit temperatures in waste repositories to less than 100°C. This limitation on repository temperatures to some extent offsets the disadvantage of creep in salt which is more of a problem at higher temperatures. It has led to a hardening of opinion in the USA in favour of wastes being retrievable for arbitrary periods ranging from 5 to 50 years. For waste to be retrievable, ventilation would have to be maintained thereby ensuring that some of the heat generated by the waste was exhausted from the repository. The penalty for retrievability is a costlier installation and an additional operational cost until the facility is finally sealed.

Presence of bitterns

Bittern is the highly concentrated solution that remains after the crystallisation of common salt (NaCl); the solution is rich in chlorides of magnesium and potassium which remain in solution at high concentrations.

The initial studies of water in salt gradually developed into the study of the physical chemistry of bitterns contained within salt deposits. The migration of bittern inclusions towards sources of heat [Roedder and Belkin, 1980], the low pH and the high percentage of salts in solution in bitterns make the salt medium highly corrosive to emplaced waste and its containers [Clynne et al., 1980]. Although lower tempera-

tures in a repository should reduce the potential for canister corrosion, this problem is being closely examined, as are the consequences of the contact of bitterns with radioactive materials in the canister [Stewart and Potter, 1979].

Geologic evidence indicates that the central mass of most preserved evaporite deposits has remained isolated from the surrounding groundwater system for millions of years [Brookins et al., 1980]. It is reasonable to assume that such a mass will retain its integrity unless a destabilising element, such as a localised heat source, is introduced that would lead to an increase in the fluid phase as bitterns. Canisters of waste provide such a localised heat source and this justifies further research and development before a salt repository is finally approved.

Hydrogeology

Groundwater systems are determined by many factors, and are complex. They must therefore be studied for each prospective disposal site, and conditions at each must be understood to a sufficient degree so that a reliable prediction of the possible interaction between the groundwater, the salt deposit and the emplaced waste can be made. This is particularly important in a mined repository at shallow depths, where groundwater tends to circulate more freely than at greater depths and could be a potential threat to the integrity of the repository.

By contrast, the hydrogeologic system poses a lesser threat to a repository of the deep-hole type, where the waste would be disposed of at depths approaching the limit of circulating waters.

Salt domes intrude through a great thickness of sediments and usually intercept several aquifers. It can often be shown that domes have undergone dissolution by groundwater. In fact, the 'cap rock' formation immediately overlying many domes is generally interpreted as the accumulation of insoluble residues previously dispersed in the salt mass [Bodenlos, 1970].

The groundwater hydrology in proximity to salt domes is usually complicated by the faulting and extensive disturbance that the sediments have undergone as a consequence of the salt intrusion. In the face of the overwhelming geologic evidence pointing to past salt dissolution, it should be established beyond doubt that salt dissolution is not taking place at any salt dome being considered as a repository for radioactive waste; an adequate understanding of the entire problem of the inter-

actions between salt domes and ground water would require additional extensive investigation [Gera, 1975].

The origin of collapse structures and breccias that vertically penetrate apparently impermeable evaporites has not been satisfactorily explained. Anderson and Kirkland [1980] postulated that the source of the dissolving water, if acting under a hydraulic head, would establish a flow cycle in which the dissolved salt provides the density gradient to maintain the movement of water and so continue dissolution of the salt. They cited the Delaware Basin in western Texas and south-east New Mexico as a good example of how brine density flow can produce dissolution chambers that collapse to form breccias. They considered that the potential for dissolution by brine flow is an inherent property of partly exhumed evaporites.

Salt with a low water content (less than 0.5 wt%) is less likely to mobilise bitterns; however, the identification of such low water-content zones with a salt deposit is not a simple matter. The water content in bedded salt and in some dome salt can exceed 3 per cent, and it has been reported that adjacent samples of bedded salt may differ widely in water content [Roedder and Belkin, 1979]. The basic method of investigation of geologic formations is the study of drillcores; because of the variability of salt formations, close drilling of a prospective site may be necessary - and all holes would subsequently require sealing against leakage.

The use of geophysical techniques to detect voids and porosity variations in salt rock has been proposed as a reliable means of investigation in the future [USDOE, 1980c]. The effectiveness of seismic techniques to detect voids and possibly pockets of water in salt rock, as distinct from salt with a high percentage of inclusions of water, has been demonstrated. However, its use is restricted to near-field conditions ahead of a mining face or to zones between two closely spaced boreholes. According to Rothemeyer [1980], the application of a newly developed high-frequency method to reflection measurements in a borehole and absorption measurements between two boreholes will identify inhomogeneous qualities of the rock up to several hundred metres away from the boreholes. The ability to discriminate between voids, changes in lithology, and moisture content variations has yet to be proved, but the method holds some promise for future investigations.

There is little doubt that some salt deposits could have properties that would meet the current specifications for a high-level waste reposi-

tory; it is equally probable that there are many that would not meet the specifications. There is a need for more site-specific research into the effect of the hydrogeologic system on the long-term integrity of each proposed repository.

10.2.2 Crystalline rock

The term 'crystalline rock' is used here for coarse-grained intrusive igneous rocks such as granite, granodiorite, diorite and gabbro; although differing in chemical composition, they have similar physical properties. Basalt (see Section 10.2.3), though generally a crystalline rock, is not an intrusive rock but is of volcanic origin.

Sweden is in the vanguard of the study of crystalline rock and intends to use it as a repository for high-level nuclear waste. Feasibility and site selection studies have been made at a number of sites and special research areas have been established at Finnsjon near Forsmark, the abandoned mine at Stripa, and Studsvik to determine the methods of investigation and criteria for confirmation of a repository site.

The construction of an underground repository in crystalline rock at depths to 5000 m can be achieved with standard drill and blast mining methods. Openings will generally be self-supporting and require only limited support measures in zones with a higher incidence of fractures or with high stress concentrations. Openings would generally be stable at repository temperatures of 300°C at atmospheric pressure. At the higher fluid pressures likely to be encountered in crystalline rock at a depth of one kilometre, some minerals will decompose in the presence of water. The rate of decomposition will depend on site-specific factors, and the process would continue until equilibrium was attained, probably within 1000 years. Mineral decomposition will probably be confined to rock exposed to groundwater in joints in the immediate vicinity of the repository and is not expected to weaken the rock structurally [Chapman, 1978]. The minerals formed by decomposition will probably include some with layer structures that will fix radionuclides by ion exchange. Although it is not yet established whether mineral decomposition will be detrimental to the confinement of high-level waste within a repository [Chapman, 1980], the current European proposals are to restrict temperatures within the repository to about 100°C [Burton and Griffin, 1981]; however, Mather et al. [1982] have claimed that there is no geochemical rationale for this restrictive limitation, as geochemical processes of waste degradation and waste/rock interaction in the hydrothermal

environment remain predictable to about 200°C.

Ventilation will be required to cool the repository to permit access during deep placement of the waste, and during any subsequent period of retrieval; it will not be required at any time other than to ensure access.

Crystalline rock would also be suitable for the disposal of high-level waste by the deep-hole method. Contrary to some reports in waste disposal literature, drilling large diameter holes to depths of 5000 m in rock at high temperatures is not beyond current technology [USDOE, 1980a]. Geothermal energy investigations in hot dry rock have included drilling 300 mm diameter holes to depths of more than 4500 m at temperatures in excess of 300°C [Pettit, 1981]. Geothermal energy investigations are confined to areas with a high thermal gradient (about 20°C per 1000 m); however, the gradient varies within the Earth's crust and depends on the composition of the crustal rocks and on the tectonic state of the crust. Greater depths could be achieved in cooler zones with a temperature gradient of 10°C per 1000 m, which is only half the gradient found in hot dry rock. These achievements provide a sound basis for further development.

As ventilation is not required in a deep drillhole repository, refractory waste forms such as ceramics and SYNROC that are stable at higher temperatures could be placed in such a repository.

Water in crystalline rock is held in fractures, the openings of which decrease with depth. Permeabilities of 10^{-8} cm s^{-1} or less, measured by water injection tests, were found at depths of more than 300 m in the granite at Stripa in Sweden [Gale et al., 1980]. In general, permeability will decrease with depth. Other advantages associated with depth include a decrease in the potential for groundwater movement and an increase in the distance along flow lines that water must travel before reaching the biosphere [KBS, 1978a]. The size of a repository is a factor in determining the amount of water that will have access to the waste. A large repository would intersect more fractures and more water would be expected to come into contact with the waste containers than is the case for a more compact repository such as a large diameter vertical hole. Sealing the deep-hole repository with a bentonite-sand mix would ensure that water entering the hole was absorbed in the bentonite; this would expand to form an impermeable barrier around waste in the hole, effectively blocking the entry of more water.

The problems of constructing a high-level waste repository in crystalline rock are well understood. Investigations are continuing into the most appropriate methods for the measurement of site-specific properties such as permeability and heat transfer, and general research is concerned with geochemistry and the adsorption characteristics of the rocks. Many crystalline rock masses seem likely to meet the current specifications for the design and construction of a mined repository for high-level waste. The technology is available for the thorough investigation of the geology and hydrogeology of crystalline rock. Apart from the time that such an investigation would take (about eight years), the project should not involve major new technical problems. About ten years would probably be required to develop and test procedures for the construction of a deep-hole repository in crystalline rock.

10.2.3 Basalt

Basalt is a volcanic lava with a texture which ranges from medium-grained crystalline to glassy. Although most basalt would be considered a fine-grained crystalline rock, its general physical properties are somewhat different from those of coarse-grained crystalline rocks such as granites. Thick bodies of basalt are generally made up of many sub-horizontal flows with a high incidence of fractures known as shrinkage fractures. The construction of an underground repository in basalt at depths similar to that for crystalline rock can be achieved by conventional drill and blast mining techniques. Openings, in general, will be self-supporting except where fractures are more numerous or where stresses are concentrated. As with granite, the openings would generally be stable at repository temperatures up to 300°C at atmospheric pressure. In a confined repository with a build-up of fluid pressure, mineral decomposition similar to that observed in granite would take place; this is not expected to weaken the rock structurally, and it may form minerals with layered structures that would fix radionuclides by ion exchange. However, until the consequences of such changes have been fully investigated, low repository temperatures of less than 100°C are preferred.

The difference in the use of basalt and crystalline rocks such as granite for repository construction is that associated with vertical thickness. Crystalline rocks emplaced by upward movement from deep within the Earth's crust extend to great depths, are homogeneous, and their composition does not change with depth. Basalt bodies are generally less thick, composition within a series of flows may vary considerably, and they may be underlain by sedimentary rock. Flows may be

interbedded with sedimentary rock, and permeable strata below and between basalt flows may be sources of water under pressure. The generally higher incidence of fractures in basalt enhances their good aquifer characteristics in many areas, and variation in the number of fractures and joints in the individual flows and tuffs of a basalt formation contributes to a complex hydrogeologic system.

Joints in basalt form when individual flows have cooled, and many joints may have been sealed by the late formation of minerals. The modelling of such a complex hydrogeologic system requires a more comprehensive investigation than would be required for a more uniformly jointed rock such as granite.

Sealing the repositories and plugging the drillholes will assume more importance in the assessment of basalt as a potential geologic medium because there are more fractures and joints in basalt than in crystalline rock. The Columbia Basalt, which occupies the Pasco Basin in Oregon, is under consideration for the disposal of high-level radioactive waste. Extensive hydrogeologic investigations to depths greater than 3000 m indicate that the fracture permeability of some of the thick basalt flows is very low; this makes them suitable for assessment as a repository [Deju and Evans, 1980].

Basalt to 3000 m thickness, a rare phenomenon, is confined to trap occurrences. The more common interbedded sediment and basalt sequences lack homogeneity and would not, prima facie, be regarded as suitable for the disposal of high-level waste.

Reservations should be maintained on the use of basalt until hydrogeologic studies on a basin by a major hydrogeological organisation produce a reliable site-specific model for the prediction of groundwater movement. More data on ion-exchange capability of site-specific basalt deposits are also required.

10.2.4 Clay and shale

Clays form a family of fine platy minerals with a range of chemical composition and crystal structure, and with excellent ion-exchange properties. The term 'clay' as a name for a geologic formation means an aggregate of predominantly clay minerals, but includes particles of other minerals and rock fragments, generally less than 0.074 mm in size (i.e. they will pass through a No. 200 sieve). Clay materials range in consistency from a spongy ooze to firm, partially consolidated materials. Clay bodies form both by deposition as sediments and as products of the weathering of rock. Consolidation of clay takes place by com-

paction under the weight of overlying sediments which expels much of the water from the clay. Consolidated clay with a layered sedimentary structure is called shale; where there is no layered structure, it is called mudstone or claystone.

Clay minerals hold a large quantity of water within their molecular structure, but this water is not released unless the molecular structure is disturbed. Clay bodies generally have a low permeability to water owing to the lack of pore spaces. Clays are structurally weak and creep under load. The eventual load to be carried by a support system for an opening in clay will be a considerable portion of the weight of the overlying material; the installation of supports to maintain openings at depth in clay is a major cost of construction.

The chemical reactions and movement of fluids in clay in response to an increase in temperature are two areas requiring further research, and maintenance of low temperatures is likely to be an essential requirement for the storage or disposal of radioactive waste in clay formations.

In Belgium, the construction of such a repository is regarded as feasible only if it is built within 250 m of the surface [Heremans, 1978]. Because of the many unknown factors associated with water migration in clay and chemical reactions in response to an increase in temperature, temperatures above 100°C in a clay repository are undesirable at this stage [Heremans, 1978]. In Italy, drilling has been undertaken with the aim of disposing of high-level waste in a deep hole in clay formations. Reports so far have provided a considerable amount of drilling data, but the feasibility of the project has not been fully assessed [Mittempergher, 1979].

The term 'shale' is applied to a range of materials which consist predominantly of quartz grains and clay minerals, and have a broad range of physical properties. Underground openings can be constructed at greater depths in shale than in clay, but the support system is likely to be a major requirement of any construction. Shales may have considerable fracture permeability, but this will be greatly reduced at depth. The regional groundwater system model would have to take account of rock types other than shale since they also influence the hydrogeologic system. Strata with much greater permeability than shale are likely to provide the major site-specific control within the groundwater system.

Near-surface in-situ heater tests are in progress in the Eleana Argillite at the Nevada Weapons Test Site, and the Conasauga Shale in

the Oak Ridge Reservation, Tennessee. A preliminary evaluation of the response of the Eleana Argillite to heating indicates that the behaviour of expandable clays is a major concern [Lappin and Olsson, 1979]; shrinkage of clay as water is driven off causes pre-existing joints to open, and results in a marked reduction in in-situ thermal conductivity. In the water-saturated Conasauga Shale, the most pronounced effect of the near-surface heater tests was the increased chemical activity in the presence of groundwater whereby metal corrosion and the alteration of borosilicate glass were considerably enhanced by refluxing steam. Despite the water-saturated nature of the shale, conduction rather than convection was the principal mode of heat transport [Krumhansl and Sandberg, 1979].

The Eleana Shale is in an elevated area of Nevada where the main water table is at a depth of about 1500 m. The information available suggests that most radionuclides would become fixed within the thick sequences of unsaturated rock above the water table. The main hydrogeologic input into this investigation should identify the perched water tables and aquicludes, and ascertain the conditions under which seepages to the surface may take place. Exceptional events such as the 500-year maximum precipitation recharge need to be considered.

10.3 Plugging Boreholes and Shafts in Waste Repositories

Plugging boreholes and shafts was the subject of a workshop organised by the OECD-NEA Coordinating Group on Geologic Disposal at Columbus, Ohio, in May 1980. The materials under investigation for plugging generally include a proportion of bentonite which contains water-expansive smectite clays. Where a repository is sited in crystalline rock or in basalt that contains fractures, a clay medium is preferred to block the movement of groundwater into or out of the repository.

Sweden proposed to use highly compacted bentonite and mixtures of bentonite and quartz grains as buffer materials in radioactive waste repositories in crystalline rock. Experiments are being conducted to examine the effects of water uptake and temperature increase on the swelling and physicochemical properties of plugging materials. The effectiveness of the sealing of fractures and joints in crystalline rock by using bentonite grouting techniques, and electrophoresis is also being evaluated [Pusch et al., 1980].

The US is carrying out shaft and borehole plugging studies in the Columbia River Basalt, and in salt in south-eastern New Mexico; the

development of cement mixes, and the long-term durability of plugs is also being investigated. Similar plugging concepts have also been developed by the FRG, the Netherlands and Belgium; in all cases a combination of cement and natural materials derived from the formations that have been penetrated has been proposed.

A maximum temperature of 100°C at the canister surface has been recommended [INFCE, 1980] for the proper functioning of backfill materials such as bentonite mixtures.

The use of brucite and periclase as high temperature sealant materials was proposed for sealing SYNROC in a deep-hole repository [Ringwood, 1978].

The OECD-NEA Co-ordinating Group on Geologic Disposal has proposed that an ad hoc group of experts should consider the functions of borehole plugging and shaft sealing, and the criteria that should be met as a preliminary step towards reaching international agreement. Considerable practical work remains to be done before international agreement on satisfactory methods of shaft and hole sealing is attained.

Although the general methods by which the plugging of boreholes and shafts may be attained are accepted, the specific methods to be used and proved for a repository will depend very much on the specific properties of the host rock.

11. ISOLATION OF BURIED WASTE

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, followed by leaching of radioactive constituents from the waste form, is generally considered to be the most probable pathway. No waste form has yet been developed that is totally insoluble in water; the greatest leaching rates could occur during the initial few hundred years after burial, owing to the local increase in the temperature of the geologic medium and contained groundwater through fission product decay heat.

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 at least 1000 years. These packages consist of the radioactive glass or ceramic, sheathed in corrosion-resistant metals such as lead and titanium [KBS, 1978a], and 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 envir-

onment which can absorb radioactive materials which might eventually be leached from the waste form. The composite waste package is surrounded by a bentonite liner during placement in the geologic medium. The host rock may provide a further barrier through chemical interaction or absorption of some radionuclides leached from the waste package [Roy, 1981]. The probability that waste material buried in an argillaceous (clay) formation will breach the barriers and enter groundwater has been estimated by fault tree analysis [D'Alessandro et al., 1980] as follows:

Time following burial	Probability of release
1000 y	$5 \times 10^{-5} - 2 \times 10^{-4}$
10 000 y	$2 \times 10^{-4} - 3 \times 10^{-3}$
100 000 y	$2 \times 10^{-3} - 2 \times 10^{-2}$

Greater probabilities of release could be expected from crystalline rocks, such as granite, which are not self-sealing.

12. SAFETY OF A HIGH-LEVEL WASTE REPOSITORY

Smith et al. [1980] reviewed several perspectives on potential risks from buried high-level wastes. Hazard indices were based on the amount of water required to dilute a unit quantity of a substance to drinking water standards. A radiotoxic hazard index [USNRC, 1976] was used to compare the total radiotoxicity of fission products and actinide elements in high-level waste, plus associated geologic medium, with that of an equal mass of a uranium ore body. The US Department of Energy [USDOE, 1980a], after Hamstra [1975], defined a relative toxicity index to compare the radiotoxicity of high-level waste from 1 t of spent LWR fuel with that of the uranium ore mined to manufacture the fuel.

The relative toxicity index for high-level waste and uranium ore is compared in Figure 3 with similarly developed toxicity indices for ranges of non-radioactive ores. The toxicity index of the high-level waste after 80 years in geologic burial is comparable with that of average mercury ore. After 200 years, it is comparable with lead ore, and after 1500 years, the hazard index of the waste is similar to that of the ore from which it came [USDOE, 1980a]. A longer period of isolation is indicated for unreprocessed fuel because of its uranium and plutonium content. It must not be inferred that 1500 years is an adequate period of isolation for high-level waste; uranium, lead and mercury ores are

chemically toxic substances also. The comparison is useful only to indicate the storage time necessary to reduce hazard levels of high-level wastes to those of familiar ores present near or at the Earth's surface.

13. CONSEQUENCE ANALYSES OF FAILURE OF A HIGH-LEVEL WASTE REPOSITORY

Generic assessments of radiological doses resulting from ingress of water into a high-level waste burial site have been conducted in Sweden [KBS, 1978a], the UK [Hill, 1979; Hill and Lawson, 1980], the US [USDOE, 1980a] and by the Commission of European Communities [Girardi et al., 1978]. These studies have assumed that water ingress to the waste occurred after 1000 years of geologic burial.

13.1 The KBS Study

The Swedish study considered high-level wastes resulting from 330 GWe y of nuclear power, generated by 13 LWRs over 20 years. A total of 9000 t U of spent fuel was reprocessed and the high-level wastes were vitrified. The vitrified waste was assumed to have been stored for 40 years before disposal to reduce heat from fission product decay. Canisters of waste were encapsulated in lead and titanium, and buried in bentonite at a depth of 500 m in a granite formation at temperatures not exceeding 80°C.

Corrosion studies indicated that groundwater would contact the vitrified waste from 1000 to 6000 years after burial, when temperatures had fallen to 25°C. The waste was assumed to have been fractured through earlier thermal stresses and its surface area increased five-fold. Leaching rates of borosilicate waste glass, measured in the laboratory at 25°C, indicated a conservative period of 30 000 years for total dissolution of the residual radionuclides. The contaminated groundwater was assumed to percolate slowly upwards through the granite rock to three locations: a source of drinking water, a lake and the Baltic Sea.

Retardation and decay of the radionuclides during passage were calculated from adsorption coefficients measured on Swedish granites. Radiological doses were estimated for hypothetical populations exposed by drinking water from deep wells near the repository, by using water from the lake to irrigate food crops, by ingestion of fish, and exposure to sea-water and coastal sediments. Maximum individual doses were received from the drinking well; doses from the lake and the ocean model pathways were lower by factors of 20 and 2000 respectively [KBS, 1978a].

Figure 4 illustrates the pattern of individual doses received over

a 30-year period for a most probable and an upper limit case. Individual radiological whole body equivalent doses derived from radionuclides from the buried waste rose to a maximum annual value of 0.15 mSv (15 mrem) after about 200 000 years. This dose is equivalent to less than 8 per cent of the annual dose from an 'average' natural background level of 2 mSv y^{-1} (200 mrem y^{-1}) [UNSCEAR, 1981]. A greater dose could be received today from the radium-226 contained in drinking water from some granite areas in Sweden. Similar low individual doses were estimated in the Swedish KBS-II study on the deep geologic burial of unreprocessed spent fuel, encapsulated in corrosion-resistant copper canisters [KBS, 1978b].

Table 7 lists the maximum radiological doses to individuals and the major radionuclides responsible for the dose from the waste repository. These maxima are not additive, as the time at which the maximum dose per nuclide was estimated to be reached ranged from 7 x 10^3 y (iodine-129) to 5 x 10^5 y (caesium-135) after the repository was filled and sealed. At the time corresponding to the maximum individual dose (2 x 10^5 y), neptunium-237 was predominant, contributing 63 per cent of the maximum, followed by uranium-233 (19 per cent) and radium-226 (15 per cent). It is of interest that the contribution from plutonium-239, a radionuclide of major public concern, was estimated at less than 0.001 per cent of the maximum dose.

The precision of radiological dose estimates in the KBS and similar generic studies depends on the validity of the mathematical model and the values of parameters used for calculation. Johanssen and Steen [1979] have claimed that uncertainties about some radionuclide adsorption coefficients of granite and its permeability could increase the KBS dose estimates by between one and two orders of magnitude. Independent reviews of the KBS studies have agreed with the leaching characteristics adopted for glass [Ross et al., 1978], and generally with the methodology and overall conclusions for a well-engineered, bedrock repository in a chosen site [NAS, 1980]. It was noted that the ability of granite to adsorb radionuclides would be far more critical if water penetrated the waste packages within the first few decades, and if bentonite was not added as overpack and backfill. In an investigation of dose estimate variability conducted for the KBS Study, Bergström [1981] recognised the importance of equilibrium constants for soil-water systems and the need for further studies on radiological intake through food chains. Confirmation of adsorption coefficient measurements of radionuclides in

granite is a major part of the in-situ geological studies underway at Stripa in Sweden [Witherspoon et al., 1981].

13.2 Other Consequence Studies

Maximum individual radiological doses from high-level waste disposal estimated in five independent generic studies have been normalised and compared in Figure 5. The consequences of repository failure in the first year after filling and decommissioning have been assessed [USDOE, 1979a], but this case was subsequently discounted as a credible situation and it was considered that detection and remedial action would protect individuals from the high doses estimated [USDOE, 1980a]. All other studies examined have assumed 1000 years of isolation for the waste before its dispersion by groundwater.

Dissolution times of the waste in groundwater, ranging from a few hundred to several tens of thousands of years, did not have a major effect on the maximum doses estimated. The effect of different food chains and the dilution mechanism indicated some advantages for coastal siting of a repository; estimated doses were between one-sixtieth and one-five hundredth of those estimated for an inland site [Hill and Lawson, 1980]. Lowest doses were estimated for waste packages disposed of on the deep ocean floor [Camplin et al., 1980], although this type of disposal of high-level waste is expressly prohibited by an international convention.

13.3 The Significance of Neptunium-237

The transfer fraction of neptunium-237 from the gastro-intestinal tract into the blood has been recognised recently as a parameter which requires further study and which could have significant influence on maximum individual radiological doses [Hill, 1979; Bergström, 1981]. Neptunium is a major contributor to estimates of the maximum dose: a value of 10^{-2} was recently recommended by the International Commission on Radiological Protection for the transfer fraction of neptunium-237 [ICRP, 1979]; by comparison, that previously recommended was 10^{-4} [ICRP, 1959]. This increased transfer fraction is reflected in the difference between the estimate of 5000 $\mu Sv\ y^{-1}$ (500 $mrem\ y^{-1}$) by Hill and Lawson [1980] and of 150 $\mu Sv\ y^{-1}$ (15 $mrem\ y^{-1}$) by KBS [1978a]. An even greater maximum individual dose of 60 $mSv\ y^{-1}$ (6 $rem\ y^{-1}$) was estimated from neptunium-237 in earlier studies [Hill & Grimwood, 1978; Hill, 1979] which used conservative assumptions in the models recognised as leading to overestimates of potential doses.

The significance of the potential dose from neptunium-237 in a

waste repository is subject to an uncertainty of up to two orders of magnitude. The increased value for the transfer fraction was based on a small number of experiments on rats; it has been recognised that the fractional absorption of trace quantities of neptunium may be a factor of ten lower, as may also be the fractional absorption of neptunium incorporated in food [ICRP, 1979]. Evaluation of factors influencing the transfer fraction for neptunium is part of the current AAEC program at the Lucas Heights Research Laboratories.

13.4 Sensitivity of Dose to Changes in Parameter Values

The sensitivity of the maximum individual annual dose to some changes in parameter values was estimated by Hill [1979] in a model with conservative parameter values. An order of magnitude alteration in parameter values in the reference case resulted in the following reduction factors on the maximum individual dose:

Parameter	Reduction factor
Sorption	20
Groundwater velocity	12
Flow path length	8.6
Leach rate	6
Dispersion coefficient	~2

Hill [1979] concluded that the rate of migration of radionuclides through the biosphere was of primary importance in determining potential dose, and that the leach rate of the waste form was of secondary concern in a wide range of circumstances.

The reduction factor is also dependent on parameter values. Table 8 illustrates application of the data to the model by Hill and Lawson [1980]. Sorption is of less importance because of the assumption that 10 per cent of the neptunium would be present either in the strongly or weakly adsorbed form. Groundwater flow at lower velocities assumes greater significance in reduction of dose. The estimated sensitivities (reduction factors) corresponding to one order of magnitude change in parameter values are: groundwater velocity (55); dispersion coefficient (7); leach rate (8); sorption (3); flowpath length (3).

13.5 Minimum Performance Criteria for Repositories

Following these generic studies, minimum performance criteria were

proposed for high-level waste packages and underground facilities in the US [USNRC, 1981]. These are both to be designed so that, assuming anticipated processes and events, and including full or partial saturation,

- the waste packages will contain all radionuclides for at least 1000 years after permanent closure;
- at any subsequent time, the annual release rate of any radionuclide into the geologic setting shall not exceed 10^{-5} per year assuming that no prior release had occurred; and
- this limitation excludes radionuclides contributing less than 0.1 per cent of the total permissible annual release.

The US Nuclear Regulatory Commission (USNRC) has requested comment on whether aims for containment and release rates should be redefined to be "as long and as low as are reasonably achievable". The Federal Republic of Germany is reported to believe that the time range for consideration of waste form stability should be limited to 10 000 years, beyond which, rational scientific predictions cannot be made [IAEA, 1980]. The US has considered it impractical to apply standards for disposal of uranium mill tailings for as long as 10 000 years [USEPA, 1980]; hazard indexes of high-level waste and uranium ores are roughly equivalent after 1500 years.

14. RADIATION DOSES AND RISKS FROM A HIGH-LEVEL WASTE REPOSITORY IN PERSPECTIVE

The basic objective of radioactive waste disposal is the protection of man and his environment from an unacceptable level of risk. For radiological protection, the International Commission on Radiological Protection (ICRP) has assumed conservatively that all radiation exposures may carry some risk of induced disease, disability or death [ICRP, 1977], the level of risk being directly proportional to the radiation dose received. Generic studies indicate that geologic burial of high-level wastes may, after several thousand years, result in an increase in the natural radiation background in the vicinity of the repository. Individuals in this vicinity may, therefore, receive a fractional increase in their normal annual dose. The risk of death from this addition may be insignificant but it is assumed not to be zero.

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, from the natural radiation background, and incurred from man-made

sources of radiation [IAEA/WHO, 1981; IAEA, 1979].

14.1 Radiation Doses from Nuclear Power and the Nuclear Fuel Cycle

An average global annual radiation dose of 0.6 μSv y^{-1} from nuclear power production has been estimated. The largest contributors to local and regional collective doses [UNSCEAR, 1981] were from uranium mining and milling (75%), nuclear power reactors (17%) and spent fuel reprocessing (7%). An average annual dose of 3 μSv y^{-1} has been estimated from nuclear energy in the UK, with maximum annual doses to members of the public during the late 1970s [NRPB, 1981] derived from reprocessing (1.5 mSv y^{-1}), nuclear reactor operation (300 μSv y^{-1}) and uranium upgrading and fuel manufacture (55 μSv y^{-1}). A lower maximum annual dose of 880 μSv y^{-1} was estimated for 1981 [BNFL, 1982]. This followed a reduction in the amount of ^{137}Cs discharged annually in liquid effluent, together with a reduced estimate of marine food consumption by the critical group. Maximum annual doses of several hundred microsieverts have been reported for UK nuclear research and development establishments [Taylor and Webb, 1978]. A maximum annual dose of 10 μSv y^{-1} has been estimated for critical groups from operation of the AAEC Lucas Heights Research Laboratories.

14.2 Radiation Doses from the Natural Background

Recent consideration of internal doses from ingestion of short-lived daughters of ^{220}Rn and ^{222}Rn [NRPB, 1981; UNSCEAR, 1981] have increased estimates of typical annual radiological doses from natural background from about 1 mSv y^{-1} (100 mrem y^{-1}) to 1.8 - 2 mSv y^{-1} (180 - 200 mrem y^{-1}). Additional radiation doses are incurred from chest and dental X-rays (0.4 and 0.2 mSv respectively), and from international air travel (Table 9).

Individuals are able to vary their natural radiation background to some extent by choice of residence and lifestyle. For example, the annual radiation dose from cosmic rays may be reduced by about 0.1 mSv y^{-1} if a person chooses to reside in Sydney (sea level) in New South Wales instead of, say, Canberra. In Australia, sunbathing for 2 hours per day on Kingscliff Beach in New South Wales can result in an annual external dose of 320 μSv y^{-1}, which is equivalent to a continual exposure rate of 3.8 mSv y^{-1}; by contrast, sunbathing for the same period on a Woronora River beach, also in New South Wales, would incur an annual external dose of 29 μSv y^{-1} [Hespe, 1979]. Thorium in beach sands at Kerala, India contributes to an abnormally high background of up

to 20 mSv y^{-1} (2100 mrem y^{-1}). Living in a brick dwelling rather than a wooden home can increase the annual individual dose by about 0.7 mSv y^{-1}. A return visit by air to the UK from Australia incurs an individual dose of about 0.3 mSv. By comparison, the maximum individual dose estimated for the waste repository is 0.13 mSv y^{-1} [KBS, 1978a]; that in the study by Hill and Lawson [1980] was 5 mSv y^{-1}. Both of the maximum doses would occur after about 200 000 years.

14.3 Risks from Radiation in Perspective

The risk of death from cancer from radiation averaged over age and sex [ICRP, 1977] is about 10^{-2} Sv^{-1} (10^{-4} rem^{-1}). Exposure to a natural radiation background of 2 mSv y^{-1} (200 mrem y^{-1}) carries, therefore, a proportionate risk of death of 2 in 100 000 per year of exposure.

A similar statistical level of risk applies to the following societal activities in the UK [Flowers, 1976; Parker, 1978]:

- smoking 30 cigarettes,
- travelling 1600 km by car,
- travelling 8000 km by plane,
- rock climbing for 30 minutes,
- canoeing for 2 hours,
- engaging in ordinary factory work for 30 weeks, and
- simply being male and aged 60 for 7 hours.

The upper limit of radiological dose estimated for the KBS repository [KBS, 1978a] and by Hill and Lawson [1980] corresponded to about one-eighth and 2.5 times the risk from these activities respectively. These levels of individual risk must be weighed against the benefit of the 330 GWe y of electrical energy produced.

15. COST OF HIGH-LEVEL WASTE DISPOSAL

The US has published [USDOE, 1979a, 1979b, 1980] economic cost estimates for the complete disposal scenario (e.g. vitrification, short-term storage, long-term storage and repository costs).

These US studies assumed manufacture of glass with a high waste loading (30 wt% calcine) and burial of waste from spent fuel 6.5 to 10 years after its discharge from the power station. These parameters resulted in estimates of temperatures of over 350°C for the buried glass. It is speculative whether such high temperatures are acceptable. They do not appear realistic, at least in the near term, as some spent fuel is likely to be 20 years old before reprocessing operations could commence in the US. However, waste loadings of 19 and 25 wt% calcine were achieved in the engineering scale US nuclear waste vitrification

project on LWR fuel five and seven years after discharge [Bonner et al., 1980].

By contrast, many European countries propose to manufacture glass at reduced waste loadings (9 - 15 wt% calcine), and to store the glass in near surface facilities for several decades to reduce the heat output of the waste through fission product decay. The waste, encapsulated in packages designed to reduce water ingress for at least 1000 years, would finally be placed in geologic formations at spacings sufficient to ensure that the temperature of the external surface of the glass does not exceed 100°C.

The US data, modified to a reduced waste and thermal loading, have been used in this study to estimate costs for the European proposals, capital and operating costs being derived from a discounted cash flow analysis. Comparative costs of the US and European strategy are listed in Table 10. Details of the technical and economic ground rules used in this assessment are given in Appendix A.

The European proposals resulted in estimates of increased costs for the vitrification operation owing to the increased volume of waste glass per tonne of spent fuel. A substantial cost penalty was estimated for extended storage of glass before geologic burial. This penalty ($US29-60/kg HM*) was based on application of US derived charges for storage and shielding of individual waste canisters. By contrast, recent unpublished French estimates for temporary storage of vitrified glass over 30-50 years were significantly lower ($14/kg HM), but involved an unspecified degree of discounting of future costs.

Greater operating charges for geologic burial were estimated for the greater volume of waste in the European case. However, a repository of a given size (800 ha) could accept a greater quantity of aged waste (137 000 tonnes), where both mechanical and thermal considerations were limiting factors. The repository capacity for fresh waste was limited to 89 700 tonnes because of temperature restrictions in the geologic medium.

16. CONCLUSIONS

(a) The studies and programs being coordinated by the OECD-NEA and IAEA are making a major contribution to the development of safe and acceptable procedures for the disposal of high-level radioactive waste from the nuclear power industry.

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* HM = heavy metal (U + Pu)

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

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

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

(e) The technology for disposing of radioactive wastes in some geologic formations is available.

(f) 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.

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

(h) 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.

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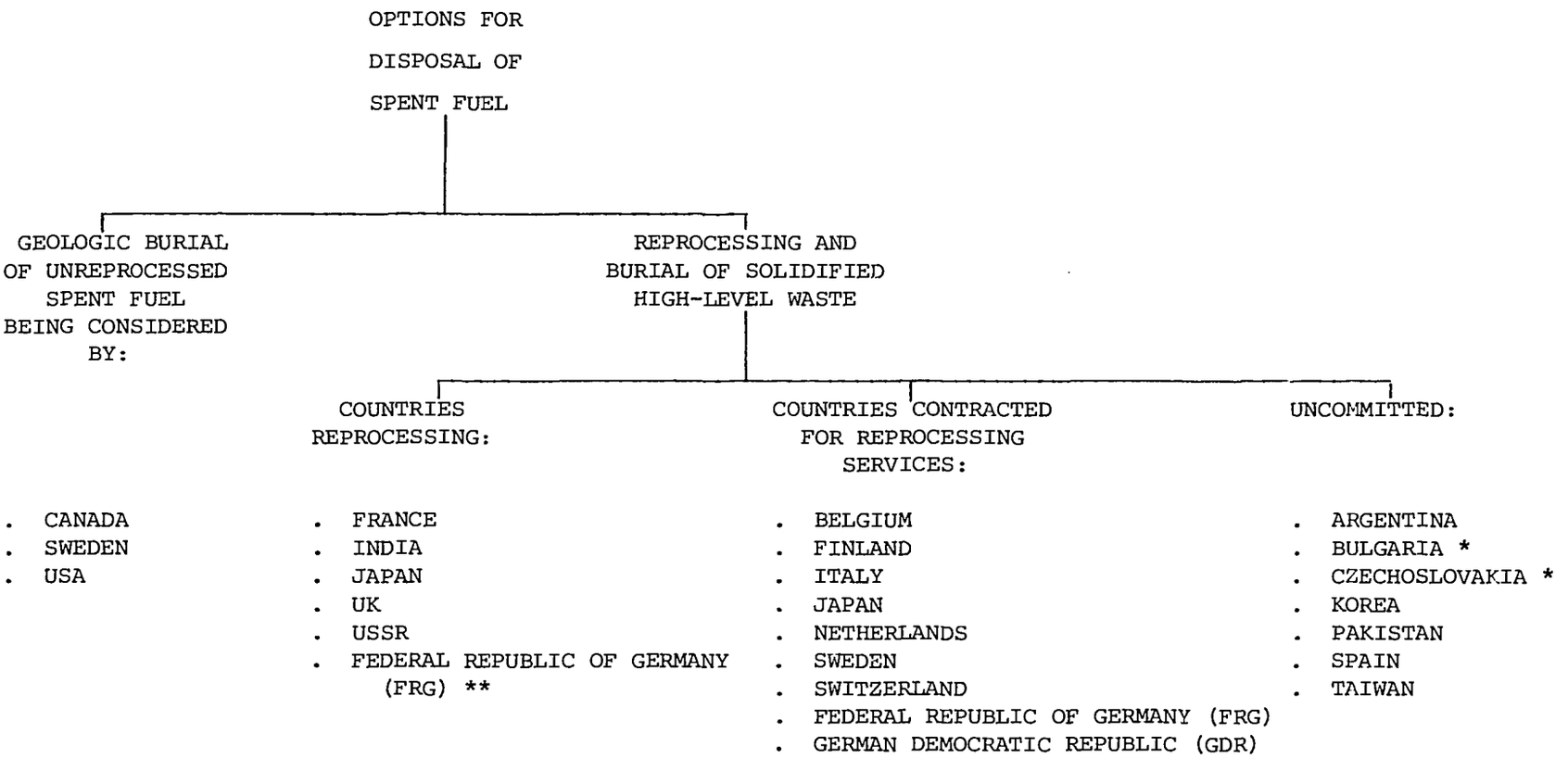
TABLE 1

ATTITUDES TOWARDS REPROCESSING IN COUNTRIES WITH NUCLEAR POWER STATIONS ABOVE 30 MW



* Believed to have options for reprocessing in the USSR.

** Plant understood to be under maintenance.

TABLE 2

NATIONAL PROGRAMS FOR REPROCESSING SPENT FUEL FROM COMMERCIAL NUCLEAR POWER GENERATION

COUNTRY	REPROCESSING CAPACITY AND PLANS
Belgium	The 60 t U y^{-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 1200 t U y^{-1} UP1 plant at Marcoule, and also since 1967 in the 900 t U y^{-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 y^{-1}. This capacity is being progressively expanded, and new plant, UP2-800, with 800 t U y^{-1} capacity is scheduled for operation by the mid-1980s. A third plant, UP3A, of capacity 800 t U y^{-1} is under construction and expected to operate in 1985. A duplicate plant, UP3B, is also under consideration. France has international reprocessing contracts involving a total of about 6000 t U of LWR fuel.
Finland	Spent fuel of Soviet origin is to be returned to the USSR for reprocessing.
Federal Republic of Germany (FRG)	The experimental 35 t U y^{-1} WAK plant at Karlsruhe, operational since 1971, was reported closed in May 1980 for repairs to equipment. Construction of a 350 t U y^{-1} plant has been proposed in Hesse State; similar plants are under consideration for the Rhineland Palatinate or Lower Saxony. A total of 1700 t U of spent LWR fuel is contracted for reprocessing in France.
India	The 60 t U y^{-1} Trombay plant, reprocessing natural uranium metal fuel, became operational in 1965. The 100 t U y^{-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 y^{-1} for spent oxide fuel from heavy water reactors is intended to be operational in the late 1980s at Kalpakkam.

TABLE 2 (Cont'd.)

COUNTRY	REPROCESSING CAPACITY AND PLANS
UK	The 1000 t U y^{-1} B204 plant reprocessed natural uranium gas-graphite fuel in the 1950s and early 1960s. The 2000 t y^{-1} B205 plant 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 1200 t U y^{-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 3100 t U of spent fuel.
USA	The 300 t U y^{-1} plant at West Valley, N.Y., operated intermittently from 1966 to closure in 1972. A novel 300 t U y^{-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 1500 t U y^{-1} plant at Barnwell, South Carolina.
USSR	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 have also negotiated for return of similar spent fuel from Finland.
Italy	The Eurex pilot plant at Saluggia has capacity equivalent to about 10-20 t U y^{-1} of LWR fuel and is used for reprocessing research and development.
Japan	A small 100-140 t U y^{-1} demonstration reprocessing plant at Tokai Mura has been in operation intermittently since 1977 on LWR fuel. A commercial 1200 t U y^{-1} plant for reprocessing LWR fuel is planned for operation in 1990. Japan has contracted for reprocessing 1600 t U LWR fuel in France and 1600 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	700 and 140 t U of LWR fuel have been contracted for reprocessing in France and the UK respectively.
Switzerland	470 t U has been contracted for reprocessing in France.

TABLE 3

CANDIDATE WASTE FORM CATEGORIES

Type	Waste Form
Glass	Porous glass Phosphate glass Borosilicate glass High silica glass Glass ceramics
Ceramic	Supercalcine SYNROC Titanates
Matrix	Metal matrixes Multi-barrier Cermets

TABLE 4

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

High-level Waste Solidification			Geologic Burial	
Country	Preferred Waste Forms	Developments	Preferred Formations	Developments
Australia	SYNROC	Non-radioactive large-scale production development, together with leaching trials at high temperatures and pressures on inactive and irradiated specimens is being conducted. Tests with specimens containing fission products and actinides are planned.	-	-
Belgium	Borosilicate glass beads in lead matrix (PAMELA Vitro-met) Borosilicate glass blocks (AVM).	Construction of a PAMELA plant at Mol to vitrify 30 L h^{-1} of HLW from former Eurochemic operation has commenced. Negotiations are proceeding for construction at Mol of vitrification plant using French technology for future reprocessing operations.	Clay	An underground laboratory under construction at Mol is planned for operation in 1982 to conduct in-situ tests in the clay deposit relevant to geologic disposal.
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 Isolation Pilot Plant (WIPP), planned for operation in 1982, will be 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 2000 for disposal of either unreprocessed 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 6000 MWe nuclear program for 30 y, with burial assumed from 2010.
German Democratic Republic (GDR)			Salt	A waste repository is being developed at Bartensleben.
Finland			Crystalline Rock	Risk assessment studies, economic evaluations.

TABLE 4 (Cont'd.)

High-level Waste solidification			Geologic Burial	
Country	Preferred Waste Forms	Developments	Preferred Formations	Developments
France	Borosilicate glass	The industrial scale AVM plant at Marcoule commenced operation in 1978. About 340 cubic metres of HLW corresponding to reprocessing of over 6000 t of natural uranium gas-graphite fuel were vitrified into 150 t of glass by early 1981. A larger plant of similar design, AVH, is scheduled to be operational at Cap La Hague in 1986 to solidify waste from up to 1600 t U y^{-1} LWR fuel, corresponding to a nuclear generation capacity of about 50 000 MWe.	Granite	It is intended to store glass blocks for 30-50 years in air cooled vaults. Subsequent burial at 1 km depth is being considered.
India	Borosilicate glass	Commissioning of vitrification plant at Tarapur commenced in late 1981. 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 y of storage in an air-cooled underground vault is planned.
Italy	Borosilicate glass	Small scale demonstration vitrification plants are under consideration.	Clay, Salt.	The sediments near the Trisaia Centre in S. Italy are under study.
Japan	Borosilicate glass, zeolite ceramics.	Laboratory scale development is planned by 1982 and a pilot scale vitrification plant for operation by 1986.	Granite	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.
Switzerland	Borosilicate glass	———	Granite	Feasibility studies for burial at depths of up to 1500 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.

TABLE 4 (Cont'd.)

High-level Waste Solidification			Geologic Burial	
Country	Preferred Waste Forms	Developments	Preferred Formations	Developments
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 will be 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 N. Scotland but were discontinued in December 1981. It is proposed to store the vitrified waste at the surface for at least 50 years.
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 for 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.	A waste isolation pilot plant (WIPP) in New Mexico is under construction in salt for operation by late 1980s as a test facility for HLW of military origin. The Nevada terminal waste isolation project in granite is intended as a disposal site for transuranic wastes and as a test site for temporary (5 year) geological emplacement of commercial spent fuel.
USSR	Aluminophosphate 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.
Federal Republic of Germany (FRG)	Borosilicate glass, glass ceramics, phosphate glasses.		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.

TABLE 5

GEOLOGICAL PROCESSES THAT COULD THEORETICALLY AFFECT A WASTE REPOSITORY

Process	Comment	Preventive Measures
Earthquake	In an area of seismic instability earthquakes may occur episodically throughout the period of desired isolation; elsewhere there should be no short-term risk.	Avoid areas with recorded significant seismicity, observed crustal stress or evidence of Holocene earthquake activity. Use appropriate design for repository.
Volcanic activity	In an area with evidence of recent (Holocene) volcanic activity episodic recurrences cannot be excluded, but other areas should be safe especially in the short term.	Avoid regions that have experienced Holocene volcanic activity.
Erosion	This may be a continuing process, the rate of which is determined by relief and availability of erosive agents. In a stable area of low to moderate relief, erosion (if any) is generally at rates of millimetres to centimetres per thousand years. Schumm [1963] gives average erosion rates in six US drainage basins of 3 to 7 cm y^{-1}.	Avoid tectonically active areas and areas of high relief; if necessary equate depth of burial to predicted rate of erosion.
Weathering of rock	Weathering of a land surface progresses very slowly. In a stable area the base of weathering would normally be lowered at a rate of millimetres to centimetres per thousand years.	Site repository beneath a mature land surface in a tectonically stable region.
Sea level changes	These may result in either (i) immersion of the site or reduced base level for drainage and erosion (rise in sea level); or (ii) accelerated erosion and lowering of base of weathering (fall in sea level). Changes may result from (a) tectonic movements (long-term except in tectonically active areas); or (b) changes in ice cap volumes (which may be significant in the order of 10^3 to 10^4 years) - see below.	(a) Avoid tectonically active areas. (b) Avoid areas 0-100 m above sea level or take special measures to ensure that immersion or damage to headworks of repository would not prejudice integrity of repository.

TABLE 5 (Cont'd.)

Process	Comment	Preventive Measures
Climatic change	Changes in climate could include reduced or increased precipitation or intensity of precipitation; reduced or increased temperature or temperature range; and greater or lesser seasonal variations. These could affect rates of erosion and weathering. Glacial cycles take of the order of 10^4 years and other significant changes of climate would take similar periods.	Site repository beneath a mature land surface in a tectonically stable area and emplace waste at a depth well below credible effects of climatic change within a period of 10^4 to 10^5 years.
Glaciation of site	Substantial changes from present day ice cover would require marked climatic change which would take thousands of years to develop. Effects of ice advance over site would be to raise water table level, possibly to scour land surface with local removal of perhaps hundreds of metres of rock, and to depress by weight of ice the land surface.	In high latitudes or altitudes increased depth of burial may be desirable. For near-sea level sites see 'Sea level changes' above.

TABLE 6

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. Will be a major source of energy in the future, i.e. rock with a thermal gradient of 20°C per 1000 m 2. Fractures are not self-sealing 3. Low permeability back-fill material required to seal repository 4. The larger the underground opening, the more fractures intersected	Austria Canada Denmark France India Japan Sweden Switzerland

TABLE 6 (Cont'd.)

Type of Formation	Advantages	Disadvantages	Investigating Countries
	5. Low thermal gradients are present in some areas (10°C per 1000 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. Highly 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	Belgium Italy

* Note: 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 6 (Cont'd.)

Type of Formation	Advantages	Disadvantages	Investigating Countries
		5. Little information is available on the migration of fluids within the clay in response to a thermal gradient	
(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

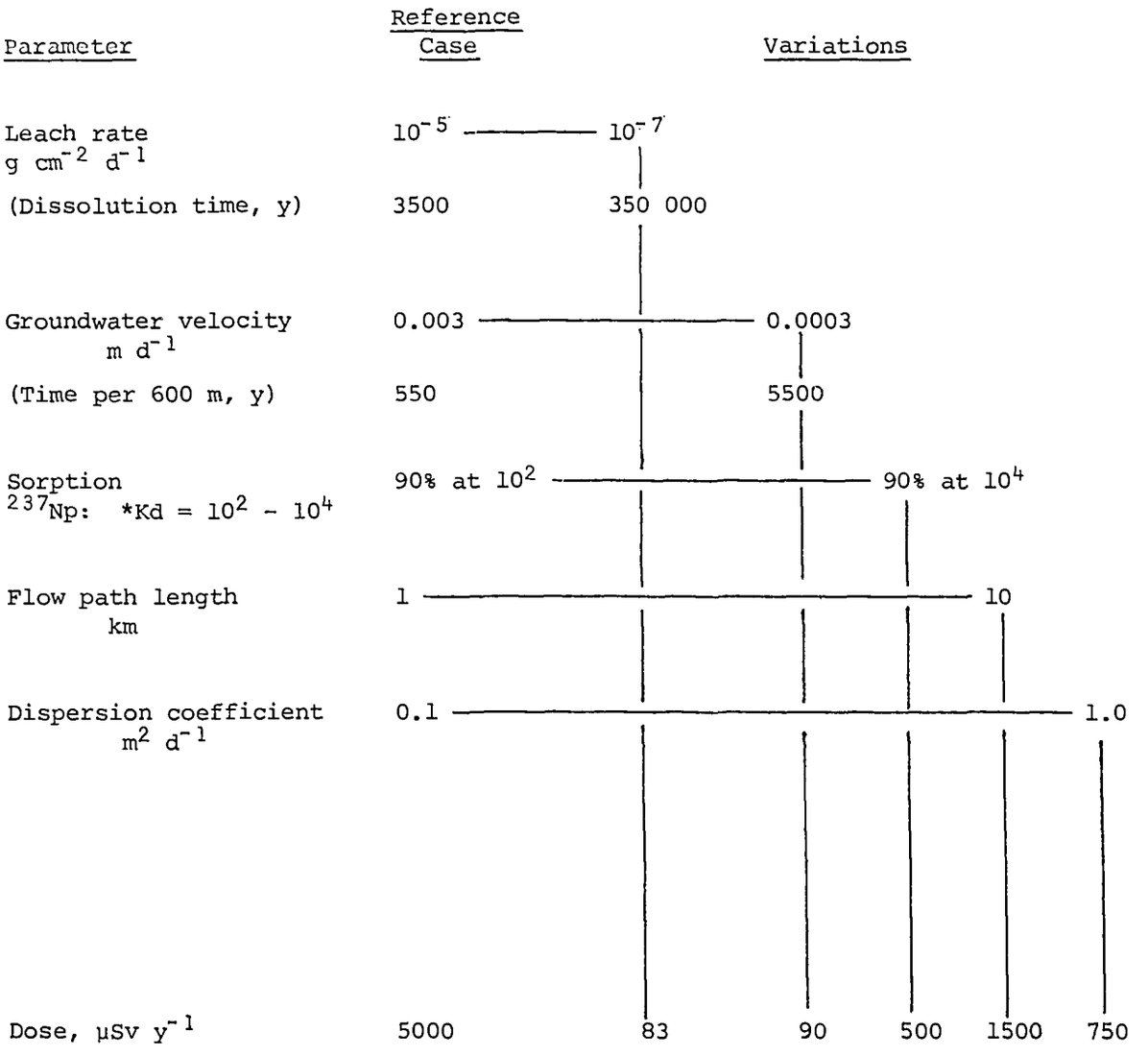
TABLE 7

MAJOR RADIATION CONTRIBUTING TO THE MAXIMUM INDIVIDUAL RADIOLOGICAL DOSE FROM A HIGH-LEVEL WASTE REPOSITORY
[After KBS, 1978]

Radionuclide	Maximum Individual Dose $\mu Sv\ y^{-1}$	Time of Maximum Dose y
^{237}Np	100	10^5
^{99}Tc	23	$1 - 2 \times 10^4$
^{226}Ra	23	8×10^4
^{233}U	23	8×10^4
^{135}Cs	6	5×10^5
^{234}U	3	5×10^4
^{129}I	0.6	$7 \times 10^3 - 7 \times 10^4$
	NOT ADDITIVE	
Maximum Dose at $\sim 10^5$ y	$\mu Sv\ y^{-1}$	%
^{237}Np	100	63
^{233}U	30	19
^{226}Ra	23	15
^{135}Cs	3	2
REST	< 1	1
	157	100%

TABLE 8

PARAMETERS AFFECTING MAXIMUM INDIVIDUAL ANNUAL DOSE
[After Hill and Lawson, 1980]



* Kd = sorption coefficient

TABLE 9

ANNUAL INDIVIDUAL RADIATION DOSE TO HUMANS

Source	Average Annual Dose mSv y^{-1}	Range mSv y^{-1}
NATURAL		
External		
Cosmic rays (sea level)	0.3	0.3 - 1.3 (2500 m)
Ground and building materials	0.3	0.2 - 20*
Internal		
Potassium-40 in body	0.2	
Water & food ingested	0.1	
Air	1.2	
Total, Natural	2.1 (200 mrem y^{-1})	2.0 - 22.8
Source: MAN-MADE RADIATION		
1 Chest X-ray	0.4	0.2 - 0.7
1 Dental X-ray	0.2	
Nuclear explosion tests	0.0003	0.02 - 0.05
Luminous watches	0.02	0.002 - 0.03
Jet air travel		
Sydney - Perth	0.03	
Sydney - London	0.14	

* Thorium in beach sands, Kerala, India.

TABLE 10

COMPARATIVE COSTS OF HIGH-LEVEL WASTE DISPOSAL (US$ 1978)

Operation	European Strategy: Glass - 10 wt% calcine *				USDOE Strategy: Glass - 30 wt% calcine *			
	Capital (10^6 $)	Annual Operating (10^6 $)	Economic life (y)	Levellised costs $/kg HM	Capital (10^6 $)	Annual operating (10^6 $)	Economic life (y)	Levellised costs $/kg HM
Predisposal Spent fuel transport & storage				~60[C]				~60[C]
Spent fuel reprocessing	700	35-59	15	197-208	700	35-59	15	197-208[C]
HLW solidification	91.4	17.9	15	20.0[C]	55	1-7	15	10.4[C]
Rail transport				3.33[C]				3.3[C]
Extended storage at solidification plant or geologic repository	127	34.0 (27.3 y)	100	29.6[F] 60.4[C]	n.a.	n.a.	n.a.	n.a.
Geologic repository	2053	65.3	25	31.95[F]	2053	33.1	16.4	38.1[F]

n.a. - Not appropriate

F - Federal funding

C - Commercial funding

HM - Heavy metal

HLW - High level waste

* N.B. See full definition of European and US strategies in Appendix A.

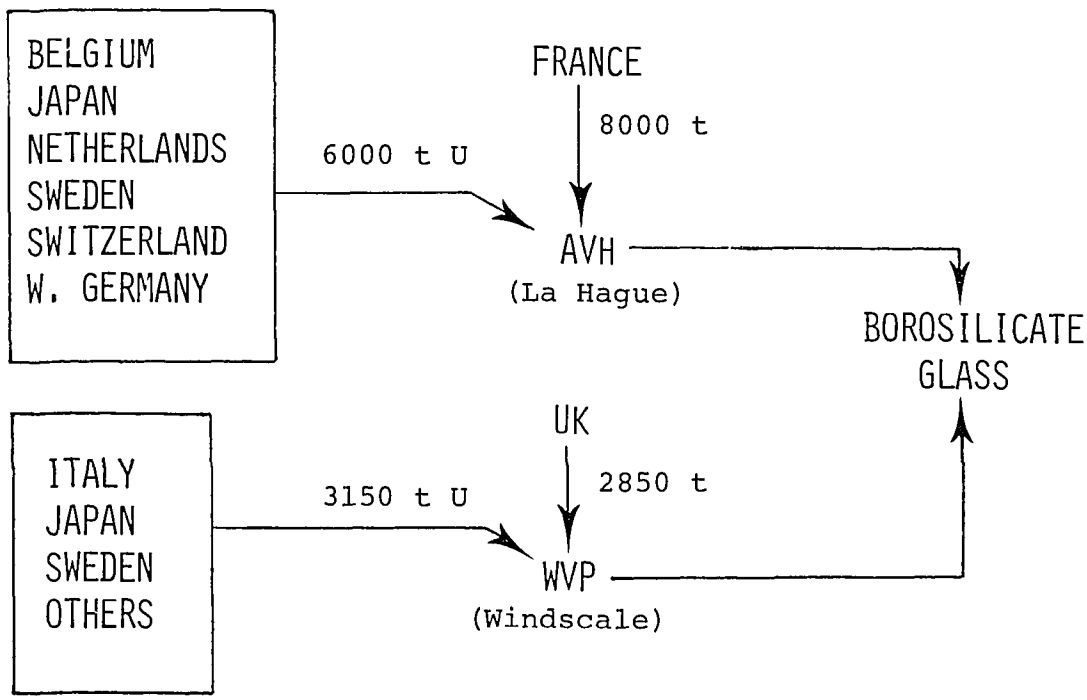


FIGURE 1. REPROCESSING COMMITMENTS BY FRANCE & UK TO 1995

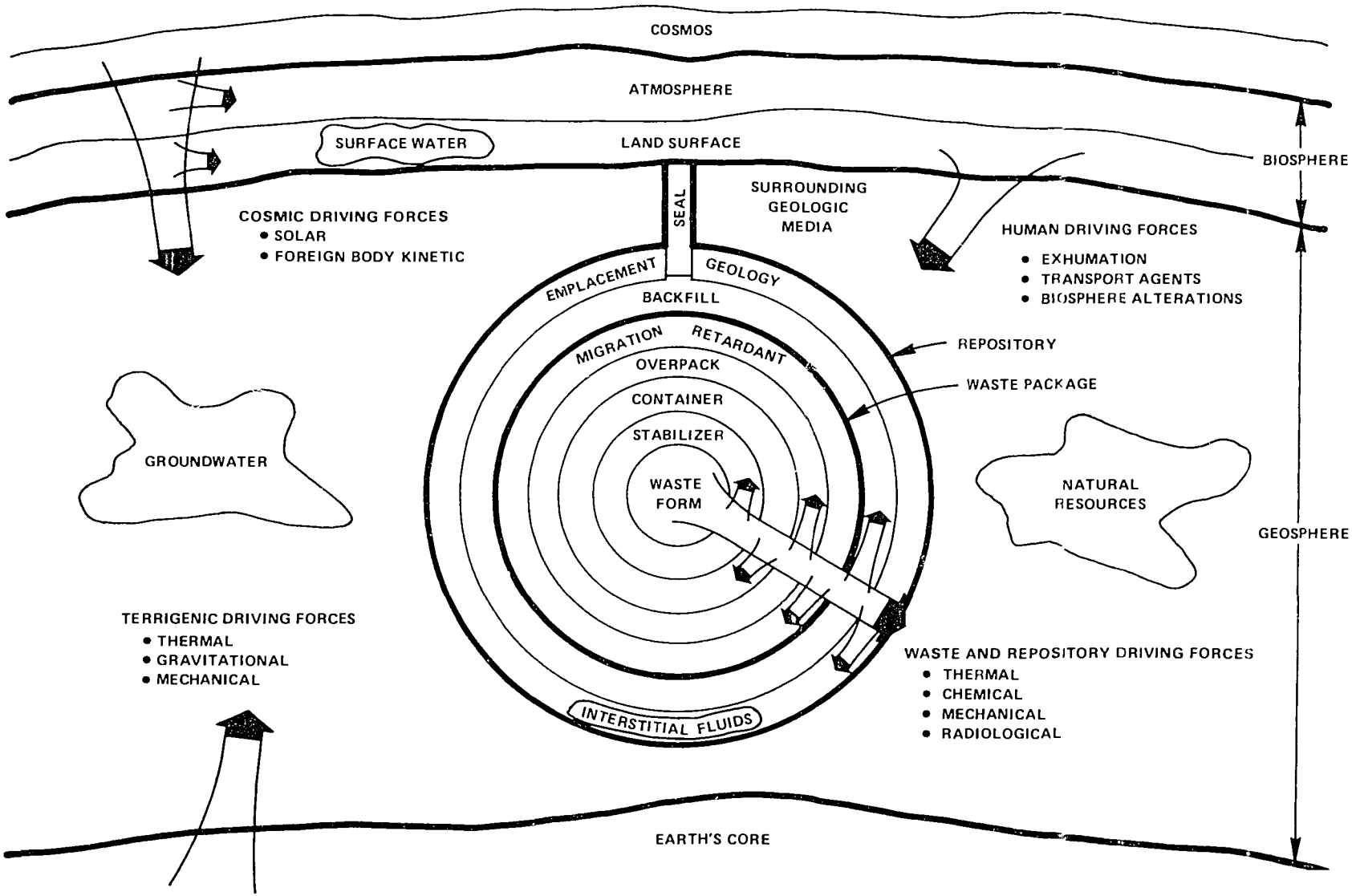


FIGURE 2. WASTE ISOLATION SYSTEM AND DRIVING FORCES (After Burkholder, 1979)

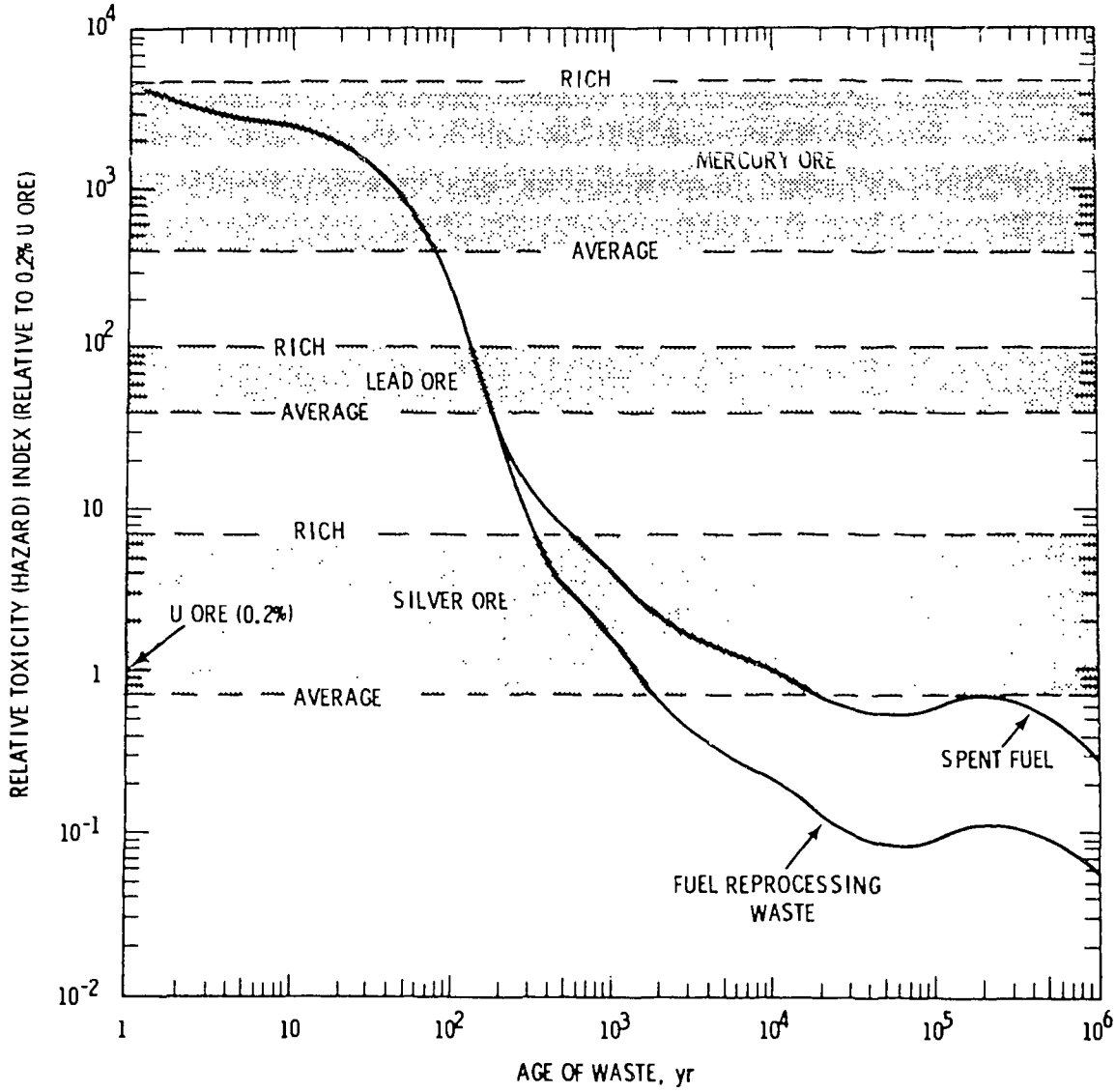


FIGURE 3. TOXICITY OF SPENT FUEL AND REPROCESSING WASTE (After USDOE 1980a)

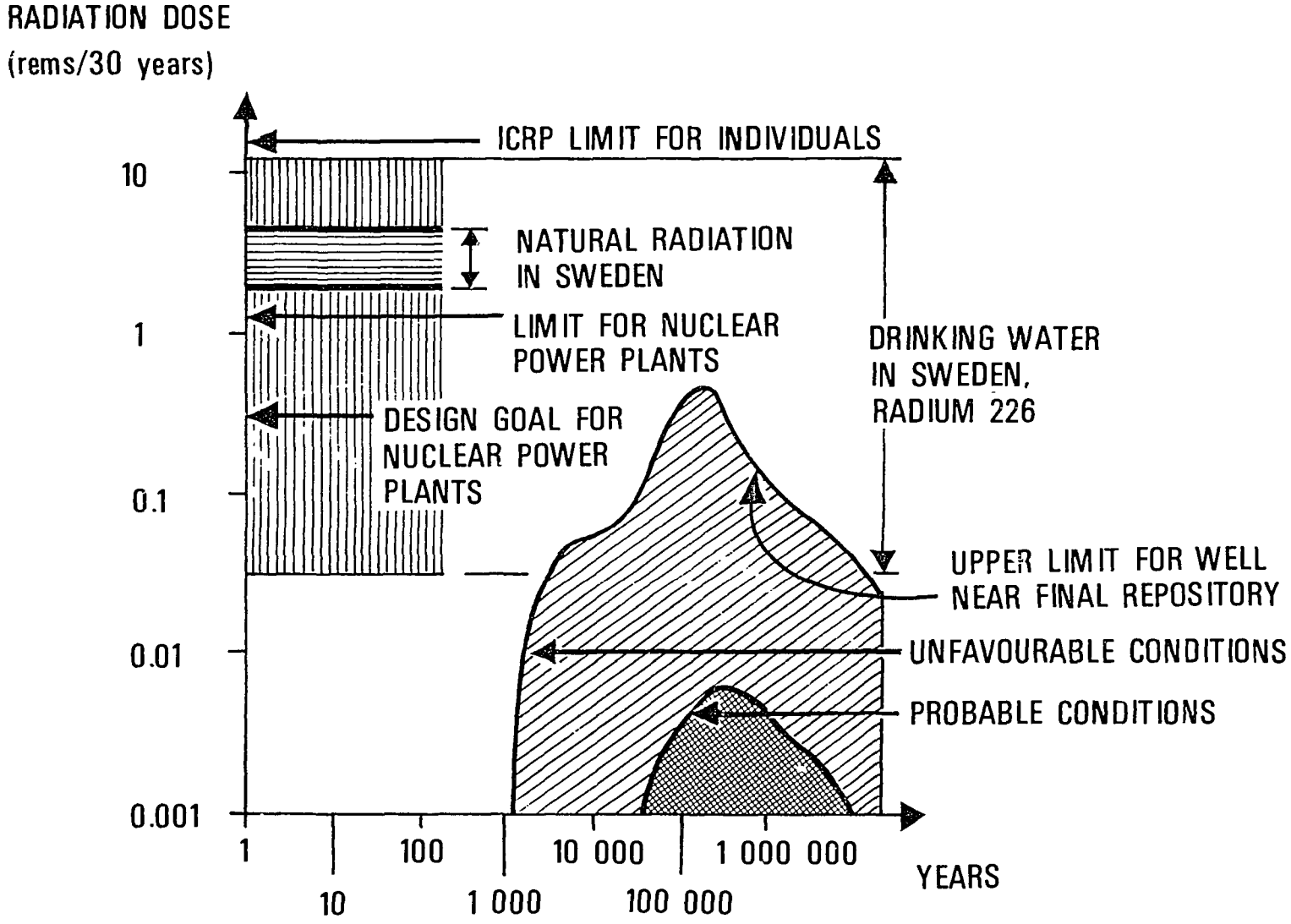


FIGURE 4. ESTIMATED INDIVIDUAL RADIATION DOSE FROM GEOLOGIC BURIAL OF HIGH-LEVEL RADIOACTIVE WASTES FROM 330 GWe y OF NUCLEAR POWER (After KBS, 1978a)

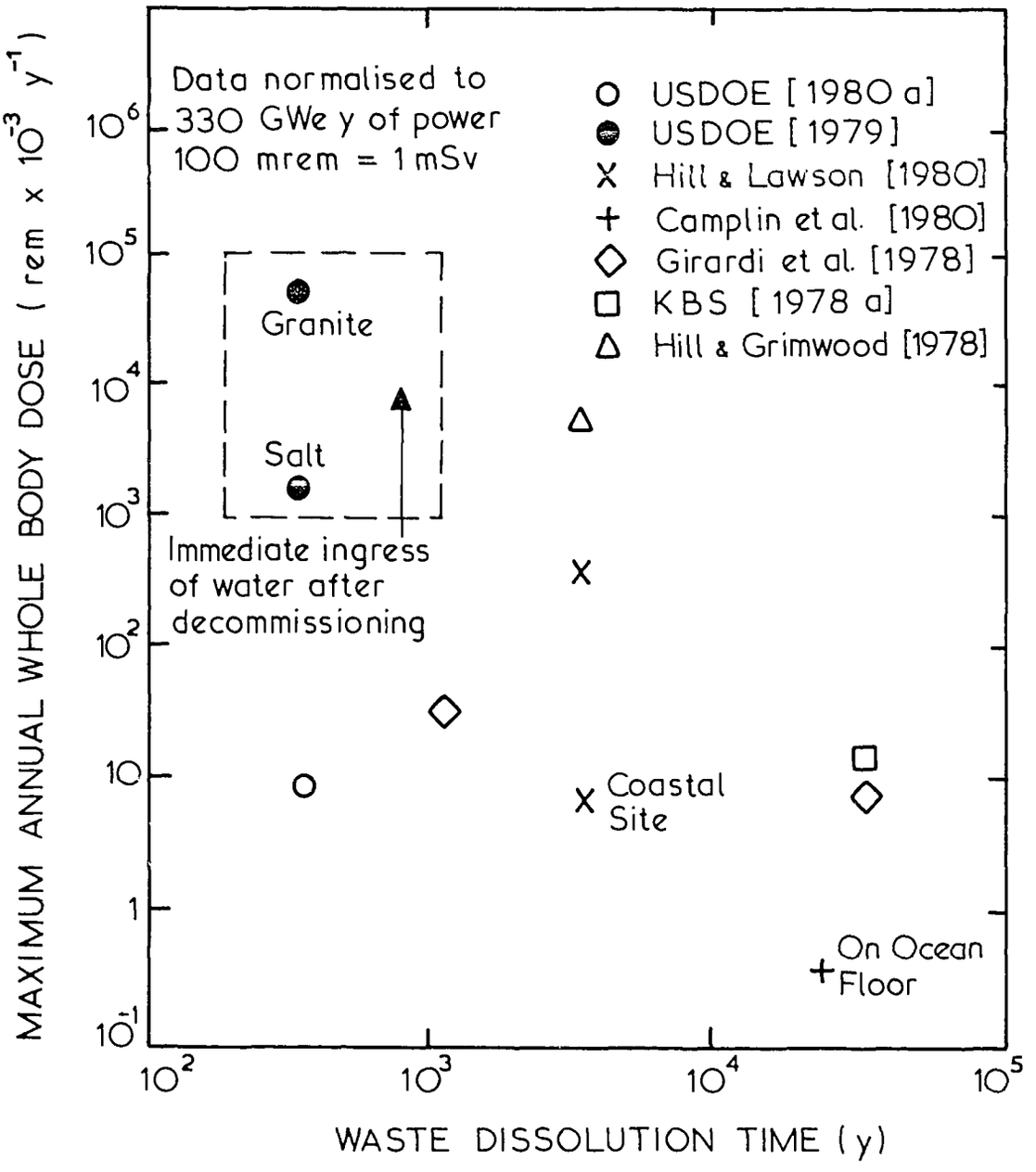


FIGURE 5. ESTIMATE OF RADIOLOGICAL DOSE FROM A HIGH-LEVEL REPOSITORY

APPENDIX A

GROUND RULES FOR COSTING HIGH-LEVEL WASTE DISPOSAL STRATEGIES

A1. STRATEGIES FOR COMPARISON

For costing purposes in this report, disposal strategies have been defined as follows:

'European strategy': Spent fuel reprocessed 10 y after discharge from the power station; vitrified high-level waste (HLW) stored for 30 y before burial in granite.

'USDOE strategy': Spent fuel reprocessed 10 y after discharge from the power station; vitrified high-level waste buried in granite without delay storage.

A2. ECONOMIC COMPARISON GROUND RULES

These follow the procedures adopted in Volume I of the five volume report 'Technology for Commercial Radioactive Waste Management' [USDOE 1979]. Briefly, the ground rules are as follows: levellised unit capital and operating costs are derived from a discounted cash flow analysis, assuming an economic life of 15 years for plant except where otherwise stated. The cost of money was taken to be 10 per cent from commercial sources and 7 per cent from US federal sources, relative prices are assumed not to change and 1976 prices were initially used but updated to 1978 by the factor 1.17 used by USDOE. Unit capital charge rates, i.e. the rates to recover capital and interest, assumed a US federal tax rate of 48 per cent, state tax rate of 6.5 per cent, insurance and property taxes of 7 per cent and a 7 per cent investment credit. For privately owned facilities, i.e. commercially funded, a bond equity ratio of 1:3 was used and bond- and stockholders' rates of return of 8 and 12 per cent respectively. For waste solidification facilities, the annual capacities were 1:3 and 2:3 and thereafter unity. The USDOE report contained a median uncertainty of ± 30 per cent for capital costs and an estimated range of +25 to -50 per cent for operating costs.

Where it is possible to defer construction and operation of any stage in a proposed waste management scheme, this will be favoured by the provision of a suitable fund accruing interest at available market rates.

A3. TECHNICAL GROUND RULES

A3.1 Scale of Operation

Estimates are based on solidifying the HLW from a 2000 t HM y^{-1} reprocessing plant, a storage facility of 2231 t HM y^{-1} for glass and a repository of 5500 t HM y^{-1} for both.

The scale of operation is based on a chemical reprocessing plant with an annual throughput of 2000 tonnes of heavy metal, i.e. uranium and plutonium, contained in the spent reactor fuel. This assumption allows the US cost data to be used directly rather than scaled. The assumed throughput is also consistent with the total reprocessing capacity the French propose to install at Cap La Hague (1600 t U y^{-1}) by the mid-1980s.

A3.2 Waste Canisters and Loading

The waste canisters are assumed to be 3 m by 0.3 m diameter, the size used by the US and Denmark. Other dimensions have been proposed, including 1.5 m by 0.4 m diameter (Sweden); and 1 m by 0.45 m (UK); 1 m by 0.5 m diameter canisters are used to receive vitrified gas-graphite wastes in France.

Waste loadings have been reported [Araki, 1981; Banba, 1981; Bonniaud et al., 1980; Burton and Griffin, 1981; ELSAM and ELKRAFT, 1981; KBS, 1978; USDOE, 1979, 1980] on variable bases ranging from wt% total calcine, wt% fission products and wt% fission products + actinides. To facilitate comparison between waste loadings, volumes per tonne of spent fuel have been derived as follows:

Country	Waste Loading in Glass		Waste Volume per t Spent Fuel m^3 t^{-1} HM
	wt% Oxides	Definition of Oxides	
Denmark	9	probably fission products	0.10
France	~13 (est)	fission products	0.10
Japan	15-30	total calcine	
Sweden	9	fission products	0.15
UK	15	fission products + actinides	0.10 (est)
USA	30	total calcine	0.06

Waste loadings assumed in this study have accordingly been 10 wt% oxides (European strategy) and 30 wt% oxides (USDOE strategy).

A3.3 Rail Transportation

Shielded containers for high-level radioactive wastes, e.g. spent fuel or solidified HLW, are generally designed for a particular heat load and, within wide margins for waste configuration, the same amount of waste can be transported.

The current design of a rail transport flask for solidified HLW has a maximum thermal rating of 50 kW and is to be operated with an approximately 40 per cent safety margin.

For this study, the levellised transport costs have been taken as those given in the USDOE report on technology for commercial radioactive waste management [USDOE, 1979].

A3.4 Storage Facility for Waste

A facility for long-term (30 y), above-ground storage of solidified HLW would have capital costs for the provision of security and surveillance, rail siding, loading and unloading systems, and so on.

The operation costs are predominantly determined by construction costs for the concrete pads and the concrete shields that provide the radiation shielding and the convective cooling for the solidified HLW. These costs are related linearly to the stored quantity of waste and relatively independent of the waste configuration (e.g. size of cylinder). The facility was assumed to have an economic life of 100 y. At each 30 year change it was assumed that one third of the concrete pads and shields were not re-usable. The storage facility was envisaged as being initially filled over a period of 9.1 y from an expanding nuclear energy program at an average rate of 2231 t HM y^{-1} (20 300 t U). The facility could receive, store and discharge three such batches (60 900 t U) during its economic life.

A3.5 Waste Repository

With respect to the European (aged) and US (fresh) waste forms, there are two major differences in the cost components for the waste repository. Because of the lower heat generating capacity of the aged glass waste, the repository loading for glass can be at the maximum determined by mechanical/operational considerations. For fresh waste the loading is restricted by its heat generation. It should be noted, however, that this is a consequence of the assumptions utilised in the USDOE study in which the repository was designed using conventional

underground construction. By employing a repository consisting of a system of deep drill-holes, in which the waste is distributed in a three-dimensional configuration, the thermal limitations may be relaxed. This option has been studied by Denmark.

The age of the waste at time of disposal is 40 years for European glass (0.4 kW/canister) and 10 years for US glass (1.7 kW/canister). The corresponding glass surface temperatures are ~85 and ~320°C above ambient temperature respectively. The repository is 600 m below ground surface, in granite, and has a floor area of 810 ha constructed over a five-year period with a potential 70 per cent utilisation for high-level waste disposal. The capacity of the repository for the European aged glass with 10 per cent waste loading is limited by a combination of operational and thermal considerations to 137 000 t HM whereas for the US fresh glass with 30 per cent waste loading the limitation was 89 700 t HM because of temperature restrictions on the supporting granite pillars within the repository. It was assumed that the waste is to be placed in holes drilled in the floors of the emplacement rooms and the holes lined with carbon steel pipe back-packed with crushed granite between this pipe and the host granite and, after emplacement, plugged at the top with concrete.

A4. APPLICATION OF GROUND RULES TO COST ESTIMATES

Cost estimates have been used on the economic ground rules and cost data extracted from the report 'Final Environmental Impact Statement on Management of Commercially Generated Radioactive Waste' [USDOE, 1980] and its supporting document [USDOE, 1979]. These data, with minor modifications, constitute the costing summary for the 'USDOE strategy' detailed in Table 10. Generation of comparative data for the 'European strategy' required adaptation for the US material to the effect of a reduced waste and thermal loading and the requirement for protracted storage before geologic burial. The waste vitrification operations used below illustrate the effect of the new ground rules.

A4.1 Comparison of Capital Costs of Vitrification

The capital costs for vitrification plants of 2000 t HM y^{-1} capacity based on European and US assumptions are summarised as follows (1976 $US x 10^6):

Component	European	US
Major equipment	7.7	4.5
Buildings	14.6	9.1
Bulk materials	13.1	7.7
Site improvements	0.3	0.2
Direct construction costs	35.7	21.5
Indirect construction costs	15.2	9.2
Total field cost	50.9	30.7
Architect-engineer services	9.2	5.5
Subtotal	60.1	36.2
Owner's cost	18.0	10.8
Total facility cost	$78.1M	$47.0M

A4.2 Comparison of Operating Costs in Vitrification

Operating costs for the European and US assumptions are as follows (1976 $US x 10^6):

Component	European	US
Direct labour	0.56	0.84
Maintenance labour	0.68	
Process materials	12.18	4.00
Utilities	0.27	0.09
Maintenance materials	0.43	0.27
Overheads	0.87	0.66
Miscellaneous	0.33	0.24
Annual operating cost (rounded)	15.3	6.1

A4.3 Comparison of Unit Costs of Vitrification ($US/kg heavy metal)

Component	European	US
Capital charge	10.1	6.1
Operating charge	7.0	2.8
Unit cost (1976 US$)	17.1	8.9
Unit cost (1978 US$)	20.0	10.4

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