AUSTRALIAN ATOMIC ENERGY COMMISSION
RESEARCH ESTABLISHMENT
LUCAS HEIGHTS

DISPOSAL OPTIONS FOR HIGH LEVEL NUCLEAR WASTE

by

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Presented at the Australian Drilling Association's Symposium on 'Drilling and the Environment', Newcastle, New South Wales, 18-20 January, 1982.

January 1982

SUMMARY

The options for the management/disposal of high level nuclear waste are discussed; it is concluded that irretrievable disposal of solidified waste is a likely end step in all disposal schemes. Disposal will probably be either in mined repositories or in deep drill holes. Two solidified waste forms - borosilicate glass and SYNROC - are considered in some detail. It is concluded that SYNROC would provide a higher level of assurance of radionuclide retention in both disposal concepts. Deep drill-hole disposal using SYNROC is an attractive concept which should be given increased attention.

1 INTRODUCTION

The world's large uranium reserves represent a concentrated energy source which can benefit mankind in many ways, not the least of which is to ease the strain on finite reserves of fossil fuels. Nuclear power for large-scale electricity production is already an important component of the energy mix in the USA, UK, France, Japan, West Germany and Canada, and many other countries have one or more nuclear power plants in operation, under construction or on order. However, the progress of this industry is being slowed by vocal opposition which concentrates on a variety of issues, including the problems of high level nuclear waste disposal, for which it is often alleged that there is no known solution.

It is true that high level waste disposal poses a challenge, true that ultimate disposal has not yet been practised or demonstrated, but not true that technically safe solutions are unavailable. Industry and governments are simply proceeding with great caution to ensure that the option or options eventually adopted are optimised in terms of both engineered safety and public acceptability.

After describing the nature of the problem, this paper broadly reviews a range of waste management / disposal options and waste forms. A disposal option for which particular expertise from the drilling industry would be necessary is included.

2 THE GENERATION OF HIGH LEVEL NUCLEAR WASTE

A typical nuclear power reactor generating 1000 MW of electrical energy on a commercial basis produces ~ 30 tonnes of spent fuel per annum. Approximately 3.3 per cent of the uranium atoms originally present will have fissioned to produce 1 t of a range of highly radioactive fission products. In addition, some of the uranium will have been converted by neutron capture and subsequent decay processes into various isotopes of the transuranic actinides neptunium (Np), plutonium (Pu), americium (Am) and curium (Cm). Fortunately, the plutonium, a potentially valuable energy source, and the residual uranium can be extracted from spent fuel by 'reprocessing', using a dissolution/solvent extraction process. This leaves a solution containing all of the fission products, 0.5 - 1 per cent of the uranium and plutonium, and relatively small amounts of Np, Am and Cm. This is the high level waste to which the title of this paper refers. The author considers that disposal of unreprocessed spent fuel as 'waste' is so unlikely to eventuate on any significant scale that it warrants no further attention here.

3. NATURE OF THE WASTE

3.1 Chemical

A typical high level liquid waste consists of a nitrate solution of more than 51 different elements, of which the most important are: various rare earths, zirconium, molybdenum, ruthenium, caesium, strontium, barium, rubidium, palladium, platinum, rhodium, tellurium, and technetium (all fission products); uranium, plutonium, neptunium, americium and curium (actinides); and iron, nickel, sodium and some other processing contaminants, including phosphate ions from the solvent extraction agent.

3.2 Radiological

Most of the fission products from uranium and plutonium are beta-gamma emitters, with half-lives ranging from microseconds through years to, in a very few cases, greater than 10^6 years. In the first few years the short half-life isotopes dominate the very high total activity, but after about 10 years the activity is due predominantly to the two caesium and strontium isotopes ^{137}Cs and ^{90}Sr. These in turn decay exponentially with 30-year half-lives. Since in exponential decay the activity decreases by factors of 10^3 and 10^6 in 10 and 20 half-lives respectively, the ^{137}Cs and ^{90}Sr activity will be very small after about 600 years. During the first 1000 years, the total activity of the waste, initially very high, will have fallen by a factor of at least 10^5 and will be only about four times that of the uranium ore from which the original mass of uranium fuel was produced. The few longer-lived fission products now contribute much less to the total activity than do the actinides.

The alpha-emitting actinides present in high level waste have half-lives ranging from 163 days to 2 x 10^6 years (excluding some even longer-lived uranium isotopes). After about 600 years, when they have begun to assume importance over the fission products, they decay only slowly - for example, by factors of 4 from 1000 to 10 000 years, 10 from 10^4 to 10^5 years and a further 10 from 10^5 to 10^7 years. However, at 1000 years, their total activity is only about three times that of the 'parent' mass of uranium ore.

3.3 Thermal

A typical high level waste liquid from spent fuel reprocessed one year after discharge from a reactor initially produces approximately 10 kW of heat per cubic metre and obviously must be continuously cooled to prevent boiling. The heat load falls by a factor of at least 10^3 in the first 1000 years.

4 MANAGEMENT OPTIONS

4.1 The Need for Eventual Disposal

High level waste is being managed quite safely in various countries mainly by water- and air-cooled storage of various waste forms in engineered storage facilities. Although the waste is very hazardous to moderately so for about 1000 years and, if accessible to man, would deserve some respect even after 10^5 - 10^6 years, its continued management/surveillance would not be technically very difficult. However, for socio-political rather than technical reasons, it is obvious that we should not commit more than about one further generation to this type of storage. There is therefore universal agreement on the need for eventual disposal well away from man's environment. However, there is a need for agreement and decision on disposal sites and on the time-scale and methodology of disposal.

4.2 Reprocessing Options

In most schemes, spent fuel discharged from reactors is stored in either local or centralised storage ponds for at least one year to allow some reduction in fission product activity and decay heat. This period could be extended to 20 years or more, a situation which currently applies in the USA but not in Europe, where reprocessing is being practised on a commercial scale. After reprocessing, the high level liquid waste is being stored, this time in water-cooled steel tanks encased underground in concrete vaults. However, such storage will not be practised on a large scale except for interim holding purposes. The next step, solidification, is designed to produce a stable solid, exemplified by borosilicate glass or SYNROC, of which the waste comprises 10 - 30 weight per cent; the solidified waste is fully sealed within a metal (usually stainless steel) canister. Again, canisters of solidified waste may be stored, in either ponds or air-cooled vaults before eventual disposal. This storage period could extend to 60 years, depending on the particular national management/disposal policy. Extended air-cooled vault storage of borosilicate waste glass canisters produced on an industrial scale is already in operation in France. At the end of the storage period, the canisters may be encased in outer canisters or 'overpacks'. For example, in Sweden (KBS, 1978) it is proposed to enclose each waste glass/steel canister in a titanium metal canister with an intervening layer of lead. Finally, 5 to 60 years after discharge of the spent reactor fuel, a 'waste package' would be ready for disposal. The dimensions of the primary canisters would usually be in the ranges 0.3 - 0.5 m diameter by 1 - 3 m long. Overpack canisters could add 0.2 m to the diameter and 0.3 m to the length.

5 SOLID WASTE FORMS

5.1 Range of Waste Forms

The simplest - and least satisfactory - solid waste form, obtained by concentrating high level waste solution and subjecting it to fluidised bed calcination at 600 - 800°C, is known as 'calcine'. It is chemically quite active and fairly soluble in water. The main objective of all waste form development is to produce a much more stable solid which, once fabricated, resists release of radionuclides into the environment. Release mechanisms which must be considered are: fragmentation during transport accidents; volatility during accidental overheating before emplacement; and aqueous leaching should water breach a disposal repository, corrode the canister and attack the waste form. In the latter case, long-term stability must be assured under a range of conditions which include temperatures above 100°C, temperature gradients, the presence of intense beta-gamma radiation fields and radiation damage arising from the radioactive decay of actinide atoms by alpha particle emission.

Borosilicate glasses are the leading 'first generation' waste forms being considered, developed or, in the notable case of France, produced on an industrial scale. The French vitrification process has also been licensed for operation in the UK and FRG. 'Alternative waste form' programs in various countries aim to develop forms with a still higher degree of assurance of radionuclide retention, possibly for use as 'second generation' waste forms. Among these are high silica glass, crystalline ceramics, and multibarrier metal matrix forms involving uncoated or coated particles of glass or ceramic dispersed in a metal. In this paper, only borosilicate glasses and the prominent alternative waste form SYNROC - a crystalline ceramic - will be described.

5.2 Borosilicate Glasses

Borosilicate glasses have been designed and developed to dissolve most of the elements of high level waste at an overall loading of up to 30 weight per cent. The glass, together with glass-forming components or frit plus the high level waste, may be melted either in situ in a stainless steel canister or in a glass furnace and then poured into the canister. Melting temperatures usually fall within the range 950 - 1150°C, depending on the particular glass composition and, if the glass is to be poured, the maximum tolerable viscosity. Stainless steel primary or overpack canisters could not be expected to last very many years in a repository which could at times be moist. However, in a Swedish study (KBS, 1978), the proposed waste glass/stainless steel/lead/titanium waste package was expected to survive for at least 500 years before moisture attack of the glass could conceivably commence. In the same study, a conceptual copper canister was expected to last for at least 5000 years.

It is claimed that borosilicate glass is very unlikely to devitrify solely as a result of temperature effects over geological times; glass also seems to be adequately resistant to gamma radiation and actinide decay damage which have little or no effect on aqueous leach rates. At temperatures up to 100°C, the leach rates in flowing water of borosilicate glasses, measured in terms of radionuclide losses, are quite low. However these rates rise with high activation energy and, by 200°C, the term 'low' no longer applies. Moreover, hydrothermal leaching of these glasses usually results in at least partial devitrification, leading to a further increase in leach rate. Because it is prudent to allow for the possibility of water moving through a repository, it follows that borosilicate glass will be an acceptable form for ultimate disposal of high level waste only if its temperature can be limited to about 150°C or if it can be guaranteed that moisture cannot contact the waste form when its temperature is above 150°C.

5.3 SYNROC

SYNROC is a crystalline ceramic waste form (Ringwood, 1978) consisting of three titanate mineral phases which are known to be highly stable in the geological environment and which, in synthetic form, can collectively take almost all the elements in high level waste into solid solution in their structures at an overall waste loading of 10 - 20 weight per cent. Dense SYNROC containing actual or simulated non-radioactive high level waste may be made by either cold-pressing and sintering ceramic powders at ~ 1350°C or preferably by hot pressing them at 1150 - 1250°C. In the latter case, it is proposed to hot press directly into a stainless steel disposal canister, a method which is being developed at Lucas Heights (Ramm and Ringwood, 1980). Final canister dimensions are undetermined but could conceivably be the same as those for borosilicate glass. Overpack canisters could be added if desired. A major aim of the developmental work at Lucas Heights is to demonstrate that, on the basis of mass of waste per unit volume, SYNROC fabrication costs would be of the same order as those for borosilicate waste glass fabrication.

The outstanding advantages of SYNROC over glass are its lower leach rate at all temperatures and its much higher temperature capability. In Lucas Heights tests on SYNROC containing non-radioactive simulated waste, it has been shown that the leach rate of caesium from SYNROC at 100°C is at least 500 times lower than that for waste glasses. Furthermore, since the activation energy for SYNROC leaching is lower, this advantage increases with increasing temperature - and, unlike glass, SYNROC suffers no structural deterioration on hydrothermal leaching. At 300°C, SYNROC leaches at about the same rate as glass at 45°C. The effect of gamma radiation on leaching is expected to be small or zero. The effect of actinide decay damage on leach rate has been inferred to be low by indirect evidence from a study of partially and fully metamict minerals of the same type as those in SYNROC (Ringwood et al, 1980) and at Lucas Heights accelerated damage studies by neutron irradiation are giving the same result.

SYNROC is therefore proving to be a potentially excellent waste form which, because of its close relationship to geologically stable natural minerals and its established high stability and leach resistance in laboratory tests, should improve the public acceptability of irretrievable geological disposal of high level nuclear waste. Moreover, SYNROC maintains this leach resistance to temperatures of at least 300°C and probably higher. Provided that radiation damage does not prove to have adverse effects on its stability, SYNROC could incorporate waste after shorter cooling periods, and at a higher level of waste addition, than for borosilicate glass, and could be disposed of in locations with higher ambient temperatures.

6 DISPOSAL OPTIONS

6.1 Range of Options

Only the two most 'popular' and likely disposal options, viz continental geological disposal in mined repositories or deep drill holes, are described in this paper. Other proposals which, in the author's opinion, are unlikely to be accepted on any significant scale, are the 'rock melt' concept, island-based geological disposal, sea-bed or sub-seabed disposal, ice-sheet disposal, deep well injection, transmutation and space disposal.

6.2 Geological Disposal Philosophy

Inherent in the idea of high level waste disposal is the need to place a multiplicity of man-made and natural barriers between the solidified waste and the biosphere. In geological disposal, these barriers include: the waste form, which must be chemically very stable but may also include sub-barriers such as particle coatings; the waste canister or canisters; impermeable or water-absorbing back-fill materials as part of the engineered repository system; and the rock strata between the repository and the biosphere. Contributing to the effectiveness of this last barrier must be: the remoteness, geological stability and expected dryness of the chosen site; the great distance for radionuclide migration; the low permeability of the host rock and surrounding strata; and the slowing of radionuclide migration by chemical absorption.

Although each country's rules for licensing and regulating waste repositories will differ in detail, the basic assurance required is that, taking into account all conceivable eventualities and making conservative (i.e. generally pessimistic) assumptions, the risk to present and future generations will be acceptably small. Without entering into detail which is beyond the scope of this paper, this certainly means that the maximum conceivable radiation dose to the 'most exposed person or persons' will be a small fraction of the radiation dose they would normally receive from natural sources. An analysis of this kind resulted from a Swedish study of an assumed waste repository 500 m deep in granite (KBS, 1978). The upper limit dose to the most exposed person over a thirty-year period was calculated as only 13 millirem per annum, occurring only after 200 000 years. This is to be compared with the annual doses in Sweden from natural background radioactivity (130 millirem) and radium in drinking water (30 millirem), and with the ICRP (International Commission on Radiological Protection) recommendation of 500 millirem per annum per person as the maximum allowable radiation dose above that received from natural sources. This limit is designed to protect individuals from measurable health effects from additional radiation.

The complex decisions involved in repository design must of course be taken as an integral part of a technically feasible, demonstrably safe, and socially acceptable management/disposal scheme. In particular, waste form and disposal methodology must be carefully matched to provide the necessary safety assurances. In this context, only one more very basic issue, viz. retrievable versus irretrievable disposal, need be mentioned. It is unlikely that retrievability of waste containers in the initial period of operation of a mined geological waste repository will be generally specified. In what is perhaps an ultra-conservative example, a recent US Nuclear Regulatory Commission draft regulation (NRC, 1981) calls for retrievability for up to 110 years from the commencement of repository operation. However, I believe that once-and-for-all, irretrievable geological disposal is the desirable and achievable goal. Although this philosophy is not incompatible with the use of relatively shallow, mined repositories, where retrievability will eventually be lost anyway, it is perhaps even more compatible with the alternative, deep drill-hole concept.

6.3 Mined Repositories

According to current US thinking (DOE, 1980), a typical mined repository would be situated about 600 m deep in granite, basalt, shale or salt. There would be up to four vertical shafts from the surface (the canistered-waste shaft, the men and materials shaft, the ventilation exhaust shaft and the mine production shaft). These would lead to a series of underground rooms, of total area around 800 ha, in which the waste packages would be placed in suitably separated drilled holes or trenches. The underground layout would be a conventional room and pillar arrangement. Of the 800 ha, the waste emplacement area would amount to no more than about 700 ha. However, thermal and mechanical loading considerations restrict the hole spacing to no less than 1.8 m centre to centre. For relatively young waste with a high thermal loading, the spacing may have to be considerably greater than this.

In the typical repository, all radioactive waste canisters would be handled remotely, according to well-established procedures. The waste packages would be received at sub-surface transfer stations where shielded transporters would remove them remotely to an emplacement room. The transporter would lower its waste package into the required hole and then return for another package. Limited access to these facilities would be allowed to maintenance personnel.

During an initial period of mandatory retrievability, the waste would be emplaced in holes lined with steel sleeves and sealed with removable concrete plugs. When retrievable disposal is no longer required, the holes would not need to be lined, and filled holes would be backfilled with excavated rock or a specially selected moisture-absorbent material such as bentonite. Filled rooms would be backfilled with a similar material.

After all rooms had been backfilled, the underground facilities would be progressively decommissioned, underground corridors backfilled and shafts filled to the surface and sealed. Repository decommissioning would be complete when the surface facilities had been decontaminated, and perhaps removed, and the repository location suitably marked. An 800 ha repository could accept the high level waste from approximately two hundred 1000 MW(e) reactors for between 5 and 15 years.

The temperatures in such repositories will rise because of fission product decay heat, the extent of the rise depending on the properties of the rock and the thermal loading of the repository. Thermal loading depends on age of waste, waste content per unit volume of waste form and packing density of canisters in the repository. Maximum temperatures will not be reached for some tens of years after emplacement and the original level will not be regained for at least 1000 years. Temperatures will increased in the order: rock-wall $<$ repository environment $<$ canister surface $<$ waste form surface $<$ waste form centre. If care is not taken to keep the original temperature level low (by not too deep disposal) and the heat loading from the waste low (by low waste loadings), waste-form temperatures, particularly at the centre-line, could easily exceed 100°C for a considerable time after disposal. If borosilicate glass is to be used under such conditions, there must be great emphasis on the design of long-life canisters to give confidence that water cannot flow past the waste form while its temperature is above about 150°C.

6.4 Deep Drill-hole Concept

6.4.1 Advantages

This concept involves the irretrievable disposal of waste canisters or packages end-to-end in 4000 - 10 000 m deep drill holes. The clearance between the package and the hole liner and between each package would be filled with a water-absorbent material such as bentonite or magnesia. The hole would be filled only to a fraction of its depth and then be backfilled with suitably dense and absorbent materials. According to Ringwood (1980), who strongly supports this proposal (on the basis of filling the lower 2500 m of 4000 m deep holes with waste canisters), the concept has the following advantages over mined repositories:

(i) Waste is located further from the biosphere in hard rock formations of very low permeability which should be easy to find at depths of 1500 m and below.

(ii) There should be less disturbance to the surrounding rock structure and hydrology.

(iii) Acceptable disposal sites should be easier to find; a waste disposal strategy based on drill holes could be implemented much more quickly; the initial capital investment would be smaller because drilling could be designed to keep pace with the rate of waste production; and public acceptability should be easier to obtain because the concept is simple and its advantages and comparative safety are readily perceivable by non-experts.

(iv) The total amount of waste in a single drill hole is much smaller than that in a large mined repository and, within 1000 years, the total radioactivity would become smaller than natural background levels arising from the normal occurrence of small amounts of uranium and thorium in the rock in the vicinity of the drill hole. In the case examined, viz. a 2500 m column of 0.5 m diameter SYNROC canisters in a 4000 m deep drill-hole, the total radioactivity falls in 1000 years to that from a uranium ore body containing 136 000 t of uranium (e.g. deposits in the Northern Territory). But, whereas the uranium in typical known ore bodies occurs within a few hundred metres of the surface in the zone of weathering and strong groundwater circulation and is usually highly leachable and mobile, the waste in a drill hole would be buried at least 1500 m deep in rock of very low permeability.

(v) This reference drill hole cited above would accommodate one year's solidified waste from eighty-three 1000 MW(e) reactors. Therefore in a country such as the USA with a large nuclear power program, the hole could be sealed within a few years of the commencement of disposal.

Other analyses of this concept include those of O'Brien et al. (1979), Crandall (1979) and DOE (1980). The first summarised feasibilities, costs, probable technological advances to the year 2000, geological uncertainties and research and development needs; the second, using an assumed hole depth of 10 000 m, recommended continuing technical development but ranked this concept fourth in preference amongst five considered, behind mined repositories, sub-seabed and space disposal; the third considered five concepts, amongst which the very deep hole concept ranked generally high on most of the assessment criteria used, but ranked low on 'status of development'.

6.4.2 Current deep drilling technology

There are two candidate deep drilling methods: big hole drilling, which was developed largely to permit underground testing of nuclear weapons; and oil field rotary drilling. Close to attainable capabilities (O'Brien et al., 1979) in hard rock include 3 m diameter holes to 3000 m by the latter method and 1.2 m diameter to 4000 m by the former. Oil field drilling has a current depth capability of 9000 m (DOE, 1980) but the diameter in the lower section of the hole would be no more than 0.3 m. A minimum diameter of 0.5 m would be necessary for waste canister disposal, which eliminates this depth of drilling at present. However, significant advances in capability and reductions in current costs are no doubt possible for both drilling methods.

6.4.3 Issues in drill-hole disposal

The following issues associated with deep drill-hole disposal require clarification:

(i) The depth of hole required to achieve the advantages claimed for this concept and whether the holes would need to be completely cased.

(ii) Probable and possible advances in deep drilling technology.

(iii) Current and projected drilling costs for various drilling methods, hole types, hole depths and hole diameters.

(iv) Prediction of the thermomechanical behaviour of rocks surrounding the deep drill hole.

(v) Stability of deep holes against inward collapse and whether the hole would need to be filled with drilling mud to prevent this.

(vi) Waste package emplacement technology.

(vii) Methods of sealing the hole.

(viii) The availability of sensors to log conditions at the bottom of a drill hole at elevated temperatures and pressures.

(ix) The availability of a waste form capable of radionuclide immobilisation at the higher ambient temperatures. (Ambient temperatures rise with depth by about 20 - 30°C per 1000 m, to give 100 - 130°C at 4000 m and 200 - 300°C at 10 000 m).

6.4.4 SYNROC in deep drill holes

Because complete dryness cannot be assumed even in a deep drill hole, borosilicate glass will not have adequate leach resistance at the higher temperatures involved to serve as the primary barrier to radionuclide dispersal. On the other hand, SYNROC has just the right higher temperature capability. Moreover at, say, 4000 m depth, where the ambient temperature is 100 - 130°C, there is scope for using hotter (shorter-cooled) waste in SYNROC than would be advisable for the disposal of glass even in shallower repositories. Thus the availability of SYNROC as a 'matched' waste form for deep drill-hole disposal should make this promising concept even more attractive. However, SYNROC would be a superior waste form for any geological disposal concept.

7 CONCLUSION

There are many management/disposal systems for the high level nuclear waste arising from electricity-generating nuclear power stations, but irretrievable geological disposal is likely to be the eventual end step in all schemes. The author believes that this will be in either mined repositories or deep drill holes or both. Of the two candidate waste forms considered here in some detail, viz. borosilicate waste glass and SYNROC, SYNROC should provide a much higher level of assurance of radionuclide retention in both disposal concepts and would be the ideal form for deep drill-hole disposal - an attractive concept which should be given increased attention as a disposal option for high level nuclear waste.

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