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OPTIONS FOR THE DISPOSAL OF HIGH-LEVEL RADIOACTIVE WASTE

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1. INTRODUCTION

The generation of large quantities of radioactivity is an inescapable consequence of the exploitation of nuclear energy. While some of these products such as plutonium, a potential reactor fuel, are valuable, most of them and especially the fission products are waste. In recent years the long-term technical management of these wastes has become increasingly accepted as one of the most important issues facing the nuclear industry. This has, for instance, been recognized in the sixth report of the United Kingdom Royal Commission on Environmental Pollution[1].

Virtually the whole of the wastes giving rise to these problems accrue at the irradiated-fuel reprocessing stage of the nuclear fuel cycle and arise as high specific activity liquid.

Their composition is predominantly fission product together with transuranics, including a relatively small amount of plutonium. Caesium-137 and strontium-90 account for most of the fission product activity and most of the heat output. They will have almost entirely decayed in the first 10^3 years but some of the transuranics are longer lived and require about 10^6 years to decay by the same proportion. Current UK practice is to store the highly active liquids. This is a safe and satisfactory interim measure on a timescale of decades leading to solidification by processes such as the vitrification process.

In the longer term it is necessary not only to have a solidified product but also to achieve a safe means of destruction or disposal of these wastes. It is not considered realistic to rely on engineered storage for their management on a time-scale of hundreds to thousands of years.

Destruction of transuranics by neutron irradiation is considered by some as a possible solution to the problem of their disposal but is not considered in this paper.

The basic objective in disposal is the achievement of such a high degree of isolation that the activity either never returns to the biosphere and man, or comes back at such low concentration due to delay and/or dilution as to present no hazard. Extra terrestrial disposal and the use of polar ice-caps have been discounted on grounds of reliability and safety, and this paper is confined to the consequences of use of two other more promising recipient media - the continental or oceanic crust and the oceans. Burial in geological formations on land (section 2) or under the ocean bed (section 3) is likely to provide the highest degree of isolation possible. In disposal on to the ocean floor (section 4) much more reliance would have to be put on a man-made physical barrier surrounding the waste form. Unlike the other two methods dilution in the ocean would be an important long-term factor in reducing activity to acceptably low concentrations.

2. THE LAND DISPOSAL OPTION

Substantial effort is being directed towards disposal of high-level radioactive wastes in a repository on land. Once emplaced in a suitable host-rock only mobile fluids, principally circulating groundwater, could transport significant quantities back to the surface. In most countries the wastes will be converted to glassy or ceramic solids having low leach-rates and encapsulated in corrosion-resistant material to minimize the risk of their return to the surface in a concentrated form. Breakdown of isolation would involve unplanned access of circulating groundwater, failure of the encapsulation and solution of the wastes from the solid. If such three-fold failure did occur, a further mechanism would delay the return of nuclides to the biosphere, because their transport in solution is inhibited by sorption on many rock-forming minerals. Confirmation of the effectiveness of sorption processes comes not only from controlled experiments, but also from the "Oklo Phenomenon" where many of the nuclides generated in "natural", water-moderated nuclear reactor zones enclosed within Precambrian sedimentary rocks at Oklo, in Gabon, migrated a negligible distance. Oklo is significant not only in demonstrating the effectiveness of sorption, but also as an illustration of the principle that, apparently despite circulating groundwater, escape of radioactive products from the reactor zones with their particular mineral assemblages was quite limited[2] and the wastes have remained isolated in a host-rock over an immense time-span, approximately 1800.10^6 years.

2.1 Potential host-rocks

The characteristics of suitable host-rocks have been reviewed extensively[3]. The fundamental requirement is that the host-rock lithology, taken in conjunction with its spatial distribution, should permit the containment of the wastes. Theoretically, any rock meeting that requirement would be suitable, but in practice the essential need for host-rocks to be free of circulating groundwater emphasizes the suitability of three main groups.

Evaporites, particularly rock-salt, have been of prime interest. Undisturbed, near-horizontal beds of salt, as well as "diapiric" structures, have been explored with the present preference for the latter because the existence of thick salt deposits indicates almost a complete absence of circulating groundwater. The thermal conductivity of salt is high so that problems of heat dissipation are reduced and as rock-salt flows plastically, stresses generated during waste emplacement will not cause the formation of permanent fractures as paths for circulating groundwater. Although rock-salt has been the preferred disposal medium, its use has been criticised because of the risk of flooding during operation of a repository. Global resources of rock-salt are so large that licensing of limited volumes for waste disposal would not significantly restrict its overall availability. Not all states having nuclear power programmes, however, have extensive salt deposits even if they wish to use them for that purpose.

Argillaceous rocks, particularly plastic clays, are also under study. They have the advantages of negligible permeability and a high sorption capacity, as well as being self-healing to a greater or lesser extent, depending upon their induration. However, their thermal conductivity is low and the effects caused by decay heat, particularly upon permeability and sorption properties, will require careful study. Although argillaceous rocks are widespread, many of the thick plastic clays occur in populous areas not suited to disposal sites.

The third group of potential host-rocks - crystalline igneous and metamorphic rocks - are attracting increasing interest. When unweathered they contain no pore-water and, although near the surface they may be traversed by water-bearing fractures, circulating groundwater is not generally encountered in them at significant depths. Experimental work has demonstrated that blocks of waste simulant and dolerite can be fused and if proved safe this simple concept would be attractive in its simplicity in its use of decay heat to bring about the weld. Crystalline rocks occupy major areas of the continents and appropriate disposal sites essentially free from the influence of deep-seated mineralizing fluids could be located without risk of sterilizing essential commodities.

2.2 Area and site selection criteria

Within any host-rock, however, local problems may exist in relation to absolute containment of wastes and detailed investigations will be required, taking account of strict criteria. Those required to select areas containing suitable formations must take account of their physiography and regional structural settings, in addition to the characteristics of the potential host-rocks. Eight sets of such criteria have been defined for use in the United Kingdom[4]. They include gross lithology and spatial distribution of the strata; physical and chemical characteristics of the strata in relation to wastes and containers; hydrogeological and hydrological conditions; seismicity; relations to other man-made structures and possible mineral deposits; effects of climatic change; leakage routes to the surface; engineering practices. Suitable areas incorporate a formation meeting a balance of the criteria appropriate to the particular host-rock and geological environment. Stable geological conditions are essential, but the time for which stability is required, $100\text{-}250.10^3$ years, is small in comparison with total geological time, upwards of 4000.10^6 years. Even in that limited time-scale, however, the present geological, climatic, hydrological and erosional regimes are unlikely to be maintained. Thus at the stage of area selection it is important to identify qualitatively the probable range of regimes to which the area may be subject. At the succeeding site selection stage the criteria must be such that the repository design should minimize the adverse effects of such changes in regime. Predictive models simulating the range of such changes and the mechanism of nuclide migration under the particular hydrogeological conditions may be of value[5].

2.3 Geological exploration and research requirements

Conventional geological exploration necessitates drilling to obtain rock cores for geotechnical examination. However, drilling creates undesirable access paths for both water and drilling fluids. Maximum use must therefore be made of surface exploration methods, including geophysical techniques, to predict the sub-surface conditions around the site. Some drilling will be essential to confirm the predictions and obtain cores for laboratory measurements. The hydrogeological conditions must also be deduced so far as possible from their surface effects, although again some observation boreholes will be essential. Although within the United Kingdom seismicity is at a low-level, some monitoring will still be necessary. Thermal disturbance to the host-rock will require investigation, whilst rock specimens must be subject to both heating and radiolysis to determine their long-term effect on geotechnical properties.

3. DISPOSAL BELOW THE OCEAN FLOOR

Disposal in geological strata below the ocean floor[6] is in many ways similar to disposal on land but has the added advantage of remoteness from human habitation and has the additional barrier of the ocean (section 4). The disposal site may be in the unconsolidated sediments extending to tens or hundreds of metres below the ocean floor, in the consolidated or lithified sediments between a few hundred and a few thousand metres, or in the igneous rocks of the oceanic crust. In addition to providing an extra barrier against container failure, burial will ensure that accidental or nefarious recovery is avoided.

3.1 The geological barrier

The effectiveness of these disposal regions as barriers depends firstly on their continuity in totally surrounding the disposed package, and secondly on the ability of the medium to adsorb migrating waste by ionic exchange. Data from the Deep Sea Drilling Project[7] have shown that the upper basaltic crust is extremely heterogeneous with breccia and rubble zones, fissures and faults which may extend several kilometres into the crust, and it is therefore not a suitable region for disposal. The lower part of the sediment layer is more or less lithified depending on the sediment facies, diagenetic processes and the overburden history. Faulting under stress produces cracks which may not be sealed because of the finite strength of the sediment. The distribution of such cracks and their effect on the gross permeability is unknown and difficult to determine so that this region is not very favourable for disposal. The unconsolidated upper sediments, however, show more promise since their plastic properties may enable stress-induced cracks to seal themselves. The high ion exchange capacity in the oxidizing environment of the deep ocean red clays can be used to fix migrating radionuclides and their low permeability can reduce the interstitial water flow near the container.

3.2 Area and site selection

To ensure minimum risk of disturbance to such a sediment barrier, the site must be in a seismically stable area and free from potential erosion, slumping or other modification of the medium over a period comparable to the life of the longest lived actinides. Furthermore, the site must avoid those areas of the ocean floor where resources such as phosphorites, manganese nodules or hydrocarbons might conceivably be exploited in the future. Prediction of the site stability can only be achieved from an understanding of all the geological processes operating there and from a careful study of its geological history during the last few million years.

Suggestions have been made that disposal should be within the subduction regions where the oceanic lithosphere plunges back into the earth's lower mantle. However, uncertainty about the details of the mechanism, the high earthquake activity and the slowness of the process must rule out this possibility. The most favourable sites appear to be in the centre of the oceanic part of the lithospheric plates, away from the active plate boundaries and from the mobile sediments of the continental margins.

3.3 The heat problem

If young waste, containing a high proportion of fission products and generating appreciable heat, is placed within the ocean bed, the container and the surrounding sediments may be considerably modified and perhaps even melted by this heat. Mass fluidization of the sediment may result leading to convective transfer of material. Alternatively the cooling of a melted aureole may enhance the containment. If local heating is an insuperable problem, the container may have to be cooled for some decades under controlled storage prior to disposal, redesigned to provide better heat transfer or incorporate a lower percentage of waste.

3.4 Emplacement

The procedures for emplacing the containers within the sediment[8] depend on the required depth of burial. A streamlined projectile containing a waste package and falling freely from the sea surface is probably the cheapest method. At a terminal velocity of 30 metres/second, a penetration of 50 metres may be possible. This may however cause an unacceptable modification of the sedimentary environment and there may be problems with sealing the entry hole. Controlled emplacement on the end of a wire would be more time-consuming but hole-closure could be effected. A more costly, but technically feasible, procedure is to drill a hole, place several or many containers in it with suitable spacers for heat dissipation, and to seal to the sediment surface. This method would be necessary for burial deeper than 50 metres, and could be used to depths of between one and two thousand metres.

3.5 Fields for future research

Little is known of the behaviour of pore-water under the thermal and hydraulic gradients existing after emplacement especially at relatively high temperatures, so that data are required on permeability characteristics in a wide range of sediment types and over a wide range of temperature and compaction pressure. The ion exchange capacity will depend on the mineral composition of the sediment, the chemical composition of the pore-water and the waste it carries and on the environmental parameters. The conditions under which extremely high temperature gradients may induce a major change in mass properties of the sediment and lead to fluidization have not been quantified. Extensive studies will be needed of possible disposal sites to establish that they are sufficiently stable and will provide effective barriers. Quantitative answers to these problems are necessary before it can be decided how thick a barrier is required and hence what emplacement technique should be used.

4. DISPOSAL ON TO THE OCEAN FLOOR

If the wastes can be packaged so as to achieve a very slow rate of release activity, it is possible that the objective of waste disposal could be fulfilled by simply laying packages of solidified wastes on the ocean floor. In addition to its simplicity, this option offers the advantage of minimal cost.

It is to be noted that even if this option proves to be impracticable, research into it would not be wasted as the possibility must be considered of radioactivity from wastes buried under the ocean floor reaching the water mass. If this occurred the same considerations of transport back to man would apply as for disposal on to the ocean floor.

4.1 Package criteria

In this option the waste packaging is an important barrier. The primary objective of packaging is to contain the waste completely whilst a significant proportion of the activity decays away. It appears technically feasible to achieve this for that part of the activity consisting of fission products with relatively short half-lives (less than 100 years) which would require an outer containment with a mean life of the order of 1000 years.

The subsequent release of activity from the waste once sea water had penetrated the outer package would depend on the leach resistance of the waste form, its physical structure, its chemical form, its resistance to radiation and thermal damage and other factors. Although further research is needed on these aspects of the waste form it is clear that the radioactivity would be released slowly over a period of at least several thousand years from a well designed package[9].

Placing the package on the ocean bed has one very great technical advantage. The excellent cooling provided by sea water circulating round the outer containment virtually eliminates the problem of melting due to self-heating and may obviate the need for intermediate storage of packages in order to allow the heat production to fall to a manageable level.

4.2 Disposal area selection

Just as for disposal below the ocean floor, the best areas for disposal on to the ocean floor would be seismically quiet and remote from present and foreseeable activities of man. Attention has therefore been concentrated on the areas of the ocean bed in deep water away from the continental shelves and the edges of the oceanic lithospheric plates. Other factors such as the sedimentation rate and the presence of mineral deposits will also need consideration.

Provided that it could be ensured that the rate of release of radioactivity into the oceans would be very slow, the oceans can be regarded - to a first approximation - as "well mixed". It must be emphasized however that there are considerable uncertainties as to the effect major climatic changes may have on oceanic circulation patterns and predictive studies must be refined to take other possibilities into account.

4.3 Effectiveness

Assuming design of packages of the waste form such that there will be no significant release of fission products or other relatively short-lived components of the waste, there remains only the long-lived components of waste, particularly actinides. Modelling of the transfer processes envisages that they will enter the water mass and ultimately be transferred through it to man, being removed only by radioactive decay and perhaps by sediments. Preliminary modelling of this type[9, 10] has identified those assumptions which give rise to the greatest uncertainty in the predicted effects of these longer-lived waste components. The areas of uncertainty include both physical and chemical processes as well as the biological aspects of the pathways back to man, notably exploitation of marine foodstuffs. A comparison with natural radicactivity in the oceans does not suffer from quite so many uncertainties and on reasonable assumptions it may be shown that average concentrations of the actinides are unlikely to exceed a small percentage of the concentration of natural radium.

4.4 Future research needs

Reference has already been made to research needs in respect of the waste form. Further work on the oceanic processes is envisaged in three main areas - the scavenging role of sediment, the dispersion and advection processes in the ocean water masses and the biological processes by which activity will predominantly return to man.

Research into sediment uptake and the stability of adsorbed activity could substantially alter the results of predictive modelling. In addition, some of this information could be directly useful in evaluating the under ocean floor burial option. Our knowledge of the deep ocean is very scanty and broad simplified assumptions have been made in present theoretical assessments. It is necessary to improve these assumptions to the point where it is reasonably certain that no unexpected pathway exists which can transport radioactivity to man at a faster rate or in a more concentrated form than has been predicted.

5. CONCLUSIONS

On the basis of current knowledge and understanding, it seems that the problem of disposal of high-level radioactive waste can, at least in principle, be overcome. Before this can become a proven reality much more research effort is needed and this should be so directed as to achieve the successive elimination of uncertainties with the aim of arriving at the optimal solution.

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