Centrifuge Modelling of Radioactive Waste Disposal: A Review

Ashok K. Gupta* and D.N. Singh[†]

Introduction

Radioactive wastes have given rise to one of the major technical and political arguments of the current decade. Safe disposal of radioactive wastes is perceived as a core issue to the future of the nuclear power industry. Nuclear reactors produce large quantities of radioactive waste of great toxicity and persistence. Radioactive wastes occur in solid, liquid and gaseous forms and contain widely different amounts and types of radioactive isotopes and thus present different degrees of concern for pollution of the environment.

Since the beginning of the nuclear age, there has been no shortage of proposals on how to isolate radioactive waste from the biosphere, but with each proposal came an array of objections. For a quick review details are presented in Table 1. The idea of burying radioactive waste in the earth's crust has been identified as most attractive and is termed as the 'best, safest long term option'. Various countries, using nuclear power, are pursuing geologic burial as the solution to their waste disposal problems. For the purpose of discussing long term waste disposal, radioactive wastes are usually subdivided into two main categories: low-level waste (LLW) and high-level waste (HLW). This subdivision is based on an estimation of the long-term health hazard posed by the radioactive wastes, which is roughly related to its content of long-lived radioisotopes, such as plutonium, that emits alpha particles and thus is highly radiotoxic if inhaled or ingested. To the first

*   Senior Lecturer, Department of Civil Engineering, North Eastern Regional Institute of Science and Technology, Nirjuli – 791109, Arunachal Pradesh, India.

†   Assistant Professor, Department of Civil Engineering, Indian Institute of Technology, Mumbai – 400076, India.

Table 1
Technical Options for Dealing with Spent Fuel (Lipschutz, 1980; Nicholas Lenssen, 1992)

Method	Process	Problems	Status
Antarctica Ice Burial	Burial in ice cap	Prohibited by international laws, uncertain behaviour of ice sheets over long periods concern over catastrophic failure	Abandoned
Geologic Burial	Burial in mined repository (hundreds of meters deep)	Difficulty predicting geology, ground water flows, and human intrusion over long time periods	Under active study by all nuclear countries as favoured strategy
Long-term Storage	Store waste indefinitely in specially constructed buildings	Depends on human institutions to monitor and control access to waste for long time periods	Not actively being studied by governments, though proposed by non-governmental groups
Reprocessing	Chemical separation of uranium and plutonium from fission products in spent fuel; radioactivity decreases by 3%	Increases volume of waste by 160-fold; poor economics; increases risk of nuclear weapons proliferation	Commercially underway in few countries, some other countries have plans to reprocess spent fuel
Seabed Burial	Burial in deep ocean sediments	Possibly prohibited by international laws; marine transport problems; heat and radiation effects upon the sediments are not well understood	Under active study by some countries
Space Disposal	Disposal into solar orbit beyond earth's gravity	Potential launch failure could contaminate whole planet; very expensive	Abandoned
Transmutation	Conversion of waste to shorter-lived isotopes through neutron bombardment	Technically uncertain whether waste stream would be reduced; very expensive	Under active study by United States, Japan, Soviet Union and France

approximation, LLW can be regarded as material that must be isolated from the biosphere for 100 to 1,000 years, after which it becomes innocuous from the point of view of its radioactivity. HLW, by contrast, must be isolated for much longer times. 10^5 to 10^6 years is the period generally used as a guide line for developing appropriate disposal concepts and techniques for HLW. The low-level waste management may include disposal concepts depending on monitoring, surveillance, and if necessary, remedial action. However, the disposal of HLW depends largely on the behaviour of natural systems.

By the end of 1995, 437 nuclear power stations (10 in India) were operating world-wide, and another around 100 (4 in India) are still under construction (Source: BARC newsletter, No. 148, May 1996). In addition to these nuclear power stations there are a large number of nuclear reactors actively used for research and military purposes. To provide these reactors with fuel, many other nuclear facilities exist such as: uranium mines, ore processing plants, fuel fabrication facilities, and fuel reprocessing plants, all joined in a complex, world-wide transportation network. All these facilities produce wastes that emanate ionising radiation at appreciably higher levels than naturally occurring materials, levels that constitute a significant health hazard to present and future generations. Because of its long-term impacts on human health and the environment, radioactive waste disposal is the main agenda for national and international debate.

This review paper presents a brief background (dimension) of the radioactive waste problem and current disposal practices. The concept of accelerated physical modelling, using a geotechnical centrifuge, has been introduced to study the migration of contaminants in soils. The current literature available on accelerated physical modelling of migration of radionuclides and transfer of heat generated by high level wastes in the subsurface environment has been reviewed.

Dimension of the Problem

Largest quantities of radioactive wastes are generated by the operation of nuclear fuel cycle either to generate electrical power or to produce plutonium for atomic weapons. Dimension of radioactive waste problem is illustrated in Fig. 1 (Milnes, 1992). The figure depicts the sequence of steps necessary to produce and use nuclear fuel along with the quantity of waste expected from the use of a 1,000 MWe reactor for one year.

The extraction of natural uranium from mined uranium ores involves grinding them to a fine powder (milling) and then leaching out the uranium with acid or alkaline solutions (depending on ore type). Chemical separation produces U_3O_8 in the form of a yellow powder called yellow cake. To produce 180 tonnes of yellow cake, needed to yield enough fuel for a 1,000
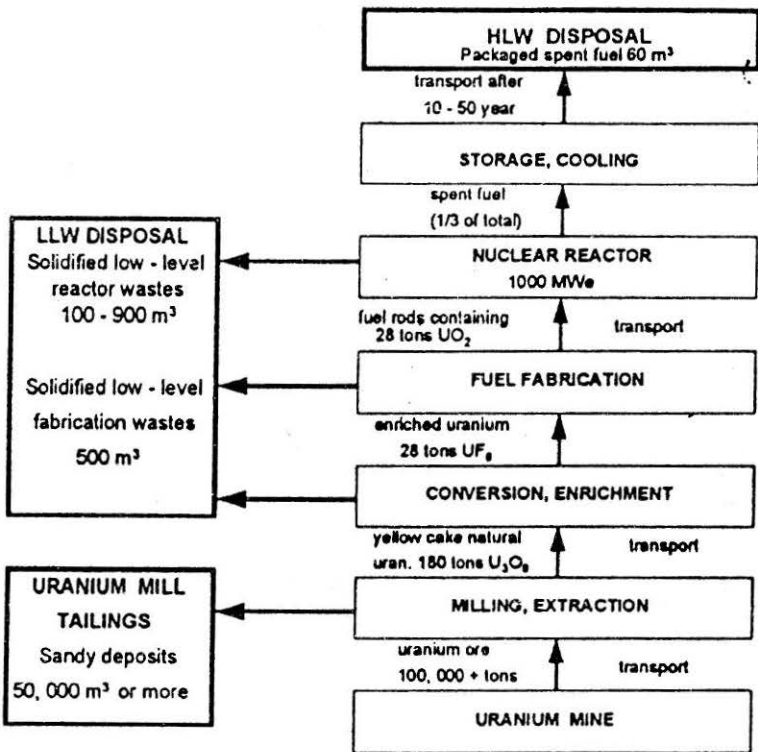
FIGURE 1 : Radioactive Wastes resulting from Nuclear Fuel Cycle

MWe nuclear power station for one year, more than 100,000 tonnes of ore are required; and more than 50,000 m³ of mill tailings are left behind which can be put in the category of LLW. These tailings contain isotopes of uranium, thorium, and radium. Out of these isotopes, Radium-226 (^{226}Ra), with a half life of 1620 years, poses the greatest environmental hazard. The maximum permissible concentration of ^{226}Ra in drinking water is 3pCi/l [the primary unit of amount of radioactivity is the curie (Ci), defined as a disintegration rate of 3.7×10^{10} disintegrations per second, picocurie (pCi) is a sub unit of Ci, 1 pCi $= 10^{-12}$ Ci or 3.7×10^{-2} disintegrations per second], which is equivalent to 10^{-9}mg/l. Thus, an extremely small amount of ^{226}Ra leached from waste tailings into ground water can cause the water to be unfit for human consumption. The yellow cake is transported to the fuel enrichment and fabrication facilities, where the production of the fuel rods results in LLW that in solid form occupies perhaps another 500 m³. The wastes arising from fuel enrichment generally contain ^{226}Ra, ^{230}Th, and ^{238}U in significant concentrations. Again, ^{226}Ra is the isotope of main concern.

Running a 1,000 MWe reactor for one year produces more LLW, in amounts between 100 to 900m^3. These wastes called reactor wastes mostly contain discarded equipment, assorted slightly radioactive refuse, and ion-exchange materials from decontamination facilities. Reactor wastes contain a variety of radionuclide species, with half-lives ranging from seconds to many decades or even longer. Of these nuclides, ^{137}Cs, ^{90}Sr and ^{60}Co, with half-lives of 33, 28, and 6 years respectively are usually regarded as posing the most significant environmental hazard. Wastes with these radionuclides need several hundred years to decay to very low radioactivity levels. At the same time, a certain amount of fuel is burnt up and has to be removed from the reactor core and placed in cooled and shielded water tanks.

About 10 tons of spent fuel are produced per reactor per year, which if it were treated as waste would have to be kept in storage for 10 to 50 years, until its heat producing capacity has died down sufficiently to allow processing and transportation, in which case it would be classified as HLW and have a volume of about 60 m^3. The spent fuel contains numerous radionuclide species. Some important radionuclides in spent reactor fuelel are ^{90}Sr (Strontium : 28 yr.), ^{137}Cs (Cesium : 33 yr.), ^{238}Pu (Plutonium : 86 yr.), ^{239}Pu (24,400 yr.), ^{240}Pu (6,580 yr.), ^{241}Am (Americium : 458 yr.), and ^{243}Am (7,950 yr.), the figures in parenthesis indicate the half-lives. These elements constitute more than 99% radioactivity of the spent fuel. Radioactive decay of these elements in the waste produces other radionuclides, known as daughter products such as ^{237}Np (Neptunium : 2,100,000 yr.), ^{226}Ra (Radium : 1620 yr.), ^{129}I (Iodine : 17,000,000 yr.), ^{99}Tc (Technetium : 210,000 yr.), and others. If these are also taken into account, the material will remain hazardous for millions of years, although at much lower radioactivity levels than will occur during the first thousand years.

Plutonium is a major component in the spent fuel, a valuable commodity which can be reprocessed by chemical means and used for power production in fast breeder reactors. The reprocessing option fundamentally affects the radioactive waste disposal problem which significantly reduces the amount of HLW to about 3 m^3 of solidified waste per reactor per year. On the other hand, the amount and variety of LLW is markedly increased to about 3,000 m^3 from the fuel cycle itself. In addition, much of the LLW will be contaminated with small amounts of plutonium, which may shift an unknown proportion of the LLW into the HLW category.

Table 2 presents approximate data related to the world-wide generation of total spent fuel from commercial nuclear plants (in 1985 and 1990), and the projections for the year 2000 (Lenssen, 1992). Most of the existing spent fuel is stored in large pools of cooling water alongside nuclear reactors. However, several other sources of radioactive waste, present and future, have not been included herein, such as: undisclosed types of military wastes, wastes

Table 2
Accumulation of Spent Fuel, in metric tonnes[#]
(Lenssen, 1992)

Country	1985	1990	2000
United States	12,601	21,800	40,400
Canada	9,121	17,700	33,900
Soviet Union	3,700	9,000	30,000
Japan	3,600	7,500	18,000
France	2,900	7,300	20,000
Germany	1,800	3,800	8,950
Sweden	1,330	2,360	5,100
India*	350	—	8,000
Others	5,879	14,540	35,315
Total	40,991	84,000	193,065

*   values in m^3 (Sethna et al., 1984).

\#   1 metric tonne of spent fuel $\cong$ 6 m^3.

derived from the decontamination and decommissioning of nuclear reactors and reprocessing plants after their expected lifetimes of 30 to 40 years, and wastes associated with the running of future reactor types, such as fast breeders and fusion reactors. As such, it is clear that our society is facing a major environmental problem which is likely to increase in complexity in future, and a solution to which is a must.

Disposal of Liquid Low-Level Waste

Present disposal trends are different than the direct release of liquid radioactive wastes into rivers, lakes or ground water as practised in the past, particularly up to the mid 1970s (Freeze and Cherry, 1979). Controlled ground percolation is still in use where some types of liquid LLW are directed to underground cribs or open ponds, from where the waste percolates down through the soil mass to the ground water table. Most radionuclides are selectively retarded by sorption on clay minerals, zeolites, etc., and the contaminant plumes are continuously monitored in surrounding wells. The crib or pond is decommissioned when the ground water concentrations of critical radionuclides exceed predefined limits. Now-a-days deep well injection systems, by which liquid LLW is pumped into aquifers at various depths, are in frequent use (Milnes, 1992). These systems are used for both LLW and HLW. Large scale field tests are conducted to study the mobility of the solutions, rates of radionuclide migration, effects of temperature increase, and

so on. However, world-wide experience with the deep injection of liquid wastes has shown that degree of confinement and migration pathways are difficult to predict and that extensive use also produces undesirable side-effects, such as upward leakage of the wastes, through abandoned unplugged wells and inducement of earthquakes as a result of increasing pore water pressures along the faults.

Deep well injection of grout, a mixture of concentrated LLW liquids and cement solids, is also in use, wherein the grout is injected under high pressure into some suitable geologic formations such as shale and salt deposits. The grout creates space for itself in the form of narrow sub horizontal fractures or cavities where it solidifies in thin sheets at various levels. Here, the radionuclides are effectively immobilised in an environment virtually free of circulating ground water.

## Disposal of Solid Low-Level Waste

Modern LLW management programme involves converting the waste into an inert solid form, reducing volume and producing manageable, standard-sized units for easy and safe transportation. A typical standard-sized unit is a canister made of steel, concrete or plastic containing particulate LLW embedded into a solid matrix of concrete, glass, bitumen or some other monolithic solid of low dispersibility. Such LLW canisters have been disposed off in five different geological environments as discussed in the following.

Above ground container storage

The wastes are placed in strong engineered containers constructed of materials such as concrete and steel situated on the ground surface (Fig. 2a). In these containers the wastes can remain in storage in areas away from population. To extend the life span of containers, these can be covered with

Average water table

(a)   (b)
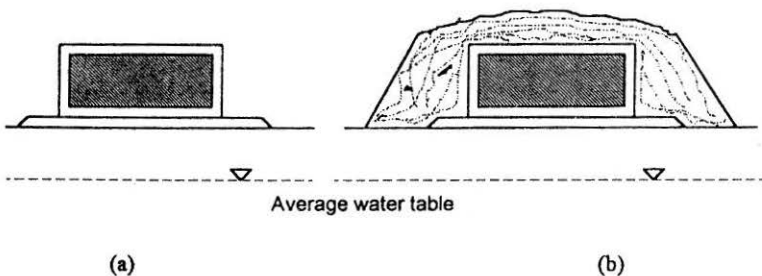
FIGURE 2 : Above Ground Container Storage of LLW
(a) Without Protection; (b) With Protection by Geologic Materials
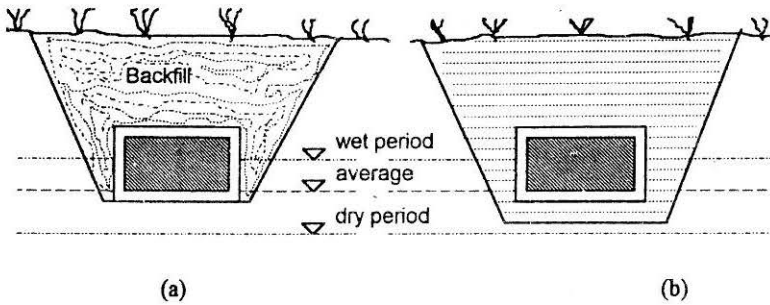
FIGURE 3 : Shallow Trench Burial of LLW
(a) With Backfill; (b) With Additional Protection provided by Engineered
Zone of Special Geological Material

properly designed earth materials (Fig. 2b). Such facilities must be under some responsible organisation to monitor deterioration of the containers. If problems arise, the containers can be repaired or the wastes can be placed in new containers.

On the ocean floor

Dumping of LLW packages into the sea has been carried out extensively in the past. At present there is a moratorium on sea dumping, pending the resolution of political problems and the synthesis of the data from an extensive research and monitoring programme aimed at learning more about the mechanisms of canister corrosion, matrix leaching, and radionuclide migration and concentration in the deep ocean (Milnes, 1992).

In shallow trenches on the land surface

The most common land based disposal strategy is shallow land burial of solid LLW in trenches few meters below ground surface, either above or below the water table (Freeze and Cherry, 1979). The earth material from the excavation is used as backfill around the containers (Fig.3a), or the fill in the excavation is designed to provide enhanced protection for the system (Fig. 3b). If the water table fluctuates within the zone of burial, there will be a possibility of container deterioration and radionuclides escaping into the environment. Nearly all burial sites in many countries fall in this category, with the water table within or just below the burial zone. Further, most of the sites are located in poor hydrogeologic settings. Therefore, subsurface migration of radionuclides from the burial zones is a common phenomena. The shallow land burial system works well in arid regions with thick deposits of porous and permeable sediments and a deep water table.
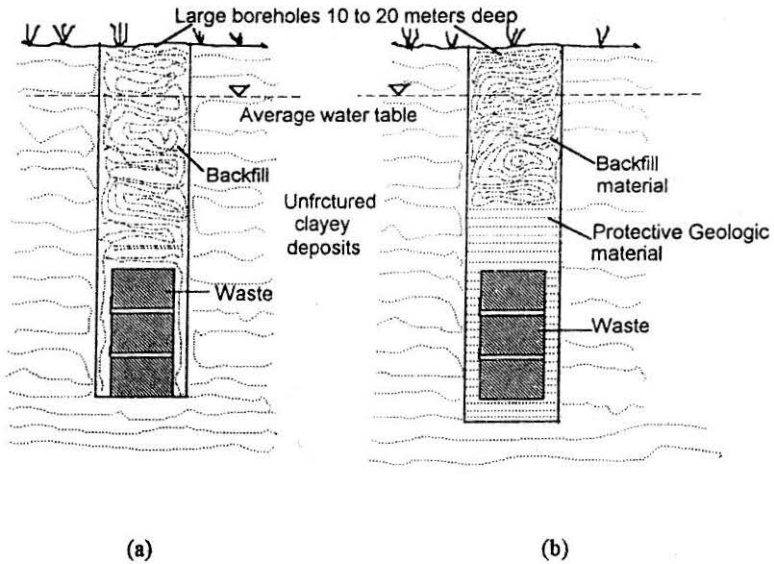
FIGURE 4 : Deeper Burial of LLW
(a) With Backfill; (b) With Protection by High-Retardation Geologic Material

Deeper burial in large diameter borehole

The canisters are buried in large holes about 10 to 20 m deep. In non-arid regions the permanent water table in these situations would normally be above the canisters. It has been proposed (Freeze and Cherry, 1979; Krauskopf, 1988) that in non-arid regions, the burial zones should be located in unfractured clayey aquitards where burial would occur at the bottom of large diameter (2 to 5m) auger holes. The hole is either simply backfilled (Fig. 4a) with material originally removed from the site, or the excavated zone around the canisters is designed with suitable geological material (Fig. 4b) to improve the long-term containment capability of the burial facility.

In shallow mined cavities in under ground rock

The use of shallow mined cavities in the underground rock for the disposal of solid LLW is favoured particularly by those countries where public opinion is against shallow land burial (Krauskopf, 1988; Milnes, 1992). The cavities may be abandoned mines or specially constructed underground facilities at depths of up to a few hundred meters. The critical factor here is the geohydrology. If water exclusion can be guaranteed for times up to 1,000 years, a high degree of safety can be attained.

Disposal of Mill Tailings

Mill tailings are the by-product of uranium mining and processing, and consist of large quantities of natural radioactive rock and soil mass. Mill tailings contain small amounts of excavated radium, which decays to emit the radioactive gas radon and other daughter products (Brookins, 1984). Uranium mill tailings are too voluminous to package or transport, therefore, create a special problem of LLW disposal.

Tailings may be placed in specially excavated pits, mines or in some other suitable locations. One reasonable disposal method is backfilling in mines with a planned period of surveillance and monitoring. The ultimate aim is to ensure that the tailings become an integral part of the natural ground surface by levelling, putting a thick soil cover, and re-vegetation etc., with a low or predictable release of radioactive contaminants. The backfilling depends very strongly on the characteristics of the mill tailings and its disposal site. Some of these are ore type, leaching process, neutralization system, impoundment structure, climate and hydrogeology (Brookins, 1984; Krauskopf, 1988). Attempts should be made to improve uranium mill tailings i.e. the removal of radium by sulphate precipitation, etc. Now-a-days, tailings are well designed and monitored. Abandoned mill tailings need to be assessed in terms of potential hazard to the environment.

Disposal of High-Level Waste

The safe disposal of high-level waste presents a much more difficult problem than low-level waste because of the much longer time for which isolation should be guaranteed. However, because of small volume of waste involved, the HLW can be carefully treated and packaged and, if need be, transported to distant localities. This has resulted in an enormous upswing in research and development in the following three areas:

(a) development of industrial-scale HLW stabilization process,

(b) world-wide search for suitable geological environments as repository sites, and

(c) assessment of the effects, rates and probabilities of geological processes and events over long periods of time.

The HLW disposal strategy consists of two broad stages called Immobilisation and Disposal in deep-mined cavities.

Immobilisation

For HLW from reprocessing, treatment can take place only after 10 to 20 years of storage to allow heat production and radioactivity to subside to manageable levels. This HLW should be immobilised by incorporating into a vitreous or a ceramic matrix which are discussed in the following:

Vitreous waste matrices are favoured at present, and their development is already on the industrial scale. Vitrification involves the mixing of solid waste with a borosilicate glass frit. The mixture is melted in a special furnace, and the liquid glass may be cast into a mould or poured directly into a metal canister. Borosilicate glass has been considered a suitable matrix for HLW because the glass, being an amorphous solid with strong inter atomic bonding and no strict atomic structure, is able to contain a variety of different elements. At moderate temperatures and pressures glass has low leachability and is resistant to structural damage from radiation, primarily the alpha particles (Kerr, 1979). However, some experiments have shown that borosilicate glasses can be highly unstable causing devitrification, alteration, fissuring and disintegration under the temperature, pressure and geochemical conditions expected in some underground repositories (Lipschutz, 1980).

Ceramics are synthetic crystalline rocks, one well known variety is called 'SYNROC' having an ordered atomic structure and that can be tailored to specific waste elements and geochemical conditions. In the radioactive waste ceramics the radionuclides are bound in the crystal lattices of the various mineral like phases and the composition of the pre treatment mixture is tailored so that treatment (e.g. hot pressing at 1,100°C) incorporates all of them into crystal structures. The stability of waste incorporated ceramics has been well accepted (Kerr, 1979). However, ceramics are much more prone to damage when the ordered crystalline structure is disturbed by radiation. Another problem is transmutation i.e., when an atom of waste decays, its daughter may have different chemical properties, thus altering the stability of the ceramic (Lipschutz, 1980).

Immobilisation of radioactive waste in an inert matrix is just one aspect of the production of a stable waste package. It is followed by encapsulation, in which the immobilised waste is placed in canisters composed of one or several shells of inert metal (e.g., chrome steel, lead, copper or titanium) or other materials (e.g., corundum). Also, after emplacement in the repository, it is envisaged to buffer the canisters with fill material (e.g., bentonite, zeolites and manganese oxides) that has special properties for retarding the migration of radionuclides, should the other barriers be breached.

Disposal in deep-mined cavities

The disposal of HLW canisters in deep underground repositories has been most consistently followed and subjected to the most extensive research and development program. It consists of a system of tunnels at depths up to 1500m in the earth's crust in which HLW canisters are emplaced individually, dimensioned and spaced to limit temperature build-up and which can be subsequently backfilled with an appropriate buffer material and permanently sealed (Brookins, 1984; Chapman and McKinley, 1987; Krauskopf, 1988). The ground above the repository would then become a protected area in which future drilling activities would be forbidden. For permanent disposal of HLW canisters, a geological system of predictable long term stability and free from circulating groundwater must be found which should not be connected with potentially valuable raw materials. The potential body of rock should exhibit a high degree of homogeneity over a vertical distance of hundreds of meters and over a horizontal area of several square kilometres. There should be no large- scale fractures in the area, and it should lie away from earthquake zones and active or dormant volcanoes. Two rock types are widely favoured as enclosing media for underground HLW repositories: rock salt and granite, but several others such as basalt, tuff, shale, and anhydrite are or have been under consideration in some very particular situations.

Salt formations (bedded salt or salt domes) are being investigated in many countries (Brookins, 1984), and underground testing under simulated repository conditions has also been carried out. Salt (i.e., mineral halite) was initially singled out for further study because of the complete dryness of most salt formations, combined with the idea that because salt is very easily soluble, the preservation of large salt deposits formed tens or thousands of millions of years ago would guarantee hydrogeological stability far into the future.. Salt is plastic in nature and flows under pressure, a property that assures that openings and joints are sealed. Salt dissipates heat well and is highly impermeable to noncirculating water. However, most salt deposits are finely laminated and contain thin interbeds of other evaporite minerals, which in the case of dome salt are generally highly contorted due to heterogeneous flow during the diapiric rise of the dome (Chapman and McKinley, 1987; Krauskopf, 1988). The specific character of these impurities is important in assessing a particular salt formation. Bedded salt contains tiny inclusions of salt-laden water called 'brine'. Salt becomes more soluble in water if heated, and this property allows brine inclusions to move through salt in the direction of the heat source (hot waste canister in this case). The role of brine and the effect of heat on a salt bed on the long term is a complex matter needing detailed investigations. Salt has poor sorption characteristics and is frequently associated with oil and gas and is itself a natural resource (Lipschutz, 1980). Now, it has been realised that a more realistic and pragmatic approach should be based on extensive in-situ experimentation and predictive modelling (Fairweather, 1991).

Granites are fairly homogeneous and fine to medium grained. Granites form well below the earth's surface, hence the porosity is low and as such, contain very little moisture. The permeability of such rocks is also low, especially for unweathered granite. Granites have high strength and high resistance to normal weathering conditions. Granites are also rigid and do not deform under most stresses at shallow depth, as well as less susceptible to heat effects than bedded salts. The main disadvantage of any hard rock including granite is that it is usually well jointed and cut through by fracture zones, which are planes of weakness for the future release of crustal stresses and pathways of easy fluid migration.

Basalt and tuff are not regarded as good repository medium because they usually are full of joints and have complex structure and natural moisture, but they are being investigated in detail because they are dense and strong and they form the bedrock of several existing nuclear facilities. Shale layers also generally contain fractures and joints that, below the water table, are full of water. High temperatures of waste canisters may cause weakening, dehydration, and crumbling of shale. Shales are being studied because they are very compact, have low permeability and solubility and possess high ionic retention.

Sub-seabed disposal

An alternative to disposal in mined geologic repositories is disposal in seabed. Stable, deep areas of the ocean floors, where thick sediment layers have been deposited and that are free from water currents, are preferred as the disposal sites. Some of these areas are considered to be among the most stable geophysical features on the earth (Chapman and McKinley, 1987; Roxburgh, 1988).

This method of disposal consists of either, drilling holes in the seabed, placing the waste canisters in the holes and then inserting a plug of inert material, or allowing the canisters, fitted with pointed ends and fins to drop vertically into the sediments at a velocity high enough to embed them deeply. Sediments fall and settle into the hole produced and thus provide a protective layer or seal. Any radioactive material leaking out of the canisters would be held by the sediments and take many years to diffuse into the sea water. Some disadvantages of seabed disposal are associated with difficulties in transporting the wastes over large distances, the possibility of radionuclide transport by sea water, and international political uncertainties.

Subsurface Radioactive Waste Transport

For radioactive waste, the main contamination pathway is the transport of waste radionuclides in the soil mass following groundwater intrusion into

a repository. Once in solution, the transport of radionuclides is similar to any conventional contaminant transport phenomena. Transport of radionuclides in porous media is mainly governed by either one or a combination of: advection, hydrodynamic dispersion and molecular diffusion (Freeze and Cherry, 1979; Pickens and Grisak, 1981; Shackelford and Daniel, 1991; Gupta and Singh, 1997). For the sake of brevity these processes are not being discussed herein.

Another important transport mechanism is the source-sink phenomena which governs the migration of radionuclides and the same is being presented in the following for the sake of completeness. The source-sink components are sorption and radioactive decay (Zimmie et al., 1994). The one-dimensional solute transport equation for radionuclide migration can be described by the following partial differential equation:

$$\frac{\partial}{\partial t}(\eta c + \gamma \bar{c}) = D\frac{\partial^2 c}{\partial x^2} - v\frac{\partial c}{\partial x} - \lambda(\eta c + \gamma \bar{c}) \tag{1}$$

where the sorption concentration $\bar{c} \cong c K_d$, c is solute concentration and K_d is the distribution factor. With this substitution Eqn. 1 becomes

$$\frac{\partial c}{\partial t}\left(1 + \frac{\gamma}{\eta}K_d\right) = D\frac{\partial^2 c}{\partial x^2} - v\frac{\partial c}{\partial x} - \lambda c\left(1 + \frac{\gamma}{\eta}K_d\right) \tag{2}$$

Substituting

$$R = \left(1 + \frac{\gamma}{\eta}K_d\right) \text{ (Freeze and Cherry, 1979), Eqn. 2 can be written as}$$

$$\frac{\partial c}{\partial t} = \left(\frac{D}{R}\right)\frac{\partial^2 c}{\partial x^2} - \left(\frac{v}{R}\right)\frac{\partial c}{\partial x} - \lambda c \tag{3}$$

where   t = time,
               η = porosity,
               γ = bulk density of soil,
               D = coefficient of hydrodynamic dispersion,
               x = distance from the source,
               v = seepage velocity,
               λ = radioactive decay, and
               R = retardation factor.

To study the migration of radioactive contaminants, the decay term (λc) should be evaluated. This term depends on the half-life of the radionuclides of interest and duration of the study. When dealing with radionuclides with short half-lives relative to the duration of the test, the decay term in the above equations should be considered. With radionuclides having long half-lives, the decay term can be neglected.

Centrifuge Modelling

Basic concepts

Centrifuge modelling is a major tool available to the geotechnical engineer. It enables the study and analysis of various geotechnical engineering problems. The basic concept of centrifuge modelling is that a model with linear dimensions scaled down by a factor of $1/N$ experiences the same magnitude and distribution of self-weight stresses as those of prototype, when subjected to a centrifugal acceleration N times greater than that due to the gravity. Further, it is possible to accelerate the flow and transport processes that govern the fate of contaminants as they pass through soils and aquifers. Large models can also be tested on large-capacity centrifuge and hence enabling inclusion of large number of heterogeneities in the model. The transport processes of contaminants occurring in centrifuge models under controlled boundary and initial conditions may be used as 'field data' to verify and improve the capabilities and efficiency of various mathematical models viz. FEM and FDM. To predict the prototype behaviour correctly from the observation of model behaviour, similarity condition must be established for the model and the prototype.

Centrifuge scaling relations

General scaling laws that govern the relationship between the model and its corresponding prototype, with respect to the contaminant transport problem, have been derived by Arulanandan et al. (1988) using dimensional analysis. Savvidou (1988) has derived scaling relationships for heat transfer through the soil mass. The basic assumption involved in the derivation of these scaling laws is that both the interstitial fluid and the soil grains are incompressible. A summary of scaling relationships is presented in Table 3. This can be noticed from the table that for flow through saturated porous media, the time for advection and diffusion in the model varies inversely as the square of acceleration scale, N. For example, at 200 g, a 24 hour centrifuge test on a model 200 times smaller represents 109 years of full-scale prototype behaviour.

For a situation where advection, dispersion and adsorption occur, physical properties that define the concentration, c, of a contaminant are given by:

Table 3
Scaling Relations for Centrifuge Experiments

Parameter	Model to Prototype Ratio
Length	$1/N$
Velocity	N
Mass-density	1
Force	$1/N^2$
Stress	1
Strain	1
Time (Dynamic)	$1/N$
Time (Seepage)	$1/N^2$
Time (Diffusion)	$1/N^2$
Time (Creep)	1
Heat transfer by pure conduction	$1/N^2$
Temperature change	1
Rate of heat generation per unit area (heat flux)	N
Heat transfer by convection	$1/N^2$
Rate of heat generation per unit volume	N^2

$$c = f\left(\mu, D^*, s, v, \sigma, \rho, g, l, d, t, \text{bulk soil properties}\right) \qquad (4)$$

where

c = concentration of the contaminant,

μ = dynamic fluid viscosity;

D^* = coefficient of molecular diffusion,

s = mass of adsorbed contaminant per unit volume,

v = interstitial flow velocity,

σ = surface tension for fluid/particle interface,

ρ = density of fluid,

g = acceleration due to gravity,

l = characteristic macroscopic length e.g. sample height,

d = characteristic microscopic length e.g. particle size, and

t = time.

The independent variable represented by 'bulk soil properties' will be assumed to be identical in both model and prototype. For the remaining ten independent variables and one dependent variable c, the following non- dimensional groups have been derived (Arulanandan et al., 1988):

$$\pi_1 = c/\rho \ \text{(Concentration number)}$$

$$\pi_2 = \rho v d/\mu \ \text{(Reynold number, } R_e)$$

$$\pi_3 = v t/l \ \text{(Advection number)}$$

$$\pi_4 = D^* t/l^2 \ \text{(Diffusion number)}$$

$$\pi_5 = \rho g l d/\sigma \ \text{(Capillary effects number)}$$

$$\pi_6 = s/\rho \ \text{(Adsorption number)}$$

$$\pi_7 = g t^2/l \ \text{(Dynamic effects number)}$$

$$\pi_8 = v d/D^* \ \text{(Peclet number, } P_e)$$

For similitude of transport processes in porous media, these eight controlling dimensionless groups must be identical both in the model (m) and the prototype (p). The conventional centrifuge conditions are

$$l_m = \left(\frac{1}{N}\right) l_p \ \text{and} \ v_m = N v_p$$

If the contaminant and the soil types are identical in both model and prototype, then the conditions

$$\rho_m = \rho_p; \ \mu_m = \mu_p; \ \text{and} \ d_m = d_p$$

and are also valid. The importance and scaling requirements of each of the eight dimensionless groups is discussed in the following paragraphs.

π_1 is the dependent non-dimensional group as it can be written as a function of the other seven groups. Strict scaling of π_1 will lead to the condition $c_m = c_p$, i.e. concentration in the model and prototype will be identical at homologous points. or, can be written as:

$$\pi_1 = f\left(\pi_2, \pi_3, \pi_4, \pi_5, \pi_6, \pi_7, \pi_8\right) \tag{5}$$

π_2 represents the dynamic similarity of fluid motion, by ensuring that the ratio between inertial and viscous forces in the fluid remain invariant. But, since flow velocities are scaled in a centrifuge model, it is not possible to maintain π_2 constant if identical materials are used in model and prototype. However, in most of the cases encountered in contaminant transport, the inertial forces are negligible in comparison to the viscous resistance (i.e. $R_e < 1$). In such cases, the condition that the Reynolds number remains invariant can be waived, and Darcy's law can be used to describe the fluid motion.

π_3 presents kinematic similarity of motion by ensuring that the groundwater seepage patterns in prototype and model are geometrically similar. The model to prototype ratio of $(v \cdot t /l)$ remains invariant when $t_m = \left(N^{-2}\right) \cdot t_p$.

π_4 indicates similarity of diffusion processes in the model and the prototype. The coefficient of diffusion for a contaminant (ionic form) in porous media is a function of both the medium and the free diffusion coefficient of the ion in solution. For an identical contaminant and identical material subjected to similar stresses in model and prototype, the condition $D^*_m = D^*_p$ should be satisfied. The model to prototype ratio of $\left(D^*t/l^2\right)$ remains invariant when $t_m = \left(N^{-2}\right) \cdot t_p$. This is in agreement with the time scale function of π_3.

π_5 describes the phenomena of capillarity. If capillary effects, such as the height of capillary rise or flow above the groundwater table are to be simulated, then π_5 must be identical in model and prototype. For identical soil and contaminant, $\sigma_m = \sigma_p$, $\rho_m = \rho_p$ and $d_m = d_p$ which leads to $g_m = N \cdot g_p$. This is consistent with the centrifugal scaling law required for similitude of the stress.

π_6 must be same for the model and the prototype for similitude of the adsorbed contaminant concentration, s. This is possible if identical materials are used and equilibrium adsorption is achieved in both model and the prototype. Equilibrium adsorption is achieved if s is only a function of solute concentration c in the pore fluid with no additional time dependence. This is often the case for flow through uniform soils. Further, for flow of certain contaminants through some types of soils (e.g. any solute through sand), adsorption is not an important factor for the transport process, and as such, π_6 can be ignored.

π_7 represents the relationship between the model acceleration and the ratio of characteristic sample length over characteristic time squared. Correct scaling of π_7 is important for modelling of dynamic events such as earthquakes or explosions where inertia forces are significant. Similitude of

π_7 leads to the conclusion that dynamic events occur N times faster in the centrifuge model than in the corresponding prototype i.e. $t_m = \left(N^{-2}\right) \cdot t_p$. This time scale factor is different for diffusion as discussed earlier. However, for most of the situations involving laminar flow through porous media, inertial forces are unimportant and π_7 need not be considered.

π_8 gives the relative importance of mechanical dispersion and molecular diffusion for the process of hydrodynamic dispersion. It is not possible to maintain π_8 constant if flow velocities are scaled (i.e. $v_m = N \cdot v_p$) and identical materials are used in the model and prototype. However, it has been shown (Bear and Verruijt, 1987) that the hydrodynamic dispersion coefficient D which is a function of flow velocity, remains invariant when the Peclet number is less than unity (i.e. $P_e < 1$), which indicates the predominance of molecular diffusion in hydrodynamic dispersion. In this case D is independent of flow velocity and hydrodynamic dispersion can be modelled without ensuring similarity of π_8, such as flow through fine-grained soils, where v is relatively very small. For values of Peclet number greater than unity (i.e. $P_e > 1$), mechanical dispersion becomes a dominant component of hydrodynamic dispersion. Since mechanical dispersion is dependent upon velocity, the whole process of hydrodynamic dispersion becomes velocity dependent and can not be correctly modelled in centrifuge experiments since $v_m = N \cdot v_p$. From practical standpoint of contaminant transport, the error due to high Peclet number is not of major concern and is often on the conservative side because the time of contaminant transport is underestimated.

Further, it has been observed (Arulanandan et al. 1988, Zimmie et al. 1994) that regardless of the Peclet number and even if any other scaling relation is invalid, the centrifuge models are useful for validating and calibrating numerical transport models.

Modelling of models

Prototype monitoring being often too costly and impractical, the concept of modelling of models has evolved to provide a check on the consistency of the centrifuge model testing scheme and to validate the scaling relationships (Taylor, 1995). Models of different heights when tested at the corresponding g levels must provide with similar extrapolated prototype results. This technique, known as 'modelling of models' has been successfully used by a number of researchers to validate the results.

Types of Problems

Study of radioactive waste migration in soil mass

Centrifuge experiments have recently been conducted to study

contaminant migration from simulated repository sites located on/in land. out of various contaminant transport phenomena, as discussed earlier, the predominance of a single phenomena depends on various factors. Some of these are of a geotechnical nature (like permeability, particle size distribution, soil mineralogy, etc.) while others are related to the nature of the waste itself (like ionic charge, diffusivity, concentration, etc.). Of great interest in radioactive waste studies is the possibility of modelling sorption and diffusion phenomena in the centrifuge. This is because low and medium level radioactive wastes are generally disposed off in repositories having low soil permeabilities.

Hensley and Schofield (1991) have investigated the technique of accelerated physical modelling of groundwater flow using a geotechnical centrifuge. A wide variety of initial and boundary conditions have been used in the centrifuge tests performed to study the migration of conservative pollutants. Prototype times of upto 30 years have been modelled during the test series, and the data generated by these tests have been used to validate the computer code, POLLUTE.

Villar and Merrifield (1992) have studied the behaviour of radioactive tracer Tc-99m in uniform fine sands in a geotechnical centrifuge.. The main advantages of using radiotracers are:

(a)   a more faithful replication of the behaviour of actual waste components,

(b)   the ease of measurement of even minute amounts of the tracer, and

(c)   the high level of test reproducibility control and the prompt reuse of the model even if sorption occurs due to short half life of the tracers.

The tracer radiation can be easily detected with small Geiger Mueller tubes (G-M tubes) attached to the centrifuge.

Villar and Merrifield (1994) have studied migration patterns of selected radioactive waste components in three soils at different Peclet numbers. A rigid aluminium box has been used to contain the soil model and a rectangular perspex box embedded in the soil surface simulates a repository. The base of perspex box has been perforated to allow solute flow into the soil, simulating a repository failure. A constant downward vertical flow has been maintained through the soil model by application of a constant hydraulic head. Miniature Druck transducers were used to monitor the pore water pressures at three heights in the soil. The radioactive plume in the soil model has been monitored by 12 miniature G-M tubes placed in the soil model. As the position of each G-M tube is known, the time required for the radioactive plume to travel between contiguous tubes is used to determine the migration

Table 4
Summary of test results showing the main characteristics of radioactive waste migration (Villar and Merrifield, 1994).

Test No.	Soil Bed	Radiotracer used	N	Migration Velocity (mm/min)	Pe
1	Silica sand	$Na^{99m}TcO_4$	100	7.64	14
2	Silica silt	$Na^{99m}TcO_4$	50	2.3	1
3	Silica silt + Illite	$Na^{99m}TcO_4$	50	1.75	4
4	Silica silt + Illite	$Na^{99m}TcO_4$	100	10.5	17
5	Silica sand + Illite	$^{90}YCl_4$	50	1.75	4
6	Silica sand + Illite	$^{90}YCl_4$	100	12.5	28

velocity of the tracer. Under non-sorptive conditions, this velocity represents the seepage velocity of water. A summary of test results is shown in Table 4.

Zimmie et al. (1994) have reported the results of a series of tests performed on silt-sized silica flour models to study radionuclide migration using I-131 as a tracer; the migration of I-131 has been detected by using an array of nine G-M tubes. The observations are noticed to be in good

Table 5
Thermal Power of Waste as a Function of Time in Watts per metric tonne of Radwaste (Zhao, 1992).

Time from reactor discharge (years)	Vitrified high-level waste	Spent nuclear fuel
10	1120	1290
100	134	284
1,000	6.8	49.4
10,000	0.6	13.5
100,000	0.1	1.0

agreement with those made by coloured dyes as tracer materials and also with finite difference modelling.

Modelling of heat transfer in soil

Modelling of heat transfer, related to the storage and disposal of heat generating high level radioactive wastes in soils is an important problem in geotechnical engineering. Both vitrified high-level waste and spent nuclear fuel generate heat from radioactive decay which decreases with time. Typical rates of heat generation and their evolution over time for these two forms of radioactive wastes are shown in Table 5.

The thermal power due to radioactive decay of wastes generates a time dependent temperature rise. The heat transfer is mainly by conduction in the repository host medium (rock, salt or clay). Convection through groundwater movement is negligible due to the deep disposal and impervious medium. The scaling relationships for heat transfer are mentioned in Table 3. The temperature evolution around heat generating wastes is characterised by a rapid temperature rise after emplacement of the waste canisters. The repository temperature elevation could be more than 30°C which will occur within 50 to 100 years for reprocessed waste (Zhao, 1992). For spent fuel, the maximum repository temperatures can be maintained for about 1,000 years or longer (Zhao, 1992).

Savvidou (1988) studied convective heat transfer in centrifuge models and 1-g models using fully saturated coarse sand and attempted to verify scaling laws by the modelling of models technique, giving a close comparison to the derived time scaling factor of N^2. Convective heat transport is found to be dominant in centrifuge models while in the 1-g tests, conduction contributes significantly. However, more model tests are required to be performed on centrifuge in order to attempt correct modelling of heat transfer in soils and rocks.

Conclusions

In this review an environmental problem of radioactive waste disposal has been highlighted with the help of statistical data. The current disposal practices for various types of radioactive wastes and the state of understanding have also been presented. Theories of various transport mechanisms for contaminants (radioactive wastes) in the subsurface environment have been discussed in brief. The usefulness of a geotechnical centrifuge has been discussed especially in relation to contaminant transport phenomena. Some problems have been identified which need further research especially with the help of accelerated physical modelling using a geotechnical centrifuge.

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