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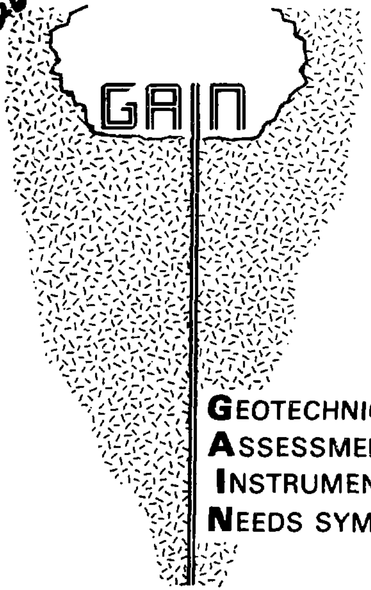

GAIN

GEOTECHNICAL
ASSESSMENT &
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NEEDS SYMPOSIUM

JULY 16-19 / 19-20, 1978

Lawrence Berkeley Laboratory
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Berkeley, California 94720

MASTER

An Appraisal of Underground Radioactive Waste Disposal in Argillaceous and Crystalline Rocks: Some Geochemical, Geomechanical, and Hydrogeological Questions

prepared by

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Crystalline and Argillaceous Rocks for Radioactive Waste Storage

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INTRODUCTION

Radioactive wastes have been generated now for more than three decades and will continue to be produced in the foreseeable future. Large quantities of these wastes exist and are currently stored at a number of surface and near-surface sites. The quantities of radioactive wastes stored at such sites are increasing each year.

Though every precaution is taken to protect the environment and man from the adverse effects of these wastes, most experts and laymen agree that near-surface storage is neither an acceptable nor practicable long-term answer. The speed and manner in which the disposal problem is resolved is likely to have major consequences, economically, politically, scientifically, and socially.

Of the many alternatives that have been considered for the disposal of these wastes, deep underground burial is the most favored. Intuitively, such disposal would seem to secure the wastes against events such as meteorological and geological changes, acts of terrorism, and political turmoil. Archeological and geological investigations thus far support such intuition. Also, there is a wealth of experience concerning underground excavation. The countries of the Organization for Economic Cooperation and Development alone construct a total of some 50,000 km of tunnels (OECD Advisory Report on Tunneling, 1970) each year, under virtually every kind of terrain, beneath land and sea, to depths approaching 4 km below surface. Some underground mines have been in operation for more than a century and many mines use excavations fifty or more years old.

The disposal of radioactive wastes, however, involves three principal factors which lie outside the realm of mining experience. First and foremost, radioactive materials must not be allowed to escape from the repository to the biosphere at levels that constitute a hazard to life. Second, containment of the wastes must be effective for unprecedented periods of time, of the order of a million years. Third, the wastes generate heat within the repository by radioactive decay.

In general, neither experience in mining nor in the fields of geomechanics, hydrogeology, and geochemistry at present are sufficient to predict the behavior of an underground repository for the storage of radioactive wastes with the degree of certainty required for the periods of time involved.

Accordingly, it is important that the necessary research be done in the fields of geochemistry, geomechanics, and hydrogeology to enable adequate predictions to be made concerning the performance of an underground repository for the disposal of radioactive wastes. Undoubtedly, the success of such repositories depends upon many factors and sciences but the first decisive questions involving geochemistry, geomechanics, and hydrogeology must be answered.

In this paper, we have endeavored to appraise the potential for the storage of radioactive wastes by burial in underground repositories and to define the more significant factors affecting the selection and design of a repository. We have tried to identify those areas in geomechanics, hydrogeology, and geochemistry where further research is needed to provide answers of the quality and certainty needed to resolve these issues.

II

STRESSES IN ROCK

The Virgin State of Stress

In general the vertical component of the virgin state of stress in rock has a value close to that given by the weight of the overburden. Departures from this may occur in areas of rapid erosion, in areas of uneven topography at shallow depths compared to local surface relief, or in and close to inclusions and intrusions of rock with mechanical properties or temperatures different from those of the surrounding rock.

A significant number of attempts has been made to measure the complete virgin state of stress in rock at different locations and depths throughout the world. These measurements have shown that the values of the horizontal components of this state of stress range from about a third to three times that of the vertical component. A compilation of many of these measurements has been done by Hoek and Brown (1977), and is shown in Figure 1. From this it can be seen that relatively high values of the horizontal components of stress tend to be a shallow phenomenon, possibly associated with the effects of rapid denudation (Voight, 1966).

The value of the vertical component of rock stress is, on average, some 2.7 times greater than the hydrostatic head of water at the same depth; that is, the value of the ratio of the hydrostatic head to the vertical stress is 0.37, as is illustrated also in Figure 1. This is a result of the ratio between the average density of rock and that of water.

The virgin state of stress at the site of any potential repository for radioactive wastes has important implications for the hydraulic transmissivity of the rock mass, the geomechanical stability of the site, and the stability of any excavations which may be made.

Hydraulic Transmissivity

To insure that such near-vertical joints and fractures as may exist in the rock at a site are not opened by the hydraulic pressure of groundwater, the minimum component of the horizontal stress must be greater than the hydrostatic pressure at all depths. The minimum value of the horizontal stress should, therefore, lie to the right of the line showing the hydrostatic pressure in Figure 1; the greater the amount by which the horizontal stress exceeds the hydrostatic pressure the lower will be the hydraulic transmissivity of near vertical joints and fractures.

Faulting and State of Stress

Potential fault movements at the site depend upon the ratio of the difference between the normal stresses and the effective normal stress on any

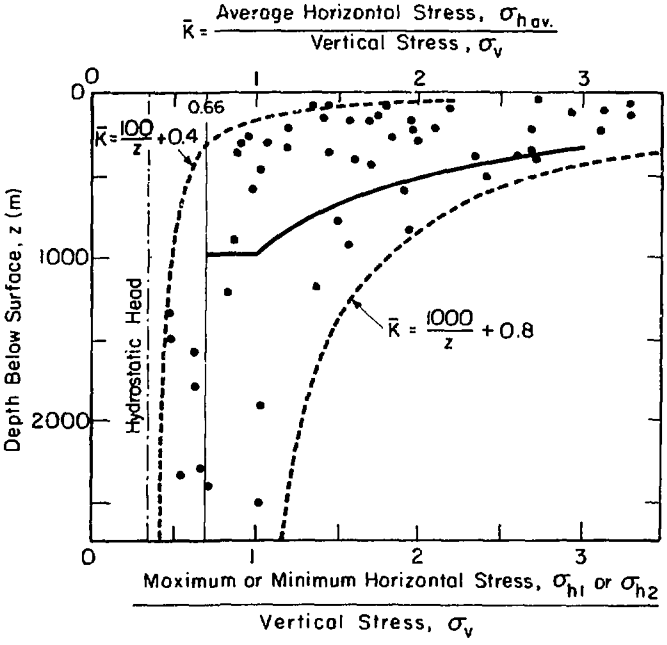

Figure 1: Compilation of measurements of the virgin state of stress in rock (Hoek and Brown, 1977)

rock surface. The effective stress is the normal stress less the hydrostatic pressure. To obviate the likelihood of faulting, especially in the presence of hydrostatic pressure, the difference between the values of the maximum and minimum components of the principal stresses should be small. Acceptable values for these components can be calculated in terms of Coulomb friction and effective stresses (Jaeger and Cook, 1976).

To preclude fault motion along existing discontinuities of any orientation, it is sufficient that:

$$(M - 1)p \leq [(\mu^2 + 1)^{1/2} + \mu]^2(L - 1)p, \tag{1}$$

where

p = hydrostatic pressure,

M = the ratio between the value of the maximum component of the virgin state of stress and the hydrostatic pressure at the same depth,

L = the ratio between the value of the minimum component of the virgin state of stress and the hydrostatic pressure at any depth,

μ = coefficient of sliding friction between rock surfaces.

This can be written as:

$$M \leq [(\mu^2 + 1)^{1/2} + \mu]^2(L - 1) + 1 = M', \tag{2}$$

and solved for M' in terms of ranges of values of L and μ. The results are given in Table 1.

Table 1: Values of M' (the ratio between the value of the maximum component of the virgin state of stress and the hydrostatic pressure at any depth) for different values of μ (the coefficient of friction) and L (the ratio between the value of the minimum component of the virgin state of stress and the hydrostatic pressure at the same depth)

μ	Values of M' for:				
	L = 1.25	L = 1.5	L = 1.75	L = 2.0	L = 2.25
0.2	1.37	1.74	2.12	2.49	2.86
0.4	1.55	2.09	2.64	3.18	3.73
0.8	2.08	3.16	4.25	5.33	6.41
1.0	2.46	3.91	5.37	6.83	8.29

If the vertical component is the maximum principal stress, the value of M is about 2.7. To preclude fault movement of a normal type, all combinations of μ and L giving values of M' less than 2.7 are not admissible; that is, the upper left portion of Table I.

As the value of μ may be as low as 0.4, the minimum value of L may be 1.78. This value of L corresponds to a ratio between the horizontal and vertical components of stress less than 0.66, if the vertical component is the maximum principal stress.

Because the coefficient of sliding friction may be greater than 0.4 high values of M' such as are shown in the lower right-hand-portion of this table may arise. These could permit high values of the maximum principal stress, without causing fault movement by frictional sliding along existing joints. However, at high values of M', movement could occur for other reasons. Laboratory tests on the strengths of intact rock (Jaeger and Cook, 1976) would suggest that this is unlikely, but size is known to have a pronounced effect on the strength of rock (Jaeger and Cook, 1976; Pratt et al., 1972), as are geological features. However, these effects are not understood well. Nevertheless, the data in Figure 1 may provide some indication of an upper limit to the maximum value of stress which can be sustained by near-surface rocks.

The upper bound to the value of the measured average horizontal virgin stress derived by Hoek and Brown (1977) can be expressed as:

$$\sigma_{hav} - \sigma_v = 25 - 0.005z, \tag{3}$$

where

σ_{hav} = the average value of the horizontal components of stress (MPa),

σ_v = the value of the vertical component, approximately 0.025z (MPa),

and

z = the depth below surface (m).

Equation (3) suggests that the maximum stress difference which rocks near the surface can sustain may be about 25 MPa. However, the definition of σ_{hav} as an average value introduces a degree of ambiguity into this interpretation. If the vertical component, σ_v, is the minimum principal stress, then $\sigma_{h1} \geq \sigma_{h2} \geq \sigma_v$, where σ_{h1} and σ_{h2} are the two horizontal principal stresses In this case, the stress difference given by equation (3) is exact for $\sigma_{h1} = \sigma_{h2}$. For $\sigma_{h2} = \sigma_v$ it becomes:

$$\frac{\sigma_{h1} + \sigma_v}{2} - \sigma_v = 25 - 0.005z \tag{4}$$

$$\sigma_{h1} - \sigma_v = 50 - 0.01z \tag{5}$$

so that the maximum stress difference, $\sigma_{h1} - \sigma_v$, is 50 MPa.

If σ_{h1} is the maximum principal stress and σ_{h2} is the minimum principal stress, $\sigma_{h1} \geq \sigma_v \geq \sigma_{h2}$ and equation (3) can be written as:

$$\frac{\sigma_{h1}}{2} - \sigma_v = 25 - 0.005z \qquad (6)$$

$$\sigma_{h1} - 2\sigma_v = 50 - 0.01z \qquad (7)$$

for the extreme case where $\sigma_{h2} = 0$, so that the maximum stress difference may be $\sigma_{h1} - \sigma_{h2} = 50 + 2\sigma_v$. Therefore, it seems reasonably safe to assume that the value of the maximum principal stress could not exceed the value of the minimum principal stress by more than 25 MPa.

The Strength of Rock around Excavations

Depending on their purpose, underground excavations can have many different configurations. Probably the most important considerations in designing the excavations for an underground repository of radioactive wastes is the safety, stability, and security of the excavations. In general, therefore, such excavations are likely to take the form of a series of adjacent, but more or less independent, tunnels. This results in simple, safe, excavations with a high degree of isolation between each tunnel.

Based on laboratory measurements of the strengths of small intact rock specimens and theoretical analyses of the stresses around tunnel-like excavations, rock failure would not appear to be a significant problem. However, it is generally accepted that such a simple approach does not accord with reality. It neglects at least two important factors; namely, the effects of size and geologic structure on the strength of the rock.

Size is thought to have a significant effect on the strength of geologic materials but there is a dearth of quantitative data on this question. Jaeger and Cook (1976) devote a chapter to this subject, discussing both experimental results and Weibull's statistical theory. Most of the experimental information that is available concerns more or less cubical specimens of coal. Evans and Pomeroy (1958) and Evans, Pomeroy and Berenbaum (1961) quote a wide range of crushing strengths for cubes of coal, the mean and modal values of which vary as

$$\sigma_c = Ka^{-d}, \qquad (8)$$

where

σ_c = the crushing strength,

K = a constant,

a = the side length of the cube,

d = an exponent with values between 0.17 and 0.32.

From a statistical analysis of case histories of pillars in coal mines, Salamon and Munro (1967) concluded that the strength of a pillar decreases inversely with size as its volume to the power 0.067, which accords well with the values for d given in equation (8) above. Data for hard rock are even more sparse than for coal. Pratt et al. (1972) obtained results, reproduced in Figure 2 for laboratory and in situ specimens of quartz diorite, showing a pronounced effect of size on strength. However, Obert et al. (1946) and Hodgson and Cook (1970) found size to have little effect on strength. Clearly, this is an important matter that cannot be settled now for want of sufficient data.

In practice, the behavior of rock around many excavations is determined by its structure and the presence of geological discontinuities (Hoek, 1977). However, little is to be gained in terms of a general, as distinct from a site-specific, attempt to evaluate this phenomenon. A worst-case analysis always results in rock failure and any less demanding theoretical assumptions, no matter how obscure they are, merely beg the question of specific data on the frequency, character, orientation, and properties of such discontinuities.

Nevertheless, it is necessary to form some idea of the magnitude of the effects of size and of geologic discontinuities on the strength of hard rock, in order to evaluate its potential as a location for an underground repository of radioactive wastes. Some guidance may be gained from an examination of the values of the field stresses known to have caused damage to tunnels in hard rock.

On the basis of observations in deep gold mines of the Witwatersrand System, Cook (1975) proposed that damage to tunnels, with a cross-section about 3 m square in argillaceous and arenaceous sediments with laboratory uniaxial compressive strengths in the range of 170 MPa to 340 MPa, begins at a value of the vertical (maximum) component of the field stress of 50 MPa and becomes dangerous at a value of about 100 MPa. Ortlepp et al. (1975) adduced observations of the onset of damage to similar tunnels totaling many hundreds of meters in length which may be interpreted as supporting this view.

In the absence of better information, it seems reasonably prudent to limit the value of the maximum principal stress at the site of any potential repository to somewhat less than 50 MPa, say, 25 MPa. This limitation is shown in Figure 1.

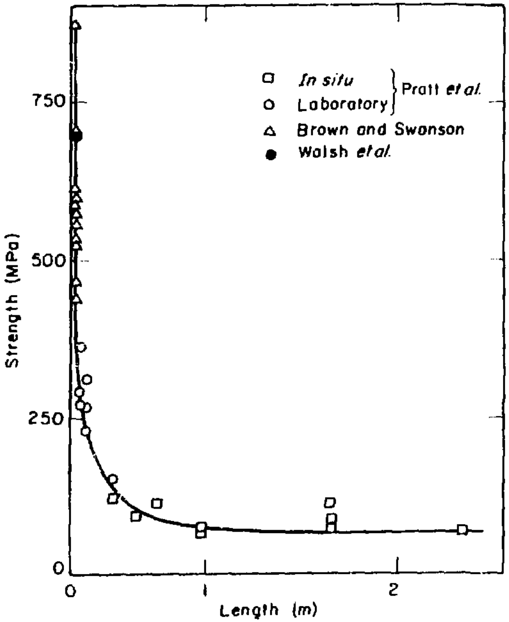

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Figure 2: A diagram illustrating the effect of size on the uniaxial compressive strength of quartz diorite (from Pratt et al., 1972).

III

IMPORTANCE OF HYDROGEOLOGY IN RADIOACTIVE WASTE ISOLATION

It is believed that the only likely means by which radioactive materials may escape from an underground waste repository to the biosphere is by transport with groundwater. Thus, an understanding of the hydrogeological factors that control groundwater movement is very important. The goal is to be able to select a site where the transport of the waste materials by the groundwater will be slow enough so that the eventual release of toxic substances to the biosphere does not constitute a hazard to life.

Permeability of a rock formation is one of the key factors that controls groundwater movement, and one would obviously prefer a repository site where the rocks possess an extremely low permeability. Salt is generally believed to be the most satisfactory rock formation from this standpoint because its permeability has been reported to be so low as to be essentially unmeasurable. In this case, the hydrogeological considerations discussed below are not a major concern and one needs to focus on other factors, such as the stability of salt at elevated temperatures. The discussion that follows is therefore only applicable to argillaceous and crystalline rocks.

Groundwater Movement

In considering the importance of hydrogeology in storing radioactive wastes in nonsaline rocks, we must consider two hydrologic regimes (a) the unsaturated zone and (b) the saturated zone. There have been some arguments proposed to support the concept of storing radioactive wastes in a repository above the water table, and thus in the unsaturated zone. The main rationale is that fluid movement would be insignificant because of the low percentage of water in the pore spaces of the rock.

It is well known among soil physicists that water ceases to move through the unsaturated zone as soon as the fluid saturation falls below some critical value referred to as the residual or immobile saturation. In a recent technical review, a National Research Council committee has concluded from an examination of field results at the Hanford Reservation that the available evidence supports the concept that a thick unsaturated zone in a semiarid or arid region can act as an effective barrier to the movement of radioactive nuclides into the biosphere (Panel on Hanford Wastes, 1978).

The critical problem that must be faced, however, is that when climatic changes increase annual rainfall or when extraordinarily heavy storms occur, water saturations can rise above the immobile value and the effectiveness of the barrier will be diminished or lost. Movement of groundwater through the area of the repository can then become a problem. This concept therefore requires that the climate remain such that the initial thickness of the unsaturated zone does not change over very long time periods.

There is also a problem involving depth of burial. Since the water table does not usually occur more than 600 m below the surface, even in arid zones, the waste repository will have to be a shallower depth to be located within the unsaturated zone. Thus, if an upward movement of the water table should take place because of changes in water saturations, the distance for radionuclides to reach the surface will only be a few hundred meters. The idea of placing a waste repository in the unsaturated zone therefore requires one to demonstrate that, regardless of future climatic conditions, the residence time for groundwater within this zone will be such that waste materials cannot reach the biosphere in hazardous amounts.

Since residence time and dilution are ultimately the governing factors in deciding where to locate an underground repository, one must also investigate the saturated zone where flow paths may be many kilometers in length. Some water migration will probably be unavoidable, but one needs to search for rock systems at depths where the velocities are low enough that the migration can be tolerated. Of course, during the construction and filling of the repository with waste material, seepage of water will be into the underground openings. However once the repository has been backfilled, groundwater will saturate the system and movement will again occur under the prevailing hydraulic gradients. There is no practical way to prevent this phenomenon. The problem therefore becomes one of selecting a repository site in terms of two key factors: (a) direction of groundwater movement, and (b) velocity of groundwater movement. A third factor, the quantity of water needed to carry radionuclides away from the repository, involves dilution and the sorption phenomena of the transported species. This will be discussed below in connection with the geochemistry of migration of radionuclides in groundwater systems.

The direction of groundwater movement varies both laterally and with depth throughout a groundwater basin. Water moves from the surface into underground layers in zones of recharge, which are the topographically higher elevations within the drainage basin. The direction of movement is essentially downward, and the depth of vertical penetration into the subsurface depends on many factors. Eventually the movement becomes more or less horizontal until a zone of discharge is reached. In a discharge area the flow lines may turn upward until the water is discharged at the surface. The distance between the point of recharge and the point of discharge can range widely, up to many tens of kilometers.

Figure 3 illustrates this concept using some simplified situations where the water-table configuration controls the location of recharge and discharge zones for water moving through a homogeneous isotropic media (Freeze and Witherspoon, 1967). Since the water table is often roughly parallel to the surface topography, we see how recharge and discharge zones differ in size depending on their position and the degree of topographic expression.

Another controlling factor is the degree of vertical inhomogeneity which would have an effect when the rocks are stratified into a sequence of layers of widely differing permeability. The more permeable layers will act as major channels for flow, and therefore, their areal extent within a basin also contributes to the location of recharge and discharge zones.

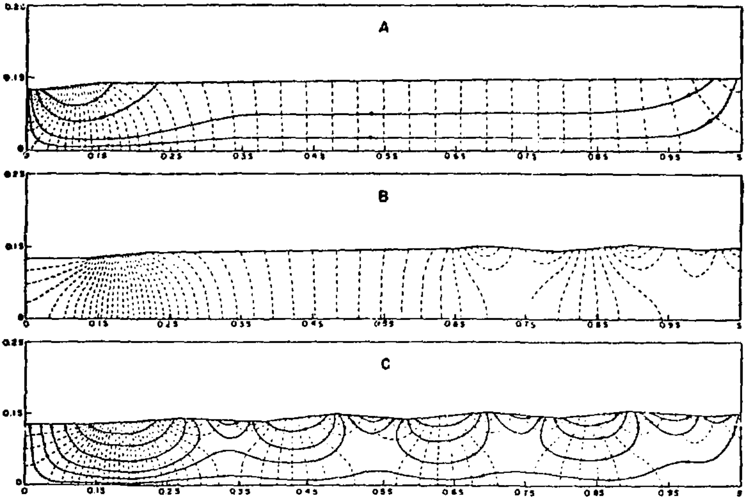

Figure 3: Effect of water-table configuration on regional groundwater flow through homogeneous isotropic media (after Freeze and Witherspoon, 1967).

Since the lengths of flow paths can vary considerably, it is important to understand the hydrogeology of a given basin in considerable detail in order to identify the complete flow field. Presumably, the optimum location for a waste repository will be in a recharge zone where the flowpath to the biosphere will be as long as possible. However, as recharge and discharge zones may change in the short term, one should look for deep geologic structures that favor long flowpaths, as these are not likely to be affected by changes in climate or topography. A very careful analysis of recharge-discharge relationships within the potential field will be needed in choosing this optimum location. The technology required for gathering the necessary field measurements of hydraulic potential to make this choice is available, and the technical problem at present is to adapt available methods and equipment to work at depths of several kilometers in fractured rock systems that are nearly impermeable.

Thermal Convection

An additional complication may be created by the temperature rise in the rocks surrounding the waste repository. The temperature increase is caused by the energy output of the waste, and its magnitude depends, among other things, on the age of the waste. An energy output of 5 kW per canister is the highest value that has been considered for waste material, and this decays rapidly to about 1 kW after some ten years.

Experience with geothermal systems (Donaldson, 1968) suggests that in fractured rocks of very small aperture, the Rayleigh number is so large that a local heat source is unlikely to cause natural circulatory convection. To establish throughflow due to a buoyancy unbalance requires recharge from the surrounding rock mass that is filled with cold groundwater. If the repository is placed in a system of nearly impermeable rock then the resistance to groundwater flow will mean that the low velocities of the regional system will control overall fluid movement. Vertical throughflow will therefore be restricted by the magnitude of available inflows from the regional system.

Groundwater Velocity

The second key factor, that of determining the velocity of groundwater flow, is a much more complex problem. The basic flow equation can be expressed in simplified form by

$$v = K \Delta h / \phi \tag{9}$$

where:

v = effective velocity, [m.sec]

K = hydraulic conductivity, [m.sec]

Δh = hydraulic gradient, [$m.m^{-1}$]

ϕ = porosity.

The problem of applying this equation to a specific groundwater system is that K can vary over a considerable range from point to point. For example, in passing from the surface to depths of thousands of meters, values of hydraulic conductivity can range over ten orders of magnitude. For underground waste storage, one obviously wants to use a rock mass with a very low value for the hydraulic conductivity. Thus, salt has been considered to be the most satisfactory rock because of its very low intrinsic permeability.

In the case of argillaceous and crystalline rocks, the hydraulic conductivity of the undisturbed matrix can also be very low. For example, shale caprocks over underground gas storage projects are routinely cored, and the measured hydraulic conductivities of the matrix are of the order of 1×10^{-13} to 1×10^{-14} $m.sec^{-1}$.* Essentially the same low magnitude permeabilities have been reported by Marine (1974) for claystones at the Savannah River Plant and by Brace (1977) for the Westerly granite.

It is a simple matter to show that rocks with hydraulic conductivities of this low magnitude provide effective barriers to migration. If we consider a hydraulic conductivity of 1×10^{-13} $m.sec^{-1}$ and assume a hydraulic gradient of 0.001 $m.m^{-1}$, which is a reasonable value, and a porositv of one percent, then equation (1) yields:

$$v = \frac{(1 \times 10^{-13})(0.001)}{0.01} = 1 \times 10^{-14} \text{ m.sec}^{-1}$$

This is equivalent to 3×10^{-10} kilometers per year, which is of no consequence even over time periods in the millions of years. Movement by diffusion would lead to even lower velocities.

In attempting to site an underground waste repository in such rocks, however, it is very likely that one will not be able to avoid discontinuities of some kind. Major features such as faults and shear zones can, of course, be avoided through careful geologic mapping, but minor fractures such as joints and fissures are pervasive features. This is especially true for crystalline rocks such as basalt and granite. It should be kept in mind, however, that our cumulative experience is derived from underground workings excavated without regard for the specific purpose of locating a nearly impermeable rock mass well below the earth's surface. The need for such a special rock condition has only arisen because of the underground radioactive waste problem.

Where fractures exist as discontinuities in an essentially impermeable rock matrix, the overall permeability will be determined by the properties of the fracture network. This can be demonstrated by considering the movement

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* A hydraulic conductivity of 1×10^{-13} $m.sec^{-1}$ converts to a permeability of 1.035×10^{-8} Darcys if water is flowing at ambient conditions.

of water through a single fracture. For laminar flow between parallel plates, it can be shown that hydraulic conductivity may be expressed by:

$$K = \frac{b^2 \rho g}{12\mu} , \qquad (10)$$

where:

b = fracture aperture, [m]

ρ = density of water, [kg.m^{-3}]

g = acceleration of gravity, [m.sec^{-2}]

and

μ = viscosity of water, [kg.m sec]

Table 2 illustrates the velocities and hydraulic conductivities that result from equations (9) and (10), using a range of apertures and assuming water at ambient conditions under a hydraulic gradient of 0.001 m.m^{-1}. By comparing these results with those cited above for an intact rock, it is apparent that water velocities in fractures will easily dominate those in the rock matrix, even when apertures are as small as 0.1 micron. It should be noted that Hadley (1976) has measured microcracks in intact Westerly granite that range in aperture from 0.01 to 10.0 microns and in length from 0.075 to 250 microns. The discontinuous nature of these microcracks explains why the measured hydraulic conductivity of the Westerly granite at 50 mPa is only 6×10^{-13} m/sec (Brace, 1977).

Table 2. Hydraulic conductivity and velocity for water flowing through a single fracture

Aperture b microns	Hydraulic Conductivity, K m.sec^{-1}	Effective velocity, v m.sec^{-1}	Effective velocity, v km.year^{-1}
0.1	8.2×10^{-9}	8.2×10^{-12}	2.6×10^{-7}
1.0	8.2×10^{-7}	8.2×10^{-10}	2.6×10^{-5}
10.0	8.2×10^{-5}	8.2×10^{-8}	2.6×10^{-3}

Although equation (10) was derived for parallel plates that are not in contact, Iwai (1976) has shown that equation (10) also holds for man-made fractures in basalt, granite, and marble under normal stresses ranging from 0 to 20 MPa. It is important to note that, at the maximum applied stress, the fractures could not be completely closed. Measured flow rates at maximum stress indicated that the effective aperture could not be reduced below about

15 microns. This is probably due to imperfect matching of opposing fracture surfaces. As fracture aperture decreases with increasing normal stress, hydraulic conductivity of fractured rock will generally decrease with depth below the surface.

Clearly, the optimum situation is a massive body of dense rock with a minimum of fractures subjected to near-lithostatic compressive stress. It is necessary to know how low the hydraulic conductivity of an undisturbed fracture can become as stress increases. One can anticipate that the minimum value will be approached asymptotically as stress increases, (Witherspoon and Gale). The stress level at which this value is first approached is therefore an important design parameter.

IV

GEOCHEMICAL CONSIDERATIONS

Scientists and engineers of several nations are actively considering the problem of long-term isolation of radioactive waste by deep burial in low permeability rocks. For various hydrological reasons cited above, the storage site will most likely be located beneath the water table. The most important aspect thereafter will be adequate containment of the radionuclides until such time as they are rendered harmless through radioactive decay. Upper limits of greater than five million years have been cited, but there is some question whether it will be possible to predict containment over such long time spans (Bredehoeft et al., 1978; Winograd, 1977). For the purpose of discussion, a one-million-year containment period is considered as a target in this section.

After wastes are buried and groundwater percolates back into the excavated regions, the radioactive waste will be subject to leaching and transport by the groundwater to the biosphere. There are four barriers to inhibit or minimize the leakage of radionuclides:

(1) an impervious and corrosion-resistant container,

(2) a waste product form highly resistant to leaching,

(3) a chemically sorbent, impermeable backfill, and

(4) a chemically sorbent and low-permeability host rock.

Arguments in support of any one or all of these four barriers have been advanced. However, a critical assessment is needed to determine what criteria will be needed to define the best barriers for a given repository, and whether or not all are required. The purpose of this section is to outline the problems pertinent to each barrier, and to determine what questions must be answered in evaluating the suitability of a storage system for long-term isolation of radionuclides from the biosphere.

Some of the questions needing resolution, for example, are: What chemical containment barriers should be emphasized? How many barriers are needed? How do we quantify their effectiveness in retarding radionuclide transport? Does the choice of one barrier affect the behavior of other barriers? To what extent do site-specific factors influence our choice of barrier? To what extent do waste pretreatment options influence our choice? Are we able to verify experimentally within a reasonable length of time that the barriers will be effective over a million years or more? A consideration regarding the effectiveness of all barriers is the ultimate concentration of the radionuclide as it enters the biosphere and subsequent concentration to toxic levels in the food chain. Careful examination of these questions is required, for a great deal is involved both with regard to the cost of a storage repository and to the proper direction of research required to resolve current uncertainties. In the following paragraphs, each barrier to migration will be considered and the potential problems identified.

Container Integrity

Container integrity is the first line of defense against the release of radionuclides. The durability of the container depends, in large measure, on its ability to resist corrosion or radiation damage under the chemical conditions found either at its interior or exterior surfaces.

Two suggestions have been made for the choice of container material, emphasizing either materials extremely resistant to chemical attack, such as fused alumina, corundum or other ceramics; or those which may be in chemical equilibrium with the subsurface environment, such as copper metal (Fyfe, 1977).

In the case of corundum, we can assume that chemical reactions proceed inwards from the exterior due to chemical attack by the groundwater. The approximate isolation period of the waste can be calculated, assuming a given dissolution rate. Reaction presumably occurs through dissolution and subsequent precipitation of secondary clay minerals, such as kaolinite or montmorillinite. Laboratory studies have demonstrated that leaching rates of corundum are very slow under near neutral pH's and near-ambient temperatures, on the order of 10^{-11} $kg.m^{-2}.sec^{-1}$. A container of corundum 0.1 m thick would take approximately 10^6 years to leach. However, intergranular attack, radiation damage, and higher temperatures may appreciably shorten container life.

The ability of copper to serve as an effective barrier to leaching over a long time period is less easily ascertained. Figure 4 is an Eh-pH diagram of the system Cu-S-C-H_2O at 25°C and one atmosphere. Under certain conditions of pH and Eh typical of some groundwaters, copper is stable. Changes in groundwater composition may affect the stability of the copper. For example, the presence of ammonium or carbonate ions may significantly increase copper solubility, and hydrogen sulfide may alter the copper to sulfide with prefeential attack along grain boundaries.

Other substances may also serve a similar role in isolating radioactive waste from the groundwater over long time spans. However, we have considered only their resistance to corrosion by groundwater. Many questions remain unanswered regarding their physical integrity, particularly the role of other physical-chemical mechanisms of degradation, such as recrystallization, chemical attack from interaction with the waste, or radiation damage. The problem of guaranteeing container integrity will require careful consideration. Specialized expertise and protracted research will be needed before there will be adequate assurance that barrier containers will contain radioactive waste for up to 10^6 years. Not only do the mechanisms of container failure have

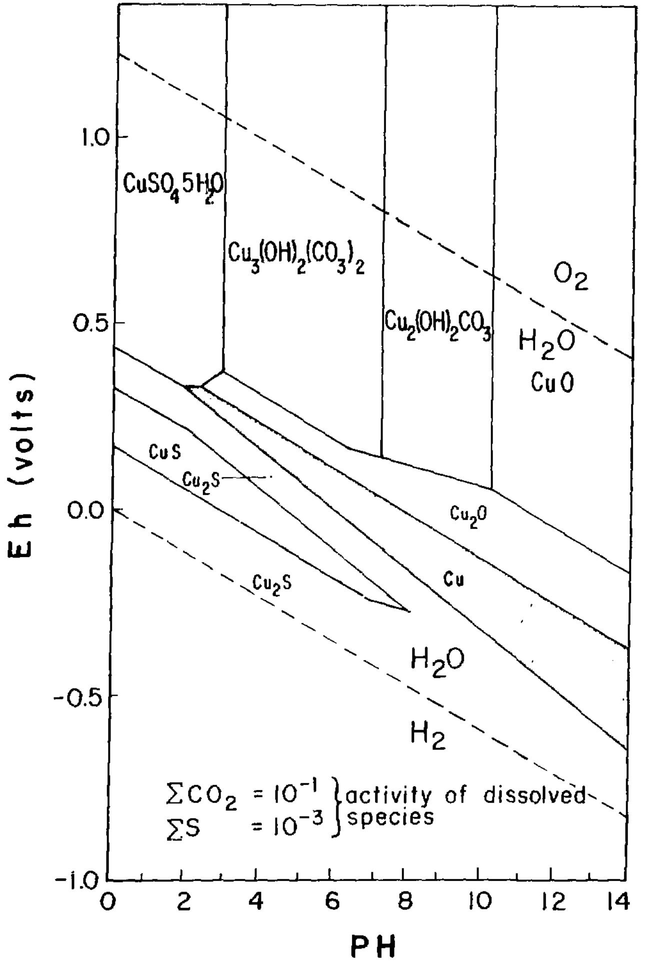

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Figure 4: Eh-pH diagram of the system Cu-S-C-O-H at 25°C and one atmosphere illustrating the stability field of copper metal (after B. B. Hanshaw, 1962).

to be identified, but quantitative predictions made on the basis of experimental observations must remain valid for up to 10^6 years. This is extremely difficult where there are no historical records to provide guidance for long range effects, other than the geological record.

Brief mention should be made of the physical integrity of the container. Ceramics are far more prone to breakage than ductile materials such as copper. Therefore, if ceramics are to be used, some means of preventing stress buildup about the container due to ground movement must be incorporated in the overall design of the repository. Otherwise premature release of the radionuclides may occur.

Leaching Resistance of the Waste Product

The second line of defense involves minimizing the leachability of the waste product. This approach is also attractive because ground movements are unlikely to enhance significantly the leaching rate of the waste other than through incremental increases in the exposed surface area caused by fracture of the waste itself. The waste product can be designed to resist leaching through minimization of surface area; such as through the preparation of glass; or through crystallization of leach resistant phases, such as crystalline silicates. The idea of producing specifically tailored, highly insoluble phases in which actinides or other radionuclides might substitute for major components (e.g., sphene) has been considered (Smyth, Vidale, and Charles, 1977).

The waste to be stored can be any of a number of substances. Currently, the most important are high-level wastes due to reprocessing such as borosilicate glass, super-calcine, and zeolites, and SURF (spent unreprocessed fuel). The nature of the waste depends on the reactor fuel cycle used, whether or not it has been processed to recover uranium and/or plutonium, and subsequent treatment to produce a solid phase. Because of the current uncertainties regarding reprocessing, the need to store SURF must be considered of prime importance.

The most important question concerning the behavior of any waste product is the rate at which radionuclides will be leached and transported away in the groundwater. The leaching mechanism will depend upon the chemical reactions that proceed between the groundwater and the waste, and between components within the waste. Figure 5 illustrates, conceptually, the vicinity of the waste canister and the kind of reactions that could occur. Table 3 summarizes the reactions and their possible significance.

With so many variables to consider; i.e., waste type and composition, backfill material, host rock environment, groundwater composition, physical configuration, time and temperature dependent factors, no simple answers are currently available that will determine and quantify the rate-controlling mechanisms.

Table 3: Waste leaching mechanisms*

A.	WASTE	SIGNIFICANCE
1.	Recrystallization Devitrification	Probably small unless catalyzed by groundwater and the temperature is high enough. Could increase or decrease solubility.
2.	Solid state diffusion	May be important.
3.	Radiation damage	May increase solubility.
B.	WASTE-GROUNDWATER	
1.	Congruent dissolution	Small - most waste materials are of complex composition.
2.	Incongruent dissolution with buildup of leached product layer.	Very possible.
3.	Incongruent dissolution with solid state diffusion	Possible.
4.	Precipitation of reactant from the groundwater.	Very possible.
C.	DISSOLVED NUCLIDE MIGRATION	
1.	Diffusion in groundwater	$J_d = -D \frac{\partial C^{\ell}_{M_i}}{\partial x} \rho_{\ell} \frac{\phi}{\tau}$ [g mol.m^{-2}.sec^{-1}]
2.	Advection by groundwater	$J_{ad} = C^{\ell}_{M_i} v_{\ell} \rho_{\ell} \phi$ [g mol.m^{-2}.sec^{-1}]
		$J_d \approx J_{ad}$ when $v_{\ell} < 10^{-8}$ m.sec^{-1}
D.	REACTION OF DISSOLVED PRODUCTS WITH BACKFILL OR ROCKS	
1.	Adsorption	Probable, depending on nuclide.
2.	Ion exchange (with solid state diffusion)	Probable.
3.	Precipitation of discrete phases	?

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* Symbols used in equations are given on page 23.

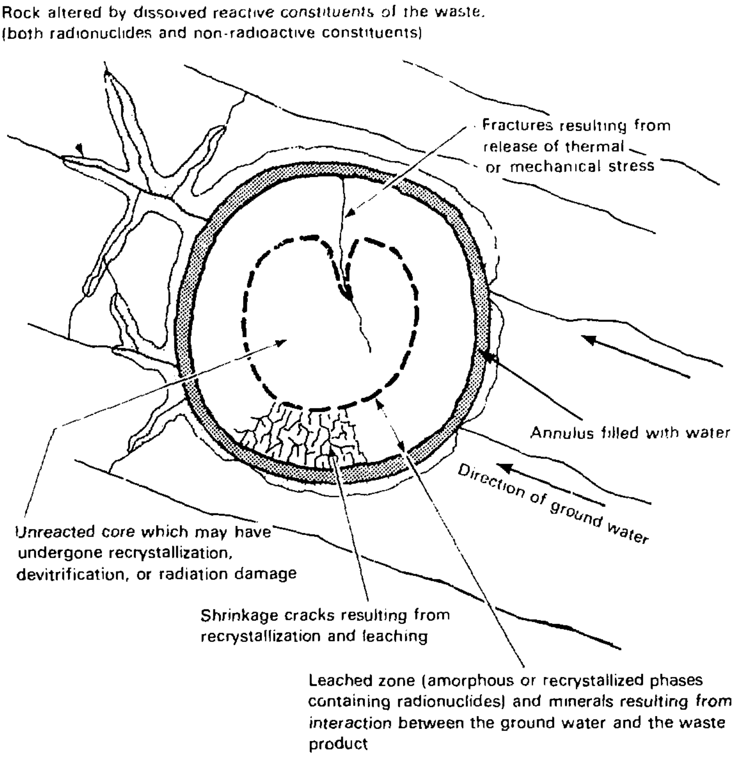

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Figure 5: Schematic diagram illustrating a hypothetical high-level waste product of cylindrical form, emplaced in a storage well.

$C^{\ell}_{M_i}$ = concentration of radionuclide M_i [g mol.kg^{-1}]

D = diffusion coefficient of radionuclide in solution [m^2.sec^{-1}]

J_d, J_{ad} = mass flow of radionuclide due to diffusion and advection, respectively [g mol.m^{-2}.sec^{-1}]

v_ℓ = average velocity of the ground water [m.sec^{-1}]

x = position coordinate measured perpendicular to the reference plane [m]

ρ_ℓ = density of ground water [kg.m^{-3}]

τ = porosity parameter [-]

ϕ = porosity of rock fractures [-]

The leaching rate for any given nuclide is a function of (a) the surface leaching rate, (b) the diffusion of the nuclide away through the surrounding water, water saturated backfill material, and/or product layer, (c) the rate of radioactive decay, and (d) transport from the site by groundwater migration through the fractured rock by advective and/or convective processes. If groundwater flow is less than 1 m.yr^{-1}, then removal of the radionuclide from the waste will occur through a combination of diffusion and ground-water transport. Diffusion may be accelerated if adsorption of the radionuclide occurs on adjacent rock mineral or grout surfaces. Stable isotopes of some elements may also be present in the ground water in significant amounts (e g., strontium and cesium) thereby complicating the problem. Determination of the rate controlling step(s) in this complex system must be made if leaching rates over long time spans are to be predicted.

It is not possible to place any limits on the rate of leaching of radionuclides over long time periods because we do not yet know enough about the kinetics of the various processes. Much of the current evidence supports low leaching rates, and hence low ultimate rates of removal of the radionuclides from the storage site. If we assume that the solubility of any given radionuclide is determined by some product phase resulting from the recrystallization of the waste, and that the radionuclide diffuses into the surrounding groundwater and is removed by advection, then we can set up a simple model to predict the approximate leaching rate and time to transport a given radionuclide away from the storage site.

Assuming that near-saturation of a radionuclide occurs rapidly in the groundwater adjacent to the waste when compared to the movement of groundwater through the storage site, then the rate at which the radionuclide is transported away from the site, the radionuclide mass flux, is a function of the saturation concentration of the nuclide and the groundwater movement through the site. The duration of leaching will be a simple function of the groundwater flow, the saturation concentration of the nuclide, the quantity initially present, and the decay constant of the radionuclide under question.

Thus, the mass of radionuclide, M_i, leached in time, t, is:

$$M_{M_i}(t),\ \text{leached} = C^{\ell}_{M_i} v_{\ell} \phi A_r \rho_{\ell} t\ , \qquad [\text{g mol}] \qquad (11)$$

where

A_r = cross sectional area of rock containing water saturated with the radionuclide, [m^2]

t = time since start of leaching. [yr]

v_ℓ = average velocity of groundwater [$m.yr^{-1}$]

The amount of radionuclide that decays during time, t, is:

$$M_{M_i}(t),\ \text{decayed} = M_{M_i}(t)[1 - e^{-\lambda t}], \qquad [\text{g mol}] \qquad (12)$$

where

$M_{M_i}(t)$ = mass of radionuclide not leached from the waste, [g mol]

λ = decay constant of the radionuclide present. [yr^{-1}]

For simplicity, let us assume that leaching starts soon after storage. Then at any time t,

$$M_{M_i}(t) = M_{M_i}(0) - M_{M_i}(t),\ \text{leached} - M_{M_i}(t),\ \text{decayed}$$

$$= M_{M_i}(0) - C^{\ell}_{M_i} v_{\ell} \phi A_r \rho_{\ell} t - M_{M_i}(t)[1 - e^{-\lambda t}]. \qquad (13)$$

where

$M_{M_i}(0)$ = quantity of radionuclide initially present. [g mol]

When all the radionuclide has been leached and decayed,

$$M_{M_i}(0) - C^{\ell}_{M_i} v_{\ell} \phi A_r \rho_{\ell} t - M_{M_i}(t)[1 - e^{-\lambda t}] = 0. \qquad (14)$$

As an example, let us consider the leaching of Plutonium - 239 in SURF from waste canisters that are 10 ft long and 12 inches in diameter. The quantity of plutonium present is ≈ 1 wt percent. Hence, the mass, $M_{M_i}(0)$, is 22.2 kg.

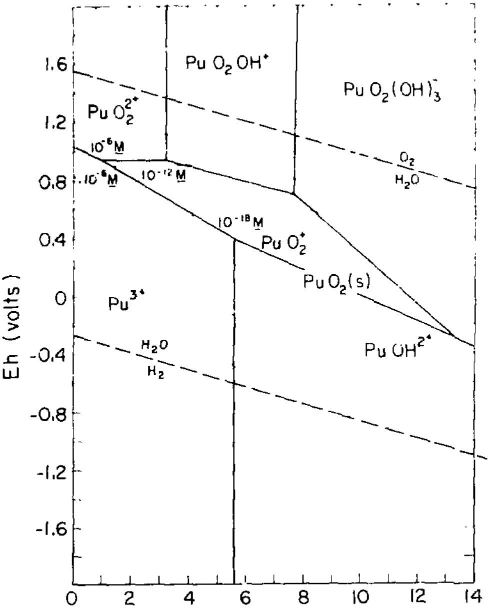

Figure 6: Eh-pH diagram at 25°C and one atmosphere showing the stability fields of hydroxyl complexes of plutonium, and plutonium oxide, $PuO_2(S)$ (Apps et al., 1977).

The approximate concentration of plutonium in solution, $C^{\ell}_{M_i}$, can be estimated from inspection of an Eh-pH diagram as shown in Figure 6 where the stability fields of plutonium oxide and various hydrolyzed species are given. Assuming PuO_2 is the stable solid phase and that the oxidation state and pH of the groundwaters are 0-400 mV S.H.E. and 6-8, respectively, we can see that the dominant hydrolyzed species in solution is $Pu(OH)^{2+}$, that Pu is in the III state, and that the solubility of PuO_2 is between 10^{-12} and 10^{-18} g mol.kg^{-1}. This diagram ignores the presence of other, possibly more stable, phases that would tend to decrease the concentration of plutonium in solution, or complexing that would tend to increase the concentration of plutonium in solution. Thus, we have the following values:

$A_r = 10,$

$M_{M_i}(0) = 93,$

$C^{\ell}_{M_i} = 10^{-12},$

$v_{\ell} = 10^{-2},$

$\lambda = 2.842 \times 10^{-5},$

$\phi = 0.05,$

$\rho_{\ell} = 10^{3}.$

Equation 14 may be solved graphically as illustrated in Figure 7. Leaching of plutonium would take place for 610,000 years.

The range of concentrations of nuclides in solution from leaching waste can be expected to fall between 10^{-3} and 10^{-12} g mol.kg^{-1} of groundwater and the average groundwater velocity through the site can be expected to fall between 10^{-3} to 10 m.yr^{-1}. This leads to a total mass flux of between 5×10^{-1} and 5×10^{-14} g mol.m^{-2}.yr^{-1}. If a SURF waste canister were to contain 1 wt percent of a very long-lived nuclide of average molecular weight (say 100), complete leaching would occur in between 4.45×10^{0} and 4.45×10^{14} years. It is quite evident from these values, that the rate of decay of the radionuclide will largely control the duration of leaching if the mass flux is less than 10^{-6} g mol.m^{-2}.yr^{-1}.

More refined models are needed to define the leaching rate during the initial phases of leaching. In particular, the rate-controlling step must be identified, and the expected maximum concentration attainable in solution. This in turn will determine the mass flux of the radionuclide that could be affected by subsequent barriers.

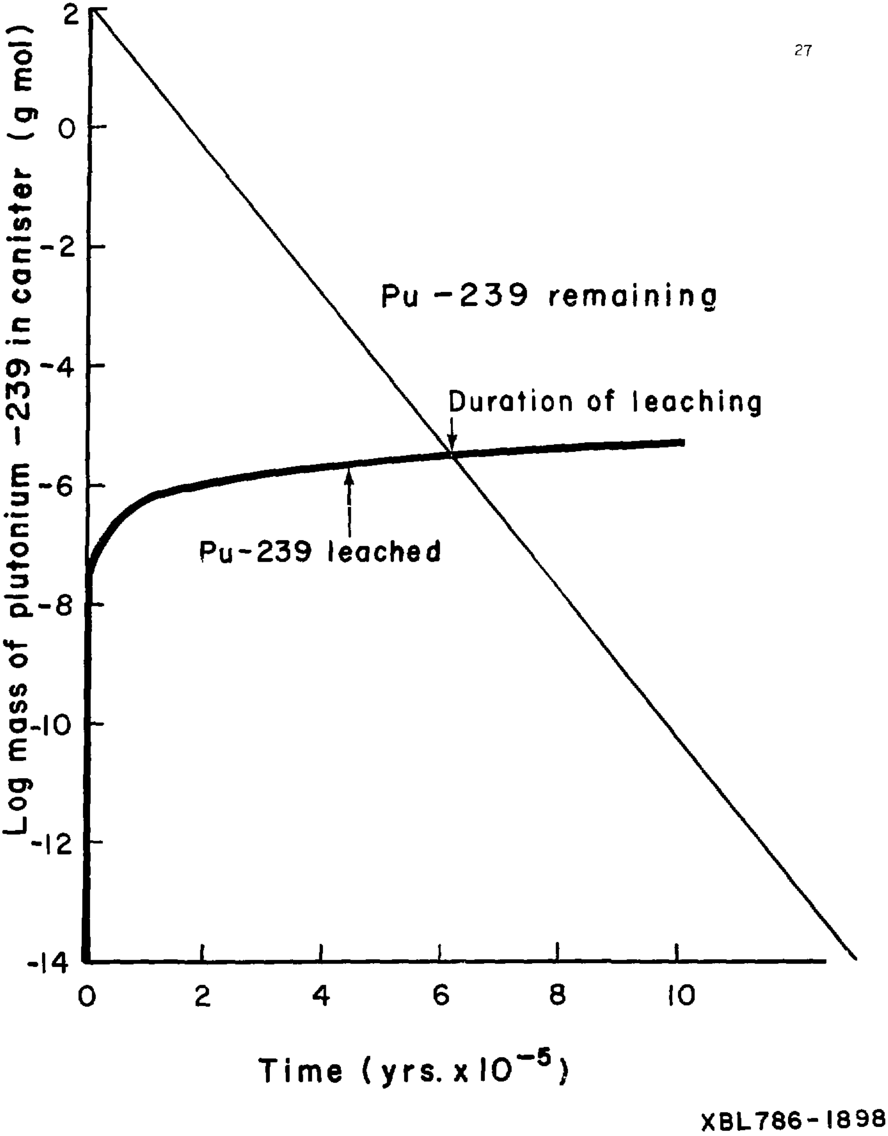

Figure 7: An approximate estimate of the time to leach plutonium from a waste container.

Chemically Sorbent Backfill

The third line of defense is a backfill material or grout which modifies the chemical composition of the invading groundwater, reacts with and sorbs leached radionuclides, and reduces still further the permeability of the local storage area. Various backfill materials have been proposed, including cements, clay, sulfides, and serpentinite. All can act as chemical sorbents upon which migrating radionuclides would be fixed, hopefully decreasing the concentration in the groundwater to imperceptible levels. Deformable backfill materials such as clays and serpentinite possess the advantage that they are not subject to fracture under stress. These could be extruded into surrounding fractures under hydraulic pressure, thus effecting an impervious seal where the repository host rocks would be most subject to increased breakage from excavation-induced stress. A deformable backfill material could also relieve directional stress around a canister, thereby decreasing the possibility of rupture.

The design of a suitable backfill material first requires an understanding of the role it is to play. If it is to be designed primarily to retard radionuclide migration, the radionuclides requiring special attention must be identified and their solubility and speciation in groundwater determined. The backfill material should be then designed specifically to maximize sorption of the species in solution. This could be done by ensuring a large surface area, many surface active sites, and special treatment to enhance adsorption. The backfill may also be designed to be chemically stable with respect to the canister material, or to react with water to provide an anhydrous environment. Such backfill materials would prevent corrosion of the exterior of the canister, but would be no defense against corrosion by the waste.

Host Rock as a Chemical Barrier

The host rock presents the final barrier against the leakage of radioactive waste to the biosphere. Radionuclides can migrate through the host rock either by transport in, or by diffusion through the groundwater filling the rock pores and fractures. Where there is pore or fracture continuity groundwater will migrate if subject to a hydraulic gradient. During migration, a radionuclide species will be subject to chemical reactions with the host rock minerals. Depending on the extent to which chemical reactions proceed, radionuclide migration may be retarded with respect to the groundwater flow.

Predicting the migration rate of radionuclides in the groundwater presents formidable difficulties because it is very difficult to perform field tests to measure radionuclide migration in low permeability rocks, and it is difficult to simulate in situ conditions in the laboratory. Predictions will depend largely upon the ability to extrapolate laboratory data to in situ conditions. The problems inherent in predicting radionuclide migration are examined in greater detail in the following two sections. The first problem is to define the chemical reactions that may occur between the radionuclide and the host rock, and the relationship of these reactions to radionuclide retardation with respect to the groundwater movement. The second is to define the influence that the host rock fabric, i.e., fracture geometry, effective porosity, and surface area have on the chemical reactions and groundwater movement.

a. Chemical Reactions with Host Rocks

Let us first relate sorption chemistry of a radionuclide with groundwater movement and the effect that sorption chemistry has on radionuclide migration rates. Then we will be in a position to draw inferences regarding the effect of sorption on migration, and the validity of using distribution coefficient, or K_d measurements in the modeling of radionuclide migration.

Let us consider a unit cubic meter of fractured rock through which groundwater can transport radionuclides:

The mass of rock = $\rho_s(1 - \phi)$, [$kg.m^{-3}$ rock]

The mass of fluid = $\rho_\ell \phi$, [$kg.m^{-3}$ rock]

where

ρ_s, ρ_ℓ = densities of solid (rock) and liquid (water), respectively, [$kg.m^{-3}$]

and

ϕ = porosity.

Following Giddings (1965), if R is the fraction of nuclide in solution at equilibrium,

Mass in solution = $M_{M_i} R$, [g mol.m^{-3} rock]

Mass sorbed = $M_{M_i}(1 - R)$. [g mol.m^{-3} rock]

The transported radionuclide reacts with the rock according to some unspecified chemical reaction. The reaction could involve adsorption, ion exchange, precipitation or coprecipitation involving any of the solid phases or aqueous species present. A generalized form of the reaction, valid for all mechanisms, and normalized to one unit of species M_i^ℓ, is as follows:

$$M_i^\ell + \sum_{j=1}^{n} \nu_{j,R} M_{j,R} = M_i^s + \sum_{j=1}^{n} \nu_{j,P} M_{j,P} , \quad (j \neq i) \tag{15}$$

At equilibrium,

$$K_i = \frac{[M_i]^s \prod_{j=1}^{n} [M_{j,P}]^{\nu_{j,P}}}{[M_i^\ell] \prod_{j=1}^{n} [M_{j,R}]^{\nu_{j,R}}}, \qquad [-] \qquad (16)$$

where [ ] denotes activity, ν is the stoichiometric coefficient, and the subscripts, R and P, denote reactants and products, respectively. Now,

$$[M_i^\ell] = C_{M_i}^\ell \gamma_{M_i}^\ell, \qquad [\text{g mol.kg}^{-1} \text{ liquid}] \quad (17)$$

$$[M_i^s] = C_{M_i}^s \gamma_{M_i}^s, \qquad [\text{g mol.kg}^{-1} \text{ solid}] \quad (18)$$

$$M_{M_i} R = C_{M_i}^\ell \rho_\ell \phi, \qquad [\text{g mol.m}^{-3} \text{ rock}] \quad (19)$$

$$M_{M_i}(1 - R) = C_{M_i}^\ell \rho_s (1 - \phi), \qquad [\text{g mol.m}^{-3} \text{ rock}] \quad (20)$$

where $C_{M_i}^\ell$ is in g mol.kg^{-1} liquid, $C_{M_i}^s$ is in g mol.kg^{-1} solid, and $\gamma_{M_i}^\ell$ and $\gamma_{M_i}^s$ are stoichiometric activity coefficients of the species M_i in the liquid and solid phases, respectively.

Rearranging (19) and (20) and substituting in (16), we get:

$$\frac{(1 - R)}{(R)} = \frac{K_i \rho_s (1 - \phi) \gamma_{M_i}^\ell \prod_{j=1}^{n} [M_{j,R}]^{\nu_{j,R}}}{\rho_\ell \phi \gamma_{M_i}^s \prod_{j=1}^{n} [M_{j,P}]^{\nu_{j,P}}} . [-] \qquad (21)$$

By substituting (16) in (21), rearranging, and collecting terms, we get

$$\frac{1}{R} = 1 + \frac{C^{s}_{M_i}}{C^{\ell}_{M_i}} \; \frac{(1 - \phi)}{\phi} \; \frac{\rho_s}{\rho_\ell} \, . \qquad [-] \qquad (22)$$

By defining a distribution coefficient, K_d, as

$$\frac{C^{s}_{M_i}(1 - \phi)}{C^{\ell}_{M_i}\rho_\ell} = K_d \, , \qquad [m^3.kg^{-1}] \qquad (23)$$

equation (22) reduces to:

$$\frac{1}{R} = 1 + K_d \frac{\rho_s}{\phi} \qquad [-] \qquad (24)$$

It is commonly believed that radionuclides are adsorbed on the surfaces of minerals by a surface ion-exchange process. If this assumption is true, then the sorption reaction is a function of both the fracture surface area and the pore surface area of the rock effectively in contact with the mobile groundwater.

Let us modify equation (16) to take into account surface adsorption processes.

$$[M_i^{ss}] = \frac{A}{N} \sum_{\alpha = 1}^{\xi} \omega_{\alpha,i}\sigma_{\alpha,i} \, , \qquad [g\ mol.kg^{-1}\ solid] \quad (25)$$

where

A = the internal surface area of the rock, $[m^2.kg^{-1}]$

N = Avogadro's number, $[g\ mol^{-1}]$

$\sigma_{\alpha,i}$ = surface site density of species i, on mineral α, $[m^{-2}]$

$\omega_{\alpha,i}$ = fraction of mineral, α, exposed. $[-]$

Substituting equation (25) into equation (22), we get:

$$\frac{1}{R} = 1 + \frac{A}{N} \frac{\sum_{i=1}^{r} \omega_{\alpha,i}\sigma_{\alpha,i}}{C^{\ell}_{M_i}} \frac{(1 - \phi)}{\phi} \frac{\rho_s}{\rho_\ell} , \qquad [-] \qquad (26)$$

where from equation (23)

$$K_d = \frac{A}{N} \frac{\sum_{i=1}^{r} \omega_{\alpha,i}\sigma_{\alpha,i}}{C^{\ell}_{M_i}} \frac{(1 - \phi)}{\rho_\ell} . \qquad [m^3.kg^{-1}] \qquad (27)$$

Moving groundwater will transport a radionuclide molecule only when it is present in the aqueous phase. If the molecules of radionuclide in the groundwater are in reversible equilibrium with those that are sorbed, then they will migrate only for the fraction of time they are in the aqueous phase or will move at a fraction ~R relative to the velocity of the groundwater. Therefore R is also equivalent to the relative migration rate of the radionuclide compared with the groundwater. Hence equations (24) and (26) relate the migration rate of the radionuclide to groundwater with the distribution coefficient, K_d.

The interesting conclusion which can be drawn here is that, for a given set of conditions, the migration rate of sorbed species with respect to the solvent, depends only upon the distribution coefficient of that species between the solid and the solvent at equilibrium. Unfortunately, the conditions under which K_d values have been determined experimentally often differ radically from those of the groundwater environment. Most K_d measurements have been made with soils using beaker or column tests. A comparison between the laboratory test conditions and those conditions anticipated adjacent to a subsurface storage repository is given in Table 4, together with estimates of the potential effect that these differences might have on K_d values.

Table 4: Factors influencing K_d for a given radionuclide, based on a comparison between conditions in a typical soil column adsorption study and conditions expected in a terminal storage repository

Parameter	Principal effect	Soil column test	Subsurface terminal storage facility	Potential effect on K_d
1. **Solution Chemistry**				
a. Major components	Ionic strength Activity coefficients Complexing	Very variable. Composition determined by conditions the test is designed to simulate	Determined by host rock chemistry and by other factors including the leaching chemistry of the waste product.	Unpredictable - Probably 10^{-3} to 10^{+3}.
b. Minor components	Complexing	Same as above.	Same as above.	
c. pH	Complexing Chemical potential	2-11, depending on the nature of the test	5-8 buffering of heterogeneous and homogeneous equilibria keep the pH range within narrow limits.	10^{-5} to 10^{+5}
d. Eh	Chemical potential	Variable, usually oxidizing, and dependent upon pH	Variable, over a narrower range, usually reducing	Up to 10^{10} or even more.
2. Radionuclide concentration and speciation	Supersaturation Polymerization Metastable equilibrium	10^{-3} to 10^{-9} g mole.kg^{-1} As ionic, polymeric, and particulate forms	Uncertain, but probably very low, depending on leaching characteristics of waste product form. Possibly as low as 10^{-9} to 10^{-12} g mole.kg^{-1} principally as ionic species.	Difficult to estimate, but could be very large for amphoteric species near the isoelectric point ($\sim 10^{6}$).
3. Flow rate	Metastable equilibrium.	$\sim 10^{-5}$ to 10^{-3} m.sec^{-1}	10^{-9} to 10^{-5} m.sec^{-1}	Lower flow rates could lead to different rate controlling transport mechanisms (e.g., ionic or molecular diffusion or advection). Could also lead to different thermodynamic controls (ion exchange vs adsorption (0 to 10^{6}).
4. Permeability	Flow rate (see above)	10^{-2} to 10 Darcys	10^{-8} to 10^{-3} Darcys	Same as above.
5. Duration	Radionuclide decay Daughter formation Front reinforcement	$\sim 10^{5}$ sec.	Up to 10^{13} sec.	None considered at this time. See also Flow rate above.
6. Surface area	Adsorption	Up to $\sim 10^{8}$ m^{2}.m^{-3} Dispersed clays, humus, fine particulates, loess	$\sim 10^{4}$ m^{2}.m^{-3} Fractures, microfractures intergranular pores	$\sim 10^{4}$
7. Path length	Dispersion	One meter	10^{3} - 10^{5} m	No anticipated effect on K_d
8. Temperature	Complexing Solubility Adsorption	25°C	10 to 300°C	Up to 10^{3}

After Apps et al., 1978.

The most serious problem connected with groundwater transport of radionuclides will be the difficulty of predicting migration rates in the field from data obtained in the laboratory. Let us consider, for example, the effect of variable adsorption and surface areas between laboratory clay and repository host rocks. The following calculations assume that at saturation all exposed surfaces adsorb one atom of a radionuclide species for every 2.5×10^{-19} m^2.

Most experimentally determined distribution coefficients are for soils, clays, and sediments. Clays are assumed to possess readily exchangeable sites of up to 100 milli equivalents.100 g^{-1} (Carrels and Christ, 1965) or approximately 2.5×10^3 equivalents . m^{-3}. The specific surface area, A, of a clay is possibly about 10^8 $m^2.m^{-3}$. A monolayer attachment on this surface would yield 6.6×10^2 g mol.m^{-3}, which is roughly in accord with measurements. In contrast, the surface area of a rock is only 10^4 $m^2.m^{-3}$, and the adsorptive capacity of typical rock-forming minerals may also be somewhat less than clays or soils.

Typical conditions for a host rock environment and a laboratory leaching study are summarized in Table 5.

Table 5: Comparison of conditions found in a laboratory sorption experiment and a repository host rock

Parameter	Unit	Sorption Experiment	Host Rock
$1 - \phi$		0.6	≈ 0.95
ρ_ℓ	[kg m^{-3}]	1×10^3	≈ 1×10^3
ρ_s	[kg m^{-3}]	2.5×10^3	≈ 2.5×10^3
$\sum_{\alpha=1}^{\xi} \omega_{\alpha,i}\sigma_{\alpha,i}$	[m^{-2}]	4×10^{18}	~ 4×10^{18}
ϕ		0.4	≈ 0.05
A	[$m^2.m^{-3}$]	10^8	≈ 10^4
N	[g mol^{-1}]	6.02×10^{23}	≈ 6.02×10^{23}

Using equation 27, the following relation can be derived:

$$\log K_d = \log \left[\frac{A}{N} \sum_{\alpha=1}^{\xi} \omega_{\alpha,i}\sigma_{\alpha,i} \cdot \frac{1-\phi}{\rho_\ell} \right] - \log C^\ell_{M_i}$$

This equation is plotted graphically in Figure 8 for two surface areas, and differing percentage site occupancies. One is typically representative of a soil (A 10^8),and the other of a rock (A = 10^4).

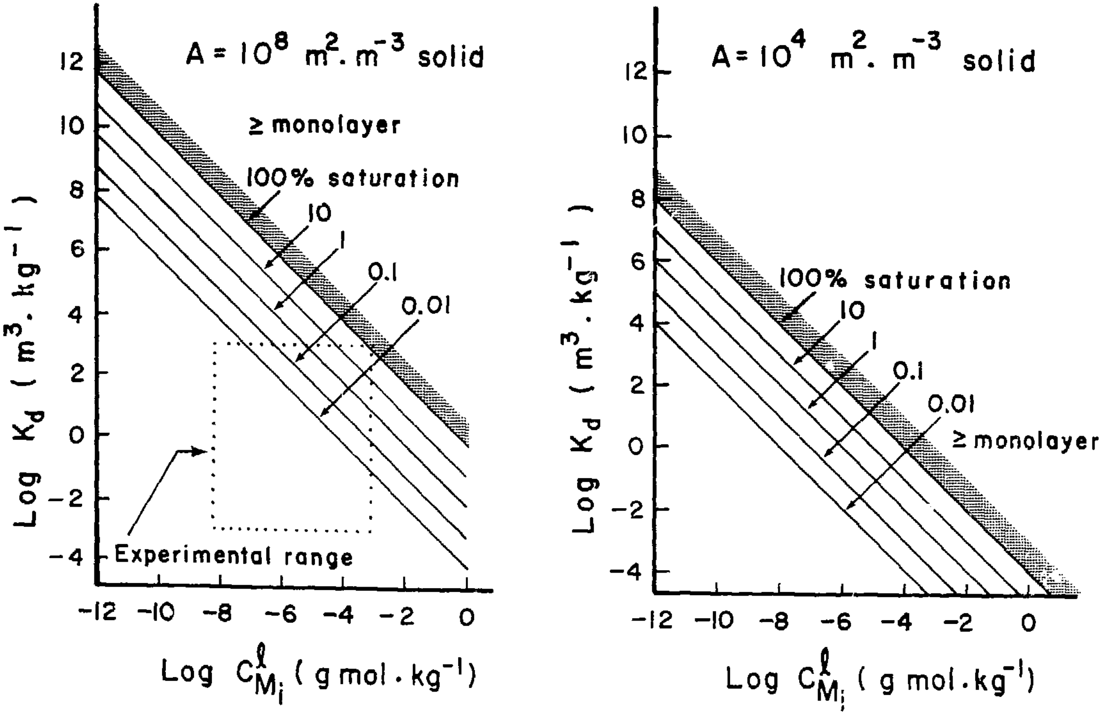

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Figure 8: Distribution coefficient, K_d, as a function of surface area, molecular site occupancy, and radionuclide concentration in solution.

The experimentally determined K_d values range from 10^{-3} to 10^2 $m^3.kg^{-1}$ (1 to 10^5 $ml.g^{-1}$). For a given site occupancy, and a given $C^{\ell}_{M_i}$, the "rock K_d" appears to be 10^4 times smaller than for soils. Thus, rocks would be much less effective than soils for retarding radionuclide movement by means of natural sorption mechanisms. However, more must be learned about the effective surface areas of rocks in their natural state.

Furthermore, we know little of the effect of long times on the sorption mechanism, a factor not easily studied through laboratory procedures. Some nuclides may remain permanently adsorbed: others may diffuse into the mineral lattice, or reprecipitate as discrete phases, thereby reducing the total equilibrium concentration in solution. The possibility that this may occur cannot be ignored if time spans of up to 10^6 years are to be considered.

b. Host Rock Geometry

It is assumed that the host rock for a terminal storage repository will be relatively impermeable. The matrix permeability of many hard rocks is so low that no significant groundwater movement can occur through the matrix (Apps et al., 1977). However, hard rocks may have significant permeability, due to ubiquitious fractures. Groundwater movement in these rocks therefore depends mainly upon fracture permeability and not, except in highly permeable sandstones, upon the matrix permeability.

The above postulate is supported indirectly by chemical evidence in the field. In low permeability rocks, alteration by migrating groundwater at low temperatures (< 100°C) usually occurs only along fracture surfaces, extending only a small distances into the adjacent rock. Alteration zones surrounding fractures generally range from less than a millimeter to a centimeter. It is presumed that this alteration takes place over time spans greater than those contemplated for the storage of radioactive waste, although useful information on this matter is hard to find in the literature.

Figure 9 illustrates schematically the groundwater environment typically expected. Fracture flow is assumed with concurrent diffusion of dissolved radionuclide species into the pores of the rocks. Some fractures oriented in the general direction of flow, and with sufficient continuity and width, will act as conduits for the groundwater flow. Others which are dead-ended or perpendicular to the hydraulic gradient will not contain any flowing water.

Although many rocks contain randomly oriented fractures, let us consider the effect of the oriented fracture, or joint geometry and effective porosity on surface area of a host rock. Assume n joint sets. Joints in each set are separated ℓ meters apart. The average fracture width of the joints in the set is w meters. Therefore the surface area exposed by the joints,

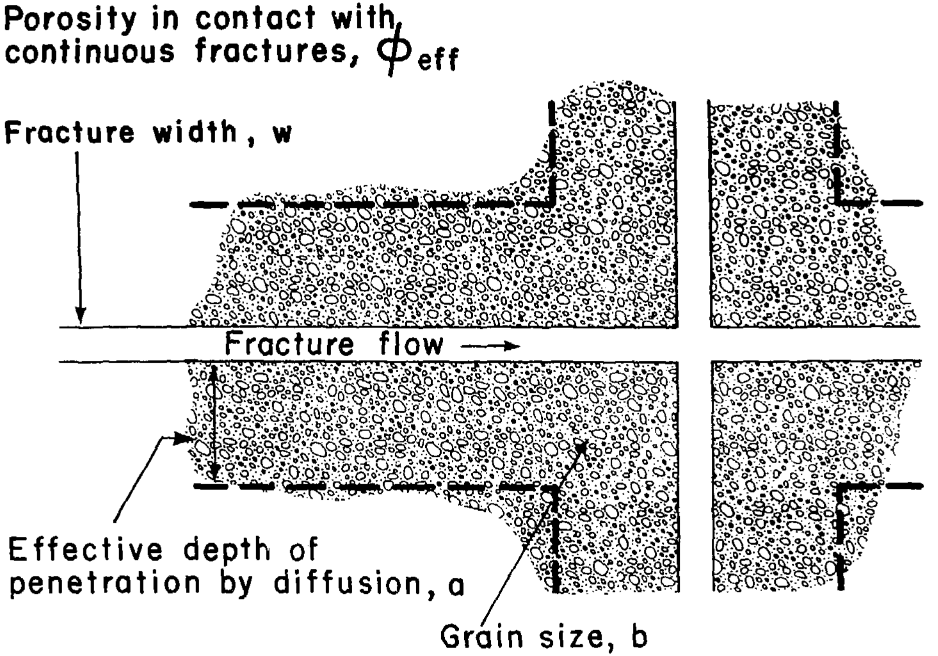

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Figure 9: Schematic diagram to illustrate the host rock geometry.

$$A_j = 2\xi_j \sum_{i=1}^{n} \frac{1}{\ell_i} , \qquad [\mathrm{m}^2.\mathrm{m}^{-3}\ \mathrm{rock}] \qquad (28)$$

where ξ_j is a surface roughness factor which ranges typically from 1 to 10. The fracture volume is

$$V_j = \sum_{i=1}^{n} \frac{w_i}{\ell_i} , \qquad [\mathrm{m}^3.\mathrm{m}^{-3}\ \mathrm{rock}] \qquad (29)$$

Each joint has associated with it a rock matrix of thickness, a, accessible by the fracture fluids. The rock matrix is made up of cubic grains of size s, and the porosity of the rock matrix adjacent to the fractures is ϕ_{eff}. Assuming $\ell >> a$; then the pore volume of the rock connected to the fracture volume is

$$V_p \simeq 2a\phi_{eff} \sum_{i=1}^{n} \frac{1}{\ell_i} \qquad [\mathrm{m}^3.\mathrm{m}^{-3}\ \mathrm{rock}] \qquad (30)$$

and the surface area of the pores connected to the fracture volume is

$$A_p \simeq \frac{12a}{s} \xi_p \sum_{i=1}^{n} \frac{1}{\ell_i} \qquad [\mathrm{m}^2.\mathrm{m}^{-3}\ \mathrm{rock}] \qquad (31)$$

where ξ_p is a combined roughness/shape factor. The total fluid volume in fractures and adjacent pores is:

$$V = \sum_{i=1}^{n} \frac{w_i}{\ell_i} + 2a\phi_{eff} \sum_{i=1}^{n} \frac{1}{\ell_i} , \qquad [\mathrm{m}^3.\mathrm{m}^{-3}\ \mathrm{rock}] \qquad (32)$$

and the total effective surface area is:

$$A = 2\xi_j + \frac{12a}{s} \xi_p \sum_{i=1}^{n} \frac{1}{\ell_i} \qquad [\mathrm{m}^2.\mathrm{m}^{-3}] \qquad (33)$$

The simple equations developed above will be used in the following subsection where the effect of rock geometry on radionuclide migration is described.

c. Effect of Chemical Sorption and Host Rock Geometry on Radionuclide Migration

Radionuclides can migrate away from the repository as dissolved, charged, or uncharged ions or molecules; as colloids; or as particulates. All species can be transported in the moving groundwater, but the ability of colloids or suspended particulates to diffuse into the pores of the rock is progressively restricted with increasing size. Thus, we can consider two limiting cases: the species that diffuse, such as molecules or ions, whose diffusion coefficients are about 10^{-9} $m^2.sec^{-1}$, and those that do not, such as suspended particulates with diffusion coefficients less than 10^{-12} $m^2.sec^{-1}$. The transported species can also be adsorbed by the mineral surfaces exposed to the groundwater. We can consider two limiting cases: no adsorption ($K_d = 0$) or complete adsorption as a monolayer ($K_d = \infty$). There are thus four limiting cases to consider. These cases and their characteristics with regard to radionuclide migration are summarized in Table 6.

Table 6: Radionuclide migration in fractured rocks

Species Type	No adsorption ($K_d = 0$)	Complete adsorption ($K_d = \infty$)
Ionic or Molecular	A1 Species can diffuse into rock pores. Therefore diffusional mixing occurs with the pore water and migration of the species is retarded in approximate proportion to the total connected water mass versus the mobile water mass in fracture conduits.	A2 Species diffuse into the rock pores and are adsorbed on all surfaces connected to fracture conduits
Suspended Particulate	B1 Species are transported with the groundwater. No diffusion into rock pores occurs.	B2 Species are adsorbed only on the surfaces of fracture conduits.

To illustrate the capacity of the host rock to adsorb radionuclides under these different conditions, let us consider a hypothetical storage repository which is leaking radionuclides into the groundwater. The repository is assumed to have the size and geometry indicated in Figure 10. This repository could contain 40,000 canisters of SURF spaced at 10-meter intervals. Because approximately 30 tons of SURF, approximately 14 canisters, would require storage for every 1000 MWe.yr of power, such a repository would serve the needs of one hundred 1000 MWe LWR plants for nearly 30 years. It is assumed that suitable hydrological criteria have been met, groundwater movement will be horizontal, and the radionuclides will be swept through a cross section of rock approximately 0.1 × 2.0 km or 2×10^5 m^2. The rock is fractured in three directions - two of the fracture sets are parallel and the third is perpendicular to the direction of groundwater flow.

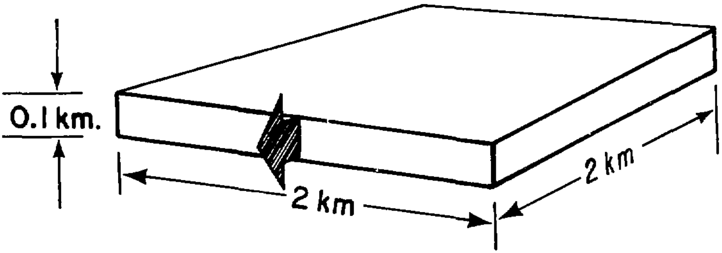

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Figure 10: Geometry of a hypothetical storage facility.

Further, let us assume the following rock physical properties:

$\xi_j, \xi_p = 1$

$v_\ell = 0.01$

$a = 0.01$

$\ell = 0.1$

$n = 0.1$

$s = 0.001$

$w = 10^{-5}$

$\phi_{eff} = 0.001$

Then, in a 1 km^3 slab:

$A_j = 6 \times 10^{10}$, $[m^2.km^{-3}]$

Area of fracture conduits = 4×10^{10}, $[m^2.km^{-3}]$

Area of other fractures = 2×10^{10}, $[m^2.km^{-3}]$

$V_j = 3 \times 10^5$, $[m^3.km^{-3}]$

Volume flowing = 2×10^5, $[m^3\ km^{-3}]$

Volume static = 1×10^5, $[m^3\ km^{-3}]$

$A_p = 3.6 \times 10^{12}$, $[m^2\ km^{-3}]$

$V_p = 6 \times 10^5$, $[m^3\ km^{-3}]$

$A = 3.66 \times 10^{12}$, $[m^2\ km^{-3}]$

$V = 9 \times 10^5$ $[m^3\ km^{-3}]$

Using the values above, we can determine the approximate distances radionuclide species will travel after one million years if it is assumed that they leach at a constant rate from the waste and the mass flux is known. Furthermore, it is assumed that radionuclides are adsorbed with one molecule for every 2.5×10^{-19} m^2 of exposed rock, and no account is taken of radioactive decay.

Case A1: Ionic or molecular transport with no adsorption

The total volume of flowing groundwater is 2×10^5 $m^3.km^{-3}$, whereas the static volume of groundwater in fractures and pores is 7×10^5 $m^3.km^{-3}$. The retardation due to molecular diffusion is

$$\frac{9 \times 10^5}{2 \times 10^5} = 4.5$$

If the groundwater travels 10 km in 10^6 years, the radionuclides will have traveled ~2.2 km.

Case A2: Ionic or molecular transport with adsorption

The internal surface of the rock is 3.66×10^{12} $m^2.km^{-3}$. Therefore the rock can adsorb on the surface 2.43×10^7 g mol of radionuclide.km^{-3} rock. The volume of rock saturated after 10^6 years, the distance away from the storage site that radionuclides will have saturated the rock, and the total mass of radionuclide transported for a given concentration of radionuclide, are given in Table 7 as a function of varying radionuclide mass fluxes.

Table 7: Distances adsorbing ionic or molecular radionuclide species travel after 10^6 years

Radionuclide Mass Flux leaving repository (g $mol.m^{-2}.yr^{-1}$)	Radionuclide Concentration leaving repository (g $mol.kg^{-1}$)	Volume Saturated (km^3 rock)	Distance Radionuclide Travels (km)	Mass Transported (g mol)
2×10^{-6}	10^{-3}	1.65×10^{-2}	8.25×10^{-2}	4×10^5
2×10^{-9}	10^{-6}	1.65×10^{-5}	8.25×10^{-5}	4×10^2
2×10^{-12}	10^{-9}	1.65×10^{-8}	8.25×10^{-8}	4×10^{-1}
2×10^{-15}	10^{-12}	1.65×10^{-11}	8.25×10^{-11}	4×10^{-4}

Case B1: Suspended particulate transport with no adsorption

Suspended particulates will migrate with the groundwater. Therefore the distance traveled after 10^6 years is 10 km.

Case B2: Suspended particulate transport with adsorption

The surface area of rock fractures exposed to flowing groundwater is 4×10^{10} $m^2.km^{-3}$. If the surface adsorption density is the same as for ionic or molecular species, then the distance away from the site that suspended particulates will have saturated the rock for given mass fluxes leaving the storage facility is given in Table 8.

Table 8: Distances adsorbing suspended particulate radionuclides travel after 10^6 years

Radionuclide Mass Flux leaving repository (g mol.m^{-2}.yr^{-1})	Radionuclide Concentration leaving repository (g mol.kg^{-1})	Volume Saturated (km^3 rock)	Distance Radionuclide Travels (km)	Mass Transported (g mol)
2×10^{-6}	10^{-3}	1.51×10^{0}	7.53×10^{0}	4×10^{5}
2×10^{-9}	10^{-6}	1.51×10^{-3}	7.53×10^{-3}	4×10^{2}
2×10^{-12}	10^{-9}	1.51×10^{-6}	7.53×10^{-6}	4×10^{-1}
2×10^{-15}	10^{-12}	1.51×10^{-9}	7.53×10^{-9}	4×10^{-4}

The above analysis does not consider intermediate cases. It is evident that partial adsorption will result in radionuclide migration intermediate between the extreme values cited. Furthermore, the analysis also ignores the effect of radioactive decay, which over the time span being considered, would have a significant impact on the distances individual nuclides would travel because of changing chemistry and the formation of nontoxic daughters (Burkholder et al., 1976).

Given the large number of variables needed to describe a rock, it is not easy to define criteria necessary for a host rock to be acceptable for a storage repository. Clearly, the adsorptive capacity of the rock is of major importance. This will be determined partly by the specific surface area of the rock, partly by the minerals exposed at the surface, and partly by the chemistry of both the groundwater and the transported nuclide, i.e., a high dispersion coefficient, or K_d value.

The radionuclide mass flux from the repository is another important parameter affecting the distance radionuclides travel and the total amount leached. Recalling that

$$J_{ad} = C^{\ell}_{M_i} v_{\ell} \rho_{\ell} \phi, \qquad [g\ mol.m^{-2}.yr^{-1}]$$

and because,

$$\rho_{\ell}\phi \simeq 1, \qquad [kg.m^{-3}]$$

if

$$v_{\ell} \simeq 0.01 \text{ to } 10, \qquad [m.yr^{-1}]$$

then

$$0.01\ C^{\ell}_{M_i} < J_{ad} < 10C^{\ell}_{M_i}. \qquad [g\ mol.m^{-2}.yr^{-1}] \qquad (34)$$

In the example given above, ϕ is low (0.0002), but it is clear that if J_{ad} were less than 10^{-6} g mol.$m^{-2}.yr^{-1}$ and the nuclide had a large distribution coefficient, a very satisfactory control over radionuclide migration would be established.

Perhaps the biggest problem will be the difficulty of characterizing a rock mass of sufficient size to encompass the distances groundwaters could travel away from the repository in 10^6 years. Volumes of many cubic kilometers would have to be sufficiently well surveyed that no significant conduit would remain unidentified. This could prove to be impossible without riddling the rock with test holes. Thus, although the host rock may prove to be a very effective barrier to nuclide migration, it will be difficult to prove this in practice.

Conclusions

Table 9 summarizes in tabular form the critical questions, problem areas and research needs in order to determine the impact of the various barriers on waste storage system integrity. It is obvious at this stage, that a multiple barrier approach will be essential, but that the roles of each barrier need to be more clearly defined.

Table 4: Status of Barrier Integrity

Barrier	Critical Questions	Problem Areas	Research	Impact on Waste Storage System
Canister	1. How long can a container of a given material contain high level waste before failure? 2. What assurance is there that experiments to determine container integrity give information that can be extrapolated to 10^6 years? 3. Will internal or external corrosion destroy the container?	1. Corrosion (pitting, galvanic stress, etc.) both with regard to ground water environment and waste. 2. Radiation damage (enhanced solubility) 3. Fracture due to stress	1. Failure analysis model 2. Potentiostatic and passivation studies. 3. Chemical compatibility 4. Impact of radiation damage.	Guaranteed container integrity for 10^6 yrs would eliminate the need for additional barriers. An order of magnitude estimate of between 10^2 - 10^6 years would have an important bearing on the choice of secondary barriers and limit the number of radionuclide species of concern.
Waste Product Leaching Characteristics	1. How do various waste products leach over a long period of time? 2. Are leaching rates acceptable given waste storage condition and characteristics of secondary barriers? 3. Do ionic species alone leach or are colloids and particulates involved? 4. What controls the leaching rate?	1. The effect of devitrification, chemical attack by ground waters, radiation damage, diffusion of radioclides in the solid state. 2. The leaching rates of various radionuclides 3. The roles of dissolution, diffusion and advection.-	1. Determination of leaching rates of the various wastes. 2. Development of models to interpret the leaching history over long time periods. 3. Design of the waste to ensure near chemical equilibrium and minimum leaching rates.	If leaching rates can be decreased to levels such that the amount leached before decay is minimal, or that the rate is so low that concentration in the ground water is below toxic levels, then other barriers would not be necessary. Knowledge of leaching rates is critical to the choice of site and host rock.
Grout or Backfill	1. What materials are best suited for retarding the leakage of radionuclides? 2. To what extent will the grout affect permeability and hence ground water flow in the vicinity of the waste? 3. Will heat change the chemical and physical properties of the grout over time? 4. What thickness is required? 5. Can backfill material protect the container from corrosion?	1. Chemical interaction between the grout and the waste 2. Physical properties (plasticity, density, permeability, surface area) 3. Chemical properties (sorption, ion exchange, oxidation state, pH, etc.)	1. Choice of suitable grout, based upon service it should perform. 2. Characterization of chemical and physical properties. 3. Behavior of grout at elevated temperatures.	Suitable grout could reduce permeability in the waste storage area to very low values, thereby retarding leaching of the radioactive waste. It could support the excavated cavity, preventing further fracturing due to stress relief. The chemical reactivity of the grout to radionuclides could lower still further the release of radionuclides to ground water migration. It could act as a means of providing a hydrostatic stress field around canisters thereby reducing the possibility of canister rupture.
Host Rock	1. Will host rocks sorb radionuclides at the low concentrations expected from the leaching of waste. 2. Do laboratory K_d measurements apply to the host rock environment? 3. Are colloids or particulates significant? 4. Are there time-dependent factors during leaching that would affect the K_d values needed to predict the sorption behavior over 10^6 years? 5. How are the sorptive properties of rocks best determined?	1. K_d's of radionuclides with respect to host rocks. 2. Characterization of the transported species. 3. Identification of sorption mechanisms. 4. Rock physical properties and how they should be measured. 5. Extrapolation of labora- data to the field.	1. Sorption mechanisms at low concentrations. 2. Measurement of surface area, porosity, permeability, fracture geometry, etc. 3. Development of models to relate laboratory measurements with field conditions.	The host rock is the last line of defense if all other barriers fail. It is also the most site-specific, since no two rocks are the same. A A definite plan needs to be established whereby the host rock can be evaluated in terms of its chemical and physical properties before development. Chemical limitations must be integrated with hydrological factors to define host rock integrity.

V

SUMMARY AND CONCLUSIONS

A satisfactory underground repository for nuclear wastes must ensure that these wastes are isolated safely from the biosphere for a period of time sufficient to ensure that their radioactivity has diminished to harmless levels.

The geomechanical stability of a suitable repository site depends in general on the absence of major geological perturbations, such as earthquake faults and volcanism, and, specifically, on a state of stress condusive to stability. A conservative interpretation of existing evidence suggests that the value of the maximum principal stress should not exceed 25 MPa and that the value of the minimum principal stress should be no less than two-thirds of this. A lithostatic state of stress at a depth of a kilometer would constitute a near ideal condition.

However, the determination of the state of stress at depth in rock is difficult and uncertain. Improved technology is required.

The excavation of the repository is not likely to pose undue problems but the question of sealing exploration boreholes, shafts and tunnels to a degree commensurate with that of the rock mass is problematical.

The most likely means by which radioactive materials from the waste could reach the biosphere is through transport by groundwater. The movement of groundwater through argillaceous and crystalline rock masses occurs along joints and fractures, pervading them. Given joints and fractures of sufficiently low hydraulic conductivity, small hydraulic gradients and long flow paths, the time taken for water from a repository to reach the biosphere may be of the order of a million years. This is regarded as a sufficient period of time for isolation.

The hydraulic conductivity of fractures in rock under stress is not understood well, and there are unresolved difficulties in measuring low conductivity in the field.

The release of radioactive materials to the biosphere is retarded by the rate of leaching of the waste by the groundwater, dilution, and sorption in the rock mass.

The mass of a radionuclide decreases by about nine orders of magnitude over a period of thirty half lives. If the product of the concentration of a radionuclide in the groundwater and the total amount of groundwater contaminated with this radionuclide is small over such a period, the duration of leaching is controlled by radioactive decay. This is an important criterion for the design and selection of waste forms.

Suspended particulate material from the waste may be transported with the groundwater and ionic or molecular transport in the absence of adsorption is retarded only by dilution. If it is necessary to retard the transport of such radionuclides, appropriate barriers will have to be engineered in the repository.

The effects of adsorption on suspended particulate and ionic or molecular transport may be of major advantage. To realize this potential advantage large surface areas of rock must be in contact with the groundwater transporting the radionuclides. For wide ranges of radionuclide concentrations leaving a repository, from 10^{-3} to 10^{-12} (g mol.kg^{-1}), the distances over which the radionuclides may move during a million years can be insignificant except in the case of suspended particulates with concentrations of 10^{-3} or more.

In conclusion, the results of this appraisal suggest that argillaceous and crystalline rocks may provide suitable sites for nuclear waste repositories. The most important factor affecting their suitability appears to be the flux of groundwater through the repository. Although intense fracturing would be advantageous in retarding migration of hazardous materials from a repository in these rocks by sorption, the hydraulic conductivities of these fractures would have to be very small and the hydraulic gradient low.

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