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MASTER

ANALYSIS OF HEAT AND MASS TRANSFER IN SUB-SEABED DISPOSAL OF NUCLEAR WASTE

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ABSTRACT

A mathematical basis is developed for the prediction of thermal and radionuclide transport in marine sediments. The theory is applied to the study of radioactive waste disposal by emplacement, in specially designed containers, well below the sediment/water interface. Numerical results are obtained for a specified model problem through use of two computer programs designed primarily for the analysis of waste disposal problems. One program (MARIAH) provides descriptions of the temperature and velocity fields induced by the presence of a container of thermally active nuclear waste. A second program (ION^{TIG}), which utilizes the results of the thermal analysis, is used to provide predictions for the migration of four representative radionuclides: ^{239}Pu, ^{135}Cs, ^{129}I, and ^{99}Tc.

INTRODUCTION

It has been suggested by Bishop and Hollister[1] that the mid-plate, mid-gyre regions of the major oceanic basins be investigated as possible repositories for high level radioactive waste. It was proposed that solidified nuclear waste be encapsulated in suitably designed containers and implanted in the seabed below the surface of the sedimentary layer. This scheme would provide a series of barriers to the release of radionuclides into the environment. One potential barrier is the marine sediment itself which is composed of very fine, porous, consolidated clay of low permeability.

The fundamental problem addressed in this paper is that of describing, mathematically, the transport of radionuclides which may inadvertently be released in marine sediments. We assume that the sediment can be adequately represented as a rigid, fluid-saturated porous medium. In general, the transport of radionuclides in such a medium involves the four fundamental processes: convection, diffusion-dispersion, sorption, and radioactive decay. Mathematical models are proposed for the description of the processes involved. Subject to rather stringent simplifying assumptions, it is possible to obtain approximate analytical solutions of the basic models, and this approach is outlined briefly. Solutions of the complete mathematical system can, however, be obtained only through the use of numerical techniques. The major portion of the paper is then devoted to the description and application of two computer programs which have been developed for the analysis of the radionuclide migration process.

One program (MARIAH[2,3]) is used to provide descriptions of the temperature and velocity fields. The analysis is conducted under the assumption that only weak concentrations of radionuclides exist in the pore water. Hence, thermal processes are not influenced by the radionuclide migration processes and can thus be analyzed separately. A second program (ION^{TIG}[4]) then utilizes the results of the thermal analysis to complete the description of the migration of radionuclides.

Preliminary results are presented for a proposed emplacement geometry. For the particular arrangement considered, it is shown that thermally induced fluid motion is relatively weak and, consequently, has little influence on the transport of radionuclides. The migration of representative radionuclides is analyzed and the associated breakthrough times computed.

Many aspects of the analyses which will subsequently be considered have been reported previously by McVey, Gartling, and Russo.[5] We have made extensive use of this earlier work in the preparation of the present paper.

REFERENCE EMPLACEMENT CONFIGURATION

In order to facilitate comparative studies, a reference emplacement configuration has been specified. The particular configuration was chosen to represent what is, at this time, believed to be a practically attainable scheme for the emplacement of nuclear waste in the seabed, and is the basis for all subsequent calculations.

A cylindrical container of solidified nuclear waste 3 m in length and 0.3 m in diameter is emplaced vertically in undisturbed sediment with the center of the container 30 m below the sediment/water interface. The sediment layer is 60 m thick and is bounded from below by a relatively impermeable layer. Temperature and pressure at the sediment/water interface are assumed constant at 1.5°C and 6×10^7Pa (600 bar), respectively, corresponding to a water depth of approximately 6000 m. The initial temperature distribution in the sediment is established by the steady geothermal heat flux (0.015 cal/m^2-s) and the constant interfacial temperature. A generic study site, with the described characteristics, has been selected in a region of the Central North Pacific.

The thermal power output of the container decays with time in response to the radioactive decay processes associated with the nuclear waste. In this paper, we assume the nuclear waste is typical reprocessed high level waste (HLW) resulting from a uranium only reprocessing cycle. The HLW is emplaced 10 years after reprocessing with reprocessing occurring 160 days after removal from the core. A normalized power history for the HLW is depicted in Fig. 1. Isotope inventories for the decay calculations were determined through use of the computer program ORIGEN.[6]

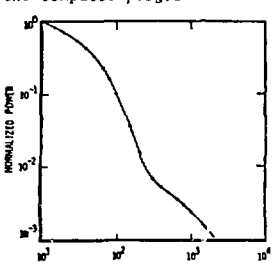

Fig. 1. Normalized Power History for HLW.

Krumhansl and Hadley[7] have reported studies dealing with thermally induced changes in the physical properties of saturated marine sediments, while Braithwaite and Molecke[8] have studied the effects of saturated sediment on corrosion of candidate container materials. Based on these studies, recommendations were made to limit the maximum temperature of the container to the range 200-250°C. Thermal analyses were then used to predict an associated maximum allowable thermal power output of 1.5 kW per container.

THERMALLY INDUCED CONVECTION

Basic Theory

In this section, we identify a mathematical model which is appropriate for the description of two-dimensional, free convection in a fluid-saturated porous medium and which forms the basis for the analytical and numerical studies to be described subsequently. The porous matrix is assumed to be rigid and the fluid incompressible, with density changes occurring only as a result of changes in the temperature according to

$$\rho = \rho_0[1 - \beta(T - T_0)] \quad , \tag{1}$$

where ρ is the fluid density, T is the temperature, β is the coefficient of thermal expansion, and the subscripts refer to reference conditions. In accordance with the Boussinesq approximation, the effects of density changes are accounted for in the buoyancy term of the equations of motion and are neglected in the continuity relation. It is also assumed that the fluid and matrix are in thermal equilibrium and that the fluid motion can be adequately described by Darcy's law. The equations of continuity, motion, and thermal transport can then be expressed, respectively, as

$$\frac{\partial v_i}{\partial x_i} = 0 \quad , \tag{2}$$

$$v_i = -\frac{k_{ij}}{\mu}\left(\frac{\partial P}{\partial x_j} + \rho g \frac{\partial z}{\partial x_j}\right) \quad , \tag{3}$$

$$\left(\rho C_p\right)_e \frac{\partial T}{\partial t} + \rho_0 C_p v_i \frac{\partial T}{\partial x_i} = \frac{\partial}{\partial x_i}\left[\left(\kappa_{e_{ij}} + \varepsilon E_{ij}\right)\frac{\partial T}{\partial x_j}\right] + Q \quad , \tag{4}$$

where the indices i and j take on the values (1,2) and summation is implied by repeated indices. The bulk volume average Darcy velocity component associated with the x_i coordinate direction is denoted by v_i, P is the pore volume average pressure, and t is the time. The permeability, effective thermal conductivity, and thermal dispersion tensors are denoted, respectively, by k_{ij}, $\kappa_{e_{ij}}$, and E_{ij}. Also, C_p, μ, ε, Q, and g are, respectively, the specific heat, viscosity, porosity, volumetric heat source, and acceleration of gravity. The elevation z is measured vertically upward. Effective properties are identified by the subscript e and are related to the fluid and solid matrix properties by the assumed relations

$$\left(\rho C_p\right)_e = \phi\rho_0 C_p + (1-\phi)\left(\rho C_p\right)_s \quad , \qquad (5)$$

$$\kappa_{e_{ij}} = \phi\kappa + (1-\phi)\kappa_{s_{ij}} \quad , \qquad (6)$$

where the subscript s refers to solid matrix properties and properties with a zero subscript or without a subscript are those of the fluid. It should be noted that we have used an extended form of the Boussinesq approximation in the development leading to Eqs. (2)-(4), in that we have not demanded that all thermophysical properties remain constant.

The mathematical system described thus far can be further simplified by combining Eqs. (2) and (3) to yield the scalar equation for pressure

$$\frac{\partial}{\partial x_i}\left(-\frac{k_{ij}}{\mu}\frac{\partial P}{\partial x_j}\right) = \frac{\partial}{\partial x_j}\left\{\frac{k_{ij}}{\mu}\rho_0[1-\beta(T-T_0)]g\frac{\partial z}{\partial x_j}\right\} \quad . \qquad (7)$$

It is usually convenient when implementing this equation to add the hydrostatic term $\rho_0 gz$ to the pressure, and to define this sum as a new pressure. The numerical method to be considered subsequently utilizes Eqs. (4) and (7) for the description of the pressure and temperature fields. Darcy's law, Eq. (5), is then used to obtain the velocity field from the pressure and temperature fields.

In order to complete the mathematical description, appropriate initial and boundary conditions must be specified for the dependent variables. Initial conditions are obtained by simply specifying initial values of pressure and temperature throughout the domain of interest. Over that portion of the boundary which encloses the hydrodynamic part of the problem, either the pressure or the velocity normal to the boundary must be specified. The specification of normal velocity is equivalent to the specification of the pressure gradient normal to the boundary. Over that portion of the boundary associated with the heat transfer portion of the problem, temperature or heat flux normal to the boundary must be specified. The latter condition is equivalent to the specification of the temperature gradient normal to the boundary. All boundary conditions can, in general, be functions of time as well as position.

Analytical Solutions

Here we wish to briefly review some previous attempts to describe, analytically, the thermal convection induced by a concentrated source in a fluid-saturated porous medium. If all physical properties are assumed constant, Eqs. (1)-(4) can be rendered non-dimensional in such a way that the only parameter involved is the Rayleigh number

$$Ra = \frac{k\rho_0^2 C_p g\beta Q'}{\kappa^2\mu} \quad , \qquad (8)$$

which is physically representative of the relative importance of buoyancy forces, viscous forces, momentum diffusion, and thermal diffusion. In Eq. (8), Q' is the energy released per unit time by the concentrated source, and all other parameters have been defined previously. Using the physical parameters for marine sediment summarized in Table I, the anticipated value for Ra is approximately 10^{-3}.

TABLE I
PHYSICAL PROPERTIES (20°C)

Property	Pore Fluid	Mineral	Waste Solid
ρ (kg/m^3)	1000.0	2980.0	2275.0
C_p (W-day/kg-°C)	0.052	0.010	0.010
κ^* (W/m-°C)	0.646	1.896	1.286
μ^* (kg/m-day)	93.590	---	---
β^* (1/°C)	3.380×10^{-4}	---	---
ϕ ---	---	0.8	---
k_{11} (m^2)	---	5×10^{-16}	---
k_{22} (m^2)	---	5×10^{-17}	---

For the steady-state, Hickox and Watts[9] obtained a numerical solution for the case of a point source in an infinite region. Results of this analysis show that when Ra << 1, the temperature distribution is virtually unaffected by the fluid motion and is essentially identical with that established by thermal conduction. Hence, the straightforward application of conduction theory is expected to provide an accurate description for the temperature distribution associated with the proposed emplacement scheme.

Bejan[10] obtained transient and steady-state solutions, for the problem described above, in terms of power series in Ra which are valid for Ra << 1. For the liriting case Ra → 0, Hickox[11] extended Bejan's results to the case of a concentrated source in a semi-infinite region below a permeable boundary on which the pressure and temperature are maintained constant. This latter case provides a reasonably accurate description of the far-field behavior for the reference emplacement.

Numerical Solutions

Computational Approach. Numerical solutions to Eqs. (3), (4), and (7) are obtained through use of the finite element computer program MARIAH which is based on the Galerkin form of the finite element method. The program structure parallels those of previous finite element computer programs developed by Gartling[12,13] and thus shares many of the conventions and capabilities of these programs. The application of the Galerkin form of the finite element method has received comprehensive treatment in previous publications[14,15] and will not be considered in detail here. Rather, the discussion will be limited to a brief description of the basic numerical procedure and some of the more important features of the computer program. Complete details concerning the construction and capabilities of the program are available in Refs. 2 and 3. In general, MARIAH can be used to analyze a rather broad class of transient or steady-state problems in arbitrarily shaped, two-dimensional (planar or axisymmetric) porous regions.

Application of the finite element approximation to Eqs. (7), (4), and (3), respectively, results in a system of discrete algebraic equations which can be written in matrix form as

$$\underline{\underline{K}}\underline{P} = \underline{F}(\underline{T}) \quad , \tag{9}$$

$$\underline{\underline{N}}\dot{\underline{T}} + \underline{\underline{D}}(\underline{v})\underline{T} + \underline{\underline{L}}\underline{T} = \underline{G} \quad , \tag{10}$$

$$\underline{v} = \underline{\underline{Q}}^{T}\underline{P} + \underline{\underline{R}}^{T}\underline{T} \quad . \tag{11}$$

Eqs. (9)-(11) represent a system of coupled, non-linear equations for the unknown vectors $\underline{P}$, $\underline{T}$, and $\underline{v}$ which are the nodal values of of pressure, temperature, and velocity. Within each element, the pressure and temperature are approximated using quadratic basis functions. The velocity components are linear within the element and discontinuous between elements.

The solution algorithms used for Eqs. (9)-(11) depend on the type of flow problem under consideration (i.e., isothermal, forced convection, free convection) and its dependence on time. For the solution of steady-state problems ($\dot{\underline{T}} = 0$), one of several methods based on Picard iteration can be selected. Transient flow problems are solved with a modified Crank-Nicholson procedure. In general, the solution methods employed in the computer program closely parallel the methods described in Ref. 12 for non-isothermal Navier-Stokes problems. In all cases, the actual solution of the matrix equations is accomplished by a frontal elimination procedure which is a special variant of Gaussian elimination.

Results for Reference Emplacement Configuration. The computational domain and boundary conditions for the numerical study of the natural convective flow induced by a thermally active container of nuclear waste are shown in Fig. 2. The container geometry, initial power, decay history, and burial configuration were described in a previous section. For computational purposes, we consider an axisymmetric region 120 m in diameter and 60 m thick, where the latter dimension corresponds to the thickness of the sedimentary layer. The diameter of the region was chosen to be sufficiently large so that the thermal field associated with the container is unaffected by the presence of the lateral boundary. This is not an unrealistic situation since, in an actual repository, waste containers would be emplaced sufficiently far apart to preclude thermal interaction. On the upper, horizontal, permeable boundary, pressure and temperature are held constant. Both the lower horizontal and vertical lateral boundaries are impermeable. The vertical boundary is insulated and the steady geothermal heat flux is applied to the lower boundary. Additional boundary conditions, required for the radionuclide migration study, are also included on Fig. 2.

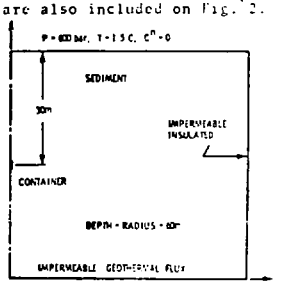

Fig. 2. Reference Emplacement Configuration.

In Table I, the material properties used in the thermal analysis are tabulated for a temperature of 20°C. Those properties indicated with an asterisk were actually allowed to vary with temperature in the analysis, in accordance with the relations given in Ref. 5.

Typical results of the numerical study are given in Figs. 3-5. In Fig. 3, selected isotherms are plotted for one and ten years after emplacement. As indicated in a previous section, a Rayleigh number of approximately 10^{3} is appropriate for this

situation. We thus expect the thermal field to be relatively unaffected by convection, as confirmed by Fig. 3. The symmetry of the isotherms indicates that the temperature distribution is established by thermal conduction. A typical computed axisymmetric streamline pattern is illustrated in Fig. 4 for an elapsed time of 10 years after emplacement. Finally, in Fig. 5, we present plots of temperature and vertical velocity versus time as calculated for a point adjacent to the container on the horizontal mid-plane. It is observed from this last figure that essentially all thermal activity has ceased as a result of thermal decay during the first 100 years of emplacement.

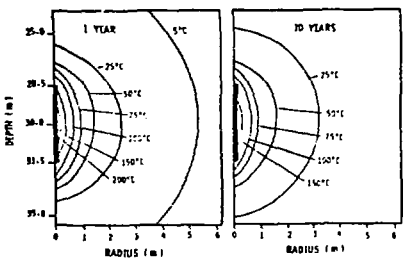

Fig. 3. Isotherm Patterns Near Container for Two Times.

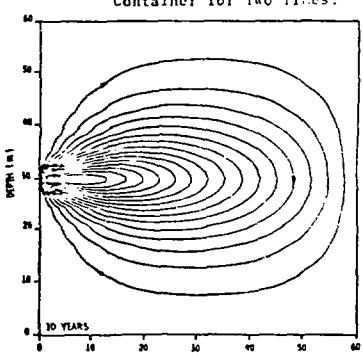

Fig. 4. Streamline Pattern at 10 Years After Emplacement.

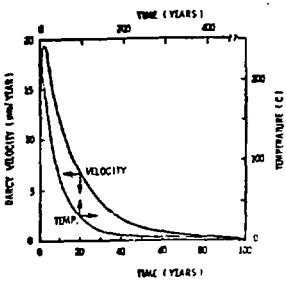

Fig. 5. Velocity and Temperature Histories on Container at a Depth of 5[illegible] m.

RADIONUCLIDE MIGRATION

Basic Theory

The development of the basic theory as well as the numerical solution of the resulting equations have been described in detail by Russo.[4] In what follows, we make extensive use of this previous work. For purposes of modeling the transport of radionuclides, it is assumed that the presence of radionuclides does not alter the fluid properties or the behavior of other nuclides and that sorption processes are reversible and describable in terms of an equilibrium distribution coefficient. Consequently, the transport of radionuclides in a porous medium by the combined effects of convection, diffusion-dispersion, sorption, and radioactive decay is described by the partial differential equation

$$\frac{\partial}{\partial t}(C^n \rho_b K^n) + \frac{\partial}{\partial x_i}\left(C^n v_i - D_{ij}^n \frac{\partial C^n}{\partial x_j}\right) = -\sum_{k=1}^{N} \lambda^{nk} K^n \rho_b C^n + \sum_{k=1}^{N} \lambda^{kn} K^k \rho_b C^k + S^n \quad , \qquad (12)$$

where C^n is the concentration for the ionic species n. In Eq. (12), K^n is the species equilibrium coefficient

$$K^n = \left(1 + \frac{1-\phi}{\phi}\right) \rho_s K_d^n \quad , \qquad (13)$$

where K_d^n is the equilibrium distribution coefficient which is, in general, a function of species concentration and temperature. For the numerical results to be described subsequently, we have neglected the temperature dependence and assumed

$$K_d^n = \frac{a_2}{1 + a_1 C^n} + \frac{a_4}{1 + a_3 C^n} \quad , \qquad (14)$$

where the a's are constants. The quantity D_i^n is the diffusion-dispersion coefficient

$$D_i^n = \alpha_{ij}^n |v_j| + D_o^n \quad , \qquad (15)$$

where α_{ij}^n are dispersion coefficients, D_o^n is the molecular diffusion coefficient, and the velocity v_i is the Darcy velocity as defined by Eq. (3). In our analysis, we have assumed $\alpha_{11}^n = \alpha_{12}^n = .61$ m and $\alpha_{21}^n = \alpha_{22}^n = 0.61$ m. Finally, λ^{nk} is the radioactive decay rate from species n to species k and S^n is a source term. All other parameters in Eq. (12) have been defined previously. The fluid velocity v_i is obtained directly from the thermal analysis.

Analytical Solutions

Numerous prior publications have dealt with the solution of various special forms of Eq. (12) for the transport of radionuclides in a one-dimensional system. We shall not attempt to review this rather large body of work, but will instead focus attention on a single analytical solution which has a more direct bearing on the problem at hand. Nuttall, Ray, and Davis[16] developed an analytical solution for the isothermal migration of a single radionuclide from a point source embedded in a semi-infinite region below a boundary on which the concentration was maintained at a constant value of zero. The processes of diffusion, sorption, and radioactive decay were included in the analysis. Convective transport was assumed negligible. In view of the rather weak convection predicted for the reference emplacement configuration, this last assumption appears reasonable. Concentration profiles and breakthrough times were predicted for the cases of instantaneous dissolution of the container and constant leakage rate from the container. The analysis is viewed as a reasonable first approximation to the far-field behavior of the migration process.

Numerical Solutions

Computational Approach. Numerical solutions to Eq. (12) were obtained with the computer program IONMIG. In IONMIG, Eq. (12) is solved in finite difference form through use of an explicit, predictor-corrector technique originated by MacCormack.[17] Stable integration of the finite difference form of the equation can, in general, require a different time step for each species. This is accomplished by selecting a global time step based on decay rate and output data considerations and an integration time step for each species which is an integer subdivision of the global time step and satisfies an appropriate stability criterion.

Decay chain species which are short-lived compared to the other chain members may be omitted by merely bypassing them in the definition of the decay chain matrix λ^{nk}. If such elements are included in the chain, the code will artificially reduce their decay constant (increase their half life) to a level for which they will disappear in several global time steps. This permits the inclusion of transition species in the chain without going to excessively small time steps. However, the instantaneous concentrations of such short-lived species will be greatly overestimated and should not be considered as part of the usable results.

In order to retain the flexibility of use provided by the options of axisymmetric geometry and variable mesh calculations, conservation differencing, which becomes rather unwieldy for these options, was not used. It is possible, therefore, that discretization and truncation errors may introduce some source error into the calculations. To partially correct for this possibility, a separate and more accurate time integration of the global quantities is performed and compared with the mesh point summations. An option to correct the mesh point values at each time step is available.

Results for Reference Emplacement Configuration. The computational domain utilized for the analysis of the radionuclide migration process is the same as that used for the thermal analysis and is depicted in Fig. 2. It is assumed that the entire contents of the waste container are initially uniformly distributed within a cylindrical region 3 m in height and 4 m in diameter centered about the location of the original container. The computer program IONMIG is then used to predict the subsequent migration of the radionuclides present in the initial inventory. Referring to Fig. 2, we note that the concentration is maintained at a value of zero at the sediment/water interface. Both the vertical and lower boundaries of the domain are considered impermeable to the transport of radionuclides.

For comparative purposes, we have considered the migration of four representative nuclides: the cationic species ^{239}Pu and ^{137}Cs, and the anionic species ^{129}I and ^{99}Tc. Plutonium is considered representa-

tive of transuranic elements with long half life and large equilibrium distribution coefficients K_d. Cesium is typical of fission products with short half life and moderate K_d. Both iodine and technetium have long half life and low K_d. Based on limited laboratory results obtained by Erickson,[18] we have assumed values for molecular diffusion and equilibrium distribution coefficients. These values can be obtained from the information contained in Table III. In order to study the migration of the four representative radionuclides, it was necessary to simultaneously consider the associated parent and daughter nuclides. Decay chains for each of the representative elements are listed in Table II along with the mass of each element present in the initial inventory for the waste container. The initial inventory is the same as that used for the thermal source calculations.

TABLE II
INITIAL INVENTORIES OF RADIONUCLIDES USED IN THE CALCULATIONS

Radionuclide	Mass in Container (kg)
^{243}Am	0.121
^{239}Np	1.0 E-7
^{243}Cm	6.92 E-5
^{239}Pu	3.58 E-2
^{235}U	1.0 E-5
^{129}I	0.305
^{129}Xe	1.178 E-4
^{99}Tc	1.13
^{99}Ru	4.32 E-5
^{137}Cs	1.33
^{137}Ba	2.01 E-7

Calculations for ^{137}Cs were carried out to 3500 years to verify that little motion took place prior to decay. Since the half life of ^{137}Cs is only 30 years, in a few hundred years virtually all the ^{137}Cs decays to ^{137}Ba, in place, and none reaches the surface. The same behavior is expected of other short-lived species with moderate values of K_d such as ^{90}Sr.

For long-lived isotopes having a high K_d, such as many forms of ^{239}Pu, the situation is similar except over much longer time scales. Fig. 6 shows the concentration of ^{239}Pu over a vertical plane containing the buried canister after 10^5 years.

TABLE III
DIFFUSION COEFFICIENTS AND PARAMETERS FOR EQUILIBRIUM DISTRIBUTION COEFFICIENTS

Radionuclide	D_0^n (m^2/yr)	a_1	a_2 (m^3/kg)	a_3	a_4
Pu	0.010	$3x10^8$	$1x10^2$	0.0	0.010
Cs	0.010	$2x10^5$	$1x10^1$	0.0	0.100
I	0.018	$1x10^4$	$1x10^{-4}$	0.0	0.0
Tc	0.010	$1x10^4$	$1x10^{-4}$	0.0	0.0

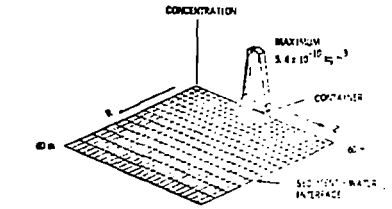

Fig. 6. Plutonium Concentration in the Sediment at 10^5 Years.

Although 4.1 half lives of ^{239}Pu have elapsed, the remaining plutonium is a little less than 1/3 of the original inventory because of the decay of ^{243}Am to ^{239}Pu. In this calculation, the Am and Np were assumed to have the same K_d as the plutonium. Data currently available indicate this is a reasonable assumption for Am but may yield too large a value for K_d for the short-lived Np. The migration rate of plutonium is so slow (a few metres per 10^5 years) that even after 10^6 years, less than 10^{-30} percent of the depleted inventory has crossed the surface. At that time, less than 10^{-10} grams of ^{239}Pu remain.

For anionic nuclides having a long half life and a very small K_d, such as ^{129}I and ^{99}Tc, the behavior is quite different. Without the retarding action of sorption, these substances diffuse through the sediment in a relatively short time (5000 years or less). Fig. 7 shows the release rate of radioactivity, at the sediment/water interface, associated with the decay of ^{129}I as a function of time. The release rate reaches a peak of 0.52 μCi/yr at about 8,000 years and then declines to approximately 1/3 of that value in 10^5 years. The specific activity of ^{129}I is low, decaying to ^{129}Xe by beta decay with a half life of 15.9 million years. Fig. 8 shows a similar release rate profile for ^{99}Tc. The peak value of 180 μCi/yr is much higher than that for ^{129}I because both the initial container inventory and the specific activity of ^{99}Tc, which beta decays to

^{99}Ru with a half life of 213,000 years, are higher.

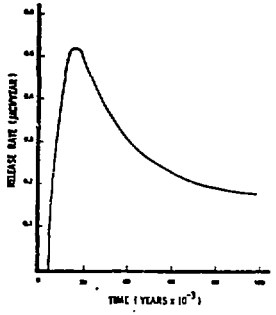

Fig. 7. Release Rate of ^{129}I from the Sediment Surface Covering a Single Container.

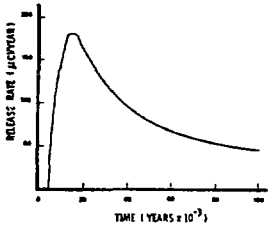

Fig. 8. Release Rate of ^{99}Tc from the Sediment Surface Covering a Single Container.

As a final observation, we note from Fig. 5 that after approximately 30 years, the maximum fluid velocity induced by thermal convection has decayed to a value typical of that due to diffusion (5-10 mm/yr). Consequently, we anticipate that convection has little effect on the migration of radionuclides in marine sediment; a conclusion which has been verified computationally.

DISCUSSION

We have developed a mathematical basis for the prediction of thermal and radionuclide transport in marine sediments. The theory was applied to the study of problems associated with the disposal of radioactive waste by emplacement in oceanic sedimentary layers well below the sediment/water interface. A reference emplacement configuration was defined and used for all analyses reported. Aside from some analytical approaches which were only briefly described, the studies reported in this paper were performed, numerically, using two computer programs which were designed especially for the analysis of the problem at hand.

Numerical results indicated that, for the proposed disposal scheme, the Rayleigh number associated with the thermally induced convective motion is expected to be quite small ($\cdot 10^{-3}$). Hence, the convective velocity is small and has only a negligible effect on the temperature distribution. Thermal conduction theory can thus be used effectively for the prediction of the temperature distribution. For the reference container, it was shown that all thermal effects decay to a negligible level in approximately 100 years.

The migration of the four representative nuclides ^{239}Pu, ^{137}Cs, ^{129}I, and ^{99}Tc was computed. For the reference problem, it was predicted that no plutonium or cesium would breach the sediment/water interface. It was, however, predicted that iodine and technitium would migrate to the interface and the associated release rates were computed.

The significance of the computed release rates in terms of human or ecological hazards can be determined only by a comprehensive analysis of the water column transport mechanisms and marine bio-system concentration mechanisms. Some perspective on the predicted release rates can be obtained, however, by a comparison with other oceanic radioactive sources. For the axisymmetric case for which the calculations were made, the area per container is based on a spacing of 120 m. This results in an average release per container of 4.5×10^{-5} μCi/yr-m^2 for ^{129}I and 1.6×10^{-2} μCi/yr-m^2 for ^{99}Tc. These fluxes compare to natural radium and radon fluxes of $3.5\text{-}8.5\times10^{-4}$ μCi/yr-m^2 for ^{226}Ra and 0.26-0.88 μCi/yr-m^2 for ^{222}Rn.[7] Based on this comparison, it may be hypothesized that the immediate exposure effects of the released ^{129}I and ^{99}Tc on benthic organisms would be negligible since they evolved in a much more intense field. Confirmation of this hypothesis awaits completion of ongoing research and experimentation on biological concentration, water column transport mechanisms, and development of pathways-to-man models.

At this point, a few final comments are offered concerning possible sources of error in our analyses. At the present, only rudimentary information is available for the thermophysical properties required in the thermal analysis. Lack of property information aside, the assumptions most likely to introduce errors into the thermal analysis are those associated with the Boussinesq approximation and the assumed

rigidity of the porous matrix. The rapid heating of a liquid confined in a rigid porous matrix of low permeability can actually produce a rather high transient pore pressure. As a result of the assumptions invoked, this behavior is not evidenced by our solutions. If the porous matrix is composed of clay particles, as is the case for marine sediments, then it is likely that some local deformation of the matrix will occur in regions of high heating rates. Questions regarding the deformational behavior of marine sediments cannot be answered conclusively until rheological properties have been accurately determined.

Accurate predictions of migration processes are also severely hampered by lack of experimental confirmation of various assumed properties. For example, insufficient information exists concerning equilibrium distribution coefficients. Recent laboratory studies[18] suggest that the mathematical form we assumed for the equilibrium distribution coefficient may be inadequate to describe the sorption of certain radionuclides, particularly iodine. Future predictive capability will depend crucially on the outcome of planned experimental research.

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