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A TWO-DIMENSIONAL FINITE-ELEMENT SOLUTION FOR THE SIMULTANEOUS TRANSPORT OF WATER AND MULTI SOLUTES THROUGH A NONHOMOGENEOUS AQUIFER UNDER TRANSIENT SATURATED-UNSATURATED FLOW CONDITIONS

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ABSTRACT

A two-dimensional finite-element model for the simultaneous solution of the transport of water and radionuclides in a nonhomogeneous anisotropic aquifer under saturated and unsaturated flow conditions is presented. The adsorption mechanism is described by a linear equilibrium isotherm. The model allows for a variety of boundary conditions; e.g. infiltration, drainage or evaporation. The solution to these equations is obtained through a Galerkin-type finite-element method using linear quadrilateral and triangular isoparametric elements, respectively. The model was tested by comparing predictive values with analytical solutions and experimental results for transient flow. Use of the model is illustrated by the analysis of the movement of hypothetical three-membered radionuclide decay chains leaching from the tailings pond of a typical uranium mill.

INTRODUCTION

Most uranium mills are located in semi-arid regions of the western United States imposing a potential threat to the environment there, including the quality of groundwater. The major concern is due to the long half-life of most radionuclides in the radioactive waste (tailings). Because of the danger, the fate and mode of travel of radionuclides downstream from their sources must be predicted. Considerable progress has been made in mathematical modelling to predict the extent and impact of groundwater contamination, but user's oriented models related to water and solute transport in a two-dimensional saturated-unsaturated media are scarce (Reeves and Duguid, 1975; Duguid and Reeves 1976).

A model with acceptable schemes for subsurface disposal of radioactive or other liquid wastes under saturated-unsaturated flow conditions is described here. An outline of the equations required to predict radionuclide migrations which are assumed to be in equilibrium with the geologic medium is given. Two test cases for validating the model are reported.

GOVERNING EQUATIONS

The continuity equation describing the transient saturated-unsaturated two-dimensional vertical plane flow of water assumed compressible in a slightly deformable unconfined aquifer in the presence of sources and sinks and under

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isothermal condition may be written as

$$L(h) = (c_m + S_w S_s)\frac{\partial h}{\partial t} - \nabla \cdot [K(\nabla h + \nabla z)] - Q = 0 \qquad (1)$$

where L is the defined differential operator; ∇ is gradient operator $\frac{\partial}{\partial x}, \frac{\partial}{\partial z}$; x and z are the horizontal and vertical (positive upwards) Cartesian coordinates (L); t is time (T); c_m is soil-moisture capacity (L^{-1}); S_w is soil-water saturation $\theta/\phi(L^0)$; θ is volumetric soil-water content (L^0); ϕ is porosity (L^0); S_s is specific storage coefficient (L^{-1}); K is hydraulic conductivity (LT^{-1}); h is soil-water pressure head (L); and Q is the strength of a source or sink function (T^{-1}).

In the derivation of Eq. (1), the specific discharge relative to the solid particles is assumed to be the one given by Darcy's law, written as

$$\bar{q} = -K\nabla(h + z) \qquad (2)$$

Note that in Eq. (1) c_m is zero for a saturated medium and S_s will be zero for a partially saturated medium, where the water density may be assumed constant. The solution of Eq. (1) will enable one to compute the soil-water content, porosity and specific discharge which are required for solving the solute-transport equation.

From the mass conservation requirement, the hydrodynamic dispersion equation describing the migrating of a solute γ in a liquid phase α through a sorbing saturated-unsaturated porous medium s may be written as

$$L(C) = \frac{\partial}{\partial t}(\theta C + \theta_s \rho_s S) - \nabla \cdot [\theta D \cdot \nabla C - C\bar{q}] - Q' = 0 \qquad (3)$$

where C is concentration of solute γ in the liquid phase α (ML^{-3}); S is concentration of solute γ in the absorbed phase s (M^0); ρ_s is soil bulk density (ML^{-3}); θ_s is volumetric fraction of the solid phase (L^0); D is coefficient of hydrodynamic dispersion (L^2T^{-1}); Q' is rate of production or removal of solute due to radioactive decay ($ML^{-3}T^{-1}$). The solution is assumed to behave like a homogeneous fluid.

The tensorial form for D for an isotropic medium with respect to dispersivity may be found in Bear (1972).

Because of the low-level nature of the radioactive liquid waste considered here, the mechanism of adsorption may be adequately described by a linear equilibrium isotherm given by

$$S = S_w K_d C \qquad (4)$$

where K_d is the distribution of coefficient (L^3M^{-1}). K_d is also function of the soil-water pH, which could be adequately handled by the model.

The transport phenomena of a radionuclide decay chain of the form A→B→C ... →N in a sorbing porous medium in both dissolved and solid states, is presumed to obey a first-order kinetic reaction under equilibrium condition expressed as

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C \rightarrow \ldots \xrightarrow{\lambda_{N-1}} N \tag{5}$$

where λ is the first-order rate constant for decay (T^{-1})

With the above considerations, the set of differential equations describing the movement of three-membered decay chains leaching for example from a liquid waste disposal pond is given by:

$$L(C_1) = \frac{\partial}{\partial t}(\theta R_{d_1} C_1) - \nabla\cdot[\theta D\cdot\nabla C_1 - C_1\bar{q}] + \lambda_1\theta R_{d_1} C_1 = 0 \tag{6a}$$

$$L(C_2) = \frac{\partial}{\partial t}(\theta R_{d_2} C_2) - \nabla\cdot[\theta D\cdot\nabla C_2 - C_2\bar{q}] + \lambda_2\theta R_{d_2} C_2 - \lambda_1\theta R_{d_1} C_1 = 0 \tag{6b}$$

$$L(C_3) = \frac{\partial}{\partial t}(\theta R_{d_3} C_3) - \nabla\cdot[\theta D\cdot\nabla C_3 - C_3\bar{q}] + \lambda_3\theta R_{d_3} C_3 - \lambda_2\theta R_{d_2} C_2 = 0 \tag{6c}$$

where

$R_d = 1 + [(1 - \phi)/\phi]\rho_s K_d$, is the retardation factor embodying the overall effect of the sorption/desorption reactions.

The finite-element method (FEM) solution schemes may be found detailed in Gureghian (1980, 1981a, 1981b). Note that Eq. (1) generates a system of ordinary differential equations with nonlinear coefficients and is solved by an iterative technique using a backward difference scheme. Eqs. (6) have a linear form and are solved in a sequential manner using a Crank-Nicolson scheme.

Test Examples

a) Saturated Case--Solute Movement (NH_4^+) with Adsorption and Nitrification.

The exact solution of the one-dimensional transport equation for NH_4^+) with nitrification under saturated steady-state flow conditions as given by Cho (1971) may be described by Eqs. (6) with the adsorption omitted in the decay components. The initial and boundary conditions selected for this example are: $C_1 = C_2 = C_3 = 0$ ($x>0$, $t=0$), $C_2 = C_3 = 0$ and $C_1 = 1$, ($x=0$, $t>0$), where subscripts 1, 2 and 3 refer to NH_4^+, NO_2^- and NO_3^-, respectively. Eqs. (6) were solved using a first-type boundary condition at $x = 0$. Figs. (1a) and (1b) show a comparison of the concentration profiles at time $t = 100$ days for three species for two cases of cell Peclet number $P_e(V\Delta x/D)$ and cell Courant number $C_r(V\Delta t/\Delta x)$, where V is the apparent velocity. Results display an acceptable level of confidence. An amplification of the numerical dispersion is witnessed in the case of NH_4^+ and NO_3^- with an increase of P_e.

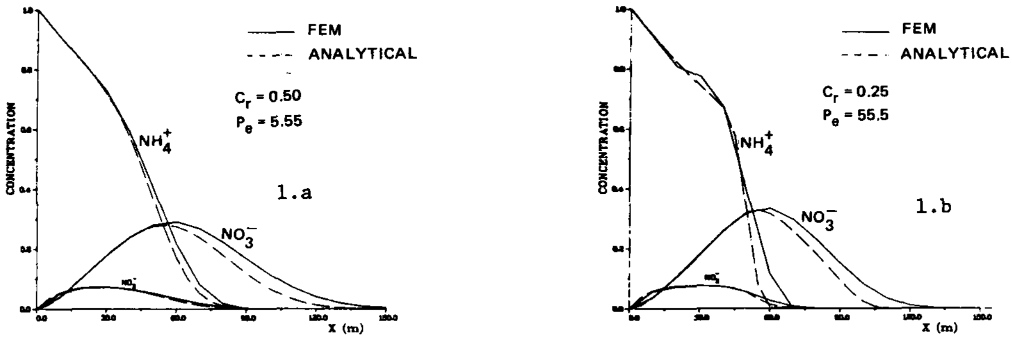

Fig. 1 Saturated nitrification solute profiles: $\lambda_1 = 0.01$/day, $\lambda_2 = 0.1$/day, $\lambda_3 = 0$; $V = 1.0$ m/day, $R_{d1} = 2$; $\Delta x = 10$m, (a) $\Delta t = 5$ days, $D = 1.8$ m^2/day; (b) $\Delta t = 2.5$ days, $D = 0.18$ m^2/day; $t = 100$ days.

b) Water and Solute Movement in a Homogeneous Unsaturated Soil.

Experimental field results for the simultaneous movement of water and a single solute (Cl^-) in a homogeneous unsaturated soil were reported by Warrick et al. (1971). The initial moisture distribution is shown by dotted lines in Fig. (2a). With an imposed boundary condition of $h = -14.5$ cm at $z = 0$, $t>0$, corresponding to the reported moisture content value, $\theta = 0.38$, Eq. (1) was solved using the same analytical expressions for the soil hydraulic properties as given in their paper. S_s and Q were set to zero, respectively. In the absence of adsorption ($R_d = 0$) and decay ($\lambda=0$), Eq. (6a) was solved using a boundary condition of the first and third type.

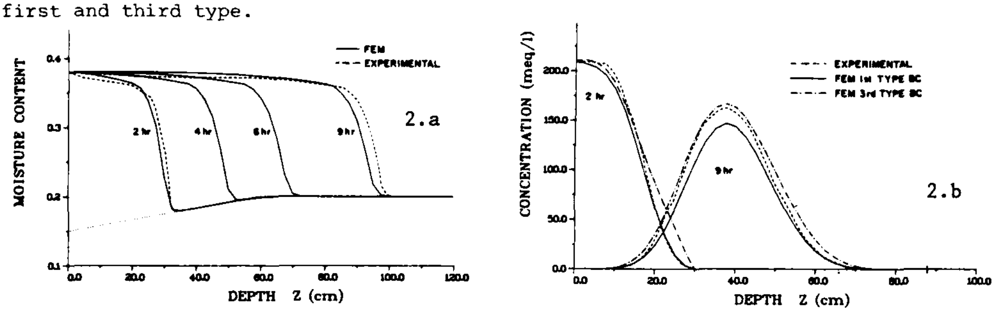

Fig. 2 (a) Moisture content profiles. (b) Concentration profiles. $\Delta z = 2.5$ cm. Initial $\Delta t = 1.98 \times 10^{-2}$ min, final $\Delta t = 4$ min. (Field data from Warrick et al. 1971)

The initial and boundary conditions in these cases are: $C = 0$ ($z \geq 0$, $t=0$), $C = 209$ mg/L ($z=0$, $0<t \leq 2.8$ hr) and $C = 0$ ($z=0$, $t>2.8$ hr) respectively. Note that in this instance, the longitudinal dispersivity was assigned a value of 1.026 cm. (See Segol (1977), van Genuchten (1980)). Figures (2a) and (3a) show a comparison of the moisture and concentration profiles for 9 hours of infiltration. In both cases, results seem to indicate a good agreement with the experimental values. Note that in the solute case, a third type boundary condition seems to yield superior results.

Application of Model to the Transport of Three-Membered Radionuclide Decay Chains

The flow domain shown in Fig. 3 refers to a vertical cross section of a sandstone aquifer (soil layer type a) with an interbedded layer of mudstone 4.7 m thick (soil layer type b), where its base and upstream boundary are impervious. The downstream boundary is in contact with a stream and the sole source of the aquifer recharge is due to the liquid leaching from the site of the disposal pond, which is 5 m deep and has a 1.2 m compacted clay liner (soil layer type c) at its bottom. Initially, the pond is subjected to a constant daily infiltration rate of 0.83 mm of H_2O until a steady-state condition is reached. Subsequently, the pond is used for a period of 10 years as a disposal site for the discharge of radioactive liquid waste from the processing of radioactive ores. It is assumed that the discharge rate remains unchanged during this time, after which the pond is restored to its initial function. The average longitudinal and lateral dispersivities correspond to 6 and 2 m, respectively; the tortuosity is 0.5; the diffusion coefficient is 8 x 10^{-4} m^2/month; the half-life and the distribution coefficients of species A, B and C are 100, 50, and 25 years and 5, 50, and 0.5 cm^3/g, respectively. The concentrations of species A, B, and C at the source are 100.0, 5.0, and 1.0 pCi/mL and the background concentrations are 10^{-7}, 10^{-6}, and 2.0 x 10^{-8} pCi/mL, respectively.

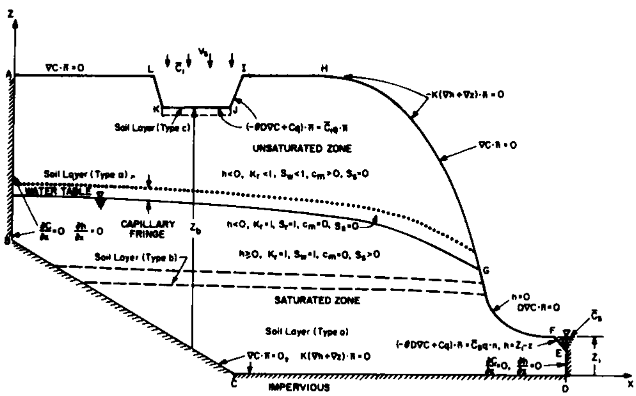

Fig. 3. Cross section of the flow domain.

Figure 4 shows the finite element grid which includes 477 mixed elements and 422 nodes. Triangles have been used in portions of the flow region susceptible to appreciable changes of h and C, respectively.

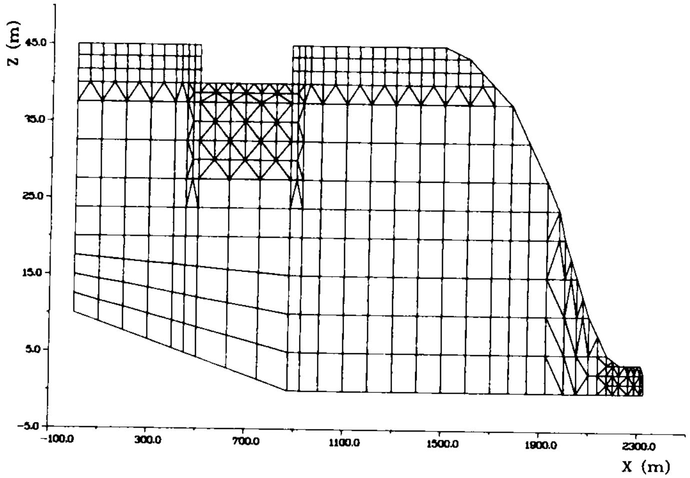

Fig. 4. Finite element grid.

Figures (5a) and (5b) show the characteristic curves of the 3 soils used in this simulation which bear a close resemblance with their counterpart located in the state of Wyoming U.S.A.

Figure (6) shows the soil-water pressure head distribution in the unsaturated zone yielded by the steady-state solution of Eq. (1). Under the circumstances, the large magnitude of h will enhance appreciably the buffering capacity of this portion of the aquifer in the presence of the radioactive liquid.

Figure (7) shows the distribution of the average specific discharge at the nodes computed in a straight forward manner.

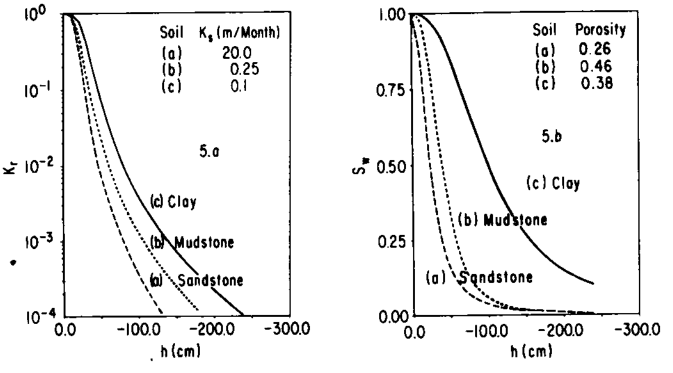

Fig. 5. (a) Relative hydraulic conductivity (K_r) and (b) soil-water saturation (S_w) both as functions of soil-water pressure head (h).

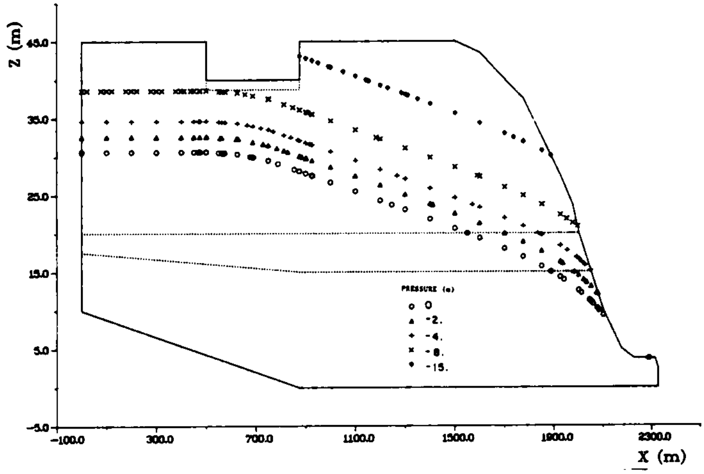

Fig. 6. The computed unsaturated soil-water regime above the water table.

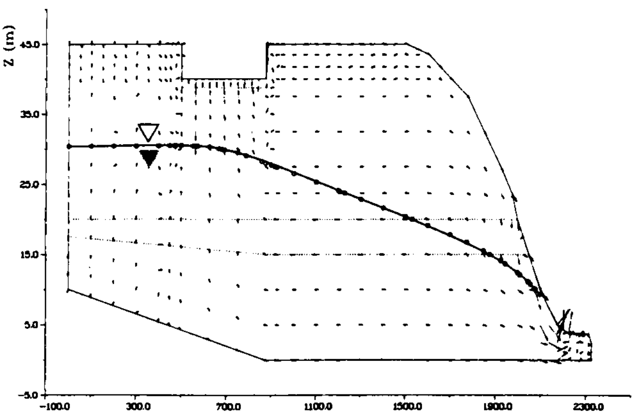

Fig. 7. Distribution of the average specific discharge.

Figures (8a), (8b) and (8c) show the concentration profiles of the hypothetical radionuclides A, B and C, respectively, at time t = 24.4 yr, almost 15 year after cessation of mill operations. An appreciable reduction of the original concentrations is witnessed: 92.3, 82.2 and 67% for species A, B and C, respectively. The rate of migration of species B, which has the highest K_d is almost negligible since the tip of the plume remains in the immediate vicinity of the pond base. The trend is migration of species C which has the lowest K_d of the three is relatively noticeable. In this case the tip of the plume is in the saturated zone of the aquifer.

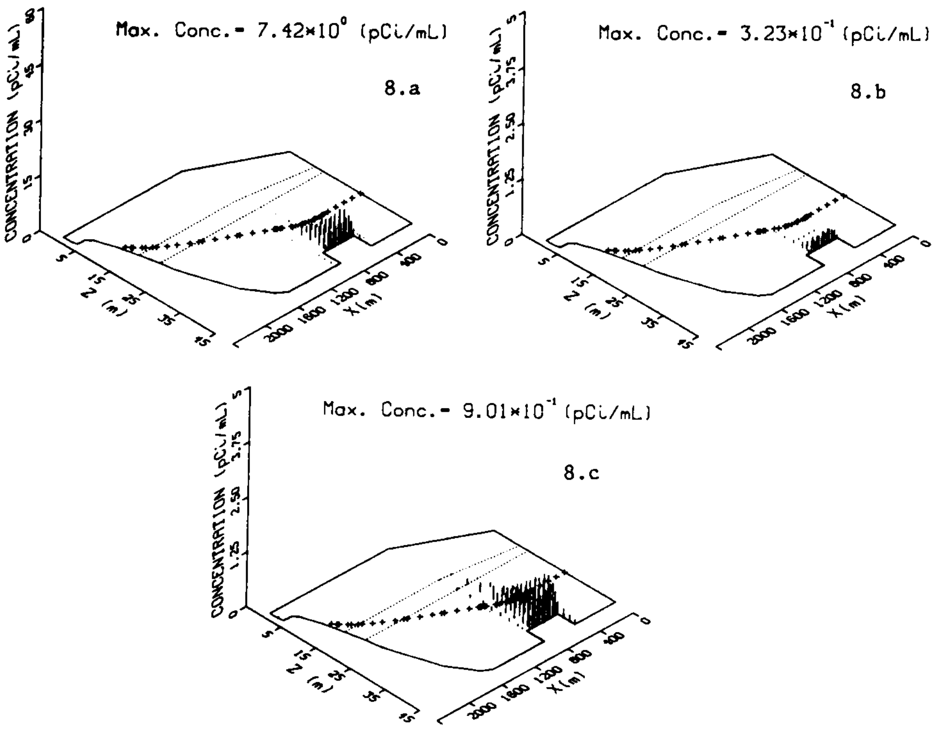

Fig. 8 a-c Concentration profiles of species A, B and C at t = 24.4 years.

Literature pertinent to this topic emphasizes the major role played by the retardation factor on the rate of migration of the radionuclides; the present analysis is supportive of this theory. The distribution factor is also pH dependent; however, this particular feature of K_d has not been considered in this instance.

CONCLUSIONS

An improved mathematical model for predicting the migration of radionuclides and their decay products in a nonhomogeneous saturated and unsaturated phreatic aquifer has been developed and is currently used by the Division of Environmental Impact Studies of ANL to evaluate the potential radioactivity releases to the groundwater at the site of uranium mills.

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