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SIMULATION OF URANIUM TRANSPORT WITH VARIABLE TEMPERATURE AND OXIDATION POTENTIAL: THE COMPUTER PROGRAM THCC

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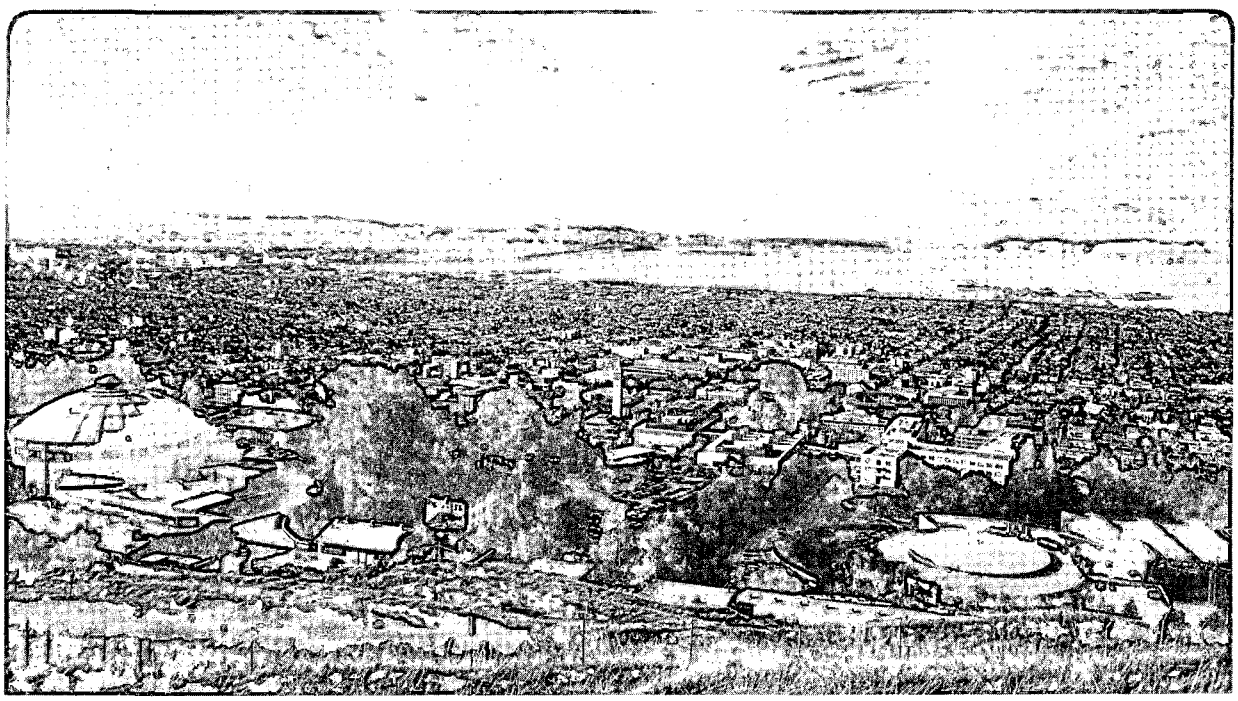

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SIMULATION OF URANIUM TRANSPORT WITH VARIABLE TEMPERATURE AND OXIDATION POTENTIAL: THE COMPUTER PROGRAM THCC

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ABSTRACT

A simulator of reactive chemical transport has been constructed with the capabilities of treating variable temperatures and variable oxidation potentials within a single simulation. Homogeneous and heterogeneous chemical reactions are simulated at temperature-dependent equilibrium, and changes of oxidation states of multivalent elements can be simulated during transport. Chemical mass action relations for formation of complexes in the fluid phase are included explicitly within the partial differential equations of transport, and a special algorithm greatly simplifies treatment of reversible precipitation of solid phases. This approach allows direct solution of the complete set of governing equations for concentrations of all aqueous species and solids affected simultaneously by chemical and physical processes. Results of example simulations of transport, along a temperature gradient, of uranium solution species under conditions of varying pH and oxidation potential and with reversible precipitation of uraninite and coffinite are presented. The examples illustrate how inclusion of variable temperature and oxidation potential in numerical simulators can enhance understanding of the chemical mechanisms affecting migration of multivalent waste elements.

INTRODUCTION

Assessment of compliance of a nuclear waste repository with regulatory standards requires the ability to account for interaction between chemistry, temperature, and mass transport. The interaction of heat flow with chemical reactions can affect the transport of chemically reactive solutes by the influence of changing temperature on chemical reaction rates and equilibria. The oxidation states and consequent chemical behavior of multivalent chemical elements are determined thermodynamically by temperature, oxidation potential, and, in many cases, hydrogen ion activity of the fluid phase. The ion activity product of water varies with temperature and so does the relation between pH and hydroxide ion activity.

Numerical simulation of migration of chemically reactive materials has received increased attention in recent years. Considerable progress has been made in the development of thermodynamically based computer programs that incorporate chemical reac-

tions, under the assumption of chemical equilibrium, into the solution of multicomponent solute transport equations. This has been accomplished, in general, by two numerical approaches. The "two-step" method iterates between solutions of the transport equations and separate solutions of the usually nonlinear algebraic equations of chemical equilibrium (e.g., [1,2,3]). The "direct" method solves the two equation sets simultaneously (e.g., [4,5,6,7,8]). Both the two-step and direct methods provide strong interaction between chemical reactions and solute transport processes (advection and diffusion). However, the current programs are constrained to operate under conditions of constant temperature (although temperature can be varied from simulation to simulation) and constant (or unspecified) oxidation potential.

It appears likely that gradients of temperature will be established quickly within packing, backfill, and geological materials near waste canisters, and eventual mixing of waste-bearing fluids with ambient fluids of different temperatures and compositions will occur. In addition, certain important waste materials (e.g., the actinide elements) can exist in several oxidation states that exhibit distinctly different chemical behaviors. Therefore, inclusion of variable temperature and oxidation potential in numerical simulators should provide increased capability for understanding the mechanisms affecting waste material migration.

A computer program named THCC (Thermo-Hydro-Chemical Coupling) simulating reactive chemical transport with temporal and spatial variations of temperature and oxidation potential is described here. Examples of simulations with the THCC program are presented.

DESCRIPTION OF THE THCC COMPUTER PROGRAM

The THCC program began as an extension of the capabilities of the CHEMTRN program [6,7], a thermodynamically based simulator of multicomponent, reactive chemical transport using the direct method of solution. However, evolution during development of THCC resulted in significant differences between the two programs in addition to the new capabilities.

Basic Functions

The THCC program simulates transport of reactive chemical species by advection with constant pore fluid velocity and by hydrodynamic dispersion or chemical diffusion in one-dimensional or cylindrically symmetric geometry. The types of chemical reactions simulated are complexation, oxidation-reduction, and dissociation of water in the aqueous phase and reversible precipitation of thermodynamically stable solid phases. Chemical reactions are described by thermodynamic mass action relations among activities of participating species. Activity coefficients are estimated by the Davies equation [9]. Equilibrium constants are temperature-dependent and are calculated from thermodynamic expressions. Simulations can be done of systems with fixed gradients of temperature, of systems with evolving thermal fields due to mixing of fluids having different temperatures, and of isothermal systems.

The set of unknowns consists of concentrations of basis species in the aqueous phase and pseudo-concentrations of precipitated solid phases. The basis species are the minimum set of aqueous species needed to describe all simulated chemical reactions plus additional nonreactive species that may be required to establish the ionic strength at a specified value. The "concentration" of a solid is the quantity (moles) of solid per unit volume of fluid phase present at any location. All aqueous species are assumed to have equal diffusion coefficients. Therefore, the concentration of each aqueous complex appearing in spatial derivatives in the governing equations of transport is replaced by a nonlinear expression, derived from a mass action relation, containing concentrations of basis species that react to form the complex. There is one transport equation for each basis species, and the transport equation for a reactive basis species involves concentrations of other basis species. The set of equations necessary to determine all unknowns is completed by a set of mass action relations, one for each solid phase included in the simulation. The treatment of equations for solid phases is described in the section, "Method of Solution".

Heat Transport

During simulations in which mixing of two fluids having different temperatures occurs, the equation of heat transport is solved first at each new time level, and new temperatures are assigned at each point in space corresponding to a finite-difference node. These temperatures and the equilibrium constants calculated from them are held constant during the subsequent iterative solution of the transport and mass action equations at the new time. Currently it is assumed that no interaction exists between temperature and fluid flow and that heats of chemical reactions do not affect the distribution of temperature. Thus, heat transport is decoupled mathematically from chemical transport and equilibrium. The equation of heat transport is solved numerically; the numerical solutions have been verified by comparison to analytical solutions.

Oxidation-Reduction Reactions

Oxidation-reduction reactions are treated by defining a hypothetical electron activity as a basis species subject to transport as are other aqueous basis species. For multivalent elements, a species in the highest oxidation state is chosen to be the basis species. Reduction to a lower oxidation state is described formally by a half-cell reaction in which the higher-valent species "reacts" with hypothetical electrons to form the lower-valent species. This process is analogous to the process by which aqueous complexes are formed by reactions between basis species. Thus, the reduced species are treated as though they are complexes of oxidized species with electrons. The oxidation potentials of initial and influent or boundary fluids are specified at input; the potentials are converted to equivalent electron activities by the relationship

$$[e] = \exp\left(-\frac{F\,Eh}{R\,T}\right) \tag{1}$$

where $[e]$ is electron activity, F is the Faraday constant, R is the gas constant, T is

absolute temperature, and Eh is the oxidation potential on the hydrogen scale. It is noted that the electron activity is an artifice that allows treatment of reduction in the same manner as complexation; there is no implication that free electrons exist in aqueous solution.

Method of Solution

The partial differential equations of heat and chemical transport are expressed as finite-difference equations using the Crank-Nicolson scheme to achieve second-order accuracy in time [10]. At each new time level, the heat equation is solved separately by LU decomposition of the resulting tridiagonal matrix of coefficients. As stated earlier, the unknowns in the chemical equations are concentrations of basis species and solids. Values of these unknowns are found iteratively by the Newton-Raphson method [11]. Residue equations for the basis species are formed by algebraically summing all terms in the finite-difference forms of the transport equations. Residue equations for the solids are formed in the following manner. At each finite-difference node the activity product (Q) of basis species formed by dissolution of a solid is computed and compared to the theoretical solubility product (K) for the solid. If the solid is present at the node, or if the solid is not present and Q is greater than K, then the residue for the solid at the node is set equal to the algebraic difference, Q minus K. On the other hand, if Q is less than or equal to K and the solid is not present at the node, the residue is set equal to zero. This procedure provides a residue equation for each solid at each node and eliminates the need to change the number of unknowns at nodes where solids have precipitated or dissolved. The procedure is considerably simpler to implement than the procedure used previously in the CHEMTRN program [6].

The residues (one for each unknown at each node) are reduced to "zero" (a small number whose upper limit of absolute magnitude is determined by an input error tolerance) iteratively by computing corrections to current values of the unknowns. Elements of the Jacobian matrix are computed from analytical expressions rather than estimated by divided differences as is done in the CHEMTRN program. The system of equations to be solved for the corrections has block tridiagonal form and is solved by use of a published software routine [12].

Convergence in the Newton-Raphson procedure provides a set of consistent values of concentrations of basis species and solids at the current time level. These concentrations simultaneously satisfy the finite-difference forms of the transport equations and the mass action relations. Concentrations of aqueous complexes and reduced species are then calculated using the final basis species concentrations.

THERMOCHEMICAL DATA

Equilibrium constants for chemical reactions must be calculated at each finite-difference node. This must be done at each new time level during simulations with evolving thermal fields, but need be done only once at the beginning of isothermal simulations and simulations involving constant gradients of temperature. The equilibrium constants

for a given reaction are calculated from the thermodynamic relation of Clarke and Glew [13]:

$$\log K(T) = A + \frac{B}{T} + C \log T + DT \tag{2}$$

where $K(T)$ is an equilibrium constant evaluated at absolute temperature T and the coefficients A, B, C, and D are related to the changes of standard Gibbs free energy, enthalpy, and heat capacity for the reaction. The coefficients used in the examples to be shown here were obtained by fitting $\log K$ values at discrete temperatures in the range 298 – 523 °K to Eq. (2). The $\log K$ values were taken from the Aqueous Solutions Data Base compiled by Phillips et al. [14], supplemented by data from other sources [15,16]. The coefficients for each reaction in a simulation are supplied as input to the program. Table I lists the chemical reactions and coefficients for Eq. (2) that were used in the example calculations presented in this paper. Preliminary calculations showed that, in the examples considered here, other possible complexes (e.g., polymeric hydroxides of U(VI) and products of partial hydrolysis of U(IV)) would be present only at negligible concentrations relative to the complexes listed in Table I. Therefore, the other complexes were omitted from the example calculations.

Table I. Chemical Reactions and Coefficients for Equation (2)

Reactions	A	B	C	D
formation of complexes:				
$H^+ + CO_3^{2-} = HCO_3^-$	−77.11	4593.3	29.04	5.944×10^{-4}
$2H^+ + CO_3^{2-} = H_2CO_3$	−40.37	5576.6	11.33	3.466×10^{-2}
$UO_2^{2+} + OH^- = UO_2OH^+$	83.00	−1107.2	−32.46	3.067×10^{-2}
$UO_2^{2+} + CO_3^{2-} = UO_2CO_3^0$	−275.36	13160.7	97.67	-1.221×10^{-3}
$UO_2^{2+} + 2CO_3^{2-} = UO_2(CO_3)_2^{2-}$	−59.82	2717.8	27.54	-1.091×10^{-3}
$UO_2^{2+} + 3CO_3^{2-} = UO_2(CO_3)_3^{4-}$	−393.77	20160.4	139.90	2.087×10^{-3}
reduction:				
$UO_2^{2+} + e^- = UO_2^+$	−356.20	13420.1	133.24	-5.502×10^{-2}
$UO_2^{2+} + 2H_2O + OH^- + 2e^- = U(OH)_5^-$	−406.50	18088.5	152.77	-7.661×10^{-2}
dissociation of water:				
$H_2O = H^+ + OH^-$	−76.99	−1506.4	31.25	-3.110×10^{-2}
dissolution of solids:				
$UO_{2(c)} = UO_2^{2+} + 2e^-$	644.66	−26101.3	−243.74	1.100×10^{-1}
$SiO_{2(am)} + 2H_2O = Si(OH)_4^0$	0.338	−840.1	0.	-7.889×10^{-4}
$USiO_{4(c)} + 2H_2O = UO_2^{2+} + Si(OH)_4^0 + 2e^-$	646.70	−27639.2	−244.23	1.089×10^{-1}

EXAMPLE CALCULATIONS

The THCC program was used to simulate transport of aqueous uranium species within a porous matrix in the presence of a radial temperature distribution varying from 90 °C at the inner boundary to 50 °C at a distance of 10 m. Dissolved uranium is transported from two different constant-concentration sources in a field of flow having a pore fluid velocity of 3×10^{-8} m/s (approximately 1 m/y), with a dispersivity of 0.05 m and a diffusion coefficient of 10^{-11} m^2/s [17,18]. In all examples shown here, the fluid present in the porous matrix has an initial composition approximating a basalt groundwater, with pH equal to 10, Eh equal to −0.36 v at 50 °C, total carbonate equal to 9×10^{-4} molar, and ionic strength equal to 0.016 molar, and is saturated with respect to amorphous silica [19,20]. Both source fluids have 9×10^{-4} molar total carbonate and ionic strength equal to 0.016 molar, but no silicic acid. Both source fluids are assumed in equilibrium with solid uraninite (U(IV) dioxide). One source fluid ("reduced") has Eh equal to −0.4 v at 90 °C and pH equal to 10 to simulate dissolution of uraninite by a fluid in equilibrium with basalt. The other source fluid ("oxidized") has Eh equal to 0.0 v at 90 °C and pH equal to 6 to simulate dissolution of uraninite by a more oxidizing fluid.

Two simulations were done with each source fluid. In one case, the only solid phase allowed to precipitate reversibly is uraninite; the presence of silicic acid in the initial groundwater is ignored. In the other case, both uraninite and coffinite (U(IV) silicate) are allowed to precipitate. Comparison of the two cases for each source fluid will demonstrate the effects on uranium concentration profiles of precipitation reactions during transport.

Each simulation ran to a simulated time of 10^7 s (approximately 0.3 y). Results of the calculations at this time are shown in Figures 1 and 2. Figure 1 shows the temperature profile, constant during all simulations, and profiles of oxidation potential (Eh) resulting

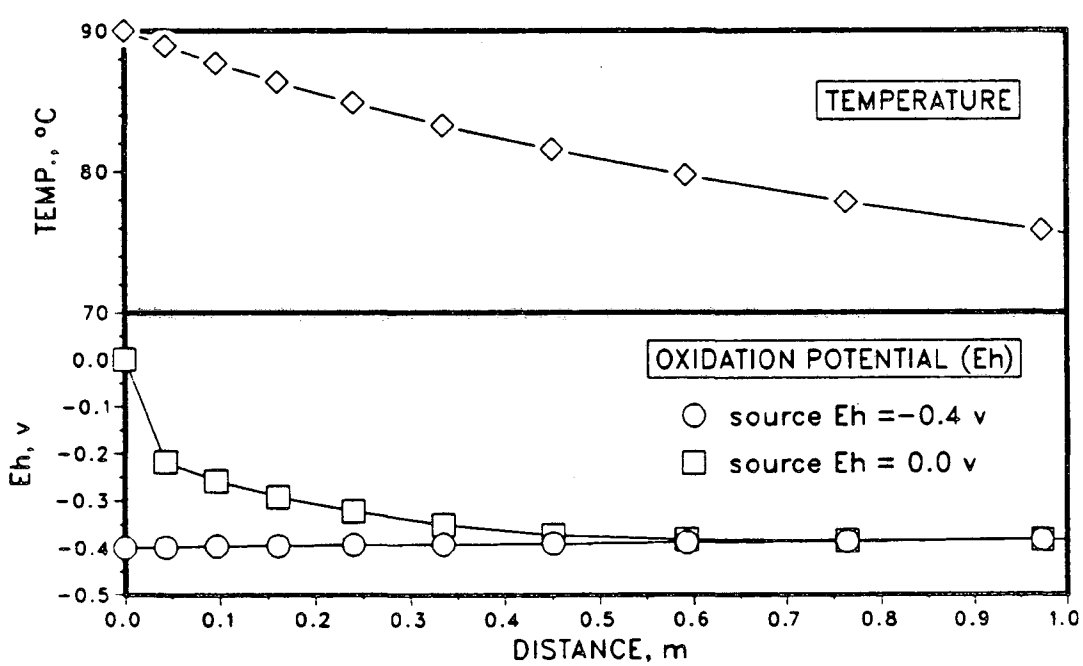

Figure 1. Temperatures and oxidation potentials at 10^7 s.

from transport, mixing, and chemical reactions of the two source fluids with the initial groundwater. Figure 2 shows profiles of hydroxide ion concentrations from the two source fluids, profiles of total dissolved uranium concentrations, and theoretical solubilities of uraninite and coffinite calculated for prevailing values of pH, Eh, and temperature. Filled symbols in the uranium concentration curves designate finite-difference nodes where precipitates of uraninite or coffinite exist.

Plots 1 and 2 in Figure 2 are uranium concentration profiles for the reduced source fluid, and plot 5 is the accompanying hydroxide concentration profile. The only significant uranium species in the reduced source fluid is the U(IV) pentahydroxide complex. In

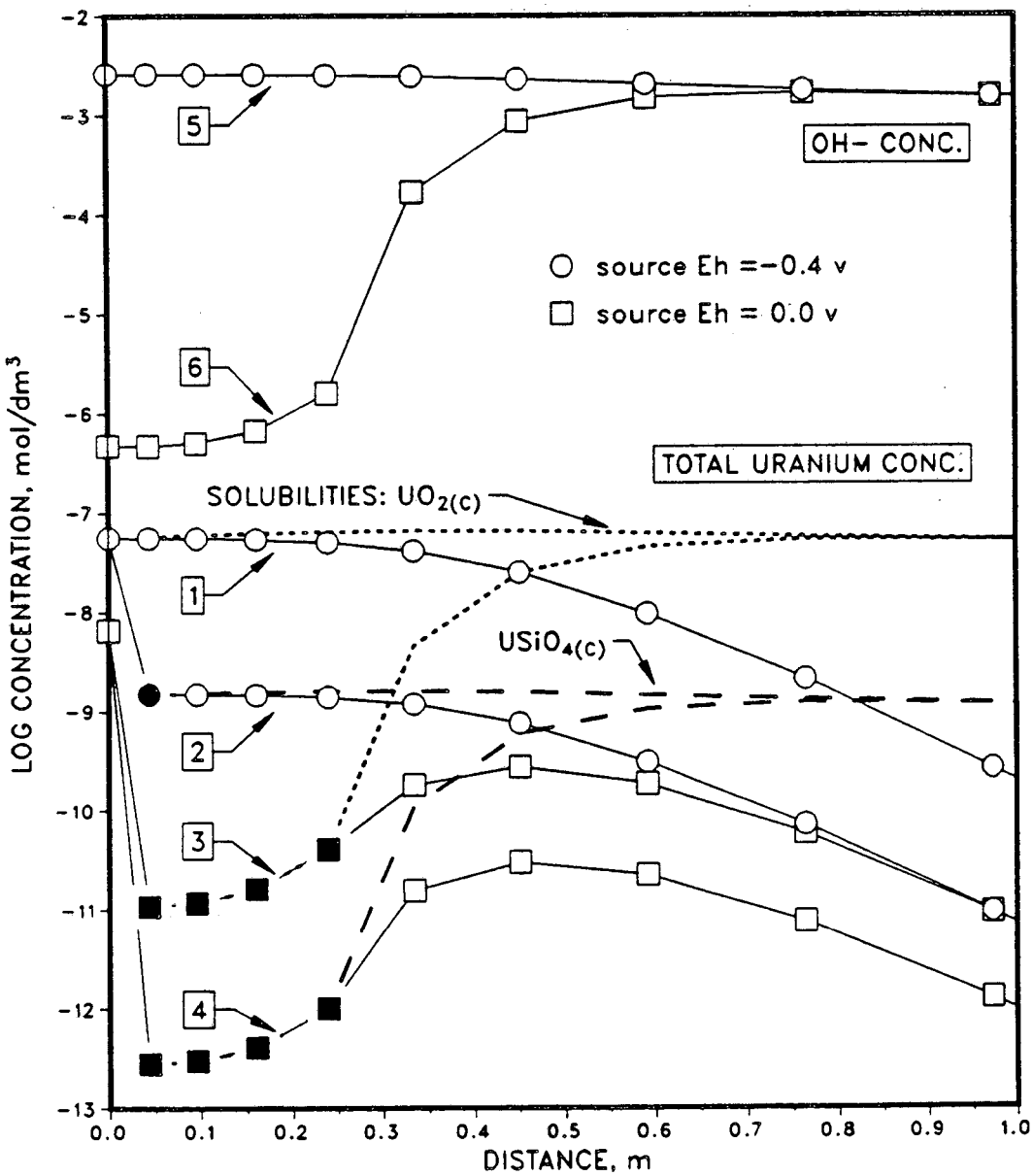

Figure 2. Total uranium concentrations, hydroxide concentrations, and solubilities of uraninite and coffinite at 10^7 s.

simulation 1, no reaction with silicic acid is simulated, and uranium is transported without precipitation of either uraninite or coffinite. In simulation 2, dissolved uranium can react with silicic acid, and coffinite has precipitated at the first node beyond the source. The concentration of dissolved uranium has decreased by a factor of 40 at this node; thereafter, the concentration distribution follows the shape of plot 1, but reduced in magnitude by this factor. In these two simulations, neither the oxidation potential nor the hydroxide concentration changes significantly between source fluids and groundwater; the reduction of uranium concentration in simulation 2 is caused only by reaction with silicic acid in the groundwater.

Plots 3 and 4 in Figure 2 are uranium concentration profiles for the oxidized source fluid. The accompanying profile of oxidation potential is shown in Figure 1 and of hydroxide ion as plot 6 in Figure 2. The uranium species in the oxidized source fluid are predominantly the mono- and bis-carbonato complexes of the U(VI) dioxo ("uranyl") cation with smaller concentrations of the uranyl monohydroxide complex, the U(V) dioxo cation, and the U(IV) pentahydroxide complex. It is noted that the total uranium concentration in the oxidized source fluid is about an order of magnitude lower than that in the reduced source fluid. In simulation 3, in which reaction with silicic acid is not simulated, uraninite has precipitated at the first four nodes beyond the source. This is caused by the decrease of oxidation potential at these nodes with the result that uraninite is more stable here than it is in the source fluid. Thus at the first node beyond the source, the dissolved uranium concentration has decreased by a factor of 500. The dominant solution species at this and higher nodes is the U(IV) pentahydroxide complex. In simulation 4, reaction between silicic acid and U(IV) is allowed and coffinite (more stable here than uraninite) has precipitated at the first four nodes beyond the source, further lowering dissolved uranium concentrations by an additional factor of 50, relative to the source concentration.

In both simulations 3 and 4 at 10^7 s, a minimum in the concentration profile of dissolved uranium has developed near the source. This has happened because at earlier times dissolved uranium was transported into the porous matrix at concentrations such that the solubilities of neither uraninite nor coffinite were exceeded in the near-source region. Under these conditions, the concentration profile of dissolved uranium decreased monotonically with distance from the source, as in the case of simulation 1. As time passed, high electron activities and silicic acid in the groundwater (which were displaced downstream by the advective flow from the source) diffused upstream until one or the other solubility product was exceeded near the source, and either uraninite or coffinite precipitated. With further passage of time, activities of electrons and silicic acid continued to increase at the nodes where precipitation occurred. Consequently, uranium concentrations at these nodes have been depressed by more precipitation in order to satisfy the constraints imposed by the solubility products. This process has resulted in the interesting phenomenon that at 10^7 s dissolved uranium in the region less than 0.45 m from the source diffuses in the upstream direction toward the zone of precipitation. This effect will further reduce the outward flow of uranium from regions downstream from the zone of precipitation.

The effects of temperature variation are not obvious in Figure 2, but nonetheless are present. For example, over the range of temperature shown in Figure 1, the equilibrium

constants for dissolution of uraninite and coffinite to form the U(IV) pentahydroxide complex change by 65 percent and 13 percent, respectively; both constants decrease with increasing temperature.

CONCLUDING REMARKS

The THCC program shares several shortcomings with its progenitor, the CHEMTRN program, as well as with other existing programs for simulating transport of reactive chemicals. However, certain numerical aspects of the transport and precipitation calculations are improved in THCC, and the program provides new capabilities with regard to variability of temperature and oxidation potential.

Prominent among the shortcomings of the THCC program is the assumption of equilibrium in groundwater chemical reactions generally, and in oxidation-reduction reactions particularly. Real groundwaters have been shown to be usually in a state of disequilibrium with respect to oxidation-reduction reactions [21], with the result that such waters often cannot be characterized by a single oxidation potential. A partial resolution of this problem has been effected in the EQ3NR speciation program, which uses multiple, "couple-specific" oxidation potentials [22]. Another prominent shortcoming of the THCC program is the assumption of constant pore fluid velocity. This assumption excludes any feedback from heterogeneous chemical reactions and temperature variations to fluid flow. A considerable obstacle to improving this situation is the lack of a general formulation or theory relating changes of porosity to changes of permeability in porous media. Other present shortcomings are the absence of a routine for calculating activity coefficients in fluids of high salinity, the lack of corrections to equilibrium constants to account for variable ionic strength, and the inability to treat nonequilibrium chemical processes including growth and decay of radioactive decay chain members.

On the other hand, the use of the Crank-Nicolson numerical scheme to achieve second-order accuracy in time, the simplified algorithm for reversible precipitation of solid phases, and the calculation of Jacobian matrix elements from analytical expressions are improvements in THCC relative to methods used in CHEMTRN. More significantly, THCC provides a new capability to treat reactive chemical transport in variable fields of temperature and oxidation potential in a unified manner within a single computer program. It is anticipated that this capability will provide a means toward increased understanding of the mechanisms of release and migration of radioactive waste materials.

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