LBNL-41639

ERNEST ORLANDO LAWRENCE
BERKELEY NATIONAL LABORATORY

EOS9nT: A TOUGH2 Module for the Simulation of Flow and Solute/Colloid Transport

G.J. Moridis, Y.S. Wu, and K. Pruess

Earth Sciences Division

April 1998

Presented at the
TOUGH98 Workshop,
Berkeley, CA,
May 4–6, 1998,
and to be published in
the Proceedings

REFERENCE COPY
Does Not
Circulate

Bldg. 50 Library - Ref.
Lawrence Berkeley National Laboratory

LBNL-41639
Copy 1

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

LBNL-41639

EOS9nT: A TOUGH2 MODULE FOR THE SIMULATION OF FLOW AND SOLUTE/COLLOID TRANSPORT

G.J. Moridis, Y.S. Wu and K. Pruess

Earth Sciences Division
Lawrence Berkeley National Laboratory
University of California
Berkeley, CA 94720

April 1998

This work was supported by the Director, Office of Civilian Radioactive Waste Management, U.S. Department of Energy, through Memorandum Purchase Order EA9013MC5X between TRW Environmental Safety Systems, Inc., and the Ernest Orlando Lawrence Berkeley National Laboratory, under Contract No. DE-AC03-76SF00098.

EOS9nT: A TOUGH2 Module for the Simulation of Flow and Solute/Colloid Transport

G.J. Moridis, Y.S. Wu and K. Pruess

Earth Sciences Division, Lawrence Berkeley National Laboratory
University of California, Berkeley, CA 94720

Abstract
EOS9nT is a new TOUGH2 module for the simulation of flow and transport of an arbitrary number *n* of tracers (solutes and/or colloids) in the subsurface. The module first solves the flow-related equations, which are comprised of (a) the Richards equation and, depending on conditions, may also include (b) the flow equation of a dense brine or aqueous suspension and/or (c) the heat equation. A second set of transport equations, corresponding to the *n* tracers, are then solved sequentially. The low concentrations of the *n* tracers are considered to have no effect on the liquid phase, thus making possible the decoupling of their equations. The first set of equations in EOS9nT provides the flow regime and account for fluid density variations due to thermal and/or solute concentration effects. The *n* tracer transport equations account for sorption, radioactive decay, advection, hydrodynamic dispersion, molecular diffusion, as well as filtration (for colloids only). EOS9nT can handle gridblocks of irregular geometry in three-dimensional domains. Preliminary results from four 1-D verification problems show an excellent agreement between the numerical predictions and the known analytical solutions.

1. Introduction

EOS9nT is a module for the TOUGH2 general-purpose fluid and heat flow simulator [*Pruess*, 1991]. It is designed to simulate the flow of fluids and the transport of an arbitrary number *n* of independent tracers (solutes and/or colloids, SCs) in complex subsurface systems involving porous and/or fractured media. EOS9nT can simulate the following scenarios:

(F1) Isothermal flow of the aqueous phase, and SC transport at concentrations too low to have any measurable effect on the water density and the flow regime.
(F2) Isothermal flow of the aqueous phase, and SC transport at concentrations sufficiently high to affect the water density and the flow regime.
(F3) Non-isothermal flow of water, and SC transport at tracer levels (i.e., at low concentrations).
(F4) Fully-coupled non-isothermal flow of water under variable density conditions, and SC transport.

2. Design, Approach and Capabilities

2.1. Modeled Processes

EOS9nT allows the modeling of the following processes:

(1) Flow of the liquid phase under saturated and/or unsaturated conditions and with variable fluid density (due to concentration, thermal and pressure effects)
(2) Transport of *n* SCs, which accounts for advection, molecular diffusion, hydrodynamic dispersion, sorption (linear, Langmuir and Freundlich), radioactive decay, and filtration (for colloids only).
(3) Heat transport, which includes advective and diffusive effects.

2.2. Assumptions and Governing Equations

Under the F1 flow regime, EOS9nT solves a single equation, the *Richards* [1931] equation, which describes the flow of water in the subsurface under saturated or unsaturated conditions. The flow of the gaseous phase is not considered. The assumptions for the F1 regime include:

(A1) The water flow is isothermal.
(A2) The concentration of the SCs is at a tracer level, i.e., too low to have any measurable effect on the flow regime.
(A3) The pressure of the gaseous phase does not deviate significantly from the reference pressure of the system.
(A4) There is no phase change.
(A5) No chemical reactions occur between the rock and the tracers, or among the tracers.

(A6) In colloid transport, the porosity and permeability of the medium are unaffected by the colloid filtration.

These assumptions allow decoupling of the flow and transport equations. The Richards equation is first solved, followed by the sequential solution of the n independent tracer transport equations.

The F2 regime differs from F1 in that assumption (A2) no longer holds true. This necessitates the solution of a SC transport equation in addition to the Richards equation, for a total of 2 coupled equations. The SC equation corresponds to the dominant species (e.g., the heavy brines, but not the accompanying radioactive tracers, in the wastes leaking from the Hanford tanks) which controls the water density dependence, and accounts for all the aforementioned phenomena. In the F3 regime, assumptions (A2) through (A6) apply, and the heat transport equation must be solved coupled with the Richards equation.

The F4 regime represents the most general scenario tractable by EOS9nT, in which the valid assumptions are (A3) through (A6). F3 necessitates the solution of 3 coupled equations, i.e., (a) the Richards equation, (b) the transport equation of the dominant species, and (c) the heat transport equation. Following the solution of the coupled equations in F2, F3 or F4, the n independent tracer transport equations are solved sequentially.

EOS9nT cannot be used for the simulation of systems in which the gas pressure is substantial, phase changes (i.e., evaporation and condensation) occur, and/or the tracer is gaseous or volatile. Such problems are tractable by the companion module EOS3nT (currently in development) which can handle gas and liquid flows in addition to all the EOS9nT capabilities.

2.3. The Time-Stepping Process

Under transient flow conditions, the time-stepping process in EOS9nT is shown in **Figure 1**, in which Δt_f is the time step in the flow equation (or the coupled equations) and $\Delta t_{t,i}$ is the time step in the transport equation of tracer i. A total of $n+1$ separate simulation times are independently tracked.

After the flow solution at $t+\Delta t_f$ is obtained, the water saturations, flow velocities and flow rates across the element interfaces are recorded. The time is then reset to t_0 in the transport equation of tracer i, and the maximum allowable $\Delta t_{t,i}$ is determined from limitations on (a) the grid Courant number, (b) the grid Peclet number, and (c) the tracer half life. The tracer transport in the Δt_f interval is simulated using a total of $m_i=\Delta t_f/\Delta t_{t,i}$ (i.e., the next largest integer of the ratio) equal time steps. The process is repeated for all n independently-tracked tracers. The time step sizes of the various tracers are not generally equal.

As the flow field becomes invariant, the size of the Δt_f in TOUGH2 increases, and convergence is achieved after a single Newtonian iteration. Contrary to the standard TOUGH2, which prints the invariant solution and stops the simulation after 10 such time steps of convergence on the first iteration, involves, EOS9nT prints the flow solution and continues solving the tracer equations. The time step in the solute transport equations becomes uniform and equal to the shortest of the $\Delta t_{t,i}$ after the flow has reached a steady state.

EOS9nT also allows solute transport simulations when the initial flow field is time-invariant. In this case, EOS9nT solves the flow field only once in order to obtain the constant water saturations, flow velocities and flow rates across the element interfaces. After printing the flow data, only the tracer transport equations are solved.

Time-step control in the flow equation (or the coupled equations) is provided internally by the Newtonian convergence criteria employed in TOUGH2 [*Pruess*, 1991]. The time-step size in the n transport equations is specific to each tracer, and is controlled by inputs which place limitations on the grid: (a) Peclet number, (b) Courant number and (c) the maximum allowable fraction of the SC half-life $T_{1/2}$ (if radioactive) for limiting the $\Delta t_{t,i}$.

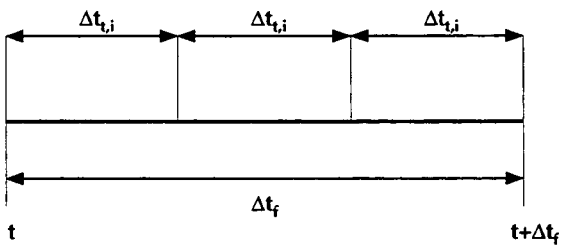

Figure 1. The time steps for flow and transport in EOS9nT.

2.4. Treatment of the Dispersion Tensor

In the treatment of the general 3-D dispersion tensor, EOS9nT follows closely the approach of the radionuclide transport module T2R3D [*Wu et al.*, 1996] for TOUGH2. Velocities are averaged by using the *'projected area weighting method'*, in which a velocity component $v_{n,i}$ (i=x,y,z) of the vector v_n is determined by vectorial summation of the components of all local connection vectors in the same direction, weighted by the projected area in that direction. Mass fraction gradients of the n SCs are evaluated using *the 'interface area weighting scheme'* [*Wu et al.*, 1996].

This approach allows the solution of the transport problem in irregular grids. EOS9nT provides an option for the removal of numerical dispersion by using the corrections proposed by *Lantz* [1971. These involve an adjustment of the dispersion coefficient, which for 1-D problems is

$$D_C = D - \frac{1}{2}U\Delta x - \frac{1}{2}\frac{U^2\Delta t}{\phi},$$

where D_C and D are the corrected and the original dispersion coefficients, respectively, U is the Darcy velocity, ϕ is the medium porosity, and Δx is the grid size.

2.5. Treatment of Colloidal Filtration

Colloidal particles moving through porous media are subject to filtration, the mechanisms of which have been the subject of several investigations, e.g., *Herzig et al.*, 1970; *Wnek et al.*, 1975; *Tien et al.*, 1979; *Corapcioglu et al.*, 1987. EOS9nT assumes conditions of *'deep filtration'*, i.e., no interaction between the colloidal particles and no effects on porosity and permeability. Under these conditions, the variation in the concentration of the retained colloids is described by the equation [*Herzig et al.*, 1970; *Dieulin*, 1982]

$$\frac{\partial\sigma}{\partial t} = \lambda UX,$$

where σ is the concentration of the retained colloid, λ is the filter coefficient, and X is the colloid mass fraction in the liquid. The linearity of the colloid equation allows its combination with the colloid transport equations. If no experimental values are available, EOS9nT computes an estimate of the parameter λ as a function of the particle and grain sizes, using the relationships of *Tien et al.* [1979].

2.6. Other Capabilities of EOS9nT

EOS9nT can simulate the transport of any combination of solutes and colloids. As with all other members of the TOUGH2 family of codes [*Pruess*, 1995], EOS9nT can handle multi-dimensional flow domains and cartesian, cylindrical or irregular grids. Initialization is possible using (a) pressure, (b) water saturation or (c) capillary pressure, and EOS9nT has the capability of determining the initial pressure and/or water saturation distribution (gravity-capillary equilibrium) in relation to an initial, areally variable, watertable elevation map.

Heterogeneity is described through the use of domain permeability modifiers, which can be externally supplied or internally generated using linear or logarithmic modifications based on random numbers. Scaling of the capillary pressures is then obtained by using the *Leverett* [1941] function.

Parameters for linear, Langmuir and Freundlich sorption isotherms are provided for all the combinations of rock/soil types and SCs.

3. Verification of EOS9nT

Four sets of simulations were conducted to verify EOS9nT against known analytical solutions of flow and solute transport. The first three test problems involved saturated regimes and invariant flow fields. The fourth test problem involved unsaturated flow and transport. The maximum allowable Peclet and Courant numbers were 2 and 1 respectively, and, when radioactive SCs were involved, the $\Delta t_{t,i}$ was not allowed to exceed $0.05T_{1/2}$. In all the verification problems, numerical dispersion was removed by using the *Lantz* [1971] corrections. It must be pointed out that the four test problems do not cover the whole range of the EOS9nT capabilities, and only reflect to-date results of a continuing effort.

3.1. Test Problem 1

This test problem involves the distribution of five independent solutes being transported at a constant pore velocity V = 0.1 m/day in a semi-infinite horizontal column of a medium with a dispersion coefficient D = 0.1 m^2/day. The solutes do not decay (λ = 0). The retardation factors R

$$R = 1 + \frac{1-\phi}{\phi S_w}\rho_R K_d$$

of the five tracers in the porous medium were 1, 2, 3, 4 and 5. In this equation ϕ = 0.3, S_w is the water saturation (=1), ρ_R is the rock

density, and K_d is the distribution coefficient. A uniform grid size of $\Delta x = 1$ m was used.

Figure 2 shows a comparison between the numerical predictions (symbols) and the analytical solutions [*Bear*, 1979] at $t = 400$ days. An excellent agreement between the two sets of solutions is observed.

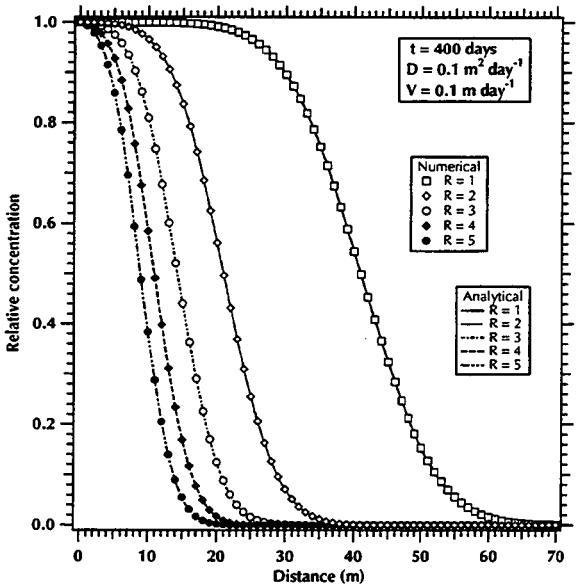

Figure 2. Analytical and numerical solutions of the concentration distribution of 5 non-radioactive tracers of variable R in a semi-infinite column (Test problem 1).

3.2. Test Problem 2

The second test problem involved the transport of three radionuclides in a saturated semi-infinite horizontal column. Water flowed at a constant $V = 0.2$ m/day, and the dispersion coefficient $D = 0.05$ m^2/day. The first radionuclide had a $R = 1$ and a $T_{1/2} = 69.32$ days. The second radionuclide differed from the first in that $R = 2$. The third radionuclide had a $R = 1.5$ and a $T_{1/2} = 693.2$ days. The porosity and discretization were the same as in Test Problem 1.

Figure 3 shows a comparison between the numerical predictions (symbols) and the analytical solutions of [*Bear*, 1979] at $t = 200$ days. The two sets of solutions are practically identical.

3.3. Test Problem 3

The third test problem involved the transport of three non-sorbing, non-radioactive colloids in a saturated semi-infinite horizontal column. Water flowed at a constant Darcy velocity of $U = 2$ m/day, $D = 1$ m^2/day, and $\phi = 0.3$. The filter coefficient λ of the three colloids were 30 m^{-1}, 100 m^{-1}, and 3000 m^{-1}. A uniform grid size of $\Delta x = 0.01$ m was used. At $t = 7600$ s, the analytical and numerical solutions in **Figure 4** practically coincide.

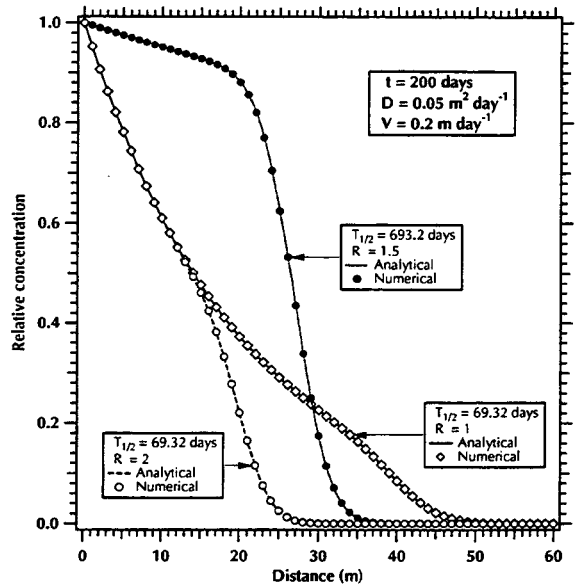

Figure 3. Analytical and numerical solutions to the problem of transport of three radioactive tracers flowing in a semi-infinite column (Test problem 2).

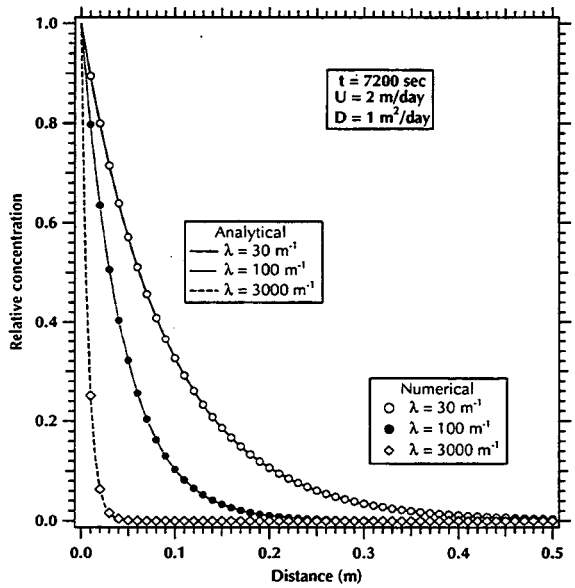

Figure 4. Analytical and numerical predictions of the concentrations of three non-sorbing, non-radioactive colloids (Test problem 3).

3.4. Test Problem 4

Test problem 4 was the horizontal infiltration problem originally solved by *Philip* [1955]. The problem was described by *Ross et al.* [1982], and was featured as Sample Problem No. 2 in the TOUGH User's Guide [*Pruess,* 1987]. A semi-infinite horizontal tube filled with a homogeneous soil is partially saturated with water. The soil porosity is $\phi = 0.45$, and the initial moisture content is $\theta = 0.2$, corresponding to a liquid saturation of $S_w = \theta/\phi = 0.44$. The liquid saturation at the $x=0$ boundary is held constant at $S_w = 1$ for $t>0$. Due to a capillary pressure differential, water infiltrates into the horizontal system. Air is considered a passive phase, and its effects are neglected.

The problem was augmented by adding two tracers to the water at the x=0 boundary. The mass fraction of the first and second tracers were 10^{-3} and 10^{-4}, respectively. The first tracer was non-decaying, and had a $R = 2$. The second tracer was non-sorbing (R =1) and had a $T_{1/2} = 1$ day.

TOUGH2 predictions using the EOS9nT module were obtained for $t = 0.01$ day, $t = 0.06$ days, and $t = 0.11$ days, at which *Ross et al.* [1982] specify exact solutions. A uniform grid size of $\Delta x = 0.002$ m was used.

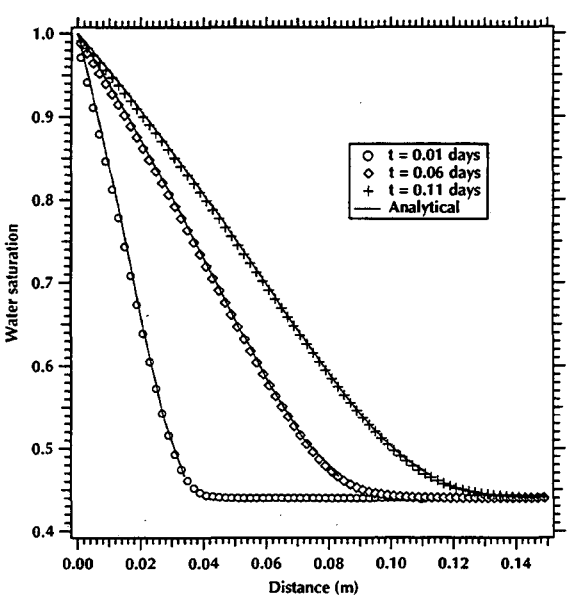

Figure 5. Analytical and numerical solutions of the water saturation distribution in Test problem 4.

In **Figure 5** we compare the numerical and analytical solutions [*Philip,* 1955] of saturation at the three observation times. The two sets of solutions are in excellent agreement. **Figures 6** and **7** show the EOS9nT solution of the concentration distributions of the two tracers at the same times. These solutions are practically identical with the ones obtained using TOUGH2 with the EOS7R module [*Oldenburg and Pruess,* 1995].

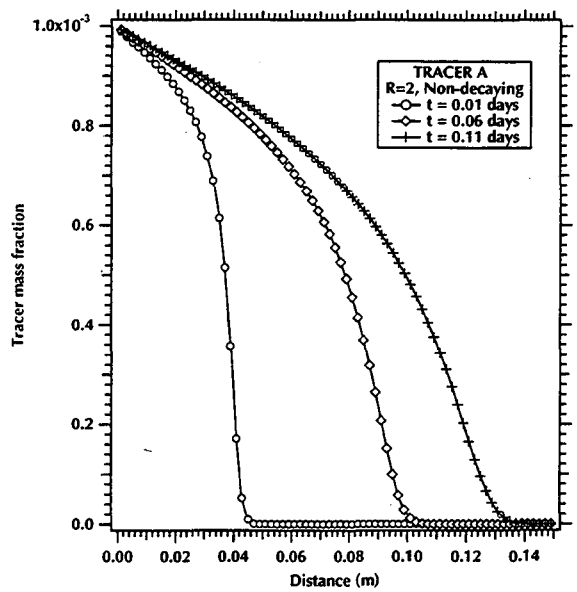

Figure 6. Numerically predicted concentration of a sorbing non-decaying tracer in Test problem 4.

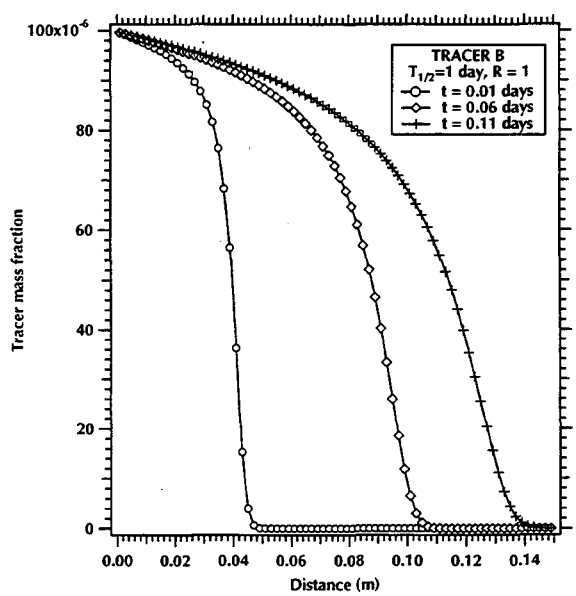

Figure 7. Numerically predicted concentration of a non-sorbing radioactive tracer in Test problem 4.

4. Conclusions

We evaluated the performance of the EOS9nT module for the TOUGH2 general-purpose fluid and heat flow simulator [*Pruess*, 1991] using a set of four 1-D test problems of flow and transport of solutes and colloids. The first three test problems, which involved SC transport in fully saturated media under steady-state flow conditions, have known analytical solutions. The fourth test problem, which involved flow and SC transport in an unsaturated medium, has a known analytical solution of saturation distribution, but no analytical solution of the SC concentration.

Comparison between the analytical solutions and the EOS9nT/TOUGH2 numerical predictions of the SC concentration distribution in the first three test problems showed that the two practically coincided. An excellent agreement was observed between the analytical and numerical solutions of saturation in the fourth test problem. In the same problem, the numerical results coincided with the numerical results obtained when using TOUGH2 with the EOS7R module [*Oldenburg and Pruess*, 1995], another module of flow and transport.

It must be clearly indicated that the evaluation results in this paper represent the (F1) flow regime only, are limited to comparisons with known solutions in 1-D systems, and do not correspond to the full range of capabilities of EOS9nT, as its evaluation is still in progress. A more thorough evaluation, including flow and transport problems in complex 2- and 3-D heterogeneous domains with variable liquid density (i.e., the (F2) and (F3) flow regimes), will be presented in a forthcoming report.

5. Acknowledgements

This work was supported by the Director, Office of Civilian Radioactive Waste Management, U.S. Department of Energy, through Memorandum Purchase Order EA9013MC5X between TRW Environmental Safety Systems, Inc., and the Ernest Orlando Lawrence Berkeley National Laboratory, under Contract No. DE-AC03-76SF00098. Drs. C. Oldenburg and S. Finsterle are thanked for their insightful review comments.

6. References

Bear, J., Hydraulics of Groundwater, McGraw-Hill, New York, 1979.

Corapcioglu, M.Y., N.M. Abboud and A. Haridas, Governing equations for particle transport in porous media, In: J. Bear and M.Y. Corapcioglu (eds.), *Advances in Transport Phenomena in Porous Media*, Series E: Applied Sciences Series No. 128, Martinus Nijhoff, Dordrecht, The Netherlands, 1987.

Dieulin, J., Filtration de colloides d' actinides par une colonne de sable argileux, *Report LHM/RD/82/83*, Paris School of Mines, Fontainebleau, 1982.

Herzig, J.P., D.M. Leclerc, and P. Le Goff, Flow of suspension through porous media, *Ind. Eng. Chem.* **62**(5), 129-157, 1970.

Lantz, R.B, Quantitative evaluation of numerical diffusion (truncation error), *Soc. Pet. Eng. J.*, **11**, 315-320, 1971.

Leverett, M.C., Capillary behavior in porous media, *Trans. AIME*, **141**, 341-358, 1941.

Oldenburg, C.M. and K. Pruess, EOS7R: Radionuclide transport for TOUGH2, *Rep. LBNL-34868*, Lawrence Berkeley Laboratory, Berkeley, CA, 1995.

Pruess, K, TOUGH User's Guide, *Rep. LBL-20700*, Lawrence Berkeley Laboratory, Berkeley, CA, 1987.

Pruess, K, TOUGH2 - A general-purpose numerical simulator for multiphase fluid and heat flow, *Rep. LBL-29400*, Lawrence Berkeley Laboratory, Berkeley, CA, 1991.

Pruess, K (ed), Proceedings of the TOUGH2 Workshop '95, *Rep. LBL-37200*, Lawrence Berkeley Laboratory, Berkeley, CA, 1995.

Philip, J.R., Numerical solution of equations of the diffusive type with diffusivity concentration dependent, *Transactions. Faraday Society*, **51**, 885-892, 1955.

Richards, L.A., Capillary conduction of liquids through porous mediums, *Physics*, **1**, 318-333, 1931.

Ross, B., J.W. Mercer, S.D. Thomas, and B.H. Lester, Benchmark problems for repository siting models, *NUREG/CR-3097*, Geotrans, Inc., Reston, VA, December 1982.

Tien, C., R.M. Turian and H. Pendse, Simulation of the dynamic behaviour of deep bed filters, *AICE J.*, **25**(3), 385-395, 1979.

Wnek, W.J., D. Gidaspow and D.T. Wasan, The role of colloid chemistry in modelling deep bed liquid filtration, *AICE J.*, **25**(3), 385-395, 1975.

Wu, Y.S. , C.F. Ahlers, P. Fraser, A. Simmons and K. Pruess, Software qualification of selected TOUGH2 modules, *Rep. LBNL-39490*, Lawrence Berkeley Laboratory, Berkeley, CA, 1996.

Ernest Orlando Lawrence Berkeley National Laboratory
One Cyclotron Road | Berkeley, California 94720

Prepared for the U.S. Department of Energy under Contract No. DE-AC03-76SF00098