Investigation of Scale and Heterogeneity Effects on Flow and Transport in Multiphase Systems

Final Report

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Statement of Problem Studied

Contamination of the subsurface environment by fluids that are immiscible with water is recognized as a commonplace occurrence. Subsurface systems that include immiscible fluids are multiphase systems; the phases present may include a solid, an aqueous, an immiscible organic, and a vapor. Fundamental processes that govern the behavior of such systems are complex. Several crucial questions remain to be answered for multiphase subsurface systems before a mature level of understanding is achieved. Among the most important of these unresolved issues are the effects of measurement scale, system dimensionality, and media heterogeneity on fundamental fluid flow and interphase mass transfer processes.

Most multiphase research to date has relied upon small-scale measurement of constitutive relations, assumptions of local equilibrium for solute distribution among phases, and the assumption of a homogeneous porous media. The purpose of this project was to investigate multiphase flow and interphase mass transfer processes as a function of scale in both homogeneous and heterogeneous porous media systems.

Summary of Most Important Results

A significant amount of new information has resulted from the work performed during this project, as is evidenced by the publications that have been attributed to this project. Some additional work is in process and I expect two or three additional papers from work completed on this project. Rather than discuss all aspects of this work, a few of the high points are summarized below.

1. Significant work was accomplished to elucidate the nonaqueous phase to aqueous phase dissolution process. This has included experimental and theoretical work that has shown that the size of a representative elementary volume (REV) for NAPL residual is relatively large compared to other properties such as porosity and grows rapidly with media heterogeneity (Mayer and Miller, 1992). The importance of this is that significant errors are expected if one uses continuum models, which are based on the assumption of the existence of an REV, for heterogeneous systems. It may well be that only stochastic approaches are reasonable for such systems, however essentially all work performed to date for multiphase systems is continuum based. This is fertile ground for continuing work that likely will be of far-ranging significance.
2. Putting the issues raised in item 1 aside, it is clear that NAPL-aqueous phase dissolution is fast for regions that are at residual saturation. Two aspects of this problem are worthy of further concern: dissolution to low concentrations typical of health-based standards, and dissolution from pools that result from subsurface heterogeneity. Partial insights into these issues are addresses in Mayer and Miller (1995), which is currently being revised and will be resubmitted in the near future.
3. It is now clear that mass transfer in the unsaturated zone is an extremely complex phenomena that is influenced by moisture characteristic history (Szatkowski et al., 1995). What we have found by examining data collected in our lab and others is that the majority of gas flow occurs through channels of large pores, which leads to diffusion-limited mass transfer resistance that is a function of moisture state and history.
4. X-ray methods developed in this project have been shown to offer an order of magnitude improvement in precision and accuracy compared to traditional dual-gamma approaches. These methods offer an exciting new approach, which is resulting in the production of data sets of a quality that can not be achieved using competing methods.
5. One application of the x-ray methods mentioned in item 4 has shown that mild heterogeneities in porous media systems exert a profound impact on fluid flow and species transport in multiphase systems. A set of intermediate-scale experiments fully describing these observations will be completed in the coming months. This finding is of profound significance, suggesting that small-scale variations can importantly effect appropriate constitutive relationships at larger scales—reinforcing the findings mentioned in item 1 for example. We will continue to explore this area in future work.

Publications and Technical Reports

Journals

1. Mayer, A.S., and C.T. Miller (1992) The Influence of Porous Media Characteristics and Measurement Scale on Pore-Scale Distributions of Residual Nonaqueous-Phase Liquids, Journal of Contaminant Hydrology, Vol. 11, No. 3/4, pp. 189–213.
2. Kandil, H., C.T. Miller, and R.W. Skaggs (1992) Modeling Long-Term Solute Transport in Drained Unsaturated Zones, Water Resources Research, Vol. 28., No. 10, pp. 2799–2809.
3. Mayer, A.S. and C.T. Miller (1993) An Experimental Investigation of Pore-Scale Distributions of Nonaqueous Phase Liquids at Residual Saturation, Transport in Porous Media, Vol. 10, No. 1, pp. 57–80.
4. Szatkowski, A., P.T. Imhoff, and C.T. Miller (1995) Development of a Correlation for Aqueous-Vapor Phase Mass Transfer in Porous Media, Journal of Contaminant Hydrology, Vol. 18, No. 1, pp. 85–106.
5. Mayer, A. S., and C. T. Miller (1995) The Influence of Mass Transfer Characteristics and Porous Media Heterogeneity on Nonaqueous Phase Dissolution, In Review.
6. McBride, J.F., and C.T. Miller (1995) Measurement of Phase Fractions in Porous Media by Using X-rays: 1. Theoretical Considerations and Optimization, In Review.
7. McBride, J.F., and C.T. Miller (1995) Measurement of Phase Fractions in Porous Media by Using X-rays: 2. Applications, In Review.
8. Barry, D.A., C.T. Miller, and K. Bajracharya (1995) Alternative Split-Operator Approach for Solving Chemical Reaction/Groundwater Transport Models, In Review.
9. Barry, D.A., C.T. Miller, and P.J. Culligan-Hensley (1995) Temporal Discretization Errors in Split-Operator Approaches to Solving Chemical Reaction/Groundwater Transport Models, In Review.

Proceedings and Book Chapters

1. Miller, C.T. and F.H. Cornew (1992) A Petrov-Galerkin method for resolving advective-dominated transport, Proceedings of Computational Methods in Water Resources IX, Denver, Colorado, Vol. 1 Numerical Methods in Water Resources, Edited by: T.F. Russell, R.E. Ewing, C.A. Brebbia, W.G. Gray, and G.F. Pinder, Computational Mechanics Publications, Southhampton and Boston, and Elsevier Applied Science, London and New York, pp. 157–164.
2. Mayer, A.S. and C.T. Miller (1992) Simulating nonaqueous phase liquid dissolution in heterogeneous porous media, Proceedings of Computational Methods in Water Resources IX, Denver, Colorado, Vol. 2 Mathematical Modeling in Water Resources, Edited by: T.F. Russell, R.E. Ewing, C.A. Brebbia, W.G. Gray, and G.F. Pinder, Computational Mechanics Publications, Southhampton and Boston, and Elsevier Applied Science, London and New York, pp. 247–254.

3. Miller, C.T., and C.T. Kelley (1994) A Comparison of Strongly Convergent Solution Schemes for Sharp-Front Infiltration Problems, Computational Methods in Water Resources X, Heidelberg, Germany, Vol. 1, Edited by: A. Peters, G. Wittum, B. Herrling, U. Meissner, C.A. Brebbia, W.G. Gray, and G.F. Pinder, Kluwer Academic Publishers, Dordrecht, The Netherlands, pp. 325–332.
4. Gilmore, P., C. Kelley, C. Miller, and G. Williams (1995) Implicit Filtering and Optimal Design Problems, In Press: Optimal Design and Control, J. Borggaard, J. Burkardt, M. Gunzburger, and J. Peterson (Editors), Birkhäuser, Boston, Massachusetts, pp. 159–176.

Published Abstracts and National Meeting Presentations

1. Kandil, H., R.W. Skaggs, and C.T. Miller (1991) Modeling long-term solute transport in the unsaturated zone, EOS Transactions, American Geophysical Union, Vol. 72., No. 44, p. 169. Presented at American Geophysical Union Fall Meeting, San Francisco, California, 9–13 December 1991.
2. Mayer, A.S., and C.T. Miller (1991) Pore size distribution and measurement scale effects on nonaqueous phase liquid morphology at residual saturation, EOS Transactions, American Geophysical Union, Vol. 72., No. 44, pp. 151–152. Presented at American Geophysical Union Fall Meeting, San Francisco, California, 9–13 December 1991.
3. Miller, C.T., M. Breve, and R.W. Skaggs (1991) Development and analysis of a method of lines solution for Richards' equation, EOS Transactions, American Geophysical Union, Vol. 72., No. 44, pp. 168–169. Presented at American Geophysical Union Fall Meeting, San Francisco, California, 9–13 December 1991.
4. Mayer, A.S., and C.T. Miller (1992) A Numerical Study of the Effects of Porous Media Heterogeneity and Nonequilibrium Relationships on Nonaqueous Phase Liquid Dissolution, EOS Transactions, American Geophysical Union, Vol. 73., No. 14, p. 133. Presented at American Geophysical Union Spring Meeting, Montreal, Canada, 11–16 May 1992.
5. Mayer, A.S., and C.T. Miller (1992b) Interactions Between Nonaqueous Phase Liquids and the Aqueous Phase as a Function of Observational Scale, Agronomy Abstracts, American Society of Agronomy. Presented at the Soil Science Society of America Annual Meeting, Minneapolis, Minnesota, 1–6 November 1992.
6. Miller, C.T., M. Breve, and R.W. Skaggs (1992) A Comparison of Adaptive Solutions for Solving Richards' Equation, EOS Transactions, American Geophysical Union, Vol. 73., No. 43, p. 240. Presented at American Geophysical Union Fall Meeting, San Francisco, California, 7–11 December 1992.
7. McBride, J.F., and C.T. Miller (1993) Dual-Radiation-Energy Fluid Saturation Measurement Using X-ray Attenuation, EOS Transactions, American Geophysical Union, Vol. 74., No. 16, p. 155. Presented at American Geophysical Union Spring Meeting, Baltimore, Maryland, 24–28 May 1993.
8. Kelley, C.T., C.T. Miller, and J.D. Grant (1993) A Comparison of Strongly Convergent Solutions Schemes for Sharp-Front Infiltration Problems, EOS Transactions, American

Geophysical Union, Vol. 74, No. 43, p. 307. Presented at American Geophysical Union Fall Meeting, San Francisco, California, 6–10 December 1993.

9. Mayer, A.S., and C.T. Miller (1993) The Role of Mass Transfer Characteristics and Heterogeneity in the Dissolution Process, EOS Transactions, American Geophysical Union, Vol. 74, No. 43, p. 289. Presented at American Geophysical Union Fall Meeting, San Francisco, California, 6–10 December 1993.
10. Barry, D. A., C. T. Miller, and K. Bajracharya, An improved split-operator algorithm for solving coupled solute transport and reaction equations, EOS Transactions, 75, in press, 1994. Presented at American Geophysical Union Fall Meeting, San Francisco, CA, December 7-11, 1994.
11. Mayer, A. S., C. T. Miller, and P. B. Calvin, Mathematical simulations of the formation and dissolution of residual nonaqueous phase liquids, Gordon Research Conference, Modeling of Flow in Permeable Media, Proctor Academy, Andover, NH, August 7-12, 1994.
12. McBride, J. F., and C. T. Miller, Optimization of phase-fraction measurements using single- and dual- photon-energy attenuation, EOS Transactions, 75(16), 148. Presented at American Geophysical Union Spring Meeting, Baltimore, MD, May 23-27, 1994.
13. McBride, J. F., and C. T. Miller, Single- and dual-energy phase fraction measurements in porous media using X-ray attenuation, Agronomy Abstracts, American Society of Agronomy. Presented at 86th Annual Meeting of the Soil Science Society of America, Seattle, WA, November 13-18, 1994.
14. Miller, C. T., A. J. Rabideau, A. S. Mayer, B. Loftis, and M. R. Houyoux, Simulating the fate and transport of groundwater contaminants, Conference on Environmental Impact Prediction, Simulation for Environmental Decision-Making, North Carolina Supercomputing Center, Research Triangle Park, NC, October 6-7, 1994.

PhD Dissertations and MS Technical Reports

1. Mayer, Alex S. (1992) An Investigation of Residual Nonaqueous Phase Liquid Dissolution in Saturated Groundwater Systems, Dissertation.
2. Cornew, Frank H. (1992) An Analysis of Methods for Modeling Advective-Dominated Transport, MS Technical Report.
3. Williams, Glenn A. (1992) An Analysis of Adaptive Finite Element Methods for Simulating Two-Dimensional Contaminant Transport in Groundwater Systems, MS Technical Report.
4. Grant, Jeffrey D. (1994) Development and Analysis of Spatially Adaptive Methods for the Numerical Solution of Richards' Equation, MS Thesis.
5. Hemmer, Paula M. (—) A Comparison of Approaches for Determination of Pressure-Saturation-Permeability Constitutive Relations for Unsaturated Porous Media Systems, MS Report in progress.
6. Arthur, Morris H. (—) An Evaluation of Complete NAPL Dissolution in Porous Media Systems, MS Report in progress.
7. McBride, John F. (—) Development and Application of X-ray Methods to Investigate Multiphase Flow and Transport Phenomena, Dissertation in progress.

Participating Scientific Personnel

1. PI: Cass T. Miller at 25% effort.
2. PhD Students supported in part: P.B. Calvin, A.S. Mayer, J.F. McBride, J.A. Pedit, A.J. Rabideau.
3. MS Students supported in part: M.H. Arthur, P.M. Hemmer, M.I. Lowry, G. Thesing, G.A. Williams.

Dissertations, theses, and reports that are credited in part to this project are annotated above. Note there is not a one-to-one correspondence between the students supported and the dissertation, theses, and report list. In some cases, students contributed to the project, but work on the project did not overlap with their dissertation or thesis. In other cases, the PI contributed significantly to work presented in a dissertation or thesis that was considered part of this project, but for which the student had other sources of support (e.g., fellowship).

Reports or Inventions

Patents will not be sought on any experimental or mathematical model developments accomplished in this work. All details of experimental procedures and computer codes have been or will be published and are considered to be in the public domain.