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6. AUTHOR(S)
John A. Trangenstein

7. PERFORMING ORGANIZATION NAMES(S) AND ADDRESS(ES)
Department of Mathematics, Box 90320
Duke University
Durham NC 27708-0320

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13. ABSTRACT *(Maximum 200 words)*
The goals of this project were to develop advanced modeling and computational methods for surfactant enhanced aquifer remediation (SEAR), and to apply the algorithms to practical recovery problems. The investigators added several features to the current modeling capabilities for SEAR.
They also developed new multigrid iterative methods for use on dynamically adaptive grids.
The research was applied to several field tests, including Hill AFB sites OU1 and OU2, MCB Camp Lejeune, the Savage Well site in New Hampshire, and a TCE site at Cape Canaveral.

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Enclosure 1

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High Resolution 3D Simulation for Surfactant Enhanced Aquifer Remediation

John Trangenstein
Duke University
Department of Mathematics
Durham, NC 17708-0320
(919) 660-2824 office
(919) 660-2821 fax
johnt@math.duke.edu

Gary Pope
The University of Texas at Austin
Petroleum Engineering Department
Austin, Texas 78712
(512) 471-3235 office
(512) 471-9678 fax
gary_pope@utpe.pe.utexas.edu

Research Objectives and Motivation

The goals of this project were to develop advanced modeling and computational methods for surfactant enhanced aquifer remediation (SEAR), and to apply the algorithms to practical recovery problems. This was an interdisciplinary project, involving a mathematician (Professor John Trangenstein at Duke University) and engineers (Professor Gary Pope and graduate students at The University of Texas at Austin).

A principal goal was to develop adaptive mesh refinement (AMR) for surfactant flooding applications. AMR has the potential to reduce the simulation cost substantially; with AMR the work typically scales as if the number of physical dimensions had been reduced by one. For example, AMR work in 2D typically scales as if it were a uniform 1D calculation, and AMR work in 3D scales as if it were a 2D calculation.

The principal difficulty with AMR for surfactant flooding lies in the choices of difference scheme for the pressure equation and iterative method for solving large linear systems defined on hierarchical grids. Difference schemes and iterative methods that work well for uniform grids do not necessarily work well on adaptive grids.

Defense/Civilian Applications

It is now generally recognized that the prevalence of NAPLs at contaminated sites is a significant impediment to aquifer restoration. Because NAPLs are typically of low aqueous solubility, these entrapped residuals may serve as long-term sources of ground water contamination. Conventional pump-and-treat remediation technologies have proved to be an ineffective and costly approach to aquifer restoration when NAPLs are present. Of particular concern are sites contaminated with chlorinated solvents, which are not typically confined to the unsaturated zone due to their large densities and low viscosities.

Chemistry of Surfactant-Enhanced Aquifer Remediation (SEAR)

SEAR is implemented by injecting dilute aqueous surfactant solutions into contaminated formations. These solutions enhance the solubilization of non-aqueous phase liquids (NAPLs) in the aqueous phase. The presence of surfactant also tends to mobilize the entrapped NAPLs. Often, polymer is injected with water and surfactant to reduce viscous instability.

One of the goals of this project was to develop improved models for surfactant-enhanced fluid behavior. These models include models for solubilization of microemulsions, NAPL adsorption, multiphase capillary pressure, and hysteresis in relative permeability and capillary pressure.

Field Test with SEAR

In a field test at Hill AFB site OU2, Gary Pope and collaborators were successful in using SEAR to remove more than 99% of the trichloroethylene from the groundwater in less than two weeks of flooding. This test used co-solvent with the surfactant to avoid phase behavior problems, and to adjust the buoyancy of the mobilized contaminant. The surfactant solubilized the contaminant extremely well, so that it could easily and quickly be flushed out by water and treated at the surface. Without a doubt, this is the largest and most successful remediation effort done in the United States. This effort demonstrates that it is possible to design a field test that is robust, efficient and low-risk.

Of course, the success of the Hill AFB test can be attributed in large part to the extensive use of modeling. This improved recovery technology requires a more complex phase behavior model.

Mathematical Structure of SEAR

Applied mathematics and careful computing can play an important role in improving the success in designing SEAR projects. The cleanup process is modeled by a system of flow equations that involve a system of conservation laws (with at least seven distinct component masses), and an elliptic equation for pressure in the wetting phase. The conservation law has a very complicated wave structure, due to nonlinearities in the phase behavior and transport properties, in addition to local linear degeneracies, eigenvector deficiencies and discontinuities in the characteristic speeds due to phase changes and history-dependence (such as irreversible adsorption, or hysteresis in relative permeabilities). The structure of these waves poses difficulties for numerical integration methods.

In a previous (NSF) project, John Trangenstein employed mathematical analysis to identify aspects of the UT surfactant model that caused the simulator to take very small timesteps; with minor modifications in the model, the simulator was able to take much larger timesteps (roughly two orders of magnitude larger). Similar analysis was used in this project to develop fluid behavior models.

Modern Numerical Methods for SEAR

The original AMR code at Duke University used an explicit second-order Godunov integrator for the component conservation equations. In that code, characteristic speeds were used to select stable timesteps, to perform characteristic tracing for second-order temporal accuracy, and to approximate solutions to Riemann problems. That Godunov integrator was somewhat expensive to operate, and very difficult to maintain. The maintenance problems were due to the use of analytical partial derivatives used to compute characteristic speeds: if modifications in the models changed the forms of the functional dependencies, then many chains of derivatives could be affected.

The previous AMR code used a mixed finite element method to discretize the pressure equation, so that the centering of the pressure and velocity fields would be consistent with the integration of the system of conservation laws. The principal difficulties in this initial application of AMR were the loss of accuracy in the mixed finite element method at coarse-fine interfaces and the unreliability of the multi-level iteration, which used multigrid as an outer iteration and Gauss-Seidel as a smoother. If the grid were not designed so that the transmissibilities were essentially the same in all coordinate directions, then the Gauss-Seidel smoother made no progress and the iterations failed to converge.

Comparison to Standard Methods

It is common in other hydrologic simulations to couple convection and diffusion, mass conservation and the pressure equation, together in a large system of nonlinear equations. The linear systems involved in this approach contain substantial asymmetry; this requires more expensive iterative methods and can lead to convergence problems. There are very few attempts to develop adaptive simulators, because of the substantial development time required, and the sophistication needed in dealing with adaptive data structures. In contrast, the approach in this project used very recently developed discretization and iteration methods; this project also separated convection from diffusion (aka physical dispersion), treating the former explicitly and the latter implicitly. The linear systems are self-adjoint and positive definite; the principal difficulty lies in the treatment of linear systems involved in local mesh refinement.

Significant Accomplishments

This project was adversely affected by a 20% budget cut in year 2, which was confined to the Duke portion of the project. The Duke University post doctoral fellow (Ilya Mishev), an expert in finite element methods and multigrid iteration, had to begin searching for a new position after only 2 months at Duke. As a result, he was unable to make any significant contributions to the iterative methods while John Trangenstein was working on surfactant modeling in year 1. Due to the budget cuts, the available manpower at Duke was changed from 14 man-months in year 1 (12 months for the post doc and 2 for John Trangenstein) to 3 man-months in years 2 and 3. This was a 79% reduction in labor at Duke. Furthermore, the loss of the post-doc's expertise in multigrid methods significantly lengthened the development time for the iterative linear algebra. Although the budget was restored in year 3 of the project, it was not possible to search for an appropriate replacement for the post-doctoral fellow for the remaining year.

Nevertheless, there are several significant accomplishments from this grant.

Improved SEAR Models

In the first year of this project, the investigators added several features to the current modeling capabilities, including NAPL adsorption, multiphase capillary pressure models and hysteresis in relative permeabilities and capillary pressure. The calculation of the irreducible phase saturations was modified to account for buoyancy effects (via the Bond number). Hysteresis is especially important, because contaminant spills can lead to spatially-varying contaminant concentrations at the start of the recovery process.

Gary Pope and Mojdeh Delshad developed significant new correlations for crucial physical properties (namely optimum salinity and optimum solubilization ratios) of microemulsions that solubilize NAPLs consisting of many components. These correlations are essential to make modeling of surfactant remediation complete and accurate for multicomponent NAPLs. Since most superfund sites are in fact contaminated by multicomponent NAPLs rather than pure NAPLs such as TCE, this work is essential to the modeling and engineering of practical cleanup projects via surfactants. These results were presented at the fall AGU meeting in December, 1997.

Gary Pope and collaborators developed a new approach to the surfactant remediation of DNAPLs that uses microemulsions of neutral buoyancy (NB) to prevent downward migration of the dense contaminants during the surfactant flood. A very precise model of the density of the microemulsion as a function of co-solvent, surfactant, electrolyte, and contaminant concentrations was developed and tested with density data from UT laboratories (Kostarelos et al., 1998). Extensive simulations were done to illustrate how to apply SEAR NB (Shook et al., 1998).

Gary Pope and collaborators developed new thermodynamic models of alcohol partition coefficients for a wide range of alcohols and DNAPLs (Dwarakanath and Pope, 1998; Wang et al., 1998) and applied these models to the design and interpretation of both laboratory and field interwell partitioning tracer tests (PITTs).

Hyperbolic Integrators

Working with Mike Edwards at UT, John Trangenstein tested some alternative second-order upwind conservative difference methods for the mass conservation equations. The final choice was to use an upstream weighting flux splitting with a TVD MUSCL limiter. This gives formal second-order accuracy in space; later modifications will give the scheme formal second-order accuracy in time.

Unfortunately, Mike Edwards left this project early in the second year of the grant.

Elliptic Solvers

In the first year of this project a post-doctoral fellow at Duke (Ilya Mishev) analytically investigated various mixed finite element methods for the numerical solution of the pressure equation on adaptively refined grids. It was at this time, before ARO budget cuts, that the Duke team made the decision to switch from the lowest-order mixed method to the hybrid mixed method for the pressure equation discretization. This decision was the scientifically correct choice, but added greatly to the development time.

Due to ARO budget cuts, Dr. Mishev was involved in this project for only the first year. He had to spend most of that time searching for a new job, instead of working on this project.

In year 2, John Trangenstein developed the hybrid mixed finite element methods for the pressure equation on adaptive grids. This method leads to discretizations of the flow equations that item are consistent with conservative second-order upwind schemes for the mass conservation equations, extend to flow problems with full tensor permeabilities, and item allow for physically reasonable scaling relationships between adaptively refined grids. Within each grid cell, the hybrid mixed finite element method satisfies Darcy's law and the divergence-free condition on the velocity exactly; between grid cells, the hybrid mixed finite element method uses a linear system to enforce equality of the volume flux. This formulation is attractive to adaptive mesh refinement because the intercell communication has an important physical interpretation and leads naturally to useful scaling relationships.

In year 2, John Trangenstein also examined alternative techniques, for applying the multigrid preconditioner to the hybrid mixed finite element method. This was unforeseen work, outside his normal area of expertise. In particular, he developed a conjugate gradient outer iteration, a multiplicative domain decomposition preconditioner between levels of refinement in the adaptive mesh, additive domain decomposition smoother between grid patches on an individual level of refinement, and incomplete Cholesky factorization relaxation on an individual grid patch. In order for conjugate gradients to work properly, both the discretization of the pressure equation and the preconditioner must be self-adjoint; the preconditioner must coalesce the eigenvalues of the original discrete elliptic operator, and the work in the preconditioner must be proportional to the number of unknowns in the problem. If designed properly, multigrid preconditioners make the condition number of the iterative system independent of the mesh size; this implies that the number of conjugate iterations needed for convergence will be independent of the mesh size. Since the work per iteration is proportional to the number of unknowns and the number of iterations is essentially independent of the number of unknowns, work required to solve the linear system is essentially proportional to the number of unknowns.

In year three, John Trangenstein developed a linear-algebraic analysis of a preconditioner for the linear system on a hierarchical grid. Because the permeability field is not necessarily smooth, he also developed an incomplete Cholesky factorization for the hybrid mixed method, to be used in place of the multigrid "smoother." Because it is planned to extend the code to distributed memory machines, additive domain decomposition was used to couple the pressure equation between grid "patches" on distinct levels of refinement. Because the hybrid mixed method employs mortar elements to determine coarse boundary pressures from fine pressures, the coarse grid is "slave" to the fine grid; as a result, it was necessary to employ overlapping domain decomposition. Between levels of refinement, the precondition uses a multiplicative domain decomposition, which looks very much like a multigrid step. The convergence of the multigrid iteration is accelerated by the use of a conjugate gradient outer iteration.

In order to simplify the debugging process, John Trangenstein stopped working with the very complicated and time-consuming surfactant model. Instead, he began working with a simple miscible displacement model, which retains all of the complexity for the linear algebra. Each of the individual pieces of the iterative linear algebra have been tested successfully. Further time is needed to get the combined algorithm to work in general.

For the continuing work on AMR multigrid methods, John Trangenstein has recently begun work with Matt Farthing, who is a graduate student at UNC working under the direction of Cass Miller.

Field Tests

Gary Pope and Mojdeh Delshad in collaboration with researchers at Rice University and Duke Engineering and Services modeled the surfactant/foam remediation of DNAPL at Hill AFB OU2 (Hirasaki et al., 1998). This 1997 field demonstration followed the highly successful SEAR demonstration in 1996 in a different part of the same DNAPL site (Brown et al., 1999). A third field test was modeled by UT in 1998 as part of the design of the SEAR at MCB Camp Lejeune where PCE was the DNAPL and the field test was completed in August, 1999. The first ever thermally enhanced SEAR was modeled and the field demonstration successfully completed in July, 1999, at Pearl Harbor. Gary Pope and Mojdeh Delshad in collaboration with researchers at Clemson University have also modeled several co-solvent remediation projects. The first ever SEAR NB field test was modeled during 1999 and the demonstration is scheduled for 2000 at the Savage Well site in New Hampshire. The swept pore volume of this surfactant flood will be 10 times larger than the previous ones. Another large SEAR was modeled for a TCE site at Cape Canaveral in collaboration with SURBEC. Thus, technology transfer in this area has been extensive.

Gary Pope and Akhil Datta-Gupta analyzed field PITT data from Hill AFB OU1 using a streamline-based inversion method (Yoon et al., 1999). More than 40 PITTs have been completed since 1996 both before and after remediation of numerous DNAPL sites across the country and most of these have been modeled at UT or in collaboration with UT as part of the field design and in several cases inverse modeling has been used subsequently as a tool in the interpretation of the field data. These PITTs have been in both vadose zones (Deeds et al., 1999; Mariner et al., 1999) and saturated zones (Young et al., 1999).

Gary Pope has also implemented and tested a mass transfer correlation which compares well with the original, high quality surfactant/NAPL experimental surfactant dissolution remediation data.

Cooperation with and Technology Transfer to Army Laboratories

The Army's Waterways Experimental Station (WES) funds Gary Pope and collaborators at the University of Texas to further develop the chemical and microbiological modeling of groundwater remediation. Gary Pope and Mojdeh Delshad have frequent technical discussions on groundwater modeling and remediation with co-solvents and surfactants with Dr. Chris McGrath at WES. Gary Pope visited WES once during 1997, and Mojdeh Delshad visited WES in 1998. During these discussions, there have been many groundwater modeling issues of vital interest to the Army clarified and Dr. McGrath has made many useful suggestions for further modeling research.

Connection to NSF Heterogeneity Characterization Project

Note that through an existing NSF grant (joint between Gary Pope and Akhil Datta-Gupta of Texas A&M), Gary Pope and Akhil Datta-Gupta are developing models for fluid flow in fractured porous media. This is an important development, of practical importance for many contaminated sites.

Connection to NSF AMR Parallelization Project

Through partial support from a project at Duke, sponsored by NSF to study shear band formation and growth, William Allard has worked with John Trangenstein to develop object-oriented message-passing paradigms for adaptive mesh refinement. On the 32-processor T3E at MCNC, Allard and Trangenstein have obtained speedup of a factor of 28 for adaptive computations of unsteady problems in linear elasticity.

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C. M. Young, R.E. Jackson, M. Jin, J. T. Londergan, P. E. Mariner, G. A. Pope, F. J. Anderson, and T. Houk, "Characterization of a TCE DNAPL Zone in Alluvium by Partitioning Tracers," Ground Water Monitoring & Remediation, XIX (1), Winter 1999.

Awards and Honors

John Trangenstein

- Invited speaker, Gordon Conference on Modeling of Flow in Permeabable Media, August 1996.
- Invited speaker, Cray Workshop on Advanced Simulation of Subsurface Flow and Contaminant Transport, December 1996.
- Invited speaker, IMA conference on Adaptive Mesh Refinement, March 1997.

Gary Pope

- Society of Petroleum Engineers Distinguished Achievement Award for Petroleum Engineering Faculty, presented at the 1996 Annual Fall meeting.
- Organized session on Simulation of DNAPL Multiphase Flow and Transport, First International Conference on Remediation of Chlorinated and Recalcitrant Compounds, Monterey, California, May 18-21, 1998.
- Inducted into National Academy of Engineering, October, 1999.

Participating Scientific Personnel

At Duke University, the participating personnel were Professor John Trangenstein, and Dr. Ilya Mishev. Dr. Mishev was a post-doctoral fellow who participated in the research during year 1; he is now working at Mobil in Dallas.

At the University of Texas at Austin, the participating personnel were Professor Gary Pope, Dr. Mojdeh Delshad and Dr. Michael Edwards. Dr. Edwards left UT early in year 2; he is now working at Stanford University.