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In Situ, Field Scale Evaluation of Surfactant Enhanced DNAPL Recovery Using a Single-Well, Push-Pull Test

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Research Objective

The overall goal of this project is to develop the single-well, “push-pull” test method as a new site characterization and feasibility assessment tool for studying the fundamental fate and transport behavior of injected surfactants and their ability to solubilize and mobilize dense nonaqueous phase liquids (DNAPLs) in the subsurface. The specific objectives are:

- (1) to develop a modified “push-pull” test for use in identifying and quantifying the effects of sorption, precipitation, and biodegradation on the fate and transport of injected surfactants,
- (2) to use the developed test method to quantify the effects of these processes on the ability of injected surfactants to solubilize and mobilize residual phase trichloroethylene, and
- (3) to demonstrate the utility of the developed test method for performing site characterization and feasibility studies for surfactant enhanced DNAPL recovery systems.

Research Progress and Implications

This report summarizes work as of June 1, 1998 (after 20 months of a 36-month project); laboratory and field work as been successfully completed for all three objectives.

The ability of the push-pull test method to characterize sorption of linear alkylbenzene sulfonate (LAS) and hexadecyl diphenyl oxide disulfonate (DOWFAX) surfactants to natural aquifer sediment was evaluated using sediment and groundwater from a TCE contaminated field site at the Building 834 operable unit at Site 300, Lawrence Livermore National Laboratory. Sorption isotherms exhibited Langmuir-type behavior with larger apparent sorption maximums for LAS than for DOWFAX. Laboratory push-pull tests were performed in unique intermediate-scale physical aquifer models (flow cells) designed and operated to approximate the radial flow field around an injection/extraction well during a field push-pull test. The models were packed with aquifer sediment from the same aquifer unit used in field push-pull tests. Results of laboratory push-pull tests indicated substantial retardation and chromatographic separation of injected LAS, but no apparent retardation or separation of injected DOWFAX. Experimental results and numerical modeling indicate that LAS and DOWFAX sorption may be kinetically-limited during transport. Field push-pull tests were successfully completed in monitoring wells D-6 and D-12, which are close to the TCE source zone but do not contain residual TCE. In these tests, 50 L of surfactant solution containing a bromide tracer were injected into the saturated zone at a depth of ~ 8 m. During the extraction phase, 100 L of test solution/groundwater mixture were extracted and analyzed for tracer, surfactant, and TCE. The field results confirmed the ability of the push-pull test method to rapidly characterize surfactant sorption to aquifer sediments in-situ. Field tests were conducted in collaboration with LLNL personnel and the EMSP project “Development of radon-222 as a natural tracer for monitoring the remediation of NAPL contamination in the subsurface”. This allowed an independent determination of residual TCE saturation in the vicinity of the monitoring wells during each test.

As part of this work, a simplified method was developed for estimating retardation factors from push-pull test results by comparing extraction phase breakthrough curves for solutes undergoing linear equilibrium sorption and a nonsorbing tracer. Using numerical simulations an extensive sensitivity analysis was performed to test the method’s performance for several cases, including push-pull tests conducted in homogeneous and heterogeneous (layered) aquifers, as well as tests conducted in wells screened across the entire saturated thickness and wells in which a packer system is used to isolate a portion of the aquifer. The simplified method proved useful in estimating retardation factors for all cases except under conditions of severe heterogeneity.

The ability of the push-pull test method to characterize surfactant enhanced solubilization of residual TCE by DOWFAX was also evaluated using sediment and groundwater from Site 300. Batch solubilization experiments indicated a linear relationship between TCE solubility and DOWFAX concentration characterized by weight- (WSR) and molar solubilization ratio (MSR) of 0.26 and 1.25, respectively. Laboratory push-pull tests were performed in physical models with both TCE-free sediment and in sediment containing 920.2 g TCE, which corresponded to a residual saturation of 5 vol. % of the pore space. In push-pull tracer tests conducted in the absence of DOWFAX, maximum aqueous phase TCE concentrations were ~ 500 mg/L. In contrast, during the first push-pull test conducted with 13,500 mg/L active DOWFAX, aqueous phase TCE concentrations increased to ~ 3,000 mg/L and TCE mass recovery increased by a factor of ~ 3. Successive push-pull tests conducted with DOWFAX yielded decreasing TCE recovery, such that by the third experiment no increase in TCE recovery was obtained. Excavation of the sediment pack confirmed the quantity of residual TCE removed by surfactant and indicated that no mobilization of residual TCE had occurred. As part of this work a novel analytical method was developed for determining TCE concentrations in the presence of large concentrations of anionic surfactants. The method uses high performance liquid chromatography with photodiode array detection to allow detection of TCE (for concentrations greater than ~ 0.1 mg/L) and surfactant by the direct injection of a single aqueous sample and substantially reduces the costs of laboratory and field experiments. Laboratory push-pull tests were conducted in collaboration with the EMSP project "Development of radon-222 as a natural tracer for monitoring the remediation of NAPL contamination in the subsurface". Measurements of dissolved radon-222 gas were used to quantify the spatial reduction in residual TCE saturation following each push-pull test with DOWFAX.

Implications

As a result of this project, the push-pull test method is ready for field deployment at any site where the use of surfactant enhanced DNAPL recovery is being considered. The method is performed in-situ using existing monitoring wells, without the need for core collection. Quantitative information on surfactant sorption or residual DNAPL solubilization potential can be obtained in only one day of field work at most sites. Because the method greatly reduces site characterization and feasibility assessment costs it should substantially reduce the time and cost of remedial action at contaminated sites.

Planned Activities

Additional experiments are in progress to characterize residual TCE solubilization by a third surfactant, Aerosol MA80-I (sodium dihexylsulfosuccinate). To produce a neutrally-bouyant injection solution, 4 wt % Aerosol MA80-I will be combined with 10% isopropanol and 0.32 wt % potassium bromide. Push-pull experiments will be conducted in physical aquifer models packed with sediment obtained from LLNL Site 300 in the presence and absence of added TCE. The effect of cation exchange between the injected sodium/potassium surfactant solution and sediment-associated calcium on the phase behavior of the surfactant and its ability to solubilize TCE will be investigated.

Other Access To Information

The following manuscript preprints are available from Dr. Istok:

- Istok, J.D., J.A. Field, M.D. Humphrey, T.E. Sawyer, and M.H. Schroth. 1998. Laboratory and field investigation of surfactant sorption using single-well, "push-pull" tests. Submitted to Ground Water.
- Field, J.A., J.D. Istok, M.D. Humphrey, T.E. Sawyer, and M.H. Schroth. 1998. Laboratory investigation of surfactant-enhanced TCE solubilization using single-well, "push-pull" tests. Submitted to Ground Water.
- Field, J.A. and T.E. Sawyer. 1998. HPLC method for simultaneous determination of TCE and anionic surfactants at high surfactant concentrations. Submitted to Journal of Chromatography.
- Schroth, M.H., J.A. Field, R. Haggerty, M.D. Humphrey, and J.D. Istok. 1998. In-situ evaluation of solute retardation using single-well push-pull tests. Submitted to Journal of Hydrology.