International Containment Technology Conference
St. Peterburg, Florida
February 9, 1997

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Advanced Hydraulic Fracturing Methods to Create In Situ Reactive Barriers

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Many contaminated areas consist of a source area and a plume. In the source area, the contaminant moves vertically downward from a release point through the vadose zone to an underlying saturated region. Where contaminants are organic liquids, NAPL may accumulate on the water table, or it may continue to migrate downward through the saturated region. In either case, NAPLs or other compounds in this area may dissolve and potentially contaminate large volumes of groundwater. The contaminant plume itself typically consists of dissolved compounds that move horizontally away from the source by ambient groundwater flow, potentially impacting supplies of drinking water or other receptors. Early developments of permeable barrier technology have focused on intercepting horizontally moving plumes with vertical structures, such as trenches, filled with reactive material capable of immobilizing or degrading dissolved contaminants (e.g. Gillam, 1996). This focus resulted in part from a need to economically treat the potentially large volumes of contaminated water in a plume, and in part from the availability of construction technology to create the vertical structures that could house reactive compounds.

Contaminant source areas, however, have thus far remained largely excluded from the application of permeable barrier technology. One reason for this is the lack of conventional construction methods for creating suitable horizontal structures that would place reactive materials in the path of downward-moving contaminants. Methods of hydraulic fracturing have been widely used to create flat-lying to gently dipping layers of granular material in unconsolidated sediments. Most applications thus far have involved filling fractures with coarse-grained sand to create permeable layers that will increase the discharge of wells recovering contaminated water or vapor (Murdoch et al., 1994). However, it is possible to fill fractures with other compounds that alter the chemical composition of the subsurface. One early application involved development and field testing micro-encapsulated sodium percarbonate (Vesper etal., 1993), a solid compound that releases oxygen and can create aerobic conditions suitable for biodegradation in the subsurface for several months.

METHODS OF HYDRAULIC FRACTURING

Hydraulic fracturing begins by injecting fluid into a borehole until the pressure exceeds a critical value and a fracture is nucleated. Coarse-grained sand, or some other granular material, is injected as a slurry while the fracture grows away from the borehole. Guar gum gel, a viscous fluid, is commonly used to facilitate transport of the granular material into the fracture. After pumping, the guar gum gel is decomposed by an enzyme added during injection and the fracture resembles a thin layer or bed in the subsurface (Murdoch et al, 1994).

The injection pressure required to create hydraulic fractures is remarkably modest. For example, at the beginning of injection during a test at 2 m depth, the pressure increased abruptly to 490

kPa (70 psi), but then decreased sharply when the fracture began to propagate. Injection pressure decreased from 200 to 100 (30 to 15 psi) throughout most of the duration of propagation (Murdoch et al, 1994). Slightly greater pressures are required to create fractures at greater depth.

Fracture Form

Conventional methods of hydraulic fracturing generally produce a single parting (multiple fractures require repeated operations) and the form of the fracture depends on the state of stress, the degree of stratigraphic layering or fabric in the enveloping formation, and perhaps other factors. In overconsolidated or bedded sediments, hydraulic fractures typically are equant to elongate in plan and dip gently towards their parent borehole (Fig. 1).
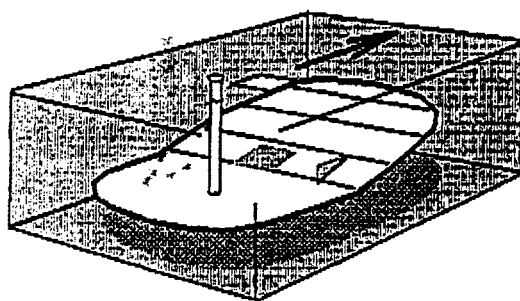

Figure 1. Idealized hydraulic fracture. Arrow points to the preferred propagation direction. White square overlies thickest area at center of fracture. Triangle shows dip angle.

In most cases, the fractures have a preferred direction of propagation so that they are asymmetric with respect to the borehole. The area of the fracture containing the thickest material, however, nearly always occurs in the vicinity of the center of the fracture, so that the propagation direction affects the distribution of injected material (Murdoch et al., 1991). Fractures typically climb in the preferred direction of propagation and will reach the ground surface along that direction if sufficient fluid is injected. The propagation direction can be related to distribution of vertical load at the ground surface, with the fractures propagating toward regions of diminished vertical load. At some locations, the vertical load has been manipulated (say, by positioning a drill rig near the injection point) to influence the propagation direction, whereas at other locations this approach has been impractical or ineffective.

Hydraulic fractures have been created between 1.2 and 16m below the ground surface during recent projects. Nearly all of them have been in silty clay, and most have been in glacial drift that probably was overconsolidated so that the form outlined above is typical. Maximum dimensions increase with depth, but are in the range of 7 to 10 m. Bulk volumes of granular material filling the fractures also increases with depth, ranging from 0.15 m³ (5 ft³) for shallow fractures to 1.25 m³ (44 ft³) for deeper ones. The average thickness of material filling a fracture ranges from 5 to 10 mm, but can be as much as 25 mm.

The sizes of the features that can be created using conventional hydraulic fracturing methods is remarkable considering the modest injection pressures and power required. However, conventional methods offer scant opportunity to influence the geometry of the fractures. Where surface loading is impractical, the direction of propagation can be difficult to anticipate, and the degree of asymmetry about the injection point cannot be influenced at all. This drawback can be of minor consequence where fractures are used for recovery because subsurface flow occurs beyond the lateral extent of the fractures so the natural variations in geometry have a only secondary effect. The drawback is more serious, however, where fractures are filled with reactive materials and used to degrade contaminants in place. In this application, the ability to influence the location of a fracture will help ensure that reactive material uniformly cuts across potential contaminant migration pathways.

Monitoring

The fracturing procedure is monitored by recording both pressure and deformation of the ground

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surface as functions of time. The ground surface over a shallow hydraulic fracture will lift to form a broad, gentle dome. The amount of uplift is similar to the fracture aperture (Murdoch et al., 1991), so the pattern of uplift can be used to infer the location and thickness of material filling the fracture at depth. Accordingly, the dome typically will be slightly asymmetric and the point of maximum uplift (the apex of the dome) will be displaced away from the injection point in the preferred direction of propagation.

Net ground displacements accompanying fracturing can be measured using optical leveling methods, or the inclination of the ground surface can be measured in real time using an array of tiltmeters. The tilt signal can be inverted to estimate uplift and, in particular, to sense the position of the apex of the dome. The tiltmeter data show that in most cases, fractures develop a preferred direction early and then continue to grow in this direction throughout propagation.

Directional Fracturing

Conventional methods of hydraulic fracturing have been modified to provide a mechanism for influencing the preferred direction of propagation of gently dipping fractures. The purpose here has been to develop a capability either to make fractures that are highly asymmetric in a particular direction, or to make fractures that are symmetric where they might otherwise be asymmetric. The first capability will allow reactive material to be placed beneath waste pits, buildings, tanks or other potential sources that cannot be penetrated by a vertical boring, whereas the second one will ensure that reactive material can be uniformly distributed in contaminated ground.

Methods of directional fracturing use an energetic water jet concurrent with slurry injection. The jet is supplied by a narrow tube that passes through a seal at the wellhead and extends to the depth of the fracture. The jet is oriented perpendicular to the tube so that its direction can be controlled by rotating the supply tube. Slurry flows down the annulus between an outer casing and the supply tube and is entrained by the jet when it reaches the depth of the fracture. The preferred direction of propagation follows the direction of the jet. During operation, tiltmeters are used to detect the propagation direction and the jet is rotated to either oppose or enhance that direction.

Field applications of the method have demonstrated the ability to induce propagation in a particular direction by directing the jet in that direction. The uplift is roughly symmetric about the injection point over most fractures created using conventional methods at a test site in the vicinity of Cincinnati, OH (Fig. 2a). Uplift that is highly asymmetric is created by operating the jet concurrent with slurry injection (Fig. 2b). Thickness of injected material is roughly proportional to the uplift, so that the injected material is concentrated to the NW of the borehole in Figure 2b. Several borings confirmed this inference.

Other field tests have involved using the directional system to improve the symmetry of the fractures. The success of this application appears to hinge on the time required to detect the direction of propagation; when propagation direction is detected early (within the first minutes of injection) it can be arrested or changed, when it is detected later the directional methods (in the current embodiment) have limited effect and the fracture continues to propagate in its preferred direction.

The directional methods developed thus far will diversify the applications of hydraulic fractures during the emplacement of reactive compounds. The system may be improved by increasing the power of the controlling jet, and experiments with this aspect of the technology are in progress.
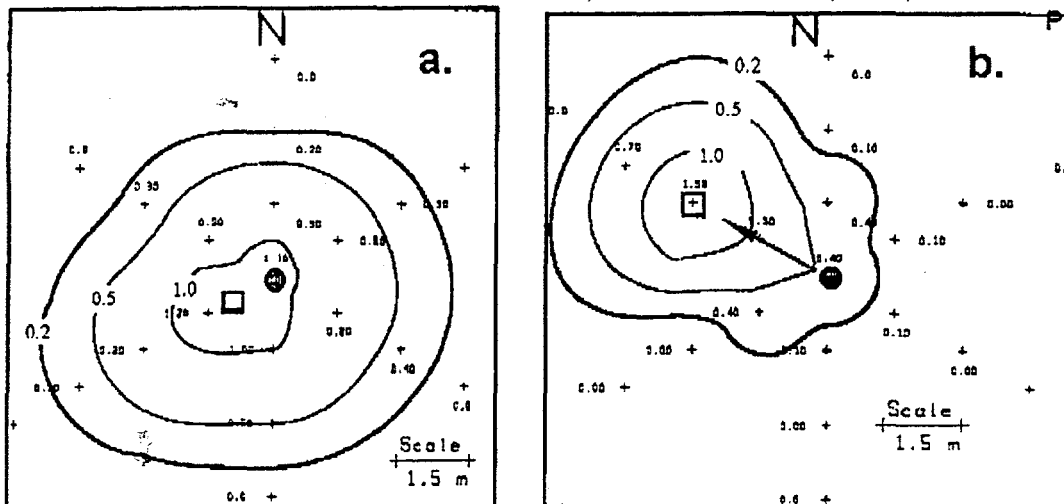
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Figure 2a. Relatively symmetric uplift (cm) over conventional fracture; b.) Asymmetric uplift over fracture created with directional method. Jet pointing in direction of arrow during slurry injection. Square: maximum uplift

APPLICATIONS

Hydraulic fractures have been used to create sheet-like layers filled with a wide range of reactive compounds, from materials that alter redox conditions, to ones that adsorb contaminants or slowly leach beneficial materials. Early tests used a solid percarbonate, which slowly releases oxygen, to create fractures that developed aerobic conditions to enhance biodegradation in the subsurface. The early work demonstrated the viability of this approach but used material created using microencapsulation methods, which was too expensive for widespread application. However, recent formulations of similar oxygen releasing compounds (e.g. Marlow, 1996) are less expensive so this application may receive renewed interest.

Applications that are currently under investigation include filling fractures with porous ceramic granules to stimulate biodegradation, with adsorptive materials that can be treated using in situ with biodegradation, or with redox-altering materials to either oxidize or reduce contaminants.

Biodegradation Using Porous Ceramics

Biodegradation at some sites can be augmented by injecting populations of organisms capable of degrading contaminants along with the nutrients they require to thrive. This approach can be facilitated by colonizing granules of a highly porous ceramic made from diatomaceous earth (trade name, Isolite) with the selected microbial population. The granules also form a permeable bed through which nutrients can be injected.

Hydraulic fractures were filled with inoculated Isolite to augment bioremediation at a site where long-chain hydrocarbons used for cutting oil resulted in TPH values in excess of 2000 mg/kg. Samples from the site indicated that the population of heterotrophic and autotrophic organisms was too sparse to accomplish degradation, according to Stavnes et al. (1996), who describe the details of this study. Moreover, the site is underlain by weathered, fine-grained sediments of low permeability so that closely spaced conventional wells would be required to effectively deliver liquid nutrients.

Laboratory studies identified and refined a consortium of cutting-oil-metabolizing bacteria culled from samples taken from the site. Bench-scale tests indicated that this consortium effectively reduced TPH values in site soils. A solution containing the bacteria consortium and nutrients was circulated through packed columns filled with Isolite, causing bacterial growth on the porous ceramic granules and effectively inoculating the Isolite for subsurface injection (Stavnes et al.,

1996).

Sets of hydraulic fractures stacked one above the other were created with the innoculated Isolite at two locations. Location 1 contained four fractures at depths of 2.4, 3.4, 4 and 4.6 m, whereas Location 2 contained only two fractures at slightly greater depths, 4.6 and 5.2 m. The fractures were filled with between 0.25 and 0.5 m^3 of Isolite (grain size: 1 to 2 mm), which created layers as thick as 2.5 cm and 6 to 8 m in maximum dimension.
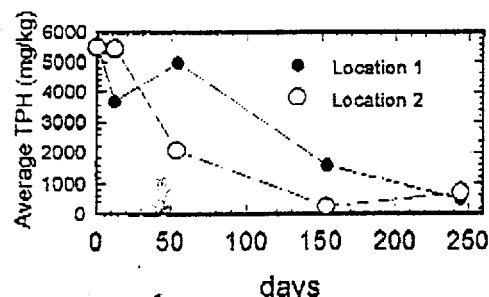

Figure 3. Average TPH concentrations from soil samples as a function of time.

The intention was to utilize the Isolite-filled fractures as in situ bioreactors that would degrade contaminants in their vicinity, although the optimal operation of the fractures was unclear. Accordingly, fractures at Location 1 were augmented by injections of nutrients, additional microbes, and a surfactant was added to increase bioavailability. Later, air was injected at Location 1 to enhance aerobic conditions. In contrast, Location 2 was left idle, although several wells were screened in the fractures and may have provided some passive aeration (Stavnes et al., 1996).

Soil samples were obtained 12, 54, 154 and 245 days after fracturing and analyzed for TPH. Samples were taken from the 11 locations in vicinity of the Location 1, and 4 locations in the vicinity of Location 2, as well as from more distant areas to act as control. The average concentrations from those sampling events indicate that the TPH concentrations remained roughly constant during the first 54 days at Location 1 and then decreased after that time (Fig. 3). Concentrations apparently decreased slightly earlier at Location 2. By day 154, average concentrations at both locations decreased to 20 percent of initial values and by day 254, the average concentrations were roughly 10 percent of initial values. Student t-tests performed on the data indicate that the reduction in concentration compared to initial measurements at both locations is statistically significant by day 154, assuming $\alpha=0.1$, and the confidence increases with time so by day 254 the reductions in concentration are significant assuming $\alpha=0.05$. Samples taken in the contaminated zone beyond the region that was unaffected by the reactive fractures yielded TPH values between 1700 and 3140 mg/kg throughout the study period and showed no systematic change in concentration with time.

Adsorption and biodegradation

It can be important in some circumstances to arrest contaminant; for example, where vertical flow is rapid enough to outpace degradation kinetics, or where contaminants (e.g. metals) cannot be degraded at all. One approach to this scenario uses fractures filled with granular activated carbon (GAC) to adsorb chlorinated solvents, coupled with techniques to periodically degrade the adsorbed solvents using biodegradation. A proof-of-concept field test has been conducted at an uncontaminated site, and a field test at a contaminated site is on going.

The proof-of-concept field test involved creating a fracture filled with GAC and inoculating the material with a consortium of TCE-degrading methanotrophic organisms. It was assumed that the GAC could adsorb TCE, and we sought to evaluate the ability of the organisms to degrade solvent without actually having any solvent in the ground. This was accomplished by injecting a mixture of air and 3 percent methane into the GAC-filled fracture for 3 months in an effort to stimulate the growth of the introduced organisms. Samples were obtained weekly and tested in small batch reactors in the laboratory for their ability to degrade TCE. The samples were also used to evaluate the activity of methane monooxygenase using the conversion of naphthalene to napthol in batch reactors, and to measure the number of TCE-degrading organisms using cell counts and SEM analyses.

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The results show that samples from the GAC-filled fracture will degrade TCE in batch reactors throughout the 3-month-long test, whereas TCE concentrations in control reactors were unchanged. Moreover, the lab tests consistently showed a conversion of naphthalene to naphthol, which indicates that methane monooxygenase was produced. These results parallel an increase in the microbial population from 10^6 to 10^7 CFU per gm GAC.

These results indicate that it is possible to maintain conditions that will favor in situ degradation of TCE using methanotrophic organisms in GAC-filled hydraulic fractures. On going field tests will evaluate the extension of these results to field conditions where TCE is present.

Redox compounds

The use of redox agents for in situ treatment of organics and inorganics is evolving as an effective means of in situ treatment (Siegrist et al., 1995; 1996, Gates et al., 1996), and implementation of in situ redox zones using hydraulic fracturing is also being explored. As of this writing, laboratory and field experiments are ongoing involving hydraulic fractures filled with redox agents for interception and destruction of DNAPL compounds, such as TCE, in low permeability media, such as silts and clays (Siegrist et al., 1995; 1996). A major part of this work involves a comparative field test that is ongoing at the DOE Portsmouth Site in Ohio where multiple test cells have been emplaced in clean and contaminated sites. These cells are being used to evaluate in situ remediation of DNAPL compounds in silty clay media by both enhanced air flushing and in situ destruction. All of the test cells involve hydraulically emplaced fractures at four to five depths in the range of 1 to 6 m with diameters of ~10 m. Two of the test cells involve fractures that form permeable barriers of either zero-valent iron particles or a special potassium permanganate grout. The work is being completed by a multi-institutional, interdisciplinary team comprised of scientists and engineers from ORNL, Lockheed Martin at Portsmouth, FRx Inc., University of Cincinnati, and the Colorado School of Mines.

The test cells at Portsmouth were installed in September, 1996, and process operation and performance monitoring occurred during fall 1996 with further experimentation planned for spring 1997. Pre- and post-operational monitoring has involved onsite/offsite analyses of more than 600 samples, coupled with 3-dimensional in situ instrumentation emplacement using direct-push and sidewall sampler devices to measure subsurface biogeochemical properties and contaminant treatment efficiency.

In the test cells containing redox reactive fractures, continuous cores were collected across the emplaced fracture zones nearly 3 months after fracture emplacement. Each core was carefully examined and dissected with analyses made for Eh, pH, TOC and WC. DNAPL compound degradation tests were also completed to define destruction efficiency and potential as a function of distance from the reactive fracture. Initial results indicate highly reactive zones were present in these two permeable barrier cells with very high DNAPL degradation efficiencies (e.g., >99%) in fracture treatment zones of varying thickness (e.g., 1 to 10 cm). Further information on this work will be available in forthcoming publications during 1997.

CONCLUSIONS

Hydraulic fracturing methods offer a mechanism for gently dipping layers of reactive compounds. Specialized methods using real-time monitoring and a high-energy jet during fracturing allow the form of the fracture to be influenced. This technique allows asymmetric fractures to be created beneath potential sources (e.g. tanks, pits, buildings) that should not be penetrated by boring.

The technique appears to be particularly versatile. Field applications thus far have created fractures filled with zero-valent iron to reductively dechlorinate halogenated hydrocarbons, or granular activated carbon to adsorb compounds. Porous ceramic granules innoculated with organisms have been used to biologically degrade cutting oil, and slowly dissolving solid

peroxides have been used to create aerobic conditions in the subsurface. The most challenging application has been to fill fractures with granules of potassium permanganate, a strong oxidant that degrades a wide range of organic chemicals. It appears that hydraulic fracturing methods will allow reactive barriers to be created with nearly any solid compound for which subsurface injection is permissible.

The flat-lying treatment zones created using these methods can be operated passively or actively to intercept vertically migrating contaminants. Both organics and inorganics can be intercepted, removed, and either immobilized or degraded. This is an exciting compliment to vertical permeable barriers and provides a means to strategically contain, and even treat in situ, contaminant source areas.

ACKNOWLEDGMENTS

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to appear in: *Proceedings of the International Containment Technology Conference and Exhibition*, Feb. 9-12, 1997, St. Petersburg, FL

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