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INTEGRATED PNEUMATIC FRACTURING AND BIOREMEDIATION FOR THE IN-SITU TREATMENT OF CONTAMINATED SOIL

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INTRODUCTION

In-situ bioremediation is often limited by the rate of transport of nutrients and electron acceptors (e.g., oxygen, nitrate) to the microorganisms, particularly in soil formations with moderate to low permeability. An investigation was conducted to integrate the process of pneumatic fracturing with bioremediation to overcome these rate limitations. Pneumatic fracturing is an innovative technology which utilizes high pressure air to create artificial fractures in the contaminated geologic formations, resulting in enhanced air flow and transport rates in the subsurface. Following the fracturing, the pneumatic fracturing system can be used to inject electron acceptors and other biological supplements directly into the formations to stimulate biodegradation.

Pneumatic fracturing is an innovative technology which enhances the in situ removal and treatment of contaminants from low permeability soil and rock formations. The process may be generally described as injecting air (or another gas) into a contaminated geologic formation at a sufficient pressure and flow rate so that artificial fractures are created. Once established, the fractures increase the permeability of the formation, thereby making contaminant removal and/or treatment more efficient. Pneumatic fracturing is designed to be integrated with other in situ treatment technologies such as vapor extraction, bioremediation, thermal injection, and pump and treat. The technology can be applied to a range of hydrogeologic conditions including vadose zones, perched water zones, and saturated zones.

Research which led to the development of pneumatic fracturing began in 1988 with bench scale studies and analytical modeling at the Hazardous Substance Management Research Center (HSMRC) located at New Jersey Institute of Technology (NJIT). The first industrial site demonstration of pneumatic fracturing was performed in 1990 at a site in Richmond, Virginia . In 1992, the technology was evaluated under the U.S. EPA Superfund Innovative Technology Evaluation (SITE) Demonstration Program at a contaminated industrial site in New Jersey for enhancement of a soil vapor extraction system [USEPA, 1993}. Pneumatic fracturing is now available commercially through Accutech Remedial Systems for applications involving soil vapor extraction and pump and treat enhancement, and is being applied to production clean-ups. Pneumatic fracturing is a patented process, and the assignee is HSMRC.

The sequence of steps required to apply the integrated pneumatic fracturing/bioremediation process are as follows. During the first step, the formation is pneumatically fractured by inserting a proprietary device known as an "HQ Injector" into a drilled well. The HQ injector applies pressurized air along a discrete two foot interval, and is subsequently repositioned at various elevations to create a fracture network. The injector is then moved to additional bore holes and the process repeated until the entire contaminated formation is fractured.

After the formation has been initially fractured with air, the second step is to introduce nutrients into the fracture network to provide substrates for enhance biological activity. This is accomplished by inserting liquid nutrients into the main injection air stream which are then dispersed throughout the already established fracture network. By maintaining a high air to liquid ratio, the liquid supplements actually become atomized during injection, which increases their ability to penetrate the formation. Additional nutrient injections are made periodically to replenish the substrates, and to provide beneficial aeration. Optionally, other biological supplements such as innoculum and/or granular media could be injected into the fracture network.

In the final step of the process, the site is operated as in situ bioremediation cell to degrade the contamination. A continuous air flow is maintained throughout the fracture network via a vacuum pump connected to a central extraction well. Outlying wells vented to the atmosphere serve as a passive source of oxygen. In the present application, air flow was purposely maintained at a low level, to minimize off-gas treatment and to maximize biological effects.

It is important to note that within the fracture network, pneumatically induced fractures will be separated by zones of unfractured soil. The distance between adjacent fractures will vary according to the geologic structure, but is generally believed to be on the order of 1 foot. As a result, a "stacked" system of microbial degradation will develop according to the availability of electron acceptors. Aerobic processes will dominate at the fracture interfaces where the oxygen supply is the highest, and to a limited distance into the soil away from the fracture. Depletion of oxygen during aerobic biodegradation and the presence of exogenous nitrate allows the formation to develop a denitrifying zone a short distance away from the fracture surface. Continued depletion of nitrate in the denitrifying zone further decreases the ambient oxidation-reduction potential and results in the formation of methanogenic populations at greater distances from the fracture. During methanogenic processes, methane is generated and diffuses to the fracture where it is removed by the sweep air extracted from the formation. The final result is contaminant degradation by a stacked series of aerobic, denitrifying, and methanogenic microbial zones which are staggered throughout the fracture network.

A project to investigate the coupling of these two technologies was sponsored by the US EPA under the Superfund Innovative Technology Evaluation (SITE) Emerging Technologies Program and an industrial sponsor. A field pilot demonstration of the integrated technologies was carried out at a gasoline refinery site over the interval of 20 months from October 1992 to May 1994.

METHODOLOGY

The site selected for the process demonstration was a gasoline bending area of a refinery located in Pennsylvania. The site stratigraphy was a surficial layer of fill comprised of clayey silts mixed with varying amounts of sand and gravel, ranging from 1 to 4 feet in thickness. The fill was underlain by a stratum of orange-tan clayey silt which extended to a depth of 9 to 10 feet below the ground surface. The consistency of the clayey silt ranged form medium stiff to stiff, indicating a high degree of over-consolidation. The upper few feet of this stratum were stained dark brown to black, apparently from infiltration of petroleum residues. The clay silt stratum graded into a gray silty sand at a depth of approximately 9 to 10 feet below the surface and extended to depths greater than 20 feet. The groundwater table was at a depth ranging from 13 to 15 feet below ground surface.

Spilt spoon boring samples were extracted and chemically analyzed to determine the extent of BTX contamination at the site. Contaminant distributions were heterogeneous, but with a general trend of total BTX concentrations of approximately 1500 mg/kg at a depth of 1 to 3 feet and decreasing with depth to less than 1 mg/kg at a depth of 9 to 11 feet. These results indicated that the pneumatic fracturing and subsequent bioremediation should focus on the soil contaminants to a depth of 7 feet. This approach also maintained an unfractured buffer of clayey silt between the shallow BTX contamination and the water bearing silty sand stratum.

The site was pneumatically fractured, and periodic injections were performed over a period of 12 months. Subsurface injections included introduction of nitrate and ammonium salt in the form of calcium ammonium nitrate to facilitate the development of aerobic, denitrifying and methanogenic biodegradation zones with respect to increasing distance from the fracture interface. Off-gases from the monitoring wells and vapor extraction well were analyzed for benzene, toluene and xylenes (BTX), oxygen, methane and carbon dioxide to evaluate process effectiveness. At the conclusion of the process demonstration, additional soil borings were carried out and samples analyzed to measure the change in the extent of site contamination as a result of the process. Carbon mass balances considering contaminant reduction, carbon dioxide evolution, methane evolution, and contaminant recovery through vapor extraction, were used to evaluate process performance.

RESULTS

Initial site characterization indicated low subsurface permeability and the presence of BTX at concentrations of up to 1500 ppm in the soil phase. Results show that fracturing increased subsurface permeability by up to 40 times within an effective radius of approximately 20 feet.

Thermodynamically, for BTX biodegradation, presence of oxygen (aerobic conditions) are more favorable for the microbial populations to effectively degrade the contaminants. In the absence of oxygen, the microbes seek an alternate electron acceptor in the form of iron, nitrate or sulfur to carry out the bioconversion. In this study nitrate served as an alternate electron acceptor and hence denitrification process, which biodegrades the organic contaminants into carbon dioxide and water, was initiated. Thermodynamically least favorable metabolic pathway for BTX biodegradation is methanogenesis and this occurs under strictly anaerobic conditions resulting in methane generation. Thus in the absence of oxygen and nitrate as electron acceptors methane generation is enhanced. This classic thermodynamic hierarchy in biodegradation pathways was observed in our field study as well. Following each nitrate injection, the methane concentration in the well decreased, while the carbon dioxide concentration increased indicating a shift towards aerobic and denitrification processes and away from methanogenesis upon the availability of an alternate electron acceptor.

After one year of sampling and monitoring, soil samples obtained from the site at the end of the demonstration show a 79% reduction in soil-phase BTX concentrations. Results from the analysis of soil samples obtained from three distinct depths of the soil bed in the pre-demonstration stage was compared with that in the post-demonstration stage. From these results the total mass of BTX removed, based on BTX concentrations was computed to be 22 kg. Based on periodic soil-gas sampling, the mass of BTX removed through vapor extraction was computed to be 3.1 kg or 11%. Vapor extraction was the predominant abiotic mode of BTX removal. The other abiotic pathways - BTX losses through fracture and amendment injections, perched water removal and passive volatilization accounted for a total of 0.8 kg (4%) based on mean BTX concentrations.

To independently ascertain the biodegradation process a mass balance was made on the nitrate introduced into the system. Based on the mass balance calculations 790 moles were depleted indicating an active denitrifying microbial population in the soil bed. Although the nitrate depletion and several positive factors such as carbon dioxide evolution following amendment injection and subsequent methane generation in the absence of available nitrate or oxygen did point towards an active biodegradation process, several constraining factors made it impractical to independently determine the mass of BTX biodegraded. Hence all possible BTX losses were accounted for, and the mass of BTX removed by biodegradation was computed by difference. Based on these calculations over 82% of the BTX reduction can be assigned to bioremediation demonstrating the synergistic effect of the two processes.

CONCLUSIONS

Pneumatic fracturing, an innovative technology for enhancing subsurface flow rates, was employed to overcome rate limitations in the transport of nutrients and electron acceptors to the microorganisms in consolidated soil, for efficient in-situ biodegradation of organic contaminants. A field-scale pilot demonstration was carried at a petroleum refinery over a period of 20 months from October 1992 to May 1994. This integrated technology has clearly been shown to be a efficient process for removal of aromatic hydrocarbons such as benzene, toluene and xylenes, especially in over-consolidated soils. Following a successful implementation of this technology at the preliminary stage, this technology is ready to be scaled up to a full-scale demonstration.

REFERENCES

US EPA, RREL, Office of Research and Development. "Accutech Pneumatic Fracturing Extraction and Hot Gas Injection, Phase I", US EPA SITE Program, Application Analysis Report, 1993.

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