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Gas production and transport during bench-scale electrical resistance heating of water and trichloroethene

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ABSTRACT

The effective remediation of chlorinated solvent source zones using in situ thermal treatment requires successful capture of gas that is produced. Replicate electrical resistance heating experiments were performed in a thin bench-scale apparatus, where water was boiled and pooled dense non-aqueous phase liquid (DNAPL) trichloroethene (TCE) and water were co-boiled in unconsolidated silica sand. Quantitative light transmission visualization was used to assess gas production and transport mechanisms. In the water boiling experiments, nucleation, growth and coalescence of the gas phase into connected channels were observed at critical gas saturations of $S_{gc} = 0.233 \pm 0.017$, which allowed for continuous gas transport out of the sand. In experiments containing a colder region above a target heated zone, condensation prevented the formation of steam channels and discrete gas clusters that mobilized into colder regions were trapped soon after discontinuous transport began. In the TCE–water experiments, co-boiling at immiscible fluid interfaces resulted in discontinuous gas transport above the DNAPL pool. Redistribution of DNAPL was also observed above the pool and at the edge of the vapor front that propagated upwards through colder regions. These results suggest that the subsurface should be heated to water boiling temperatures to facilitate gas transport from specific locations of DNAPL to extraction points and reduce the potential for DNAPL redistribution. Decreases in electric current were observed at the onset of gas phase production, which suggests that coupled electrical current and temperature measurements may provide a reliable metric to assess gas phase development.

1. Introduction

Remediation of dense non-aqueous phase liquid (DNAPL) source zones at heterogeneous sites can be inhibited by flow bypassing of injected amendments, which can lead to back diffusion and rebound from less permeable zones following treatment (Stroo et al., 2012). Thermal remediation methods can overcome these mass transfer limitations and treat less permeable zones because spatial variations in thermal and electrical properties are typically less than spatial variations in aqueous-phase permeability (Stroo et al., 2012; Triplett Kingston et al., 2010). In situ thermal treatment (ISTT) technologies are being increasingly used to treat DNAPL source zones, with electrical resistance heating (ERH) technologies accounting for over half of the applications presented in a recent review (Triplett Kingston et al., 2010).

During ISTT, increased subsurface temperatures enhance volatile organic compound (VOC) vaporization, volatilization, dissolution, desorption and DNAPL mobility, which are controlled by vapor pressure, Henry's law constant, aqueous solubility, soil–water partition coefficient, viscosity and interfacial tension with water, respectively (e.g., USEPA, 2004; USACE, 2009; Triplett Kingston et al., 2014). Although each of these mechanisms contributes to DNAPL removal, gas capture of vaporized VOCs is the dominant mechanism of removal during a thermal remediation application (e.g., Baker and

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Hiester, 2009; Beyke and Fleming, 2005; Heron et al., 2005, 2006, 2013; Vermeulen and McGee, 2000). As such, the effectiveness of ISTT requires that temperatures are high enough within the treatment volume to vaporize VOCs and establish connected gas transport pathways to soil vapor or multiphase extraction points (e.g., Baker and Hiester, 2009; Heron et al., 2006; McGee et al., 2004). In other words, the application of heat makes conditions more favorable for removal, but most thermal methods are still extraction techniques (e.g., Beyke and Fleming, 2005; Stroo et al., 2012). Consequently, failure to produce or remove gas from the subsurface can lead to poor remediation performance.

Previous studies have highlighted the importance of establishing connected gas channels for advective transport between the DNAPL source zone and extraction points (e.g., Heron et al., 1998; Martin and Kueper, 2011) and that subsurface temperatures should be increased to the boiling point of groundwater to facilitate the formation of steam conduits (e.g., Baker and Hiester, 2009; Heron et al., 2006). Martin and Kueper (2011) further exemplified this in the presence of low-permeability layers, where the absence of steam conduits resulted in local accumulation and lateral migration of co-boiled vapor leading to reduced mass removal. There is a need to understand the gas saturations at which connected pathways can be created during ISTT, and how parameters monitored during ERH, such as temperature or electric current (USACE, 2009), can be affected by gas production and transport mechanisms. These parameters could provide performance metrics that focus on gas-based removal without direct measurement of gas saturations.

A potential limitation of a thermal remediation application is convective heat loss out of the treatment volume as a result of rapid groundwater fluxes in more permeable zones, which can prevent sites from achieving water boiling temperatures (Stroo et al., 2012). For example, Triplett Kingston et al. (2012) presented results from an ERH field site where maximum representative temperatures were limited to below 90 °C as a result of convective heat loss. Furthermore, subsurface temperatures can be lower in the areas furthest from electrode or heater locations at earlier times due to non-uniform power distributions (e.g., Carrigan and Nitao, 2000; McGee and Donaldson, 2009). Condensation in these colder zones prior to vapor extraction can be detrimental to performance. For example, Baker and Hiester (2009) observed both the partial collapse of a steam front due to increased groundwater flux during thermal conductive heating (TCH) experiments, as well as DNAPL condensation at the steam front as it propagated through colder zones. The risk of condensation and subsequent dissolution of DNAPL following buoyant transport of discontinuous gas clusters (i.e., as opposed to continuous gas channels) into colder zones was also identified by McGee et al. (2004) and Krol et al. (2011a). The impacts of condensation on gas transport through these colder zones must be understood in order to inform the design of electrode or heater networks, vapor extraction wells and systems to control convective heat transfer within the treatment volume.

The objectives of this research were to: (i) measure gas saturations during water boiling and trichloroethene (TCE)–water co-boiling, (ii) identify regions of gas transport as continuous channels or discontinuous buoyant clusters, (iii) determine how gas production and transport changes electric current and temperature measurements, and (iv) investigate the effects of a colder zone on gas transport and condensation. Bench-scale ERH experiments were performed using quantitative light transmission visualization to assess gas dynamics in porous media for water boiling and TCE–water co-boiling.

2. Background

2.1. Electrical resistance heating

ERH is an ISTT technique that has the potential to remediate soil and groundwater contaminated by VOCs and semi-volatile organic compounds (sVOCs) such as chlorinated solvents and petroleum hydrocarbons. Low frequency (60 Hz) three- or six-phase electrical power is supplied to a network of subsurface electrodes, which are out of phase such that the resulting voltage potentials induce alternating current (AC) and resistive heating can be approximated by Ohm's law:

$$J = \sigma E \tag{1}$$

where J is the current density, σ is the effective subsurface electrical conductivity and E is the electric field intensity. Conceptually, current flow through the subsurface at low frequencies ($<10^6$ Hz) is primarily the result of ionic conduction through the pore water, and kinetic energy transferred to the pore water from free charge carriers accelerated by the electric field causes resistive heating (McGee and Vermeulen, 2007; Vermeulen and McGee, 2000). As such, the maximum temperature that can be achieved by resistive heating is limited to the boiling point of the pore water (e.g., Triplett Kingston et al., 2014). The power dissipated by resistive heating is dependent on the subsurface electrical conductivity and root-mean-squared (RMS) electric field intensity (e.g., Carrigan and Nitao, 2000; Krol et al., 2011b; McGee and Vermeulen, 2007; Vermeulen and McGee, 2000):

$$U = \sigma |E|^2 \tag{2}$$

where U is the resistive heating power density. Because electrical conductivities and field intensities vary in time and space, numerical models used to simulate ERH calculate transient electric potential distributions and electric field intensities from the current conservation equation to obtain resistive heating power densities (e.g., Carrigan and Nitao, 2000; Krol et al., 2011b; McGee and Vermeulen, 2007). Transient temperature distributions are calculated from the energy conservation equation, which can include resistive heat (Eq. (2)), thermal conduction, thermal convection, latent heat and heat accumulation terms (e.g., Carrigan and Nitao, 2000; Krol et al., 2011b; McGee and Vermeulen, 2007; McGee et al., 2004). Numerical models have shown that resistive heating power densities, subsurface temperatures and gas production are highest in the vicinity of the electrodes, which must be monitored to avoid dry-out and reduction in electrical conductivity (Carrigan and Nitao, 2000; Krol et al., 2011b; McGee and Donaldson, 2009; McGee and Vermeulen, 2007). Conductive and convective heat transfer, as well as the