



# Factors affecting gas migration and contaminant redistribution in heterogeneous porous media subject to electrical resistance heating



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## ARTICLE INFO

### Article history:

Received 6 August 2015

Received in revised form 20 October 2015

Accepted 29 October 2015

Available online 30 October 2015

### Keywords:

Groundwater remediation

Gas migration

Gas condensation

Nonaqueous phase liquid

Thermal remediation

Electrical resistance heating

## ABSTRACT

A series of intermediate-scale laboratory experiments were completed in a two-dimensional flow cell to investigate gas production and migration during the application of electrical resistance heating (ERH) for the removal of dense non-aqueous phase liquids (DNAPLs). Experiments consisted of heating water in homogeneous silica sand and heating 270 mL of trichloroethene (TCE) and chloroform (CF) DNAPL pools in heterogeneous silica sands, both under flowing groundwater conditions. Spatial and temporal distributions of temperature were measured using thermocouples and observations of gas production and migration were collected using front-face image capture throughout the experiments. Post-treatment soil samples were collected and analyzed to assess DNAPL removal. Results of experiments performed in homogeneous sand subject to different groundwater flow rates showed that high groundwater velocities can limit subsurface heating rates. In the DNAPL pool experiments, temperatures increased to achieve DNAPL–water co-boiling, creating estimated gas volumes of 131 and 114 L that originated from the TCE and CF pools, respectively. Produced gas migrated vertically, entered a coarse sand lens and subsequently migrated laterally beneath an overlying capillary barrier to outside the heated treatment zone where 31–56% of the original DNAPL condensed back into a DNAPL phase. These findings demonstrate that layered heterogeneity can potentially facilitate the transport of contaminants outside the treatment zone by mobilization and condensation of gas phases during ERH applications. This underscores the need for vapor phase recovery and/or control mechanisms below the water table during application of ERH in heterogeneous porous media during the co-boiling stage, which occurs prior to reaching the boiling point of water.

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## 1. Introduction

Dense non-aqueous phase liquids (DNAPLs) such as chlorinated solvents, creosote, coal tar and polychlorinated biphenyls can act as long-term sources of soil and groundwater contamination upon release to the subsurface. In situ thermal treatment (ISTT) technologies are one group of technologies with the potential to treat DNAPL-impacted sites (Stroo et al., 2012; Triplett Kingston et al., 2014). ISTT technologies include

thermal conductive heating (TCH), steam enhanced extraction (SEE) and electrical resistance heating (ERH). During the application of an ISTT technology, subsurface temperatures are increased to boil water and DNAPL. While decreased DNAPL–water interfacial tension and decreased DNAPL viscosity may aid recovery by increasing DNAPL mobility, removal of organic compounds is primarily through boiling and subsequent recovery of the gas phase using vapor or multi-phase extraction systems.

During ERH applications, heating results from the resistance of the subsurface to alternating electric current that passes between networks of electrodes. Because of its reliance on electric current flow through the pore water by ionic conduction

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(Vermeulen and McGee, 2000), maximum subsurface temperatures during ERH applications are limited to the boiling point of water, which increases with increasing depth below the water table. However, gases can be produced at temperatures below the boiling point of water due to the exsolution of dissolved gases (Hegele and Mumford, 2015) or volatile organic compounds (VOCs) (Krol et al., 2011b), as well as the co-boiling of water and DNAPL (Burghardt and Kueper, 2008). DNAPL–water co-boiling occurs because the vapor pressures of two immiscible liquids are additive at their interface, and both contribute to the total gas pressure (De Voe and Udell, 1998; Martin and Kueper, 2011; Triplett Kingston et al., 2014; Zhao et al., 2014). As such, gas production occurs at the co-boiling temperature, which is lower than the temperature of either the water or DNAPL boiling temperatures.

Heating during ISTT applications can be limited by high groundwater fluxes that transport warmer groundwater away from a target heated zone and cooler water into it (Stroo et al., 2012; Triplett Kingston et al., 2014). Beyke and Fleming (2005) presented results from an ERH field site where an unexpected high groundwater velocity of 305 cm/day through high hydraulic conductivity zones required additional hydraulic control and power input to the subsurface to reach target temperatures. Water boiling temperatures in a portion of the treatment zone during a TCH pilot test in fractured rock at a former Naval Air Warfare Center (NAWC) in New Jersey, USA, were not achieved because of inflowing cold groundwater (Lebron et al., 2012). Despite the recognition of high groundwater fluxes having the potential to limit ISTT performance, there have been relatively few studies (Krol et al., 2011a,b, 2014) that have incorporated flowing groundwater and their focus was not on cooling effects.

ISTT technologies are often applied to sites consisting of interbedded fine and coarse grained lenses and layers. In contrast, many laboratory studies have investigated ISTT in homogeneous porous media (Heron et al., 1998; Burghardt and Kueper, 2008; Krol et al., 2011a; Liu et al., 2013; Liu et al., 2014; Hegele and Mumford, 2014; Zhao et al., 2014). In addition to controlling advection and convection of the aqueous phase (Krol et al., 2014), permeability differences between layers will decrease local heating rates as a result of increased groundwater fluxes in high-permeability layers, and will promote the accumulation and lateral migration of gas beneath low-permeability layers that act as capillary barriers. Martin and Kueper (2011) conducted a laboratory study that included layers of fine and coarse sand, and observed gas production and accumulation under a fine sand layer during the co-boiling of water and pooled DNAPL. If gas capture is not sufficient, lateral gas migration facilitated by low-permeability layers has the potential to transport gas outside of an ISTT treatment zone, where VOCs can condense to form a DNAPL phase. For this reason, successful ISTT design must include a gas collection system that prevents the migration of gases to cooler regions of the subsurface (Heron et al., 2005, 2009, 2013).

The goal of this study is to investigate the effect of high- and low-permeability layers on heating and gas migration during ERH. Specific objectives are to investigate i) the influence of high groundwater flux on heating rate; ii) lateral gas migration due to the accumulation of gas under low-permeability layers during DNAPL–water co-boiling; and iii) contaminant redistribution by gas migration and subsequent condensation outside of a target heated zone. These objectives were addressed using

a series of intermediate-scale two-dimensional laboratory experiments, during which ERH was applied, temperatures and electric currents were measured, and gas formation was monitored during DNAPL–water co-boiling using digital images.

## 2. Background

### 2.1. Gas production during heating

The increase in subsurface temperatures during ERH of a DNAPL source occurs in four stages: i) DNAPL–water heating, ii) DNAPL–water co-boiling, iii) water heating, and iv) water boiling (Burghardt and Kueper, 2008; Zhao et al., 2014). During the first heating stage energy is converted to sensible heat, which increases the temperature of the water, DNAPL and solid grains. The rate of temperature increase is controlled by the input energy, heat transport, groundwater flux, and the heat capacities of the water, DNAPL and solid grains. In the absence of significant exsolution of dissolved gases, gas is first formed during the second stage (DNAPL–water co-boiling). This stage begins at the temperature (co-boiling temperature) where the sum of the vapor pressures of the water and DNAPL is equal to the sum of the liquid water pressure and the capillary pressure:

$$P_w^v + P_n^v = P_w + P_c \quad (1)$$

where  $P_w^v$  and  $P_n^v$  are the vapor pressures of water and DNAPL respectively (Pa),  $P_w$  is the liquid water pressure (Pa), and  $P_c$  is the capillary pressure (Pa). Co-boiling temperatures increase with depth below the water table as water pressure increases. The vapor pressures of water and DNAPL increase with increasing temperature, and can be approximated using the Antoine equation:

$$\log P_i^v = A_i - \frac{B_i}{T + C_i} \quad (2)$$

where  $P_i^v$  is the vapor pressure of compound  $i$  (Pa), and  $A_i$ ,  $B_i$  and  $C_i$  are compound-specific empirical constants (Yaws et al., 2005). Example co-boiling temperatures, calculated using Eqs. (1) and (2), are shown in Fig. 1 as a function of the total gas pressure ( $P_w + P_c$ ). Total gas pressure increases with increasing depth or decreasing pore throat size. During DNAPL–water co-boiling, energy is converted to latent heat as a gas phase mixture of water and VOC is produced, and no increase in temperature at the DNAPL–water interface is expected. Periods of constant temperature, referred to as temperature plateaus, have been observed in laboratory experiments of DNAPL–water co-boiling when temperatures were measured in the immediate vicinity of DNAPL (Udell, 1998; Burghardt and Kueper, 2008; Martin and Kueper, 2011); however, these temperature plateaus become less distinct as temperature measurements are made further from the DNAPL–water interface (Zhao et al., 2014). The third stage (water heating) begins once the DNAPL is depleted and gas production ceases, as energy is again converted to sensible heat and the temperature of the water and solid grains is increased. The fourth stage (water boiling) begins when temperatures reach the water boiling point (Eq. (1) with  $P_n^v = 0$ ), and a second temperature plateau occurs as energy is converted to latent heat during steam production.

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