



Gas formation in sand and clay during electrical resistance heating



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ABSTRACT

A series of electrical resistance heating (ERH) experiments were performed in a two-dimensional test cell to investigate gas production within clay lenses composed of specified mass fractions of kaolin and #20–30 silica sand (40%, 70% and 100% kaolin by mass). Temperature, electrical, photographic and piezometric data were collected during the heating of the test cell by ERH, followed by local electrical resistance measurements during cooling. In each experiment the clay lens heated more rapidly than the surrounding sand, with gas production originating at the clay-sand interface and subsequently extending outwards. The Waxman-Smits model was used to interpret electrical properties to assess the production of gas within the lens. Gas saturations immediately after ERH were estimated in the lens interiors as $S_g = 0.38 \pm 0.07$, 0.25 ± 0.05 and 0.09 ± 0.02 (for the 40%, 70% and 100% clay mass fractions). Gas saturations sufficient to produce a connected gas phase can be generated during ERH in lenses containing moderate amounts of clay.

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1. Introduction

Soil and groundwater contamination by dense, non-aqueous phase liquids (DNAPLs) such as chlorinated solvents, creosote, coal tar and polychlorinated biphenyls (PCBs) is an environmental concern throughout industrialized areas of the world. Field, laboratory and numerical modeling studies have demonstrated that DNAPL migration in the subsurface and the resulting architecture within a DNAPL source zone are strongly influenced by permeability variations at a variety of scales [1–3]. In saturated porous media with DNAPL non-wetting with respect to water, horizontal, low permeability layers and lenses provide interfaces upon which lateral spreading and pooling of DNAPL can occur. Over time, components of the DNAPL will diffuse into these capillary barriers, resulting in the sequestration of volatile organic compounds (VOCs). Back diffusion from these low permeability features during the late life-cycle stage of a DNAPL source zone may result in plume persistence and ultimately dictate the time scale of remediation [4–7].

VOCs in low-permeability features are difficult to access using injection-based remediation technologies such as in-situ bioremediation (ISB) and in-situ chemical oxidation (ISCO). Where variations in permeability exist within a treatment zone, high-permeability areas will be preferentially treated, while low-

permeability areas may retain high VOC concentrations. In-situ thermal treatment (ISTT) technologies, such as electrical resistance heating (ERH) and thermal conductive heating (TCH), are often considered for treating sites with low-permeability media because electrical and thermal conductivity vary over a smaller range than hydraulic conductivity [8]. ISTT relies upon gas production caused by elevated subsurface temperatures to remove VOCs. During heating, gas bubbles nucleate and grow until a connected gas phase is produced, which is then extracted and treated ex-situ.

ISTT technologies have been applied at contaminated sites [9,10] but there have been relatively few laboratory-scale investigations to assess fundamental mechanisms. Most experimental studies have focused on homogeneous sand, including investigations of VOC removal with heating time [11], DNAPL-water co-boiling [12,11,13], and gas development and migration [14]. Some laboratory studies have examined the removal of DNAPL from layered sands [15–17], silty soil [18], fractured clay [19] and rock [20,21], along with field-scale studies that included low-permeability media [22,23]. Martin et al. [24] specifically addressed the presence of clay lenses surrounded by sand and demonstrated that the preferential heating of the clay during ERH can produce large quantities of gas at the interface between the clay and sand. However, for enhanced removal of VOCs from a clay lens, the formation of gas at the lens interface is not sufficient, as gas must also be generated within the lens.

Gas production in clay has not been well studied in the context of ISTT, although there have been investigations related to nuclear

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waste repositories [25,26]. Gas behavior is examined in these studies in terms of a threshold pressure, above which a connected gas pathway is formed allowing gas mobilization through consolidated clay. The threshold pressure for bentonite is typically on the order of 10–17 MPa [27] and depends on the swelling pressure of the bentonite [28], water saturation [25,29] and amount of sand present in the clay blend [30]. Clays such as kaolin with lower swelling pressures have lower threshold pressures [31], and the threshold pressure decreases with increasing sand content [32–34]. These studies highlight that connected gas pathways can exist within clay lenses at low to moderate gas saturations, but the formation of those pathways is sensitive to the composition of the lens.

The objectives of this laboratory study are to: (i) determine if gas is produced in the interior of clay and sandy clay lenses during ERH, (ii) quantify gas saturations in the lens and assess the potential for the development of connected gas pathways and (iii) determine the sensitivity of gas saturations to the sand content of clay lenses.

2. Theory

2.1. Ohmic heating

ERH elevates subsurface temperatures by means of Ohmic heating whereby electrical energy is converted to thermal energy by passing current through the soil. The amount of thermal energy that can be produced is described by Ohm's law at low frequency for a passive resistor as [35]:

$$P_{ERH} = RI^2 \quad (1)$$

where P_{ERH} is the power dissipated in the ERH process (W), R is the resistance of the material between the electrodes (Ω), and I is the current passing through the heating zone (A). Both the power dissipated and current represent root-mean-square (rms) values. Ohm's law can also be represented in terms of electrical resistance:

$$R = \frac{V_{rms}}{I_{rms}} \quad (2)$$

where V is the applied electric potential (V). Resistance can be expressed as an index (R_o/R) with respect to resistance at some initial state (R_o).

2.2. Gas formation

In order for gas to be formed in a water-saturated porous medium, the vapor pressure of the liquid must equal or exceed the sum of the water pressure and a representative capillary pressure [9]:

$$P_g \geq P_w + P_c^* \quad (3)$$

where P_g is the gas (vapor) pressure at which gas forms (Pa), P_w is the water pressure (Pa) and P_c^* is the representative capillary pressure (Pa) required for nucleation. Gas first forms in microcavities on the surface of the grains, where preferential contact angles due to cavity geometry or local wetting characteristics result in a P_c^* that is lower than that found on grain surfaces, making gas nucleation at these sites favorable [36]. Once nucleated, a gas bubble can expand, filling the pore in which it was formed. This leads to internal drainage whereby the gas (non-wetting fluid) is introduced in a dispersed fashion and displaces the water (wetting fluid) from the porous medium [37–39]. The relative uniformity of internal drainage produces a relatively uniform change in electrical properties, which allows for bulk fluid saturation measurement through the monitoring of changes in electrical resistance.

The capillary pressure that has to be overcome for bubble expansion into surrounding pores is described by the Laplace equation [40]:

$$P_c = \gamma \left(\frac{1}{r_1} + \frac{1}{r_2} \right) \quad (4)$$

where P_c is the entry pressure (Pa) of the pore throat, γ is the interfacial tension (N m^{-1}) between the gas and the surrounding water, and r_1 and r_2 are the primary radii of curvature of the gas-water interface. Radii will be smaller in fine grained media than in coarse grained media, resulting in a higher capillary pressure required for gas expansion and drainage of water. It is expected that the establishment of connected gas pathways at a macroscopic scale would require a capillary pressure greater than or equal to the macroscopic air-water entry pressure of the porous medium. For example, connected gas pathways have been observed in clay and clay-sand mixtures at gas saturations between 0.07 and 0.15 [41]. The air entry value varies substantially between clay and sand. Air-water entry pressures greater than 70 kPa have been reported for kaolin at room temperature [42–44], which highlights the minimum potential air entry pressure for the clay samples used in this research, and entry pressures between 26 and 41 kPa have been reported for mixtures of sand and kaolin with clay fractions between 0.45 and 0.85 [34]. In comparison, an air-water entry pressure of 1.2 kPa has been reported for #20/30 silica sand [45]. Air-water entry pressures will decrease with increasing temperature, proportional to a decrease in interfacial tension. For example, the interfacial tension between air and water at 20 °C is 0.072 N/m while the interfacial tension between steam and water at 100 °C is 0.056 N m^{-1} [46].

During ERH, the increase in gas pressure required to exceed the sum of water pressure and capillary pressure is achieved by an increase in temperature. The relationship between vapor pressure and temperature can be described by the Clausius-Clapeyron equation [47]:

$$\ln \left(\frac{P_i^v}{P_i^{v,o}} \right) = \frac{\Delta h_{vap}}{R_g} \left(\frac{1}{T^0} - \frac{1}{T} \right) \quad (5)$$

where P_i^v is the vapor pressure of liquid i (Pa), T is the temperature (K), R_g is the universal gas constant ($8.314 \text{ J K}^{-1} \text{ mol}^{-1}$), Δh_{vap} is the heat of vaporization of the liquid, equal to 2260 kJ/kg for water [47] at 100 °C and is a weak function of temperature, and $P_i^{v,o}$ is a reference vapor pressure (Pa) measured at a reference temperature T^0 (K). Gas production will require temperatures greater than 100 °C for increased water pressure (due to depth below the water table) and when capillary pressures are high (due to smaller grain sizes). For example, for a water pressure of 150 kPa and a capillary pressure of 2 kPa (152 kPa total), Eq. (3) is satisfied at a temperature of 111 °C. However, for a water pressure of 150 kPa and a capillary pressure of 70 kPa (220 kPa total), Eq. (3) is satisfied at a temperature of 123 °C.

2.3. Gas saturation measurements

Gas saturation in a porous medium can be estimated using the Waxman-Smiths model [48] that relates changes in resistivity to water saturation. Used in the petroleum industry to estimate saturations of non-conductive oil, it can also be applied to any non-conductive fluid [49]. The Waxman-Smiths model can be expressed as [50]:

$$\sigma_{bulk} = \theta^m S_w^n \left(\sigma_w + \frac{B_T Q_V}{S_w} \right) \quad (6)$$

where σ_{bulk} is the bulk electrical conductivity of the formation (S m^{-1}), θ is the porosity (-), σ_w is the electrical conductivity of

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