Available online at www.sciencedirect.com
SCIENCE DIRECT®

Journal of Contaminant Hydrology 86 (2006) 32–52

JOURNAL OF
Contaminant
Hydrology

www.elsevier.com/locate/jconhyd

Detection of advected, reacting redox fronts from self-potential measurements

Alexis Maineult [a,*], Yves Bernabé [a], Philippe Ackerer [b]

[a] *Institut de Physique du Globe de Strasbourg, CNRS, Université Louis Pasteur, 5 rue Descartes, 67000 Strasbourg, France*
[b] *Institut de Mécanique des Fluides et des Solides de Strasbourg, CNRS, Université Louis Pasteur, 2 rue Boussingault, 67000 Strasbourg, France*

Received 13 July 2005; received in revised form 8 February 2006; accepted 9 February 2006
Available online 20 March 2006

Abstract

We report on an experiment aimed at testing the use of self-potential measurements to monitor the motion and mixing of redox reactants advected through a well-controlled, laboratory-scale, artificial aquifer system. A rectangular, plastic tank was filled up with water-saturated sand and an array of unpolarizable electrodes was installed in the sand body. A nearly uniform, steady-state flow was established by tilting the tank and controlling the water level in reservoirs at both ends. Then, we simultaneously injected a known quantity of $KMnO_4$ and $FeCl_2$, respectively, into two separate compartments forming the upstream reservoir. We thus generated two abrupt fronts, one oxidizing and the other reducing, which subsequently travelled in parallel by advection through the sand body. The $KMnO_4$ and $FeCl_2$ solutions were in contact and reacted with each other in a region located along the median vertical plane parallel to the flow direction. During flow, the electrical potential differences between each electrode and a reference located in the downstream reservoir were recorded. In the unreacted $FeCl_2$ region the electric potential showed sudden variations successively occurring at increasing distances in the flow direction, associated with the passage of the $FeCl_2$ front. These signals essentially corresponded to the junction potential produced by the difference in ionic mobility of Fe^{2+} and Cl^-. In the unreacted $KMnO_4$ region sharp signals, but with much smaller amplitudes, were also observed. Near the vertical median plane on the $FeCl_2$ side, we observed a second front associated with the spreading of the reaction zone. The shape and evolution of the reaction zone was largely controlled by the precipitation of $Fe(OH)_3$.

Keywords: Solute transport; Redox reaction; Self-potential; Junction potential; Porous media

---

[*] Corresponding author. Present address: Institute for Geology, Mineralogy and Geophysics, Ruhr-University Bochum, D-44780 Bochum, Germany. Tel.: +49 234 322 32 77; fax: +49 234 321 41 81.
E-mail address: alexis.maineult@ruhr-uni-bochum.de (A. Maineult).

0169-7722/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.jconhyd.2006.02.007

1. Introduction

Geophysical methods can be very helpful for the resolution of hydrogeological problems (e.g., Fetter, 2001, and references therein). One important advantage of geophysical surveys is that they can cover broad areas with densely spaced measurement points. Among the various geophysical techniques, self-potential (SP) monitoring is particularly useful since it is directly sensitive to changes in groundwater flow, chemistry, or thermodynamics, owing to coupling of the electric potential to gradients of, among other things, hydraulic head (i.e., electrokinetic coupling), concentration or temperature (e.g., Marshall and Madden, 1959; Nourbehecht, 1963; Corwin and Hoover, 1979). SP monitoring is an extension of SP sounding. It consists in continuously recording the electrical potential difference between unpolarizable electrodes implemented at the surface and a reference electrode, generally placed at a remote, unperturbed location. SP sounding and SP monitoring have been successfully applied to various hydrological problems, such as underground flow characterization (e.g., Ogilvy et al., 1969; Perrier et al., 1999; Titov et al., 2002; Trique et al., 2002; Rizzo et al., 2004; Revil et al., 2005), estimation of the water table position (e.g., Fournier, 1989; Birch, 1998; Sailhac and Marquis, 2001; Darnet et al., 2003; Revil et al., 2004), or characterization of fluid circulation within volcanic or geothermal systems (e.g., Anderson and Johnson, 1976; Corwin and Hoover, 1979; Revil and Pezard, 1998; Marquis et al., 2002; Zlotnicki and Nishida, 2003; Darnet et al., 2004).

Redox processes are thought to generate strong self-potential anomalies (i.e., several hundreds of millivolts). Sato and Mooney (1960) developed the so-called “geobattery” model to explain the electric signals associated with underground, electronically conductive bodies – such as ore deposits (e.g., Goldie, 2002) or graphitic seams (Bigalke and Grabner, 1997) – which connect oxidized waters near the Earth’s surface to reduced waters at depth. Strong negative SP anomalies have also been observed over contaminant plumes leaking from waste landfills (e.g., Weigel, 1989; Hämman et al., 1997; Nyquist and Corry, 2002; Naudet et al., 2003, 2004). These anomalies are correlated with the redox front that separates the reduced, microbial biodegradation zone (reduced) near the disposal, and the surrounding oxidized area (e.g., Christensen et al., 2000, 2001). The origin of the SP signal over such contaminant plumes are thought to have two main components (Naudet et al., 2003): one associated with the electrokinetic response to the subsurface flow, and the other produced by variations of the redox potential. A linearly increasing relation between the latter and the in situ redox potential was observed by Naudet et al. (2003, 2004) and Naudet and Revil (2005). The physical phenomena causing this correlation have not been clearly identified to date. Naudet and Revil (2005) proposed that bacterial biofilms acted as electron conductors connecting the reduced and oxidized zones.

Laboratory experiments performed using well-controlled, reproducible artificial aquifer systems (i.e., sand-filled tanks, through which fluid flow is generated) can be useful to investigate the effect of the relevant parameters with great accuracy. These parameters can be varied independently or in combination, thus providing a wealth of information relevant to field situations. In the context of SP monitoring, this approach was pioneered by Ahmad (1964), who used it to characterize the electrokinetic properties of Saint-Peter sand. Laboratory SP experiments have also been applied to various environmental problems. For example, Suski et al. (2004) tested a method to interpret pumping tests, and Naudet and Revil (2005) investigated the possible role of bacteria in redox-generated SP signals. In previous papers (Maineult et al., 2004, 2005), we reported on series of experiments aimed at isolating and characterizing the main components of the SP signals recorded. In the field, it is often very difficult to separate the various, simultaneous sources of the SP signals (e.g., Kulessa et al., 2003; Darnet et al., 2004).