Electrokinetically based remediation of chlorinated ethenes in heterogeneous soils



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Motivation and aim

Interesting and encouraging results have been obtained when applying electrokinetics to soil contaminated with chlorinated ethenes

Many subsurfaces are heterogeneous, which complicates remediation

This work focus on identifying important knowledge and knowledge gaps in order to engineer robust EK technologies for remediation of heterogeneous sub-surfaces contaminated with chlorinated ethenes

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Approach





Method

Literature survey. Journal papers and reports from Danish in-situ remediations

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Evaluation and discussion also involve experiences from the project group members from adjacent fields of experience

EK in combination with other remediation methods



Lima, A.T.; Hofmann, A.; Reynolds, D.R.; Ptacek, C.J.; Van Cappellen, P.; Ottosen, L.M.; Pamukcu, S., Alshawabekh, A., O'Carroll, D.M., Riis, C., Cox, E., Gent, D.B., Landis, R., Wang, J., Chowdhury, A.I.A., Secord, E.L., Sanchez-Hachair, A., 2017. Environmental Electrokinetics for a sustainable subsurface. Chemosphere 181, 122-133

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EK combined with other techniques for remediation of soil contaminated with chlorinated ethenes:

Method	Remediation	Substance transported by	Transport process	Refs
EK	Transport to treatment zone	Chlorinated ethenes	EO	(Bruell et al. 1992), (Ho et al 1997), (Ho et al. 1999b), (Ho et al. 1999a), (Athmer 2004), (Weng et al 2003), (Chang et al 2006), (Athmer 2014)
EK-BIO	In situ biotic reduction	Bacteria Substrate	EP or EO EM	(DeFlaun & Condee 1997) (Rabbi et al. 2000), (Wu et al. 2012)
		Substrate and bacteria	EM, EP and/or EO	(Hansen et al. 2015), (Mao et al. 2012a)
EK-ISCO	In situ abiotic oxidation	Permanganate	EM	(Chowdhury et al 2017b),
		Persulfate	EM	(Yang & Yeh, 2011), (Chowdhury et al 2017a)
EK-ZVI	Abiotic	Groundwater	HF	(Roh et al. 2000). (Moon
	reduction	(EK-ZVI as reactive barrier)		et al, 2005), (Chang et al. 2006), (Chen et al. 2010), (Huang & Cheng, 2012)



EK-Transport processes





Transport velocities?
Distribution of electric field in subsurface?



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EK transport velocities



Transport velocity [cm/d]

Driving force [V/cm]

EK transport velocity $[cm^2/(V \cdot d)]$

EK-BIO

	Transport of	EK transport velocity (cm ² /V day)	Velosity limiting factors
Electromigration	Electron donor	Lactate: • Fine sand ~5 (Wu et al 2007) • Clay ~3-4 (Wu et al 2007) • Subsurface clay 2.6 (Mao et. al 2012)	Dissolution/precipitationBiological activity
Electroosmosis	Advection: • Bacteria • Chlorinated ethenes	Bacteria: • Sand ~0.2 (Wick et al 2004) • Clay <0.08 (Wick et al 2004) TCE • Clay soil: 1-2 (Weng et al. 2003), (Chang et al. 2006)	 Zeta potential soil (particle size, pH and conductivity soil solution) Biological activity Solubility
Electrophoresis	Bacteria	 Sand ~0.05 (Wick et al 2004) Clay ~0.24 (Wick et al 2004) 	 Zeta potential bacteria (pH and conductivity) Soil (adhesion, pore size)

EK-ISCO

	EK transport of	Transport velocity (cm ² /V day)	Specific properties	Velocity limiting factors
Electromigration + Electroosmosis	Persulfate (S ₂ O ₈ ²⁻)	EM Silt:4.6 (Chowdhury et al 2017b) Sand: 12 (Mikkola et al. 2008) Silt: 19.8 (Mikkola et al. 2008) EO Fine grained: 2.4 (Fan et al. 2016)	Activation by: heat, mineral-based activators, UV light, and high pH Crucial to prevent development of an acidic front	Activation by soil constituents
Electromigration	Permanganate ion (MnO ₄ ⁻)	Sand/silt: 21 (Chowdhury et al 2017a) Pottery clay: 2.3 (Hodges et al 2013)	Crucial to prevent development of an acidic front	Precipitation MnO ₂ (s)

Transport velocities of 1-5 cm/day in clay soils with a hydraulic conductivity of $1\cdot 10^{-8}$ m/s require a hydraulic gradient of 2-12 m/m

..... or EK

In lab scale EK-BIO and EK-TAP have proven to supply bacteria, electron donor and chemical oxidants into fine-porous materials

Thus, EK enables BIO and TAP to work in these soils due to the transport of chemicals and bacteria into these otherwise problematic soils

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Major questions when moving from lab to field

In-situ remediation is always grey box action, as we cannot know every detail of the subsurface

The remediation methods needs to be robust to meet different scenarios, which may not even be known when designing and planning the action

Do we have the necessary basic knowledge in relation to EK?



EK transport velocity

 $(cm^2/V day)$

EM Lactate

- Fine sand ~5 (Wu et al 2007)
- Clay ~3-4 (Wu et al 2007)
- Subsurface clay 2.6 (Mao et. al 2012)

EO Bacteria

- Sand ~0.2 (Wick et al 2004)
- Clay < 0.08 (Wick et al 2004)

EO TCE

• Clay soil: 1-2 (Weng et al. 2003), (Chang et al. 2006)

EP Bacteria

- Sand ~0.05 (Wick et al 2004)
- Clay ~0.24 (Wick et al 2004)





EK transport of	Transport velocity (cm ² /V day)
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Permanganate ion (MnO ₄ ⁻)	EM Sand/silt: 21 (Chowdhury et al 2017a) Pottery clay: 2.3 (Hodges et al 2013)



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Electric field distribution in layered subsurface

We know:

- Generally electrical conductivity is low in sand compared to clay
- Electroosmotic flow is low in sand compared to clay
- Hydraulic conductivity is low in clay compared to sand
- The electric field is strongest where the electrical conductivity is highest

We aim at:

• Distributing chemical reagents and bacteria into the clay

Theoretically we obtain:

• From the general knowledge the electric field and thus the distribution of reagents and bacteria will be into the clay

But:

- The electric field distribution will be uneven
- The electrical conductivity in the soil is changing over time during EK-BIO and EK-TAP (unevenly)
- The pollutants are transported
- Hydraulic flow may prevail in some layers



Electric field distribution over time?





What happens at the boundary between sand lenses and clay?

J Appl Electrochem (2010) 40:1113–1121 DOI 10.1007/s10800-010-0075-0

ORIGINAL PAPER

Influence of boundary conditions on transient excess pore pressure during electrokinetic applications in soils

Laura Gabrieli · Akram N. Alshawabkeh

A primary challenge for engineering field implementation of EK is the development of non-linear geo- and physico-chemical conditions between the electrodes.

These conditions result in a non-uniform EO flow that is heterogeneous and transient (time and space dependent). Heterogeneous geochemical conditions result in transient fluid flow and development of negative or positive pore pressure

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EK supply of MnO₄⁻ **into heterogeneous matrices**



Electrokinetic Migration of Permanganate Through Low-Permeability Media

by David A. Reynolds¹, Edward H. Jones², Mike Gillen², Ismail Yusoff³, and David G. Thomas⁴

Conducted experiments in a two-dimensional sandbox packed with vertical layers of coarse sand and silt contaminated with aqueous TCE.

EK successfully delivered MnO_4^- throughout the silt cross-section while without EK the MnO_4^- was delivery only to the edges of the silt layer fringes.





Concluding remark

EK delivery of reactants and bacteria is superior in fine porous soils

This calls for utilization in practice as many other methods are inefficient in such soils

When developing EK techniques it is highly important not to simplify the theoretical basis to an extent where the remediation process is not reflected – overcome challenges and explore to full potential

With sufficient theoretical basis, EK techniques can be engineered to robust in-situ techniques coping with heterogeneous soil systems

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Thank you for your attention

