



Recent advances in surfactant-enhanced In-Situ Chemical Oxidation for the remediation of non-aqueous phase liquid contaminated soils and aquifers

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HIGHLIGHTS

- Understanding of S-ISCO chemical kinetics saves remediation time and resources.
- S-ISCO approached for NAPL and/or source zone remediation technology.
- S-ISCO is effective and provides near permanent solution for residual NAPLs.
- Versatile, cheap and non-food grade surfactants is highly needed.

ARTICLE INFO

Article history:

Received 11 March 2017

Received in revised form 9 August 2017

Accepted 17 August 2017

Available online 26 August 2017

Keywords:

NAPLs

S-ISCO

Surfactants

Oxidant and activators

ABSTRACT

Protecting soils and groundwater from contaminants is a global challenge and remediating contaminated sites may require decades and many resources. Non-aqueous phase liquids (NAPLs) resulting from oil and gas exploration or drilling, coal gas plants, leaking storage tanks and wood treatment facilities are major contaminants of subsurface soil and groundwater. In-Situ Chemical Oxidation (ISCO) has certain limitations with respect to cost, time, and rebound of contaminants when it is applied to remediation of NAPLs. The development of plant-based biodegradable and environmentally friendly surfactants has opened the door to developing an innovative Surfactant Enhanced In-Situ Chemical Oxidation (S-ISCO) system for remediating hydrocarbon contaminated soils and aquifers. This review highlights the state of the art of this technology and compares it to ISCO. An overview on the three most important components of S-ISCO – surfactant/co-solvent mixtures, the catalyst and/or the oxidant and the free radicals is presented. The major factors affecting the application of S-ISCO for the remediation of contaminated sites are highlighted. This review delivers a detailed explanation on the recent case studies and field applications of S-ISCO and factors affecting its implementation.

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Contents

1. Introduction.....	304
2. State-of-the-art of In-situ Chemical Oxidation.....	305

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<http://dx.doi.org/10.1016/j.eti.2017.08.004>

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2.1.	Advances of Surfactant Enhanced In-situ Chemical Oxidation	305
2.1.1.	Surfactant Enhanced Product Recovery (SEPR)	308
2.1.2.	Advantages of S-ISCO.....	308
2.2.	Surfactants/co-solvents mixtures for NAPL removal	309
2.2.1.	Mechanism of surfactants for the removal of NAPL using S-ISCO.....	309
2.2.2.	Evaluation of surfactants used for S-ISCO and their limitations	311
2.3.	Oxidants and catalyst/activators	311
2.3.1.	Activators/Catalyst chemicals and reagents	312
2.3.2.	Commonly used oxidants.....	312
2.4.	S-ISCO treatability studies	315
3.	Subsurface properties affecting the efficiency of S-ISCO	315
4.	S-ISCO field demonstrations and performances	316
4.1.	Former manufactured gas plant	316
4.2.	S-ISCO application for residential and other sites.....	317
4.3.	SEPR and S-ISCO sequential field application.....	317
4.4.	Ex-situ field application of S-ISCO	320
5.	Conclusion and future outlook.....	320
	References	320

1. Introduction

Volatile organic compounds (VOCs) are amongst the world's most widely occurring environmental contaminants of subsurface soil, groundwater and sediments because of the ubiquitous production and use of organic solvents and hydrocarbon fuels (Rivett et al., 2005). Sources of VOCs contamination range from localized point sources (e.g. leaking underground storage tanks and landfill sites) to non-point-source or diffuse pollution associated with industrial, agricultural or defence sites (Siegrist et al., 2011; Stroo et al., 2012; USEPA, 2016). The sources include fuel stations, dry cleaners, petroleum drilling, nuclear and military installations. A number of contaminated soils and groundwater sites and a range of remediation technologies have been reported (Bekele and Naidu, 2015; Dugan et al., 2010; EthicalChem, 2016b,k; Hoag, 2009; Siegrist et al., 2011). To safeguard public health and the environment, there is a need for cost-effective and rapid remedial action and an urgent need for further research and innovative technologies. Existing contaminated sites and recent spills will remain on-going serious problems. For instance, in the USA an estimated 294,000 contaminated sites with clean-up costs of \$209 billion are required from 2004 to 2033 (Siegrist et al., 2011).

If high concentrations of VOCs are present in soil, they are toxic to bacteria and recalcitrant to biodegradation (Paria, 2008). Contaminants occur in the subsurface environment dissolved in water, sorbed to soil surfaces, as soil gas, and in non-aqueous phase liquids (NAPLs). NAPLs are water immiscible and almost always embrace refractory compounds. NAPLs generally fall into two groups based on their density: light non-aqueous phase liquids (LNAPLs); and dense non-aqueous phase liquids (DNAPLs). LNAPLs are relatively soluble and volatile and tend to be relatively mobile, while DNAPLs typically have the opposite properties and tend to adsorb onto soil, and have low solubility in water (CLU-IN, 2016; Müller and Sedláčková, 2003; Mulligan et al., 2001; Siegrist et al., 2011). Common examples of DNAPLs include chlorinated solvents such as trichloroethene (TCE), tetrachloroethylene (PCE or Perc); creosote, coal tar, TCA whereas LNAPLs are usually fuel products, petroleum hydrocarbons (TPHs) and gasoline. Due to their water immiscibility and tendency to be sorbed to soil materials, effective remedial technologies for NAPLs have been difficult to implement.

After being spilt on soil, migration of NAPL occurs by gravitational and capillary forces. It becomes a non-continuous phase and forms residual mobile globules (Mulligan et al., 2001). Globules are entrapped in pores by capillary forces in soil resulting in residual saturation. Permeation of NAPLs results in their retention in the unsaturated zone. Further infiltration could trap NAPLs in the saturated zone. Consequently, the dissolution of NAPLs by residual saturation by water infiltrating gradually through the unsaturated zone to the water table pollutes the groundwater (Müller and Sedláčková, 2003).

Recently, the remediation of NAPLs in groundwater using the pump-and-treat system has been improved to create more effective and sustainable technologies of in-situ plume treatment. More recently source zone remediation has been developed (EthicalChem, 2016b; Siegrist et al., 2011; Stroo et al., 2012). In-situ chemical oxidation (ISCO) (Bekele and Naidu, 2015; Siegrist et al., 2011), in-situ surfactant/co-solvent flushing (Dugan et al., 2010; Mao et al., 2015; Strbak, 2000), surfactant enhanced aquifer remediation (SEAR) (Abriola et al., 2005), in-situ thermal treatment (Kingston et al., 2010), in-situ bioremediation (Kuppusamy et al., 2016) and in-situ chemical reduction (Stroo et al., 2012; Wit, 2013) were reported to remediate source zone NAPLs contamination.

The application of ISCO to remediate NAPLs is limited to the dissolved phase in the subsurface, and is not suitable to remediate sorbed or residual contaminants, ganglia of NAPLs in the unsaturated soils and source zone NAPLs (Brebbia, 2013; Collins, 2012; Dugan et al., 2010; Hoag and Collins, 2011; Siegrist et al., 2011; Stroo et al., 2012; Wang et al., 2013). In the last decade Surfactant Enhanced In-Situ Chemical Oxidation (S-ISCO) has been extensively used for in-situ remediation technologies for removing NAPLs from soil and groundwater (Brebbia, 2013; EthicalChem, 2016d,e,f; Lanoue et al., 2011). ISCO involves the injection or direct mixing of reactive chemical oxidants directly into the contaminated medium (soil or groundwater) to destroy chemical contaminants in place. S-ISCO involves the reactive transport of surfactant and co-solvent

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