



Review

Fenton based remediation of polycyclic aromatic hydrocarbons-contaminated soils

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ABSTRACT

This paper aims to review the applications of Fenton based treatments specifically for polycyclic aromatic hydrocarbons-contaminated soils. An overview of the background and principles of Fenton treatment catalysed by both homogenous (conventional and modified Fenton) and heterogeneous (Fenton-like) catalysts is firstly presented. Laboratory and field soil remediation studies are then discussed in terms of efficiency, kinetics and associated factors. Four main scopes of integrated Fenton treatments, i.e. physical-Fenton, biological-Fenton, electro-Fenton and photo-Fenton are also reviewed in this paper. For each of these integrated remediation technologies, the theoretical background and mechanisms are detailed alongside with achievable removal efficiencies for polycyclic aromatic hydrocarbons in contaminated soils compared to sole Fenton treatment. Finally, the environmental impacts of Fenton based soil treatments are documented and discussed.

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Nomenclature

(OH)C ₆ H ₆ ·	hydroxycyclohexadienyl radical	HEIDA	disodium ethanoldiglycine
[H ₃ O ₂] ⁺	oxonium ions	HMW	high molecular weight
≡Fe	surface iron species	HO ₂	perhydroxyl radical
AC	alternating current	HPCD	hydroxylpropyl β-CD
ANT	anthracene	HP-MF	hydrogen peroxide-modified Fenton
AOPs	advanced oxidation processes	ISCO	in situ chemical oxidation
BAA	benzo[a]anthracene	KH ₂ PO ₄	potassium dihydrogen phosphate
BAP	benzo[a]pyrene	K _{ow}	octanol–water partition coefficient
BAP-1,6-dione	benzo[a]pyrene-1,6-dione	LMW	low molecular weight
BAP-6,12-dione	benzo[a]pyrene-6,12-dione	MCD	methylated β-CD
BBF	benzo[b]fluoranthene	MGP	manufactured gas plants
CDs	cyclodextrins	NAP	naphthalene
CHR	chrysene	NAPL	non-aqueous phase liquid
CMC	critical micelle concentrations	NOM	natural organic matter
CMCD	carboxymethyl β-CD	NTA	nitritotriacetic acid
CO ₂	carbon dioxide	O ₂ ^{·−}	superoxide radical
CP-MF	calcium peroxide-modified Fenton	OH·	hydroxyl radical
DAA	dibenzo[a,h]anthracene	PAH	polycyclic aromatic hydrocarbons
DC	direct current	PHE	phenanthrene
DNAPL	dense non-aqueous phase liquid	PKO	palm kernel oil
DTPA	diethylene triamine pentaacetic acid	PYR	pyrene
EDTA	ethylenediaminetetraacetic acid	SBR	sequential batch reactor
EK	electrokinetic	SDS	sodium dodecyl sulphate
EK-Fenton	electrokinetic-Fenton	SOM	soil organic matter
EOF	electroosmotic flow	SPS	sodium persulfate
Fe ²⁺	ferrous ion	TOC	total organic carbon
Fe ³⁺	ferric ion	UV	ultra-violet
Fe ₃ O ₄	magnetite	VOCs	volatile organic compounds
Fe-DTPA	iron chelated diethylene triamine pentaacetic acid	α-Fe ₁₀ O ₁₅ ·9H ₂ O	ferrihydrite
Fe-EDTA	iron chelated ethylenediaminetetraacetic acid	α-FeOOH	goethite
FL	fluorene	α-Fe ₂ O ₃	hematite
FLT	fluoranthene		
H ₂ O ₂	hydrogen peroxide		
H ₂ SO ₄	sulphuric acid		

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of several hundred individual compounds containing at least two condensed aromatic rings that are known to be toxic, mutagenic, carcinogenic and teratogenic. They are common pollutants in the environment, and particularly high concentrations are found at sites contaminated with coal tar and creosote, especially as the heritage from the manufactured gas plants (MGP) and wood treatment facilities of the last few centuries. In the United States, for example, there are 1500 former MGP with estimated clean up costs ranging from \$1 million to \$10 millions per site (Electric Power Research Institute, March 2010). At seriously contaminated sites, the contamination level of PAHs has been reported to be as high as 300 g kg^{−1} soil (Kanaly and Harayama, 2000). PAHs are very persistent showing extended natural attenuation times. The half life of PAHs in contaminated soils can be as long as 5.7 years and 9.1 years for low molecular weight (LMW) and high molecular weight (HMW) PAHs respectively (Wild et al., 1991), thus demonstrating the need for remediation processes to accelerate the clean-up of PAH-contaminated soils. PAHs in soil can be strongly sorbed to soil organic matter (SOM), encapsulated in soil mineral, and can also be present in dense non-aqueous phase liquid (DNAPL), which makes the remediation process difficult.

Many types of remediation technologies have been explored for the removal of PAHs from soils, involving solely one or a combina-

tion of physical, chemical, biological and thermal processes (Gan et al., 2009). Faster and more efficient degradation of recalcitrant compounds such as PAHs can be achieved using advanced oxidation processes (AOPs) (Kawahara et al., 1995; Nam et al., 2001; Watts et al., 2002). Fenton treatment is a widely studied technique that is classified under AOPs. The Fenton reagent was developed by Henry John Horstman Fenton in the 1890s (Fenton, 1893, 1894a,b). It has been commonly used as an oxidising agent for organic contaminants in industrial wastewaters since 1960s (Huang et al., 1993; Bautista et al., 2008). However, studies on its potential applications in soil remediation only started in 1990s on a laboratory scale (Tyre et al., 1991; Watts et al., 1991, 1993, 1994; Ravikumur and Gurol, 1994; Karkala and Watts, 1997), and the first study of a Fenton based treatment for PAH-contaminated soil was conducted by Martens and Frankenberg in 1995 using surfactant (Martens and Frankenberg, 1995).

Over the last two decades, Fenton treatment has emerged as a viable remediation technology for PAH-contaminated soils. Several reviews on various Fenton based treatments for contaminated soils have been published (Munter, 2001; Watts and Teel, 2005; di Palma, 2005; Pignatello et al., 2006). In these works, PAHs have been grouped as hydrophobic or semi-volatile contaminants and the discussions presented were broadly for hydrophobic or semi-volatile contaminants. For PAHs specifically, Rivas (2006) in his review on chemical oxidation treatments for soil PAHs briefly discussed the overall considerations in

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