

Review

Polycyclic aromatic hydrocarbons sorbed on soils: A short review of chemical oxidation based treatments

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Abstract

A brief review is completed on the chemical oxidation of PAHs contaminated soils. Previously, the adsorption and extraction processes are also documented and discussed. The tree main technologies reported in the specialised literature include the use of ozone, hydrogen peroxide (with or without ferrous iron addition) and high temperature pressurized water (both in sub- and super-critical conditions) in the presence of an added oxidant like hydrogen peroxide, oxygen, persulfate, etc. Each process is detailed by the influence of the main operating variables reported in the literature (i.e. presence of organic matter, soil type, reagents dosage, etc.), the kinetics and the description of integrated treatments (i.e. chemical oxidation + biodegradation).

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of compounds that consist of two or more fused benzene rings. They are the by-products of incomplete combustion or pyrolysis of virtually all organic matter. PAHs are considered to be the most widely distributed class of potent human carcinogens present in the environment, and a number of them are catalogued as potential carcinogens by various international agencies [1]. Additionally, other non-cancer-causing effects have not been deeply investigated yet. In aquatic systems, PAHs increase their toxicity with the increasing molecular weight. Although the rate of uptake from the environment is variable among species, bioaccumulation tends to be rapid [2].

PAHs are characterized by their palpable hydrophobic nature. Consequently, these species tend to be adsorbed on solid particulates, especially on the organic fraction of the solids [3]. Accordingly, developing techniques intended to remediate contaminated soils has to consider all the features associated to the adsorption–desorption stages and availability of PAHs to added chemicals.

2. The sorption stage

The sorption of hydrophobic organic compounds (as in the case of PAHs) onto soils has been widely investigated by a number of authors [4–11].

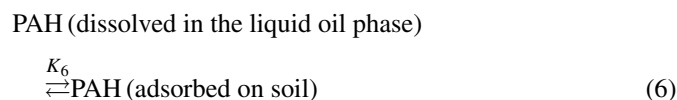
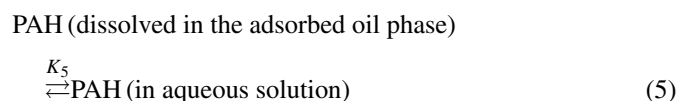
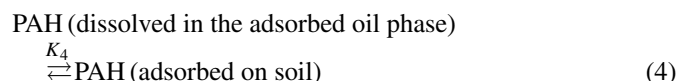
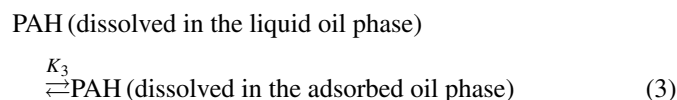
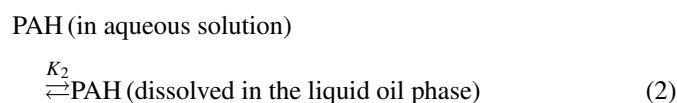
Broadly speaking the most influencing parameters affecting PAHs sorption onto soils are the solubility of the different PAHs and their organic fraction. Other potential factors exerting some influence on the adsorption stage like temperature, salinity or the presence of dissolved organic matter are in fact variables which influence the solubility of the aromatic compounds.

Intimately related to the adsorption process is the sequestration phenomenon. The sequestration is an aging stage in which hydrophobic adsorbed substances show a declining availability to both bioremediation and chemical extraction. The occurrence of sequestration is of significant importance since the toxicity values of sequestered molecules are, obviously, quite different from those assigned to dissolved substances or weakly adsorbed molecules. Physically this phenomenon consists in a migration of adsorbed substances into condensed organic matter and inaccessible micropores present in the geosorbent [7]. Luthy et al. [12] suggested the interaction of several surfaces and matrixes with hydrophobic substances in the sequestration process. Thus, these authors suggest accounting for the presence of inorganic micropores, organic matter in the solid, combustion carbon particulates and spilled organic liquids (i.e. oils). The presence of sub-phases in the solid involves a complex mechanism of mass transfer phenomena that are normally studied at the macroscopic scale by means of static or dynamic experiments [9]. In the majority of cases the adsorption of PAHs on soils follows a linear isotherm [8], i.e., the sorption stage is similar to the partition of non-ionic substances between a hydrophobic and a hydrophilic phase. Nevertheless, non-linear behaviour has also been reported [13]. Luthy et al. [12] give some general rules of thumb to account for the experimental isotherm shapes. These

authors differentiate between five sorption mechanism hypotheses, however, in natural processes more than one case might simultaneously occur, consequently mixing the expected macroscopic results. The five potential cases reported were:

- A. Sorption into amorphous organic matter or non-aqueous phase liquids. Characterized by fast kinetics, linear isotherm, low activation energy and sorption heat with a high chemical extractability.
- B. Sorption into condensed organic polymeric matter or combustion residues. The kinetics is slow showing hysteresis in the adsorption–desorption curves. Upon reaching equilibrium conditions, the isotherm is linear. The activation energy is usually high as well as the heat of sorption. The solvent extractability is catalogued as low.
- C. Adsorption onto water wet organic surfaces. Is a fast process showing non-linear isotherm shapes and low activation energy and low to high (depending on contaminant hydrophobicity) sorption heat. The sorbate can be easily extracted by the adequate solvent.
- D. Adsorption to water wet inorganic surfaces. Characterized by fast kinetics and linear isotherm. The thermodynamic parameters are quite similar to the previous case.
- E. Adsorption into the microporous structure of the solid. This situation is similar to case B with analogous characteristics.

Other authors, however, only consider the soil particulate as a whole entity (organic and inorganic fractions of the soil are taken as a unique entity). Other components not being part of the soil (i.e. oils, dissolved organic matter, etc.) and influencing the adsorption process can also be adsorbed or not onto the solid. Thus, Walter et al. [9] propose a lumped model to account for the transference of PAHs in a three phase system according to



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