



Experimental and theoretical investigation of the removal organic pollutants from contaminated soils using superheated water



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ABSTRACT

The experimental and theoretical remediation of polycyclic aromatic hydrocarbons (PAHs) contaminated soil has been investigated by superheated water extraction. In this study, effect of three parameters on extraction efficiency has been investigated including the extraction temperature, time, and flow rate. Water temperature was varied between 100 and 165 °C, extraction time between 5 and 15 min, and flow rate between 1 and 2.5 mL/min. Specifically, this study examines the effectiveness of different parameters on the removal of phenanthrene (a representative PAH) from the soils. More than 83% extraction of the phenanthrene from contaminated soil was observed at 165 °C for 15 min and flow rate of 1.75 mL/min at a constant pressure of 20 bar. Also, a mathematical model for soil remediation was developed which predicted the experimental data very well. Both the model and experimental results suggested that the extraction efficiency was extremely dependent on extraction temperature and time.

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1. Introduction

One of the most significant contaminants found in polluted sites is polycyclic aromatic hydrocarbons (PAHs) [1]. PAHs are toxic hydrophobic organic contaminants, which consist of two or more fused aromatic rings [2]. PAHs are micro-pollutants that because of their toxicity and their mutagenic properties have been listed as most dangerous pollutants and because of their hydrophobic nature, PAHs are highly resistant to environmental degradation [3,4]. The U.S. Environmental Protection Agency has categorized 16 PAHs as priority environmental contaminants [5]. Due to their persistence in the environment through contamination of soils, sediments and water, remediating these pollutants is necessary [6]. When the concentrations of the pollutants have come down to the desired level, the remediation is considered to be successful [7].

Some of researchers have studied the remediation of PAHs contaminated soil using the phytoremediation and bioremediation process [4]. Unfortunately, removal of PAHs (especially higher molecular weight species) in this process are often poor, although bioremediation is a very cost-effective technique [8]. Phenanthrene (PHE) that is a tricyclic aromatic hydrocarbon with three-fused rings in an angular fashion, is commonly used as a model compound for PAH bioremediation studies [5].

Superheated water extraction (SWE) is a new extracting method that has attracted the attention of many researchers [9,10]. SWE is a novel and powerful technique based on the use of water. The term “superheated water” is referred to the region of condensed phase of water between the temperature ranges of 100–374 °C [11]. In this region water is much less polar and organic compounds are much more soluble in it than at ambient conditions [12]. The ability of water to extract nonpolar organics such as polychlorinated biphenyls (PCBs) and PAHs is linked to the fact that the polarity of water can be reduced significantly with rising temperature. The dielectric constant (ϵ) of water at 25 °C is nearly 80, so it is a polar solvent. With enough pressure to maintain water in the liquid phase at elevated temperature, the dielectric constant decreases to 27 at 250 °C, this is between those of methanol ($\epsilon=33$) and ethanol ($\epsilon=24$) at 25 °C [13]. These temperature effects mean that superheated water can have a solvating power very similar to organic solvents which can dissolve a wide range of medium to low polarity analytes [14].

Superheated water has very special characteristics; such as dramatically decreasing of dielectric constant, viscosity and surface tension with increasing temperature under enough pressures applied to maintain water in liquid phase [15]. Moreover, water is environmentally friendly, easily available, non-toxic and it can easily be obtained and disposed off. Hence, SWE has steadily become an efficient and low cost method of extraction for less-polar organic components from environmental soil, sediments and plant materials [16].

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Nomenclature

c_i	solute concentration in the bulk fluid phase (g/cm ³)
c_s	solute concentration in the surface of particle (g/cm ³)
c_{is}	solute concentration in the fluid phase at the particle surface (g/cm ³)
D_{AB}	binary diffusion coefficient of phenanthrene in superheated water (cm ² /s)
d_b	bed diameter (cm)
D_1	axial dispersion coefficient (cm ² /s)
d_p	particle diameter (cm)
E_{exp}	experimental data
E_{model}	model predicted data
k_f	external mass transfer coefficient
k_p	volumetric partition coefficient of the solute between the solid and the fluid phase
k_d	thermodynamics partitioning coefficient
M_B	molecular weight of fluid
Pe	Peclet number
Re	Reynolds number
Sc	Schmidt number
Sh	Sherwood number
T	temperature (K)
t	time (s)
u_z	superficial velocity (cm/s)
V_A	molar volume of the solute at its normal boiling point (cm ³ /mol)
z	bed height (cm)
α	specific surface (m ² /m ³)
ε	void volume fraction
ρ	density of water (kg/L)
φ	association factor for the solvent
μ	viscosity (Pa s)

SWE has been reported for the extraction of PAHs [4,7,8,17], PCBs [18] and pesticides [8] from contaminated soil. However, the extraction of PAHs in contaminated soil for practical or commercial applications has not been sufficiently carried out [5] and superheated water extraction modeling is still in its infancy. The extraction of materials from solid matrices in packed beds forms the basis of most present day Research on superheated water extraction, Therefore a better understanding of the basic phenomena which controls the process of superheated water extraction is required.

Aim of this study is investigation of remediation of phenanthrene as a polycyclic aromatic hydrocarbon (PAH) from contaminated soil by extraction using superheated water and to develop a mathematical model for simulate the remediation of contaminants from soil and to describe the behavior of superheated water extraction system.

2. Materials and methods

2.1. Chemicals

Doubly distilled water purified through a Milli-Q deionizing unit (Millipore, Bedford, MA) was used as the extraction solvent. Phenanthrene (purity >98%) was purchased from Sigma Aldrich. Methanol (purity >99%) was used to prepare samples and standard for UV analysis and to prepare spiking solutions.

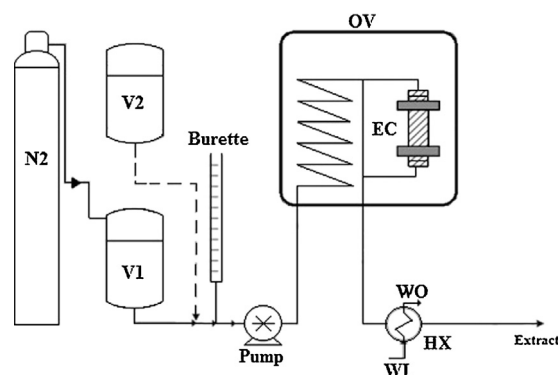


Fig. 1. Schematic diagram of superheated water extraction system (IROST): V1: water reservoir; OV: oven; EC: extraction cell; HX: heat exchanger; WI: cooling water in; WO: cooling water out.

Table 1

Experimental design for remediation of phenanthrene from contaminated soil.

Experiments	Water temperature (°C)	Extraction time (min)	Flow rate (mL/min)
Temperature effect	100	15	1.75
	110		
	120		
	125		
	130		
	140		
	145		
	150		
Time effect	155	5	1.75
		10	
		15	
Flow rate effect	155	15	1
			1.75
			2
			2.5

2.2. Contaminated soil

In this study, sand (used in sand baths) was used to simulate soil. Phenanthrene has been spiked in to soil to prepare contaminated soil. 100 mg phenanthrene was inserted in to 100 mL of methanol to make the spike solution. The spiking solution was added in to the 100 g soil and mixed by rotating mixture for 2 h, then the contaminated soil was dried for 24 h and maintained for 3 days.

2.3. Superheated water extraction process

A schematic diagram of superheated water extraction apparatus is presented in Fig. 1. The lab-scale superheated water extractor was described in a previous work [19]. Doubly distilled water was first purged for 20 min with nitrogen to remove dissolved oxygen and then filled in to the pump. 5 g of contaminated soil was packed into the extraction cell and the extraction cell was assembled in the oven and the pump was turned on and let pressure up to 20 bar. Then, the pump was turned off and temperature of oven was increased to the required temperature, this process took about 15 min. Then the pump was turned on and the flow rate was adjusted at the desired rate. Water was passed vertically down-flow through the extraction cell. To investigate the effect of temperature, flow rate of water and extraction time, experiments were performed according to the set of experimental designs presented in Table 1. The counting of extraction time was started after the oven

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