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Fluidized countercurrent solvent extraction of oil pollutants from contaminated soil. Part 1: Fluid mechanics

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ABSTRACT

Liquid–solid countercurrent solvent extraction is a potential application way for the removal of oil pollutant from contaminated soil. This study focuses primarily on the fluidization performance of liquid–solid two-phase flow in an extraction column (2.1 m × 50 mm diameter), with the solvent and solid flow rates ranging from 10 to 800 L h⁻¹ and from 8 to 107 kg h⁻¹, respectively. Several key hydrodynamic parameters, including pressure drop, solid holdup, superficial velocity of the solvent and solid, and dimensionless slip velocity, were investigated experimentally. At a given solid flow rate, a uniform axial distribution of solid holdup was observed at low solvent flow rate, whereas the axial solid holdup gradient increased with the solvent flow rate. The appearance of a solid holdup gradient was mainly attributed to the presence of small particles. An operating curve for the soil remediation by countercurrent solvent extraction was obtained according to the critical flow rate of the solvent and solid for predicting the occurrence of flooding and the appearance of the solid holdup gradient.

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Keywords: Solvent extraction; Countercurrent flow; Fluidization; Solid holdup; Pressure drop

1. Introduction

In recent years, petroleum contamination in soil has received increasing social concern all over the world. The remediation of the foul soil accordingly has become an urgent task for the removal of oil pollutant from contaminated soil. Liquid–solid countercurrent solvent extraction is a potential technique, particularly for soil historically contaminated in a high oil concentration (Lian et al., 2008) (up to 140 g kg⁻¹) and large scale application. As prerequisite, a full understanding of the fluidization characteristic in extraction column is necessary and inevitable; unfortunately, it so far remains still scanty due to

the inherent complexity of liquid–solid two-phase flow especially countercurrent behavior. Solvent extraction technology for soil remediation usually involves three key problems: (1) the selection of an effective and environment-friendly solvent; (2) the development of proper equipment, and (3) the recycling of the solvent in system. In addition, cost in equipment investment and operation, solid feeding, sludge discharging and flow control and scale-up effect would also become huge challenges for operational-scale application.

During the past decades, considerable efforts have been devoted to solvent selection (Viglianti et al., 2006; Gong et al., 2005; Lau et al., 2014) and solvent recycling (Ahn et al., 2008;

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Gong et al., 2007; Wu et al., 2011). In general case, solvent could be conveniently recovered and reused by distillation with a satisfactory recovery rate. The pollutant organic components could be used for industrial materials or fuel oil. However, few studies have reported on the development of large-scale industrial equipment for continuous operation. Most of the existing studies on soil cleanup via solvent extraction were carried out on a bench scale under precisely controlled conditions (e.g., temperature and pressure), in which the mass transfer was facilitated by shakers and ultrasonication (Wu et al., 2011, 2013a,b; Zhou and Zhu, 2007; Silva et al., 2005). Although a few studies (U.S. Environmental Protection Agency, 1990) with larger treatment capacities (up to $45 \text{ kg soil h}^{-1}$) had also been conducted, they were often run in batches at a packed column or mix tank. Obviously, the continuous operation has remarkable competitive advantages over the batch operation with large productivity, high efficiency, and low energy consumption. Most recently, Li et al. (2012) demonstrated the high efficiency of crosscurrent and countercurrent solvent extraction, where up to 97% of heavy crude oil was removed from contaminated soil. Future works are required to extend this technology from the laboratory to industrial application for dealing with larger volumes of contaminated soil. One technical bottleneck in such scale-up is equipment design and the associated control system, including solid feeding, liquid pumping, sludge discharging and flow control. These issues are usually ignored in laboratory experiments, but remain major challenges in actual remediation.

As it is well known, on lacking an essential understanding of fluid dynamics, it is very difficult even infeasible for designing a complicated continuous operation system such as continuous liquid–solid countercurrent solvent extraction column. For example, the balance between feeding and discharging rates requires prior knowledge on the pressure drop of continuous fluid, as well as the sedimentation velocity and the particle size distribution of solid particles, and these factors would determine the hydrodynamic characteristic and mass transfer performance inside the extraction column. To our knowledge, no research has reported in the literature about the fluidization hydromechanics of a liquid–solid two-phase countercurrent flow system for soil remediation engineering.

In our previous studies, a remediation technology including solvent extraction, solvent recycling and biodegradation processes was conceptualized for removing high concentration of aged oil pollutant from soil (Wu, 2012). The total petroleum hydrocarbons were decreased rapidly from 140 to 14 g kg^{-1} via solvent extraction using TU-A solvent (mixture of hexane and pentane, 4:1, v/v) successively, the residue oil was intensively biodegraded to less than 4 g kg^{-1} . The net consumption of solvent and water were 53 L and 200 L for treating 1 ton of foul soil, respectively. However, all experiments were carried out on a bench scale and the conceptualized process was only an idealistically prediction under no consideration of scale-up effect.

On basis of the aforementioned studies, this paper aims at understanding the countercurrent fluid mechanisms of organic solvent and solid particles in an extraction column ($2.1 \text{ m} \times 50 \text{ mm}$ diameter). To facilitate the experimental investigation, in this work, the glass beads were used as solid particles instead of practical soil considering their approximate density and settling properties (Garside and Al-Dibouni, 1977) and the diameters of the glass beads span from $95 \mu\text{m}$ to $285 \mu\text{m}$. The objective of the present study is expected as

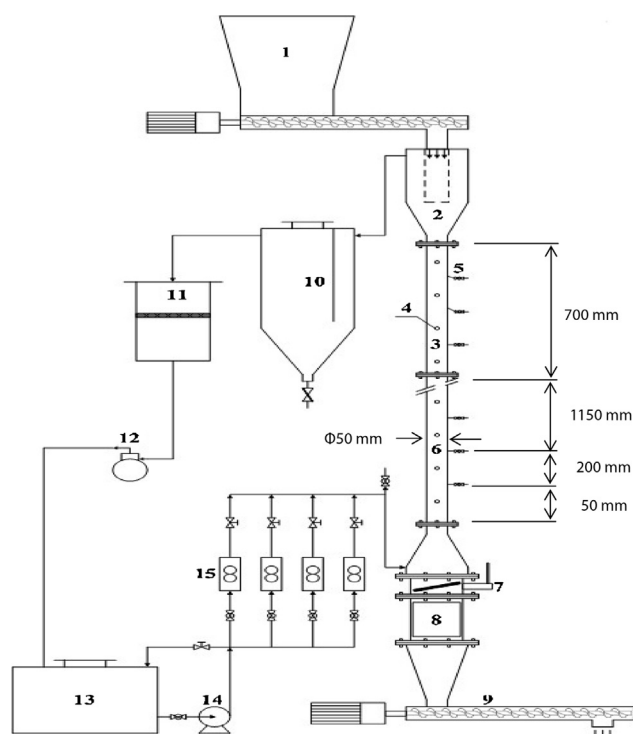


Fig. 1 – Schematic of the countercurrent flow experimental setup. (1) Solid feeder, (2) jacketed column head, (3) column part 1, (4) pressure tap, (5) sampling point, (6) column part 2, (7) controlling valve, (8) bottom viewport, (9) dumping tube, (10) buffer, (11) filter, (12) recycling pump, (13) solvent tank, (14) solvent pump, and (15) flow meter.

follows: (i) to establish a continuous countercurrent extraction system and assess its basic hydro-mechanical performance, and (ii) to determine the operating curves of the continuous solvent extraction processes.

2. Materials and methods

2.1. Experimental setup

The composite TU-A solvent used in this work consisted of hexane and pentane (4:1), of which density and viscosity are 710 kg m^{-3} and 0.45 cp , respectively, as described by Wu et al. (2011). There were three kinds of glass beads with narrow particle size distribution, of which the average diameters were $285 \mu\text{m}$, $170 \mu\text{m}$ and $95 \mu\text{m}$ with the density of 2400 kg m^{-3} , and each type was used at a time. The size distribution of the glass beads obtained by laser particle analyzer is shown in Fig. SM-1. Hydromechanical experiments were carried out in a fluidized two-phase flow system with continuous countercurrent flow column, flow rate controllers, and solvent recycling units (Fig. 1) at temperature 298.15 K and atmospheric pressure. The system ran for about 15 min to obtain stability before reading the pressure gauges and undertake samplings. The column (2.1 m height \times 50 mm diameter) was constructed with two parts connected by flanges, on which 22 taps were placed homogeneously for sampling and pressure tests. The column was initially wet with TU-A solvent. Solids were fed from the top of the column through a solid feeder (Fig. SM-2). This feeder was composed of a double-helix feeding tube driven by a direct-current brushless speed motor (Fig. SM-3) and a hopper with an arch breaker for stirring solids (Fig. SM-4).

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