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Research article

Simultaneous application of chemical oxidation and extraction processes is effective at remediating soil Co-contaminated with petroleum and heavy metals

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ABSTRACT

Chemical extraction and oxidation processes to clean up heavy metals and hydrocarbon from soil have a higher remediation efficiency and take less time than other remediation processes. In batch extraction/oxidation process, 3% hydrogen peroxide (H₂O₂) and 0.1 M ethylenediaminetetraacetic acid (EDTA) could remove approximately 70% of the petroleum and 60% of the Cu and Pb in the soil, respectively. In particular, petroleum was effectively oxidized by H₂O₂ without addition of any catalysts through dissolution of Fe oxides in natural soils. Furthermore, heavy metals bound to Fe-Mn oxyhydroxides could be extracted by metal-EDTA as well as Fe-EDTA complexation due to the high affinity of EDTA for metals. However, the strong binding of Fe-EDTA inhibited the oxidation of petroleum in the extraction-oxidation sequential process because Fe was removed during the extraction process with EDTA. The oxidation-extraction sequential process did not significantly enhance the extraction of heavy metals from soil, because a small portion of heavy metals remained bound to organic matter. Overall, simultaneous application of oxidation and extraction processes resulted in highly efficient removal of both contaminants; this approach can be used to remove co-contaminants from soil in a short amount of time at a reasonable cost.

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1. Introduction

Remediation of soils co-contaminated with organic compounds and heavy metals as well as either of these types of contaminant alone is considered an important environmental issue (Atagana, 2011; Sprocati et al., 2012; Zhou et al., 2014). In Korea, the incidence of co-contamination of soils around factory sites, open storage spaces for ore and scrap metal, waste landfills, and traffic facilities with petroleum and heavy metals has increased over the past few. Multiple remediation process are required to remove mixed contaminants, because each contaminant has different characteristics as well as removal mechanisms (Cang et al., 2013;

Colacicco et al., 2010). Heavy metals including Cd, Cu, Ni, Pb, and Zn can be removed via mass transfer into aqueous phase using inorganic acidic solution or chelating agents by extraction, desorption, and dissolution from soils (Dermont et al., 2008; Yoo et al., 2013, 2015a). Petroleum-related organic compounds can be removed by applying oxidizing or surfactant solutions to soil (Khalladi et al., 2009; Lee et al., 2005; Urum et al., 2004), and contaminants can be completely oxidized to carbon dioxide and water molecules by bioremediation or chemical oxidation.

Among the available remediation processes for cleaning up co-contaminated soil, cost effective and eco-friendly bioremediation has been used to simultaneously remove co-contaminants including heavy metals and organic compounds from soils (Alisi et al., 2009; Dong et al., 2013; Gutierrez-Gines et al., 2014; Park et al., 2013; Sprocati et al., 2012; Sun et al., 2011). However, the performance of the technique is highly dependent on the surrounding environment, and remediation takes a long time. Furthermore, some studies have reported that bioremediation is

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not effective in certain co-contaminated soils because toxic metals may inhibit the biodegradation of hydrocarbons (Alisi et al., 2009; Gutierrez-Gines et al., 2014; Singh and Cameotra, 2013). Another remediation technique to clean up soils co-contaminated with petroleum and heavy metals, electrokinetic remediation, has been studied over the past decade.

Electrokinetic technique can be used to remove co-contaminants both in situ and ex situ (Dong et al., 2013; Maturi and Reddy, 2006).

However, this technique is more suited to fine grained soils than other soil types, and treatment takes a long time. In general, chemical extraction/oxidation processes are preferred over *ex-situ* electrokinetic processes in sandy soil. Chemical extraction/oxidation processes can result in effective removal of organic compounds and heavy metals in soil via direct contact between the contaminants and extractants/oxidants within a short period of time when stirring is performed (Eve, 1998). In particular, co-contaminants, including heavy metals and organic compounds, can be removed via direct application of extracting agents and/or surfactant either successively or concurrently to soil. Khodadoust et al. (2005) investigated the effect of sequential extraction using various metal and organic extractants from field soil co-contaminated with phenanthrene, Pb, and Zn. They found that individual or sequential application of metal extractants and surfactants mainly extracted metal and organic contaminants respectively, regardless of application sequence. Lo et al. (2012) found that sequential HCl extraction and persulfate oxidation by a Fe-EDDS treatment process was effective at removing both heavy metals and polycyclic aromatic hydrocarbons (PAHs) from river sediment. However, more studies are required to investigate the effect of the application sequence of extraction and oxidation processes to remove contaminants from co-contaminated soil.

Laboratory-scale chemical extraction/oxidation processes were applied to clean up soil co-contaminated with petroleum hydrocarbons and heavy metals. It has been investigated that the effect of application sequence of chemical extraction and oxidation on the overall performance of the technique. In particular, three sequences – extraction–oxidation, oxidation–extraction, and simultaneous application – were designed to evaluate remediation of co-contaminated soil. Prior to sequential application, individual batch washing and oxidation experiments were performed to select the most suitable agents.

2. Materials & methods

2.1. Soil sample

Soil co-contaminated with diesel, Cu, and Pb was sampled from a military site in Korea. Air-dried soil with a particle size <2 mm after sieving was used for all experiments. Soil moisture content was analyzed by weight loss at 105 °C for 24 h. Soil texture and organic matter were determined by ASTM D421 and D2974, respectively. Soil pH of a 1:5 w/v mixture of soil-deionized (DI) water mixture prepared according to Korean Standard Test Method (KSTM ES 07302) was measured using a pH/ISE meter (Istek, 735P, Korea). Cation exchange capacity (CEC) of soil was determined by method 9081 of USEPA SW-846 (USEPA, 1986). TPH was extracted from soil according to KSTM ES 07552. A mass of 10.0 g of soil was extracted with 50 mL of dichloromethane by ultrasonication. The mixture was concentrated using a rotary evaporator. The concentration of TPH in the concentrate was measured by a gas chromatograph fitted with a flame ionization detector (GC/FID, Varian, CP-3900, USA). Metals in soil were extracted by *aqua regia*, and the extract was filtrated using 5B filter paper. The filtrate was used to analyze metal concentrations in soil using inductively-coupled

plasma optical emission spectroscopy (ICP-OES, Agilent, 720-ES, USA). The concentration of heavy metals and TPH in untreated soil was averaged across 10 samples due to the significant heterogeneity of the field soil sample.

Fractionation of heavy metals in soil was determined by the standards, measurement, and testing program (SM&T) three-step sequential extraction procedure, which is summarized in Table 1. This procedure involves sequential extraction of trace metals by specific conditions and chemicals, and consists of four fractions: bound to soluble and exchangeable fraction (F1), bound to Fe-Mn (oxy)hydroxides fraction (F2), bound to organic matter and sulfides fraction (F3), and residual (F4) (Bacon and Davidson, 2008; Quevauviller et al., 1997; Rauret et al., 1999).

2.2. Batch washing and oxidation

A mass of 20 ± 0.001 g of soil sample was added into 250 mL Erlenmeyer flasks filled with 40 mL of each washing solution. Oxidants including 3% hydrogen peroxide (H_2O_2), 0.1 M potassium permanganate (KMnO_4), and oxone (potassium peroxymonosulfate; $\text{HKO}_5\text{S} \cdot 0.5\text{HKO}_4\text{S} \cdot 0.5\text{K}_2\text{O}_4\text{S}$) were used for removal of TPH, and 0.1 M ethylenediaminetetraacetic acid (EDTA) and citric acid and inorganic acids including 0.1 M hydrochloric acid (HCl), nitric acid (HNO_3), and sulfuric acid (H_2SO_4) were used as extracting agents for metals. Mixtures were shaken in a horizontal shaker at 150 rpm at 20 °C for 60 min. After treatment, mixtures were centrifuged at 5500 rpm for 5 min, and filtered through 5B filter paper. Residual soil was used for further analysis. All experiments were carried out in duplicate and average values were calculated.

2.3. Sequence of extraction and oxidation

We evaluated the removal efficiency of metals and TPH from soil according to various sequences of application of chemical extraction and oxidation processes. Three different cases were evaluated: extraction after oxidation, oxidation after extraction, and simultaneous oxidation and extraction using 3% H_2O_2 and 0.1 M EDTA. A mixture of 20 ± 0.001 g of soil and 40 mL of extracting/oxidizing solution was shaken in a horizontal shaker at 150 rpm at 20 °C for 30 min. After washing, analysis was performed as described previously.

3. Results and discussion

3.1. Screening of oxidants on batch tests

Table 2 shows the initial concentrations of contaminants and physicochemical properties of co-contaminated soil. The initial concentration of TPH, Cu, and Pb in untreated soil was approximately 3,300, 230, and 450 mg/kg, respectively, which greatly exceeds Korean regulation levels for soil quality.

Fig. 1 shows the efficiency of removal of TPH and heavy metals from soil after batch treatment using various oxidants for TPH and extractants for heavy metals. Oxidation using oxone, H_2O_2 , and KMnO_4 removed approximately 53.2%, 72.9%, and 67.5% of TPH, respectively (Fig. 1(a)). Among these oxidants, H_2O_2 is a well-known oxidant of hydrocarbons, and generates hydroxyl radicals with a redox potential of 2.8 V via the following reactions (Neyens and Baeyens, 2003; Ojinnaka et al., 2012; Watts and Teel, 2005) for *in situ* soil and groundwater remediation



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